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Abstract

Neutron cross section measurements are reviewed for the production of long-lived isotopes in fusion reactor materials. Samples were irradiated at 14.5-14.8 MeV at the RTNSII facility. Long-lived reaction products were detected using gamma and x-ray spectroscopy, liquid scintillation spectrometry, and accelerator mass spectrometry. Radiochemical separations were performed for many samples prior to analysis. Results are summarized for reactions leading to ^{26}Al (720,000 y), ^{53}Mn (3,700,000 y), ^{55}Fe (2.73 y), ^{63}Ni (100 y), ^{59}Ni (76,000 y), ^{91}Nb (700 y), and ^{94}Nb (20,300 y).

Introduction

The production of long lived isotopes in fusion reactor materials is of concern due to restrictions which may be placed on the disposal of such radioactive materials. Such activation data are also useful for neutron dosimetry, plasma diagnostics, and reactor maintenance applications. Unfortunately, the necessary neutron activation cross sections are generally not well-known. Although in principle measurements are needed at all neutron energies up to 14 MeV, many of the most important long-lived products are formed by (n,2n) reactions which have thresholds of 10 MeV or higher. Hence, for fusion reactor spectra of interest, it is usually sufficient to measure the production cross sections near 14 MeV. Consequently, we have irradiated a wide range of materials with high fluences of 14 MeV neutrons and have used a variety of techniques to detect the long-lived activation products. By including dosimetry materials with each irradiation, neutron cross sections have been measured relative to well-known reactions. The measured cross sections can then be used to predict activity levels in fusion reactors in order to evaluate restrictions which may have to be placed on the use of various materials.

Irradiation and Dosimetry

The reactions which we are studying are listed in Table I. Samples consisted of the pure metals Al, Fe, Mo, Cu, Zr, Ni, and Nb and separated isotopes of ^{56}Fe, ^{60}Ni, ^{64}Ni, ^{94}Zr, and ^{94}Mo. All of the separated isotopes were obtained from Oak Ridge National Laboratory in the form of metallic powder which was then pressed into discs or placed in thin aluminum tubing for irradiation. Samples measured 1.5-3.0 mm diameter and 1-14 mm long, depending on the amount of material available and the expected strength of the activation product.

The samples were irradiated during several different runs over a period of several years at the Rotating Target Neutron Source (RTNS II) at Lawrence Livermore National Laboratory. Some of the smaller samples were irradiated very close to the source allowing us to obtain fluences of about 10^{17} n/cm^2 in about one week of operation. Larger samples were irradiated at much larger distances from the source by piggybacking with different irradiations over periods up to 7 months. In this way we were able to obtain fluences up to $2x10^{16}$n/cm^2. In each case, dosimetry materials were included with each irradiation and neutron fluence maps were generated. Shorter irradiations used the ^{93}Nb(n,2n)^{92m}Nb (10 d) reaction which has a nearly flat energy response near 14 MeV with a cross section of 463 mb (±4%).[1] For the long irradiations, the ^{54}Fe(n,p)^{54}Mn (312 d) reaction was used to determine the fluence. The cross section for this reaction drops from 331 mb at 14.5 MeV to 303 mb at 14.8 MeV, with an uncertainty of ±7%.[1] The mean neutron energy at each sample is determined by the deuteron beam energy, energy loss in the tritium targets, and reaction kinematics. Calculations of the neutron spectra as a function of angle were recently published.[2] Careful measurements of the geometry for each irradiation, as well as the neutron dosimetry mapping, permitted us to determine the average neutron energy and fluence for each sample; more detailed results have been published previously.[3]

Previous Measurements

Activation cross sections for 22 reactions to shorter-lived reaction products measured at the RTNS II were recently published.[1] Measurements were reported previously for the ^{27}Al(n,2n)^{26}Al reaction to the ground state (720,000 y) and isomeric state (6.34 s)[4] and for the ^{54}Fe(n,2n)^{53}Fe(β)^{53}Mn ($3.7x10^6$ y)[5]. These reactions have thresholds near 14 MeV and have been shown to be very useful for plasma diagnostics.[6] Due to the steep energy dependence near threshold, fusion activation calculations are very dependent on the plasma temperature and the presence of neutral beams. We have recently published results for the production of ^{91}Nb (700 y) and ^{94}Nb (20,400 y) from natural Mo and ^{94}Mo.[2] All of these measurements were performed by gamma spectroscopy, although ^{26}Al was also measured using the relatively new technique of accelerator mass spectrometry.

