MASTER

Program: Fusion Program — Development and Technology

ERDA Task: Task 27 — Helium Generation in Fusion Reactor Materials

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Reporting Period: October 1976 - March 1977 **Category:**

General Order: 09077

Subaccount: 10000

189a Number: 16801

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I. PROJECT OBJECTIVES

1. Determine the helium generation rates and spectrum-integrated (n, α) cross sections of a number of pure elements, isotopes, and alloys in neutron environments being used for fusion reactor development.
2. Use this information to predict the helium production rates in any selected fusion reactor wall materials.
3. Develop helium accumulation fluence monitors for measuring neutron fluences in fusion experimental and blanket irradiations.

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II. SUMMARY

During this report period a topical report entitled "A Neutron Fluence Map for the Rotating Target Neutron Source," (AI-ERDA-13188) was completed and distributed. The report, which included R. A. Van Konynenburg of Lawrence Livermore Laboratory (LLL) and R. R. Heinrich of Argonne National Laboratory (ANL) as coauthors, provided a detailed description of the fluence distribution for the high-flux region of the Rotating Target Neutron Source (RTNS-I) at LLL. The report also included new 14.8-MeV neutron cross section data for the Ni, Nb, and Au dosimetry reactions. These results are summarized in Section III.

Section III also includes a description of the preliminary analysis of wire rings included in the irradiation capsule for helium accumulation fluence dosimetry. The results show non-uniformities in the neutron fluence distribution characterizing this irradiation, and point out the value of helium accumulation fluence dosimetry for characterizing high energy (~ 5 to 30 MeV) neutron fields. The helium accumulation fluence data are being used to adjust the neutron fluence map for calculation of final helium generation cross sections from the other RTNS-irradiated pure element specimens.

Most of the specimens irradiated in the RTNS-I experiment have been analyzed for helium, and preliminary results are presented in Section IV.B. Before performing the mass spectrometric helium analyses, rough calculations were made to estimate the fractions of helium generated by 15-MeV neutron-induced reactions that would be lost by α -recoil. It was found that for thin samples (less than ~ 5 mils thick), a significant fraction of the helium would be lost, and in the case of stacked thin foils, this helium would be injected into adjacent foils. For the "chunk" specimens in this experiment, however, the surface material that was depleted (or enhanced) by α -recoil effects could generally be etched away with acids, leaving a core region for helium analysis that required no corrections for this phenomenon. Details of the calculations are presented in Section IV.A.

Preparations are being made to fabricate and irradiate a larger package containing 17 pure elements, four or more alloys, and a number of separated isotopes, in the broad-band neutron spectrum provided by the d-Be source at

the Crocker Nuclear Laboratory of the University of California at Davis, California (UC-Davis). This package, described in Section V, will contain an extensive set of radiometric dosimetry foils with different energy sensitivities, plus eleven wires of five pure elements for helium accumulation fluence dosimetry. Personnel from ANL and LLL will participate in this irradiation, from which it is planned to produce both an extensive set of helium generation cross sections and a detailed characterization of the d-Be neutron environment.

Two other irradiation experiments have been initiated in support of the d-Be irradiation. A second RTNS-I irradiation has been performed, with a capsule geometry similar to that planned for the d-Be experiment. This will provide a reference point for interpretation of the dosimetry and helium generation results from the more complex d-Be neutron spectrum. Two capsules were also loaded for irradiation in the High Flux Isotopes Reactor (HFIR) at Oak Ridge National Laboratory, in order to correlate the total helium generation in the d-Be environment with that fraction produced by the low energy neutron component. These experiments are described briefly in Section VI.

Personnel from Atomics International have participated in the ERDA-DMFE Task Group on Damage Analysis and Fundamental Studies. Section VII provides some details of the scope and activities of the task group.

Section VIII summarizes the concluding work of two other Task 27 programs, for which the funding was terminated at the end of FY 1976. These programs were unrelated to the present project scope, but were completed as a part of the continuing Task 27 program.

III. RTNS-I NEUTRON FLUENCE DOSIMETRY

The accurate determination of spectrum-integrated helium generation cross sections requires an accurate characterization of the neutron fluence over the volume occupied by the irradiation samples. This led to the design of detailed dosimetry procedures for an irradiation of several pure elements at the Rotating Target Neutron Source (RTNS-I) at the Lawrence Livermore Laboratory (LLL). Analysis of the foil activation dosimetry information has provided the most detailed study yet undertaken of the innermost fluence variation of the RTNS-I neutron field. It also has provided a procedure for determining neutron fluence profiles in other neutron irradiations, and has given new relative cross sections for the neutron dosimetry reactions used in this experiment.

A second potentially valuable method of characterizing high energy (~ 5 to 30 MeV) neutron environments was also investigated. This method, termed "Helium Accumulation Fluence Dosimetry," is based on the post-irradiation measurement of the neutron-induced helium production in segments of a suitable dosimetry material. Detailed fluence profiles can be determined by mapping the relative helium generation in the segments. Total neutron fluences can then be obtained by folding in the total helium production cross sections. While this analysis method is more time-consuming than activation analysis, it has several advantages that make it a valuable dosimetry tool complementary to foil activation techniques. These include the ability to use finer segments, the potential for obtaining neutron energy information by selecting several dosimetry materials that have helium-generation cross sections with different neutron energy sensitivities (also available to foil activation), and the lack of dependence on the time-history of the irradiation. The time-history is important for activation analysis, on the other hand, because short-lived activities may provide no more than flux measurements of the latter part of the irradiation, rather than fluence measurements integrated over the whole irradiation period. Partial analysis of the helium generated in pure element wire rings irradiated in the RTNS-I experiment has demonstrated the value of the helium accumulation technique.

