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SCIENTIFIC COMMITTEE

Pablo Arenillas, CNEA, Argentina
Dirk Arnold, PTB, Germany
Christophe Bobin, LNE-LNHB, France
Philippe Cassette, LNE-LNHB, France
Jeffrey Cessna, NIST, USA
Teresa Crespo, CIEMAT, Spain
Pierino De Felice, ENEA, Italy
Eduardo García-Toraño, CIEMAT, Spain
Arvic Harms, IAEA, International
Yoshio Hino, NMIJ/AIST, Japan
Mikael Hult, EC-JRC-IRMM, EU
Simon Jerome, NPL, UK
Lisa Karam, NIST, USA
John Keightley, NPL, UK
Mark Kellett, LNE-LNHB, France
Matjaž Korun, IJS, Slovenia
Karsten Kossert, PTB, Germany
Marie-Christine Lépy, LNE-LNHB, France
Aurelian Luca, IFIN-HH, Romania
Franz Josef Maringer, BEV, Austria
Xavier Mougeot, LNE-LNHB, France
Tae Soon Park, KRISS, Korea
Stefaan Pommé, EC-JRC-IRMM, EU
Guy Ratel, BIPM, International
Octavian Sima, Univ. Bucharest, Romania
Michael Unterweger, NIST, USA
Freda van Wyngaardt, ANSTO, Australia
Uwe Wätjen, IRMM, Belgium
Mike Woods, IRMC, UK
Akira Yunoki, NMIJ/AIST, Japan
Brian Zimmerman, NIST, USA

CONFERENCE ORGANISATION

Scientific Secretary
(abstracts, manuscripts)

Pablo Arenillas
Comisión Nacional de Energía Atómica
Presbítero Juan Gonzáles y Aragón N°15,
Ezeiza, Argentina
Phone: +54 11 4125 8218
e-mail: science@icrm2017.com

Local Organizing Committee
(CNEA- Argentina)

Pablo Arenillas
Juan Carlos Furnari
Amanda Iglicki
Christian Balpardo
Gabriela Cerutti
Claudia Guardo
Melania Kikuchi
Daniela Guevara Vallese
Silvia Chinen
Carlos Demasi
Mariano Solari

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WELCOME MESSAGE

It is a great pleasure to welcome you to the 21st International Conference of the International Committee for Radionuclide Metrology. The conference takes place at Hotel Panamericano in the city centre of Buenos Aires, Argentina, from 15th to 18th of May 2017.

The goal of this international scientific conference is the exchange of information on the development of techniques and applications of radionuclide metrology, and to encourage international co-operation. This biennial conference was recently held in June 2015 in Vienna, Austria (ICRM2015).

The conference will include oral and poster presentations and business meetings of the ICRM Working Groups. After a peer review process, selected conference papers will be published in Applies Radiation and Isotopes.

Additional activities like the General Meeting of ICRM delegates and social events for accompanying persons are offered.

Again, welcome to ICRM2017 and thank you for attending!



Dirk Arnold
President ICRM



Pablo Arenillas
2017 Scientific Secretary

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THE VENUE: HOTEL PANAMERICANO

Located in the city center, opposite to the obelisco, the Teatro Colón and Avenida 9 de Julio. One block from the mythical Corrientes Avenue, house of the most famous theaters and shows in the country. Just a 10 minutes walk from the fashionable Recoleta.

Address: Carlos Pellegrini 551, Buenos Aires, Argentina

Phone: (+54 11) 4348 5000



BUENOS AIRES: THE CITY

Santa María del Buen Ayre, founded by Spain in the sixteenth century, forced its way by ambition and strength, becoming since the late nineteenth century in a cultural reference in South America. In its suburbs the tango was born, Intangible Heritage of Humanity, present in each of its bars, cafes and "bodegones", where the table is served with the incomparable meat of the pampas and local Malbec wines.

It has one the largest concentration of bookstores and theaters worldwide, many of them open almost all night, every night. It has the most intense sports experience of the world in his football derbies, with the echo of Maradona and Messi in the grandstands. Today receives college students across the continent because of its widespread, free and excellent Universidad de Buenos Aires, which gave Argentina four Nobel Prizes of science and peace.

City where the Pope Francis and the Queen Máxima of the Netherlands were born, also saw the Madres de Plaza de Mayo demand for their missing children. This diversity characterizes its population, native and immigrant, latinamerican, european and asian, who knows hospitality and courtesy despite being among the twenty most populated in the world. Its expression is the architectural eclecticism of the hundred neighborhoods ("barrios") of the city: from the little and colorful Italian-fashioned houses in La Boca, to the Museum Mile, with avenues of Madrid style and Parisian palaces.

For booking assistance: icrm2017@armotour.com.ar

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<table>
<tr><th>Sunday 14 May</th><th>Monday 15 May</th><th>Tuesday 16 May</th><th>Wednesday 17 May</th><th>Thursday 18 May</th><th>Friday 19 May</th></tr>
<tr><td rowspan="3"></td><td>Registration desk open</td><td>Registration desk open</td><td>Registration desk open</td><td></td><td rowspan="3"></td></tr>
<tr><td rowspan="3">Opening ceremony / Intro
International Metrology
Radionuclide Metrology in America
AIM Session
Coffee break
MSRM Session</td><td rowspan="3">GS Session
Coffee break
WG GS
ABS Session</td><td rowspan="3">RMT Session I
RMT Session II
Coffee break.
I Session
WG RMT</td><td>Registration desk open</td></tr>
<tr><td rowspan="2">ND Session I
Coffee break.
ND Session II
WG ND</td><td rowspan="3">ICRM General Meeting</td></tr>
<tr><td>Lunch</td><td>Lunch</td><td>Lunch</td><td>Lunch</td></tr>
<tr><td rowspan="3"></td><td rowspan="3">QA Session
RMLS Session
Coffee break
SP Session
WG RMLS</td><td>WG AS
WG BS
LSC Session
Coffee break.
WG LSC</td><td rowspan="3">Open Seminar</td><td rowspan="2">LL Session
WG LL
Coffee break.
Closing ceremony
ICRM Executive Board Meeting I</td><td rowspan="3">ICRM Executive Board Meeting II</td></tr>
<tr><td>Registration desk open</td><td>Informal reception</td></tr>
<tr><td></td><td>Reception</td><td></td><td>Tango class
Conference dinner</td></tr>
</table>

ABS	Alpha-particle and Beta-particle Spectrometry
AIM	Aspects of International Metrology
GS	Gamma-ray Spectrometry
I	Intercomparisons
LL	Low-level radioactivity measurement techniques
LSC	Liquid Scintillation Counting techniques
MSRM	Measurement Standards and Reference Materials
ND	Nuclear decay Data
QA	Quality Assurance and uncertainty evaluation
RMLS	Radionuclide Metrology in Life Sciences
RMT	Radionuclide Metrology Techniques
SP	Source Preparation techniques
WG	Working Group

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CONFERENCE DESCRIPTION

The goal of this international scientific conference is the exchange of information on the development of techniques and applications of radionuclide metrology, and to encourage international co-operation. This biennial conference was recently held in June 2015 in Vienna, Austria (ICRM2015).

The conference will include oral and poster presentations and business meetings of the ICRM Working Groups. After a peer review process, selected conference papers will be published in Applies Radiation and Isotopes. Additional activities for the General Meeting of ICRM members and social events for accompanying persons are offered.

CONFERENCE TOPICS

The conference is focused on developments in all fields of radionuclide metrology and activity measurement techniques including its applications. The scientific sessions of the conference are:

- Aspects of international metrology
- Intercomparisons
- Radionuclide metrology techniques
- Liquid scintillation counting techniques
- Source preparation techniques
- Measurement standards and reference materials
- Alpha-particle spectrometry
- Beta-particle spectrometry
- Gamma-ray spectrometry
- Nuclear decay data
- Radionuclide metrology in life sciences
- Low-level radioactivity measurement techniques
- Quality assurance and uncertainty evaluation in radioactivity measurements

LIABILITY

The 21st International Conference on Radionuclide Metrology and its Applications or the local organizing committee or the Comisión Nacional de Energía Atómica or the Hotel Panamericano cannot be held responsible for any personal injury, loss, damage, accident to private property or additional expenses incurred as a result of delays or charges in air, rail, sea, road or other services, strikes, sickness, weather or any other cause. Participants are advised to have their own insurance.

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SOCIAL PROGRAMME

The local organizing committee is glad to offer the participants the following social programme:

EVENT	DATE / TIME
Informal get-together	Sun 14th May, 17 hs
Reception	Mon 15th May, 20 hs
Tango Class / Tango Show Dinner	Wed 17th May, 20hs



We are enjoying a Tango Class before the Conference Dinner at Piazzolla Tango located in Florida 165.

ACCOMPANYING PERSONS SOCIAL PROGRAMME

Itinerary Tuesday 16th.

We will visit the historical part of Buenos Aires City, from Plaza de Mayo and Manzana de las Luces, going on to Avenida de Mayo till the Congreso (Parliament), where all the historical events take place. After that we will enter in the Buenos Aires of the 1810, visiting San Telmo neighbourhood, with its typical houses, and cobblestones streets. Our tour will end in the most colourful neighbourhood, La Boca. We will take a walk around and discover the typical Conventillos, and the life that used to have this part of the city in 1930.

Itinerary Thursday 18th.

We will enter in to the aristocracy of Buenos Aires starting at Recoleta NeighbourWe will enter in the aristocracy of Buenos Aires starting at Recoleta neighbourhood, where is situated one of the 3 cemetery museums around the world. Also there
we will visit Evita's tomb, and the church of Nuestra Señora del Pilar. Going on to the north we will walk around the Palermo park and know other part of Buenos Aires, the most trendy one, Palermo Soho and Hollywood, full of design shops, and bars.

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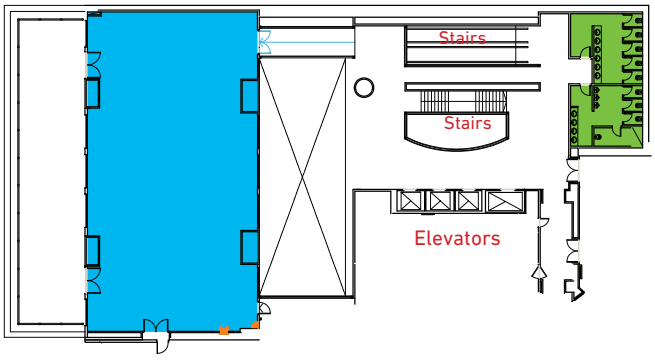
CONFERENCE VENUE MAP

Mezzanine Everyday Lunch & Reception (Mo, 15th Apr 20hs)



1st Basement



References

- Toilets
- Salón Río Paraná: Conferences
- Salón Los Jardines: Everyday Lunch & Reception (Mo, 15 Apr 20hs)
- Salón Río Colorado: Reviews
- Salón Amazonas: Posters & Exhibitions

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BUENOS AIRES METROPOLITAN AREA MAP



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BUENOS AIRES SUBWAY MAP



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DETAILED PROGRAMME

Sunday, 14th May 2017

17:20 – 20:00

Registration desk open
&
Informal reception

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Monday, 15th May 2017

8:20	**Registration desk open**

9:00	**Open Ceremony**	
9:20	International Metrology. The meter convention and the Inter-american metrology system *Laiz, H.*	
9:40	Radionuclide Metrology in America *Karam, L.*	

Session: Aspects of International Metrology *Chairpersons: Ratel, G.; Karam, L.*		
10:00	Is decay constant? *Pommé, S.; et al*	O-008
10:20	Methodology for activity comparison of thorium-227 via the international reference system (SIR) *Keightley, J. D.; Pearce, A. K.; Pommé, S.; Michotte C.*	O-094
10:40	**Poster presentation: AIM** *Chairpersons: Ratel, G.*	
On the comparison of radon standard using ionization chambers: an attempt to improve the reliability of the SIR response for radioactive gas *Pierre, S.; Sabot, B.; Cassette, P.; Liang, J.; Le Garrérès, I.; Rigoulay, F.; Courte, S.; Ratel, G.; Lourenço, V.; Ferreux, L.*		P-057

10:45	**Coffee break and posters**

Session: Measurement Standards and Reference Materials *Chairpersons: Karam, L.; Harms, A.*		
11:15	Isolation and purification of Protactinium-231 *Jerome, S. M.; Ivanov, P.; Russell, B. C.; Happel, S.*	O-037
11:35	New Thorium-229 amount content standard reference material *Essex, R.; Mann, J.; Williams, R.; Kinman, W.; Hubert, A.; Bennet, M.*	O-077
12:05	Interlaboratory comparison of a new norm reference material obtained from treatment of drinking water *Wiedner, H.; Maringer, F. J.*	O-155
12:25	**Poster presentation: MSRM** *Chairpersons: Karam, L.*	
Analysis of actinides in the insoluble residues after decomposition by various techniques *Benedik, L.; Trdin, M.*		P-081
Homogeneity of wheat flour in 5 ml containers for certificated reference materials *Furukawa, R.; Unno, Y.; Miura, T.; Yunoki, A.; Hachinohe M.; Hamamatsu, S.*		P-084
Production of spiked vegetation samples containing gamma-emitting radionuclides to participate in intercomparison tests *Santos de Souza, P.; Clain, A. F.*		P-138

12:35	**Lunch**

	Conference Photograph

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Monday, 15th May 2017

Session: Quality Assurance and uncertainty evaluation *Chairpersons: Korun, M.; Pibida, L.*		
14:00	Proficiency test exercises for CTBT particulate radionuclide laboratories *Nakashima, N.; Duran, E. B.; Auer, M.*	O-074
14:20	A review of the TAEK proficiency test on natural and anthropogenic radionuclides activities in black tea *Yeltepe, E.; et al*	O-142
14:40	Results of a regional measurement comparison exercise for I-131 and Y-90 using radionuclide calibrators *Oropesa, P.; Arenillas, P.; De Oliveira, E. A.; Oyarzun, C. H.; García, L.; Guardo, C.; Iwahara, A.; da Cruz, P. A. L.; da Silva, C. J.; Moreno, Y.*	O-143
15:00	**Poster presentation: QA** *Chairpersons: Korun, M.*	
Systematic influences on the areas of peaks in gamma-ray spectra, having a large statistical uncertainty *Korun, M.; et al*		P-005
An alternative approach to the decision threshold *Korun, M.; Vodenik, B.; Zorko, B.*		P-024
Determining acceptable limits of uncertainties in urinary ^{238}U measurements by using the new LTUM method *Guichet, C.; Lecoix, G.; Hurtgen, C.*		P-039
Comparison of intercomparison results of gamma ray spectrometry of spiked and real samples *Glavič-Cindro, D.*		P-041
Rapid and accurate assessment of the activity measurements in Brazilian hospitals and clinics *de Oliveira, A. E.*		P-151

Session: Radionuclide Metrology in Life Sciences *Chairpersons: Cessna, J. T.; Yunoki, A.*		
15:15	Assessing the absolute quantitative accuracy of positron emission tomography for Cu-64 using traceable calibrated phantoms *Zimmerman, B. E.; Bergeron, D. E.; Edgerton, J. P.*	O-002
15:35	Quantitative imaging, dosimetry and metrology; where do national metrology institutes fit in? *Fenwick, A. J.; Merrett, J. L.; Ferreira, K. M.; Robinson, A. P.*	O-146
16:00	**Poster presentation: RMLS** *Chairpersons: Cessna, J. T.; Yunoki, A.*	
Utilizing normalized manufacturer's calibration factors for an ionization chamber with depleted gas *Lubbe, J.; Simpson, B. R. S.; van Staden, M. J.; van Rooy, M. W.*		P-016
Development of the Australian standard for Ge-68 by two liquid scintillation counting methods *van Wyngaardt, W.M.; Smith, M. L.; Jackson, T. W.; Howe, B.; Reinhard: M. I.*		P-022
Impurities in Tc-99m radiopharmaceutical solution obtained from Mo-100 in cyclotron *Tymiński, Z.; Saganowski, P.; Pawlak, D.; Wojdowska, W.; Dziel, T.; Kołakowska, E.; Listkowska, A.*		P-042

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Monday, 15th May 2017

A movable precision ionization chamber: the transfer ionization reference chamber *Juget, F.; Nedjadi, Y.; Buchillier, T.; Duran, T.; Bochud, F.; Bailat, C.*	P-088
Extended utilization of a ^{68}Ge/^{68}Ga reference source as a mock ^{18}F source in the ionization chamber calibration *Yamada, T.; Ishizu, H.*	P-073
An EGSNRC model for the Vinten ionization chamber *Galea, R.; Townson, R.; Tessier F.; El Gamal, I.*	P-105
^{99m}Tc by ^{99}Mo produced at the ENEA-FNG facility of 14 Mev neutrons *Capogni, M.; De Felice, P.; Fazio, A.; Quintieri, L.; Pietropaolo, A.; Pillon, M.; Pizzuto, A.*	P-110
Iodine-123 intercomparison exercises: assess measurement capability in the UK hospitals *Ferreira, K.; Fenwick, A.*	P-112
Measurement of ^{18}F activity by NaI(Tl) well type scintillation spectrometer *Vukanac, I.; Kandić, A.; Djurašević, M.; Šešlak, B.; Ciraj Bjelac, O.; Antić, V.*	P-125
Ga-68 activity calibrations for nuclear medicine applications in Cuba *Oropesa Verdecia, P.; García Rodríguez, L.; Serra Águila, R. A.; Moreno León, Y.; Jénez Magaña Y., Cassette P.*	P-137
New and updated calibration factors for the NPL secondary standard radionuclide calibrator *Fenwick , A. J.; Ferreira, K. M.; Pearce, A. K.; Keightley, J. D.*	P-148

16:20	**Coffee break and posters**

Session: Source Preparation techniques *Chairpersons: García-Toraño, E.; Jerome, S.*		
17:20	Qualification of a precision pattern dispenser *Van Ammel, R.; et al*	O-020
17:40	**Poster presentation: SP** *Chairpersons: García-Toraño, E.*	
Verification of ^{137}Cs determination in large volume seawater using Cu-Hexacyanoferrate cartridge filters *Visetpotanakit, S.; Khrautongkieo, C.*		P-011
Activity standardization of ^{210}Pb by liquid scintillation counting method *Havelka, M.*		P-097
The method of production and quality control of volume multigamma sources with different matrices *Listkowska, A.; Lech, E.; Saganowski, P.; Tymiński, Z.; Ziemek, T.; Dziel, T.; Kołakowska, E.*		P-101

17:45	**Working Group: Radionuclide Metrology in Life Sciences**

18:45	**End of Conference day 1**

20:20 22:00	Reception

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Tuesday, 16th May 2017

8:20	**Registration desk open**

	Session: Gamma-ray Spectrometry *Chairpersons: Sima, O.; Maringer, F. J.*	
9:00	Consistency of photon emission intensities for efficiency calibration of gamma-ray spectrometers in the energy range from 20 keV to 80 keV *Lépy, M.C.; Ménesguen, Y.; Riffaud, J.*	O-058
9:20	On homogeneity approximation in calibration of gamma-spectrometry assessment of bulk samples *Sima, O.*	O-067
9:40	Determination of the absolute photon emission intensities of some gamma rays of Ho-116m *Peyrés,V.*	O-133
10:00	**Poster presentation: GS** *Chairpersons: Sima, O.; Maringer, F. J.*	
	Determination of a HPGe detector efficiency curve by Monte Carlo simulation in the energy interval from 20 to 1408 keV *Haoran, L.; Zhang Jian, Z.; Zeshu, L.; Juncheng, L.*	P-012
	Determination of the neutron activation profile of core drill samples by gamma-ray spectrometry *Gurau, D.; S. Boden, S.; Sima, O.; Stanga, D.*	P-014
	Impact evaluations of activity measurement results for clearance operations *Yeh , C. H.; Yuan, M. C.*	P-021
	GEANT4 based Monte Carlo simulation for verifying the modified sum-peak method *Aso, T.; Ogata, Y.; Makino, R.*	P-034
	Well type vs coaxial type HPGe detector. The significance of self-absorption corrections in Pb-210 geochronology *Iurian, A. R.; Millward, G.; Sima, O.; Taylor, A.; Blake, W.*	P-043
	Portable real time in-situ gamma-ray analysis system *Wei, C.; et al.*	P-047
	Determination of absolute peak and total efficiency functions for Ge-detectors by iteration calculation using a mixed radionuclide source *Ishizu, H.; Yamada, T.*	P-048
	Subtraction of natural radionuclides contribution from gamma spectra measured by HPGe detector *Petr Kovar, A.; Jaroslav Solc, B.*	P-064
	Determination of ^{134}Cs activity by the sum-peak method via a well type of Ge detector *Ogata, Y.; Itadzu, H.; Kojima, S.*	P-066
	Performance of the segmented HPGe detector in kriss *Han, J.; Lee, K. B.; Lee, J. M.; Lee, S. H.;Park, T. S.; Oh, J. S.*	P-068
	Gamma-ray spectrometry analysis software environment *Lutter, G.; Hult, M.; Tzika, F.; Stroh, H.; Marissens, G.*	P-083
	Improvement of the activity measurement method for solid dosimeters emitting x-rays *Domergue, C.; Riffaud, J.; Philibert, H.; Destouches, C.; Thiollay, N.; Girard, J. M.; Carcreff, H.; Vigneau, O.; Gravier, L.*	P-103
	Uncertainty of determination of Tb-158 in the RBMK nuclear reactor waste *Plukis, A.; Barkauskas, V.; Druteikienė, R.; Duškesas, G.; Germanas, D.; Gudelis, A.; Juodis, L.; Plukienė, R.; Remeikis, V.*	P-106
	SUMCOR: cascade summing correction for volumetric sources applying MCNP6 *Dias, M. S.; Semmler, R.; Moreira, D. S.; Koskinas, M. F.*	P-114

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Tuesday, 16th May 2017

	Coincidence summing corrections for volume ^{60}Co sources measured by scintillation NaI(Tl) spectrometers *Kandić, A.; Šešlak, B.; Vukanac, I.; Djurašević, M.; Jevremović, A.*	P-127
10:30	**Coffee break and posters**	
11:00	**Working Group: Gamma-ray Spectrometry**	
	Session: Alpha-particle and Beta-particle Spectrometry *Chairpersons: Pommé, S.; Mougeot, X.*	
12:00	Activity determination of ^{60}Co and the importance of its beta spectrum *Kossert, K.; Marganiec-Gałązka, J.; Mougeot, X.; Nähle, O.*	O-006
12:20	Concept design of a time-of-flight spectrometer for the measurement of the energy of alpha particles *García-Toraño, E.*	O-013
12:40	A new code for improved calculations of electron capture transitions *Mougeot, X.*	O-051
13:00	**Poster presentation: ABS** *Chairpersons: Pommé, S.*	
	Relative conversion electron emission probabilities of Pu isotopes derived from ice spectrometry *Pommé, S.; Pöllänen, R.; Paepen, J.*	P-009
	The case of long-range alpha particles from ^{212}Po revisited *Sánchez, A. M.*	P-096
	Alpha-particle spectrometry: the importance of measurement setup conditions and sample preparation in nuclear decay data measurements *Dirican, A.*	P-134
13:10	**Lunch**	
14:10	**Working Group: Alpha-particle Spectrometry**	
14:40	**Working Group: Beta-particle Spectrometry**	
	Session: Liquid Scintillation Counting techniques *Chairpersons: Kossert, K.; Zimmerman, B.*	
15:10	Activity determination of Ge/Ga by means of 4π(č) β-γ coincidence counting *Marganiec-Gałązka, J.; Kossert, K.; Nähle, O.*	O-007
15:30	Absolute standardizations of ^{99m}Tc and ^{57}Co by 4πβγ liquid scintillation counting for SIRTI and SIR comparisons *van Rooy, M. W.; van Staden, M. J.; Simpson, B. R. S.; Lubbe, J.*	O-017
15:50	Standardization of ^{68}Ge/^{68}Ga using the 4πβ-γ coincidence method based on Cherenkov counting *Bobin, C.; Thiam, C.; Bouchard, J.*	O-052
16:10	Comparison of calculation of the activity of a H-3 source measured by the LSC-TDCR method *Cassette, P.; et al*	O-053

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Tuesday, 16th May 2017

16:30	**Poster presentation: LSC** *Chairpersons: Kossert, K.; Zimmerman, B.*	
	IFIN-HH progress in tritium standardization by the LSC-TDCR method, proved in international comparisons *Antohe, A.; Sahagia, M.; Luca, A.; Ioan, M. R.; Cassette, P.*	P-023
	Standardization of Praseodymium-142 activity concentration using CIEMAT/NIST tritium efficiency tracing method *Yücel, H.; Yeltepe, E.*	P-036
	Diffusion lengths and partition coefficients of ^{131m}Xe and ^{85}Kr in MAKROFOL N and MAKROFOL DE *Mitev, K.; Cassette, P.; Tartès, I.; Georgiev, S.; Dimitrova, I.; Pressyanov, D.*	P-050
	A new 4π(LS)-gamma coincidence system using the TDCR system in the beta channel at CMI *Sochorová, J.; Auerbach, P.*	P-075
	Use of the TDCR method for the standardization of radionuclides in the FTMC *Gudelis, A.; Gaigalaitė, L.; Gorina, I.; Butkus, P.*	P-107
	Ionic liquids as solvents for LSC: Pyranine as wavelength shifter to enhance Cherenkov-light detection *Mirenda, M.; Ferreyra, C.; Rodrigues, D.; Arenillas, P.; Sarmiento, G.; Krimer, N.*	P-122
	Standardization of Fe-55 by TDCR efficiency fitting using EGS5MPI Monte Carlo code *Sato, Y.*	P-139

16:45	**Coffee break and posters**

17:40	**Working Group: Liquid Scintillation Counting techniques**

18:40	**End of Conference day 2**

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Wednesday, 17th May 2017

8:20	**Registration desk open**

	Session: Radionuclide Metrology Techniques *Chairpersons: Keightley, J.; Park, T. S.*	
9:00	Monte Carlo modeling of live-timed anticoincidence counting (LTAC) for Cu-64 *Bergeron, D. E.; Fitzgerald, R.*	O-031
9:20	Activity determination of ^{67}Ga using 4πβ–γ coincidence counting *Nähle, O.*	O-045
9:40	Investigation of γ-γ coincidence counting using the National Nuclear Array (NANA) as a primary standard *Collins, S. M.; Shearman, R.; Keightley, J. D.; Regan, P. H.*	O-100
10:00	**Poster presentation: RMT (I)** *Chairpersons: Keightley, J.; Park, T. S.*	
	Standardization of ^{67}Cu and calibration of the ionization chamber impurities and decay scheme problems *Sahagia, M.; Luca, A.; Ioan, M. R.; Antohe, A.; Ivan,C.*	P-004
	Measurement for accurate extrapolation in 4πβ-γ coincidence counting method using plastic scintillator *Unno, Y.; Sanami, T.; Sasaki, S.; Hagiwara, M.; Yunoki, A.*	P-082
	68(Ge+Ga) activity standardization by 4πβ(LS)-γ(NaI(Tl)) anticoincidence counting measurements *da Silva, C. J.; da Cruz, P. A. L.; Iwahara, A.; Loureiro, J. S.*	P-087
	Disintegration rate and gamma ray probability per decay measurement of Cu-64 *Yamazaki, I. M.; Koskinas, M. F.; Moreira, D. S.; Takeda, M. N.; Dias, M. S.*	P-089
	^{134}Cs activity standardization by 4π (LS)-γ(NaI(Tl)) anticoincidence counting and submission to international reference system *dos S. Gomes, R.; da Silva, C. J.; da Cruz, P. A. L.; Iwahara, A.; Loureiro, J. S.*	P-130
	Russian national primary activity standard extensive renewal, first results *Alexeev, I.; Kharitonov, I.; Sepman, S.; Shilnikova, T.; Tereschenko, E.; Trofimchuk, S.; Zanevsky, A.; Zhukov, G.*	P-044
	Standardization of Cd-109 by three methods *Yuan, M. C.; Yeh, C. Y.; Chu, W. H.*	P-027
	Different techniques for ^{198}Au activity measurements in gold foils *Tereshchenko, E.; Moiseev, N.; Didyk A.*	P-070
	"Influence method", a useful tool for radiation measurements *R.E. Mayer, R. E.; Rios, I. J.*	P-156
	Measurements of response-ratio of ionization chamber flowed with dry air to with P-10 gas in the calibration of gas monitors *Yunoki, A.; Kawada, Y.; Hino, Y.*	P-061
	Development of the internal gas proportional counting system of the beta radioactive gas *Hwang, S.; Lee, B. J. M.; Lee, K. B.; Park, T. S.*	P-120
10:20	**Poster presentation: RMT (II)** *Chairpersons: De Felice, P.; Bobin, C.*	
	Fast digital 4πβ-4πγ coincidence counting with offline analysis at IRA-METAS *Durán, M. T.; Nedjadi, Y.; Juget, F.; Bochud, F.; Bailat, C.*	P-080
	Standardization of ^{67} Ga by digital coincidence counting *Balpardo, C.; Arenillas, P.; Roteta, M.*	P-118
	A new digital 4πγ system with a NaI well-type detector at LMR-CNEA *Rossi, M.; Balpardo, C.; Arenillas, P.; Cerutti, G.; Ferrari, M. C.*	P-149
	Revisiting the standardization of ^{134}Cs: updated ionization chamber calibration factors and new measurements of absolute γ-ray emission intensities *Collins, S. M.; Keightley, J. D.*	P-095

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Wednesday, 17th May 2017

Absolute standardization of ^{54}Mn by sum-peak method using HPGe detectors *de Araújo, M. T. F.; et al*	P-098
Standardization of ^{133}Ba by sum-peak method *da Silva, R. L.; et al*	P-109
Application of the sum-peak method to activity standardizations of ^{152}Eu sources in LNMRI(BR) *Ferreira Filho, A.; et al*	P-115
Metrology for decommissioning nuclear facilities – outcomes of joint research project within the European metrology research program *Šuráň, J.; et al*	P-128
Development of the absolute standardization apparatus for Radon-222 activity. *Liang, J. C.; Wang, L. Y.; Li, Z. X.*	P-38
Activity measurement of 222Rn gas for a key comparison. *Kim, B.J.; Lee, J.M.; Lee, K.B.; Park, T.S.; Kim, B.C.*	P-141
The large-area multi-wire proportional counter for 2pi alpha and beta emission measurement at the NIM China *Zhang, M.; Zhang, J. W.; Duan, L. M., Tu, Z.; Yang, Z. J.; Liang, J. C.; Liu, H. R.*	P-1
Efficiency transfer method applied to surface beta contamination measurements. *Stanga,D. ; De Felice, P.; Capogni, M.*	P-15
Large area alpha sources with a LIP: integral counting and spectral distortions. *Fitzgerald, R.; King, L.*	P-117

10:45	**Coffee break and posters**

	Session: Intercomparisons *Chairpersons: G. Ratel; L. Karam*	
11:30	Comparison of ^{131}I activity measurements at the NCBJ RC POLATOM and the ENEA-INMRI linked to the BIPM SIR system *Ziemek,T.; Capogni, M.; Ratel, G.; Broda, R.; Dziel,T.; Fazio, A.; Listkowska, A.; Kołakowska, E.*	O-28
11:50	Validation of the extension of the SIR to beta emitters. *Ratel,G. ; Los Arcos, J.M.; Laureano-Perez, L.; Rodríguez-Barquero, L.; Courte,S.*	O-86
12:10	Results of an international comparison of activity measurements of ^{68}Ge/^{68}Ga *Cessna,J.T. ; et. al.*	O-104

12:30	**Working Group: Radionuclide Metrology Techniques**

14:00	**End of Conference day 3**

14:30	**Open Seminar by Canberra-Mirion** ISOCS/LabSOCS methodology True coincidence summing SAGe Well New features in Genie Cosmic Guard Transportable Radiation Analysis System

20:00 23:00	**Tango class & Conference Dinner at Piazzolla Tango**

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Thursday, 18th May 2017

9:00	**Registration desk open**	

Session: Nuclear decay data (I) *Chairpersons: M. Kellett; Y. Hino*		
9:20	Differences in published half-life values for ^{207}Bi. *Pibida, L.; King, L. E.*	O-3
9:40	Direct measurement of the electron capture probability ratios of Fe-55. *Loidl,M.; Rodrigues, M.*	O-56
10:00	Measurement of 103mRh x-ray emission intensities and evaluation of the decay scheme. *Riffaud,J. ; Cassette, P.; Lacour, D.; Lourenço, V.; Tartès, I; Kellett, M.A.; Corbel, M.; Lépy,M.-C.; Domergue, C.; Destouches, C.; Carcreff, H.; Vigneau, O.*	O-59

10:20	**Poster presentation: ND (I)** *Chairpersons: M. Kellett*	
	Study of ^{243}Am decay. *Caro Marroyo, B.; Martín Sánchez, A.; Jurado Vargas, M.; García-Toraño, E.; Roteta, M.*	P-65
	Two determinations of the Ge-68 half-life. *Bergeron, D.E.; Cessna, J.T.; Zimmerman, B.E.*	P-116
	Standardization and half-life of Zr-89. *García-Toraño,E. ; Roteta, M.; Peyrés, V.; Mejuto, M.; Sánchez-Cabezudo, A.;Romero, E.*	P-124
	^{230}U nuclear decay data evaluation. *Luca, A.; Ioan, M.R.*	P-140
10:30	**Coffee break and posters**	

Session: Nuclear decay data (II) *Chairpersons: S. Pommé, E. García-Toraño*		
11:10	Recommendations for new decay data measurements arising from the decay data evaluation project (DDEP). *Kellett, M.A.*	O-54
11:30	Precise test of internal-conversion theory: transitions measured in nine nuclei spanning 45 ≤ Z ≤ 78. *Hardy, J.C.; Nica, N.; Iacob, V.E.; Trzhaskovskaya, M.B.*	O-85
11:50	**Poster presentation: ND (II)** *Chairpersons: S. Pommé*	
	Yields measurement of photons 186,21 keV and KX (RN) in Ra-226 decay. *Dryák, P.; Havelka, M.; Mazánová, M.*	P-18
	Decay data for the positron emission tomography imaging radionuclide I-124: a DDEP evaluation. *Zimmerman, B. E.*	P-32

11:55	**Working Group: Nuclear decay data**	

12:55	**Lunch**	

Session: Low-level radioactivity measurement techniques *Chairpersons: M. Hult, D. Arnold*		
14:00	Evaluation of the Radon and Thoron interference on the performance of the compact aerosol monitoring device. *Glavič–Cindro, D.; Petrovič, T.; Brodnik, D.; Vencelj, M.; Korun, M.; Bell, S. J.; Keightley, L.; Woods, S.*	O-33

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Thursday, 18th May 2017

14:20	Characterization of an ultra-low background point contact HPGe-well detector. *Hult, M.; Marissens, G.; Stroh, H.; Lutter, G.; Tzika, F.*	O-69
14:40	Low-level laboratory measurement of Radio-Xenon: beta-gamma versus gamma measurements. *Delaune,O.; Cagniant, A.; Douysset, G.; Fontaine,J.-P.; Gross, P.; Le Petit, G.*	O-78
15:00	**Poster presentation: LL** *Chairpersons: M. Hult, D. Arnold*	
	On the sequential separation and quantification of ^{237}Np, ^{241}Am, Th, Pu, and U isotopes in environmental and urine samples. *Vasile, M.; Jacobs, K.; Zhuxin, L.; Verrezen, F.; Bruggeman, M.*	P-46
	SPALAX ng: a breakthrough in RadioXenon field measurement. *Cagniant, A.; Topin, S.; Le Petit, G.; Delaune, O.; Gross, P.; Philippe, T.; Piwowarczyk, J.-C.; Douysset, G.*	P-79
	Determination of ^{22}Na activity in air and rainwater samples by gamma-ray spectrometry. *Savva, M.I.; Karangelos, M.J.; Anagnostakis, M.J.*	P-92
	Low-level direct measurement of U-238 in environmental water using state-of-the art gamma spectrometry. *Douysset, G.; Cagniant, A.; Delaune, O.; Greiner,V.*	P-93
	A complete analysis method of norm building materials by gamma spectrometry with HPGe detectors. *Quintana, B.; Santamaría, R.; Pedrosa, M.C.; Vázquez-Canelas, L.; Díaz-Rivera, N.; Sanjuan, M. A.; Puertas, F.; Gascó, C.*	P-126
	Measurements of environment samples in well-type detectors in the Modane Underground Laboratory. *de Vismes Ott, A.; Cagnat, X.; Gurriara, R.*	P-129
	Applications of low-level underground gamma-ray spectrometry at IAEA's Environment Laboratories. *Osvath, I.; Comanducci, J.F.; Fujak, M.; Harms, A.V.; Levy, I.; Mc Ginnity, P.; Pham,M.K.*	P-153
	Radon in drinking water: review and evaluation of activity measurement methods. *Wiedner, H.; Lotter, K.; Maringer, F. J.; Friedmann, H.*	P-154
	Natural radionuclide activity concentrations in soils and vegetation at an uranium mining site. *Charro, E.; Moyano, A.*	P-157
15:20	**Working Group: Low-level radioactivity measurement techniques**	
16:20	**Coffee break and posters**	
17:00	**Best poster award & Closing ceremony**	
17:45 20:45	**ICRM Executive Board Meeting I**	

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Friday, 19th May 2017

9:00 - 13:00	**ICRM General Meeting**
13:00	**Lunch**
14:00 16:00	**ICRM Executive Board Meeting II**

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PARTICIPANTS

Pablo Arenillas
National Atomic Energy Commission
Argentina
arenilla@cae.cnea.gov.ar

Yi – Chun Lin
INER
Taiwan, Province of China
jasmine@iner.gov.tw

Chin – Hsien – Yeh
INER
Taiwan, Province of China
chyeh@iner.gov.tw

Jubong Han
UST
Republic of Korea
Donbosco73@hanmail.net

Marcos Izarra
Nucleoelectrica Argentina sa
Argentina
mizarra@na-sa.com.ar

German Mosteiro
Nucleoelectrica Argentina sa
Argentina
gmosteiro@na-sa.com.ar

Ricardo Smitd
Nucleoelectrica Argentina sa
Argentina
rsmidt@na-sa.com.ar

Marcelo Jofre
Nucleoelectrica Argentina sa
Argentina
mjofre@na-sa.com.ar

Leonardo Verdún
Nucleoelectrica Argentina sa
Argentina
lverdun@na-sa.com.ar

Walter Ponce
Nucleoelectrica Argentina sa
Argentina
wponce@na-sa.com.ar

Paola Lujan Echeverría
Nucleoelectrica Argentina sa
Argentina
pecheverria@na-sa.com.ar

Verónica Bernetti
Nucleoelectrica Argentina sa
Argentina
vbernetti@na-sa.com.ar

Rita Beatriz Bejarano
Nucleoelectrica Argentina sa
Argentina
rbejarano@na-sa.com.ar

Pierino De Felice
Enea-Inmri
Italy
pierino.defelice@enea.it

Olivier Delaune
CEA
France
olivier.delaune@cea.fr

Guilhem Douysset
CEA
France
guilhem.douysset@cea.fr

Begoña Quintana Arnés
University of Salamanca
Spain
quintana@usal.es

Emin Yeltepe
TAEK-SANAEM
Turkey
emin.yeltepe@taek.gov.tr

Inés Marilina Basso
National Atomic Energy Commission
Argentina
basso@cae.cnea.gov.ar

Miguel Roteta
CIEMAT
Spain
miguelroteta@ciemat.es

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Eduardo García-Toraño
CIEMAT
Spain
e.garciatorano@ciemat

Raphael bekuochukwu udeogu
University of ibadan Nigeria
Nigeria
kollymaster1@gmail.com

Yasuhiro Unno
NMIJ/AIST
Japan
y.unno@aist.go.jp

Boshi Lian
Beijing Shucheng S&T Development Co. Ltd
China
bslian@sc-st.com

Zhaoli Shi
Beijing Shucheng S&T Development Co. Ltd
China
zlshi@sc-st.com

Roberto E. Mayer
National Atomic Energy Commission
Argentina
mayer@cab.cnea.gov.ar

Selva Bertola
National Atomic Energy Commission
Argentina
selvadepeche@hotmail.com

Youcef Nedjadi
Institute of Radiation Physics
Switzerland
youcef.nedjadi@chuv.ch

Rodrigo Amor
National Atomic Energy Commission
Argentina
rodrigoamor85@hotmail.com

Marianela Lobo
National Atomic Energy Commission
Argentina
marianela_sl@hotmail.com

Roberto Llovera
National Atomic Energy Commission
Argentina
llovera@cae.cnea.gov.ar

Gabriela Lujan Cerutti
National Atomic Energy Commission
Argentina
cerutti@cae.cnea.gov.ar

Esteban Cirello
National Atomic Energy Commission
Argentina
estebancirello@gmail.com

Maria Clara Ferrari
National Atomic Energy Commission
Argentina
mcferrari@cae.cnea.gov.ar

Claudia Guardo
National Atomic Energy Commission
Argentina
ccguardo@cae.cnea.gov.ar

Evgeny Taskaev
Eckert & Ziegler Analytics
United States
evgeny.taskaev@ezag.com

Dirk Arnold
Physikalisch-Technische Bundesanstalt
Germany
Dirk.Arnold@ptb.de

Michel Bruggeman
SCK-CEN Studiecentrium voor Kernenergie
Belgium
mbruggem@sckcen.be

Tsukasa Aso
National Institute of Technology, Toyama College
Japan
tsukasa.aso@gmail.com

Aleksandar Kandic
Institute of Nuclear Sciences "Vinca"
Serbia
akandic@vinca.rs

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Lisa Karam
NIST
United States
lisa.karam@nist.gov

Justyna Marganiec- Galazka
PTB
Germany
justyna.marganiec@ptb.de

Raphael Galea
National Research Council of Canada
Canada
raphael.galea@nrc-cnrc.gc.ca

Fabio Andre Ludolf Cacais
LNMRI/IRD
Brazil
facacais@gmail.com

Denise Simões Moreira
IPEN/CNEN
Brazil
denise.moreira@ipen.br

Marina Fallone Koskinas
IPEN - CNEN/SP
Brazil
koskinas@ipen.br

Andrew Robinson
National Physical Laboratory
United Kingdom
andrew.robinson@npl.co.uk

Sean Collins
National Physical Laboratory
United Kingdom
smc1@npl.co.uk

Mario Pablo Rossi
National Atomic Energy Commission
Argentina
mprossi@cae.cnea.gov.ar

Andrew Fenwick
National Physical Laborartory
United Kingdom
andrew.fenwick@npl.co.uk

John C. Hardy
Cyclotron Institute, Texas A&M University
United States
hardy@comp.tamu.edu

Rodolfo Gurriaran
IRSN
France
rodolfo.gurriaran@irsn.fr

Anne de VIsmes Ott
IRSN
France
anne.de-vismes@irsn.fr

Marios Anagnostakis
NED/NTUA
Greece
managno@nuclear.ntua.gr

Mauro Da Silva Dias
IPEN-CNEN/SP
Brazil
msdias@ipen.br

Maria Candida Moreira de Almeida
CNEN-IRD -RJ
Brazil
candida@cnen.gov.br

Sanghoon Hwang
Korea Research Institute of Standards and Science
Republic of Korea
shhwang@kriss.re.kr

Virginia Peyres
CIEMAT
Spain
virginia.peyres@ciemat.es

Miriam Tainá Ferreira de Araújo
Coppe - UFRJ
Brazil
miriamtaina01@gmail.com

Kelley Ferreira
NPL
United Kingdom
kelley.ferreira@npl.co.uk

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Uwe Wätjen
private, retired from EC-JRC-IRMM
Belgium
uwe.watjen@telenet.be

Franz Josef Maringer
BEV
Austria
franz-josef.maringer@bev.gv.at

Octavian Sima
University of Bucharest
Romania
Octavian.Sima@partner.kit.edu

Christian Balpardo
National Atomic Energy Commission
Argentina
balpardo@cae.cnea.gov.ar

Eliana Depaoli
National Atomic Energy Commission
Argentina
edepaoli@cae.cnea.gov.ar

Anna Listkowska
NCBJ RC POLATOM
Poland
a.listkowska@polatom.pl

Milton William van Rooy
NMISA
South Africa
mvrooy@nmisa.org

Joline Lubbe
NMISA
South Africa
jlubbe@nmisa.org

Yasushi Sato
National Metrology Institute of Japan
Japan
yss.sato@aist.go.jp

Freda van Wyngaardt
ANSTO
Australia
freda.vanwyngaardt@ansto.gov.au

Dario Pablo Rodrigues Ferreira Maltez
National Atomic Energy Commission
Argentina
darodrig@tandar.cnea.gov.ar

Jeffrey Morrison
Department of Homeland Security
United States
Statesjeffrey.morrison@hq.dhs.gov

Anderson Ricardo Leiras dos Santos
LMNRI/IRD/CNEN
Brazil
andersonleiras@hotmail.com

Andra-Rada Iurian
Plymouth University
United Kingdom
andra.iurian@plymouth.ac.uk

Zbigniew Tyminski
RC POLATOM
Poland
z.tyminski@polatom.pl

Hannah Wiedner
BEV
Austria
hannah.wiedner@bev.gv.at

Alejandro Martin Sanchez
University of Extremadura
Spain
ams@unex.es

Karsten Kossert
PTB
Germany
karsten.kossert@ptb.de

Ole Nähle
Physikalisch-Technische Bundesanstalt
Germany
ole.j.naehle@ptb.de

Miroslav Havelka
Czech Metrology Institute
Czech Republic
mhavelka@cmi.cz

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Krasimir Mitev
Sofia University St. Kliment Ohrdiski
Bulgaria
kmitev@phys.uni-sofia.bg

Yoshimune Ogata
Nagoya University
Japan
ogata.yoshimune@b.mbox.nagoya-u.ac.jp

Roberto Poledna
CNEN
Brazil
miriamtaina@hotmail.com

Ivan Daniel Viotto
Nucleoelectrica Argentina s.a.
Argentina
iviotto@na-sa.com.ar

Esteban Stoll
Nucleoelectrica Argentina s.a.
Argentina
estoll@na-sa.com.ar

Mauricio López
Nucleoelectrica Argentina s.a.
Argentina
maulopez@na-sa.com.ar

Jorge Francisco Rostagno
Nucleoelectrica Argentina s.a.
Argentina
jrostagno@na-sa.com.ar

Lorena Ana Zabala
Nucleoelectrica Argentina s.a.
Argentina
lzabala@na-sa.com.ar

Yoshio Hino
NMIJ/AIST
Japan
y.hino@aist.go.jp

Akira Yunoki
National Metrology Institute of Japan
Japan
a.yunoki@aist.go.jp

Ljudmila Benedik
Jožef Stefan Institute
Slovenia
ljudmila.benedik@ijs.si

Arunas Gudelis
FTMC
Lithuania
arunas.gudelis@ftmc.lt

Haluf Yücel
Ankara University, Institute of Nuclear Sciences
Turkey
haluk.yucel@ankara.edu.tr

Christian Lucks
Federal Institute for Radiation Protection
Germany
clucks@bfs.de

Carlos José Da Silva
Instituto de Radioproteção e Dosimetria
Brazil
carlos@ird.gov.br

Paulo Alberto Lima da Cruz
LNMRI/IRD/CNEN
Brazil
palcruz@ird.gov.br

Alfredo Lopes Ferreira Filho
LNMRI/IRD/CNEN
Brazil
alfredo@ird.gov.br

Simon Jerome
National Physical Laboratory
United Kingdom
simon.jerome@npl.co.uk

Tsukasa Aso
National Institute of Technology
Japan
aso@nc-toyama.ac.jp

Mikael Hult
EC-Joint Research Centre
Belgium
mikael.hult@ec.europa.eu

ICRM2017.indb 27 09/05/2017 12:36:50

Christophe Domergue
CEA CADARACHE
France
christophe.domergue@cea.fr

Jiri Suran
Czech Metrology Institute
Czech Republic
jsuran@cmi.cz

Monika Mazánová
Czech Metrology Institute
Czech Republic
mmazanova@cmi.cz

Denis Glavič-Cindro
Jožef Stefan Institute
Slovenia
denis.cindro@ijs.si

James Zickefoose
Canberra
United States
james.zickefoose@canberra.com

Estela Maria de Oliveira
IDR
Brazil
estela@ird.gov.br

Jonathan Riffaud
LNE-LNHB
France
jonathan.riffaud@cea.fr

Matjaž Aleš Korun
Jožef Stefan Institute
Slovenia
matjaz.korun@ijs.si

Stefaan Pommé
Joint Research Centre
Belgium
stefaan.pomme@ec.europa.eu

Marie-Christine Lépy
LNE-LNHB
France
marie-christine.lepy@cea.fr

Christophe Bobin
LNE-LNHB
France
christophe.bobin@cea.fr

Jana Sochorova
Czech Metrology Institute
Czech Republic
jsochorova2@cmi.cz

Philippe Cassette
LNE-LNHB
France
philippe.cassette@cea.fr

Xavier Mougeot
LNE-LNHB
France
xavier.mougeot@cea.fr

Sylvie Pierre
LNE-LNHB
France
sylvie.pierre@cea.fr

Martin Loidl
LNE-LNHB
France
martin.loidl@cea.fr

Jaroslav Solc
Czech Metrology Institute
Czech Republic
jsolc@cmi.cz

Mark Kellett
LNE-LNHB
France
mark.kellett@cea.fr

Marine Wansek
CEA
France
marine.wansek@cea.fr

Andrei Antohe
IFIN-HH
Romania
antohe@nipne.ro

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Mihail-Razvan Ioan
IFIN-HH
Romania
razvan.ioan@nipne.ro

Doru Stanga
IFIN-HH
Romania
doru@nipne.ro

Daniela Gurau
IFIN-HH
Romania
daniela.gurau@ymail.com

Antonio Eduardo de Oliveira
IRD/CNEN
Brazil
aedu@ird.gov.br

Aurelian Luca
IFIN-HH
Romania
aluca@nipne.ro

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SPONSORS

National Atomic Energy Commision
Buenos Aires
Argentina

National Institute of Industrial Technology
Buenos Aires
Argentina

Aadee SA
Buenos Aires
Argentina

Armotour
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Argentina

Caen SyS Systems and Spectroscopy Solutions
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Eckert + Ziegler Isotopes Group
Braunschweig
Germany

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Buenos Aires
Argentina



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Buenos Aires
Argentina



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Argentina



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Netherlands



Veccsa SA
Buenos Aires
Argentina

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ABSTRACTS

Please be aware that the following abstracts which were submitted for the evaluation process and shall only serve as an information guide for this conference and have no claim for a scientific publication. The presentations of the conference can deviate from these. Furthermore, the abstracts can differ from the submitted forms due to the formatting for this book.

