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DEFECT PRODUCTION EFFICIENCY IN THERMAL
NEUTRON IRRADIATED COPPER AND MOLYBDENUM

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DEFECT PRODUCTION EFFICIENCIES IN THERMAL NEUTRON IRRADIATED COPPER AND MOLYBDENUM

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ABSTRACT

We have derived the primary recoil spectra for thermal neutron capture in copper and molybdenum, and have calculated the damage energy cross-sections needed to determine the defect production efficiencies from measured resistivity damage rates. These efficiencies, which are in excellent agreement with fully dynamic computer simulations of collision cascades, are compared to the predictions of displacement functions which are currently in use.

1. INTRODUCTION

The ability to predict the number of defects produced by an energetic recoil atom with an energy T is of fundamental importance to kinetic models of radiation-induced property changes in metals and alloys. The number of defects, $\nu(T)$, depends upon the damage energy deposited by the primary knock-on atom (PKO) and the threshold energy for producing atomic displacements. At this time, asymptotic forms of the modified Kinchin-Pease expression are used to obtain the number of Frenkel-pairs produced by a PKO with an energy T ; namely,

$$\nu(T) = \frac{K\hat{E}}{2T_A} \quad (1)$$

where $\hat{E}$ is the damage energy, T_A is the effective displacement energy and K is an efficiency factor.

Electron¹, ion² and neutron³ irradiations of a number of metals demonstrate that the efficiency factor, K , in Equation (1) is an energy dependent parameter which varies between close to 1 at recoil energies slightly above threshold to .3 to .5 at recoil energies greater than 100 times threshold. Knowing the value of K as a function of recoil energy is necessary for the development of displacement

functions for use in radiation damage modeling.

The parameters for displacement functions which have recently been published in the literature have been fit to the results of charged particle irradiations in the intermediate recoil energy region where the efficiency is changing significantly with energy.^{4,5} Because the primary recoil energy spectra which are characteristic of these ion irradiations are broadly distributed about the mean recoil energy, errors can be introduced into the calculated displacement function. These errors could significantly alter the estimates of the radiation damage expected in many components of fusion reactors.

It has been known for many years that thermal neutron capture in metals provides a source of intermediate energy recoils.⁶ The inability to accurately calculate the thermal neutron capture recoil spectrum, however, has prevented researchers from fully utilizing the results of thermal neutron damage experiments. In this work, the primary recoil spectra for thermal neutron capture in copper and molybdenum are calculated, and the damage energy cross-sections needed to determine the defect production efficiency from the measured resis-

tivity damage rates are presented. These results are compared to the predictions of the displacement functions which are currently in use.

2. CALCULATION OF THERMAL NEUTRON RECOIL ENERGIES

We have developed a model which allows us to overcome the lack of experimental gamma-ray data which has prevented us from calculating accurate recoil energies in the past. In this model, we use the experimental gamma-ray transition probabilities when they are available, and use a statistical nuclear calculation to obtain the gamma-ray transition probabilities into and out of the unresolved continuum. The physics of this statistical method are described elsewhere.⁶ The gamma-ray spectra and multiplicity (gammas per capture) which we calculate are in excellent agreement with experimental observations.

The results of the thermal neutron recoil calculations for molybdenum and copper are given in Table I. In this table, $T_{1/2}$ is the median recoil energy which is defined as the recoil energy above which half of the damage energy is produced. Figures 1 and 2 show the integral recoil spectra from electrons, thermal neutrons and reactor neutrons in both metals. It is apparent from these figures that thermal neutrons produce a very narrowly distributed recoil distribution in the intermediate energy region.

Table I:

Element	$\langle E \rangle$ (eV)	$T_{1/2}$ (eV)	Damage Energy Cross-Section (barns x eV)
Copper	392	480	1276
Molybdenum	178	240	410

3. DISCUSSION

Jung and co-workers have recently derived displacement functions for aluminum, copper,

Fig. 1. Integral damage energies in Mo.

Fig. 2. Integral damage energies in Cu.

platinum, vanadium, niobium and molybdenum from the resistivity damage rate results of electron, light ion and neutron irradiations.⁴

These functions describe the low energy, intermediate energy and high energy recoil regions with different functional forms. At low energy, the damage function was fit to the results of electron irradiations. At intermediate energy, ion-irradiation data was used, and for high recoil energies the results of reactor and fusion neutron irradiations were used.

Another approach was taken by Simons,⁵ who used a method known as damage functional analysis to derive analytic functions for the PKO energy dependence of Frenkel-pair production in copper, silver, molybdenum and tungsten. The damage analysis method involves solving integral equations of the form

$$N_i = \int_{T_0}^{T_m} P_i(T) \nu(T) dT \quad (3)$$

where N_i is the number of Frenkel-pairs in the i th particle recoil spectrum per unit fluence, $P_i(T)$ is the probability that an incident particle of type i produces a PKO with an energy T (the primary recoil spectrum) and $\nu(T)$ is the displacement function to be determined. In copper, N_i was calculated at low recoil energies from the results of computer simulations. At intermediate recoil energies in copper, N_i was determined from ion irradiations, and at high recoil energies N_i was determined from fission and fusion neutron experiments.

