We are IntechOpen, the world's leading publisher of Open Access books
Built by scientists, for scientists

6,900
Open access books available

186,000
International authors and editors

200M
Downloads

Our authors are among the

154
Countries delivered to

TOP 1%
most cited scientists

12.2%
Contributors from top 500 universities

CLARIVATE ANALYTICS
BOOK CITATION INDEX
INDEXED

WEB OF SCIENCE™

Selection of our books indexed in the Book Citation Index in Web of Science™ Core Collection (BKCI)

Interested in publishing with us?
Contact book.department@intechopen.com

Numbers displayed above are based on latest data collected.
For more information visit www.intechopen.com

7

Low-Waste and Proliferation-Free Production of Medical Radioisotopes in Solution and Molten-Salt Reactors

D. Yu. Chuvilin, V. E. Khvostionov,
D. V. Markovskij, V. A. Pavshouk and V. A. Zagryadsky
NRC "Kurchatov Institute"
Russian Federation

1. Introduction

There is an old dispute what is more valuable in work of atomic power stations - the electric power or by-products of its operation - produced stable and radioactive isotopes. Among the experts in reactors physics and techniques, an opinion prevails that nuclear reactors make profit exclusively from electric power production on atomic power stations. However some specialists consider this opinion rather far from a real state of things.

In the early nineties on the initiative of the American Council for Energy Awareness, USCEA, detailed research of influence of nuclear and radiating technologies (except for defensive) on national economy of the USA (Rabotnov, 1999) was undertaken. It consisted of two parts. In one part applications of radioactive materials in the industry, medicine and scientific researches were only considered, in the other - actually nuclear power, that is electric power production at atomic power stations. Results have surprised both authors of the report, and customers. First, the scale of figures. It appeared, that the total annual volume of the business connected with radioisotopes application in the United States made 257 billion dollars in 1991. Secondly, this sum appeared three and a half times more than the full cost of the nuclear electric power (73 billion dollars). Radioisotopes are used in eight various branches, 3.7 million persons of almost five hundred specialties are employed in this activity that makes about three percent of full employment in the USA. The total product of 330 billion dollars means that the nuclear-power and radiation complex of the USA looks as the eleventh on size a world industrial state.

Actually nuclear reactors are "generators" of a large amount of artificial radioisotopes, basic of which are fission products and actinium series. It is assumed to consider as fission products not only the radioisotopes got directly as a result of heavy nuclei fissions, but also the radioisotopes formed as a result of radioactive transformations and nuclear reactions of type (n, γ), (n, 2n), (n, p), etc. on radioactive and stable nuclei of fission products.

The chapter presents low-waste technologies of medical fragment radioisotopes production. Schemes of fragment radioisotopes production in reactors with fuel in the form of uranium salts solution or with molten-salt fuel on the basis of metal fluorides LiF, BeF_2, UF_4 are considered. The aggregate state of liquid fuel opens the possibility of selective extraction of

fragment radioisotopes $^{99}Mo/^{99m}Tc$, ^{89}Sr, $^{131,133}I$, ^{133}Xe, etc. from fuel not involving both ^{235}U and the basic group of fragment elements. Radiochemical processing of the irradiated uranium typical for solid fuel systems is as a result excluded, that finally should lead to decrease in an exit of a highly active waste and reduction of the fission materials flow in technological process.

2. Application of fragment radioisotopes in medicine

One of the most scale consumers of radioisotopes is nuclear medicine where they are applied in diagnostics and therapy of heavy diseases. Production of medical radioisotopes has turned to an important branch of the industry sharing more than 50 % of annual radioisotopes production all over the world. Today more than 160 radioisotopes of 80 chemical elements are produced by means of nuclear reactors and charged particles accelerators. The nomenclature of preparations labeled with radioisotopes permanently extends, new diagnostic instruments are developed. High efficiency of radioisotopes use in medicine is confirmed by long-term clinical practice. Radioisotope methods are used in oncology, cardiology, hematology, urology, nephrology, etc.

The simplest and cheapest way of scale radioisotopes production is based on fission reaction (n,f). The basic medicine radioisotopes formed as a result of uranium-235 fission by neutrons are ^{137}Cs, ^{131}I, ^{133}Xe, ^{90}Sr and ^{99}Mo.

^{99}Mo is one of the most demanded radioisotopes in nuclear medicine. It is used in $^{99}Mo/^{99m}Tc$ generators widely applied in the world at early diagnostics of oncologic, cardiovascular and a number of other diseases. More than 80 % of radiodiagnostic procedures in the world are carried out by preparations labeled with ^{99m}Tc (IAEA, 1999). From the IAEA data, approximately 15 millions of diagnostic procedures were carried out with ^{99m}Tc annually in the world ten years ago, from them 7 millions in Europe and 8 millions in the USA. The total world consumption of $^{99}Mo/^{99m}Tc$ for that period was estimated at a level of 6 000 Ci a week (6 day pre-calibration). The prediction of ^{99}Mo demand till 2006 made in works (Ball, 1999; Ball, Nordyke & Brown, 2002) is illustrated in the diagram in figure 1. About 30 millions of diagnostic procedures are carried out with ^{99m}Tc in the world annually today (Hansell, 2008). The world consumption of $^{99}Mo/^{99m}Tc$ has exceeded 12 000 Ci a week, and annual volume of the world market of ^{99m}Tc consumption is estimated as US$ 3.7 billion (including cost of medical services).

The basic producers of ^{99}Mo are Canada, EU and South Africa, which supply about 95 % of this radioisotope in the world. The technology of ^{99}Mo production is based on irradiation in research reactors and subsequent radiochemical processing of the targets from highly-enriched uranium. Table 1 provides further information on the major ^{99}Mo producing reactors (Westmacott, 2010).

Recent series of the not planned stops of the reactors producing ^{99}Mo, mainly due to significant age of these reactors, have shown to the world imperfection of the existing system of supplying this radioisotope and have caused growing concern for the future of nuclear medicine. The faults in ^{99}Mo production had negative effect on treatment of oncologic patients in Europe and Northern America. For example, during these stops, supply of $^{99}Mo/^{99m}Tc$ generators in some British hospitals has reduced to 30 % from normal. The doctors had to choose, who from the patients more than others requires the procedures with ^{99m}Tc.

Reactor name	Location	Annual operating days	Normal production per week[1]	Weekly % of world demand	Fuel/ Targets[2]	Date of first commis-sioning
BR-2	Belgium	140	5 200[3]	25-65	HEU/HEU	1961
HFR	Netherlands	300	4 680	35-70	LEU/HEU	1961
LVR-15[4]	Czech Rep.	-	>600	-	HEU[5]/HEU	1957
MARIA[4]	Poland	-	700-1 500	-	HEU/HEU	1974
NRU	Canada	300	4 680	35-70	LEU/HEU	1957
OPAL	Australia	290	1 000-1 500	-[6]	LEU/LEU	2007
OSIRIS	France	180	1 200	10-20	LEU/HEU	1966
SAFARI-1	South Africa	305	2 500	10-30	LEU/HEU[7]	1965
RA-3	Argentina	230	200	<2	LEU/LEU	1967

1. Six-day curies end of processing. In some cases, the maximum production can be substantially higher that the values listed here for normal production.
2. Fuel elements and targets are classified as either LEU, containing less than 20% of ^{235}U, or HEU, which contains greater than 20% ^{235}U (in some cases greater than 95%).
3. Does not account for increase in capacity since April 2010 with the installation of additional irradiation capacity. This increases BR-2 available capacity to approximately 7 800 six-day curies EOP: however it is not yet clear what "normal" production will be at the facility.
4. These reactors started production in 2010, - so some data is not available yet.
5. The LVR-15 reactor uses fuel elements that are enriched to 36% ^{235}U.
6. The OPAL reactor started production in 2007 for domestic use but has not yet exported significant amounts.
7. SAFARl-1 is in the process of converting to using LEU targets and expects to have completed conversion in 2010.

Table 1. Major ^{99}Mo producing reactors

An attempt to solve the problem of ^{99}Mo production by building two specialized reactors MAPLE in Canada was not successful - May 16, 2008 these reactors were finally stopped because of the made design mistakes. Against steady ageing of the research reactors which operation time sometimes exceeds 50 years (reactor NRU), this problem becomes sharper.

Besides, the isotope crisis has drawn attention to one more acute problem – production technology. The fission fragment method is a basis of current ^{99}Mo production. A highly-enriched uranium target is irradiated in a reactor, and then processed by one of the known radiochemical ways. ^{99}Mo, which specific activity reaches several tens thousand Ci per gram of molybdenum is separated from fission products (Gerasimov, Kiselev & Lantcov, 1989). More than 95 % of ^{99}Mo are produced using highly-enriched uranium (HEU), ~90 % of ^{235}U. Commercial producers of ^{99}Mo spend about 50 kg of HEU annually.

The weakest feature of this way of ^{99}Mo production is extremely inefficient use of uranium. Only ≈0.4 % of ^{235}U is used for production of ^{99}Mo, and the other part is directed in waste. Moreover, at fission of uranium a wide spectrum of fragments is formed, which total activity at the moment of irradiation end exceeds activity of ^{99}Mo by two orders of value. That results in necessity of contamination of plenty radioactive waste, including long-lived ones.

