



Preparation of ^{134}Cs sealed source based on WO_3 adsorbent for radiometric calibration purposes

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ABSTRACT

Cesium-134 artylon sealed source based on inorganic adsorbent (as a core) was easily prepared. The sealed source was in the form of an artylon disk constituted of two halves; base and top. Cesium-134 was adsorbed by equilibration of $^{134}\text{CsCl}$ solution with WO_3 adsorbent at pH 4 for 24 h at 25 ± 1 °C. Then, the liquid phase was decanted and checked for the remaining activity. Thereafter, the ^{134}Cs -loaded WO_3 was dried at 100 °C for 24 h. A definite weight of the ^{134}Cs -loaded WO_3 adsorbent was placed in the central cavity of the base half-disk. Then the two half-disks were assembled using a commercial epoxy adhesive. The activity values of the sealed source calculated by using the absolute efficiency curve and DETEFF software were found to be closest (7.81×10^{-2} and 7.70×10^{-2} μCi , respectively) at a 5 cm source-detector distance. After encapsulation and radiometric standardization, the sealed source was checked radiometrically before release to the local market for calibration purposes of radiation measurement and identification instruments.

1. Introduction

Radioisotopes that are fully encapsulated in containers without contact with the outside are termed sealed sources [1]. Depending on radioisotope and activity level, sealed sources can be used in a wide- variety applications. According to the activity level, sealed sources can be classified as [2]:

1. Low-radioactivity sources (μCi 's). Examples: those containing ^{129}I and other containing a mixture of radioisotopes (e.g., $^{45}\text{Mn}/^{57}\text{Co}/^{65}\text{Zn}/^{137}\text{Cs}/^{155}\text{Eu}$) that are used for the calibration of radiometric instruments.
2. Intermediate-radioactivity sources (mCi 's): Such sources may be permanently installed in instruments. Examples: those containing ^{137}Cs for liquid scintillation counters and these containing ^{63}Ni for electron capture detectors of gas chromatographs.

3. High-radioactivity gamma sources: usually have radioactivities ranging from Curies to several thousand Curies. Examples: Those of ^{192}Ir used to inspect cracks in steel pipes and those containing ^{60}Co and ^{137}Cs that are used for food irradiation and sterilization of medical supplies.

Standard sources of radionuclides emitting multi-gamma rays in a wide energy range are preferred for determination of several efficiency values (at different energies) per a measurement e.g., ^{152}Eu and ^{154}Eu sources [3]. However, the use of standard sources of single-photon radionuclides, e.g., ^{51}Cr and ^{137}Cs sources, achieves only determination of a single efficiency value per a measurement, such sources are advantageous for the high accuracy measurements because of the absence of true coincidence summation and overlapping of the adjacent energy peaks which may present in the case of multi-photon sources [4].

The core of a sealed source can be produced by pipetting drops of the radioactive solution, which are then dried by IR irradiation [5] or by other deposition methods such as electrode deposition and sublimation/chemisorption [6], freeze drying deposition [7].

The radioactive nuclide(s) can be loaded onto organic or inorganic ion-exchangers e.g., ^{137}Cs was loaded onto sulfophenol formaldehyde cation exchanger [8] and onto 6-tungstocerate(IV) ion exchanger. Chemical precipitation can be used to form the radioactive core, e.g., coprecipitation of ^{60}Co and $\text{Al}(\text{OH})_3$ [9]. Poly (acrylamide-acrylic acid)-montmorillonite composite [P(AAm-MA)-M] was synthesized and used for preparing ^{60}Co / ^{65}Zn / $^{134,137}\text{Cs}$ mixed sealed source. The synthesis procedure was based on γ -induced polymerization of polyacrylamide (PAAm) in the presence of acrylic acid (AA), clay minerals (montmorillonite, M), and *N,N'*-methylenebisacrylamide (NMBA) as a cross-linker [1]. A ^{137}Cs -loaded sealed source was prepared by contacting silicotitanate gel with a ^{137}Cs solution at pH 7 for 48 h at 25°C, drying at 100°C for 24 h, placing as a radioactive core inside a central cylindrical cavity in a base Perspex half-disc, and assembling a top Perspex half-disc with the base one using epoxy adhesive. The sealed source prepared was standardized using a γ -ray spectrometer and various calculation methods including DETEFF code and different ways of the efficiency transfer concept. For higher accuracy, the effect of the HPGe detector contamination background at different measurement distances was studied. Such prepared sealed source was found to have a ^{137}Cs activity of 0.98 ± 0.06 kBq [10].

