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Temperature Dependence of Swelling in Type 316 Stainless Steel Irradiated in HFIR*

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Abstract

The temperature dependence of swelling was investigated in solution-annealed (SA) and 20% cold-worked (CW) type 316 stainless steel irradiated to 30 dpa at 300 to 600°C in the High Flux Isotope Reactor (HFIR). At irradiation temperatures ≤400°C, a high concentration (2 to 4×10^{23} m^{-3}) of small bubbles (1.5 to 4.5 nm diam) formed uniformly in the matrix. Swelling was low (<0.2%) in both SA and CW materials irradiated to 30 dpa. In the SA 316, cavity size increased but the number density decreased with increasing irradiation temperature above 500°C. At 500°C, there was a mixture of bubbles and voids, but at 600°C, most of the cavities were voids. Maximum swelling (~5%) occurred at 500°C. By contrast, cavities in 20% CW specimens were much smaller, with diameters of 6 and 9 nm at 500 and 600°C, respectively, suggesting that they were primarily bubbles. The cavity number density in the CW 316 at both 500 and 600°C (~1×10^{22} m^{-3}) was about one order of magnitude less than at 400°C. Swelling increased slightly as irradiation temperature increased, peaking at 600°C (0.3%). These results indicate that SA 316 swells more than CW 316 at 500 and 600°C, but both SA and CW 316 are resistant to void swelling in HFIR at 400°C and below to 30 dpa.

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1. Introduction

The radiation resistance of structural materials is a crucial factor in the development of fusion reactors. The irradiation damage caused by fusion neutrons is characterized by atomic displacement-damage and helium produced by transmutation reactions. The displacement-damage effects in type 316 stainless steel have been extensively studied by irradiating the steels in fast breeder reactors. The effects of transmuted helium have been investigated by irradiation in HFIR (High Flux Isotope Reactor), a mixed-spectrum reactor in which thermal neutrons react with ^{58}Ni to produce helium [1–3]. However, most of these data have been obtained at temperatures which are much higher (≥500°C) than those expected in the proposed FER (Fusion Experimental Reactor). Swelling in the presence of helium has not yet been examined for the low temperature operating range of the FER.

The objective of this paper is to investigate the swelling behavior in type 316 stainless steel irradiated in HFIR to 36 dpa at 300 to 600°C. This work has been performed under the U.S./Japan collaborative program for fusion materials research.

2. Experimental

The chemical composition of the type 316 stainless steel used in this experiment is given in Table 1. After 3-mm-diam transmission electron microscopy (TEM) disks were punched from the cold-rolled 0.27-mm-thick sheet, they were solution annealed for 30 min at 1050°C. To produce 20% CW materials, the steel (0.34 mm in thickness) was annealed for 30 min at 1050°C prior to a final 20% reduction by cold rolling, after which the disks were punched from the sheet. The surfaces of the disk were lightly mechanically polished to 0.25-mm-thick sheet to prevent strain prior to irradiation.

The disks were irradiated in the JP-1, -3, -6, and -7 capsules of HFIR at 300 to 600°C up to 36 dpa (2327 appm He). After irradiation, disks were electropolished in a twin-jet Tenupol unit and examined with a JEM 2000FX high-resolution electron microscope operating at 200 kV. Carbon-extraction replica films were also prepared from the TEM disks to analyze the composition of precipitates. These analyses were made using x-ray energy dispersive spectroscopy (XEDS) and a TN 5500 computer to obtain quantitative compositions.

3. Experimental Results

3.1 Cavity Distribution

The cavities and cavity size distributions observed in SA and CW specimens irradiated at 300 to 600°C in HFIR are shown in Figs. 1 and 2, respectively. At 300°C, a uniform distribution of fine cavities (1 to 5 nm in diameter) was observed in the matrix of both CW and SA steels; densities were $\sim 10^{23}$ m^{-3}. The sharp, narrow peaks in the size distributions suggest that most of these fine cavities are subcritical bubbles [Fig. 2(a) and (b)]. The swelling in both materials at 300°C was less than about 0.14%. Cavity distributions in the matrix of SA and CW steels at 400°C are similar to those observed at 300°C, except that some larger cavities (4 to 8 nm in diameter) are also present. The larger faceted cavities are probably evolving into supercritical voids. The size-distribution histograms show a broadening toward larger sizes, suggestive of an incipient bimodal distribution. These size distributions suggest a critical size of 4 to 5 nm [4]. The swelling in SA and CW at 400°C was 0.22 and 0.14%, respectively.

