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ABSTRACT

Investigations of helium effects in metals at the Savannah River Laboratory have been carried out by introducing helium by radioactive decay of tritium. This process does not create concurrent radiation damage, such as accompanies ion implantation and (n,α) reactions. The process has its own peculiarities, however, which partially mask and interact with the helium effect of interest. The distribution and local concentration of helium and tritium, which are responsible for changes in mechanical properties and fracture mode, are controlled by the large difference in solubility and diffusivity between the two atoms and by their differing interaction energies with lattice defects, impurities, and internal boundaries. Furthermore, in all investigations with helium generated from tritium decay, some tritium and deuterium are always present. Consequently, property changes include tritium-helium

interaction effects to some extent. Results of investigations with several austenitic stainless steels, Armco iron, and niobium single crystals illustrate the variety of phenomena and some of the complex interactions that can be encountered.

INTRODUCTION

Structural components in fission and fusion reactors may be degraded because of helium accumulation in the metals as a result of tritium decay, ion implantation, or (n,α) reactions. Consequently, these processes can limit the useful lifetimes of reactor components, affect reactor operation, or preclude some reactor repair techniques. Control and mitigation of helium damage, therefore, could have a significant impact on reactor operation and costs.

The investigations of tritium-helium effects on mechanical properties of metals and alloys at the Savannah River Laboratory contribute to an understanding of helium damage and solution of these problems. Tensile and fracture mechanics specimens have been exposed to tritium under various pressure and temperature conditions. By testing after aging for up to ten years, the changes in properties may be related to helium concentration. The next section discusses the effects of helium on the tensile properties, fracture resistance, and crack growth properties of several types of austenitic stainless steels. Later sections discuss the helium effects on Armco iron and niobium.

AUSTENITIC STAINLESS STEELS

Helium effects in austenitic stainless steels have been studied extensively over the past decade.[1-7] In all instances, there was residual tritium present, as well as a small deuterium concentration, both of which also affect the mechanical properties. Isolation of the helium effect by outgassing to remove tritium and deuterium is only partially effective in stainless steels because diffusivity of the hydrogen isotopes is too low (10^{-10} to 10^{-12} cm^2/sec) to allow complete outgasing at acceptable temperatures (300 to 400 K). Annealing of dislocation substructures and rearrangement of helium can occur and alter machanical properties if the temperature is high enough ($\geq$670 K) to outgas all the tritium in a reasonably short time (six months for example). Consequently, all studies of helium effects on the properties of stainless steel require careful interpretation and comparison with both control specimens and specimens containing stable hydrogen isotopes.

Tensile Properties

The earliest studies with Type 304L and 309S austenitic stainless steels, were conducted with thin (0.25-mm thick) specimens that contained either 30 appm (atomic parts per million) helium and 1700 appm of hydrogen isotopes or 100 appm helium and 870 appm of hydrogen.[1,2] Small reductions in ductility were observed in both alloys.[1,2] In contrast, yield strengths increased by 35 MPa for Type 309S and by 75 MPa for Type 304L steel, with 30 appm helium. When the helium content was 100 appm, the increases in yield strength were 118 MPa for Type 309S and 106 MPa for Type 304L steel. An anneal of one-half hour at 973 K caused recovery of the yield strength, but ductility was reduced to near zero, and the fracture mode was 100% intergranular.

In a second study, 3.5-mm diameter specimens of high-energy-rate-forged (HERF) Nitronic®-40 and Type 304L stainless steel were exposed to tritium and then stored for 5.5 years[3-5] (Tables I and II). This exposure procedure produced a 1000-μm deep peripheral zone of helium and tritium with a peak helium concentration of ~2300 appm in the Nitronic®-40.[5] Tensile ductility of the Nitronic®-40 was reduced to near zero (Table II). The same treatment produced only a moderate ductility loss in Type 304L steel. The peak helium concentration was 1200 appm in this case. An additional 5-years' storage of a second Type 304L specimen caused a further increase in yield strength (Table I), and the ductility of the Type 304L steel appeared to have recovered. This result should be viewed cautiously, however, as only one specimen was tested at each aging time.

These tensile specimens do not always show an increase in yield strength after exposure to hydrogen or tritium because only a relatively narrow peripheral area has been affected by the hydrogen. In contrast, the sheet specimens discussed above had a uniform hydrogen concentration across the entire cross section.

