POSSIBILITIES OF A NOVEL APPROACH TO OBTAIN ^{223}RA BY IRRADIATING A TARGET CONTAINING ALUMINIUM OXIDE (III) AS A MATERIAL WITH HIGH SORPTION PROPERTIES

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Abstract – This study presents a novel approach for production of a perspective radionuclide ^{223}Ra, utilizing aluminium oxide (III) as a material with exceptional sorption properties. The method involves the optimized preparation of aluminium oxide (III) to maximize its sorption capacity for ^{223}Ra, followed by a series of controlled reactions to ensure the stability and availability of the final product. Preliminary results indicate significant improvements in the yield and purity of ^{223}Ra, making this method a promising alternative for producing high-quality product for medical applications.

INTRODUCTION

Radium chloride is similar in chemical properties to calcium chloride, so it also forms a complex compound with the bone mineral hydroxyapatite. This means that the introduction of an α-decaying radium isotope into the human body in the form of chloride will selectively affect bone tissue, in particular metastatic foci of prostate cancer in the bones. The high linear energy value of α-particle transfer of about 80 keV/μm leads to a high amount of double-strand breaks in DNA molecules and causes a strong cytotoxic effect. So, such therapy can be used for the selective destruction of cancer cells without affecting healthy organs and tissues [1].

One of the isotopes for radionuclide therapy of bone metastasis in castration-resistant prostate cancer is the ^{223}Ra isotope. The usage of ^{223}Ra is based on the high accumulation of its chloride in osseous issue foci. As for the nuclear parameters of ^{223}Ra, it releases four high-energy alpha particles, 93% of which are formed during the decay of short-lived nuclides [1].

The demand for this radionuclide exceeds supply, since only one of the existing methods for obtaining ^{223}Ra is currently widely used, which is based on the generator method of its production from ^{227}Ac.

The purpose of this research is to develop a new promising method for producing ^{223}Ra using aluminium oxide (III) as a substance with high sorption properties. This material will serve as a target in a research nuclear reactor for the production of ^{223}Ra.

MATERIALS AND METHODS

The overall process is expected to be as follows:

$$^{222}\mathrm{Rn} + \mathrm{n} = {}^{223}\mathrm{Rn} + \gamma \text{ - neutron capture (1)}$$

$$^{222}\mathrm{Rn} + \mathrm{e}^{-} = {}^{223}\mathrm{Fr} + \nu_{\mathrm{e}} \text{ - electron capture (2)}$$

$$^{223}\mathrm{Fr} + \mathrm{e}^{-} = {}^{223}\mathrm{Ra} + \nu_{\mathrm{e}} \text{ - electron capture (3)}$$

The theoretical calculation was carried out by formulating a system of linear differential equations and subsequently solving it using Wolfram Mathematica, a computational software environment for modelling and performing mathematical iterations:

$$\frac{dN(^{222}Rn)}{dt} = -\lambda(^{222}Rn) \cdot N\left(^{222}Rn\right) - \sigma_s \cdot \Phi_s \cdot N\left(^{222}Rn\right) \text{ (4);}$$

$$\frac{dN(^{223}Rn)}{dt} = \sigma_s \cdot \Phi_s \cdot N\left(^{222}Rn\right) - \lambda(^{223}Rn) \cdot N\left(^{223}Rn\right) \text{ (5);}$$

$$\frac{dN(^{223}Fr)}{dt} = \lambda(^{223}Rn) \cdot N\left(^{223}Rn\right) - \lambda(^{223}Fr) \cdot N\left(^{223}Fr\right) \text{ (6);}$$

$$\frac{dN(^{223}Ra)}{dt} = \lambda(^{223}Fr) \cdot N\left(^{223}Fr\right) - \lambda(^{223}Ra) \cdot N\left(^{223}Ra\right) \text{ (7)}$$

where N is the number of nuclei, λ is the decay constant, σ_s is the corresponding neutron cross section and resonance integral, Φ_s is the neutron flux.

The initial radioactivity of ^{222}Rn was determined by γ-spectrometric measurements to be 1570±80 Bq.

The simulation parameters included a Pu-Be neutron source with an average fast neutron flux of $3.17 \cdot 10^6$ cm^{-2}s^{-1} and mean neutron energy of 3.4 MeV based on characteristics of Pu-Be spectra, typically ranging 1-10 MeV with an average of ≈3-5 MeV, corresponding to the characteristics of the ISO 8529-1 reference neutron field specifications.

