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Evaluation of Sn-117m Production in the SM-3 High-Flux Reactor

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Abstract. Due to its electron emission, the ^{117m}Sn isotope is an important radionuclide for therapeutic applications, showing promise in the palliative treatment of painful metastases in bone cancers. Also, ^{117m}Sn emits a specific 158.6 keV gamma ray which allows imaging for targeting and also for dosimetric purposes. The SM-3 reactor is a 100 MWth high flux reactor able to produce a wide range of radioisotopes for various applications in nuclear medicine and industry. In this work, we investigated the modeling of ^{117m}Sn production in the SM-3 high flux reactor. Calculations are performed using the ChainSolver 2.34 code. The calculations showed that the specific activity of ^{117m}Sn was about 10 Ci.g^{-1} during 5 full power irradiation days.

Keywords. ^{117m}Sn, SM-3 high flux reactor, Radioisotope production, ChainSolver 2.34 code.

INTRODUCTION

The Tin-117m isotope, also referred to as ^{117m}Sn, is a highly significant radioisotope renowned for its therapeutic applications. With a half-life of 13.6 days and a gamma energy emission of 158.6 keV, it holds great potential in the field of medicine. Being a beta emitter, ^{117m}Sn emits high-energy, slow-converting electrons at 127, 129, and 152 keV, with a short effective range of 0.22 to 0.29 mm in tissue. This unique characteristic allows for the elimination of malignancies while minimizing damage to bone marrow

and other healthy tissues (Ponsard et al., 2009).

Moreover, the gamma photons released at 158.6 keV enable imaging, making it invaluable for precise targeting in medical diagnostics and treatment.
The current focus is on demonstrating the feasibility of producing ^{117m}Sn in the SM-3 high-flux reactor located in Russia. This production effort aims to supply other laboratories with a substantial quantity of high specific activity Tin-117m, thereby fostering collaborative research and enabling the development of a joint production plan for this radioisotope. By increasing the availability of Tin-117m, researchers can explore its therapeutic potential in a broader range of applications and further enhance its contributions to medical advancements.

PRODUCTION OF ^{117m}Sn

Therapeutic radiopharmaceuticals that rely on specific beta emissions of radio-nuclides, often accompanied by imaging photons, are widely used in the field. Radiotherapy has two goals: to effectively relieve pain caused by bone metastases and to significantly enhance patients' quality of life. From a diagnostic viewpoint, ^{117m}Sn is a good radioisotope candidate since its rays have energy of 158.6 keV, which is comparable to that of ^{99m}Tc. This tin isomer is a perfect replacement for ^{99m}Tc because it has a half-life of 13.6 days and decays towards a stable state with a certain amount of conversion electrons (Shearer et al., 1988).
^{99m}Tc is always contaminated with ^{99}Mo, which leads to unnecessary doses of radiation given to patients during radiopharmaceutical injection (Dantas et al., 2005).
The inclusion of the beta emitter ^{99}Mo in the radiopharmaceutical solution injected into the patient constitutes a superfluous radiation dosage; the dose coefficient for ^{99}Mo is about 50 times more than that of ^{99m}Tc (Hassanpour et al., 2019). ^{117}Sn is suggested for palliative therapy of bone metastases discomfort (Młyńczyk et al., 2020).
As other radioisotopes the use of ^{117m}Sn for therapeutic purposes is characterized by the targeted administration of radiation doses to target organs, as well as by its low toxicity and short half-life.
Unlike beta emitters, ^{117m}Sn emits slow gamma mono-energetic with a short range, enabling the elimination of cancerous cells while causing minimal harm to healthy tissues, including bone marrow. Additionally, the emitted gamma photons at 158.6 keV (86% intensity) can be employed to monitor the distribution and administration of the radiopharmaceutical within the patient (Shearer et al., 1988).
The production of ^{117m}Sn is achievable in a nuclear reactor through two main methods: the radiative capture of thermal neutrons on targets enriched in ^{116}Sn (^{116}Sn(n, γ)^{117m}Sn) or the inelastic neutron scattering on ^{117}Sn-enriched targets (^{117}Sn(nf, n'γ)^{117m}Sn). The former reaction, accomplished using thermal neutrons, exhibits a cross-section of approximately 6 ± 2 mb (IAEA, 2003).The latter reaction, involving fast neutrons with threshold energy of 318 keV, presents a significantly higher cross-section of around 222 ± 16 mb (Mirzadeh et al., 1977).
According to Mirzadeh et al. (Mirzadeh et al., 1977), the inelastic neutron scattering on ^{117}Sn-enriched targets is the most favorable reaction for the production of ^{117m}Sn in high-flux

reactors due to the substantial gain in cross-section. Figure 1 illustrates the ^{117m}Sn production chain.