Liquid Scintillation Measurements

Several new measurements have been completed using liquid scintillation spectrometry to detect radioisotopes which decay by electron capture or low-energy beta decay. In most cases, the desired long-lived activities cannot be easily detected due to the presence of other shorter-lived activities from the sample or impurities. Hence, radiochemistry was used to separate the desired activities prior to analysis. Gamma and x-ray spectroscopy was used to monitor the separations. Samples were generally dissolved in acid and separated using ion-exchange

reactions. Complete chemical procedures have been published elsewhere.[7] Identification of the desired activity was confirmed by analysis of the liquid scintillation energy spectrum. Standard solutions of ^{55}Fe and ^{63}Ni were prepared to match the concentration (quenching) expected for the samples. In the case of ^{59}Ni, the spectrum was assumed to be similar to that of ^{55}Fe since the x-rays and conversion electrons only differ in energy by about 1 keV. The ^{59}Ni x-rays were counted using a thin, intrinsic Ge detector which was calibrated using ^{54}Mn, ^{55}Fe, ^{137}Cs, and ^{241}Am standards.

^{55}Fe Measurements

^{55}Fe has a halflife of 2.73±.03 y and decays by electron capture producing Auger electrons and x-rays near 6 keV.[8] ^{55}Fe was produced by the ^{56}Fe(n,2n) reaction from natural Fe and isotopically separated ^{56}Fe(99.87%). The principal unwanted activities in the samples were ^{51}Cr, ^{54}Mn, ^{57}Co, and ^{58}Co. The first two were produced by reactions on ^{54}Fe, principally in the natural Fe target whereas the Co activities were observed from Co and Ni impurities in the ^{56}Fe samples. All of these undesirable activities were easily removed by ion-exchange.[7]

Figure 1 shows the liquid scintillation spectra for ^{55}Fe both from our samples and the standard solution. The data are given in Table II. As can be seen, our results for the ^{56}Fe(n,2n)^{55}Fe cross sections near 14.8 MeV have a standard deviation of only 2.6%.

^{63}Ni Measurements

^{63}Ni has a halflife of 100.1±2.1 y and decays by β^- emission with an endpoint energy of 66 keV (average energy = 17 keV).[8] ^{63}Ni was produced by the ^{64}Ni(n,2n) and ^{63}Cu(n,p) reactions. The ^{64}Ni was enriched to 93.57% and the copper had the natural abundance of 69.17% for ^{63}Cu. The ^{64}Ni samples contained undesirable activities of ^{54}Mn, ^{57}Co, ^{58}Co, and ^{60}Co, whereas the Cu samples contained mainly ^{60}Co. All of these unwanted activities were easily removed by ion-exchange separations.[7]

For both reactions, no residual activities were detected by gamma spectroscopy. Samples were prepared from the ion-exchange column wash and all of the unwanted activites could be accounted for within a few percent. Figure 2 shows the liquid scintillation spectra for each case, both before and after chemical separations, as well as the spectra from the standard solutions. As can be seen, the spectra from the separated materials closely match that of the standards. The neutron cross section data for each sample are listed in Table II. Again we note that the scatter in the data is only 1.5% for both the Ni and Cu samples.

^{59}Ni Measurements

^{59}Ni has a halflife of 7.5±1.3x10^{4} y and decays by electron capture emitting about 6 keV Auger electrons and 7 keV x-rays.[8] ^{59}Ni was produced from the ^{60}Ni(n,2n) reaction using separated isotopes of ^{60}Ni (99.81%). The principal unwanted activities were ^{54}Mn, ^{57}Co, ^{58}Co, and ^{60}Co.

The chemical procedures were initially the same as for ^{63}Ni. Although the Co activities were readily separated by ion-exchange, we were only able to remove about 99.7% of the ^{54}Mn which left a residual activity comparable to that from ^{59}Ni. Furthermore, the spectrum may contain a contribution from ^{63}Ni. In spite of the low ^{64}Ni content (less than 0.02%), our present measurements of the (n,2n) reactions indicate that the ^{63}Ni activity could be comparable with that of ^{59}Ni.

