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HELIUM DAMAGE IN AUSTENITIC STAINLESS STEELS*

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ABSTRACT

Helium produced by tritium decay was first shown to embrittle austenitic stainless steel at ambient temperature in tensile specimens of Nitronic®-40 steel (Armco, Inc.). A long-term study was initiated to study this form of helium damage in five austenitic alloys. Results from this study have been analyzed by the J-integral technique and show a decrease in ductile fracture toughness with increasing He-3 concentration. Sustained-load cracking tests indicate that the stress intensity required to initiate and propagate a crack also decreases with increasing He-3 concentration.

INTRODUCTION

Helium generated from radioactive decay of tritium has been shown to cause significant changes in the deformation and fracture of austenitic stainless steels at room temperature (1-2). Following our first observation of severe helium embrittlement in HERF Nitronic®-40 (Armco Steel Co),[1] a second investigation was undertaken which included HERF Type 304L, 316, Nitronic®-40, A-286 and annealed A-286 (modified) stainless steels. In every alloy, fracture toughness as evaluated by the J-integrated technique was reduced for samples tested after tritium exposure.[2] Additional mechanical tests have been completed on companion specimens stored an additional 15 months to generate higher helium concentrations. These latter tests demonstrate that the fracture toughness of the five stainless steels was not lowered further by the additional 15 months' aging, suggesting that combined tritium-helium damage is a complex function of tritium-helium concentrations and distribution. The new tests also

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show that the stress intensity for sustained-load cracking is reduced in two of the steels [Nitronic®-40 and A-286], and that significant recovery of toughness takes place in these five steels when residual tritium is removed by outgassing.

Experimental Procedure

Notched C-shaped specimens 3.8 mm thick and 25 mm outer radius[3] were machined from five stainless steels: Type 304L, Type 316, Nitronic®-40, A-286 and a modified A-286 (Table 1). The modified A-286 was in the annealed condition, all others were high-energy rate-forged (HERF). Specimens were not fatigue precracked. The specimens were exposed to tritium at 61 MPa pressure at 423 K for six months.

Calculated tritium concentrations were 270 mol T_2/m^3 at the surface and 60 mol T_2/m^3 at the center based on the exposure conditions and the solubility relation derived by Louthan and Derrick from permeation measurements on several stainless steels.[4] There are, however, significant differences in hydrogen (tritium) solubility among stainless steels exposed to hydrogen under identical conditions.[5 6] 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 earlier work.[5 6] Tritium concentration ratios from the autoradiography were 0.89 for Type 316, 0.73 for A-286 and 1.3 for Nitronic®-40 and the modified A-286. Tritium and helium contents averaged over the plane at the notch root are plotted in Figure 1 for aging times up to 30 months. These values are based on the Louthan-Derrick relation and the decay constant for tritium of 1.801×10^{-9} sec^{-1}. Data points in Figure 2 have been adjusted for each alloy on the basis of the autoradiography taking solubility in Type 304L equal to the Louthan-Derrick value.

Fracture Toughness

Tensile tests were made on specimens of each steel immediately after exposure and following 15 and 30 months' storage at 273 K. 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.[3] Crack initiation was assumed to begin at maximum load in all cases — an assumption that has been verified in all five alloys by observation of a burst of tritium coincident with maximum load. Reported J-integral values have been corrected for combined bending and tensile loading.[7] J-integral at maximum load (J_m) and deflection at maximum load are

given in Table 2. Note that in our earlier report of the initial stage of this investigation,[2] the data for Type 316 and A-286 steels were reversed due to improper labelling of the as-received materials.

The data demonstrate that the combined helium-tritium has degraded the mechanical properties 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 decrease in tritium content had little effect on J_m, as seen in Figure 2. 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 (640kJ/m^2). The modified A-286 and HERF Nitronic®-40 appear to have been degraded more severly 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 A-286 (modified) was probably associated with microstructural differences arising from thermomechanical or thermal treatment. Although differences in composition between the two varieties of A-286 were small, the difference may contribute also to the relative damage by small changes in tritium solubility or distribution.

In contrast to behavior of J_m with aging, deflection at maximum load continued to decrease as aging time was extended from 15 to 30 months (Table 2). This means that maximum load increased since J_m increased slightly in each alloy.

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 A-286 (modified) fractured by intergranular separation in all conditions except for the control specimens (Figure 3a). The fraction of intergranular separation increased with aging time in HERF Nitronic®-40 (Figure 4a) and HERF A-286 (Figure 5a). Fracture of Type 304L stainless steel was transgranular along austenite-martensite and twin boundaries with some secondary cracking and intergranular separation (Figure 6a). Fracture of Type 316 stainless steel was by microvoid coalescence under all conditions with some evidence of transgranular fracture as in Type 304L steel (Figure 7a).

