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# PHOTONUCLEAR PRODUCTION OF $^{99}$Mo/$^{99m}$Tc-GENERATOR WITH USING EFFECT SZILARD-CHALMERS AND ELECTROLYSIS METHOD

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The possibility of photonuclear production of $^{99}$Mo medical radioisotope using recoil nuclei of molybdenite nanoparticles from reaction $^{100}$Mo(γ,n)$^{99}$Mo was investigated. The enrichment of radioactive isotopes is carried out by the effect of Szilard-Chalmers. Molybdenite nanoparticles were irradiated by bremsstrahlung with $E_{max}$ = 39 MeV. The recoil nuclei of $^{99}$Mo are separated by electrolysis. The yield of $^{99}$Mo from the extractable phase amounted to ~3%. This technology will allow producing 0.8 GBq/g of $^{99m}$Tc with high specific activity per day. Key words: linear electron accelerator, photonuclear production, electrolysis, specific activity, recoil nuclei, molybdenite nanoparticles.

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## INTRODUCTION

The main technology for the mass production of $^{99}$Mo for obtaining $^{99m}$Tc is the $^{235}$U fission method in research nuclear reactors. $^{99m}$Tc ($T_{1/2}$=6 h) is produced from the decay of the $^{99}$Mo ($T_{1/2}$=66 h). $^{99m}$Tc produces a 140 keV gamma-ray and it is an ideal isotope for nuclear medicine [1]. $^{99}$Mo is used in the preparation of the $^{99}$Mo/$^{99m}$Tc-generator. Usually, $^{99}$Mo is produced either by neutron bombardment of $MoO_3$ or as nuclear fission of enriched uranium [2]. A significant difference between these two procedures is that $^{99}$Mo obtained from fission is "carrier free". This allows for the production of $^{99}$Mo with a specific activity of tens of thousands of Ci/g. But this technology has a number of problems: 1 – the formation of a significant amount of radioactive waste that requires immobilization; 2 – reducing the spread of nuclear weapons.

Canada, Europe, and the United States are trying to develop new technologies for the production of $^{99m}$Tc without the use of highly enriched uranium to obtain a $^{99}$Mo/$^{99m}$Tc-generator. When developing such technologies, such issues as disposal of radioactive waste, target material, radiochemistry, post-isotope separation, commercialization, etc., are necessarily considered.

Therefore, recent attention has been focused on the development of alternative methods for the production of this isotope. This is confirmed by the latest International Atomic Energy (IAEA) grant "Production Tc-99m and Tc-99m-generators beyond fission and cyclotron method" (2021), which was carried out by the authors of this article.

One of the promising methods for the production of $^{99}$Mo is the photonuclear method using electron accelerators and a photonic converter to obtain a photon beam that is directed to the Mo target. As a result of the nuclear reaction $^{100}$Mo(γ,n)$^{99}$Mo, the isotope $^{99}$Mo is produced [3, 4]. The method of photonuclear production of $^{99}$Mo is characterized by considerable advantages.

The (γ,n)-reaction does not be accompanied by a change in nuclear charge. A high concentration of $^{99}$Mo is required for the manufacturing of $^{99m}$Tc/$^{99}$Mo-generators. These generators will promote the successful using $^{99m}$Tc in nuclear medicine.

A significant disadvantage of accelerator technologies compared to reactor technologies is the low specific activity of the $^{99}$Mo isotope in the target. Accordingly, the procedure for separating the produced isotope is quite complex and costly.

The specific activity of radionuclide $^{99}$Mo can be increased in many cases by using the Szilard-Chalmers process [5]. The enrichment of radioactive isotopes is been carried out using the effect of Szilard-Chalmers. This process is based on the fact that after a photonuclear reaction, the daughter nucleus can have sufficient kinetic energy to leave an activated target. Such high-energy nuclei can accumulate in a liquid or solid medium. Since the range of molybdenum recoil nuclei is several nanometers, the effect of knocking out nuclei is significant only in the presence of a target with small geometric dimensions. The target dimensions should correspond to the same order as the range of nuclei in the target medium. An example of such a target can be a thin molybdenum foil.