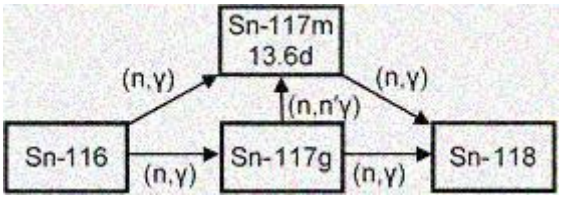

Fig.1. ^{117m}Sn Production chain.

THE HIGH FLUX SM-3 REACTOR

The SM-3 of the Research Institute of Atomic Reactors (RIAR), Dimitrovgrad is a pool-type reactor, meaning that the reactor core is located within a pool of water. This design allows for efficient cooling of the reactor and easy maintenance of the core. The SM-3 reactor is high-flux water-vessel reactor with a thermal power of 100 MW is designed mainly to produce transuranic elements and radioactive isotopes of lighter elements and to irradiate samples of reactor materials and study their properties during irradiation (Golovanov et al., 2006).

The SM-3 reactor is fueled with highly enriched (90% enrichment) Dioxide Uranium UO2. The reactor uses a graphite moderator and a Beryllium reflector to slow down neutrons and increase the likelihood of fission. The reactor is also equipped with several safety systems, including a passive emergency cooling system, to ensure that the reactor remains safe and stable during operation. The reactor has an extremely compact core with a metallic Beryllium reflector, housed in a steel reactor vessel and consisting of 28 fuel assemblies. The fuel assemblies are rod-type, cross-sectional cross-shaped fuel rods. The fuel composition is, dispersed in a copper matrix with the addition of Beryllium Bronze (Diakov, 2014).

The SM-3 reactor is primarily used for the production of medical isotopes, as well as for research and development purposes. The reactor produces a number of different isotopes, including Molybdenum-99 (^{99}Mo), which is used in the production of Technetium-99m (^{99m}Tc). In addition to medical isotope production, the SM-3 reactor is also used for materials testing and neutron activation analysis. The reactor's high thermal output and intense neutron flux make it an ideal facility for these types of experiments.

The SM-3 high flux reactor generates thermal neutron flux density exceeding 10^{15} n cm^{-2} s^{-1}. This high thermal neutron flux provides a crucial potential for radioisotope synthesis for use in industry and medicine. Horizontal cross-section of core geometrical of SM-3 reactor is shown in Fig. 2 (Tarasov et al., 2007). The SM-3 reactor has been in operation since 1973 and has a long history of safe and reliable operation. The SM-3 reactor is a valuable resource for medical isotope production, research and development, and materials testing. Its efficient design and high thermal output make it an ideal facility for these types of activities. The SM-3 reactor offers a high thermal flux up to 10^{15} n cm^{-2} s^{-1}.

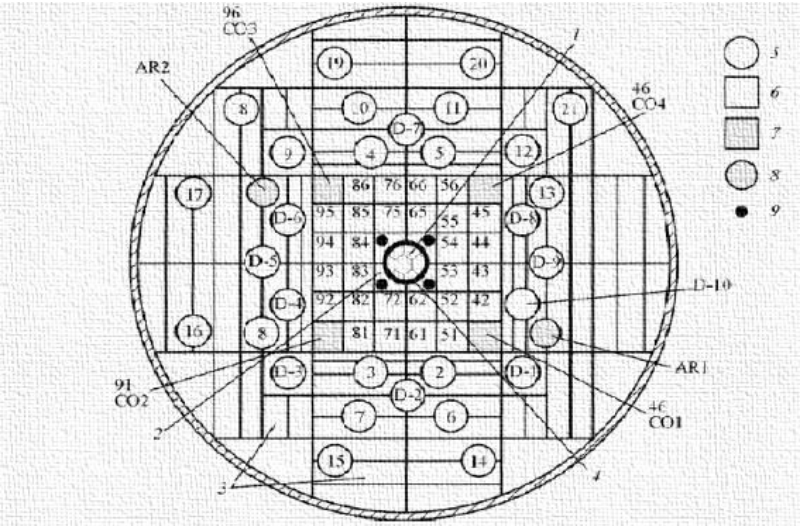

Fig.2. Map of the SM-3 reactor: (1) central block of transmutation targets, (2) beryllium bushes, (3) beryllium block of the reflector, (4) central compensating controller, (5) channels, (6) core sub-channels, (7) compensating controllers, (8) automatic controllers, and (9) safety rods (Tarasov et al., 2007).

