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IMPROVED SWELLING RESISTANCE FOR PCA AUSTENITIC STAINLESS STEEL UNDER HFIR IRRADIATION THROUGH MICROSTRUCTURAL CONTROL[*]

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Six microstructural variants of Prime Candidate Alloy (PCA) were evaluated for swelling resistance during HFIR irradiation, together with several heats of type 316 stainless steel (316). Swelling was negligible in all the steels at 300°C after ~44 dpa. At 500 to 600°C 25%-cold-worked PCA showed better void swelling resistance than type 316 at ~44 dpa. There was less swelling variability among alloys at 400°C, but again 25%-cold-worked PCA was the best. Microstructurally, swelling resistance correlated with development of fine, stable bubbles whereas high swelling was due to coarser distributions of bubbles becoming unstable and converting to voids (bias-driven cavities).

1. INTRODUCTION

A prime candidate alloy (PCA) currently undergoing study for fusion reactor applications is a titanium-modified austenitic alloy which demonstrates superior void swelling resistance under EBR-II irradiation.[1,2] The present work is part of the data base for developing swelling resistance reliable for the first wall of a Magnetic Fusion Energy (MFE) reactor.[3] Previous work introduced the alloy design and development of preirradiation microstructural variants and corresponding thermomechanical treatments of the PCA.[4] Seven microstructural variants were produced in that first stage of the development. Six were selected for the evaluation stage to scope the swelling resistance during HFIR irradiation.

Table 1. Alloy Compositions (weight percent)

Elements	Alloys			
	DO-heat 316	N-Lot 316	R1-heat 316 + Ti	PCA
Fe	Bal	Bal	Bal	Bal
Ni	13.0	13.5	12.0	16.2
Cr	18.0	16.5	17.0	14.0
Ti	0.05	--	0.23	0.24
Mo	2.6	2.5	2.5	2.3
Mn	1.9	1.6	0.5	1.8
C	0.05	0.05	0.06	0.05
Si	0.8	0.5	0.4	0.4
P	0.01	0.09	0.01	0.01
S	0.016	0.006	0.013	0.003
N	0.05	0.006	0.006	0.01

2. EXPERIMENTAL

Compositions of the PCA alloys and three other comparison materials designated DO-heat and N-lot 316s, and the R1-heat of 316 + Ti are given in Table 1. Designations, descriptions, and thermomechanical pretreatments for the preirradiation microstructural variants of the PCA are shown in Table 2. Standard 3-mm-diam disks were punched from 0.254-mm-thick sheet stock. PCA and 20%-cold-worked (CW) 316 (N-lot) disks were irradiated in experiments HFIR-CTR-30, -31, and -32, at temperatures of 300, 400, 500, and 600°C (verified by temperature monitors). More detail is available elsewhere.[5,6] Cavity volume fraction (cvf) swelling was measured via transmission electron microscopy (TEM), described elsewhere.[7] Data are also included on solution annealed (SA) and CW 316 (DO-heat) and CW 316 + Ti (R1-heat) from previous experiments[7,8] ranging in temperature from 325 to 755°C and in fluence from ~3 to 69 dpa (HFIR-SS-2 through -6 and HFIR-CTR-9 through -13). Calculated irradiation temperatures from the older HFIR experiments have been corrected[9] upwards by 50 to 75°C. All displacement damage (dpa) calculations in this work include the recent correction

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Table 2. Alloy Microstructural Variants and Thermomechanical Pretreatments

Designation	Microstructure	Treatment
PCA-A1	Very low dislocation density	50%-CW + SA for 15 min at 1175°C
PCA-A2	Moderately high dislocation density	SA (A1 treatment) + 10% CW
PCA-A3	Very high dislocation density	SA (A1 treatment) + 25% CW
PCA-B1	Low dislocation density; medium-g.b. and coarse matrix MC	SA (A1 treatment) + 8 h at 800°C at 900°C
PCA-B2	Very high dislocation density; medium-coarse g.b. and fine matrix MC	SA (A1 treatment) + 8 h at 800°C + 25% CW + 2 h at 750°C
PCA-C	Very high dislocation density; fine g.b. and matrix MC	SA (A1 treatment) + 25% CW + 2 h at 750°C

