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Investigation of Mo-99 radioisotope production by d-Li neutron source

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ABSTRACT

A plan of an advanced fusion neutron source (A-FNS) by using d-Li reaction is in progress at Rokkasho in Japan. We investigate multipurpose usages of the A-FNS in addition to fusion material irradiation test. Production of medical isotope ^{99}Mo is considered as one of the usages. We conducted a conceptual study on a module for radioisotope production which was composed of a neutron spectrum shifter and a neutron reflector. We examined impacts of materials of the shifter and reflector on amounts of the ^{99}Mo production, and their thicknesses. It was concluded that beryllium is the most suitable material both for the shifter and the reflector from the viewpoint of the ^{99}Mo production. It was shown that we produced an enough amount of the ^{99}Mo for the demand in Japan. We can apply natural molybdenum for this purpose. It was also shown that we could use a part of irradiation capsules in high flux test module, which was for the fusion material irradiation test originally, by using isotopically enriched ^{100}Mo to meet that.

1. Introduction

An advanced fusion neutron source (A-FNS) is under planning at Rokkasho in Japan [1]. We work on an early realization of neutron source operations at the A-FNS by utilizations of resources in the International Fusion Materials Irradiation Facility Engineering Validation and Engineering Design Activities (IFMIF/EVEDA) [2,3]. In Europe, a similar project of a neutron irradiation facility which is called as International Fusion Material Irradiation Facility - DEMO Oriented NEutron Source (IFMIF-DONES) is also in progress [4]. The A-FNS has an accelerator providing a 40 MeV deuteron beam of 125 mA and a Li target. It produces a large amount of neutrons due to the Li(d,xn) stripping reaction, and has the neutron flux of about 6.1×10^{14} n/cm^2/s at a back plate of the Li target with a broad energy peak around 14 MeV. The main purpose of the A-FNS is the fusion material irradiation test. In order to apply the A-FNS with multipurpose usages, we have proposed a production of medical isotope ^{99}Mo. The ^{99}Mo is a parent nucleus of ^{99m}Tc, which is used for the nuclear medicine diagnosis. The ^{99m}Tc is produced by β^- decay of the ^{99}Mo with half-life of 65.94 h. Then, the ^{99}Mo is one of the most important isotopes in nuclear medicine. The ^{99}Mo used in Japan are supplied only from nuclear reactors in foreign countries [5]. The domestic production of the ^{99}Mo is required because of the avoidance of the risk of the supply disruption because of transportation troubles, aging of the facility, etc. The demand of the ^{99}Mo in Japan is about 84 TBq/week before shipment from the producer's facility in Japan on assumption of domestic production without any loss of activity during the transportation from abroad [6]. A new production method is proposed by using the $^{100}Mo(n,2n)^{99}Mo$ reaction with accelerator [7]. We show that the A-FNS has an enough potential to produce the ^{99}Mo over its demand in Japan by applying the $^{100}Mo(n,2n)^{99}Mo$ reaction [8]. We have investigated the usefulness of the $^{98}Mo(n,\gamma)^{99}Mo$ reaction with a usage of natural molybdenum (^{nat}Mo) from the viewpoint of cost reduction. In this study, we conduct a conceptual study on a module for radioisotope production (RI module) newly designed for the A-FNS which is composed of a neutron spectrum shifter and a neutron reflector through analyses of the production of the ^{99}Mo. We calculate the amount of the ^{99}Mo production with the RI module and investigate the possibility of the ^{99}Mo production to fulfill the demand in Japan.

2. Calculation

2.1. Production reaction of ^{99}Mo

Two production paths of the ^{99}Mo by neutron irradiations can be considered as follows: the $^{98}Mo(n,\gamma)^{99}Mo$ and $^{100}Mo(n,2n)^{99}Mo$ reactions. The cross sections of these reactions are shown in Fig. 1 [9]. The $^{100}Mo(n,2n)^{99}Mo$ reaction is induced by fast neutrons above about 8.4 MeV. The $^{98}Mo(n,\gamma)^{99}Mo$ reaction is induced by lower energy neutrons and has large resonances in about 12 eV and 0.4–10 keV energy region.