As the irradiation temperature increased to 500°C, cavity evolution became significantly different between SA and CW material. Cavity sizes in SA material irradiated at 500°C are much larger than at 400°C, while those in CW material are only slightly larger than at 400°C [Fig. 1(e) and (f)]. Both size distributions of SA and CW material at this temperature show sharp, narrow peaks at smaller sizes (<5 nm) that suggest these are subcritical bubbles, [Fig. (2(e) and (f)]. However, SA material develops a broad tail toward larger sizes that suggests a significant number of subcritical bubbles have converted to bias-driven voids. The concentration of cavities in SA and CW decreases by about one order of magnitude compared to 400°C. The swelling in SA and CW was 1.09 and 0.16%, respectively. In SA material at 500°C, cavity appearance suggests that the largest ones are being affected by the electropolishing and therefore are eliminated from swelling estimates. This would lead to an underestimate of the swelling determined by TEM. Swelling values obtained by immersion density measurements were larger than those obtained by TEM; swelling in SA and CW was 5.1 and 0.87%, respectively.

At 600°C, large, faceted voids (10 to 65 nm in diameter) were observed in SA 316, whereas a uniform distribution of much smaller cavities (5 to 10 nm in diameter) was found in the matrix of CW 316 [Fig. 1(g) and (h)]. The cavity size distribution of SA 316 shows a broad distribution with a tail toward larger sizes, with the peak at about 30 nm, whereas the cavity size distribution in CW is sharp and narrow with a peak at 10 nm [Fig. 2(g) and (h)]. The concentration of voids decreased by three orders of magnitude in SA, but decreased only slightly in CW. The swelling decreases to 1% in SA but slightly increases in CW (0.29%) at 600°C compared to 500°C. However, polishing effects could also cause many large voids to be missed in TEM estimates of swelling at 500°C.

3.2 Dislocation Loop Evolution

The dislocation structure in all specimens consisted of network and Frank faulted loops. Loops are a particularly important factor in assessing the effect of point defects produced by irradiation. The faulted Frank loops were therefore imaged separately from the network, using a high-resolution dark-field method suggested by Okamoto and Harkness [5].

A high concentration of dislocation loops was found in SA and CW at 300°C. The dislocation loop density increased slightly at 400°C, but then decreased dramatically at 500°C. Loop growth was greater at irradiation temperatures of 500°C and higher, and many apparently have unfaulted to form a network. At 600°C, only a loose tangle of dislocation network was found; no loops were observed by using the high-resolution dark-field method.

3.3 Precipitation

The precipitates formed in type 316 stainless steel during HFIR irradiation were quite sensitive to the irradiation temperature. No precipitation was observed in either SA or CW at 300°C. Precipitates did develop at 400, 500, and 600°C, as shown in Fig. 3. A low concentration of small intragranular particles were observed at 400°C via Moiré fringes [see Fig. 3(a)]. These particles were found to be the chromium-rich $M_{23}C_6$ type phase, as shown via XEDS analysis on an extraction replica in Fig. 3(b); Moiré fringe spacing was also consistent with the crystal structure information for $M_{23}C_6$ [6,7]. Some precipitates were also observed along grain boundaries, and there was no difference in precipitation between SA and CW materials. None of these $M_{23}C_6$ precipitates showed any association with cavities.