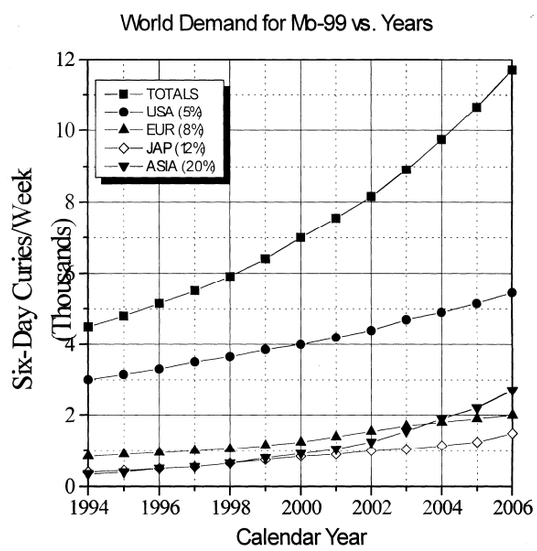


Fig. 1. Prediction of growth of demand for ^{99}Mo in the world (Ball, 1999; Ball, Nordyke & Brown, 2002)

In Fig. 2 the basic stages of the existing ^{99}Mo production technology are shown - from manufacturing of a uranium target to delivering the final product to the consumer.

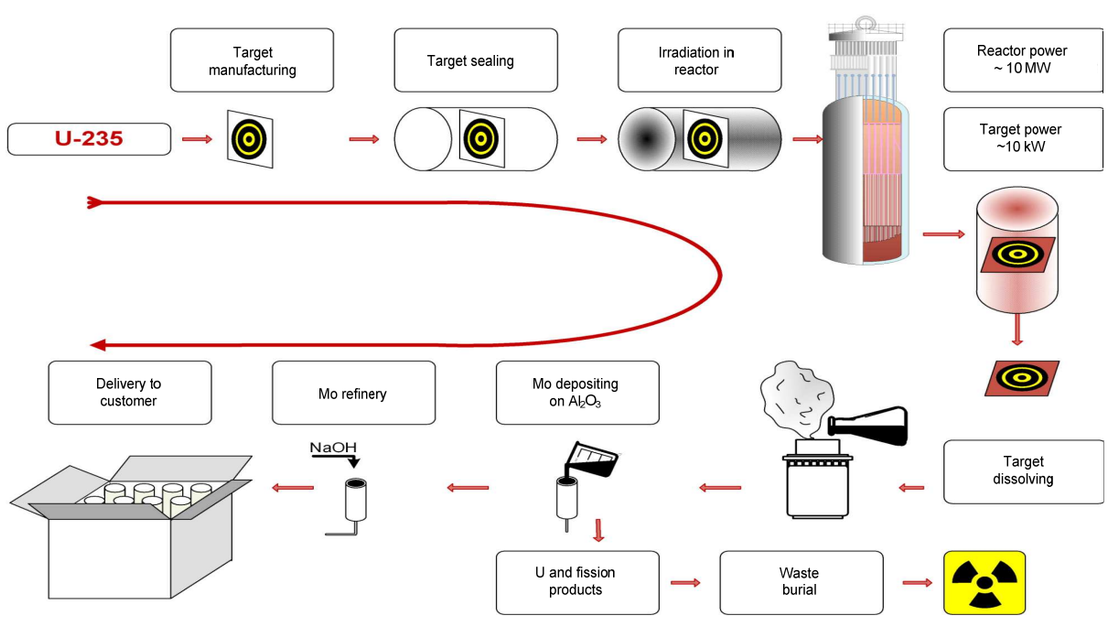


Fig. 2. Basic stages of the existing ^{99}Mo production technology

The third problem of the traditional ^{99}Mo production is that its target waste can be easily transformed by known chemical methods to metal HEU - material used in manufacture of nuclear military loads. Orientation of modern ^{99}Mo production on the use of highly-enriched uranium, against gradual deducing of HEU from a civil turn according to the IAEA "non-proliferation" concept, creates additional risks for ^{99}Mo consumers. Thereupon expected rapid development of the nuclear medicine using ^{235}U fission products can lead the next years to serious problems with ensuring the efficiency of the international monitoring system for non-proliferation of nuclear materials.

In the international community there is a consensus about necessity to limit or even to bring to naught HEU turn in the civil sphere. Programs, in particular, the program RERTR (Reduced Enrichment for Research and Test Reactors) on reduction of HEU turn in peace sectors of economy are accepted. The research reactors used for production of ^{99}Mo will be gradually converted to low-enriched uranium fuel (LEU) and LEU-targets. Risks from use of uranium of high enrichment will start to constrain sooner or later, and then can even stop this production completely.

The world crisis of ^{99}Mo production gives an exclusive chance for realization of alternative methods of this radioisotope production, free from disadvantages of the modern technologies and ensuring large-scale low-waste production of ^{99}Mo based on the use of low-enriched uranium.

What could be alternative? The choice, in effect, is limited: building of a new multi-purpose research reactor, development of accelerating methods or building of inexpensive and simple in operation solution reactors. Taking into account inertia of the licensing system and time for designing and building the facilities, the choice needs to be made the next years.

It would seem, the most real alternative to HEU for today is conversion of a research reactor core for work with LEU-fuel and introduction of new types of targets of low-enriched uranium. However the "target" technology characterized by the lowest efficiency of use of uranium, most likely will not give competitive advantages as the use of LEU will inevitably lead to involving for manufacture of a considerable amount of irradiated uranium. It will result in additional loadings on radiochemical processing and in considerable amount of radioactive waste that, obviously, will raise production cost price. The most research reactors now producing the basic amount of ^{99}Mo are depreciated and for their maintenance in operating and safe condition more and more considerable efforts and expenses are required. The next years these reactors will demand capital reconstruction or will be deduced from operation. Other alternative technologies based on neutron activation of molybdenum isotope ^{98}Mo or on uranium photo-fission at accelerators, are not effective for large-scale production and can be applied only for satisfying regional needs.

Renunciation of the traditional technology of uranium targets irradiation and transition to technologies based on specific physical-chemical properties of liquid nuclear fuel, ensuring an opportunity to extract ^{99}Mo directly from the reactor fuel leaving uranium and fission fragments in it, allow to exclude a problem of highly-enriched uranium contamination and to simplify the problem of handling the radiochemical processing waste radically. Water solutions of uranium salts or melts of fluoride salts can be used as liquid nuclear fuel.

The aggregate state of liquid fuel opens the possibility of selective extraction of fragment radioisotopes ^{99}Mo/^{99m}Tc, ^{89}Sr, ^{131,133}I, ^{133}Xe, etc. from fuel not involving both ^{235}U and the basic group of fragment elements. It will lead to sharp decrease in the flow of nuclear

materials in the technological process and in the yield of highly active waste. Estimations show, for example, that a solution reactor of 200 kW power is capable to produce more than 1000 Ci/w of ^{99}Mo with calibration for 6-days from the moment of delivery.

The technology based on use of homogeneous liquid nuclear fuel possesses a number of obvious advantages in comparison with a "target" method:

- All fuel of such a reactor is a "target";
- There is no necessity for creation of special facility for manufacturing and radiochemical processing of targets;
- ^{99}Mo is selectively taken from solution fuel by sorption on special filters or spontaneously leaves the molten salt fuel in the form of flying fluorides;
- Uranium and the basic highly active fission fragments remain in fuel;
- Reactors with homogeneous fuel possess immanent nuclear safety (negative thermal reactivity factor);
- Research mini-reactor "Argus" with solution fuel in the form of UO_2SO_4 has operated in Russia for about 30 years;
- Research reactor MSRE of power 8 MW with molten salt fluoride fuel $LiF-BeF_2-UF_4$ successfully operated in the USA within four years, when the effect of spontaneous exit of a part of fission fragments in the gas phase was observed (including ^{99}Mo);
- There are no problems with reactor operation on LEU.

Now development and creation of a specialized nuclear-chemical complex for commercial production of fragment radioisotopes ^{99}Mo, ^{89}Sr, ^{133}Xe, ^{131}I, ^{133}I, etc. for satisfaction of growing requirements of nuclear medicine look actual.

3. Production of fragment radioisotopes with help of molten salt fluoride nuclear fuel

Transition from the traditional technology of uranium targets irradiation to the technology based on an unique effect of spontaneous exit of fragment ^{99}Mo from molten salt fluoride fuel to the gas phase (Grimes, 1970), observed in the late sixties in the experimental reactor MSRE (ORNL) allows to simplify a problem of waste disposal at production of fragment radioisotopes considerably. According to the results of measurements, more than 50 % of all molybdenum formed at fission of uranium leaves melts. The most of other fragment elements, such as Zr, Ba, Sr, Cs, Br, I, rare-earth elements and all uranium having steady well soluble in fuel salt compounds remain in melts.

The idea of use of molten salt nuclear fuel in the reactor technology was proposed at the beginning of the 50-ties at creation of small power autonomous energy sources. A distinctive feature of this kind of fuel is the possibility of continuous correction of the chemical structure and control of nuclear-physical, chemical and thermal processes at its work in the nuclear reactor.

The molten salt reactor concept was realized at creation of experimental reactors ARE (Aircraft Reactor Experiment) by power of 2.5 MW and MSRE (Molten-Salt Reactor Experiment) of 7.3 MW. The first experimental molten salt reactor, ARE, constructed in the USA in 1954, worked with salt composition $NaF-ZrF_4-UF_4$ at temperature 860°C (Bettis e.a., 1957). The basic task of the experiment was confirmation of serviceability and stability of the reactor with circulating fuel. MSRE operated in Oak Ridge National Laboratory (ORNL)

within four years in the late 60s (Haubenreich & Engel, 1970) with fuel $^{7}LiF-BeF_2-ZrF_4-UF_4$ at temperature 650°C. The purposes of MSRE construction consisted in working out the molten salt nuclear fuel technology, checking the serviceability of some structure units, study of neutronics characteristics of this reactor type. Corrosion steady nickel alloy Hastelloy-N was developed, some questions of processing and cleaning the salt were solved during reactor operation, and its nuclear safety was shown.

Significant researches of molten salt coolants were performed in the Soviet Union (Novikov et al., 1990). The structural materials on the basis of nickel, corrosion stable in fluorides are created, the pump, heat exchanging devices of various purposes etc are developed and checked in conditions of stand tests. Now these researches proceed, mainly within the framework of the international project "Generation-IV" on creation of new generation nuclear reactors.

