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**NEUTRON IRRADIATION EFFECTS
IN FCC, BCC, AND CPH METALS**

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J. L. Brimhall and B. Mastel

INTRODUCTION

Neutron irradiation of materials is known to produce lattice defects that are resolvable by transmission electron microscopy. A number of factors are known to influence the type and magnitude of defects produced in a particular metal subjected to neutron bombardment. These include temperature of irradiation, purity of metal, total exposure, neutron flux, and energy spectrum of the neutrons. This report briefly reviews some of the work performed at Battelle-Northwest on radiation effects on metals. Three different metals--high-purity nickel, molybdenum and rhenium--have been irradiated under identical flux and temperature conditions and a comparison of the damage state in the different materials has been made as a function of total-exposure, irradiation temperature, and post-irradiation annealing treatments using transmission electron microscopy.

SUMMARY

It has been established that size and size distribution of radiation-induced point defect clusters can be directly related to the homologous temperature of irradiation. Similar damage structures in different materials can be induced by irradiating at equivalent homologous temperature. However, the role of the material parameters in affecting cluster size remains to be explored. Postirradiation

annealing behavior is highly dependent on the initial damage state; it appears that large clusters and a distribution of cluster sizes is necessary for significant loop formation during annealing.

DISCUSSION

Samples were in the form of thin discs 0.5 in. diam by 0.003 in. thick. Samples were irradiated to exposure in the range 1×10^{18} to 1×10^{20} nvt at a temperature of 50 °C unless otherwise specified. For comparison purposes, the range 0.5×10^{19} to 1×10^{19} nvt will generally be used. The materials were of a purity 99.9+% or better.

Figure 1 illustrates the damage formed in these materials after a fluence of approximately 6×10^{18} nvt ($E > 1$ MeV) at 50 °C. The irradiation temperature in terms of the homologous temperature is shown. The damage in each metal is characterized by a high density of point defect clusters. The average size of the clusters varies from 35 Å in rhenium to 60 Å in molybdenum and to 100 Å in nickel. The density of cluster defects is inversely proportional to their size. Rhenium has a density of $4 \times 10^{16}/\text{cm}^3$ with nickel $2 \times 10^{16}/\text{cm}^3$ and molybdenum in between. Because of inaccuracies in foil thickness the density values could vary by $\pm 20\%$.

A saturation limit in the density of clusters as a function of exposure

Neg 0661943-2

Ni ($0.187 T_M$)Mo ($0.11 T_M$)Re ($0.093 T_M$)Irradiated 6×10^{18} nvt ($E > 1$ MeV) at 50°C

FIGURE 1. Series of Micrographs Showing the Change in Size of the Point Defect Clusters in Nickel, Molybdenum and Rhenium with Changes in the Homologous Temperature of Irradiation. Samples Were Irradiated to Exposure of 6×10^{18} nvt ($E \geq 1$ MeV) at 50°C .

is reached in the materials. Molybdenum has been most extensively studied and saturation in the density of clusters occurs at approximately 1×10^{19} nvt. Further irradiation produces interaction of the clusters resulting in a very complex structure as illustrated in Figure 2. Increasing the exposure from 1×10^{19} to 1×10^{20} nvt produces coagulation of the spots and development of visible loops, eventually leading to a tangled dislocation

network. The density of the smallest clusters is actually lower at 1×10^{20} than at 1×10^{19} nvt. Although data on the other two materials is not as extensive, nickel appears to reach saturation in the range 0.5×10^{19} to 1×10^{19} and rhenium has not reached saturation at a level of 1.5×10^{19} .

One major block to meaningful interpretation is the inability to determine the nature of these defects. Dark field analysis has been attempted.

Neg 0662153-7

 1×10^{19} 5×10^{19} 1×10^{20}

Molybdenum, Irradiated at 50 °C

FIGURE 2. Series of Micrographs Illustrating the Interaction of the Clusters and Development of a Tangled, Dislocation Network with Increasing Neutron Exposure in Molybdenum.

Because there appears to be some controversy among experts on the correct interpretation of dark field contrast, it is not proper now to say that all are vacancies or interstitials or a mixture of both.

From work on molybdenum irradiated at elevated temperatures and molybdenum given postirradiation annealing treatments where large interstitial loops have definitely been identified, at least some of the small clusters can be inferred as interstitial in nature. The only point defect that could

have significant mobility in these materials at this temperature would be the interstitial. One interpretation of the difference in the damage characteristics of the three metals can be traced to a difference in the melting point; hence, differences in mobility of the point defects. The increased atomic mobility in nickel compared to that of rhenium could lead to the observed increase in cluster size. The differences are subtle but real.

