Development of a Tandem Generator System $^{229}$Th/$^{225}$Ac/$^{213}$Bi for Repeated Production of Short-Lived $\alpha$- Emitting Radionuclides

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Abstract—$^{226}$Th was isolated from a long-stored $^{233}$U sample using an anion exchanger and 8 M HCl. Thorium was separated from macroamounts of inactive impurities on an anion exchanger in aqueous-methanol HNO$_3$ solutions. A tandem generator system was developed for repeated isolation of $^{225}$Ac ($\alpha$, 10 days) from the parent radionuclide $^{229}$Th (7.3 $\times$ 10$^3$ years) sorbed on an anion-exchange column, with the subsequent sorption of Ac on a cation-exchange column and repeated elution of $^{213}$Bi ($\alpha$, 45.6 min) with dilute HCl and HBr solutions. The $\gamma$-ray spectra of the radionuclides at various steps of their isolation are presented. Alternative procedures for production and isolation of $^{225}$Ac and $^{213}$Bi are described. The application potential and advantages of the newly developed tandem generator system are discussed.

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Much attention is given today to the development of procedures for production and isolation of short-lived radionuclides with the aim of their use in scientific research, in nuclear medicine, and as tracers in analysis of various natural objects. $\alpha$- Emitting radionuclides show much promise in nuclear medicine for diagnostics and targeted radiotherapy of tumor diseases [1–6]. Depending on particular problem, relatively “long-lived” and short-lived $\alpha$- emitting radionuclides are used, with the half-life ranging from several days ($^{225}$Ac, $^{223}$Ra, $^{223}$Es) to several hours ($^{211}$At, $^{212}$Pb/$^{212}$Bi, $^{215}$Fm) and minutes ($^{211}$Pb/$^{211}$Bi, $^{213}$Bi/$^{213}$Po).

Thanks to favorable nuclear-physical characteristics, $^{225}$Ac ($T_{1/2}$ = 10 days) is one of the most promising $\alpha$- emitting radionuclides for use in radiotherapy [7, 8]. $^{225}$Ac is the member of the radioactive family of neptunium:

$^{227}$Np ($\alpha$, 2.14 $\times$ 10$^6$ years) $\rightarrow$ $^{223}$Pa ($\beta$, 27.4 days)

$\rightarrow$ $^{223}$U ($\alpha$, 1.6 $\times$ 10$^5$ years) $\rightarrow$ $^{229}$Th ($\alpha$, 7.3 $\times$ 10$^3$ years)

$\rightarrow$ $^{225}$Ra ($\beta$, 14.9 days) $\rightarrow$ $^{225}$Ac ($\alpha$, 10 days)

$\rightarrow$ $^{221}$Fr ($\alpha$, 4.9 min) $\rightarrow$ $^{217}$At ($\alpha$, 0.032 s)

$\rightarrow$ $^{213}$Bi ($45.6$ min)$\rightarrow$ $^{211}$Po ($4 \times 10^{-5}$ s)

$\rightarrow$ $^{211}$Po ($\alpha$, 209$^+$Tl ($2.2$ min)$\rightarrow$ $^{209}$Pb ($\beta$, 3.25 h)

$\rightarrow$ $^{206}$Bi (stable)

The $\alpha$-decay of $^{225}$Ac produces three short-lived $\alpha$- emitting radionuclides: $^{221}$Fr, $^{217}$At, $^{213}$Bi/$^{213}$Po; the mean energy of all the $\alpha$-particles is 6.9 MeV.

Actinium-225 is usually produced on accelerators by irradiation of U, Th, or $^{226}$Ra with high-energy protons [9–11]. In the course of irradiation, a large number of other $\alpha$- and $\gamma$- emitters, including longer-lived radionuclides, are formed along with $^{225}$Ac in side reactions. Their separation and isolation of $^{225}$Ac from the irradiated material require the use of a set of various chemical methods, which makes the procedure expensive and time-consuming.

$^{225}$Ac can also be produced by a photonuclear reaction $^{226}$Ra($\gamma$, $n$)$^{225}$Ra ($\beta$, 14.8 days) $\rightarrow$ $^{225}$Ac on a microtron [12] or linear electron accelerator [13]. An apparent disadvantage of this procedure for producing $^{225}$Ac is that the target used is highly radioactive and emanating.

The most convenient and promising procedure for producing $^{225}$Ac is the generator procedure of its isolation from the parent radionuclide $^{229}$Th formed by decay of $^{233}$U, which is produced in nuclear reactors by the reaction $^{212}$Th($n$, $\gamma$) $\rightarrow$ $^{233}$Th($\beta$, 23.5 min) $\rightarrow$ $^{233}$Pa ($\beta$, 27.4 days) $\rightarrow$ $^{233}$U. Apparently, an alternative procedure for obtaining $^{233}$U can be its isolation from long-stored $^{237}$Np.

The $^{229}$Th/$^{225}$Ra/$^{225}$Ac generator was developed on
RESULTS AND DISCUSSION

Isolation and purification of $^{229}$Th. To separate $^{229}$Th from $^{235}$U, we used a well-known separation procedure on an anion exchanger in concentrated HCl solutions. The nitric acid solution containing $^{233}$U was evaporated to wet residue which was then dissolved in 2 ml of 8 M HCl. The hydrochloric acid solution (sorbate) was passed through a column containing 0.5 g of Dowex 1 × 8 anion exchanger, preliminarily washed with 8 M HCl. After passing the sorbate through the column, an additional 18-ml portion of 8 M HCl was passed. More than 99% of Th and its decay products (Ra, Ac, etc.) were washed out with 6 ml of the eluate, whereas U remained on the column (Fig. 1). Uranium

The goal of this study was the development of a generator system for repeated isolation of $^{225}$Ac from $^{229}$Th sorbed on an anion-exchange column by elution with 8 M HNO$_3$ followed by sorption of $^{225}$Ac on a cation-exchange column and repeated elution of $^{213}$Bi with dilute solutions of HCl and HBr. $^{229}$Th was isolated from long-stored solutions containing $^{233}$U and was purified to remove a number of macroimpurities using two anion-exchange columns and 8 M HCl and HNO$_3$/CH$_3$OH solutions, respectively.

EXPERIMENTAL

Chemicals, radionuclides, radiometric measurements. All the chemicals used in the experiments (HCl, HBr, HNO$_3$, CH$_3$OH) were of chemically pure grade. Dowex 50 × 8 cation exchanger and Dowex 1 × 8 anion exchanger of particle size 100–200 and 200–400 mesh (Serva) were thoroughly washed with water to remove the fine fraction and were dried in air. The radionuclides $^{229}$Th, $^{225}$Ra, $^{225}$Ac, and $^{213}$Bi were isolated from long-stored nitric acid solutions containing $^{233}$U, following the procedures described below. When choosing optimum conditions for repeated isolation of $^{213}$Bi, we also used short-lived isotopes $^{211}$Pb and $^{211,212}$Bi, which were repeatedly isolated from $^{227}$Ac–$^{211}$Pb/$^{211}$Bi and $^{225}$Ra–$^{212}$Pb/$^{212}$Bi generators developed previously [17–21].

Radiometric measurements were performed on a γ-ray installation with a Na(Tl)I crystal and on a UMF-2000 α-ray counter. The radionuclides were identified by the main γ-quantum energies using a Ge(Li) detector connected to an NUC-810 multichannel analyzer (Merion-X Ltd., Hungary).