At 500 and 600°C, there was a large difference in precipitation between SA and CW 316. Many precipitates at 500°C were observed throughout the matrix in SA 316 and were associated with voids of similar size. The average diameter and number density of precipitates were 37 nm (15- to 65-nm range) and 2.4×10^{21} m^{-3}, respectively. By contrast, precipitates formed in CW specimens at 500°C were similar to those observed at 400°C. The XEDS spectrum of an extracted particle from SA 316 irradiated at 500°C is presented in Fig. 3(e) and the composition is listed in Table 2. This precipitate was identified as the M_6C type phase whose major components are Ni, Cr, Fe, Si, and Mo. The composition agrees well with the characteristic M_6C composition reported by Lee et al. and others [8–10]. The precipitate microstructural evolution in CW 316 at 600°C is similar to that found at 500°C. However, for the same comparison, precipitates in SA 316 coarsened considerably with increased temperature [cf. Fig. 3(b) and (c)]. These precipitates had an average diameter of 160 nm (100- to 270-nm range) and number density of 2.3×10^{19} m^{-3}. Despite increased precipitate sizes, the associated voids were not much larger than those found at 500°C, which means that at 600°C, voids are smaller than their associated precipitates. Figure 3(f) shows an XEDS spectrum from a typical extracted particle and from SA 316 at 600°C, and again, the phase is identified as M_6C. Compositionally, this phase contains about 30 wt % each of Cr and Ni, 17% Mo, and 7% Si, and the lattice parameter is a_0 = 1.10 nm. Comparison of the M_6C composition at 500 and 600°C (Table 2 and Fig. 3) shows that the molybdenum content of the phase has increased considerably with irradiation temperature. Most of the precipitates observed on this replica were identified as M_6C phase. Other phases like γ′, G, and Laves, which have been reported elsewhere [8–11], were not found in the present work; $M_{23}C_6$ (τ) phase was found only at 400°C. The probability of microsegregation within the huge precipitates on the replica was examined, but results indicate a uniform distribution of elements for the electron probe size used (~65 nm).

Data on irradiation conditions and microstructures on 316 stainless steel irradiated in HFIR are summarized in Table 3. Temperature dependence of cavity swelling is shown in Fig. 4; swelling data for 20% CW DO-heat and N-lot determined at various fluences by Maziasz and Braski [4] are also included.

4. Discussion

In the present work, a peak in swelling occurs at 500°C in SA material (Fig. 4). On the other hand, swelling in CW material does not show a strong dependence on irradiation temperature. Compositional differences in precipitate phases also suggest that maximum radiation induced segregation (RIS) of solute elements correlates with the swelling maximum observed in SA 316 at 500°C.

4.1 SA Material

At temperatures below 400°C, swelling is low in SA 316 because the cavity size is small and the cavity concentration is high. Calculations of gas atom accommodation by the cavity structure suggest that most of the fine cavities are equilibrium bubbles. The high concentration of helium bubbles and loops makes the critical cavity size large to suppress conversion of bubbles to voids [12].

At 500°C, the average cavity size is about five times that at 400°C, while the cavity number density is about one order of magnitude less than at 400°C. More than half of the cavities present appear to be larger than their critical size and thus have converted into bias-driven voids [4,13]. Bubble growth and/or coalescence are more likely at higher temperatures. Coalescence coarsening of cavities with increasing temperature could contribute to a reduction in the critical cavity size as well as make bubbles suddenly larger than their critical size, both of which would increase conversion of bubbles to voids. The coarse M_6C phase particles which develop at 500°C should also directly enhance the growth rate of attached voids by acting as point defect collectors [14, 15]. Furthermore, these M_6C particles in SA 316 may preferentially absorb interstitials to increase the net bias of the material, further increasing swelling. The XEDS shows that the M_6C at 500°C is modified by RIS, with slightly more Ni and Si, and much less Mo than at 600°C. An interstitial bias for these particles has been suggested on the basis of their strong chemical affinity for Si and Ni which migrate interstitially when RIS is intense [11]. The microstructure observed in SA 316 at 500°C after ~30 dpa suggests that swelling will continue to increase at higher dose.

At 600°C, few Frank loops remain in the SA 316 matrix, and some of the largest voids are found at climbing dislocations. Increased bulk recombination with temperature would reduce long-range migration of interstitials to reduce loop nucleation. Moreover, bubble nucleation decreases and growth increases at higher temperatures, leading to a coarser microstructure, particularly if bubbles nucleate along the reduced concentration of network dislocations. Together, these changes in bubble density would lead to a large decrease in void density. Even though voids are larger at 600°C, there are so few that swelling is less at 500°C.

