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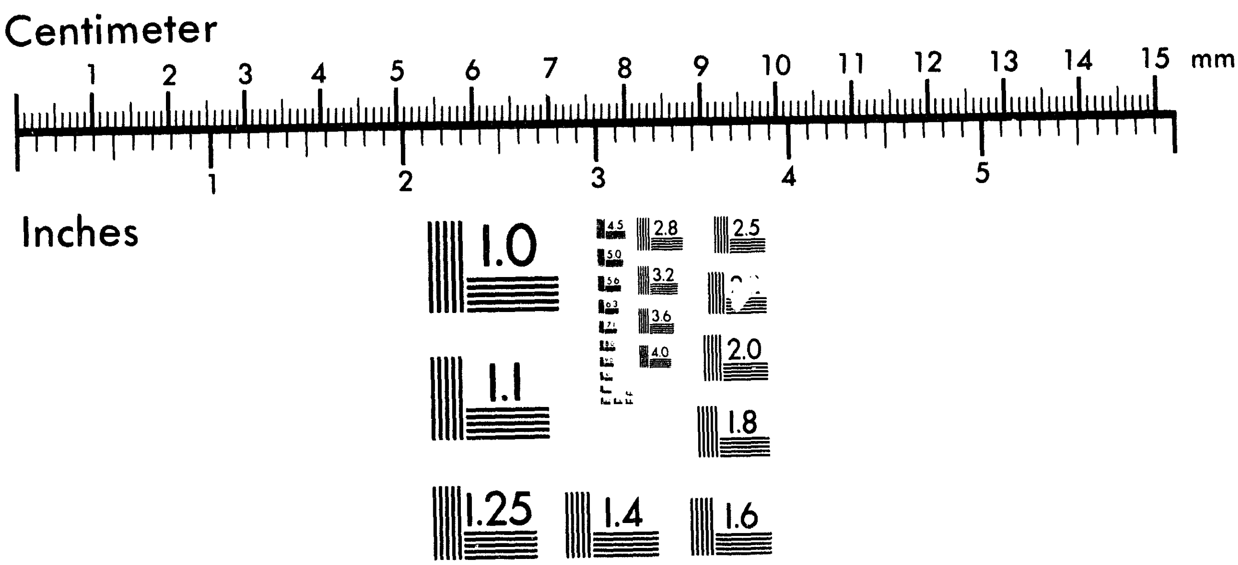

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POST-IRRADIATION DEFORMATION CHARACTERISTICS OF HEAVY-ION IRRADIATED 304L SS

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ABSTRACT: Post-irradiation deformation behavior in Ni-ion-irradiated 304L stainless steel (SS) is examined as a function of radiation dose and deformation temperature. For similar strain levels, specimens exhibit a transition from dislocation slip to deformation-induced twinning at 25°C with increasing radiation dose. At 288°C twinning is no longer observed and highly localized slip occurs by the formation of narrow "channels" containing a reduced defect density. The observations are discussed in terms of radiation-induced defect character and expected deformation mechanisms.

KEYWORDS: Ion-irradiation, radiation damage, stainless steel, deformation twinning, dislocation channeling

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Significant hardening and reduced ductility can occur in metallic alloys as a result of high-energy particle bombardment. This has become a particular concern in commercial light water reactors (LWRs) where neutron irradiation exposure has promoted premature failure of stainless alloy reactor core components[1]. There appears to be a critical radiation dose above which an increased susceptibility to irradiation assisted stress-corrosion cracking (IASCC) is observed. Recent research has indicated that radiation induced segregation does not fully explain IASCC susceptibility in many stainless steels[2] and that radiation hardening may play a role in the cracking process[2-6]. Greater understanding of deformation processes in irradiated microstructures may help in providing some answers to the increasing number of questions surrounding IASCC.