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 specimen. A specimen of each alloy that had been tritium charged and

aged 20 months was 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,[8] whereas tritium was removed from the specimens. As seen in Figure 8, J_m recovered in all five steels. Recovery of J_m was greatest in HERF A-286, HERF Type 316 and annealed A-286 (modified) and was least in Nitronic®-40.

Fracture modes were changed to microvoid coalescence by outgassing to remove tritium in HERF A-286 (Figure 5b) and annealed A-286 (modified) (Figure 3b). Fracture modes tended to remain unchanged in the other three alloys: intergranular separation in Nitronic®-40 (Figure 4b), microvoid coalescence in Type 316 (Figure 7b) and transgranular fracture in Type 304L steel (Figure 6b). These observations are consistent with recovery of J_m noted above for the two varieties of A-286.

Sustained-Load Cracking

Although ductile fracture toughness measurements are useful for evaluating an alloy's susceptibility to hydrogen damage, a design parameter based on J-integral results has not been established. Critical design data are usually determined from sustained-load cracking tests that provide the threshold stress intensity for slow crack growth. Because of the importance of threshold data, an attempt was made to measure the effect of increasing helium content on the stress intensity necessary to initiate and propagate a crack in the C-shape samples. The samples had been in storage at 273 K for 22 months.

Eight C-shaped specimens (2 each of the four HERF alloys) were placed under sustained load with the fixture shown in Figure 9. Samples of each alloy were loaded to a level approximately 20 to 30 percent less than the maximum load that companion samples had sustained in the 15-month tensile test. These maximum loads were converted to equivalent stress intensities at the notch and are given in Table 3 along with the stress intensities chosen for sustained loading.[9]

A crack initiated in the first of two Nitronic®-40 specimen immediately upon loading to a stress intensity of 66 MPa $\sqrt{m}$, about 25 percent less than the maximum load sustained by 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 Nitronic®-40 sample was loaded to a stress intensity of 60 MPa $\sqrt{m}$, 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 both A-286 specimens at stress intensities of 60 and 55 MPa $\sqrt{m}$. The cracks were observed about two hours after loading, but unlike cracks in the Nitronic®-40 specimen, had not propagated completely across the width of the samples. Neither Type 304L nor Type 316 samples have cracked after nine months under sustained load.

The C-shaped samples used in this study were not designed for sustained-load cracking tests, and do not satisfy ASTM testing requirements.[10] 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).

For example, Nitronic®-40 samples sustained a load of about 4450 N (81 MPa $\sqrt{m}$) 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 (60 MPa $\sqrt{m}$) before cracking. A 33 percent increase in average helium concentration (32 vs. 24 moles $^3He/m^3$) resulted in a 25 percent decrease in the stress intensity required for crack initiation. The fracture mode of the 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.

The results for A-286 were similar with the samples aged 22 months displaying a stress intensity for crack initiation 20 percent less than the corresponding stress intensity level at 15 months. Since crack propagation ceased in these samples well before back edge effects became significant, it was possible to calculate a stress intensity at crack arrest (in the same manner that threshold stress intensities are determined from standard fracture mechanics samples). The stress intensity at crack arrest ranged from 35 to 38 MPa $\sqrt{m}$, significantly less than the 55 MPa $\sqrt{m}$ required to initiate crack growth.

SUMMARY AND CONCLUSIONS

This investigation demonstrates that the fracture toughness (evaluated by the J-integral technique on C-shaped specimens) is reduced by the combined presence of tritium and helium in five stainless steels. There appears to be a maximum or saturation level for combined tritium-helium damage that is dependent on the alloy and its thermomechanical treatment. The relative as well as absolute amounts of tritium and helium damage vary among the alloys and are least in HERF Type 316 stainless steel and greatest in HERF Nitronic®-40.

These results show that both crack initiation and propagation occur at lower stress intensity levels in the presence of helium. The stress intensity for crack propagation is further lowered by increasing helium concentration. Samples of both Nitronic®-40 and A-286 are being baked out at 423 K so that sustained-load tests may be repeated with specimen that contain only the helium decay product.

There were large differences in the extent of recovery of fracture toughness among the alloys following outgassing to remove tritium. These differences suggest that tritium-helium damage is a complex function of microstructure and may depend on interaction between tritium and helium in the alloys.

Currently, detailed transmission electron microscopic examination of the specimens is under way to identify structural changes characteristic of helium and combined tritium-helium damage. Completion of this microstructural examination is necessary to determine the mechanism of tritium-helium damage.

ACKNOWLEDGEMENT

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TABLE 1

Nominal Alloy Compositions (Weight Percent)

Alloy	C*	Mn*	Si*	Cr	Ni	Mo	N	Other
304L	0.03	2.00	1.00	19	10	-	-	-
316	0.08	2.00	1.00	17	12	25	-	-
Nitronic®-40	0.08	9.00	1.00	21	6	-	0.3	-
A286	0.08	1.35	0.70	15	26	1.25	-	**
A286 (modified)	0.03	0.20	0.10	15	30	1.25	-	***

* Maximum
** 2.15 Ti, 0.30 V, 0.20 Al, and 0.003 B.
*** 2.15 Ti, 0.25 V, 0.25 Al, and 0.002 B.