2. Material and methods

2.1 Reagents

All chemical reagents used in this work were of AR grade. Multichannel analyzer coupled with a high-purity germanium (HPGe) coaxial detector of "Model 2000", Canberra Series, made in USA, was used for radiometric assay and identification of gamma-ray emitters. It was calibrated using a mixed sealed source of the radioisotopes: ^{155}Eu (86.5 and 105.3 keV) / ^{57}Co (122.1 and 136.5 keV) / ^{137}Cs (662 keV), ^{54}Mn (834.8 keV) / ^{65}Zn (1115.5 keV).

2.2 Radiotracer stock solution

To obtain ^{134}Cs radiotracer, samples of CsCl (0.20 g each) were wrapped in cleaned aluminum foils and placed into aluminum can. Aluminum can was then tightly sealed by cold welding, passed a water-proof test and subsequently irradiated in the Egyptian Second Research Reactor (ETRR-II) for 4 h at a thermal neutron flux of 1×10^{14} n.cm⁻². s⁻¹ to obtain ^{134}Cs radionuclide via $^{133}\text{Cs}(n,\gamma)^{134}\text{Cs}$ nuclear reaction. The irradiated CsCl was dissolved in distilled water. The necessary dilutions were performed to carry out batch uptake studies with 1×10^{-4} M $^{134}\text{CsCl}$.

2.3 Preparation of ^{134}Cs sealed source

10 mL of ^{134}Cs solution of pH 4 was added to a pre-conditioned 0.1 g of WO_3 adsorbent in a glass vial and left for 24 h at $25 \pm 1^\circ\text{C}$. Then, the liquid phase was decanted and checked for the remaining activity. Thereafter, the ^{134}Cs -loaded WO_3 was dried at 100 °C for 24 h. The sealed source capsule was made from Artylon and composed of two parts; the base disc and the top disc, which finally were assembled to each other, after placing the required amount of ^{134}Cs -loaded WO_3 . It is worth mentioning that the packed ^{134}Cs -loaded WO_3 was definitely placed in the cylindrical cavity of the base disc as a radioactive core of the sealed source.

In order to fix the core material at a definite geometry, the space above the packed matrix was then filled with slurry of a commercial epoxy adhesive till the top of the cavity. The base disc was left for 1 h for the solidification of the adhesive slurry. Thereafter, both of the top and the base discs were assembled by using the same commercial epoxy adhesive. The whole assembly was left again for 1 h to ensure the complete solidification. The finally prepared sealed source was radiometrically assayed using the γ -ray spectrometer to accurately detect its activity (using the absolute efficiency curve), to be further used in the radiometric calibration purposes. In addition, DETEFF software was used to calculate the sealed source activity to be compared with that calculated from the absolute efficiency curve at different source-HPGe detector distances.

3. RESULTS AND DISCUSSION

3.1 Nuclear reactions occurring during neutron irradiation in the reactor

The cross sections of $^{133}\text{Cs}(n,\gamma)^{134m}\text{Cs}$ and $^{133}\text{Cs}(n,\gamma)^{134}\text{Cs}$ are 2.6 and 30 barns, respectively [11]. Due to the great differences between half-lives of ^{134m}Cs and ^{134}Cs (2.9 h and 2.06 y, respectively) [3], cooling the irradiated targets for 48 h is fair

enough for ^{134m}Cs decay. Fig. 1 shows nuclear decay scheme of ^{134m}Cs and ^{134}Cs . According to Fig. 1, ^{134m}Cs decays via isomeric transition (IT) to ^{134}Cs . Only $3 \times 10^{-4} \%$ of ^{134}Cs decays via electron capture (EC) to the stable ^{134}Xe , while the remaining ratio of it decays via β^- -emission to the stable ^{134}Ba [3]. In the present work the irradiated targets were cooled for 7 days before processing.

From nuclear purity point of view, Cs_2O and CsOH are preferred as targets for neutron irradiation. In the present work, CsCl was selected because of its much lower price and the absence of any adverse effect on the required purpose (preparation and calibration of ^{134}Cs sealed source) as illustrated below. When CsCl targets are irradiated in nuclear

reactors secondary nuclear reactions with fast neutrons occur; $^{35}\text{Cl}(n,p)^{35}\text{S}$ and $^{35}\text{Cl}(n,\alpha)^{32}\text{P}$ with cross sections (averaged over ^{235}U fission neutron spectrum) of 78 and 8.8 millibarns and threshold neutron energies of 2.3 and 2.7 MeV, respectively [11, 12]. Thus, both of ^{35}S and ^{32}P can only present as trace impurities and, in addition, their level can be further decreased by choosing the suitable irradiation site of the highest possible thermal neutron flux ratio. It is worth mentioning that both of ^{35}S ($t_{1/2} = 37.82$ d) and ^{32}P ($t_{1/2} = 14.26$ d) are pure β^- -emitters and, as a result, have no interference with the γ -radiometric assay of ^{134}Cs sealed sources since the sealed source capsule serves as a shield for β^- -particles.