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INTERNATIONAL METROLOGY. THE METER CONVENTION AND THE INTER-AMERICAN METROLOGY SYSTEM

Laiz, H.

INTI (AR)

The acceptance of measurements results at a global level requires the use of a common system of units and the agreement on procedures to establish the equivalence of measurements.
The Meter Convention, signed in 1875, is the framework in which 58 member states and 41 associates agree on a common system of units, the so called International System of Units (the SI). This is not a static system; just now we are facing a significant change of its fundation. After 2018 all the base units will be defined by fixing the value of set of constants of nature.
But international recognition needs more. In 1999 an agreement was signed by the National Metrology Institutes from countries members of the Convention. This agreement stablished that in order to have its Calibration and Measurements Capabilities recognized worldwide it is necessary to participate in interlaboratory comparisons and have in operation a Quality System with and external evaluation.
The Sistema Interamericano de Metrology (SIM) plays a crucial role in the operation of the CIPM-MRA, by organizing regional comparisons and reviewing the QS of the NMIs and Designated Institutes of the Americas.
This talk will provide an overview of the structure of the Meter Convention and the future changes in the SI. It will also describe how SIM support the international recognition of the calibrations and measurements made in the Americas.

Corresponding author's email address: **laiz@inti.gob.ar**

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SIM IONIZING RADIATION LABORATORIES IN SIM: RADIONUCLIDE METROLOGY IN THE AMERICAS

Karam, L. R.

NIST (US)

The Sistema Interamericano de Metrologia (Inter-American Metrology System, or SIM) is the Regional Metrology Organization (RMO) for the Americas (except, currently, for Cuba). As is the case with other RMOs, SIM's goals are to promote an integrated measurement infrastructure in the region to support a variety of activities including trade and economic development. All nations are expected to need ionizing radiation measurement traceability for health-care applications (e.g., mammography, radiation therapies), and many countries also look to the ionizing radiation metrology community to support manufacturing, device sterilization and radiation processing. Although the IAEA supports some of the smaller economies to meet their traceability needs, most require a domestic source (often supported by the IAEA) for their ionizing radiation measurement standards.

Distributed among five sub-regions (NORAMET, CAMET, ANDIMET, SURAMET and CARIMET), National Measurement Institutes (NMIs) have or are planning to designate laboratories (Designated Institutions, or DIs) to develop and provide the measurement standards for ionizing radiation, including radioactivity. DIs are frequently laboratories originally involved in nuclear energy activities. Two NMIs, the NRC in Canada and the NIST in the USA, have not designated laboratories but develop and provide measurement standards in ionizing radiation themselves.

In order to facilitate interactions among the various SIM laboratories in radionuclide metrology, the SIM technical committee for ionizing radiation, Metrology Working Group 6 (MWG 6), provides a forum for technical exchanges and learning opportunities for established laboratories as well as for those designated laboratories which, though technically experienced, may be new to metrology and their role in the international metrology infrastructure. The role of the technical committee and its members in enabling the establishment and awareness of radionuclide metrology throughout SIM will be considered.

This talk will provide an overview of the various laboratories in SIM that currently or anticipate providing radioactivity standards for their relevant economies and stakeholder base, their facilities, and their participation in the activities of the CIPM MRA (including in international measurement comparisons). Particular attention will be given activities supporting customer responsiveness, verifiability of measurements, improvements in technologies/techniques, and improvements in metrological infrastructure (the four strategic goals of MWG 6).

Corresponding author's email address: **lisa.karam@nist.gov**

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008

IS DECAY CONSTANT?

Pommé [1] S. *et al.*

[1] JRC (EU), 2 collaboration of NMIs

The common measurement system for radioactivity is solidly based on the famous exponential-decay law and doubt raised on the invariability of decay constants touches the foundations of nuclear physics, the establishment and international equivalence of the SI unit becquerel, quantitative predictions of activity and radiation dose, and accurate dating through nuclear chronometers. The issue at hand concerns the observation of permille-sized modulations in particular decay rate measurements repeated over several years and the interpretation that they are caused by variations in the decay constants rather than instabilities in the detection system. Claims were made about a new interaction – the fifth force – by which neutrinos could affect decay constants, thus predicting changes in decay rates in correlation with the variations of the solar neutrino flux due to the seasonal changes in Earth–Sun distance. Other hypotheses have been launched finding against constancy of decay rates, e.g. suggesting that periodic changes in beta radioactivity is due to interactions with slow neutrinos as part of dark matter, such as the relic neutrinos produced at the early stages of the existence of the Universe.

In recent work, a unique collection of more than 60 data sets of long-term decay rate measurements was obtained from 14 nuclear laboratories and investigated for the presence of systematic annual modulations. The claim that the seasonal variations in orbital proximity to the Sun cause modulations of decay constants at the permille level has been investigated and disproved. The decay constants of alpha, beta and electron capture decaying sources is invariable to Earth–Sun distance within 0.008% for most nuclides.

In a next step, which is the subject of the current work, the data sets will be checked for oscillations at other frequencies, such as 11-year cycles allegedly associated with internal solar rotation. Conclusions will be drawn whether there are consequences for the establishment of the becquerel and the various applications of the exponential-decay law.

Corresponding author's email address:

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A HISTORY OF THE CIEMAT-NIST METHOD OF STANDARIZATION OF RADIONUCLIDES

030

Coursey, B. M.[1] , Grau Malonda, A.[2]

[1] NIST (US), [2] CIEMAT (ES)

Liquid scintillation detectors have been used in radiation detectors since the 1950's. They were little used in radionuclide metrology laboratories for two chief reasons. LS detectors exhibit a threshold for detection of low energy electrons, resulting in a non-detection probability, and this threshold is unique to each scintillator formulation. Additionally, a large number of radionuclides of interest are not soluble in the organic solvents used in the liquid scintillators. These fundamental problems in physics and physical chemistry were first addressed in a systematic 5-year study by investigators from the CIEMAT and the NIST with support from the U.S.-Spain Joint Committee for Scientific and Technological Cooperation. The ambitious aim of this project was to develop procedures for the standardization of any radionuclide by liquid scintillation counting. The four objectives stated in the proposal were: to obtain new models and computer codes for different decay modes, to stabilize inorganic species in liquid scintillators, to measure selected nuclides to validate the computational methods, and to investigate the basic processes of interaction of charged particles with liquids.

This 5-year collaboration resulted in a radionuclidic metrological technique for standardization of pure-beta particle emitters that was rapidly adopted and expanded upon by other NMIs, including the BIPM, and became known as the CIEMAT-NIST method. The success of the CIEMAT-NIST method has led to increased harmonization within NMIs for a range of beta-emitting radionuclides.

It has also contributed to the availability of effective radiopharmaceuticals for health sciences applications including bone palliation and radioimmunotherapy.

This paper will look at the literature on LS counting that influenced the development of the method and the systematic studies that were carried out during the five years supported by the US-Spain collaboration.

Corresponding author's email address: **bert.coursey@nist.gov**

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METHODOLOGY FOR ACTIVITY COMPARISON OF THORIUM-227 VIA THE INTERNATIONAL REFERENCE SYSTEM (SIR)

094

Keightley, J.D. [1]; Pearce, A.K.[2] ; Pommé, S, [2]; Michotte, C. [3]

[1] NPL(UK), [2] JRC-Geel(EU), [3] BIPM

A current branch of research in radio-immunotherapy is focused on the development a pipeline of α-emitting radiopharmaceuticals, where a payload of ^{227}Th is chelated to monoclonal antibodies (or other proteins) and coupled with conjugation technologies to provide "Targeted Thorium Conjugates", used to target hematological cancer cells and various tumor types. Th-227 (half-life 18.7 d) α-decays to ^{223}Ra (half-life 11.4 d) which subsequently decays via a series of short-lived α-emitters to the β-emitter ^{211}Pb (half-life 36.1 min), followed by a branching of short-lived α- and β-emitters to stable ^{207}Pb. The attractiveness of ^{227}Th for cancer treatment (5 α-emitters in its decay chain) is accompanied by significant complications in measurement of its activity using re-entrant ionization chambers (ICs) as employed in NMIs and at the BIPM in the International Reference System (SIR).

These complications arise since the solution of differential equations describing the activity of ^{227}Th progeny as a function of time reveals that equilibrium is not reached before the ^{227}Th itself has decayed appreciably. Furthermore the activity ratios of the progeny below ^{211}Pb (with respect to the first daughter ^{223}Ra) are such that one cannot simply consider the ^{227}Th as a simple ^{227}Th/^{223}Ra system (i.e.: unsupported ^{223}Ra will equilibrate with its progeny within approximately 10 hours, but ^{223}Ra supported by ^{227}Th takes approximately 10 days to reach a similar level of equilibration with its progeny). Accurate measurement within the SIR requires precise knowledge of the time (and efficacy) of the chemical separation of ^{227}Th from its progeny, and repeated measurements of an ampoule submitted to the SIR over an extended period on the decay/ingrowth curve are required in order to accurately determine the ^{227}Th activity at the "effective" separation time.

This subject has been the focus of discussions at recent CCRI(II) KCWG meetings, where we recognized the need for a suitable robust mechanism for inter-NMI activity comparison for ^{227}Th. This manuscript presents a methodology which considers the concept of a "time dependent equivalent activity" in the SIR in which the total current induced (at a particular measurement time) is partitioned into that induced by each of the progeny. Each contribution is calculated from the predicted relative progeny activity, the associated γ- and X-photon emission intensities, the β- and conversion-electron intensities along with the SIR efficiency curves for photons and electrons determined via the "SIRIC" software package. An iterative procedure is utilized to minimize the residuals between the measured current and the sum of the predicted progeny-specific responses, yielding an estimate of the ^{227}Th activity at the effective separation time.

This methodology requires that when submitting ^{227}Th to the SIR, NMIs must provide additional data to the BIPM including details (and time) of the actual chemical separation procedure employed along with results from of a series of γ-spectrometry measurements required to determine the effective separation time. The precision on the estimate of the separation time propagates directly into the uncertainty of the calculated equivalent activity and ultimately on achievable degrees of equivalence. A recommended procedure for calculation of the effective separation time is presented along with an example uncertainty budget. The accuracy of the equivalent activity of a given SIR submission will depend on the availability of the SIR to make the required repeated measurements, and a discussion on the implications regarding uncertainties on the KCRV and degrees of equivalence is presented. The methodology presented will also provide guidance for NMIs to maintain their own "in-house" ^{227}Th IC calibration factors, for subsequent dissemination of ^{227}Th standards to their respective user communities.

Corresponding author's email address: **john.keightley@npl.co.uk**

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ON THE COMPARISON OF RADON STANDARD USING IONISATION CHAMBERS: AN ATTEMPT TO IMPROVE THE RELIABILITY OF THE SIR RESPONSE FOR RADIOACTIVE GAS

057

Pierre, S. [1]; Sabot, B. [1]; Cassette, P. [1]; Liang, J. [3]; Le Garrérès, I. [1]; Rigoulay, F. [1]; Courte, S. [2]; Ratel, G. [2]; Lourenço, V. [1]; Ferreux, L. [1].

[1] CEA, LIST (FR), [2] BIPM, [3] NIM (CN)

The primary activity measurement system of ^{222}Rn, based on the defined solid angle alpha counting (ASD) of a cryogenic solid source, allows the production of a radon standard in precisely manufactured metal containers or in flame-sealed glass ampoules. The metal containers are generally preferred as they have a well-defined geometry allowing the non-destructive control of the activity of the source by gamma-ray spectrometry. Nevertheless, preparation of radon standards in flame-sealed ampoules is sometimes necessary, especially for comparisons within the Système International de Référence (SIR) of the Bureau International des Poids et Mesures (BIPM). In this system, which is the cornerstone of the international traceability of radon standards, the ampoule is placed inside a well-type re-entrant ionization chamber (IC), and the equivalent activity is measured against very stable radium sources. As this measurement method is based on the detection of photons, its reliability requires that the variability of the ampoules has a negligible influence on the detector response. This is probably true (and admitted) for the measurement of radionuclides with strong gamma emission, but this is more questionable for radionuclides with lowphoton emission intensity, where the response of the IC is mostly due to bremsstrahlung from electrons emitted by decaying nuclides or released by photon interaction. In the case of ^{222}Rn, the problem is even more complicated, as the gamma and electron emission is mostly due to the radon progenies, which are attached on the walls and the neck of the ampoule and thus can be considered as a surface source.
In this paper we studied the influence of the glass thickness and the sealing point position on the IC response to radon. The glass thickness was quantified by a photon absorption method using a ^{241}Am point source, and the global study of the detector response was quantified by measuring various ^{222}Rn standard ampoules with different glass thicknesses, internal volumes and positions of the sealing point. The measurements were made on three different ICs at LNHB, and also on the SIR IC. These experiments showed that the standard deviation between ampoules containing the same reference activity is more than two times higher than the standard uncertainties on the activity of each ampoule, and thus, that this effect could lead to erroneous conclusions about the compatibility of the samples, if this variability is not accounted for. A reduction of this variability by a factor of two was observed when using a coaxial copper filter around the ampoule, at the price of a reduction of the sensitivity of the IC. The paper concludes on the necessity to add an uncertainty term for the measurement of ^{222}Rn ampoules in the SIR system, in order to take into account the variability between the ampoules. Remediation techniques are also proposed, like the use of filters or the development of a well-defined metal container for gas measurement in the SIR chamber, but also on the use of other robust measurement techniques for comparing the activity of this kind of radioactive gas.

Corresponding author's email address: **sylvie.pierre@cea.fr**

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ISOLATION AND PURIFICATION OF PROTACTINIUM-231

037

Jerome, S.M.[1]; Ivanov, P.[1]; Russell, B.C.[1]; Happel, S.[2]

[1]NPL(UK), [2]Triskem (FR)

Protactinium-231 is one of the lesser known actinides, yet the measurement of this radionuclide is central to dating studies in both paleoclimate and nuclear forensics measurements; furthermore, it is important as the immediate parent nuclide of the ^{227}Ac decay chain. In this paper, we present the preparatory work for an upcoming CCRI(II) supplementary comparison of this radionuclide.

Protactinium-231 presents some unique measurement challenges, due to the nature of its chemical behavior for which there is a single systematic study[1], the complexity of its decay scheme and the generally unsatisfactory state of the decay data of this radionuclide[2].

The material used in this work was of poorly known provenance, and it was necessary to carry out a chemical purification of this material, prior to use. It was known that this material had been recovered from a solid source of ^{231}Pa in the early 1990s, and it was likely to contain stable element impurities. In addition, it was important to separate and recover ^{227}Ac for half-life studies of this radionuclide. Protactinium does, however have an affinity for medium size alcohols, that have been used in the isolation of this nuclide in the past[1]. A new extraction chromatography resin, TK 400 has been developed for the separation of ^{231}Pa, which was tested at NPL. The aims of this study were to (i) recover ^{231}Pa in high yield, due to it's scarcity, (ii) remove and recover (in high yield) the ^{227}Ac, ^{227}Th and ^{223}Ra decay products for other projects, and (iii) to remove stable element impurities, suspected to be present.

The distribution coefficients (k_d) for Pa (using ^{233}Pa) and a number of elements suspected or known to be in the original material (Fe, Ni, Cu, Zn, Y (*stable proxy for Ac*), Ba (*stable proxy for Ra*), Ra, Ac, Th, U) and, for completeness, Np were tested by measuring their extraction onto TK400 resin from varying strengths of HCl; the individual elements were measured either by collision cell ICP-MS (Fe, Ni, Cu, Zn, Y, Ba, Th, U) or γ-spectrometry for Ra, Ac, ^{233}Pa and Np). The outcome of this study was to show that these elements had k_d values of 1-3 across the concentration range, with the exception of Pa which showed a maximum k_d of ~100 at 9-10 M HCl. Protactinium was recovered from the resin with 6M HCl and stabilized with traces of fluoride ion and oxalic acid.

The existing ^{231}Pa bearing material was converted into a carrier-free solution in 10M HCl, which was then purified using the TK-400 resin. A recovery of ~75-95% was achieved for the original sources used as the source material for this work. Measured by γ- and α-spectrometry, purity was excellent with <0.2% contamination from Ac, Th and Ra is a single separation of Pa; to increase the purity a second separation was carried out to reduce the levels of daughter nuclides to insignificance. The original aims of the study were achieved; the recovery of ^{231}Pa was good; the decay products were recovered in good yield (>75%) and stable element impurities were removed.

This separation described was used to prepare pure ^{231}Pa with a known separation time for the upcoming CCRI(II) comparison; part of this work will be to achieve a new measurements of the decay data, especially the half-life of ^{231}Pa. It will also be used in future studies (2017-2018) of the ^{234}Th/^{234m}Pa/^{234g}Pa system to determine the branching ratio and half-life of ^{234g}Pa.

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[2]Arinc, A., 2011. http://www.nucleide.org/DDEP_WG/Nuclides/Pa-231_com.pdf

Corresponding author's email address: **simon.jerome@npl.co.uk**

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NEW THORIUM-229 AMOUNT CONTENT STANDARD REFERENCE MATERIAL

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Essex, R. [1]; Mann, J. [1]; Williams, R. [2]; Kinman, W. [3]; Hubert, A. [4]; Bennet, M.[5]

[1] NIST (US), [2] LLNL (US), [3] LANL (US), [4] CEA (FR), [5]ANL (US)

Thorium isotope amount ratios and ^{229}Th amount content were measured for a new Standard Reference Material, SRM 3059. This SRM is primarily intended for use in nuclear forensic analyses as an isotope dilution mass spectrometry (IDMS) spike for measurement of trace-level Th in uranium materials. Thorium mass fraction and U purification age measurement capabilities of particular interest to the nuclear forensic analytical community. IDMS represents a precise and accurate method for determining trace concentrations of Th and one of the most robust and widely used radiochronometric "clocks" is the ^{234}U-^{230}Th parent-daughter system. A significant source of uncertainty for this clock is quantifying the amount of ^{230}Th daughter product produced by radioactive decay of the parent isotope. This isotope is frequently measured by IDMS using an isotopically enriched ^{229}Th material that is calibrated "in-house" or using a certified ^{229}Th radioactivity standard and converting massic activity to amount content using published half-lives.

The stock material for the SRM 3059 was created from a ^{229}Th master solution that was previously characterized as the starting material for NIST radioactivity standard, SRM 4328C. Two ampoules of the concentrated master solution were combined and quantitatively diluted to create the stock solution which was dispensed to specially prepared high-purity quartz glass ampoules (3 mL / unit). The ampoules were flamed sealed immediately after the solution was dispensed and a subset of the ampoules was randomly selected for Th isotopic composition and Th amount content analyses. The Th amount content was measured by reverse IDMS. Thorium reverse-spike solutions were prepared from a well characterized high-purity Th metal (> 99.9% ^{232}Th). A total of 14 units of the ^{229}Th were analyzed by IDMS using 3 separate preparations of the Th reverse spike and 11 un-spiked samples from 8 units were analyzed for Th isotopic composition. Thorium isotope amount ratios were measured at 3 separate laboratories on multi-collector inductively coupled plasma mass spectrometers. These measurements included Th isotope amount ratios of the ^{229}Th spike, the ^{232}Th reverse spike, and the IDMS sample mixes. Mass bias corrections for measured isotope ratios were determined using interspersed measurements of CRM U010 or CRM U500.

Analysis results did not indicate any statistically significant heterogeneity in the SRM units or any bias between results for the 3 IDMS spikes. Accordingly, the data were combined to assign certificate values and to estimate measurement uncertainties that are consistent with international standards for certified reference materials. The weighted mean value and expanded uncertainty (k = 2) for the ^{229}Th amount content of the SRM 3059 spike is (0.11498 ± 0.00015) nmols • g^{-1}. The weighted mean value and expanded uncertainty (k = 2) for the $n(^{230}\text{Th})/n(^{229}\text{Th})$ isotope amount ratio is (0.0000518 ± 0.0000026) and the $n(^{232}\text{Th})/n(^{229}\text{Th})$ amount ratio is (0.0003815 ± 0.0000088). The certified amount content for SRM 3059 is traceable to the mol and represents a 7-fold improvement in uncertainty relative to an amount content value derived from counting standard SRM 4328C.

Corresponding author's email address: **Richard.Essex@NIST.gov**

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INTERLABORATORY COMPARISON OF A NEW NORM REFERENCE MATERIAL OBTAINED FROM TREATMENT OF DRINKING WATER

155

Wiedner H., Maringer F. J.

BEV (AT)

Naturally occurring radionuclides such as ^{40}K and the decay products of the primordial radionuclides ^{232}Th, ^{235}U and ^{238}U are present in many natural resources. Naturally occurring radioactive materials (NORM) containing these radionuclides are exploited by industrial endeavors and often exceed the exemption limits of the activity concentration for radionuclides of the U and Th series, depending on mineral composition and geological origin. Industrial and public activities are generating a significant portion of NORM waste and can enhance the potential of radiation exposure of workers and the public, therefore, necessitating a careful determination of activity content. NORM emits many gamma-rays of different, and sometimes interfering, energies that have to be measured and analyzed by an expert and sample activity often barely exceeds the background, requiring long measuring times. For the traceable determination of activity concentrations in NORM, reference materials with a well-established activity concentration are essential.

This paper outlines the process of the characterization of a new NORM reference material as well as the results of an interlaboratory comparison using this new reference material.

The new reference material is contaminated sand obtained from the backflush of a drinking water treatment facility in Lower Austria with significantly elevated levels of ^{226}Ra. The original material was sifted, dried for a week at 105 °C and carefully homogenized using a three dimensional shaker mixer. 200 g portions of the grain size fraction of 0.8 – 2 mm were filled into polypropylene sample containers and measured on an HPGe detector to verify homogeneity of the sample collective. For the characterization process, methods developed in the European Metrology Research Project MetroNORM have been used. MetroNORM focused on creating traceable, accurate, and standardized measurement methods, reference materials and systems for application in the concerned industries.

Samples of the fully characterized reference material were sent to European laboratories concerned with radioactivity measurements. The paper discusses the results, specific requirements, problems and solutions that were found during the characterization process as well as the subsequent interlaboratory comparison evaluation.

Corresponding author's email address: **Hannah.wiedner@bev.gv.at**

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ANALYSIS OF ACTINIDES IN THE INSOLUBLE RESIDUES AFTER DECOMPOSITION BY VARIOUS TECHNIQUES

Benedik, L. ; Trdin, M.

JSI (SI)

One of the most important requirements in radiochemical analysis is digestion of a sample before radiochemical separation of the radionuclide(s) of interest. It is well known that soil samples are often considered one of the most difficult matrices to digest. Natural radionuclides are incorporated into the structure of difficulty soluble minerals – such as zirconia, apatite, titanite, allanite etc. On the other hand, most authors presume that the artificial radionuclides are attached to the surface of the sample particles. Currently available techniques for the determination of radionuclides in solid samples can take up to several days if acid digestion utilizing mineral acids is used to decompose the samples. Alternatively, thermal fusion and microwave digestion are known as a tool to shorten the digestion process. However, in the vast majority of digestion methods, the solid residue remains and is removed prior to radiochemical separation. A digestion method allowing a sample to be loaded on a separation resin without any residue after its decomposition is therefore of great interest.

The aim of our study was to compare different dissolution techniques for the determination of actinides in environmental samples: conventional wet dissolution with mixtures of acids HNO_3, $HClO_4$ and HF, microwave dissolution using HNO_3 and HF, and thermal fusion utilizing Li borate as a fusion agent. The results obtained showed that dissolution without any residue was achieved only when the third method was used. Thus, no filtration was needed prior to the loading of the sample on the resin. For checking the content of uranium and thorium in the residue after dissolution, radiochemical neutron activation analysis was used, while the content of radium, americium and plutonium radioisotopes in the residue was tested by alpha-particle spectrometry. The results obtained showed that depending on the geological properties, insoluble residues contained up to 30% of natural radionuclides.

Corresponding author's email address:

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HOMOGENEITY OF WHEAT FLOUR IN 5 ML CONTAINERS FOR CERTIFIED REFERENCE MATERIALS

084

Furukawa, R.[1]; Unno, Y.[1]; Miura, T.[1]; Yunoki, A.[1]; Hachinohe, M.[2]; Hamamatsu, S.[2]

[1]NMIJ (JP), [2]NFRI (JP)

Many laboratories measure activity using high-purity germanium (HPGe) detectors. They use some certified reference material (CRM) from a specific environment to check the validity of their measurement devices. We have produced a CRM from the environment using brown rice contained in a 100 ml *polypropylene* container (Unno et al., 2014). Such a samples is used to check the validity of measurement devices by many laboratories that use coaxial HPGe and cylindrical scintillation detectors. We plan to produce a CRM in 5 ml container for well-type detectors.

For this purpose, we characterized the homogeneity of small samples. In this study, we selected wheat and brown rice as the candidate materials for the CRM. The certified value is expected to be below 100 Bq kg^{-1}.

The samples were harvested in Fukushima Prefecture. In the case of rice, we filled several 100 ml containers with 81g and several 5 ml containers with 4 g of rice grains. The wheat grains were milled for homogeneity. For the wheat flour samples, 100 ml containers were filled with 81 g and the 5 ml containers were filled with 3.8 g of flour. We measured the samples using an HPGe detector with the model GEM130108(ORTEC) for the 100 ml containers and a detector with the model GWL150-15-S(ORTEC) for the 5 ml containers. The measurement times were 80,000s for the 100 ml containers, and 200,000s for the 5 ml containers. The brown rice was obtained from the same lot as the samples used in the 2015 proficiency testing (Furukawa et al., 2016).

For the brown rice, the standard deviation of the Cs-137 activity for the 100 ml containers was 1.22% (N = 20), and the standard deviation for the 5 ml containers was 4.35% (N = 20). The statistical uncertainties were 0.9 % (k = 1) for the 100 ml containers and 1.8 % for 5 ml container. In the case of wheat flour, standard deviations of Cs-137 activity of 100 ml container was 1.29% (N = 60), and the standard deviation of the 5 ml containers was 1.69% (N = 15). The statistical uncertainties were 0.7% for the 100 ml containers and 1.3% for 5 ml containers.

In the 5 ml containers, the wheat flour samples were more homogeneous than the brown rice samples. This means that wheat flour is more suitable for the 5 ml CRM than brown rice. Difference of the activities of the brown rice samples in the 5 ml containers was larger. This may be caused by the inhomogeneity of the grains and the deviation in sample height. The uncertainty in sample height was calculated at about 2.7% (Unno, 2016).

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Corresponding author's email address: **rio.furukawa@aist.go.jp**

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PRODUCTION OF SPIKED VEGETATION SAMPLES CONTAINING GAMMA-EMITTING RADIONUCLIDES TO PARTICIPATE IN INTERCOMPARISON TESTS

138

Santos de Souza, P. ; Clain, A.F

IRD/CNEN (BR)

The production of environmental samples such as soil, sediment, water and vegetation with radionuclides for intercomparison tests is a very important contribution to environmental monitoring. Laboratories that carry out such monitoring need to demonstrate that their results are reliable. The National Laboratory for Ionizing Radiation Metrology (LNMRI) by means of National Intercomparison Program (PNI) produces and distributes environmental samples containing radionuclides used to check the environmental measurement laboratory performances. This work demonstrates the feasibility of producing vegetation (grass) samples containing ^{60}Co, ^{65}Zn, ^{134}Cs, and ^{137}Cs by the spiked sample method for the PNI. The preparation and the statistical tests followed 34 and 35 ISO Guide recommendations. The grass samples were dried, ground and passed through a sieve of 250 µm and homogenized. 500 g of vegetation was treated in each procedure. Samples were treated by two different spike sample procedures: homogenizing of the radioactive solution containing vegetation by hand and; and homogenizing of the radioactive solution containing the vegetation in a rotatory evaporator. In both procedures samples were dried in an oven. The reference values for activity concentration of the radionuclides in the grass had a range of 593 Bq/kg to 683 Bq/kg. After gamma spectrometry analysis the results of both procedures were compared as accuracy, uncertainty, homogeneity and stability. The accuracy, uncertainty and short time stability from both methods were similar but the homogeneity test using the rotatory evaporator was not approved for the radionuclides ^{60}Co and^{134}Cs. Both procedures of preparation showed similar results related to reference values. Agitation by hand proved to be more homogeneous than rotatory evaporator procedure. Based on comparisons between both procedures, the manual agitation procedure was chosen for the grass sample production for PNI. The accuracy of the procedure had a range between -1.1 and 5.1% and the uncertainty between 0.6 and 6.2 %.

Corresponding author's email address: **poliana.ssouza@yahoo.com.br**

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PROFICIENCY TEST EXERCISES FOR CTBT PARTICULATE RADIONUCLIDE LABORATORIES

074

Nakashima, N.; Duran, E. B.; Auer M.

CTBTO Preparatory Commission

Since 2000, the Provisional Technical Secretariat (PTS) of the Comprehensive Nuclear Test Ban Treaty (CTBT) Organization has organized an annual Proficiency Test Exercise (PTE) for 16 radionuclide laboratories supporting its network of radionuclide monitoring stations. Sixty-three of the 80 International Monitoring System (IMS) radionuclide stations comprising this network have been certified against stringent technical requirements and are delivering standard data products to national data centers. The IMS laboratories re-analyze samples (using HPGe gamma-ray spectrometry) from these stations on a periodic basis for quality assurance, and provide more in depth measurements to clarify the presence or absence of fission products and/or activation products. The PTEs support the monitoring of the quality of analytical results provided by certified laboratories and assures the data quality from an uncertified laboratory during the certification process.

The PTEs use the same sampling media as used at the IMS stations, but PTEs samples are spiked with CTBT-relevant nuclides with certified activities. Recent PTEs, the spike solution were produced by irradiation of natural uranium. Participating laboratories report the identified nuclides, the activities, the activity concentration at assumed sampling period, and zero time. This requires the laboratory to address complex decay correction.

The paper presents the primary challenges in organizing and conducting the PTEs and how these are being addressed by the PTS. The paper also describes common problems encountered in analysis of these samples and the main challenges with the identification and quantification of nuclides in the PTEs and how the PTE results are used by participating laboratories to improve their analytical techniques and data quality.

Corresponding author's email address: **naoko.nakashima@ctbto.org**

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A REVIEW OF THE TAEK PROFICIENCY TEST ON NATURAL AND ANTHROPOGENIC RADIONUCLIDES ACTIVITIES IN BLACK TEA

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Yeltepe, E. *et al.*

TAEK(TR), JRC-Geel(EC), PTB(DE)

Nuclear weapon tests, nuclear power reactor accidents like Chernobyl and Fukushima and other radiological emergency situations require a sound network of radioactivity measurement laboratories. As contributors of this network the university and institute laboratories in Turkey require proficiency tests (PTs) to demonstrate the reliability of their analyses. Most of the PTs on radioactivity measurements are organized by institutes from outside Turkey with often limitations on the number of participants. It is desirable for Turkish laboratories to participate more often in PTs. TAEK being the EURAMET delegate in Turkey on ionizing radiation metrology, is dedicated to organize PTs on a regular basis to enable the national laboratories to check their analysis methods and help them in their accreditation processes. This paper will describe the organization and establishment of the reference values of the a PT on radioactivity measurement of Turkish black tea organized by Saraykoy Nuclear Research and Training Center (SANAEM) of Turkish Atomic Energy Authority (TAEK) in 2015.

The objective of the PT was to determine the ^{137}Cs, ^{40}K and ^{90}Sr massic activity in black tea powder samples. The bulk material consisting of tea produced in 2014 was mixed with contaminated tea withdrawn from the market after the Chernobyl accident, then milled, remixed thoroughly and sieved down to 250 µm particle size. A total of 60 samples were prepared and 20 of them were distributed to participating laboratories. Two samples were sent to JRC-Geel and PTB-Germany each for the determination of the reference values. The reference values were calculated as the unweighted average of the TAEK, PTB and JRC results. Homogeneity of the final mix was tested with 10 randomly taken samples and outlier, normal distribution, unimodality and ANOVA tests were performed. A minimum of 25 g sample intake is required to achieve homogeneity values of 1.8 %, 2.3 % and 2.5 % for ^{137}Cs, ^{40}K and ^{90}Sr, respectively. The reference massic activities of three radionuclides (1702 ± 30 Bq kg-1, 539 ± 14 Bq kg-1 and 155 ± 16 Bq kg-1 (all dry weight) for 137Cs, 40K, and 90Sr, respectively for confidence level k=2) were measured using gamma-ray spectrometry and liquid scintillation efficiency tracing (CIEMAT/NIST) method. The uncertainty budget includes the gamma spectrometric uncertainties such as those introduced by weighing, efficiency calibration, nuclear data and true coincidence summing correction, as well as uncertainties due to homogeneity. 18 laboratories reported 44 results. The evaluation of the results was done based on the accuracy and precision criteria adopted by IAEA Proficiency Testing Group. The percentage of acceptable scores was 67 %. Only 4 laboratories reported ^{90}Sr results due to the lack of source preparation setups or measurement systems. The performance of the participating laboratories indicates that there was an improvement in the success rate compared to the previous exercise (49 % acceptable results) although the labs need more exercises and proficiency tests to ensure the quality of the analytical data they produce. Recommendations have been provided to some of the labs to improve their measurement setups and techniques. TAEK-SANAEM plans to continue these proficiency tests periodically.

Corresponding author's email address: **emin.yeltepe@taek.gov.tr**

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RESULTS OF A REGIONAL MEASUREMENT COMPARISON EXERCISE FOR I-131 AND Y-90 USING RADIONUCLIDE CALIBRATORS

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Oropesa, P.[1]; Arenillas, P.[2]; De Oliveira, E. A.[3]; Oyarzun, C.H.[4]; García, L.[1]; Guardo, C.[2]; Iwahara, A.[3]; da Cruz, P. A. L.[3]; da Silva, C. J.[3]; Moreno, Y.[1]

[1]CENTIS (CU), [2]CNEA (AR), [3]CNEN-IRD-LNMRI (BR), [4]CCHEN (CH)

A measurement comparison exercise for activity measurement or therapeutic radionuclides in dose calibrators in use during radiopharmaceutical production and applications is organized in the Latin American and Caribbean Region in the frame of the ARCAL RLA 6074 Project and with the support of the International Atomic Energy Agency. The developed protocol takes into account the supporting metrological, radiopharmaceutical manufacture and distributing infrastructures that are available in the participants' countries and the possibility of sharing these infrastructures and measurement capabilities at the regional basis to provide calibration services for radionuclide calibrators in the countries of the Region. Therefore, the regional comparison contemplates fourth sub-regions with fourth metrology institutions acting as pilot institutions: one pilot institution per sub-region. Namely, the Southern Sub-region groups participant institutions from Argentina, Uruguay, Paraguay and Peru being CNEA the pilot institution; the Northern Sub-region is compounded by participants from Colombia, Costa Rica, Cuba, Ecuador, Guatemala, Mexico and Nicaragua and CENTIS acts as pilot. The third and fourth sub-regions are formed for the participants from Brazil and Chile, respectively, being the CNEN-IRD-LNMRI and the CCHEN the correspondingly pilot institutions.

The regional comparison is aimed at the evaluation of the current regional measurement capabilities of I-131 - the most employed therapeutic radionuclide at regional level- and also of the Y-90 - as a perspective therapeutic radionuclide - using radionuclide calibrators. The evaluation is performed on the basis of the statistical sample conformed by the results reported by the participating radiopharmacies and nuclear medicine organizations.

Moreover, with the objective of supporting the comparability of the I-131 and Y-90 activity measurements performed in the four sub-regions a comparison measurement exercise is also organized between the four pilot institutions: the measurement equivalence between the sub-regions is supported by the demonstrated measurement equivalence between the institutions acting as pilot at sub-regional level. The CNEA acts as pilot for the exercise between pilot institutions.

The discussion of the results of the regional comparison is performed with emphasis in common problems and observed differences between sub-regions, taking into account calibrator types or radionuclides between other characteristics as well as in the recommended actions to attain the improvement of the evaluated regional measurement capabilities.

Corresponding author's email address: **poropesa@centis.edu.cu**

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SYSTEMATIC INFLUENCES ON THE AREAS OF PEAKS IN GAMMA-RAY SPECTRA, HAVING A LARGE STATISTICAL UNCERTAINTY

005

Korun, M. *et al.*

SCK-CEN(BE), Tsinghua University(CN), SURO(CZ), UPC(ES), KRISS(KR), FTMC(LT), IFIN-HH(RO), INN(RS), JSI(SI), NPL(UK)

In computerized gamma-ray peak analysis procedures, systematic effects on the peak areas may appear when small peaks are evaluated. They occur when the peaks are located on the basis of the spectral shape and originate in the interference between the peak and the random fluctuations of the continuous background. Their magnitude depends on the software, mostly on the widths of the spectral regions used for determining the height of the continuous background under the peaks.

To investigate the general relevance of the described effect, within the LLRMT Working Group of the ICRM an exercise was performed which comprised the analysis of gamma-ray spectra. The exercise was aimed to analyze peaks with poor counting statistics. A set of 35 spectra measured with a ^{226}Ra source in repeatable conditions was made available to the community engaged in gamma-ray spectrometry for analyses with the peak-analysis software of their choice.

The systematic influence, expressed in the form of a correction factor, was calculated from the ratio between the expected peak area, which is calculated from areas of large peaks having a small relative uncertainty, and the peak areas with large relative uncertainties, taking into account the ratios of the intensities and detection probabilities. For a peak, appearing at a specific energy, the mean correction factor was calculated as an average of the values obtained from the 35 spectra. Analogously, the mean relative uncertainty was obtained as the mean of the relative uncertainties obtained from peaks at that energy. The result of one analysis of the set of spectra is the mean correction factor as a function of the mean relative uncertainty [1].

Fifteen sets of peak-analysis data were reported, comprising peak data and their uncertainties. Ten of them were obtained by the same software, Genie 2000 and four of them by Gamma Vision, AGNES, APTEC and the Gamma-W software. One data set was obtained by manual analysis. Two of the data sets could not be used, because the dispersion of data was too large. The number of peaks reported in the energy range where the correction factor could be determined varied between 47 and 102. The maximum value for the mean relative uncertainty reported (after rejection of outliers) varied between 25 % and 150 %.

It is a common feature of all data sets, that the correction factor assumes the value of unity at the relative uncertainty around 1%. However when the relative uncertainty increases, it may depart from unity. Six data sets did not exhibit any systematic departure from unity within the range of uncertainties reported, however, the maximum relative uncertainty reported in three of these data sets did not exceed 40%, therefore here the systematic influence is not expected to emerge. Seven data sets revealed a systematic decrease of the correction factors at large relative uncertainties. In all these data sets the maximum reported uncertainty exceeded 40%. The relative uncertainty, where the systematic departure starts, varies between 10 % and 30 %. The scattering of data in the energy range, where the systematic influence appears, does not allow extracting any dependence of the correction factor on the relative uncertainty except a linear one.

In the contribution the results will be presented in more detail regarding the software used for spectra evaluation, dispersion of correction factors and systematic departures from unity.

The 35 gamma-ray spectra, together with the Excel file for calculating the mean values and for presenting the results, remain in the link:
https://www.dropbox.com/sh/f9ws2t6g5mn0nqd/AABiO5-yq1ICf76EYdvMCZq6a?dl=0

1. Korun, M., Vodenik, B., Zorko, B., *Appl. Radiat. Isot.*, 105 (2015) 60.

Corresponding author's email address: **matjaz.korun@ijs.si**

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AN ALTERNATIVE APPROACH TO THE DECISION THRESHOLD

024

Korun, M.; Vodenik, B.; Zorko, B.

JSI (SI)

The standard ISO 11929 defines the decision threshold as *a value of the estimator of the measurand, which when exceeded by a result of an actual measurement using a given measurement procedure of a measurand quantifying a physical effect, one decides that the effect is present* [1]. Further, the standard prescribes to calculate it as the null measurement uncertainty, times a factor, given by the coverage probability.

According to the method of calculation, the decision threshold is an assessed value of a quantity, which is calculated from an uncertainty. Therefore, in general, it depends on both, the conversion factor and the indication and their uncertainties as well. However, since the decision threshold is calculated at the value of the indication zero, it depends only on the uncertainty of the indication and not on the uncertainty of the conversion factor. The evaluation at the quantity valued zero is not a part of the definition, therefore it would be reasonable to widen the definition with the specification, that the estimator of the measurand is calculated at the conventional quantity of zero. Namely, according to the definition only, already the uncertainty, associated with any measurement result can serve as a decision threshold.

Alternatively, given the definition in the standard, the method of calculation could be modified. Namely, in the existing method of calculation, the expected value of the conversion factor is used for calculating the decision threshold. Since the true value of the conversion factor may be larger from the expected value with the probability of about 50%, also the decision threshold is underestimated with this probability. It follows then, that although the decision thresholds are calculated for a large probability for not making errors of type I, they may be still underestimated with the probability of 50%. A change of the conversion factor for 5% changes the probability for type-I errors for 0.8%, if the decision threshold is calculated for the probability $\alpha = 5\%$.

