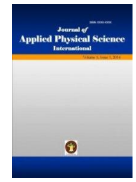
Journal of Applied Physical Science International

10(5): 241-246, 2018
ISSN: 2395-5260 (P), ISSN: 2395-5279 (O)



THE ABSOLUTELY STANDARDIZATION METHODS OF ^{153}Sm FOR CALIBRATING NUCLEAR MEDICINE INSTRUMENTS IN INDONESIA

GATOT WURDIYANTO[1*], HERMAWAN CANDRA[1] AND AGUNG AGUSBUDIMAN[1]

[1]Center for Technology of Radiation Safety and Metrology (PTKMR), National Nuclear Energy agency of Indonesia (BATAN), Jalan Lebak Bulus Raya, No. 49, Jakarta Selatan, 12440, Indonesia.

AUTHORS' CONTRIBUTIONS

This work was carried out in collaboration between all authors. All authors read and approved the final manuscript.

ARTICLE INFORMATION

Reviewers:
(1) Sebastião David Santos-Filho, State University, Rio de Janeiro, Brasil.
(2) Marcus Oliveira Ifba, Federal Institute of Bahia, Salvador, Brazil.

Received: 20 October 2018
Accepted: 26 December 2018
Published: 16 January 2019

Method Article

ABSTRACT

The Standardization absolutely of radioactive sources ^{153}Sm to calibrate the nuclear medicine equipment had been carried out in PTKMR-BATAN. This is necessary because the radioactive sources used in the field of nuclear medicine has a very short half-life in other that to obtain a quality measurement results require special treatment. Besides that, the use of nuclear medicine techniques in Indonesia develop rapidly. All the radioactive sources were prepared by gravimetric methods using KERN ABT 220-5DM type balance, traceable to SI. Standardization of ^{153}Sm has been carried out 4phi beta (LS) – gamma coincidence methods, while the impurities has been carried out by gamma spectrometry methods which calibrated using ^{133}Ba, ^{152}Eu, and ^{241}Am. The result of absolutely measurement was 88.48 kBq/g with 1.36 % of the expanded uncertainty, in covered factor, k = 2. This value to be used for calibrating a Capintec CRC-7BT radionuclide calibrator which is a secondary standard instrument in PTKMR - BATAN. The results show that calibration factor for Capintec CRC-7BT dose calibrator is 1.05 by about 4.6 % of the expanded uncertainties.

Keywords: Standardization; 4phi beta (LS)-gamma coincidence; dose calibrator; short half-life; ^{153}Sm.

1. INTRODUCTION

Utilization of nuclear technology in the health sector is growing rapidly in Indonesia. More than 30 hospitals have been utilizing this technology as a means of activities, and various types of radionuclides that are used also more varied. The use of radioactive substances are part of the nuclear technology relatively quickly perceived by society. This is due to radioactive substances have specific properties, not shared by other elements. By utilizing the properties of the radioactive, so many complex issues that can be simplified so the solution becomes easier. Based on the duties and functions as a national reference

**Corresponding author: Email: gatot_w@batan.go.id;*

laboratory in the field of the measurement of radioactivity, the Centre for Technology of Radiation Safety and Metrology (PTKMR-BATAN) has to be able to provide a liquid standard source with various types that qualify as standard reference materials so that results of the measurement and testing of samples has a value of accurate, precise and traceable to the International System.

In a previous study, it has been reported standardization source ^{125}I, ^{131}I, ^{99m}Tc and ^{18}F to calibrate equipment used in nuclear medicine Indonesia [1]. The radionuclides of ^{131}I, ^{99m}Tc and ^{18}F were standardized by the gamma-ray spectrometry method, but ^{125}I is absolutely standardized by photon–photon coincidence counting methods. At present, ^{153}Sm radionuclides were absolutely standardized by 4πβ(LS)-γ coincidence counting system.

The PTKMR-BATAN Secondary Standard Ionization Chamber is a Capintec CRC-7BT radionuclide calibrator that is used as a working chamber for the routine dissemination of activity standards for photon emitting radionuclides. The calibrator is checked routinely to ensure constancy and relative accuracy using standard sources from national metrology institutes (NMIs), namely the Physikalisch-Technische Bundesanstalt (PTB), the National Bureau of Standards (NBS, now the National Institute of Standards and Technology, NIST) and the National Measurement Institute of Japan (NMIJ). In this case, the PTKMR-BATAN Secondary Standard Ionization Chamber is calibrated by ^{153}Sm which absolutely standardized by 4πβ(LS)-γ coincidence counting system.