TABLE 2

Tritium and Helium Effects on Fracture Toughness

Alloy	History	Deflection at Max. Load, mm	Fracture Toughness Jm, kJ/m^2
304L	C	3.71	1280
HERF	T	1.55	360
	C + A15	3.33	1120
	T + A15	1.32	190
	C + A30	3.43	1230
	T + A30	0.97	200
316	C	3.51**	1500**
HERF	T	2.39	890
	C + A15	2.97	950
	T + A15	2.03	630
	C + A30	3.15	1360
	T + A30	1.70	650
Nitronic®-40	C	2.67	1200
HERF	T	1.50	480
	C + A15	2.62	960
	T + A15	1.02	75
	C + A30	3.00	1470
	T + A30	0.84	180
A286	C	1.95**	610**
HERF	T	1.80	530
	C + A15	1.70	750
	T + A15	1.14	130
	C + A30	1.35	380
	T + A30	0.81	140
A286	C	2.86	840
(Modified)	T	1.50	350
Annealed	C + A15	2.79**	680**
	T + A15	0.84**	40**
	C + A30	2.74**	870**
	T + A30	0.66**	100**

* C - Control
C + A15 - Control + Aged 15 months.
C + A30 - Control + Aged 30 months.
T - Tritium charged.
T + A15 - Tritium charged + Aged 15 months.
T + A30 - Tritium charged + Aged 30 months.
** Single specimen. All others in duplicate.

TABLE 3

Maximum Loads and Equivalent Stress Intensities Sustained in 15-Month Test. Loads and Stress Intensities for Sustained-Load Study After 22-Months' Storage.

	15-Month Test		22-Month Test	
	Load(N)	Stress Intensity (MPa√m)	Load(N)	Stress Intensity (MPa√m)
A-286				
#1	3740	68	3310	60*
#2	3420	63	3000	55*
Nitronic®-40				
#1	4580	84	3600	66**
#2	A310	79	3310	60*
304L				
#1	3310	60	2710	50†
#2	3250	59	3000	55†
316				
#1	4270	78	3600	66†
#2	5250	96	3960	71†

* Crack growth observed within one to two hours after loading.
** Crack growth observed immediately upon load.
† No crack propagation.