It follows then, that the simplicity of calculation of the decision threshold comes at a price. The described deficiency could be improved by changing the method of calculation in such a way, that the decision threshold would depend on the uncertainty of the conversion factor. Following the method of calculation of the decision threshold, it should be proportional to the uncertainty at some value of the measurand y, being different from zero:

$y^* = k1_{-\alpha}\, u(y)$.

The value y should be determined in the definition of the decision threshold, in terms of e.g. the decision threshold itself (what leads to a calculation similar to that of the detection limit) and/or the null measurement uncertainty. In the presentation the decision threshold calculated as

$y^* = k1_{-\alpha}\, u[y{=}u(0)]$,

will be described. Here the uncertainty of the measurand is evaluated at its value being equal to the null measurement uncertainty and not at zero. The difference with respect to the decision threshold, calculated according to the standard, will be presented as a function of the uncertainty of the conversion factor.

[1] ISO, 2010. ISO 11919:2010(E) Determination of the characteristic limits (decision threshold, detection limit and limits of the confidence interval) for measurements of ionizing radiation – Fundamentals and application, ISO Geneva.

Corresponding author's email address: **matjaz.korun@ijs.si**

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DETERMINING ACCEPTABLE LIMITS OF UNCERTAINTIES IN URINARY ^{238}U MEASUREMENTS BY USING THE NEW LTUM METHOD.

039

Guichet, C. [1-2]; Lecoix, G. [1-2]; Hurtgen, C. [1]

[1]PROCORAD (FR), [2]CEA(FR)

PROCORAD organizes for more than 20 years proficiency tests in radiotoxicology to estimate the performance of the laboratories in term of accuracy. The results of these intercomparisons also show a wide range of uncertainties values provided by these laboratories. The regulatory obligations related to accreditation, which require that the laboratory shall define the performance requirements for measurement uncertainty and regularly review estimates of measurement uncertainty, and the lack of published or required acceptable limits in radiotoxicology led PROCORAD to propose uncertainties acceptable limits from the "state of art". For this purpose, PROCORAD used a new method to estimate the uncertainties based only on external quality assessment results: the Long-Term Uncertainties Method (LTUM). This method is based on a linear regression model to express the laboratories values as a function of the target value during a long period. The uncertainty LTUM components are the long-term analytical coefficient of variation and the long-term bias. The LTUM is used by several proficiency testing suppliers in bioassay. This method was applied for ^{238}U in urines using alpha-spectrometry methods and mass measurement by different methods (ICP-MS, KPA and fluorimetry) with the results of 5 years of intercomparisons results provided by Procorad. The LTUM was previously validated for actinides in faecal ashes, ^{3}H and γ/X emitters in urines. The acceptable limits were defined by using the medians calculation of uncertainties provided by LTUM: 19% for the activity measurement by alpha spectrometry and 22% for the mass measurement. The LTUM results were compared to the uncertainties provided by the laboratories. The results provided by laboratories shows an underestimation of the uncertainties calculated by laboratories, independently of the principle of measurement, indicating that all influencing factors are not always taken into account in the uncertainties measurements. These results are in accordance with the results obtained with actinides in faecal ashes, ^{3}H and γ/X emitters in urines. The most important component in the uncertainties calculation is the long term reproductibility for 35% of laboratories (^{238}U activity measurement) and 37% of laboratories (^{238}U measurement by mass). This result is similar to those obtained with ^{239}Pu in faecal ashes and ^{3}H in urines. We compared the results of uncertainties for each laboratory who was giving results in the two principles of measurement. The uncertainties are very similar for the ICP MS users (ratio of uncertainties between 0.8 and 1.1 for 83% of laboratories), indicating that the uncertainties calculated with the same method are not dependent of the technic used but more dependent of the laboratory. But there is a big difference between uncertainties for KPA and fluorimetry users (ratio between 4.1 for 50 % of KPA users and 6.3 for 100% of fluorimetry users). The LTUM is a convenient and simple method to use to evaluate the uncertainties with only the external quality assessment schemes in radiotoxicology.

Corresponding author's email address: **claude.guichet@cea.fr**

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COMPARISON OF INTERCOMPARISON RESULTS OF GAMMA RAY SPECTROMETRY OF SPIKED AND REAL SAMPLES

041

Glavič-Cindro, D.

IJS(SI)

Laboratories engaged in measurements of activities of gamma-ray emitters with high resolution gamma-ray spectrometry in samples from the living and working environment, food and feeding stuff, chemicals, building and raw materials in frame of different monitoring programmes or for the needs of regulatory bodies, have to deliver reliable, traceable and comparable results.
The common accepted way to proof and manifest the quality of the measurement results to different regulatory bodies or customers is be accredited according to the ISO/IEC 17025 standard for the measurements performed. For laboratories and for accreditation bodies the most important proof of the quality of work performed in laboratories is participation with comparable and accepted results in different international intercomparison schemes or proficiency tests.
In the field of gamma-ray spectrometry this tool for checking and confirming the quality of measurement results has already been recognized more than 30 years ago. For example the IAEA's Marine Environment Laboratory has been assisting laboratories in Analytical Quality Control Services (AQCS) for the analysis of radionuclides in the marine environment since the early seventies. In Italy a series of laboratory proficiency tests have been held from 1980 in the framework of the Italian National Environmental Radioactivity Surveillance Network. Since 1989, the National Physical Laboratory (NPL), UK, conducts intercomparison exercises on environmental radioactivity measurements aiming at assessing the measurement capabilities of the laboratories involved in alpha, beta and gamma environmental radioactivity. Similarly in France the first intercomparisons took place in 1978.
The quality and comparability of results of different intercomparison schemes depend strongly on way of the sample preparation. There are two main different approaches of preparing the comparison samples. One approach is that sample matrix, usually aqueous solution, is spiked with certified reference material containing radionuclides of known and traceable activities. In this case the reference values are mostly determined gravimetrically. The other possibility is that the reference material is prepared from a bulk material taken in the contaminated environment. In this case the reference value can be determined statistically on the basis of the measurement results of the participating laboratories or by the measurements of selected reference laboratories.
In this paper different intercomparison schemes will be presented and compared. The attention will be focused on the way how the reference values were determined and its influence on the spread of the values, reported by participating laboratories. One of the main conclusions is that the preparation of real environmental comparison samples and evaluation of reference values and results from participating laboratories is complex process and usually lasts long time. Due to this reason the final reports are often issued with delay, therefore the lessons learned by participation laboratories could not be promptly implemented in their regular work.

Corresponding author's email address: **denis.cindro@ijs.si**

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RAPID AND ACCURATE ASSESSMENT OF THE ACTIVITY MEASUREMENTS IN BRAZILIAN HOSPITALS AND CLINICS

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de Oliveira A. E. ***et al.***

CNEN (BR), CENTIS (CU), CNEA (AR), CCHEN (CH)

The nuclear medicine activities in Brazil are decisively influenced by the ability and mobility of the country to provide radiopharmaceuticals to Nuclear Medicine Services-NMS, taking into account the land area of about 8.5 million square kilometers, and with some cities more distant that 4000 km of others.
In this work, we intend to confirm that having traceability in measurements consistent with the National Metrology of Radiation Ionizing Laboratory - LNMRI, which is the National Metrology Institute of Brazil, the Radiopharmaceuticals Producers Centers are able to serve as a link in the evaluation of measurements of national NMS.
As a strategy, the Institute of Energy and Nuclear Research, IPEN distributed sources of I-131 (half-life: 8.0233 d) to 33 distant SMN over 3,300 km from North to South.
All sources were previously measured at IPEN's radionuclide calibrator on the same day between 14:49 and 15:59 h and were received by NMS across the country until late afternoon the next day.
Additionally IPEN sent to LNMRI three radioactive sources of I-131 from the same solution, after measurement. These sources were calibrated in LNMRI reference system (Ionization Chamber well type) and had their activity confirmed by measuring at CIEMAT / NIST system.
Correction factor related to LNMRI was obtained. This factor, were applied to measurements performed by IPEN to evaluate the performance of measurements of SMN.
The difference between the reference values from sources sent to the IRD and IPEN measurements were less than 1.5%.
The results of comparisons of IPEN and SMN measurements are in accordance with the Brazilian Standard that requires differences smaller than 10%. However, most of the results have difference lower than 2%, showing a measurement quality to I-131 consistent with IAEA-TRS454 - Radioactivity Measurement for Quality Assurance in Nuclear Medicine for NMS.
With over 420 MSN around the country, this methodology allows evaluating the measurement conditions for radioactive sources with half-life equal or bigger then F-18 (This is provided at the same day). IPEN difference of measurement related to LNMRI for most radiopharmaceuticals distributed is less than 2%.

Corresponding author's email address: **aedu@ird.gov.br**

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DEVELOPING RAPID RESPONSE CAPACITY IN THE ALMERA NETWORK OF ANALYTICAL LABORATORIES

152

Osvath, I.; Tarjan, S.; Pitois, A.

IAEA Environment Laboratories

The International Atomic Energy Agency has established in 1995 the Network of Analytical Laboratories for the Measurement of Environmental Radioactivity (ALMERA) with the objective to ensure that reliable data can be reported by the member laboratories in a timely manner in case of accidental or intentional radioactive contamination of the environment. In 20 years the network developed from 24 laboratories in 15 countries to 158 laboratories in 85 countries and continues to grow at a rate of up to eight laboratories every year. The IAEA supports member laboratories to improve, maintain and document the quality of the analytical data they produce in both routine and emergency situations. Professional networking, communication and capacity building through exchange of expertise are key to the development of the network and its participation in methodological development and IAEA reference material certification. Given the broad interests of member laboratories, the technical and strategic challenges are to support through network activities the development and use of methods improving precision, accuracy, sensitivity and response time. A group of ALMERA laboratories has also registered into the IAEA's Response and Assistance Network (RANET), which provides international assistance, upon request from a State, following a nuclear or radiological incident or emergency.

In the wake of the Fukushima Daiichi Nuclear Power Plant accident there was increased interest in rapid assessment methods. This interest was addressed by the IAEA through specific training courses, coordination of joint development and validation of rapid radioanalytical methods and organisation of customized proficiency tests (PTs). The yearly PTs involve six environmental and food samples, typically prepared by spiking natural matrix materials with anthropogenic and natural radionuclides. The radionuclides and levels are chosen so as to be relevant to the measurement of environmental radioactivity of different origins and to continuously challenge the participants. Radiochemical and counting techniques are comprehensively tested, with a reporting time of the order of several months. In addition, rapid reporting is requested for selected radionuclides and samples three days after the registered reception of the set of PT samples. This paper analyses the performance of the network laboratories in these rapid measurements over a period of three years, points out the strengths and areas needing improvement and describes the way the latter are addressed through methodological guidance and training.

Corresponding author's email address:

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ASSESSING THE ABSOLUTE QUANTITATIVE ACCURACY OF POSITRON EMISSION TOMOGRAPHY FOR CU-64 USING TRACEABLE CALIBRATED PHANTOMS

002

Zimmerman, B. E.; Bergeron, D. E.; Edgerton, J. P.

NIST (USA), Appalachian State Univ. (USA)

The use of Position Emission Tomography (PET) as a quantitative tool in the diagnosis and treatment of disease is due in part to the rapid advances in instrumentation and software. However, the true quantitative utility of this modality can only be realized when the entire system, which includes both the scanner and activity calibrator, are calibrated on an absolute basis against national standards. The ability to calibrate the activity calibrator in a traceable way is well-established, but is rarely implemented clinically. Routine calibration of the PET scanner is generally carried out clinically using a phantom containing an aliquot of a F-18 solution whose activity was determined with an activity calibrator. For clinical applications that use only relative measurements (i.e., measurement of ratios in different regions at a single time point), this lack of absolute calibration has a minor influence on the clinical decisions being made. However, for applications in which the activity value is important, such as in internal dosimetry for treatment planning, or when the comparability of data across multiple clinical sites at multiple time points is critical, as in a clinical trial for new drugs, absolute calibration is required.

Copper-64 is an example of a radionuclide for which such an absolute calibration is needed. Its decay properties make it ideal for use in "theranostic" radiopharmaceuticals, which can act as both diagnostic and therapeutic agents. Image-based dosimetry is needed for optimized treatment planning with these types of drugs. The effect that the decay properties of a particular radionuclide has on the quantitative accuracy of PET, particularly for nuclides with complex decay schemes such as Cu-64, has not been systematically studied. Moreover, the uncertainties associated with extracting activity values from PET imaging data for one radionuclide when the system is calibrated for another have not been previously considered in a way consistent with the GUM.

In the first study of its kind, we have used liquid phantoms with activity concentrations traceable to our primary standards to calibrate the NIST PET-CT scanner for F-18. The calibration was monitored over the course of about 8 months using calibrated Ge-68 solid phantoms. Calibrated Cu-64 phantoms of different configurations and having combined standard uncertainties on the contained activity of less than 1.5 % were prepared and scanned at different time points to assess how accurately the Cu-64 activity values could be extracted from the images. The analysis revealed biases in the recovery of the activity concentration of about -9 %. Data from the calibrated F-18 and Ge-68 phantom measurements could be used to renormalize the Cu-64 data, which greatly reduced the bias, albeit with a larger uncertainty. With this method, the extracted activity values were consistent with 100 % recovery for uniform phantoms.

This talk will discuss the methods used for absolute calibration and monitoring of the scanner, as well as for determining the recovery of the Cu-64 activity, focusing on the assessment of uncertainties in the entire imaging process.

Corresponding author's email address: **bez@nist.gov**

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QUANTITATIVE IMAGING, DOSIMETRY AND METROLOGY; WHERE DO NATIONAL METROLOGY INSTITUTES FIT IN?

146

Fenwick, A. J.[1-2]; Merrett, J. L.[1-3]; Ferreira, K. M.[1]; Robinson, A. P.[1]

[1]NPL (UK), [2]Cardiff University (UK), [3]University of Surrey (UK)

Quantitative Imaging (QI) in nuclear medicine is far from a novel concept. Since the inception of Single Photon Emission Computed Tomography (SPECT) there has been a desire to obtain more than just qualitative information from medical images. From the outset in Positron Emission computed Tomography (PET) QI has been used to great effect in diagnostic studies. Given the suggested importance of these imaging techniques in the diagnosis, staging and treatment of cancers it seems somewhat amiss that National Metrology Institutes (NMIs) have had very little input into this area to date, and this has led to other organisations taking on the traditional role of a metrology institute in providing standards, traceability and comparability in this field. So where does the NMI fit in?

There is a demonstrable need for standardisation in QI that is currently not being addressed adequately. QI is essential for the correct determination of dose to patients undergoing Nuclear Medicine administrations be it for diagnostic, Therapeutic or Theranostic applications. Organisations such as the American Society of Nuclear Medicine (SNM) and the European Association of Nuclear Medicine (EANM) have undertaken exercises to establish comparability of measurements, especially in PET. These exercises often lack the metrological basis required to demonstrate traceability to national standards and although developed with the intention of providing comparability, particularly in clinical trials, it is difficult to determine if datasets are truly comparable, and if so to what extent.

NMIs sit in a unique position in that they have the means and expertise to offer truly traceable measurements alongside world-class dataset analysis techniques. This paper discusses progress and developments in the field of QI within the NMI community in order to address the traceability aspect of such measurements along with a summary of work still to be done.

Corresponding author's email address: **andrew.fenwick@npl.co.uk**

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UTILISING NORMALIZED MANUFACTURER'S CALIBRATION FACTORS FOR AN IONIZATION CHAMBER WITH DEPLETED GAS

016

Lubbe, J.; Simpson, B.R.S.; van Staden, M.J.; van Rooy, M.W.

NMISA (ZA)

The NMISA Radioactivity Standards Laboratory maintains South Africa's national measurement standard for radioactivity. As for other national metrology laboratories, primary measurement standards obtained from absolute measurement methods are checked for uniformity and equivalence by regular participation in international and regional key comparisons. Due to the fleeting nature of radioactivity, the NMISA absolute standardizations of gamma-emitting radionuclides are maintained through the calibration of secondary standard instrumentation such as high purity germanium detectors and an ionization chamber. This traceability chain of activity measurements is disseminated to the user community via these instruments. The work presented here focuses on NMISA's Isocal IV well-type ionization chamber (IC) that is connected to the manufacturer's supplied electrometer.

A summary is given of the ionization chamber operation and calibration process. A calibration normalization scheme, which was introduced after repairs to the chamber were undertaken a few years after purchase, is then described. The scheme was devised after it was found that less current was produced from a ^{60}Co standard (after decay correction) than before, by assuming that this was due to lower N_2 gas pressure than that normally produced by the manufacturer. A value for the normalization factor of 0.9157 was determined from an accurately standardized solution of ^{60}Co. This implies that the chamber gas pressure was depleted by about 9 %. The applicability of the procedure is justified through analysis of some results from a comprehensive simulation of the chamber undertaken in an independent study [1]. For verification purposes, normalized calibration factors derived from factors supplied by the manufacturer have been compared against those obtained through absolute standardization made at NMISA for a variety of radionuclides. The results are presented and show good agreement within the specified uncertainties in all cases.

Most measurements made for the nuclear medicine community are secondary, made with the IC using the radionuclide specific calibration factors. Where absolute standardizations on certain radionuclides have not yet been undertaken to allow for the NMISA to derive a factor, the manufacturer's normalized factor can be utilised with confidence. Radionuclides which have been measured successfully for nuclear medicine clients are ^{18}F, ^{67}Ga, ^{68}Ga/^{68}Ge, ^{90}Y, ^{99}Mo, ^{99m}Tc, ^{123}I, ^{131}I and ^{137}Cs. Other medical radionuclides that can be measured with the IC are ^{57}Co, ^{59}Fe and ^{177}Lu, which have been verified though international comparisons. To summarise, the derived normalization factors allow NMISA to indirectly make use of the manufacturer's supplied calibration factors when necessary. It has been demonstrated that the procedure extracts activity values to an accuracy of between 0.5 % and 3 %, depending on the radionuclide.

[1] D.I. Kryeziu, PhD dissertation submitted to Atomic Institute of the Austrian Universities, Vienna, Austria (2006)

Corresponding author's email address: **jlubbe@nmisa.org**

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DEVELOPMENT OF THE AUSTRALIAN STANDARD FOR GE-68 BY TWO LIQUID SCINTILLATION COUNTING METHODS

022

Van Wyngaardt, W. M.; Smith, M.L.; Jackson, T.W.; Howe, B.; Reinhard, M.I.

ANSTO (AU)

Ge-68 is a pure electron capture radionuclide that decays to the ground state of Ga-68 with a half-life of 271 days. Ga-68 decays by a combination of positron emission (88.9%) and electron capture (11.1%), predominantly to the ground state of Zn-68 (96.6%). Five excited levels of Zn-68 are also populated, which de-excite primarily by the 1077 keV γ(1,0) transition (3.2%).
Ge-68 provides a convenient long-lived surrogate for calibration in quantitative PET imaging. In addition, Ge-68 is used to manufacture generators that can provide a steady supply of Ga-68 for nuclear medicine without the need for an on-site cyclotron. Significant advances have been made in the application of Ga-68 for both therapy and diagnostics in recent years. Accurate measurement standards of Ga-68 and Ge-68 are therefore essential for instrument calibration to aid with consistency in clinical trials and patient treatment. In response to these needs, the CCRI(II) organized an international key comparison, CCRI(II)-K2.Ge-68, of activity measurement of a Ge-68 solution. ANSTO standardized the comparison solution by two liquid scintillation counting (LSC) methods to prevent complications that arise due to volatilization of Germanium when dry sources are prepared. Measurements for the 4π(LS)β-γ coincidence method were made using the ANSTO 3-phototube LSC system with a NaI(Tl) detector comprising the gamma channel. Data acquisition was performed using the FPGA Acquisition and Software Event Analysis (FASEA) System. Efficiency variation in the beta channel was by threshold discrimination, with the lowest beta threshold set to about 20 keV to exclude all electron capture emissions. The gamma window was set over the 511 keV annihilation peak. Further details on the measurements and corrections applied will be provided.
The solution was also standardized using the Triple-to-Double Coincidence Ratio (TDCR) efficiency calculation method using the same LSC detection and FASEA data acquisition systems. The detection threshold was set just below the single-electron peak to ensure the detection of emissions related to both electron capture and positron emission decays. The GeGa68 TDCR program developed by Philippe Cassette of LNHB was applied for data analysis. The uncertainties obtained with the 4π(LS) β-γ and TDCR methods were 0.54% and 0.61% respectively, with the results agreeing to within 0.2%. The standard was transferred to the ANSTO TPA Secondary Standard Ionization Chamber. Internationally traceable Australian Certified Reference Materials (ACRMs) for Ge-68 can now be provided to local users. The contribution to ionization current by low energy Ge-68 emissions is expected to be very small, so that standards of Ge-68 can be used for the calibration of dose calibrators for Ga-68. A project is being planned to experimentally determine the magnitude of this correction factor.

Corresponding author's email address: **freda.vanwyngaardt@ansto.gov.au**

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IMPURITIES IN Tc-99m RADIOPHARMACEUTICAL SOLUTION OBTAINED FROM Mo-100 IN CYCLOTRON

042

Tymiński, Z.; Saganowski, P.; Pawlak, D.; Wojdowska, W.; Dziel, T.; Kołakowska, E.; Listkowska, A.

NCBJ RC POLATOM (PL)

This work aim was to investigate the concentration of gamma emitting impurities in cyclotron produced ^{99m}Tc for radiopharmaceutical purposes.
^{99m}Tc radioisotope produced from irradiation of ^{99}Mo in nuclear reactor is widely used in medicine either in diagnosis or therapy treatment. The crisis of ^{99}Mo supply arouse the interest in alternative methods of ^{99m}Tc obtaining. This situation, besides the tendency to reduce production based on high enriched uranium, impel the research program of IAEA No 17419 "Accelerator-based alternatives to non-HEU production of Mo-99/Tc-99m" in which the NCBJ RC POLATOM also participated. As results the ^{99m}Tc solution derived from high enriched molybdenum carries pellets irradiated in GE PET-trace 840 cyclotron (at Heavy Ion Laboratory of University of Warsaw) was investigated.
According to the quality control regulations the radiopharmaceuticals must be evaluated for all possible radionuclidic and radiochemical impurities. Accelerated 16 MeV protons interacting with pellets in ^{100}Mo(p,2n) reaction with moderate cross section can produce impurities such as ^{93}Tc, ^{93m}Tc, ^{94}Tc, ^{94m}Tc, ^{95}Tc, ^{95m}Tc, ^{96}Tc, ^{96}Nb, ^{97}Nb, ^{97m}Tc and ^{99}Mo. For Tc-99m separation from Mo-99 and additional purification the method with the highest Tc recovery in the shortest time was selected. Among 3 different methods tested in RC POLATOM the one based on AnaLig Tc-02 resin was chosen (recovery >76%). The measurements of Tc solutions from different stages of purification process were performed. For the quality assessment of gamma emitters an HPGe high resolution gamma spectrometer was used. The measuring system have been previously calibrated in relation to the National Standard of Radionuclide Activity in Poland held in Radioisotope Centre POLATOM. The gamma spectra were analyzed by GENIE2000 software based on the photopeak area integration method. For not detected gamma emitting impurities the detection limits and decision threshold were calculated.
The total activity of gamma emitters in final radiopharmaceutical product did not exceed 1.6 x 10^{-2} % of ^{99m}Tc activity. The most critical impurities such as ^{99}Mo, ^{96}Nb, ^{97}Nb and ^{95}Tc reached the concentration at the same order of magnitude as specification limits which were proposed as 5.0 x 10^{-3} % for those isotopes. The rest of impurities had the concentration at least one order of magnitude lower than the assumed limits. The studies have confirmed that purity of the final products in terms of gamma emitters with emission lines in range of 50 - 2000 keV met the proposed requirements.

Corresponding author's email address: **zbigniew.tyminski@polatom.pl**

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EXTENDED UTILIZATION OF A ^{68}GE/^{68}GA REFERENCE SOURCE AS A MOCK ^{18}F SOURCE IN THE IONIZATION CHAMBER CALIBRTATION

073

Yamada, T.; Ishizu, H.

JRIA(JP)

Positron emitter of ^{18}F is widely used for medical imaging by the positron emission tomography (PET). Activity of this nuclide is generally measured by use of several types of well-type ionization chambers. Though calibration factors of these chambers shall cover a range of individual radionuclides, it would be not easy to calibrate using short-lived nuclide. Consequently type testing calibration for a master chamber using a ^{18}F reference source or mathematical calculation approach using a response function is adapted. In this case differences of responses among chambers are likely to be checked by ^{137}Cs source for each chamber before shipment. However difference of response for ^{137}Cs might not reflect that for ^{18}F. As another approach Zimmerman and Cessna developed a traceable calibration methodology for solid ^{68}Ge/^{68}Ga sources used as a calibration surrogate for ^{18}F in radionuclide activity calibrators [1][2]. In their method the source was cross-calibrated for equivalent ^{18}F content activity by direct comparison with the national standard for ^{18}F in the same geometry. Their method is practically useful for users who use the same model of chamber that was used in calibration of a reference source. On the other hand since such an equivalent ^{18}F activity would be depend on response function of the chamber, miss calibration with unexpected uncertainty might occur in use of other models of chamber that have different constructions (e.g. wall thickness, type of gas and its pressure). In addition it would mean that unexpected error arise even in use of the same model of chamber. In the present study some effects and uncertainties arise from the difference of constructions between the chambers that were used in the source calibration and practically in use were evaluated to extend utilization of the mock source.

In order to evaluate effects arise from the difference of constrictions among chambers, Monte-Carlo simulation using EGS5 was carried out. In the present study isotope calibrator model IGC7 (Hitachi, Ltd) was modeled by EGS5. Response ratios ^{18}F to ^{68}Ge/^{68}Ga were calculated for various chamber wall thicknesses and pressures of gas conditions. Consequently variations in response ratio for these conditions were less than 0.3% and 0.5% for gas pressure ranging from 0.6MPa to 1.0MPa and aluminum wall thickness from 1.6mm to 2.4mm, respectively. As shown in these results it should be noted that equivalent ^{18}F content activity that is determined by use of the same model chamber equates to real reference value of 18F with taking into account of tolerances of these specification of the chamber. In addition response ratios were also calculated for three different models of chambers (IGC, CRC15R(Capintec), NAK1(Fuji Electric) in the same manner. As a result 1% difference was found between NAk1 and other two models of chamber. Since NAK1 has thin SUS wall of 0.1mm, even difference of β-particle energy between ^{18}F ($E_{\beta max}$=0.634) and ^{68}Ga($E_{\beta max}$=1.899) might cause this significant difference. In the present study measurement using a Mock ^{18}F consists of ^{68}Ge/^{68}Ga solution was also carried out. Activity of ^{68}Ge was absolutely measured by the 4βπ++4πγ sum counting technique [3]. Equivalent ^{18}F activity was determined in the similar manner shown in a previous paper by Zimmerman BE and Cessna JT [1]. In order to evaluate extendibility of the use of mock source, measurements were carried out in the several different conditions. The measured results including uncertainty evaluation will be shown.

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[2] Zimmerman BE, et.al.(2014) *Appl. Radiat. Isot.*87:5-9
[3] Yamada T, et.al.(2016) *Appl. Radiat. Isot.* 109: 325-329

Corresponding author's email address:

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A MOVABLE PRECISION IONIZATION CHAMBER: THE TRANSFER IONIZATION REFERENCE CHAMBER

088

Juget, F.; Nedjadi, Y.; Buchillier, T.; Duran, T.; Bochud, F.; Bailat, C.

IRP (CH)

In Switzerland, the activity unit is maintained by the "chambre d'ionisation de reference" (CIR) at the Institute of Radiation Physics (IRA) which is the designated institute by the Swiss Institute of Metrology for the unit of activity (Bq). The CIR activity measurements are traceable to the international reference system at BIPM for 21 Calibration and Measurement Capabilities (CMC). The relative standard uncertainties of the CIR activities are under 0.5 %.

As part of a wider project to modernise our reference ionization chamber and to broaden our secondary measurement capabilities, a project to develop a portable precision ionization chamber was started in 2012. The goal of the project is to have an instrument in order to measure short-lived radionuclides in hospitals or radioisotope production centres. This mobile instrument, completed in 2016, consists of a Centronics IG12 U-shaped cylindrical ionization chamber, filled with Argon at 2 MPa pressure. The chamber is not shielded to facilitate transport but a 2-inch NaI(Tl) guard detector, positioned above it, monitors continuously the surrounding background. The ionization current is measured directly with a Keithley 6517B electrometer in charge mode via the loading of its internal capacitor for specific time slots. The instrument is controlled with a Labview code which manages the data acquisition, corrects for background, decays during measurements and current saturation effects, and finally outputs the estimated current and its associated statistics. The saturation current correction was determined using very high activity Tc-99m sources and cross-checked with Co-57, Co-60 and Cs-137 sources to ensure a consistent behaviour for different gamma energy ranges.

Optimisation of the TCIR to ensure a good reproducibility has been achieved through defining an appropriate vial, filling volume and position in the well chamber, as well as monitoring the external background with an NaI probe which enables the rejection of measurements in case of too high surrounding background. The calibration factors have been determined using solutions with activities measured with the CIR. The TCIR is currently calibrated for F-18, C-11, O-15, N-13, I-131, Tc-99m and is able to measure isotope activities within 1% standard uncertainty. Measurements on site at the Lausanne University Hospital (CHUV) in the nuclear medicine department have been performed for Tc-99m and F-18 confirming the good agreement with the activity measured using the onsite dose calibrators of CHUV.

The TCIR will typically offer new services to nuclear medicine departments and cyclotron facilities. Short-lived radionuclide calibration traceability will be possible. The TCIR will insure the metrology of short-lived radionuclides and improve their traceability in Switzerland.

Corresponding author's email address: **frederic.juget@chuv.ch**

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AN EGSNRC MODEL FOR THE VINTEN IONIZATION CHAMBER

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Galea, R.; Townson, R.; Tessier, F.; El Gamal, I.

NRC (CA)

Radionuclide calibrators are used extensively in radiopharmacies, nuclear medicine departments and National Metrological Institutes (NMI) for the accurate determination of calibration factors of many radionuclides. The National Physical Laboratory (NPL) Vinten 671 chamber is available at several NMIs and its design is reflected in the commercial NPL Fidelis secondary standard radionuclide calibrator found in many hospitals in the UK. One of the earliest Vinten chambers ever built has accumulated a long measurement record in radionuclide metrology at the NRC. Owing to this longevity and its ubiquitous use, it was selected as a proving ground for a new Vinten model built with the EGSnrc software, the de facto gold standard for Monte Carlo (MC) radiation transport simulation. The chamber was previously studied using the legacy EGS4 code, but critical improvements in electron transport algorithms and the recent implementation of detailed radionuclide decay schemes prompts us to revisit modelling using EGSnrc. We validated our new computational Vinten model against measurements of radionuclide artifacts whose activity was determined by absolute methods. The MC and experimental calibration factors are found to agree at the percent level, in absolute terms.

The response of the Vinten chamber is first calculated as a function of gamma energy, from which in principle the calibration factor for any particular gamma emitting isotope can be estimated. But more strikingly, the explicit simulation of radionuclide decay now permits the direct determination of a calibration factor, including additional effects due to all decay paths of the radionuclide, e.g., high energy beta particles reaching the sensitive volume, bremsstrahlung photons from the initial beta emission, etc. As we demonstrate, such a computational tool begs to address key questions in radionuclide metrology: In what way and how much is the uncertainty of a particular parameter affecting the measurement? What is the calibration factor for a radionuclide not previously measured? What is the calibration factor for a non-standard measurement geometry? What is the effect of radio-impurities on the measurement of a radionuclide?

Corresponding author's email address: **raphael.galea@nrc-cnrc.gc.ca**

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^{99M}TC BY ^{99}MO PRODUCED AT THE ENEA-FNG FACILITY OF 14 MEV NEUTRONS

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Capogni, M.[1-2]; De Felice, P.[1-2]; Fazio, A.[1-2]; Quintieri, L.[1-2]; Pietropaolo, A. [1]; Pillon, M.[3]; Pizzuto, A.[1]

[1]ENEA, [2]ENEA-INMRI, [3]ENEA-FSN (IT)

^{99m}Tc ($T_{1/2}$ = 6.0067(10) h), an isomer of ^{99}Tc emitting γ-ray with energy E_γ=140 keV, is the most widely used radionuclide for diagnosis in nuclear medicine (NM). Europe is the second largest consumer of ^{99m}Tc, accounting for more than 20% of the global market.
^{99m}Tc is derived by the parent radionuclide ^{99}Mo[$T_{1/2}$ = 2.77479(6) d] which is usually produced in nuclear research reactors. A severe supply crisis of ^{99}Mo production [1] in 2008-2009, due to by now aged reactors, with the result in delaying or canceling important diagnostic tests for many patients, highlighted the fragility of the current production chain for this important medical radionuclide.
Following the idea for producing short-lived radionuclides by 14 MeV intense neutron beams recently developed at the Japan Atomic Energy Agency [2], a test for producing ^{99m}Tc from ^{99}Mo, irradiating a natural Molybdenum (Mo-nat) target with the 14 MeV neutron beam of the Frascati Neutron Generator (FNG), was carried out at ENEA, within a scientific collaboration between the Italian National Institute of Radiation Metrology (INMRI), located at the ENEA-Casaccia R.C., and the FNG research laboratory at the ENEA-Frascati R.C.
The experiment was suitably designed according the predictions obtained with Fluka code by simulating the irradiation with the FNG neutron beam of a cylindrical target of Mo-nat powder in a plastic container (2.4 cm^3 containing 4.7 g of natural Molybdenum).
The experimental measurement has been carried out on a target as the simulated one and choosing an irradiation time of about 15 min, with the 14 MeV neutron beam of FNG facility, at an emission rate R=2.9 10^{10} n/s. The activity of the irradiated Mo-nat target was then measured with two high energy resolution HPGe coaxial detectors, both at the FNG laboratory and at INMRI laboratory, where the 4πγ well-type NaI(Tl) 5''x5'' detector has been also used. The ^{99}Mo activity in the irradiated sample measured at high-metrological standard level was estimated to be 4.9 10^4 Bq, at the end of irradiation (EoI) time.
This experiment opens interesting perspectives in ^{99}Mo/^{99m}Tc alternative production in Italy to the classical reactor chain and could be a valid subject of study for further applications in NM.

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[2] Yasuki Nagai, 2014. ^{99}Mo production via ^{100}Mo(n,2n)^{99}Mo using accelerator neutrons. EPJ Web of Conferences 66, 10007. doi:10.1051/epjconf/20146610007

Corresponding author's email address: **marco.capogni@enea.it; antonino.pietropaolo@enea.it; lina.quintieri@enea.it**

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IODINE-123 INTERCOMPARISON EXERCISES: ASSESS MEASUREMENT CAPABILITY IN THE UK HOSPITALS

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Ferreira, K.; Fenwick , A.

NPL (UK)

In Nuclear Medicine, radionuclide calibrators are used to measure the activity before administration to the patient for diagnostic procedures. Two comparison exercises have been performed with ^{123}I to assess the UK hospitals measurement capabilities using radionuclide calibrators.
The comparison exercises were performed in 1999 and 2015. The sources for the 1999 exercise were supplied by NA on behalf of NPL. The solutions were prepared from a bulk solution from which nominal activities were dispensed into 10 ml P6 vials. The sources for the 2015 comparison exercise were prepared and supplied by GE Healthcare on behalf of NPL. The solutions were prepared from a bulk stock solution from which nominal activities were dispensed into 10ml Type 1+ coated Schott© vials. The vials were measured by the suppliers (NA and GE Healthcare) using an in-house calibrator before dispatch to the participants to ensure consistency between samples. A subset of vials were sent to NPL for standardization of the solution by means of ionisation chamber previously calibrated for the radionuclide of interest and to check homogeneity. The subset was chosen to represent 20 % of the overall sample size and it was randomly chosen.
The comparison exercise performed in 1999, showed the fact that small variations in wall thickness of either the sample container or the radionuclide calibrator could result in large variations in the response of the radionuclide calibrator, due to the significant amount of characteristic x-rays, with energies between 27 and 32 keV emitted from ^{123}I. The Institute of Physics and Engineering in Medicine (IPEM) proposed that the administered activity to patients for diagnostic procedures should be within 5%. The exercise performed in 1999 showed that only 66 % of the participants could measure the solution to within 10 % of the standardized value and only 29 % could measure within 5% of the certificated value. The exercise performed in 2015 showed improvement in the hospitals measurement capability, 97 % of participants reported results within 10 % of the certificated activity and 85 % of the participants within the 5 % limit recommended by the IPEM. Moreover 55% of the results were within 2% of the true value. Additionally, the participants were asked to transfer the solution to other containers, such as plastic syringes or other clinically useful geometries, and to report the measurement results to NPL. The use of the "Thomson" copper filter showed to be useful in decreasing the response variability between the different containers measured.
The realization of these exercises are useful to provide the hospitals with traceability to national primary standards and assure measurement capability. Additionally, the obtained results are essential to identify measurement problems and provide possible solutions.

Corresponding author's email address:

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MEASUREMENT OF ^{18}F ACTIVITY BY NAI(TL) WELL TYPE SCINTILATION SPECTROMETER

Vukanac, I.[1]; Kandić, A.[1]; Djurašević, M.[1]; Šešlak, B.[1]; Ciraj Bjelac, O.[1]; Antić, V.[2]

[1]INS, University of Belgrade (RS); [2]CNM, University Clinical Centre of Serbia (RS)

Intercomparative measurements/testing of radionuclide activity calibrators (RC) were performed in October 2015 in a frame of the project Quality Control and Metrological Traceability of Radionuclide Activity calibrators used in Nuclear Medicine in Serbia. Testing of the RC used in nuclear medicine centers in Serbia was performed with ^{99m}Tc, as well as with ^{18}F for two RC devices.
For the purpose of this comparison it was necessary to establish the reliable method/procedure for ^{18}F activity measurement. Since the standard absolute/primary methods for ^{18}F measurements were unavailable, activity of ^{18}F was measured by gamma-ray spectrometry in accordance with laboratory's capabilities.
^{18}F solution was measured by means of NaI(Tl) well-type scintillation spectrometer, in a vial (test tube) geometry, and specific activity was absolutely determined by sum-peak method. Net count rates were corrected for background and decay during measurement. Sample was measured consecutively 80 times, and spectra were recorded with MCA Maestro, Ortec.
Method was established and its validity was tested by using the certified solution of ^{18}F with reference activity of 185 MBq/ml.
Due to high value of solution activity, dead time was higher than 50% at the beginning of the measurement sequence. Influence of the dead time was analyzed and only those spectra where dead time was les than 5% were included in final calculation.
Direct sum-peak method was applied by using the formed system of count rate equations in two manners. Firstly, activity was calculated assuming that angular correlation factor (W) is equal to unity. Secondly, formed system of equation was numerically solved, and factor W was calculated for each of 80 spectra. In both cases total count rate were extrapolated to 0 keV.
In the first calculations, discrepancies between reference and measured values of ^{18}F activity ranged from 3 % to 6 %, with decreasing trend with dead time, while in second case those discrepancies were slightly lower, from 0.1% to 5%, but without any trend.
Uncertainty budget included statistical uncertainties of net count rates in recorded spectra, and uncertainty of solution weighing. Since the uncertainty of final result was calculated as an uncertainty of series of measurements, it was less then 1%.
Analysis of the obtained results showed that, standard direct sum-peak method, improved with numerical approach for solving of system of count rate equation could be considered as a reliable method for determination of ^{18}F activity by scintillation gamma spectrometry.

Corresponding author's email address:

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GA-68 ACTIVITY CALIBRATIONS FOR NUCLEAR MEDICINE APPLICATIONS IN CUBA

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Oropesa Verdecia P.[1]; García Rodríguez L.[1]; Serra Águila R. A.[1]; Moreno León Y.[1]; Jénez Magaña Y.[1]; Cassette P.[2]

[1]CENTIS (CU), [2]CEA/LIST/LNHB (FR)

In this paper the realization and dissemination of the Ga-68 activity measurement unit in Cuba is presented. Firstly, the implementation of the Triple to Double Coincidence Ratio (TDCR) method for the calibration of the activity concentration of a Ga-68 solution using solution's samples of 40 -50 mg contained in low potassium content borosilicate glass vials with 15 ml of ULTIMAGOLDTM liquid scintillation cocktail, a commercial liquid scintillation counter HIDEXTM and a FORTRAN code developed by the CEA-LIST-LNHB for the calculation of the Ga-68 counting efficiencies in the given measurement system is described. The relative expanded uncertainty (k=2) associated to the Ga-68 activity concentration obtained with the TDCR method was equal to 2 %. This uncertainty estimate was confirmed applying the method to a standard solution of Na-22 - a positron emitter with similar to the Ga-68 disintegration scheme from the point of view of type and energy range of emitted particles detected in the used measurement system. Indeed, the observed difference between the measured Na-22 activity concentration by the TDCR method and the corresponding value certified by the NIST was 0.15 %.

Outcome of the transferring of the Ga-68 activity unit realized with the TDCR method to the secondary standard radionuclide calibrator, CAPINTEC CRCTM 15R are also shown. The transferred geometries of interest in nuclear medicine were 15 R glass vial with 6 ml of radioactive solution and plastic syringes of 2.5 and 5 ml capacity containing 2 ml of radioactive solution both, respectively. The new calibration factors determined for the Ga-68 measurements in the secondary standard radionuclide calibrator consisted in a combination of the manufacturer dial setting (416) and the calculated correction factors for the transferring geometries, namely, 0.936, 0.894 and 0.892 factors with relative expanded uncertainty (k=2) equal to 4.6, 3.2 and 3.0 %, for the 15 R glass vial and plastic syringes of 2.5 and 5 ml capacity, correspondingly.

Finally, the calibration results of three radionuclide calibrators, two of model CAPINTECTM CRC-25PET, and another instrument of CAPINTECTM CRC-15R model, for their used during Ga-68 measurements in PET applications in a hospital, are presented. Specifically, new dial settings were determined with relative expanded uncertainties (k=2) ranging from 4.9 to 5.5 %, depending on the reference geometry and the specific calibrated calibrator.

Corresponding author's email address: **poropesa@centis.edu.cu**

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NEW AND UPDATED CALIBRATION FACTORS FOR THE NPL SECONDARY STANDARD RADIONUCLIDE CALIBRATOR

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Fenwick, A. J.; Ferreira, K. M.; Pearce, A. K.; Keightley J. D.

NPL (UK)

The provision and dissemination of radioactivity standards within the UK is largely dependent on the suite of ionisation chambers maintained at the NPL. These chambers are vital for underpinning work in the medical, industrial, low level and nuclear data fields. The NPL provides ongoing support for the commercially available 'Fidelis' secondary standard radionuclide calibrator (previously marketed as the Vinten 671/271, ISOCAL IV or NPL-CRC and based on the NPL designed IG-42 chamber marketed by Centronic Ltd.). This ionisation chamber is used throughout the world both by industry and national metrology institutes to demonstrate traceability to NPL and subsequently BIPM.

The NPL maintains two secondary standard ionisation chamber systems which have been characterised over several decades, and a commercially available Capintec CRC-25R Radionuclide Calibrator which provides a reference to clinically relevant measurements. These systems allow rapid dissemination of radioactivity standards to the user community by means of providing calibrated solutions, calibration of solutions sent into NPL, or by dissemination of calibration factors to users. Additionally, the provision of calibration factors for the Vinten 671 system allows NMIs and other designated institutes to rapidly compare primary measurements with the NPL and other institutes without the need for a worldwide comparison exercise.

This paper provides an updated list of calibration factors for the NPL secondary standard ionisation chambers and commercial calibrators for over 80 radionuclides including; ^{64}Cu, ^{18}F, ^{11}C, ^{68}Ge/^{68}Ga, ^{177}Lu, ^{131}I, ^{123}I & ^{75}Se. Calibration factors are included for a range of commonly used geometries and include volume correction factors where possible. Full details of the source holders used on the Vinten 671 system are also included to allow exact replication of measurements.

Corresponding author's email address:

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QUALIFICATION OF A PRECISION PATTERN DISPENSER

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Van Ammel, R.[1]; Sobiech-Matura, K.[1]; Vercammen, T.[2]; Castelein, P.[3]; Wuyts, R.[2]; Paul, J.[3]

[1]JRC-Geel, [2]SampleQ, [3]Flamac, a division of SIM

The European Commission Joint Research Centre in Geel (JRC-Geel) is on a regular basis preparing radioactive sources. These sources can be used as calibration standards or reference sources in different applications, e.g. proficiency testing of laboratories monitoring radioactivity in the environment organized by JRC-Geel.
In order to automate the sample preparation process a precision pattern dispenser was designed to reproducibly dispense radioactive solutions. The set-up should be able to dispense preset amounts of stock solution in an automated and reproducible way in predefined positions or patterns. Correct dispensing should be ensured by defining the positions of ampoules or special source holders precisely. The set-up must also be able to dispense preset amount of liquid according to designed patterns on air filters having a maximum size of 60x60 cm. During all manipulations no evaporation of stock solution can be afforded in order not to compromise its concentration. The liquids to be handled are mainly acidic solutions so all parts must be resistant to the acidic environment. The number of instrument parts that come in contact with the radioactive solution must be kept to a minimum and should be easily replaceable to reduce the amount of radioactive waste.
The precision pattern dispenser is composed of a commercially available sample handling unit that makes use of syringes to handle the liquid. This unit is coupled to an XYZ-table where e.g. air filters can be fixed on. A custom-made interface (protocol generator software) is used to design dispensing patterns on filters of different sizes and shapes. The combination of the filter and the pattern, called protocol, is sent to the software steering the sampling unit and the XYZ-table. The mass or volume to be dispensed and the position where it has to be dispensed is easily introducible and made visible in the protocol generator.
Qualification tests were conducted to evaluate the performance of the instrument and to assess the compliance with the requirements. The two most important parameters tested were trueness and precision. The maximum values set up in the specifications were 2 % for trueness and 1 % for precision.
Tests were performed by dispensing water into glass ampoules. For each test, a set of 10 ampoules was dried, weighed and put in a tray. After dispensing the preset amount of water in the ampoules, they were weighed again. The calculated mass difference gave the exact amount of liquid dispensed. The tests were carried out using six different syringes (volumes ranging from 50 µL to 5 mL) dispensing a variable water volume from 20 µL to 4 mL. The results for trueness varied between 0.14 % and 1.67 %. For the precision they varied between 0.02 % and 0.45 %. For small dispensed volumes the values for trueness and precision were higher compared to the bigger volumes.
All other parameters described above were examined and found to be compliant with the specifications. The results of the tests demonstrate that the precision pattern dispenser can dispense liquid within the required specifications. It can also be used to prepare air filters with an almost homogenous distribution of activity within a predefined area. The precision pattern dispenser makes it possible to prepare sources for different applications in a fast and accurate way.

