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MICROSTRUCTURAL DESIGN OF PCA AUSTENITIC STAINLESS STEEL FOR IMPROVED RESISTANCE TO HELIUM EMBRITTLEMENT UNDER HFIR IRRADIATION*

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Several variants of Prime Candidate Alloy (PCA) with different preirradiation thermal-mechanical treatments were irradiated in HFIR and were evaluated for embrittlement resistance via disk-bend tensile testing. Comparison tests were made on two heats of 20%-cold-worked type 316 stainless steel. None of the alloys were brittle after irradiation at 300 to 400°C to ~44 dpa and helium levels of 3000 to ~3600 at. ppm. However, all were quite brittle after similar exposure at 600°C. Embrittlement varied with alloy and pretreatment for irradiation to 44 dpa at 500°C and to 22 dpa at 600°C. Better relative embrittlement resistance among PCA variants was found in alloys which contained prior grain boundary MC carbide particles that remained stable under irradiation.

1. INTRODUCTION

Titanium or niobium modifications are known to improve the helium embrittlement resistance of austenitic stainless steels under neutron irradiation.[1-3] In 1967, Rowcliffe et al.[1] and Martin and Weir[2] conjectured that the reason for such resistance was interfacial helium bubble trapping by g.b. MC carbides. Kesternich and Rothaut[4] recently demonstrated embrittlement resistance resulting from helium trapping at fine matrix MC particles in helium preinjected and creep tested DIN 1.4970 (15 wt % Ni–15 Cr–0.1 C–1.3 Mo–0.3 Ti stainless steel).

Helium embrittlement resistance at the higher helium generation rates expected for fusion (12–15 at. ppm He/dpa) is an important concern for first-wall lifetime at higher temperatures. In 1981, preirradiation microstructural variants of PCA were designed and produced, with optimized distributions of grain boundary titanium (MC) carbide, intended for better helium embrittlement resistance in fusion reactor service.[5] An important feature was incorporation of our understanding of phase stability under neutron irradiation.[6,7] The present work is intended to evaluate relative embrittlement resistance through bend testing of various specimens irradiated in HFIR at the high helium generation rates of 20 to 80 at. ppm/dpa.

2. EXPERIMENTAL

Steel compositions appear in Table I. The designations and descriptions of the PCA pretreatment variants are given elsewhere in this publication.[8] Standard 3-mm-diam disks were punched from 0.254-mm-thick sheet stock. Four disks of each of these were irradiated in HFIR at 300, 400, 500, and 600°C to fluences producing 10.5 to ~44 dpa.[8] After irradiation, selected disks were bend tested at their irradiation temperature. Details of the disk bend testing and calculation of bend ductilities can be found elsewhere.[9-11] Scanning electron microscopy (SEM) and optical stereomicroscopy were used to examine cracks or fractures on

Table I. Alloy Compositions[a] (wt %)

	Cr	Ni	Si	C	P
PCA[b]	14.0	16.2	0.4	0.05	0.01
ref. 316	17.3	12.4	0.7	0.05	0.03
N-lot 316	16.5	13.5	0.5	0.05	0.01

[a]1.6–1.8 Mn, 2.3–2.5 Mo, bal Fe.
[b]0.24 Ti.

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tested disks. Transmission electron microscopy (TEM) was used to observe grain boundary microstructures in identically irradiated, but untested disks.[8]

3. RESULTS AND DISCUSSION

3.1 Disk bend testing

Disk bend ductilities as functions of irradiation temperature (equal to test temperature) and fluence for the various alloy and pretreatment variants (47 disks) are shown in Fig. 1. The data symbols also indicate fracture behavior (opened symbols — no cracks; filled symbols — intergranular cracking). The data are separated into general trend bands indicating behavior judged relatively better or worse. Figure 2 shows SEM of obvious examples for fracture behavior of specimens judged better or worse. In general, PCA-B1 and -B2 as well as CW 316 (ref.-heat) exhibited better behavior at higher temperatures and fluences than the other specimens.

