Generator of Actinium-228 and a Study of the Sorption of Actinium by Carbon Nanomaterials

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Abstract—A method has been developed for isolation of ^{228}Ac from aged samples of natural ^{232}Th. Macroscopic amounts of the thorium bulk (20 g) were separated by its extraction by a solution of di(2-ethylhexyl)phosphoric acid in toluene from 4 M HNO_3 solutions, with microamounts of ^{228}Ra and ^{228}Ac remaining in the aqueous phase. ^{228}Ac was selectively extracted from the resulting solution by the method of extraction chromatography on a column with RE resin sorbent, and ^{228}Ra quantitatively remained in the 4 M HNO_3 eluate. ^{228}Ac was washed out of the column with 0.05 M HNO_3, it could be used as a radioisotope for research purposes. After the accumulation of the daughter ^{228}Ac the solution of ^{228}Ra in 4 M HNO_3 was repeatedly passed through an identical column. The resulting ^{228}Ac was used in the study to examine its sorption on commercial and modified carbon nanomaterials (CNMs) instead of the difficultly accessible ^{225}Ac. It is shown that the application of CNMs as carriers of actinium isotopes for nuclear medicine purposes is promising.

Keywords: actinium-228, radioisotope generators, extraction chromatography

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INTRODUCTION

The targeted alpha-therapy in nuclear medicine frequently uses the ^{225}Ac that undergoes the fast alpha-decays [1–3]. This isotope is obtained by different methods, including that in which thorium targets are irradiated with high-energy protons. However, this method does not find wide application because the number of suitable accelerators in the world is exceedingly small, which gives rise to problems with the availability of the given isotope [4]. In addition, ^{225}Ac is not quite convenient for pre-clinical trials because of emitting gamma photons in low yield (not more tham 1% at 100 keV). At the same time, the gamma-emitting isotope ^{228}Ac ($T_{1/2}$ = 6.1 h) is present in the radioactive decay series of natural thorium as a product formed in the decay of a mother isotope ^{228}Ra and can be isolated in the radiochemically pure form. To advantages of ^{228}Ac, isolated from ^{228}Ra belong the long lifetime of the mother nuclide ($T_{1/2}$ = 5.7 yrs) and the absence of long-lived daughter nuclide. The activity of ^{228}Ra in the secular equilibrium with ^{232}Th is about 1.6 kBq per gram of thorium nitrate. Therefore, with ^{228}Ra isolated from gram amounts of thorium nitrate, ^{228}Ac can be dayly extracted from the generator with activity sufficient for carrying out studies, including those performed in vivo (several and tens of kBq).

Various ways to separate Th, Ra, and Ac are known from the literature [4–10]. ^{228}Ra and ^{232}Th are, as a rule, separated via coprecipitation of radium and its extraction in various systems [4, 5]. Actinium and radium are separated by ion-exchange chromatography [6, 7], extraction [8], selective sorption [9], and extraction chromatography [4, 6,10]. In these studies, small amounts of ^{232}Th or trace amounts of ^{229}Th have been used. At the same time, there is no published information about isolation of ^{228}Ac from large amounts of ^{232}Th (tens of grams).
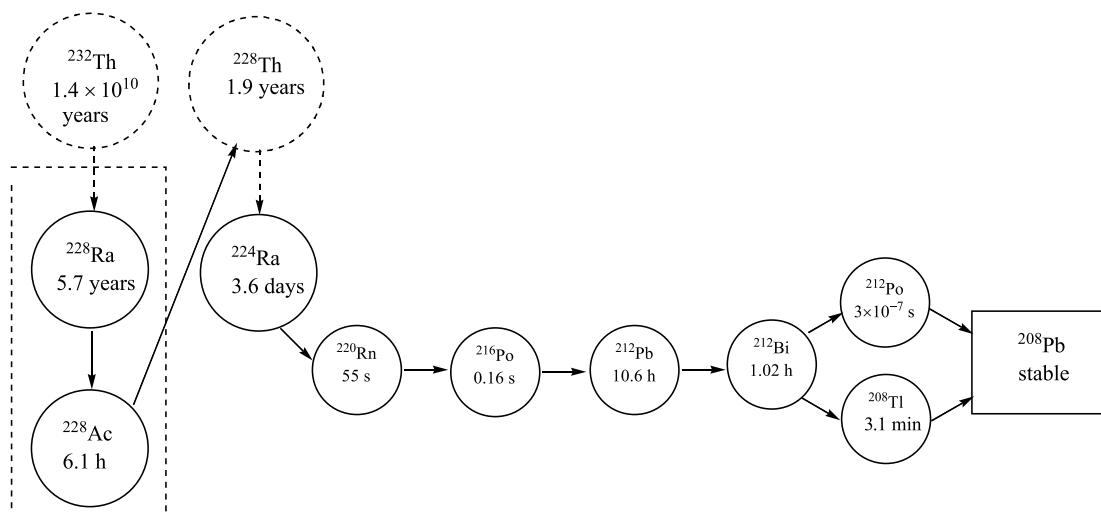
Fig. 1. ^{232}Th decay series containing the generator pair ^{228}Ra/^{228}Ac

