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Development of methods for the preparation of radiopure ^{82}Se sources for the SuperNEMO neutrinoless double-beta decay experiment

https://doi.org/10.1515/ract-2019-3129
Received March 4, 2019; accepted May 16, 2019; published online June 15, 2019

Abstract: A radiochemical method for producing ^{82}Se sources with an ultra-low level of contamination of natural radionuclides (^{40}K, decay products of ^{232}Th and ^{238}U) has been developed based on cation-exchange chromatographic purification with reverse removal of impurities. It includes chromatographic separation (purification), reduction, conditioning (which includes decantation,

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centrifugation, washing, grinding, and drying), and ^{82}Se foil production. The conditioning stage, during which highly dispersed elemental selenium is obtained by the reduction of purified selenious acid (H_2SeO_3) with sulfur dioxide (SO_2) represents the crucial step in the preparation of radiopure ^{82}Se samples. The natural selenium (600 g) was first produced in this procedure in order to refine the method. The technique developed was then used to produce 2.5 kg of radiopure enriched selenium (^{82}Se). The produced ^{82}Se samples were wrapped in polyethylene (12 µm thick) and radionuclides present in the sample were analyzed with the BiPo-3 detector. The radiopurity of the plastic materials (chromatographic column material and polypropylene chemical vessels), which were used at all stages, was determined by instrumental neutron activation analysis. The radiopurity of the ^{82}Se foils was checked by measurements with the BiPo-3 spectrometer, which confirmed the high purity of the final product. The measured contamination level for ^{208}Tl was 8–54 µBq/kg, and for ^{214}Bi the detection limit of 600 µBq/kg has been reached.

Keywords: Selenium-82, ion exchange chromatography, purification, mass spectrometry, neutron activation analysis, SuperNEMO, LSM Modane.

1 Introduction

Selenium-82 is one of the most suitable isotopes for studying double-beta decay (ββ decay) and searching for its neutrinoless mode (0νββ). Interest in studies of 0νββ decay has increased significantly in recent years since the discovery of neutrino oscillations. NEMO-3 was one of the leading underground experiments for ββ decay studies in different isotopes (^{48}Ca, ^{82}Se, ^{96}Zr, ^{100}Mo, ^{116}Cd, ^{130}Te, ^{150}Nd) with a total mass of about 10 kg. The experiment used a unique tracking-calorimetric technique to measure the full topology of ββ decay and obtained a number of important ββ decay results [1–3]. The SuperNEMO project extrapolates the successful NEMO-3 technique to a next-generation of 0νββ-experiments, planning to reach a sensitivity $t_{1/2}^{0\nu\beta\beta} \sim 10^{26}$ years with ~100 kg of enriched isotope (^{82}Se, ^{150}Nd) [4].

Use of ultrapure materials with respect to impurities of ^{40}K, ^{232}Th, ^{238}U is a key factor in the success of low-background underground experiments. Therefore, all materials used (e. g. construction materials, detector shielding, photomultipliers, cables, etc.) are carefully selected after screening for radioactive impurities. Inductively coupled plasma mass spectrometry (ICP-MS) [5], instrumental neutron activation analysis (INAA) [6, 7], and ultra-low background HPGe and BiPo-3 detectors have frequently been used [7–10] for the determination of the radiopurity of materials.

The most critical factor is the ultra-high radiopurity of the ββ source foils. The SuperNEMO spectrometer is designed in such a way that all radio-decays in source foils are registered with maximal efficiency. Unfortunately, this equally applies to the useful ββ signal and the background from radioactive impurities in foils. This background in the ROI can degrade critically the sensitivity of the experiment. In the SuperNEMO detector the ββ-sources, i. e. the enriched selenium (^{82}Se), are wrapped in 12 µm-thick polyethylene. The radiopurity of the NEMO-3 experiment's ^{82}Se foils was measured precisely: ^{208}Tl − 0.39 ± 0.01 mBq/kg, ^{214}Bi − 1.5 ± 0.04 mBq/kg (1σ statistical errors are given) [11]. A significant reduction (down to ~ µBq/kg level) in the radioactive contamination of the ^{82}Se was one of the main tasks of the SuperNEMO R&D.

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The work presented here concentrates on the development of the radiochemical purification and conditioning) of enriched selenium (^{82}Se) samples produced in a “standard” state, and also on radioanalytical studies of radiopure (conditioned) ^{82}Se produced for the preparation of new ^{82}Se foils. For the “standard” state we mean samples with certain characteristics: isotopically enriched, chemically pure, radiochemically pure, in a physico-chemical state of elemental selenium, and with characteristic particle size of the powder of up to 5 µm.