The formation of recoil nuclei occurs due to two processes: photonuclear reactions on molybdenum nuclei and a cascade of elastic collisions of recoil nuclei and target nuclei. The first process includes the reactions (γ,n), (γ,2n), (γ,3n), the probability of which increases significantly with increasing photon energy. The second process leads to the knocking out of the target of molybdenum nuclei, which are part of the target itself. It should be noted that since the daughter nucleus of the photonuclear reaction on molybdenum and the molybdenum nucleus of the target have practically the same masses, due to the kinematics of the elastic scattering process, the energy transfer from the driving nucleus to the knocked-out nucleus will be significant.

These radionuclides in form of labeled compounds or conjugates of suitable biomolecules in aqueous or solid-liquid suspension are at various stages of development/clinical investigation. The use of liquid suspensions offers a significant advantage in the reusability of the target.

The aim of the present investigation is to work out photonuclear technology for the production of a high specific activity $^{99}$Mo with using of molybdenite nanoparticles and the effect of Szilard-Chalmers.

## EXPERIMENTAL TECHNIQUE

The objects of study were molybdenite nanoparticles (US Research Nanomaterials, Inc., 15…80 nm). The samples of molybdenite nanoparticles were activated by bremsstrahlung from a linear electron accelerator with E = 39 MeV and I = 4 A within 2 hours. Isotope activity was measured by a Ge(Li)-detector with a volume of 50 $cm^3$ and resolution of 3.2 at a 1332 keV line. Nuclear reaction $^{100}Mo(\gamma,n)^{99}Mo$ was used to obtain $^{99m}Tc$.

Extraction problem of technetium. An electrolysis method has been developed for the extraction of $^{99m}Tc$. The parameters of technetium electrolysis were the following: the current is ~150 mA/$cm^2$, electrolyte temperature -30°C. The increase of productivity of electrochemical deposition of the technetium on the carbon cathode is reached by means of the use of a rotating electrode. The diffusion limiting current $i_d$ for a rotating electrode:

$$i_d = 0{,}62\ F\ D^{2/3}\omega^{1/2}\upsilon^{-1/6}\ C_o,$$

where $i_d$ – an angular speed of rotation of an electrode; $\upsilon$ – kinematic velocity; $C_o$ – concentration of a required component in volume; D – diffusion factor; and F – factor Fano; $\omega$ – an angular speed of rotation of an electrode. Therefore changing the speed of rotation of an electrode, we can change the limiting current density.

The electrolysis of irradiated molybdenite was carried out in hydrogen peroxide presence, in the exercise intensive stirring of an electrolyte, and of temperature variations of the solution. To isolate $^{99m}Tc$, electrolysis was carried out in a 2N sodium hydroxide solution. The possibility of separating $^{99m}Tc$ from a NaOH solution is shown. The part deposited on the cathode is about 3%.

An electrolysis method gives the possibility to produce not only of $^{99m}Tc$, and of $^{99m}Tc/^{99}Mo$-generators.

## RESULTS AND DISCUSSION

The energy of the recoil nucleus depends on the pulse gamma-ray, neutron pulse emitted, and the angle between the directions of doing these pulses. Where $E_r$ – recoil energy molybdenum atom, the M is the mass of molybdenum atom; m – a mass of the neutron; the Q – energy nuclear reaction; c – the speed of light; θ – the angle between the directions of the neutron is-started-up and the incident photon; $E_\gamma$ – energy gamma-quantum and has the following form:

$$E_r = \frac{ME_\gamma^2}{2(M+m)^2c^2} + \frac{m^2}{2(M+m)^2}\left\{(E_\gamma+Q)2(M+m) - \frac{E_\gamma^2}{c^2}\right\} -$$

$$- \frac{E_\gamma Mm\cos\theta}{2c(M+m)^2}\left\{\frac{(E_\gamma+Q)2(M+m) - \frac{E_\gamma^2}{c^2}}{Mm}\right\}^{1/2},$$

The estimate of medial energy of neutrons for gamma-radiation with the maximum energy of 39 MeV of reaction $^{100}Mo(\gamma,n)^{99}Mo$ is equal to 450 keV (Fig. 1). Therefore medial energy of recoil nuclei of $^{99}Mo$ is equal to 4.5 keV. Recoil nuclei $^{99}Mo$ can leave nanoparticles of $MoS_2$ from the depth of 4.5 nm (Fig. 2).

