Production of ^{91}Nb, ^{94}Nb, and ^{95}Nb from Mo by 14.5—14.8 MeV neutrons

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Production cross sections for ^{94}Nb(2.03×10^4 yr) have been measured from enriched ^{94}Mo and ^{nat}Mo. The samples were irradiated by 14.5—14.8 MeV neutrons and the cross sections were measured by gamma spectroscopy. The cross section for the ^{95}Mo(n,np + d)^{94}Nb reaction was deduced from the difference in ^{94}Nb activity in the natural and enriched Mo samples. Cross sections were also determined for the ^{95}Mo(n,p)^{95}Nb, ^{92}Mo(n,x)^{91}Nb^m, and ^{98}Mo(n,α)^{95}Zr reactions, all being in good agreement with previous data. Our data can also be used to estimate the production of the long-lived (700 yr) ground state of ^{91}Nb^g from ^{92}Mo.

INTRODUCTION

Fusion reactor materials will be exposed to high fluxes of 14 MeV neutrons which will produce a variety of long-lived activation products. These long-lived isotopes are of concern since they are difficult to dispose of under current waste disposal guidelines. Unfortunately, many of the production cross sections are not well known, so that it is difficult to assess the importance of various reactions or the necessity of tailoring fusion materials to minimize these long-lived isotopes. Consequently, we have undertaken to measure some of these reaction cross sections. Previously, we have reported measurements for the reactions ^{27}Al(n,2n)^{26}Al (7.2×10^5 yr) (Ref. 1) and ^{54}Fe(n,2n)^{53}Fe which decays to ^{53}Mn (3.7×10^6 yr) (Ref. 2). Cross sections for 22 reactions to shorter-lived isotopes have also been reported previously.[3] In the present paper, measurements are reported for various reactions on Mo, with particular interest in the production of ^{94}Nb (2.03×10^4 yr). Data are also presented for the ^{95}Mo(n,p)^{95}Nb, ^{92}Mo(n,x)^{91}Nb^m, and ^{98}Mo(n,α)^{95}Zr reactions. ^{91}Nb^m is also of interest to fusion waste activation since this isomeric state decays 96.5% of the time to the long-lived ground state ^{91}Nb^g (about 700 yr).

EXPERIMENTAL TECHNIQUE

The enriched Mo samples (92% ^{94}Mo) were obtained from Oak Ridge National Laboratory in the form of pressed metallic powder. Samples of natural Mo were sliced from a metal rod (Johnson Matthey, 99.99% purity). Both materials were in the form of disks about 3 mm in diameter by 1 mm thick. The density of the enriched Mo was about 60% that of natural Mo. The disks were then included with various other materials in a sample tube about 1.3 cm long which was irradiated at the Rotating Target Neutron Source II (RTNS II) at Lawrence Livermore National Laboratory. Two samples of each material were located at different positions in the tube. The sample tube was mounted on a radius of 0.85 cm where 0 cm is the incident deuteron beam axis at the RTNS II. Natural and enriched Mo samples were located at (z,θ) coordinates of (0.65 cm, 52°) and (1.5 cm, 30°), respectively, where z is the axial distance from the source. These parameters can be used to determine the average neutron energies and spread in neutron energies. The neutron energy spectra were calculated for the RTNS II as shown in Fig. 1. The calculations assumed a 360 keV deuteron beam incident on a TiT_2 target and the known dependence of energy loss,[4] the d-t cross sections, and angular distributions.[5] By averaging these spectra over the geometry of our samples, we estimate that the average neutron energies are 14.6 and 14.8 MeV and that the

