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CASCADE COLLAPSE IN COPPER AND NICKEL.

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Molecular dynamic computer simulations in Cu and Ni have suggested that (i) within a displacement cascade the material exists in a highly excited, perhaps even molten, state and (ii) many atoms are transported from the cascade region by replacement collision sequences. This model offers a simple mechanism for explaining dislocation loop formation from a displacement cascade that is based on resolidification kinetics. The molten zone resolidifies from the periphery and contracts inward. Initially the difference in the atomic density of the cascade region and the lattice is small and a perfect crystal regrows. As resolidification proceeds, the difference in density increases due to the higher fraction of missing atoms, and vacancies begin to "freeze-out". The distribution of the vacancies depends on the rate at which the resolidification front moves. At slow rates the vacancies become concentrated near the cascade center, creating a high vacancy supersaturation which favors loop formation. At high solidification rates, the vacancies are distributed throughout the cascade volume and loop formation is less likely. This mechanism is similar to zone refining.

The validity of this mechanism was studied by using transmission electron microscopy to investigate the damage structures produced in Cu, Ni and Ni-alloys by heavy-ion irradiations. The development of the damage structure and the fate of individual defects as the ion dose was increased was investigated at 30 and 300 K. The irradiations and the electron microscopy were performed *in situ* in the HVEM-Accelerator Facility at Argonne National Laboratory. In all materials dislocation loops formed at 30 K, indicating that the loop formation process is athermal, and occurs within the lifetime of the thermal spike. The low temperature loop densities were approximately 50% lower than those at room temperature, in agreement with the idea that the rate of energy dissipation is important. The slower cooling at room temperature provides more time for "zone-refining" and more vacancies are formed near the cascade center. Also, as the irradiation proceeds, new loops were produced and pre-existing ones disappeared, moved or changed Burgers vector. These effects are readily understood in terms of the local melting model. That is, assuming that an existing loop is engulfed and hence destroyed by a second displacement cascade, then this new cascade volume will have a probability for forming a loop which can have a different position and/or Burgers vector from the first loop.

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Abstract : In-situ TEM studies of the development of the damage structure produced by heavy ion irradiations have been performed in copper and nickel to investigate the possibility that melting occurs in local regions within displacement cascades. These experiments reveal that as the ion dose increases additional loops form from isolated displacement cascades, but more surprisingly some of the pre-existing loops are annihilated, change position, size and/or Burgers vector. It was also found that the probability for loop formation and the defect image size are greater in copper than in nickel even at temperatures well below stage III. It will be demonstrated that these observations provide supporting evidence, albeit indirect, that local melting occurs within the cascade core. These results will be compared to the molecular dynamic computer simulations of the damage created by low energy self-ions in copper and nickel.

Introduction.

The response of a material to bombardment with energetic particles, neutrons or heavy-ions, is known to be dependent on material parameters (e.g. structure, impurity concentration, cohesive properties etc) and irradiation parameters (flux, fluence, irradiation temperature, ion mass and energy etc). However, despite the large body of information that has now been gathered the question as to why materials respond differently under irradiation remains unanswered. In this paper we address the specific issue of why the probability for loop formation from displacement cascades is material sensitive ^[1]. For this study copper and nickel were selected because they have the same structure and density, and the cascade dynamics are expected to be similar. However, the materials respond differently to heavy-ion and neutron irradiation. For example, bombardment with energetic heavy-ions produces more loops in copper than in nickel. For room temperature irradiations with 50 keV Kr⁺ ions approximately 60 % of the cascades form dislocation loops in copper ^[2] as compared to 30 % in nickel ^[3]. It is also found that the average and the maximum loop size is greater in copper than in nickel, indicating that a higher fraction of the vacancies are retained in dislocation loops in copper than in nickel. The efficiency for retaining the created vacancies in a loop is approximately 20 % in copper compared to 10 % in nickel ^[2]. A similar materials dependence on loop formation probability is obtained when the irradiations are performed below the main stage I recovery peak, although the probability is decreased by approximately 50 % from the room temperature value in both materials.^{[4][5]}

Molecular dynamic computer simulation results.

