A SEARCH FOR LONG–LIVED RADIONUCLIDES PRODUCED BY FAST–NEUTRON IRRADIATIONS OF COPPER, SILVER, EUROPIUM, TERBIUM, AND HAFNIUM*

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ABSTRACT

Identical sample packets, each containing samples of elemental copper, silver, europium, terbium, and hafnium, as well as titanium, iron and nickel as dosimeters, have been irradiated in three distinct accelerator neutron fields (at Argonne National Laboratory and Los Alamos National Laboratory in the U.S.A., and Japan Atomic Energy Research Institute, Tokai, Japan) as part of an interlaboratory research collaboration to search for the production of long–lived radionuclides for fusion waste disposal applications. This paper is a progress report on this project. To date, we have detected the following activities, and have obtained preliminary experimental cross section values for several of these: Ag–106m,108m,110m; Eu–150m,152g,154; Tb–158,160; and Hf–175,178m2,179m2,181.

I. INTRODUCTION

A recommendation was made at the 16th Meeting of the International Nuclear Data Committee (INDC), held at Beijing, People's Republic of China, 12–16 October 1987, to establish a Coordinated Research Program (CRP) entitled "Measurements and Calculations of Activation Cross Sections for Long–Lived Radionuclides Important for Radioactive Waste Estimates in Fusion Reactor Technology". The list of reactions to which the attention of this CRP is devoted is given in Table I. These reactions have been identified as being potentially troublesome in fusion nuclear waste disposal considerations. The present investigation was undertaken in support of this project.

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Table I: List of Activation Reactions Included in the IAEA Coordinated Research Program

Reaction	Half–Life (years)
Al–27(n,2n)Al–26	7.2(5)[1]
Cu–63(n,p)Ni–63	100.1
Mo–94(n,p)Nb–94	2.03(4)
Ag–109(n,2n)Ag–108m	127
Hf–179(n,2n)Hf–178m2	31
W–182(n,n'α)Hf–178m2	31
Eu–151(n,2n)Eu–150m	35.8
Eu–153(n,2n)Eu–152g	13.33
Tb–159(n,2n)Tb–158	150
Dy–158(n,p)Tb–158	150
Ir–193(n,2n)Ir–192m2	241
Re–187(n,2n)Re–186m	2.0(5)

[1] 7.2(5) signifies 7.2×10^5.

The original plan of this work was to conduct measurements solely at Argonne National Laboratory (ANL), employing the Fast Neutron Generator Accelerator Facility (FNG) [1] and the thick–target Be–9(d,n)B–10 reaction as an intense neutron source [2]. Using 7–MeV deuterons, this source produces a continuous spectrum of neutrons from < 200 keV to 11.4 MeV. It was reasoned that integral activation data acquired from such measurements would be useful to supplement the 14–MeV results likely to be provided by other participants in this CRP. As the work at Argonne progressed, two collaborators from other laboratories (RCH and YI) joined the Argonne investigators (JWM, DLS and LRG) in this endeavor. The scope of this work was then expanded to include irradiations at Los Alamos National Laboratory (LANL), in the quasi–monoenergetic 10–MeV neutron field produced by the H–1(t,n)He–3 reaction [3] at the LANL Ion Beam Facility [4], and at the Japan Atomic Energy Research Institute (JAERI), Tokai, in the 14–MeV field produced by the JAERI Fusion Neutronics Source Facility (FNS) [5].

As a prelude to the experimental program, nuclear–model cross–section calculations were performed for the reactions in Table I using the pre–compound, statistical model code ALICE [6]. For the (n,2n) processes, the total cross–section strengths derived by this method were allocated among the isomeric states according to the distribution of J in the Gilbert and Cameron level density formula [7]. This is a crude approximation, but it does serve to reduce the weight of high–spin states. These calculations indicated that it would not be practical to produce sufficient activity for reliable measurements (given the available irradiation conditions) for those CRP reactions involving aluminum, molybdenum, dysprosium, tungsten, rhenium, and iridium. Thus, it was decided to abandon these for the present, and to concentrate entirely on investigating those radionuclides produced by fast–neutron irradiation of copper, silver, europium, terbium, and hafnium. Calculated results for several reactions associated with the selected elements are given in Table II, in terms of spectrum–average cross sections for the primary neutrons from two of the accelerator neutron sources employed in this investigation (at ANL and LANL).