TABLE I

Mechanical Properties of HERF Type 304L Stainless Steel

Exposure Conditions	Strength, MPa Yield	Strength, MPa Ultimate	Total Elongation, %	Fracture Strain
NONE	630	790	32	1.83
30 MPa H_2 at 470 K, 56 days	600	840	35	0.87
48 MPa T_2 at 345 K, 17 months and aged 5.5 years at 250 K	620	745	33	0.73
48 MPa T_2 at 345 K, 17 months and aged 11.5 years at 250 K	660	765	32	1.47

TABLE II

Mechanical Properties of HERF Nitronic®-40 Stainless Steel

Exposure Conditions	Strength, MPa Yield	Strength, MPa Ultimate	Total Elongation, %	Fracture Strain
NONE (3)	800	930	29	1.20
30 MPa H_2 at 470 K, 56 days and aged 7 years at 250 K	860	990	28	0.76
69 MPa T_2 at 470 K, 60 days	830	940	32	0.73
48 MPa T_2 at 345 K, 17 months and aged 5.5 years at 250 K	870	925	6	0.10

Tensile specimens of Nitronic®-50 and weld metal of Type 304L stainless steel reveal similar changes in properties due to helium.[6] The concentration of helium was calculated to be ~670 appm at the surface and fall to zero within the first 1000 μm.

Transmission electron microscopy of sheet specimens of Type 304L and 309S stainless steel [1,2] and of the HERF Type 304L and Nitronic®-40 steels[3] revealed defects with strain contrast of around 5 nm diameter. These may be interpreted as helium bubbles. Their size and number density are consistent with the estimated helium content in the several specimens. The defects are found throughout the microstructure, not just on internal boundaries. Where heated to 973 K for 30 minutes, larger helium bubbles (50 nm diameter) are found which lie predominantly on boundaries or dislocation networks.[1,2]

Taken together, the mechanical property changes and microscopy show an unmistakeable helium effect on tensile properties of austenitic stainless steels. Yield strength is increased and ductility is reduced. Differences in specimen shape and exposure conditions among the several investigations preclude a quantitative comparison of the data.

Fracture Resistance

Fracture resistance was evaluated by the J-integral method in a series of five austenitic stainless steels.[7,8] The steels were Type 304L, 316, Nitronic®-40, A-286, and JBK-75. All were HERF except the JBK-75 which had been annealed. Specimens were exposed to tritium at 61 MPa pressure at 423 K for six months.

Calculated tritium concentrations were 1900 appm at the surface and 420 appm at the center based on the exposure conditions and the solubility relation derived by Louthan and Derrick from permeation measurements on several stainless steels.[9] There are, however, significant differences in hydrogen (tritium) solubility among stainless steels exposed to hydrogen under identical conditions.[10,11] Tritium autoradiography on cross sections of specimens of each of the steels in the current study shows that the relative tritium concentrations differed and that the ratios of tritium contents relative to HERF Type 304L agreed reasonably well with the ratios of hydrogen contents measured in the solubility studies. Tritium and helium contents, averaged over the plane at the notch root for aging times up to 30 months, were based on the Louthan-Derrick relation and the decay constant for tritium of 1.801×10^{-9} sec^{-1}. Data points in Figure 1 have been adjusted for each alloy on the basis of the autoradiography. In Figure 1 the tritium solubility of Type 304L is assumed equal to the Louthan-Derrick value.

J-integral tests were made on specimens of each steel immediatly after exposure and following storage at 273 K for 15 and 30 months. Control specimens were exposed to air at 423 K for six months and stored at the same low temperature as the exposed specimens. Tensile tests were made in air at room temperature and the load-deflection curves were analyzed by the J-integral technique.