3.1 Behavior of fission fragments in molten salt fuel

In MSRE, the basic group of fragment elements, such as Zr, Ba, Sr, Cs, Br, I and rare earth elements, having well soluble in salt compounds remained in fuel. Insoluble compounds deposited on the surface of structural materials of the reactor facility (graphite, Hastelloy-N). Krypton and xenon, having low solubility in salt, spontaneously or under bubbling of fuel by inert gas left the salt melt. Moreover it was found, that a small group of elements, so-called "noble" metals Mo, Nb, Te, Ru in the form of volatile fluorides or aerosols entered gas covering free surfaces of salt. About half of these elements deposited on surfaces of reactor structural materials and less than 1% remained in fuel salt.

In a series of ORNL 1966-1970 reports the results of numerous measurements of fuel structure, researches of structural material samples and gas tests from MSRE contour are published. An analysis of these experimental data has allowed us to reveal certain laws of distribution of some fragment elements in the reactor facility, to put forward a hypothesis of the mechanism of "noble" metals exit to gas volume above the salt melt and on this basis to propose a new way of fragment ^{99}Mo production.

In particular, in (Rosenthal et al., 1969) (see tab. 2) the balance of radionuclides in reactor MSRE is shown. Significant fraction of ^{99}Mo, ^{132}Te, ^{103}Ru entered gas covering free surfaces of salt. The detection of significant amount of ^{89}Sr in the gas volume above the salt surface is connected with its predecessor ^{89}Kr entering into gas.

Nuclide	Contents (% from calculated value)				
	Fuel	Graphite	Hastelloy-N	Evacuated gases	Total yield
^{99}Mo	0.17	9.0	28	50	87
^{132}Te	0.47	5.1	14	74	94
^{129}Te	0.40	5.6	17	31	54
^{103}Ru	0.033	3.5	3.7	49	56
^{95}Nb	0.001	41	18	11	70
^{89}Sr	83	17	8.5	0.11	109

Table 2. Distribution of fission products in reactor MSRE

The assumption was made, that these fission fragments arising in extremely electron-scarce states, in the process of their thermalization catch electrons, passing a number of unstable, but volatile valent states (Kirslis & Blankenship, 1967). If the reactor structural materials are rather inert, and the fuel does not contain significant amount of other absorbers of radiolytic fluoride, some part of Mo, Te and Ru will oxide up to maximal fluorides and pass to the gas phase.

In work (Rosenthal et al., 1968), 50 g salt samples were located in hermetic reactionary volumes connected to a gas contour, from which helium or mix of helium with hydrogen could be made to fuel melt. For melting of fuel the reactionary volume was heated up to 600°C. The gas moved in the tube at the flow rate 10 - 15 $cm^3 \cdot min^{-1}$, allowing both blowing the salt melt mirror and bubbling the fuel. After passage of reactionary volume the gas was evacuated through a system of filters including few sections of fission fragments catching – metal-ceramic filter, sorbent on the basis of NaF and chemical sorbent. After reactor operation of 32 000 MW×h the filters were removed, each section was separately dissolved in acid and the solutions were analyzed on the contents of fission fragments and ^{235}U. Some measurement results of the salt radioisotope structure are given in table 3.

Calculated by us total activities of radioisotopes collected in all elements of filtration system, normalized to initial activity in salt test are shown in the same table. The cumulative yields of radioisotopes at ^{235}U fission by thermal neutrons, their half-decay periods, chemical formulas of maximal fluorides of elements and boiling temperatures of these chemical compounds are also shown here.

Parameter	^{103}Ru	^{132}Te	^{95}Nb	^{99}Mo	^{95}Zr	^{144}Ce	^{140}Ba
$T_{1/2}$	39.36 days	78.2 h	34.97 days	66.0 h	64.02 days	284.4 days	12.75 days
Cumulative yield of nuclide at fission of ^{235}U,%	3.04	4.28	6.5	6.15	6.29	5.47	6.08
Cross section of the reaction (n,γ), barn	7.71		<7	1.73	0.49	2.6	1.6
Nuclide specific activity in melt, $decay \cdot min^{-1} \cdot g^{-1}$	$7.5 \cdot 10^6$	$1.7 \cdot 10^8$	10^9	$1.2 \cdot 10^9$	$1.2 \cdot 10^{11}$	$7.3 \cdot 10^{10}$	$1.6 \cdot 10^{11}$
Nuclide concentration in melt, g^{-1}	$6.1 \cdot 10^{11}$	$1.2 \cdot 10^{12}$	$7.3 \cdot 10^{13}$	$6.8 \cdot 10^{12}$	$1.6 \cdot 10^{16}$	$4.3 \cdot 10^{16}$	$4.2 \cdot 10^{15}$
Ratio of total nuclide activity in filters to its activity in 1 g of initial melt, rel. un.	5.6	0.7	≈0.02	1.1	$1.1 \cdot 10^{-4}$	$2.0 \cdot 10^{-5}$	$1.2 \cdot 10^{-5}$
Maximal fluorides of nuclides	RuF_5	TeF_6	NbF_5	MoF_6	ZrF_4	CeF_3	BaF_2
Boiling temperature of maximal fluorides at saturated vapor pressure 760 mm, °C	272	-38.9	233	36.2	903	2327	2260

Table 3. Radioisotope structure of MSRE fuel ("blowing" of a salt mirror with helium)

First of all one may indicate a wide scatter of fission fragments concentrations in the initial salt sample and a possibility to divide them into two groups by this parameter. The concentrations of ^{99}Mo, ^{132}Te, ^{95}Nb, ^{103}Ru are lower by two - four orders of value than the concentrations of ^{89}Sr, ^{141,144}Ce, ^{95}Zr, ^{140}Ba, though cumulative yields of these fragments differ not more than twice. It is impossible to explain the so large difference by burning up of fragments in the reactor, at the given cross sections. The most probable reason is connected to individual behavior of chemical compounds of these elements in molten salt fuel.

In table 3 there are two radioisotopes ^{95}Nb and ^{95}Zr presenting the generically connected pair in the decay ^{95}Kr→^{95}Rb→^{95}Sr→^{95}Y→^{95}Zr→^{95}Nb→^{95}Mo. At long irradiation of fuel, these radioisotopes should be in balance, i.e. their activities should be equal. The difference of ^{95}Nb and ^{95}Zr activities by more than two orders of value means, that ^{95}Nb mainly leaves melt, and ^{95}Zr remains in fuel.

The observable difference in concentrations of the two groups of radioisotopes specified in table 3 can be explained by exit of ^{99}Mo, ^{132}Te, ^{95}Nb, ^{103}Ru to the gas phase in form of volatile fluorides, boiling temperature of which is much lower than the temperature of salt melt. Thus refractory fluorides ^{95}Zr, ^{141,144}Ce, ^{140}Ba mainly remain in salt.

Molybdenum has a few stable fluorides, which can be formed under γ-irradiation of the nuclear reactor - MoF_3, MoF_4, MoF_5, MoF_6. At the fuel melt temperature ≈600^{0}C, significant fractions of tetra- and pentafluoride of molybdenum MoF_4, MoF_5 and also molybdenum hexafluoride MoF_6 (boiling temperature of 36.2^{0}C (Nikolsky, 1998) are in the gas phase. In case these compounds arise near to borders with the gas phase, they can leave the melt.

In work (Kirslis & Blankenship, 1969), the degree of fission fragments penetration in graphite of MSRE core was investigated. As well as for the previous experiment, we were interested by penetration into graphite of generically connected pair ^{95}Zr→^{95}Nb. It follows from experimental data that ^{95}Nb concentration in graphite has appeared approximately by three orders of value higher, than ^{95}Zr. Their distribution on depth of the graphite sample is shown in figure 3.

It is known, that the radioactive atoms formed as a result of nuclear transformations, partially appear in chemical compound, at which they were formed, as so-called "parent" compound (Nesmeyanov, 1978). Taking into account that zirconium has sole stable fluoride - ZrF_4, we estimated the possibility of preservation of the initial chemical form of a molecule as tetra fluoride after decay of a parent nucleus ^{95}Zr and formation on its place of ^{95}Nb nucleus.

The energy of the exited molecule, which can be spent on break of the chemical binding, is equal:

$$E_B = E\frac{M_R}{M + M_R}$$

where E is recoil energy, M is mass of the recoil atom, M_R - mass of the rest of molecule. 0.44E will be spent on break of the chemical binding for a molecule ZrF_4, i.e. about a half of ^{95}Zr recoil energy.

At β-decay of ^{95}Zr basically two groups of β-particles with average energies of 121 and 110 keV (Burrows, 1993) are formed. Recoil energy of ^{95}Nb for β-particles with energy 121 keV makes 0.77 eV and for 110 keV – 0.69 eV. Besides at decay of ^{95}Zr the γ-quanta with energy about 750 keV are emitted. Thus the recoil energy of ^{95}Nb will make 3.2 eV.