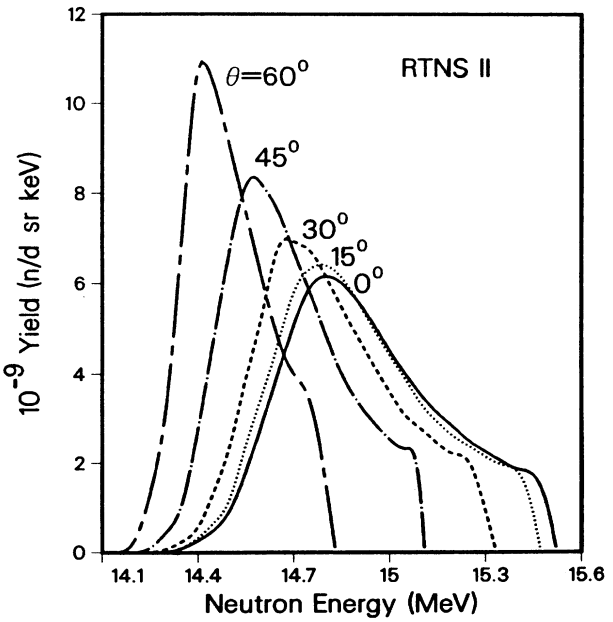

FIG. 1. Calculated neutron energy spectra as a function of angle for the RTNS II facility assuming a 360 keV deuteron beam incident on a TiT_2 target.

spread in the neutron energy distributions is about 400 keV full width at half maximum (FWHM). However, this spread in neutron energies is not very important since the reaction cross sections which we are measuring are expected to change very slowly near 14.7 MeV.

The sample tube contained iron dosimetry foils to determine the neutron fluence at the location of each sample. Using the iron cross sections determined in Ref. 3, we were able to construct a map of the neutron fluences. Fluences were also determined at 173 other locations surrounding the RTNS II target. The fluence map agreed with data in our region within 5%; however, only the local dosimetry need be considered for the present analysis and we estimate that the neutron fluences are accurate to about 5%, the largest uncertainty being our normalization to ^{93}Nb(n,2n),[3] which is known to have a cross section of 463 mb (±4%) with a flat energy dependence near 14 MeV.[6] Corrections were also made for the detailed beam history since the samples were irradiated along with other experiments for only 81 days over a period of about seven months. A computer program was used to follow the daily production and decay of each isotope so that all activities could be corrected to saturation levels. These corrections were only significant for the shorter-lived isotopes. The measured neutron fluences ranged from $(0.53-1.47)\times10^{18}$ neutrons/cm^2, depending on the position with respect to the source.

The presence of ^{94}Nb in the Mo samples was determined directly by Ge gamma spectroscopy. ^{94}Nb is known to have two gamma rays at 702.6 and 871.1 keV, both with 100% intensities.[7] All four samples were counted at six different decay times over a period of 480 days following the irradiation. Although no particular interferences or variations in the counting rates were observed, the background due to other activities declined substantially. It was necessary to count each sample for several days to obtain counting statistics less than 1%. The samples were counted in a well-defined geometry at about 10 cm from the face of a 25% efficient coaxial detector. Coincidence gamma summing for the 702—871 keV gamma cascase is estimated to be about 1%. Corrections were also made for gamma self-absorption, which averaged 2% for the enriched samples and 3.4% for the natural samples. The efficiency of the gamma detector was determined by reference to standard gamma sources from the National Bureau of Standards with an estimated absolute uncertainty of 1.5%.

The half-life of ^{94}Nb is reported to be $(2.03\pm0.16)\times10^4$ yr.[8] This uncertainty of 8% in the half-life unfortunately is directly carried over into an uncertainty in the measured cross section, since for such a long-lived activity the cross section is proportional to the measured specific activity times the half-life.

^{91}Nb^m was measured by the 105 and 1205 keV gammas, which have intensities of 0.58 and about 3.5%, respectively, and a half-life of 61 days.[7] Although the intensity of the 1205 keV gamma is not well known, the intensity of the 105 keV gamma is known to 3% and our measurements agree with the above 1205/105 ratio to within 1%. The isomeric state decays to the long-lived ground state, as discussed below. ^{95}Zr was determined by gamma counting the 724 and 756 keV gamma rays, which have intensities of 43.7% and 55.3%, respectively, and a half-life of 64.02 days.[7]

TABLE I. Isotopic composition of Mo samples (in %).