(increase) due to helium production from nickel transmutations, as noted by Greenwood.[6] The dpa values for HFIR-CTR-30, -31, and -32 ranged from ~10.5 dpa (calculated from dosimetry[6]) to ~44 dpa (estimated, pending dosimetry). Helium levels range up to about 3000 at. ppm for the type 316s and up to ~3600 at. ppm for the PCA after ~44 dpa.

3. RESULTS

3.1 Swelling

Of the six PCA variants examined PCA-A2, -B1, and -C were eliminated from further consideration because of poor swelling resistance after ~10.5 dpa at 500 and 600°C. The data are plotted as functions of temperature in Fig. 1 and are tabulated elsewhere.[10] The PCA variants and CW 316 (N-lot) show increased swelling with increased irradiation temperature. Study of PCA-A1 was continued as a higher swelling base line against which to gauge swelling resistance at higher fluence. A trend band for the CW (DO-heat) and CW 316 + Ti (R1-heat) data in this temperature and fluence range is also included in Fig. 1. At temperatures above 500°C, these alloys show better swelling resistance than the PCA variants or CW 316 (N-lot). However, below 450 to 500°C, the situation is reversed.

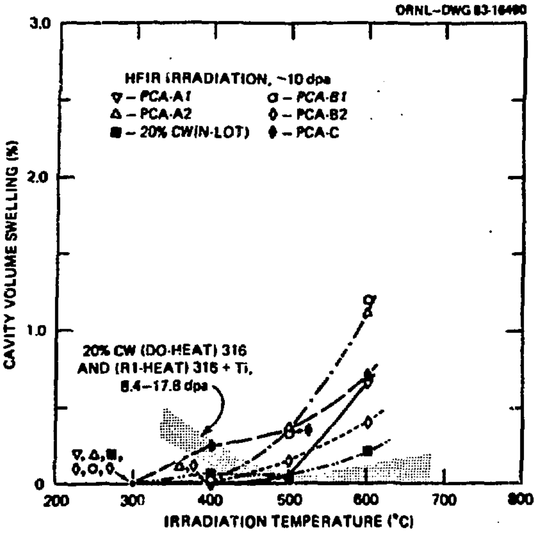

FIGURE 1

Swelling as functions of temperature for PCA variants and several 20%-cold-worked steels irradiated to lower fluences in HFIR.

The swelling values for PCA-A1, -B2, and -A3 and CW 316 (N-lot) are shown as functions of temperature [Fig. 2(a)] and fluence [Fig. 2(b)]. Trend bands for data on SA and CW 316 (DO-heat) are included for comparison. The swelling of the PCA variants and CW 316 (N-lot) is low at 400°C and negligible at 300°C, even at ~44 dpa. The temperature dependencies of swelling in PCA-A1 and CW 316 (N-lot) are weak and the levels of swelling low at ~22 dpa and are roughly parallel to the dependencies found at ~10 dpa. The PCA-A3 has the least temperature dependence at ~22 dpa at the lowest swelling. All of these alloys develop much stronger temperature dependencies at ~44 dpa. Swelling of the PCA variants peaks at 500°C, whereas that of [illegible]