Studies concerned with the possibility of targeted delivery of actinium examined the sorption of ^{225}Ac on various supports in radiopharmpreparations. For example, it has been shown that it is possible to use liposomal carriers [11–14] or nanoparticles based on inorganic substances: $LaPO_4$ [15–19], $InPO_4$ [20], $GdVO_4$ [21], and TiO_2 [22].

At the same time, application of carbon nanomaterials (CNMs) seems to be promising because of the important properties: potential biocompatibility of their separate forms, accessibility in industrial amounts, low cost as compared with vector molecules, and the possibility of varying their biodistribution upon an appropriate modification. However, the evidence about using CNMs as actinium supports is extremely scarce [23, 24]. In this context, the goal of the present study was to develop a method for quantitative isolation of ^{228}Ac as a product of ^{228}Ra decay from natural thorium, with their subsequent separation and using the isolated ^{228}Ac in the radiochemically pure state to examine the sorption of actinium on CNM sample with varied structure and physicochemical properties.

EXPERIMENTAL

Development of a procedure for separation of radiochemically pure ^{228}Ac from natural thorium. In the study, we used thorium nitrate (weighed portions with masses of up to 20 g) with decay products accumulated during 60 years. All the chemical reagents used in the study were not less than chemically pure.

The purified ^{228}Ac preparation was obtained in several stages. In the first stage, macroamounts ^{232}Th and accumulated ^{228}Th are separated from a mixture of their decay products (Fig. 1) via extraction of thorium by a 1 : 1 solution of di(2-ethylhexyl)phosphoric acid (HDEHP) in toluene from 4 M HNO_3 solutions. The choce of the extraction agent was governed by earlier studies [4] in which it was found that the best reagent among those examined for separation of ^{232}Th from products formed in its irradiation by high-energy protons (and also of radium and actinium) is HDEHP, which extracts ^{232}Th from nitrate solutions in a wide range of HNO_3 concentrations [25–27]. The phases were vigorously agitated for 5 min, centrifuged, and then separated in a separating funnel. The extraction was performed three times, and the aqueous solution with ^{228}Ra was passed, to be fuller purified to remove thorium, through a column with extraction-chromatographic sorbent LN (Triskem Int., France, based on HDEHP, particles size 100–150 mesh). The sorbent firmly retains thorium in a wide range of HNO_3 concentrations, with radium and actinium not retained by this sorbent at HNO_3 concentrations exceeding 0.2 M [28].

The mixture of nuclides, remaining after the separation of thorium isotopes contains ^{228}Ra (in which ^{228}Ac accumulates) and also ^{224}Ra (in equilibrium with other decay products, including gaseous ^{220}Rn) and the stable ^{208}Pb. A single-stage isolation of ^{228}Ac from a mixture of this kind is a complicated task, and deep purification to remove ^{224}Ra is necessary for obtaining the radiochemically pure ^{228}Ac. For this reason, the

mixture was kept for one month till the complete decay of ^{224}Ra. In the final stage, ^{228}Ac was separated from ^{228}Ra, ^{208}Pb, and newly formed ^{228}Th. For this purpose, a solution of ^{228}Ra in HNO$_3$ was evaporated to dryness in a glass flask on an electric hot plate, dissolved in 4 M HNO$_3$; and the solution was placed in a column with RE extraction-chromatographic sorbent (Triskem Int., France, based on carbamoylmethylphosphine oxide and tributyl phosphate) with volume of 2 mL. ^{228}Ra and ^{228}Ac were separated by selective elution from the column: first, ^{228}Ra was eluted with a 4 M HNO$_3$ solution, and then ^{228}Ac, in 0.05 M HNO$_3$, with fractions collected each having a volume of 1 mL The purity of ^{228}Ac was determined by registering its gamma-spectrum during three days. To perform experiments on sorption of actinium on CNMs, the ^{228}Ac eluate in 0.05 M HNO$_3$ was evaporated to dryness and dissolved in several mL of a phosphate buffered saline (PBS) with pH 7.