Several national metrology institutes have standardized of ^{153}Sm using absolute methods, such as PTB, measured ^{153}Sm using 4π (PPC)β – γ coincidence counting and by 4πβ liquid scintillation counting (LSC) spectrometry [2]; NPL, measured ^{153}Sm using 4π (PC)β – γ coincidence counting with a proportional counter operating at atmospheric pressure and a NaI crystal used as a gamma detector [3]. National Center for Nuclear Research Radioisotope center POLATOM, measured ^{153}Sm by 4π(LS) – γ coincidence and anticoincidence [4].

^{153}Sm was found in 1953 by the Swiss chemist Jean Charles de. ^{153}Sm is highly effective in reducing the pain of secondary cancers that have spread to bone, sold as Quadramet, is a chelated complex of a radioisotope of the element samarium with EDTMP (ethylene diamine tetramethylene phosphonate). These radionuclides are also very effective commonly used in lung cancer, prostate cancer, breast cancer, and osteosarcoma. This paper describes the procedures used to standardizes the activity of ^{153}Sm, and the results of the measurements used to calibrate the nuclear medicine equipment's in Indonesia.

The use of radiopharmaceuticals for the treatment of pallation pain in patients with suffering from bone metastatic pain have been reported by several previous researchers, such as Finlay et al. [5]; Lam et al. [6]; Gkialas et al. [7].

Radionuclide ^{153}Sm emit β^- decays with half-live 1.9285 (5) days to ^{153}Eu with most gamma energy at 69 keV and 103 keV [8].

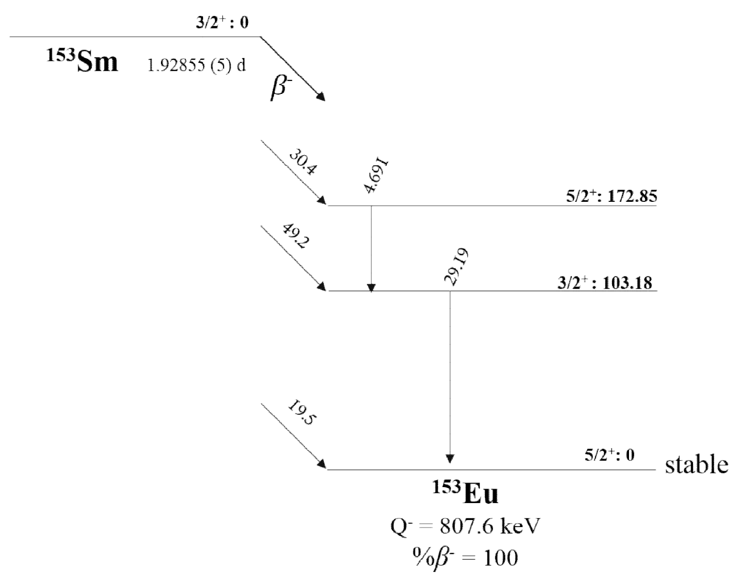


Fig. 1. Simplified decay scheme of ^{153}Sm

2. EXPERIMENTAL METHOD

2.1 Source Preparation

The ^{153}Sm source used in this experiment is produced by the neutron bombardment of isotopically enriched $^{152}Sm_2O_3$ in nuclear reactor, multipurpose Siwabessy reactor, Serpong-Indonesia. Samples for activity measurements were prepared by gravimetric method using a calibrated scales semi-micro balance. Three sets of ^{153}Sm samples were prepared from the initial stock solution. The first set is used for the absolute activity measurement using a 4πβ(LS)-γ coincidence counting method [9,10]. Three LS samples were prepared in 20 mL glass vial each contained 10 mL of Ultima Gold® cocktail solution. The second set of samples are used for the impurities test using a gamma spectrometer system. Three-point sources of ^{153}Sm were prepared for this set. For the last set, two samples were prepared in ampoules to calibrate the dose calibrator unit. Vials that are used are made by Wheaton, USA and are made of borosilicate glass, with a volume of 20 ml, height of 55 mm, outer diameter of 30 mm, and thickness of 1 mm. The point sources had a sample diameter of about 4 mm, and the weight varied between 10 and 15 mg. After drying under a heat lamp for about 30 minutes, they were then covered with a film of mylar with a ± 25 □g/cm^2 thickness. The samples was measured in the activity range of 10^3 Bq to 4.0 10^5 Bq. The density of the solution was measured as 1.033 g/ml.