The resonance integrals and cross-section values are derived from the nuclear data library [2] and shown in table 1. The total resonance integral for the dominant 3.0–3.8 MeV resonance with peaking at 3.4 MeV with σ ≈ 0.42 barn are calculated via trapezoidal integration with Lorentzian broadening.

Table 1.Resonance integral calculation for ^{222}Rn(n,γ) reaction

Energy, MeV	Cross section, barn	Resonance integral contribution, barn	Total resonance integral, barn
3.2	0.25	$(9.68 \pm 2.1) \cdot 10^{-3}$	$(59.1 \pm 5.1) \cdot 10^{-3}$
3.4	0.42	$(23.1 \pm 3.5) \cdot 10^{-3}$	
3.6	0.25	$(19.2 \pm 2.9) \cdot 10^{-3}$	
3.8	0.05	$(8.1 \pm 1.7) \cdot 10^{-3}$	

The stage of preparation of the object of study for subsequent experiments included passing derivatographic analysis. This type of analysis is designed to improve the sorption capacity of the substance under study. Given that substantial moisture may accumulate on the material's surface during transportation and storage in a warehouse, this could lead to a reduction of the sorption efficiency of the object under study. It was found that the best temperature to which the powder should be heated is 400°C. Preparation is carried out by means of derivatographic analysis, during which excess moisture of 3.8% is removed from the substance. This study showed that the method of thermal treatment increases the sorption properties of the material. The method includes irradiation of the original aluminium oxide by means of plasma-chemical synthesis in a unique installation, in which gaseous ^{222}Rn is sprayed over the entire surface of the sorbent with the exposure time of about 30 minutes. The exposure time was selected so that the amount of radon sorbed during synthesis was more than 50% from the amount sprayed.

Based on the data obtained, a linear function was drawn which was necessary for a visual representation of the loss of powder mass with increasing temperature (Figure 1).

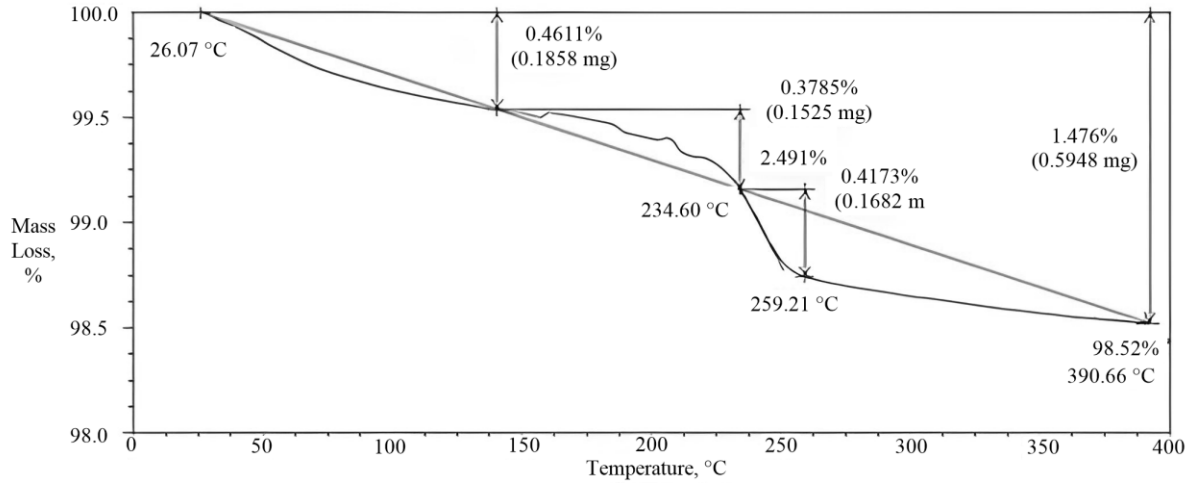

Figure 1. Results of derivatographic analysis

The experimental setup «Plasma module based on high-frequency generator VChG8-60/13-01» was used in the work, with the help of which plasma-chemical synthesis of aluminium oxide (III) with ^{222}Rn was carried out.

