PROCEEDINGS OF THE INTERNATIONAL CONFERENCE ON CRYSTAL LATTICE DEFECTS, 1962, CONFERENCE
JOURNAL OF THE PHYSICAL SOCIETY OF JAPAN VOL. 18, SUPPLEMENT III, 1963

Energy Release in Irradiated Copper

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A very intensive program of energy release measurements associated with Stage I recovery of reactor irradiated copper has been undertaken during the past five years at Oak Ridge[1),2),3)]. During this period some modifications of experimental technique have occurred, but in general the experiment has been performed in the following way. The sample, usually of the order of 30 grams, has been suspended by a copper-constantan thermocouple. During the bombardment, varying from one week to three weeks, a heat switch has been closed keeping the sample in thermal contact with the cryostat wall so that its temperature was maintained below 22°K. Following the bombardment the heat switch was opened isolating the sample from the cryostat wall. Heat was applied to the sample by means of nuclear heating from the reactor. This heat (mostly γ rays) is homogeneously absorbed in the sample so that equilibrium conditions are always present. This means that it is possible to determine specific heats by the measurement of a time-temperature curve; the specific heat being proportional to the slope of the time-temperature curve. Mathematically the following equation exists:

$$C_p \Delta T = \gamma \Delta t$$

where T is the temperature, t is the time, C_p is the specific heat, and γ is the heat input, and

$$C_p = \gamma \frac{\Delta t}{\Delta T} \rightarrow \gamma \frac{dt}{dT} .$$

Thus, in the limit the exact specific heat can be determined by this method, whereas in the classical scheme of specific heat determination only the average specific heat in a temperature interval could be determined.

The heat switch and the thermocouple are the key items in this technique.

More recently, stored energy measurements have been made on B^{10} doped copper. A considerable greater amount of damage was introduced by the fissions of the B^{10} making for a relatively easy measurement. In addition, it was possible to measure the resistivity associated with the release of the stored energy. The sample design is as shown in Fig. 1.

The hollow cylinder was selected as it was easiest to cool by convection while the slit was added to increase the L/A ratio important to the resistivity measurements. The thermocouple and potential probes are also shown. The heat switch was also a novel idea. It is illustrated in Fig. 2.

