PNL-SA-18521

Received by 271

OCT 0 4 1990

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PNL-SA--18521

DE91 000155

August 1990

Presented at the
Seventh ASTM-EURATOM Symposium
on Reactor Dosimetry
Strasbourg, France
August 27-31, 1990

Work supported by
the U. S. Department of Energy
under Contract DE-AC06-76RLO 1830

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Measurements of Activation Cross Sections for Fusion Reactor Applications

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ABSTRACT

Neutron activation cross section data are needed for fusion reactor materials to facilitate neutron dosimetry and to determine activity levels for waste disposal, safety, and reactor maintenance applications. Measurements are reviewed for 36 different reactions, many leading to long-lived nuclides. Irradiations were performed at 14 MeV at the RTNS II to fluences of 10^{18} n/cm². Activities were determined by either gamma and x-ray analysis or liquid scintillation counting following radiochemical separations to remove interfering activities. Results are compared to calculations with the ALICF computer code. Activity levels are predicted for the STARFIRE fusion reactor design and compared to previous estimates. Other work is summarized including rare earth irradiations in Be(d,n), H(t,n), and T(d,n) accelerator-based neutron sources with average energies of 3, 10, and 14 MeV, respectively.

INTRODUCTION

Neutron activation cross sections are needed for fusion reactor materials to facilitate neutron dosimetry and to determine activity levels for waste disposal, safety, and reactor maintenance applications.¹ Although such data are widely available for fission reactor applications, the high fluences of higher energy (14 MeV) neutrons found in fusion reactors favor different reaction channels leading to different radioisotopes. In particular, (n,2n) and charged particle reactions are favored, thus tending to produce lower mass isotopes than the target material, whereas fission reactor irradiations tend to produce higher mass products through successive (n,gamma) capture reactions.

Unfortunately, we do not have any sources of high-energy neutrons at the fusion reactor levels of 10^{15} n/cm²-s. Hence, we cannot experimentally verify theoretical predictions^{2,3} of the production of radioisotopes that are produced by two or more

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successive reactions. To illustrate the problem, suppose that all of the (n,2n) and charged particle reactions have a cross section of about 1 barn (10^{-24} cm²) and that the 14 MeV fusion reactor flux is about 10^{15} n/cm². If all the products are long-lived, then in 1 year we will produce an atomic ratio of 0.01 for the primary product, 10^{-4} for the reaction product from that isotope, and so on. On the experimental side, the most intense source of 14-MeV neutrons was the Rotating Target Neutron Source II (no longer in operation) with a flux of about 10^{12} n/cm². This means that in a 2-week irradiation we can produce a neutron fluence of about 10^{18} n/cm², which is only capable of producing atom ratios of 10^{-6} for the primary reaction product and only 10^{-12} for the secondary, and so on. Although, in some cases we can measure each sequential reaction rate separately, this is only rarely possible since many of the intermediate isotopes are radioactive. Hence, we have little hope of directly measuring all of the predicted activation products barring the operation of a high-flux, high-energy neutron source.

Consequently, we have attempted to measure the primary reaction products at 14 MeV on a variety of fusion reactor materials. Comparisons between measured and calculated reaction rates can then help to refine our predictions of activities.

OVERVIEW OF MEASUREMENTS

Measurements have been performed at a wide range of neutron energies from thermal up to 450 MeV. Results presented in this paper will focus on measurements of the production of long-lived isotopes near 14 MeV and attempts to measure long-lived isotopes in rare earth materials. Previously, we have reported measurements of activation cross sections of 24 other reactions at 14 MeV, including the ^{27}Al and $^{54}\text{Fe}(n,2n)$ reactions^{5,6} that are useful for fusion plasma diagnostics. Above 14 MeV, neutron dosimetry cross sections have been measured and tested at energies up to 40 MeV in Be(d,n) fields,^{7,8} including new work presented separately in these proceedings.⁹ At higher energies, spallation cross sections have been measured for Al and Cu^{10,11} for irradiations at spallation neutron sources.

MEASUREMENTS NEAR 14 MEV

In the absence of any high-flux source of 14-MeV neutrons, we have irradiated samples in a variety of available facilities. Prior to decommissioning in 1987, many different samples were irradiated at the Rotating Target Neutron Source II at Lawrence Livermore National Laboratory. Fluences up to 10^{18} n/cm² were obtained during 2-week irradiations in close geometry as well as during prolonged irradiations at more remote locations.^{12,4} The source and neutron spectra have been described previously.^{12,4} Data have

been published^{13,14} for 12 different reactions leading to long-lived isotopes. The results are summarized in Table I. Results have been compared to previous measurements and theoretical calculations with the ALICE computer code,¹⁵ and the ratio of calculated to experimental values (C/E) are listed in Table I along with the data, half-lives, and references.