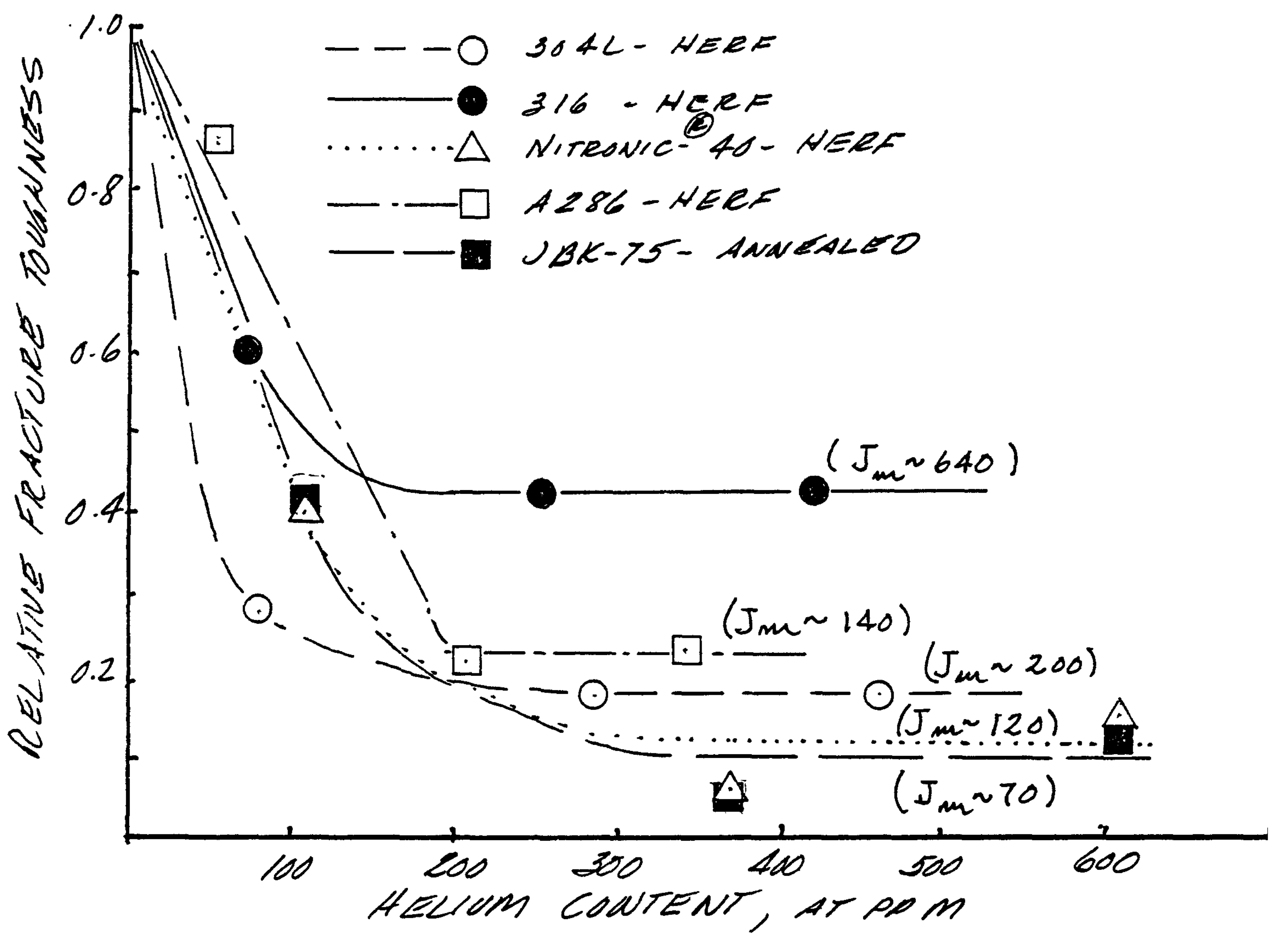

FIGURE 1. Reduced Fracture Toughness Due to Helium

Crack initiation was assumed to begin at maximum load (J_m) in all cases, an assumption that has been verified in all five alloys by observation of a burst of tritium coincident with maximum load.

The data demonstrate that the combined helium-tritium has degraded the fracture resistance of all five steels. Fracture toughness decreases to less than fifty percent of the value for the control specimens during the first 15 months aging. Further increases in helium content and concurrent decreases in tritium content had little effect on J_m. HERF Type 316 stainless steel was more resistant to helium damage than the other steels in terms of both relative J_m (0.43) and absolute value of J_m (640 kJ/m^2). The JBK-75 and HERF Nitronic®-40 appear to have been degraded more severely than the other steels. Our earlier study had shown a larger helium effect in the HERF Nitronic®-40 than in HERF Type 304L steel that was tentatively attributed to the larger tritium and helium contents in the HERF Nitronic®-40.[2] The present results suggest, however, that there is a difference in the severity of tritium-helium damage in these steels which is dependent on aging time and is a complex function of the separate tritium and helium damage mechanisms and interaction between them. The difference in severity of helium damage between HERF A-286 and annealed JBK-75 was probably associated with microstructural differences arising from thermomechanical or thermal treatment.