The temperature dependence of the bend ductilities indicated virtually no embrittlement due to irradiations of ~22 and 44 dpa at 300 to 400°C [Fig. 1(a,b)]. Consistent with this, optical stereomicroscopy indicated considerable

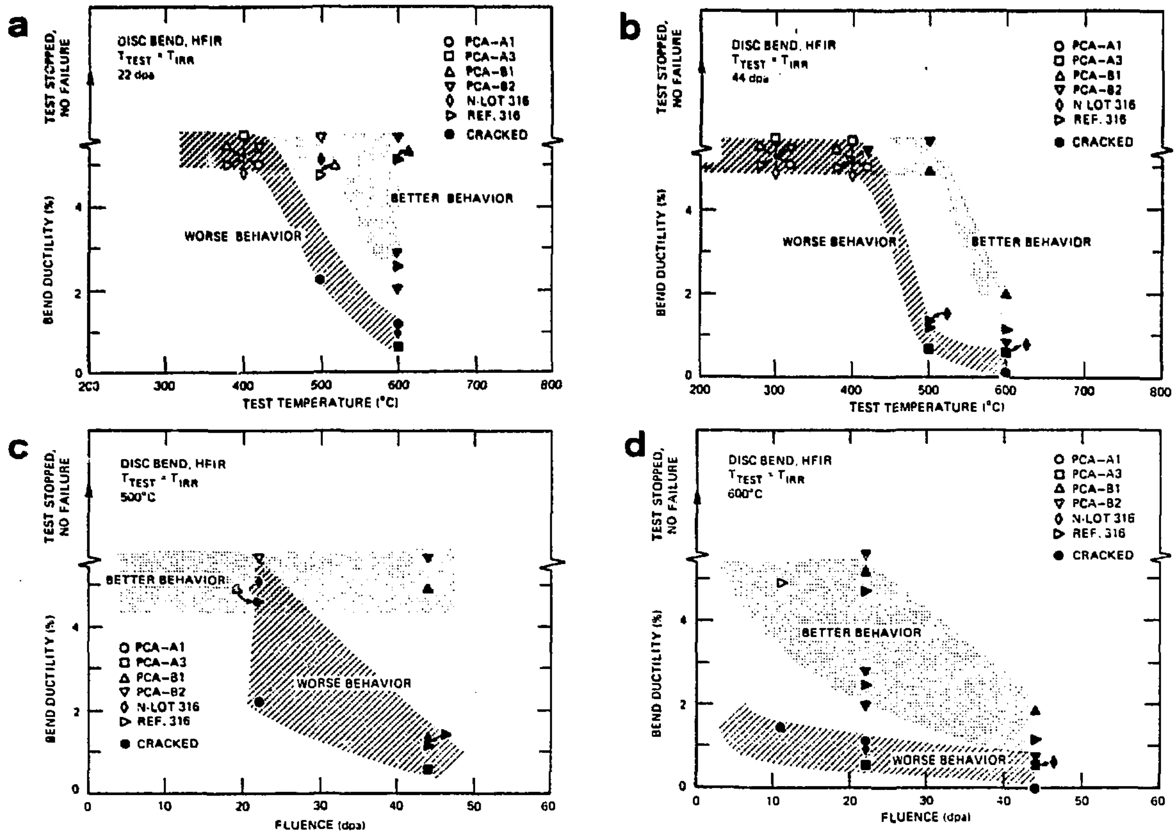

FIGURE 1

Plots of bend ductility as functions of irradiation temperature at (a) ~22 dpa and (b) 44 dpa and as functions of fluence at (c) 500°C and (d) 600°C for the alloys indicated after irradiation in HFIR. Note that the helium content (due to higher nickel content) is higher in PCA (~3600 at. ppm) than the CW 316s (~3000 at. ppm) after exposure to ~44 dpa. Scatter bands indicate relatively better or worse behavior as judged from both fracture behavior and bend ductility.