2.2. Calculation model of A-FNS and RI module

In this calculation, we modify the IFMIF test cell design model for

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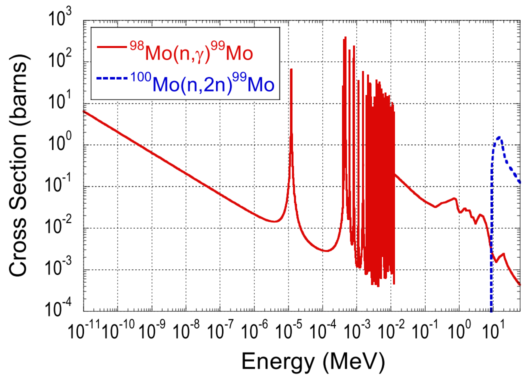

Fig. 1. Cross sections of ^{99}Mo production.

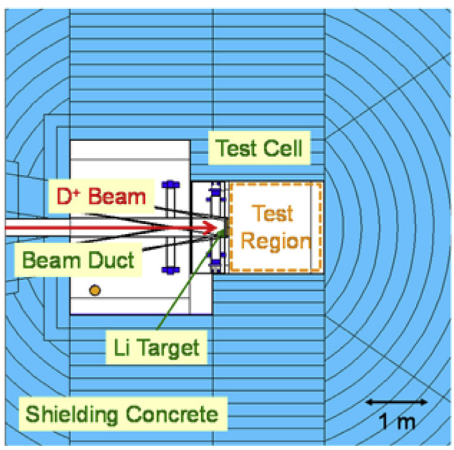

Fig. 2. Calculation model on test cell in A-FNS.

the A-FNS [10]. The calculation model is shown in Fig. 2. The beam duct is reduced to one from two in the IFMIF test cell design model, because one d^+ beam enters vertically on the Li target in the case of the A-FNS. The beam footprint is 20 cm in width and 5 cm in height, which is the same as the IFMIF design. The Li target assembly and beam duct are composed of the Eurofer steel and stainless steel SS316L. The test cell is surrounded by the shielding concrete and has the test region behind the Li target.

Fig. 3 shows a calculation model of the RI module. This module consists of a $^{nat}MoO_3$ sample [11], a neutron spectrum shifter, and a neutron reflector. The density of $^{nat}MoO_3$ is 4.69 g/cm^3. Natural molybdenum has seven stable isotopes: ^{92}Mo (14.53%), ^{94}Mo (9.15%), ^{95}Mo (15.84%), ^{96}Mo (16.67%), ^{97}Mo (9.60%), ^{98}Mo (24.39%) and ^{100}Mo (9.82%). The surface area of the sample is the same as that of the beam footprint in which neutrons are produced, except for a case as described in Section 3.3. The thickness of the sample is varied with the

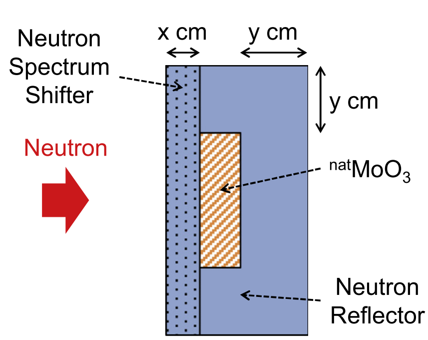

Fig. 3. Calculation model of RI module. x cm is thickness of neutron spectrum shifter and y cm is thickness of reflector.