To further illustrate the effect of irradiation temperature, we can compare

the same metal, in this case rhenium, irradiated at two different temperatures, shown in Figure 3. The size of the clusters in rhenium has increased by a factor of 2 to 3 by increasing the irradiation temperature from 50 to 500 °C.

In addition, we can compare two dissimilar metals irradiated at equivalent homologous temperatures. Figure 4 compares nickel irradiated at 50 °C ($0.19 T_m$) with rhenium irradiated at 500 °C ($0.22 T_m$). Although there is an apparent difference in density (much of this can be accounted for by the difference in foil thickness), the size of

the cluster is quite similar. Therefore, by irradiating dissimilar metals at similar homologous temperatures, similar damage structures can be induced even in metals of different crystal structure.

It has also been postulated that the small black spots (clusters) are a vacancy-type defect and are a direct result of clustering of vacancies located near the core of the displacement spikes. The difference in cluster size would then reflect the difference in size of the spikes due to differences in material parameters such as atomic

Neg 0662357-1

1.5×10^{19} at 50 °C
($0.093 T_M$)

2.0×10^{19} at 500 °C
($0.22 T_M$)

Effect of Temperature of Irradiation on Rhenium

FIGURE 3. Point Defect Clusters Formed in Rhenium after Exposure at two Different Temperatures Showing Increase in Size with Increase in Temperature.

Neg 0662357-2

Ni
 1×10^{19} nvt at 50 °C
 (0.187 T_M)

Re
 2×10^{19} nvt at 500 °C
 (0.22 T_M)

FIGURE 4. Point Defect Clusters in Nickel and Rhenium Irradiated at Nearly Equivalent Homologous Temperatures Showing the Similarity in the Size of the Clusters.

mass and threshold energies of the different metals. Even in this case, however, the very short-range mobility of the vacancies in the region of the spike may be significant.

Annealing of the radiation damage was similar except for one significant difference in one of the metals. Figure 5 illustrates the damage state after annealing in nickel, molybdenum and rhenium respectively. All samples were previously irradiated to 6×10^{18} nvt at 50 °C. The annealing temperatures in terms of homologous temperature are shown. These were 1000 °C for rhenium, 750 °C for molybdenum, and 450 °C for nickel.

In rhenium and molybdenum, there is no growth of the defect clusters into resolvable dislocation loops, rather a gradual decrease in the density of clusters occurs. In nickel, however, there is growth of the clusters into resolvable dislocation loops as well as a concurrent decrease in the density of the clusters and/or loops.

The disappearance of the clusters at this temperature can be explained through the breakup of vacancy clusters and annihilation at sinks, in particular the interstitial clusters. In addition, however, a significant fraction of the point defects in nickel must be migrating to loops of their own

Neg 0661943-3

Ni ($0.41 T_M$) Mo ($0.35 T_M$) Re ($0.37 T_M$)Irradiated 6×10^{18} nvt and Annealed

FIGURE 5. Series of Micrographs of Nickel, Molybdenum, and Rhenium Which had been Irradiated to 6×10^{18} nvt Showing the Change in Size and Density of the Point Defect Clusters after Annealing. Dislocation Loops are Observed only in Nickel after Postirradiation Annealing.

kind to account for the loop growth. This process is analogous to that observed in quenched metals in which large loops are observed to grow at the expense of smaller loops. This apparently happens only in the irradiated nickel because the clusters are initially larger in the nickel (in fact, some resolvable loops are observed in the as-irradiated nickel.)

A very similar situation is noted in molybdenum containing carbon as impurity. Figure 6 compares postirradiation annealing behavior in pure molybdenum with that containing carbon. Significantly larger clusters are seen in the carbon bearing as compared to the high-purity molybdenum. After 750°C there are relatively distinct large dislocation loops in the molybdenum

HIGH PURITY MOLYBDENUM, <10 ppm CARBON

Neg 0640055

Nonirradiated

As irradiated

Annealed 575 °C

Annealed 750 °C

Annealed 900 °C

MOLYBDENUM CONTAINING 450 ppm CARBON

Nonirradiated

As irradiated

Annealed 575 °C

Annealed 750 °C

Annealed 900 °C

FIGURE 6. Series of Micrographs Showing the Differences in the Annealing Behavior of Irradiated Molybdenum of Two Different Purities.

containing carbon. This behavior is directly analogous to that observed in nickel. Loop growth can occur when loops have attained a sufficiently large size in the as-irradiated state and when cluster sizes cover a sufficiently wide distribution. In high-purity molybdenum and rhenium where there are very small clusters of uniform size, there is no tendency for certain loops to grow at the expense

of others. These observations do not differentiate between loop growth by lattice migration of vacancies or by some self-climb process. These conclusions also only apply to that exposure range $\leq 10^{19}$ nvt. If high-purity molybdenum is irradiated to higher exposure such as that shown in Figure 2, then large dislocation loops are formed during annealing.

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