

APPLICATION OF NUCLEAR METHODS

USE OF MOLYBDENITE NANOPARTICLES FOR PHOTONUCLEAR PRODUCTION OF TECHNETIUM-99m

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The possibility production of ⁹⁹Mo radioisotope with using recoil nuclei of molybdenite nanoparticles from reaction ¹⁰⁰Mo(γ,n)⁹⁹Mo was investigated. The use of thermally stable molybdenite allows solving the problem of thermal loads of high-power electron beams. The enrichment of radioactive isotopes carried out by the effect of Szilard-Chalmers. Molybdenite nanoparticles were irradiated by bremsstrahlung with E_{max}=39 MeV. The recoil nuclei of ⁹⁹Mo separated by electrolysis at low concentration. The yield of ⁹⁹Mo from extractable phase amounted 3% for size nanoparticles 350 nm.

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INTRODUCTION

^{99m}Tc (T_{1/2}=6 h) is being produced from the decay of the ⁹⁹Mo (T_{1/2}=66 h). ^{99m}Tc produces a 140 keV gamma ray and it is an ideal isotope for nuclear medicine imaging [1]. ⁹⁹Mo is using in the preparation of ⁹⁹Mo-^{99m}Tc generator. Usually ⁹⁹Mo is being produced either by neutron bombardment of MoO₃ or as nuclear fission of enriched uranium [1]. A significant difference between of these two procedures is that ⁹⁹Mo obtained from fission is “carrier free”. This allows to producing of ⁹⁹Mo with a specific activity of tens of thousands of Ci/g. The method of photonuclear production of ⁹⁹Mo characterized by considerable advantages [2 - 4]. The (γ,n)-reaction does not be accompanied by a change in nuclear charge. Therefore, the enrichment of radioactive isotopes is been carried out using the effect of Szilard-Chalmers. The high concentration of ⁹⁹Mo is required for manufacturing of ^{99m}Tc-⁹⁹Mo generators. These generators will promote successful using ^{99m}Tc in nuclear medicine.

A feature of this study is the use of electrolysis to separate of ^{99m}Tc. The electrolysis method allows the separation of ^{99m}Tc from solutions with a lower specific concentration of ⁹⁹Mo than, for example, when the extraction of ^{99m}Tc using methyl ethyl ketone [2 - 4].

Currently molybdenum disulfide intensively investigating d. This use molybdenum disulfide in optoelectric

device, to molecular and biomolecular sensors, for hydrodesulfurization catalyst, catalyst for hydrogen evolution reaction, catalyst for water splitting and chemical reaction, various bioapplications [5, 6]. Another application is photothermal therapy, which takes advantage of its good dispersibility and superior absorbance of near infrared. A molybdenum disulfide dispersion shows 7.8 times higher absorbance than graphene oxide, and its extinction coefficients at 800 nm is 29.2 l·g⁻¹·cm⁻¹, which is higher than gold nanorods. Visualisation of certain organs or cells is one of the most important functions for diagnosis and treatment. Based on the large atomic number of Mo, MoS₂ flakes as image contrast agent are using for computed tomogram [5].

The purpose of the present investigation is the production of a high specific activity ⁹⁹Mo with using of molybdenite nanoparticles and the effect of Szilard-Chalmers.

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. The energy of the recoil nucleus E_r expressed by the following expression:

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

where E_r – recoil energy molybdenum atom; the M is the mass of molybdenum atom; m – 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

The estimate of medial energy of neutrons for gamma radiation with the maximum energy of 39 MeV of reaction ¹⁰⁰Mo(γ,n)⁹⁹Mo is equal to 450 keV (Fig. 1) [7]. Therefore, medial energy of recoil nuclei of ⁹⁹Mo is equal to 4.5 keV. Recoil nuclei ⁹⁹Mo can leave nanoparticles of MoS₂ from the depth of 4.5 nm (Fig. 2).

Nuclear reaction ¹⁰⁰Mo(γ,n)⁹⁹Mo was used to obtain ^{99m}Tc. Samples of molybdenite nanoparticles were activated by bremsstrahlung from a linear electron accelerator with E = 39 MeV and I = 4 μA within 2 hours. The size of molybdenite nanoparticles approximately was 300...400 nm.

Isotope activity is measured by a Ge(Li)-detector with the energy resolution of 3.2 at 1333 keV line (Fig. 3). It is known that the diffusion coefficients of molybdenum and technetium in molybdenite are different [8]. This feature is using for separation of ^{99m}Tc.

Thus, chemical reagents with affinity to technetium [9] are using.

