PREPRINT UCRL- 76765

Lawrence Livermore Laboratory

CONF-751026--7

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September 22, 1975

This paper was prepared for submittal to the International Conference on Radiation Effects and Tritium Technology for Fusion Reactors, October 1-3, 1975, Gatlinburg, Tennessee

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ABSTRACT

High purity copper foils were irradiated with 14.8-MeV neutrons from the rotating target neutron source facility at LLL. The average energy of the neutrons was 14.75 ± 0.1 MeV, and the average fluence was 7.0×10^{16} n/cm^2. After irradiation each foil was heated to the melting point and the released helium was measured by a mass spectrometer of special design. Isochronal heating was carried out on several samples to establish the type and temperature of maximum release. Calculated cross sections from the literature for the (n,α) and $(n,n'\alpha)$ nuclear reactions were used, and the predicted amount of helium was consistently about 0.5 of that actually measured. Because there is very little data on helium generation in metals irradiated with high energy neutrons, these results are important and will be related to potential CTR materials.

*This work was performed under the auspices of the U.S. Energy Research & Development Administration, under contract No. W-7405-Eng-48.

INTRODUCTION

Because helium atoms are not very soluble in metals and alloys, they tend to form bubbles as the temperature increases. These bubbles play an important role in such destructive processes as void-swelling and embrittlement of materials subjected to neutron irradiation. Numerous studies[1] have dealt with the diffusion of helium as a function of temperature. However, the effect of void-swelling, embrittlement, and related phenomena cannot be completely understood until the amount of helium produced for a known neutron fluence can be predicted.

Recent information[2] about helium production at the low neutron energies (1 to 3 MeV) characteristic of fission reactors emphasizes the need for the same data at higher energies. Unfortunately, very little data is available on helium generation in material exposed to 14.8-MeV neutron bombardment. There are indications that helium generation in certain metals during this high energy bombardment can be 1000 times greater than that observed in fission irradiation.

The atomic fraction of helium produced during neutron damage is related to neutron energy through the total cross section of the various nuclear reactions according to the relation

$$He_{(at.\ fraction)} = \sigma_T \Phi ,$$

where Φ is the neutron fluence (n/cm^2) and σ_T is the sum of all cross sections of neutron reactions that generate helium, such as (n,α) and $(n,n'\alpha)$, times their respective isotopic fraction. For a given fluence, the helium content is proportional to the total cross section σ_T. By measuring the helium content and the neutron fluence, the total cross section can be calculated and compared with values measured by activation techniques or computed from theoretical data. In some metals the helium content can be precisely determined by counting those radioactive species produced along with the helium. This method is not applicable when nuclear reactions produce only stable isotopes.

In such cases, direct mass spectrometric measurements of the amount of helium resulting from 14-MeV neutron bombardment would be useful. Copper, because of its well-characterized physical properties, was chosen for this preliminary study in which we developed equipment and procedures. Subsequent to this study, helium generation in refractory metals such as niobium, vanadium and molybdenum are planned.

EXPERIMENTAL METHODS

Disks, 12.7 mm in diameter and 0.05 mm thick, were cut from Marz grade copper foils. These samples were carefully cleaned with methyl alcohol before loading into the target holder of the rotating target neutron source (RTNS) at LLL. The irradiation procedure and beam characteristics are reported elsewhere.[3] After irradiation, the samples were gamma-ray counted to determine the amount of ^{60}Co produced from the $^{63}Cu(n,\alpha)^{60}Co$ reaction. The amount of helium was measured by heating each disk to the melting point in a specially designed mass spectrometer.[4] This low-resolution, high-sensitivity mass spectrometer can reach pressures lower than 0.13 μPa (10^{-9} Torr) and detect as few as 10^{10} atoms of helium. To maximize the amount of information gained from this study, either an isochronal or an isothermal mode of heating was used prior to the final melting of the sample. The samples were held at the melting point for a period of time (~15 min) sufficient to release all the helium. Several unirradiated disks were run to establish that the background level of helium was negligible.