Much of the current understanding of deformation process in

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irradiated metals is derived from neutron-irradiated copper single crystals[7-11], while few investigations[12,13] can be found on deformation of irradiated polycrystalline alloys, such as 304 stainless steel (SS), in the dose and temperature ranges relevant to commercial LWRs. In general, prior studies with copper found that glide dislocations generated during post-irradiation deformation interacted with loops formed during radiation. This resulted in the annihilation of the loops and produced "channels" relatively free of defects. Further deformation was concentrated in these obstacle-free channels while the surrounding material remained undeformed. Compared to copper, 304 SS has a much lower stacking fault energy (SFE) making direct comparisons of the two metals unreliable. Deformation behavior in unirradiated metals has been shown to depend on SFE and this trend is expected to occur in irradiated metals as well.

In order to investigate the process of deformation in irradiated stainless steel microstructures, 304L SS samples were irradiated to various dose levels with Ni^{++} ions at a temperature of 500°C. Heavy-ion irradiation can produce defect structures similar to the relevant neutron defect structures, and allows the generation of samples in a relatively short time period without the expense and hazards of neutron irradiation. Following irradiation, the samples were deformed in tension at room temperature or at 288°C. Microstructural changes were then documented with electron microscopy. Key issues concerning the deformation behavior of these samples will be discussed with respect to the observed mechanisms.

Experimental

Low-carbon commercial purity 304L SS (Fe-18.6Cr-8.9Ni-1.8Mn-0.46Si-0.016C-0.083N) tensile samples and 3-mm discs were prepared from 0.7-mm-thick sheet material. Cold-worked sheet material was initially heat treated at 900°C for 2 hours to produce recrystallized grains ~40μm in diameter. Electric discharge machining was used to produce the tensile samples as illustrated in Figure 1. The gage sections of the tensile samples were ground to 600-grit emery paper and electropolished in a 5% $HClO_4$/95% methanol solution to remove surface damage. Two-mm diameter areas on the gage section were ion-irradiated with Ni^{++} at an

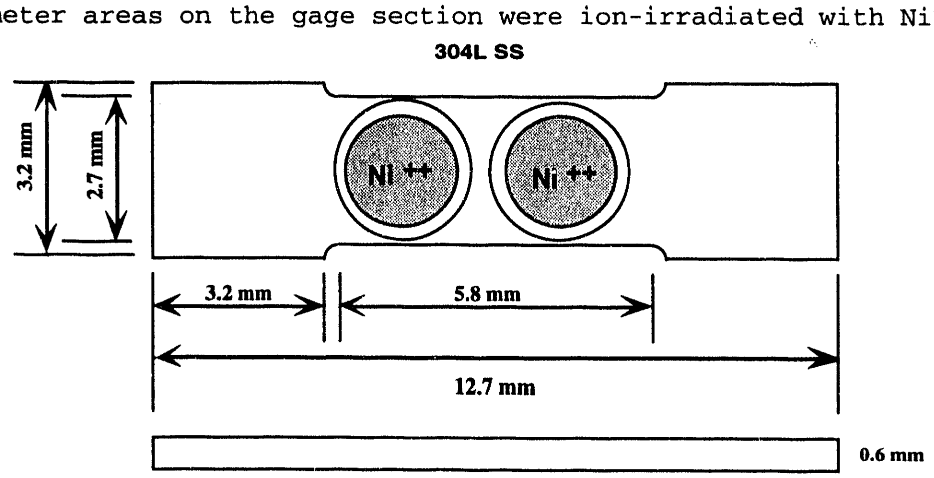


FIG. 1--Schematic of ion irradiation tensile sample

accelerating voltage of 5 MeV in a tandem accelerator. A damage rate of 1.6×10^{-3} dpa/s was used such that a level of 1 dpa was achieved in approximately 10 minutes. Following ion irradiation, the samples were deformed in tension at 25 or 288°C using strain rates ranging from 2×10^{-4} s^{-1} to 2.5×10^{-6} s^{-1}. Reported strains are based on total elongation of the samples not accounting for the difference in deformation between the irradiated and unirradiated regions. Post-deformation scanning electron microscope (SEM) examination of the surface was first performed, followed by transmission electron microscopy (TEM) examination. Foil preparation consisted of removing approximately 0.5μm from the bombarded surface (halfway through the total irradiation depth) and then thinning from the back side with the same solution used for electropolishing. The microstructures of the samples were examined in a conventional TEM operated at 120 KeV.