Table II: Calculated Spectrum–average Cross Sections (in Millibarns) for Primary Neutrons Associated with the Indicated Neutron Fields[1]

Reaction	H(t,n)[2]	Be(d,n)[3]
Ag–107(n,2n)Ag–106m	87	2
Ag–109(n,2n)Ag–108m	151	3
Eu–151(n,2n)Eu–150m	1264	30
Eu–153(n,2n)Eu–152g	647	12
Tb–159(n,2n)Tb–158	1400	54
Hf–176(n,2n)Hf–175	1571	34
Hf–179(n,2n)Hf–178m2	22	1
Hf–180(n,2n)Hf–179m2	159	5

1 Based on ALICE calculations as described in Section I.

2 For this experiment primary neutrons from the H(t,n) source have energies in the vicinity of 10 MeV [3].

3 For this experiment continuum neutrons from the ANL thick–target Be(d,n) source have energies ranging from < 200 keV to 11.4 MeV [2].

II. SAMPLES

Since it was essential for the success of this experiment to acquire the maximum possible neutron doses in the time available for each of the sample materials, it was decided to irradiate the samples simultaneously in a sample packet that also contained dosimeter foils for determining the neutron fluence. Several identical sample packets were prepared (as shown in Fig. 1) for irradiation at the three accelerator facilities. These packets were 2.54–cm in diameter and $\leq$ 2 cm thick. The europium, terbium, and hafnium samples were in the form of oxides, since these rare–earth materials are otherwise rather reactive. These oxides were encapsulated in sealed plastic containers which defined the sample geometries and prevented any loss of material. All the other sample materials were in the form of metal foils. Several nickel samples were included in each packet in order to measure the neutron fluence gradients (see Fig. 1). A group of copper, nickel, iron, and titanium foils, placed at the rear of the sample packet, provided nine reactions with well–determined excitation functions that could be used to either confirm or determine the neutron spectrum. The chemical purity of all samples was > 99.9%.

III. IRRADIATION PROCEDURES

A single sample packet was placed on the beam line at zero degrees for the irradiations which took place at ANL and LANL. At ANL, the individual samples were located between 5–7 cm from the target, while at LANL the corresponding distances were 4–6 cm. Two sample packets were irradiated at JAERI (one to be retained in Japan for activity analysis and the other to be returned to the U.S.A.). These packets were