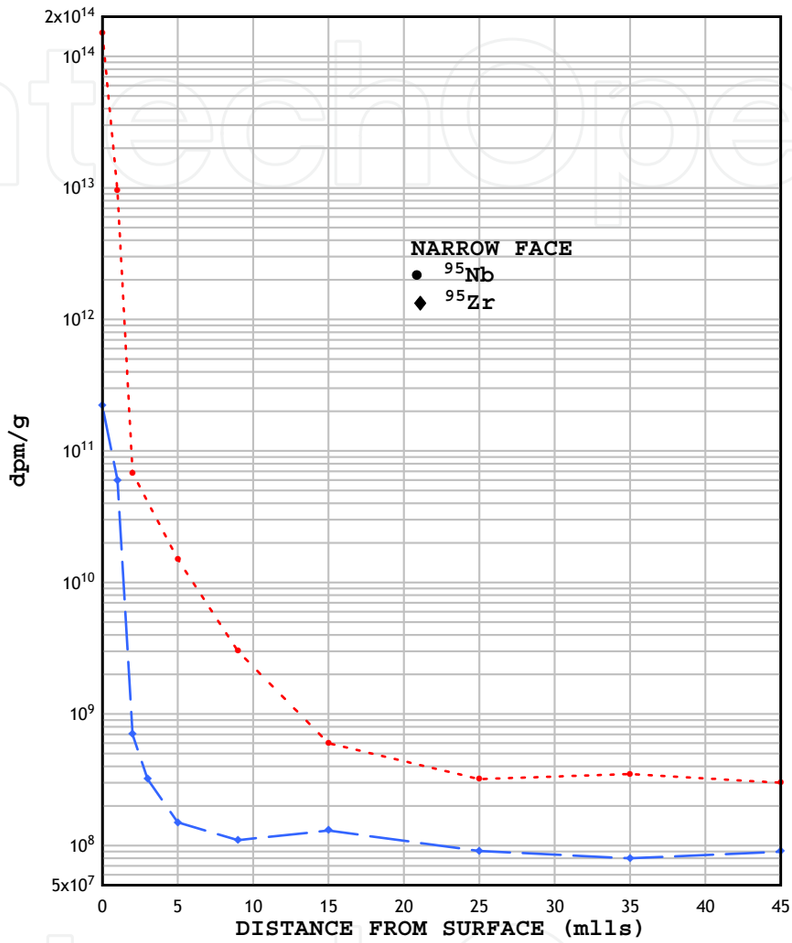


Fig. 3. Distribution of ^{95}Zr and ^{95}Nb activities on depth of graphite sample contacted with salt melt in the reactor MSRE core (Kirslis & Blankenship, 1969)

As the binding energy of atoms in molecules is 2-5 eV (Nesmeyanov, 1978), in most cases of ^{95}Zr decay in a molecule $^{95}ZrF_4$ the formed new atom ^{95}Nb will be kept in the chemical form of the parent compound, i.e. as a molecule $^{95}NbF_4$. The further fate of niobium tetra fluoride $^{95}NbF_4$ is determined by thermodynamics and radiation-chemical processes in molten salt fuel. In the reactor γ-field formation of niobium pent fluoride $^{95}NbF_5$ is possible, which at temperature ≈600°C will be in gas form ($T_{boiling}$ = 234°C), and at presence of the phase border "liquid – gas" it will leave fuel melt, penetrating, including, into open gas pores of graphite, which is in contact with molten salt fuel. The boiling temperature of zirconium fluoride is $T_{boiling}$ = 912°C, that is much higher than the fuel temperature. Therefore $^{95}ZrF_4$ does not enter gas bubbles and, accordingly, does not penetrate into pores of graphite that explains significant difference of ^{95}Zr and ^{95}Nb contents in graphite.

In our opinion, the observed experimental results can be explained by formation and exit of easily volatile fluorides of noble metals Mo, Nb, Te from molten salt fuel to the gas phase. In detail the mechanism of ^{99}Mo exit from molten salt fuel is considered in (Zagryadsky et al., 2008; Menshikov & Chuvilin, 2008).

The proposed radiochemical model of "noble" metals exit from fluoride fuel in a gas phase assumes, that chemical processes in fuel $LiF\text{-}BeF_2\text{-}UF_4$ vary sharply in the field of an ionizing radiation and are determined by a number of active particles - a complex ion BeF_4^2, radicals BeF_4^-, $Be_2F_8^{3-}$ and a number of other ions. The fragment molybdenum formed in molten-salt fuel at uranium fission exists in the melt mainly in the form of complex ions, such, for example, as MoF_5^{2-}. Reacting with ions $Be_2F_8^{3-}$ within characteristic time $\tau \approx 3 \cdot 10^{-4}$ s, molybdenum ions can form at temperature ≈ 600°C volatile fluorides MoF_4, MoF_5, MoF_6 which, being near to borders with a gas phase will leave the melt. Due to presence in a gas phase of radiolitic fluorine, molecules $^{99}MoF_4$, $^{99}MoF_5$ are oxidized to a higher fluoride $^{99}MoF_6$ which is delivered by helium flow into catching system along transport communications.

3.2 Advantages and possible variants of realization of fragment ^{99}Mo production on the basis of molten salt nuclear fuel

Basing on the results of experiments performed within the framework of the research program at the reactor MSRE and the hypothesis of the mechanism of ^{99}Mo exit from molten salt fuel, we proposed a new way of fragment ^{99}Mo production, using a spontaneous (or stimulated by bubbling of fuel with inert gas) exit of fragment ^{99}Mo from molten salt fuel to the gas phase. For the first time this idea was mentioned in (Chuvilin & Zagryadsky, 1998).

The aggregate condition of fuel on the basis of Li, Be and U fluorides allows to take ^{99}Mo, ^{89}Sr, ^{133}Xe from salt melt selectively, leaving ^{235}U, and the basic group of fragment elements. As a result inherent for solid fuel systems radiochemical processing of the irradiated uranium is excluded, what at the end should result in reduction of highly active waste and of the flow of fission materials in the technological process.

Principally the process of ^{99}Mo production in a research reactor can be organized as follows (Chuvilin & Zagryadskiy, 1997). One of fuel assemblies of the reactor core is replaced with a loop facility filled with molten salt fuel, attached to a gas contour. As a result of ^{235}U fission, in the salt melt ^{99}Mo is generated, which moves to the salt - gas border and passes to the gas phase above the salt melt surface in form of fluorides and aerosols. ^{99}Mo removal can be intensified by bubbling of helium or argon through salt.

The collected in free volume of the loop system gas-aerosol fraction is removed by purge of the cavity with inert gas. The gas - carrier containing aerosols and volatile fluorides of molybdenum enters the catching system, which can be realized as a set of filters, chemical sorbents or freezing traps. Cleaned from fragment elements, inert gas recirculated in the loop system. For ^{99}Mo removal, the filters periodically are directed to radiochemical processing. Accompanying molybdenum fragment elements deposited in the catching system are utilized after an appropriate procedure of extraction. The basic scheme of fragment ^{99}Mo production in the molten salt reactor is shown in Fig. 4.

The possible design solutions of the loop system will be determined by the technology of ^{99}Mo removal from molten salt fuel. At the first stage of its introduction it is expedient to use elementary devices that will allow checking up the basic technical decisions, investigating radionuclide purity of the product, estimating economic parameters of the proposed way of ^{99}Mo production.

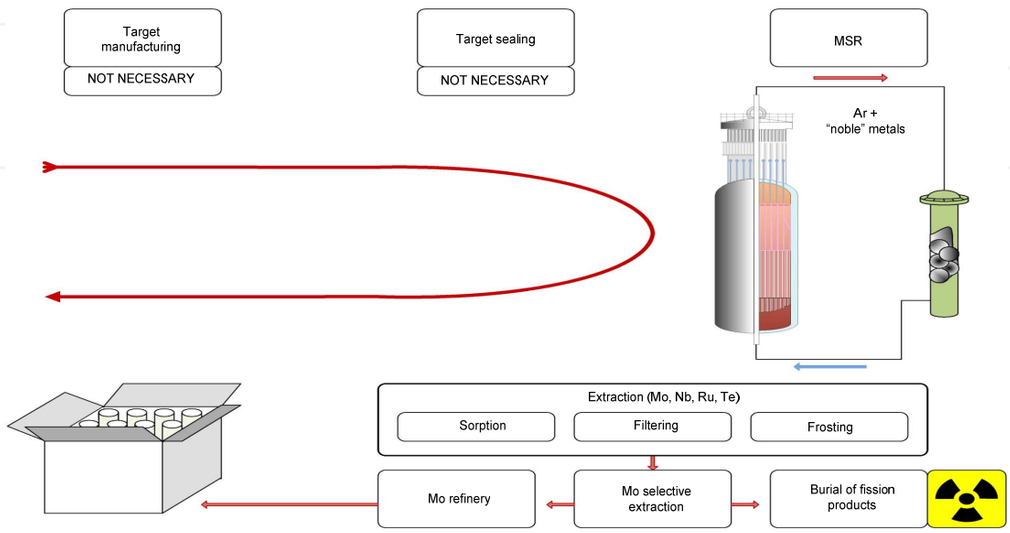


Fig. 4. Scheme of fragment ^{99}Mo production in a molten salt reactor

3.3 Experimental ampoule loop system "RAMUS"

For demonstration of physical practicability of the proposed new technology of ^{99}Mo production, in the research reactor IR-8 of the NRC "Kurchatov Institute" experimental ampoule loop system "RAMUS" with molten salt fluoride fuel is created, where the process of ^{99}Mo production will be realized according to (Chuvilin & Zagryadskiy, 1997). It is planned to determine the efficiency of ^{99}Mo extraction from fuel, possibility of transporting molybdenum fluoride compounds from the reactor core to the accumulating zone for extraction from the gas – carrier flow, and other parameters. Besides that it is planned to produce as well two other accompanying radioisotopes of medical purpose: ^{89}Sr, ^{133}Xe in the loop system "RAMUS".

The loop system "RAMUS" will be placed in the beryllium reflector of IR-8 having neutron flux $\approx 10^{13}$ n·cm^{-2}·s^{-1}. The loop system has the following characteristics:

Molar structure of fuel composition, %	$66LiF\text{-}33.9BeF_2\text{-}0.15UF_4$
Enrichment by isotope ^{235}U, %	90
Volume of salt, cm^3	450
Power of reactor IR-8, MW	8
Heat generation in fuel salt, kW	3.2
Range of salt working temperatures, ^{o}C	613 - 680

The basic scheme of the ampoule part of the loop system "RAMUS" is shown in Fig. 5. The radioisotopes ^{99}Mo, ^{89}Sr, ^{133}Xe will be evacuated from molten salt fuel by bubbling of melt with helium. After passage of gas-carrier along the communications and filters, in which

decay and depositing of short-lived fission products take place, the target radioisotopes are caught in depositing devices, from which they are extracted subsequently.