2.2 Standardization Method

The coincidence counting method was used to determine the activity of ^{153}Sm. The general equation is shown below:

$$\rho_\beta = A\left[\varepsilon_\beta + \frac{(1-\varepsilon_\beta)(\alpha\varepsilon_{ce}+\varepsilon_{\beta\gamma})}{1+\alpha}\right] \quad (1)$$

$$\rho_\gamma = A\frac{\varepsilon_\gamma}{1+\alpha} \quad (2)$$

$$\rho_{\beta\gamma} = A\left[\frac{\varepsilon_\beta\varepsilon_\gamma}{1+\alpha}\right] + (1-\varepsilon_\beta)\varepsilon_c \quad (3)$$

where A is the activity of radionuclide, ρ_β,ρ_γ, and $\rho_{\beta\gamma}$ are the count rate for beta, gamma, and beta-gamma coincidence, respectively. The efficiency of beta, gamma, and beta gamma coincidence are represented respectively as ε_β, ε_γ, and ε_c. This basic equation was applied to the decay scheme of ^{153}Sm with condition that the activity is determined based on the coincidence events of beta and gamma within the 103 keV gamma energy window.

A 4πβ(LS)-γ coincidence counting system was used in this experiment. The system consists of two NaI(Tl) detector for gamma detection and two PMTs for beta detection. The schematic diagram for the system is shown in Fig. 2.

For each measurement of LS samples, three data files, two for the beta-ray detection channel and one for gamma-ray detection channel, are generated. The files were then used as input data on the offline data processing software. All analysis process, such as gamma gate and beta discrimination setting as well as the selection of coincidence events, can be done by using the software. Detail of system configuration and software architectures used for digital data processing together have been described in detail by K.B. Lee et al. [9].

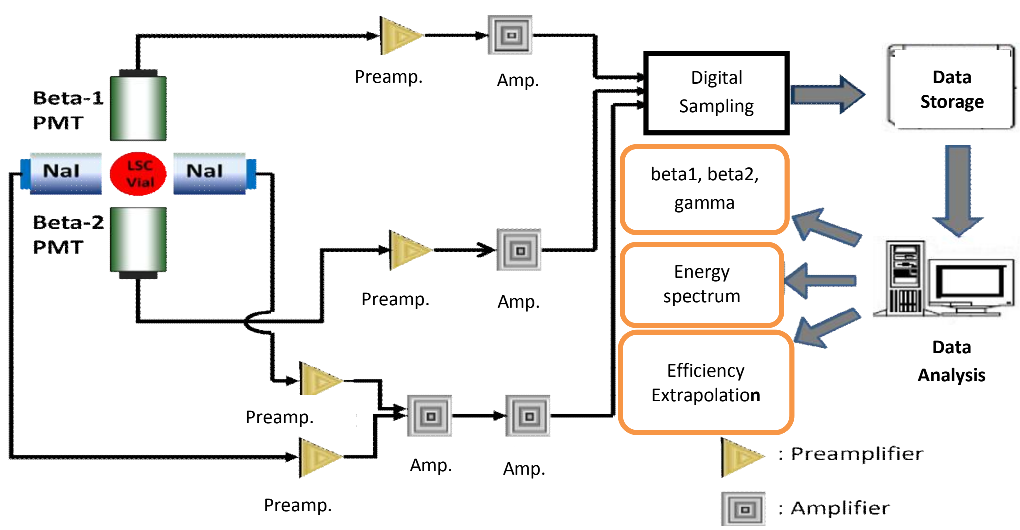


Fig. 2. The schematic diagram for the 4πβ(LS)-γ coincidence counting system

2.3 Impurity Measurement

The impurity of ^{153}Sm were measured by gamma spectrometry. The detector is an HPGe model GC1018 (Canberra, USA), which has a relative efficiency of 10.3% with an energy resolution of 1.69 keV FWHM at 1332.5 keV. The detector is equipped with a model 2002CSL pre-amplifier, and a Canberra model 2020 amplifier and operates at a bias voltage of +4500 V. Signals from the detector are processed by the Canberra gamma spectrum analysis system using GENIE 2000 software (Canberra Industries, USA). The source-to-detector distance was 25 cm. As described above, the gamma-ray spectrometry system was first calibrated using standard sources of ^{133}Ba, ^{152}Eu, and ^{241}Am, that have traceability to the SI. Three sets of measurements were made with a counting time of 60 minutes in each case. Impurities were also measured many times after 5, 10, and 15 times of ^{153}Sm half-lives' using gamma spectrometry.