Results

Radiation-Induced Microstructures

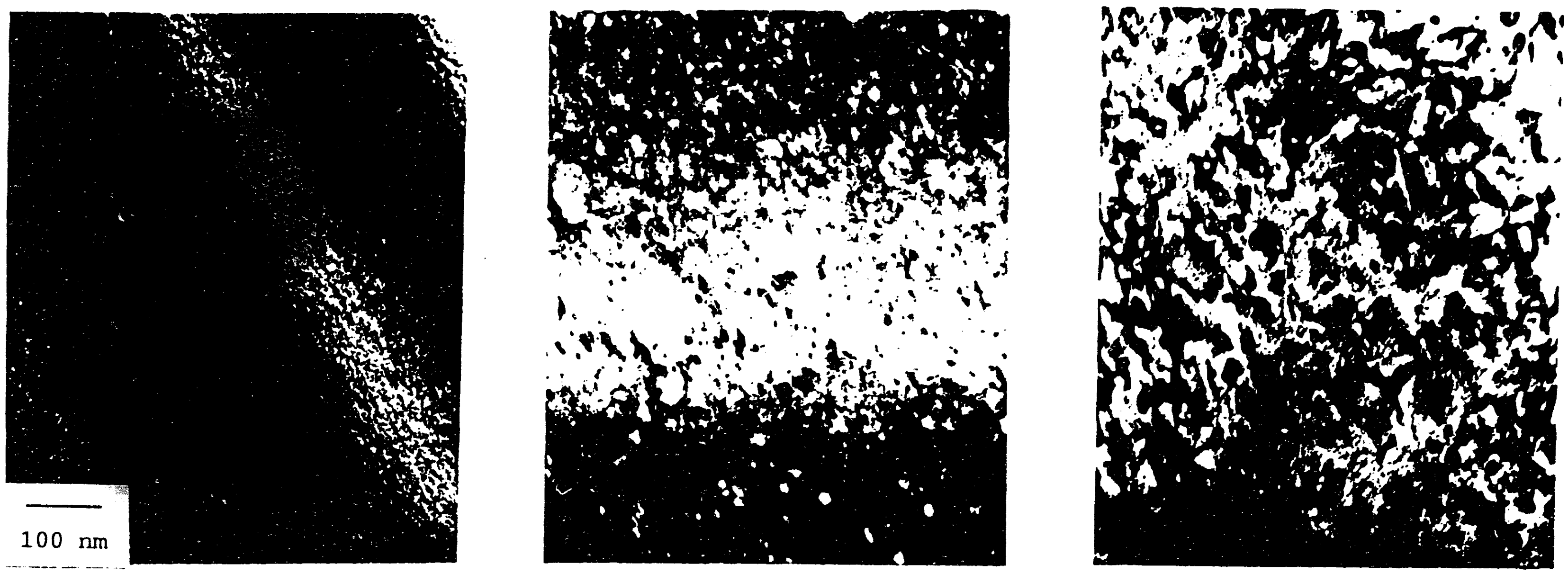
Nickel-ion irradiation has a significant effect on the microstructure of the 304L SS. Figure 2 presents TEM micrographs of the 0, 1, and 5 dpa microstructures, while Table 1 provides a summary of the defect character. The unirradiated microstructure (Figure 2a) has a very low density of randomly distributed perfect dislocations indicating that the alloy has been fully recrystallized.

The 1 dpa sample (Figure 2b) shows a uniform distribution of isolated defects. The defects consist of faulted Frank dislocation loops and black spot damage (small loops and defect clusters). Frank dislocation loops form from the coalescence of vacancies or interstitials on {111} type planes. The Frank loops observed were believed to be interstitial due to higher diffusivity of interstitials at typical irradiation temperatures[14].

The samples irradiated to 5 dpa (Figure 2c) have a significantly more complex microstructure. The microstructure consists of a mixture of larger Frank loops, unfaulted perfect loops, tangles of dislocations and a much lower density of black spot damage. Perfect loops which are glissile form by the growth and unfaulting of Frank loops[13].

