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Preparation and Evaluation of Mixed Gamma Sealed Source for Calibration Purposes

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ABSTRACT

The sealed sources containing a mixture of 370±5% kBq (~10±5% µCi) ^{137}Cs, 740±5% kBq (~20±5% µCi) ^{60}Co, and 740±5% kBq (~20±5% µCi) ^{152}Eu based on inorganic exchangers, preparations were made, standards were met, and quality checks were passed satisfactorily. ZrSiW matrix retaining ^{137}Cs, ^{60}Co, and ^{152}Eu, the core of the sealed source for the appropriate radioisotope was cleaned and dried before use. A circular disc-shaped acrylic canister was filled with a calibrated quantity of the matrix. Before being sealed, the source underwent quality control inspections, followed by radiometric standardization and encapsulation, before being placed on the local market to calibrate radiation detection and measurement equipment.

INTRODUCTION

A sealed radioactive source is a radioactive material that has been enclosed in a metallic or another container without interaction between the radioisotope substance and the environment outside [1]. Radioactive sources with known activities and radionuclides must be used to precisely calibrate all detector systems for monitoring radioactivity in the various application areas.

In single- or mixed radionuclide-sealed sources, radioisotopes including ^{241}Am, ^{133}Ba, ^{182}Ta, ^{57}Co, ^{226}Ra, ^{110m}Ag, ^{137}Cs, ^{54}Mn, ^{22}Na, ^{60}Co, and ^{152}Eu are used to calibrate gamma-ray spectrometers [2]. Depending on the source usage, the activity of a sealed source can range from kilo Becquerels for calibration sources to thousands of Curies for industrial usage [3, 4].

For crucial applications in several sectors, there are many various sizes, shapes, and activities of sealed radioactive sources. They are utilized in the industrial sector for mass flow measure, flow meter detection, the creation of plastic pipes shrinking, radiograph for crack detection in steel tubes, gauging liquid and solid thickness, evaluation of mineral resources, food sterilization, detection of smoke, and many other tasks [4-5]. They are frequently used in medical applications such as bone density assessment, eye and prostate cancer treatment, teletherapy, and brachytherapy to treat malignant diseases. [4, 6]. They are usually used in research to calibrate radiation detectors and spectrometers and for elemental analysis, like XRF and neutron-activation studies.

Standard radionuclide sources generating multi-gamma rays over a broad energy range, such as ^{152}Eu and ^{154}Eu sources, are chosen for determining multiple efficiency values (at various energies) per measurement [7]. The lack of accurate coincident summation and the merging of neighboring gamma energy peaks, which can happen with multi-photon sources, makes ordinary single-photon radionuclide sources, such ^{51}Cr, and ^{137}Cs sources, beneficial for high-accuracy [8].

Different methods can be used to create the core of sealed sources, including pipetting drops of a radioactive solution that are then dried by IR irradiation [9], as well as other deposition procedures like electrodeposition and sublimation/chemosorption [4], freeze-drying deposition, and others [10]. It is possible to feed the radioactive nuclide(s) onto inorganic or organic ion exchangers [11].

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This research aims to make sealed sources based on inorganic sorbents using simple techniques that don't require any special knowledge (such as electrodeposition, implantation, distillation, electrospraying, etc.) and where the level of radioactivity is easy to control.

This project also seeks to complete the entire preparation operations using commercially accessible and easily made materials with trustworthy qualities under mechanical, thermal, and radiation exposure circumstances that are more severe than those encountered in typical work and use.

EXPERIMENTAL

Chemicals and equipment

The present experiments employed only AR-grade chemicals. To measure radioactivity-level, a high-purity germanium coaxial detector (HPGe) of the GX2518 model was used in conjunction with a multichannel analyzer (MCA) of the "Inspector 2000" type, Canberra Series, manufactured in the USA. All solutions were prepared with distilled water. Also, all washing was done with distilled water.