Fracture modes were altered by tritium charging and aging the specimens. In all cases, except HERF Type 316 stainless steel, varying amounts of intergranular separation accompanied the tritium-helium damage. Annealed JBK-75 fractured by intergranular separation in all conditions except for the control specimens. The fraction of intergranular separation increased with aging time in HERF Nitronic®-40 and HERF A-286. Fracture of HERF Type 304L stainless steel was transgranular along austenite-martensite and twin boundaries with some secondary cracking and intergranular separation. Fracture of HERF Type 316 stainless steel was by microvoid coalescence under all conditions with some evidence of transgranular fracture as in HERF Type 304L steel.

The relative contributions of tritium and helium to the total damage were estimated by comparing results from tritium-charged, aged and outgassed specimens with results from tritium-charged and aged specimens. A specimen of each alloy that had been tritium charged and aged 20 months was partially outgassed by heating in air at 423 K for four and a half months. At this temperature, helium does not diffuse over distances of more than 10 to 100 nm because helium is easily trapped by lattice defects,[12] whereas tritium was partially removed from the specimens. Recovery of J_m was greatest in HERF A-286, HERF Type 316 and JBK-75 and was least in HERF Nitronic®-40, Figure 2.

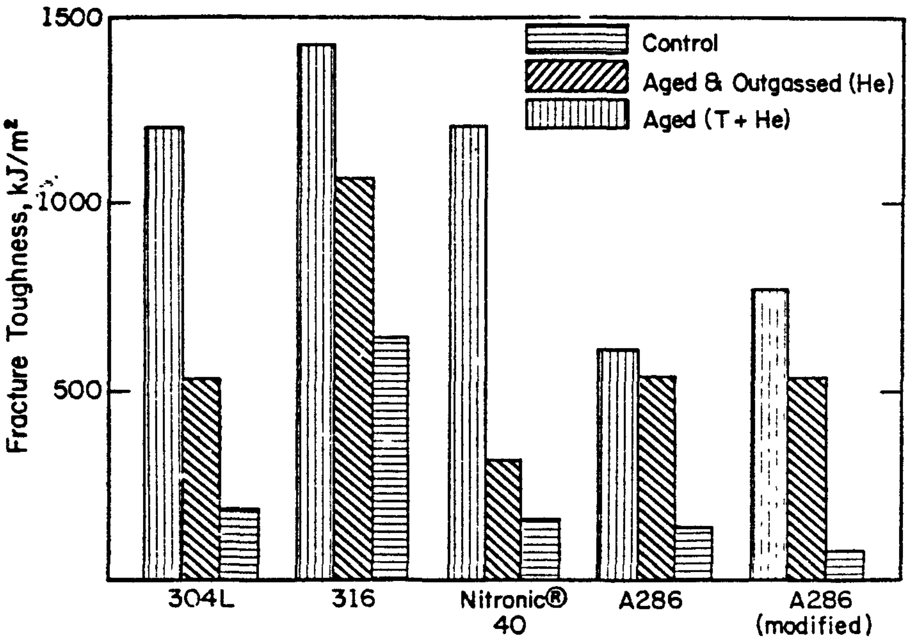

FIGURE 2. Effect of Tritium Outgassing on Fracture Toughness of Stainless Steels

Fracture mode reverted to microvoid coalescence following outgassing to remove tritium in HERF A-286 and annealed JBK-75. Fracture modes tended to remain unchanged in the other three alloys: intergranular separation in HERF Nitronic®-40, microvoid coalescence in HERF Type 316, and transgranular fracture in HERF Type 304L steel. These observations are consistent with recovery of J_m noted above for HERF A-286 and annealed JBK-75.