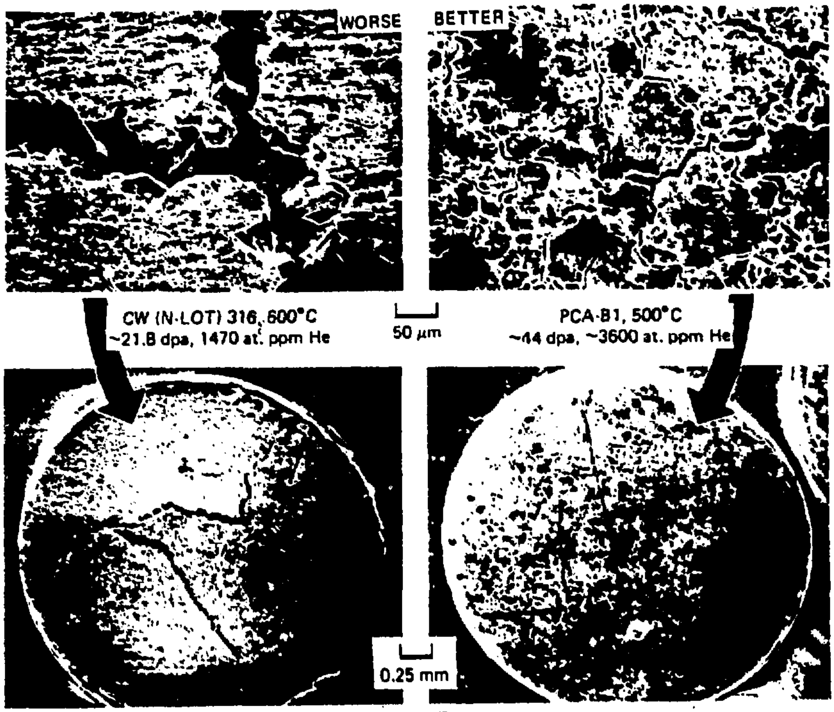

FIGURE 2
SEM fractography at high and low magnifications of HFIR-irradiated and bend tested ($T_{test} = T_{irr}$) disks and that failed (i.e., CW 316 N-lot designated worse) and passed (i.e., PCA-B1 designated better) for comparison. Irradiation conditions are indicated.

cupping of the disks to conform to the punch during testing and SEM showed no cracking. At 500 to 600°C, embrittlement increased with increased temperature and fluence, but varied among the specimens. PCA-B1 (solution annealed plus 8 h at 800°C plus 8 h at 900°C) and -B2 (solution annealed plus 8 h at 800°C plus 25%-cold-work plus 2 h at 750°C) consistently show better ductility and embrittlement resistance than PCA-A3 (25%-cold worked), -A1 (solution annealed), or CW 316s at 500 to 600°C. This is particularly true at 500°C after ~44 dpa (see for instance, PCA-B1 in Fig. 2). At 500°C and ~22 dpa, CW 316 (ref.-heat), PCA-B1, and -B2 all have similar behavior; this is also the case for 600°C at both ~22 dpa (with multiple specimen testing) and ~44 dpa. The CW 316 (ref-heat) appears to be less prone to embrittlement (postirradiation tensile testing) than other heats of type 316 (primarily the DO-heat) irradiated in HFIR.[12] The heat-to-heat embrittlement variation may be due to residual elemental differences (i.e., P content). Furthermore, helium production in CW 316 (ref.-heat) is considerally less than PCA in HFIR due to a 22 to 25% lower nickel content (see Table I).

