# Production of High-Specific Activity Radionuclides Using the SM-High-Flux Reactor

Y. A. KARELIN, YU.G. TOPOROV, V.T. FILIMONOV, V.A.TARASOV,
F.Z. VAHETOV, R.A. KUZNETSOV, V.M. LEBEDEV, M.I.MELNIK, V.D.GAVRILOV
State Scientific Center of Russian Federation
Research Institute of Atomic Reactors (SSC RF RIAR)
433510 Dimitrovgrad-10, Russia

**SUMMARY.** The development of HSA radionuclides production technologies is one of the directions of SSC RIAR activity, and the high flux research reactor SM, having neutron flux density up to $2.10^{15}$ $cm^{-2}s^{-1}$ in a wide range of neutron spectra hardness, plays the principal role in this development. Using of high-flux reactor for radionuclide production provides the following advantages:

- production of radionuclides with extremly high specific activity,
- decreasing of impurities content in iradiated targets (both, radioactive and non-radioactive),
- cost-effective using of expensive isotopically enriched target materials.

The production technologies of $^{33}P$, $^{153}Gd$, $^{188}W$, $^{63}Ni$, $^{55,59}Fe$, $^{113,117m,119m}Sn$, $^{89}Sr$, applied in industry, nuclear medicine, research, etc., were developed by RIAR during last 5-10 years. The research work includes the development of calculation procedures for radionuclide reactor accumulation forecast, experimental measuring of neutron cross-sections, the development of irradiated materials reprocessing, isolation and purification of radionuclides. The principal results are reviewed in the report

## 1. INTRODUCTION

One of the important characteristics of radionuclide preparations is their specific activity (SA) i.e. the amount of radionuclide per volume or mass unit of a preparation. An increase of specific activity allows for the implementation of the following advantages:

1. Decrease of the amount of starting material used for production of activity unit that is important for the usage of isotope-enriched (i.e. very expensive) materials.
2. Decrease of the relative amount of radionuclide impurities.
3. Decrease of the dimensions of a source that is important for the production of ionization sources applied for medicine and radiation flaw-detection.
4. Reduction of self-absorption of «useful» radiation in the source material that is essential for β-particles and low-energy gamma-radiation.
5. New fields of application of radionuclides, particularly in medicine.

There are also other advantages of high-specific activity preparations that should be indicated.

A natural way of production of high-specific activity preparations is accumulation of «carrier-free» radionuclides using accelerators. This method allows for the production of radionuclides with the specific activity close to the theoretical value. At the same time this method is characterized by a relatively low productivity (usually, up to several Ci). An essentially higher productivity can be achieved through the production of radionuclides using neutron irradiation in nuclear reactors. In this case high-flux research reactors play the main role. However, in this case an increase of specific activity requires investigation of parameters of the accumulation process for this or that radionuclide taking account of the increase of the rate of all nuclear reactions including burnup rate of the useful radionuclide as well as the formation rate of impurity radionuclides. In other words, the development

methods for accumulation of useful radionuclides with the required properties in high-flux nuclear reactors is a complicated optimization task. The given paper presents the main investigation results on the possibility to produce high-specific activity preparations using the SM-high-flux research reactor.

## 2. BRIEF DISCRIPTION OF THE SM-REACTOR

The SM-high-flux research reactor (Klinov et al (1)) is intended for accumulation of TUE and radioactive isotopes of lighter elements, for irradiation of specimens of reactor materials to study their properties under irradiation and for investigations in the field of nuclear physics. The reactor design implements a concept of production of high density of thermal neutron flux in a moderating trap with a hard neutron spectrum situated in the center of the core.

The reactor core is located in a steel vessel having a diameter of 1.46m and a height of 7.33m. The vessel is designed for an operating pressure of 5MPa. Uranium dioxide of 90% - enrichment dispersed in the copper matrix is used as fuel.

The reactor is equipped with a wide range of experimental facilities. In addition to 27 cells in the neutron trap, there are 30 vertical channels located at a different distance from the core in the beryllium reflector.

## 3. SPECIFIC FEATURES OF RADIONUCLIDE PRODUCTION IN A HIGH-FLUX REACTOR

### 3.1 BURNUP OF A USEFUL RADIONUCLIDE

On irradiation in a high-flux reactor the burnup rate of a radionuclide, due to its interaction with neutrons, can appear comparable with that of its radioactive decay. A possibility to consider this effect correctly through calculations is determined by the availability of neutron cross-sections of radionuclides. At SSC RIAR this problem was solved experimentally by measuring total neutron cross-sections of radionuclides with the neutron spectrometer (Belanova et al (2)) and specially prepared specimens. The neutron spectrometer was installed in the horizontal channel of the SM-reactor and the «time-of-flight» method was implemented. The advantages of the method were supplemented by:

- possibility of accumulation of specimens with the required characteristics by the irradiation of special targets in the SM reactor;
- availability of necessary radiochemical technologies for the production of specimens;
- combination of other methods, e.g. such as radiometry and mass-spectrometry, used for the analysis of specimens.