Synthesis of zirconium silica tungstate (ZrSiW)

To prepare ZrSiW gel, 50 ml of 0.04 mol L^{-1} sodium tungstate (Na2WO4.2H2O) and 50 ml of 0.28 mol L^{-1} sodium silicate (Na2SiO3) were combined. The tungstate/silicate mixture was then added dropwise to 50 ml of 0.04 mol L^{-1} heated zirconium oxychloride (ZrOCl2.8H2O, dissolved in 0.2 mol L^{-1} at pH 8 for about 30 min. The precipitate was washed using distilled water several times until a constant pH was obtained. ZrSiW was then dried at 50 °C for 48 hours before being ground into particle size measuring 0.12 to 0.24 mm [12].

Batch distribution investigations

Individually, the distribution pattern of ^{134}Cs, ^{60}Co, and ^{152}Eu was examined between 10 ml (V) 1×10^{-4} M of the radioisotope in various concentrations of HCl acid solution (10^{-4}, 10^{-3}, 10^{-2}, 0.1, 0.5, and 1 M) and 0.1 gm of ZrSiW gel (m). After shaking the two stages all night, they were allowed to settle down for the radioisotope experiment.

By replacing counting rates in 1 ml of the appropriate radioisotope solution without the adsorbent and after equilibrium (A_o and A_e, respectively), the distribution coefficient values were calculated in the equation.

$$K_d = \frac{A_o - A_e}{A_e} \times \frac{V}{m} \qquad \text{mL/g}$$

Where,

A_o = beginning activity, A_e = activity after equilibrium, V = solution volume (10 mL), and m = ion exchanger weight (0.1 g).

The attaining adsorption equilibrium time of ^{60}Co, ^{134}Cs, and ^{152}Eu was determined by calculating the distribution coefficient values for each isotope as a function of time at pH four and at different time intervals (1, 4, 8, 12, 18, 25, 30, 35, 40, 45 and 50 hours) in the same other conditions mentioned above.

^{137}Cs, ^{60}Co and ^{152}Eu sealed source core preparation

To 0.5 g of ZrSiW as a cation exchanger, 20 ml of a pH 4 mixed solution containing 370±5% kBq (~10±5% µCi) ^{137}Cs, radionuclide purity 99.9%, 740±5% kBq (~20±5% µCi) ^{60}Co, radionuclide purity 99.9%), and 740±5% kBq (~20±5% µCi) ^{152}Eu, radionuclide purity 99.9%. A water bath thermostat shaker set to 25 °C was used to shake an intermediate for 24 h. After that, the liquid phase was separated and the radioactivity in it was examined. ZrSiW was dried out at 50 °C for one day, washed with 30 ml distilled water, and finally, dried once more.

Packing and encapsulation of the prepared sealed source core

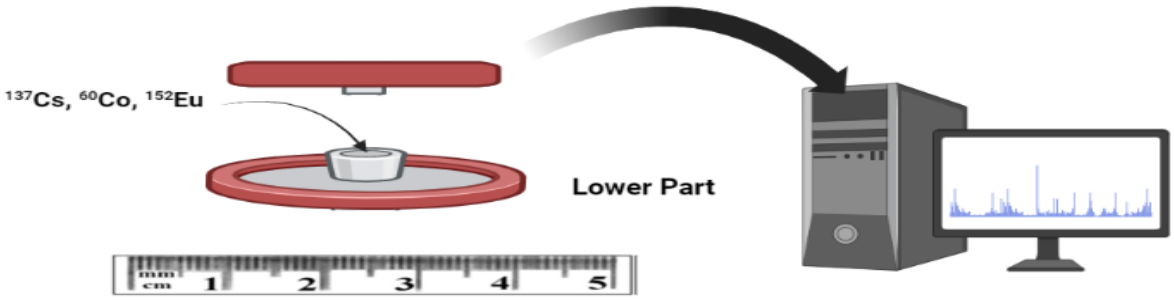

Fig. (1): Schematic diagram of the capsule components used to construct the mixed ^{137}Cs, ^{60}Co, and ^{152}Eu sealed source.