Examination of the data in Table I reveals rather large uncertainties in some of the measurements and rather large deviations from unity in the C/E ratios. The uncertainties in the nuclear decay data¹⁶ are described in detail in Table II. As can be seen, the half-lives and x-ray intensities are substantial, especially for ⁵⁹Ni, ⁹¹Nb, and ⁹³Mo. Comparison with Table I shows that the decay data uncertainties are usually the largest part of the total error, the remainder being counting statistics and efficiency calibrations. In general, this is a serious problem in attempts to measure any of the longer-lived isotopes, and we note a need to review and improve the nuclear decay data.

The C/E ratios, which compare ALICE calculations with the experimental data, may indicate problems with the nuclear data. In particular, the case of ⁹¹Nb is quite unusual with a C/E value of only 0.33, whereas more generally ALICE appears to overpredict the (n,2n) cross sections. We note that the half-life is only known to about 20%. If, in fact, the half-life were closer to 300 years rather than 680 years, our results would be in better agreement with the theoretical predictions. A similar situation is found for ⁵⁹Ni, where the C/E value is 6.0 and the half-life is only known to about 20%. If the half-life were 3×10^5 or longer, the results would agree with calculations and systematics of other nickel isotopes. Obviously, more measurements of the cross sections and half-lives are needed to resolve these questions.

PREDICTED FUSION REACTOR ACTIVITIES

The measured activation cross sections in Table I can be used to predict the production of these long-lived isotopes in operational fusion reactors. We have performed such calculations using the first wall spectrum of the STARFIRE reactor design,¹⁷ and the results are listed in Table III. In most cases the 14 MeV cross section is all that is needed for this calculation due to the high thresholds of the (n,2n) reactions. In the case of ⁶³Cu(n,p)⁶³Ni and ⁹⁴Mo(n,p)⁹⁴Nb, lower energy neutrons do contribute and we have used the ALICE calculation to estimate these contributions. We have not made any allowance for burnup of the isotopes, as has been done in previous calculations;^{2,3} however, we estimate that such effects would be small for these isotopes. The calculations assumed six full-power years operation followed by a 3000-day cooling period. These results can be compared to previous calculations as well as to landfill disposal requirements.

OTHER WORK IN PROGRESS

Argonne is participating in a Coordinated Research Program sponsored by the International Atomic Energy Agency to measure the production of long-lived activities in rare earth materials. The experimenters and facilities include J.W. Meadows and D.L. Smith at Argonne, using a $\text{Be}(d,n)$ neutron spectrum produced by 7-MeV deuterons stopped in a thick beryllium target; R.C. Haight at Los Alamos National Laboratory, using 10-MeV neutrons from the $\text{H}(t,n)$ reaction; and Y. Ikeda at the Japan Atomic Energy Research Establishment, using the 14-MeV $\text{T}(d,n)$ Fast Neutron Source. Foils and pressed powders of copper, silver, europium, terbium, and hafnium, as well as titanium, iron, and nickel dosimeters, were irradiated in each facility. The reactions being studied are listed in Table IV. Preliminary results have been obtained for each irradiation; however, data have not been corrected for interferences from low-energy neutrons nor for multiple scattering effects. Estimates show that such effects are less than 7% for all reactions except for $^{153}\text{Eu}(n,2n)^{152}\text{Eu}$ where the $^{151}\text{Eu}(n,g)$ reaction may produce up to half of the observed activity due to the very high thermal cross section (5900 barns). In the case of $^{179}\text{Hf}(n,2n)^{178\text{m}}\text{Hf}$, the activity has not been observed after nearly 2 years decay due to the higher activities of other hafnium isotopes.

FUTURE WORK

Other measurements have been made at the $\text{Be}(d,n)$ facility at Argonne and work is in progress to determine cross sections for the reactions $^{93}\text{Nb}(n,n')^{93\text{m}}\text{Nb}$ (16.1 y), $^{63}\text{Cu}(n,p)^{63}\text{Ni}$ (100 y), and $^{60}\text{Ni}(n,p)^{60}\text{Co}$ (5.27 y). Other materials were also irradiated to high fluences at RTNSII at 14 MeV and analyses are planned for the reactions $^{94}\text{Zr}(n,2n)^{93}\text{Zr}$ (1.5×10^6 y) and $^{93}\text{Nb}(n,2n)^{92}\text{Nb}$ (3.6×10^7 y). As discussed previously, measurements are needed for many other reactions to cover the gamut of fusion reactor materials.