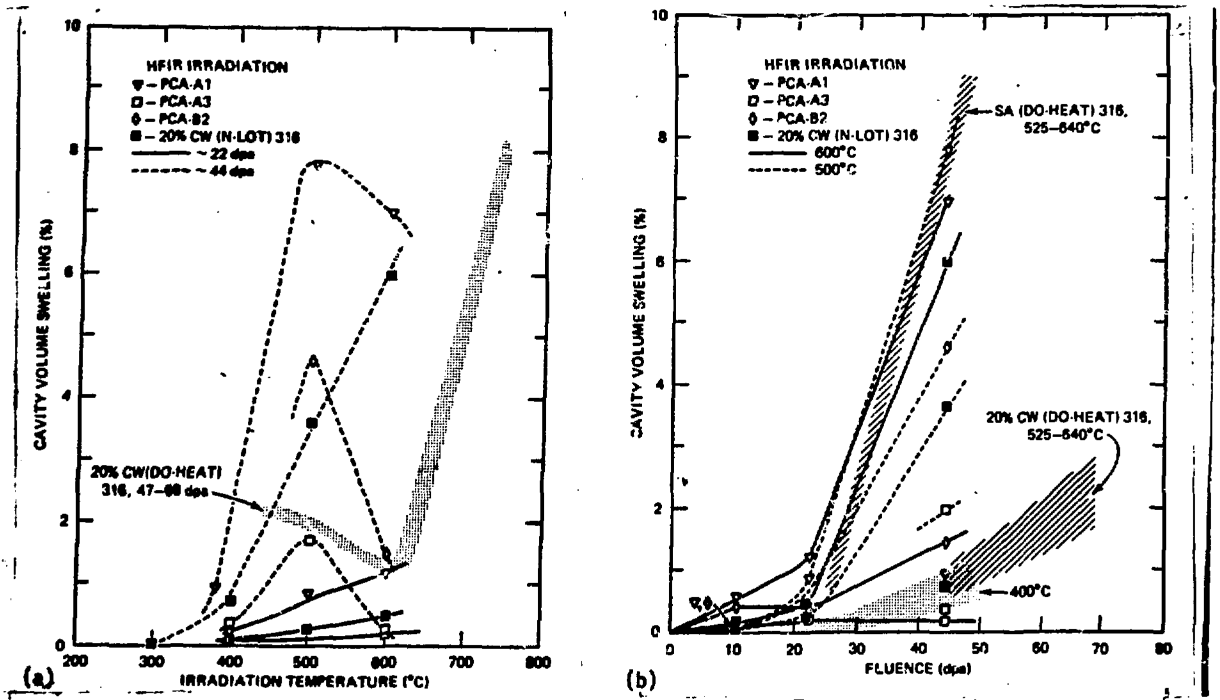

FIGURE 2

Swelling of several HFIR-irradiated PCA variants and type 316s (a) as functions of temperature at 22 and 44 dpa and (b) as functions of fluence at 400 to 640°C.

CW 316 (N-lot) increases monotonically from 400 to 600°C. Of the PCA variants, the swelling resistance of PCA-A3 is by far the best, from 400 to 600°C. PCA-A3 is also considerably better than CW 316 (N-lot) at 500 to 600°C at 44 dpa. PCA-A1, -B2, and CW 316 (N-lot) are clearly less resistant than CW 316 (DO-heat) at higher fluences below 600°C.

At 400°C, PCA-A1, -A3, and CW 316 (N-lot) all swell at less than 0.04%/dpa, with the latter two being the lowest. With increasing fluence, swelling rates increase rapidly after a low swelling transient of about 20 to 25 dpa at both 500 and 600°C for PCA-A1 and CW 316 (N-lot) and only at 500°C for PCA-B2. These alloys roughly follow the high swelling behavior of SA 316 (DO-heat) irradiated at 525 to 640°C, with swelling rates ranging from 0.16 to 0.37%/dpa. PCA-A3 at 500°C and PCA-B2 at 600°C simply extend their lower fluence behavior with no additional upturn, similar to CW 316 (DO-heat) irradiated at 525 to 640°C. PCA-A3 shows virtually no increase in swelling from ~22 to ~44 dpa at 600°C, and hence an increased transient period.