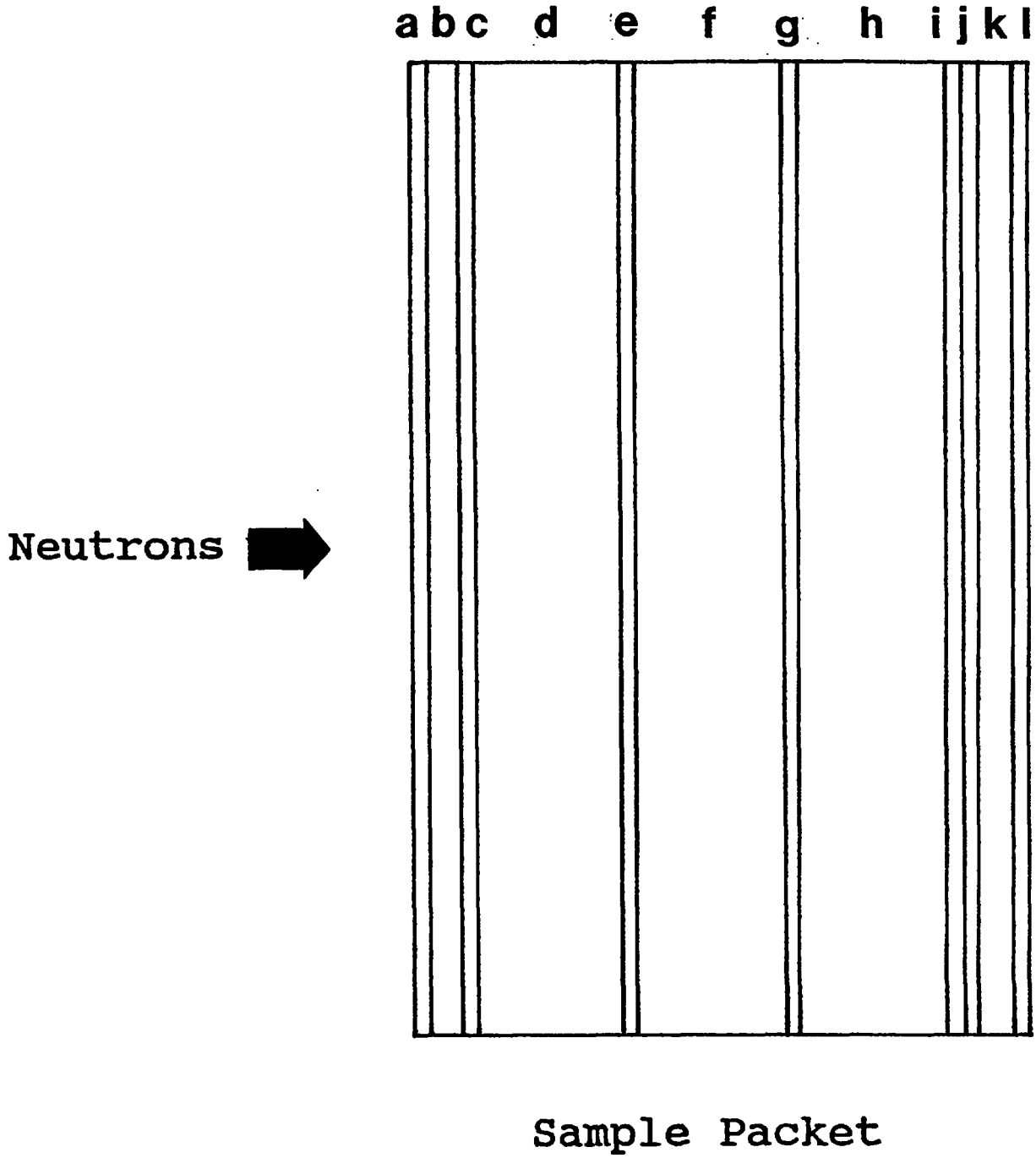

Fig. 1: Schematic diagram for the sample packets used in each of the neutron irradiations. The samples materials are as follows: a = Ni(1), b = Ag, c = Ni(2), d = HfO_2, e = Ni(3), f = Tb_4O_7, g = Ni(4), h = Eu_2O_3, i = Ni(5), j = Cu, k = Fe, l = Ti. The oxide samples are contained in plastic capsules.

Table III

Decay Properties and Other Data [10,11]
for the Radionuclides and Nuclear Reactions
Considered in this Experiment

Activity	Reaction	Half-Life	Principal γ Energy (MeV)	γ Branch (%)	Isotopic Abund. (%)	σ thermal (b)	Iγ (b)	Jπ
Ag–106m	Ag–107(n,2n)	8.46±.10 d	0.512	88±3	51.8	—	—	6+
Ag–108m	Ag–109(n,2n)	127±21 y	0.434	90.5±.6	48.2	—	—	6+
	Ag–107(n,γ)				51.2	0.33	1.2	
Ag–110m	Ag–109(n,γ)	249.8±.04 d	0.658	94.6±1.9	48.2	4.7	72.3	6+
Eu–150m	Eu–151(n,2n)	35.8±1.0 y	0.334	94.0±1.9	46.8	—	—	0–
Eu–152g	Eu–153(n,2n)	13.33±.04 y	0.344	26.6±.5	52.2	—	—	8–
	Eu–151(n,γ)				47.8	5900	1510	
Eu–154	Eu–153(n,γ)	8.8±.1 y	1.275	35.5±.7	52.2	312	1420	—
Tb–158	Tb–159(n,2n)	150±30 y	0.944	43 ± 3	100.0	—	—	—
Tb–160	Tb–159(n,γ)	72.3±.2 d	0.879	29.8±1.5	100.0	23.4	418	—
Hf–175	Hf–176(n,2n)	70±2 d	0.343	87.0±.5	5.2	—	—	—
	Hf–174(n,γ)				0.16	561	436	
Hf–178m2	Hf–179(n,2n)	31±1 y	0.326	94.1±1.9	13.7	—	—	16
	Hf–177(n,γ)				18.6	.0002	?	—
Hf–179m2	Hf–180(n,2n)	25.1±.3 d	0.453	66±3	35.2	—	—	25/2
	Hf–178(n,γ)				27.1	?	?	
Hf–181	Hf–180(n,γ)	42.39±.06 d	0.482	80.6±.7	35.2	13.0	350	—