Using a neutron spectrometer, the total cross-sections of more than 70 stable and radioactive nuclides were measured that made it possible to optimize the conditions of their production.

Fig. 1 gives an example of calculations of $^{113}Sn$ yield with or without consideration (radioactive decay only) of burnup in the course of irradiation (Toporov et al (3)).

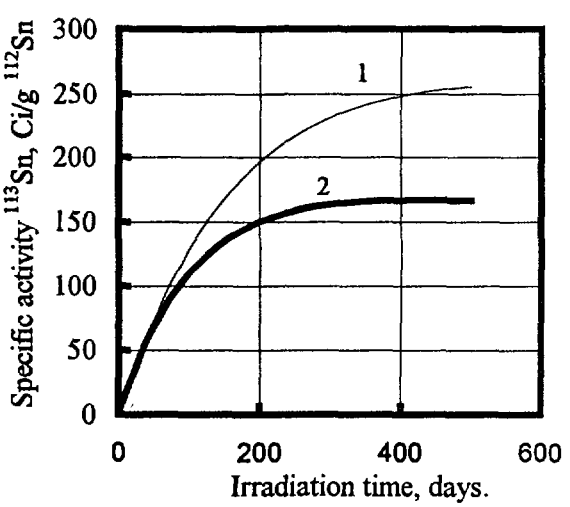


Fig.1 Specific activity $^{113}Sn$ vs irradiation time. 1- without consideration of $^{113}Sn$ "burn-up", 2- with consideration of $^{113}Sn$ "burn-up"

The presented data illustrate the real burnup range of a useful radionuclide in the high-flux reactor.

### 3.2 SENSITIVITY TO A NEUTRON SPECTRUM

The accumulation of some radionuclides with high values of resonance integrals is very sensitive not only to the amplitude of the neutron flux density but to the neutron spectrum as well. Let us remind the neutron constants of nuclides involved into the $^{252}Cf$ accumulation chain.

The resonance integrals of the «key»-nuclides, $^{246}Cm$ and $^{248}Cm$, are approximately two orders of magnitude higher than their thermal cross-sections, while «intermediate» $^{249}Bk$ and "useful" $^{252}Cf$ have approximately equal cross-section values in the thermal and resonance range of neutron energies (Anufriev et al (4)). This determines the following:

- decrease of the thermal neutron flux density at a constant resonance neutron flux density (i.e. increase of the neutron spectrum hardness) can maintain the accumulation rate of $^{252}Cf$ and reduce the rate of its loss;
- increase of the resonance neutron flux density is more efficient than that of thermal neutrons;
- certain effects can be achieved by changing the neutron spectrum in the process of irradiation.

In 1992 during the last reconstruction of the SM-high-flux reactor the neutron trap design was changed so that the epithermal neutron flux density increased by about 15%. In order to test experimentally the effect of this change on $^{252}Cf$ yield, two similar targets with heavy curium isotopes were irradiated. One of these target was irradiated before the reconstruction and the other - after the reconstruction. Fig. 2 shows calculation and experimental data on $^{252}Cf$ yield (Klinov et al (5)). One can see that «hardening» of neutron spectrum led to a marked increase in californium yield per 1 g of starting materials.

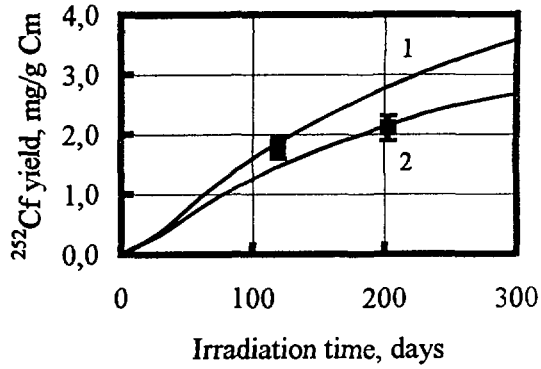


Fig.2 Comparison of 252Cf yield during the heavy curium irradiation on neutron trap SM reactor. 1-after neutron trap reconstruction, 2- before reconstruction, ▮ - experiment