SPECIFIC ACTIVITIES OF ^{117m}Sn IN THE SM-3 HIGH FLUX REACTOR

ChainSolver code description

ChainSolver code is designed to do quick transmutation simulations of samples exposed to nuclear reactor radiation (Romanov et al., 2005 ; Romanov, 2003). The software assesses the temporal evolution of nuclide density as a function of burnup, decay, and accumulation. Thermal neutron flux reduction, isotope resonance self-shielding during irradiation, and the irradiation schedule are all considered. The fundamental assumption used in transmutation calculations is that changes in the irradiated material structure have no significant influence on the nuclear reactor characteristics (Romanov et al., 2005).

In this study, we used the "ChainSolver" transmutation calculation code (Romanov, 2003), in order to simulate the neutron transmutation of Sn in a nuclear power reactor where the thermal, resonance and fast neutron flux at the irradiation position is of the order of:

$\Phi_{th} = 1.51 \times 10^{15}$; $\Phi_r = 1.32 \times 10^{14}$ and $\Phi_f = 1.34 \times 10^{15}$n cm^{-2} s^{-1}, respectively.

This program is based on the resolution of the differential equations of radioactive decay. The resulting equation takes into account the disappearances and appearances of nuclei i of concentration $N_i(t)$ over a time interval dt around the time t (Romanov et al., 2005):

$$\frac{dN_i}{dt} = -N_i\mu_i + \sum_{j\neq i} k_{ji}N_j \quad (1)$$

Where N_i is the number of nuclei i and μ_i is the constant of i-th state withdrawal:

$$\mu_i = \lambda_i + \sum \sigma_g \phi_g k_g \quad (2)$$

Where: λ_i is the half-live. σ_g is the burnout cross-section. ϕ_g is the neutron flux. k_g is the self-shielding or depression blocking factors. g is the neutron energy group. K_{ji} is the constant of transformation from j-th isotope to i-th.
The program interface utilizes a visual component in Delphi that allows the display of multiple pages on a single main form.

Calculation process

Generally, the work with the program ChainSolver requires to know special and precise parameters defining the material to be studied (composition and mass) and the irradiation site such as values of flux; thermal, resonance and fast and the power of irradiation (Romanov, 2003).
The program does not require entering information about the geometry, composition and dimensions of the reactor.
In this work, for the evaluation of the nuclear transmutations of Tin (Sn) (Romanov, 2003; Romanov et al., 2005), we introduced:

- The names of the different isotopes that make up this material (Sn-116 and Sn-117g) and even the irradiation products (Sn-117m, Sn-117g and Sn-118) through which the program will produce the reaction chain on the "Chain" page with pre-existing nuclear data in the code's data library; such as effective cross-section, decay constant, etc... This program also allows us to modify these data according to the circumstances and the new data.
- The concentration of all the isotopes initially present in the material to be studied and the total mass of this material in the "Conditions" page.
- The time and power of the radiation as well as the three components of the neutron flux (thermal, resonance and fast) must be specified on the "times" page. Knowing that this program allows us to determine the temperature during the irradiation.
- Stored in the ChainSolver code, the data file provides nuclear constants and decay data for about 2800 nuclides with atomic weights ranging from 1 to 293 (nuclear charges ranging from 1 to 118) and chemical element properties.
- The file contains information on the fission product yields for thermal and rapid neutron driven fission of 22 heavy isotopes (Romanov et al., 2005).

RESULTS AND DISCUSSION

Since fast neutron fluxes are required for the production of ^{117m}Sn from irradiation of ^{117}Sn targets, irradiation positions inside SM-3 reactor offer the possibility to maximize the production yield of ^{117m}Sn within a 5 day full reactor operating power of about 100 MWth.
The calculations of ^{117m}Sn yields and specific activities were performed using ChainSolver 2.34 code (Romanov, 2003; Romanov et al., 2005), a nuclear transmutation and decay code. These calculations were carried out for the irradiation of 200 mg tin metal target with an enrichment of 92.23% ^{117}Sn and 7.54% ^{116}Sn in the neutron trap of the SM-3 reactor.
The neutron flux (n $cm^{-2}.s^{-1}$) was as follow:

- Thermal neutrons, $\Phi_{th} = 1.51 \times 10^{15}$;
- Resonance neutrons, $\Phi_r = 1.32 \times 10^{14}$;
- Fast neutrons, $\Phi_f = 1.34 \times 10^{15}$.