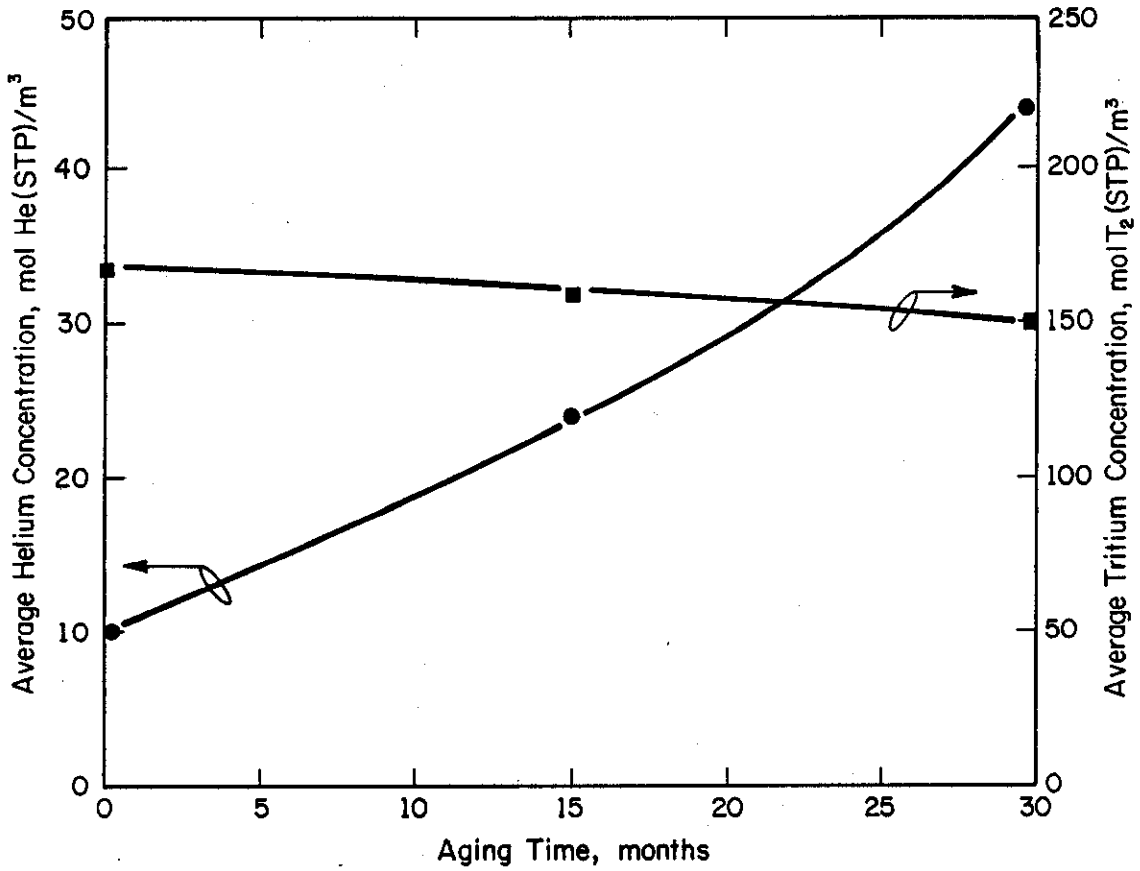


FIGURE 1. Average Helium and Tritium Concentrations in 304L Stainless Steel After Tritium Charging and Aging