The molecular dynamic computer simulation code employed in these calculations was the modified version of Superglob^[6]. The pair potentials used were the Gibson II form of the Born-Mayer ^[7] potential for Cu and the Johnson-Erginsoy potential ^[8] for nickel. These potentials tend to underestimate and overestimate the melting temperature of copper and nickel respectively, and thus will exaggerate the actual differences. Nevertheless the potentials are useful because their computational efficiency is important for the simulations of high energy events. The recoil event is studied by giving all the cascade energy to one atom and then following all subsequent events until a near quiescent state is reached. For the initial studies the lattice temperature was 0 K. The final distribution of the point defects has the interstitials surrounding the vacancy rich cascade center. The interstitials are produced by replacement collision sequences.

The most striking result from these computer simulations is the observation of local melting in the central region of the cascade core ^{[9][10][11]}. Figure 1 which is a cross-sectional view through the displacement cascade, shows the damage distribution generated by a 5 keV self-ion in copper (a) and nickel (b) as a function of time after the event. Initially, the central region of the cascade core is in a highly disturbed configuration in both materials. The radial pair correlation functions, Figure 2, show that the (200) peaks do not exist initially and the distribution for copper is identical to that for simulated liquid copper. With time order is restored to this region. However, an interesting difference is noted between copper and nickel in that the disordered state persists longer in copper than in nickel (compare Figures 1 b and d, and Figure 2).

This picture of the cascade has suggested that the formation of a vacancy loop from a displacement cascade is related to the rate of energy removal from the molten zone. Resolidification of the molten zone occurs initially at the cooler periphery. The degree of perfection of the regrown material depends on the degree of atomic rearrangement that can take place at the interface. For sufficiently slow regrowth rates there will be sufficient time for atomic rearrangement to occur and a defect free structure will be produced. In this situation as the solidification front proceeds inward, the atomic density fluctuations in the liquid increase due to the missing atoms, and vacancies will begin to "freeze out". A high concentration of vacancies will therefore be obtained near the cascade center, creating a situation favorable for loop formation. At faster resolidification rates where there is insufficient time for atomic rearrangement at the liquid/solid interface the vacancies are created throughout the cascade volume, creating a situation less favorable for loop formation. In the case of nickel and copper, the condition for loop formation (high vacancy concentration in the cascade center) is obtained more frequently in copper because the cascade core region remains above the melting temperature longer than in nickel. This is a consequence of the lower melting temperature of copper and implies that the loop formation probability should scale with the melting temperature of the material; this issue is addressed in a separate paper^[2].

The computer simulations have also been performed for different lattice temperatures^[11]. At higher lattice temperatures the resolidification is slower allowing for more perfect regrowth in the initial stages and therefore a higher vacancy concentration near the cascade center. Thus conditions favorable for loop formation are more readily obtained in irradiations performed at room temperature

than at 30 K; this is consistent with the experimental observations.

Electron microscopy results

High-purity samples of copper and nickel were irradiated in the electron transparent condition *in-situ* in the HVEM-Accelerator Facility at Argonne National Laboratory ^[12]. The availability of a liquid helium, double-tilt sample stage for the HVEM, and the coupling of the microscope to an accelerator affords one the capability of observing the behavior of individual defects as the irradiation dose and temperature are altered. For these experiments the ion fluence ranged from $2 - 175 \times 10^{15}$ ions m^{-2} and the ion flux was 2×10^{14} ions $m^{-2} sec^{-1}$. Copper samples were irradiated with 50 keV Kr^{+} ions and the nickel sample with either 50 keV Kr^{+} , 50 keV Ni^{+} or 100 keV Ni^{+} ions.

The irradiations and the electron microscopy were performed at sample temperatures of 30 and 300 K. At 30 K there is no long range point defect mobility in either material, whereas at 300K vacancies are mobile in Cu, but have only limited mobility in Ni.