TABLE 1--Defect Character

Dose	Defect type	Defect Sizes	Defect Densities
1 dpa	Black spot Frank Loops	< 5 nm to 10 nm	9×10^{15} cm^{-3}
5 dpa	Black spot Frank Loops Perfect Loops Dislocation Tangles	< 5 nm to 30 nm	3×10^{16} cm^{-3}



(a) (b) (c)

FIG. 2--TEM micrographs of the (a) unirradiated (b) irradiated to 1 dpa and (c) irradiated to 5 dpa microstructures

Interactions of the growing loops leads to the formation of the dislocation tangles. The weak beam TEM micrograph shown in Figure 3 of the 5 dpa sample shows more clearly the presence of the faulted Frank loops.

Deformation Microstructures

Surface examination of the samples after tensile deformation indicated that fundamental differences existed in deformation characteristics between the irradiated and unirradiated regions. All of the samples examined in the SEM exhibited steps or ledges on the surface following deformation (Figure 4). Within each sample, the density of ledges varied greatly from grain to grain. However some general trends with strain, dose level, and temperature were observed. A higher density of ledges was seen in the 0 and 1 dpa samples compared to the 5 dpa samples at equivalent strain when deformed at 25°C. The density of ledges in the 0 and 1 dpa samples appeared to saturate at a strain of about 2%, becoming more pronounced in straining up to 10% (Figure 4a). In comparison, the density of steps nearly doubled in the 5 dpa sample in straining from 5% to 10%. Ledges in the 5 dpa sample deformed to 10% at 25°C (Figure 4b) were not as distinct as in the unirradiated case, which may have been a result of lower strain in the irradiated region caused by radiation-induced hardening. Straining to 10% at 288°C (Figure 4c) produced a high density of very faint ledges which are much shallower than in the sample deformed at 25°C.

The highly localized deformation, indicated by the surface steps, was verified using TEM. Representative microstructures from deformed unirradiated and irradiated specimens are illustrated in Figure 5. The unirradiated material deformed to approximately 5% at room temperature shows a highly ordered network of dislocations as indicated by the dark bands of contrast in Figure 5a. At lower strains, the highly planar nature of deformation in this alloy is illustrated by the formation of a large number of dislocation pile-ups on the grain boundaries. Stacking