### 3.3 SENSITIVITY TO THE REACTOR OPERATING CONDITIONS

As a rule the duration of the reactor operation campaign is determined by its physical features, in the first place, by the fuel burnup rate. In a high-flux reactor the power density in fuel is high (for SM in average - 2 MW/l) that limits the campaign duration. Thus, the SM operation duration till the next loading is about 10 days followed by a break for 1.5-2.0 days for fuel reloading. Such operating conditions influence the accumulation of radionuclides with a short half-life or those which have precursors with a short half-life. Fig. 3 gives an example of this influence presenting a dependence of $^{188}W$ (which is formed from $^{186}W$ through intermediate $^{187}W$ (T1/2=2.37 h)) specific activity taking account of the real operation schedule of the reactor (Toporov et al (6))

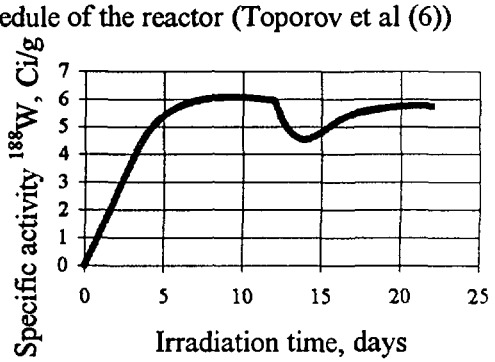


Fig.3 Specific activity $^{188}W$ vs irradiation time during real reactor schedule.

These examples illustrate the specific features of the radionuclide accumulation in high-flux reactors and prove the necessity of scientific support of practical work. At SSC RIAR such scientific support is provided with a special software complex, by the possibility to perform mock-up experiments and by qualified and experienced personnel. 30-year experience of radionuclide production made it possible for us to supply radionuclide products to 5 continents, the quality of these products compares well with that of the developed countries. A brief overview of the production methods of some of the products you will find below.

**$^{33}$P.** SSC RIAR uses a «traditional» technology for the production of this radionuclide, including the irradiation of $^{33}$S in the reactor, separation of $^{33}$S from the target material by vacuum distillation, phosphorus transfer into solution followed by its ion-exchange purification from impurities. The specific feature of the RIAR technology is the use of the «hard» neutron spectrum of the SM-high-flux reactor core that increases the $^{33}$P yield up to 7-8 CI/gS and improves essentially the quality of the preparation due to the reduction of the amount of impurities.

$^{33}$P-orthophosphoric carrier-free acid has a molar activity of 4200-5000 Ci/mmole (theoretical value - 5200 Ci/mmole), polyphosphate activity contribution is less than 0.1-0.2% and total impurity concentration is less than 0.02g/l.

**$^{153}$Gd.** (Tarasov et al (7)) Natural europium is used as starting material for production of $^{153}$Gd. The calculated and experimental data presented indicate to a possibility to optimize the irradiation conditions of europium target so that on completion of irradiation $^{153}$Gd under production has specific activity not less than 120-150 Ci/g (up to 100 Ci/g on completion of its reprocessing) and its yield is 6-7 Ci/g of europium.The production procedure is based on the europium cementation with sodium amalgam followed by the extraction-chromatography of $^{153}$Gd from impurities..
The preparation can be delivered to a customer as oxide (in the form of pellets as well) or chloride. An essential part is used for the production of low-energy photon-radiation sources applied for bone-densitometers, densitometers, etc. At present at RIAR the work is being performed on the development of linear sources with an activity from 50mCi to 1Ci with an active part length up to 350-500mm and a diameter of 2-6mm.

**$^{188}$W.** The production of $^{188}$W with a required specific activity value is possible only using a high-flux reactor while this radionuclide is formed from enriched $^{186}$W through a double neutron capture through the reaction $^{186}$W(n,$\gamma$ )$^{187}$W(n, $\gamma$) $^{188}$W.

At SSC RIAR $WO_3$ of no less than 98.5% enrichment $^{186}$W is used as starting material. Post-irradiation reprocessing includes dissolution of wolfram oxide in sodium hydroxide solution, cation-exchange purification of wolfram from impurities (if necessary) and production of wolfram oxide or sodium wolframate solution in sodium hydroxide. Specific activity is not less than 5Ci/g.

**$^{199m}$Sn.** We have developed an unconventional technology of the post-irradiation processing of irradiated tin based on recovering of tin up to a metal state with metal aluminium from citrate solutions. It was shown, that in this case a satisfactory purification is achieved from a large group of impurities including antimony radionuclides. For a deeper purification from antimony radionuclides an anion-exchange in the Dowex-1 - HCl system can be used. $^{119m}$Sn specific activity is 0.5-1.0 Ci/g, radionuclide impurities content is less than 1%.

The developed technology is characterized by a simple remote operation. It can be used for the production of other tin radionuclides, e.g. $^{113}$Sn and $^{117m}$Sn.