The fluence dependence of bend ductilities shows that all of the alloys eventually are embrittled at 600°C. However, continued embrittlement resistance was achieved in PCA-B1 and -B2 at 500°C [Fig. 1(c,d)]. At 600°C, severe embrittlement occurred in PCA-AI after only ~11 dpa and ~550 at. ppm He. At ~22 dpa, PCA-A1, -A3, and CW 316 (N-lot) are all similarly embrittled at 600°C and grow only slightly worse as fluence increases [see Fig. 1(d)]. These disks show very little cupping and very large, opened intergranular cracks via SEM [see CW 316 (N-lot) in Fig. 2], as well as clearly defined and sharp drops in their load versus displacement curves.[10] By comparison, PCA-B1, -B2, and CW 316 (ref.-heat) show less embrittlement at 22 dpa judged by higher ductilities prior to catastrophic cracking than the other disks. PCA-B1 and -B2 show only small intergranular cracks in SEM (see Fig. 2) and they exhibit no sharp load drop (load-displacement curve) even after ~44 dpa at 500°C. Their ability to resist rapid intergranular cracking is a sign that some ductility was retained, especially compared to the other alloys.

3.2 Grain boundary microstructural observations

Grain boundary TEM was obtained from irradiated, untested disks. All alloys irradiated at 300 to 400°C showed similar tiny bubbles. However, at 500 and 600°C, both grain boundary bubble and precipitation development varied significantly with alloy and pretreatment variations. Figure 3 shows that increased helium generation under neutron irradiation increases bubble nucleation at the grain boundaries, for CW 316 (DO-heat) irradiated at 525 to 550°C in EBR-II and HFIR. Similar steels irradiated in EBR-II show no evidence of embrittlement below about 650°C to very high fluences[12] These observations indicate that embrittlement and g.b. bubble formation are more the result of helium buildup than displacement damage level. However, the variation of embrittlement among the alloys irradiated in HFIR indicated that differences in bubble and precipitate structure at boundaries also strongly influence embrittlement.

In this work, only PCA-B1 and -B2 contained g.b. precipitates (MC) prior to irradiation. These precipitates were produced by aging for 8 h at 800°C after solution annealing.[5] The as-fabricated g.b. MC precipitate microstructure can be seen for PCA-B1 in Fig. 4(a,c). During irradiation the medium-coarse g.b. MC precipitation developed via pretreatments in PCA-B1 and -B2 remained stable and virtually unchanged up to 44 dpa at 300 to 500°C and up to ~22 dpa at 600°C. The latter case is shown in Fig. 4(b,d). By contrast, fine MC developed via pretreatments in PCA-C (25%-cold-worked plus 2 h at 750°C) dissolved after only 11 dpa at 600°C. Neither PCA-A1 nor -A3 develop any g.b. MC under similar irradiations. The g.b. MC stability was also fairly independent of matrix void formation. At

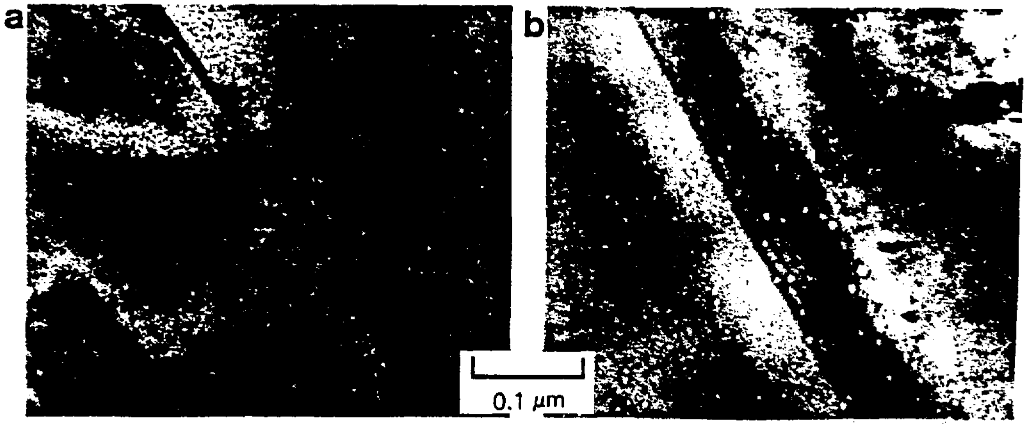

FIGURE 3
Grain boundary bubbles in 20%-cold-worked 316 (DO-heat) irradiated at 525 to 550°C in (a) EBR-II to 36 dpa and ~22 at. ppm He and (b) HFIR to 17.8 dpa and 1020 at. ppm He.