The development of the damage structure as a function of ion dose was determined from comparison of micrographs taken in the same sample area and under the same diffraction conditions. The Burgers vector of the defect was determined by using a combination of the $g \cdot b = 0$ invisibility condition and by observing the change in image symmetry for different operating reflections and then comparing these results with computer simulated images^[13].

Initially the defect production rate is linear with increasing ion dose,

but as the irradiation proceeds to higher doses ($> 10^{15}$ ions m^{-2}) the production rate decreases. This trend was observed in copper and nickel for all irradiation conditions employed. An example of this behavior is shown in Figure 3 for the room temperature irradiations of nickel with 50 and 100 keV Ni^+ ions. The reason for the change in defect production rate as the irradiation proceeds can be understood by comparing the same area of the sample at different ion doses, Figures 4 and 5. In Figure 4 the same area of a Cu sample after an irradiation at 30 K with 50 keV Kr^+ ions to a total ion dose of a) 4×10^{15} ions m^{-2} and b) 6×10^{15} ions m^{-2} are compared. The dislocation loops indicated by arrowheads are present at the lower ion dose, but have disappeared after the second irradiation. One possible explanation for this loop loss is that the loops are glissile and have slipped to the foil surface where they are annihilated. However, the two loops, marked "F", have been identified as having a Burgers vector of the type $a/3\langle 111 \rangle$; that is, they are sessile Frank loops. For these loops to slip to the foil surface they would have to be "affected" by the second ion irradiation, converting to perfect loops which then glide to the foil surface. While this is a possible explanation for this effect, it cannot account for the observation that loops can have a different Burgers vector after the second irradiation. An example of this is shown in Figure 5 in which dark-field micrographs taken with different operating reflections before and after the second irradiation are presented. The loop marked by the arrowhead in Figure 5 is seen to have different image characteristics after the second irradiation. The l -vector which is drawn from the center of the black lobe to the center of the white lobe has shifted by about 90° and the operating reflections in which the image exhibits strong and weak contrast are reversed. These changes in image characteristics are consistent with the loop Burgers vector changing from either $b = a/3 [111]$ or $b = a/3 [\bar{1}\bar{1}\bar{1}]$ to a Frank loop having a Burgers vector of either $b = a/3 [\bar{1}11]$ or $b = a/3 [1\bar{1}1]$.

Again some defects were observed to have been annihilated by the second irradiation, one example is indicated by the arrow.

In addition to the loss of some loops and the change in Burgers vector of others it was also observed that the second irradiation caused some neighboring loops to coalesce, while yet others changed their position and/or size.

The magnitude of these effects has been quantified in terms of a cross-sectional area, σ , for irradiations in copper and nickel. The cross-sectional area and the effective cross-sectional diameter, d , were determined from the relationship:

$$\sigma = \pi d^2 / 4 = N_A / N_I (1/\Delta\phi).$$

In this expression N_A is the number of defects affected, N_I is the number of defects present after the initial dose ϕ_1 , and $\Delta\phi$ ($= \phi_2 - \phi_1$) is the incremental ion dose. Values for the cross-sectional area and diameter are presented in Table 1 for a variety of irradiation conditions of copper and nickel. The magnitude for the effective cross-sectional diameter ranges from 7.0 nm (for the 50 keV Kr⁺ ion irradiation of copper at 30 K) to a value of 1.4 nm (for the 100 keV Ni⁺ ion irradiation of nickel). These values are of the same order of magnitude as the average image diameters which are also quoted in Table 1. From this it appears that for these irradiation conditions the second cascade must fall very close to the pre-existing dislocation loop if that loop is to be affected. Alternatively, the cross-sections are larger but with a smaller efficiency for loop removal. The fraction of loops affected by a following irradiation increases with the total ion dose. The percentage of loops affected is higher in copper than in nickel

which probably reflects that the higher loop density in copper.

