RFNC-VNIIEF CAPABILITIES TO PRODUCTION HIGH PURE ISOTOPES FOR SCIENTIFIC AND MEDICAL APPLICATIONS

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ABSTRACT

In the technical paper there is presented the information on the basic equipment and more than thirty-year experience of RFNC-VNIIEF activities in the sphere of producing highly enriched isotopes of actinide elements – thorium, uranium, neptunium, plutonium, americium and curium – for scientific researches and practical applications.
Electromagnetic separator and radiochemical methods provide obtaining of superpure isotope samples for nuclear-physical radiometric and mass-spectrometric equipment, and also as tracers when analyzing environmental contamination.
There are presented the structure of the laboratory occupied with these isotopes electromagnetic separation as well as the nomenclature and characteristics of the specimens supplied.
There are stated science and engineering elaborations of technologies aimed at producing alpha-ray radiating radionuclides – thorium-229, thorium-228, actinium-225, radium-224 – for the purpose of anti-cancer therapy using bismuth-212 and bismuth-213 produced by the specially developed generators.
There are presented the basic directions of cooperation with other Russian Institutes in developing this promising line of conversion.

INTRODUCTION

The Russian Federal Nuclear Center founded in 1946 to design Russian nuclear weapons has turned into the largest scientific research center, to great extent influencing the modern science development in the various fields of the fundamental and applied research. The research in the field of experimental nuclear physics performed on the experimental base and measuring technique concentrated in the Institute for nuclear and radiation physics are of great importance in RFNC activities.

The main fields of these research are as follows:

- Study of fission and fusion nuclear reactions, measurement of nuclear data for a wide range of elements and construction materials, measurement of critical mass, study of critical mass configurations, measurement of integral nuclear constants.
- Creation, development and application techniques of radiation characteristics measurement for pulse and stationary regimes.
- Research in the field of radiochemistry, mass-spectrum and isotope analyses using super pure actinide isotopes, including transuranium elements, development of radiochemical measurement techniques and technologies for different purposes.
- Design and creation of the laboratory experimental base for nuclear physics research on the basis of electrophysics and nuclear physics facilities, using radioisotopes.
- Study of possibility of transuranium elements transmutation, the latter containing in radioactive wastes of nuclear power engineering, measurements of nuclear constants, necessary for transmutation process development.

• Monitoring of environmental contamination (soil, underground waters, etc.) with actinide elements, creation of technology for the contaminated areas, decontamination, monitoring of residual actinide content.

To provide this research program, the actinide isotopes electromagnetic separation laboratory that operates a mass-separator, S-2 type, equipped with personnel radiation protection to produce super pure isotopes of uranium, plutonium, americium and curium was set up in 1967 in the Radiochemical Department. There is also a group dealing with production of layers and targets of these isotopes for nuclear physics experiments. The isotope production of the Radiochemical Department was delivered to different Russian research institutes to perform the state program on nuclear constants measurements for actinide elements. Not long ago this production became available for the foreign laboratories, as well.

THE MAIN CHARACTERISTICS OF S-2 MASS-SEPARATOR

The main characteristics of mass separator S-2 are described in a series of publications and are shown in table I.

Table I. Basic parameters of the S-2 mass separator

Magnetic field form	$H=H_0r_0/r$
Average trajectory radius, r_0	1000 mm
Average intensity of magnetic field, H_0	4500 Oe
Ion beam angle in magnetic field	114.6° (2 rad)
Total length of average trajectory	6000 mm
Dispersion for 1% relative mass difference	20 mm
Accelerating voltage	up to 45 kV
Ion source	plasma-type with filamentary longitudinal cathode, bicrucible
Maximum temperature of crucibles	1000°C
Crucible capacity (large) (small)	10 g 1 g
Ion source current	up to 10 mA
Isotope receiver	slit-type with a variable number of boxes
Material utilization factor during separation	~5%
Evacuation system	four steam-oil apparatus of 5000 1/s pumping rate and mechanical pumps
Working vacuum	$2\cdot10^{-6}$-10^{-5} Torr
Electromagnet weight	18 t
Power	50 kW