During design of the loop system "RAMUS" some technical decisions have required to be experimentally substantiated. In particular, the data on formation and carrying over of aerosol particles in the process of molten salt bubbling by inert gases and spontaneous thermocondensation of salt component vapors are practically absent in the literature. At bubbling the large interphase surface on "liquid-gas" border is created that promotes an intensification of mass-exchange processes, and also a more complete chemical interaction of gases with liquids.

The experiments on formation and carrying over of aerosol particles by helium in the process of molten salt fuel bubbling were performed to make more precise the parameters of the unit "RAMUS". The regime of the vertical bubble flow in molten salt $66LiF\text{-}34BeF_2$ was investigated at temperature 530-630°C. The experiments are carried out on a model installation reproducing the dimensions of the ampoule "RAMUS". Weight and height of the salt melt made 900 g and 450 mm, respectively. For creation of the gas bubble flow, a tube was immersed in the salt melt, on which bottom end cylindrical sprayers with face or lateral holes were installed. 6 types of sprayers were tested. The stabilized helium flow rate varied in the range of 1.5-4.5 $cm^3\,s^{-1}$.

The rate of carrying over of aerosol particles was determined with help of the photometer FAN-A. Besides that the aerosols were taken out on filters and then the deposit was weighted and analyzed by ICP-AES and DTA-TDA methods.

Foam was observed on the liquid surface in the experiments with bubbling of the salt melt by helium. It was possible to get the minimal height of the foam layer – 15-18 mm with the sprayer of three holes ∅=0.5 mm, L = 0.5 mm located uniformly on the circle. The mass concentration of aerosol particles did not exceed 100 $mg{\cdot}m^{-3}$, and the rate of their carrying over - 0.1 $\mu g{\cdot}s^{-1}$. The size distribution of aerosol particles has appeared bimodal. The first mode (superfine aerosol, average particle diameter ≈0.05 microns) is caused by spontaneous thermocondensation of salt components vapors at their cooling from 630°C to 60°C. 90÷95 percents of aerosol particles mass consist of beryllium difluoride. That is because BeF_2 vapors pressure at temperature 630^{0}C more than 50 times exceeds LiF vapors pressure.

The second mode of submicron aerosol is caused by collapsing of bubbles on the boundary surface "foam – gas" and corresponding dispersion of liquid. The diameter of particles taken out from an ampoule does not exceed 4 microns, at average diameter 0.5 microns. Their chemical structure coincides with the structure of the salt melt. It means that the carrying over of salt components in form of aerosols at bubbling of melt is determined by dispersion of liquid at collapsing of bubbles on the surface of two phase boundary, and the contribution of BeF_2 superfine particles generated at vapors spontaneous thermocondensation makes less than 5% of weight.

The received experimental results have allowed making a conclusion that for the planned term of the loop system "RAMUS" operation in the reactor IR-8 channel the convective carrying over of salt components in form of vapors and aerosols will be negligible small and will not influence gas-dynamic characteristics of communications and the gas flow filtration system of the loop system.

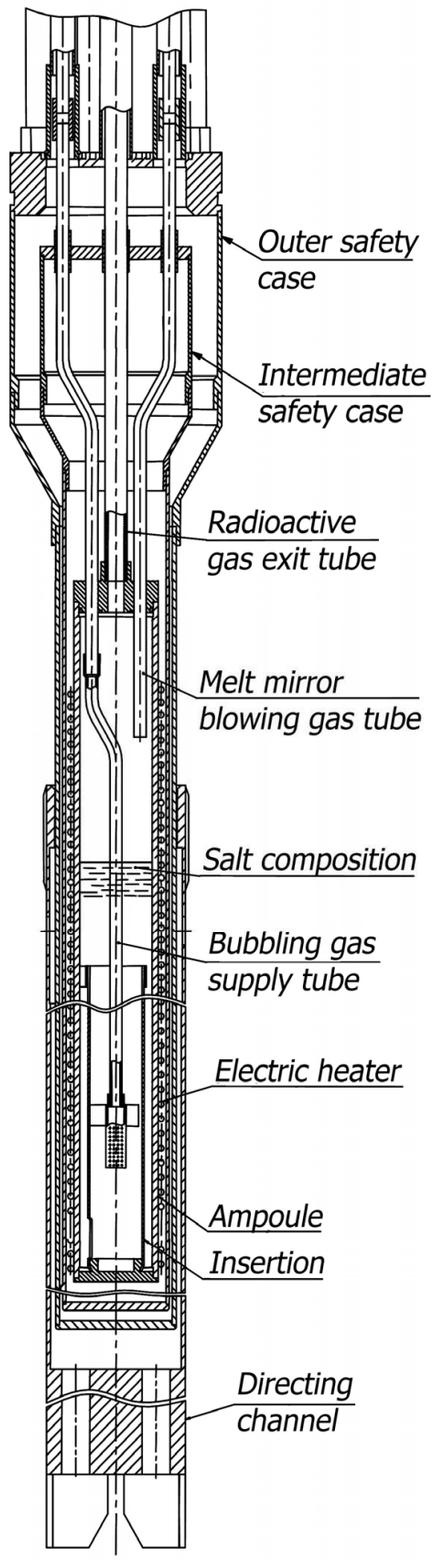


Fig. 5. General scheme of the ampoule part of the loop system "RAMUS"

In other series of the experiments, the possibility of MoF_6 loss at its transport along technological communications of the loop system was studied depending on temperature. Sorption of MoF_6 from the gas mixture flow (He + MoF_6) with the contents of molybdenum hexafluoride in the range 10^{12} - 10^{13} cm^{-3} on granulated NaF was also investigated. The content of MoF_6 in the gas mixture corresponds to the calculated concentration of fragment molybdenum in the loop system "RAMUS". The infra-red Fourier-spectrometer FSM-1202 is used for MoF_6 measurement in the gas mixture. The experiments are carried out for the middle and near infra-red areas of the spectrum, with use of multirunning flask with the optical way 4.8 m long, which has allowed measuring MoF_6 concentration down to 10^{12} cm^{-3}. The measurements have shown that deep extraction of MoF_6 from the gas flow takes place even at high flow rates of the mixture up to 10 $cm^3 \cdot s^{-1}$.

The process of MoF_6 desorption from the sorbent surface becomes noticeable at temperature 100^oC. However, as in reactor conditions the temperature at location of the column with sorbent will not exceed 60°C, the effect of MoF_6 desorption in the loop system "RAMUS" could be neglected. The measurements of MoF_6 losses during its transport along metal communications specially prepared in fluoride atmosphere have shown that they are within uncertainty limits of the spectrometer FSM-1202, and the loss of target product could be neglected.

Thus, the carried out pre-reactor experiments have allowed estimating a number of technological parameters of the loop system "RAMUS", choosing the optimal design of pneumatic sprayers and the regime of molten salt bubbling, predicting possible losses of MoF_6 at its transport along gas communications of the loop system.

4. Production of ^{99}Mo in solution reactors

The reactor is named "solution" if its fuel is in the form of water solution of uranyl-sulphate UO_2SO_4 or uranyl-nitrate $[UO_2(NO_3)_2]$ (Kolesov, 1999). At production of ^{99}Mo in the solution reactor, the necessity of target manufacturing and their radiochemical processing disappears. All the reactor fuel is a target. ^{99}Mo is taken from solution by sorption on special sorbent.

Russell Ball (companies Babcock & Wilcox and Ball Systems, the USA) has proposed a design of the liquid homogeneous reactor MIPR (Medical Isotope Production Reactor) for production of radioisotopes, in particular ^{99}Mo (Ball, 1999). This reactor can work on uranylnitrate solution in water with low-enriched ^{235}U (to 19 %) as fuel composition. At power of 200 kW it is capable to produce by estimations up to 2000 Ci of ^{99}Mo a day. The proposed design of a reactor and its characteristics allows receiving the permission to its placing even in the centre of big cities. The power of a solution reactor is more than 10 times less than of a traditional research reactor and demands hundreds times smaller amount of ^{235}U for production of equal amount of ^{99}Mo. According to estimations, building and physical start-up of a reactor will take 36 months with the total cost from 6 to 26 million US dollars.

4.1 Production of fragment ^{99}Mo on basis of solution reactor ARGUS

"Argus" is a thermal homogeneous solution reactor, operating at stationary power up to 20 kW in the NRC "Kurchatov Institute". Water solution of uranylsulphate by volume 21.1 l

with uranium concentration 81.3 g/l and enrichment by ^{235}U 90 % (Pavshouk & Chuvilin, 2005; Afanasev et al., 1986) is used as fuel.

The fuel solution is located in the case - a welded cylinder with a hemispherical bottom and a flat cover in which a cooling coil is located. Vertical "dry" channels are placed in the case: central and two symmetric peripheral channels in which control and safety rods are located. The reactor case is surrounded by a lateral and bottom face graphite reflector. Gaseous radiolysis products of fuel solution are regenerated by means of the system including a catalytic recombiner, which together with the case forms a tight system excluding leak of fission products to the environment. The recombination system is based on the principle of natural circulation of the gas mix along the contour. The solution type of the reactor provides conditions of the maximum safety which is guaranteed by the big negative reactivity effect and the optimum concentration of uranium in solution, leading to self-regulation of the reactor facility.