Corresponding author's email address: **Raf.VAN-AMMEL@ec.europa.eu**

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VERIFICATION OF ^{137}CS DETERMINATION IN LARGE VOLUME SEAWATER USING CU-HEXACYANOFERRATE CARTRIDGE FILTERS

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Visetpotanakit, S.; Khrautongkieo C.

OAP (TH)

A rapid method to determine ^{137C}s in seawater using Cu-hexacyanoferrate ion exchanger has been developed and verified for the purpose of environmental radiation monitoring (for both normal situations and any radiological accident). The Cu-hexacyanoferrate precipitate supported cotton-wound cartridge filters were prepared by submerging filters in $K_4[FeCN_6]$ and $Cu(NO_3)_2$ solutions. After rinsing and drying the filters were ready to be used as the ion exchanger. For the method verification, the prepared filters were fabricated in the filtration set equipped with a low flowrate pumping system. The filters were used to absorb cesium from spiked synthetic seawater containing 0.191 ± 0.002 Bq/L of ^{137}Cs activity by passing various large volumes i.e. 200L, 300L and 400L over the filters with flow rate of 240 L/hr. The filter samples were detached and taken for a sample preparation step. The samples were dried and converted to ash through a heating step and directly measured with gamma-ray analysis using a HPGe detector. The result showed accuracy with a bias below ± 25 %, in the range from - 24.63% to + 3.29 % and the Cs collection efficiency varied from 48.09 - 94.82%. The variation in results was due to some loss of Cu-hexacyanoferrate and unstable flowrate during filtration, this can be improved by enhancing the quality of the prepared Cu-hexacyanoferrate filters, controlling and reducing the flowrate during filtration. This developed method is cost-effective and easy to perform in sampling fields and more importantly can be optimised for accuracy with filtration time i.e. sample volume and flowrate, depending upon situations and purposes for data use.

Corresponding author's email address: **suputtra.v@oap.go.th**

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ACTIVITY STANDARDISATION OF ^{210}PB BY LIQUID SCINTILLATION COUNTING METHOD

097

Havelka, M.

CMI (CZ)

The standardisation of ^{210}Pb is usually complicated by the presence of daughter products, ^{210}Bi and ^{210}Po. However, if the standardised solution contains ^{210}Pb in the radioactive equilibrium with ^{210}Bi and ^{210}Po, it is possible to utilise the activity determination of ^{210}Po for standardisation of mother nuclide ^{210}Pb. ^{210}Po, as pure α particle emitting nuclide, can be easy standardised after the separation from ^{210}Pb solution by 4πα-liquid scintillation counting, where unit detection efficiency for alpha decay is expected. For the separation of ^{210}Po from ^{210}Pb solution, two methods based on precipitation of Pb, eventually Pb with Bi, remaining Po in solution, were introduced.
The first one used precipitation of Pb in form of $Pb(NO_3)_2$ from mixture of acetic acid-toluene-water-HNO_3, where approximately 99% of ^{210}Po and ^{210}Bi remained in the solution. Subsequently, both fractions, precipitate and solution, were used for source preparation for LSC. The source originating from Pb precipitate was counted immediately after separation in order to calculate ^{210}Po separation loss, the source with ^{210}Po was counted after ^{210}Bi decay to determine ^{210}Po activity. The ^{210}Pb activity result with overall standard uncertainty less than 0.45% was reached.
The second separation method was based on co-precipitation of Bi and Pb with KCl from organic solvent mixture, whereas ^{210}Bi with ^{210}Pb was fixed to the precipitate, while Po remained practically in the solution. Advantage of this method is that the LSC sources could be counted immediately after the radionuclide separation, however in this case the determination of separation loss was more complicated. The results obtained by both methods will be presented.

Corresponding author's email address: **mhavelka@cmi.cz**

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THE METHOD OF PRODUCTION AND QUALITY CONTROL OF VOLUME MULTIGAMMA SOURCES WITH DIFFERENT MATRICES

101

Listkowska, A.; Lech, E.; Saganowski, P.; Tymiński, Z.; Ziemek, T.; Dziel, T.; Kołakowska, E.

NCBJ RC POLATOM (PL)

The standard sources in Marinelli beakers (volume sources) are widely used for calibration of scintillation or HPGe detectors e.g. in industry, environmental protection, dosimetry of the ionising radiation and nuclear medicine. Measurements of specific materials should be preceded by calibrating with standards with matrices densities and geometries similar to the real sample.
As a depositary of the National Standard of Radionuclides Activity in Poland Laboratory of Radioactivity Standards (LRS) in Radioisotope Centre POLATOM produces standard solutions and different types of radioactive sources, among others for calibration of gamma spectrometers. There is a need for new types of volume sources to cover diversified customers' requests. The aim of this work was to develop a set of new compositions of used matrices based on epoxy resins and to establish a validated production and QC procedure for this kind of sources. These matrices should be solid, homogeneous and long-lasting in order to use them as the filling of Marinelli bakers.
In this study the optimal proportions of the components of the matrices were determined, with aim of getting densities higher than water equivalent. Homogeneity of final materials was studied with use of ^{85}Sr spiking and measurements with the scintillation counter with NaI(Tl) detector. After confirming the homogeneity and the permanence of the matrix the multigamma Marinelli beaker standards (including ^{241}Am, ^{109}Cd, ^{57}Co, ^{51}Cr, ^{113}Sn, ^{85}Sr, ^{137}Cs, ^{54}Mn, ^{65}Zn and ^{60}Co) were prepared in order to calibrate 3 HPGe detectors: GEM20P4, GX1520, GX2018. The activity of these sources was determined with reference to the National Standard of Radionuclides Activity in Poland, which is stored and used in LRS. For the quality control purpose calibrated germanium detectors were used for the measurement of new standards with known activities. The maximum difference of measured values and the values calculated from radioactive concentration of used solutions standardized with absolute methods in the National Standard of Radionuclides Activity didn't exceed 2.5 %. These values are fulfilling the requirements for standard sources therefore this type of standards will be implemented for the production in RC POLATOM.

Corresponding author's email address: **a.listkowska@polatom.pl**

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CONSISTENCY OF PHOTON EMISSION INTENSITIES FOR EFFICIENCY CALIBRATION OF GAMMA-RAY SPECTROMETERS IN THE ENERGY RANGE FROM 20 KEV TO 80 KEV

058

Lépy, M.-C.; Ménesguen, Y.; Riffaud J.

CEA, LIST,LNE-LNHB (FR)

Barium-133 and americium-241 are frequently used to calibrate gamma-ray spectrometers in the low-energy range, where only a few gamma-emitting radionuclides are available for this purpose. Systematic deviations with relative differences of about 2-3% between the full-energy peak (FEP) efficiencies experimentally obtained for the peaks at 53 keV (^{133}Ba) and 59.6 keV (^{241}Am) were observed, despite the small energy gap between them. Moreover, the relative uncertainty associated to the emission intensity of the 79.6-keV line of ^{133}Ba is 7 %.

It was then decided to examine in details the experimental FEP calibration in the low-energy range for different high-purity germanium (HPGe) detectors, with different sizes (coaxial 100 cm^3, planar 2 cm^3 and 40 mm^3) and to compare two different approaches:

i.The conventional method, using standard radioactive sources was applied for the two large detectors. Careful analysis of the peaks shape (modelled by a Gaussian for gamma rays or by a Voigt for X rays), including tailing due to scattering in the source was performed in order to derive experimental efficiencies with about 1 % relative standard uncertainties. Complementary information was also obtained by Monte Carlo simulation using the PENELOPE code, after optimization of the geometrical parameters.

ii.For the last detector, characterized by a small size and high energy resolution (full-width at half-maximum (FWHM) of 115 eV at 5.9 keV), a dedicated experiment was performed using a tunable monochromatic photon beam and a reference proportional counter to determine the thickness of absorbing materials inside the detector. Thus the FEP efficiency could be derived without reference to radioactive standards. This calibration result could be compared to the one obtained in a previous experiment [1] and discussed.

This was used to measure photon emission intensities of several low-emitting photon radionuclides, including ^{133}Ba, and compared to the tabulated values in the NUCLEIDE database.

[1] Plagnard, J., Bobin, C., Lépy, M.-C., 2007a. Accurate efficiency calibration of a low-energy HPGe detector using a monochromatic X-ray source. X-ray Spectrometry 36, 191–198.

Corresponding author's email address:

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ON HOMOGENEITY APPROXIMATION IN CALIBRATION OF GAMMA-SPECTROMETRY ASSESSMENT OF BULK SAMPLES

067

Sima, O.

PD-UB (RO)

Accurate efficiency calibration of gamma-ray spectrometers for the assessment of solid samples is a challenging task. It can be done by using liquid or water equivalent gel calibration sources, or standards with specific matrices prepared by spiking. In both cases, the effects of the differences in matrix and density between the sample of interest and the standard are subsequently corrected using self-attenuation factors. This calibration procedure is perfectly adequate if the samples (and standards) are homogeneous with respect to matrix and activity distribution. However, specific bulk samples (e.g. sediment, soil) have an intrinsic inhomogeneity, comprising of several components with various matrices and specific activities. In a recent study [1] we developed a Monte Carlo model for the evaluation of the efficiency considering that the sample is composed from a collection of elementary building blocks, each with given matrix and specific activity. We have shown that the efficiency depends on the characteristics of the intrinsic inhomogeneity, in particular on the dimensions of the grains, in specific cases the dependence being significant for low energy photons.

In the present work, using Monte Carlo simulation, we extend the study by evaluating the bias in the efficiency (and consequently in the activity assessment) resulting from the application of the homogeneity approximation for samples with intrinsic inhomogeneity. In a first approach the equivalent homogeneous matrix, obtained theoretically from the total mass of each component present in the sample, is used for the evaluation of self-attenuation corrections. In the second approach, the linear attenuation coefficients are assumed to be evaluated using transmission experiments. In both cases, the bias of the efficiency due to the application of the homogeneity approximation decreases if the size of the elementary building blocks decreases. If the components have similar activities and matrices or the linear attenuation coefficient is low for each component, the bias is generally negligible for blocks of dimensions lower than 0.1 mm. However, in the particular case when one of the components has a much higher specific activity and high density and atomic number, the bias can be significant. For example, in a soil sample in which pitchblende grains of 0.1 mm size are included, the bias in the efficiency is about 20 % at 46.5 keV and about 4 % at 186 keV, in the case when the pitchblende mass fraction in the sample equals 5 %, and the ratio of activity of a pitchblende grain to a common soil grain is 2000. The bias is due to the fact that most of the photons are emitted by the nuclides included in the pitchblende grains, being thus strongly attenuated before exiting the grain, whereas in the case of the homogeneous matrix the same total activity would be distributed in a matrix in which the total pitchblende mass is diluted in the soil and the resulting photon attenuation is lower.

[1]. O. Sima, To be presented at the ICRM-LLRMT Conference, Seattle, September 26-30, 2016

Corresponding author's email address: **octavian.sima@partner.kit.edu**

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DETERMINATION OF THE ABSOLUTE PHOTON EMISSION INTENSITIES OF SOME GAMMA RAYS OF HO-116M

133

Peyrés, V.

CIEMAT (ES)

The metastable nuclide ^{166m}Ho disintegrates by beta minus emission to several excited levels of ^{166}Er. In the subsequent de-excitation, a large number of gamma-rays (more than 50) are emitted. The combination of a relatively long period and the high counting efficiency in gas detectors makes it a possible candidate as component of a reference source for checking the stability of ionization chambers, in replacement of ^{226}Ra sources which are commonly used to this purpose. It is also convenient for energy calibration of Ge and other gamma detectors.
A set of gamma ray probabilities from this nuclide have been measured at CIEMAT using a well characterized Ge detector. Point sources were produced by gravimetric methods from a reference solution standardized in the frame of the EURAMET.RI (II)-K2.Ho-166m activity comparison.
The procedure included the calculation of the detection efficiency by combination of two Monte Carlo calculations, one for the specific energy of the gamma transition under study and a second including the complete decay scheme. Coincidence-summing factors were obtained from the results of both calculations. Peak analysis was carried out using the GRILS program from the GANAAS package (IAEA). Several measurements were carried out at a source-to-detector distance of 15 cm and typical counting times were about 5×10^4 s.
The results obtained for more than 10 gamma emissions are compared to reference values. In general, uncertainties are lower than those from previous measurements.

Corresponding author's email address: **virginia.peyres@ciemat.es**

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DETERMINATION OF A HPGE DETECTOR EFFICIENCY CURVE BY MONTE CARLO SIMULATION IN THE ENERGY INTERVAL FROM 20 TO 1408 KEV

012

Haoran, L.[1]; Jian Z.[1]; Zeshu, L.[2]; Juncheng, L.[1]

[1] NIM; [2] CDC (CH)

A new extended range coaxial Ge detector (model number GX6520) made by CANBERRA has been installed in National Institute of Metrology (NIM). To achieve a higher accuracy of activity determination of γ-emitting radionuclides, it is necessary to employ a precise calibration for the detector. In this paper, we report a careful calibration of GX6520 detector by Monte Carlo simulation in the energy interval from 20 to 1408 keV.

To run Monte Carlo simulation, a detector model was created using DETEFF code. Since the detector parameters specified by the manufacturer are known with insufficient accuracy, a simple way to access a better knowledge of detetor's dimensions can be by X-ray radiography. The following parameters were obtained by X-ray radiography: crystal diameter, crystal length, window diameter, window thickness, window-to-crystal distance, crystal hole diameter, crystal hole length, mount cup diameter and thickness, end-cap diameter and thickness, radius of the upper edge rounding. Due to the inherent limitation of DETEFF code, we didn't describe mount cup in the detector model even when the values were known. To estimate the thickness of germanium dead layer, we adjusted the model to match the experimental results of point sources. The window material acknowledged by the manufacturer is carbon composite, but without detailed composition. For this reason, we compared the difference for different window materials in Monte Carlo simulations. The results indicated no significant difference between materials with close atomic number and with same density. The relative deviation of detector efficiency for Co-60 as well as Cs-137 is less than 0.5% between carbon and beryllium window. As a result, a pure carbon window with density of 2.26 g/cm^3 was chosen for final usage instead of real composition in the detector model.

A set of calibrated point sources were used to validate the Monte Carlo calculations. The sources were placed on the axis of the detector. To avoid a heavy coincidence summing effect, we set the source-detector distance to be 25 cm for each measurement. Full energy peak efficiency calibration was carried out in the energy range of 59.5 to 1408 keV using both NIM and PTB standard point like sources with the following radionuclides: Am-241, Cd-109, Ba-133, Eu-152, Cs-134, Cs-137, Co-60.

The results of Monte Carlo simulation and experimental calibration show good agreement between each other. The relative deviation between simulated and experimental efficiencies is within 1.5 % in most cases, only except at 122.06 keV, which has a relative deviation of 1.9%. The results of the above suggest that the detector model is useful for accurate calculations of efficiencies for HPGe detectors.

Corresponding author's email address: **liuhr@nim.ac.cn**

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DETERMINATION OF THE NEUTRON ACTIVATION PROFILE OF CORE DRILL SAMPLES BY GAMMA-RAY SPECTROMETRY

014

Gurau, D.[1]; Bodenb, S.[2]; Simac, O.[3]; Stanga, D.[1]

[1] IFIN-HH, Magurele, (RO), [2] SCK-CEN, (BE), [3] UB, (RO)

In decommissioning projects, it is highly desirable to know the activity profile of the in-depth contamination or activation of concrete building structures to optimize radioactive waste management. Core drill sampling followed by gamma spectrometry measurements is the only way for determining the activity distribution of radionuclides produced by neutron activation inside the biological shield of nuclear reactors. Moreover, the core sampling is used as a reference method for validating the results obtained by Non Destructive Assays based on In-Situ Gamma Spectrometry. For decommissioning projects, the major objectives in determining the activity profile of core samples are: (i) finding a suitable function for the activity depth distribution and calculating its parameters; (ii) computing the effective contamination depth for which the specific activity of the core sample is smaller than the clearance level and (iii) computing the effective total activity of core samples.

This paper provides new guidelines for determining the activity profile of core drill samples considering that: (i) a function can be assumed for the one-dimensional activity depth distribution from physical considerations and (ii) a polynomial function can be used assuming that the activity depth distribution is a smooth and continuous function which can be well approximated by a polynomial.

The determination of the activity profile is based on the measurement by gamma spectrometry of the activity of cylindrical slices (discs) cut from the core sample. We consider a disc located between coordinates *xi* and *xi+1* of the core sample which was cut so that its base is perpendicular to the core axis. For thin discs (assuming a linear activity depth distribution), we have

$$\Lambda_i/m_i = (1/d_i)\int_{x_i}^{x_{i+1}} \Lambda_s(x)dx = \Lambda_s(\overline{x}_i) = f(x_i, d_i, p_1, p_2, ..., p_m)$$

where Λ_j and m_i are the activity and the mass of the disc, $\Lambda_s(x)$ represents the specific activity of the core-drill sample as a function of $x, f(x, p_1, ...p_m)$, is the model function assumed for $\Lambda_s(x), p_1, p_2, ...p_m$ are the parameters of the function, $\overline{x}_i = (x_i + x_{i+1})/2$, $d_i = x_{i+1} - x_i$, $i=1,...n$ and $n > m$. From Eq. (1), an overdetermined set of n equation is obtained. For thick discs Eq. (1) becomes: $\Lambda_i/m_i = F(x_i, d_i, p_1, p_2, ..., p_m)$ where $F(x_i, d_i, p_1, p_2, .., p_m) = (1/d_i)\int_{x_i}^{x_{i+1}} f(x, p_1, p_2, ..., p_m)dx$ The effective contamination depth, *xCL*, was determined from equation $f(x, p_1, ...p_m) - \Lambda_{CL} = 0$ where ΛCL is the clearance level (Bq/g). The effective total activity of the core sample, Λ_T, was calculated by means of the following equation $f(x, p_1, ...p_m) - \Lambda_{CL} = 0$ where ρ and S are, respectively, the density and base surface of the core sample. To calculate the parameters $p_1, p_2...p_m$, least square fitting was applied including weighted total least squares method to account for the uncertainties of x_i. In the first step, we used Trendline function from Excel to perform exponential and polynomial fits. The results were refined by means of Matlab software.

The determination of the activity profile based on the gamma spectrometry was applied to core drill samples taken from the biological shield of the VVR-S reactor block from Magurele-Romania. Data obtained by gamma spectrometry were fitted with exponential and polynomial functions. The total activity of the core sample and the effective contamination depth were determined together with their standard uncertainties. The propagation of distributions using MC simulation was used and a Matlab script was written to compute these uncertainties taking into account the uncertainties of both Λi and xi.

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Corresponding author's email address: **doru@nipne.ro**

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IMPACT EVALUATIONS OF ACTIVITY MEASUREMENT RESULTS FOR CLEARANCE OPERATIONS

021

Yeh, C.H.; Yuan, M.C.

INER (TW)

Inside the barrels which contained metal tubes and metal slices, there were point sources, rod sources and slice sources of Cs-137 and Co-60 to simulate the low-level radiowaste whose radioactivity was non-homogeneous distribution in order to observe the standard deviation (σ) of the counting rate as well as the maximum ratios of the counting rate against the relative average values of the ten detectors of the 4π plastic scintillation counter. These two homogeneity indexes indicated the distribution homogeneity status of the radioactivity of the radiowaste in the whole barrel. When the whole barrel radioactivity was homogeneously distributed, the standard deviation(σ) was less than 0.5% and the maximum ratios of the counting rate against the relative average values were less than 1.5. When the whole barrel radioactivity was not homogeneously distributed, the maximum differences of Cs-137 and Co-60 were >26%; when the whole barrel radioactivity was homogeneously distributed, the maximum differences of Cs-137 and Co-60 were respectively >31% and >48%. When the hotspots distribution centered at the center of the whole barrel, the shielding effect of the materials made the two homogeneity indexes smaller but the differences with the standard radioactivity appeared larger. To avoid the clearance operations caused by the release recommendations complying with the IAEA Safety Standards Series No. RS-G-1.7, it is necessary to use the survey meters which are highly sensitive to confirm the actual radioactivity distribution along the horizontal axis and vertical axis of the whole barrel.

Corresponding author's email address: **chyeh@iner.gov.tw**

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D GEANT4 BASED MONTE CARLO SIMULATION FOR VERIFYING THE MODIFIED SUM-PEAK METHOD

034

Aso, T.[1]; Ogata, Y.[2]; Makino, R.[1]

[1] Elec. and Comp. Eng. NIT-Toyama, [2] RIC Nagoya Univ. (JP)

The sum-peak method is a kind of the absolute measurement methods of radioactivity, and is applicable for the radionuclides which emits more than one photon in coincidence. The fundamental formula of the conventional sum-peak method is;

$$N_0 = \left(\frac{N_1 N_2}{N_{12}} + N_t \right) \overline{w}(0) \quad (1)$$

where N_0 is the activity of the source, N_1 and N_2 are the full energy peak count rates, N_{12} is the sum peak count rate, N_t is the total count rate, is the angular correlation, and subscripts 1 and 2 donate the corresponding gamma rays. However, since the formula includes N_t, it is not applicable to samples containing other radionuclides. To overcome the problem, the authors developed the modified sum-peak method which remove Nt from the above formula and was presented in the previous conference [1]. The formula is;

$$N'_0 = \frac{N_1 N_2}{N_{12}} \overline{w}(0) \quad (2)$$

and N'_0 converges at N_0 with increasing the source-to-detector (S-D) distance, which corresponds to decreasing N1. The method was experimentally confirmed via a p-type HP-Ge detector and an n-type one using ^{60}Co, ^{22}Na and ^{134}Cs point sources.

The applicability of the modified sum-peak method needs to be verified in many experimental conditions. In order to efficiently verify the method in various conditions, a Geant4 based Monte Carlo simulation has been applied. At first, the simulation modeled HP-Ge detector of n-type. The basic parameters were taken from the datasheet of GMX-25190 Ortec. The source of radionuclide was placed at a certain distance from the surface of the detector. The deposited energy in the Ge crystal inside the HP-Ge detector was integrated for each decay. Then detected energy was recalculated by emulating detector response according to the energy dependent energy resolution of the detector. Finally, the gamma-ray spectrum was collected as the series of detected energy in each decay. The peak count rate was obtained by fitting energy spectrum around the peak with a composite function of exponential and Gaussian. The simulations were repeated while varying the S-D distance along the axis of the detector. The physics-list in Geant4 defines physics processes to be applied for particles. Our physics-list takes into account for all particles and interactions including the radioactive decay of isotopes, electromagnetic physics for gamma, electron and positron. In this study, the time stamp of each decay was omitted, so that the activity corresponds to the number of simulated decays.

Firstly, the relative efficiency (RE) was estimated in the simulation. It was roughly consistent with the measured RE. Secondly, the simulations had been performed by assuming a ^{60}Co point source with various S-D distances. N_0 and the extrapolated value of N'_0 in the limit of N_1->0 agreed with the number of decays in the simulation. Therefore, the simulation has been validated to apply to studying the modified sum-peak method in various source conditions.

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Corresponding author's email address: **aso@nc-toyama.ac.jp**

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WELL TYPE VS COAXIAL TYPE HPGe DETECTOR. THE SIGNIFICANCE OF SELF-ABSORPTION CORRECTIONS IN Pb-210 GEOCHRONOLOGY

043

Iurian, A-R.[1]; Millward, G.[1]; Sima, O.[2]; Taylor, A.[1]; Blake, W.[1]

[1]SGEES-PU (UK), [2]PD-UB (RO)

High resolution gamma-ray spectrometry is widely used for radionuclide determination in different sample matrices, and a wide range of radiation detector systems are nowadays available. Pb-210 determination for geochronological applications is very often performed by gamma-ray spectrometry as it is a fast and non-destructive method. However, it is often the case that only small amount of sample is available for each sediment section within the core length and a well type detector is necessary to be employed for Pb-210 measurements (either singular or in combination with a coaxial type detector). The paper aims to (i) discuss the advantages and disadvantages of employing well type vs coaxial type detectors for Pb-210 activity determinations in geochronological applications; (ii) outline the importance of self-attenuation corrections for both well and coaxial type detectors, even when an 'in-house' soil standard of similar matrix with the samples is used for efficiency calibration of the gamma detectors; and (iii) to give a practical example on the contribution of self-attenuation corrections factors on the derived geochronological data and sedimentation rates for two sediment cores from east Cornwall, UK, when using both vial and cylindrical geometries. An efficiency transfer approach was applied for Pb-210 activity measurements in salt marsh sediment samples. The self-absorption correction factors were determined following the procedure of [1] for vial geometries measured on well type detector, or based on the transmission method proposed by [2] for cylindrical geometries measured on coaxial type detector. The Pb-210 experimental efficiency was obtained by measurement of a soil standard prepared 'in-house' from a multinuclide standard solution supplied by Eckert&Ziegler. The quality assurance of the measurements was established through gamma-ray spectrometry analysis of two IAEA Proficiency Test materials containing known Pb-210 activity concentrations, and Student t-test agreement was found for both geometries at 95% confidence level. The consequence of employing well type or coaxial type detectors for Pb-210 activity determinations in geochronology applications is further on discussed. Nineteen sediment layers of a salt marsh monolith were dried, homogenized and encapsulated in both vial and cylindrical geometries for Pb-210 determination. The Student's t-test showed agreement at 95% confidence level between decay-corrected Pb-210 activity values obtained for the 19 sediment samples in vial and cylindrical geometries. However, the combined standard uncertainty of the measurement result at 1σ confidence level and for 48h measurement time reached 4-7% for cylindrical geometry, and 7-14% for the vial geometry. Values of experimental efficiency transfer factors (ETF) for vial geometry were about 7 - 11% lower than the ETF values for cylindrical geometry, showing a higher self-attenuation effect for the coaxial detector compared with the well detector. The geochronological data were derived for two salt marsh sediment sequences using (i) Pb-210 measurements by gamma-ray spectrometry, and (ii) vial and cylindrical geometries from two combined sediment cores in each site. Differences in apparent density (up to 30 - 73%) and chemical composition (e.g. for Fe, Si, Na, C, Ca) were found between the 'in-house' soil standard and the sediment samples from the two study sites. Neglecting Pb-210 self-attenuation corrections can lead to different ages and/or sedimentation rates when using Pb-210 dating approach.

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Corresponding author's email address: **andra.iurian@plymouth.ac.uk**

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PORTABLE REAL TIME IN-SITU GAMMA-RAY ANALYSIS SYSTEM

047

Wei, C.[1]; Morris, K.[2]; et al.

[1]FDA (USA); [2]Canberra Industries (USA)

This presentation communicates a collaboration effort between Canberra Industries and the US FDA Winchester Engineering and Analytical Center (WEAC) in developing rapid turnaround and high throughput analytical systems for radionuclide analysis. Conventional gamma-ray analysis demands laboratory analysts to process samples, place the processed samples in a gamma-ray spectrometry system for gamma-ray counting and performing data processing and analysis for laboratory conclusions besides ensuring quality control requirements are met. The instruments used for quantitative gamma-ray analysis are normally too heavy to be mobile and to be utilized for field applications. The presented portable gamma-ray analysis system realized the following design features: free of gamma-ray spectroscopy knowledge and experience requirements of an end user; light weight, modular, scalable, mobile; robust; user friendly; laboratory analytical data quality; real time in-situ and high throughput sample data collections and analysis. The system includes a field unit and a central laboratory instrument controller. It allows automatic data transmission from the field unit to the central laboratory controller when both units are connected to the internet. The field unit consists of a modular and scalable cart, a CeBr3 gamma-ray spectrometer with a modular and scalable shield, a camera, a GPS tag, and an instrument controller with a user friendly graphic user interface (GUI) enabling data collection with a few simple operation steps of an end user. The camera is used for capturing how a sample is prepared and filled into a counting container by an end user and photo files will be transmitted with the rest of data files as one of the quality control measures. The central laboratory instrument controller's application software features simple steps needed to receive and populate data collected from field for efficient data processing and analysis by central laboratory analysts. The uncertainty estimate of the system at the limit of quantification (LOQ) yielded 17% for both Cs-134 and Cs-137, and 19% for I-131 at k = 1. The average minimum detectable activity concentration (MDC) is 36 Bq/kg for Cs-134, 34 Bq/kg for Cs-137, and 35 Bq/kg for I-131 based on data collected with 20 minute counting of each sample obtained from 25 water blanks, 50 food matrix blanks with 19 different matrices, and 59 spiked food samples with 15 different matrices. A comparison of the above MDC data to the FDA's recommended derived intervention levels (DILs), 1200 Bq/kg for Cs-134 plus Cs-137 and 170 Bq/kg for I-131, indicated that the system's data quality objectives are adequate for radiological or nuclear emergency response or a targeted surveillance program for analyzing samples that could be contaminated due to a nuclear incident such as Fukushima nuclear power plant accident. The implementation of such a system will dramatically increase sample analysis throughput, reduce sample analysis turnaround time from a week or weeks to less than a day, and therefore address the current radio-analytical expertise shortage issue and increase the capacity in responding to a radiological or nuclear incident where gamma-ray analysis is needed.

Corresponding author's email address: **cong.wei@fda.hhs.gov**

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DETERMINATION OF ABSOLUTE PEAK AND TOTAL EFFICIENCY FUNCTIONS FOR GE-DETECTORS BY ITERATION CALCULATION USING A MIXED RADIONUCLIDE SOURCE

048

Ishizu, H.; Yamada T.

JRIA(JP)

True coincidence summing may arise in case of nuclides which emit two or more gamma-rays within resolving time of germanium detector, especially when the source is positioned close to the detector. In such a case, not only absolute peak efficiency but also total efficiency should be required to correct effects of coincidence summing. In the conventional approach, single gamma-emitting nuclides are preferably used to avoid summing effects to determine these two kinds of efficiency functions. However, it would be difficult to exclude some summing nuclides such as ^{60}Co or ^{88}Y in order to obtain a peak efficiency curve for a wide energy range, arising error to determine an efficiency function due to large uncertainties of summing correction. In addition, since there are few available non-summing nuclides especially for high energy range, it is difficult to obtain total efficiencies accurately by the conventional approach. Instead of such a conventional technique, Monte-Carlo simulation was adapted by several authors [1]. However it might be not easy to reproduce a real geometry of the detector (e.g. dead layer thickness and its aging variation, sizes of a detector, impurities of constitutional materials). As another approach, Blaauw developed a direct calibration technique of absolute peak and total efficiency curves using a multiple gamma-emitting nuclide of ^{82}Br [2]. In our previous study, these two functions of a well-type germanium detector were successfully determined using ^{75}Se and ^{134}Cs sources in place of ^{82}Br [3], though very large amount of coincidence summing effects were found in the measurement. In use of these two nuclides, however, it would be difficult to determine efficiency functions accurately in wider energy range 1.3 MeV and over. In order to determine efficiency functions in an energy range from 70 keV to 2700 keV by use of not only single peaks but also sum peaks without extrapolation approximating approach, other nuclides were additionally adapted in the present study. Blaauw also suggested that the direct calibration method could be applied for mixed nuclide reference source with a suitable combination of nuclides [4]. However since suitable choice of nuclides and adequate activity fractions of each nuclide in the method are still unclear, we studied applicability for combination of other nuclides and optimal activity levels in this technique.

In the present study, the absolute peak and the total efficiency functions of a well-type germanium detector were determined using a point source of mixed nuclides with several combinations of ^{60}Co, ^{75}Se, ^{88}Y, ^{134}Cs or others. In the present measurements, 1mm thickness of brass inner liner for X-ray attenuation was used in order to simplify the iteration algorithm based on maximum likelihood estimation. The iteration calculations for directly estimating coefficients of efficiency curves were carried out by use of home-made program coded with VBA and the solver add-in in ExcelTM. As a result, efficiencies in an energy range from 70 keV to 2700 keV were obtained by using the spectra. In order to verify the choice of nuclides mentioned above, the direct calibration approach was also applied to several types of combinations of selected peaks obtained by the same spectra. This method using such mixed nuclides might be also applied to an ordinary cylindrical p-type germanium detector with some source conditions including extended sources.

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Corresponding author's email address: **ishizu-hidetake@jrias.or.jp**

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SUBTRACTION OF NATURAL RADIONUCLIDES CONTRIBUTION FROM GAMMA SPECTRA MEASURED BY HPGE DETECTOR

064

Petr Kovar, A.; Jaroslav Solc B.

CMI (CZ)

Within the European joint research project "Metrology for radiological early warning networks in Europe" a problem had to be solved regarding high contribution of natural radionuclides to gamma spectra measured by HPGe detector. Because the contribution of artificial radionuclides only is usable for early warning purposes and the contribution of natural radionuclides is highly variable from sample to sample, a method had to be used enabling subtraction for each measured spectrum.

The method was developed for measurement of airborne radioactivity concentrations of aerosols using modular portable aerosol collection device based on the mechanically-cooled HPGe detector ORTEC IDM-200-V (rel. efficiency 50%, FWHM 2 keV) calibrated for filters in close measuring geometry. Principle of the method, so called "software shielding", is to determine detector pulse-height spectra of pre-selected natural radionuclides by Monte Carlo (MC) simulations and then subtract, in successive steps, each spectrum from measured spectra.

The following work was done:

(a) Creation of a precise MC model of the whole measuring geometry including the detector, lead shielding, sample holder, etc.

(b) Calculation of detector pulse-height spectra for radionuclides Bi-214, Pb-214, Pb-212 and Tl-208, taking into account true summation effect.

(c) Subtraction of contribution of natural radiation to the measured spectra performed as follows:

-Subtraction of cosmic background from the measured spectrum;

-For each pre-selected natural radionuclide:

-Determination of the net area of one or a few suitable full-energy peaks of the natural radionuclide in the measured spectrum;

-Matching the MC-simulated spectrum to the measured one in such a way that net areas of the suitable full-energy peaks equal to those in the measured spectrum;

-Subtraction of the matched MC-simulated spectrum from the measured one.

The method was validated by addition spectra of spiked filters with different activity levels of Cs-137 and Co-60 to the measured spectra and experimental detection limits evaluation.

Comparison of measured and calculated detection limits confirmed their reduction by the factor of 5 for Cs-137 and 10 for Co-60.

The "software shielding" can be applied to all measurements where contribution of natural radionuclides disturbs evaluation of gamma spectra when identifying artificial radionuclides and determining their activities, e.g. radioactive waste measurement or ground radioactivity monitoring after a nuclear accident.

Corresponding author's email address: **pkovar@cmi.cz**

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DETERMINATION OF 134CS ACTIVITY BY THE SUM-PEAK METHOD VIA A WELL TYPE OF GE DETECTOR

066

Ogata, Y.[1]; Itadzu, H.[2]; Kojima, S.[3]

[1] RIC, Nagoya Univ., [2] SEIKO EG&G, [3] Aichi Med. Univ. (JP)

Since 134Cs emits more than ten gamma-rays per a disintegration, one should thoroughly pay attention to the true coincidence sum (TCS) effect. Especially, the effect is severe for well-type of detectors. Alternatively we tied to measure the activity by means of the sum-peak method which contrariwise uses the TCS effect. The formula of the sum-peak method is as;

$$N_0 = \left(\frac{N_1 N_2}{N_{12}} + N_t \right) \overline{w}(0) \qquad (1)$$

where N_0 is the activity of the source, N_1 and N_2 are the full energy peak count rates, N_{12} is the sum peak count rate, N_t is the total count rate, $\overline{w}(0)$ is the angular correlation, and subscripts 1 and 2 donate the corresponding gamma rays. For well-type of detectors, $\overline{w}(0)$ equals unity. Appling the sum-peak method to ^{134}Cs activity determination, the count rates of 605 keV peak, 794 keV peak and their sum-peak was used as well as the total count rate. Whereas, since the sum-peak method needs the total count rate, it cannot be applied when an object contains another radionuclide. Conversely, our concern is to determine the activity of objects sampled in Fukushima after the accident. The samples mainly contain not only ^{134}Cs but also ^{137}Cs. If the contribution by ^{137}Cs to the total counts could be approximated, the activity of ^{134}Cs would be determined by the sum-peak method by subtracting the contribution. The total counts originated from ^{137}Cs would be guessed from its 662 keV peak counts by measuring ^{137}Cs single source. Subtracting the contribution of ^{137}Cs to the total counts of the mixture source, then the sum-peak method would be applied to estimate ^{134}Cs activity. However, the technique would be affected by the ratio of the activities of ^{137}Cs to ^{134}Cs.

The sources, ^{134}Cs, ^{137}Cs and their mixture, were prepared from the standard solutions calibrated by Japan Radioisotope Association (JRIA). From few microliters to 3 cm^3 of aqueous solution containing the radiocesium(s) were put into polyethylene tubes. The volume of used well type HP-Ge detector (GWL-120230, Ortec) was 120 cm^3. The gamma spectrum ranged from 0 to 4 MeV was converted to 8 kch digital data with an ADC (EASY-MCA-8K, Ortec), and the spectrum was analyzed using a software (Spectrum Navigator, SEIKO EG&G).

When ^{134}Cs single source was measured, the activity estimated via the sum-peak method was confirmed to agree well with that of the source. Then the contribution of ^{137}Cs to the total counts depended on the sample volume was assessed as the function of 662 keV peak counts using ^{137}Cs single source. Using the results, the total counts from ^{134}Cs of the mixture source was calculated by subtracting the total counts of ^{137}Cs from the total counts of the mixture source. Consequently, activity of ^{134}Cs was estimated via the sum-peak method. The applicable ratio of ^{137}Cs to ^{134}Cs for this technique will be experimentally verified.

The sum-peak method does not require standard sources; usually those are rather expensive. It was performed that even if an object includes another radionuclide, ^{134}Cs activity can be estimated via the sum-peak method by the appropriate subtraction. A performance of this technique will be presented.

Corresponding author's email address: **ogata.yoshimune@b.mbox.nagoya-u.ac.jp**

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PERFORMACE OF THE SEGMENTED HPGE DETECTOR IN KRISS

068

Han, J. ; Lee, K. B.; Lee, J.M,; Lee, S.H.; Park, T.S.; Oh, J. S.

UST(KR), KRISS(KR)

The segmented HPGe coaxial detector was set up with the digitized acquisition system (DAQ). The detector was manufactured by CANBERRA with dimension, diameter 80 mm and length 90 mm. The crystal was electrically segmented 6 longitudinally and 4 azimuthally. The 24 segments were grounded and one hole in the cylinder was biased to 5000 V. This detector is cooled down by the liquid nitrogen with the 11 liters dewar.
The DAQ was composed of the digitizer (50 M sampling/s), Field-programmable gate array (FPGA), and Real time Operating system. They were evaluated that The Full width half maximum(FWHM), rise time, signal characteristics, and spectrum of the radionuclide source.
All of these performances would be related with the gamma ray tracking system which we build up. The voltage of the cell was histogrammed and FWHM at the 662 keV was 7.58 keV. This voltage was defined to be the difference of the data value at the maximum and minimum. These data were triggered and shaped and histogramed by the paw(CERN) base composed program.
The paw based program handled the raw data from the segmented HPGe detector. The first program is triggering the raw data and selects the main signal and mirror signal. This program also calculates the base voltage which is averaged by the several data. These data were not increased or decreased but only fluctuated and then averaged. This gave the base voltage at the each data set. One data set is composed of 2000 data which is transferred and calculated by the DAQ system. These data were shaped by the second program. The deconvolution method of fast pulse shaping was used for this program. This gives the trap zooidal signal which is shaped. These shaped signals were histogrammed and evaluated by the third program.
The FWHM at the 662 keV was evaluated as 2.46 keV at the a1 cell. The rise time is related with where the interaction happens. The energy which is absorbed at the interaction was independent on the rise time. The gamma ray tracking system is using this rise time and the energy which is absorbed.
The exact evaluation of the absorbed energy and the shape during the rise time gives where the interaction happens in the specific cell.
The analog system using shaping amp and multi-channel analyzer from ORTEC was used for comparing FWHM. The FWHM of analog and digital was 1.88 and 2.46 at a1 cell with 662 keV.
The a2 cell gave 1.57 and 2.18 FWHM, respectively.
All segmented cells are using different amps one another. The azimuthal and longitudinal position can be calculated by the comparison of the mirror signals. The a1 and a2 cells have the same voltage height against the 662 keV. But the voltage which is produced by the 662 keV was different. This difficulty was solved by calibrating on the energy. The Cs-137 and Co-60 were used for the source. These had the 662, 1173, and 1332 keV energies. The positon of three energies was evaluated by the Gaussian fitting for the peaks. The linear fitting about three points gave two parameters, A for the slope and B for the value at the zero energy position. The A and B of a1 cell were 7432.19 +/- 8.637 and 1.97973 +/- 1.269, respectively.
Two values of a2 cell were 6766.34 +/- 0.08843 and 1.06543 +/- 0.01428.

Corresponding author's email address: **donbosco73@naver.com**

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GAMMA-RAY SPECTROMETRY ANALYSIS SOFTWARE ENVIRONMENT

083

Lutter, G.; Hult, M.; Tzika, F.; Stroh, H.; Marissens G.

JRC-Geel (BE)

The low-level radioactivity group at JRC-Geel performs around 250 measurements per year using low-background and ultra-low-background High Purity Germanium detectors. Currently, 13 HPGe detectors are operated in the laboratory HADES, which is located 225 m underground at the premises of the Belgian nuclear research centre SCK-CEN, and 3 detectors are operated above ground at JRC-Geel's site. All detectors have distinct characteristics (type, size and manufacturer) and a wide range of samples are measured with different geometries, masses or matrices. In addition, the facilities of the group are accessible to external researchers through the transnational access scheme EUFRAT (European facility for nuclear reaction and decay data measurements). Currently, 13 external projects are ongoing or have been finalised through the EUFRAT access scheme.

To enable precise and robust analyses, an analysis software environment has been developed. The spectrum analyses are performed using Canberra's Genie 2000. For the efficiency calibration of the detector systems, computational methods are implemented. This is due to the wide variety of samples, which limits the availability of suitable reference materials. The determination of detector response is performed using either absolute Monte Carlo calculation or Monte Carlo efficiency transfer methods based on validated Monte Carlo models of the individual HPGe systems. The detector models have been developed using the EGSnrc software toolkit. The EGSnrc is widely regarded as the most accurate Monte Carlo code for electron and photon transport simulations, nevertheless, it is not including, by default, the possibility to reproduce radionuclide decays and no coincidence summing corrections can be calculated. These corrections are fundamental in many cases in gamma-ray spectrometry. At JRC-Geel, a radionuclide decay generator has been developed and implemented in the EGSnrc code, using directly the nuclear data provided by the Decay Data Evaluation Project. The final activity calculations are realised by specifically developed analysis software, based on an MS Excel sheet with VBA scripts, which reads and uses as input the Canberra Genie 2000 data and the Monte Carlo output files. The ISO11929-3 from 2000 and the ISO11929 from 2010 standards have been implemented in the analysis software. The analysis software is user-friendly and has been used by non gamma-ray spectrometry experts as well, which was triggered by the transnational access scheme. To ensure the robustness of the software in respect to user-effect different features in the analysis have been set to help and warn the user of possible problems (bad peak fitting, possible interference with other radionuclides).

The paper will describe the radionuclide decay generator, its performance and the different features of the analysis software developed at JRC-Geel.

Corresponding author's email address: **guillaume.lutter@ec.europa.eu**

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IMPROVEMENT OF THE ACTIVITY MEASUREMENT METHOD FOR SOLID DOSIMETERS EMITTING X-RAYS

103

Domergue, C.[1]; Riffaud, J.[2]; Philibert, H.[1]; Destouches, C.[1]; Thiollay, N.[1]; Girard, J. M.[1]; Carcreff, H.[3]; Vigneau, O.[3]; Gravier, L.[3]

[1]CEA, DEN, DER (FR); [2]CEA, LIST (FR); [3]CEA, DEN, DRSN (FR)

The MADERE platform, operated by the Laboratoire de Dosimétrie, Capteurs et Instrumentation (LDCI: Instrumentation Sensors and Dosimetry Laboratory) at CEA Cadarache, aims at measuring the specific radio-activity ($Bq.mg^{-1}$) of irradiated solid samples used as neutron dosimeters. The platform is accredited by the French accreditation Committee (COFRAC) for specific activity measurement of γ- and X-ray emitters. Measurements are carried out for reactor dosimetry analysis in support of experimental programs performed in CEA's research reactors and in support of the vessel surveillance program of the French PWR fleet. MADERE will also ensure the dosimetry of the future Jules Horowitz Reactor dedicated to material testing.