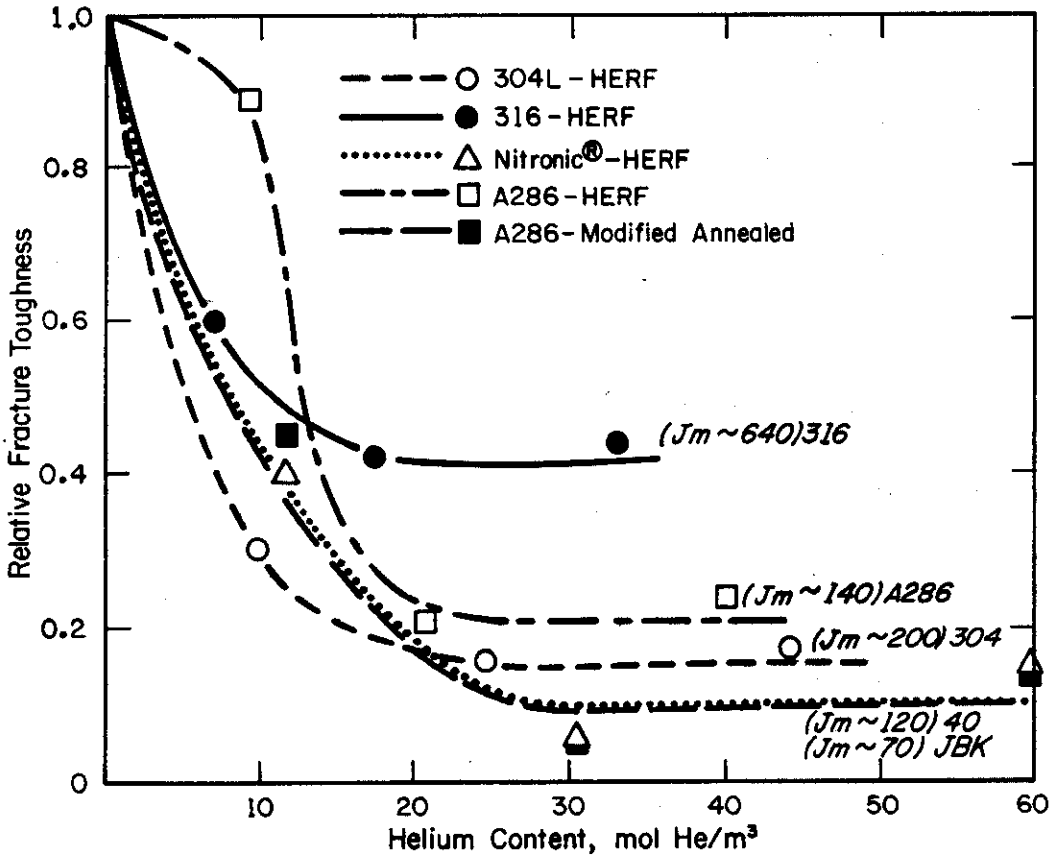
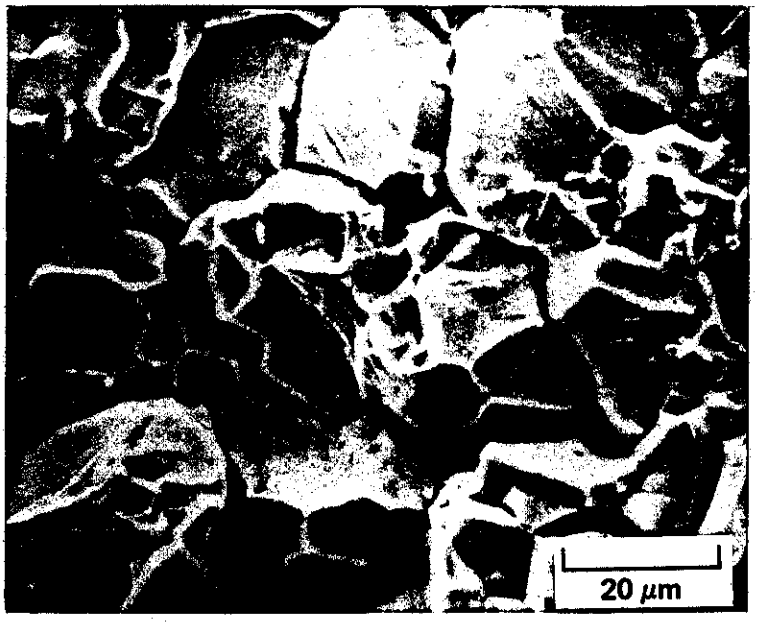
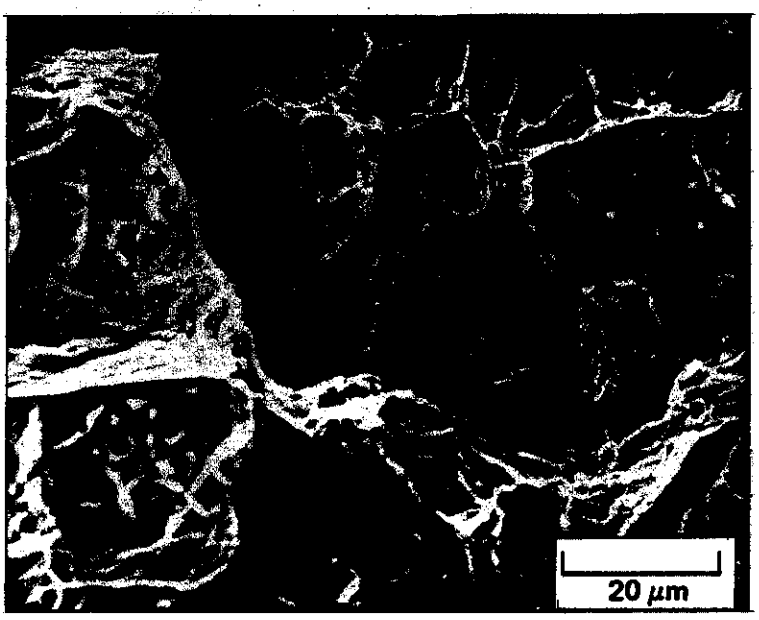