A. DESCRIPTION OF IRRADIATION ASSEMBLY

The geometry of the stainless steel helium-generation assembly irradiated in RTNS-I is shown in Figure 1. It contained two aluminum wire rings, one copper wire ring, and many small pure element and alloy specimens, sandwiched between two stacks of three thin dosimetry foils (Au, Nb, and Ni). A stack of five thicker foils (Au, Nb, Ni, Fe, and Pt) and an iron ring were placed behind. The assembly was irradiated for 77.55 h, giving a total neutron fluence of $2.6 \times 10^{17} \text{ cm}^{-2}$ at the capsule front center. It was then disassembled and the dosimetry foils were segmented for γ -ray counting at Argonne National Laboratory and LLL and for weighing at ANL and AI. The wire rings were etched and segmented, and most of the segments, plus the helium specimens, have been weighed and analyzed for their helium content by high sensitivity gas mass spectrometry.

B. FLUENCE MAP FROM FOIL ACTIVATION MEASUREMENTS

The foil counting and weighing results were combined with the irradiation time-history to calculate the neutron fluences received by the individual dosimetry foil segments. These fluences and the foil segment geometries were then used to construct a fluence map for the space occupied by the AI helium generation capsule for this irradiation. The mapping procedure is described in detail in the report "A Neutron Fluence Map for the Rotating Target Neutron Source,"⁽¹⁾ completed during this report period, and coauthored by R. A. Van Konynenburg (LLL) and R. R. Heinrich (ANL).

The map was constructed by first calculating the offset of the irradiation assembly relative to the 0° neutron source axis. This offset was determined from recording microphotometer scans of autoradiograph negatives made of the assembly after the irradiation, and from quadratic surface fits to the average fluences calculated for the gold foil segments. This information was then used to calculate radial neutron fluence profiles for the gold foils sandwiching the helium specimens, in the form:

$$\Phi(R) = C_1 + C_2 R + C_3 R^2 + C_4 R^3, \quad \dots (1)$$

FIGURE 1a
PHOTOMACROGRAPH
OF LOADED INTERIOR

FIGURE 1b
DESCRIPTION OF PHOTOMACROGRAPH

9077-4001

Figure 1. Helium-Generation Assembly Irradiated at the RTNS-I

where R is the radial distance from the 0° neutron source axis. The constants were evaluated by integrating $\Phi(R)$ numerically over the area of each foil segment, and solving several sets of simultaneous equations. The results are shown in Figure 2, where they have been corrected for neutron attenuation to represent "free space" profiles.

6506-4010B

Figure 2. Attenuation-Corrected (Free-Space) Radial Cubic Fits to the Front and Middle Gold Dosimetry Foil Data

The calculations were then extended to find a single three-dimensional expression that reproduced the fluence profiles of all nine Au, Nb, and Ni dosimetry foils:

$$\Phi(R, \Delta Z) = \alpha \left[C_1 + C_2 \beta R + C_3 (\beta R)^2 + C_4 (\beta R)^3 \right]. \quad \dots (2)$$

Here ΔZ is the axial distance from the front face of the capsule, β is a correction factor for the changing radial shape of the fluence curve as a function of ΔZ , and α is the normalization of the radial profile as a function of the distance from the neutron source. Equation 2, which reduces to the front gold foil radial profile at that foil position (for the constants chosen), is plotted for the middle gold foil radial profile as a dashed line in Figure 2.

The axial ($R = 0$) dependence of $\Phi(R, \Delta Z)$ is shown as the solid line in Figure 3. This figure also includes the axial fluences determined for all nine foils. These fluences were obtained from Equation 2 and normalized by equating the integral of Equation 2 with the total measured foil fluences. This normalization eliminated the Z -dependence of Equation 2, and thus gave the measured Z -dependence of the axial neutron fluence. This Z -dependence was found to be linear over the 2-mm thickness of the capsule, and is attributable to the finite neutron source spot size.

C. DOSIMETRY REACTION CROSS SECTIONS FOR 14.8-MeV NEUTRONS

Figure 3 presents a direct comparison of the axial neutron fluences derived from the Au, Nb, and Ni dosimetry foils. This comparison shows that the cross sections used for the $^{58}\text{Ni}(n, p)^{58}\text{Co}$ and $^{58}\text{Ni}(n, 2n)^{57}\text{Ni}$ reactions⁽²⁾ are $(7.4 \pm 2.3)\%$ high relative to the $^{197}\text{Au}(n, 2n)^{196}\text{Au}$ and $^{93}\text{Nb}(n, 2n)^{92\text{m}}\text{Nb}$ cross sections.^(3, 4, 5) This information made it possible to calculate relative cross sections for these reactions. This required further analysis, however, because the two nickel cross sections are strongly energy-dependent over the experimental geometry, and the incident neutron energy at any point in the capsule was an average over a large reaction solid angle.