Among the measured samples, niobium and rhodium, respectively via inelastic neutron diffusion reactions $^{93}Nb(n,n')^{93m}Nb$ and $^{103}Rh(n,n')^{103m}Rh$, provide information for reactor fast neutron spectrum characterization between 0.5 and 5 MeV, which is the main energy range taken into account for neutron damages to materials.

The activity measurements are carried out by X-ray spectrometry with a relatively high uncertainty up to 6%, to be compared with roughly 1% for standard γ measurements. However the required accuracy measurements should be better than 3% in order to draw pertinent conclusions on calculation schemes and nuclear data used in reactor physics. It must be noted that MADERE performs X-ray spectrometry on solid-state samples, which requires several corrective factors while other laboratories work with liquid-state samples resulting from dissolution of the dosimeters.

Therefore, these last years, the CEA has launched a specific program to improve and validate the X-ray spectrometry measurements methodology and to benchmark MADERE measurements with techniques based on dosimeter dissolution performed by the Laboratoire d'Analyses Radiochimiques et Chimiques (LARC: Radio-chemical and Chemical Analyses Laboratory).

After a description of the X-ray spectrometry measurement process in the MADERE platform, the paper will present the related research program and the main results already obtained to validate the measurement methods. A particular focus will be proposed on the following actions:

a) Development of Monte Carlo simulation (using TRIPOLI4 and GEANT4) to evaluate correction factors (self-fluorescence, self-absorption...) and to improve detector calibration,

b) Improvement of nuclear data with experimental campaigns performed at SOLEIL synchrotron facility (for fluorescence yields measurements) and at ISIS reactor,

c) Experimental validation of self-absorption corrections as a function of sample thickness, based on the measurement of several tens of dosimeters irradiated at the EOLE research reactor,

d) Comparison between solid and liquid niobium sample measurement techniques (^{93m}Nb).

Prospects of these studies will then be presented, assuming that the ultimate goal is to decrease relative standard uncertainties associated to activity measurement down to 3%.

Corresponding author's email address: **christophe.domergue@cea.fr**

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UNCERTAINTY OF DETERMINATION OF Tb-158 IN THE RBMK NUCLEAR REACTOR WASTE

106

Plukis, A.; Barkauskas, V.; Druteikienė, R.; Duškesas, G.; Germanas, D.; Gudelis, A.; Juodis, L.; Plukienė, R.; Remeikis, V.

FTMC (LT)

Optimal characterization of radioactive waste from decommissioning of a nuclear power plant (NPP) is based on the use of the method of scaling factors, in which a massic activity of difficult-to-measure nuclides (alpha-, beta-, and long lived X-ray emitters) in the waste is assessed by measurement of the massic activity of easy-to-measure nuclides (strong gamma-ray emitters). In general, assessment of the massic activity of weak gamma-ray emitter can be done both ways: by using a scaling factor of its activity with a strong gamma emitter, if the origin of a weak gamma emitter is the same as the one of the strong gamma-ray emitter, this scaling factor can be derived by modelling; or by direct measurement based on some calibration, for which modelling of the gamma-ray spectrum from all the nuclides in the mixture is useful. In any case, it is important to establish the origin of gamma-ray emitters, which are presented in the spectrum.

Tb-158 has a complex decay scheme: it decays to Gd-158 through the (ec/ß+) channel with the branching ratio of 0.834 and through (ß-) channel to Dy-158 (branching ratio 0.166). The detection of Tb-158 is possible with a gamma-ray spectrometer at the following energies when the quanta emission probability is above 5% (data taken from the ENDF/B-VII.1 database): 944.2 keV (emission probability 0.4390), 962.1 keV (0.2027), 79.5 keV (0.1163), 181.9 keV (0.0991) and 780.2 keV (0.0956). The half-life of Tb-158 is 180 y; therefore, it is important for decommissioning.

The measurement of low active Tb-158 in the presence of the other activation and fission products such as Co-60, Eu-152, Eu-154, Eu-155, and Ho-166m poses a serious difficulty for the non-destructive analysis of the waste from the RBMK-1500 reactors using a gamma-ray spectrometry due to an influence of Compton scattering and interferences at the full-energy peaks.

In this work, the activity of Tb-158 in the mixture of gamma-ray emitters was measured in the waste samples from the Ignalina NPP Unit I RBMK-1500 reactor using a gamma-ray spectrometry. The coincidence-summing effect was taken into account. Detectors were efficiency calibrated using reference materials traceable to NMI. The GammaVision software (v. 6.06) was applied for spectra acquisition and analysis. The combined standard uncertainty of Tb-158 activity did not exceed 14% (k = 2) in analyzed samples.

For the calibration of particular Tb-158 activity and determination of uncertainty, modelling of activity of the gamma-ray emitters was done. The simplified 4x4 core plateau fragment (SCALE 6.1) was used for description of the neutron fluence in different parts of the RBMK-1500 reactor. Reactor core with uniformly distributed fuel of average burnup of 12 MWd/kg and realistic distribution of rods of control protection system in the active core was modeled using standard materials composition. The parameters, which mostly influence the distribution and activities of Tb-158 during nuclear plant operation, were investigated.

The results of calculation of the massic activity of gamma-ray emitters were used for interpretation of the total gamma-ray spectrum and determination of Tb-158 massic activity uncertainty in the waste of RBMK-1500 reactor.

Corresponding author's email address: **arturas.plukis@ftmc.lt**

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SUMCOR: CASCADE SUMMING CORRECTION FOR VOLUMETRIC SOURCES APPLYING MCNP6

114

Dias, M.; Semmler, R.; Moreira, D.; Koskinas M.

IPEN-CNEN/SP (BR)

There is a continuing effort inside the ICRM community to improve the codes for calculating the cascade summing corrections, for samples of different geometries and decay scheme characteristics. Reports on comparisons were presented at the 2011, 2013 and 2015 ICRM meetings, showing the evolution of the codes [1-3].
Within this context, the LMN (Nuclear Metrology Laboratory) in São Paulo has developed a code, called SUMCOR, for calculating the cascade summing correction, which incorporates several desirable features as follows:

- Uses MCNP6 to model volumetric sources, running in multi-core processor;
- Points are randomly chosen inside the source and tracked individually to the HPGe detector;
- All cascades are followed according to the decay scheme, for each individual points chosen inside the source, according to equations given by ref. [4];
- Total and peak efficiencies are calculated for all energies included in the cascades;
- Decay scheme data are taken from NUCLEIDE;
- Metastable transitions are considered, according to equations given by ref. [5];
- X-rays from electron capture and internal conversion processes are considered;
- LS Ratio method can be compared to exact calculation, following the equations given by ref. [4];
- Comparison has been performed with recent exercises organized by the Gamma-Ray Spectrometry Working Group and reported in references [2] and [3].

Following the procedure described in reference [3], the cascade summing corrections were calculated for a point source of ^{133}Ba and the results agreed with the average values shown in Table 3 from reference [3], within the standard deviation for each gamma-ray energy.
In the case of volumetric sources, the procedure followed reference [2] for ^{60}Co uniformly distributed in water, soil and filter. The results showed average deviations of: 3.2, 0.6 and 0.2 %, respectively when compared to Table 5 from reference [2].

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Corresponding author's email address: **msdias@ipen.br**

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COINCIDENCE SUMMING CORRECTIONS FOR VOLUME ^{60}CO SOURCES MEASURED BY SCINTILLATION NAI(TL) SPECTROMETERS

Kandić, A.; Šešlak, B.; Vukanac, I.; Djurašević, M.; Jevremović A.

INS-UB (RS)

The gamma spectrometry often deals with the problem of coincidence summing effects and therefore with determining the corresponding correction factors. This paper presents the method for obtaining the coincidence summing correction factors for ^{60}Co measured by scintillation NaI(Tl) spectrometer in two different experimental settings.
In the first part of experiment, a well-type scintillation NaI(Tl) spectrometer - Canberra 802 - 4W was used, and correction factors for test tube measurement geometry with different volume of standardized solution were determined. In the second part, the scintillation NaI (Tl) spectrometer - Canberra 802 was used, and correction factors for different measurement geometries of volume sources were determined.
In both parts of the experiment, the systems of equations were settled up, and as input data net area in peaks at energies 1 173 keV and 1 332 keV were used, as well as the net area in the entire spectrum (upto approximately 2.700 keV), where in the data were corrected on the background, a dead time, and the spectra were extrapolated to 0 keV. The systems were solved in the program Mathematica 5.
For well-type scintillation NaI(Tl) spectrometer - Canberra 802 - 4W, determination of correction factors was quite simple, because all measurements of 60Co solution were performed in precisely defined measurement geometries.
On the other hand, for scintillation NaI(Tl) spectrometer Canberra 802 - 4, there were some difficulties. Namely, it was necessary to determine the correction factors for different measurement geometries. In order to avoid making a large number of standards with different geometries and matrices, numerous measurements of ^{60}Co point source, in different positions in relation to the detector itself, were carried out. Analysis of obtained values indicates the spatial dependence of the correction factors: exponential axial dependence and linear radial dependence. Further analysis showed that a correction factor for a particular geometry of the sample can be determined as the arithmetic mean of correction factors for the four characteristic positions, with an estimated uncertainty due to this approximation less than 1 %. It should be mentioned that, based on previous experience, the estimated uncertainty due to different chemical composition of the used matrices is also less than 1%.

Corresponding author's email address:

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ACTIVITY DETERMINATION OF ^{60}CO AND THE IMPORTANCE OF ITS BETA SPECTRUM

006

Kossert, K.[1]; Marganiec-Gałązka, J.[1]; Mougeot, X. [2]; Nähle, O.[1]

[1]PTB (DE); [2]LNHB (FR)

Cobalt-60 was measured by means of 4πβ(LS)-γ coincidence counting with a liquid scintillation counter in the beta channel. Currently, this technique is being developed as new activity standardization method at PTB. Validation measurements were carried with a revived 4πβ-γ coincidence counting system with a proportional counter in the β channel.
Additional measurements were carried out in liquid scintillation counters with two and three photomultiplier tubes, respectively. The counting efficiencies for these systems were determined using CIEMAT/NIST efficiency tracing and the triple-to-double coincidence ratio (TDCR) method, respectively. Both methods require computed beta spectra and other information about the nuclear decay as input data.
The results of both coincidence counting techniques were in very good agreement and yield a robust reference value. In contrast to coincidence counting, the TDCR method and CIEMAT/NIST efficiency tracing yield results which depend to a certain extent on the above-mentioned input data. The initial activity concentration determined with these techniques was found to be systematically lower than the results from coincidence counting. In addition, the results from TDCR and CIEMAT/NIST show some inconsistency.
The discrepancies were resolved by applying new beta spectrum calculations for the allowed beta transitions of ^{60}Co. Usage of calculations, which take screening effects as well the atomic exchange effect into account, leads to good agreement between all four methods. Results of the LS methods are consistent when applying efficiency variation over a wide efficiency range. In this way, the combination of techniques allows an important validation of calculated and measured beta spectra. Improvements for beta spectra measurements as well as for corresponding calculation techniques are main tasks in the ongoing EMPIR project MetroBeta.

The uncertainties as well as pros and cons of the methods are discussed in detail.

Corresponding author's email address: **karsten.kossert@ptb.de**

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CONCEPT DESIGN OF A TIME-OF-FLIGHT SPECTROMETER FOR THE MEASUREMENT OF THE ENERGY OF ALPHA PARTICLES

013

García-Toraño, E.

CIEMAT (ES)

The knowledge of the energies of the alpha particles emitted in the radioactive decay of a nuclide is a key factor in the construction of its decay scheme. Following an intensive work of measurement and normalization of existing data carried out at BIPM by A. Rytz in 1973, a catalog containing the energies of alpha particles for many radioactive nuclides was made available. The nuclear data evaluation community regularly provides reliable data references. Virtually all existing data are based on measurements made by magnetic spectrometry. Absolute measurements cannot be done with Si detectors because of the pulse height defect, and cryogenic detectors are still been tested for this and other purposes.
An alternative solution would be the use of time-of-flight detectors. This paper discusses the main aspects to be considered in the design of such detectors, including:

- Basic numerical data for the energies considered (~ 4 to 8 MeV).
- Timing (Start and Stop) detector components: foil structure and homogeneity, electrostatic detectors, multichannel plates, fast scintillators and others.
- Effect of solid angle variation, flight distance, radioactive source, stability and other issues in the detector performance.
- Electronic setup.

Based on data presented, it can be estimated that an energy resolution in the order of 1.5 to 2.5 keV would be attainable.
Reference will be made to the first and only prototype developed for this purpose by Frolov at VNIIM in 1972.

Corresponding author's email address: **e.garciatorano@ciemat.es**

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A NEW CODE FOR IMPROVED CALCULATIONS OF ELECTRON CAPTURE TRANSITIONS

051

Mougeot, X.

CEA, LIST (LNE-LNHB) (FR)

Electron capture transition properties are crucial features when evaluating the decay schemes of radionuclides. In modern evaluations from the Decay Data Evaluation Project (DDEP), the LogFT and EC-capture programs are commonly used for determining branching ratios and capture probabilities. However, these codes are limited by their models, using incomplete data tables and analytical estimates, and can only treat allowed, first- and second- forbidden unique transitions. These limitations lead to a lack of accuracy and this work aims at improving this situation.

A new code has been developed for improving the calculations, based on Behrens and Bühring formalism [1]. In previous work, the study of the exchange effect in beta decays led to a precise determination of atomic relativistic electron wave functions [2]. These wave functions are used for calculating the atomic effects, which are of high importance in the context of electron capture transitions. The change in the nuclear charge induced by the transition leads to an imperfect atomic overlap. The indistinguishability of electrons allows an exchange effect. These two effects are usually combined into a single overlap and exchange correction. Two approaches were considered and unified in the same formalism: Bahcall's approach [3] which only considers the first three s orbitals and uses a closure relation, allowing an approximate inclusion of shake-up and shake-off; and Vatai's approach [4] which considers the contribution of each orbital. This latter approach, though being more comprehensive, does not account for shake-up and shake-off which create secondary vacancies, thus Crasemann's formalism [5] has been included to estimate the probability of secondary electrons for each possible captured electron. Finally, as the capture process produces the daughter atom in an excited state, the influence of the hole on the bound wave functions, called the inner hole effect, was estimated by first order perturbation theory.

Allowed and forbidden unique transitions are calculated explicitly and no limitation exists on the transition order, while forbidden non-unique transitions are determined using the ξ approximation. Ratios of capture probabilities are determined for each orbital, as well as capture probabilities assuming PK + PL + PM + ... = 1. Electron capture to beta plus ratios are also determined by means of previously improved beta calculations. The calculation of log ft values and the propagation of uncertainties from the input parameters are also included.

Preliminary results have already been shown during the last ICRM conference for ^{138}La decay [6], highlighting the interest for these improved calculations. The present contribution will describe this new code and its validation against existing nuclear data.

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Corresponding author's email address: **xavier.mougeot@cea.fr**

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RELATIVE CONVERSION ELECTRON EMISSION PROBABILITIES OF PU ISOTOPES DERIVED FROM ICE SPECTROMETRY

009

Pommé, S.[1]; Pöllänen, R.[2]; Paepen, J.[1]

[1]IRMM (EU); [2]STUK (FI)

In previous work, novel conversion electron spectrometry set-ups at STUK and JRC have been described. They make use of a windowless Peltier-cooled silicon drift detector (SDD), which is well suited for measuring low-energy x-rays and electrons. Internal conversion electron (ICE) spectra have been taken of thin plutonium sources, respectively enriched in the isotopes Pu-238, 239, and 240. The spectra show major ICE peaks between 20 and 50 keV, with an energy resolution of 0.47 keV. The shape of the conversion electron peaks is comparable to alpha peaks, with typical tailing due to energy loss in the absorbing materials. The x-ray peaks are comparably sharp and their position is invariable to source thickness.
In this work, the relative emission probabilities for the L and M+ ICE peaks for the 43.469 keV (Pu-238), 51.624 keV (Pu-239) and 45.244keV (Pu-240) gamma transitions are investigated. To this end, independent deconvolution software tools have been developed at JRC and STUK to fit simultaneously the x-ray and conversion electron peaks obtained with the SDD. The ICE peak areas are determined and relative intensities are derived.
The software is also tested on mixed plutonium spectra to assess the potential accuracy of the spectral deconvolution as a tool for determining the isotopic composition. This could be used as a complementary technique to alpha spectrometry for nuclear safety applications. Resolving the alpha peaks of Pu-239 and Pu-240 is particularly challenging due to the limited energy resolution of alpha spectrometry. Since the concentration of Pu-239 is key to evaluating the fissility of a Pu mixture, it should be advantageous to use ICE spectrometry in addition. The characteristic conversion electron peaks of the Pu isotopes are well separated in energy and integration of the corresponding peak areas should yield a good indication of the isotopic ratio.

Corresponding author's email address:

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THE CASE OF LONG-RANGE ALPHA PARTICLES FROM ^{212}PO REVISITED

096

Sánchez, A. M.

UEx (ES)

Among the radiations from ^{212}Bi sources, Rutherford and Woods noted in 1916 (Evans, 1985) a few particles whose range is about 11.6 cm in air, or markedly in excess of the 8.6 cm range of the well-known 8.78437-MeV alpha-particle emissions (Browne and Firestone, 1986) from ^{212}Po to ^{208}Pb. It is now well stablished that these long-range alpha particles are emitted from excited levels of ^{212}Po. In this nucleus the lifetime for alpha decay is comparable with the life time for gamma decay. Consequently a small fraction of the excited ^{212}Po nuclei will undergo alpha decay directly from an excited level. The relative intensities and energies of the known long-range alpha-particle groups of ^{212}Po were summarized by Rytz (1953). Scarce revisions of these data have been found in the bibliography (Emery and Kane, 1960). But even in the most recent compilations (Martin, 2007) no new values have been found.

New measurements have been made of these long-range alpha particles in our laboratory, and then a check of the energy emissions and their intensities. The measurements were performed by alpha-particle spectrometry with a low-resolution silicon detector inside a vacuum chamber. The source used was a ^{232}U (in equilibrium with its daughters) collimated disc, 20 mm^2 active area, made by electrodeposition, and provided by CIEMAT. A PIPS detector from Camberra, with 50 mm^2 active area, was used, and 65 mm for the source-to-detector distance.

Due to the short life of the ^{212}Po levels, coincidence summing is produced between the emitted alpha particles and the beta particles coming from the ^{212}Bi ground level, making difficult the analysis of the region above 8.78437 MeV, where the long-range particles are expected (Martín Sánchez et al., 1990). To avoid these interferences, measurements including a magnet between source and detector were also performed. Results show that the values reached for energies and intensities can be checked with the existing tabulated values.

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Corresponding author's email address: **ams@unex.es**

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ALPHA-PARTICLE SPECTROMETRY: THE IMPORTANCE OF MEASUREMENT SETUP CONDITIONS AND SAMPLE PREPARATION IN NUCLEAR DECAY DATA MEASUREMENTS

134

Dirican, A.

TAEK-SANAEM (TR)

Alpha-particle counting at defined solid angle (DSA) spectrometers are used for the determination of alpha emission probabilities, relative half-lives, branching factors and energies. This technique is used in the case of primary standardization and mostly performed by ion-implanted silicon (PIPS) detectors. Most important features of this technique are geometrically calculated efficiency, low background and energy spectra of each alpha decaying nuclides. Counting by a systems with defined solid angle is one of the oldest and most accurate primary standardization methods for the measurement of the disintegration rate of alpha emitting radionuclides. For β- rays the method is inaccurate. But, for alpha-particles (1.83 – 11.65 MeV), accuracies better than ± 0.1%. This counting system has been applied in many emission probability studies, standardization exercises and half-life determinations of long-lived radionuclides. In this paper required measurement setup conditions, sample preparation (drop deposition), source density distribution by using Cyclone Storage Phosphor Imaging system, measurements of geometrical parameters of DSA, solid angle calculation procedure and spectrum deconvolution of alpha-particle counting were explained for DSA-high resolution alpha particle spectrometry at TAEK-SANAEM. Preliminary works prior to measurement were explained. The acceptance test of our first DSA instrument was carried out with a source of ^{241}Am. This source was prepared by aliquot of a mother standard reference material solution which was purchased from NIST. These information can be useful for dilution alpha spectrometric measurements. Source was prepared by dropping solution on to a flat watch glass (thickness: 1mm, diameters: 30mm, 15mm). After determination of source density distribution and thickness, it was measured with new DSA instrument at TAEK-SANAEM. Measurement uncertainty components were also presented.

Corresponding author's email address: **abdullah.dirican@taek.gov.tr**

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ACTIVITY DETERMINATION OF ^{68}GE/^{68}GA BY MEANS OF 4π(Č)β-γ COINCIDENCE COUNTING

007

Marganiec-Gałązka, J.; Kossert, K.; Nähle, O.

PTB (DE)

An important imaging technique used in nuclear medicine is the positron emission tomography (PET) which allows observing metabolic processes in the body. It plays a fundamental role in oncology, neuroimaging, cardiology and other medical disciplines. The radionuclides used for PET scanning are typically short-lived isotopes with half-lives between a few minutes up to several hours. Short half-lives are mandatory to minimize the patient's radiation exposure and, hence production facilities like cyclotrons must be close to the PET imaging system.
Germanium-68 is an electron-capture nuclide which emits rather low-energetic X-rays and Auger electrons. It disintegrates to ^{68}Ga which decays predominantly (88.88%) by positron emission. The half-life of ^{68}Ga is 67.83 min and, consequently, this isotope can be used for PET imaging. Gallium-68 can be extracted chemically from sources of the longer-lived ^{68}Ge ($T_{1/2}$ = 270.95 d). In this way, ^{68}Ge/^{68}Ga generators allow usage of PET imaging even if no cyclotron is available. Since ^{68}Ge is longer lived and not seen by conventional PET scanners, ^{68}Ge/^{68}Ga sources are often used a surrogates for ^{68}Ga or even other PET isotopes when calibrating ionization chambers or PET scanners using phantoms. Thus, there is a great demand on accurate activity measurements of ^{68}Ge/^{68}Ga sources.
In this work, the activity concentration of a ^{68}Ge/^{68}Ga solution was measured using a new 4πβ-γ coincidence counting system which comprises a TDCR counter with an automated sample changer and an NaI scintillation detector which is located below the polyethylene vial in the optical chamber. The vials were filled with an aqueous solution and, hence, the β-channel corresponds to a Čerenkov detector. High-energetic positrons from the β^+-decay of ^{68}Ga may create Čerenkov light when moving in a dielectric and transparent medium, whereas the low-energetic electrons and photons which are ejected as a consequence of ^{68}Ge electron capture are discriminated in a natural way. The requisite efficiency variation was carried out with the aid of grey filters (natural density filters: from 0.15ND to 1.2ND) which were placed around and underneath the vials. The data were evaluated for double (D) and triple (T) coincidences as the β-channel, and second order polynomial functions were fitted to the data. The function was finally extrapolated to a β-efficiency of 100%.
The application of this method requires knowledge about the overall beta plus emission intensity which yields a major uncertainty component of about 0.47% to the overall relative standard uncertainty which was found to be 0.82%.
The measurements were carried out within the scope of the international CCRI(II)-K2.Ge-68 comparison.

Corresponding author's email address: **justyna.marganiec@ptb.de**

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ABSOLUTE STANDARDIZATIONS OF ^{99m}Tc AND ^{57}Co BY 4πBY LIQUID SCINTILLATION COUNTING FOR SIRTI AND SIR COMPARISONS

017

Van Rooy, M.W.; M.J. Van Staden, M.J.; Simpson, B.R.S.; Lubbe, J.

NMISA (ZA)

During 2015, the National Metrology Institute of South Africa (NMISA) performed absolute standardizations of ^{99m}Tc and 57Co solutions via the 4π(LS)β-γ coincidence counting technique for inter-comparison purposes. Both radionuclides are used in the nuclear medicine industry; ^{99m}Tc for diagnostics and the longer-lived ^{57}Co as a surrogate for ^{99m}Tc for γ-camera calibrations, due to its similar γ-ray energy and intensity. The literature indicates that these two radionuclides are particularly difficult to standardize accurately.

We provide the first detailed report on the absolute standardization of ^{99m}Tc by liquid scintillation counting (LSC). The absolute standardization of a ^{99m}Tc stock solution formed part of the BIPM.RI(II)-K4.Tc-99m International Reference System Transfer Instrument (SIRTI) comparison which took place a few months later. This was the first participation by NMISA in the SIRTI comparisons. The standardized solution was used to determine a radionuclide calibration factor for ^{99m}Tc in the standard geometry of the NMISA secondary standard ionization chamber (IC). Consequently, NMISA IC measurements were used to report ^{99m}Tc activity values during the SIRTI comparison. The absolute standardization of ^{99m}Tc by LSC resulted in a relatively high standard uncertainty of 1.5 %. Due to the low detection efficiency of 1.73 – 2.13 keV conversion electrons in the β-channel (maximum efficiency of 16 %), the biggest contribution to the total uncertainty stems from the estimation of the rate vs. efficiency functional form over the large extrapolation range, contributing by 1.3 %. We undertook a simulation based on a detection efficiency analysis in an attempt to predict the functional form and the effect on the corresponding extrapolated activity value. The simulation involved two simple methods to estimate the detection efficiency of the 140.51 keV γ-ray interacting with the liquid scintillator, using its theoretically generated Compton spectrum. Results from the simulation showed that the deviation from linearity observed with the experimental results was due to the interacting γ-ray with the liquid scintillator. This makes the standardization of ^{99m}Tc more problematic by LSC than proportional counting, where a linear fit is normally obtained. The NMISA SIRTI equivalent activity is 1.9 % higher than the Key Comparison Reference Value (KCRV) and in agreement within two standard deviations.

The absolute standardization of a ^{57}Co stock solution, performed earlier in 2015, formed part of the BIPM.RI(II)-K1.Co-57 comparison. Consequently, an ampoule containing a solution of ^{57}Co was sent to the Bureau International des Poids et Mesures (BIPM) for measurement in the SIR. This submission supercedes the 1985 participation by NMISA for ^{57}Co. To aid the standardization, a detection efficiency analysis was undertaken to get an indication of the rate vs. efficiency function to be expected. A summary is presented. Experimentally, source data from the standardization were analyzed with a γ-window around the 122 and 136 keV photopeaks in a NaI detector, encompassing the ~100 keV iodine escape peak. The functional form of the extrapolation curve and corresponding extrapolated activity values were investigated by varying the β-efficiency range and distribution electronically from ~30 % to a maximum of ~86 %. Slight curvature was observed. The main contribution to the total standard uncertainty of 0.22 % came from the uncertainty in the fitting procedure which was 0.2 %. Results from a second γ-window, similar to the first but with a lower threshold set above the iodine escape peak also showed curvature, with a ~0.4 % difference between extrapolated average activity values from second and first order polynomials. Results from five counting sources showed a 1 % spread in extrapolated activity values. Results from the second γ-window showed no improvement and were therefore not used for the final activity calculation. The NMISA SIR equivalent activity is 1.1 % higher than the KCRV and in agreement within four standard deviations. This result indicates that undertaking an absolute standardization of ^{57}Co with LSC in the 4π channel is comparable to the proportional counting technique.

Corresponding author's email address: **mvrooy@nmisa.org**

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STANDARDIZATION OF $^{68}Ge/^{68}Ga$ USING THE 4πβ-γ COINCIDENCE METHOD BASED ON CHERENKOV COUNTING

052

Bobin, C.; Thiam, C.; Bouchard, J.

CEA, (FR)

For several years, Cherenkov counting has been investigated in radionuclide metrology as an alternative to liquid scintillation (LS) for activity measurements based on the Triple to Double Coincidence Ratio (TDCR) method. For that purpose, new modellings of light emission were developed in order to take into account the physical properties characterizing Cherenkov effect. The detection efficiency is directly related to the Cherenkov threshold which depends on the refractive index of the transparent medium (~ 260 keV in aqueous solutions). Cherenkov light emission is anisotropic: it is emitted from the surface of a cone at an angle with respect to the trajectory of charged particles. This anisotropy is at the origin of the difference observed on TDCR counting between polyethylene (diffusive wall) and glass vials.

As no LS cocktail is required, the interest of Cherenkov counting is the possibility to carry out measurements directly in aqueous solutions. It is also possible to take advantage of the Cherenkov threshold for discrimination purpose: for instance, the standardization of ^{32}P ($E_\beta max$ ~ 1710 keV) in aqueous solutions is not affected by the usual ^{35}S impurity (E_β max ~ 167 keV). This property was recently used in the standardization of $^{68}Ge/^{68}Ga$ based on the 4π β-γ coincidence method in the framework of the international comparison conducted by NIST. In that case, only interactions given by β+ events resulting from 68Ga (E_β max ~ 1899 keV) are counted in the β-channel because electron-capture events from ^{68}Ge have lower energies (maximal values of about 10 keV) than the Cherenkov threshold in aqueous solutions. The 4π(CH)β-γ coincidence method is implemented using the emission of 511-keV photons resulting from β+ annihilation without the need of a model of light emission which is a prerequisite of TDCR-Cherenkov counting.

For this first participation to an international comparison using 4π(CH)β-γ coincidence measurements, the three-photomultipliers apparatus (designed for TDCR counting) was set in the β-channel and coupled to a NaI(Tl) scintillation detector in the γ-channel. Cherenkov measurements were carried out using polyethylene and glass vials yielding maximum detection efficiencies of about 70 %. The extrapolation technique was carried out by defocusing the photomultipliers for the variation of the detection efficiency in the β-channel. Compared to a classical proportional counter, coincidence counting is not only sensitive to the β-detector efficiency but it is also significantly impacted by the Cherenkov threshold. Thus 4π(LS)β-γ coincidence measurements (based on liquid scintillation) were also carried out to verify potential biases due to the extrapolation technique implemented in the case of 4π(CH)β-γ counting. Comparisons with simulations based on the Monte Carlo code Geant4 are also reported. The modelling was designed to take into account the emission of Cherenkov photons in each transparent component of the β-detector (aqueous solution, glass vial and photomultiplier window) and by considering their respective optical properties.

Corresponding author's email address: **christophe.bobin@cea.fr**

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COMPARISON OF CALCULATION OF THE ACTIVITY OF A H-3 SOURCE MEASURED BY THE LSC-TDCR METHOD

053

Cassette, P.[1] and the 17 participants to the comparison[2]

[1]CEA, LIST, LNE-LNHB, (FR); [2] CNEA, ANSTO, IRD, NRC, NIM, CENTIS, IRMM, PTB, ENEA, KRISS, FTMC, POLATOM, IFIN, VNIIM, METAS, NPL, NIST

The aim of this comparison exercise, recorded as CCRI(II) S-12 comparison, was to compare the calculation methods used at the National Metrology Institutes to determine the value and the standard uncertainty of the activity of a $^{3}H_{2}O$ liquid scintillation source (LSC) using the same experimental data set collected at the LNE-LNHB with a triple to double coincidence ratio (TDCR) counter and to analyze the reasons for the observed differences. The measurement results were sent to the participants as raw data acquisition files and summarized in a spreadsheet. The detection efficiency has been varied using two different methods: by adding coaxial grey filters around the source and by defocusing the photomultiplier tubes (PMT). All participants reported their results in terms of activity of the measured source with a detailed uncertainty budget and also provided details on the models and methods used to calculate the detection efficiency and the associated uncertainty.

The coordinator received contributions from 17 laboratories. The results, analyzed according to the CCRI(II) procedure were quite consistent, and no outlier was identified. Various indicators were used to analyze the data (arithmetic mean, weighed mean, median and power-moderated mean) and all these indicators gave the same reference value, within their associated uncertainties. The relative standard deviation of the reported results is 0.17 %, to be compared with a value of 0.4 % observed, after rejection of an outlier, in the CCRI(II) tritiated water activity comparison organized by BIPM in 2009.

The causes of observed dispersions are discussed in the paper, like the ionization quenching factor, the electron stopping power, the asymmetry of the photomultiplier tubes and the software package used. No influence of the calculation of the beta spectrum was identified. The relative standard uncertainties reported by the participants varies from 0.4 % to 2 %, and the causes of this dispersion are discussed in the paper.

As expected, even if all laboratories analyzed the same data set, some variability of the results was observed and thus, an output of this exercise is also an estimation of the dispersion of the measurement results to be expected in LSC, even when the measurement conditions are well defined

Corresponding author's email address: **philippe.cassette@cea.fr**

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IFIN-HH PROGRESS IN TRITIUM STANDARDIZATION BY THE LSC-TDCR METHOD, PROVED IN INTERNATIONAL COMPARISONS

023

Antohe, A.[1]; Sahagia, M.[1]; Luca, A.[1]; Ioan, M. [1]; Cassette, P.[2]

[1] IFIN-HH (RO), [2] LNE-LNHB (FR)

This paper presents the progress obtained in the standardization of ^{3}H, by using the LSC-TDCR method and the equipment existing in both IFIN-HH and LNE-LNHB. The evolution in preparation and measurement of samples, processing and analysis of experimental data, finalized by reporting the results: activity or activity concentration of a solution and its combined standard uncertainty is described. The method and basic results of three ^{3}H international comparisons, involving our laboratories, are presented, such as follows. *(i).* CCRI(II)-K2.H-3 2009, key comparison, pilot laboratory LNE-LNHB, preparing and distributing the solution (Ivan et al. ARI 68, 1543-1545, 2010; G. Ratel, BIPM report, 2015); *(ii).* CCRI(II).PS.ESIR.2014 (BIPM, 2015), pilot study, solution prepared and sent to BIPM by IFIN-HH and *(iii)* the supplementary comparison CCRI(II)-S12.H-3, pilot laboratory LNE-LNHB, who prepared, measured a LS vial with 3H and sent the measurement data to the participants. Some improvements are underlined.

- Use of the software's, both of them written by P. Cassette, the TDCR06b in cases (*i* and *ii)* and the improved TDCR07c, taking into account the assimetry of the PMs, in case (*iii*).

- Careful reading and application of recommendations from the two essential papers on the LSC method [Broda et al. Metrologia 44(2007) S36-S52 and Kosssert et al. Metrologia 52(2015) S172-S190] in the comparison (*iii*).

-Improvement of the sample preparation and measurement. In the (*i*) case, the type A uncertainty was 0.63%, in the case (*ii*) it was 0.37%; in both cases only grey filters were used for efficiency variation. In the case (*iii*), it arrived at 0.48%, resulting from the calculation of mean activity from grey filters and defocusing measurements.

-Evaluation of the significant influence of the, *kB* parameter over the calculated double and triple coincidence efficiencies, chosen as *kB*=0.011 cm/MeV in (*i* and *ii*) and *kB*=0.010 cm/MeV in (*iii*). The analysis of this influence was consistent, resulting in all cases as a type B uncertainty of about 0.7% for a *kB* value variation of 0.001 cm/MeV, comparable with that reported in (Cassette et al. ARI 109,41-43, 2016), 0.64%. In the (*iii*) case, the systematic study of the curve Activity vs. TDCR slope was performed.

-Analysis of the statistical model influence, by comparing Poisson and Polya statistics L=0.05, arriving at 0.34% type B uncertainty.

-The reported combined standard uncertainties were respectively: u_c = 0.88% in (i), with the difference *D* from KCRV of 1.8%; In the cases (*ii*) and (*iii*) they are u_c =0.79 % and respectively a lower value, under analysis at the comparisons' organizers.

Corresponding author's email address: **antohe@nipne.ro**

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STANDARDIZATION OF PRASEODYMIUM-142 ACTIVITY CONCENTRATION USING CIEMAT/NIST TRITIUM EFFICIENCY TRACING METHOD

036

Yücel, H.[1], Yeltepe, E.[2]

[1]AU-NBE (TR), [2] TAEK -SANAEM (TR)

In medicine, pure high energy beta emitting radionuclides or beta emitters with low energy, low intensity gamma rays are particularly preferred in brachytherapy techniques to limit the dose transferred to the affected region. Among them, praseodymium-142 (^{142}Pr) can also be potentially used as high dose rate brachytherapy sources due to its suitable decay properties such as high beta energy, $E_{\beta max}$=834.2keV(96.3%) and its half-life(19.12h). But this radionuclide has also an unwanted high gamma energy emission but with low intensity,i.e., E γ =1576keV(3.7%). On the other hand, literature survey shows that the standardization of ^{142}Pr radionuclide activity is still scarce, and hence the radioactivity standardization methods to be used for ^{142}Pr will contribute to the radionuclide metrology field in an essential way.

In this study, several mg of high-purity Pr2O3 in a flame sealed-ampoule was irradiated in a high thermal neutron flux(at about 10^{12} n•cm^{-2}•s^{-1}) of Istanbul Technical University 250 kW TRIGA Mark II Research Reactor for 5 minutes. A set of irradiated oxide samples was then dissolved by adding 2 mL HNO_3 and 1 mL HCl acids with the help of ultrasonic bath. The activity of 142Pr produced via ^{141}Pr(n,γ)^{142}Pr reaction in the samples was diluted two times to reduce at an activity level of about 100 Bq in each 20mL low K-glass vial and then total 18 vials were measured for 30 min periods by using LSC technique. Measurements were performed in turn two Quantulus 1220 devices(Perkin Elmer) installed in AU-NBE and TAEK-SANAEM for the comparison aims. Before the standardization, ^{3}H efficiency tracing method was employed to obtain the interested radionuclide efficiency. To this end, tritium tablets having a known activity was mixed with a 15 mL of Ultima Gold® plus 1mL de-ionized water, and by adding 0 µl to 170 µL nitrometan(NM), thus providing a polynomial ^{3}H-quenching curve plotted to SQP(E) parameter.

CN2005 code was used to calculate the radionuclide efficiency against ^{3}H efficiency with a quenching parameter kB of 0.0075 cm•MeV^{-1}. As of Dec.15,2015,15:30pm the activities of ^{142}Pr were measured to be 24.04 ± 0.44 MBq/g, 24.08 ± 0.29 MBq/g, 24.12 ± 0.44 MBq/g and 22.62 ± 1.63 MBq/g in the samples. When the standardization of the solutions were performed by CIEMAT-NIST(CN-LSC) method, a combined relative standard uncertainty is 1.25%(±1σ). Then a gamma ray spectrometry (HPGe-GS) with use of a well-type, 44.8% relative efficient HPGe detector(Canberra GCW4023 model) was used to measure the count rates of 1575.6 keV photopeak (3.7%) of 142Pr for which the efficiency value was estimated from Monte Carlo Programs (GESPECOR and PENELOPE). For the case of the results from HPGe-GS method, a combined relative standard uncertainty of 7.8%(±1σ) is five times higher than the uncertainty of ^{142}Pr standardization using CN-LSC method. Thus CN-LSC can referred to be the better one for ^{142}Pr standardization. The detail of the methods and the uncertainty budgets will be discussed.

Corresponding author's email address: **haluk.yucel@ankara.edu.tr**

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DIFFUSION LENGHTS AND PARTITION COEFFICIENTS OF ^{131M}XE AND ^{85}KR IN MAKROFOL N AND MAKROFOL DE

050

Mitev, K.[1]; Cassette, P. [2]; Tartès, I.[2]; Georgiev, S.[1]; Dimitrova, I.[1]; Pressyanov, D.[1]

[1]Sofia University, (BG); [2]LNE-LNHB, (FR)

A large number of isotopes of the radioactive noble gases (RNG) Kr and Xe are produced by nuclear fission and thus are good tracers of civil or military nuclear activities. Some isotopes like ^{85}Kr and ^{133}Xe are released to the atmosphere during the normal operation of nuclear reactors and these are considered as key nuclides for reporting emissions from nuclear facilities. Large amount of RNGs are released to the atmosphere in the case of nuclear accident or during a nuclear weapon test. Therefore, adequate metrological assurance and RNG measurement techniques are important during the normal operation of nuclear facilities and in the case of accidents.

In the last decade it was found that some polycarbonate materials like Makrofol DE and Makrolon have high absorption ability to noble gases. We have proposed methods for measurement of the concentrations of Kr and Xe in air or water based on absorption in polycarbonates in a previous publication. More recently, another polycarbonate material named Makrofol N was found to have even higher absorption ability to RNGs. Currently, there is a need of precise characterization of the absorption ability of these materials and of studies on the application of the materials in RNG measurements and/or metrological assurance.

The objective of this work is to try to fill this gap by: 1. Development of experimental set-ups for exposure of polycarbonates to precisely known RNG concentrations; 2. Application of precise techniques like liquid scintillation counting (LSC) and triple to double coincidence ratio method (TDCR) in LSC for measurement of the activity absorbed in the polycarbonates; 3. Application of a previously developed analytical model for estimation of the diffusion length (L_d) and the partition coefficient (K) of RNGs in the polycarbonate from the activity measurement and the exposure data.

To achieve this goal, we exposed Makrofol N foils to referent ^{85}Kr and ^{131m}Xe activity concentrations in air and water. The RNG activity was then determined using the TDCR method in LSC. We have shown previously that the activity of ^{222}Rn in polycarbonate foils can be measured absolutely by the TDCR method using toluene-based scintillator which dissolves the Makrofol N foils. We demonstrate here that the same approach is applicable to ^{85}Kr and ^{131}mXe measurements, which means that the absorbed activity of these isotopes in the polycarbonate can be measured with relative standard uncertainty better than 0.5%. We have performed exposure experiments and studies of the desorption of ^{85}Kr and ^{131m}Xe from Makrofol N and DE foils and we have determined: L_d, K from air and K from water for ^{85}Kr in Makrofol N and Makrofol DE; L_d and K from air for ^{131m}Xe in Makrofol N and Makrofol DE.

To the best of our knowledge, the experimental approach proposed in this work allows the most accurate determination of ^{85}Kr and ^{131m}Xe absorption properties of polycarbonates. The diffusion length (L_d) and the partition coefficient (K) of ^{85}Kr and ^{131m}Xe in Makrofol N polycarbonate were previously unknown and are determined for the first time in this work. This allows the characterization and quantification of the remarkable RNG absorption properties of this material.

Corresponding author's email address: **kmitev@phys.uni-sofia.bg**

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A NEW 4 π (LS)-GAMMA COINCIDENCE SYSTEM USING THE TDCR SYSTEM IN THE BETA CHANNEL AT CMI

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Sochorová, J.; Auerbach, P.

CMI(CZ)

A new 4π(LS)-γ coincidence system (TDCRG) was built at the CMI as a universal device enabling measurement using the TDCR method and 4π(LS)-γ coincidence measurement. The beta counting channel – the TDCR system - consists of three Hamamatsu R331-05 photomultipliers placed in a steel case with a custom made rapid voltage divider and preamplifier and affixed to specially made optical chamber. Insertion of sources (vial Perkin Elmer 20ml) from above is performed using a stepper motor. The thin cylindrical filter can be controllably pushed onto the vial in order to change the efficiency of detection in beta channel. In gamma counting channel, either of the two NaI(Tl) detectors (NaI(Tl) ET Enterprises 9266 KB, 2'x1' or Canberra 802, 3'x3') can be used, depending on the decay scheme of measured radionuclide. The signal delivered by counting channels is processed in the FPGA multifunction card NI USB-7885R. The home made code (LabVIEW FPGA module) for beta channel is similar to the classic MAC3 module with outputs from photomultiplier A, B, and C, double coincidences AB, AC and BC and their logical sum, triple coincidence ABC and live time. Extendable dead time is used. The processing of gamma channel and beta-gamma coincidences can use both fixed and extendable dead time in gamma channel. All the outputs are recorded in an Excel table and then processed in accordance with the procedures of conventional coincidence method.
Features and measurement capabilities of beta channel were verified by measuring a set of ^{3}H sources. The range of efficiency corresponding to different filter positions was 0.40 – 0.50.
The specific activity of ^{3}H solution was determined using the TDCR part of TDCRG system with a combined uncertainty of 0.6 % and was in good agreement (0.3%) with values measured using the existing CMI TDCR device, which was used in CCRI(II)-K2.H-3 comparison.
For verification of parameters of gamma and beta-gamma coincidence channels, a measurement of ^{60}Co and ^{54}Mn solutions was performed. Different settings of gamma channel were used for efficiency extrapolation measurement to minimize the measurement uncertainty. The resulting activity values were in good agreement with the values obtained using conventional 4πβ-γ coincidence system with gas proportional counter (PC) in beta channel, which is routinely used for ^{60}Co and ^{54}Mn standardisation in CMI. The difference of the activity value did not exceed 0.25%.

Corresponding author's email address: **jsochorova2@cmi.cz**

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USE OF THE TDCR METHOD FOR THE STANDARDIZATION OF RADIONUCLIDES IN THE FTMC

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Gudelis, A. ; Gaigalaitė, L. ;Gorina, I.; Butkus, P.

FTMC(LT)

In the National metrology institute, FTMC, the equipment which uses the TDCR method for electron capture and beta-emitters standardization is based on the MAC3 electronic module and a three photomultiplier liquid scintillation counting apparatus. By now, the following radionuclides were standardized: H-3, C-14, Cl-36, Ni-63, Sr-90, Tc-99 and I-129. The Ionizing Radiation Metrology Laboratory participated in the H-3 activity comparison between FTMC, LNE-LNHB and VNIIM in 2013. This allowed the comparison of the FTMC values to the CCRI(II)-K2.H-3 2009 H-3 international comparison through the established traceability chain.
Tritium has been standardized with the combined relative standard uncertainty of 0.7% (k = 1). This radionuclide is important for activities undertaken in the Ph. D. studies programs on "Research into radionuclides transfer from a near-surface radioactive waste repository to the environment" and Application and development of LSC methods for standardization of radionuclides". The tritiated water solutions prepared after the application of TDCR method are used for calibrations of an ultra low-level liquid scintillation counter Quantulus-1220. Hence, the TDCR method, as a primary method, enables the quality assurance and quality control of the secondary radiation measuring equippment. The LSC Quantulus-1220 is used in environmental radioactivity, radiation protection and decommissioning programs.
FTMC has organized two comparisons in the field of tritium activity measurements in water, in 2013 and 2015. The results of these comparisons showed that the determination of tritium in water was not for all participating laboratories a straightforward task. The comparisons revealed problems in both calibration and uncertainty evaluation.
FTMC provides support to Lithuanian laboratories via preparation of comparisons' samples traceable to the national standard of radionuclide activity.