FIGURE 2. Reduced Fracture Toughness Due to Helium Content

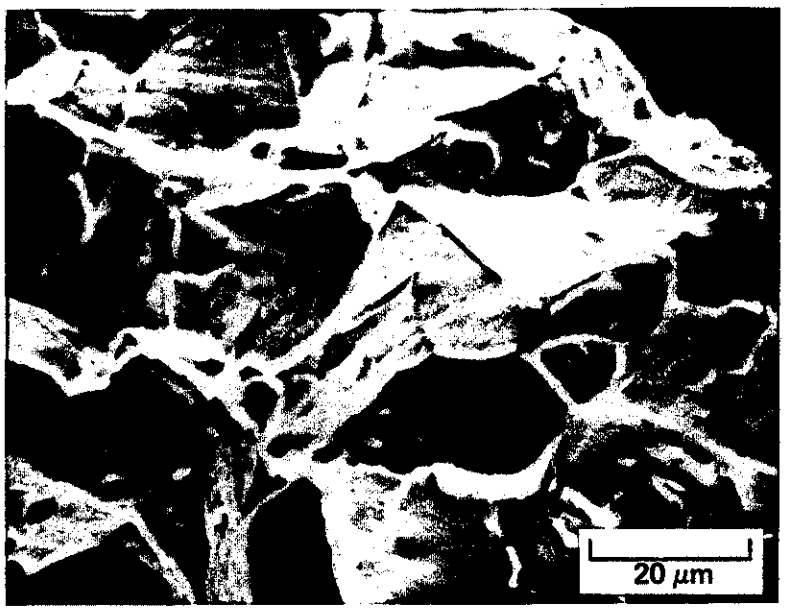


a) Tritium Charged and Aged 30 Months

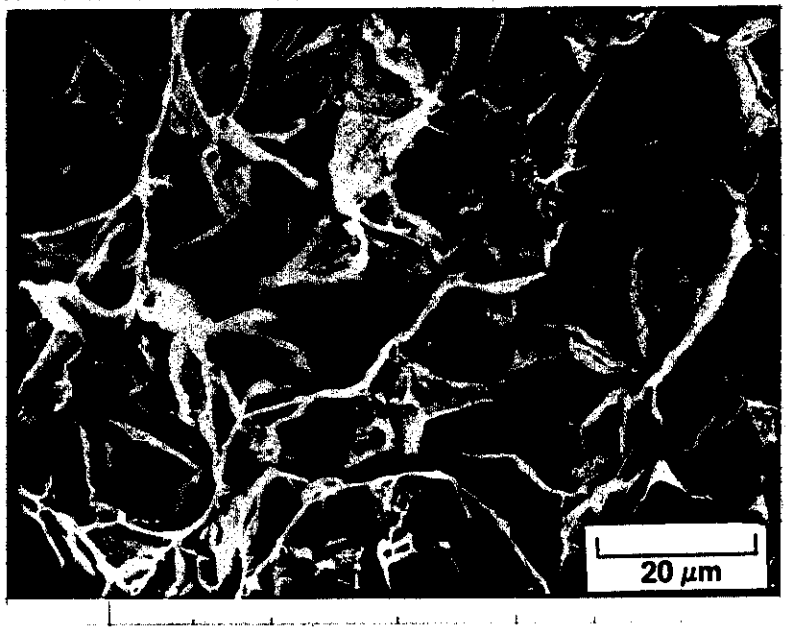


b) Tritium Charged, Aged and Outgassed

FIGURE 3. Fracture Modes in Annealed A-286 (Modified) Stainless Steel

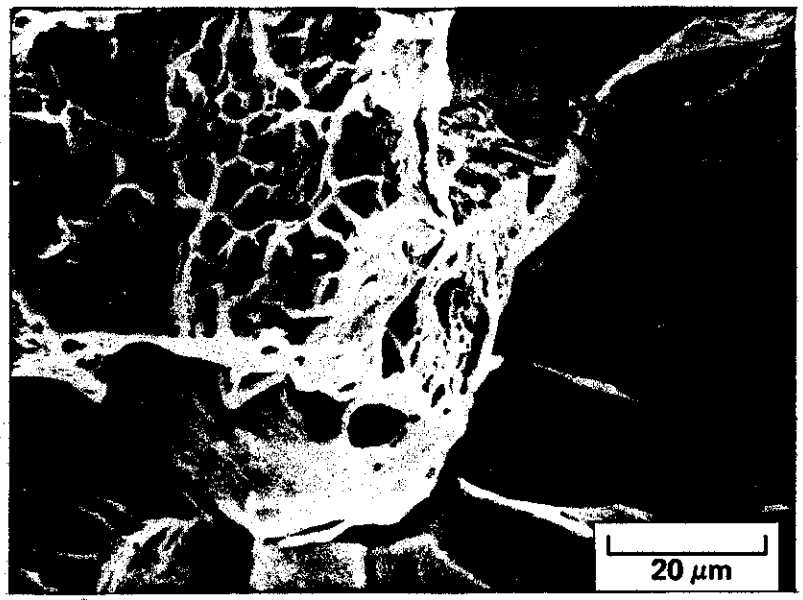


a) Tritium Charged and Aged

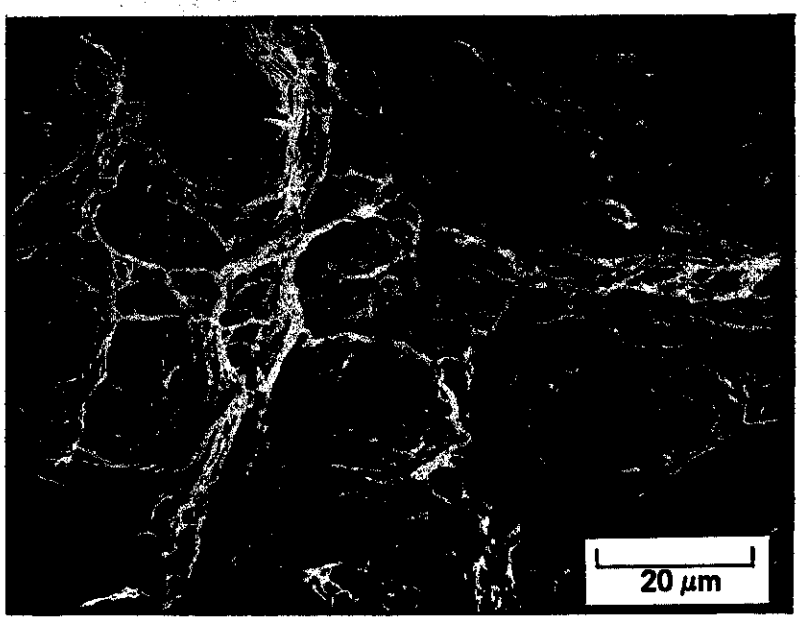


b) Tritium Charged, Aged and Outgassed