Since the middle of the 90s years the National Research Centre "Kurchatov Institute" has been working out a technology and creating a demonstration nuclear-technological complex of fragment ^{99}Mo production on the solution reactor "Argus". The aggregate state of the reactor "Argus" fuel allows ^{99}Mo extraction directly from fuel solution without influence of structure and characteristics of the reactor core. For that, after reactor operation on power solution is pumped through special sorbent on the basis of titanium oxide which provides the first stage of ^{99}Mo extraction from all mass of fission products. Thus uranium is not absorbed, and fuel solution completely comes back to the reactor case. All amount of the uranium which has undergone fission reaction, necessary for maintenance of reactor power, is spent for production of the target isotope.

This newest technology of ^{99}Mo production allows lowering the necessary reactor power 100 and more times as compared to the traditional technology that not only results in reduction of radioactive waste production, but also supposes reactor locating in the occupied areas and promotes solving the licensing problems. Thus efficiency of ^{235}U use comes practically to 100 % while in target technologies only 0.4 % of ^{235}U are spent for getting the product, and its other part after target processing, as a rule, goes to waste (Starkov, 1996). The necessity of uranium targets manufacturing is, besides, excluded. The developed technology allows to solve an actual problem of uranium enrichment reduction in production of ^{99}Mo and to eliminate public concern in connection with non-proliferation problem. The reactor which core is water uranylsulphate solution is used for accumulation of ^{235}U fission fragments. Thus the yield of fragment ^{99}Mo per fission of ^{235}U makes 6 %. Accumulation of ^{99}Mo in the core during the reactor operation on power of 20 kW is shown in Fig. 6.

For demonstrating the practicability of ^{99}Mo extraction technology directly from fuel solution, the demonstration experimental complex of the reactor "Argus" presented in Fig. 7 and including a reactor loop, transportation means and hot cells equipment for purifying ^{99}Mo from fission products has been designed and put into operation.

As a result of reactor experiments the possibility of ^{99}Mo extraction from reactor solution fuel has been shown for the first time, and also the following technological process has been developed. The reactor "Argus" operates at power within 5 days. After some endurance, the fuel solution is pumped through sorbent within 3-6 hours. For returning of the rests of fuel

solution into the reactor case the reactor loop together with the sorption column is washed out by the condensate which has been accumulated in a system of radiolysis products regeneration during power operating time of the reactor. Then the column is remotely placed in a transport container and goes to hot cells, where after sorbent washing by acid solutions radionuclide ^{99}Mo is desorbed with alkali. For getting demanded radionuclide purity two stages of purification with help of refinery columns filled with the same sorbent of type "Thermoxid" are taken.

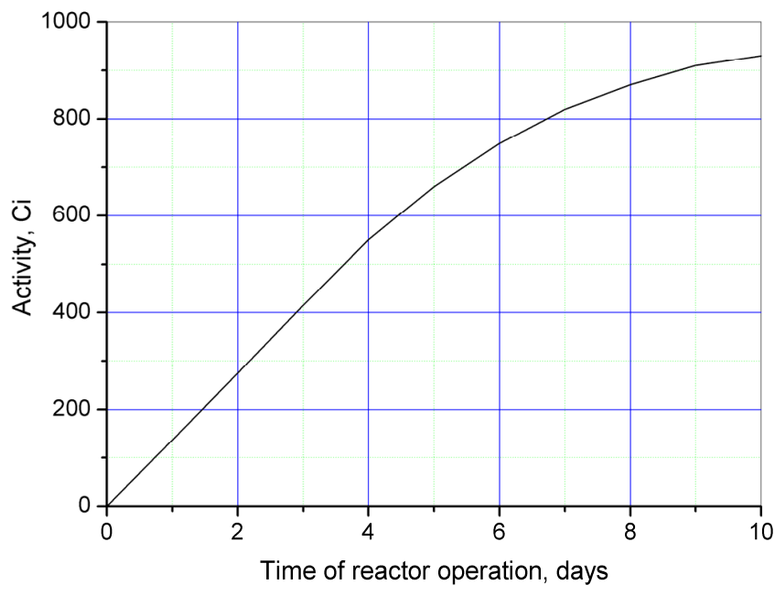


Fig. 6. Activity of ^{99}Mo in the core depending on reactor operating time

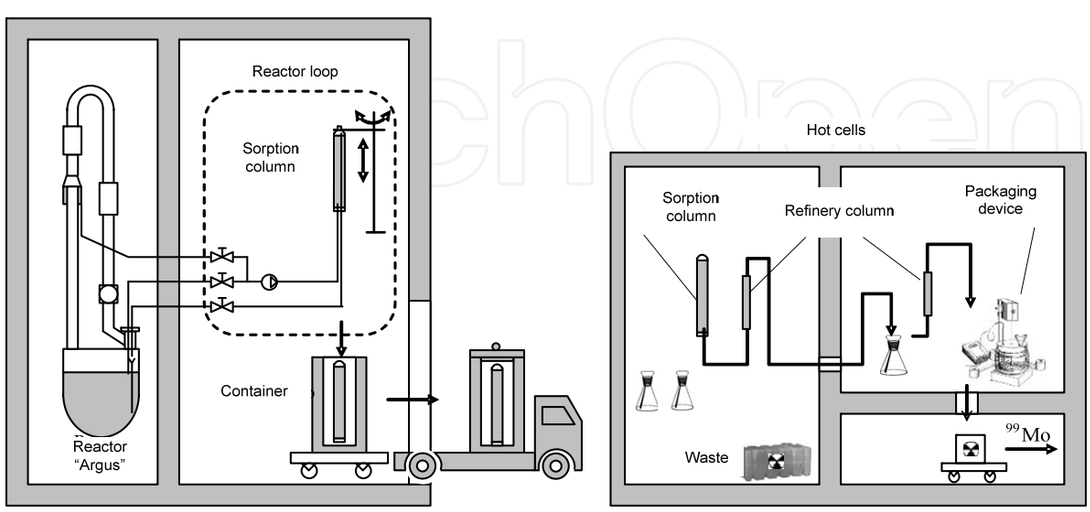


Fig. 7. Transport-technological scheme of demonstration complex at reactor "Argus"

Using the created demonstration equipment reactor experiments are performed on production of ^{99}Mo samples and working out of solution technologies. Quality certification of the samples was performed in Europe (Institute of radioactive elements, Belgium), and in the USA (Argonne National Laboratory) where the impurity content in the sample more than 10 times less than in the international requirements (see table 4) was confirmed. The maximum activity of samples made ~ 10 Ci.

On the basis of ^{99}Mo produced in the solution reactor "Argus", generators of ^{99m}Tc with the standard column filled with sorbent of type "Thermoxid" have been made. Eluted ^{99m}Tc has been investigated on injection suitability in State Scientific Center "Institute of Biophysics" of the Russian Federation and has received a good rating as for radionuclide purity, and absence of macro impurity of stable elements (manganese).

Nuclide	European requirements	Results of the samples analyses	
		IRE (Belgium)	ANL (USA)
α-activity $\alpha/^{99}Mo$	$<1.0\cdot10^{-9}$	$1.4\cdot10^{-10}$	$< 7.1\cdot10^{-10}$
β-activity $^{89}Sr/^{99}Mo$ $^{90}Sr/^{99}Mo$	 $<6.0\cdot10^{-7}$ $<6.0\cdot10^{-8}$	 $6.4\cdot10^{-9}$ $6.4\cdot10^{-10}$	 $1.2\cdot10^{-8}$ $7.8\cdot10^{-11}$
γ- activity $^{131}I/^{99}Mo$ $^{103}Ru/^{99}Mo$	 $<5.0\cdot10^{-5}$ $<5.0\cdot10^{-5}$	 $1.1\cdot10^{-5}$ $0.9\cdot10^{-6}$	 $4.2\cdot10^{-7}$ $1.4\cdot10^{-7}$
Others γ-, β- $(\beta-,\gamma-)_{\Sigma}/^{99}Mo$ $^{132}Te/^{99}Mo$ $^{133}I/^{99}Mo$ $^{125}Sb/^{99}Mo$	 $<1.0\cdot10^{-4}$	 $<1.0\cdot10^{-4}$ $4.4\cdot10^{-5}$ $3.4\cdot10^{-7}$ $2.7\cdot10^{-7}$	 $<3.8\cdot10^{-7}$ $8.9\cdot10^{-8}$ - $2.1\cdot10^{-7}$

Table 4. Results of ^{99}Mo samples quality analysis

4.2 Technology of radionuclide ^{89}Sr production in the solution reactor

One of the most effective ways is production of ^{89}Sr from uranium fission products, but formation of impurity ^{90}Sr makes impossible practical application of this way when uranium targets are used. The solution reactor type gives a unique possibility of solving this problem.

The method of fragment radionuclide ^{89}Sr extraction is based on a possibility during fission reaction in nuclear fuel of solution reactor to influence the genetic predecessors of a target radioisotope resulted in nuclear transformations of fission fragments in a decay chain of elements with the mass number 89: $^{89}Se \rightarrow ^{89}Br \rightarrow ^{89}Kr \rightarrow ^{89}Rb \rightarrow ^{89}Sr$. The earlier experimental researches have shown that practically all long-living isotopes of krypton and xenon leave in the gas phase of solution reactors (Loboda et al., 1989). The basic decay chains of the fission products, leading to formation of strontium radioisotopes with a half-life period of gaseous predecessors more than 1 second, are presented in Fig. 8.

Apparently from consideration of the basic decay chains leading to formation of radioactive strontium isotopes, the half-life period of the gaseous predecessor (^{89}Kr) of a target radionuclide ^{89}Sr makes 3.2 minutes, and the half-life period of a gaseous predecessor (^{90}Kr) of the main impurity radionuclide Sr^{90} is essentially less and makes 33 s. Using this circumstance, in the isolated volume of a gas after corresponding endurance it is possible to reach demanded value of the activity ratio of target gaseous predecessors and impurity strontium radionuclides, and then to pump over the gas mix through filters in accumulating volume in which after decay of predecessors ^{89}Sr will accumulate (Loboda et al., 1989; Abalin et al., 2000).