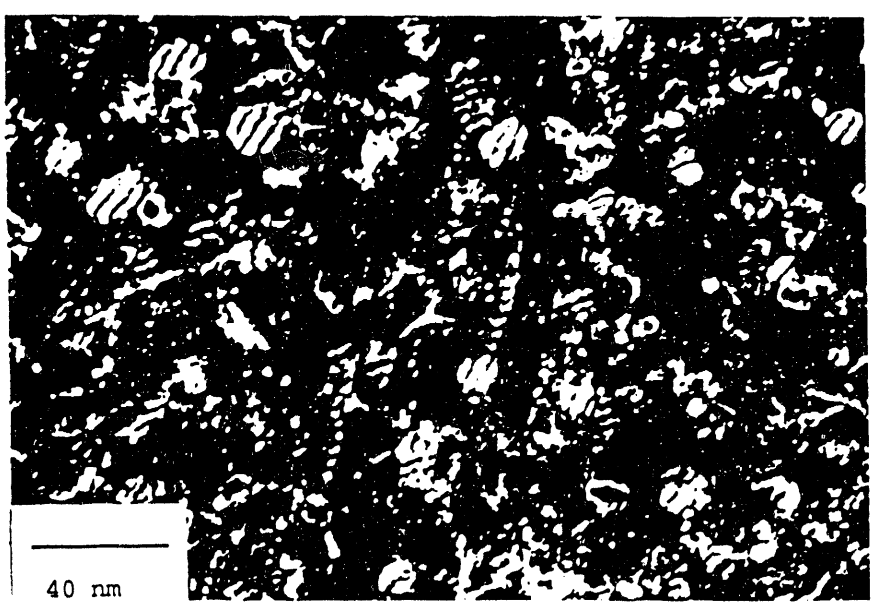
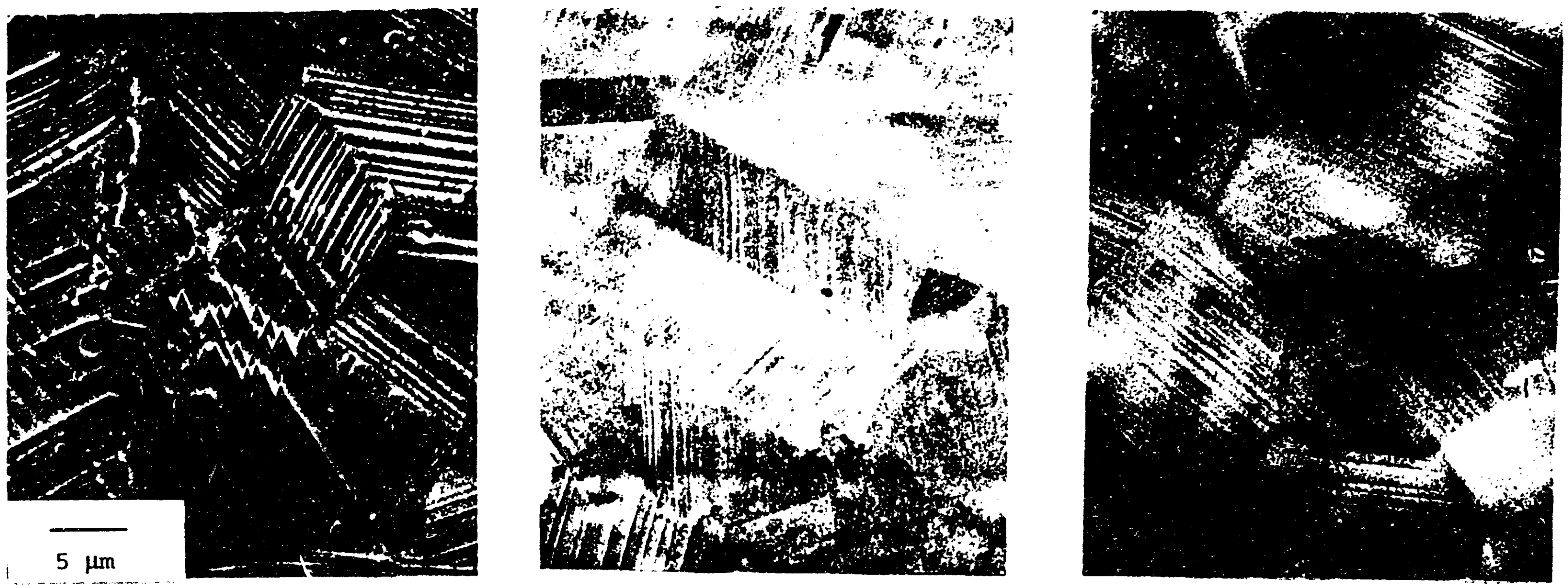
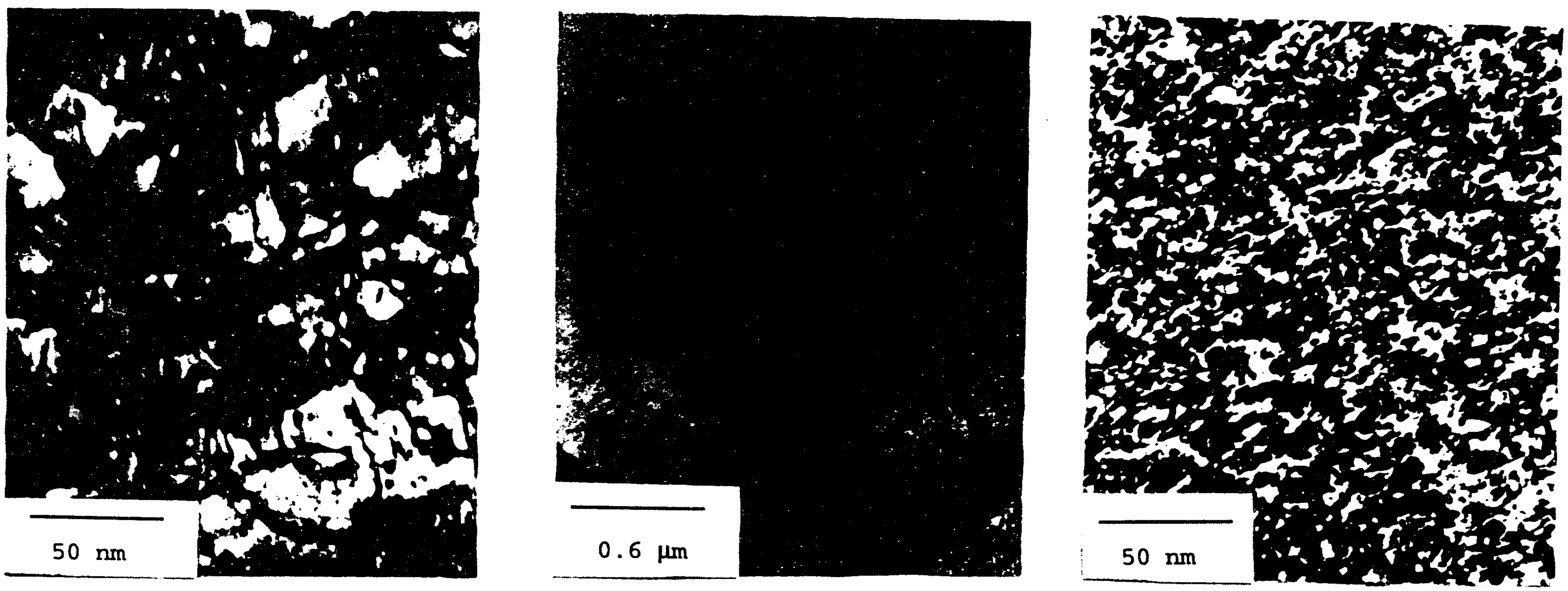