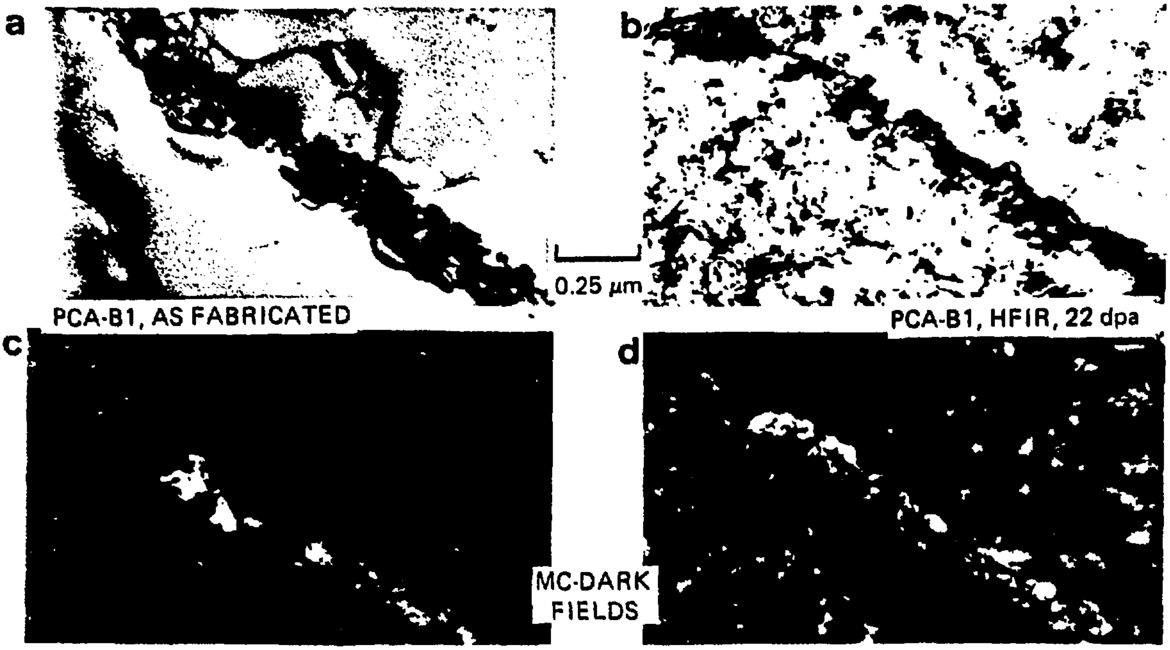

FIGURE 4
A comparison of grain boundary MC in PCA-B1 as fabricated (a) and (c), and after HFIR irradiation at 600°C (b) and (d) to demonstrate its stability under irradiation.

600°C and 22 dpa PCA-B1 develops high void swelling and radiation-induced precipitation, whereas PCA-B2 develops much less swelling and enhanced thermal precipitation (MC) under irradiation. However, both retain their g.b. MC distributions. The medium-coarse MC in both PCA-B1 and -B2 does dissolve after ~44 dpa at 600°C, possibly due to g.b. migration which occurs due to differential void swelling in adjacent grains. When the g.b. MC was stable, it caused much finer g.b. helium bubble distributions due to trapping at the interphase boundaries, particularly when compared to g.b.'s without MC (cf PCA-A1 and -B2 in Fig. 5). Bubbles are almost unresolvable at these MC particles at lower fluence.