Consequently, we decided to count the 7 keV x-rays from the decay of ^{59}Ni which have an intensity of 33.1±1.7%.[8] Samples were prepared from the purified Ni elutant by evaporating several milligrams of Ni onto an aluminum counting plate. Net corrections on the order of 10% were applied for self-absorption and scattering. In this case ^{63}Ni does not interfere since the β^- decay produces no Co x-rays. Data are listed in Table II. Uncertainites are larger for this reaction due to poorer counting statistics and the large (17%) uncertainty in the halflife.[8]

Comparison With Previous Data

All of the measured neutron cross sections are summarized in Table III, where they are also compared with previous data. In the case of the ^{56}Fe(n,2n)^{55}Fe reaction, our data are seen to agree quite well with previous data by Wenusch[9], Frehaut[10], Molla[11], and Kozyr[12]. The measurement by Joensson et al.[13] is clearly much lower. Our ^{63}Cu(n,p)^{63}Ni cross sections are somewhat lower than previous measurements.[14,15] However, these are measurements of the total proton production cross section which includes other possible reactions and hence are expected to be larger than our measurement. The measurement by Molla et al.[11], is directly comparable to ours since they measured the β^- activity from ^{63}Ni. However, their cross section has a relatively large uncertainty (40%) compared to our measurement (6.7%). No previous data were found for the ^{60}Ni(n,2n)^{59}Ni or ^{64}Ni(n,2n)^{63}Ni reactions.

Discussion and Conclusions

The measured neutron cross sections can be used to calculate the production of long-lived isotopes in fusion reactor materials, as shown in Table IV. These calculations are for the STARFIRE[16] fusion reactor design for a six year operating time (21.6 MW-y/m^2); however, effects due to thermal neutrons have been neglected due to uncertainties in shielding calculations. In most cases, the activation is mainly due to the interaction with 14 MeV neutrons since there is no significant

neutron flux above the typical threshold energies of 7-13 MeV. However, in the case of the ^{63}Cu(n,p) reaction, lower energy neutrons increase the production of ^{63}Ni significantly. Since we do not know the energy-dependent cross section for this reaction, we assumed that the relative contribution from lower energy neutrons would be about equal to that from the 14 MeV neutron flux, as reported previously.[17] Calculations were not included for the ^{27}Al and ^{54}Fe(n,2n) reactions since the activities are crucially dependent on the plasma temperature and neutral beam heating parameters.[6]

The fusion reactor activities reported in Table IV can be compared to previous estimates.[17] This comparison is not straightforward since we have neglected effects due to thermal neutrons. However, it is not certain that the relatively high thermal fluxes given for the STARFIRE[16] first-wall spectrum should be used for bulk materials due to neutron shielding effects. If we compare our measurements with the 14 MeV cross sections used in that study, we find that the ^{63}Cu(n,p) values are 80% higher, the ^{60}Ni(n,2n) values are 3.0 times higher, and the ^{94}Mo(n,p) values are about 50% lower. Values for the other reactions are quite similar. Hence, we would expect that the activity levels would scale accordingly if we included secondary effects due to thermal neutrons and side reactions as done in reference 17. The implications of these activities for fusion reactor design depend crucially on the details of the design as well as our assumptions regarding future regulations for the disposal of waste materials.

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Figure Captions

1. Liquid scintillation spectra for ^{55}Fe from the ^{56}Fe(n,2n) reaction: The initial dissolution (DS); the ion-exchanged solution (IX); and the ^{55}Fe standard (STD).

2. Liquid scintillation spectra for ^{63}Ni from the ^{63}Cu(n,p) and ^{64}Ni(n,2n) reactions:

The initial dissolutions (DS); ion-exchange products (IX); and ^{63}Ni standard (STD).