Corresponding author's email address:**gaigalaite@ftmc.lt**

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IONIC LIQUIDS AS SOLVENTS FOR LSC: PYRANINE AS WAVELENGTH SHIFTER TO ENHANCE ČERENKOV-LIGTH DETECTION

122

Mirenda, M.[1]; Ferreyra, C.[2]; Rodrigues, D.[1]; Arenillas, P.[2]; Sarmiento, G.[1]; Krimer, N.[1]

[1] GQ-CNEA (AR), [2] LMR-CNEA (AR)

Ilonic liquids (ILs) are organic salts that have melting points near to room temperature, taken 100ºC as an arbitrary upper limit [1]. The organic constituent ions give them distinctive physicochemical properties that clearly differ from traditional organic solvents, most of them toxic, volatile and flammable. Their high thermal stability, low flammability and moderate conductivity are some of the remarkable features that justify its nascent application, e.g. in nuclear technology [2].

We have been already shown that ILs are suitable solvents for liquid scintillation counting (LSC) [4]. The specific activity of a 18F solution (emitting positrons up to 634 keV) has been standardized by means of the TDCR- Čerenkov method [3], using 1-Butyl-3-methylimidazolium Cloride (BmimCl). This IL (with a refraction index of ~1.5) allowed a detection efficiency of 7 %.

In the present work we have synthesized a novel IL, the 1-Butyl-3-methylimidazolium 8-hydroxypyrene-1,3,6-trisulfonate (BmimHPTS), with the aim to enhance the amount of detected Čerenkov-light. The strategy was to use the 8-hydroxypyrene-1,3,6-trisulfonate anion, most commonly referred as pyranine, as a wavelength shifter. This dye is suitable to re-absorb the Čerenkov radiation in the UV region (low photocathode responsivity) and to re-emit it as fluorescence in the blue region (optimum photocathode responsivity).

First, we perform an exhaustive photophysical characterization of different BmimHPTS/BmimCl mixtures in order to find the optimal concentration of pyranine that gives the highest fluorescence signal. The analysis of absorption and steady-state emission spectra enables us to find the adequate balance between fluorescence intensity and self-absorption.

Second, we used the TDCR system at the LMR-CNEA [5] in order to optimize the pyranine concentration for differences in vial geometries. As a result, the detection efficiency for ^{18}F, using the TDCR–Čerenkov method, increases up to 15 % for 1.5×10^{-4} M of BmimHPTS in BmimCl. This result shows no significant difference compared with the activity concentration of the same ^{18}F solution using the classical TDCR method with a commercial scintillation cocktail.

Third, we perform several control experiments to safely discard another kind of luminescence that could eventually cause coincidences in the photomultipliers. In particular, no coincidence events above the background were observed for measurements with ^{14}C in the same experimental conditions.

The fact that the activity of high energy β particles emitters can be quantified in ILs mixture by TDCR–Čerenkov technique makes them a promising cocktail for future applications in LSC measurements, especially for cases where high water solubility cocktails are required.

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Corresponding author's email address: **darodrig@tandar.cnea.gov.ar**

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STANDARDIZATION OF FE-55 BY TDCR EFFICIENCY FITTING USING EGS5MPI MONTE CARLO CODE

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Sato, Y.

NMIJ(JP)

IWe developed efficiency fitting program for TDCR counting data to standardize Fe-55 as an electron capture decaying nuclide. Experimental data were fitted with parameters of activity, kB value and ratios of free parameters on the plane of efficiency and TDCR value in this method.

The efficiency fitting program for beta-emitting nuclide was modified for applying electron capture decaying nuclides. Eleven paths of rearrangement was considered and probability of energy deposition for Kα, Kβ and L X-rays were estimated by EGS5MPI Monte Carlo code on parallel computing environment. Functions of FJON, BIRKS, EFFIC2, EffenEC, probaEC and Quant_EC from TDCRB-2 and CN2004 programs were transplanted and modified for the fitting program. Escape events of X-rays were stored in the arrays of deposition energy and number of paths of rearrangement was compressed. Atomic data were taken from CNEC.nuc file of CN2004.

Fe-55 solution of about 0.3 mL and pure water of about 0.7 mL was mixed with Ultima Gold™ scintillation cocktail of 15 mL in a 20 mL vial. Triple coincidence count rates and double coincidence count rates of sources were measured by a TDCR counter that consisted of three photomultipliers for photon detection and NIM modules for coincidence counting in AIST. Typical double count rate was about 1200 cps and TDCR value was about 0.2. The probability of full energy deposition of Kα, Kβ and L X-rays calculated by EGS5MPI code were 98.8 %, 98.1 % and 100 %, respectively. Specific activity of the solution was 7.962 ± 0.159 kBq/g (k=2) by the developed efficiency fitting program.

On the other hand, the sources was also measured by PerkinElmer Tri-Carb 3110 TR. The detection efficiency was calculated by CN2004 program and specific activity of the solution was 7.932 ± 0.190 kBq/g (k=2).

Both of specific activities were in good agreement within uncertainties. In near future, KL1L2L3M model will be applied for TDCR efficiency fitting.

Corresponding author's email address: **yss.sato@aist.go.jp**

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Y-90 STANDARDIZATION BY CIEMAT/NIST LIQUID SCINTILLATION METHOD

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de Oliveira, A. E.; *et al.*

CNEN-IRD-LNMRI (BR); CENTIS (CU); CNEA (AR); CCHEN (CH)

Radionuclide ^{90}Y ($T_{1/2}$ =64.4 h, 100% β^- decay, E_{max} 2.28 MeV) is a widely used for therapy. It is a pure beta emitter and of growing significance in therapy, especially liver cancer, and it is obtained from ^{90}Sr, a high yielded fission product. It is used for cancer brachytherapy and as silicate colloid for the relieving the pain of arthritis in larger synovial joints. The measurement of administered activity before and residual activity after treatment in radionuclide calibrators is required to determine total activity delivered to the patient. In this sense, the absolute calibration of Y-90 is essential to provide traceability to the measurements in Brazilian hospitals, clinics, and industrial radiopharmacy that provide samples to the treatments.

The CIEMAT/NIST method based on liquid scintillation technique was used for the primary standardization using commercial cocktails of Hisafe and Ultimagold. Commercial WALLAC 1414 Liquid Scintillation Spectrometer with two phototubes in coincidence was used for the measurements. Several tests were conducted to verify some factors that could influence the results such as: cocktail scintillator volume, which ranged from 10 to 20 mL and intensity of the samples which ranged from 10^5-10^6 counts.

Two sets of sources from two different solutions were prepared to verify the consistency of results by determining the calibration figures of an IG11 ionization chamber. One sample was provided by The Isotope Center of Cuba, CENTIS-DMR as part of a program to support development and harmonization of measurements in hospitals and clinics. The other sample was provided by Brazilian radiopharmaceutical Producer. The chemical form of both solutions was $^{90}YCl_3$ in 0.04M and 0.1M in HCl, respectively. The combined standard uncertainties were less than 1% and the results agreed within one standard deviation value.

The anticoincidence method using ^{60}Co as a tracer was also performed but the results did not show consistency and studies are being conducted to verify these discrepancies.

Corresponding author's email address: **aedu@ird.gov.br**

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MONTE CARLO MODELLING OF LIVE-TIMED ANTICOINCIDENCE COUNTING (LTAC) FOR CU-64

031

Bergeron, D.E.; Fitzgerald, R.

NIST (USA)

Copper-64 decays by positron emission, beta decay, and electron capture with a half-life of 12.7004(20) h (Bé *et al.*, 2011). Interest in Cu-64 stems mostly from its promise as a "theranostic" radionuclide. Copper nimbly coordinates to a vast assortment of complexing agents, allowing for very specific chemical targeting of biologically active sites. Its β^+ branch (≈ 17.5 %) allows for quantitative imaging by positron emission tomography (PET), while its β^- branch (≈ 38.5 %) provides significant local therapeutic dose ($E_{\beta max}$ ≈ 580 keV).
NIST recently standardized a Cu-64 solution by live-timed anticoincidence counting (LTAC), with confirmatory measurements by the CIEMAT-NIST efficiency tracing (CNET) and triple-to-double coincidence ratio (TDCR) methods of liquid scintillation counting. The LTAC-determined standard activity was used to determine a calibration factor for the Vinten 671 ionization chamber maintained at NIST. This calibration factor provided an indirect link to the international intercomparison described by Bé *et al.* (2012).
As part of the LTAC standardization, Monte Carlo simulations of the experiment were performed with a GEANT4 package previously described by Fitzgerald (2016). Simulations were optimized to reproduce the experimental beta (liquid scintillation) and gamma (NaI(Tl)) spectra.
Gating over the 1345 keV gamma peak (from the electron capture transition), the LTAC data were analyzed by applying linear and quadratic fits to the simulated data over the inefficiency range in which experimental data were collected and calculating a correction factor (= 0.9956(6)).
With additional gates set over the annihilation photon peaks, a single-parameter scaling approach was also used to analyze the LTAC data. Data were analyzed with several different weights applied to the gates to generate a combined "effective inefficiency". It was clear that while the Monte Carlo model more easily reproduced beta contributions to the inefficiency plots, the scaling approach was less robust than extrapolation methods, resulting in consistently lower recovered activities with much larger (≈ 2×) uncertainties.

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Fitzgerald, R., 2016. ARI 109, 308.

Corresponding author's email address:

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ACTIVITY DETERMINATION OF ^{67}Ga USING 4πβ–γ COINCIDENCE COUNTING

045

Nähle, O.

PTB (DE)

Gallium-67 decays by electron capture to ^{67}Zn. For several reasons the activity standardisation of this nuclide is challenging and considered to be one of the most difficult nuclides in the generic grouping tables to support CMC entries. At first, the half-life of 3.2613(5) d is rather short and requires careful planning of the efficiency variation techniques needed for 4πβ–γ coincidence counting. Another difficulty is the existence of a delayed excited state of ^{67}Zn with a half-life of 9 µs that decays to the ground state via a γ transition which mainly leads to the ejection of conversion electrons and thus, inevitably is seen in the β channel with high efficiency. Especially for coincidence counting with non-extending dead-times, this can easily lead to an over-estimation of activity of more than 1% depending on measurement conditions and initial activity. This might partly explain discrepancies observed in intercomparisons.
Funck [1] theoretically derived formulae to correct the effects of delayed states when using coincidence counting and non-extendable dead-times. There is one effect that can be overcome by using a dead-time about 10 times longer than the half-life of the delayed stated, but another rate-dependent effect still remains. To test these corrections a series of measurements continuing for more than 3 weeks with activities ranging from 25 kBq in the beginning down to 250 Bq was carried out and the dead-time was varied with set values of 4 µs, 8 µs, 16 µs, 32 µs and 64 µs. Seven sources – covered with increasing number of absorbing foils – were measured in a coincidence system with a proportional counter (PC) operated at atmospheric pressure and one source was measured in a system with a pressurized proportional counter (PPC) with different threshold settings to vary the efficiency.
Every single measurement was corrected for the rate-dependent effect making assumptions on detection efficiencies and taking nuclide data into account. After compiling all results an extrapolation to infinite dead-time was carried out. It is shown that the correction formulae work, resulting in consistent results deviating not more than two per-mille. A final uncertainty of 0.52% for the activity determination was reached and an ampoule was sent to the SIR to have the measurement result included in the key comparison database. The result is used to establish new calibration factors for the PTB ionization chambers as a secondary standard.

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Corresponding author's email address: **ole.j.naehle@ptb.de**

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INVESTIGATION OF γ-γ COINCIDENCE COUNTING USING THE NATIONAL NUCLEAR ARRAY (NANA) AS A PRIMARY STANDARD

100

Collins, S. M.[1]; Shearman, R.[1-2]; Keightley, J. D.[1]; Regan, P. H.[1-2]

[1]NPL (UK), [2]US (UK)

The National Physical Laboratory (NPL) has recently installed and commissioned the National Nuclear Array (NANA), a multi-detector γ-ray spectrometer array [1]. The complete array utilises 12 $LaBr_3$(Ce) scintillation detectors coupled to fast timing photomultiplier tubes as its intrinsic γ-ray spectrometers. The primary purpose of the array for nuclear structure and nuclear decay data measurements, however the array's coincidence counting capabilities also allow it to be used for the primary standardisation of radionuclides that emit two or more γ-rays in cascade using the γ-ray energy coincidence (γ-γ) counting technique. In the current work, we report on the application of NANA as a primary standardisation system by γ-γ coincidence counting. The β^- decay of ^{60}Co to ^{60}Ni has been selected for this investigation as the intense spin/parity 4^+ --> 2^+ --> 0^+ cascade of pure, stretched E2 transitions energy 1173.2 keV and 1332.5 keV provides an ideal decay scheme for the application of the coincidence counting method for primary standard activity evaluation. Measurements were performed using eight individual $LaBr_3$(Ce) detectors placed in a central symmetrical ring of the NANA frame. This provided relative, coincident angles between the individual detectors oft $\Delta\theta$ = 45°, 90°, 135°, 180°, 225°, 270° and 315°. The front face of the detectors were placed at 40 mm from a centrally located source of ^{60}Co. The time stamped pulse trains of the individual detectors were processed and stored in a list mode format using a CAEN V1751, 1 GHz digitizer. A series of five dried drop deposited sources were measured for 24 hours each. The coincidence gating was performed offline with logical gated conditions on the 1173.2 keV and 1332.5 keV γ rays. For each $LaBr_3$(Ce) detector at θ = 0, the activity was determined for each detector pair around the symmetrical ring, using the calculation method described by Voltovitsky and Naudus [2]. This provided a total of 56 coincidence activity determinations per measurement. The mean average activity at each angle showed a strong relationship to the inverse of the γ-ray angular correlation function of the decay of ^{60}Co. The angular correlation correction has been investigated using Monte Carlo simulations to account for the angular smearing and extended 2-dimensional source geometry. The source material solution used was also standardised using the NPL 4π(Liquid scintillation)-γ digital coincidence counting system as a comparison. The two distinct absolute counting techniques have determined consistent results (reduced x^2= 0.58) for the ^{60}Co solution of 330.92 (86) kBq.g-1 (k = 1) and 333.3 (30) kBq.g-1 (k = 1) by 4π(LS)-γ DCC and γ-γ coincidence counting respectively.

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Corresponding author's email address: **sean.collins@npl.co.uk**

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STANDARDIZATION OF ^{67}Cu AND CALIBRATION OF THE IONISATION CHAMBER. IMPURITIES AND DECAY SCHEME PROBLEMS

004

Sahagia, M; Luca, A.; Ioan M. R.; Antohe, A.; Ivan C.

IFIN-HH (RO)

This paper describes the absolute standardization of ^{67}Cu, a beta-gamma decaying radionuclide with a triangular decay scheme, by the 4πβ(PC)-γ coincidence method, and use of the solution for the calibration of the secondary standard, CENTRONIC IG12/20A ionization chamber. To our knowledge, no results regarding its standardization were published until now. Three specific difficulties were overcome in our measurements. (i) The excited 93.31 keV level of the daughter ^{67}Zn has a 9.10 μs life, what implies a special application of the method in standardization. We avoided the counting of the 93.31 keV gamma-rays in the gamma channel and applied the corrections for the non coincident conversion electrons and gamma-rays in the beta channel counting. (ii) A recent publication of Chen et al., in Physical Review C 92, 044330 (2015), reconsiders significantly the classical decay scheme parameters published in the most recent ENSDF evaluation (Junde et al., Nucl. Data Sheets 106, 159 (2005)). The influence of these new decay data on measurements uncertainty is analyzed. (iii) The measurements by gamma-ray spectrometry concluded that the available ^{67}Cu solution for measurement had a significant content of impurities. They were determined and their influence on the ^{67}Cu activity was calculated. The most important impurity was ^{67}Ga (the activity ratio ^{67}Ga/^{67}Cu was about 3.6 % at the reference date and time), an electron capture decaying radionuclide, which emits common energy gamma-rays with ^{67}Cu (up to 393 keV) and a few higher energy gamma-rays with very low emission intensities, allowing its measurement. The CENTRONIC IG12/20A ionization chamber experimental calibration with the standardized solution needed to apply corrections for the impurity content. In parallel, its detection efficiency for ^{67}Cu was calculated, considering the influences of the decay scheme parameters. The two types of results were compared.

The conclusions of all measurements were:

- The result of the absolute standardization is less influenced by the decay scheme parameters and impurities, than the measurements by gamma-ray spectrometry and ionization chamber, due to: (a) in the coincidence method, efficiency extrapolation variant, there was no influence of beta branching ratios and emission intensity of the main 185 keV gamma-rays, and (b) less influence of impurity.
- In the gamma-ray and ionization chamber measurements, important influences of emission intensities of ^{67}Cu and ^{67}Ga impurity gamma-rays occured.

Corresponding author's email address: **msahagia@nipne.ro**

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MEASUREMENT FOR ACCURATE EXTRAPOLATION IN 4πβ-γ COINCIDENCE COUNTING METHOD USING PLASTIC SCINTILLATOR

082

Unno, Y.[1]; Sanami, T.[2]; Sasaki, S.[2]; Hagiwara, M.[2]; Yunoki, A.[1]

[1]NMIJ (JP); [2]KEK (JP)

For accurate measurement of absolute value in radioactivity, apparatus to conduct the 4πβ-γ coincidence counting method is usually composed of beta ray and gamma ray detector. Uncertainty related with the extrapolation technique in beta ray counting efficiency to determine the absolute value is often a dominant source of uncertainty. Higher beta ray counting efficiency make the extrapolation uncertainty smaller as shown in our previous study [1]. We aimed to develop new apparatus for the 4πβ-γ coincidence counting with high beta ray counting efficiency. A plastic scintillation detector was chosen for the beta ray counting because the high counting efficiency was realized in a way that samples were placed in the sealed plastic scintillator as well as Kawada (2004) [2]. Furthermore, pulse height distribution corresponding to emitted beta ray energy spectrum could be obtained. In our previous setup [3], the plastic scintillator (ELJEN, EJ-212) was optically connected with two facing large photomultiplier (Hamamatsu, H3177-50) via a light guide. In the present study, another thimble-sized photomultiplier (Hamamatsu, R9880U-210) was selected as a result of competition in the beta ray counting efficiency that was 93 % to radioactivity of Cs-134 (previously, 74 %). Combination with a gamma ray detector was considered in the point of size of photomultiplier. The high efficiency was achieved by direct optical contact to two facing photomultipliers in addition to inherent high sensitivity of the photomultiplier. It means that large solid angle and short flight length of scintillation light strengthened signals to be detected. The facing setup of two photomultipliers were useful to reject self-noise of the photomultiplier and clarify threshold level in pulse height distribution.
To use this beta ray detector in the 4πβ-γ coincidence counting method with sufficient counting statistics, a through-hole type NaI(Tl) scintillation detector was used. The through-hole have a 20 mm diameter at the center of side of Φ3" x 3" size cylindrical NaI(Tl) crystal. The beta ray detector was inserted in the through-hole completely. The hole made pulse height resolution obtained by the NaI(Tl) scintillation detector worse actually (17% in FWHM at 662keV), but that had no concern for the accuracy. The extrapolation uncertainty for absolute measurement of Cs-134 radioactivity was evaluated at 0.3 % (k=1) at most. The result was comparable to ones in Kawada (2004) [2]. In discussion about lower level limit to detect low energy beta ray, the result was reasonably explained by calculation from the number of produced scintillation photons to electrical pulse heights.

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Corresponding author's email address: **y.unno@aist.go.jp**

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^{68}(GE+GA) ACTIVITY STANDARDIZATION BY 4∏β(LS)-Γ(NAI(TL)) ANTICOINCIDENCE COUNTING MEASUREMENTS

087

da Silva, C. J.; da Cruz, P. A. L.; Iwahara, A.; Loureiro, J. S.

LNMRI/IRD (BR)

The radionuclide ^{68}Ge decays 100% by electron capture emitting characteristics X-rays and Auger electrons with energy lower than 11 keV. ^{68}Ga, its daughter, decays by positron emission and electron capture emitting characteristics X-rays and Auger electrons with energy lower than 10 keV and 1077 keV gamma ray of low intensity (3.235%).
Due to these decay characteristics, ^{68}Ge in equilibrium with its daughter ^{68}Ga, is a potential surrogate of ^{18}F not only in the checking of the radionuclide calibrators but also in quality control tests of diagnostic medical imaging in nuclear medicine devices. Besides, the half-life of ^{68}Ge (270.95 ± 0.16) d is much longer than the ^{18}F half-life (1.8288 ± 0.0003) h.
A ^{68}(Ge+Ga) solution has been standardized at National Institute of Ionizing Radiation Metrology (LNMRI), in Brazil, in the frame of a Bureau International des Poids et Mesures (BIPM) international key comparison piloted by National Institute of Standards and Technology (NIST/USA).
The method used was the 4πβ(LS)-γ(NaI(Tl)) anticoincidence method with live-timed and extended dead-time and the result was validated by 4πβ(LS)-γ(NaI(Tl)) coincidence and liquid scintillation Triple to Double Coincidence Ratio (TDCR) measurement methods.
Samples for measurements of the three methods were prepared in 15 mL scintillation cocktails of commercial Optiphase 'HiSafe' 3 and Ultima Gold, manufactured by PerkinElmer, Inc. USA, and contained in low potassium content glass vials.
The differences of the activity concentration values of coincidence and TDCR measurements from the anticoincidence result were +1.6% and +0.78%, respectively, which were within experimental evaluated uncertainties at 95% level of confidence (coverage factor k=2). The combined standard uncertainties were 0.53, 0.87 and 0.82% for anticoincidence, coincidence and TDCR methods, respectively, at k=2.
Thus, LNMRI is able to provide standard sources of ^{68}(Ge+Ga) with relative standard uncertainty less than 1% to medical community can calibrate their measuring instruments with uncertainty within the levels required by regulatory authorities.

Corresponding author's email address: **carlos@ird.gov.br**

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DISINTEGRATION RATE AND GAMMA RAY PROBABILITY PER DECAY MEASUREMENT OF Cu-64

089

Yamazaki, I. M.; Koskinas, M. F.; Moreira, D. S.; Takeda, M. N.; Dias, M. S.

IPEN-CNEN/SP (BR)

This work aims to present the ^{64}Cu standardization method developed in the Nuclear Metrology Laboratory (LMN) at the IPEN-CNEN/SP, using a 4πβ-γ coincidence system, and the measurement of the gamma-ray emission probability per decay of 1345.7 keV of ^{64}Cu by means a REGe gamma-ray spectrometer. The 4πβ-γ coincidence system consists of a gas-flow proportional counter with 4π geometry and using 90% Ar + 10% CH_4 gas at 0.1 MPa, as the β detector, coupled to two NaI(Tl) crystals for the gamma-ray emission detection. The events were registered by means of a Time to Amplitude Converter (TAC), associated with a Multi-Channel Analyzer. Two gamma-ray windows were set for the coincidence measurements, one including the positron-annihilation quanta and the other located at the total absorption energy peak of the 1345.7 keV gamma-ray transition. The ^{64}Cu was obtained by irradiating 0.3 mg of metallic copper in the IEA-R1 research reactor under a $1x10^{13}$ cm^{-2} s^{-1} thermal neutron flux. To obtain the ^{64}Cu solution, the foil irradiated was dissolved in 20 µl of HNO_3 65% and, then, it was diluted in 20 mL of 0.1N HCl. The sources to be measured in the 4πβ-γ system were prepared by dropping known aliquots of the radioactive solution onto a 20 $µg.cm^{-2}$ thick Collodion film. This film had been, previously, coated on both sides with a 10 $µg.cm^{-2}$ thick gold layer in order to render the film conductive. A seeding agent (CYASTAT SN) was used for improving the deposit uniformity and the sources were dried under a nitrogen jet at 45 °C. The accurate source mass determination was performed using a Mettler XP56 balance by the pycnometer technique. A bi parametric linear square fit was performed in order to determine the activity. The 4π detector efficiencies were changed by using external absorbers over and below the radioactive sources. The absorbers were made of Collodion films, previously, coated with a 20 $µg.cm^{-2}$ gold layer and aluminum foils. To obtain the extrapolation curve, the efficiency parameter was varied in the range from 99 % to 94 %, for the 511 keV window, and in the range from 59 % to 13 %, for the 1345.7 keV window.

The gamma-ray full efficiency peak curve of the germanium spectrometer was measured at 17.9 cm of the source-detector distance, in the energy range from 244 keV to 2754 keV, by measuring flamed-sealed ampoules of ^{24}Na, ^{60}Co, ^{133}Ba,^{137}Cs and ^{152}Eu standardized at the LMN. The sample for measurement in the spectrometer was prepared in flame-sealed ampoule with 1 mL of the diluted solution.

Monte Carlo simulation, by means of ESQUEMA code, was applied to simulate all the radiation interaction processes in the coincidence system described by the radionuclide decay scheme, considering all materials and geometries of the experimental setup, as well as the characteristics of the radioactive sources; the predicted behavior of the extrapolation curve was compared with the experimental curve.

The emission probability per decay of 1345.7 keV gamma ray of ^{64}Cu was measured by the absolute method. The result obtained from the average of two distinct irradiations was (0.478 ± 0.018) % and it is in agreement with the literature.

Corresponding author's email address: **koskinas@ipen.br**

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^{134}CS ACTIVITY STANDARDIZATION BY 4∏ (LS)-Γ(NAI(TL)) ANTICOINCIDENCE COUNTING AND SUBMISSION TO INTERNATIONAL REFERENCE SYSTEM

130

Gomes, R. dos S.; da Silva, C. J.; da Cruz, P. A. L.; Iwahara, A.; Loureiro, J. S.

LNMRI/IRD (BR)

The National Laboratory for Ionizing Radiation Metrology (LNMRI) that belongs to Institute of Radioprotection and Dosimetry (IRD) has been working since 2005 with the live-timed anticoincidence counting for activity standardization. During this period LNMRI performed 50 primary standardization of 26 radionuclides with different decay schemes. Recently a ^{134}Cs radioactive solution was purchased from a commercial supplier in order to establish its primary standardization. ^{134}Cs is a beta gamma emitter with half-life of 754.006 days and gamma emission with 242.7 to 1365,2 keV energy. This radionuclide is very important for gamma spectrometry calibration and study of gamma sum effect in spectrum analysis. In Brazil this radionuclide is used as standard in environmental, water and food control. LNMRI supplies users with this standard solution and also in water matrix for comparison program in which take part 22 laboratories. The primary standardization of ^{134}Cs is very important due its potential use in radionuclide metrology as a tracer.

In order to perform the measurement of this radionuclide three independent methods were used: coincidence counting, anticoincidence counting and CIEMAT/NIST method. Samples for measurements of the three methods were prepared in 15 mL scintillation cocktails of commercial Optiphase 'HiSafe' 3 and Ultima Gold, manufactured by PerkinElmer, Inc. USA, and contained in low potassium content glass vials. The results of activity concentration values presented very close agreement, within 0.3%.

Since 1984 until now there are 15 submissions of ^{134}Cs samples to the International Reference System (SIR/BIPM) but only three show results with agreement better than 0.1%. The LNMRI submission performed in 2015 is in perfect consistent with reference value for k =1, around 0.07%, and we hope that it improve the value of CRV. The LNMRI activity submitted to SIR/BIPM was the value obtained by live-timed anticoincidence counting.

In order to memorize the primary standardization results, the LNMRI/IRD secondary reference system with well type ionization chamber was calibrated with this solution by the determination of calibration figure (A/Bq) for NIST and LNMRI standard ampoules.

This paper presents a detailed discussion of measurement used by each independent method.

Corresponding author's email address: **carlos@ird.gov.br**

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RUSSIAN NATIONAL PRIMARY ACTIVITY STANDARD EXTENSIVE RENEWAL, FIRST RESULTS

044

Alexeev, I.; Kharitonov,I. ;Sepman, S.; Shilnikova, T.; Tereschenko, E.; Trofimchuk, S. ; Zanevsky, A.; Zhukov, G.

VNIIM (RU)

VNIIM Ionizing radiation department took part in Russian national program of national standards upgrade in order to provide modern technology and scientific background for the primary national activity standard. Almost all instruments of the primary standard underwent a deep upgrade in the frame of the program. Several new methods were implemented in the new instruments, which were incorporated into the standard. The upgrade program lasted from 2012 and was finished in 2016. Here we are presenting a general overview of the instruments and methods of the primary standard, the ideas of the instruments architecture and first results of the comparisons and instruments features exploration.

The set of primary standard instruments includes: 4π β and γ absolute counters, β-γ and Kx-γ coincidence instruments, a calorimeter and a precalculated ionization chamber. Two instruments were completely new to the standard and were designed and built during the standard upgrade: a liquid scintillation counter implementing the TDCR method and a 4πγ two NaI crystal absolute counter. Other instruments were partly upgraded. All new instruments were designed using modern digital architecture and most of old instruments got digital signal processing electronics too. A unified counting registration channel has been designed. The central piece of this design is an industry-produced digitizer, which allows simultaneous pulses registration at multiple channels. All instruments were equipped with identical 12bit 250 MHz digitizers. The offline data processing mode was chosen with complete events data set storing at the experiment control computer hard drive. All coincidence instruments were equipped with these unified registration channels. The 4πγ original two-crystal design counter utilizes such registration channels too, in order to account properly for the coincidence events. The 4πγ instrument detector consists of two 200 mm diameter and 100 mm high NaI coaxially placed crystals. The top edge of the lower crystal has a cylindrical hallow for the radionuclide source placement. The prototype of this instrument successfully passed the SIRTI key comparison of ^{99m}Tc and ^{18}F. The overview of the instruments cross-comparisons and their metrological properties investigation is presented. Various technological and methodological problems, which were encountered during the primary standard modernization, are discussed.

Corresponding author's email address: **i.v.alekseev@vniim.ru**

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STANDARDIZATION OF CD-109 BY THREE METHODS

027

Yuan, M.C.; Yeh, C. Y.; Chu, W.H.

INER (TW)

Cd-109 is quite a useful nuclide for the calibration of X-ray spectroscopy, gamma-ray spectroscopy or as an excitation source in X-ray fluorescence; therefore, to standardize this nuclide is meaningful. In this work, 4π(PC)e-X coincidence counting, 4π(LS)ce counting and the CIEMAT/NIST methods were used to determine the radioactivity of the Cd-109. The Cd-109 source solution was bought from CMI/ Czech Republic with the certificate of 59.96(2.2%) MBq/g. To make proper counting rates for these counting systems, the Cd-109 solution was diluted by a factor of 100 for the 4π(PC)e-X system, and diluted by a factor of 2000 for the 4π(LS) system.

10 VYNS samples were prepared for the 4π(PC)e-X counting. Each sample would be sandwiched between aluminum foils of thicknesses from 0.6 to 3.2 mg/cm2 to avoid the auger electrons and to change the counting efficiency. A counting result of 60.33(0.75%) MBq/g was obtained after the counting data being corrected by the dead time, resolving time, background, decay time, decay scheme, dilute factor and extrapolated the counting efficiency of the conversion electron to 100% by a two-order polynomial equation.

A Perkin Elmer 2900TR scintillation spectrometer was used to perform the 4π(LS)ce counting and CIEMAT/NIST counting. In the 4π(LS)ce counting, three samples were prepared and each contained 15 mL Ultima Gold AB scintillator and 0.1 mL Cd-109 solution in a polyethylene vial. Electron energy spectrum of each sample was accumulated. Through analyzing the peak area of the conversion electrons and considering the contribution of the low energy tail of the conversion electron peak, high energy tail of the auger electron peak, the 88 keV gamma rays and the background, the emission rate of the conversion electron could be obtained. And then corrected by the decay scheme and the dilute factor the counting result of 61.16(0.56%) MBq/g was observed.

In the CIEMAT/NIST counting, a set of H-3 tracer quench standard was built by using the NIST 4297F SRM, and the CN2003 program was adopted to calculate efficiency relations between the Cd-109 and H-3. Six samples were arranged for this counting. Each sample contained 15 mL Ultima Gold AB scintillator, 0.5 mL water, 0.1 mL Cd-109 solution and around 10 μL nitro-methane in a 20 mL glass vial. The average result of the 6 samples measured by the CIEMAT/NIST method was 61.07(0.61%) MBq/g. Counting results of the three methods showed good consistence in 95% confidence level and their weighted mean was 60.93(0.22) MBq/g. This weighted mean would be used to calibrate the well type ionization chambers, IG11-N20, IG-11-A20 and ISOCAL-IV, in 5 mL ampoule geometry at INER to maintain the Cd-109 radioactivity measurement standard.

Corresponding author's email address: **mcyuan@iner.gov.tw**

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DIFFERENT TECHNIQUES FOR ^{198}AU ACTIVITY MEASUREMENTS IN GOLD FOILS

070

Tereshchenko, E.; Moiseev, N.; Didyk, A.

VNIIM, (RU)

The activity of ^{198}Au in thin sources prepared from a solutions can be determined with high accuracy (0.3%) using the 4πβ-γ-coincidence technique. Measurement conditions will change vastly when dealing measurements of ^{198}Au activity in gold foils. The foils are the sources which generally have significant self-absorption not only beta-particles and conversion electrons but gamma-photons. Furthermore, gamma-photons absorption leads to photo-electrons nascency whose energy is equal to that of the conversion electrons. Therefore, the measurement of the ^{198}Au activity in the foils is a separate independent problem.

The present report is focused at comparison of different techniques for gold foils activity measurements The result of ^{198}Au activity measurement in metallic foil obtained by two different coincidence techniques (4πβ(LS)-γ(NaI) and 4πβ(Plastic)-γ(NaI)) and 4πγ(NaI) counting is described. The 4πβ(LS)-γ(NaI) coincidence setup developed at D.I. Mendeleyev Institute for Metrology was described at Proceedings of the 20th International Conference on Radionuclide Metrology and its Application (2016, Int. J. Appl. Radiat. Isot. vol. 109, 378-381).

The 4πβ(Plastic)-γ(NaI) coincidence technique is used at VNIIMs neutron laboratory for thermal neutron fields parameters investigation. For the 4πβ(LS)-γ(NaI) absolute activity measurement gold foil placed into liquid scintillation detector (10 ml Ultima Gold) for detecting beta-particles. In order to decrease the beta-participle background the double coincidence technique (two PMT tube) was used. The colored films with variated transparency and high voltage variation were used for the beta efficiency extrapolation in the 30-78% range.

For 4πβ(Plastic)-γ(NaI) absolute activity measurement gold foil placed between plastic scintillation beta-detector and NaI gamma-detector. For the beta efficiency extrapolation in the 20-50% range were used next processes: source-detector distance changing, thin filter for beta and low threshold variation.

The results obtained are in good coincidence. The combined uncertainty (K=2) of results is estimated in the range 1-2 % for each method.

Corresponding author's email address: **tee@vniim.ru**

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"INFLUENCE METHOD", A USEFUL TOOL FOR RADIATION MEASUREMENTS

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Mayer, R. E.; Ríos, I. J.

CNEA (AR); UNCUYO (AR)

The "Influence Method" is conceived for the absolute determination of a nuclear particle flux in the absence of known detector efficiency and without the need to register coincidences of any kind. This method exploits the influence of the presence of one detector in the count rate of another detector, when they are placed one behind the other and define statistical estimators for the absolute number of incident particles and for the efficiency.
The method and its detailed statistical treatment have been published in 2015, while in 2016 its application to the measurement of a moderated neutron flux was published, along with the extension of the method to multiple detectors. Another application published dealt with the implementation of the method to experimentally assess the "space charge effect" in proportional counters undergoing irradiation by intense radiation bursts.
The current presentation deals with the philosophy of the method, which allows to deal with a problem with a number k of unknowns, through the employment of k detectors (whenever possible), placed in such a way that their successive influences yield correlated measurements. The ensuing equations would allow to find statistical estimators for each unknown parameter.

Corresponding author's email address: **mayer@cab.cnea.gov.ar**

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MEASUREMENTS OF RESPONSE-RATIO OF IONIZATION CHAMBER FLOWED WITH DRY AIR TO WITH P-10 GAS IN THE CALIBRATION OF GAS MONITORS

061

Yunoki, A.; Kawada, Y.; Hino, Y.

AIST/NMIJ(JP)

Gas-flow type ionization chambers (ICs) are often used for monitoring radioactivity concentration of gaseous effluent from nuclear facilities and of air in work places of radioisotope laboratory. Since the component of the gas is mostly air in practice, such gas monitor should be calibrated under the condition filled with air.

In NMIJ, standardization of radioactive gases are performed by the differential counting method using several proportional counters with different length [Yunoki, *et. al.*, ARI 68 (2010) 1340-1343], in which P-10 gas (argon 90%+methane 10%) is used as the counting gas. The response of the IC as filled with P-10 gas can be easily done by circulating P-10 gas mixed with radioactive gas (e.g.: ^{85}Kr) through a gas-loop in which the differential gas counters are connected to the IC in series. In order to obtain the chamber response for air as ionizing gas, however, the response-ratio of the IC with dry air to with P-10 gas should be considered.

The chamber response is dependent on the W-value and mass-density of filling gas and other conditions. To make experimental determination of the response-ratio, we firstly prepared several small vessels whose volumes are the same (10 cm^3) and less than 1/100 of the chamber volume, and filled all the vessels with same ^{85}Kr-gas by circulating the Kr-gas after connecting all small vessels and ^{85}Kr gas reservoir in series. The activity concentration of ^{85}Kr-gas was approximately 10^4 Bq cm^{-3}. After filling ^{85}Kr gas to every small vessel with same size, the valves of both sides of each vessel were closed, and thereby we obtain several same ^{85}Kr gas-sources. Radioactivity containing in the each vessel is enough to operate the ionization chamber. One of the reference gas sources thus prepared was connected to ionization chamber filled with pure P-10 gas in series, and mixed with ^{85}Kr gas by circulating the gases about 20 min. The output current of the IC was measured with PC-controlled precision pico-ammeter after the output retained constant. Next, chamber gas was changed to dray air, and similar measurements were performed using another fresh ^{85}Kr source. Prior to the second measurement, it is important to confirm the memory effect is not significant by checking background. The ratio of output currents with dry air and with P-10 gas gives directly the response ratio. In case of 1.5 L IC designed for gas monitor (Ohkura Corp. Type I-4096), which are most commonly used in Japan for this purpose, the result of the response ratio with dry air to with P-10 gas was determined to be 0.688 0.003. Similar technique can be applied to another gaseous nuclides such as ^{133}Xe, ^{41}Ar etc. and any size ICs for gas monitoring purpose. The content of krypton gas is very small in volume (1/150 in mol.) as compared with the chamber gases (dry air, P-10 gas), but we have discussed the possible effects due to the Penning ionization, and it was confirmed such phenomenon does not give appreciable change of the results. As a supplemental experiment, we measured the ratio of output currents causing external irradiation of gamma-rays from a ^{226}Ra sealed source, and the response-ratio was 0.681 which was very near to the results obtained with the present study. Similar technique can be applied to another gaseous radionuclides and to any chamber with different size.

Corresponding author's email address: **a.yunoki@aist.go.jp**

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DEVELOPMENT OF THE INTERNAL GAS PROPORTIONAL COUNTING SYSTEM OF THE BETA RADIOACTIVE GAS

120

Wang, S. H.; Lee, B. J. M.; Lee, K. B.; Park, T. S.

KRISS (KR)

A length-compensated internal gas counting system has been developed to measure the absolute radioactivity of the novel gases at NPL, PTB, LNHB and NIST. Recently, we are developing a new primary system of the beta radioactive gas by the length-compensated internal gas counting to be used as the standardization of the radioactive novel gases at Korea Research Institute of Standards and Science (KRISS).
The proportional counting system consists of three stainless tubes of 25 mm inner diameter and 26.5 mm outer diameter with different length (150 mm, 200 mm and 350 mm), respectively. 15 μm thick gold coated tungsten wires are located in the center of the tubes to collect the ionized electron by the radioactive gases. The tubes are placed in airtight chamber to be kept the vacuum condition and filled the ionizing gas and the radiative gas. The pressure reaches 0.6 Torr within 5 minutes operation of the vacuum pumps, which consists the dry pump and turbo pump.
The geant4 simulation with the realistic design was performed to evaluate the systematic uncertainty. In addition, the commissioning operation is performing. In this presentation, the new system of the internal gas proportional counting system and the properties of the new system of KRISS will be discussed.

Corresponding author's email address:

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FAST DIGITAL 4πβ-4πγ COINCIDENCE COUNTING WITH OFFLINE ANALYSIS AT IRA-METAS

080

Durán, M. T.; Nedjadi, Y.; Juget, F.; Bochud, F.; Bailat, C.

IR (CH)

IRA-METAS launched recently a project to digitize all the data acquisition systems it uses for primary radionuclide standardizations. It is well-known that the digital approach presents numerous advantages over the traditional analog electronics such as information lossless, scalability, on-line and/or off-line data processing, and it is also a solution to the growing difficulties to renew or repair ageing modules.
As a first step in this wider program, our institute set-up a 4πβ-4πγ digital coincidence counting system, with FPGA-based commercial boards from National Instruments (NI) to perform data acquisition and off-line data analysis. Choosing all components and software from the same supplier provides a full compact and consistent electronic system.
The detection unit consists of plastic scintillators optically coupled to a low-noise photomultiplier tube, which are housed within a 5"x5" NaI(Tl) well detector. The charge-sensitive pre-amplified pulse trains from the two detectors are digitized through a high speed double input digitizer adapter module linked to a Kintex-7 FPGA. Raw-data acquisitions were performed at 100MS/s with a 12 bit resolution though they can be carried out at 800 MS/s maximum sampling rate. Sample voltages above a selectable threshold and a 10 ns resolution time stamp are acquired for each event. A timing and synchronization control card with high precision and stability oscillator ensures triggering, timing and operating synchronicity. The modules together with the controller PC are connected to a high-bandwidth chassis. Data are streamed to an external hard-drive RAID (redundant array of inexpensive disks) where they are stored. Data can be transferred at up to 8 GB/s but the storage in the RAID is limited to 3.6 GB/s.
Off-line analyses routines were developed to process the pulse trains. Customized digital filters are used to shape the signals, eliminate noise and extract time and pulse amplitude information in both beta and gamma channels. Energy and time spectroscopy, and the efficiency extrapolation method are performed using an additional code.
To demonstrate and validate the capacity of this system to standardize the activity of radioisotopes, we compare its predictions for the activities of ^{18}F, ^{133}Ba and ^{166m}Ho solutions with the results of coincidence counting systems with analog electronics, as well as with the activities measured using other primary standardization methods.

Corresponding author's email address: **teresa.duran@chuv.ch**

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STANDARDIZATION OF ^{67}Ga BY DIGITAL COINCIDENCE COUNTING

Balpardo, C.[1]; Arenillas, P.[1]; Roteta, M.[2]

[1] CNEA(AR); [2] CIEMAT(ES)

The nuclide ^{67}Ga is widely used in Nuclear Medicine for diagnostic purposes. The extensive use of this nuclide in nuclear medicine is due to the short half-life and the energies of its gamma rays It decays by electron capture with a half-life of 3.259 days to ^{67}Zn followed by gamma de-excitation. The 93 keV excited level has a half-life of 9.1 μs, which makes its absolute standardization difficult. Digital Coincidence Counting is a very useful tool applied to standardize by different techniques: This paper discusses the standardization of ^{67}Ga by DCC method in different ways like: 4πβ-γ(NaI) coincidence counting with efficiency extrapolation to infinite dead time, 4πβ-γ(NaI) coincidence with two-dimensional extrapolation, 4πβ-γ(NaI) anticoincidence counting with extended dead time and 4πγ measurements and compares the results and their uncertainties. Especially relevant is the discussion about the type of dead time to be imposed on the system We analyzed the results of extrapolation to infinite fixed dead time with those obtained by extensible dead time and the convenience of using one or other kind of dead time depending on the activity is discussed. Pressurized proportional counter has been used as electron detector. In addition, the half-life of the 93 keV excited level has been measured using two methods: Time spectra between photons of 300 and 93 keV and Time spectra between photons of 300 keV and conversion electrons.

Corresponding author's email address: **balpardo@cae.cnea.gov.ar**

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A NEW DIGITAL 4πγ SYSTEM WITH A NaI WELL-TYPE DETECTOR AT LMR-CNEA

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Rossi, M.; Balpardo, C.; Arenillas, P.; Cerutti, G.; Ferrari, M. C.

CNEA(AR)

A 4πγ integral counting system with a NaI(Tl) well-type detector and a digital interface to take measurement data was implemented at the Radioisotope Metrology Laboratory (LMR-CNEA). In the last years our laboratory has been adapting its primary systems to new digital standards. We use a NIM module with a FPGA from ULS to digitize the signal from the detector. The 4πγ system is very useful for radionuclides with complex decay schemes and with many gamma and X rays emissions. Each energy efficiency was computed by Monte Carlo simulations and the total efficiency was calculated considering all the decay branches. A code was developed to analyse data, this program take the files generated by the digitizer module, imposes and correct for dead time and calculate sources activities and its uncertainties. This primary measurement method has many advantages, accepts different sources geometries, short measurements times making it ideal for use with radioisotopes in medical applications. Three radionuclides were measured, ^{113}Sn, ^{192}Ir and ^{131}I, and compare with gamma ray spectrometry. The Iodine's result was used to calibrate the LMR's activimeter, this device is used to calibrate the equipment of Argentine hospitals.