This paper is a continuation of our previous work [12] concerning chromatographic separation of a small quantity (100 g) of selenium and preparation of selenium oxide (SeO_2) as the final product. In this paper we present the developed method for production of radiopure ^{82}Se in large quantities (1.5 kg and 1 kg batches from JINR Dubna and France, respectively) in the appropriate form (elemental selenium powder) required for the production of thin ^{82}Se foils. Therefore, special attention has been paid to a number of new aspects of the technology:

- Creation of a new chromatographic system with a new larger ion-exchange column optimized for obtaining kilogram quantities of highly radiopure selenium;
- Development of a technique and construction of a facility (system, equipment) for reduction of purified selenious acid to an elemental state;
- Preparation of the final product by decantation, centrifugation, washing, grinding and drying.

Particular attention has been paid to materials from which chemical vessels and instruments are made. These materials should be as pure as possible with respect to radioactive impurities (^{40}K, decay products of ^{232}Th and ^{238}U) and sufficiently resistant to chemical reagents. It should be noted that it is extremely difficult to find chemical vessels from materials which meet the requirements of modern low-background experiments.

The radiopurity of plastic materials of the chromatographic column (PEEK – chromatographic column material, Teflon – column filter) and polypropylene chemical vessels (glasses, bottles, centrifuge tubes) was determined by instrumental neutron activation analysis (INAA). The radiopurity of the final ^{82}Se wrapped in the foils was measured by the BiPo-3 spectrometer [10] (operating in the Canfranc underground laboratory in Spain).

The distribution coefficients of a series of lanthanides (^{88}Y, ^{139}Ce, ^{143}Pm, ^{167}Tm, ^{169}Yb, ^{173}Lu), ^{60}Co and ^{137}Cs (a potassium analog) on the DOWEX 50W × 8 resin, used to produce the high radiopurity selenium, were also determined.

2 Experiment

2.1 Chemical reagents, materials and equipment

The following reagents, materials and equipment were used: bidistilled water; nitric acid (qualification “osch 18-4”, GOST 11125-84); sulfur dioxide – SO_2 (GOST 2918-79, purity – 99.9 %); isopropyl alcohol (qualification “osch”, STP TU COMR 2-018-06); natural technical selenium (^{nat}Se) CT-1 (GOST 10298-79); initial enriched selenium ($^{82}SeO_2$) was obtained in two batches. first part of $^{82}SeO_2$ (isotope composition: ^{82}Se-99.88 %, ^{74}Se-0.01 %, ^{76}Se-0.01 %, ^{77}Se-0.01 %, ^{78}Se-0.04 %, ^{80}Se-0.05 %) purchased in JSC “PA Electrochemical Plant” (Russia) and labeled as “Dubna”; second part of $^{82}SeO_2$ (isotope composition: ^{82}Se-96.1 %, ^{80}Se-3.9 %) purchased in “CHEMGAS” (France) and labeled as “France”; cation-exchange resin DOWEX 50W × 8 (200–400 mesh, catalog No.217514, Fluka/Sigma-Aldrich, Germany); chemical vessels made of polypropylene (glasses, bottles, centrifuge tube); chromatographic column material: PEEK – polyether ether ketone (Elmica, Russia) – material of the chromatographic column (internal diameter of 28 mm and an operating length of 500 mm); three-way valves (stopcock 3-way PFA 6 mm OD, catalog No. 30503-10, Cole-Parmer, USA); peristaltic pumps (Masterflex L/S Computer-Compatible Digital Drive; 600 rpm, catalog No.07551-00, Cole-Parmer, USA); pump tubes (catalog No.77390-60, 4/6 mm Teflon (PFTE) tubing, Cole-Parmer, USA); magnetic stirrer with heating (MSH-30D, Daihan Scientific, Korea); Centrifuge (CL-40, Thermo scientific, USA); centrifuge tube with a volume of 750 mL (catalog No.75003699, Thermo scientific, USA); Homogenizer (HG-15D-Set-B, Daihan Scientific, Korea); vacuum drying cabinet (VDL 23, Binder, Germany); ICP mass spectrometer (x-7, Thermo elemental, USA); ICP atomic emission spectrometer (ICAP-6500, Thermo scientific, USA); AEROTRAK Handheld Particle Counter TSI 9303-01.

