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Positron-lifetime study of copper irradiated by *d*-T fusion neutrons

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Positron-lifetime characteristics have been investigated for samples of pure annealed copper irradiated by *d*-T fusion neutrons to a fluence of 5×10^{16} n/cm^2. It is found that the general characteristics of the data are similar to those obtained from proton irradiation under similar circumstances. While there are minor differences in the dose dependence, trap lifetime, and recovery between the two irradiations, there is quantitative agreement between the trapping-rate data and calculated damage production when damage from recoils below 50 keV is excluded.

I. INTRODUCTION

The production of damage in materials irradiated by energetic particles is a subject of practical interest to the development of both the fusion and fast-fission reactor. The present experiment was performed to expand the scope of previous work in which the energy dependence and dose dependence of radiation damage from protons was studied.[1] In the proton-irradiation experiment copper samples irradiated near room temperature by protons at energies between 24 and 4.5 MeV were analyzed with positron-lifetime measurements and it was concluded that the low-energy recoils, below 55 keV, did not produce defects which survived as positron traps with a short lifetime. Since the primary knock-on atom's recoil spectrum is very similar for high-energy protons and neutrons of comparable energies, the present experiment was done to add information about the survival of the defects produced by low-energy recoils in this energy range and also to demonstrate the similarity, or lack of similarity, between the effects obtained with proton and neutron irradiations.

II. EXPERIMENTAL METHOD

Samples of MARZ-grade copper from MRC, Inc., were prepared in disks 19 mm in diameter by 0.252 ± 0.003 mm which were annealed in an atmosphere of argon and hydrogen at 650 °C for two hours and electropolished to their final thickness. Positron-lifetime measurements of these samples show only one lifetime which was used as the value for the free-annihilation lifetime in the trapping-model analysis.

Irradiations were done with *d*-T fusion neutrons (14 MeV) from the rotating-target neutron source at Lawrence Livermore Laboratory (RTNS-I).[2] This accelerator provides a spatially small source of fusion neutrons radiating isotropically from the source. The samples were irradiated simultaneously for the same time period with samples placed closer to the source receiving higher doses.

Samples were irradiated in pairs with spaces between them to obtain the dose-rate variation. In this geometry the dose rate varied from 3×10^{11} $n\ \mathrm{cm}^{-2}\ \mathrm{s}^{-1}$ to 6×10^{8} $n\ \mathrm{cm}^{-2}\ \mathrm{s}^{-1}$. The temperature of the samples was held at 5 °C during the irradiations and the samples were stored at 25 °C until measurement. This schedule was purposely chosen so that the temperature history of these samples would be similar to those reported in Ref. 1. The neutron fluence was measured for each of the samples by measuring the activity in niobium foils placed adjacent to each sample position. For those samples that were close to the source, the average of foils immediately in front of and behind the sample was used to compensate for the variation of neutron flux over the sample thickness. In the closest samples, the fore-aft variation in dose was less than 15%. Three separate irradiations were performed, two resulting in fluences of about 4×10^{16} $n\,\mathrm{cm}^{-2}$ for the samples irradiated at the highest rate and one shorter irradiation in which the high-rate sample received 1.7×10^{16} $n\,\mathrm{cm}^{-2}$. The lower-fluence–higher-rate sample set was obtained in order to separate dose-rate effects from overall dose-level effects in the final analysis.

Positron-lifetime distributions were measured in this experiment with a coincidence spectrometer consisting of two truncated cones of NE111 mounted on RCA C31024 phototubes. Timing discrimination was done with Ortec 473A constant fraction discriminators, and conventional fast-slow discrimination was used to identify valid start and stop events. The resolution of this system is 160 ps full width at half maximum obtained with a ^{60}Co source and the same energy windows as used in the positron measurement. A similar system was first reported in Ref. 3. Because energetic neutron irradiation of copper produces small quantities of ^{60}Co in the samples, a stronger positron source of 20 μCi of ^{22}Na was used in these measurements than in the previous studies of proton-irradiated copper. With this source, the overall background levels were higher but the contribution of the sample activation to the total coincidence was limited to less than 2% of the total. Subtracting the contribution of the ^{60}Co did not change the results of the analysis of test spectra and so no correction for this effect was made during the analysis of the data for lifetimes and trapping rates. The contribution of annihilations in the source was 4% of the total coincidence counts above background and was included in the fitting of the model to the spectrum.