The sample chamber wall is maintained

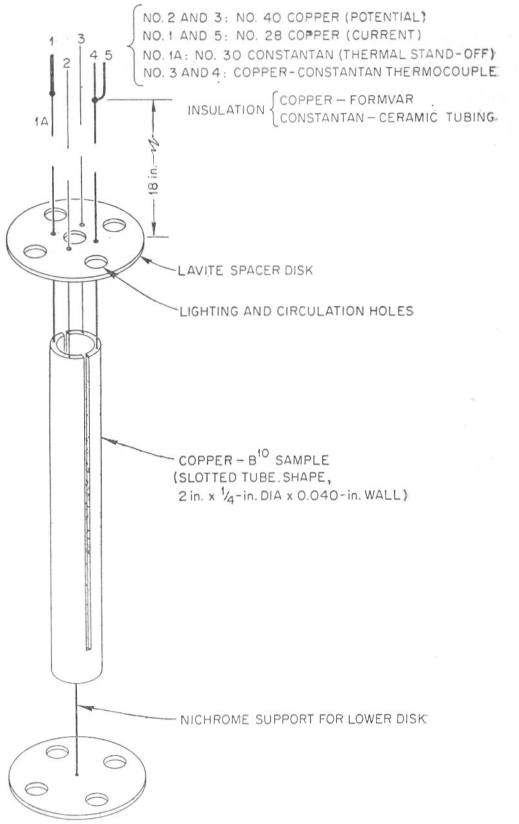


Fig. 1

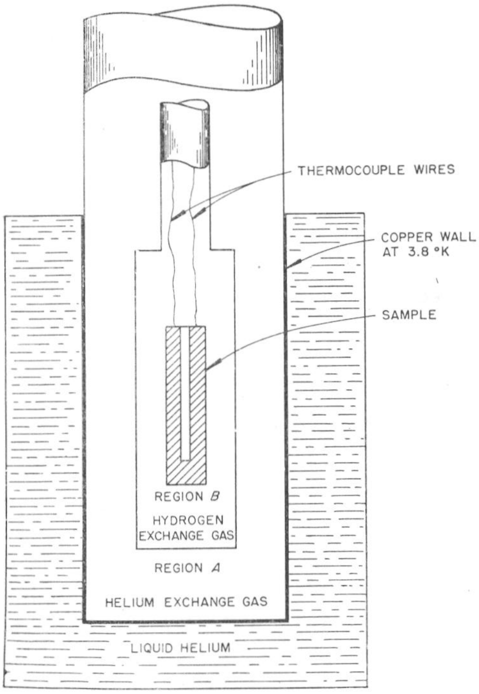


Fig. 2

at 3.8°K throughout the experiment. Helium exchange gas is added to region A. The vapor pressure was adjusted so that wall temperature of region B was at 10°K when the switch was closed. At this temperature hydrogen has a vapor pressure of about 100 microns and a sample temperature of about 20°K resulted. The switch was opened by dumping sufficient helium into region A to condense liquid at 3.8°K. At this temperature the vapor pressure of hydrogen is of the order of 10^{-2} microns, and the sample was isolated. In practice, some α-particles were produced and escaped during the bombardment so that a small amount of helium was generated in region B. This was not too serious, however, as the resulting heat leak was relatively small in the interesting temperature region and was reproducible. The reproducibility arose from the fact that region B was a static system throughout the experiment. This particular heat switch had the advantage of relative simplicity along with an additional advantage that the entire region B contained exchange gas (hydrogen) during the cool down so that the thermocouple quickly attained steady state conditions.

A time-temperature curve was measured following the bombardment. The slope of this curve is plotted as a function of absolute temperature in Fig. 3. Three such measurements were made following the bombardment. Only one of these, run A, contains stored energy. It can, however, be noted that there is excellent reproducibility in run 2 and run 3. The difference between run 1 and runs 2 and 3 represents the energy release. To evaluate this difference, it is necessary to know the effective heat input. It has been pointed out that the formation of α-particles results in a heat leak. The effective gamma heating, γ', can be evaluated from equation

$$\gamma'=\gamma-\delta=C_p\frac{dT}{dt}$$

since C_p, the specific heat is well known with the value given by Giaugue and Mead[4] being used here. Since the other variable dT/dt has been determined experimentally, the value of γ' can be readily determined. These values are shown as a function of temperature in Fig. 4.

The stored energy released in run A can be included in the specific heat term if it is treated as a specific energy release ε where the stored energy E is given by