In comparison to PCA-B1 or -B2, CW 316 (N-lot) developed bubbles and little g.b. precipitation at 500°C, and bubble and M_6C precipitate structures that coarsened considerably with fluence at 600°C. By contrast, grain boundaries in CW 316 (ref.-heat) have fairly uniform and stable dispersions of medium-coarse M_6C particles[13] after ~10 dpa in HFIR at 400 to 550°C. This observation again relates embrittlement resistance to g.b. precipitate stability for these heats of CW 316.

3.3 Bend test — microstructural correlation and summary

This work emphasizes that embrittlement resistance was anticipated and did correlate with the stability and beneficial bubble refinement of g.b. MC in the PCA alloys at 500 to 600°C. PCA-B1 and -B2, despite their higher helium content, also show consistently better embrittlement resistance than the CW 316s, especially N-lot. The CW 316 (ref.-heat) may also benefit from better stability of g.b. M_6C. At 400°C and below, embrittlement does not appear to be a problem for any of these alloys. Precipitate-

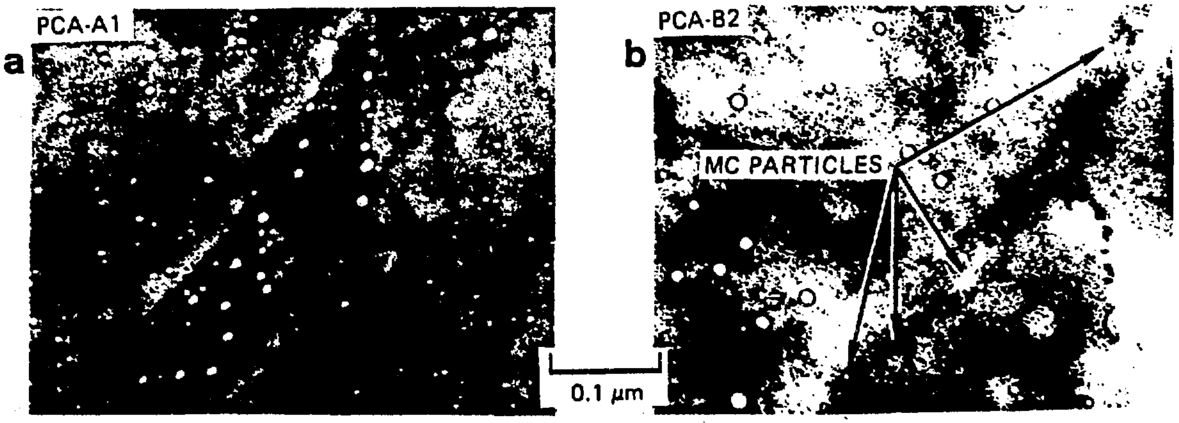

FIGURE 5
A comparison of grain boundary bubble structures in (a) PCA-A1 with no MC and (b) PCA-B2 irradiated in HFIR at 600°C to ~22 dpa (~1760 at. ppm He) with prior grain boundary MC.

free g.b.'s covered with many bubbles consistently appear quite brittle and prone to cracking for all alloys at 500 and 600°C. Stable grain boundary MC, and hence embrittlement resistance, does not naturally develop under irradiation, but must be produced by appropriate thermal mechanical treatments before irradiation. Because the MC instability appears to correlate with g.b. migration under irradiation, g.b. MC stability should be improved with more swelling resistant grains. Therefore, a new PCA microstructure, PCA-B3, has been developed to combine the void swelling resistance of PCA-A3 (ref. 8) with the g.b. MC structures of PCA-B1 and -B2. PCA-B3 is produced by aging solution-annealed material (PCA-A1) for 8 h at 800°C and then cold working 25%.

4. SUMMARY

The bend test results suggest qualitatively the possibility that embrittlement can be diminished by suitable design and control of the microstructure. These results must be supported by more engineering relevant mechanical testing to verify the embrittlement resistance and to qualify these alloy conditions for fusion reactor applications.

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