3. RESULTS AND DISCUSSION

The result of the efficiency calibration by using ^{133}Ba, ^{152}Eu, and ^{241}Am standard sources is shown by the curve in Fig. 3. The energy used to make efficiency calibration curve as much as nine of energies, respectively 59.5409, 80.9979, 121.7817, 244.70, 344.28, and 356.013 keV [8]. This is done to improve the quality measurements for more accurate, precision and traceable. Efficiency calibration curve, estimated by fourth polynomial equation, founded the efficiency, y is = $-2E\text{-}12x^4 + 2E\text{-}09x^3 - 7E\text{-}07x^2 + 0.0001x - 0.0028$ with the correlation coefficient, $R^2 = 1$ and the fitting uncertainty of 1.5%.

Impurities check by gamma spectrometry system giving a result that the nuclide impurity is 0.001% of ^{155}Eu in ^{153}Sm at initial time. There are no other photon emissions detected on the ^{153}Sm source solution. Activity of ^{153}Sm was determined from efficiency extrapolation taking a plot the $\rho_\beta\rho_\gamma/\rho_c$ as a function of $(\rho_\gamma/\rho_c - 1)$ [9]. The extrapolation curves were obtained from the measurement at 103 keV gamma gate. The efficiency range of 80% - 90% shows a very linear trend and was used to extrapolate to 100% β-counting efficiency. The extrapolation results are shown in Fig. 4. The value of the uncertainty budgets for 4πβ(LS)-γ coincidence counting system is shown in Table 1, while the uncertainty values on the calibrating of Cap Intec dose calibrator is shown in Table 2.

A final activity results of 88.48 kBq/g at reference time was obtained with uncertainty 1.36% at k=2. The result was used as a reference to determine the calibration factor of Radioisotope Calibrator Capintec CRC-7BT which is the secondary standard instrument at PTKMR – BATAN. A factor 1.05 was obtained as the calibration factor of ^{153}Sm measurement with the Cap Intec CRC-7BT.

Table 1. Uncertainty budget for standardization of ^{153}Sm

Parameter	Relative standard uncertainty (%)
SD of counting	0.36
Weighing	0.05
Half-life of ^{153}Sm	0.02
Efficiency extrapolation	0.54
β-β resolving time	0.30
Combined standard uncertainty	0.68
Expanded uncertainty	1.36