2.2 Production of radiopure selenium (purification of selenium)

The complete purification procedure consists of four main stages:

- Preparation of the initial solution of selenious acid (H_2SeO_3);
- Cation-exchange chromatographic separation (purification);

- Reduction of purified selenious acid to an elemental state (Se);
- Conditioning (decantation, centrifugation, washing, grinding and drying) of the final product.

All processes were conducted in a specially prepared clean room equipped with air supply, exhaust and recirculation ventilation systems, air intake devices, air distributors with final filters, air regulating devices, sensor equipment and automation elements, etc. The level of dust particles in the room was measured to be ~100 000 (≥0.5 μm) per m^3 and zero in the laminar box (measurement by the AERO-TRAK Handheld Particle Counter TSI 9303-01) where the chromatography stage was carried out.

2.2.1 Preparation of initial solution of H_2SeO_3

The selenium was divided into portions for purification. The volume of one portion of initial selenious acid solution was 5 L, with selenium concentration of 0.5 mol/L. In the case of natural selenium (^{nat}Se), the solution was prepared by dissolving 200 g of selenium in concentrated nitric acid, evaporating to SeO_2, followed by dissolution in water. In the case of enriched selenium (99.88 % of ^{82}Se), 285 g of selenium oxide ($^{82}SeO_2$) was dissolved in water (equivalent to 200 g of selenium, taking into account mass fraction corrections). The obtained solutions were then filtered.

2.2.2 Purification of selenium

The basic aspects of chromatographic purification of selenium for low-background measurements are described in our previous article [12]. Figure 1 shows the purification system with a reverse flow used in this work. The main changes from the previously used system relate to the components and the overall organization of the system. The unit is specially adapted for multiple repetitions of purification cycles for separate small portions in order to purify, in total, kilogram quantities of selenium.

A chromatographic column (item 10, Figure 1) is filled with cation exchange resin DOWEX 50W × 8 (200–400 mesh). The purification system uses PTFE pumping tubes and connection hoses, and the flow of liquid is ensured by a peristaltic pump. Particular attention is paid to the selection of three-way valves, which must be sufficiently resistant to increased pressure (~5 atm) and to HNO_3 (5 mol/L). Before starting the work, the resin loaded in the column was thoroughly washed (reverse direction) with HNO_3 (2 L, 5 mol/L) and water (0.5 L).

Each purification cycle of a single portion of selenium (200 g) lasted about 3 h and included the following operations, presented in Figure 1:

1. Passing of unpurified selenious acid (5 L, 0.5 mol/L) solution through the column at a rate 40 mL/min in the forward direction (vessel 1→column→vessel 4).
2. Washing the column with HNO_3 (1 L, 0.1 mol/L) in the forward direction (vessel 2→column→vessel 4).
   The solutions from vessels 1 and 2 were eluted from the column and collected in a plastic vessel 4 for the purified H_2SeO_3 (6 L).
3. Elution of impurities adsorbed on the resin in the reverse direction (vessel 6→column→vessel 3) with HNO_3 (6 L, 5 mol/L).
4. Rinsing the resin from the HNO_3 (5 mol/L) residuals in the reverse direction (vessel 5→column→vessel 3) with HNO_3 (1 L, 0.1 mol/L).