Extraction problem of technetium. The parameters of technetium electrolysis were the following: the current is $\sim 150 \text{ mA/cm}^2$, electrolyte temperature – 30°C . Electrolysis carried out in a quartz cell. The increase of productivity of electrochemical deposition of the technetium on the carbon cathode reached by means of the use of a rotating electrode. The diffusion limiting current i_d for a rotating electrode:

$$i_d = 0.62 F D^{2/3} \omega^{1/2} \nu^{-1/6} C_o,$$

where ω – an angular speed of rotation of an electrode; ν – kinematic viscosity; C_o – concentration of a required component in volume and D – diffusion factor. Therefore changing the speed of rotation of an electrode, we can change the limiting current density.

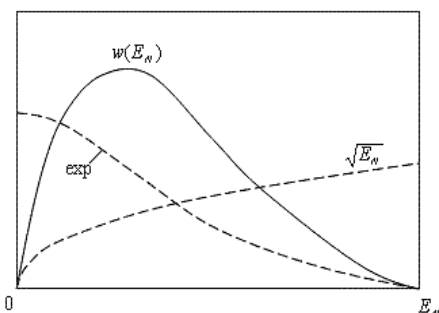


Fig. 1. The energy distribution of photo nucleons predicted by the statistical theory [3]

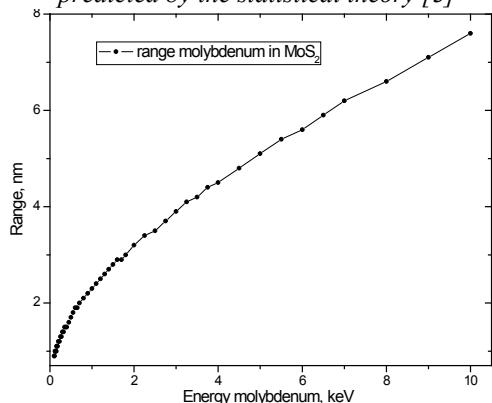


Fig. 2. ^{99}Mo ranges in natural molybdenum disulfide

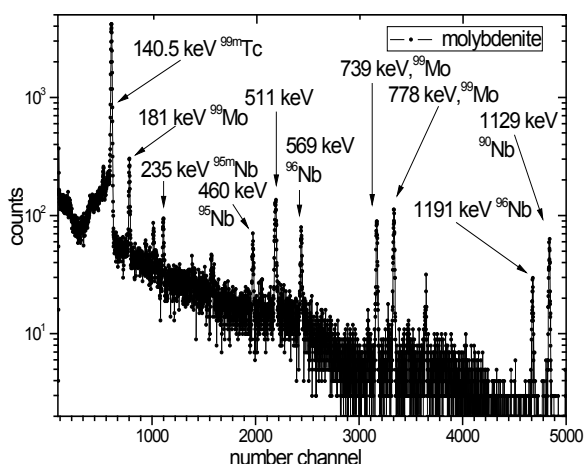


Fig. 3. The spectrum of molybdenite nanoparticles after irradiate by bremsstrahlung with $E_{max}=39 \text{ MeV}$

Wettability of molybdenite increases with the reduction of the size nanoparticles. Electrolysis oxidation

oxidizes the molybdenite surface slightly and oxygen becomes attached to the surface of the molybdenite layer (Fig. 4). 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 [10]. Molybdenite is hydrophobic. On the other hand for molybdenite, with electrolysis oxidation, molybdenum oxide produced but surface keeps weak hydrophobic since molybdenum oxide is soluble. With retention in electrolyte after electrolysis, surface keeps weak hydrophobic since a still small amount of molybdenum oxide stayed on the surface.

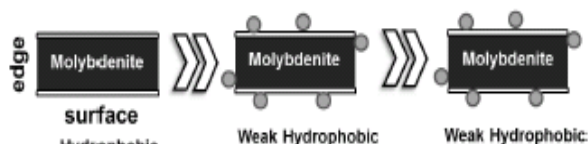


Fig. 4. Properties of molybdenum disulfide during electrolysis

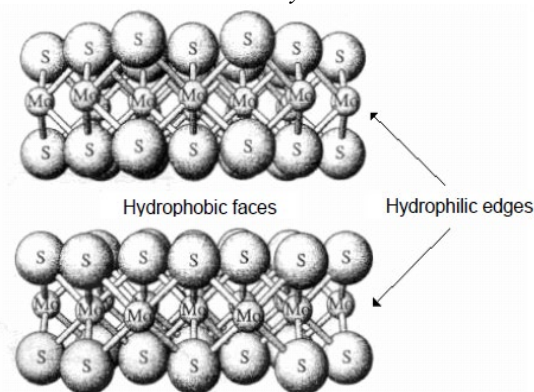


Fig. 5. Structure of molybdenum disulfide

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

The method of electrolysis works with a great low specific concentration of ^{99m}Tc . The specific concentration of ^{99m}Tc is in 100 less for this case than at use of extraction with the help of methyl-ethyl ketone (Fig. 6) [11, 12].