The neutron gas temperature was 493 K (Kuznetsov et al., 2012). The capture cross-section of the reaction ^{116}Sn (n, γ)^{117}Sn and ^{117}Sn(n, γ)^{118}Sn is 124 mb and 1070 mb, respectively (Ponsard et al., 2009). Effective cross-sections for the inelastic scattering reaction ^{117}Sn (nf, n′γ) ^{117mSn} were calculated using the reported cross-section of 222 mb (Mirzadeh et al., 1977). The inelastic scattering reaction has a threshold of 318 keV and as such, the appropriate flux to use for ^{117m}Sn production is the total flux above 318 keV.

The progression of the specific activity of ^{117m}Sn is illustrated in figure 3, depicting its production under various flux conditions: total flux, thermal flux, resonant flux, and fast flux. It is noteworthy that a significant portion, approximately 85.527%, of the specific activity originates from inelastic interactions triggered by fast neutrons.

Conversely, the impact of thermal neutrons is minimal, with a mere 0.188% contribution. This outcome can be attributed to the enriched presence of ^{117}Sn, resulting from the enrichment process, and its comparatively lower capture cross-section compared to other reactions.

It can be seen that high ^{117m}Sn specific activities are achieved in the SM-3 high-flux reactor within just 5 full power irradiation days. The ChainSolver calculations showed that the specific activity of ^{117m}Sn was about 10 Ci.g^{-1} during a 5-day irradiation period.

These specific activities remain within the acceptable range for 10 days of decay after the end of irradiation, when they reach a minimum of 6 Ci.g^{-1}, i.e. the sufficient level of specific activity (6 - 10 Ci.g^{-1}) to reduce the concentration of ^{117m}Sn in the patient's body and reduce its chemical toxicity (Ponsard et al. 2009).

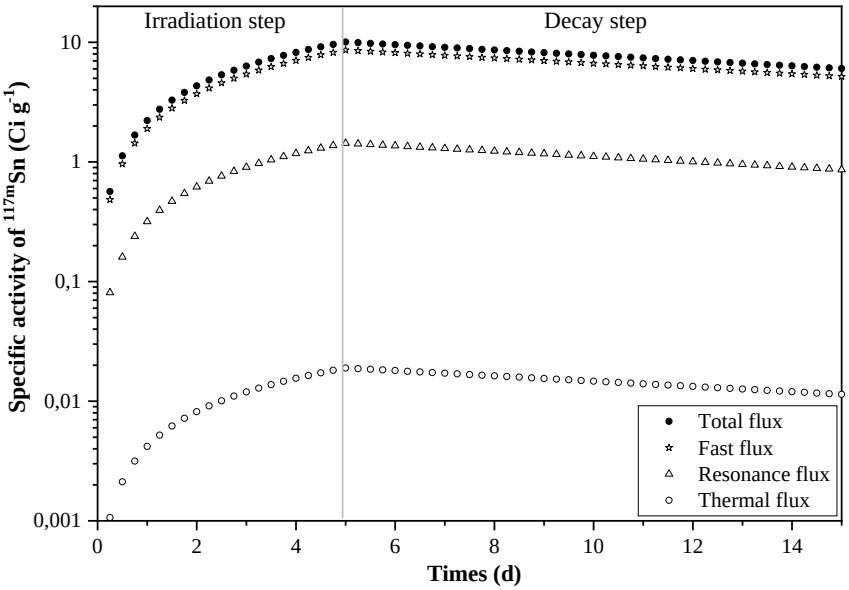

Fig.3. ^{117m}Sn production and decay timeline in a high neutron flux (n cm^{-2}.s^{-1}) values: Total 2.98×10^{15}, thermal neutrons (1.51 × 10^{15}), resonance neutrons (1.32 × 10^{14}), and fast neutrons (1.34 × 10^{15}).

4 CONCLUSION

In this paper we have studied the production of the ^{117m}Sn radioisotope in irradiation of a 92% ^{117}Sn enriched metal targets target in the high-flux SM-3 reactor for 5 effective days at a fast neutrons flux of $\Phi_f = 1.34 \times 10^{15}$ n cm^{-2} s^{-1}. The specific activity achieved is about 10 Ci.g^{-1}.

The calculation was performed using the ChainSolver 2.34 code. The high specific activity obtained in a short irradiation time is due to the high fast flux present in the SM-3 reactor, which can play a key role in the production of this important radioisotope used for medical purposes.

Further work is the experiment production of the ^{117m}Sn in the SM-3 reactor in order to compare with calculated data.

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