Isotope	Natural[a]	Enriched[b]
92	14.84	0.71
94	9.25	92.03
95	15.92	5.18
96	16.68	0.83
97	9.55	0.40
98	24.13	0.67
100	9.63	0.19

[a]Reference 9.
[b]Oak Ridge National Laboratory, Isotope Sales Division.

The isotopic composition of the Mo samples was determined for the enriched ^{94}Mo by Oak Ridge National Laboratory and the natural Mo was assumed to have the natural abundances as given in the literature.[9] Both compositions are listed in Table I. These data are needed in order to separate the relative contributions to the production of ^{94}Nb from both ^{94}Mo and ^{95}Mo. ^{95}Mo can produce ^{94}Nb by the (n,np) or (n,d) reactions, both of which are energetically possible at 14 MeV. It is also possible that there is a weak contribution from the ^{96}Mo(n,t) reaction; however, this cross section is expected to be weak at 14 MeV and there is only 0.83% ^{96}Mo in the enriched samples. This contribution was thus neglected and we solved for the relative isotopic cross sections using simultaneous equations for the known abundances of each isotope in the natural and enriched samples. The relatively large uncertainties for the ^{95}Mo cross sections are mainly due to 4% counting uncertainties in the natural Mo values, which are magnified (12%) in the separation of the isotopic cross sections.

The analysis of the ^{95}Nb activity is complicated by the fact that ^{95}Zr decays to ^{95}Nb. Hence it was necessary to subtract this contribution both during and after the irradiation. ^{95}Nb activity was determined by gamma counting the 766 keV gamma, which has an intensity of 99.8% and a half-life of 35.06 days.[7] The enhancement of the ^{95}Nb activity from the decay of ^{95}Zr following the irradiation was easily determined by following both activities over several half-lives and the uncertainty on the ^{95}Nb activity at the end of irradiation is only 2%. The enhancement due to ^{95}Zr decay during the irradiation was calculated using the ^{95}Zr activity and Mo isotopic ratios in Table I. In the enriched ^{94}Mo samples, ^{95}Mo comprises 5.18% while ^{98}Mo is only 0.67%. Hence, the contribution from the ^{98}Mo(n,α)^{95}Zr reaction is only about 1% for these samples.

DISCUSSION OF RESULTS

The measured cross sections are presented in Table II and shown in Figs. 2 and 3. As discussed above, the major sources of the uncertainties include our absolute normalization for the neutron fluence measurements, which

TABLE II. Measured cross sections (mb) for Mo.

Reaction	14.55	14.60	14.78	14.80	±%[a]
^{94}Mo(n,p)^{94}Nb	57.2		53.1		10
^{nat}Mo(n,x)^{94}Nb[b]		7.9		7.8	11
^{95}Mo(n,x)^{94}Nb[c]		16.3		18.3	15
^{95}Mo(n,p)^{95}Nb	40.4		37.1		6
^{92}Mo(n,x)^{91}Nb^{m} [d]	157.0	153.0	145.0	145.0	7
^{98}Mo(n,α)^{95}Zr	6.56	6.56	6.24	6.21	6
^{92}Mo(n,x)^{91}Nb^{g} [e]	≈300				
^{nat}Mo(n,x)^{91}Nb^{g} [e]	≈45				

[a]Major sources of uncertainty include neutron fluence (5%), ^{94}Nb half-life (8%), efficiency (1.5%), statistics (1%), and deconvolution of ^{95}Mo, (12%) for ^{94}Nb and (2%) for ^{95}Nb.
[b]Sum of reactions from ^{94,95,96}Mo.
[c]Sum of (n, d + np + pn) reactions.
[d]Sum of (n, 2n + d + np + pn) reactions.
[e]Values include contribution from ^{91}Nb^{m}.

has an estimated uncertainty of 5%, and the 8% uncertainty in the half-life of ^{94}Nb. All of the uncertainties are summarized in Table II. Relative uncertainties are only about 3%, except for the ^{nat}Mo and ^{95}Mo reactions to ^{94}Nb, which include a 4% counting uncertainty and a 12% deconvolution uncertainty, respectively.