A schematic diagram representing the capsule constituents used for the preparation of the desired sealed source is shown in Figure 1. A suitable amount of ZrSiW gel loaded with the desired radioisotopes was filled in the cylindrical chamber with a 3 mm diameter located at the button part of the capsule, as shown in Figure 1. A puddle of epoxy glue was used to cover the space above the filled matrix, with the hardener diglycidyl ether of bisphenol A+ 4,4-diamino diphenyl methane to secure the core material to a specific shape and prevent it from spreading out. The adhesive slurry was allowed to solidify for 1 hour on the inner disc. The inner and outer walls of the disc were filled in any gaps between the two discs caused by minute surface defects with a thin coat of epoxy resin before compression to stop any potential radioactive leaks. The extra epoxy resin was swiftly replaced before compaction caused it to harden. To confirm that there had been no change in the produced sealed source activity as a result of any potential loss or geometric change of the radioactive matrix throughout the production steps indicated above, the radioactivity-levels of the sealed sources were measured once more. Before performing the quality control procedures discussed below, the contained sources were allowed to solidify to confirm that the sticky component had hardened completely.

Quality assurance tests

Wipe test

This test takes place through swabs of the enclosed sources with cotton wool soaked in acetone and then monitoring for radioactive emissions to check for surface contamination.

Impact test

The sources were lowered onto a solid, horizontal surface from a height of nine meters, and their deformations were then visually examined [13].

Leakage test

The source was dipped 5 cm below the water level in a 95°C water bath. During a 5-minute interval, the water bath was inspected for any bubble emanation from the source [14,15].

Leaching test

For the leaching test, the sealed source was submerged in a 50°C water bath for 5 hours. The water was then examined for radioactive leaks after that. [16].

Radioactivity measurement

Additionally, the radioactivity of the produced sealed source was measured to ensure that no changes in activity happened following the quality assurance tests.

Finalizing the manufacturing process

The produced sealed source was then covered with a label bearing the radiation warning symbol, the radionuclide it contained, as well as the activity and calibration date of the radioactivity.

RESULTS AND DECISION

^{137}Cs, ^{60}Co and ^{152}Eu uptake on ZrSiW

The distribution coefficient of ^{134}Cs, ^{60}Co, and ^{152}Eu at 1×10^{-4} M of each between HCl acid medium and ZrSiW gel is shown in Figure 2. At pH 3.0, ^{137}Cs have the greatest K_d value (2700 mL/g). Also, at pH 3.0, the highest K_d values for ^{60}Co and ^{152}Eu are 890 and 950 mL/g, respectively. It is recognized that cesium always occurs as a Cs^+ ion in an aqueous solution [17]. It is known that heteropolyacids exhibit high selectivity for Cs^+ ions through a chemical sorption mechanism involving structural modifications in the sorbent [18], which may be the primary uptake mechanism in the acidic pH range (especially for the heteropolyacid salt of the gel material).

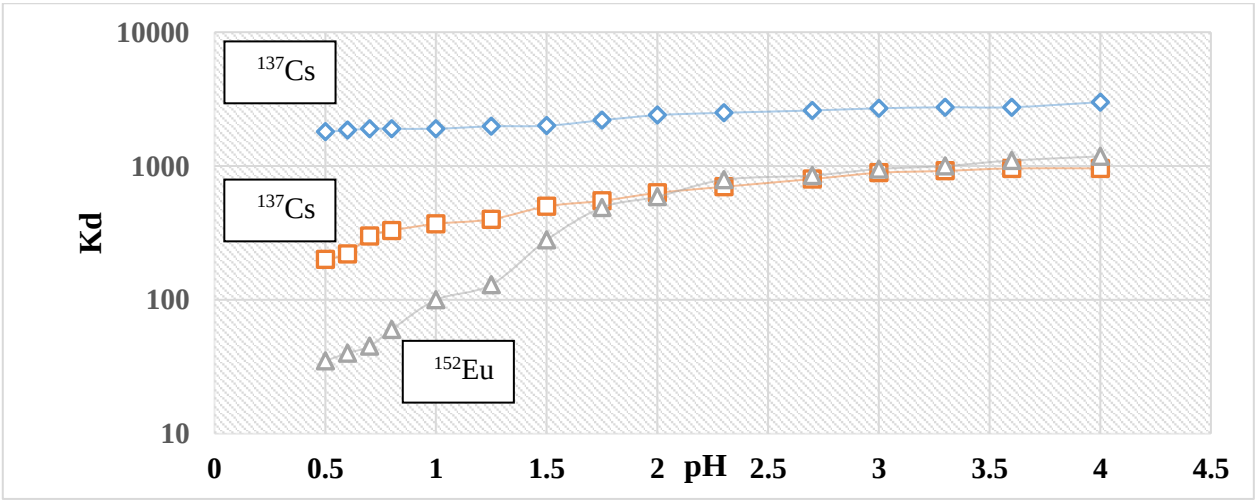

Fig. (2): The distribution coefficient of ^{137}Cs, ^{60}Co, and ^{152}Eu (1×10^{-4} M of each between HCl acid medium and ZrSiW gel.