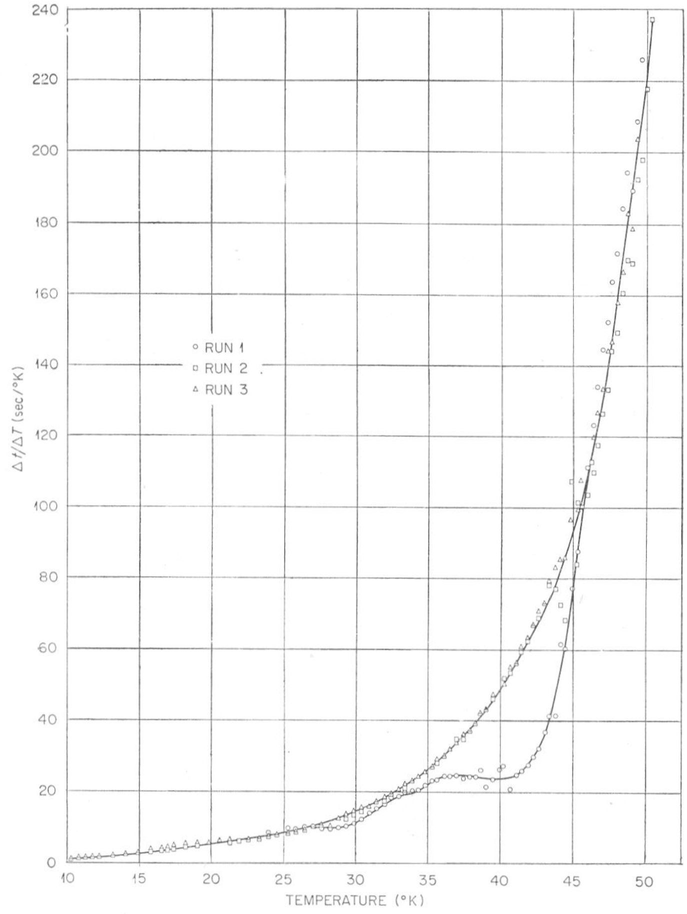
$$E=\int \varepsilon dT\,.$$

Thus, run A can be represented mathematically as

$$(C_p-\varepsilon)dT_A=\gamma' dt_A$$

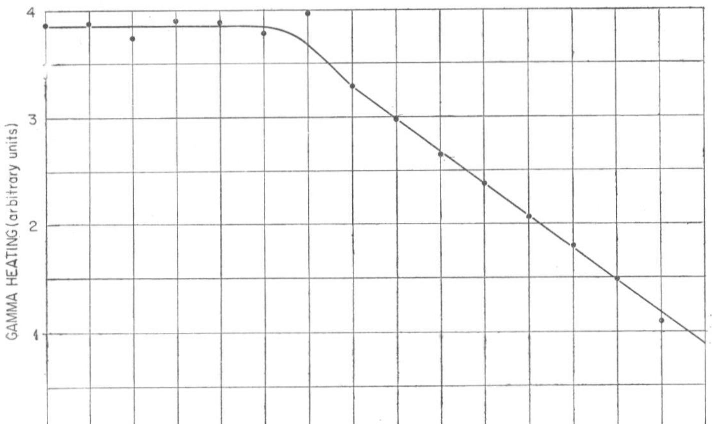
or $$\varepsilon=C_p-\gamma'\left(\frac{dt}{dT}\right)_A=\gamma'\left\{\left(\frac{dt}{dT}\right)_B-\left(\frac{dt}{dT}\right)_A\right\}\,.$$

Thus, the specific energy release ε is just equal to the effective heat input multiplied by the difference in the slope of the heating curve for run 1 and run 2 (or run 3). In Fig. 5 the lower curve is $\gamma'\left(\frac{dt}{dT}\right)_A$ and the upper curve is the specific heat. The shaded area is then the energy released, E or $\int\gamma'\left\{\left(\frac{dt}{dT}\right)_B-\left(\frac{dt}{dT}\right)_A\right\}dT$. This experimental value is found to be 4.54 cal/mol for one week bombardment (4×10^{17} thermal neutrons). Now following this determination, the sample was recooled and bombarded and an exact repetition of the stored



240
220
200
180
160
140
120
100
80
60
40
20
0
Δt/ΔT (sec/°K)
○ RUN 1
□ RUN 2
△ RUN 3
10
15
20
25
30
35
40
45
50
TEMPERATURE (°K)

Fig. 3



4
3
2
1
0
GAMMA HEATING (arbitrary units)
20
24
28
32
36
40
44
48
TEMPERATURE (°K)

Fig. 4

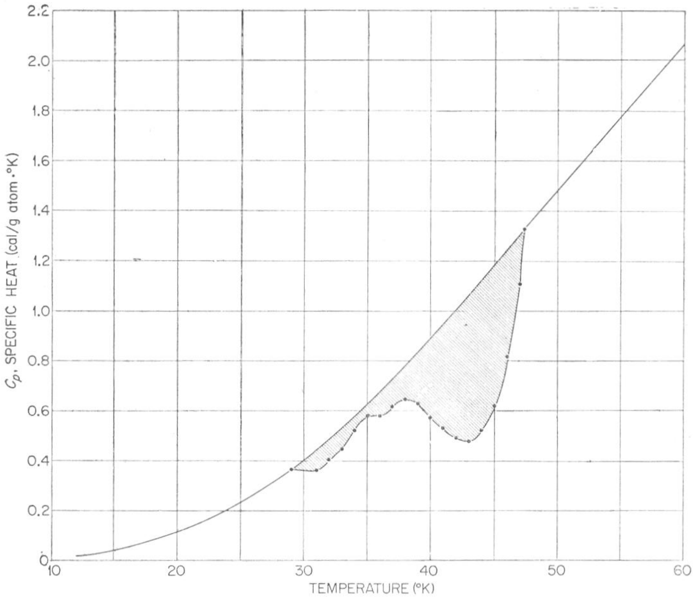


Fig. 5

energy run was made. This time, however, the resistance was read before and after the warm-up. It was found that the resistivity decreased by 1.77×10^{-8} ohm cm.