An experimental loop system consisting of a closed technological loop with rotation pumps, valves and vacuum–tight connecting units was used for taking gas samples from the reactor free volume. The hydraulic scheme of the experimental installation is shown in Figure 9.

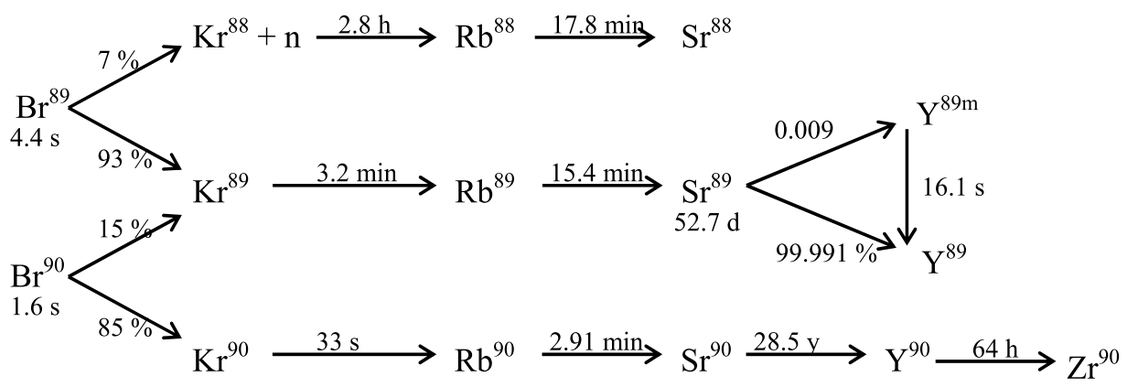


Fig. 8. Decay chains of fission products with atom masses 89 and 90

The experiments confirming the possibility of ^{89}Sr extraction from fuel solution of the reactor "Argus" were performed under the following scheme. The reactor operated at power from 5 to 20 kW within 20 minutes. Then after endurance within 5-6 minutes the gas-air mix was pumped over through a column filled with Rashig rings. The reactor loop mentioned above was used. Pumping time of the gas mix was 5 minutes, which is enough for filling the column. The columns were removed after 2-3 days of gaseous fission products decay. In the laboratory the aggregate taken from the column was washed out with solution of hydrochloric acid for extraction of ^{89}Sr and some accompanying fission products.

A series of experiments for confirmation of fragment ^{89}Sr production using exit of the gaseous predecessor ^{89}Kr from fuel solution was performed. Samples of ^{89}Sr chloride of high radionuclide purity are got in which ^{90}Sr impurity content has made less than $5 \cdot 10^{-4}$ %. The typical content of the produced ^{89}Sr chloride solution is given in Table 5.

With the obtained experimental estimations of ^{89}Sr yield in a solution reactor we can compare the specific productivities of reactors BR-10 (Russia, Obninsk) (Zvonarev et al., 1997), BOR-60, BR2 (Koonen, 1999).) with "Argus" reactor, normalized to the target mass and reactor power. For the sake of comparison, we accept the mass of ^{235}U in the solution equal to 1600 g, as a target. Table 6 shows specific yields of ^{89}Sr in various reactor installations.

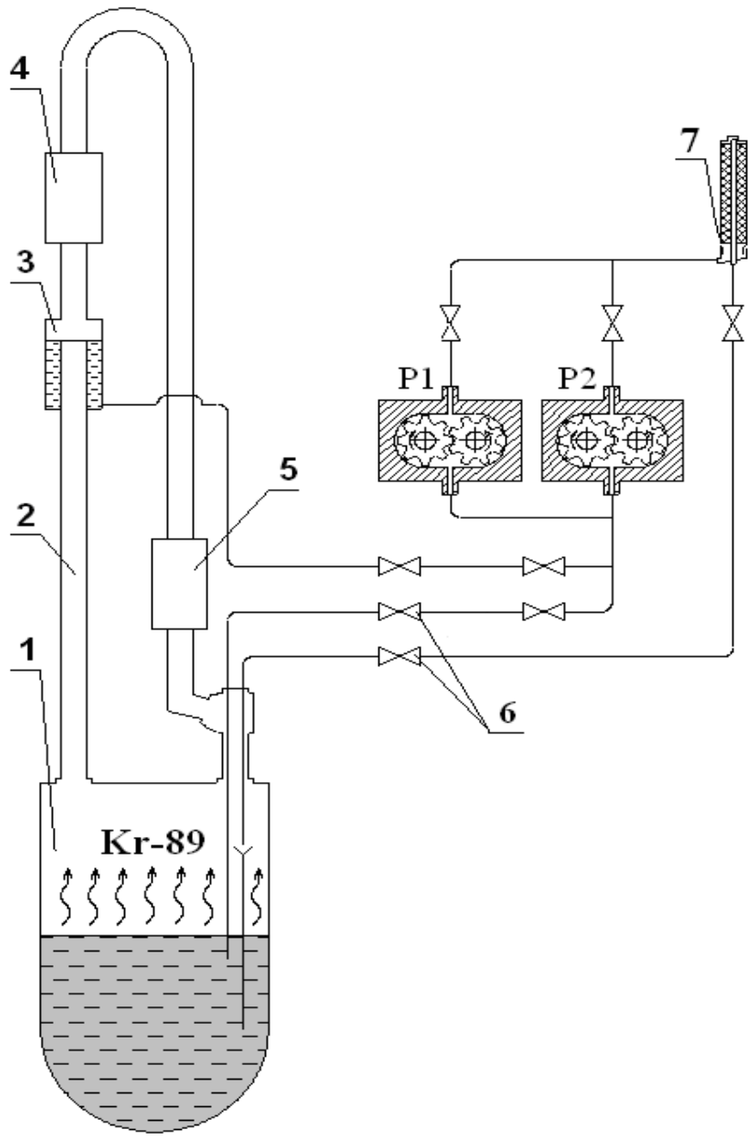


Fig. 9. Scheme of reactor "Argus" with experimental loop. 1 – reactor core; 2 – system of catalytic regeneration; 3 – accumulator of condensate; 4 - heat exchanger; 5 – catalytic recombiner; 6 – valves; 7 – sorption column; P1, P2 – pumps.

Radionuclide	Activity, relative units.		
	HEPA	MKF-F	MKF-U
Sr-89	1	1	1
Cs-137	$1.2 \cdot 10^{-2}$	-	-
Ba-140	$2.3 \cdot 10^{-2}$	$4.5 \cdot 10^{-4}$	$4.5 \cdot 10^{-6}$
Ce-141	$5.4 \cdot 10^{-4}$	-	-

Table 5. Typical content of the produced ^{89}Sr chloride solution

Specific yield of ^{89}Sr, MBq/g/kW			
BR-10	BOR-60	BR2	"Argus"
$7.4 \cdot 10^{-3}$	0.01	0.4	45.0

Table 6. Specific yields of ^{89}Sr in various reactor facilities

The advantage of the 20 kW solution reactor is obvious and undeniable. The expected "Argus" reactor productivity of $(1.5\text{-}1.8) \cdot 10^3$ GBq of ^{89}Sr a year is comparable with that of the 60-MW BOR-60 Reactor ($\approx 4 \cdot 10^3$ GBq/y). However, it is worth noting that ^{89}Sr in a solution reactor is a byproduct from its "main" production of medical $^{99}Mo/^{99}Tc$. The target radionuclide separation occurs spontaneously; a majority of fission fragments remains in the fuel, and the buildup of radioactive waste with the new technology is very low. The separated impurity radionuclides ^{133}Xe and ^{135}Xe can also be used in medicine, while ^{137}Cs is a valuable radionuclide for technical applications.

The unique properties of the solution reactor fuel open the doors for this type of nuclear installations to the nuclear medicine industry. Separation of radionuclides directly from fuel solution allows using the low power (50 kW) reactor for production of ^{99}Mo with the minimum yield of $1.85 \cdot 10^4$ GBq/day, 700 GBq/week of ^{133}Xe, as well as a number of other valuable radionuclides (Burrows, 1993; Zagryadsky et al., 2008).

So, the new method for medical ^{89}Sr production in the reactor with solution fuel is proposed which is characterized by simplicity, high production efficiency and low buildup of radioactive waste. The main advantages of the new technology were validated by numerous experiments. Basic features of the method are as follows:

- the mechanism for ^{89}Sr delivery to the sorption volume of the "Argus" reactor experimental loop is based on transport of gaseous ^{89}Sr predecessor – radionuclide ^{89}Kr;
- the radionuclide impurity composition of $^{89}SrCl_2$ solution includes elements with nuclear masses ^{137}, ^{140} and ^{141}, which have gaseous elements in their decay lines;
- filtration of the gas flow with mechanical aerosol filters reduces the content of radionuclide impurities 10^2-10^3 times;
- cleaning of ^{89}Sr chloride solution in chromatographic columns with DOWEX-50×8 or Sr-Resin ensures full removal of ^{137}Cs from the solution and significant reduction of $^{140}Ba/^{140}La$ impurity;
- ^{90}Sr impurity in $^{89}SrCl_2$ solution was not detected with the measuring instruments within the sensitivity limits of ($\approx 5 \cdot 10^{-4}$ %).