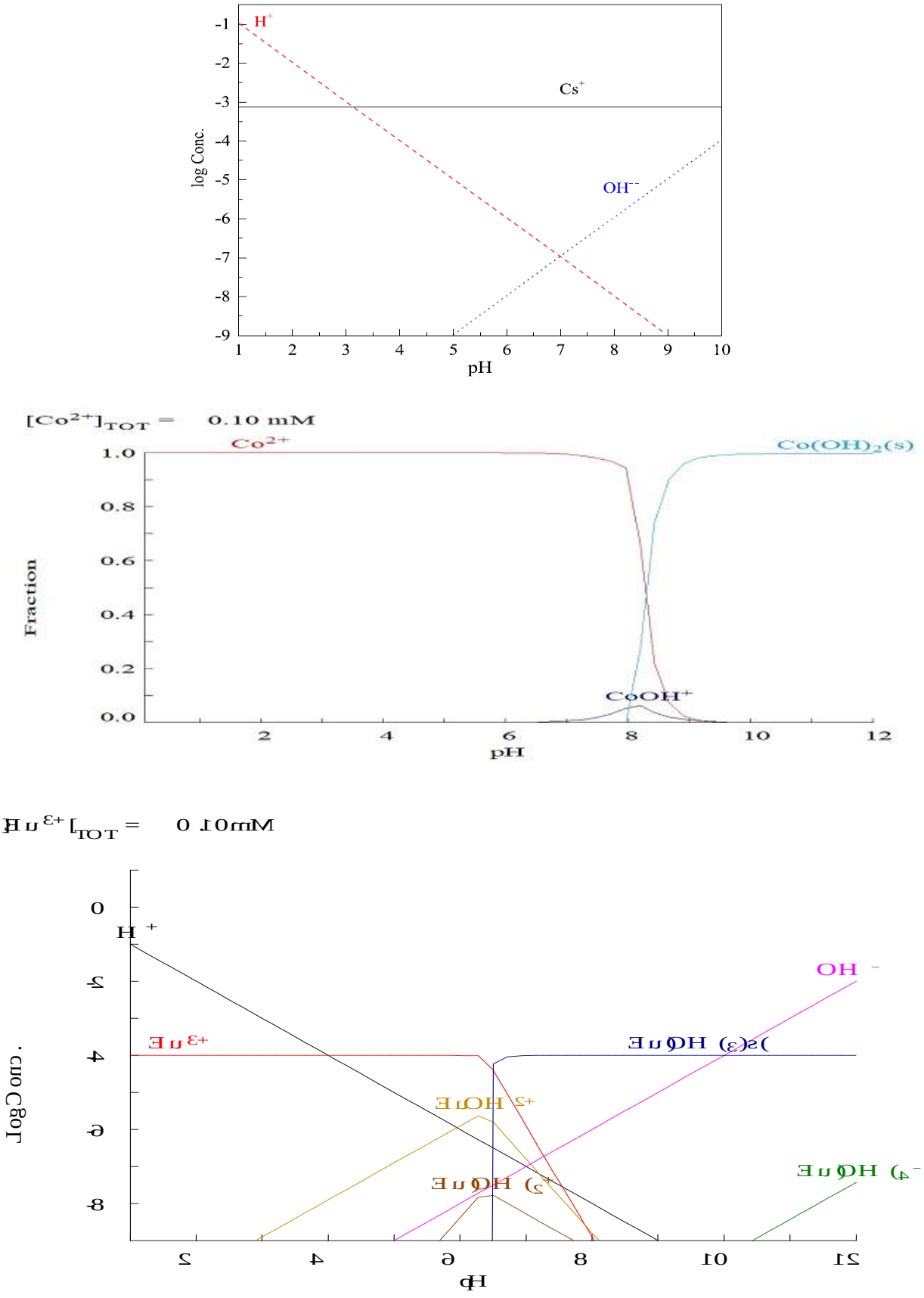

Fig. (3): Chemical species distribution of Cs^{+}, Co^{2+}, and Eu^{3+} species ions as a function of pH.