The positron-lifetime spectra were analyzed with two models, the single-lifetime model and the trapping model in a least-squares-fitting code described in Ref. 1. This analysis is different from the two-lifetime models used by some researchers in that including the trapping model specifically in the fit allows the bulk lifetime to be fixed during the analysis while the trapping rate and trap lifetime vary. This method of constraint is not possible during the fitting process when the analysis is done in terms of two uncorrelated lifetimes. Fitting the trapping model directly has the disadvantage that the quality of the fit is determined most sensitively by inspecting the residual spectrum. This is less convenient to report for each sample as model deficiencies show up most strongly as structure in the residual spectrum rather than changes in the overall value of chi squared. In the analysis of the data reported here there was pronounced structure in the residual spectrum from a single-lifetime fit of spectra of the samples containing traps. In the trapping-model fits the residual structure could be completely eliminated. Model differences therefore were observed. The advantage of applying physically reasonable constraints in the trapping model leads us to prefer the trapping-model analysis.

The analysis for trapping rates was done with the bulk lifetime fixed at the value measured for a well-annealed sample and varying the trapping rate and trap lifetime. In the case where the trap lifetime varied about some constant, the analysis was redone with the trap lifetime fixed as before. This procedure does not significantly change the value of the individual trapping rates but does reduce the scatter in the trapping-rate values. This reduction can be understood by recognizing that there is coupling in the shape of the lifetime spectra between the values of the trapping rate and the trap lifetime resulting from both of these quantities appearing in the term for the strength of the long-lifetime component of the trapping. Thus more accurate trapping-rate data can be obtained with a fixed trap lifetime, but care must be taken that this procedure does not obscure changes in the trap lifetime which are indicative of changes in traps.

III. RESULTS

Positron-lifetime spectra were obtained for all of the irradiated samples from three separate irradiations. The lifetime spectra were first analyzed using a model which contained a single lifetime. This model also contained as parameters, the system parameters (the centroid, resolution width, and slope of the wings of the resolution function) and the strength of the source annihilation component. The lifetime of the source component was fixed at 405 ps from other measurements. The results of the single-lifetime fits to the data are presented in Fig. 1. The single-lifetime data have one outstanding feature, that the lifetime values are increasing as a function of increasing neutron fluence for all of the samples indicating that either saturation of the measurement by high-trap density has not been obtained or that the trap lifetime is changing with dose. Trapping-model analysis will show that the trap lifetime is reasonably constant, however; as a consequence of not obtaining saturation, the trap lifetime cannot be determined from the saturation lifetime independently from the trapping model.

To define the parameters of the trap, the trapping rate, and trap lifetime, the data were analyzed with a single-trap model. In the analysis, the lifetime of the free-positron annihilation was set at 120 ps, the value obtained from repeated measurements of well-annealed copper samples. The source and system parameters were treated in the same manner as for the single-lifetime

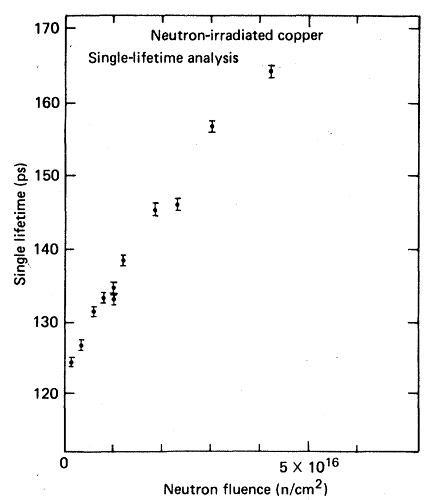

FIG. 1. Positron lifetime versus dose deduced from a single-lifetime analysis of copper irradiated by d-T fusion neutrons. The lifetime is still increasing at the highest dose obtained with no sign of saturation. The errors are derived from the fitting procedure.