Discussion

The molecular dynamic computer simulations of low energy displacement events in copper and nickel have shown that local regions of the central core of the cascade exists in a molten state. As the molten zone resolidifies vacancies are created. The distribution of vacancies depends on the amount of atomic rearrangement that occurs at the solidification front. The faster the resolidification the less perfect the regrowth and the more dispersed the vacancy distribution. If sufficient time is allowed for atomic rearrangement at the liquid/metal interface the vacancies will be formed near the cascade center, creating a situation favoring loop formation. This model for loop formation from a displacement cascade provides an explanation for the material and temperature dependence of the loop formation probability that is observed experimentally in copper and nickel. These dependences can also be satisfactorily explained within the precipitation and growth^[14], and the thermotransport models^[15] for loop formation from a displacement cascade. However, the loss of loops, change in Burgers vector, size and location during continued irradiation are most readily understood in terms of the molten zone model. A possible scenario is: If a new cascade is created such that it engulfs the pre-existing loop, that loop will be destroyed and the vacancies from the loop will further decrease the atomic density within that volume. A new loop will be created from this volume with some probability which will be greater than from a single isolated displacement cascade, but will still be relatively low because of the low probability of a loop forming from an isolated displacement cascade. The new loop may have a different Burgers vector, location or size from the original loop. Although this

experimental evidence does not directly verify the existence of a local molten zone within displacement cascades in copper and nickel, it does provide strong supporting evidence.

From our understanding of the changes in the dislocation loop population during irradiation, the decrease in the defect production rate with increasing ion dose can be understood (Figure 3). Initially, the defect production rate depends on the formation of dislocation loops from individual displacement cascade. As the ion dose increases there will be some loops annihilated when impacted by a second ion, some enhanced loop formation will also occur when these fresh cascades spatially overlap with previously created uncollapsed cascades. These effects are probably about equal in magnitude. At the ion doses where the defect production rate begins to decrease the dominant effects will be the annihilation/ reformation of loops and loop coalescence processes that occur when a fresh cascade lands in the vicinity of a pre-existing loop.

Conclusions

By studying the fate of pre-existing dislocation loops during continued irradiation it has been found that new loops appear from isolated displacement cascades, but surprisingly some loops are annihilated, some coalesce with neighbors, while others are replaced with new loops that have a different Burgers vector, size or position. These effects are most readily accounted for within the framework of the local melting model that has been developed from molecular dynamic computer simulations of low energy cascades in copper and nickel.

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Figure captions

Figure 1 Comparison of the projections of a (100) cross-sectional slab near the center of a 5 keV displacement cascade in (a) copper and (b) nickel as a function of time.

Figure 2. Radial pair correlation functions of the atoms in the disordered zones of displacement cascades in (a) Cu and (b) Ni as a function of time. A comparison with the correlation function for simulated liquid copper is made in (a).

Figure 3. Defect density as a function of ion dose for nickel irradiated at 300 K with 50 keV and 100 keV Ni⁺ ions.

Figure 4. Dark field micrographs comparing the same area of a copper foil irradiated and examined at 30 K. The total ion dose was in (a) 4 and (b) 8×10^{15} ions m⁻². The diffracting vector was (200).

Figure 5. Dark field micrographs comparing the same area of a copper foil irradiated at 300 K with 50 keV Kr⁺ ions. The total ion dose was (A) 4 and (B) 8×10^{15} ions m⁻². The diffracting vectors were (1) 200, (2) 220 and (3) $\bar{2}20$.

Table 1. Incremental ion dose, percentage of loops affected by the ion dose increment, cross-sectional area and effective diameter and the average image diameter for loops produced by 50 and 100 keV Ni⁺ ion irradiations in nickel and 50 keV Kr⁺ ion irradiations in copper at 30 and 300 K.