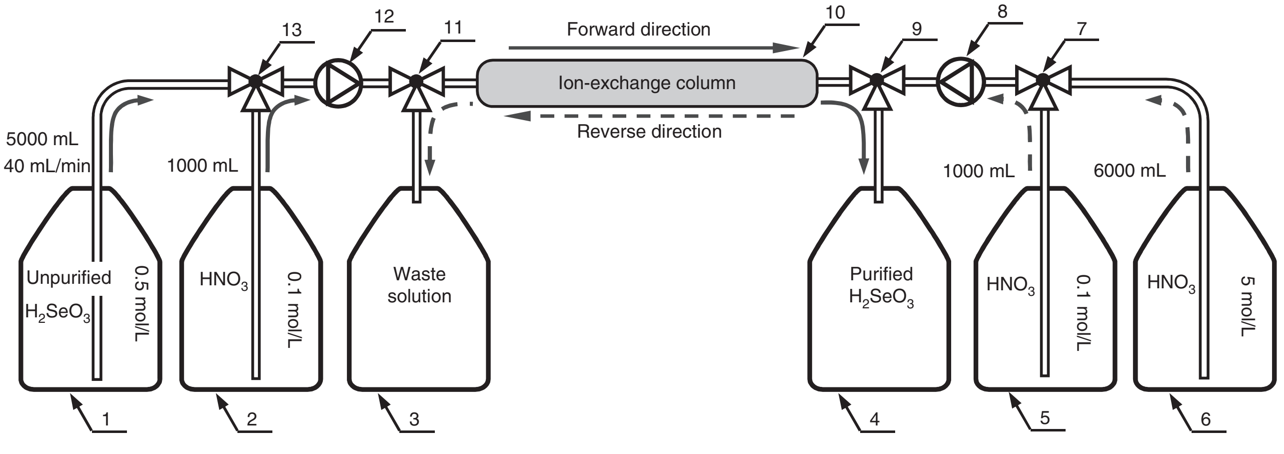

Figure 1: Chromatographic separation (purification) scheme of selenium by the reverse method.
1–6 – polypropylene vessels with the solutions; 7, 9, 11, 13 – triple valves; 8, 12 – peristaltic pumps; 10 – ion exchange column.

These solutions from vessels 6 and 5 were eluted through the column and collected in a separate vessel 3 (7 L waste (impurities) solution).

After the last operation, the purification system is ready to process the next portion of selenium.

2.2.3 Reduction of selenious acid to the elemental state

In order to obtain elemental selenium the purified selenious acid (vessel 4, Figure 1) was reduced with sulfur dioxide (SO_2) according to the reaction

$$H_2SeO_3 + 2\ SO_2 + H_2O \rightarrow Se + 2\ H_2SO_4. \qquad (1)$$

Figure 2 shows the scheme of the reduction system used. A purified solution of H_2SeO_3 (vessel 4, Figure 1) was placed in vessel 6 (6 L; 5 L of original + 1 L of washing solution) and saturated through the surface with sulfur dioxide.

To increase the absorption of SO_2, the solution was agitated with a magnetic stirrer. The flow of SO_2 was regulated (valve 2 and rotameter 3, Figure 2) to provide almost complete absorption of SO_2 with a small excess of gas released by bubbles in vessel 8. The average flow of SO_2 was ≈10 L/h. The reduction of one portion took 6–7 h. The saturation of the solution through the surface is more convenient than bubbling due to the good solubility of SO_2, otherwise the tube is quickly clogged with reduced selenium. The use of gas (SO_2) in the recovery process is preferable for our purposes to using other reagents (liquid, solid) since it minimizes radioactive contamination of the final product.

2.2.4 Conditioning

The reduced elemental selenium was separated from the residual solution (Figure 2, vessel 6) by decanting and centrifugation, and then was collected in 750 mL centrifuge tubes. About 200 g of selenium (a single processed portion) was stored in each tube (~300 mL of the wet material). Even small residues of sulfuric acid formed in the process of selenium reduction create serious difficulties in the process of making the selenium source. Therefore, the selenium was further washed, first 10 times with distilled water, and then 5 times with isopropyl alcohol. For this purpose, water (or isopropyl alcohol) was added to the centrifuge tube with a single selenium portion until the tube was completely filled (750 mL mark), then gently blended with a Teflon stick, decanted and centrifuged.

During the reduction, elemental selenium was recovered in the form of tiny particles (<10 nm) which, over time, especially during centrifugation, coalesce into large crumbs and flakes of millimeter portions. They are not suitable for the production of thin ^{82}Se foils. Therefore, a final milling of the selenium was carried out with a homogenizer in isopropyl alcohol until a homogeneous suspension was obtained (~5 min, ~500 rpm). After grinding, the characteristic size of selenium particles was of the order of μm, which is suitable for the production of thin foils (average thickness of 300 μm). The use of surfactant isopropyl alcohol in the final stages of washing and grinding also reduces the adhesion of selenium. In addition, because of its good volatility, isopropyl alcohol facilitates the drying process.

Selenium drying was carried out for a day in a vacuum cabinet, without heating, at an average vacuum of ~5 mbar.