FIG. 3--Weak beam dark field micrograph of 304L SS ion-irradiated to 5 dpa



(a) (b) (c)

Fig. 4--SEM Micrographs of the 304L SS surface: (a) unirradiated and deformed to 5% at 25°C (b) irradiated to 5 dpa and deformed to 5% at 25°C (c) irradiated to 5 dpa and deformed to 10% at 288°C



(a) (b) (c)

FIG. 5--Microstructures of deformed 304L SS samples: (a) unirradiated (b) irradiated deformed at 25°C and (c) irradiated and deformed at 288°C

faults were also present in many regions of the samples with some extending across the width of the grain.

The dramatic influence of the irradiated microstructure on deformation characteristics was seen in the 5 dpa sample. Twinning was observed following room temperature deformation to only 5% strain (Figure 5b). Twin density and width varied from grain to grain with a high density of planar faults present in the 5 dpa. Twin spacing was approximately 0.2μm and the width of the twins was between 50 and 200nm. Upon straining to 10%, most grains showed twinning on the primary slip plane with a few grains also twinning on secondary planes. There is a noticeable decrease in the number of stacking faults with increasing strain, which suggests that the twin nucleation began with the formation of stacking faults. Samples irradiated to 1 dpa and deformed at room temperature showed principally dislocation slip, planar faults, and only a limited amount of twinning. The twins that did develop were extremely narrow (< 20nm) and their spacing was approximately 0.1μm.

Deformation behavior of irradiated 304L SS at 288°C indicates a transition from twinning to one of highly localized slip. This change in deformation is illustrated in Figure 5c, which shows a narrow channel with a reduced density of defects. This channel, although not completely free of defects, appears similar to dislocation channels observed in earlier investigations[7-11]. The channels were very faint and incompletely cleared with a width of approximately 15nm and spacing of 150nm. They were present in many of the grains observed and their spacing was relatively uniform. Twins were not present in samples deformed at the slower strain rate of $2.5 \times 10^{-6} s^{-1}$.

Discussion

Radiation hardening in ion-irradiated 304L SS is promoted by the presence of a high population of dislocation loops and defect clusters introduced by particle bombardment. These defects act as barriers to dislocation glide resulting in increased flow stress. At lower temperatures, the flow stress increase is sufficient to induce deformation twinning shortly following the onset of plastic strain in microstructures containing large overlapping defects. Microstructures containing a lower density of small isolated defects do not contain enough barriers to glide to raise the flow stress above the twin stress, and therefore dislocation slip is the dominant deformation mode. With increasing temperature, the flow stress for glide is reduced so the twin stress is never reached. Thermal activation leads to the formation of channels in the alloy with a reduced density of defects. Deformation is concentrated in these channels with the surrounding hardened microstructure remaining undeformed.