positioned symmetrically on opposite sides of the beam line at equal distances from the target, so that they would experience equivalent neutron fields and receive about the same dose. The distance from the neutron source to the front surfaces of the packets was 7 cm. They were oriented at an angle of 43 degrees relative to the incident deuteron beam.

The measurements at ANL and JAERI were entirely passive, in the sense that no data were recorded in real time with any active detectors. The dosimeter reactions employed to passively determine the total neutron fluence were as follows: Ti–46,47,48(n,p)Sc–46,47,48, Fe–54(n,p)Mn–54, Fe–54(n,α)Cr–51, Ni–58(n,p)Co–58, and Cu–63(n,α)Co–60. At LANL, a fission chamber fluence monitor was also employed during the irradiations as an auxiliary neutron monitor. This detector contained a uranium deposit with a total mass of 991 μg of uranium. The isotopic composition was: U–238 (99.585%); U–235 (0.415%), others negligible. The irradiation at ANL consisted of about 110 hours of cumulative exposure, occurring over a period of nearly a month during

Table IV

Fluence as Measured by
Uncorrected Spectral Index Reactions[1]

Reaction	Fluence (neutrons/cm^2) ANL (x10^{15})	LANL (x10^{13})
U–238(n,f)	—	2.26
Cu–63(n,α)Co–60	0.578	1.67
Ni–58(n,p)Co–58	0.681	2.14
Fe–54(n,p)Mn–54	0.649	2.12
Fe–54(n,α)Cr–51	0.647	1.47
Ti–48(n,p)Sc–48	0.679	1.30
Ti–47(n,p)Sc–47	0.586	1.44
Ti–46(n,p)Sc–46	0.632	1.74

[1] Note that the Ti, Fe, Ni and Cu foils used for spectral indexing (labeled "i", "j", "k", and "l" in Fig. 1) all have about the same geometry factors since they are close together in the packets. Furthermore, they were positioned very near to the U–238 deposit during the LANL irradiation.

November and December 1988. Preliminary estimates of the neutron doses to the samples from the ANL exposures fall in the range $(0.58 - 0.68) \times 10^{15}$ neutrons/cm^2 (as deduced from the spectral index results given in Table IV). The irradiation at LANL involved about 14 hours of cumulative exposure, gained in two irradiations during October and November 1988. The LANL irradiations involved doses to the samples which appear to be in the range $(1.3 - 2.3) \times 10^{13}$ neutrons/cm^2 (see Table IV). The JAERI irradiations took place during the time period 20–23 June 1989. A total of 32 hours (divided into four 8–hour intervals) of beam time were employed in the experiment, resulting in an estimated dose of 2×10^{14} neutrons/cm^2.

IV. ACTIVITY MEASUREMENTS

One sample packet is being retained at JAERI for activity analysis. Otherwise, all sample counting is taking place at ANL. Since each of the sample irradiations involved time intervals extending from several days to several weeks (often with extensive interruptions), and since the counting of some of the samples was delayed to accommodate sample transport back to Argonne (from LANL and JAERI), no attempt has been made in the ANL counting to study any radionuclides with half–lives shorter than a few days. Table III shows the activities that are being observed at ANL in this experiment and indicates some of their principal features. All of the activity measurements involve gamma–ray counting with germanium detectors, except for Ni–63 which is a beta emitter with no gamma rays. Analysis of the Ni–63 activity has been deferred, pending availability of support to fund the liquid scintillation counting analysis that will be needed

for this purpose. As of this writing, some of the longer–lived gamma–ray activities are still not measurable for the samples irradiated at ANL and LANL because of interference from shorter–lived species. The error for most of the activity measurements is 2–4%. The samples irradiated at JAERI have not yet been counted at ANL.