Isotope activity is measured by a Ge(Li)-detector (Fig. 3). It is known that the diffusion coefficients of molybdenum and technetium in molybdenite are different [6]. This feature was used for the separation of technetium-99m. Thus chemical reagents with affinity to technetium [7] were used.

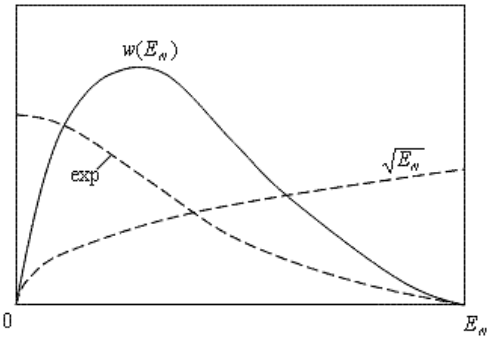


*Fig. 1. The energy distribution of photo nucleons predicted by the statistical theory [3]*

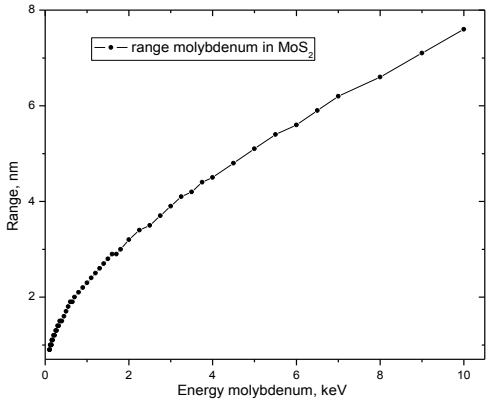


*Fig. 2. $^{99}Mo$ ranges in natural molybdenum disulfide*

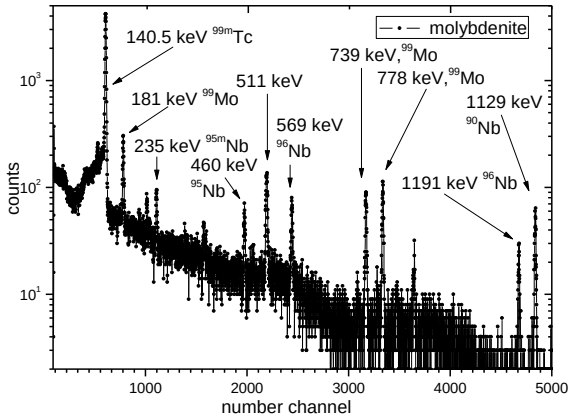


*Fig. 3. The spectrum of irradiate nano-$MoS_2$*

The wettability of molybdenite increases with the reduction of the size of nanoparticles. Electrolysis oxidation oxidizes the molybdenite surface slightly and oxygen becomes attached to the surface of the molybdenite layer. With prolonged retention after electrolysis oxidation, molybdenum oxide can dissolve as molybdenum oxide ions, such as $MoO_2^{-4}$. As a result, the molybdenite surface becomes hydrophobic with prolonged retention. On the other hand for molybdenite, with electrolysis oxidation, molybdenum oxide is produced but the surface keeps weak hydrophobic since molybdenum oxide is soluble. With retention in electrolyte after electrolysis, the surface keeps weak hydrophobic since a still small amount of molybdenum oxide stayed on the surface (Fig. 4).

The crystalline structure of molybdenite with hydrophobic faces and hydrophilic edges is shown (Fig. 5).

The method of electrolysis works with a great low specific concentration of $^{99m}$Tc [8]. The specific concentration of $^{99m}$Tc is 100 less for this case than at the use of extraction with the help of methyl ethyl ketone (Fig. 6).