To further investigate ^{137}Cs, ^{60}Co, and ^{152}Eu with ZrSiW gel matrix, pH 3 (equivalent to $1X10^{-3}M$ HCl) was selected as the ideal condition. The speciation of Cs+, Co2+, and Eu3+ ions in an aqueous solution at various pH values, 10-4 mol/L of metal ion concentration, and ambient temperature was carried out using the MEDUSA software [19], which offers information about the distribution of species as a function of pH as seen in Fig. 3. It is clear that Co^{2+} predominates in an HCl acid solution, while $Co(OH)_2$ is present in lesser amounts. The Co^{2+} content is above 98 % at pH 3.0. In the HCl acid medium, Eu (III) is the dominant species, as opposed to $EuCl^{2+}$, which has a low concentration (15%) at pH five and then drops to a very low value at pH 3.0). Eu^{3+} makes up more than 98% of the solution at pH 3.0.

Co^{2+} and Eu^{3+} uptake was attributed to surface complexation via ion exchange and/or electrostatic attraction with the ZrSiW gel surface, depending on pH value. At pH 3.0, electrostatic attraction is expected between the positively charged Co(II) and Eu(III) species (Figure 3) and the negatively charged ZrSiW gel surface [20-22].

Characteristics of the material used in manufacturing the sealed source

The base material of the core

In general, organic ion exchangers are less chemically stable than inorganic sorbents in the presence of ionizing radiation [23, 24]. Since H_2, CO_2, CO, O_2, CH_4, SO_2, N_2, N_2O, and NO gases are created during radiolysis of the organic exchanger matrix, pressure increases results in a crack in the built sealed source, and the risk of explosion and fire increases when using high radiation doses [25-26]. On the other hand, it has been discovered that inorganic sorbents containing radionuclides with high-specific activity are stable over time [27, 28].

ZrSiW gel matrix might be employed effectively as the main component of $^{188}W/^{188}Re$ radioisotope generators because of its high chemical stability toward radiation. Also, it had good radiochemical stability and maintained its sorption capability even after prolonged radiation exposure [12, 29].

Epoxy adhesive

Since epoxy compounds fulfill the requirement for radiation stability as they have excellent mechanical and sticking strength, durability in alkaline and acidic solutions, and negligible leach and gas emission even with using high activity, they are promising materials for the immobilization of radioactive waste as opposed to organic ion-exchange resins [30, 31].

Aromatic amine-cured epoxy resins have greater thermal stabilities and are more radiation-resistant because epoxy diamines create surfaces with metals with superior mechanical characteristics. Because each of their amine groups has two hydrogen atoms, which make up the reactive component of the hardener, primary amines having NH2 groups are favored. 4,4-diamino diphenyl methane was used as a hardener in this work [32, 33].

Acrylic material

Acrylic is a beneficial material *for a wide range of applications. It has desirable physical properties* such as (i) mechanical *tensile strength of* 75 mpa and flexural *strength of* 115 mpa (ii) thermal properties where 40 ^0C is the minimum service temperature while 80 ^0C is the maximum service temperature softening point of 110 ^0C and linear expansion 77 x 10^{-5} (iii) acrylic is very resistant to temperature changes *and* (iv) acrylic plastic has outstanding chemical resistance [34].

It is used to build sealed radioactive sources due to its ideal properties of having excellent mechanical and thermal capabilities, resistance to both corrosion and chemicals, and radiation stability.

By building a framework, the core chamber to be in the needed geometrical and dimensions and packing it with the matrix loaded with the required radioactivity, the core preparation procedures described in this work can be used to produce a sealed source of different shapes that is suitable for the necessary application [35-37]. Additionally, the entire sealed source assembly (capsule form) is modifiable as needed [38,39].