Table I: Long-lived Activities in Fusion Materials

Isotope	Half-life,y	Reactions	Status
^{14}C	5730	$^{14}N(n,p)$	in progress
^{26}Al	$7.2x10^5$	$^{27}Al(n,2n)$	Ref 4
^{53}Mn	$3.7x10^6$	$^{54}Fe(n,2n)\beta$	Ref 5
		$^{54}Fe(n,d+np)$	Planned
^{55}Fe	2.73	$^{56}Fe(n,2n)$	This work
^{63}Ni	100.	$^{63}Cu(n,p)$	This work
		$^{64}Ni(n,2n)$	This work
^{59}Ni	$7.6x10^4$	$^{60}Ni(n,2n)$	This work
^{93}Zr	$1.5x10^6$	$^{94}Zr(n,2n)$	In progress
		$^{93}Nb(n,p)$	In progress
^{92}Nb	$3.2x10^7$	$^{93}Nb(n,2n)$	In progress
^{91}Nb	700.	$^{92}Nb(n,2n)$	Ref. 2
^{94}Nb	$2.0x10^4$	$^{94}Nb(n,p)$	Ref. 2
^{93}Mo	3500.	$^{94}Mo(n,2n)$	In progress

Table II: Results for $^{56}Fe(n,2n)^{55}Fe$

Sample	Energy[a] MeV	Fluence $10^{17}n/cm^2$	At. Ratio 10^{-8}	σ mb[b]
^{56}Fe-1	14.83	1.88	8.58	456.
^{56}Fe-2	14.82	1.61	7.34	456.
^{56}Fe-3	14.85	1.31	5.86	448.
Fe-1	14.68	10.30	48.6	472.
Fe-2	14.81	4.53	19.9	440.

Results for $^{63}Cu(n,p)^{63}Ni$

Sample	Energy[a] MeV	Fluence $10^{17}n/cm^2$	At. Ratio 10^{-8}	σ mb[c]
Cu-1	14.65	1.17	6.40	54.7
Cu-2	14.65	1.14	6.16	54.0
Cu-3	14.82	0.462	2.45	53.0

Results for $^{64}Ni(n,2n)^{63}Ni$

Sample	Energy[a] MeV	Fluence $10^{17}n/cm^2$	At. Ratio 10^{-7}	σ mb[c]
^{64}Ni-1	14.85	1.96	1.90	967.
^{64}Ni-2	14.83	1.65	1.57	948.

Results for $^{60}Ni(n,2n)^{59}Ni$

Sample	Energy[a] MeV	Fluence $10^{17}n/cm^2$	At. Ratio 10^{-8}	σ mb[d]
^{60}Ni-1	14.81	1.26	1.57	124±9
^{60}Ni-2	14.83	1.78	1.73	97±6
^{60}Ni-3	14.82	1.34	0.95	84±9

[a]Mean energy; width ≈0.5 MeV

[b]Uncert.: Stat. 1%, eff. 3%, std. 5%, $T_{1/2}$ 1.1%, fluence 5%, net 7.8%

[c]Uncert.: Stat. 1%, eff. 3%, std. 2.5%, $T_{1/2}$ 2.0%, fluence 5%, net 6.7%

[d]Uncert.: Stat. above, eff. 4%, $T_{1/2}$ 17%, fluence 5%, net 19-23%

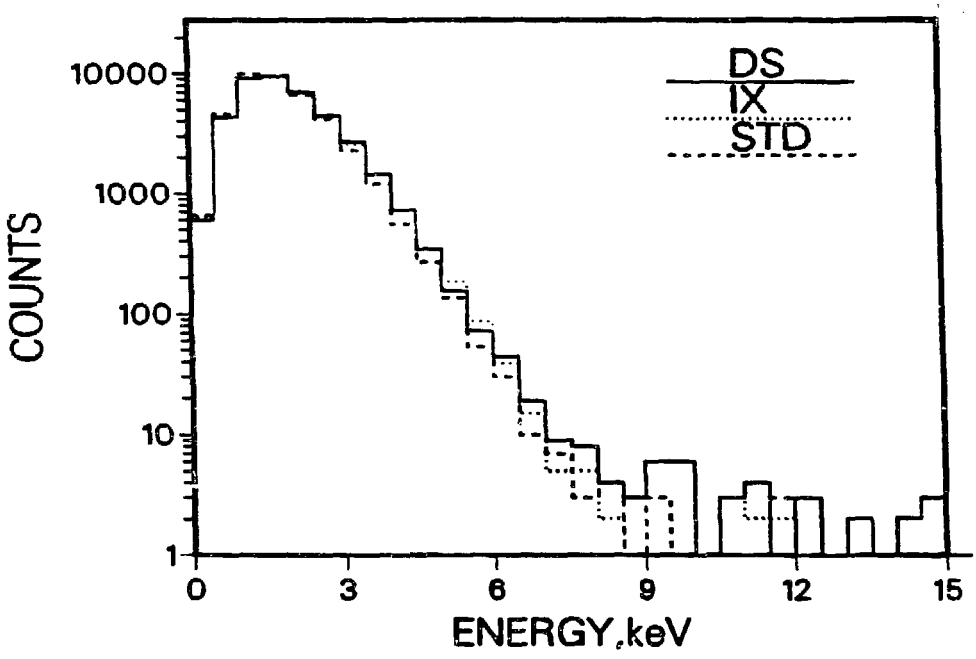
Table III: Summary and Comparison of Results with Previous Data

