THE USE OF MOLYBDENUM OXIDE NANOPARTICLES FOR PRODUCTION OF FREE ISOTOPE Mo-99

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The possibility of photonuclear production of ^{99}Mo medical radioisotopes using recoil nuclei of nanoparticles MoO_3 from reaction ^{100}Mo(γ,n)^{99}Mo was investigated. (γ,n)-reaction does not be accompanied by change in nuclear charge. Therefore, the enrichment of radioactive isotopes is being carried out using methods based on the effect of Szilard-Chalmers. The highest concentration of ^{99}Mo is required for manufacturing of ^{99m}Tc-^{99}Mo generators. These generators will promote successful using ^{99m}Tc in nuclear medicine. MoO_3 nanoparticles of size 13…80 nm were placed in isopropyl alcohol and ethylene glycol. The colloidal solution of MoO_3 is achieved by sonication. The colloidal solution of nanoparticles of MoO_3 was irradiated by bremsstrahlung with E_{max} = 12.5 MeV. The recoil nuclei of ^{99}Mo were separated by means of diantipyrylmethane in sulfuric acid solution. Yield of ^{99}Mo from extractable phase amounted ~ 4%, in consequence of the high density of MoO_3 nanoparticles in a colloidal solution.

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INTRODUCTION

^{99m}Tc ($T_{1/2}$=6 h) is being produced from the decay of the ^{99}Mo ($T_{1/2}$=66 h). ^{99m}Tc produces a single 140 keV gamma ray and it is an ideal isotope for nuclear medicine imaging [1]. ^{99}Mo is used in the preparation of ^{99}Mo-^{99m}Tc generator. Usually ^{99}Mo is being produced either by neutron bombardment of MoO_3 or as nuclear fission of enriched uranium [1]. A significant difference of these two procedures is that ^{99}Mo obtained from fission is "carrier free". This allows to produce of ^{99}Mo with a specific activity of tens thousands of Ci/g.

Production of ^{99m}Tc and ^{99}Mo by charging particle bombardment is also possible [2]. The method of photonuclear production of ^{99}Mo is being characterized by considerable advantages especially [3].

In generator systems the ^{99}Mo is normally adsorbed onto an alumina column and the less strongly bound $^{99m}TcO_4^-$ is eluted with isotonic saline solutions. However, the limited adsorption capacity of alumina for Mo(VI) requires the use of very high specific activities of ^{99}Mo (~$3\cdot10^4$ Ci/g). To avoid this drawback, alternative methods of generator preparation with using low specific activities have been proposed [4].

Nevertheless, efforts which have been directed at obtaining of a high specific activity of producing isotopes were undertaken for an appreciable time interval [5]. These preparations of radioactive elements with high specific activity are necessary for nuclear medicine. (γ,n)-reaction does not be accompanied by change in nuclear charge. Therefore, the enrichment of radioactive isotopes is being carried out using methods based on the effect of Szilard-Chalmers. The highest concentration of ^{99}Mo is required for manufacturing of ^{99m}Tc-^{99}Mo generators. These generators will promote successful using ^{99m}Tc in nuclear medicine.

The purpose of the present article is the production of a high specific activity ^{99}Mo on the basis of nanoparticles of molybdenum oxide and using of the effect of Szilard-Chalmers.

RESULTS AND DISCUSSION

Molybdenum oxide nanoparticles (US Research Nanomaterials, Inc, USA, Orthorhombic crystal) of size 13…80 nm and weight of 80 mg were placed in isopropyl alcohol and ethylene glycol. The colloidal solution of MoO_3 was obtained by treating the nanoparticle suspension sonicated. The colloidal solution of nanoparticles of MoO_3 were irradiated by bremsstrahlung with E_{max} = 12.5 MeV.