Gamma-spectrometry. The content of thorium, radium, and actinium, in the samples was found by the gamma-spectrometry on a spectrometer equipped with a GR 3818 high-purity germanium detector (Canberra Industries, United States). Thorium was identified by the gamma-peak of ^{228}Th with energy of 84.4 keV. The content of ^{228}Ra was determined by using the additionally introduced gamma-emitting ^{226}Ra (186 keV, 3.6%) obtained via dissolution of ^{226}RaBr$_2$ in 0.01 M HCl. ^{228}Ac was determined from the lines at 338 (11.3%) and 911 (25.8%) keV.

Characterization of CNMs. We used in the study the following commercial samples of CNMs: powdered detonation nanodiamonds (DNDs) of UDA-TAN brand (Federal State Unitary Enterprise SKTB Tekhnolog, Russia), aqueous suspension of partly reuced graphite oxide (GO), and powdered multiwall carbon nanotubes (MWCNTs) (Nanotekhtsentr OOO, Tambov, Russia). The morphology of CNM particles was determined by high-resolution transmission electron microscopy (HR-TEM) on a JEM-2100F/Cs/GIF instrument (JEOL, Japan). The specific surface of the CNMs was determined by the method of thermal desorption of nitrogen in terms of the BET model. The content of impurity elements in the CNMs was found by mass spectrometry with inductively coupled plasma (ICP-MS) (X Series 2, Thermo Scientific, United States). The elemental composition of the CNM surface was determined by the method of X-ray photoelectron spectroscopy (XPS) (Kratos Axis Ultra DLD, Kratos

Analytical, Great Britain). The hydrodynamic diameter of CNM particles in hydrosols was determined by the method of dynamic light scattering (DLS) on a ZetaSizer Nano ZS (Red Badge, 633 nm) ZEN 3600 analyzer (Malvern Instruments, United States). Acid functional groups on the CNM surface were determined by potentiometric titration.

The surface of the CNM samples was modified by the procedures described in [29, 30]. DNDs were oxidized in a 3 : 1 mixture H$_2$SO$_4$/HNO$_3$ at 120°C for 24 h, and GO and MWCNTs were hydrogenated in the Ar + 10% H$_2$ atmosphere at 800°C for 5 h.

Study of the sorption behavior of ^{228}Ac on CNM. In a study of the sorption of Ac(III) on CNMs, a 100-μL portion of a CNM suspension was placed in polypropylene test tubes with a cover, and 900 μL of the resulting solution of ^{228}Ac was added. The test tubes were shaken on a TS-100 shaker (Biosan, Latvia), and after that the mixture was centrifuged on a CM-50 centrifuge (Eppendorf, United States) for 20 min at 18000 g. Then, the gamma-spectrum of the ^{228}Ac solution was recorded, and the counting rate of an aliquot of the solution was compared with that for a reference sample. To determine the amount of desorption, to the solution and precipitate remaining in the test tube was added a solution of bovine serum albumin (BSA) in PBS, with the content of BSA in the resulting solution being 40 g/L. The contents of the test tube were stirred up and placed on a shaker, after which the phases were separated by centrifuging for subsequent radiometric measurements.