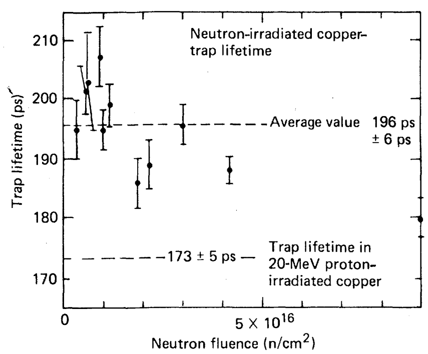

FIG. 2. Trapped-positron lifetimes for d-T neutron-irradiated copper. The lifetime values appear to exhibit some slight dose dependence; however, the data are statistically consistent with a random distribution about a constant value. All of the values for the trap lifetime determined in this experiment are significantly higher than the value determined in Ref. 1 for copper irradiated by protons. The errors are derived from the fit.

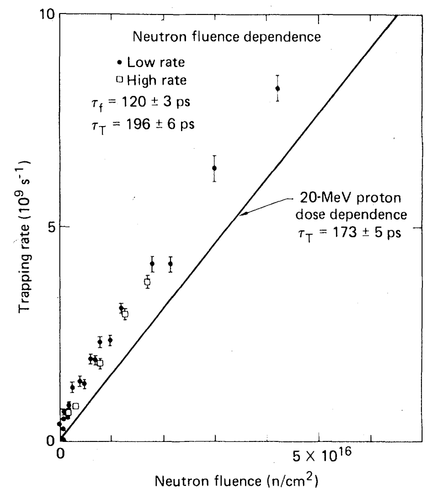

FIG. 3. Trapping rate determined by fitting the trapping model to data obtained from copper irradiated with d-T fusion neutrons. The data represented by dark circles were all obtained under similar irradiation conditions where low-dose samples were irradiated at low rates and high-dose samples were irradiated at high rates. The data represented by the open squares were obtained by irradiating at a higher dose rate for a shorter time in order to determine that the low-dose behavior of the trapping rate was not due to irradiation-rate effects. The errors are derived from the fit. The solid line is from proton-irradiation-damage results from Ref. 1.

model described above. Analysis of the samples was carried out with both the trapping rate and trap lifetime as adjustable parameters of the fit. The quality of the fit to data from damaged samples with the trapping model was always better than that obtained on the same spectrum with the single-lifetime model. The magnitude of the improvement depended on the size of the contribution of trapping to the overall lifetime, with the most improvement obtained for cases where the trapping rate and the annihilation rate for free positrons were similar. The improvement in fit resulted in improvements in chi-squared of as much as 40% and resulted in residual data-model differences with no discernable structure over the whole spectrum range. The smooth residual spectra are evidence that a second trap with a separate distinct lifetime was not observed.

The results of the determination of the trap lifetime are shown in Fig. 2. The trap-lifetime values may have some slight dose dependence with about the same magnitude as the accuracy of the lifetimes themselves. However, the data are statistically consistent with no dose dependence and an average value of the lifetime of 196 ± 6 ps.

Since the trap lifetimes determined from separate fits of the individual spectra may reasonably be considered to define a constant average trap lifetime, the values for trapping rates were obtained by fitting the spectra with the single-trap model with the trap lifetime fixed at the average of the previously determined values. As the trapping rate was determined in each of the fits with fixed or variable trap lifetime, the effect of this procedure can be directly assessed. In no case did fixing the trap lifetime to the average value result in a change of more than 8% in the trapping-rate result. The values for trapping rates determined with a fixed trap lifetime are presented in Fig. 3. The data are from analysis of all three sample sets. The three sample sets include one high-rate–low-dose irradiation so that the nonlinear dose dependence of the trapping rate at low dose is identified as an effect of the low total dose rather than the lower dose rate. The overall consistency of the data from the three irradiations is quite good.