The average neutron energy incident upon the capsule was calculated at each of four foil positions, by performing a coarse numerical integration over the area of the RTNS-I source spot. For these calculations, the source spot was assumed to be 10 mm in diameter, and the capsule front face was assumed to be 3.5 mm from the source. The large neutron energy variation introduced by this close proximity to the source is readily seen: the axial position on the front nickel dosimetry foil, for example, intercepted neutrons with source laboratory angles of 0° to 54° . This angular variation corresponds to laboratory neutron energies of 15.0 to 14.6 MeV, respectively, as determined from

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Figure 3. Attenuation-Corrected Axial Neutron Fluence Profile

reaction kinematics.⁽⁶⁾ These neutron energies are also averages, derived from the neutron energy distribution produced by deuterons slowing down in the thick titanium-tritium target. The average neutron energy was determined for each foil position by folding incremental source area and solid angle factors into the laboratory neutron energy variation, assuming a uniform deuteron beam profile. The average axial neutron energies were found to be (neglecting uncertainties) 14.81, 14.84, and 14.86 MeV at the front nickel, middle nickel, and back niobium foils, respectively. (See Figure 3 for the foil geometry.) The average neutron energy at the edge of the front nickel foil was calculated to be 14.69 MeV. The 0.12-MeV average neutron energy variation across this foil is consistent with the experimentally-determined average energy variations between the front nickel foil segments.⁽¹⁾

The new nickel cross section values were determined by multiplying the off-axis cross section values used in the fluence calculations, extrapolated to 0° , by the normalization factor (0.931 ± 0.020) obtained from the axial fluence comparison with the gold and niobium results. The axial cross section extrapolation was made by extrapolating the experimental neutron energies (derived from the nickel foil segment disintegration ratios) to 0° , and finding the corresponding cross sections in the same tabulations as used for the fluence calculations.⁽²⁾ This procedure gave the two nickel cross sections, even though only one set of fluence values was obtained from the nickel foils, because the ratio of the disintegration rates at each energy provided a direct measure of the cross section ratio at that energy. As expected, the experimentally-determined average axial neutron energy (15.0 ± 0.1 MeV) did not agree with the value determined from kinematics (14.8 MeV), because of the cross section discrepancy.

The relative dosimetry reaction cross sections determined from this experiment are listed in Table 1, Column 3, normalized to the 458-mb $^{93}\text{Nb}(n, 2n)^{92m}\text{Nb}$ cross section. The quoted uncertainties are relative values, omitting the $\pm 5\%$ niobium cross section uncertainty.

TABLE 1
NEUTRON CROSS SECTIONS OF ^{58}Ni , ^{93}Nb ,
AND ^{197}Au AT 14.8 AND 15.0 MeV

Reaction	Cross Sections (mb) and 1σ Relative Uncertainties			
	14.8 MeV		15.0 MeV	
	Literature Values	This Work Normalized to Nb (Ref. 5)	Unpublished Data (Ref. 7)	This Work Adjusted to 15 MeV*
$^{58}\text{Ni}(n, p)^{58}\text{Co}$	329 (Ref. 2)	294 ± 8	283	289 ± 8
$^{58}\text{Ni}(n, 2n)^{57}\text{Ni}$	35.1 (Ref. 2)	34.9 ± 1.3	—	38.2 ± 1.4
$^{197}\text{Au}(n, 2n)^{196}\text{Au}$	2110 (Ref. 3, 4)	2110 ± 40	2150	2160 ± 40
$^{93}\text{Nb}(n, 2n)^{92m}\text{Nb}$	458 (Ref. 5)	458 [†]	469	469 [†]

*See text

†The results of this work (Columns 3 and 5) are normalized to the Nb cross section values in References 5 and 7 (Columns 2 and 4), respectively.

Table 1 also includes a comparison with a recent unpublished set of cross sections measured by Nethaway⁽⁷⁾ at 15.0 MeV. For this comparison, the present results (Column 3) were adjusted to 15.0 MeV and renormalized to Nethaway's $^{93}\text{Nb}(n, 2n)^{92\text{m}}\text{Nb}$ cross section measurements. The adjustment was made by assuming constant $^{93}\text{Nb}(n, 2n)^{92\text{m}}\text{Nb}$ and $^{197}\text{Au}(n, 2n)^{196}\text{Au}$ cross sections in this energy region, and by using the energy dependences given by the ENDF/B-IV evaluations⁽²⁾ for the $^{58}\text{Ni}(n, p)^{58}\text{Co}$ and $^{58}\text{Ni}(n, 2n)^{57}\text{Ni}$ cross sections. The available experimental literature supports the adoption of these energy dependences for the four reactions. Both the $^{197}\text{Au}(n, 2n)^{196}\text{Au}$ and $^{58}\text{Ni}(n, p)^{58}\text{Co}$ cross sections determined in the present work are consistent with Nethaway's results.

D. HELIUM ACCUMULATION FLUENCE DOSIMETRY

The aluminum and copper wire rings were incorporated in the irradiation assembly both for helium generation cross section measurements and for use as helium accumulation fluence monitors. Helium accumulation fluence dosimetry offers a potentially valuable technique for characterizing fusion neutron environments, and this experiment provided an initial application of the technique. The analysis was simplified in this experiment by the fact that the wire rings were essentially symmetrically placed within the irradiation assembly. This meant that the incident neutron energy, and hence the helium production cross section, could be assumed constant over the entire volume of each wire ring.