3.2 Microstructural development

Over 50 TEM disks were examined, and the total microstructure (i.e., dislocation, precipitate, cavity, and grain boundary components) of each was observed in detail. Only a brief summary of these results appear in this paper; more details can be found elsewhere,[10,11] and will be published later.

In these samples, swelling greater than about 0.5 to 1.0% appears to be due to formation of large voids (or bias-driven cavities[12]) initiated at small helium bubbles (or stable cavities), particularly at higher fluences at 400 to

600°C. A basis for distinguishing between voids and bubbles experimentally has been suggested previously,[13] even though both can be described by the more general term of cavity. For example, at 600°C CW 316 (N-lot) and PCA-A1 begin forming voids at ~10.5 dpa[10,11] which then lead to the highly swollen void microstructures found at ~44 dpa and shown in Fig. 3(a) and (b), respectively. By contrast, large voids do not develop in PCA-A3 after similar exposure, as shown in Fig. 3(c). Although many fine bubbles can be found at lower fluences in the PCA-A1 and CW 316 (N-lot), they do not remain stable, but coarsen with increasing fluence and convert to a combination of large matrix voids and precipitate-associated voids. Many more (up to a factor of 50) fine bubbles develop early in PCA-A3, which appear stable and do not appreciably coarsen or convert to voids. The microstructure of PCA-A3 at ~22 dpa is nearly identical to that shown at ~44 dpa in Fig. 3(c).

Microstructural dependence of void formation on temperature from 300 to 600°C is shown in Fig. 4 for PCA-A3 irradiated to ~44 dpa. At 300°C, bubbles are barely resolvable (<2 nm in diameter). High concentrations ($>10^{23}$ m^{-3}) of fine (4 to 5 nm in diameter) bubbles remain stable at 400 and 600°C [Fig. 4(b,d) respectively]. At 500°C, both matrix and precipitate-associated voids form.

Void swelling differences found in Fig. 3 at 600°C correlate with differences observed in the evolution of the precipitate and dislocation components among these alloys. Both PCA-A1 and CW 316 (N-lot) develop higher dislocation concentrations, many more Frank interstitial loops, and radiation-induced phases (RIP), (γ' (Ni_3Si) in this case) early in their irradiation histories compared to PCA-A3. These in turn correlate with the onset of early bimodal cavity distributions in the matrix. In CW 316 (N-lot), the Frank loops remain in the microstructure to ~22 dpa at 600°C and to ~44 dpa in PCA-A1. In both of these steels, the γ' forms early and then dissolves, giving way at higher fluence to coarse η (M_6C) in the CW 316 (N-lot) and to coarse η and/or G phase in the PCA-A1. Fairly coarse voids then develop in association with these coarse precipitates. By contrast, a considerable amount of fine MC, very few Frank loops, and considerable dislocation recovery develop in PCA-A3 at ~22 dpa and then these remain unchanged with increasing fluence. The