As the working substance in the ionic source of S-2 mass-separator the anhydrous trichlorides of the actinide elements used in the amount of about 1 gram are utilized. The efficiency of the working substance application in the one separation cycle is in the average about 5% of the initial mass, all the isotopes of the element separated accumulating simultaneously in the isotope receivers. The mass-separator configuration envisages the possibility of the dispersed

substance collection from the inner surfaces, thus, from 50 % to 80% of the initial mass can be reused for separation. The enriched isotopes are accumulated in the receivers made of super pure copper or aluminium. The isotopes are collected with the help of the nitric or hydrochloric acids. The radiochemical methods provide obtaining super pure preparations, free from mould materials and radioactive contamination by the other elements, the chromatographic and extraction methods to determine the separating substance content being used. The quality of the isotopes enriched and the isotopes contamination content are determined only by the effects connected with chromatism and dispersion in the electromagnetic system of S-2 installation .

THE NOMENCLATURE OF THE ISOTOPES PRODUCED.

To certify the highly-enriched preparations produced, the methods of mass- and alpha-spectrometry are used. The mass-spectrometer MI3340 type with the source of thermal ionization allows to determine isotope contamination at the level of $5 \cdot 10^{-7}$, the sensitivity of the analysis for uranium being 1500 atoms/ion. The accuracy of the isotope analyses reaches 0.1% relat.(2v) for ratios > 10^{-2}, and 1-3% relat.(2v) for ratios<10^{-4}. To determine isotopes like ^{232}U, ^{236}Pu, ^{238}Pu, ^{242}Cm, ^{244}Cm, alpha-spectrometry methods are applied in the highly enriched preparations.

For some nuclear-physical experiments it is essential to possess isotopes preparations of high chemical purity, the content of contamination being at the level not more than 10^{-7}-10^{-8}. The methods developed in the Radiochemical Department provide such a degree of purification from contamination with the other actinides. The content of the other elements in the preparations delivered is not more than 0.1 %. The monitoring of chemical contamination is exercised through mass-spectrometer usage with a spark ionic source and absolute measurements of contaminators concentration.
The nomenclature of the isotopes delivered and their characteristics are shown in the table 2. In the lines *) the characteristics of the preparations obtained by the double fusion process are shown. To monitor the environmental contamination and purification technologies, the following super pure isotopes are suggested (see table 3.):

METHODS OF SAMPLES PRODUCTION

Depending on nuclear-physical experiment designation and edition, various configurations of the targets and sources, not to mention the methods of plotting the active substance on carrier of different material are being used. The application of the plotting methods with volatile organic compounds of transuranium elements in the vapour phase are of impressive results.

Such methods were developed for thorium, uranium, americium, curium and provided highly-efficient and even precipitation of the substance on the carrier of various materials: aluminum and its alloys, nickel, copper, bronze, titanium, zirconium, glass, quartz, ceramics. For americium the most considerable results were obtained on ferrum and steel.
The technologies of the plotting uranium and trasuranium isotopes on the super thin (~ 2μm) metal carrier with the additional protection by polymeric and metal coverings are developed. This helps to increase the safety of the activities performed with such layers and decrease the substance losses during the measurements.

Table II. Isotope contents of highly enriched samples

Isotope	Main isotope contents and impurities (as atoms. %)					
Uranium	232	233	234	235	236	238
U-233	$0.5\cdot10^{-3}$ act.	99.97	0.03	$0.1\cdot10^{-2}$	$<10^{-3}$	$0.1\cdot10^{-2}$
U-234		15.3	84.52	0.13	$1.3\cdot10^{-3}$	$4\cdot10^{-2}$
U-235		$1.7\cdot10^{-4}$	$2.6\cdot10^{-3}$	99.9923	$4.5\cdot10^{-3}$	$4.3\cdot10^{-4}$
U-236			$< 1.27\cdot10^{-2}$	1.41	97.81	0.763
U-238		$< 1.8\cdot10^{-4}$	$<1.58\cdot10^{-4}$	$<4.03\cdot10^{-4}$	$<2.82\cdot10^{-4}$	99.9990

Isotope	Main isotope contents and impurities (as atoms. %)	
Neptunium	236	237
Np-237	<0.01	99.99