amount of the sample. The spectrum shifter moderates fast neutrons to lower energy neutrons. It increases the production of the ^{99}Mo due to the ^{98}Mo(n,γ)^{99}Mo reaction, while it decreases that due to the ^{100}Mo (n,2n)^{99}Mo reaction. The neutron reflector increases both of fast and low energy neutrons in the samples. The shifter and reflector consist of the same material for simplicity. We apply those with graphite, Be, Pb, H_2O, and D_2O. For comparison, we also investigate the case without the shifter and reflector. The RI module is placed in the test region in the test cell of the A-FNS as shown in Figs 4(A)-(C). High Flux Test Module (HFTM) is a module for the fusion reactor material irradiation test and Middle Flux Test Module (MFTM) is a module for creep fatigue test and tritium release test in the IFMIF model. Furthermore, we investigate the use of a part of irradiation capsules in the HFTM for the RI production. Fig. 5 shows a layout of the test modules in the case using the HFTM capsules and its size. Low Flux Test Module (LFTM) is a flexible test module for various experiments and is designed as several vacant containers in the IFMIF model. The $^{nat}MoO_3$ samples are put in the capsule. We also perform another calculation using isotopically enriched $^{100}MoO_3$ samples in order to utilize fast neutrons directly because there are no Be shifter and reflector which produce lower energy neutrons in the HFTM.

2.3. Calculation code and nuclear data

The calculations are carried out by using the McDeLicious-11 Monte Carlo code [12] which has been developed as an extension of MCNP5 code [13] in order to simulate the d-Li reaction. General purpose file FENDL-3.1b [9] is used as the nuclear data library of the neutron transport calculation, and activation file FENDL/A-3.0 [9] is used as that of the calculation of the reaction rates for the ^{99}Mo production.

3. Result and discussion

3.1. Case (A) Only RI module

Fig. 6 shows the reaction rates for the ^{99}Mo production as a function of the shifter thickness in the case (A) Only RI module. The reaction rate of the ^{nat}Mo(n,x)^{99}Mo reaction is a sum of that of the ^{98}Mo(n,γ)^{99}Mo reaction and that of the ^{100}Mo(n,2n)^{99}Mo reaction with consideration for their abundance. The weight of the $^{nat}MoO_3$ sample is about 750 g (net weight of the ^{nat}Mo is 500 g), and the thickness of the sample is about 1.6 cm. The reflector thickness is 20 cm. In all cases, the reaction rate of the ^{100}Mo(n,2n)^{99}Mo reaction decreases with the shifter thickness. Except for 0 cm of the shifter thickness, the reaction rates in the case without the shifter and reflector ((a) Nothing in Fig. 6) is larger than those in the other cases because fast neutrons are shielded and lose their energies by the shifter. For 0 cm, the reaction rates in the other cases are slightly larger by reflection of the neutrons. The reaction rate of the ^{98}Mo(n,γ)^{99}Mo reaction in the case without the shifter and reflector is smaller than the other cases because the shifter and reflector produce lower energy neutrons. Especially in the case of Be, the reaction rate of the ^{98}Mo(n,γ)^{99}Mo reaction increases clearly with the shifter thickness from 0 cm to 6 cm and decreases with that from 6 cm to 10 cm. The reaction rates of the ^{nat}Mo(n,x)^{99}Mo reaction represent the maximum value at 3 cm of the shifter thickness by the sum of these two effects. The maximum effect of the shifter is achieved in the case of Be. However, the differences are very small from 0 cm to 4 cm in this case. Then the shifter is not necessary in the case (A). Comparing the reaction rates of the ^{nat}Mo(n,x)^{99}Mo reaction at 0 cm of the shifter thickness which means only reflector, the case of Be shows the largest value. Be also gives the maximum effect as the reflector.

Fig. 7 shows the comparison between the neutron flux spectra in the $^{nat}MoO_3$ sample with the Be shifter of 3 cm thickness and the Be reflector of 20 cm thickness and that without ones. The lethargy width is defined as $\ln(E_{high}/E_{low})$ where E_{high} is a upper value and E_{low} is a lower value in an energy bin. When there are no shifter and reflector, high