There are no reported activation measurements of the cross section for the ^{94}Mo(n,p)^{94}Nb reaction. Haight *et al.*[10] have measured the total proton emission spectrum at 14.7 MeV and report a total cross section of 124±15 mb. However, this value includes other possible reaction channels such as (n,np) and (n,2p). Bramlitt *et al.*[11] measured the (n,p) cross section to the 6.6 m isomer of ^{94}Nb (which decays to the ground state) at 15 MeV and report a value of 6±1.5 mb. Both of these measurements are consistent with our results. Gardner *et al.*[12] have performed theoretical calculations of the (n,p) cross section and predict a value of 53.8 mb at 14.5 MeV, in excellent agreement with our value. We have also used the THRESH2 (Ref. 13) computer code to predict the relative cross sections of (n,px) reactions. THRESH2 is based on a semiempirical fit to available neutron reaction data for a broad range of elements. Although the results may only be reliable to a factor of 2, the code is quite useful for estimating the magnitude of various cross sections. In the present case, THRESH2 predicts that the ^{94}Mo(n,p)^{94}Nb reaction represents about 70% of the total (n,px) cross section near 14.7 MeV. Hence, it is quite reasonable that Haight *et al.*[10] report a significantly higher cross section for the (n,px) reaction. Qaim[14] reports similar ratios for other Mo isotopes.

There are no data reported in the literature for direct comparison with our measurements of the ^{nat}Mo and ^{95}Mo reactions to ^{94}Nb. Haight *et al.*[10] report a value of 8±3 mb and Qaim[14] estimated a value of 4±3 mb for the the ^{95}Mo(n,d) reactions to ^{94}Nb; however, our values are

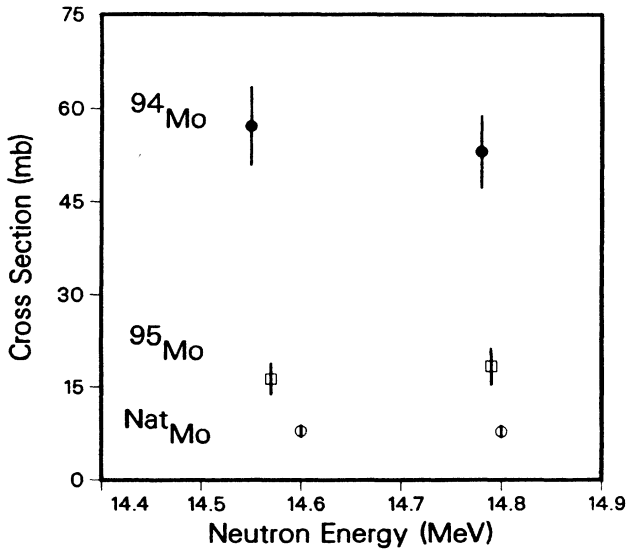

FIG. 2. Data are shown for the production of ^{94}Nb from ^{94}Mo (solid circles), ^{95}Mo (squares), and ^{nat}Mo (circles).