Recovery of the radiation-induced damage from room temperature to 600 °C was measured and analyzed with a single-lifetime model. The data are presented in Fig. 4. Shown with the data obtained from neutron-irradiated copper are data taken from Ref. 1 for copper irradiated with energetic protons.

A convenient quantity which may be used to compare the relationship of defect-production calculations and experiments using different irradiation sources is the damage-energy cross section. Damage energy is the portion of energy from a specific recoiling atom which is given up to scattering other atoms and is calculated by the theory

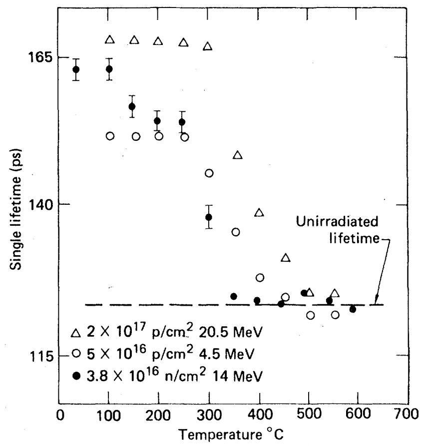

FIG. 4. Single-lifetime fits to the data from copper samples isochronally annealed. The data from the present experiment and those from Ref. 1 are displayed together. The errors indicated for some of the data may be taken as typical for all the data and are derived from the fit.

of Lindhard.[4] To calculate the damage energy one requires a knowledge of the recoil spectrum produced by radiation source.

The calculation of the neutron-recoil spectrum is made using experimental data which have been tabulated for each of the possible neutron-copper reactions. While there exist minor differences among the values of these cross sections stored in the various available libraries, the overall consistency in the final values for the damage energy is good.[5] For proton calculations, a similar library of experimental cross sections does not exist. Consequently, the recoil-energy–spectrum calculations for protons are derived from nuclear models and some simplifying assumptions.[6,7] Thus the proton-recoil calulations are more removed from the primary data and may not be as accurate as those for neutrons.

Damage-energy values for *d*-T fusion neutron and for protons are in Table I. The damage-energy values are calculated for several threshold energies, one (70 eV) obtained from twice the value of the average energy for producing a displacement, and three near the threshold value determined in Ref. 1 (55 keV). The damage-energy values calculated by retaining all the recoils above the lower displacement threshold are 1.7 times larger in 20-MeV proton-irradiated samples than in the samples irradiated by the same fluence of 14-MeV neutrons. If the threshold model is correct, and the damage energy is calculated using only recoils above 50 keV then the trapping rate for the neutron-irradiated samples would be slightly lower than for the proton-irradiated samples. This result does not change substantially when comparing 14-MeV neutrons and 20-MeV protons even when the threshold is changed to 20 or 100 keV. There is greater sensitivity when comparing 5-MeV protons and 14-MeV neutrons over that same range of thresholds. Consequently, the direct comparison of trapping rates and damage energy for these cases can be another test of the existence of a threshold in the recoil spectrum and can even aid in determining the value of that threshold.

TABLE I. Calculated damage-energy cross-section values and experimentally determined trapping rates per unit fluence for *d*-T fusion neutrons and protons of various energies. The ratio of the values calculated for a given threshold may be compared with the ratio of trapping rates per unit dose, to identify an effective threshold for trap production. Such a comparison among proton-irradiated samples in Ref. 1 leads to a threshold of 55 keV for proton-irradiated samples.

Threshold for integration	Damage-energy cross section (10^{-25} MeV cm^2) 100 keV	50 keV	20 keV	70 eV	Trapping rate per unit fluence 10^{-7} s^{-1} particle^{-1} cm^2
Proton energy (MeV)					
25	0.280	0.305	0.319	0.470	
20	0.248	0.260	0.277	0.478	1.5 ± 0.10
15	0.193	0.216	0.241	0.527	
10	0.131	0.173	0.217	0.688	
5	0.103	0.180	0.289	1.121	1.0 ± 0.2
Neutron energy					
14.5 MeV	0.250	0.263	0.270	0.277	1.7 ± 0.35