RESULTS AND DISCUSSION

Development of a procedure for separation of radiochemically pure ^{228}Ac from natural thorium. It was shown that ^{228,232}Th is extracted with HDEHP from a 4 M HNO$_3$ solution nearly quantitatively (99%), with no ^{228}Ra extracted in this case. An additional purification of ^{228}Ra remaining in the aqueous phase to remove thorium was performed by its passing through a column with extraction-chromatographic sorbent LN, with 99.9% ^{228}Ra and not more than 0.01% ^{228,232}Th contained in the eluate. ^{228}Ra and ^{228}Ac were separated by the extraction-chromatographic method on the RE sorbent. It can be seen in Fig. 2, which shows a chromatogram, that ^{228}Ra is quantitatively extracted in 2–5-mL fractions and ^{228}Ac in 13–16-mL fractions. The flow rate of the eluate was 1 mL/min, which makes it possible to separate ^{228}Ra and ^{228}Ac in 20 min. It was
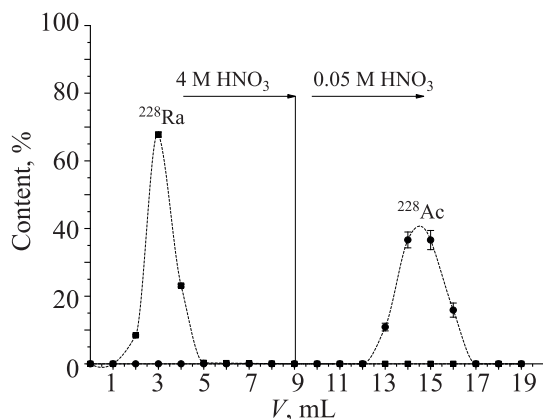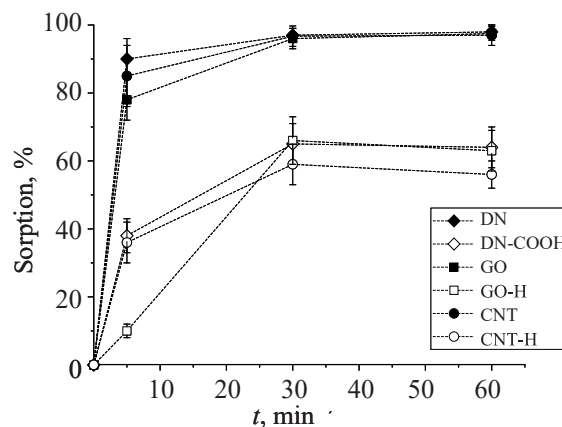
Fig. 2. Extractive-chromatographic separation of ^{228}Ra and ^{228}Ac on RE sorbent in HNO$_3$. Column volume 2 mL, diameter 0.7 cm.

Fig. 3. Kinetics of Ac(III) sorption on CNM in PBS at 25°C, m/V ratio 100 μg/mL.

found here that the yield of ^{228}Ac is nearly quantitative (not less than 95% relative to the amount placed on the column), with the separation factor of ^{228}Ra and ^{228}Ac being 2.2×10^4. According to [31], ^{208}Pb is eluted from the column together with ^{228}Ra. Thus, we developed a method for separation ^{228}Ac in the radiochemically pure state from large amounts of natural thorium. The isolated ^{228}Ac can be used in scientific studies, including *in vivo* experiments.

It was found that, with the developed method implemented, the loss of ^{228}Ra does not exceed 0.005% of the amount placed in the column. Therefore, the starting ^{228}Ra preparation can be repeatedly used. For this purpose, the fractions from 1 to 9 mL were evaporated and again dissolved in 1 mL of 4 M HNO$_3$. It was shown that the results are reproducible because ^{228}Ac could be eluted from the generator we developed not less than 20 times. In a prolonged use of the solution of ^{228}Ra, there occurs accumulation of ^{228}Th occurs due to the decay of ^{228}Ac, but ^{228}Th is quantitatively sorbed on the column with RE in the process of separation of ^{228}Ra and ^{228}Ac [31]. It is supposed that the columns with the sorbent are intended for single use, and, therefore, ^{228}Th does not contaminate the solution of ^{228}Ac being prepared. In the many-years use of the generator, the extract of ^{232}Th can be repeatedly used to extract ^{228}Ra accumulated in it.

Characterization of CNM samples. The physicochemical properties of the commercial and modified CNM samples are presented in Table 1. It can be seen in the table that the CNMs used in the study significantly changed from each other both in structure

and in composition. For example, DNDs are constituted by spherical particles, GO, by planar nanoparticles, and CNTs have micrometer threadlike structure with a internal hollow. Also the CNT samples had different specific surface areas which were 240, 700, and 160 m^2/g for DNDs, GO, and CNTs, respectively. It was found that, among the CNMs under study, DNDs have the smallest amount of impurities (up to 1.4 mg/g). It was shown that modification of samples in their oxidation and hydrogenation led to a change in the surface composition of CNMs according to the results of XPS and potentiometric titration. For example, the oxidation of a DND sample resulted in that the number of carboxy groups became 3 times larger (from 330 to 990 μmol/g).

Study of ^{228}Ac sorption onto CNMs. The kinetics of ^{228}Ac sorption on various CNM samples in PBS was examined (Fig. 3). It can be seen in Fig. 2 that the sorption equilibrium is reached in 30 min for all the CNM samples for all studied CNMs. The sorption in 60 min was nearly quantitative (not less than 95% for DND, GO, and CNT samples, the limiting amount of sorption being about 60% for all the CNM samples.

It was found the decrease between the CNM mass and the solution volume from 100 to 10 μg/mL does not lead to a change in the amount of sorption. This will make it possible to substantially reduce the mass of the *in vivo* introduced conjugate Ac(III)@CNM.