	Present		Previous		
Reaction	E_n	σ,mb	E_n	σ,mb	Ref.
$^{56}Fe(n,2n)^{55}Fe$	14.8	454±35	14.0	440±90	9
			14.3	410±33	10
			14.7	440±40	11
			14.6	480±50	12
			15.1	190±40	13
$^{63}Cu(n,p)^{63}Ni$	14.7	54±4	14.7	125±50	11
$^{63}Cu(n,px)^{a}$			14.0	105±9	14
			14.1	149±30	15
$^{64}Ni(n,2n)^{63}Ni$	14.8	958±64		none	
$^{60}Ni(n,2n)^{59}Ni$	14.8	104±25		none	
$^{94}Mo(n,p)^{94}Nb$	14.7	55±6		none	
$^{Nat}Mo(n,x)^{94}Nb$	14.7	7.8±0.8		none	
$^{Nat}Mo(n,x)^{91}Nb$	14.7	≈45		none	

[a]Total proton yield from ^{63}Cu includes other reactions

Table IV: Fusion Reactor Activation Calculations
(STARFIRE design, 21.6 MW-y/m^2, 3000 day cooling)

Material	Isotope	Activity,mCi/cc
Iron	^{55}Fe	25,000.
Copper	^{63}Ni	1795.
Nickel	^{63}Ni	227.
	^{59}Ni	0.99
Molybdenum	^{91}Nb	122.
	^{94}Nb	0.77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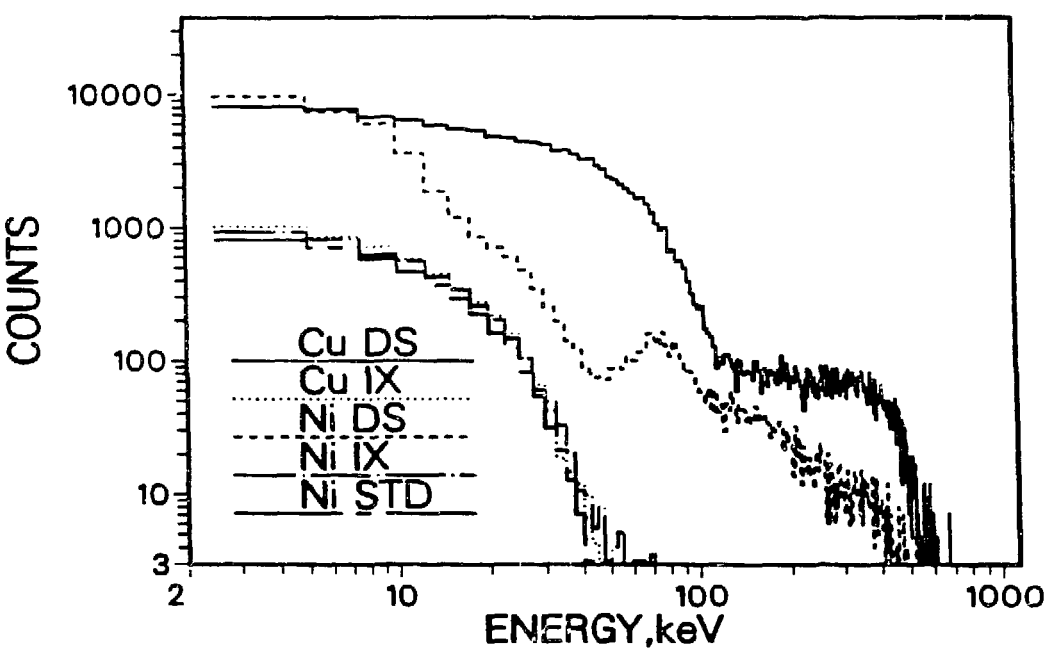
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