4.2 CW Material

Swelling in CW 316 is low and does not depend much on temperature. At 300 and 400°C, cavity behavior is similar in SA and CW material [Fig. 1(a–d)]. Both have a high concentration of fine helium bubbles in the matrix, and swelling is minimal. However, SA and CW materials behave differently at 500 and 600°C. Many small cavities (below 10 nm in diameter) are uniformly dispersed along the dislocation lines in CW 316. The high density of dislocations from cold work provide more bubble nucleation sites, since most subcritical bubbles are observed to be attached to dislocations. Calculations to account for partitioning of helium atoms suggest that most of these small cavities are equilibrium bubbles, not voids. The high concentrated of helium bubbles could then act as strong sinks to increase the critical cavity size and thus suppress their conversion into voids. These same bubble sinks would also dilute RIS [14] to produce enhanced thermal M_6C rather than the modified M_6C observed in SA 316 at 500°C. This change may further contribute to reduced swelling in the CW 316.

Finally, data in Fig. 4 show that swelling varies from heat to heat for 20% CW 316 stainless steels irradiated in HFIR to below 30 dpa (cross-hatched region). The data suggest, however, that for a swelling-resistant heat of 316 stainless steel, 20% CW can effectively suppress swelling to below 0.5% during HFIR irradiation at 600°C to 36 dpa.

5. Conclusions

Solution-annealed and 20% CW type 316 stainless steels have been irradiated in HFIR to 36 dpa and 2327 appm He at temperatures of 300 to 600°C. Cavity development was examined by transmission electron microscopy. The conclusions are as follows:

1. Swelling in solution-annealed 316 stainless steel depends strongly on temperature, with a maximum at 500°C.

2. Swelling in 20% CW material, increases slightly with increasing irradiation temperature, but is much lower than solution-annealed material, particularly at 500 and 600°C.

3. 20% CW is more effective at extending the incubation time for swelling resistance at temperatures at 500°C and above.

4. The temperature dependence of void swelling in both SA and CW 316 correlates well with differences observed in cavity, precipitate, and dislocation microstructures.

5. Swelling data on heat-to-heat variations of type 316 stainless steel suggest that swelling in 20% CW type 316 stainless steel is less than 0.5% when irradiated in HFIR to ~36 dpa at 600°C and below. Both SA and CW 316 are resistant to void swelling at 400°C and below.

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Table 1. Chemical Composition of Type 316 Stainless Steel

Content, wt %												
C	Si	Mn	P	S	Ni	Cr	Mo	Ti	Nb	B	N	Co
0.058	0.61	1.80	0.028	0.003	13.52	16.75	2.46	0.005	<0.1			<0.1

Table 2. Data on the Chemical Composition of Extracted Particles

Position Analyzed	Composition, wt %								
	Fe	Ni	Cr	Mn	Ti	Si	P	Mo	V
400°C (d)	15.61	2.09	63.15	2.40	0.05	1.64	0.52	6.24	1.97
500°C (e)	18.29	31.51	30.56	0.55	0.05	9.75	1.03	6.84	1.06
600°C (f)	10.37	28.61	28.81	0.02	0.12	8.27	1.07	20.37	2.01

Table 3. Microstructural Data on Alloy 316 Irradiated in HFIR

Heat Treatment	Irradiation Condition			Data						
				Cavity			Precipitate		Loop	
	Temperature (°C)	Damage (dpa)[a]	Helium Content (appm)	Diameter (nm)	Concentration (m^{-3})	Swelling[b] (%)	Diameter (nm)	Concentration (m^{-3})	Diameter (nm)	Concentration (m^{-3})
SA[c]	300	33.0	2116	1.9	3.9×10^{23}	0.13			10.3	1.3×10^{22}
CW[d]	300			2.0	3.7×10^{23}	0.15			10.4	1.5×10^{22}
SA	400	33.4	2142	2.3	3.2×10^{23}	0.22			16.0	2.5×10^{22}
CW	400			2.3	2.3×10^{23}	0.14			11.6	4.5×10^{22}
SA	500	34.0	2187	13.3	8.9×10^{21}	1.09[e]	37	2.4×10^{21}	21.1	1.2×10^{20}
CW	500			6.1	1.3×10^{22}	0.16			29.3	3.5×10^{20}
SA	600	36.1	2327	33.9	4.8×10^{20}	1.00	160	2.3×10^{19}		
CW	600			8.8	8.2×10^{21}	0.29				

[a]Displacements per atom.

[b]Swelling calculated from cavity volume fraction.