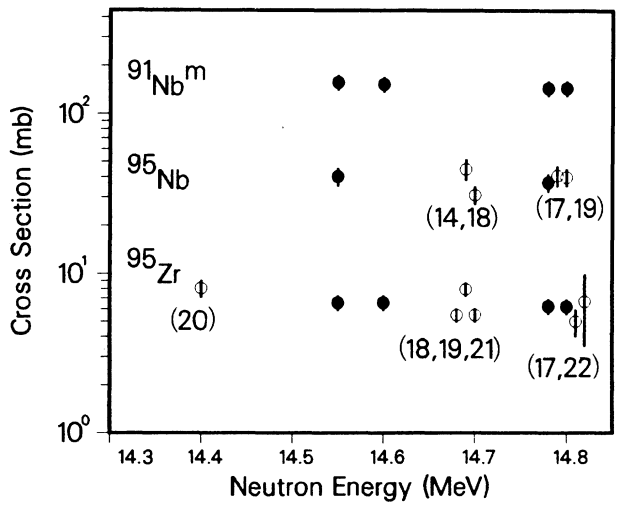

FIG. 3. Data (solid circles) for the ^{92}Mo(n, 2n + d + np) ^{91}Nb^{m}, ^{95}Mo(n,p)^{95}Nb, and ^{98}Mo(n,α)^{95}Zr reactions are compared to previous data (open circles) for which references are denoted in parentheses.

higher since we also include the (n,np) and (n,pn) reactions. THRESH2 predicts values of 40—50 mb for the ^{95}Mo reactions (n, d + np + pn), which is somewhat higher than our data. However, THRESH2 also overpredicts the ^{94}Mo(n,p) reaction at about 87 mb. THRESH2 also predicts that the possible contribution of ^{96}Mo to ^{94}Nb via the (n,t) reaction is only about 10% of the ^{95}Mo reactions to ^{94}Nb. Hence, this is not important in our enriched samples since ^{96}Mo is only 0.83% and the reaction thus contributes less than 0.1%. In the natural Mo samples, the ^{96}Mo contribution is estimated to contribute less than 2% of the total and thus has only a small effect ($<1\%$) on our separation of the ^{95}Mo reactions from the ^{94}Mo(n,p) reaction.

Our results for the production of ^{91}Nb^m are due to a combination of the (n,2n) and (n, d + pn + np) reactions from ^{92}Mo. In the (n,2n) case, ^{91}Mo decays rapidly to ^{91}Nb. Unfortunately, it is difficult to compare our results to previous data due to the presence of isomeric states for both ^{91}Nb and ^{91}Mo. Previous data[15,16] shows that the ^{92}Mo(n,2n) reaction proceeds mainly to the ground state of ^{91}Mo. Since ^{91}Mo^g decays mainly to the ground state of ^{91}Nb, this part of the reaction would not be observable in our data. Thus, although the total (n,2n) cross section is about 160 mb, only about 10—20 mb proceeds to ^{91}Nb^m, which we are measuring.[15] Consequently, our data imply that most of our production of ^{91}Nb^m is due to the (n, d + np + pn) reaction mechanism rather than the (n,2n) reaction. No previous data for this reaction are known. In any case, we note that the isomeric state of ^{91}Nb^m decays 96.5% of the time to the ground state, which is also of interest to fusion reactor activation since the ground state half-life is about 700 yr. Our cross sections to the isomeric state can thus be used to estimate the production of the long-lived ground state, as follows. Using previous data, the total (n,2n) cross section is about 160 mb. Our data imply that the (n,d +) reactions must be about 140 mb, if we subtract about 10 mb for the (n,2n) branch to the isomeric level. Adding these two results gives a total production cross section for the long-lived ground state of ^{91}Nb^g from ^{92}Mo of about 300 mb near 14.7 MeV. Considering the abundance of ^{92}Mo in natural Mo, the production cross section would be about 45 mb for ^{nat}Mo.