Corresponding author's email address: **balpardo@cae.cnea.gov.ar**

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REVISITING THE STANDARDISATION OF ^{134}CS: UPDATED IONISATION CHAMBER CALIBRATION FACTORS AND NEW MEASURMENTS OF ABSOLUTE γ-RAY EMISSION INTENSITIES.

095

Collins, S. M.; Keightley, J. D.

NPL (UK)

Caesium-134 is a widely used radionuclide for the full-energy peak detection efficiency calibration of γ-ray spectrometers due to its 2.0652 a half-life and the numerous intense γ-rays emitted following β- decay to ^{133}Ba. Standards of ^{134}Cs are routinely provided by NPL using our secondary standard ionization chamber (SSIC) systems, via calibration factors derived directly from primary standards performed as part of the CCRI(II)-K2.Cs-134 comparison exercise [1].
Recent work on updating the photon and electron response curves for the NPL SSIC systems (using the SIRIC software package [2]) revealed a discrepancy of the order of 0.7 % between the new "SIRIC-predicted" ^{134}Cs calibration factors and those historically employed for ^{134}Cs. In order to investigate the matter, a series of LS vials were prepared from a stock solution of a radio-chemically pure ^{134}Cs solution, and a primary standardization was performed using the NPL 4π(LS)-γ(HPGe) digital coincidence counting (DCC) system. A series of ampoules were also prepared using the absolutely standardized ^{134}Cs solution for assay on the NPL SSIC and HPGe γ-spectrometry systems respectively.
The 4π(LS)-γ(HPGe) coincidence counting efficiency-extrapolations were performed using three γ-gate settings: single gates on the 604 keV and 795 keV γ-ray emissions respectively, as well as a simultaneous multi-dimensional extrapolation on both gates, with excellent agreement between the resulting activity concentrations, yielding an activity concentration of 330.3(10) kBq.g^{-1} (k=1) at the reference time. Calibration factors for the NPL SSIC systems were derived from this newly standardized material, revealing a 0.7% bias from the historic calibration factors (which yielded a result of 332.7(23) kBq.g-1 (k=1)).
Measurements were performed on the HPGe γ-ray spectrometer previously calibrated for its full-energy peak detection efficiency with a suite of more than 20 other radionuclides directly traceable to NPL primary standards (NPL did not include ^{134}Cs in the determination of the HPGe efficiency calibration curve). The activity concentration determined from the most intense γ-rays (using emission intensities from the most recent evaluation from the DDEP) were on average 0.69 % lower than the activity determined from the historic SSIC calibration factors.
Updated calibration factors for the NPL SSIC systems (FIDELIS, ISOCAL IV) are reported, as are new measurements of the absolute γ-ray emission intensities for ^{134}Cs, along with full uncertainty budgets.
It is intended that this manuscript serves as a reminder for NMIs to not only concentrate on "novel" or "emerging" radionuclides, but to continuously question and revisit existing calibrations and standardizations for more commonly used radionuclides.

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Corresponding author's email address: **sean.collins@npl.co.uk**

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ABSOLUTE STANDARDIZATION OF ^{54}Mn BY SUM-PEAK METHOD USING HPGE DETECTORS

098

de Araújo, M.T.F.[1,2]; Poledna, R.[1]; Delgado, J. U.[1]; de Paula, E.; Leiras, A.; Lopes, R. T; Ferreira Filho, A.L.[1,2]; da Silva, R. L.[1]; de Almeida, M. C. M.[1]; De Oliveira, A. E.[1], de Veras, E. V.[1], Rangel, J.[1]

[1] LNMRI/IRD/CNEN–RJ-(BR), [2] COPPE/UFRJ-RJ-(BR)

IThe ^{54}Mn is a radionuclide that decays by electron capture to the intermediate level of 834, 85 keV with a 99.97% of the emission probability and later decays to ^{54}Cr. It has a half-life ($T_{1/2}$) of 312.19 days, which facilitates laboratory work such as verification of system stability, or even making efficiency curves for detectors. In this way, the LNMRI aims to use the ^{54}Mn radionuclide as standard work considering that the energy of the main range is a useful critical region for the activities developed in the laboratory. Many international literatures make use of 54Mn properties in their analyzes such as half-life measure, absolute standardization by 4π (PC)-γ coincidence, decay data analysis among others. Due to present a simplified decay scheme and to provide a discrete range of 834 keV, the absolute sum-peak method can be applied considering the sum of 5 keV x-ray with 834.55 keV gamma ray enabling to determine the activity for the radionuclide of interest.

The absolute determination by the sum-peak method is necessary because it enables rapid determination of decay parameters without the requirement of an electronic and very elaborate experimental apparatus. They were used two different semiconductor detectors with distincts efficiencies. The results obtained were compared with other absolute methods showing consistent activities values with uncertainties below 1%. Although the ^{54}Mn has been standardized by other absolute methods according to the available literature, it is the first time that the absolute standardization was performed using the sum-peak method.

The comparison criterion, the ^{54}Mn activity value was also determined using the efficiency curve relative method and it was made using some gamma standards available in LNMRI whose energy ranges are applicable to this work. They were obtained good results with uncertainties around 2%. In general, both methods have proved quite adequate for determination of the ^{54}Mn radioisotope activity, contributing to the improvement of Laboratory measurements obtaining lower uncertainties.

Corresponding author's email address: **miriamtaina@hotmail.com**

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STANDARDIZATION OF ^{133}BA BY SUM-PEAK METHOD

da Silva, R. L.[1]; de Almeida, M. C. M.[2]; Delgado, J. U.[1]; Poledna, R.[1]; Da Silva, C. J.[1]; Cruz, P. A. L.[1]; Iwahara, A.[1]; de Oliveira, A. E.[1]; de Araújo, M.T.F.; Trindade, O. L.[1]; de Veras, E. V.[1]; Rangel, J.[1]

[1] IRD / CNEN-RJ (BR), [2] CNEN-RJ (BR)

The National Laboratory of Ionizing Radiation Metrology (LNMRI/IRD/CNEN) is responsible for the custody and dissemination of radioactive standards throughout the country. The LMNRI provides radioactive reference standards with various geometries with activity values and their associated uncertainties that meet the needs of end users. Several absolute and relative calibration methods are used routinely. The advantages of calibrations by absolute methods are: they do not require standards, the precision and the uncertainties are small. As ^{133}Ba has half-life ($T_{1/2}$) of 10.54 years and emits multiple gamma rays is usually used in the detectors calibration in research laboratories. It decays 100% by electron capture and emits about 14 lines of gamma and X-rays, generating several coincidences between them. ^{133}Ba is still used as surrogate for ^{131}I for checking the calibration number of radionuclide calibrators used in nuclear medicine despite this procedure is metrologically not recommended.

The Committee Consultatif des Rayonnments Ionisants, of the Bureau International des Poids et Mesures (CCRI/BIPM) presented by CCRI II / 11-10 document described radionuclide measurement methods with their estimated respective uncertainties. There are three methods of standardization for ^{133}Ba in this document of which two methods are considered difficult. In the late 60's some researchers described an absolute calibration method for the activity, known as sum-peak method. This method enables direct standardization of radionuclide activity that emits two photons in coincidence.

The purpose of this paper is the ^{133}Ba standardization by peak-sum method using an HPGe germanium detector and point sources of low activities with the subsequent evaluation of their X and gamma probabilities. For this calibration were used the coincidence of characteristic X-rays (Kα1) with some gamma rays coming from its decay. The result of ^{133}Ba activity obtained by sum-peak was compared to those obtained by anticoincidence and liquid scintillation CIEMAT/NIST methods. The final results produced consistent values, within a standard uncertainty, and uncertainties lower than 0.30% (k = 1) showing that this sum-peak method can be used with reliability for direct calibration of multi-gamma radionuclides

Corresponding author's email address: **marcandida@yahoo.com.br**

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APPLICATION OF THE SUM-PEAK METHOD TO ACTIVITY STANDARDIZATIONS OF ^{152}EU SOURCES IN LNMRI(BR)

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Ferreira Filho, A.[1,2]; da Silva, R. [1]; Tadeu Lopes, R. [2] *et al.*

[1] LNMRI/IRD(BR) [2] COPPE/UFRJ(BR)

Activity determination by γ-ray spectrometry is a valuable aid which is widely used where radioactive sources are applied. The sum-peak method is an absolute measurement technique that uses coincidence counting and γ-ray spectrometry. Coincidence counting itself has been used as an absolute method by composing the detection system of pair detectors. The pair detectors register β – γ, γ – γ, X – (X, γ), etc in coincidence and the radioactivity of an unknown source is determined using the counts in coincidence or anticoincidence between the detector signals. The sum-peak method is based on the same principle as coincidence counting, but requires only one γ-ray detector with spectroscopy electronics. It is a primary activity standardization procedure for radionuclides emitting more than one photon in coincidence. It is based on the fact that when two photons, temporarily coincidents, interact with a detector, giving each one its energy to the detector, the detector response is proportional to the total deposited energy. The merit of the sum-peak method lies in the simplicity of both the technique and the detection system. The sum-peak method requires a single detector with spectroscopy electronics, while the coincidence counting method requires two or more detectors with timing modules. The sum-peak method also has an advantage when compared with the relative measurement technique using γ-ray spectroscopy, which is frequently used to determine the radioactivity of an unknown source. The relative measurement requires a precise energy-efficiency calibration of the spectroscopy system. Hence, it is influenced by the experimental conditions of the source-to-detector geometry, the γ-ray energy, and the source radioactivity. The method normally requires the knowledge of the total count rate as well as the peak count rates and the sum peak count rate. Most commonly, it is applied to point sources measured in a close geometry, which improves the statistics of the sum-peak area. In this study, the sum-peak counting method was applied for the absolute activity standardization of five ^{152}Eu sources, using GeHP detector. The ^{152}Eu 13-year very complex decay scheme, with two branches of decay, disintegrates into ^{152}Sm by electron capture (72,1%) and positron emission (0,027%), and into ^{152}Gd by beta minus emission (27,9%). The daughter nucleus ^{152}Sm has 19 excited levels as the ^{152}Gd has 15. About 132 γ-ray transitions are known to follow the decay, with intensities above 10^{-5} photons per parent decay. The particular interest in this radionuclide is explained by its wide use for energy and efficiency calibration of semiconductor gamma spectrometers since its energy range of the emitted photons is broad, that is, 40–1769 keV, making this radionuclide suitable for the aforementioned purpose. The branch of decay used here was that of Sm^{152}, as it is practically all by electron capture and show the characteristic x-rays, $k_{\alpha 1}$, $k_{\alpha 2}$, with energy of 39,5 keV and 40,1 keV. In the photon spectrometry, these two x-ray, with so close energies, appear as a single peak. And so they have a high probability of coincidence with a γ-ray in all the energy range of the ^{152}Sm decay. The coincidence between this X-ray peak and the 121 keV gamma ray, γ_{1-0} (Sm), were used. The areas of the peaks of energy and coincidences were determined using the deconvolution program COLEGRAN developed by LNHB. The result was compared with the calibration performed by ionization chamber traceable to BIPM. The results presented show uncertainty values below 0.30%.

Corresponding author's email address: **alfredo@ird.gov.br**

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METROLOGY FOR DECOMMISSIONING NUCLEAR FACILITIES PARTIAL OUTCOMES OF JOINT RESEARCH PROJECT WITHIN THE EUROPEAN METROLOGY RESEARCH PROGRAM

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Jiří Šuráň *et al*

CMI(CZ), JRC(EC), LNE(FR), NPL(UK), PTB(DE), SCK•CEN(BE), STUK(FI), ANDRA(FR), EDF(FR), NUVIA(CZ)

The nuclear industry in Europe is facing an immediate and major challenge with the decommissioning of old nuclear facilities and the planned shut downs which will incur enormous costs regarding the accurate characterisation and disposal of the decommissioned materials.
Therefore, there is a need for the implementation of new and traceable measurement technologies and techniques to select the appropriate release or disposal route of radioactive wastes which potentially could reduce the costs during the decommissioning process.
This paper addresses some of the innovative outcomes of the project "Metrology for Decommissioning Nuclear Facilities" funded by the European Metrology Research Program, as shown in the list below.

- Dissolution procedures for graphite matrices, to enable radiochemists to validate measurement techniques for the mapping of contamination inside nuclear facilities
- Spectral characteristics of air radioluminescence investigation for remote mapping of alpha radiation inside nuclear facilities
- Improved contamination depth distribution mapping of nuclear facilities structures by integration of data from gamma spectroscopy, surface beta and dose rate measurements by progressive geostatistical modelling techniques
- Validation of Monte Carlo detection efficiency transfer for waste pre-selection and free release measurement system characterized by unique, patented lead-free shielding, and improved throughput, accuracy, reliability, modularity, and mobility
- Facilities and methods for the metrological characterization of Raman distributed temperature sensing systems for long term repository sites integrity monitoring
- Production of blank reference materials of concrete, steel and graphite together with a series of standard sources, which will be combined for the validation of radiochemical procedures

The objective of the paper is to demonstrate improved parameters and expected impact.

Corresponding author's email address: **jsuran@cmi.cz**

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DEVELOPMENT OF THE ABSOLUTE STANDARDIZATION APPARATUS FOR RADON-222 ACTIVITY

038

Liang, J. C.; Wang, L. Y.; Li, Z. X.

NIM (CN); SNST/USC (CN)

Radon and its progenies lead to more than 50% of natural radiation dose, so national metrology institutes have been developing absolute measuring apparatus to provide primary standardization for calibration utilization especially in the radiation protection field. Several methods including internal gas counting, defined angle method and liquid scintillation method etc. has been used to realize the precise standardization of radon radioactivity. At present, from the view of accuracy, the defined angle method is undoubtedly the most precise one in all candidate methods. So, several national institutes including LNHB, PTM, IRA-METAS and KRISS etc. have developed their primary system with this kind of method.
In order to meet the traceability requirements of radon radioactivity, an apparatus based on the defined solid angle method has also been established in the national institute of metrology, China. Two home-made vacuum sealed radium-226 sources with radioactivity of 3MBq and 0.5MBq are used to provide high-purity radon gas to minimize the impact from gas impurities. The distance of source to diaphragm and the radius of diaphragm are separately measured by the precise Four Coordinate Measuring Machine and a laser tracking interferometer measuring system. The diameters of the condensed radon sources are evaluated by the cyclone storage phosphor equipment with the autoradiography method, the temperature on the cold finger is adjusted from 35K to 80K, the radioactivity condensed vary from 50kBq to 2.8MBq and the result shows the diameter to be (4.1±0.8) mm. The defined solid angle is calculated depending on precisely measured geometrical dimensions by several algorithms including Monte Carlo simulation and the results are equivalent in uncertainty. Alpha particle's scattering behavior in the measuring chamber has also been simulated by geant4 Code and the preliminary result shows a total scattering alpha particle ratio up to 0.27% at the edge of diaphragm when the geometry factor of $1.101(1)\times10^{-3}$ is used. As a result, a combined standard uncertainty of less than 0.5% for radon radioactivity primary measurement is achieved in NIM's apparatus.

Corresponding author's email address: **liangjc@nim.ac.cn**

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ACTIVITY MEASUREMENT OF ^{222}RN GAS FOR A KEY COMPARISON

Kima, B.J.[2]; Lee, J.M.[1,2]; Lee, K.B.[1,2]; Park, T.S.[1]; Kim, B.C.[1]

[1] KRISS (KR); [2] UST (KR)

The Korea Research Institute of Standards and Science (KRISS) developed a primary measurement standard for radioactivity of gaseous radon in 2012. The primary measurement of ^{222}Rn activity is based on the defined solid angle counting method. This method condenses ^{222}Rn gas molecules at a cold point and uses the alpha energy spectroscopy to determine the activity. In 2015, we have participated in the key comparison (KC) of activity measurements of ^{222}Rn, termed as CCRI(II)-K2. Rn-222, for proving the international equivalence of the ^{222}Rn measurement. This paper describes the measurement procedure that we employed, and measurement results that we obtained with the KC source. We also include in this paper a detailed evaluation of the measurement uncertainty of the activity of ^{222}Rn gas.

For a preliminary study we received an empty container for KC from the LNHB, the KC pilot laboratory, in order to modify the input part of the primary system and calibrate a Ge-detector. The ^{222}Rn gas flown into the main chamber of the primary system from a ^{226}Ra powder source was measured by a Si-detector to obtain thirty sequential energy spectra of alpha particles from the condensed radon. The radon gas was then transferred into the empty container connected to the primary system and was used for the calibration of the Ge-detector. We obtained calibration factors at six representative gamma-ray energies from the radon decay products at radioactive equilibrium using the primary measurement results.

Immediately after receiving the real KC source distributed by the LNHB, we measured the source activity using the Ge-detector calibrated at the preliminary study and then connected the source to the primary measurement system. The primary measurement was carried out in an identical way as in the preliminary study after the radon gas was transferred into the main chamber of the primary system. The residual radon gas in the source container after the transfer was measured to determine its activity with the Ge-detector. The main sources of measurement uncertainty were considered the count rate (0.05%), the geometry factor (0.1%), the reproducibility of our primary measurement system (0.1%) and the spectrum analysis (0.07%). The uncertainty due to the residual radon gas in the container was negligible. The relative combined uncertainty was therefore estimated 0.17%.

Corresponding author's email address: **jmlee@kriss.re.kr**

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THE LARGE-AREA MULTI-WIRE PROPORTIONAL COUNTER FOR 2PI ALPHA AND BETA EMISSION MEASUREMENT AT THE NIM CHINA

001

Zhang, M.[1]; Zhang, J. W.[2]; Duan, L. M.[2], Tu, Z.[3]; Yang, Z. J.[1]; Liang, J. C.[1]; Liu, H. R.[1]

[1]NIM (CN); [2]USTC (CN); [3]IMP/CAS (CN)

We established an alpha/beta emission rate counting system at the National Institute of Metrology (NIM, China), which comprises of a large-area windowless multi-wire proportional counter and a custom-built digital pulse processing module, to realize the dissemination of the emission rate values to the surface contamination monitors.

The counter is designed to have an effective detection area of 340 mm 240 mm, and allow collecting data at pressures up to 0.2 MPa by introducing four clamps at each corner of the chamber. The anode gold-plated tungsten wires are 20 um in diameter, and are in array separated by 25 mm. A custom-built signal processing system has been developed to replace the conventional electronic units, which consists of a FPGA based acquisition card with a 16-bit resolution at a sampling rate of 100 MSPS combining with a pulse height analysis routine.

The performances of the detector, including the high-voltage plateaus for alpha/beta source measurement and the homogeneity of response over the whole chamber, were measured. The measured high-voltage plateau slopes are 0.30% and 0.35% over a range of 100 V, and the plateau lengths are 900 V and 600 V, for an Am-241 alpha emitter and Sr-90/Y-90 beta emitter, respectively. The measured count rates for a small Pu-239 alpha emitter showed that the results at different positions are constant within 0.3%, expect for those performed at the outer edges, and a good homogeneity of response over the whole chamber has been achieved.

In order to validate the measurement ability of the counter, a series of certificated Am-241 and Sr-90/Y-90 sources, which are direct traceable to PTB through the DAKKS and comply with the requirements for traceability to NIST with dimensions of 150 mm 100 mm, were measured. By taking into account of the influence of background subtraction, threshold setting and dead time corrections, the percent differences between the measured results and the given certificated emission rate values show good agreements within the uncertainty range. Further, the same sources were also measured at pressures of 0.5 and 1.5 times of the atmospheric pressure. This is no significant dependence of the measured emission rate on the operating pressures for both alpha and beta measurements found within the uncertainties.

A digital pulse processing module has been developed to pick up the shape of the pulse, using a FPGA based acquisition card with a 16-bit resolution at a sampling rate of 100 MSPS considering the pulse widths (several micro seconds) from gas detector after the shaping amplifier. The stored data are handled and processed by a pulse height analysis routine, where the reproduced pulse is visible, and the non-extending dead time algorithm is taken in the data analysis program. Further, the energy spectrum can be obtained, and threshold extrapolation to low energy counting for beta measurement is easily realized by changing the threshold setting in the software. The digital pulse processing module in our laboratory has been proved by comparing with the results using the conventional electronic units.

A brief introduction on the alpha/beta emission rate counting system at the NIM China is presented above, including the design of the detector and the data acquisition system, preliminary results on the performance measurements and emission rates for a series of certificated large-area alpha and beta emitters. The system still needs to be validated by participation at international comparisons with the other NMIs or DI in the frame work of CCRI or RMOs.

Corresponding author's email address: **zhming@nim.ac.cn**

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EFFICIENCY TRANSFER METHOD APPLIED TO SURFACE BETA CONTAMINATION MEASUREMENTS

015

Stanga, D.[1]; De Felice, P.[2]; Capogni, M.[2]

[1] IFIN-HH (RO), [2] INMRI, ENEA (IT)

The new edition of the international standard ISO 7503 (2016) provides a methodology for calibrating contamination monitors but it does not provide a methodology for estimating the activity per unit area of contamination sources and its standard uncertainty. It stipulates that the direct measurement of the surface contamination is difficult because it is difficult to estimate the efficiency of actual contamination sources and its standard uncertainty. However, it describes a conservative approach for evaluating the efficiency of contamination sources but this approach has not a strong theoretical basis.

In this paper, the efficiency transfer method is applied to the direct measurement of surface beta contamination using contamination monitors. The method of detection efficiency transfer is based on the following equation $\varepsilon_a^{(calc)} = T(a/calib)\,\varepsilon_{calib}^{(exp)}$ where $\varepsilon_a^{(calc)}$ is the calculated value of the detection efficiency in the measurement of actual contamination sources, $\varepsilon_{calib}^{(exp)}$ is the detection efficiency measured in the calibration of contamination monitors and *T(a/calib)* is the efficiency transfer factor from the calibration measurement (index *calib*) to the actual measurement (index *a*) of the contamination source. *T(a/calib)* is defined by the ratio of the detection efficiencies ($T(a/calib) = \varepsilon_a^{(model)}/\varepsilon_{calib}^{(model)}$) computed on the basis of a model developed in this work for calculating the detection efficiency for beta radiation. This model is based on previous results obtained in numerical modeling of electron transport in planar geometry. According to this model, the detection efficiency, ε_{det}, is given by

$$\varepsilon_{det} = f_c p_\beta \int_0^{x_{max}} \varepsilon_p(x + s_{air} + s_w) f(x) dx$$

where pβ is the probability of detecting and recording of beta particles reaching the sensible volume of the detector, *f(x)* and x_{max} are the activity depth distribution and the thickness of the active layer of the source, sair is thickness in mg/cm2 of the air gap between source and detector window, s_w is the thickness of the detector window and fc is a correction factor. By means of Eq. (2), the efficiency transfer factor can be calculated and used for determining the activity per unit area, ΛS, of the contamination source. Thus, we have $\Lambda_S = (R - B)/[S(T\,\varepsilon_{calib}^{(exp)} + \varepsilon_g)]$ where *R* is the counting rate provided by the contamination monitor in the measurement of the contamination source, *B* is the background, *S* is the surface of the detector window and ε_g represents the detection efficiency for gamma radiation.

The efficiency transfer method was applied to the measurement of two ^{60}Co and two ^{137}Cs sources (with known activities measured by gamma spectrometry) which mimic contamination sources encountered in practice (especially in nuclear industry). The activity per unit area of the sources was estimated by means of the contamination monitor FH 40G provided with the contamination probe FHZ 742 (plastic scintillation detector with window area of 10x12.5 cm^2). In the first step, the detection efficiency of the instrument for beta (including conversion electrons) and gamma radiation were determined using calibration sources of ^{60}Co and ^{137}Cs constructed from aluminized aluminum foils. In the second step, *R* and *B* were measured by means of FH 40G contamination monitor. After estimating the efficiency transfer factor, the activity of the four sources was calculated using the formula mentioned above. A Matlab script was written for computing both Λ*S* and its standard uncertainty. The measurement results obtained by means of the efficiency transfer method agree within one standard uncertainty with the measurement results obtained by gamma spectrometry. This validates the efficiency transfer method applied to surface beta contamination measurements.

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Corresponding author's email address: **doru@nipne.ro**

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LARGE AREA ALPHA SOURCES WITH A LIP: INTEGRAL COUNTING AND SPECTRAL DISTORTIONS

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Fitzgerald, R.; King, L.

NIST (US)

Calibration of large area sources for surface emission of alpha and beta particles is essential for measuring performance of deployed instruments used in radiological protection, as well as to provide traceability for calibration sources. Predominantly, such calibration sources are nominally flat and either circular or rectangular in shape, with radii or sides of a few centimeters and thicknesses of a few millimeters. The NIST large area proportional counters that are used for calibrations were themselves characterized, and are periodically validated, using sources of similar, flat, geometry, making these detectors appropriate for calibrating flat sources. However, more recent sources that have been sent to NIST for calibration sometimes take the form of a circular disc having a raised lip around the edge of several millimeters both in height and radius. Alpha emission measurements of these sources produce energy spectra that differ qualitatively from corresponding spectra resulting from measurements of flat surfaces. The object of this work was to devise a method by which the correct alpha emission rate could be recovered from the distorted spectra.

This work was carried out both experimentally and through Monte Carlo simulation. The experimental approach was to use flat sources of Pu-239 to obtain multiple spectra, analyzed using our standard spectral extrapolation-integration method, averaged as the nominal truth value. Aluminum rings were constructed and stacked on top of the source such that the assembled geometry would approximate sources with a lip.

Spectra obtained from the ring-topped sources showed the same qualitative spectral distortions as true lipped sources that had been received for calibration. Namely, the resulting spectra displayed an order-of-magnitude enhancement in low-energy region, new low-energy structure, and also a peak shift to higher energy (larger pulse-height). Two extrapolation methods were attempted to recover the true emission rate. The first method, similar to our usual method, consistently over-estimated the emission rate. However, using a short extrapolation to the origin proved satisfactory, with biases of +0.12 % and – 0.40 % for 5 mm and 10-mm lips respectively. The uncertainty was 0.7 % for the 10 mm lip.

Monte Carlo simulations of the radiation transport using Geant4, coupled with modelling of the electric field distortions caused by the lip revealed the causes of both the low-energy structure and positive offset of the alpha peak resulting from the lips. The former is caused by interactions of the alphas with the lip walls. The origin of the increased peak pulse height was found to be distortions in the electric field, causing an enhancement in the electron multiplication within the counter. As a result of this study, we can explain numerous aspects of the distorted spectra, and confidently calibrate large area alpha sources with a lip.

Corresponding author's email address: **ryan.fitzgerald@nist.gov**

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COMPARISON OF ^{131}I ACTIVITY MEASUREMENTS AT THE NCBJ RC POLATOM AND THE ENEA-INMRI LINKED TO THE BIPM SIR SYSTEM

028

Ziemek, T.[1]; Capogni, M.[2]; Ratel, G.[3]; Broda, R.[1]; Dziel, T.[1]; Fazio, A.[2]; Listkowska, A[1]; Kołakowska, E.[1]

[1]NCBJ RC POLATOM (PL); [2]ENEA-INMRI (IT); [3]BIPM (FR)

Radioactive iodine ^{131}I is largely diffused in Nuclear Medicine for the treatment of an overactive thyroid in a so called hyperthyroidism disease. Due to both the β^- and γ components in its decay scheme, ^{131}I is very useful both for therapy and diagnostic. Its wide application in SPECT QI requires a more precise and accurate activity standard of this radionuclide.
In order to face to the increasing request coming from Nuclear Medicine departments of own country for calibrating instruments devoted to the ^{131}I activity measurements and provide a robust traceability in this kind of measurements, a bilateral comparison between the Italian National Institute of Ionizing Radiation Metrology, belonging to ENEA (Italy), ENEA-INMRI, and the National Centre for Nuclear Research Radioisotope Centre POLATOM (Poland) on activity measurements of a solution of ^{131}I was organized in the year 2015 and piloted by POLATOM. The aim was to link both the ENEA-INMRI and POLATOM to the BIPM International Reference System (SIR) as none of the laboratories did participated earlier in the ^{131}I activity comparison with BIPM.
A master ^{131}I solution was prepared at POLATOM with sufficient activity per gram for measurements at ENEA-INMRI and at BIPM. The solution was diluted and a set of twelve sources in Ultima Gold scintillator was prepared. Sources were measured at POLATOM by the 4π(LS)-γ coincidence and anticoincidence method in two various counters developed in-house. The first one with two PM-tubes in beta channel and two NaI(Tl) detectors in gamma channel was equipped with an analogue electronics modules. The second one with a TDCR system and a NaI(Tl) detector in gamma channel was equipped with the FPGA XILINX Spartan-3AN digital board. Sources were measured also by the CIEMAT/NIST method in the Tri-Carb 2810 TR scintillation counter.
Three ready-to-measure sources were sent to ENEA-INMRI and measured using the 4π(LS)-γ coincidence method in a system with a triple LS-detector and a NaI(Tl) detector in gamma channel, that was equipped with the CAEN Desktop digitizer DT5720. The sources were measured also using the TDCR method in a Hidex 300SL 'Metro' version counter and a home-made portable TDCR counter developed at ENEA-INMRI. An ampoule with the master ^{131}I solution was also sent to ENEA-INMRI exclusively for measurement in two calibrated ionization chambers.
All results obtained by both laboratories were in excellent agreement. The degree of equivalence between two laboratories was $D_{ij} = A_i - A_j = 0.11$ kBq/g with its expanded uncertainty Uij = 2.55 kBq/g (k = 2).
One glass SIR ampoule contained the 131I master solution was sent by POLATOM to the BIPM SIR. The determined SIR equivalent activity shows a reasonably good agreement with the key comparison reference value (KCRV). In order to enable the entry of the ENEA-INMRI result in the SIR database, through the measurements of the ampoule sent by the POLATOM, the bilateral comparison was registered as a EURAMET.RI(II)-K2.I-131 key-comparison.

Corresponding author's email address: **tomasz.ziemek@polatom.pl; marco.capogni@enea.it**

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VALIDATION OF THE EXTENSION OF THE SIR TO BETA EMITTERS

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Ratel, G.[1]; Los Arcos, J. M.[1]; Laureano-Perez, L.[2]; Rodríguez-Barquero, L.[1]; Courte, S.[1]

[1]BIPM (FR); [2]NIST (US)

The pilot comparison of only ^{63}Ni solutions initiated by the Working Group "Extension of the Système International de Référence (SIR) to β and α emitters" (ESWG(II)) of the Comité Consultatif des Rayonnements Ionisants CCRI(II), to test the use of liquid-scintillation counting systems available at the BIPM, to extend the scope of the SIR towards pure β-emitters has shown promising results. These preliminary results were already presented at the ICRM 2013 conference in Antwerp.
However, some discrepancies were detected at that time, for which the Working Group judged necessary to undertake additional investigation. To this purpose and to understand the universality of the method to be used it was decided to redo measurements of ^{63}Ni solutions and to extend the study to ^{3}H, ^{14}C and ^{55}Fe. A protocol has been established and circulated among the NMIs asking them for their willingness to participate. Fourteen laboratories, ANSTO (Australia), CIEMAT (Spain), ENEA (Italy), IFIN-HH (Romania), IRMM (European Community), LNE-LNHB (France), MKEH (Hungary), NIM (China), NIST (United States of America), NMIJ (Japan), NPL (United Kingdom), NRCC (Canada), PTB (Germany) and RC-POLATOM (Poland) agreed initially to take part by submitting all or part of the indicated radionuclides. Unfortunately, the measurements had to be postponed due to an unforeseen breakdown of the BIPM microbalance which had to be replaced. In the meantime two additional laboratories declared their readiness to take part in the exercise, i.e. the BARC (India) and the NPL (United Kingdom). To carry out the extensive measurements necessary to fulfil the proposed task the BIPM will rely on the help of several external (wo)man-power from the CIEMAT, the NIST and the NIM.
The goals of the comparison are to: 1) analyse the results obtained by two different approaches, the "cross-efficiency method" developed at the CIEMAT, and the "apparent activity", a simple method proposed by the NMISA and based on the TDCR technique; 2) characterize the benefits of both methods; and 3) try to evaluate the most appropriate one to achieve the foreseen goal.
The measurements will be carried out with three commercial liquid-scintillation spectrometers, LS 6000TA from Beckman, TriCarb 2910TR and Quantulus 1220 from Perkin Elmer and the BIPM home-made TDCR set-up. At the same time, three different scintillators, based on di-isopropylnaphtalene (DIPN), Linear Alkylbenzene (LAB) or Pseudocumene, i.e. Ultima Gold®, Opti-Fluor® and Insta-Gel Plus® respectively will be used as well as two different volumes, 10 mL and 15 mL. Further, the suitability and the robustness of the quenching parameters provided by the three commercial devices will be tested.
This paper will detail the measurements and present with a particular care the uncertainty evaluation taken into account possible correlations.

Corresponding author's email address: **gratel@bipm.org**

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RESULTS OF AN INTERNATIONAL COMPARISON OF ACTIVITY MEASUREMENTS OF ^{68}GE/^{68}GA

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Cessna, J.T. *et. al.*

NIST (US)

There has been increasing interest in the use of ^{68}Ge/^{68}Ga as a surrogate for ^{18}F in quantitative imaging, as well as increasing interest in the use of ^{68}Ga for radionuclide-based radiotherapy for certain types of cancers. Accurate administrations of drugs using this radionuclide require accurate standards against which instrumentation used in the clinics and radiopharmacies can be calibrated. Germanium-68 decays with a half-life of 270.95(26) days by pure electron capture to the ground state of ^{68}Ga. Gallium-68 decays by both positron emission and by electron capture, mainly to the ground state of ^{68}Zn with a half-life of 67.83(20) minutes. Additionally, there is decay to a 1077 keV excited state with a probability of about 3 % and decay to several higher excited states with a combined probability less than 0.4 %. This decay scheme makes it suitable for analysis using a variety of techniques, including liquid-scintillation counting and coincidence counting.
Several national metrology institutes have reported measuring either ^{68}Ge/^{68}Ga or the daughter 68Ga alone. However, there had been no submissions to the International Reference System (SIR). The first international comparison for this radionuclide was organized and carried out. This paper presents a detailed discussion of the results of the comparison.
Germanium-68 solutions prepared at NIST were distributed to 18 participating institutions. A sample was also delivered to the BIPM for measurement in the SIR. The measurand for the comparison was activity of ^{68}Ge per unit mass. Results were received from 17 laboratories, comprised of 29 results by 8 primary methods and 7 measurements by 3 secondary methods. Each participant identified a single value, either a single method or combination of results, for calculation of a proposed comparison reference value (CRV).
The submitted values were evaluated by the Power Moderated Mean method and a comparison reference value is proposed. With the exception of one laboratory, identified as an outlier, the results are in statistical agreement with the proposed CRV. The discussion includes results of evaluations by other methods. No obvious trend could be identified in the results with respect to the method used. The results include methods not previously published for this radionuclide.

Corresponding author's email address: **jcessna@nist.gov**

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DIFFERENCES IN PUBLISHED HALF-LIFE VALUES FOR ^{207}Bi

003

Pibida, L.; King L. E.

NIST (US)

In 1973 the National Institute of Standards and Technology (NIST) developed a standard reference material (SRM) for ^{207}Bi. SRM point sources were prepared as part of that work. One of these sources was used over the years to calibrate high purity germanium (HPGe) detectors. This source was consistently used since the year 2000 to calibrate different HPGe detectors at different source-to-detector distances.

Last year (2015) a new detector geometry was calibrated using this same ^{207}Bi SRM point source and a discrepancy was noted when measuring the full-energy peak efficiency as a function of photon energy. In addition to the ^{207}Bi source, several other SRMs that included different radionuclides with photon energies ranging from 45 keV to 2.6 MeV were used to perform this calibration. The half-life that was initially used to decay correct the activity of the ^{207}Bi source was obtained from the Decay Data Evaluation Project (DDEP), which provides a value for the half-life of 32.9 (14) years. In the process of trying to understand the discrepancies observed in the efficiency values calculated using ^{207}Bi and other radionuclides, with photon energies close to the ^{207}Bi values, a separate independent value of the half-life of 31.55 (4) years published by the Evaluated Nuclear Data Structure File (ENSDF) was used to perform the decay correction. Prior to 2015, the ENSDF value of the half-life for ^{207}Bi had been used in the process of determining all the efficiency curves for the HPGe detectors

These observed differences in the values of the half-life lead us to revise all the efficiency curves measured from 2000 to 2016. In the process of doing so it was noted that the ^{207}Bi measured efficiency values were in better agreement with the efficiency values obtained using other radionuclides when the ENSDF half-life value was used. The differences when using the DDPE half-life value ranged from 2.5 % to 5.5 % (depending on the year when the measurements were performed) while the difference using the ENSDF value range from 0.5 % to 2 %.

During this time NIST re-evaluated many of the half-life measurements for long-lived radionuclides and reported a new half-life for ^{207}Bi of 11395 ± 18 days. The differences when using this new NIST half-life value ranged from 0.2 % to 1 %.

As a result of these measurements a review of the evaluation of the ^{207}Bi half-life value is recommended. This talk will discuss the full-energy peak efficiency measurements performed with the different HPGe detectors and results obtained when using the different ^{207}Bi half-life values.

Corresponding author's email address: **leticia.pibida@nist.gov**

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DIRECT MEASUREMENT OF THE ELECTRON CAPTURE PROBABILITY RATIOS OF FE-55

056

Loidl, M.; Rodrigues, M.

CEA, LIST, (FR)

The electron capture probability ratios PL/PK and PM/PK of ^{55}Fe have been measured with an electroplated ^{55}Fe source enclosed in the gold absorber of a metallic magnetic calorimeter (MMC). These detectors, a class of cryogenic detectors operating at 10 – 20 mK, had previously been used for the precise measurement of the shapes of low energy beta spectra. The absorber dimensions were chosen such that the detection efficiency for Mn K X-rays is larger than 99.99 % and the energy threshold of the detector lies below 50 eV. Since all electrons and photons emitted by the source are absorbed in the detector, the detection efficiency is virtually 100 % for K, L and M captures, whose X-ray and Auger electron emission energies sum up to 6.54 keV, 769 eV and 84 eV respectively. The energy of N captures (~ 3 eV) lies far below the energy threshold of the detector.

The absorber containing the source was attached to the paramagnetic Au:Er temperature sensor of an MMC device optimized for the absorber heat capacity. The sensor, exposed to a permanent magnetic field, transduces the temperature rise caused by the energy deposit of a decay within the absorber to a variation of magnetization. A superconducting quantum interference device (SQUID) reading out the sensor magnetization, and a specific SQUID electronics produce a voltage proportional to the energy deposit. The energy spectrum generated from the recorded pulse heights exhibits clearly distinct peaks for the K, L and M captures. The peak surfaces correspond to P_L/P_K = 0.1173 and P_M/P_K = 0.0181. Assuming P_N = 0.0006 according to Bé *et al.* 2006, this translates to P_K = 0.8802 (33), P_L = 0.1032 (11) and P_M = 0.0160 (4). Comparing these values to the recommended values P_K = 0.8853 (16), P_L = 0.0983 (13) and P_M = 0.0157 (6) from Bé *et al.* 2006, based on calculation, reveals a slight discrepancy for P_K and a somewhat larger discrepancy for PL. They are, however, compatible with the experimental values from Pengra et al. 1972, P_K = 0.881 (4), P_L = 0.103 (4) and P_{M+} = 0.0161 (8).

It is the first time that a cryogenic detector was used for the measurement of the ratios of electron capture probabilities. This new measurement technique has the advantage that it does not rely on the knowledge of the fluorescence yields, as it is the case of several other measurement techniques, or any other data of questionable reliability. It is furthermore, to our knowledge, the only measurement of P_M/P_K since the work of Pengra *et al*. in 1972.

M. M. Bé *et al.*, Table of Radionuclides, Monographie BIPM-5, Vol. 3 (2006)
Pengra *et al*., Phys. Rev. C 5 (1972) 2007

Corresponding author's email address: **martin.loidl@cea.fr**

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MEASUREMENT OF ^{103M}RH X-RAY EMISSION INTENSITIES AND EVALUATION OF THE DECAY SCHEME

059

Riffaud, J.[1]; Cassette, P.[1]; Lacour, D.[1]; Lourenço, V.[1]; Tartès, I.[1]; Kellett, M.A.[1]; Corbel, M.[1]; Lépy, M.C.[1]; Domergue, C.[2]; Destouches, C.[2]; Carcreff, H.[3]; Vigneau, O.[4]

[1] CEA, LIST, LNE-LNHB (FR) [2] CEA, DEN, DER (FR) [3] CEA, DEN, DRSN (FR) [4] CEA, DEN, DEC (FR)

Rhodium is used for dosimetry in nuclear reactors since the inelastic scattering reaction ^{103}Rh(n, n')^{103m}Rh is particularly useful to characterize the neutrons with energy around 1 MeV. For the dosimetry applications, the activity measurement of ^{103m}Rh is conventionally performed by spectrometry of low-energy photons. Unfortunately, the ^{103m}Rh decay scheme is rather poorly known. Its half-life is short (56.115 (6) min) and the transition from the isomeric state to the ground state of ^{103}Rh is highly converted, resulting in the emission of X-rays and Auger electrons. The K X-ray emission intensities (20 and 22 keV) are specified with relative standard uncertainties between 5.5% and 7%. In addition to the low energy of emitted photons, these large uncertainties induce difficulties to get reliable dosimetry results with low uncertainties. So, a new experiment has been initiated to improve the decay data of 103mRh. The procedure includes an absolute activity measurement, followed by X-ray spectrometry measurements, on the same sample, to determine the photon emission intensities. A specific irradiation of rhodium samples was conducted at the reactor ISIS, CEA Saclay. For this experiment the samples consisted of a rhodium(III) chloride powder ($RhCl_3$) and a pure rhodium pellet. Due to the ^{103m}Rh short half-life, the chemical preparation of the samples for measurements was performed immediately after irradiation. The rhodium chloride powder is soluble in water and allows samples to be readily prepared for measurements. The pellet was dissolved according to a procedure established and validated before irradiation.The activity measurement was performed by liquid scintillation (LS) counting. After dissolution, drops of the rhodium solution were mixed with a commercial liquid scintillator and the scintillating sources were measured using the triple-to-double coincidence ratio (TDCR). In parallel, the X-ray photon emission intensities were measured using a high-purity germanium detector. The experimental efficiency calibration is established for a reference geometry, using standard point sources with a relative standard uncertainty of 1.5-2 % around 20 keV. Monte Carlo simulations are used to refine the efficiency calibration curve due to the lack of standard sources in the required low energy range and also to compute the correction factors of the efficiency calibration to take into account the change in geometry, since the measured Rh samples are not point sources. In order to accurately determine the K X-ray emission intensities, the peak shape analysis takes into account the scattering effect (i.e. low-energy tailing). Internal conversion coefficients, conversion and Auger electron emission intensities and other atomic parameters will be calculated to improve the activity measurement. These new results will be incorporate into a new evaluation of 103mRh decay scheme, with a significant reduction of the photon emission intensity uncertainties expected.

Corresponding author's email address: **jonathan.riffaud@cea.fr**

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STUDY OF ^{243}AM DECAY

065

Caro Marroyo,B. ; Martín Sánchez, A.; Jurado Vargas, M.; García-Toraño, E.; Roteta, M.

UEx(SP), CIEMAT(SP)

Some time ago, the Research Project "Updated Decay Data Library for Actinides" highlighted the need to review several nuclear data for some actinides, such as ^{243}Am. These evaluations requested new measurements of the half-life and some alpha-particle and gamma-ray emission probabilities of this nuclide. The latest published data of gamma-ray emission probabilities of this nuclide were done in 1996. During those measurements, and using an alpha-gamma coincidence system, three new gamma transitions were found: 46.84, 98.36 and 102.02 keV. In the last evaluation by Decay Data Evaluation Project the gamma-ray emission probabilities of 46.84 and 102.02 transitions were not included. One of the aims of the present work was to find experimental evidences that confirm the existence of these two transitions as well as reviewing some of the gamma-ray emission probabilities previously reported. For that purpose, gamma-ray spectrometry and alpha-gamma coincidence measurements were used in this work. A vacuum chamber, a PIPS detector for alpha particles and a LEGe detector coupled to the back of the chamber to register gamma rays have been used. Preamplifier outputs from both detectors were connected to a digital module N1728 B from CAEN. The measurements were carried out for 132 days without interruptions. Our results confirm the existence of the two mentioned transitions. Pure experimental P_γ were determined by gamma-ray spectrometry for 31.3, 43.53, 46.84, 74.66, 86.71 and 141.9 keV transitions. On the other hand, combined P_γ values were calculated from experimental $P_{\gamma|\alpha}$ obtained by gamma-alpha coincidences and a 90° measuring angle and the isotropic reference transition probabilities recommended in the Nuclear Data Tables. These last probabilities were calculated for 43.53, 46.84, 40.62, 55.18, 74.66, 86.71, 98.36, 102.02, 117.84 and 141.89 keV transitions. In most cases, our results were in good agreement with the reference data.

Corresponding author's email address: **ams@unex.es**

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TWO DETERMINATIONS OF THE GE-68 HALF-LIFE

Bergeron, D.E.; Cessna, J.T. ; Zimmerman, B.E.

NIST (US)

Ge-68 decays by electron capture to the ground state of ^{68}Ga, which in turn decays with a half-life of 67.83(20) min., mostly by positron emission (89 %) (Bé *et al*., 2013). In nuclear medicine, ^{68}Ge is used to generate ^{68}Ga for imaging by positron emission tomography (PET). Further, sealed sources containing ^{68}Ge have been adopted as long-lived calibration surrogates for the more common PET nuclide, ^{18}F.