Sustained Load Crack Growth

Sustained-load cracking tests were made to measure the effect of increasing helium content on the stress intensity necessary to initiate and propagate a crack in samples which had been in storage 273 K for 22 months.

A crack initiated in the first two HERF Nitronic®-40 specimens immediately upon loading to 3600 N or about 25 percent less than the maximum load sustained by HERF Nitronic®-40 samples in the 15-month tensile tests. Without a further increase in load, the crack propagated to the back edge of the specimen in less than a second. The remaining HERF Nitronic®-40 sample was loaded to 3310 N without immediate crack initiation. Examination of the stressed sample one hour later revealed that a crack had initiated and propagated in the same manner as before.

Crack initiation and growth were also noted in two HERF A-286 specimens at loads of 3300 N and 3000 N. The cracks were observed about two hours after loading, but unlike cracks in the HERF Nitronic®-40 specimen, had not propagated completely across the width of the samples. Neither HERF Type 304L nor HERF Type 316 samples have cracked after 33 months under sustained load.

The samples used in this study were not designed for sustained-load cracking tests, and do not satisfy ASTM testing requirements.[13] Therefore, the stress intensity required to initiate and propagate cracks in these samples is not a valid threshold stress intensity. The results do, however provide relative values that can be compared to the stress intensities necessary to initiate and propagate cracks in companion specimens aged for various times (i.e., as a function of helium content), as seen in Figure 3.

For example, HERF Nitronic®-40 samples sustained a load of about 4450 N in the 15-month tensile test before cracking catastrophically, in exactly the same manner as samples placed under sustained load after 22 months. However, the latter samples could sustain only 3338 N before cracking. A 33 percent increase in average helium concentration (225 vs 169 at ppm He) resulted in a 25 percent decrease in the load requried for crack initiation. The fracture mode of the HERF Nitronic®-40 under sustained load was intergranular, the same as in the 15- and 30-month tensile tests. In contrast, the fracture mode of HERF A-286 was microvoid coalescence under sustained load but intergranular in the tensile tests.