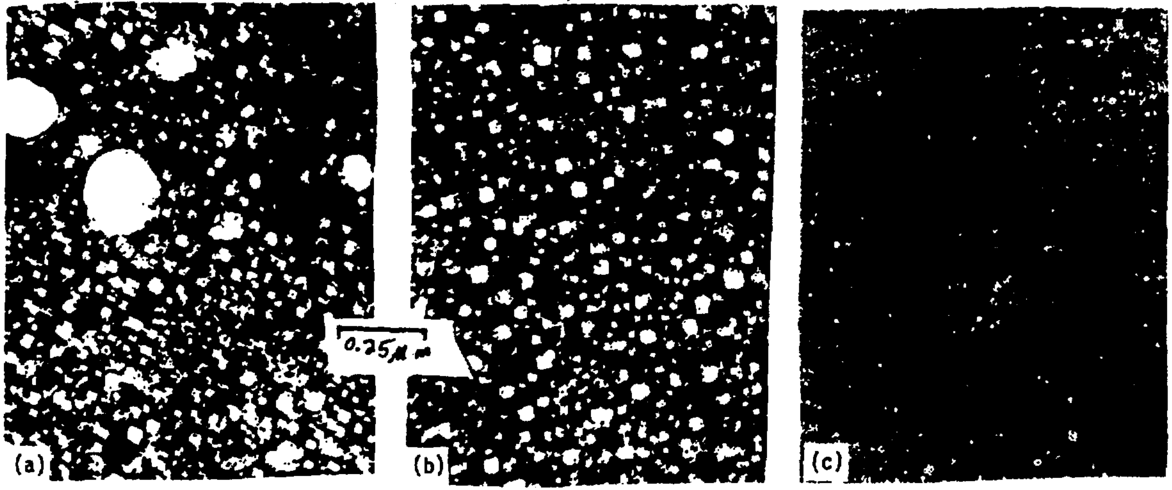

FIGURE 3
Cavity microstructures of stainless steels irradiated in HFIR at 600°C to ~44 dpa. (a) CW 316 (N-lot), (b) PCA-A1, (c) PCA-A3.

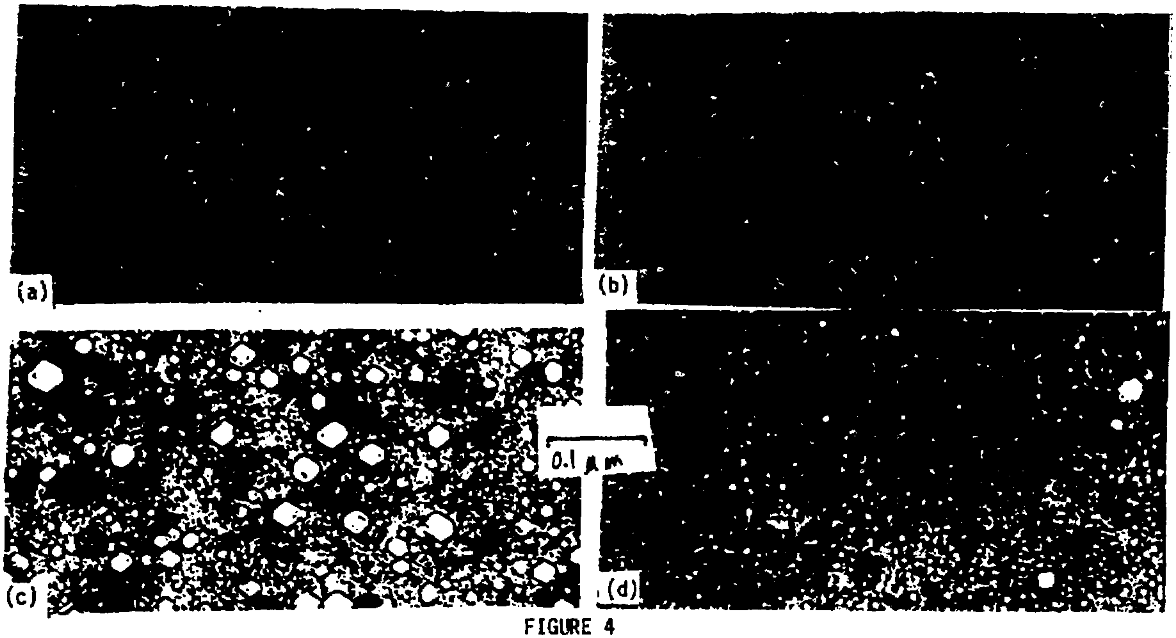

FIGURE 4
Cavity microstructures of PCA-A3 irradiated in HFIR to ~44 dpa at (a) 300°C, (b) 400°C, (c) 500°C, and (d) 600°C. Note the void formation at 500°C.