Our results for the ^{95}Mo(n,p)^{95}Nb reaction agree very well with the literature. Artem'ev *et al.*[17] report a value of 40±5 mb at 14.8 MeV, Qaim[14] reports 31±4 mb at 14.7 MeV, Fukuda[18] reports 45 mb at 14.6 MeV, Amemiya[19] 41.1±3.6 mb at 14.8 MeV, and Gardner *et al.*[12] calculate 39.5 mb at 14.5 MeV using a semiempirical model. Haight *et al.*[10] measured the total proton production cross section and report 84±10 mb at 14.8 MeV. As discussed previously, this value includes other possible reaction channels, as estimated by Qaim.[14] THRESH2 predicts that the (n,p) channel is only about 70% of the total; hence, the value of Haight *et al.* is in reasonable agreement with our results.

The ^{98}Mo(n,α)^{95}Zr reaction has been measured previously by Lu *et al.*,[20] who report 8.1±1.0 mb at 14.4 MeV and by Artem'ev *et al.*,[17] who report 5±1 mb at 14.8 MeV. Rahman *et al.*,[21] Fukuda[18], and Amemiya[19] also report values near 5, 8, and 5.5 mb, respectively, near 14.7 MeV. All of these values are in reasonable agreement with our results, especially since the cross section is expected to decline with energy. Helium production measurements at RTNS II by Kneff *et al.*[22] also estimate that the ^{98}Mo cross section is 6.7±3.2 mb at 14.8 MeV, in excellent agreement with our results.

Rahman *et al.*[21] recently reviewed various reactions on Mo isotopes and report data in the 5.9—9.6 MeV energy range. Our data agree quite well with the overall trend of these reactions.

CONCLUSIONS

Our cross section measurements can be used to predict the production of ^{91}Nb and ^{94}Nb at fusion reactors. However, these calculations are complicated by variations in specific reactor designs, the influence of other side reactions which we did not measure, and the effects of burnup in lengthy irradiations. More detailed calculations are thus required, as has been done recently for the STARFIRE and MARS reactor designs.[23,24] If we compare our cross sections to those used during recent activation calculations,[24] then near 14 MeV we find that our values for the production of ^{94}Nb are about 12% higher for ^{94}Mo(n,p), 55% higher for ^{95}Mo(n, d + np), and about 23% higher for natural Mo. In the case of ^{91}Nb, our values are about 30% lower than previous calculations. Consequently, we would expect that the production of ^{94}Nb would be increased by about 23% and ^{91}Nb decreased by about 30% for any fusion reactor calculation. If we only consider the fast neutron flux in the STARFIRE reactor design, then for an operation of six years at first wall loading of 3.6 MW/m^2 (21.6 MW yr/m^2), Mo would produce about 75 μCi/g (755 μCi/cm^3) of ^{94}Nb and 12 mCi/g (124 mCi/cm^3) of ^{91}Nb. These values are similar to those reported recently; however, these calculations also included the growth and decay of contributing side chains. In the case of ^{94}Nb, the isomeric cross sections were assumed to be equal to the ground state values. This is not a good assumption and has the effect of doubling the predicted activities of ^{94}Nb. However, this is balanced by the fact that the STARFIRE design includes a significant thermal neutron flux which produces a significant (50%) burnup of ^{94}Nb. Thus the two effects nearly cancel, making our calculations appear to be similar to the previous ones.[24] This is not the case for the MARS design (which has no significant thermal flux) and our activities are abut half those reported previously. In the case of ^{91}Nb, our values are indeed about 30% lower than before, as would be expected.

^{91}Nb^g has a shorter half-life and decays by electron capture producing only low energy x rays rather than the energetic gammas of ^{94}Nb. Hence, ^{91}Nb^g is less of a concern that ^{94}Nb; nevertheless, it contributes, along with ^{94}Nb and ^{93}Mo, to the problem of disposing of Mo-containing alloys. The impact of these cross sections is dependent on specific reactor designs and requires more detailed calculations. We also note that Mo will generate other long-lived isotopes, such as ^{93}Mo (3500 yr), ^{93}Zr (1.5×10^6 yr), and ^{92}Nb (3.7×10^7 yr), and cross sections to these isotopes are also relatively unknown. Work on these and

other long-lived products from other materials is currently in progress.

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