ACKNOWLEDGEMENTS

This work was supported by the Office of Fusion Energy, U.S. Department of Energy.

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Table I Production of Long-Lived Isotopes Near 14.8 MeV

<u>Reaction</u>	<u>$T_{1/2}$, y</u>	<u>σ, mb</u>	<u>C/E^a</u>
$^{50}\text{V}(n, 2n)^{49}\text{V}^b$	0.90	258 ± 39	2.8
$^{56}\text{Fe}(n, 2n)^{55}\text{Fe}$	2.7	454 ± 35	1.4
$^{63}\text{Cu}(n, p)^{63}\text{Ni}$	100.	54 ± 4	0.91
$^{64}\text{Ni}(n, 2n)^{63}\text{Ni}$	100.	958 ± 64	1.2
$^{60}\text{Ni}(n, 2n)^{59}\text{Ni}$	7.5×10^4	104 ± 25	6.0
$^{94}\text{Mo}(n, p)^{94}\text{Nb}$	2.0×10^4	55 ± 6	0.82
$\text{Nat}_{\text{Mo}}(n, x)^{94}\text{Nb}$	2.0×10^4	7.8 ± 0.8	-
$^{92}\text{Mo}(n, 2n\beta)^{91}\text{Nb}$	680. (?)	603 ± 119	0.33
$^{94}\text{Mo}(n, 2n)^{93}\text{Mo}$	3500.	550 ± 136	2.5
$^{94}\text{Mo}(n, d+np)^{93\text{m}}\text{Nb}$	16.1	5.7 ± 0.9	2.0
$^{95}\text{Mo}(n, t+nd)^{93\text{m}}\text{Nb}$	16.1	1.36 ± 0.27	-
$\text{Nat}_{\text{Mo}}(n, x)^{93\text{m}}\text{Nb}$	16.1	0.75 ± 0.11	-

^aRatio of ALICE calculation to experimental measurement

^bVanadium data measured at 14.3 MeV

Table II Nuclear Decay Data for Long-Lived Isotopes

<u>Isotope</u>	<u>Half-life, y</u>	<u>Radiation, keV</u>	<u>Branching, %</u>	<u>Net, %</u>
^{49}V	0.90 ± 0.04	X_K 4.5-4.9	0.196 ± 0.017	9.8
^{59}Ni	$7.5 \pm 1.3 \times 10^4$	X_K 6.9	29.5 ± 1.5	18.2
^{63}Ni	100.1 ± 2.0	β^- 66.	100.	2.0
^{91}Nb	680 ± 130	X_K 15.7	54.2 ± 1.8	19.4
^{93m}Nb	16.13 ± 0.15	X_K 16.6	8.98 ± 0.19	2.3
^{93}Mo	3500 ± 700	X_K 16.6	62.0 ± 2.3	20.3

Table III Calculated Fusion Reactor Activities
(STARFIRE First Wall, 21.6 MW-y/m^2 , 3000 d decay)

<u>Material</u>	<u>Isotopes</u>	<u>Half-life, y</u>	<u>Activity, mCi/cc</u>
Vanadium	^{49}V	0.9	2.8
Molybdenum	^{91}Nb	680.	243.
	^{93m}Nb	16.1	106.
	^{94}Nb	20,000.	0.8
	^{93}Mo	3500.	28.
Iron	^{55}Fe	2.7	25,000.
Copper	^{63}Ni	100.	1,795.
Nickel	^{63}Ni	100.	227.
	^{59}Ni	75,000.	1.

Table IV Long-Lived Rare Earth Isotopes
 [Irradiations at Be(d,n), 10, and 14 MeV]

<u>Reaction</u>	<u>Half-life</u>	<u>Comments (1-2 year decay)</u>
$^{107}\text{Ag}(n,2n)^{106\text{m}}\text{Ag}$	8.5 d	Measurements complete
$^{109}\text{Ag}(n,2n)^{108\text{m}}\text{Ag}$	127 y	Only seen at 14 MeV
$^{151}\text{Eu}(n,2n)^{150\text{g}}\text{Eu}$	35.8 y	Measurements complete
$^{153}\text{Eu}(n,2n)^{152}\text{Eu}$	13.3 y	Strong interference $^{151}\text{Eu}(n,g)$
$^{159}\text{Tb}(n,2n)^{158}\text{Tb}$	150 y	Measurements complete
$^{176}\text{Hf}(n,2n)^{175}\text{Hf}$	70 d	Measurements complete
$^{179}\text{Hf}(n,2n)^{178\text{m}2}\text{Hf}$	31 y	Not seen - very weak
$^{180}\text{Hf}(n,2n)^{179\text{m}2}\text{Hf}$	25 d	Measurements complete

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