IV. DISCUSSION

The evidence that positrons trap at defects in copper which are formed by deformation, irradiation damage, and thermal effects is extensive.[1,8-15] Of particular interest is a study of proton-irradiated copper in which energetic protons were used to damage copper for subsequent positron-lifetime analysis.[1] This irradiation was done at $-4\,^{\circ}$C, just above the temperature at which stage-three recovery is observed. The irradiated samples were analyzed with positron lifetimes and the trapping model and only one trap was observed, the lifetime of which was 173 ± 5 ps. In a subsequent experiment with different copper samples,[16] proton-irradiated copper samples contained two traps with different lifetimes, one at 173 ± 5 ps and a second at 450 ± 20 ps. In those samples, the longer lifetime is identified as vacancy-impurity clusters, the formation of which may be controlled by the impurity levels in the copper samples.

In the first experiment above, the dose dependence and proton-energy dependence of the trapping rate were measured. In the analysis of those experiments it was found that a threshold in the defect-production calculation had to be set at high recoil energy (55 keV) in order to describe adequately the dependence of the trapping rate of the short-lifetime trap on varying proton energy. The conclusion that there are low-energy recoils that do not contribute to the formation of the short-lifetime trap observed in copper and the close similarity of the high-energy parts of the recoil spectrum produced by energetic protons and d-T fusion neutrons suggest that similar effects might be observed for both irradiations.

Comparing the results obtained in this experiment with those obtained in proton-irradiation experiments, we find both similarities and differences. The value found for the trap lifetime in the neutron irradiation, for example, is larger than for the proton case. The neutron-dose dependence is not completely linear whereas that for protons is linear. On the other hand, the absolute values of the trapping rate per unit dose calculated for the higher-dose irradiations is nearly equal in neutron and proton irradiations.

The trap lifetime found in the present experiment is 196 ± 6 ps which may be compared with 173 ± 5 ps for proton irradiation and 175 ± 5 ps for a sample damaged by repeated flexing. These values were all obtained in the same lifetime spectrometer and with the same model for analysis. The values for the proton-irradiated and flexed samples were determined both by single-lifetime analysis of saturated samples and by a trapping-model analysis. While these values do not differ greatly, the differences among the data are greater than the errors that were obtained by repeated measurement and statistical analysis. Thus there is evidence of some differences between the traps produced in the neutron irradiation and those produced in proton irradiations or flexing.

The data available in the literature for traps produced by deformation exhibit similar differences, spanning the range of values found here. In fact deformation-produced traps have been measured with lifetimes similar to either the proton or neutron one. Since the deformation studies were performed by various experimenters, the small differences in those data may be due to some systematic differences in the collection or analysis of the data. This cannot be the explanation for the present case, however, since all the samples were measured and analyzed in the same manner and repeat measurements were highly reproducible. The explanation must be found in differences in the trap.

For copper irradiated by protons at 16 MeV and neutrons at 14 MeV and observed in a transmission electron microscope, the size and distribution of the visible defects is identical.[17] Also the recoil spectra and consequently the distribution of damage energy for proton and neutron irradiations are very similar for energies above a few tens of keV. However, there is substantially more damage energy found in low-energy recoils in the proton irradiation than in the neutron irradiation. This means that during the irradiation there are more isolated vacancies and interstitials in the sample during proton irradiation. The dose and energy dependence of the trapping rate for proton-irradiated copper was interpreted in Ref. 1 to indicate that the contribution to the trapping rate of the recoils below a 55-keV threshold was negligible. This implies that the defects that are produced by low-energy recoils are free to migrate and to annihilate at sinks during the irradiation and the period afterwards until the samples are measured. Therefore it is possible that the difference between the two trap lifetimes reflects some small change in defect geometry caused by the flux of free-point defects acting on the defect structure during proton but not neutron irradiation.

