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Standardization of ^{142}Pr activity concentration

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HIGHLIGHTS

- Production of ^{142}Pr radionuclide was carried out in a research reactor.
- Standardization of the ^{142}Pr solution with three different standardization methods.
- Lowest uncertainty given by CIEMAT/NIST efficiency tracing method.
- Ionization chamber calibration curve used for the ^{142}Pr activity determination.

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ABSTRACT

^{142}Pr (praseodymium-142), a potential radionuclide for brachytherapy, was produced after irradiating high purity natural Pr_2O_3 powder in a research reactor. The irradiated powder was then dissolved in acids and diluted for measurement. Several radioactivity measurement methods were used in the standardization of ^{142}Pr radionuclide. This work reports the results of standardization of ^{142}Pr with CIEMAT/NIST efficiency tracing method, gamma spectrometry and a calibrated ionization chamber. The activity measured from different standardization methods are compatible with each other within the uncertainty limits. The CIEMAT/NIST efficiency tracing method gives the lowest uncertainty and is a reliable method for the standardization of this radionuclide.

1. Introduction

Brachytherapy is a well-known cancer therapy technique in which a radioactive source is placed near the tumour to irradiate the cancer cells locally while giving reduced dose to the surrounding healthy tissue. Radioactive source is usually encapsulated in a metal or plastic tube, sent to the tissue of interest through a catheter and taken out after the treatment without the risk of degradation of the source in the body. In brachytherapy, the most common gamma emitter sources are ^{192}Ir and ^{125}I but the most frequently used beta-emitter sources include ^{32}P, ^{90}Sr/^{90}Y, ^{188}Re and ^{106}Rh (Sadegh et al., 1999; Baghani et al., 2013). These beta emitters are chosen for certain characteristics. Firstly, the radionuclide should emit beta radiation with enough range to deliver the required dose within a predetermined volume without subjecting the surrounding healthy tissue to excessive radiation. Secondly, if there is also gamma radiation accompanying the beta particle, this needs to have insignificant dose effect compared to the beta radiation. In addition, the radionuclide should have high specific activity and be produced in high activity relatively easily. Research on potential radionuclides shows ^{142}Pr with a half-life of 19.14(4) h is a candidate with relatively high average beta particle energy of 0.83 MeV and low intensity gamma component. ^{142}Pr predominantly decays by emitting two beta particles with maximum energies 585.8 keV and 2161.6 keV having emission probabilities 3.7 (4) % and 96.3(4) %, respectively. ^{142}Pr emits also a high energy gamma photon with energy 1575.77 keV but with low emission probability 3.7(4) % (DDEP, 2017; Bakht and Sadeghi, 2011). ^{142}Pr also has the possibility to be used in targeted radionuclide therapy chelated it with certain molecules that are selectively consumed more by tumour cells. The gamma component of ^{142}Pr with its high energy(~1576 keV) is easily detectable in the tissue thus making it a suitable radioisotope for use in biological distribution studies (Bakht and Sadeghi, 2011). One of the essential aspects of any radionuclide treatment is the use of properly calibrated sources (Suntharalingam et al., 2005). However, literature survey shows that standardization studies of the activity of this potential radio-therapeutic radionuclide is not extensive. This work presents the results of various activity standardization methods for ^{142}Pr. Such measurements are an important prerequisite for potential application of this isotope in radiotherapy.

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2. Experimental

2.1. Production of ^{142}Pr in a research reactor

Praseodymium occurs in Nature as a mono isotope with a mass number 141. It is a malleable, silvery lanthanide metal. It has a relatively high thermal neutron capture cross section of $\sigma_{th} = 11.4$ b when compared to the other currently used beta sources such as ^{32}P ($\sigma_{th} = 0.18$ b) and ^{90}Y ($\sigma_{th} = 1.28$ b). This relatively high cross section makes production of practical ^{142}Pr sources in a low flux research reactor possible. In this study, ^{142}Pr was obtained via $^{141}Pr(n,\gamma)^{142}Pr$ reaction by irradiating natural Pr_2O_3 powder in flame-sealed glass ampoule in the central irradiation tube of TRIGA MARK II research reactor of Istanbul Technical University.

A flame-sealed ampoule of Pr_2O_3 (Sigma-Aldrich, 99.5% purity) was opened in a specially designed glove box with dry nitrogen gas flushing inside. This was done to lower the humidity inside the box so that the slightly hygroscopic praseodymium oxide would not draw moisture and the mass of the sample was as accurate as possible. A high precision balance and a box of SiO_2 desiccant was also placed inside the glove box. Three borosilicate glass ampoules were filled with 0.1452(2) g, 0.2870(3) g and 0.5020(6) g natural Pr_2O_3 powder labelled as Pr-1, Pr-2 and Pr-3, respectively. The ampoules were then flame sealed inside a fume hood with a manually operated glass ampoule sealer. The theoretical activity that can be produced after the irradiation may be calculated according to the following equation assuming a uniform irradiation of the sample (Lenihan and Thomson, 1965):

$$A(Bq) = 0.6022 \cdot \sigma \cdot \Phi \cdot \frac{w\theta}{M} \cdot \left(1 - e^{-\lambda t_{irr}}\right) \cdot D_c \cdot K_c \quad (1)$$

where A is the activity in Bq after an irradiation period of t_{irr}, Φ is the thermal neutron flux, σ is the thermal neutron capture cross section, w is the mass of the isotope in the irradiated sample, θ is the isotopic abundance of the target isotope in the sample, λ is the decay constant of ^{142}Pr and M is the atomic mass of the target isotope. The expected activity of the ^{142}Pr is about 40 MBq after a 5-min irradiation of the Pr-2 sample with a thermal neutron flux of 10^{12} neutrons·cm^{-2}·s^{-1} after 2 h of cool down following the irradiation according to the above equation. A decay factor of $D_c = e^{-\lambda t_{cool}}$ can be used to calculate the activity after cool-down period(t_{cool}), and $K_c = \lambda t_m/(1 - e^{-\lambda t_m})$ is also for correction during the counting losses with t_m is the measuring true time (Karadag et al., 2014).