Isotope	Main isotope contents and impurities (as atoms. %)					
Plutonium	238	239	240	241	242	244
Pu-238	99.6	0.4	0.015			
Pu-239		99.5 99.997	0.5 $2\cdot10^{-3}$	 $2\cdot10^{-4}$	 $1\cdot10^{-4}$	
Pu-240		$4\cdot10^{-3}$ $3\cdot10^{-2}$	99.9 90.7	$6\cdot10^{-2}$ 0.2		
Pu-241		$<10^{-3}$ $<10^{-4}$	0.17 $3\cdot10^{-4}$	99.6 99.998*	0.23 $1.6\cdot10^{-3}$	$<10^{-3}$ $<10^{-4}$
Pu-242		10^{-3} $5\cdot10^{-4}$	0.1 $5\cdot10^{-4}$	2.0 0.04	97.8 99.96	$7\cdot10^{-4}$ $5\cdot10^{-4}$
Pu-244		$5\cdot10^{-2}$ $3\cdot10^{-4}$	0.4 0.3	$5\cdot10^{-2}$ $7\cdot10^{-3}$	1.8 0.7	97.8 98.9

Isotope	Main isotope contents and impurities (as atoms. %)		
Americium	241	242m	243
Am-241	99.99		
Am-242m	30.5 13.0	55.1 85.6	18.2 1.6
Am-243	0.7 $1.6\cdot10^{-3}$	0.1 $4\cdot10^{-4}$	99.2 99.998

Isotope	Main isotope contents and impurities (as atoms. %)					
Curium	243	244	245	246	247	248
Cm-243	93.3 99.99*	0.6 $8\cdot10^{-3}$	$4\cdot10^{-4}$ $5\cdot10^{-5}$	0.6 10^{-4}	$<10^{-2}$ $<10^{-4}$	$<10^{-2}$ $<10^{-4}$
Cm-244	$1.5\cdot10^{-2}$	99.3	$6\cdot10^{-2}$	$4\cdot10^{-3}$	$<10^{-3}$	$<10^{-2}$
Cm-245		1.3 $6\cdot10^{-3}$	98.4 99.998*	0.3 $2.5\cdot10^{-3}$		
Cm-246		$<10^{-2}$ $<10^{-2}$	6 $8\cdot10^{-3}$	98.0 99.8	$<10^{-2}$ $<10^{-2}$	$<10^{-2}$ $<10^{-2}$
Cm-247	0.5	12 2.7	7 0.8	7 5	72.3 90.2	1.4 0.5
Cm-248	0.1 0.1	2.8 1.7	0.04	1.24 0.7	0.1	 97.5

Table III. Isotopic contain of uranium, plutonium and americium tracers for environment measurements

Uranium-233		99,9472%	(mass)
	Uranium-232	(0,5 ±0,1)	$\cdot 10^{-3}$%
	Uranium-234	(0,5035±0,0019)	$\cdot 10^{-1}$%
	Uranium-235	(0,108±0,002)	$\cdot 10^{-2}$%
	Uranium-238	(0,133±0,002)	$\cdot 10^{-2}$%
Plutonium-242		99,98%	(mass)
	Plutonium-238	(5±1)	$\cdot 10^{-5}$%
	Plutonium-239	(6±1)	$\cdot 10^{-3}$%
	Plutonium-240	(7±1)	$\cdot 10^{-4}$%
	Plutonium-241	(8±1)	$\cdot 10^{-3}$%
	Plutonium-242	<(2±1)	$\cdot 10^{-4}$%
Americium-243		99,998%	(mass)
	Americium-241	(1,6±0,4)	$\cdot 10^{-3}$%
	Americium-242m	(4 ±1)	$\cdot 10^{-4}$%
	Curium-244	<(7 ±2)	$\cdot 10^{-2}$% (act.)

STABLE ISOTOPE PRODUCTION

For the last years RFNC has turned a new leaf in its work: in 1993 the activities on stable isotopes and extremely pure substances production were organized with participation of firm "Rinvers-N" (N.Novgorod).