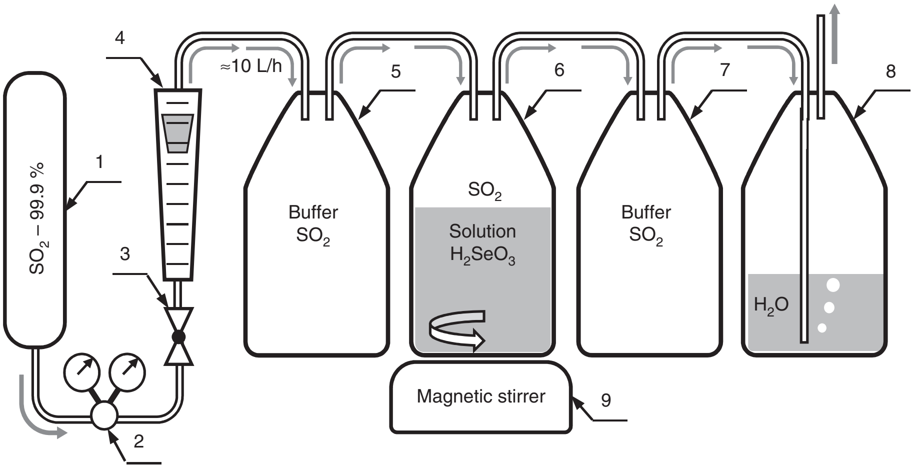

Figure 2: The Scheme for the reduction system of selenious acid (H_2SeO_3) to an elemental state (Se). 1 – a cylinder with SO_2, 2 – a reducer, 3 – a control valve, 4 – a rotameter, 5–8 – polypropylene vessels (10 L), 9 – a magnetic stirrer.

For each selenium portion (200 g), all washing, grinding and drying processes were carried out in the same centrifuge tube.

During conditioning, we tried to reduce the speed and time of centrifugation (typical value ~1300 rpm for 2 min), and, if possible, to replace this procedure with settling the solution; this reduces the formation of coarse-grained selenium and reduces the grinding time. The grinding process can lead to contamination of selenium by the attrition products of the homogenizer nozzles made of stainless steel (Fe, Cr, Ni), which in turn can contain radioactive impurities.

It should be emphasized that reduction and washing leads to the removal of thorium, uranium, radium, actinium and potassium as they remain in solution.

A total of 600 g of natural selenium was purified and conditioned in order to test and refine the method. The resin in the column was then renewed and used to purify 2.5 kg of enriched selenium (^{82}Se).

2.3 Measurement of the distribution coefficients of radionuclides

The following radionuclides were used as indicators: ^{60}Co ($t_{1/2}=5.27$ years), ^{88}Y ($t_{1/2}=106.63$ days), ^{137}Cs ($t_{1/2}=30.08$ years), ^{139}Ce ($t_{1/2}=137.64$ days), ^{143}Pm ($t_{1/2}=265$ days), ^{167}Tm ($t_{1/2}=9.25$ days), ^{169}Yb ($t_{1/2}=32.02$ days), ^{173}Lu ($t_{1/2}=1.37$ years) [13]. Radionuclides ^{88}Y, ^{139}Ce, ^{143}Pm, ^{167}Tm, ^{169}Yb, and ^{173}Lu were obtained by irradiating a tantalum target with 660 MeV protons [14]. The ^{60}Co and ^{137}Cs radionuclides were of commercial origin.

To determine the distribution coefficients (K_D), the static adsorption method was used. Certain volumes of selenium acid solution, H_2O, and 10 µL of the prepared radionuclide cocktail were added to a sample of 50 mg resin (DOWEX 50W × 8) and placed in a 1.5 mL centrifugation tube (“Eppendorf”, Germany) so that the volume of each sample was 1 mL. The samples were then thoroughly mixed and left to stand for 24 h to achieve equilibrium. Then, centrifugation was carried out and the radioactivity of the solution was measured with a HPGe γ-spectrometer. The distribution coefficients were calculated using the equation:

$$K_D = \frac{A_{1\ \mathrm{g}}(\mathrm{resin})}{A_{1\ \mathrm{mL}}(\mathrm{solution})} = \frac{A_{50\ \mathrm{mg}}(\mathrm{resin})}{A_{50\ \mu\mathrm{L}}(\mathrm{solution})}, \qquad (2)$$

where $A_{50\ \mathrm{mg}}$ (resin) is the radioactivity (Bq) of 50 mg of resin, and $A_{50\ \mu\mathrm{L}}$ (solution) is the radioactivity (Bq) of 50 µL of the solution. To reduce uncertainties in the determination of these values, samples were weighed immediately after preparation and after fluid extraction.