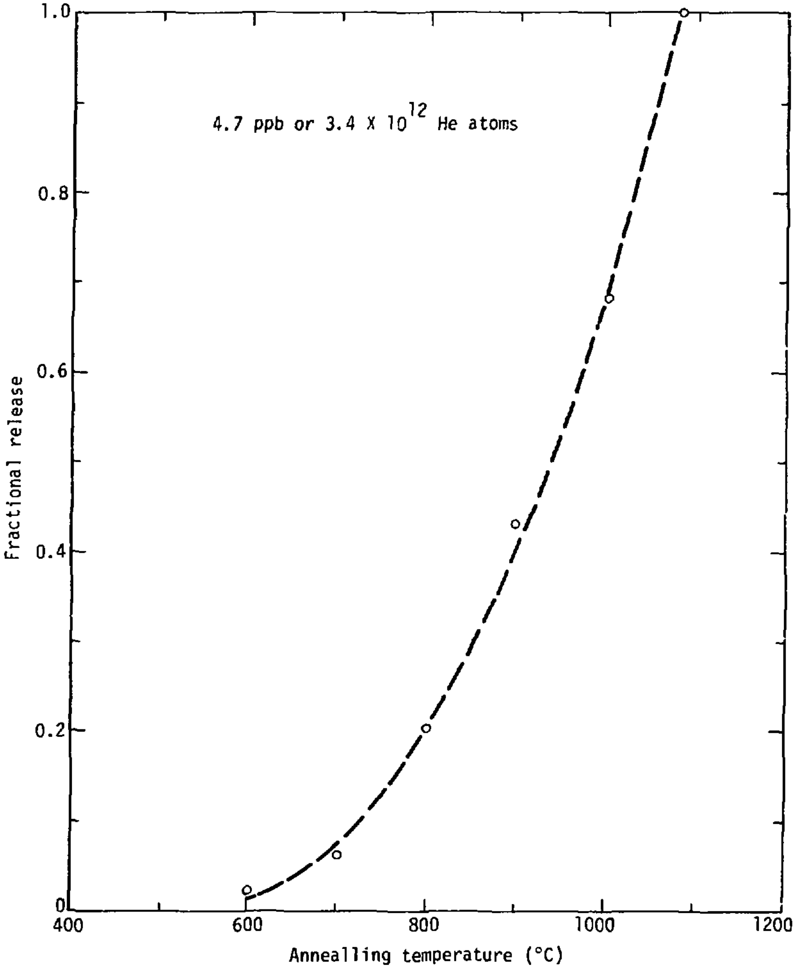
RESULTS

The amount of helium varied from $1.8 \cdot 10^{12}$ to 5.5×10^{12} atoms, more than enough to accurately measure the cross section. Five copper disks were irradiated simultaneously and had an average fluence of 7.0×10^{16} n/cm^2. Another copper sample was irradiated to a fluence of 1.08×10^{17} n/cm^2.

Figures 1 and 2 show typical isochronal and isothermal release of helium from copper. The helium is tenaciously held within the solid until nearly 0.5 of the melting temperature is reached (Fig. 1). The isothermal release is similar to that of other metals. There is an initial accelerated outgassing followed by a rapidly decreasing rate of release. The isothermal curves do not fit those expected from the outgassing of a disk with an initial homogeneous distribution of helium. So, even at these low concentrations (2 to 3 at. ppb), the release is apparently influenced by the formation of slow moving bubbles.

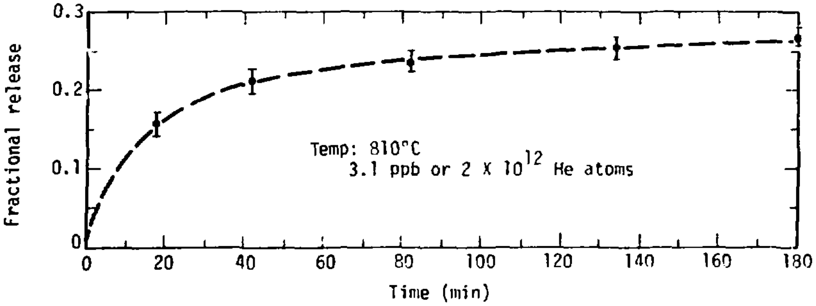
It would be instructive to know if the helium content is a linear function of fluence. As there are only two data points for fluence, we used the ^{60}Co count from each of the five disks. The ^{60}Co count should be directly proportional to the neutron fluence, and therefore proportional to the amount of helium generated. In Fig. 3, the amount of helium released is plotted vs the ^{60}Co count. In each case, the ^{60}Co was measured with a precision of approximately 0.2%. The absolute accuracy of the measurement is estimated to be ·5 . Even with the scatter in Fig. 3, the linear relation seems valid.

Table 1 shows the helium content of each disk with the calculated total cross section. The five disks have an average cross section of 54 mb. This value is very close to 55 mb for the specimen irradiated to a slightly different fluence. The overall uncertainty of these results is estimated to be ±10%. The important comparison is that of the average experimental value (54 mb) to the value obtained by activation techniques. The main source of helium will be the (n,α) reactions, because the (n,n'α) cross section is too low to be significant. The (n,α) cross sections determined by activation are 34 mb for ^{63}Cu and approximately 21 mb for ^{65}Cu.[5] Taking the isotope fraction into account, the total activation cross section should be about 30 mb. Obviously, the amount of helium experimentally measured is a factor of 1.8 higher than that computed from activation cross sections. We believe the experimentally derived value to be valid because our recent measurements of helium