Monitoring radioactivity and the prepared sealed source quality

Using calibrated weights in the order of milligrams of ZrSiW matrix, a sealed source containing a mixture of 370±5% kBq (~10±5% µCi) ^{137}Cs, 740±5% kBq (~20±5% µCi) ^{60}Co, and 740±5% kBq (~20±5% µCi) ^{152}Eu was prepared. This mixture was shown via radiometric assessment using the efficiency-calibrated MCA.

There was no evidence of a radioactivity loss or change in the prepared sealed source's measured activity throughout any phase of the preparation process or during any of the quality control tests. A high-purity germanium coaxial detector (HPGe) of the GX2518 model was used in conjunction with a multichannel analyzer (MCA) of the "Inspector 2000" model, Canberra Series, built in the USA, to count the prepared sealed source for roughly five minutes. Figure 4 shows the gamma-ray spectrum of ^{137}Cs, ^{60}Co, and ^{152}Eu sealed source at 5 cm source – HPGe detector distance. The spectrum includes the main gamma energy peaks of the used radioisotopes. Table (1) represents the main gamma energy peaks of the prepared sealed source.

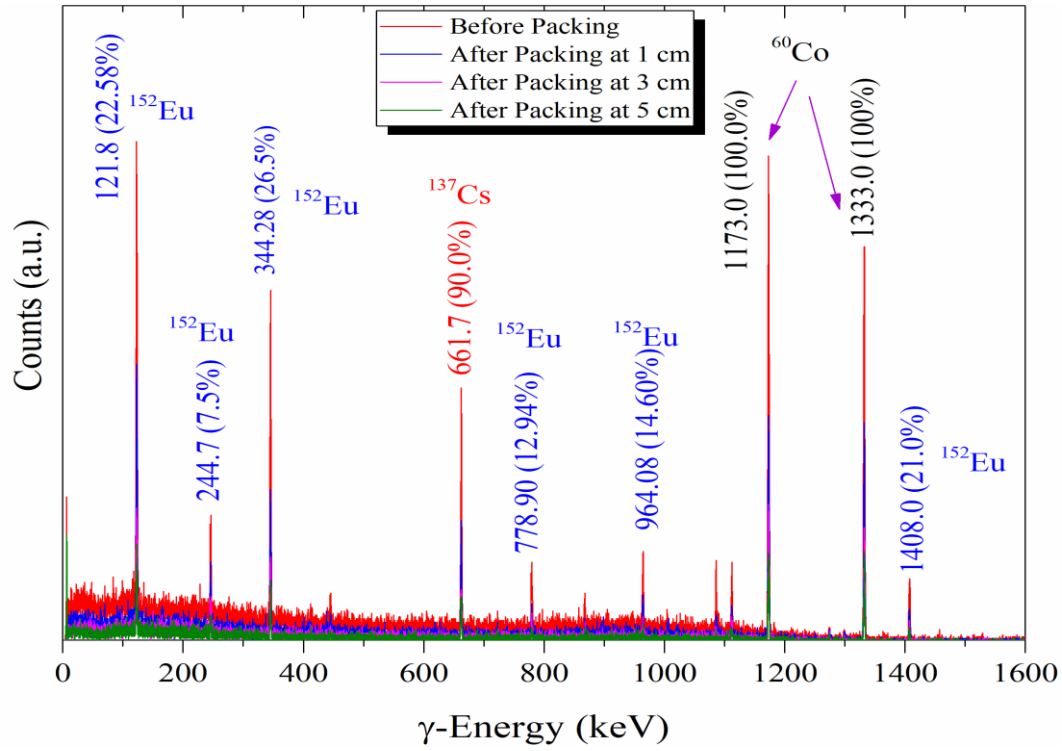

Fig. (4): Measured gamma-spectrum of the prepared sealed source.