The Pr-2 ampoule was sent to the reactor core for irradiation for 5 min since the expected activity after irradiation was not high to take stricter radiation protection measures while there would be enough ^{142}Pr radioactivity left for measurement after transport and sample preparation procedures. After irradiation, the sample was measured in the radionuclide metrology laboratories in TAEK-SNRTC and those in AU-NBE in Ankara, respectively. The ampoule was opened and the powder inside was dissolved by adding 2 mL concentrated HNO_3 and 1 mL 10 M HCl to the ampoule in an ultrasonic bath. The solution became a homogeneous liquid with bright green colour. This radioactive solution was transferred to another borosilicate ampoule and the ampoule was flame sealed as the master solution. The master solution was then portioned into subsamples for analysis.

2.2. Calibrated ionization chamber measurements

The master solution was divided into two approximately equal portions in flame sealed ampoules. One of these ampoules with a solution mass of 2.3733 (18) g was used in the calibrated re-entrant (well-type) ionization chamber measurements. The calibrated ionization chamber was not calibrated with ^{142}Pr so the efficiency for this radionuclide was found by using the efficiency curve constructed in an earlier work (Yeltepe et al., 2016). The fitting functions for the beta and gamma efficiency curves are given in the following equations.

Table 1
Decay parameters and the total efficiency for gamma rays and beta particles from ^{142}Pr.

Decay Emission	Energy (keV)	Gamma or beta emission probability	Total efficiency	Calibration factor (k_{IK})
Gamma	508.159(22)	0.00024(3)	0.6654(96)	8.40(12)
Gamma	641.280(9)	0.000022(8)		
Gamma	1575.771(10)	0.037(4)		
Beta	182.5(6) (average)	0.037(4)		
Beta	833.9(7) (average)	0.963(4)		

$$\varepsilon_\beta(E) = 0.05054E^{1.71242} + 1.15153E^{11.80786} \quad (2)$$

$$\varepsilon_\gamma(E) = 11.36598E^{0.64275}e^{-\left(\frac{E}{0,14491}\right)^{-0.85810}} + 0.09624E^{-0,58492}e^{-\left(\frac{E}{0.02666}\right)^{-3.17963}} \quad (3)$$

Where $\varepsilon_\beta(E)$ is the beta efficiency, $\varepsilon_\gamma(E)$ is the gamma efficiency and E is the energy in MeV. The average beta energies are used in the fitting of the curve taken from Decay Data Evaluation Project website (DDEP, 2017). The total efficiency for ^{142}Pr is given in Table 1 together with the emission intensities of the beta and gammas. Total efficiency is the sum of each beta and gamma efficiency multiplied by the corresponding emission probability. The total efficiency for ^{142}Pr is calculated by summing the efficiencies according to the respective beta and gamma emission intensities given in Table 1.

The equation used in the calculation of the activity is given below:

$$a = \frac{I_{net}}{m} k_{IK} C_{Geom} C_{imp} C_{decay} C_{dur} \frac{A_{Ref}}{I_{Ref}} \quad (4)$$

Where a is the specific activity in MBq/g, I_{net} is the average background subtracted current of a set of 30 measurements with 10 s duration in picoamperes(pA), m is the mass of the solution in grams, k_{IK} is the calibration factor obtained from the total efficiency, C_{geom} is the geometry correction factor, C_{imp} is the radionuclidic impurity correction factor, C_{decay} is the decay correction due to the time passed between reference time and time of start pf counting, C_{dur} is the decay correction factor during current measurement, A_{ref} is the activity of the reference source (^{137}Cs) in MBq and I_{ref} (in pA) is the net current due to the reference source. The details of each of these correction factors are explained in detail elsewhere (Yeltepe et al., 2016). The calibration factor k_{IK} is calculated according to the following equation:

$$k_{IK} = \frac{\varepsilon_{ref}}{\varepsilon_{total}} = \frac{5.789(67)\text{pA/MBq}}{\varepsilon_{total}} \quad (5)$$

where, ε_{ref} is the efficiency of the reference source and ε_{total} is the efficiency of the measured radionuclide calculated from the efficiency curve.

The current was measured intermittently for at least 40 h (about two half-lives) constituting 150 sets of thirty 10 s measurements.

2.3. Liquid scintillation counting with CIEMAT/NIST efficiency tracing method

CIEMAT/NIST efficiency tracing technique is a powerful activity standardization method especially for beta or positron emitting and electron capture radionuclides. For instance, standardization of ^{129}I, ^{151}Sm, and $^{166\,m}Ho$ activity concentration was successfully carried out using the CIEMAT/NIST efficiency trancing method (Altzitzoglou and Rozkov, 2016). The details of the method is given elsewhere in a comprehensive manner (Broda et al., 2007; Malonda and Garcia-Toraño, 1982). Briefly, the counting efficiency of a liquid scintillation counter with two nearly identical photomultipliers may be expressed in terms of three quantities. One is the normalized beta spectrum, the