FIGURE 4. Fracture Modes in HERF Nitronic®-40 Stainless Steel

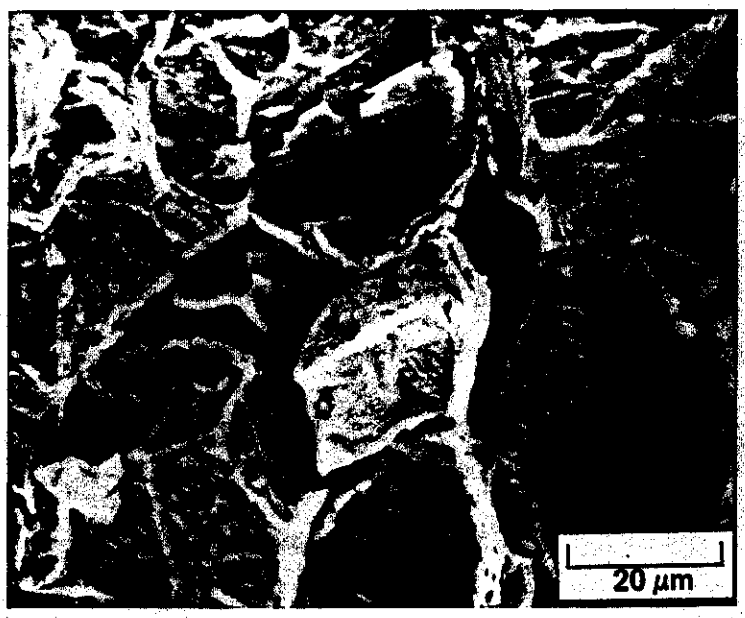


a) Tritium Charged and Aged

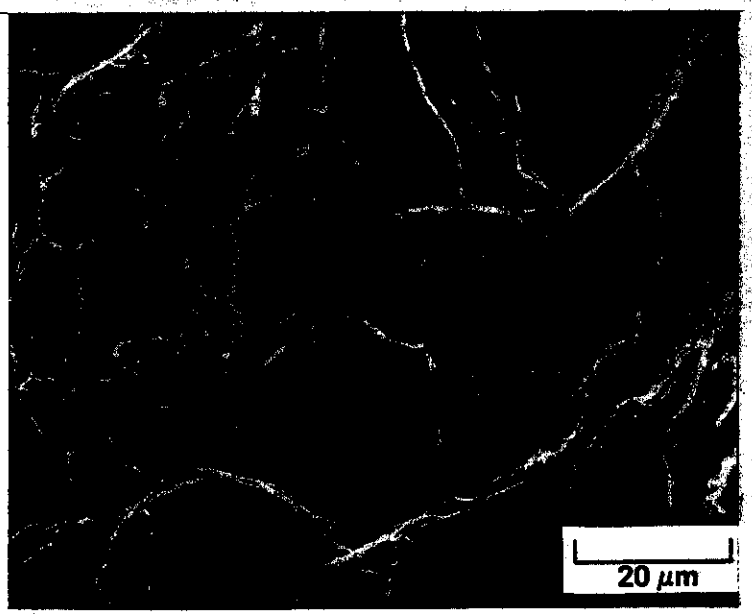


b) Tritium Charged, Aged and Outgassed

FIGURE 5. Fracture Modes in HERF A-286 Stainless Steel

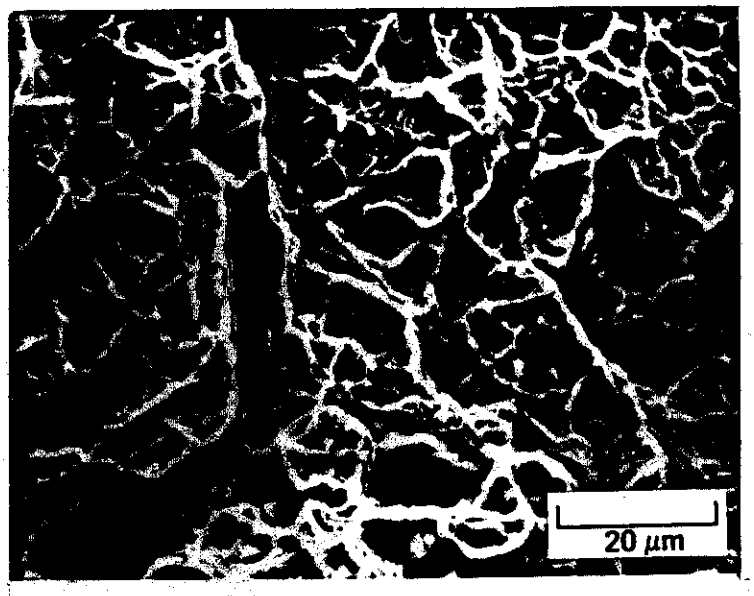


a) Tritium Charged, and Aged 30 Months

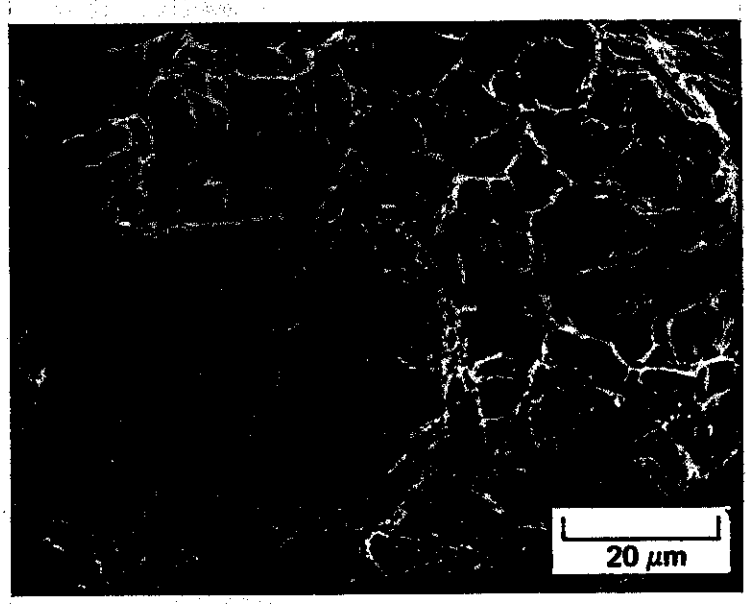


b) Tritium Charged, Aged and Outgassed