Canberra GC1518 coaxial germanium detector with a registration efficiency of 15% and a resolution of 1.8 keV at 1.33 MeV was used in the work [3]. Spectrometric analysis made it possible to determine the isotopic composition of powdered targets based on aluminium oxide (III).

The second stage of the study was the analysis of the target after bombardment with neutrons of a certain energy in the field of a neutron generator, which was a hermetically sealed cylindrical shell-moderator, inside which five neutron sources were located at the same level in

the geometric centre for uniform irradiation. Inside there were two plutonium-beryllium and three americium-lithium sources creating an ambient equivalent of neutron radiation dose of about 32 mSv/hour inside the box.

For a Pu-Be neutron source with 3.4 MeV mean energy, self-absorption effects reduce target radionuclide yield by up to 40% in thick (>1 mm) targets due to neutron flux attenuation. The optimal uncorrected thickness range is 0.1–0.3 mm, where yield losses remain below 5%.

RESULTS

The solution of the system of differential equations (4)-(7) gives us the final number of nuclei of the isotope ^{223}Ra that should have been formed during irradiation, which amounted to $4.6 \cdot 10^2$ atoms. So, the possibility of producing the radium-223 isotope using a neutron flux of a discrete energy spectrum is shown.

The aim was to accumulate in the studied samples the target isotope ^{223}Ra, which is a product of the daughter decay of ^{231}Pa with characteristic energy peaks of 269.46 keV, 154.21 keV, 323.87 keV, 338.28 keV, 122.32 keV, 445.03 keV. The identification results of the accumulation of radioactivities after the irradiation are presented in the table 2.

Table 2. Identification of the gamma radiation spectrum

Isotope	Reliability of identification	Activity, Bq
^{214}Bi	0.994	3200±200
^{214}Pb	0.875	2200±100
^{226}Ra	0.998	0.83±0.06
^{227}Th	0.371	0.21±0.01
^{235}U	0.541	0.051±0.003
^{238}U	0.904	1.4±0.1

DISCUSSION

^{226}Ra and ^{227}Th can be produced through successive neutron capture and electron capture processes. We found that the irradiation time during which the target was subjected to the neutron generator field together with accompanying nuclear processes lead to the formation of corresponding nuclides.

^{226}Ra with gross activity of about 0.83 Bq and ^{227}Th with gross activity of about 0.21 Bq, which is a parent nucleus of ^{223}Ra, were identified in low concentration during the gamma-spectrometric analysis. The detected spectrum contains low-intensity lines from the ^{227}Ra isotope with characteristic energy peaks of 283.68 keV and 258.3 keV. Characteristic line of ^{227}Th with

the energy peak of 256.23 keV was detected in the object of study. It is impossible to unambiguously establish the content of radium isotopes in the studied samples according to the graph obtained during γ-spectrometric analysis with the corresponding peaks with a certain energy due to their low activity. A line with the energy of 87.3 keV was detected in the spectrum of the studied powders. It is not possible to unambiguously identify the X-ray spectrum of ^{223}Ra.

CONCLUSION

Thus, with the activity of the radionuclide ^{222}Rn 1570±80 Bq, the activity of the produced ^{223}Ra at a given neutron flux of $3.17 \cdot 10^{6}$ $cm^{-2}s^{-1}$ is calculated to be 0.47 Bq. This study considers a promising method for producing the ^{223}Ra isotope. γ-spectrometric analysis revealed the presence of actinium, thorium, and radium isotopes and their decay products. For a comprehensive analysis of the composition of the research object, it is necessary to adjust the measuring instruments and systems for better efficiency in the selected geometry. Changing the geometric parameters of the target will improve the output parameters of the neutron flux and improve the production of the target ^{223}Ra isotope. The upcoming studies are planned to be carried out at a research nuclear reactor which will allow us to get closer to producing the target isotope ^{223}Ra.

REFERENCES

1. Butkaluk P.S., Obtaining experimental samples of alpha-emitting radionuclides for medical purposes // Collection of works of JSC SRC RIAR, 2018.
2. TENDL-2019: Evaluated Nuclear Data Library – Rn-222 neutron cross-sections (MT102) // Data set, 2019.
3. Boyko V.I., Zherin I.I., Karatayev V.D, Methods and instruments for measuring nuclear and other radioactive materials: a tutorial // Publishing house of the International Scientific and Technical Center, Moscow, 2011.