[c]Solution annealed.

[d]Cold worked.

[e]Estimation of all these cavities would lead to higher swelling of 5 to 10%.

LIST OF FIGURES

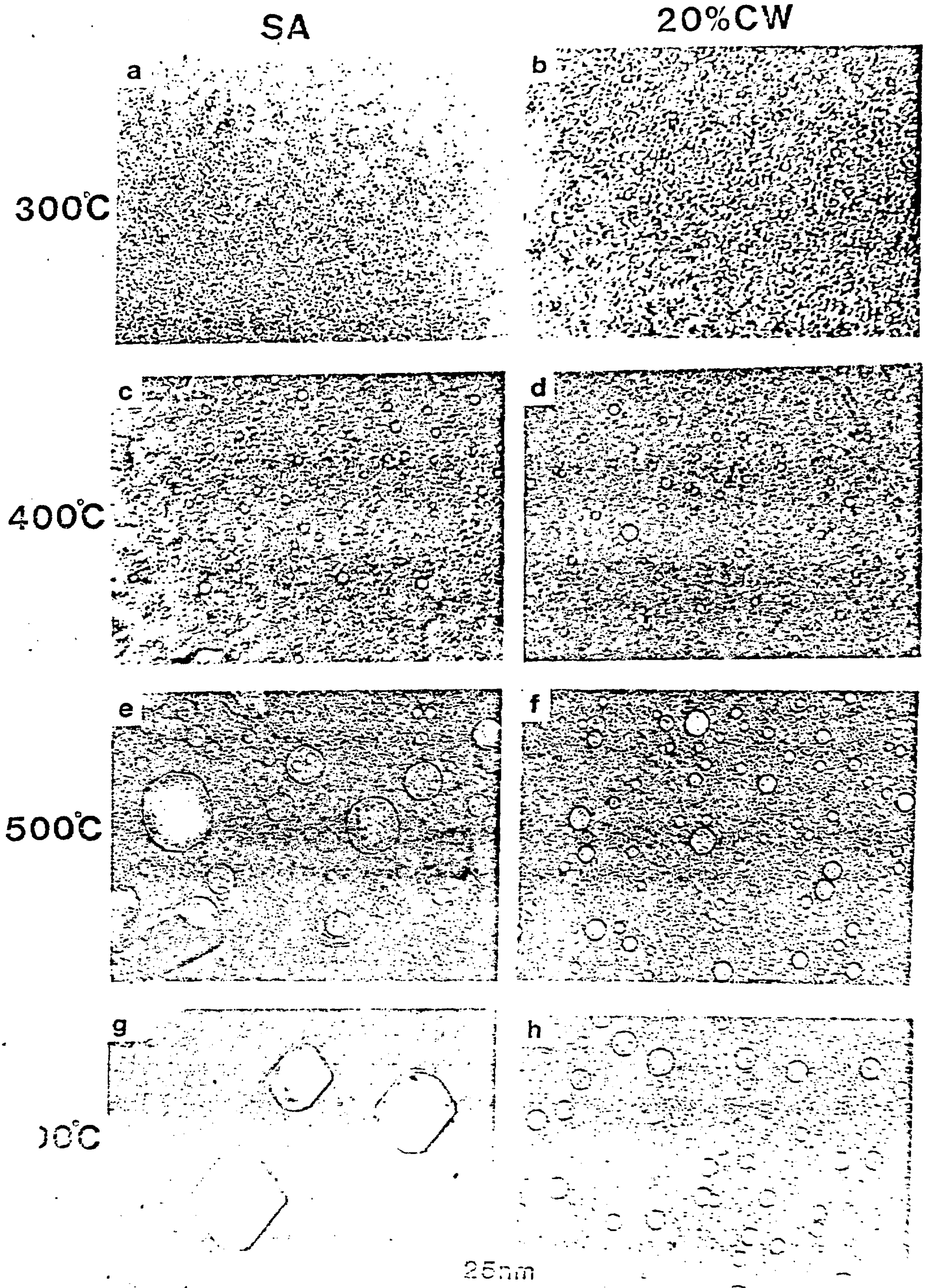

Fig. 1. A comparison of cavities for SA (a,c,e,g) and 20% CW (b,d,f,h) type 316 stainless steel irradiated in HFIR at 300 to 600°C up to 36 dpa and to 2327 appm He.