The copper, inner aluminum, and outer aluminum rings were etched and cut into 25, 9, and 14 segments, respectively. Most of these segments have been analyzed for helium, but a few copper specimens are presently at ANL for radiometric analysis of the $^{63}\text{Cu}(n, \alpha)^{60}\text{Co}$ reaction. Preliminary information from the copper ring specimens is shown in Figure 4, as a function of angular orientation about the capsule center (in the plane perpendicular to the 0° neutron source axis). The aluminum ring results provide a similar picture. The fluence profile calculated from the dosimetry foil map for the location of the copper ring is superimposed on Figure 4 as the solid curve.

Comparison of the fluence profile with the helium results (normalized in Figure 4 by dividing the helium results by the average calculated cross section)

Figure 4. Preliminary Helium Generation Results and Neutron Fluence Calculations for the Copper Wire Ring

leads to two observations. First, the offset of the capsule from the 0° neutron source axis, as determined from the radiometric dosimetry foils, is too small. The dashed curve in Figure 4, obtained by increasing the assumed vertical capsule offset from 0.6 to 1.0 mm, provides a much better fit to the measured helium data values. Second, the fluence profile is somewhat nonsymmetrical, reflecting non-uniformities in the time-averaged deuteron beam profile. Further analysis indicates that the beam profile was slightly elongated, with some additional structure.

The information obtained from these three rings, plus that from the iron ring, will be used to adjust the dosimetry foil fluence map to reflect the integrated neutron field non-uniformities. This revised map will then be combined with the helium generation data for the pure element helium generation specimens (see Section IV.B) to obtain final helium-production cross sections.

The use of the wire rings to "fine tune" the fluence map demonstrates the value of helium accumulation fluence dosimetry as a complementary tool to foil

activation for neutron characterization in this energy region. The use of wire samples of various elements in ring and other geometries will be expanded in future irradiations. To this end, it is increasingly important to determine accurately the total helium-generation cross sections for other candidate helium dosimetry materials. Multiple elements with different neutron energy sensitivities will be irradiated together in future experiments, and the results will be analyzed using unfolding techniques to obtain neutron energy information. Such information is particularly needed in the neutron environments of the d-Be and d-Li sources, whose energy distributions change significantly over the sample area. Those elements presently being evaluated include ^6Li , ^7Li , ^{10}B , Al, Fe, Ni, Cu, and Au.

IV. HELIUM GENERATION CROSS SECTIONS FOR D-T NEUTRONS

All of the samples irradiated at the RTNS-I in May 1975 for helium generation measurements have been prepared for mass spectrometric helium measurements, and most have been analyzed. The preparations included acid-etching to calculated fractional masses, to remove all possible α -recoil effects from the neutron-induced reactions (see Section IV.A), segmenting several of the samples for multiple helium measurements, and post-etch final weighing using a substitution weighing scheme with standard weights traceable to the U. S. National Bureau of Standards. Additional samples of unirradiated material from each element were similarly prepared. Preliminary cross sections were then calculated from the helium results by combining them with the average neutron fluences at the sample locations, calculated from the dosimetry foil fluence map.⁽¹⁾ These values will be refined when the fluence map has been revised to account for the more detailed fluence information derived from the helium accumulation fluence data obtained from the wire rings, and when corrections are made for recoil effects where they were not removed by etching.

The measurement of helium production cross sections by the mass-spectrometric technique depends on the analysis of the total amount of helium retained by the irradiated sample. The effects of recoiling helium are therefore an important consideration, since they can deplete or enrich the helium content of samples to be analyzed. This is expected to be a small correction to a sample in which the helium recoil range is a small fraction of the sample dimensions; it can be further reduced or eliminated by appropriate etching of the sample surface. However, there are several cases in which recoil effects can introduce large measurement errors. These include, for example, the use of very small sample sizes in which the α -particles have large recoil ranges, and the irradiation of a sample with a low helium-production cross section in close proximity to a material with a high cross section. An investigation of helium recoil was thus initiated, both to estimate the magnitude of the effect on experimental measurements, and as a guide for the design of experiments to minimize the influence of recoil effects.

A. RECOIL CALCULATIONS

Rough calculations have been made to obtain estimates of the effect of recoils on helium retention in samples irradiated at the RTNS-I. The irradiation geometry was approximated by unidirectional, monoenergetic neutron bombardment of a flat slab of thickness w . The fraction of reaction-produced α particles remaining in the slab can be represented by:

$$F_{\alpha}(E_n) = \frac{2\pi \int_0^{\pi} \sigma(\phi) \left[1 - \left| \frac{R(\phi, E_n) \cos \phi}{w} \right| \right] \sin \phi \, d\phi}{2\pi \int_0^{\pi} \sigma(\phi) \sin \phi \, d\phi} \quad \dots\dots (3)$$

This expression assumes a single α -production reaction channel populating a single final state, and it ignores edge effects. Neutron attenuation in the slab, which would have a negligible effect on the results, is also ignored. The angle ϕ is the α -particle laboratory recoil angle measured relative to the beam direction; E_1 is the laboratory neutron energy; $\sigma(\phi)$ is the differential reaction cross section; and R is the range of the α particle. The range is a function of the recoil energy, which in turn is readily expressed as a function of E_1 and ϕ [or alternatively by the center-of-mass (CM) angle θ] using reaction kinematics. The bracket in Equation 3 must be set equal to zero in the regime $R \cos \phi > w$, if it exists in the case of interest.