It was shown by a study of the desorption of ^{228}Ac from samples of commercial CNMs in a solution of BSA in BP that up to 30–40% of actinium is desorbed in 30 min, after which the amount of desorption does not change significantly during 150 min (Fig. 4). At the
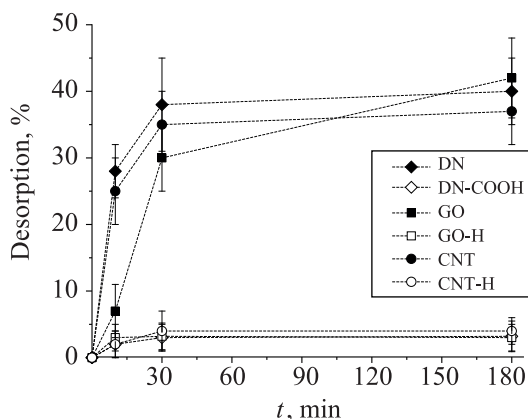
Fig. 4. Kinetics of Ac(III) desorption from CNM in BSA solution (40 g/L) in PBS at 25°C, *m/V* ratio 100 μg/mL.

same time, it was found that the desorption of ^{228}Ac from DND-COOH, GO-H, and CNT-H samples under the same conditions does not exceed 5% in 3 h. The change in the behavior of Ac(III) in its sorption from PBS and desorption from BSA in PBS for the commercial DNDs, GO, and CNT and modified DND-COOH, GO-H, and CNT-H can be accounted for by various factors. The sorption of Ac(III) on a commercial sample of DNDs probably occurs by two mechanisms: firm chemical binding of ^{228}Ac with carboxy groups and physical adsorption onto other groups on the DND surface. In the case of DND oxidation to DND-COOH, these groups are substituted by carboxy groups, which negatively affects the total amount of adsorption because of the

Table 1. Results of characterization of CNM samples

Commercial CNM samples			
Characteristics	DNDs	GO	CNTs
Particle size of initial samples according to HR-TEM, nm	3–10	Nanosheets 2 Sheets > 10^2	Length > 2×10^4 Diameter 30 Wall thickness 5–10
Specific surface area, m^2/g	240	700	160
Total content of impurities according to ICP-MS data, mg/g	1.4	3.0	14.0
Main impurities (more than 0.1 mg/g) and the content of these, mg/g	Fe 0.538 Ti 0.459 K 0.156	Ti 2.600 Al 0.124	Mo 6.880 Co 5.830 Al 0.635 Ni 0.156
Elemental composition of the surface according to XPS	C_{sp3} 92.3% O 7.7% N 1.0%	C_{sp2} 77.4% C_{sp3} 7.9% O 14.7%	C_{sp2} 99.0% O 1.0%
Size of particles and aggregates of these in hydrosols, nm	100	Nanosheets 2 Sheet[a]	[a]
Number of –COOH groups according to titration data, μmol/g	330	–	–
Modified CNM samples			
Characteristics	DND-COOH	GO-H	CNT-H
Number of –COOH groups according to titration data, μmol/g	990	–	–
Size of particles and aggregates of these in hydrosols, nm	95	200 and 700	150 and 650
Elemental composition of the surface according to XPS data	C_{sp3} 88.7% O 9.9% N 1.4%	C_{sp2} 72.3% C_{sp3} 24.6% O 3.1%	C_{sp2} 99.4% O 0.6%

[a] It is impossible to determine by the DLS method if at least one of the linear dimensions of the particle exceeds 1000 nm.

disappearance of its physical component, but positively affects the strength of binding between the conjugate and actinium in a solution of BSA in PBS. Interpretation of changes of this kind is difficult for GO and GO-H, and a further study of the ^{228}Ac sorption mechanism on these samples is necessary. As for the interaction of ^{228}Ac with CNTs, two mechanisms are probably operative in the sorption: sorption of ions within the tube hollows, as shown previously in [32], and the physical sorption on impurity metals (Table 1) contained at tube ends. In the hydrogenation, of CNTs, the catalyst is removed, which can account for the decrease in the total amount of sorption and the increase in the stability against desorption.

To conclude, it can be noted that all the CNMs are promising for application in nuclear medicine as a carrier for actinium. In the case of limited amounts of radioactive isotopes of actinium, it is appropriate to use DNDs, GO, and CNTs for which the sorption is nearly quantitative, whereas DND-COOH, GO-H, and CNT-H are the most suitable for *in vivo* studies, For these the amount of desorption in 3 h does not exceed 5% in solutions of a model biological medium, whereas for the commercial CNMs under study it is about of 40%.

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CONFLICT OF INTEREST

The authors state that they have no conflict of interest.

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