FIGURE 6. Fracture Modes in HERF Type 304L Stainless Steel



a) Tritium Charged and Aged 30 Months



b) Tritium Charged, Aged and Outgassed

FIGURE 7. Fracture Modes in HERF Type 316 Stainless Steel

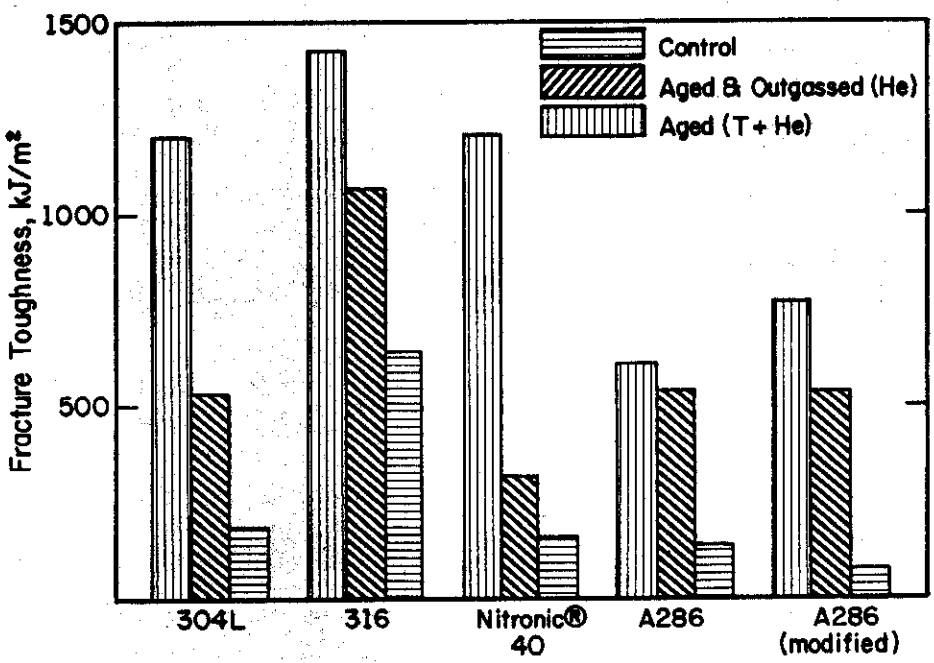


FIGURE 8. Effect of Tritium Outgassing on Fracture Toughness of Stainless Steels

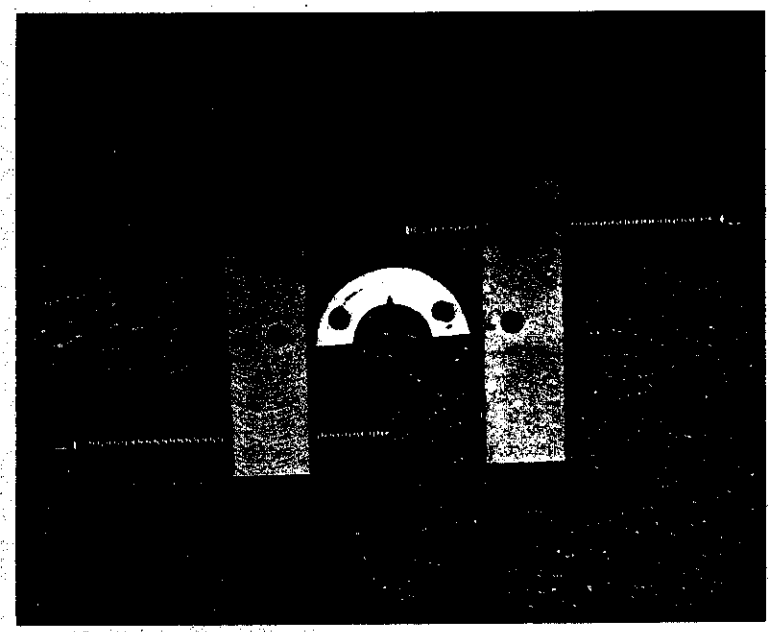


FIGURE 9. Loading Fixture for C-Shaped Samples