V. DATA ANALYSIS.

The analyses performed to date on the data include corrections for activity decay and approximate corrections for geometry and neutron absorption. Information required for the determination of these corrections has been obtained from Refs. 8–11. It should be noted from Table III that there are significant uncertainties in the half–lives of several of the radionuclides and, to a lesser degree, in the branching ratios. This will have a marked effect on the ultimate accuracies that can be expected for the derived cross sections.

So far, no corrections have been applied for neutron multiple scattering in the samples, and for the background produced by target–associated reactions or by room–return neutrons. These matters are presently under investigation, and it is clear that they will involve nontrivial corrections. The sample packets are thick (transmissions of about 90%), implying multiple scattering corrections on the order of 10%. Background corrections at the ANL Be(d,n) facility are small. Previous measurements have shown that room–return in the MeV energy range is not a serious problem. Furthermore, neutrons produced by other target associated reactions are overwhelmed by the Be(d,n) yield and, in any case, are included in the measured spectrum. Corrections for these backgrounds are small and are based on previous measurements. For the LANL H(t,n) source, there is a significant neutron yield from (t,n) reactions with the target structure. These neutrons extend to energies well above the H(t,n) peak. The correction for a particular reaction depends on the shape of the excitation function and may be rather large.

Room–return at thermal and epithermal energies does present a troublesome problem in correcting the ANL and LANL data and may, upon examination, turn out to be a problem for the JAERI data. Some activities are produced by both the (n,γ) and (n,2n) reactions. The (n,γ) cross section is quite small in the MeV region and usually causes no serious problem in the measurement of (n,2n) cross sections. However, these materials have rather large thermal capture cross sections and resonance capture integrals (see Table III), so a small thermal and epithermal component in the total neutron spectrum can cause a serious problem. Five of the radionuclides observed can be produced by both reactions. Two of them (Hf–178m2 and Hf–179m2) should present no problem as these states have very high spins (see Table III) and are unlikely to be populated by s– or p–wave capture, but the other three may require correction. Fortunately, four activities were observed that can only be produced by (n,γ) reactions, and these can be used to estimate the corrections for the others. A preliminary estimate of the thermal fluence was made using the thin absorber approximation plus the capture data in Table III. The samples involved are by no means thin, but fairly consistent results were obtained. The thermal and epithermal fluence was about 0.01% of the total for the ANL measurement, and about 0.02% for the LANL measurement. At these levels, only the Eu–152g yield will need a significant correction. Unfortunately, the thermal capture cross section and resonance integral are large and most of that activity is produced by the (n,γ) reaction. It will be very difficult to obtain reliable Eu–153(n,2n) cross sections without using separated isotopes.

The neutron fluence, based on idealized spectra, was calculated for the eight spectral–index reactions, and the agreement indicates how well the actual neutron spectra agree with the assumed spectra. For the ANL measurement, the calculated fluences are fairly consistent (see Table IV). The lowest and highest values differ by less than 20%, and this should improve once the multiple scattering corrections are made. For the LANL measurement, the lowest and highest values differ by nearly 75% (see Table IV). This is much greater than the expected multiple scattering effect, and it is most probably due to the undetermined target–associated neutron background.

VI. CURRENT STATUS

Preliminary cross sections have been derived from the measurements at ANL and LANL, but we choose to not report these data because several important corrections have not been applied and characterization of the neutron spectra associated with these measurements is still incomplete. These results will also eventually be supplemented by data on the production of at least two additional long–lived activities, once the counting of the samples is complete, and by 14–MeV results from the samples irradiated at JAERI.

VII. PENDING WORK

Several matters must be attended to before this experiment will have been completed. The counting of long–lived activities for the samples from the ANL and LANL irradiations is still pending, as well as all sample counting for the JAERI exposures. More detailed corrections for neutron absorption and geometric effects are required, and the effects of multiple scattering must be estimated. The detailed nature of the neutron fields employed in this investigation has to be determined in order to calculate certain corrections associated with both primary and secondary neutron fields, and to refine the estimates of interfering contributions from (n,γ) reactions which come about mainly from lower–energy neutrons.

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