one-to-one spatial correspondence between rafts of fine bubbles and clusters of fine MC particles can be seen in Fig. 5. Comparing PCA-A1 and -A3 at 600°C and ~44 dpa, the absence of voids correlates with maximum fine MC precipitation and minimum formation of coarse, radiation-induced solute segregation (RIS)-induced or -modified phases. Fine, stable MC is evident at both 400 and 600°C after ~44 dpa, but is minimal at 500°C and mixed with coarser phases associated with voids. These correlations are consistent among the other PCA variants considered only at low fluence.[10,11]

4. DISCUSSION

The comprehensive microstructural data reveal at least several expected and unexpected mechanisms influencing swelling resistance. It is expected that large differences in point defect sink strengths between alloys should influence both the critical radius for conversion of bubbles (or stable cavities) to voids and the bifurcation of possible cavity evolution into high- or low-swelling paths.[4,12,14] Furthermore, it is also expected that such large differences in cavity evolution can affect precipitation, leading to enhanced thermal precipitation (like MC) when RIS is suppressed or diluted in a refined, cavity sink-dominated system.[13] Similar correlations between void formation and phase evolution are also observed over a large body of data on PCA by Imeson et al.[13] It seems unexpected, at least from previous neutron data, that Frank loop formation should be so variable under the same irradiation conditions, and so strongly correlate with void development and especially RIS-induced γ' formation at higher temperatures. Much yet needs to be understood about the effects of helium on overall microstructural/microchemical evolution.

5. CONCLUSIONS

1. PCA-A2 (10% CW), -B1 (SA plus double aged), and -C (25% CW plus aged) were eliminated from further consideration due to rapid swelling

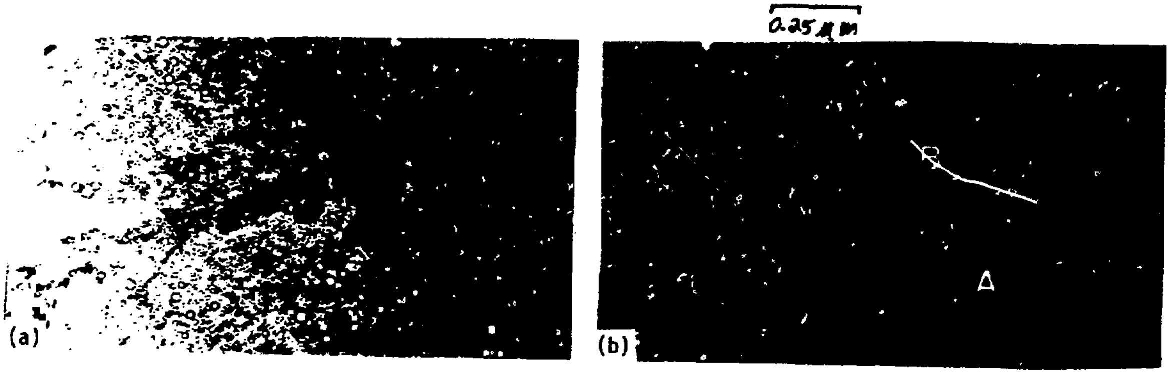

FIGURE 5
Microstructural correlation of (a) patches of fine bubbles in bright field and (b) clusters of fine MC particles in precipitate dark field in PCA-A3 irradiated in HFIR at 600°C to ~44 dpa.

after only ~10 dpa at 500 to 600°C. After ~44 dpa, PCA-A1 (SA) exhibited the highest swelling from 400 to 600°C, nearly as high as SA 316 (DO-heat).

2. At higher fluences in HFIR, the void swelling resistance of PCA-A3 is similar or better than CW 316 (DO-heat), and better than CW 316 (N-lot) from 500 to 600°C. PCA-A3 is more resistant to swelling than CW 316 (DO-heat) below 500°C.

3. Swelling resistance under high-helium generation correlates directly with the formation of high concentrations of stable, fine bubbles that resist conversion to voids. Other correlation factors are the presence of low network dislocation concentrations, few Frank loops, and a suppression of RIS effects on precipitation (which promote MC in the PCA).

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