The dose dependence obtained from the three neutron irradiations is shown in Fig. 3 along with the dose-dependence fit for 20-MeV proton data from Ref. 1. The trapping rate increases much more rapidly at low doses than at higher dose levels with the transition from the steeper slope to the more gradual occurring at about 5×10^{15} n/cm^2. A test to determine if the nonlinear dose dependence was actually a dose rate dependence is shown by the open squares. Those samples were irradiated at a high rate for a short time so that if the effect were a dose-rate effect one would expect the open squares to fall below the other points. That the data are all consistent confirms that the effect is a dose effect only.

A possible explanation of this effect is that there is partial saturation of impurity sinks in the sample by mobile-point defects during the irradiation. Since there are relatively few point defects produced by the neutron-damage mechanism, there may be traps such as impurity–single-vacancy complexes formed in the neutron-irradiated samples which would not survive in the proton-irradiation environment due to the much higher flux of point defects. If such traps were formed, they would make an additional contribution to the trapping rate obtained from the trap formed by the high-energy recoils. Also the extra contribution would tend to a steady state as new impurity atoms were included and the existing traps were destroyed by capture of additional point defects. In this interpretation, the trap lifetime determined in the analysis would be a composite of the high-dose lifetime and one characteristic of the second trap. The very slight difference between the low-dose trap lifetime and the high-dose one also supports this interpretation.

The slope of the dose dependence at high doses is about the same for the proton and the neutron irradiations and the absolute values of the neutron-induced trap rate are less than 20% larger than the proton-induced trap rate. Taking these data along with the damage-energy results in Table I, we conclude that a model of damage energy which uses a threshold in the recoil energy at 50 keV is necessary in order to explain the positron trapping-rate data. The experimentally determined slope for the dose dependence is also shown in Table I and it can be seen that the relative value of the measured production of traps and the calculated production of damage energy are in best agreement when the values calculated for 50 keV are compared with the experimental data. In fact the agreement between the relative values is quantitatively correct, the calculation and experiment agreeing within error. Considering the deficiencies in the proton-damage calculations discussed earlier, the agreement is surprisingly good.

This comparison of damage rates obtained in two separate experiments at dose rates differing by at least 2 orders of magnitude firmly establishes that the threshold concept is valid and that the contribution of the point defects in the two irradiation conditions is to modify in small ways the geometry and dose dependence of the major defect structure. The point defects may become part of large defects which are the result of migration and growth and may not appear as independent entities unless stabilized by some other feature in the metal such as an impurity.

Finally, the recovery of the defect structure shown in Fig. 4 suggests an additional low-temperature stage in the neutron-irradiated samples which does not appear in either of the proton-data sets. This stage may be the annealing of the postulated impurity defect structure which is the possible cause of the nonlinearity in the dose dependence in the neutron irradiation. Similar impurity effects have been observed in positron studies of the recovery of deformed copper.[18] The major recovery of the defect structure is at the same temperature at the recovery of the proton-irradiation samples, as would be expected from the other similar characteristics of these defects. The major stages in the irradiated deformation samples are similar to those found in the deformation studies where the defect structure is thought to be predominantly dislocation loops and vacancy clusters. A detailed comparison of the data of proton-irradiation and deformation is given in Ref. 1.

V. SUMMARY

The positron-lifetime characteristics have been investigated for pure annealed copper samples that have been damaged by irradiation with d-T fusion neutrons. The data obtained are similar to those from proton irradiation under similar circumstances. The positron lifetime determined for the trap differs slightly from the proton and neutron cases and there is an additional nonlinear component to the dose dependence of the trapping rate determined in the neutron experiment. The nonlinear dose dependence and extra stage in the recovery of the lifetime observed in the neutron-irradiated samples may be related to additional defects which are produced. The absence of these effects and the slightly shorter lifetime of the trap in the proton-irradiated samples may be due to a modification of the defect structure by the higher flux of freely migrating point defects in charged-particle irradiations. Comparison of the damage-production rates and the trap-production rates is quantitatively consistent with a model that excludes the damage produced by low-energy recoils below 50 keV from directly resulting in positron-trapping defects. Such a model, first suggested on the basis of the proton energy dependence of trap production, is now firmly established.

ACKNOWLEDGMENT

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