ion dose x10 ¹² ions cm ⁻²	ion dose increment $\Delta\phi$ 10 ¹² cm ⁻²	% loops affected	cross- sectional area, σ 10 ⁻¹⁴ cm ⁻²	cross- sectional diameter d (nm)	average image diameter (nm)
<u>100 keV Ni⁺ → Ni</u>					
0.19	0.38	1.8	4.12	2.29	2.68
0.57	0.58	1.0	1.58	1.42	2.68
1.15	1.15	2.5	2.13	1.65	2.68
2.30	1.15	2.5	2.29	1.71	2.68
3.45	1.14	2.2	1.81	1.52	2.68
4.59					
<u>50 keV Ni⁺ → Ni</u>					
0.6	0.42	2.8	6.68	2.91	
1.02	0.98	3.4	3.45	2.10	
2.0					
<u>50 keV Kr⁺ → Cu at 30 K</u>					
0.4	0.2	7.8	39.0	7.0	3.76
0.6					
<u>50 keV Kr⁺ → Cu at 300 K</u>					
0.4	0.4	5.3	13.0	4.1	3.76
0.8					

References.

1. English C.A., and Jenkins, M.L., "Insight into cascade processes arising from studies of cascade collapse," Mats Science Forum, vols. 15 - 18, 1987. pp 1003 - 1022.
2. Smalinskas, K., Robertson, I.M., Averback, R.S., and Kirk M.A., "The formation of vacancy loops from Kr⁺ ion irradiations of Cu-Ni alloys," these proceedings.
3. Vetrano, J.S., "Heavy-ion damage in Cu, Ni and Ni-alloys," Ph.D. Thesis, University of Illinois, Urbana-Champaign (1990).
4. Vetrano, J.S., Robertson, I.M., and Kirk, M.A., "Experimental evidence favoring local melting within heavy ion generated displacement cascades in copper," Scripta Met., vol 24, 1990, pp 157-162.
5. Kirk, M.A., Robertson, I.M., Jenkins, M.L., English, C.A., Black, T.J., and Vetrano, J.S., "The collapse of defect cascades to dislocation loops," J.Nucl. Mat., vol.149, 1987, pp 21 - 28.
6. King, W.E. and Benedek.R., "Molecular Dynamics Simulation of Low Energy Displacement Cascades in Cu," J. Nucl. Materials, vol. 117, 1983, pp. 26 - 35.
7. Gibson,J.B., Goland, A.N., Milgram, M. and Vineyard, G. H., "Dynamics of radiation damage," Phys. Rev. vol 120, 1960, pp. A1229 - A1253.
8. Erginsoy,C., Vineyard, G.H., and Englert, A., "Dynamics of Radiation Damage in a Body-Centered cubic lattice," Phys. Rev., vol. 133, 1964, pp. A595 - A606.
9. de la Rubia, T.D., Averback, R.S., Benedek, R. and King, W.E., "Role of thermal spikes in energetic displacement cascades," Phy. Rev.Let., vol. 59, 1987, pp. 1930 - 1933.
10. de la Rubia,T.D., Smalinskas,K., Averback, R.S., Robertson, I.M., and Benedek, R., "Mechanisms of cascade collapse," Mat. Res. Soc. Symp. Proc. , vol. 138, 1989, pp 29 - 34.
11. de la rubia, T.D. Averback, R.S., Hsieh, Horngming, and Benedek, R., "Molecular dynamics simulation of displacement cascades in Cu and Ni: thermal spike behavior," J.Material Res., vol 4, 1989, pp 579 - 586.
12. Taylor, A., Allen, C.W. and Ryan, E.A., "The HVEM-Tandem accelerator facility at Argonne National Laboratory," Nucl. Insts and Methods, vols. B24/25, 1987, pp. 598 - 602.

13. Saldin, D.K., Stathopoulos, A.Y. and Whelan, M.J., " Computer simulations of loop images in FCC materials," Phil. Trans. Roy. Soc., vol 292, 1979, pp. 523 - 535.

14. Praminak D., and Seidman, D.N., " The irradiation of Tungsten with metallic diatomic molecular ions : Atomic Resolution Observations of Depleted Zones," Nucl. Inst. and Meth., vol. 209-210, 1983, pp.453 - 459.

15. Protasov, V.I., and Chudinov, V.G., " Evolution of Cascade Region during the Thermal Spike," Rad Effects, vol66, 1982, pp. 1 - 7.