2.4 Instrumental neutron activation analysis of plastic materials

Determination of thorium and uranium contamination in plastic materials of the chromatographic column (PEEK – column material, 2 Teflon samples – construction material and filter for the column) and polypropylene chemical vessels (glasses, bottles, centrifuge tubes) was carried out by INAA. Prepared samples of the plastic materials together with standard reference samples were irradiated at the IBR-2 reactor (JINR, Dubna) in a flux of epithermal neutrons of 2.2×10^{11} cm^{-2} s^{-1}. The standard materials IAEA 433 (marine sediment) and BCR 667 (estuarine sediment) were used as reference samples. The radioactivity of the samples and standards was measured with a HPGe detector GC5519 (Canberra, USA) with relative registration efficiency of 55 %. The collection and processing of gamma-spectra were carried out using Genie-2000 software [15].

2.5 Measurements with a BiPo-3 spectrometer

The BiPo-3 detector is a low radioactivity detector dedicated to the measurement of ^{208}Tl and ^{214}Bi in thin materials. The detector, running in the Canfranc Underground Laboratory since 2013, was initially developed to measure the radiopurity of the ββ-source foils for the SuperNEMO experiment. A comprehensive description of the design and performance of the detector, as well as the results of the radioactivity measurements of the first ^{82}Se foils prepared for the SuperNEMO experiment, can be found in [9]. The BiPo-3 detector represents at present the most sensitive radiometrics detector for radiopurity measurements of samples wrapped in foils. The obtained detection limits were 2 µBq/kg (90 % CL) for ^{208}Tl and 140 µBq/kg (90 % CL) for ^{214}Bi after 6 months of measurement.

2.6 Source foil production

The prepared source foils from the radiopure (conditioned) natural selenium (600 g) and enriched ^{82}Se (2.5 kg) were made in LAPP (Annecy, France); details of the foil production can be found in [16]. The surface density of the foil is ~55 mg/cm^2. The foils were carefully packed, transported by car from Annecy to the Canfranc underground laboratory, unpacked and installed in the BiPo-3 detector. It is necessary to emphasize once again that all materials, substances, and tools used in the foil preparation procedures have been carefully screened using low-background HPGe spectrometers and the BiPo-3 (foil) detector in order to select samples with the best radiopurity. All the details of the foil production protocol will be presented in a dedicated publication.

3 Results and discussion

The radiopurity of the final product (at a given radiopurity of the initial selenium) depends on the following factors:

- The characteristics of the chromatographic process (distribution coefficients, parameters of the column, the flow of solutions, etc.). In such chromatographic separation systems, it is usually possible to achieve very high purification coefficients. Our experience shows that the obtained purification coefficients on thorium and uranium in the chromatographic system exceed 10^5.
- Radiopurity of the liquid chemical reagents (bi-distilled water, nitric acid and isopropyl alcohol).
- Radiopurity of plastic materials of the chromatographic column and polypropylene chemical vessels.
- General organization of the procedure; for example, one needs to minimize the number of operations with higher risk of contamination of the final product.

3.1 Chemical yield of selenium

The chemical yield of the selenium recovery in the chromatographic purification procedure is more than 99 %. The main losses occur during conditioning. The residue is collected for subsequent extraction of ^{82}Se. The losses during chromatography are negligible – less than 0.1 %, according to the ICP-AES/MS analysis of the (waste (impurities) solution (vessel 3 in Figure 1).

3.2 Distribution coefficients of radionuclides

The distribution coefficients (K_D) of lanthanides (^{88}Y, ^{139}Ce, ^{143}Pm, ^{167}Tm, ^{169}Yb, ^{173}Lu) and technogenic radionuclides – ^{60}Co and ^{137}Cs (a potassium analog) in the DOWEX 50W × 8 – H_2SeO_3 system were determined experimentally (Table 1). It can be seen that the K_D values of rare-earth elements with decreasing concentration of H_2SeO_3 (from 4 to 0.5 mol/L) exceed 10^4.

Table 1: Distribution coefficients (K_D) of lanthanides, as well as cobalt and cesium on the cation – exchange resin DOWEX 50W × 8 at various concentrations (C) of selenious acid in solutions.

C(H_2SeO_3), mol/L	**Distribution coefficients (K_D) of radionuclides**			
	^{60}Co	**^{88}Y**	**^{137}Cs**	**^{139}Ce**
4	400	>10^4	55	>10^4
3	1350	>10^4	160	>10^4
1	6700	>10^4	970	>10^4
0.5	~10^4	>10^4	1400	>10^4
	^{143}Pm	**^{167}Tm**	**^{169}Yb**	**^{173}Lu**
4	>10^4	>10^4	~10^4	~10^4
3	>10^4	>10^4	>10^4	>10^4
1	>10^4	>10^4	>10^4	>10^4
0.5	>10^4	>10^4	>10^4	>10^4

The K_D values of cobalt and caesium are lower, but with a decrease in the concentration of selenious acid to a working concentration of 0.5 mol/L, the coefficients increase significantly, exceeding the critical values with respect to U. Thus, when the solution passes in the forward direction in a volume exceeding 100 times the volume of the column, the radionuclides and their analogs do not enter the purified solution.