**$^{51}$Cr.** Metal chromium, enriched in $^{50}$Cr nuclide up to 90%, is used for the production of this radionuclide. The irradiation in the high-flux reactor during one campaign allows for the production of a preparation with a specific activity of metal of 1000-2000 Ci/g. The specific feature of the production process of $^{51}$Cr preparations ($^{51}CrCl_3$ or $Na_2{}^{51}CrO_4$) is that

in the course of irradiation an essential part of metal transforms into a hard-dissoluble form (probably, chromium nitride) and, therefore, dissolution of the irradiated material is performed in concentrated chlorohydric acid with phosphoric acid additions. The next purification of chromium (in the form of chromate-ions) from phosphate-ions is conducted by the anion-exchange method. The radionuclide purity of $^{51}Cr$ preparations under production is higher than 99.99%, content of non-radioactive impurities in the preparation solution corresponds to the usual level of impurities in highly-pure acids of the analytical grade.

**$^{55}Fe$, $^{54}Mn$** (Filimonov et al (8)) In order to produce these radionuclides, highly-enriched $^{54}Fe$ is used as starting material. $^{55}Fe$ specific activity is not less than 60Ci/g after irradiation in the SM-reactor trap during 200 days of its efficient operation. Simultaneously, a marked amount of carrier-free $^{54}Mn$ (0.8-1Ci/g of iron) is formed through (n,p) reaction. The reprocessing of irradiated targets, after their dissolution in 4-6 mole/l chlorohydric acid, includes a selective separation of $^{55}Fe$ from impurities (MN, Cr and Co) by the extraction-chromatography method in «three-n-butyl phosphate-HCl» or «three-n-octylamin-HCl» systems or by co-precipitation of manganese with iron-hydroxide (after chromium stabilization in 6-valent state) followed by the separation of $^{55}Fe$, $^{60}Co$ and $^{54}Fe$ using the extraction-chromatography method.

**$^{63}Ni$.** The production of this radionuclide with practically acceptable specific activity (>10Ci/g) is possible only using highly-enriched $^{62}Ni$ as starting material, the irradiation duration being not less than 200-250 efficient operation days in the position with the highest thermal neutron flux density ($>1.10^{15}cm^{-2}s^{-1}$).

The radiochemical reprocessing of an irradiated target involves $^{63}Ni$ purification from radionuclides of $^{59}Fe$ and $^{60}Co$ by the extraction-chromatography method and purification from $^{51}Cr$ impurity by the electrochemical precipitation method.

**OTHER RADIONUCLIDES WITH HIGH SPECIFIC ACTIVITY.** In addition to the above- mentioned radionuclides, SCC RIAR can produce other radionuclide preparations and sources with the required high specific activity that is possible only using high-flux reactors.

## ACKNOWLEDGMENTS

This work was supported by Minatom of Russia

## REFERENCES

1) Klinov A.V. et al. "Redisign of SM-2 Reactor Facility", Nov.16-20,1992, Vienna. Technical Commitee Meeting on Management of Research Reactor Ageing.
2) Belanova T.S. et al. 1976, Dimitrovgrad, "Time of flight neutron spectrometer on a reactor SM-2". Preprint RIAR, П-6 (272).
3) Yu.G.Toporov, F.Z.Vakhetov, O.I.Andreyev, R.A.Kuznetsov.. Radionuclides $^{113}Sn$ and $^{119m}Sn$: reactor production and chemical reprocessing of irradiated targets. //, Proc. of 3rd Topical Meeting on Industrial Radiation and Radioisotopes Measurements and Application (IRRMA'96), October 6-9, 1996, Raleigh, NC, p.196.
4) Anufriev V.A. et al.. Total neutron cross-section $^{249}Bk$ on energy range 0.02-0.46 eV//Atomnaja energija, V.55, 1983 r., p.320
5) A.V.Klinov, Yu.G.Toporov. Optimization of $^{252}Cf$ accumulation in SM reactor.// Proc. of 3rd Topical Meeting on Industrial Radiation and Radioisotopes Measurements and Application (IRRMA'96), October 6-9, 1996, Raleigh, NC, p.182
6) Yu.G.Toporov, V.A.Tarasov, R.A.Kuznetsov, G.V.Goncharova. Production of tungsten-188 preparation // Ibid., p.195

7) V.A.Tarasov, Yu.G.Toporov, V.T.Filimonov, A.A.Yadovin, V.M.Lebedev, R.A.Kuznetsov, M.I.Melnik, Ya.N.Gordeev, Ye.A.Karelin. High-specific activity gadolinium-153 production technology.// Ibid., p.193

8) V.T.Filimonov, R.A.Kuznetsov, A.N.Pakhomov, O.I.Andreyev. Application of extraction chromatography in HSA $^{55,59}Fe$, $^{54}Mn$, $^{63}Ni$, $^{58}Co$ production technology. // Ibid, p.186.