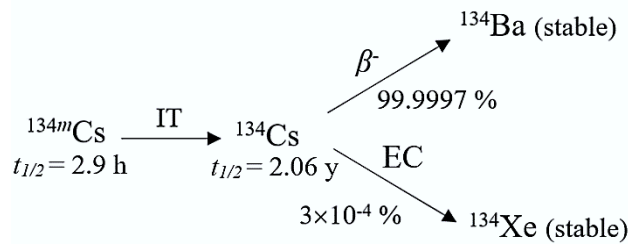


Fig. 1 Nuclear decay scheme of ^{134m}Cs and ^{134}Cs

3.2 Preparation, counting and calibration of the ^{134}Cs sealed source

It was found that the maximum uptake of ^{134}Cs was achieved at pH 4 ($96 \pm 2 \%$). Thus, a bulk amount of WO_3 adsorbent was pre-conditioned by washing thoroughly with HCl solution of pH 4 and then 0.1 g of it was weighed to be batch equilibrated with 10 mL of $^{134}\text{CsCl}$ solution

of pH 4. The core of ^{134}Cs sealed source was ^{134}Cs -loaded WO_3 (dried at 100°C), which was packed in artylon disc Fig. 2. The activity of the prepared ^{134}Cs was calculated by counting on HPGe detector of multichannel analyzer at different distances (0, 5, 10 and 15 cm) and using the absolute efficiency curve of the HPGe and DETEFF software [13, 14] Table 1.

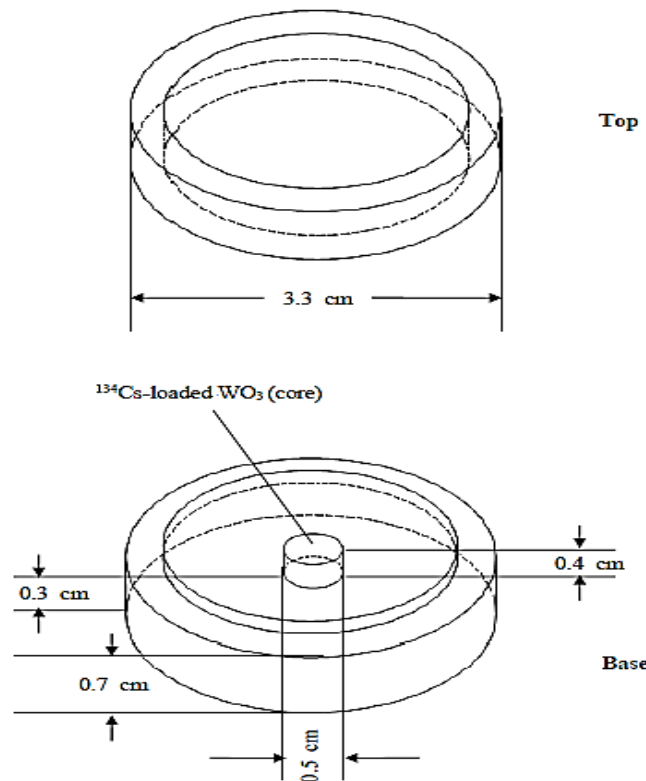


Fig. 2 ^{134}Cs sealed source

Table 1. ^{134}Cs sealed source activity calculation using the absolute efficiency curve of the HPGe detector and DETEFF

Source-detector distance (cm)	^{134}Cs count rate (area under 604.7 keV), counts/1000 s	Absolute efficiency at 604.7 keV		^{134}Cs sealed source activity, μCi	
		Absolute efficiency curve	DETEFF software	Absolute efficiency curve	DETEFF software
0	1.12×10^5	4.90×10^{-2}	3.71×10^{-2}	6.32×10^{-2}	7.30×10^{-2}
5	1.79×10^4	6.34×10^{-3}	5.86×10^{-3}	7.81×10^{-2}	7.70×10^{-2}
10	5.963×10^3	2.17×10^{-3}	2.21×10^{-3}	7.62×10^{-2}	6.77×10^{-2}
15	3.39×10^3	1.04×10^{-3}	1.20×10^{-3}	6.26×10^{-2}	7.44×10^{-2}

DETEFF software assumes a homogeneous distribution of the radioactive material inside the source volume, so, the uniform slab is adopted to describe the activity depth distribution. Thus, the source is assumed to be consisted of cylindrical slabs, in which each slab has a defined individual thickness, homogeneous density and fraction of the total homogeneously distributed activity [14]. In DETEFF software to obtain the absolute efficiency of a γ -line, detector definite dimensions and components should be introduced, in addition to data of the radioactive source (γ -ray line, geometry, dimensions, source-detector distance, etc).