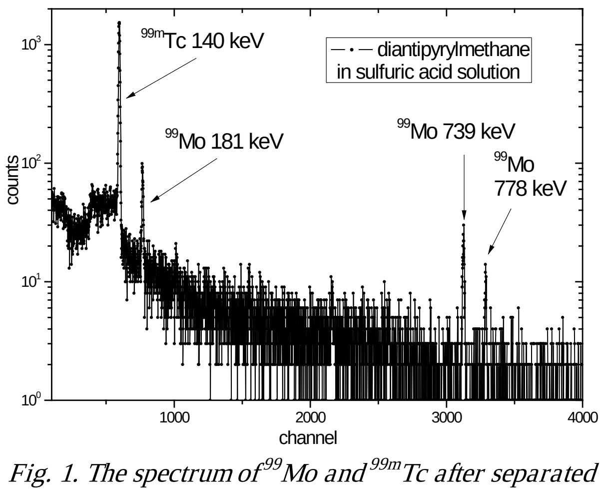

Fig. 1. The spectrum of ^{99}Mo and ^{99m}Tc after separated from colloidal solution of MoO_3 nanoparticles

After activation of samples and standards the activity of radioisotopes obtained in reactions ^{100}Mo(γ,n)^{99}Mo has been measured by Ge(Li)-detector with volume 50 cm^3 and with energy resolution 3.2 keV in the area of 1332 keV. The recoil nuclei of ^{99}Mo were separated by means of diantipyrylmethane in sulfuric acid solution (Fig. 1).

The energy spectrum of neutrons depends on the incident bremsstrahlung, target material, and cross section of the photonuclear reaction. The evaporation model for compound nuclei predicts that the emitted neutron energy distribution approaches the form of a Maxwell distribution [7 - 9]:

$$w(E_n) = const\,\frac{E_n}{\theta^2}\exp(-\frac{E_n}{\theta}),$$

where $\theta = [(E_\gamma - B_n)/a]^{1/2}$, B_n – separation energy of neutron, E_γ – bremsstrahlung energy. The constant a define of speed of ascending of density of levels of a nucleus at increasing of energy. The experimental estimate of this constant is a ≈ A/15 MeV^{-1}.

The estimate of the medial energy of neutrons for a gamma radiation with the maximum energy of 12.5 MeV the reaction $^{100}Mo(\gamma,n)^{99}Mo$ is equal 400 keV [6 - 9]. Therefore the medial energy of recoil nuclei of ^{99}Mo is equal 4 keV. Recoil nuclei ^{99}Mo can leave nanoparticles of MoO_3 from a depth of 4.8 mm (Fig. 2). For medial radius of MoO_3 nanoparticles 23 nm the part of recoil nuclei, which can go out into a solution, is 10.4%. Yield of ^{99}Mo out of extractable phase amounted ~4%, due to the high density of molybdenum nanoparticles in a colloidal solution.

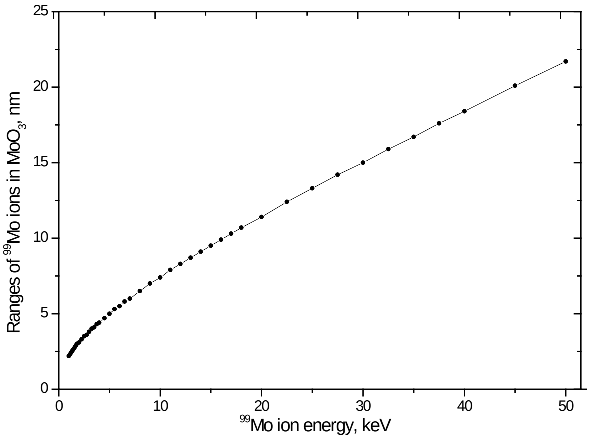

Fig. 2. ^{99}Mo ranges in natural molybdenum oxide

It is necessary to notice that MoO_3 nanoparticles in the process of an irradiating have been covered by a stratum of ethylene glycol, at least, 0.5 nm. Therefore the considerable part of the recoil nucleus of atoms of the natural molybdenum after scattering of bremsstrahlung cannot go out of MoO_3 nanoparticles [10]. It will promote the production of ^{99}Mo with a high specific activity.

Irradiating 1 g Mo target for a day using 10 kW electron LINAC would result in 1.68 GBq/g [10].

However, the problem of heat generation in the target and the problem of obtaining of the maximum specific activity in extracts of ^{99}Mo limited the capacity of electronic accelerator for the above parameters up to 1 GBq/g [11]. Therefore, the use MoO_3 nanoparticles with size 15 nm and of gamma radiation with E_{max}=25 MeV on 10 kW electron accelerator will allow to produce 0.8 GBq/g per day of ^{99}Mo with high specific activity, which is necessary for manufacturing generators ^{99m}Tc-^{99}Mo.