Two substitutions were used to simplify the numerical evaluation of Equation 3. First, the differential cross section is more conveniently expressed as a function of the CM angle θ :

$$\sigma(\phi) = \sigma(\theta) \frac{d \cos \theta}{d \cos \phi} \quad \dots\dots (4)$$

where

$$\frac{d \cos \theta}{d \cos \phi} = \frac{(\sqrt{G^2 - \sin^2 \phi} + \cos \phi)^2}{G \sqrt{G^2 - \sin^2 \phi}} \quad \dots\dots (5)$$

and

$$G = \left(\frac{m_1 + m_2}{m_1} \right) \sqrt{\frac{m_1}{m_3} \left(\frac{m_4}{m_3 + m_4} \right) \left[\left(\frac{m_2}{m_1 + m_2} \right) E_1 + Q \right] \frac{1}{E_1}}. \quad \dots (6)$$

Here Q is the reaction Q value, and m_1 , m_2 , m_3 , and m_4 are the masses of the neutron, target nucleus, α particle, and heavy residual nucleus, respectively.

The second substitution replaces the integrals in Equation 3 with numerical summations over small angular increments, since $\sigma(\theta)$ and R are generally not available as analytical functions. Choosing an angular increment $2\delta\phi$ sufficiently small that $\sigma(\phi)$ and R remain approximately constant over that interval, $F_\alpha(E_n)$ can be written approximately as:

$$F_\alpha(E_n) \approx \frac{4\pi \sum_i \sigma_i(\theta) \left(\frac{d \cos \theta}{d \cos \phi} \right)_i \left[1 - \left| \frac{R(\phi_i, E_n) \cos \phi_i \cos \delta\phi_i}{w} \right| \right] \sin \phi_i \sin \delta\phi_i}{4\pi \sum_i \sigma_i(\theta) \left(\frac{d \cos \theta}{d \cos \phi} \right)_i \sin \phi_i \sin \delta\phi_i} \quad \dots (7)$$

The denominator is simply the total reaction cross section. Equation 7 can be extended to multiple reaction channels, final states, isotopes, and neutron energies by weighted averages of appropriate sets of F_α 's.

Equation 7 has been evaluated for a few representative cases by making some simplifying assumptions. First, only the (n, α_0) reaction channel was considered, so the F_α 's calculated are upper limits of the helium lost by recoil. This can be refined later by including level density considerations. Second, since the relative differential cross sections are generally not known for the reactions of interest, they were assumed to be isotropic in the CM system. The uncertainties introduced by this estimate should be small, since the greater helium loss out the back surface from a forward-peaked distribution will be partially compensated by a smaller loss out the front. Later calculations can incorporate statistical model predictions of the relative cross sections. Third, the angular distribution of incident neutrons, associated with the close sample proximity to the RTNS-I source, was ignored. Further calculations have shown this to be unimportant for the present estimates. An average neutron energy of 14.8 MeV was assumed. Fourth, edge effects were

incorporated for those samples where the areas of the sides were comparable to the front surface area. They were estimated by first noting that the assumption of an isotropic CM helium recoil distribution yielded nearly isotropic laboratory helium recoil distributions for the intermediate-mass target nuclei considered. Equation 7 was thus used with w replaced by the other sample dimensions. This approximation is less accurate for forward-peaked recoil distributions, but is partially compensated by the distribution of incident neutron angles.

The results are shown in Table 2. This table also includes gold foil results calculated for the (n, α') reaction channel representing an average CM α energy of 19.3 MeV, estimated from the available experimental literature.^(8, 9) Although these results are not substantially different from the (n, α_0) case, this may not be true for other materials. The amount of helium lost from each sample by recoil is seen to be a significant fraction of the helium produced in each case, at least for the reaction channels considered. The recoil effects could, however, be completely removed from most of the chunk and wire samples by etching. The amount of surface material to be removed from each irradiated specimen was calculated from α -recoil range considerations, and most of the specimens were subsequently etched. Exceptions were the 5-mil gold and platinum foils, which were too thin to etch usefully, the diamonds, which were difficult to etch, and the zirconium samples, which were so small that all the material was needed to provide enough helium for the mass spectrometric analysis. Recoil corrections will have to be made to the helium generation measurements for these specimens.

B. HELIUM GENERATION CROSS SECTIONS

Preliminary results of the helium analyses and helium generation cross sections are summarized in Table 3. Over 100 helium analyses have actually been conducted, both on the RTNS-I-irradiated material, and on unirradiated material from the same lots. Only in the cases of carbon (diamond) and molybdenum did the unirradiated material contain a measureable amount of helium, and even then the amounts were much less than those generated by the irradiation. One irradiated specimen of each element was also analyzed for ^3He , and, as expected, none was detected ($<5 \times 10^{-11}$ atom fraction in all cases).

TABLE 2
ESTIMATED α -RECOIL LOSSES FROM REPRESENTATIVE
RTNS-IRRADIATED SAMPLES

Sample Material	Sample Shape*	Thickness (mils)	Reaction	Estimated α Escape Before Etching† (%)
Au	Foil	5	(n, α_0)	27
	Foil	5	(n, α')	24
	Foil	0.5	(n, α_0)	91
	Foil	0.5	(n, α')	90
Nb	Foil	5	(n, α_0)	33
	Foil	1	(n, α_0)	85
	Chunk ENb-1	30	(n, α_0)	10
V	Chunk EV-1	30	(n, α_0)	6
	Chunk EV-2	30	(n, α_0)	6
Al	Wire	18	(n, α_0)	13
Cu	Wire	24	(n, α_0)	8

* Chunk and wire samples assumed to have square areal cross sections for these calculations.