3.3 Radiopurity of plastic materials

INAA was used to determine the radiopurity of the plastic materials used: the chromatographic column and polypropylene chemical vessels, as explained in the Section 2.4. Table 2 presents the INAA results for the plastic materials that are in direct contact with solutions: PEEK (chromatographic column material), Teflon (filter for the column and construction material), polypropylene vessels (glasses, bottles, centrifuge tubes) and polyethylene terephthalate (often used for making chemical vessels). The last two materials were not used in the setup but were measured for completeness and comparison.

The most radiopure plastic materials are usually Teflon and high-pressure polyethylene, although it should be noted that there can be exceptions to this rule. Typically, low-density polyethylene and polypropylene have approximately the same level of contamination in thorium and uranium. Polypropylene is still more mechanically resistant than polyethylene, and therefore in many cases its use is preferable. The obtained results show that the materials used are reliable for the whole

Table 2: Radiopurity of various plastic samples measured by INAA.

Samples	Mass, g	Irradiation/measurement time, h	Th content, g/g	U content, g/g
PEEK	0.49	93/0.5–1.5	$(3.8\pm0.7)\times10^{-8}$	$(4.3\pm0.2)\times10^{-8}$
Teflon (filter for column)	0.69	93/0.5–1.5	$(4.9\pm0.8)\times10^{-8}$	$(4.8\pm0.2)\times10^{-8}$
Teflon – material for construction	1.10	71/0.5–7	$<3\times10^{-10}$	$<9\times10^{-11}$
Glass, PP	0.37	93/0.5–1.5	$(5.5\pm1.0)\times10^{-8}$	$(1.3\pm0.1)\times10^{-7}$
Bottle 5 L, PP	0.17	71/0.5–10	$<2.9\times10^{-9}$	$<1.4\times10^{-9}$
Bottle 10 L, PP	0.19	71/0.5–10	$<2.6\times10^{-9}$	$<1.1\times10^{-9}$
Centrifuge tube, PP	2.76	71/0.5–10	$(6.9\pm0.4)\times10^{-9}$	$(2.7\pm0.2)\times10^{-9}$
Polyethylene terephthalate	0.18	71/0.5–10	$<2.4\times10^{-8}$	$<2.6\times10^{-8}$

PEEK, Polyether ether ketone; PP, polypropylene.

of our purification procedure. Of course, it is better to use the purest material (if available), however, the contamination levels in the selected plastic materials do not exceed ~10^{-8} g/g for uranium. From our point of view, such levels are not critical for vessels and materials that are used repeatedly, especially for a chromatographic column. The surface of such materials will be cleaned during multiple cycles and will be in balance with the solutions used in the purification process. The radiopurity of the materials will be reduced to the radiopurity of the water and chemicals used in the purification process. As a result, we used chemical vessels made of polypropylene (with the exception of pump hoses from teflon). According to Table 2, these were the purest products available to us from the relevant materials, and their use should not substantially affect the radiopurity of the resulting selenium samples. It should be noted that INAA is, because of good sensitivity and the ability to obtain results relatively quickly, one of the most successful methods for analysis of thorium, uranium and potassium in plastic materials.

3.4 Purity of liquid chemical reagents

Data on the purity of bi-distilled water and nitric acid can be found in our previous work [12]. The bi-distilled water does not contain thorium and uranium (detection limits were $<4.6\times10^{-13}$ g/g and $<1.7\times10^{-13}$ g/g, respectively). ICP-AES/MS analysis of isopropyl alcohol (1 L of isopropyl alcohol was evaporated in a Teflon vessel at a temperature of 50 °C; the residue remaining in the pot was analyzed with ICP-AES/MS) showed that the content of thorium and uranium was 6.7×10^{-14} g/g and 1.4×10^{-13} g/g, respectively. As can be seen, the content of Th and U is very low, therefore it should not affect the purity of the selenium samples produced.