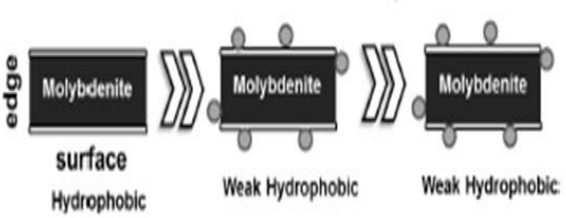


*Fig. 4. Properties of $MoS_2$ during electrolysis*

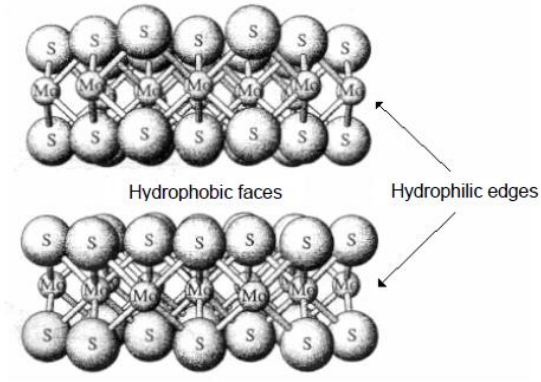


*Fig. 5. Structure of molybdenum disulfide*

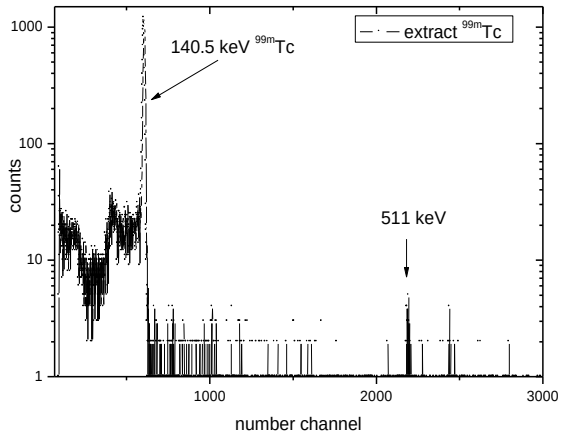


*Fig. 6. The spectrum of extract $^{99m}$Tc*

## CONCLUSIONS

The possibility of photonuclear production of $^{99}$Mo by using recoil nuclei of molybdenite nanoparticles from reaction $^{100}$Mo(γ,n)$^{99}$Mo has been found.

The use of thermally stable molybdenite allows for solving the problem of thermal loads when using high-power electron beams.

The electrolysis of molybdenite nanoparticles allows the separation of technetium-99m at low concentrations.

The use of $MoS_2$ nanoparticles with size 13…80 nm and of bremsstrahlung with $E_{max}$=25 MeV on a 10 kW electron accelerator will allow producing 0.8 GBq/g of $^{99m}$Tc with high specific activity per day. It simplifies the use of $^{99m}$Tc in medical institutions.

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## ФОТОЯДЕРНЕ ВИРОБНИЦТВО $^{99}$Mo/$^{99m}$Tc-ГЕНЕРАТОРА З ВИКОРИСТАННЯМ РЕАКЦІЇ СЦІЛАРДА-ЧАЛМЕРСА ТА МЕТОДУ ЕЛЕКТРОЛІЗУ

***М.П. Дикий, Ю.В. Ляшко, О.П. Медведєва, Д.В. Медведєв, В.Л. Уваров***

Досліджена можливість фотоядерного виробництва медичного ізотопу $^{99}$Mo з використанням ядер віддачі наночастинок молібденіту з реакції $^{100}$Mo(γ,n)$^{99}$Mo. Збагачення радіоактивними ізотопами здійснюється за реакції Сціларда-Чалмерса. Наночастинки молібденіту були опроміненні гальмівним випромінюванням на ЛПЕ з $E_{max}$=39 МеВ. Ядра віддачі $^{99}$Mo були виділені методом електролізу. Вихід $^{99}$Mo з фази екстракції становив ~3%. Ця технологія дозволить виробляти ~0,8 ГБк/г $^{99m}$Tc з високою питомою активністю за добу.