† The numbers for the (n, α_0) cases are strictly upper limits, since only the maximum possible α -recoil energy was considered. For most irradiated helium generation specimens, all recoil effects were removed by etching away the surface material with acids.

TABLE 3
PRELIMINARY HELIUM GENERATION CROSS SECTIONS FOR
14.8-MeV NEUTRONS

Material	Number of Samples Analyzed	Average Concentration of Helium Measured (appb)	Preliminary Cross Section (mb)
C*	2	155	900 [†]
Al	21	24	140
Ti	2	7.3	39
V	3	2.7	15
Fe	4	8.7	46
Ni	3	18	97
Cu	22	7.9	51
Zr*	2	2.4	10
Nb	2	3.3	17
Mo	3	3.6	16
Pt*	2	1.5	8
Au*	2	5.7	37
316SS	5	10.3	56

* Corrections for α -recoil effects have not been made for C, Zr, Pt, and Au, where they could not be removed by etching, but they are expected to increase the cross sections only slightly.

† The carbon cross section includes helium production by the $^{12}\text{C}(n, n\alpha)^8\text{Be}$ reaction, which yields 3 α particles.

The cross section results, presented in the last column of Table 3, provide the helium production rates for most of the elements of interest to magnetic fusion reactor designers. The estimated uncertainties of all the preliminary cross section results are ~ 10 to 20% . The data in the final column of Table 3 are total helium generation cross sections, integrated over all isotopes and reactions that produce the helium. Most of the helium is expected to come from simple (n, α) reactions, with a smaller contribution from $(n, n\alpha)$ reactions. In the case of carbon, the 900 mb cross section is due largely to the contribution from the $^{12}\text{C}(n, n\alpha)^8\text{Be}$ reaction, which effectively produces three α particles.

The fact that the helium generation cross section of aluminum (~ 140 mb) is significantly larger than the well-known $^{27}\text{Al}(n, \alpha)^{24}\text{Na}$ cross section (~ 118 mb) is attributed to the $^{27}\text{Al}(n, n\alpha)^{23}\text{Na}$ reaction, which appears to have a cross section of ~ 20 mb. This is in close agreement with a value of ~ 16 mb independently predicted in recent unpublished calculations by F. M. Mann and R. E. Schenter (HEDL) using a Hauser-Feshbach nuclear optical model. ⁽¹⁰⁾

An important correlation can be made between some of the pure element helium production cross sections and the 56-mb effective cross section measured for N-Lot Fast Flux Test Facility (FFTF) Type 316 stainless steel. When the C, Fe, Ni, and Mo cross sections from Table 3 are weighted in proportion to their compositions in the steel, they give a combined cross section of 54 mb. The chromium cross section, which has not yet been measured in this program, was assumed to be the same as that of iron in this calculation. The excellent agreement between the sum of the components and the measured aggregate amount provides further confidence in the results from Type 316 stainless steel, and indicates that the minor constituents of steel, other than carbon, contribute little to the total helium-production rate in a 14.8-MeV neutron spectrum.

V. PREPARATIONS FOR A d-Be IRRADIATION

Preparations are underway for a d-Be neutron irradiation at the Crocker Nuclear Laboratory (UC-Davis). Current plans call for a 96-hour irradiation at a deuteron beam energy of 30 MeV, to be run as a joint experiment by AI personnel, R. R. Heinrich and L. R. Greenwood of ANL, and M. W. Guinan of LLL. The ANL and LLL personnel will participate in the neutron dosimetry aspect of the experiment, from which a detailed characterization of the source will be obtained.

A. HELIUM-GENERATION SPECIMENS

The capsule, as presently designed, will contain approximately 100 helium-generation specimens of elements, alloys, and separated isotopes. These include 17 different elements (Li, B, C, Al, Si, Ti, V, Cr, Mn, Fe, Ni, Cu, Zr, Nb, Mo, Sn, and Au), and a number of fusion reactor candidate alloys (316 SS, TZM, Nb-1%Zr, Mo-0.5%Ti, and possibly some Sn-based alloys). The samples will be distributed in three flat layers, sandwiched between four stacks of dosimetry foils. The capsule will be placed approximately 1 cm behind the beryllium target, in order to obtain maximum neutron fluences at all locations in the capsule, while minimizing the effect of the finite neutron source size on the dosimetry analysis.

The capsule will also contain eleven rings of the elements Al, Fe, Ni, Cu, and Au, concentric with the deuteron beam axis, for helium accumulation fluence dosimetry. These rings will be segmented later for helium analysis, as was done in the RTNS-I experiment, and the results will complement the foil activation fluence dosimetry. There will be at least two rings of different diameters for each element, in order to examine the use of this technique for extracting neutron energy spectrum information. The wires will generally be 40 mils (1 mm) in diameter, which will give enough material to permit etching to remove the recoil effects produced by the high-energy component of the d-Be neutron spectrum.