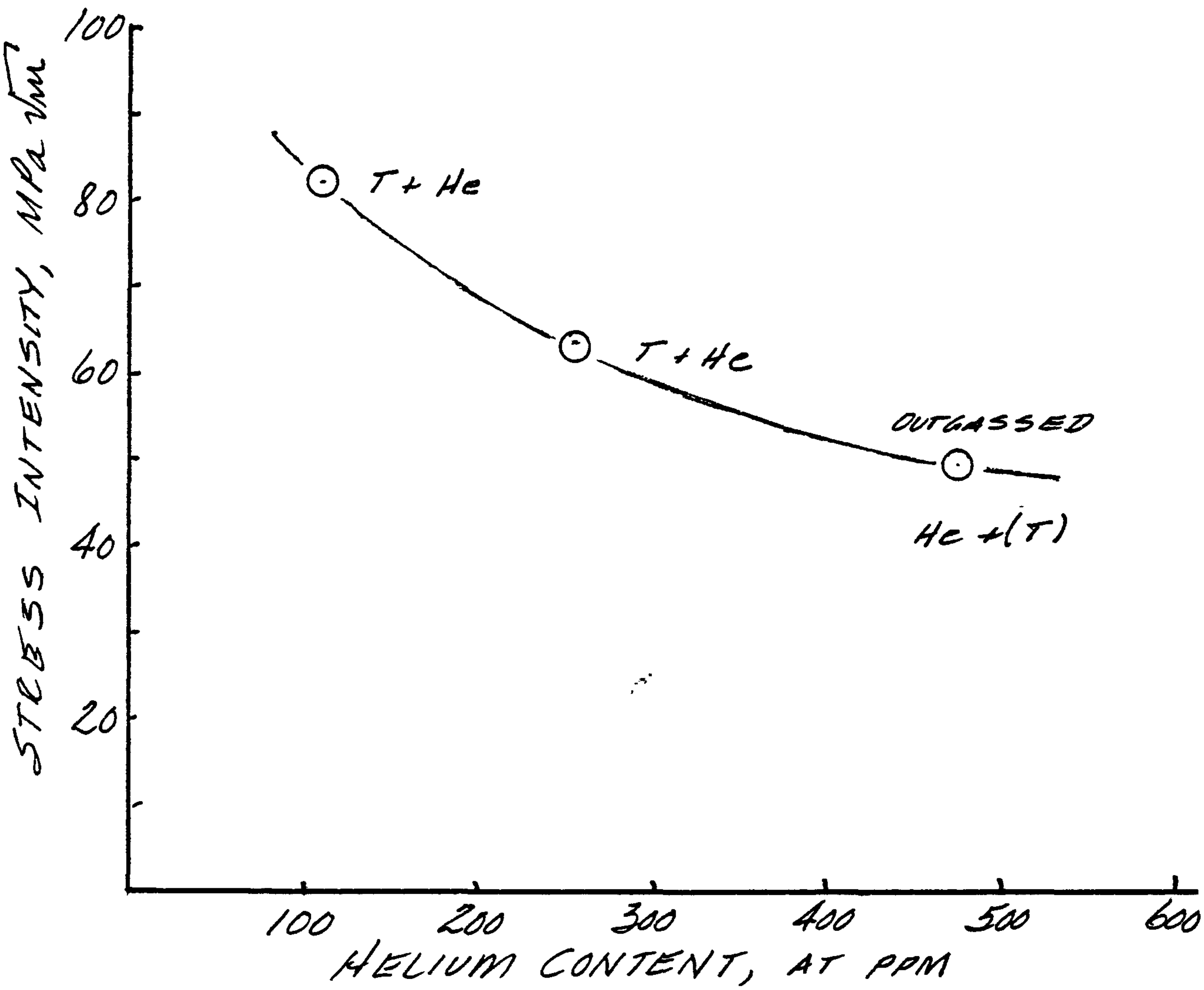

FIGURE 3. Stress Intensity For Crack Initiation in HERF Nitronic®-40

ARMCO IRON

Reduced yield and tensile strengths were observed in tensile specimens of Armco iron at all test temperatures between 295 and 1000 K.[14] The specimens contained about 5 appm helium and a small but undetermined quantity of residual tritium that remained after offgassing at 373 K. Ductility was reduced also, except at 400 K which is in the "Blue Brittle" range where specimens with helium (and tritium) were more ductile than control specimens. These reduced strengths were attributed to weakened dislocation interaction with carbon and nitrogen because of the stronger binding of helium and tritium with carbon and nitrogen.

NIOBIUM

A simpler situation was studied with single crystals of pure niobium.[15] Tritium exposure and outgassing at 673 K yielded helium contents of 340 to 1675 appm plus residual tritium. Yield strength increases were seen at test temperatures of 100 to 500 K which were due to helium clusters and punched out dislocation loops. Helium bubbles developed during annealing at temperatures over 100 K and strengthened the niobium also. In both regions, dislocation shearing of the clusters or bubbles could account for the observed strengthening. In this case, the helium effect is free from competing interactions that affect strengthening also.

SUMMARY AND CONCLUSIONS

Helium damage arising from decay of dissolved tritium has been observed in several varieties of austenitic stainless steel and in Armco iron and pure niobium. The strengthening and losses in fracture resistance and ductility are associated with defects of about 50 nm size for temperatures below 500 K. Each defect is presumed to contain helium atoms and vacancies. The size and numbers of defects can account for the helium present. At temperatures of approximately 1000 K, helium agglomerates into distinct bubbles in both niobium and stainless steel. Bubbles lie on internal boundaries or dislocation networks. In stainless steel, this condition promotes fractures on internal interfaces, weakened by the presence of the helium bubbles.

Loss of fracture resistance at room temperature appears to reach a lower bound and is not reduced further by additional helium beyond 300-400 appm. This behavior is consistent with a hardening model where the "precipitate" is sheared by advancing dislocations and where there are increasing numbers of "precipitates" of uniform size. The magnitude of the yield strength increase in austenitic steels and niobium supports this model also.[1,15]

Helium accumulation within reactor components by any of the three mechanisms may have a negative impact on the useful life of the structure or its repair. Reduced fracture resistance at high helium levels must be taken into account in assessing safety margins in structures, for example. Repair or replacement procedures based on welding may encounter difficulties, as seen for example, in the almost total loss of ductility following heating to 1000 K with 100 appm helium present. Substantially more data under other experimental conditions are needed to evaluate the seriousness of these issues and to define the helium concentrations which limit repair procedures.

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