In RFNC-VNIIEF an experimental area of high-speed centrifuges for isotope separation by centrifugal method has been already put into operation. The main task of the experimental area is mastering isotope centrifugal separation by RFNC-VNIIEF specialists and training staff for fulfilling the basic project on creation of the powerful production, the realization of which started in 1996.

The project purpose is to utilize the equipment and technology developed by Design Office "GAZ", the city of Nizhny Novgorod. The production being organized uses the new gas centrifuges models capable to efficiently separate both heavy and light isotopes.
The centrifuges are united into the technological blocks, designed constructively in such a way that they allow to actively reorganize the lines on different isotopes production. It, in its turn, will help to meet the demands of the market and enlarge the range of the isotopes production.

The initial program of our experimental production envisaged the simultaneous output up to 6 different isotopes of sulfur, xenon, krypton, cadmium and zink. The capacities of the main production will allow to simultaneously produce up to 30 different isotopes, see table IV.

Table 4. Main characteristics of producing stable isotopes.

Substance	Isotope or compound	Atom's ratio of isotope (%)	Mass ratio of substance (%)
Ge-76	metal	65-95	99.8
Ge-76	oxide	85-99.7	99.8
Cd-112, Cd-114, Cd-116	metal	60-99	99.95
Cd-112, Cd-114, Cd-116	oxide	40-96	99.95
Kr-78, Kr-80, Kr-82, Kr-83,Kr-84, Kr-86	Elementary	90-99	99.9
Xe-124, Xe-126, Xe-128, Xe-129, Xe-130, Xe-131, Xe-132, Xe-134, Xe-136	Elementary	90-99	99.9
Ni-58	metal	77-99.9	99.95
Ni-58	oxide	88-99.9	99.93
Se-78	Elementary	74-98	99.9
S-33, S-34, S-36	Elementary	10-98	99.8
Te-126	metal	60-99	99.95
Te-126	oxide	60-94	99.95
Cr-50	metal	80-97.7	99.85
Cr-50	oxide	80-97.7	99.85
Zn, impoverished in isotope Zn-64	metal	<1	99.9
Zn-64, Zn-66, Zn-67, Zn-68, Zn-70	metal	>34-99.5	99.95
Zn-64, Zn-66, Zn-67, Zn-68, Zn-70	oxide	>34-99.2	99.85

ISOTOPES FOR MEDICAL APPLICATIONS

RFNC-VNIIEF has lately paid particular attention to the development of the program of producing radionuclides for medical purposes.

In connection with the conversion of defense trend there is being developed the technology of thorium, radium and actinium production to get bismuth-212 and bismuth –213 so that it is used as a compound of radioisotopes to be applied in cancer therapy. This direction is seemed to be promising as clinical researches of medical preparations containing bismuth-212 and bismuth-213 performed in a set of countries made it possible to get impressing results at treating such diseases as quinsy and chronic leukemia, melanoma etc.

To produce actinium-225, there is used thorium-229 accumulated within the long years (up to 50 years) of storing uranium-233 containing small admixtures of uranium-232. The developed technologies provide for practically complete extraction of thorium isotopes and return of uranium-233 with low technological losses.

The developed radiochemical techniques and corresponding laboratory equipment will provide for the possibility of monthly production and transportation to customers of up to 80-100 mCi of actinium-225 and up to 2-2,5Ci of radium-224 (bismuth-212).

It is assumed that there will be organized the corresponding laboratory and services ensuring quick delivery of radioactive specimens (preparations) to medical research centers. This program is being developed jointly with a set of other scientific centers of Russia.

Now we can monthly produce some tens of mCi of actinium-225 and radium-224 and pass this nuclide to medical research centers.

CONCLUSIONS

The highly enriched and super pure isotopes produced in RFNC are of the unique characteristics on enrichment. They are applied in lots of laboratories and research centers both of Europe and Asia, with which we are in close contact and business relations. We possess transport containers that meet all the international rules of radioactive substances transportation. The articles produced and delivered by us containing highly enriched isotopes have been highly approved by the customers. In recent years our products have been used for the creation of a new generation of standards of nuclear materials for the programs dealing with non-proliferation control.
We would be pleased to find new customers.

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