B. FOIL ACTIVATION DOSIMETRY

The foil activation neutron dosimetry for this experiment will be substantially expanded over that used for the RTNS-I irradiation, in line with the

increased complexity of the neutron field generated by the d-Be source. Several different dosimetry foils will be used, based on their varying energy sensitivities to neutron-induced reactions. The foils will be segmented and counted following the irradiation, and the results will be combined to unfold the neutron fluence and energy spectra as a function of position in the capsule. The primary dosimetry foils will be four stacks of six 1- to 2-mil-thick elemental foils (Au, Co, Nb, Ni, Zr, Fe) sandwiching the capsule components, and producing eleven to fifteen neutron-induced dosimetry reactions. These will be supplemented by combinations of seven other foils (Sc, Al, Y, ^{238}U , ^{235}U , Tm_2O_3 , and perhaps In), placed before and following the capsule assembly. These will provide another twelve possible reactions from which to unfold the neutron energy spectra.

Current plans also call for additional helium accumulation fluence wires and foil stacks at positions well behind the capsule, to characterize the d-Be neutron environment at more distant locations. The activation analysis will also be extended to include several of the small helium-generation specimens, in order to determine additional neutron-induced cross sections and to identify other possible candidates for future activation analysis dosimetry.

VI. SUPPORT IRRADIATIONS FOR d-Be CHARACTERIZATION

The large energy and fluence variations characteristic of the d-Be neutron field require a significant amount of spectrum unfolding of the neutron dosimetry data, as described in Section V.B. This is further complicated by the general lack of neutron cross section information above 15 MeV. It thus is essential to compare the d-Be (and subsequently d-Li) dosimetry information with the information obtained from other, more readily characterized, neutron environments. To this end, two additional irradiations have been undertaken.

A. SECOND RTNS-I IRRADIATION

A second capsule has been irradiated for 76.7 hours at the RTNS-I facility at LLL. This capsule, to be described in detail in a later report, contained substantially more neutron dosimetry and helium analysis material than the previous RTNS-I assembly (described in Section III.A). The set of dosimetry foils used in the second irradiation (Au, Nb, Ni, Zr, Co, Fe) are expected to produce the bulk of the energy-sensitive neutron dosimetry information in the d-Be irradiation, and have been incorporated in a capsule geometry similar to that planned for d-Be. The capsule also included 10 wire rings of the elements Al, Cu, Au, Ni, Fe, and Ti, to expand the development and application of helium accumulation fluence dosimetry. Over one hundred other specimens were included for helium generation determination. The latter specimens included 8 additional pure elements, 21 separated isotopes, and 4 alloys of interest as fusion reactor first-wall candidates. These will be used to obtain further helium generation cross section information in a D-T environment. Several specimens are also being counted radiometrically at ANL for the subsequent determination of other neutron-induced cross sections. These results could lead to adoption of additional neutron activation reactions for dosimetry analysis.

The analyses of the activated foils and specimens are being undertaken jointly by personnel from AI, ANL, and LLL.

B. HFIR IRRADIATION

Previous d-Be neutron dosimetry measurements⁽¹¹⁾ have shown that up to half of the neutrons from 30-MeV deuteron beam bombardment of a beryllium

target have energies below 7 MeV. While the lowest energy neutrons are assumed to generate no measurable helium concentrations in most of the pure element specimens, this significant low-energy flux and the possibility of helium-generating impurities in the specimen materials warrant an experimental verification of this assumption. Two helium-generation capsules have thus been loaded in the High Flux Isotopes Reactor (HFIR) at the Oak Ridge National Laboratory for a 5-cycle irradiation in a subassembly provided by F. W. Wiffen. These capsules contain many of the same materials to be used in the d-Be irradiation. The 130 HFIR specimens include 17 pure elements, 4 separated isotopes, 7 stainless-steel alloys of varying nickel content, and one compound (SiO_2). Subsequent analysis of these specimens will provide low neutron energy helium generation information needed for the analysis of the d-Be neutron environment.

VII. TASK GROUP ON DAMAGE ANALYSIS AND FUNDAMENTAL STUDIES

Harry Farrar IV was invited by the Materials and Radiation Effects Branch (ERDA-DMFE) to serve as a member of the Task Group on Damage Analysis and Fundamental Studies. The first meeting of the Task Group was held in Germantown in October 1976, and it was determined that AI should also participate as a working member in Subtask Group A, Environment Characterization. The work scope of the subtask group includes the characterization of DMFE irradiation environments in terms of neutron flux, fluence, spectral distribution, physical parameters, and gamma-ray influence, in order to provide a standardized basis for intercomparison of damage data.

Exact responsibilities for further participation were defined, and personnel from AI participated in second and third meetings of the Task Group at Los Alamos in January 1977 and at Santa Barbara in March 1977. A final report for Subtask Group A was submitted to the Task Group and to ERDA.

VIII. CONCLUDING WORK ON TASK 27-1 AND TASK 27-3 PROGRAMS

At the end of FY 1976, the funding for Tasks 27-1 and 27-3 was terminated. The final wrap-up work was completed as a part of the continuing Task 27 program, and is reported here by the principal investigators: D. W. Keefer and J. T. Roberts (Task 27-1) and D. W. Keefer and A. G. Pard (Task 27-3).

A. DEVELOPMENT AND TESTING OF NONMETALLIC REFRACTORIES FOR FUSION REACTORS

The project objectives of Task 27-1 were to develop and test ceramic insulator coatings required in the development of the Theta Pinch-type fusion reactor, and to determine the effect of radiation on properties of nonmetallic refractories for use in fusion reactors.