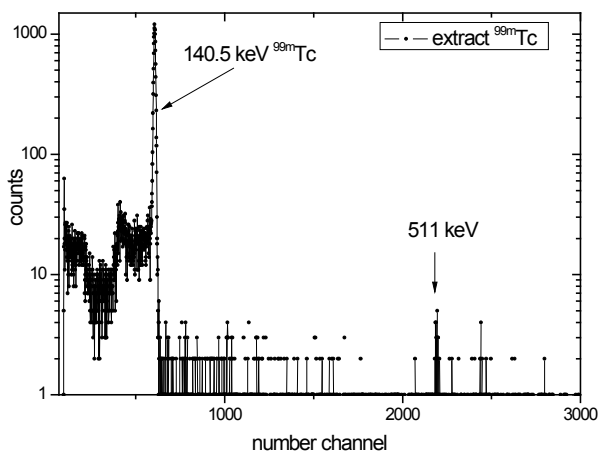


Fig. 6. The spectrum of extract ^{99m}Tc

Protons with energy 30 MeV used for production of ^{99}Mo from nanoparticles of MoS_2 [13, 14].

Due to unexpected outages or planned and unplanned reactor shutdown, significant ^{99m}Tc shortages appeared as a problem since 2008.

We note the tendency for the production of the ^{99}Mo isotope at charged particle accelerators. The new methods have a number of advantages compared with the production of ^{99}Mo at the reactors. One can note the almost absence of radioactive waste, as well as the more economic characteristics of the production of the ^{99}Mo isotope [15].

However, there are problems of heat removal from the target. For this reason, sapphire, a ceramic material with one of the highest thermal conductivities ($60 \text{ Wm}^{-1}\cdot\text{K}^{-1}$), and synthetic diamond (CVD), a material with an extremely high thermal conductivity (up to $2000 \text{ Wm}^{-1}\cdot\text{K}^{-1}$), are proposed as the components of the target backing plate. In order to minimize the thermal resistance related to the ceramic part (again, to optimize the heat exchange of the target), its thickness was minimized [16].

Note that US-based NorthStar Medical Radioisotopes and Belgium's Ion Beam Applications (IBA) have signed a contract under which IBA would supply up to eight Rhodotron TT300 HE electron beam accelerators to NorthStar, which has issued purchase orders for the first two units. Six more will be delivered in coming years. This accelerator have very high power and energy resolution: 125 kW, 40 MeV, ~5% energy spread.

CONCLUSIONS

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

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

The electrolysis of molybdenite nanoparticles allows separating technetium-99m at low concentration.

The use MoS_2 nanoparticles with size 15 nm and of bremsstrahlung with $E_{\text{max}}=25 \text{ MeV}$ on 10 kW electron accelerator will allow producing 0.8 GBq/g per day of $^{99\text{m}}\text{Tc}$ with a high specific activity. It simplifies the use of $^{99\text{m}}\text{Tc}$ in medical institutions.

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ИСПОЛЬЗОВАНИЕ НАНОЧАСТИЦ МОЛИБДЕНИТА ДЛЯ ФОТОЯДЕРНОГО ПРОИЗВОДСТВА ТЕХНЕЦИЯ-99m

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Исследована возможность получения радиоиотопа ^{99}Mo с использованием ядер отдачи наночастиц молибденита из реакции $^{100}\text{Mo}(\gamma, n)^{99}\text{Mo}$. Использование термостойкого молибденита позволяет решить проблему тепловых нагрузок мощных электронных пучков. Обогащение радиоактивных изотопов осуществлялось с помощью эффекта Сциларда-Чалмерса. Наночастицы молибденита облучались тормозным излучением с $E_{\text{max}}=39$ МэВ. Ядра отдачи ^{99}Mo были отделены электролизом при низкой концентрации. Выход ^{99}Mo из экстрагируемой фазы составил $\sim 3\%$ для наночастиц размером 350 нм.

ВИКОРИСТАННЯ НАНОЧАСТИНОК МОЛІБДЕНІТУ ДЛЯ ФОТОЯДЕРНОГО ВИРОБНИЦТВА ТЕХНЕЦІЯ-99m

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Досліджено можливість отримання радіоіотопа ^{99}Mo з використанням ядер віддачі наночастинок молибденіту з реакції $^{100}\text{Mo}(\gamma, n)^{99}\text{Mo}$. Використання термостійкого молибденіту дозволяє вирішити проблему теплових навантажень потужних електронних пучків. Збагачення радіоактивних ізотопів здійснювалося за допомогою ефекту Сциларда-Чалмерса. Наночастки молибденіту опромінювалися гальмівним випромінюванням з $E_{\text{max}}=39$ МеВ. Ядра віддачі ^{99}Mo були відокремлені електролізом при низькій концентрації. Вихід ^{99}Mo з екстрагрованої фази склав $\sim 3\%$ для наночастинок розміром 350 нм.