Reactor neutron recoils span several decades in energy. In this regime where the efficiency is relatively constant, there is little error involved in calculating the efficiency from the initial damage rate measurements. However, when the recoil spectrum principally lies in the region where the efficiency is rapidly changing, as in the case of ion-irradiations, significant errors can result. Although Simons reduces the effect of these errors by best fitting an integral equation over a large range of energies, the possibility of error is still great. In Jung's work, on the other hand, the intermediate recoil energy regions are almost entirely fit with light ion data, and the errors in the resulting damage functions could be quite large. The question, then, is: How well do the damage functions derived by Jung and Simons reproduce the actual decrease in efficiency observed in the intermediate recoil energy regime? A major problem with both methods is the reliance on ion data to study the intermediate recoil energies.

The efficiency factor, K , can be determined from the resistivity damage rate as

$$K = (d\rho/d\phi) 2T_d/\sigma_{de} \rho_f \quad (4)$$

where $(d\rho/d\phi)$ is the initial resistivity damage rate, σ_{de} is the damage energy cross-section and ρ_f is the Frenkel-pair resistivity³. Using the resistivity damage rate data^{3,8,9,10,11} and Equation (4) we can calculate the efficiency factor in copper for a large number of electron and neutron irradiations. These efficiency factors have been plotted in Figure 3 as a function of the median recoil energy normalized to the threshold energy. Also plotted in this figure are the predicted efficiencies from the displacement functions of Jung and Simons', along with the experimental results of ion irradiations.

Figure 3 clearly demonstrates that K decreases between two times threshold and one hundred times threshold. The efficiency of thermal neutron recoils is less than that for ions with similar median recoil energies. This is not surprising, since, as was mentioned earlier, the recoil spectra of the ions is distributed over a large energy range. In fact, Simons' damage function, which averages the data over the entire recoil spectra, brings these efficiencies down considerably and is in reasonable agreement with the thermal neutron data.

The deviation of Simons' damage function at low recoil energies is due to the fact that the parameters for the function were determined at these energies from the computer simulation data of Schiffgens and Borquin.⁷ If these parameters had been fit to electron data and the correct thermal neutron spectrum the errors in this region would be substantially reduced.

Jung's calculated efficiencies for copper show large deviations from the data in the intermediate recoil energy region. Jung's predictions also indicate a strange, double hump between two and one hundred times the threshold energy. Though there is not an a

Fig. 3. Damage efficiencies in Cu.

priori reason why one should expect a smooth, monotonic behavior for the efficiency function, one is hard put to explain such drastic behavior. It might be argued that the displacement threshold in copper is anisotropic to such a large extent that the defect production saturates between two and ten times threshold and then suddenly increases again at higher energies. The data, however, fail to support such an argument. The deviation is most likely due to either experimental errors or to the method used in interpreting the ion data.

We can compare the predictions of both of these damage functions to the experimental number of Frenkel-pairs, N_f , per neutron capture. Using the damage rate data⁹ with the relation

$$(d\rho/d\phi)/\rho_f \sigma_{n\gamma} \equiv N_f \equiv \int_0^{T_m} P(T) v(T) dT \quad (5)$$

where $\sigma_{n\gamma}$ is the capture cross-section, $P(T)$ is the primary recoil spectrum and $v(T)$ is the damage function we obtain an experimental value on copper of 2.51 Frenkel-pairs per capture compared to Jung's predicted 5.33 Frenkel-pairs per capture and Simons'

predicted 3.03 pairs per capture.

Using the damage rate data^{3,12,13,14} and eq. (4) we have calculated K in molybdenum. These values are plotted in Figure 4 as a function of the normalized median recoil energy. Also plotted are the predictions of Jung's and Simons' displacement functions, as well as the results of fully dynamic computer simulations. As was the case in copper, the efficiency for defect production in molybdenum appears to decrease between two and one hundred times the threshold energy.

The initial rise in efficiency near the threshold energy is well reproduced by both of the displacement functions: a manifestation of the fact that both functions were fit to the electron data. Simons' function, however, overestimates the efficiency between two and ten times the threshold energy. This is most likely due to the fact that Simons only used the thermal neutron point in this region. At the time that Simons fit to the thermal neutron data, the recoil spectrum and median recoil energy were not known.

Jung's function, which was fit to ion data, comes much closer to fitting the thermal neutron point in molybdenum. Once again,

Fig. 4. Damage efficiencies in Mo.

however, Jung predicts much higher efficiencies at intermediate recoil energies.

The computer calculations, which fit the electron and thermal neutron results reasonably well, have the same slope in the intermediate recoil energy range as Simons' prediction. Using the damage rate data¹³ for molybdenum with a value for ρ_f of $13 \times 10^{-6} \Omega \cdot m$, we obtain the experimental number of Frenkel-pairs per thermal neutron capture of 1.17. This compares to Simons' prediction of 1.46 and Jung's prediction of 1.21 Frenkel-pairs per capture.

4. CONCLUSION

The primary recoil spectra for thermal neutron capture in copper and molybdenum have been calculated. The results of thermal neutron irradiations of these metals have been interpreted with these new recoil energies. The thermal neutron efficiencies derived from the resistivity damage rate measurements have been compared to the predictions of the most recent damage functions. The data clearly show that the efficiency for defect production in these metals decreases between a few to one hundred times the threshold energy. Though two previously proposed damage functions exhibit decreases in efficiency, the damage

function proposed by Jung and co-workers does not fit the data as well as the damage function described by Simons.

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