During this report period, the mechanisms of glass coating - metal substrate adherence and the mechanisms affecting coating bubble content were examined. In general, these have been found to be strong functions of the chemistry of both the substrate surface and the coating. At elevated temperatures, where metal ions in the glass and the substrate are mobile, exchange occurs at the glass-metal interface. The bubble content, adherence of a glass coating, and degree of coating crystallinity are related to the metal interchange established at the interface just prior to solidification. In the case of Hastelloy X substrates, this interchange probably involves only iron and chromium, since oxidation experiments performed elsewhere show that only these two elements migrate from the metal to form oxides.

Lack of adherence of a coating is believed to be the result of inability of the coating to "wet" the substrate. This failure is brought on by depletion of oxides at the interface. This can occur during firing or during subsequent aging treatments. X-ray diffraction measurements of substrate oxide content and type after firing and aging substantiate this view.

The gas bubble content of a coating is dependent on a number of factors. Higher firing and aging temperatures assist in entrapped-gas escape by lowering the coating viscosity. In the case of firing in helium, as opposed to air, the more mobile helium can escape during cooling, and the coating is found to be

relatively bubble-free without aging. Evolution of oxygen created by oxide breakup contributes to bubble formation during both firing and aging.

Changes in crystalline content of glasses during post-firing aging have been observed as a function of firing atmosphere. These are believed to depend on the valency states of Cr ions present, which in turn depend on the oxygen content available in the glass at the time of solidification. For example, Cr in glass fired in air may exist in the highest valency state, Cr^{6+} . As such, the Cr may deter the formation of BAS_2 , thus causing the observed low BAS_2 content. The helium-fired glasses should have lower oxygen content and Cr of a lower valency state. This can account for the increases in the amount of BAS_2 detected in those glasses.

The results obtained in the course of this program can be summarized as follows:

- 1) Al glass insulator coatings were found to be adherent to stainless steel, Hastelloy X, and Nb-1%Zr
- 2) Understanding of the mechanisms of coating adherence, bubble content, and crystalline content was developed
- 3) The coatings were found to have high dielectric breakdown strength at temperatures up to 800°C ; the breakdown strength was not degraded in a reducing atmosphere
- 4) The softening temperature of the glass was found to be over 800°C
- 5) The coatings were found to have a low 14-MeV neutron sputtering rate: 2.3×10^{-4} atoms/incident neutron
- 6) A high rate of release of helium produced during an 800°C 14-MeV neutron irradiation was observed
- 7) The coatings were resistant to charged particle damage at 300°C
- 8) The coatings were found to be effective as tritium barriers at temperatures up to 800°C .

B. IRRADIATION DAMAGE IN STRUCTURAL MATERIALS FOR FUSION REACTORS

The project objective of Task 27-3 was to participate in the BCC Ion Damage Correlation Experiment.

During this report period the microstructures of two molybdenum samples, proton irradiated at 800°C, were examined. Information pertaining to the void populations is given in Table 4, along with results obtained previously at 900°C.

The results obtained in the course of AI's participation in the BCC Ion Correlation Experiment can be summarized as follows: The void size increases with irradiation temperature, while the void density decreases. At 800°C the void size is very small and there is no apparent increase in size with increasing damage. The void density does, however, increase in the course of irradiation. Void densities of the magnitude seen here have previously been observed in molybdenum and other BCC metals. At 900°C the void size increases with damage; the void density increases with damage up to 6 displacements per atom (dpa), after which it remains constant up to 16 dpa.

There is no indication of an incubation period for swelling at 900°C, and from Table 4 the swelling rate is about 0.35%/dpa. Absence of an incubation period at 800°C has been assumed, and the two data points from Table 4 give an approximate swelling rate of 0.14%/dpa.

For both irradiation temperatures, many of the void images appear as squares on the photomicrographs. At 900°C, where the voids are most easily visible, many of the void images are truncated. Although the orientation of the cube faces was not determined in this work, previous investigators have found voids in BCC metals to be bounded by {100} planes. Truncation can occur on both {110} and {111} planes.

In addition to voids, the microstructure contains a tangled array of dislocation lines and a low density of loops. In some instances, regions roughly 0.03- μ m wide adjacent to grain boundaries are found to be denuded of voids, except for the occasional appearance of large voids. The latter are several times the average size of voids observed within the main void population.

There is no evidence of a void superlattice in any of the specimens irradiated in this work. This is unusual since such superlattices have been observed

TABLE 4
VOID FORMATION IN PROTON-IRRADIATED MOLYBDENUM

Irradiation Temperature (°C)	Proton Fluence (10^{23} p/m ²)	Displacement Damage (dpa)	Damage Rate (10^{-4} dpa/s)	Average Void Diameter (10^{-8} cm)	Void Density (m ⁻³)	Swelling (%)	Swelling Rate (%/dpa)	TEM Foil Level (10^{-4} cm)
800	6.75	8	2.1	36	4.8×10^{23}	1.4	~0.14	1.2
800	10.1	12	2.2	35	5.9×10^{23}	1.5		1.1
900	2.73	3.1	2.0	140	5.9×10^{21}	1.1	0.35	1.0
900	5.46	6.0	1.9	140	1.2×10^{22}	2.9		0.9
900	10.9	16	2.5	170	1.1×10^{22}	5.1		1.6

in molybdenum following both neutron and heavy ion irradiations. It is worth mentioning that the substantially different nature of the damage created by protons may have an influence on the formation of void superlattices.

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