Table (1): Characteristics and gamma-energy measurements of the constructed sealed source

Nuclide	Half-life (y)	Calibration Date	Activity (MBq)	E_γ (keV)	Iγ (%)
^{60}Co	5.27	Sep. 2019	24.0	1173.228 1332.492	100.0 100.0
^{137}Cs	30	Sep. 2019	28.0	661.6570	90.0
^{152}Eu	13.54	Sep. 2019	28.0	121.8 244.7 344.28 778.90 964.08 1408.013	22.58 7.50 26.5 12.94 4.60 21.0

REFERENCES

[1] McGill University, Montreal, Quebec, Canada (2005), Radiation Safety Policy Manual, 5.1th ed., Environmental Safety Office, 200.

[2] B. Jarosievitz, (1996) Gamma-spectrometry of extended sources for analyzing environmental samples,. 1

[3] H. Wyllie, (1989) The preparation of radioactive sources with radioactivities of less than 110 kilobecquerels, in, Australian Nuclear Science and Technology Organisation.

[4] P. Techniques, (2006) Quality Control of Sealed Radioactive Sources of Palladium-103, Iodine-125, Iridium-192, and Ytterbium-169, in, IAEA, VIENNA.

[5] DOE (Department of Energy), (2000) The Current and Planned Low-Level Waste Disposed Capacity Report, Revision 2, Office of Environmental Management, USA.

[6] C. Mathew, M. Majali, S. Balakrishnan, (2002) A novel approach for the adsorption of iodine-125 on silver wire as matrix for brachytherapy source for the treatment of eye and prostate cancer, Appl. Radiat. Isot., 57, 359-367. 4

[7] S. Chu, The Lund/LBNL (1999) nuclear data search, http://nucleardata. nuclear. lu. se/nucleardata/toi/index. asp.

[8] G. Gilmore, (2008) Practical gamma-ray spectroscopy, John Wiley & Sons.

[9] A. El Abd, M. Mostafa, M. El-Amir, (2012) Production of ^{152,154} Eu mixed sources for calibrations of gamma-ray spectrometers, J. Radioanal. Nucl. Chem., 293, 255-260.

[10] J. De Sanoit, B. Leprince, C. Bobin, J. Bouchard, (2004) Freeze-drying applied to radioactive source preparation, Appl. Radiat. Isot., 61, 1391-1395.

[11] B. Zaitsev, A. Grivkova, M. Glazunov, (1962) The use of ion-exchange materials for the preparation of low activity radiation sources, The Soviet J. Atomic Energy, 11, 1075-1078.

[12] M. Aydia, H. El-Said, A. El-Sadek, K. El-Azony, (2018) Preparation and characterization of zirconium silico^{188}W-tungstate as a base material for 188W/188Re generator, Appl. Radiat. Isot., 142, 203-210.

[13] D.A. Martinez, C.L. Abeyta, L.E. Leonard, J.A. Tompkins, S.J. Leonard, (2002) Development and Certification of a Special form Capsule for Sealed Sources to Facilitate Transportation and Storage as Special form Material, Los Alamos National Laboratory-OSRP documents, in Los Alamos National Laboratory-OSRP Documents, LA-UR-02-433. 11

[14] I.O.f. (1979) Standardization, Sealed Radioactive Sources-Leak Test Methods, International Organization for Standardization.

[15] ISO, (1999), Radiation Protection-Sealed Radioactive Sources-General Requirements and Classification, ISO 2919, 2nd edition.

[16] J. Tompkins, (2003) Control and Disposition of Sealed Sources: Relative to a Campus Setting, in Los Alamos National Laboratory-OSRP Documents, LA-UR-03-1016. 13

[17] G. Schweitzer, L. Pesterfield, (2010) The Aqueous Chemistry of the Elements, 434 Oxford University Press, New York.

[18] Y. Park, W.S. Shin, S.-J. Choi, (2013) Ammonium salt of heteropoly acid immobilized on mesoporous silica (SBA-15): an efficient ion exchanger for cesium ion, Chem. Eng. J., 220, 204-213.

[19] Puigdomenech, I., Zagorodni, A., Wang, M., (2010) Program MEDUSA (Make equilibrium diagrams using sophisticated algorithms). Royal Institute of Technology. Inorg. Chem. 10644.

[20] W. Xiangke, D. Wenming, D. Xiongxin, W. Aixia, D. Jinzhou, T. Zuyi, (2000) Sorption and desorption of Eu and Yb on alumina: mechanisms and effect of fulvic acid, Appl. Radiat. Isot., 52, 165-173.