Recently, NIST prepared several ^{68}Ge sources for measurement on a pressurized ionization chamber (ICB, 1 half-life ampoule) and a NaI(Tl) well counter (gwc, 3 half-life ampoules), both equipped with automatic sample changers. The sources were measured approximately once a week for a period of nearly 100 weeks. Each ^{68}Ge measurement was accompanied by appropriate background and reference source (^{226}Ra for ICB, ^{129}I for gwc) measurements.

From the resulting data, values for the half-life of ^{68}Ge were calculated, giving $T_{1/2}$ = 271.03(14) d and $T_{1/2}$ = 271.09(17) d (k = 1) from the ICB and gwc, respectively.

This paper will describe in full the details of the uncertainty analyses, which look beyond fit uncertainties to identify uncertainty components acting on multiple timescales. We believe the final uncertainties to be conservative but realistic. We will show fit residuals to illustrate that they are without discernible structure.

The half-lives we report are in accord with the evaluated value of 270.95(26) d (Bé *et al*., 2013), which owes mostly to just two determinations, by Waters *et al*. (1981) and Schönfeld *et al*. (1994). This paper will discuss our measured values in the context of earlier determinations and describe the impact that including them would have on a new evaluation. We hope that this contribution will promote confidence in the nuclear data for ^{68}Ge, a radionuclide of extreme and growing importance in medicine.

Bé *et al*., 2013. Monographie BIPM-5. Table of Radionuclides Vol. 7 – A = 14 to 245.
Schönfeld *et al*., 1994. *Int. J. Appl. Radiat. Isotop.* 45, 955.
Waters *et al*., 1981. *Int. J. Appl. Radiat. Isotop.* 32, 757.

Corresponding author's email address:

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STANDARDIZATION AND HALF-LIFE OF ZR-89

García-Toraño, E. ; Roteta, M.; Peyrés, V; Mejuto, M. ; Sánchez-Cabezudo, A.; Romero, E.

CIEMAT (ES)

The nuclide ^{89}Zr disintegrates by beta plus emission (22.8 %) and by electron capture (77.2%) to ^{89}Y. More than 99% decays feed a metastable level of ^{89}Y at 909 keV. The recommended half-life is 78.42 (13) h, one of the longest among current commercially available beta plus emitters which allows imaging studies up to a 1 week after the injection of a ^{89}Zr-based probe. Pre-clinical studies of ^{89}Zr-based immuno-PET are in progress. This paper describes the standardization and half-life determination of this nuclide.

The standardization was done using the following techniques:

1.Liquid Scintillation Counting (LSC), using TDCR and CIEMAT/NIST methods and HISAFE 3 as scintillation cocktail. TDCR and CIEMAT/NIST methods were used.

2.4πγ counting with a 7 cm. × 7 cm. NaI(Tl) well type detector.

3.Coincidence counting between positrons and 511 keV photons using LSC in the beta channel and a NaI(Tl) crystal in the gamma channel. The system is controlled by a digital acquisition unit.

Half-life determination was done by fitting the decay curve obtained with different batches of material and using two measurement systems:

a. A well-type ionization chamber IG11 with digital registering of the ionization current.

b. A Ge spectrometer with gain stabilization and periodic ROI registering.

The ^{89}Zr decay was followed for time intervals up to 6 half-lives. The provisional value for its half-life is 78.33(8) h, slightly shorter but in agreement with recommended values.

Corresponding author's email address: **e.garciatorano@ciemat.es**

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^{230}U NUCLEAR DECAY DATA EVALUATION

140

Luca, A.; Ioan, M.R.

IFIN-HH (RO)

The ^{230}U decay chain is composed of 8 radionuclides; starting with ^{214}Po, this series has common components with the ^{238}U natural radioactive series. Most of the radionuclides from the ^{230}U decay chain are alpha-particle emitters, with potential to be used in targeted alpha therapy (Morgenstern et al., Appl. Radiat. Isot. 66 (2008) 1275).

In the frame of the Decay Data Evaluation Project (DDEP) and the IAEA CRP F41029 "Nuclear data for charged-particle monitor reactions and medical isotope production" (Report IAEA / INDC (NDS)-0675, April 2015), a decision was taken: to evaluate/update the previous decay data evaluations of the ^{230}U decay chain: ^{230}U and ^{226}Th by IFIN-HH (Romania), and from ^{222}Ra to ^{210}Po - by CEA/LNE-LNHB (France).

The paper describes the main results obtained in the new ^{230}U decay data evaluation (the first DDEP evaluation of this radionuclide, which is a 100% alpha-particle emitter), performed according to the DDEP procedures.

For this aim, the published data from more than 10 references were identified, analyzed and compiled. The evaluated half-life value of ^{230}U, 20.23 (2) days, was the weighted mean of the measurements published in the literature. The adopted decay energy (Q) is from the most recent atomic mass evaluation file available, Wang *et al*., *Chin. Phys.* C36 (2012) 1603.

The adopted levels energy, spin and parity values were obtained by combining the information from the mass-chain evaluation for A=230, most recently published (however, 20 years ago) in Nucl. Data Sheets (Y.A. Akovali, vol. 77 (1996) 433), and the results of Koua Aka *et al.*, *Radiochim. Acta* 68 (1995) 155.

The high-quality measurements of half-life and of alpha particles energy and emission probabilites, published a few years ago by Pommé *et al.* (Appl. Radiat. Isot. 70 (2012) 1900) and Marouli *et al.* (Appl. Radiat. Isot. 70 (2012) 2270), respectively, were very important for this new evaluation.

The BrIcc computer code (Kibédi *et al.*, Nucl. Instrum. Meth. Phys. Res. A 589 (2008) 202), was used to determine the internal conversion coefficients associated to the gamma-ray transitions. The results obtained for the energy and emission probabilities of the main X-rays following the ^{230}U decay are also provided.

The main difficulties encountered and approached in the evaluation process were:

-the necessity of an updated ^{226}Th scheme of nuclear levels populated by the ^{230}U alpha decay;

-the lack of experimental measurements, especially for the half-life (only two measurements published, one from 1948) and the extremely low emission intensities of most of the gamma-rays (down to less than 10^{-4} per 100 decays).

The consistency of the ^{230}U decay scheme was finally tested by using the tools of the SAISINUC software, produced by CEA/LNE-LNHB. The evaluation results will be reported to IAEA and DDEP for peer-review and, afterwards, the data sets will be included in their nuclear databases available to the international users.

Corresponding author's email address: **aluca@nipne.ro**

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RECOMMENDATIONS FOR NEW DECAY DATA MEASUREMENTS ARISING FROM THE DECAY DATA EVALUATION PROJECT (DDEP)

054

Kellett, M.A.

CEA, List, Laboratoire National Henri Becquerel (LNE-LNHB), (FR)

The Decay Data Evaluation Project (DDEP) has been established for over twenty years and has produced recommended decay data for over 220 radionuclides. The quality of these data has been recognised by the Consultative Committee for Ionizing Radiation (CCRI) of the Bureau International des Poids et Mesures (BIPM), who has recommended their use by national metrology institutes (NMIs) performing international comparison exercises.
DDEP evaluators perform a critical assessment of available experimental data as a fundamental part of any decay scheme evaluation, allowing shortcomings to be identified in the measured data. Recommendations can thus be made on where future measurement activities should be targeted. An earlier assessment [1] has already been carried out on the eighty-five actinide decay scheme evaluations performed under the auspices of an International Atomic Energy Agency (IAEA) Coordinated Research Project (CRP) and has proven to be of interest to the measurement community. Hence a similar exercise has been carried out on the remaining radionuclides.
Details on the quality of the available experimental data for each radionuclide will be presented, with recommendations for new data measurements.

[1] M.A. Kellett, 2012. "Assessment of actinide decay data evaluations: Findings of an IAEA Coordinated Research Project", *Appl. Radiat. Isot.*, 70, 1919.

Corresponding author's email address: **mark.kellett@cea.fr**

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PRECISE TEST OF INTERNAL-CONVERSION THEORY: TRANSITIONS MEASURED IN NINE NUCLEI SPANNING 45 ≤ Z ≤ 78

085

Hardy, J.C.[1]; Nica, N.[1]; Iacob, V.E.[1] ; Trzhaskovskaya, M.B.[2]

[1]Cyclotron Institute (US), [2] Petersburg Nuclear Physics Institute (RU)

The internal conversion coefficient (ICC) is a critical property of each nuclear electromagnetic transition, and is an essential ingredient in the construction of nuclear decay schemes. Even so, ICCs have only rarely been measured precisely. Instead, most decay schemes depend on ICCs obtained from theory – either by means of tabulations or on-line calculations – which are applied to measured relative gamma-ray intensities to determine total transition intensities. The accuracy of the largely untested calculations was a concern for decades.

In 2002, Raman et al. published [1] a careful study that compared the then-available ICC calculations with one another and with a compilation of all published measurements of ICC values. This revealed systematic discrepancies of up to 3% between theory and experiment but, since most measurements were far from precise, it was difficult to draw a firm conclusion. One particularly awkward question remained unanswered: How should the atomic vacancy created by the departing electron be treated? Some calculations took account of the vacancy; others did not. And the experimental results then available could not definitively distinguish between them. Starting in 2003, we embarked on a program to resolve this dilemma. The treatment of the vacancy has its greatest impact for high multiplicity transitions with low energy; it also varies as a function of the atomic number Z [2]. Consequently we have made precise measurements of K-shell conversion coefficients, α_K, for *E3* and *M4* transitions whose energy is close to the K-shell binding energy; and these measurements have by now encompassed transitions in nuclei over a wide range of atomic numbers from $Z = 45$ to 78 ($111 \leq A \leq 197$). Using an HPGe detector whose high-precision efficiency calibration has been described previously [3], we determine α_K from the measured intensity ratio of K x rays to γ rays for a transition of interest. Of course this technique requires that the radioactive source being used is dominated by a single transition that can convert in the K shell.

To date, we have measured and reported on four *M4* transitions – in ^{197}Pt [4], ^{193}Ir [5], ^{137}Ba [6] and ^{119}Sn [2] – and two *E3* transitions – in ^{134}Cs [6] and ^{111}Cd [7]. Two more *M4* transitions – in ^{125}Te and ^{127}Te – have been measured more recently and will be reported here. A measurement of another E3 transition – in ^{103}Rh – is currently in progress. The precision of the extracted results ranges from 0.5% to 1.9%. In all cases they disagree with ICC calculations that ignore the atomic hole, and are statistically consistent with Dirac-Fock calculations in which the atomic hole has been incorporated according to the "frozen orbital" approximation [8].

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[6] N. Nica *et al.*, *Phys. Rev.* C 77 (2008) 034306.
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Corresponding author's email address: **hardy@comp.tamu.edu**

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YIELDS MEASUREMENT OF PHOTONS 186,21 keV and KX (Rn) in Ra-226 DECAY

018

Dryák, P.; Havelka, M.; Mazánová, M.

CMI (CR)

Emission probabilities of 186.21 keV and KX(Rn) photons were measured by two calibrated HPGe detectors. The detector GC4018 is coaxial type, FWHM 1.8 keV for 1332 keV, relative efficiency 40%. The second one is type GCX, FWHM 1.8 keV for 1332 keV, relative efficiency 41%. Detectors are connected to the preamplifiers 2002CSL and modules LYNX. All systems are Canberra's products. Correction of the dead time (max. 0.4%) and pile-up effect was performed electronically by modules LYNX.

The efficiency was computed with MCNP code and validated using experimental points. Measurement of the yield 186.21 keV was done with two sources with Ra-226: one was an ampoule with 1 ml solution (activity 45.5 kBq) prepared in the year 1990 and transferable to Hoenigschmid Ra-226 standards, the second was prepared from NPL solution in the form of spectrometric CMI standard (activity 7.85 kBq). The source-detector distances were typically 25 or 26 cm.

The yield KX(Rn) in Ra-226 decay were determined from spectra without X-ray from daughter products (RaA, RaB, RaC, RaD). An absence of X-ray (Po, Bi) in measured spectra is demonstrated. The special Ra-226 source (type RF produced by CMI) was used: Ra-226 is incorporated into the Si gel foil with emanation power better than 0.998. There were used both detectors. The distances source-detector were 3 and 10 cm. The volume around detectors was intensively ventilated to remove any radon from the air surrounding the detector.

The relative X-ray intensities of radon (KXα2, KXα1, KXβ1, KXβ2) and gamma intensity were established and then absolute yields provided Y(186.21 keV)=0.03555(25).

The obtained value of yield of gamma 186.21 keV are in agreement with DDEP. A standard uncertainty of the measured KX(Rn) yields are from 1.4 to 2.4 %. They are less about of 6 % from DDEP.

The experiments were carried out in the frame EMRP project Metrology for processing materials with high natural radioactivity 'MetroNORM' (http://metronorm-emrp.eu/). The EMRP is jointly founded by the EMRP participating countries within Euramet and the European Union.

Corresponding author's email address: **pdryak@cmi.cz**

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DECAY DATA FOR THE POSITRON EMISSION TOMOGRAPHY IMAGING RADIONUCLIDE I-124: A DDEP EVALUATION

032

Zimmerman, B. E.

NIST (US)

Despite the fact that the majority of Positron Emission Tomography (PET) studies continue to be carried out using F-18, an increasing number of positron-emitting radionuclides are being developed for specific applications. One such nuclide is I-124, which is being investigated as a PET imaging surrogate for dosimetry assessments of the widely-used therapeutic radionuclide I-131. In addition, I-124 labeled compounds, such as [^{124}I]-M-iodobenzylguanidine ([^{124}I]-MIBG), are finding use in a wide variety of applications, such as cardiac imaging. Unlike the "traditional" PET radionuclides (e.g., ^{11}C, ^{13}N, ^{15}O, ^{18}F), whose half-lives are on the order of minutes, longer-lived radionuclides such as ^{124}I ($T_{1/2}$ = 4.17 d) are able to image physiological process that have longer time scales.

This presentation will discuss the details of a recent evaluation of the decay data for I-124 carried out under the auspices of the Decay Data Evaluation Project (DDEP). An evaluation was carried out by the Evaluated Nuclear Structure Data File (ENSDF) in 2008, however this is the first such evaluation for this radionuclide done by DDEP. While the ENSDF evaluation relied primarily on one study conducted in 1998, the present evaluation considered four primary sources in arriving at many of the recommended values.

Applying the DDEP methodology to the available literature data, a total of 86 gamma-rays emitted from 28 levels were considered. Three new levels not included in the previous evaluation have been added, along with several new gamma-rays. From these data, new recommended Pγ values have been determined, along with a new set of recommended $P_{\beta+}$ and P_{EC} values, that are consistent with experiment. Evaluated spin and parity assignments, logft values, and other pertinent nuclear and atomic data relevant to decay studies are provided, including a new evaluated half-life of 4.177(16) days.

Recommendations for additional experiments to help improve the current available data set will be presented.

Corresponding author's email address: **bez@nist.gov**

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NUCLEAR DECAY DATA EVALUATIONS FOR ^{56}CO, ^{181M2}TA, ^{196M2}AU, ^{199M}HG

040

Chechev, V.P. ; Kuzmenko, N.K.

KRI (RUS)

Nuclear decay data evaluations within the Decay Data Evaluation Project (DDEP) are presented for the radionuclides of ^{56}Co, ^{181m2}Ta, ^{196m2}Au, ^{199m}Hg. The previous DDEP evaluation was done only for ^{56}Co. It was placed on the DDEP web site in 2004 and published in the BIPM-5 monograph [1]. The experimental data published up to 2016 were taken into account in the current evaluations as well as other information: compilations, analyses, corrections.

In addition, there are some decay characteristics which undergone to the revision for all considered nuclides during the intervening period of 2004 - 2016. These are internal conversion coefficients (ICCs) and energy values related to the decay energy, Q.

For gamma-ray transition probability evaluations, the ICCs were used being interpolated with the BrIcc computer program [2] from the tables of Band *et al.* [3]. It has provided more accurate decay scheme intensity balance relations. As well, updated Q-values were taken from the 2012 atomic mass evaluation by Wang *et al.* [4] leading to the appropriate amendments to the nuclear transition energy values.

The updated evaluations were obtained using the approaches and methodology adopted by the working group of the DDEP cooperation. The reliability of the obtained evaluated data was checked for each nuclide by comparison of the total average emission energies, per decay, calculated from the present evaluated data, with the corresponding Q-values from mass tables [4].

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Corresponding author's email address: **chechev@khlopin.ru**

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EVALUATION OF THE RADON AND THORON INTERFERENCE ON THE PERFORMANCE OF THE COMPACT AEROSOL MONITORING DEVICE

033

Glavič–Cindro, D.; Petrovič, T.; Brodnik, D.; Vencelj, M.; Korun, M.; James Bell, S.; Keightley, L.; Woods, S.

IJS (SI), NPL (UK)

The main aim of the European metrological research project Metrology for radiological early warning networks in Europe (MetroERM), launched in June 2014, is to improve, optimize and harmonize the metrological foundation of measurements of fundamental radiological quantities such as airborne radioactivity concentrations, ground contamination levels and ambient dose equivalent rate in real-time. With the development of new methods for air sampling and new measurement systems (for rapid radioactive airborne concentration measurements also at low concentrations) that can be easily transported to locations of interest, the availability of accurate information on airborne radionuclide content, including nuclide-specific information provided by early warning networks in real time, will be considerably improved.

In the frame of this project a compact portable aerosol sampling and measurement device was developed at Jožef Stefan Institute. The system incorporates a $CeBr_3$ scintillation detector positioned centrally within a concertinaed filter assembly and an improved high flow rate air pump with a stable flow rate up to 200 m^3/h. It provides continuous on-line low level airborne radioactive particulate monitoring for field station use, connected directly via 3G network communication to a remote diagnostic center. The calibration and validation of the compact aerosol monitoring device was performed at the National Physical Laboratory (NPL) through use of filters spiked by a certified mixed nuclide solution containing a set of radionuclides with 14 prominent gamma-ray lines between 60 keV and 1836 keV (Am-241, Cd-109, Co-57, Ce-139, Cr-51, Sn-113, Sr-85, Cs 137, Mn-54, Y 88, Zn-65, Co-60 and Y-88). To make the distribution of activity over the filter as homogenous as possible, filters were spiked by drop-deposition of the solution on the filter cylinders along 11 equidistant rings.

In addition to validation of the system, also evaluation of the effect of the background radiation of natural radionuclides on the measurements of the airborne radioactivity with the compact aerosol monitoring device has to be performed. Therefore the study of radon and thoron interference with the overall nuclide identification performance in $CeBr_3$-based gamma spectrometry was included.

The measurements and the evaluation of the radon and thoron interference on the performance of the compact aerosol monitoring device will be presented and discussed.

Corresponding author's email address: **denis.cindro@ijs.si**

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CHARACTERISATION OF AN ULTRA LOW BACKGROUND POINT CONTACT HPGE-WELL DETECTOR

069

Hult, M.; Marissens, G.; Stroh, H.; Lutter, G.; Tzika, F.

EC, JRC-Geel (BE)

The workhorses of most radionuclide laboratories today are the HPGe-detectors. The technology to fabricate these High Purity Diodes has improved a lot since the first crystal was produced in the early 1960ies. In recent years, the technology to produce thick planar detectors with a small point contact anode at the back has led to detectors with better resolution and more stable operation. The latest development is to transfer this technology to the production of well-crystals. Canberra's trade-name of these detectors are SAGe™ -well detectors (Small Anode Germanium). Traditional well-crystals are known to have the lowest (worst) resolution amongst standard HPGe-detector but with this new technology there is a significant improvement particularly at lower energies.

This paper will describe the first ultra-low-background SAGe-well detector installed in a semi-deep underground laboratory (HADES at a depth of 500 m water equivalent). The well's diameter available for samples is 16 mm and its depth 40 mm. The crystal height is 85 mm and its diameter 90.5 mm and the Ge mass 2.8 kg. The endcap is made from ultrapure Al. The shield is composed of (from inside to out) 9 cm of electrolytic copper, 5 cm ULB lead (2 Bq kg^{-1} of ^{210}Pb), 15 cm "normal" lead (50 Bq kg^{-1} ^{210}Pb). The measured resolution is: ^{241}Am (59.5 keV): 0.72 keV; ^{137}Cs (661.7 keV): 1.33 keV; ^{60}Co (1173.2 keV): 1.79 keV; 60Co (1332.5 keV): 1.94 keV. The background count rate in the interval 40-27000 keV, when installed in a lead/copper shield underground, is 800 cpd (counts per day) which equals 290 cpd per kg Ge. Seeing that cosmogenic activation (^{65}Zn, $^{57,58,60}Co$ etc.) is still present in the crystal and copper shield, it is expected that the background will continue to decrease in the coming years. The detector and its shield were taken underground in July 2015.

The paper will describe the work to quantify the background contributions from the following sources: muons, neutrons, radioimpurities/activation in the shield, radioimpurities/activation in the detector and radon. This work involves using different shields and techniques to single out individual sources.

JRC-Geel performs reference measurements in all fields where radioactivity measurements are needed. The detector is expected to contribute significantly to projects where measurements of mono-energetic gamma-ray emitting radionuclides in small samples are required. Examples of such projects are ^{137}Cs in sea water samples and ^{241}Am in reference materials. In addition quantitative data on the improvement in detection limits and decision thresholds, compared to existing detector-systems, will be given.

Corresponding author's email address: **mikael.hult@ec.europa.eu**

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LOW-LEVEL LABORATORY MEASUREMENT OF RADIO-XENON: BETA-GAMMA VERSUS GAMMA MEASUREMENTS

078

Delaune, O.; Cagniant, A.; Douysset, G.; Fontaine, J. P.; Gross, P.; Le Petit, G.

CEA, DAM, DIF (FR)

Detection of nuclear explosions on the whole planet is a challenging issue for all countries that have signed the Comprehensive nuclear Test Ban Treaty (CTBT). Especially if the explosion occurs within underground cave, as only few radionuclides may escape from it. Noble gases, such as Xenon, diffuse more easily than other radionuclides through rocks and then may be released in the atmosphere. They are then detected by radionuclides stations from the CTBT organization.

Four radio-Xenon with suitable half-lives are considered: ^{131m}Xe, ^{133}Xe, ^{133m}Xe and ^{135}Xe. Two different setups have been designed by CEA for the radio-Xenon laboratory measurements:

- A double-sided cell with an active volume of 13 cm^3 made of stainless steel and 0.6 mm thick entrance windows in carbon.
- A PIPSBox cell designed in collaboration with the CANBERRA company. Its active volume is 10.8 cm^3 and it is composed by two face-to-face Silicon detectors and 0.6 thick entrance windows in carbon. Both Silicon detectors are sensitive to low-energy beta (from about 30 keV) and have a rather good resolution (11 keV (FWHM) at 130 keV).

In addition, an ultralow background gamma spectrometer, named Gamma^3, has been developed in CEA. It is composed of three hyper-pure Germanium detectors (HPGe). Its shielding —made of several layers of low-activity lead coupled to a layer of borated polyethylene and of cosmic veto covering 95% of external surface— allows reaching very low background integral count rate, such as 0.047 cps/kgGe (40-2700 keV) which is equivalent to the background of detectors located at 10 meters of water equivalent depth.

Radio-Xenon are usually measured by gamma spectrometry. Coupling electron detection capacities allows minimizing the radionuclides interferences and a significant reduction of the minimum detectable activity (MDA), especially for metastable radio-Xenon. Thus, MDA for a 3-days long measurement goes from 3.1 to 0.8 mBq for ^{133m}Xe and from 2.3 to 0.6 mBq for ^{131m}Xe corresponding to a sensitivity increased by a factor 4. Adding a second HPGe, so that the PIPSBox be in sandwich between both detectors, allows to goes down to 0.4 and 0.3 mBq for ^{133m}Xe and ^{131m}Xe respectively. For non-metastable radionuclides, using β/γ measurement allows to reduce MDAs by a factor 2 (^{133}Xe) and 4 (^{135}Xe).

The presentation will show the performance of the Gamma^3 spectrometer and the MDA that are obtained. Then, the gain of using the PIPSBox for reducing MDAs will be presented.

Corresponding author's email address: **olivier.delaune@cea.fr**

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ON THE SEQUENTIAL SEPARATION AND QUANTIFICATION OF ^{237}NP, ^{241}AM, TH, PU, AND U ISOTOPES IN ENVIRONMENTAL AND URINE SAMPLES

046

Vasile, M.[1]; Jacobs, K.[1]; Zhuxin, L.[2]; Verrezen, F.[1]; Bruggeman, M.[1]

[1] SCK-CEN (BE) [2] University Nantes (FR)

Radioactivity analyses based on low-level alpha spectrometry are quite expensive because of the labour-intensive chemical separations that are required to minimise interferences in the alpha spectrum. We investigated a one-pass-through sequential separation (as opposed to several individual chemical separations) technique using stacked chromatography columns with TEVA – TRU resins for the separation of ^{237}Np, ^{241}Am, Th, Pu and U from environmental and urine samples. The element specific factions are obtained by elution from the chromatographic columns with different reagents. The procedure was adapted from Triskem (03/03/14) and Maxwell, S.L. (2008). The counting planchets are prepared by electrodeposition. Special attention must be paid when using ^{232}U as a tracer since it decays to ^{228}Th and will contribute to the activity concentration of ^{228}Th in the thorium sample fraction. This problem can be avoided by purifying the ^{232}U tracer prior to its use following the procedure described by Sill (1974). Since also ^{229}Th is used as tracer, its daughter product ^{225}Ac will follow the Am fraction and will interfere with the analysis of Cm. The sequential separation and subsequent radionuclide analyses were validated using reference materials (IAEA-447 and IAEA-434) for solid environmental samples and well known tracers for liquid samples (water and urine).
The sequential separation technique proves to be successful and gives similar results as those obtained by individual separations hence allowing more cost effective analyses of these radionuclides by alpha spectrometry.
Since the assay of actinides relies more and more on mass spectrometry we will discuss a comparison between alpha-particle spectrometry and ICP-MS results obtained using the sequential separation method. Advantages and disadvantages will be discussed for the both measurement techniques.

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Corresponding author's email address: **mvasile@sckcen.be**

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SPALAX NG: A BREAKTHROUGH IN RADIOXENON FIELD MEASUREMENT

079

Cagniant,A.; Topin, S.; Le Petit, G.; Delaune, O.; Gross, P.; Philippe, T.; Piwowarczyk, J.-C.; Douysset, G.

CEA, DAM, DIF (FR)

The International Monitoring System (IMS) of the Comprehensive nuclear Test Ban Treaty Organization (CTBTO) aims to detect any nuclear explosion releasing at least 1 kT eq. TNT. It is composed of hydro-acoustic, seismic, infrasonic and radionuclide stations spread all around the world. The 135, 133m, 133 and 131m radioactive isotopes of xenon (or radioxenon) releases in the environment are key indicators of nuclear activity. Therefore, IMS radionuclide stations include radioactive xenon sampling and measurement capabilities. The design of these stations was let to CTBT state-parties, and three types of station were installed in the early 2000s: the SAUNA (from Sweden), the ARIX (from Russia) and the SPALAX (from France).
Following several years of R&D, CEA developed:

(i) A new high efficiency Xe adsorbent (silver doped zeolite Ag@ZSM5) for a more efficient gas concentration process (in terms of capacity, xenon recovery yield and power consumption),

(ii) A new high resolution photon/electron detection setup (HPGe detector coupled with Si detectors), designed to drastically reduce radioxenon Minimum Detectable Activity and minimize radionuclide interference during the measurement.

(iii) A new air-treatment gas line composed of two compressors coupled to 8 permeation membranes, in order to increase the rapidity, reliability and the efficiency of xenon gas production,
Thanks to these developments and to the lessons learned from 10 years of SPALAX stations operations, CEA initiated in 2015 the development of a New Generation of SPALAX stations. From the collaboration between CEA and the Cegelec company, the conception of the future station was completed in July 2016 and the first industrial prototype SPALAX NG station should be delivered by the end of June 2017.
The performance obtained with a laboratory developed prototype installed at Bruyères-le-Châtel will be presented. The new station produces a xenon sample equivalent to approximately 70 m^3 of air every 8h (when previous stations produced 20 m^3 of air from a 24h sampling period). The resulting Minimum Detectable Concentrations of the prototype are greatly reduced for the metastable radioxenon (131m- and 133m-Xe) as they reach 0.4 mBq/m^3 (against 1.5 and 4 mBq/m^3).
Thanks to the high resolution of the implemented electron and photon detectors, interferences between radionuclides are greatly reduced, which preserve the trace detection capability of the station even in the case of 133-Xe or 222-Rn consequent level of activity in the sample. Which is a great advantage compared to β/γ spectrometer-equipped stations.
Finally, radioxenon field-measurement improvements from air sampling will be demonstrated.

Corresponding author's email address: **antoine.cagniant@cea.fr**

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DETERMINATION OF ^{22}Na ACTIVITY IN AIR AND RAINWATER SAMPLES BY GAMMA-RAY SPECTROMETRY

092

Savva, M.I.; Karangelos, M.J.; Anagnostakis, M.J.

National Technical University of Athens, (GR)

The activity concentration of cosmogenic radionuclides in atmospheric aerosol has been used in many applications, such as air mass transport models and solar activity reconstructions. One of the most frequently studied cosmogenic radionuclides is ^{7}Be, while ^{22}Na can also be measured but presents challenges due to its very low activity concentration of the order of 1 μBq/m^3.
Natural radionuclides such as short lived radon daughters and the 7Be have been analyzed in aerosols at the Nuclear Engineering Department of the National Technical University of Athens (NED-NTUA) since many years, with high accuracy and precision. During this research, an investigation regarding the detection of ^{22}Na and the determination of ^{7}Be/^{22}Na ratio has been conducted. Sampling and analysis methods were appropriately selected to allow for the detection of the low activity of ^{22}Na. In order to study the role of precipitation in surface air ^{22}Na activity concentration, a procedure for collecting and analyzing rainwater was also developed.
Air sampling was performed using a high volume (~80m^3/h) air sampler and high efficiency (>98%) glass fiber filters 8"x10". In this study, sampling covering a 3-year period is presented, with 4 – 10 days of sampling per month (average flow 1300 L/min, volumes sampled 7500 – 18700 m^3). Plastic basins, covering an area of about 1 m^2, were installed on the roof of NED-NTUA building for the rainwater collection. The basins were continuously exposed to precipitation, while rainfall height was simultaneously monitored. Sampling covered a one year period (2015) with monthly intervals (volumes sampled 6 – 64 L). Rainwater samples were chemically treated using cation exchange resin for the extraction of Na cations.
The determination of the activity concentration of ^{22}Na was performed by means of gamma-ray spectroscopy, through its 1274.5 keV photos. The apparatus consists of an eXtended Range Ge detector (XtRa) 104% rel. eff. housed in an old steel shield and equipped with a Compton Suppression System (CSS). The CSS consists of a NaI(Tl) annulus guard detector (Ø195x267mm) and a NaI(Tl) plug detector (Ø103x51mm). The electronic set-up of the apparatus allows for the simultaneous collection of both the suppressed (anti-coincidence) and the unsuppressed (normal) spectrum. The spectrum that provides the best detection limit and measurement uncertainty may then be selected. Since the guard detector can be used both in anti-coincidence and coincidence mode with the primary detector, the possibility of ^{22}Na detection through γ-γ coincidence of its 511 keV and 1274.5keV photons, was also examined.
With the techniques used ^{22}Na was detected in air samples, although with relatively high uncertainties (>50%). The lower limit of detection for ^{22}Na was calculated to be of the order of 0.8 μBq/m^3 in air samples and of the order of 0.6 mBq/L in rainwater samples.

Corresponding author's email address: **msavva@nuclear.ntua.gr**

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LOW-LEVEL DIRECT MEASUREMENT OF U-238 IN ENVIRONMENTAL WATER USING STATE-OF-THE ART GAMMA SPECTROMETRY

093

Douysset, G. ; Cagniant, A. ; Delaune, O.; Greiner, V.

CEA, (FR)

Detection and measurement of uranium in environmental water is of primary importance for site monitoring in normal and incidental situation. Current recommended analytical methods for measurement of this parameter involve fluorimetry, mass spectrometry or possibly alpha spectrometry. Detection limits ranging from ~1 ng/l to ~10 µg/l are achieved with these techniques at the cost of time-consuming radiochemical procedures and/or large financial investments. Even though implementation of direct gamma spectrometry would be more efficient for these aspects, until now this technique has not demonstrated sufficient sensitivity. Hampered by the low gamma emission of U-238, minimum detectable concentrations as large as 160 µg/l (~1.8 Bq/l for U-238) are typically encountered in routine operation of a standard gamma spectrometer.

A state-of-art gamma spectrometry facility has been designed and setup at our surface-level laboratory. This instrument, also known as Gamma3, is exhibiting both optimized low background and very large detection efficiency. Optimized active shielding (cosmic veto + radon mitigation system) and passive shielding (radiopure Pb layers + borated polyethylene layer) lead to drastic reduction of telluric and cosmic components of the background which is now as low as 2.9 cpm (range: 20-2500 keV). Gamma3 comprises three separated moveable Ge γ-spectrometers (CANBERRA BEGe5030P type) that can accommodate any sample geometry so as to optimize detection solid-angle. The individual spectrometers can operate either in single, additive, coincidence/anti-coincidence mode thanks to implementation of XIA Pixie4 digital electronics.

For water analysis, a dedicated large volume square bottle is placed in close contact with the three crystals. This configuration has been optimized by Monte Carlo simulation so as it enhances the detection efficiency by a factor of ~3.5 compared to the standard cylindrical 500 ml (SG500) as measured on a single coaxial detector. Taking benefit of the enhanced sensitivity and the reduced background, detection limits for uranium as low as 10 µg/l (~0.12 Bq/l for U-238) are achievable without pre-concentration of the sample. The latest WHO's (World Health Organization) provisionary guideline value for maximum uranium concentration in drinking water is 30 µg/l. Therefore gamma spectrometry, even though not as sensitive as some other advanced techniques, begins to be competitive for simple assessment of uranium content of water samples. It also offers the advantage to monitor in a single run some relevant decay products of U-238 such as Ra-226 or Pb-210. This presentation describes the measurement system, its optimization and calibration. Performances are illustrated in several situations.

Corresponding author's email address: **guilhem.douysset@cea.fr**

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A COMPLETE ANALYSIS METHOD OF NORM BUILDING MATERIALS BY GAMMA SPECTROMETRY WITH HPGE DETECTORS

126

Quintana, B.[1]; Santamaría, R.[1]; Pedrosa, M.C.[1]; Vázquez-Canelas, L.[1]; Díaz-Rivera, N.[1]; Sanjuan, M. A.[2]; Puertas, F.[3]; Gascó, C.[4]

[1] LRI, Universidad de Salamanca (ES); [2] Instituto Español del Cemento y sus Aplicaciones (ES); [3] CSIC (ES); [4] CIEMAT (ES)

The use of NORM wastes for building materials requires procedures easy to implement and flexible enough to take into account the material characteristics, which provide reliable measurements of low-level activities of natural radionuclides as ^{210}Pb, $^{226/228}$Ra, ^{230}Th, $^{235/238}$U. Low-level background gamma spectrometry with HPGe detectors allows for the simultaneous determination of the most of them, but the analysis of γ-ray spectra from natural radionuclides is somewhat tricky since it often involves de-convolution of overlapped full-absorption peaks. In addition, these materials show different compositions and densities, even shapes when in solid form, requiring efficiency calibrations for each case. Coincidence summing is also a challenge when dealing with such a wide variety of measuring conditions. In this paper, all these aspects are tackled, first, by applying a new method that implements a genetic algorithm to de-convolute full-absorption peaks in γ-ray spectra, which is included in a complete gamma analysis tool: GALEA, and, second, by using Monte Carlo simulations to calculate efficiencies and coincidence-summing corrections, which take into account material composition. To reach a complete and consistent analysis of the spectrum, GALEA considers possible secular equilibrium among radionuclides and automatically achieves their identification. The global statistical coherence on the activities calculated from the different emissions of the same radionuclide located in a spectrum is preserved through a preliminary analysis. The main issues associated to the spectral analysis of natural radionuclides are also tackled in this work. Regarding Monte Carlo simulations, they are carried out with GEANT4, tool that allows for an easy implementation of measurement geometries and matrix characteristics. The full method has been applied to NORM building materials ranging from ceramics to concrete and NORM wastes used in building materials as fly and bottom ashes, clinker and phosphogypsums, being validated with reference data. Results show the method to provide reliable results in a fast way for any kind of NORM samples.

Corresponding author's email address: **quintana@usal.es**

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MEASUREMENTS OF ENVIRONMENT SAMPLES IN WELL-TYPE DETECTORS IN THE MODANE UNDERGROUND LABORATORY

129

de Vismes Ott, A. ; Cagnat, X.; Gurriaran, R.

IRSN (FR)

The French Institute of Radioprotection and Nuclear Safety (IRSN) is responsible for performing the radiological environment surveillance in France. In this context radioactivity in environment samples is measured at trace levels in the laboratory of environmental radioactivity measurement by low level gamma ray spectrometry.
During the last decades metrology developments have been made in concert with sampling improvements in order to deal with the globally decreasing levels of the anthropogenic radioactivity in the French environment.
In Orsay the 15 HPGe detectors are made of materials especially selected for their low radioactivity. The telluric component of the background is also reduced thanks to lead shields, and especially a 20 m^2 shielded room made of 10 cm lead and 0.5 cm of OFHC copper. Air renewing systems prevent the presence of radon progeny. And finally the cosmic ray induced background is also diminished thanks to the shallow location of our equipments but also with anti-cosmic devices equipping 6 of our detectors.
One of the detectors is also equipped with an anti-Compton system which decreases the background induced by the sample itself and is particularly suitable for biological samples containing large amount of ^{40}K whose Compton continuum makes the determination of the ^{137}Cs activity difficult.
This paper presents the use of the very low background detectors we installed in the deep underground laboratory in Modane (LSM) focusing on both well-types detectors. These detectors combine a very low background thanks to their location under 1700 m of rock and a very high efficiency thanks to their typical shape. Both features lead to very low detection limits. This is why they are used for the measurement of environment samples characterized by low activity concentrations or very small quantities, leading to very low activity per sample to measure.
These measurements combine the requirements and difficulties related to trace level measurements, such as a very precise determination of the background to be subtracted from the measurement, but also the requirements and difficulties related to the measurements in well-type detectors, such as the efficiency calibration and how the traceability to the primary standards is established, or the coincidence summing corrections made by Monte Carlo simulations.
This paper will describe how these requirements are met and how the difficulties are overcome. It will show the achieved performances in terms of detection limits illustrated with measurement results of various environment matrices: sea waters in the marine environment, suspended sediments sampled in rivers, or aerosol filters and rain or cloud waters in the atmospheric field.

Corresponding author's email address:

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APPLICATIONS OF LOW-LEVEL UNDERGROUND GAMMA-RAY SPECTROMERY AT IAEA'S ENVIRONMENT LABORATORIES

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Osvath, I.; Comanducci, J.F. ; Fujak, M.; Harms, A.V.; Levy, I.; Mc Ginnity, P.; Pham, M.K.

IAEA Environment Laboratories, (MC)

The International Atomic Energy Agency operates a low-level shallow underground counting laboratory in Monaco. This facility, situated at a depth of 35 m water equivalent, is primarily used for gamma-ray spectrometric measurements of samples of marine environmental and atmospheric origin. It hosts seven low-background Germanium detectors, including large volume well, coaxial and broad-energy type detectors. Careful selection of materials for the passive shielding, Radon reduction and the addition of a cosmic veto background reduction system and a Compton suppression system are used to further reduce the background and improve the detection limit and sample throughput.

These facilities are used in the characterization of reference materials of marine origin for natural and anthropogenic radionuclides in matrices such as sediment, seawater and marine biological materials (seaweed, mollusks, fish); in testing or analysis of proficiency test and interlaboratory comparison samples, for natural and anthropogenic radionuclides, mainly in marine matrices; in analyses of marine samples collected in environmental radiological assessment projects and in oceanographic and climate studies using radiotracers; and in analyses of large volume aerosol samples and of atmospheric deposition samples collected in Monaco for atmosphere-sea surface transfer studies and in support of Monaco's atmospheric monitoring programme. The low background and the stability of the systems support increased accuracy and precision of the measurement results.

This paper presents the recent improvements and summarizes the characteristics of the facility and the gamma spectrometric systems and provides an overview of the performances achieved in the different types of measurements performed in this laboratory. It outlines the analytical challenges and the operational solutions adopted for different types of applications, with respect to optimizing counting geometries, calibration methods and corrections. These are illustrated through examples such as sediment dating, deep ocean seawater analyses, interlaboratory comparisons in support of sea area monitoring offshore Fukushima, etc. Finally the paper presents plans for future developments of IAEA's underground facility in Monaco.

Corresponding author's email address:

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RADON IN DRINKING WATER: REVIEW AND EVALUATION OF ACTIVITY MEASUREMENT METHODS

154

Wiedner, H.; Lotter, K.; Maringer, F. J.; Friedmann, H.

University of Vienna, (AT)

Radon is estimated to cause between 3 % and 14 % of all lung cancer cases, depending on the average radon level in the country, making it the second cause for lung cancer after smoking (WHO, Fact sheet N°291, 2014). For Europe, this corresponds to about 15 to 20 thousand people dying per year due to lung cancer caused by radon exposure. ^{222}Rn, the progeny of ^{226}Ra, emanates from the soil into dwellings and is breathed by the occupants.
In most cases, radon in water has no relevant influence on the exposure of the public. In specific cases, like in areas with elevated geogenic radon potential, the effects could be of relevance. The effect of radon in water is less investigated and the correlation between the geogenic radon potential and the local radon concentration in ground and spring water has not been studied extensively yet. Sampling of water sources is relatively easy in wells and springs, compared to soil gas radon exhalation measurements. The correlation of the results of the water measurements and the local radon potential is discussed in this paper. Furthermore, it is investigated if the influence of radon in water in Austria is of concern in areas with elevated radon potential.
This paper offers a review of existing activity measurement methods of radon in water as well as the results of radon activity concentration measurements of water samples taken in regions with elevated radon potential in Austria. The method applied in this paper uses air flowing through a one liter water sample, accumulating approx. 95 % of the radon in the water. The radon rich air flows into a pre-evacuated 10 l ionization chamber. The ionization current is measured by a high resistance electrometer. This method is quick and easy and can also be applied in the field. Furthermore, this paper offers an investigation of the recovery rate of radon in water, using different water temperatures and underpressures as well as drinking water spiked with traceable amounts of ^{226}Ra.
Traceable calibration of all involved measurement devices has been established. Special attention has been given to the traceability of the method and uncertainty evaluation of the applied method.

Corresponding author's email address: **hannah.wiedner@bev.gv.at**

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NATURAL RADIONUCLIDE ACTIVITY CONCENTRATIONS IN SOILS AND VEGETATION AT AN URANIUM MINING SITE

Charro, E. ; Moyano, A.

Universidad de Valladolid, (SP)

The objective of this work was to investigate the uptake of several radionuclides in uranium mining-impacted soils in Central-West of Spain by the vegetation characteristic of a dehesa ecosystem. Therefore the aim of the present investigation was to determine the accumulation of ^{238}U, ^{226}Ra, ^{210}Pb, ^{232}Th and ^{224}Ra by native vegetation, given the existence of uranium mine tailings in the area. First, a radiological characterization of the granitic area was done in order to choose the sampling and control points. Second, analysis of the physicochemical parameters as pH, texture, organic carbon and Ca, Na, K, Mg in the soil samples was performed. Third, the activity concentration for ^{238}U, ^{226}Ra, ^{210}Pb, ^{232}Th, and ^{224}Ra was measured in soil and vegetation samples using a Canberra n-type HPGe gamma-ray spectrometer. The influence of the mine, the physicochemical parameters of the soils and the type of vegetation was analysed in order to justify the accumulation of the radionuclides in the plants. Given that there was no evidence of a higher content of radionuclides in the vegetation collected in the points closer to the tailings, the radionuclide contents in the plants could be explained in terms of the type of vegetation.

A comparison of the ratios [^{226}Ra/^{210}Pb] in soil and vegetation samples shows that the values in soils are close to the equilibrium but the same ratio in the different parts of the plants were found lower, being the activity concentration of ^{210}Pb higher of ^{226}Ra.

An analysis of the distribution of the radionuclide activity concentration in five grain size fractions of the soils has been also performed in order to discuss the mobility or the availability of some radionuclides by the plants. The values of the concentration and the ratios of the radionuclides under study do not show a clear preference for a particular soil fraction.

The transfer factor (TF) of the natural radionuclides in different parts of the native plants (leaves, branches, twigs, and others) was evaluated. A preferential uptake of ^{210}Pb and ^{226}Ra by the plants, particularly by the trees of the *Quercus* species (*Quercus pyrenaica* and *Quercus ilex rotundifolia*) has been observed. From this data, a correlation between the TFs of ^{226}Ra and ^{210}Pb of the different parts of the plants has been found. According to our data, the TFs for ^{226}Ra and ^{210}Pb in Quercus tree species were found higher than those for the other plants (*Pinus pinaster, Rubur ulmifolius* and *Populus sp.*). Also, translocation factors (TLF) for ^{226}Ra and ^{210}Pb were also evaluated for the different parts of the plants. In conclusion, no evidence of the influence of the radionuclide concentration in soils with higher TF has been found. However, the analysis of the results suggest that the main influence for the TF was the type of plants, and in particular the specie of the tree, with special attention to the rate of growth of the tree.

Corresponding author's email address: **echarro@dce.uva.es**

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