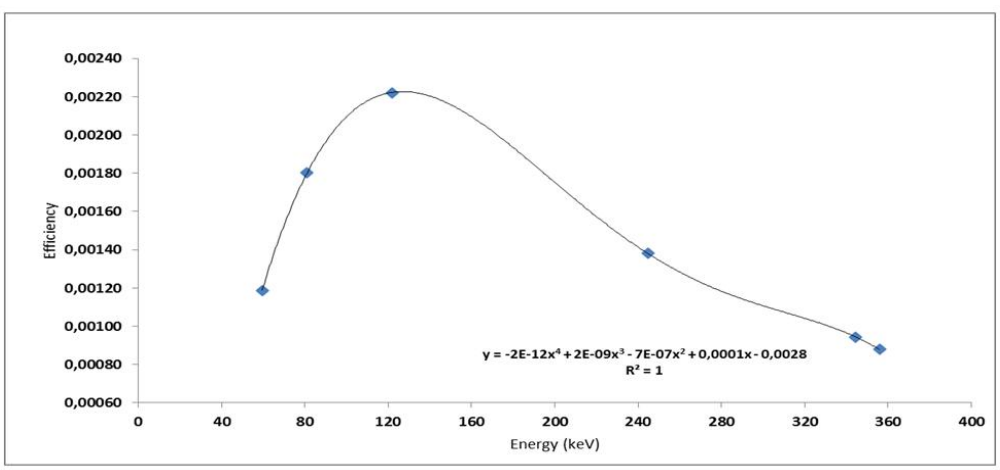


Fig. 3. Efficiency curve of gamma spectrometer system

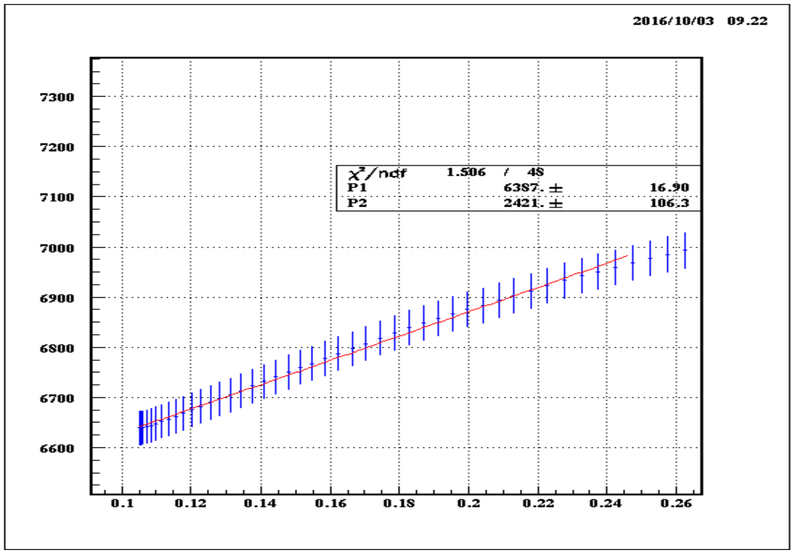


Fig. 4. Efficiency curve with superimposed fitting line for ^{153}Sm

Table 2. Uncertainty components for the R-value determined dose calibrator for ^{153}Sm

Source of uncertainty	Standard uncertainty components (%)	
	^{153}Sm	Type
Standard source	0.68	B
Half-life of sample	0.02	B
Statistics of counting	0.40	A
Detector response	1.155	B
Accuracy of reading	1.732	B
Repeatability	0.577	B
Non-linearity	0.35	B
Mass	0.05	B
Combined standard uncertainty	2.31	B
Expanded uncertainty (k = 2)	4.62	B

4. CONCLUSION

The radionuclide ^{153}Sm has been standardized using the 4πβ(LS)-γ coincidence counting with digital sampling method. The result was used as primary standard to calibrate the secondary standard instrument at PTKMR-BATAN, and we found the calibration factor for Capintec CRC-7BT dose calibrator is 1.05 with 4.62% of the expanded uncertainties. PTKMR-BATAN, can calibrate the equipment in field of nuclear medicine using a short half-life of ^{153}Sm standard source, so that the application of nuclear technology in health sectors can be done with the secure and safe for workers, communities and environment.

ACKNOWLEDGEMENTS

The authors would like to thank the Government of Indonesia , especially the leaders of all ranks BATAN which has provided the opportunity to successfully complete this study.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

REFERENCES

1. Gatot Wurdiyanto, Hermawan Candra. The standardization methods of radioactive sources (^{125}I, ^{131}I, ^{99m}Tc, and ^{18}F) for calibrating nuclear medicine equipment in Indonesia. Journal of Physic: Conference Series. 2016;694:012060.
2. Schotzig U, Schonfeld E, Gunther E, Klein R, Schrader H. Standardization and decay data of ^{153}Sm. Applied Radiation and Isotopes. 1999; 51:169–175.
3. Bowles NE, Woods SA, Woods DH, Jerome SM, Wood MJ, De Lavison P, Lineham S, Keightley J, Poupaki I. Standardization of ^{153}Sm. Applied Radiation and Isotopes. 1998; 49:1345–1347.
4. Dziel T, Broda R, Ziemek T, Muklanowics A, Listkowska A. Standardization of Sm-153 solution by absolute methods. Appl. Radiat. Isotope. 2014;87:19-23.
5. Finlay LG, Mason MD, Shelley M. Radioisotopes for pallation of metastatic bone

cancer; Systematic review. Lancer Oncol. 2005;6:392-400.
6. Lam MG, de Klerk JM, van Rijk PP, Zoonenberg BA. Bone seeking radiopharmacaeticals for pallation of pain in cancer patients with osseous matastases. Anticancer Agents Med. Chem. 2007;7:381-397.
7. Gkialas I, Iordanidou L, Galanakis I, Giannnopoulos S. The use of radioisotopes for pallaiation of metastatic bone pain. J. BIJON. 2008;13:177-183.
8. Bé, et al. Table of radionuclides. Monographie BIPM-5 Vol. 1 – A=1 to 150, Bureau International des Poids et Mesures, Sèvres; 2004.
9. Lee KB, et al. Application of digital sampling techniques for 4πβ(LS)-γ coincidence counting. Nucl. Instrum. Methods. 2011;626-627:72-76.
10. Casette P, Tartes I. Experimental study of the influence of the counter and scintillator on the universal curves in the cross-efficiency method in LSC. Appl. Radiat. Isotope. 2014;87:32-37.