[21] X. Wang, W. Dong, G. Wang, Z. Tao, (2002) Sorption and desorption of Co (II) on alumina:: mechanisms and effect of humic substances, Appl. Radiat. Isot., 56, 765-771.

[22] X. Wang, D. Xu, L. Chen, X. Tan, X. Zhou, A. Ren, C. Chen, (2006) Sorption and complexation of Eu (III) on alumina: Effects of pH, ionic strength, humic acid and chelating resin on

kinetic dissociation study, Appl. Radiat. Isot., 64, 414-421.

[23] M. Qureshi, K. Varshney, Inorganic ion exchangers in chemical analysis, CRC press Florida, 1991.

[24] K. Pillay, (1986) A review of the radiation stability of ion exchange materials, J. Radioanal. Nucl. Chem., 102, 247-268.

[25] A. Kazanjian, M. Killion, Radiation effects on amberlite IRA-938 and bio-rad AG MP-50 ion exchange resins, in, Rockwell International Corp., 1982.

[26] A.R. Kazanjian, J. Stevens, Radiation effects on Dowex MSC-1, Amberlite 252, and Duolite C-264 ion exchange resins, in, Rockwell International Corp., 1983.

[27] L. Zsinka, L. Szirtes, V. Stenger, (1970) Investigation of the resistance of chromium phosphate ion exchanger against radiation, in, Inst. of Isotopes, Budapest.

[28] L. Zsinka, L. Szirtes, J. Mink, A. Kalman, (1974) Effect of γ-radiation on various synthetic inorganic ion exchangers, J. Inorg. Nucl. Chem., 36, 1147-1152.

[29] H. El-Said, A. El-Sadek, M. Aydia, K. El-Azony, (2018) Zirconium silicotungstate matrix as a prospective sorbent material for the preparation of $^{113}Sn/^{113m}In$ generator, J. Radioanal. Nucl. Chem., 317,1341-1347.

[30] P. Platonov, Y.I. Shtrombakh, O. Chugunov, N. Lobanov, V. Aleksandrov, V. Alekseev, V. Semochkin, V. Denisov, V. Kochkin, (2002) Immobilization of radionuclides in irradiated reactor-graphite blocks, At. Energ., 92, 475-481.

[31] Y.I. Shtrombakh, P. Platonov, N. Lobanov, O. Chugunov, V. Aleksandrov, O. Zinov'ev, (2005) Epoxy compounds for immobilizing radioactive wastes, At. Energ., 98, 331-333.

[32] A. Roche, J. Bouchet, S. Bentadjine, (2002) Formation of epoxy-diamine/metal interphases, Int. J. Adhes. Adhes., 22 431-441.

[33] C. Augustsson, NM epoxihandbok, Nils Malmgren AB, 2004. 29

[34] S. Ammar, I.W. Ma, K. Ramesh, S. Ramesh, (2019) Polymers-based nanocomposite coatings, in: Nanomaterials-Based Coatings, Elsevier, 9-39.

[35] International Atomic Energy Agency (IAEA), 2000, Vienna, Categorization of Radiation Sources, IAEA-TECDOC-1191.

[36] EPA (U.S. Environmental Protection Agency), 2002., Industrial Applications of Sealed Radiation Sources and Alternative Non Nuclear Technologies, Final Report, Contract 68-D-00-210, Work Assignment E210 1-16, Office of Radiation and Indoor Air, Washington, DC.

[37] M. Mostafa, M. El-Amir, (2008) Preparation of ^{137}Cs and ^{60}Co sealed sources based on inorganic sorbents for radiometric calibration purposes, Colloids and Surfaces A: Physicochemical and Engineering Aspects, 317, 687-693.

[38] International Atomic Energy Agency (IAEA), 2004, Vienna, Management of Disused Long Lived Sealed Radioactive Sources (LLSRS), IAEA TECDOC-1357.

[39] R.S. Hassan, M.A. Eid, A.A. El-Sadek, Muhammad S. Mansy, (2019) Preparation, characterization and application of Mg/Fe Hydrotalcite as gamma sealed source for spectroscopic measurements, Applied radiation and isotopes 151,74.