5. Conclusions

The key problem of modern medical radionuclide production in research nuclear reactors - considerable amount of a highly active waste and use in technology of highly-enriched uranium - can be solved by transition to use of homogeneous liquid nuclear fuel enriched by isotope of uranium ^{235}U less than 20 %. It is known (Basmanov, E. et al., 1998), that conversion traditional «target» technology of fragment radioisotopes production from HEU to LEU in a target cycle not only does not solve a problem of a radioactive waste (the yield

of fission products per unit ^{99}Mo activity is the same), but also aggravates it: the amount of the irradiated uranium, the radioactive waste accompanying processing of more uranium for the same ^{99}Mo yield, both accumulation of ^{239}Pu and other transuranic elements increase in times. That increases the product cost and difficulty of disposal. A target turnover cycle of fission materials is absent in the proposed concept of medical isotopes production. Therefore, on the one hand, the overwhelming part of the waste connected with fission of uranium appears "locked" in a reactor core or a loop installation, and, on the other hand, there is no problem of fission materials proliferation in a target cycle. The question of conversion of the considered reactors with liquid fuel from HEU to LEU does not concern actually technology of medical isotopes production, and should be considered within the general strategy of nuclear reactors safety. Technical possibility of such conversion does not call doubts.

The accumulated operational experience with solution and molten salt fuel, presence of the developed experimental base provide a possibility for performing a broad spectrum of technological researches directed on creation and realization of new methods for production of radionuclides and radiopharmaceuticals for nuclear medicine.

Carried out in the NRC "Kurchatov Institute" model experiments at the solution reactor "Argus" have confirmed basic provisions of the new law-waste technologies of ^{99}Mo and ^{89}Sr production.

This newest technology of the most important medical radionuclide production allows to lower necessary power of a reactor 100 and more times comparing to traditional technology that not only leads to decrease in total radioactive wastes of production, but also supposes placing the reactor in the occupied areas and promotes solving the licensing problems. The efficiency of ^{235}U use comes practically to 100 % while traditional target technologies use only 0.4 % of ^{235}U for reception of the final product, and other part of uranium after target processing, as a rule, goes to utilization.

6. References

Abalin, S. et al. (2000). Method of strontium-89 radionuclide reception. *Patent of Russian Federation # 2155399*, 2000

Abalin, S. et al. (2002). Method of Strontium-89 Radioisotope Production. *US Patent 6,456,680*. September 24, 2002.

Afanasev, N., Benevolenskij, A., Ventsel, O. et al. (1986). Reactor "Argus" for laboratories of nuclear-physical methods of the analysis and the control, *Atomic energy* (Russian), 1986, v. 61, iss. 1, pp. 7-9.

Ball, R. (1999). *IAEA-TECDOC-1065*, IAEA, Vienna, 1999, pp.5-17.

Ball, R., Nordyke, H. & Brown, R. (2002). Considerations in the Design of a High Power Medical Isotope Production Reactor, *Abstracts at the 24th International Meeting RERTR*, Argentina on November 3-8, 2002.

Basmanov, E. et al. (1998). Management of radioactive waste from ^{99}Mo production. *IAEA-TECDOC-1051*, Vienna, November 1998, p. 39.

Bettis, E., Schroeder, R., A.Cristy G. et al. (1957). The Aircraft Reactor Experiment. Design and Construction. *Nucl. Sci. Engng*. 1957, v.2, N 6, pp. 804-826.

Burrows, T. (1993). Nuclear Data Sheets Update for A = 95. *Nuclear Data Sheet,* 68, 1993.

Chuvilin, D. & Zagryadskiy V. (1997). The low radioactive waste method of a fragment Mo-99 production. *Proc. of the Third European Workshop on Chemistry, Energy and the Environment*, Estoril, Portugal, 25-28 May 1997, pp. 381-388.

Chuvilin, D. & Zagryadsky, V. (1998). Method of reception of isotope ^{99}Mo. *Patent of Russian Federation #2102807*. 1998

Gerasimov, A., Kiselev, G. & Lantcov M. (1989). Reception of ^{99}Mo in nuclear reactors. *Atomic Energy* (Russian), v.67, iss. 2, 1989, pp. 104-108 (in Russian).

Grimes, W. (1970). Molten-salt reactor chemistry. *Nucl. Appl. Technol.* 1970, v.8, N 2, pp. 137-155.

Hansell, C. (2008). Nuclear medicine's double hazard. Imperiled Treatment and the Risk of Terrorism. *Nonproliferation Review*, v.15, No. 2, July 2008.

Haubenreich, P. & Engel, J. (1970). Experience with the Molten-Salt Reactor Experiment. *Nucl.Appl. Technol.* 1970, v.8, N 2, pp.107-140.

IAEA (1999). Production Technologies for Molybdenum-99 and Technetium-99m, *IAEA-TECDOC-1065*, 1999.

Kirslis, S. & Blankenship, F. (1967). *Report ORNL-4076*, March 1967, p. 51.

Kirslis, S. & Blankenship, F. (1969). *Report ORNL-4344*, February 1969, p. 119.

Koonen, E. (1999). The BR2 Reactor, a Major European Radioisotope Producer. *Int. Conf. GLOBAL'99 "Nuclear Technology - Bridging the Millennia"*, August 29 - September 3, 1999.

Kolesov, V. (1999). Aperiodic pulse reactors. Sarov, RFNC VNIIEF, 1999 (in Russian).

Loboda, S. et al. (1989). Exit of fission products from a fuel of solution reactor. *Atomic energy* (Russian), v. 67, iss. 6, December 1989, pp. 432-433 (in Russian).

Menshikov, L. & Chuvilin, D. (2008). *Preprint IAE-6526/12*, (2008) (in Russian).

Nesmeyanov, A. (1978). Radiochemistry. M., Chemistry, 1978 (in Russian).

Nikolsky, B. (Ed(s).). (1998). *Directory of the chemist*. Vol. 1. Ed. Chemistry, Leningrad branch, 1971, (in Russian).

Novikov, V. et al. (1990). Molten salt NEU: prospects and problems. M., Energoatomizdat, 1990 (in Russian).

Pavshouk, V. & Chuvilin, D. (2005). Reception of radionuclides – fission fragments of nuclear fuel. In: *Book of Isotopes*. V. 1. Properties. Reception. Application. M, Phyzmatlit, 2005, p. 524 (in Russian).

Rabotnov, N. (1999). Radiation pharmacology - revolution in public health services. *Nuclear society* № 2-3, (September 1999) pp. 35-38 (in Russian).

Rosenthal, M., Briggs, R. & Kasten, P. (1968). *Report ORNL-4254*, August 1968, pp.101-107.

Rosenthal, M., Briggs, R. & Kasten, P. (1969). *Report ORNL-4344*, February 1969, p.136.

Starkov, O. (1996). Production of radioisotopes for medicine and scientific researches. SSC-RF PhEI:50 Obninsk, 1996, pp. 359-363 (in Russian).

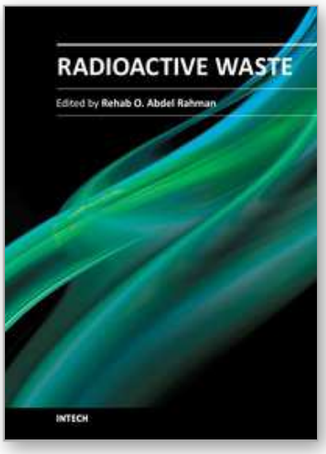
Westmacott, C. (2010). The Supply of Medical Radioisotopes. An Economic Study of the Molybden-99 Supply Chain, *OECD*, 2010, NEA No6967

Zagryadsky, V., Menshikov, L. & Chuvilin, D. (2008) *Preprint IAE-6518/4*, 2008 (in Russian).

Zvonarev , A. et al. (1997). Reception of ^{89}Sr in fast reactors. *Atomic Energy* (Russian), v.82, iss.5, May 1997, pp. 396-399 (in Russian).

IntechOpen

IntechOpen



Radioactive Waste

Edited by Dr. Rehab Abdel Rahman

ISBN 978-953-51-0551-0

Hard cover, 502 pages

Publisher InTech

Published online 25, April, 2012

Published in print edition April, 2012

The safe management of nuclear and radioactive wastes is a subject that has recently received considerable recognition due to the huge volume of accumulative wastes and the increased public awareness of the hazards of these wastes. This book aims to cover the practice and research efforts that are currently conducted to deal with the technical difficulties in different radioactive waste management activities and to introduce to the non-technical factors that can affect the management practice. The collective contribution of esteem international experts has covered the science and technology of different management activities. The authors have introduced to the management system, illustrate how old management practices and radioactive accident can affect the environment and summarize the knowledge gained from current management practice and results of research efforts for using some innovative technologies in both pre-disposal and disposal activities.

How to reference

In order to correctly reference this scholarly work, feel free to copy and paste the following:

D. Yu. Chuvilin, V. E. Khvostionov, D. V. Markovskij, V. A. Pavshouk and V. A. Zagryadsky (2012). Low-Waste and Proliferation-Free Production of Medical Radioisotopes in Solution and Molten-Salt Reactors, Radioactive Waste, Dr. Rehab Abdel Rahman (Ed.), ISBN: 978-953-51-0551-0, InTech, Available from: http://www.intechopen.com/books/radioactive-waste/low-waste-and-proliferation-free-production-of-medical-isotopes-in-solution-and-molten-salt-reactors

INTECH
open science | open minds

InTech Europe
University Campus STeP Ri
Slavka Krautzeka 83/A
51000 Rijeka, Croatia
Phone: +385 (51) 770 447
Fax: +385 (51) 686 166
www.intechopen.com

InTech China
Unit 405, Office Block, Hotel Equatorial Shanghai
No.65, Yan An Road (West), Shanghai, 200040, China
中国上海市延安西路65号上海国际贵都大饭店办公楼405单元
Phone: +86-21-62489820
Fax: +86-21-62489821

© 2012 The Author(s). Licensee IntechOpen. This is an open access article distributed under the terms of the Creative Commons Attribution 3.0 License, which permits unrestricted use, distribution, and reproduction in any medium, provided the original work is properly cited.