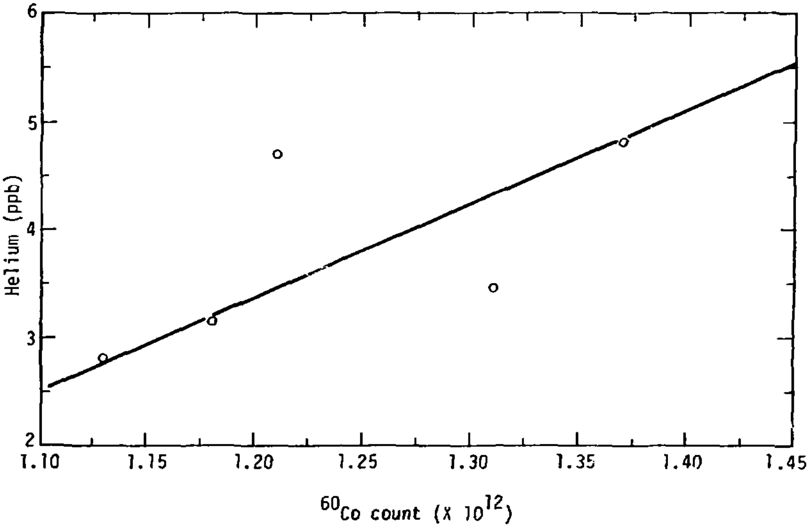
4.7 ppb or 3.4 X 10^12 He atoms
Fractional release
1.0
0.8
0.6
0.4
0.2
0
400
600
800
1000
1200
Annealling temperature (°C)

Fig. 1



0.3
0.2
0.1
0
Fractional release
Temp: 810°C
3.1 ppb or 2 X 10^12 He atoms
0
20
40
60
80
100
120
140
160
180
Time (min)

Fig. 2



6
5
4
3
2
Helium (ppb)
1.10
1.15
1.20
1.25
1.30
1.35
1.40
1.45
60Co count (X 10^12)

Fig. 3

Table 1. Helium content and calculated total cross section of copper disks.

Sample no.	Type of anneal	Amount of helium (ppb)	Fluence	σ_T (mb)
1	isochronal	4.7	7.0×10^{16}	67
2	isochronal	4.8	7.0×10^{16}	68
3	isothermal	3.1	7.0×10^{16}	45
4	isothermal	2.8	7.0×10^{16}	40
5	isothermal	3.4	7.0×10^{16}	49
6	isothermal	5.9	1.08×10^{17}	55
			Average	54
			Uncertainty	±10%

generation in aluminum agree very well with that predicted by the activation cross sections. Aluminum occurs as a single isotope, and its (n,α) cross section is the best known in this energy range.

A possible explanation of the discrepancy in copper might be the presence of impurities within or on the surface of the metal. Most elements in the parts per million range will not increase the helium content significantly. Nevertheless, there are certain elements such as carbon and boron with (n,α) cross sections large enough (~1 b), that when present in parts per million, may make a significant contribution to the inventory of helium. For this reason several copper disks were examined by spectrographic and combustion analysis-mass spectrometry techniques to determine whether boron, carbon or nitrogen might be present in amounts large enough to account for the excess helium.

The analyses are given in Table 2. The carbon contamination might be suspected as a source of helium, because the $^{12}C(n,n')3\alpha$ reaction has

Table 2. The impurity analysis of copper disks (ppm).

Detected	Not detected (limits of detection)
Ag 6	U >1050
Si >6	As, Th >600
Ca 3	Ca, Na >400
Al 1	Ba, Cd, Hg, In, P, Sb, An >100
Mg >1	Bi, Ge, Sb, Sn >40
Be >1	Ga, Co, Cr, Mn, Mo, Nb, V, Sr 15
C 86[a]	B, Ti, Fe >4
	Ni >3

[a]This analysis was by combustion analysis - mass spectrometry.

a cross section of 0.28 b: effectively 0.8 b in terms of helium production. However, the bulk carbon analysis, 86 ppm weight (450 at. ppm), could only contribute 0.025 ppb helium from a fluence of $7.0 \cdot 10^{16}$ cm^{-2}, which is a few percent of the amount actually measured. Other low-Z elements have cross sections in the 1-b range, but their concentration also appears to be low. We did not determine if surface contamination is a source of helium. Surface carbon for example, might be a source of helium injected into the metal to a depth of up to 10 μm. To contribute to the helium actually measured however, the carbon layer would have to be over 1 μm thick, much more than expected after our preirradiation clean-up procedure. This seems to negate the probability of impurities contributing to the excess helium generated in the copper.

The discrepancy remains unresolved. The consequence of this preliminary study of helium generation in metals by 14.8-MeV neutron bombardment is to point out the possibility of error in predicting atomic fraction of helium using activation cross sections and to emphasize the importance of high purity, especially low-Z elements, when considering materials in the design of fusion reactors.

ACKNOWLEDGMENT

We appreciate the suggestions and critical review of the manuscript by Ralph H. Condit and Clarence L. Hoenig.

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FIGURE CAPTIONS

Fig. 1. An isochronal plot of helium release from a copper disk. The time at each temperature was 15 min.

Fig. 2. An isothermal plot of the helium release from a copper disk at 810°C.

Fig. 3. The measured helium released from each disk is plotted as a function of the ^{60}Co count that is directly proportional to the neutron fluence.

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