Twin formation in ion-irradiated 304L stainless steel at room temperature is in contrast to the dislocation channels observed in neutron-irradiated copper[7-11] and quenched aluminum[15,16] both of which have higher SFE. However, Suzuki et al.[17] pointed out that for vacancy loops (as in quenched aluminum), annihilation requires the passage of only a single partial dislocation to unfault the loop, while interstitial loops require the passage of two partial dislocations. It was shown that large interstitial loops in proton-irradiated materials were very stable against elimination by the interaction with glide dislocations and large loops were cut by glide dislocations as opposed to being completely eliminated. This suggests that large stable interstitial loops in the ion-irradiated 304L SS may be very effective in blocking dislocation motion and promotes twin nucleation at lower temperature.

Stacking fault energy plays a key role in deformation behavior. The low stacking fault energy of 304 SS, ~18 mJ/m^2[18] prompts much more planar slip behavior than Cu with a SFE of ~70 mJ/m^2[19]. Extended dislocations form in low SFE metals that inhibit cross-slip and produce large dislocation pile-ups on the primary slip planes. At higher strains, when conjugate slip systems become active, the intersection of dislocations leads to rapid hardening. Twinning takes place when the stress for slip becomes greater than the stress to nucleate a twin, above approximately 15% strain in the unirradiated microstructure. High defect densities in irradiated metals may be analogous to a pre-hardened microstructure. Stresses to nucleate twins are reached rather readily due to stress concentrations in heavily defected microstructures. However, the stress to propagate twins is much higher[19] restricting twin thickness. New twins nucleate in preference to significant growth of existing twins as seen in the ion-irradiated 304L SS.

Defect nature is also important in determining the mode of deformation. Venables[20] showed how the interaction of glide dislocations on the primary slip plane with a large prismatic loops on a secondary plane can lead to the formation of a twin. Deformation twinning models involve the formation of a pole dislocation and creation of a faulted layer[21], one reason why low SFE alloys are more prone to twin. The 5 dpa samples in the present investigation have a wider distribution of loop sizes. The larger Frank loops may provide likely sites for twin nucleation. In contrast, the 1 dpa samples have lower defect densities, consisting mainly of small Frank loops and defect clusters. Thus, there are fewer sites for twin nucleation and fewer obstacles to enhance stress concentration.

With increasing temperature, the likelihood of dislocations bypassing obstacles by climb processes is enhanced. However, the formation of what appears to be channels in 304L SS at 288°C when they fail to form at room temperature is still not completely clear. The disappearance of only a portion of the defects within the channels suggests glide dislocations are selectively interacting with the loops present. Large interstitial loops were shown to be cut rather than annihilated by glide dislocations[17]. This may result in only the smaller loops being cleared. Detailed analysis of the loops has not been performed in this study due to the complexity of the microstructure. Loop clearing should also be aided by the ability of dislocations to climb at higher temperatures. Climb should enhance cross-slip of glide dislocations and aid in the annihilation of defects on conjugate slip planes. Similar channeling has been observed in proton irradiated ultra high purity (UHP) 304 SS deformed at 300°C[6], while both twinning and channeling were observed in ion bombarded samples following fatigue[23]. Deformation characteristics in these experiments most closely parallel the conditions of the thin irradiated region examined in the present study.

CONCLUSIONS

The presence of a high density of defects in Ni-ion-irradiated 304L stainless steel leads to a transition in deformation behavior with increasing dose and temperature. Dislocation glide is the primary mode of deformation at room temperature in the unirradited condition, and at low radiation dose. With increasing dose at 25°C, deformation twinning becomes the dominant mode of strain accommodation, while at 288°C highly localized slip occurs along paths with lower defect density. These transitions appear to correlate principally with the lower SFE of 304L SS and defect nature.

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