This decrease in resistivity can be used as a criterion for the amount of damage introduced into the sample. Thus, this experiment has shown that an energy release of 4.54 cal/mol will be associated with a recovery of 1.77×10^{-2} $\mu\Omega$cm. Thus,

$$\frac{E}{\Delta\rho}=\frac{4.54}{1.77\times10^{-2}}=\frac{256\ \text{cal/mol}}{\mu\Omega\text{cm}}=\frac{4.01\ \text{cal/gm}}{\mu\Omega\text{cm}}\,.$$

References

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DISCUSSION

Koehler, J. S.: Do you have any opinions regarding the fact that the close pair annealing stages are not observed in neutron irradiated copper?

Blewitt, T. H.: In the case of neutron irradiation localized regions with high defect densities arise, in contrast to the electron irradiation results where single pairs occur and it may be that the stress field arising from the large cluster results in changes in the migration energy of the defects. It should be noted that copper seems unique in this regard as substructure has been seen in most other metals in the stage I annealing peak.

Seeger, A.: Would Dr. Blewitt care to comment on the difference between the Granato-Nilan stored energy value of 7.1 cal/g/$\mu\Omega$cm and his value of 4.0 cal/g/$\mu\Omega$ cm?

Blewitt, T. H.: I have really had my say on our method and results. I obviously believe that our results are correct and I might add that from the similarity of the irradiation it would seem the same results should be obtained. The group at University of Illinois must feel the same way, so I should like Prof. Koehler to have his say also.

Mendelssohn, K.: Do the values of negative specific heat traced in your diagram correspond to equilibrium conditions in the metal at each temperature?

Blewitt, T. H.: I don't know for sure, but I certainly hope so. The fact that negative specific heat curve approximates the shape of the differential isochronal annealing curve would offer some evidence in support of equilibrium. Furthermore some measurements have been made at different reactor power levels which seem to support this view also.

Hasiguti, R. R.: You mentioned that you found I_B, I_C and I_D sub-stages in your stored energy spectrum. I understand the reason why I_A is missing. But how about I_E?

Blewitt, T. H.: Since a long bombardment was used it is probable that stage I_E is submerged in stage I_D.

Schmid, E. K. H.: I should like to make a short comment to the interesting results of Dr. Blewitt. It concerns the irradiation of Cu- and Ag-whiskers with α-rays. Ivoboda in our Institute in Vienna has found that the resistivity is raised with the dose of irradiation and that creep resistance was lowered. Whereas non irradiated samples did not show any creep, they began to elongate immediately after approaching the Po target. Elongations more than 1% could be detected.

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Recovery Stages in Noble Metals

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In this paper the recovery processes occurring in the temperature range of stage III and IV after irradiation, deformation, and quenching will be discussed. A comparison of the activation energies measured in this temperature range and a consideration of the kinetics during recovery suggest the migration of interstitials in stage III and the migration of vacancies in stage IV.

Point defects, which are created in metals either by irradiation or deformation at low temperatures, or by quenching from high temperatures anneal out with increasing temperatures in five different recovery stages[1]. Stage V, where recrystallisation and recovery of hardening occurs, will not be discussed here. Numerous experimental and theoretical works have been published in the past ten years, dealing with the identification of point defects and their attribution to particular recovery stages. There is considerable disagreement in the interpretation of results, especially as far as interstitial migration is concerned, which has been attributed to stage I (at very low temperatures) and to stage III (just below room temperature) by different authors (c.f. (2) to (6)).

According to the calculations by Huntington[7], the migration energy of an interstitial in copper amounts to 0.1–0.3 eV. Activation energies of this order are found in stage I (20° to 70°K), so that the migration of interstitials was frequently attributed to stage I (I_E in copper). This, however, leads to contradictions in the interpretation of the remaining recovery stages[2),3)]. Recent calculations[8] yield a value of 0.5 eV for the migration energy of interstitials (dumb-bell configuration) in copper, which is in good agreement with the experimental value of