CONCLUSIONS

The possibility of photonuclear production of ^{99}Mo by using recoil nuclei of MoO_3 nanoparticles that obtained by reaction $^{100}Mo(\gamma,n)^{99}Mo$ has been found. The colloidal solution of MoO_3 in isopropyl alcohol or ethylene glycol was used which was obtained by means of sonicating of nanoparticle suspension. The recoil nuclei of ^{99}Mo after irradiation of the solution by bremsstrahlung were separated by means of diantipyrylmethane in sulfuric acid solution. As a result, there is prepared with high specific activity of ^{99}Mo that can be used for making generators ^{99m}Tc-^{99}Mo on the basis of Al_2O_3.

The use MoO_3 nanoparticles with size 15 nm and of bremsstrahlung with E_{max}=25 MeV on 10 kW electron accelerator will allow to produce 0.8 GBq/g per day of ^{99}Mo with high specific activity, which is necessary for manufacturing generators ^{99m}Tc–^{99}Mo. It simplifies use of ^{99m}Tc in medical institutions.

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ИСПОЛЬЗОВАНИЕ НАНОЧАСТИЦ ОКСИДА МОЛИБДЕНА ДЛЯ ПРОИЗВОДСТВА СВОБОДНОГО ИЗОТОПА Mo-99

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Была исследована возможность фотоядерного производства медицинского радиоизотопа ^{99}Mo при использовании ядер отдачи из наночастиц MoO_3 по реакции ^{100}Mo(γ,n)^{99}Mo. (γ,n)-реакции не сопровождаются изменением заряда ядра, поэтому обогащение радиоактивных изотопов проводилось с использованием методов, основанных на эффекте Сциларда-Чалмерса. Высокая концентрация ^{99}Mo требуется для изготовления генераторов ^{99м}Tc-^{99}Mo. Эти генераторы позволят успешно использовать ^{99м}Tc в ядерной медицине. MoO_3 – наночастицы размером 13…80 нм были помещены в изопропиловый спирт и этиленгликоль. Коллоидный раствор MoO_3 достигался обработкой ультразвуком. Коллоидный раствор наночастиц MoO_3 был облучен тормозным излучением с $E_{макс}$ = 12,5 МэВ. Ядра отдачи ^{99}Mo были выделены из сернокислого раствора с использованием диантипирилметана. Выход ^{99}Mo в экстрагируемую фазу составил ~ 4%, что обусловлено высокой плотностью наночастиц молибдена в коллоидном растворе.

ВИКОРИСТАННЯ НАНОЧАСТИНОК ОКСИДУ МОЛІБДЕНУ ДЛЯ ВИРОБНИЦТВА ВІЛЬНОГО ІЗОТОПУ Mo-99

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Була досліджена можливість фотоядерного виробництва медичного радіоізотопу ^{99}Mo при використанні ядер віддачі з наночастинок MoO_3 з реакції ^{100}Mo(γ,n)^{99}Mo. (γ,n)-реакції не супроводжуються зміною заряду ядра, тому збагачення радіоактивних ізотопів проводилося з використанням методів, заснованих на ефекті Сциларда-Чалмерса. Висока концентрація ^{99}Mo потрібно для виготовлення генераторів ^{99м}Tc-^{99}Mo. Ці генератори дозволять успішно використовувати ^{99м}Tc в ядерній медицині. MoO_3 – наночастинки розміром 13…80 нм були поміщені в ізопропіловий спирт і етиленгліколь. Колоїдний розчин MoO_3 досягався обробкою ультразвуком. Колоїдний розчин наночастинок MoO_3 був опромінений гальмівним випромінюванням з $E_{макс}$ = 12,5 МеВ. Ядра віддачі ^{99}Mo були виділені з сірчанокислого розчину з використанням діантіпірілметана. Вихід ^{99}Mo в екстрагуєму фазу склав ~ 4%, що зумовлено високою щільністю наночастинок молібдену в колоїдному розчині.