3.5 Measurement of selenium foils with the BiPo-3 spectrometer

Unfortunately, the ICP-AES/MS analyses were not sensitive enough to determine K, Th and U levels in the initial and final (purified) selenium samples (both the natural and the enriched). Only detection limits for the K, Th and U were obtained. For thorium and uranium detection limits obtained by ICP-MS were 8×10^{-9} g/g and 3×10^{-10} g/g for the natural Se and for the enriched selenium (^{82}Se) samples, respectively. Further work is in progress to analyze these samples by ICP-MS and accelerator mass spectrometry. The obtained detection limit for K by ICP-AES was 10^{-7} g/g.

The source foils containing ~150 g of natural selenium (^{nat}Se) and 2.5 kg of purified (conditioned) ^{82}Se were measured, however, with the BiPo-3 spectrometer (Table 3).

Measurements (with interruptions) took place between December 2016 and August 2017, with a total exposure of ~130 days. The results obtained (Table 3) show lower ^{208}Tl (^{232}Th) and ^{214}Bi (^{238}U) levels for enriched selenium (^{82}Se) than for the natural (^{nat}Se) sample which was the first sample used for optimization of the method. This

Table 3: Results of ^{208}Tl and ^{214}Bi in the purified ^{82}Se samples wrapped in polyethylene foils measured by the BiPo-3 detector (90 % CL).

Origin of selenium foils	The specific activity, μBq/kg	
	^{228}Th (^{232}Th)/^{208}Tl	^{226}Ra/^{214}Bi
High radiopurity (conditioned) ^{nat}Se	19-336/7-121	<870/<870
High radiopurity (conditioned) ^{82}Se “Dubna”	22-150/8-54	<600/<600
High radiopurity (conditioned) ^{82}Se “France”	<294/<106	<1370/<1370

shows the suitability of the technique. The radionuclide levels in ^{82}Se (purified and conditioned in JINR Dubna) foils which will be used in the SuperNEMO experiment, are lower by factors of about 10 (^{208}Tl) and 3 (^{214}Bi) than in the ^{82}Se foil used in the NEMO-3 experiment [11].

4 Conclusions

The methods for the preparation of radiopure ^{82}Se sources for the SuperNEMO neutrinoless double-beta decay experiment have been developed. The chromatographic separation, reduction and conditioning have been used consistently to produce 2.5 kg (from total 6.23 kg) of highly dispersed, high radiopurity (conditioned) ^{82}Se. Grinding and washing were used additionally to obtain the highly dispersed powder (particle size ~1 µm) of elemental selenium as the final product, required for the production of thin source foils.

Key features of the proposed technology should be noted:

- use of ion-exchange chromatography for elution of impurities with reverse method;
- minimum number of chemical reagents (bidistilled water, nitric acid and isopropyl alcohol) used;
- use of sulfur dioxide (SO_2) as a reducing agent for obtaining of elemental selenium;
- high chemical yield (>99 %), unique radionuclide purity and high dispersion of final product.

The productivity of technology proposed is 300 g of ^{82}Se per day, which makes it possible to obtain tens and hundreds of kilograms of purified (conditioned) selenium.

Particular attention was paid to the selection of pure materials, liquid chemical reagents (water, nitric acid and isopropyl alcohol) and plastic vessels which were used at all stages. One needs also to highlight the methodological studies of the various plastic materials radiopurity (ultra-low contaminations of thorium and uranium) performed by INAA method, which is found to be the best for these purposes. It has been shown, it is preferable to use products from Teflon and, if necessary, polypropylene.

The radiopurity of the final ^{82}Se foils has been measured by BiPo-3 spectrometer. The levels of impurities were found to be 8–54 µBq/kg level for ^{208}Tl (the ^{232}Th chain), and ≤600 µBq/kg for ^{214}Bi (the ^{238}U chain). This corresponds to an improvement in the radiopurity of the selenium foils by about a factor of 10 for ^{208}Tl and at least by about a factor of 3 for ^{214}Bi in comparison with the NEMO-3 experiment.

The ^{82}Se source foils produced by described techniques were approved for using in SuperNEMO, an experiment aimed to search for neutrinoless double-beta decay of ^{82}Se. The foils have been installed in the Demonstrator (the first module of the experiment) and data-taking will start at the end of 2019.

Acknowledgments: We acknowledge support by the staff of the Modane Underground Laboratory and by the MEYS of the Czech Republic (grant number EF16_013/0001733), APVV in Slovakia, CNRS/IN2P3 and Labex Enigmass (A. Remoto) in France, RFBR in Russia (Project No. 16-52-16018 NCNIL a and No. 19-52-16002 NCNIL a), STFC in the UK and NSF in the USA.

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