Activity of the sealed source calculated from efficiency calibration curve, A_{EC} , and that calculated from DETEFF software, A_{DET} , can be calculated, respectively as:

$$A_{EC} = \frac{C_{Cs}^D}{E_{EC}^D I t_c} \quad (3.35)$$

$$A_{DET} = \frac{C_{Cs}^D}{E_{DET}^D I t_c} \quad (3.36)$$

Where,

C_{Cs}^D : area under 604.7-keV photopeak at source-HPGe detector distance D .

E_{EC}^D : absolute efficiency determined from absolute efficiency curve for 604.7-keV photopeak at source-HPGe detector distance D .

I : intensity of 604.7 photopeak (0.9762) ⁽³⁾.

t_c : Count time.

E_{DET}^D : absolute efficiency determined by using DETEFF software for 604.7-keV photopeak at source-HPGe detector distance D .

According to data compiled in Table 1, the ^{134}Cs sealed source activity values (calculated by using the absolute efficiency curve and DETEFF software) were

found to be closest at a 5 cm source-detector distance; 7.81×10^{-2} and 7.70×10^{-2} μCi , respectively. Such calibrated sealed source can further be used for efficiency calibration of other gamma-ray spectrometers.

Fig. 3 shows the gamma-ray spectrum of ^{134}Cs sealed source at 5 cm source-HPGe detector distance. The spectrum includes the photopeaks of ^{134}Cs : 475, 563, 569, 604.7 (main peak), 796, 802, 1039, 1168 and 1365 keV confirming its high radionuclidic purity.

3.3 Nuclear safety aspects

If a crucial damage of the sealed source occurs (leading to the spread-out of the radioactive core (^{134}Cs -loaded WO_3)), the following sequence should be followed in the containing room:

- All fans or air conditioners should be shut down.
- Windows should be closed.
- The area containing the leaked radioactive material should be surrounded by marking tapes.
- All the present people should leave, close all room entries.
- Warning signs should be placed in front of all entries.
- All the involved people should make a check for personal radioactive contamination and do not leave the work place before complete decontamination.
- Then, the leaked radioactive material should be removed carefully and placed in a special waste bag by an authorized intervention person under supervision of a radiation protection officer.
- The contaminated area should be swapped with 5 M NaOH solution many times (till complete decontamination), washed thoroughly with water and finally dried.

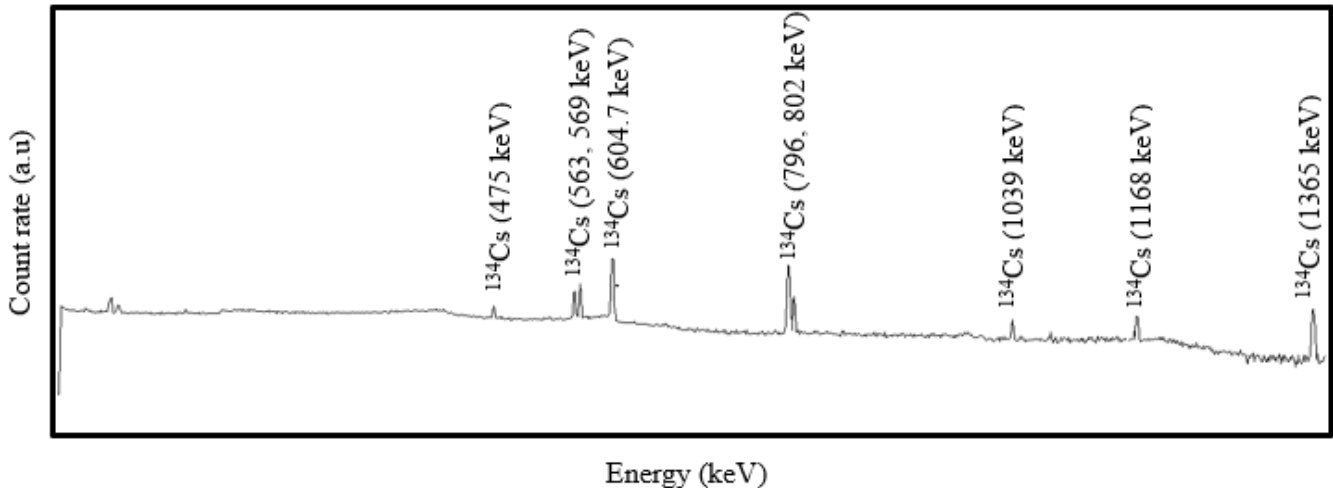


Fig. 3 Gamma-ray spectrum of ^{134}Cs sealed source at 5 cm source-HPGe detector distance

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