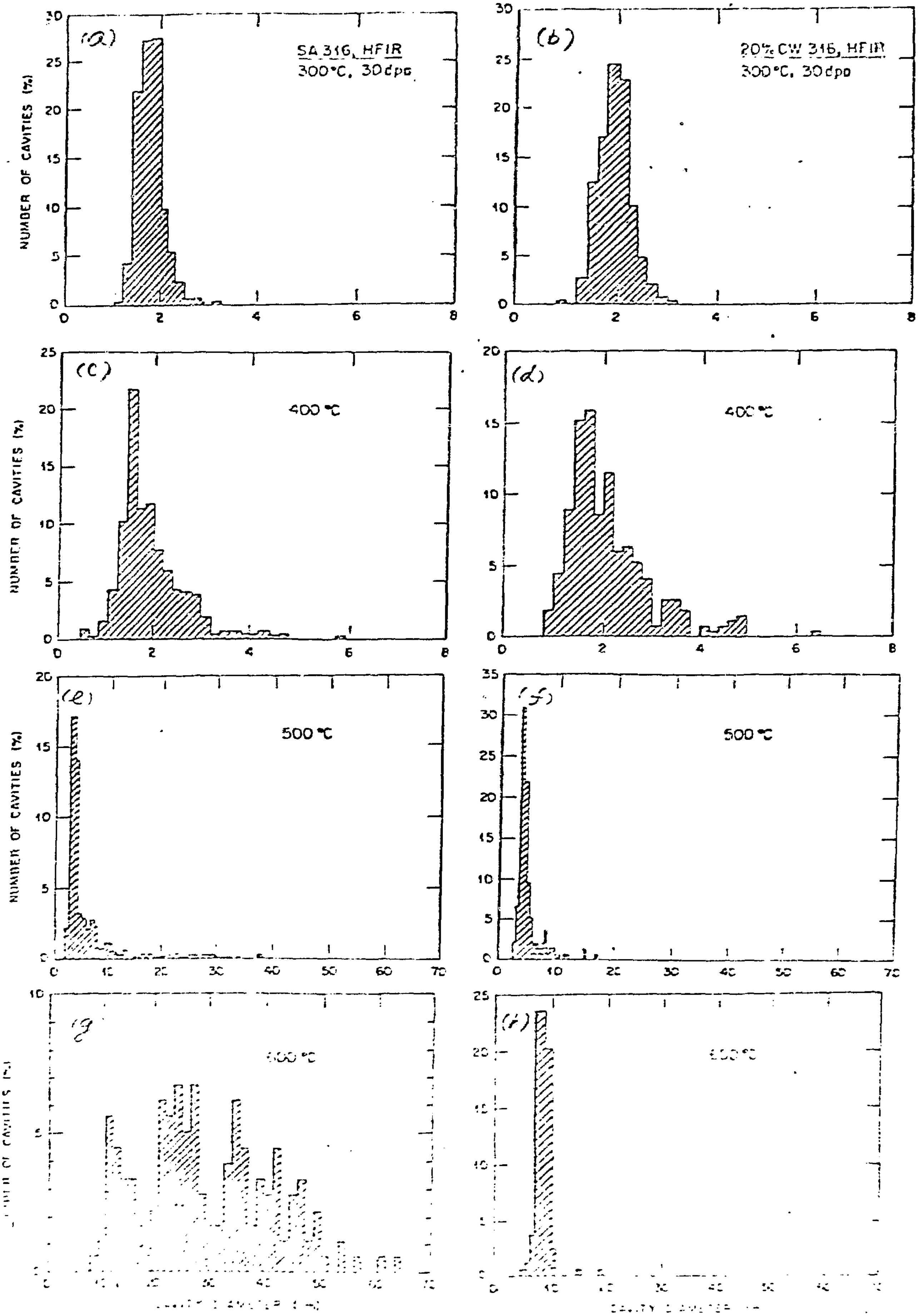

Fig. 2. Histograms of cavity size distribution in SA (a,c,e,g) and 20% CW (b,d,f,h) type 316 stainless steel irradiated up to 36 dpa in HFIR at 300 to 600°C.

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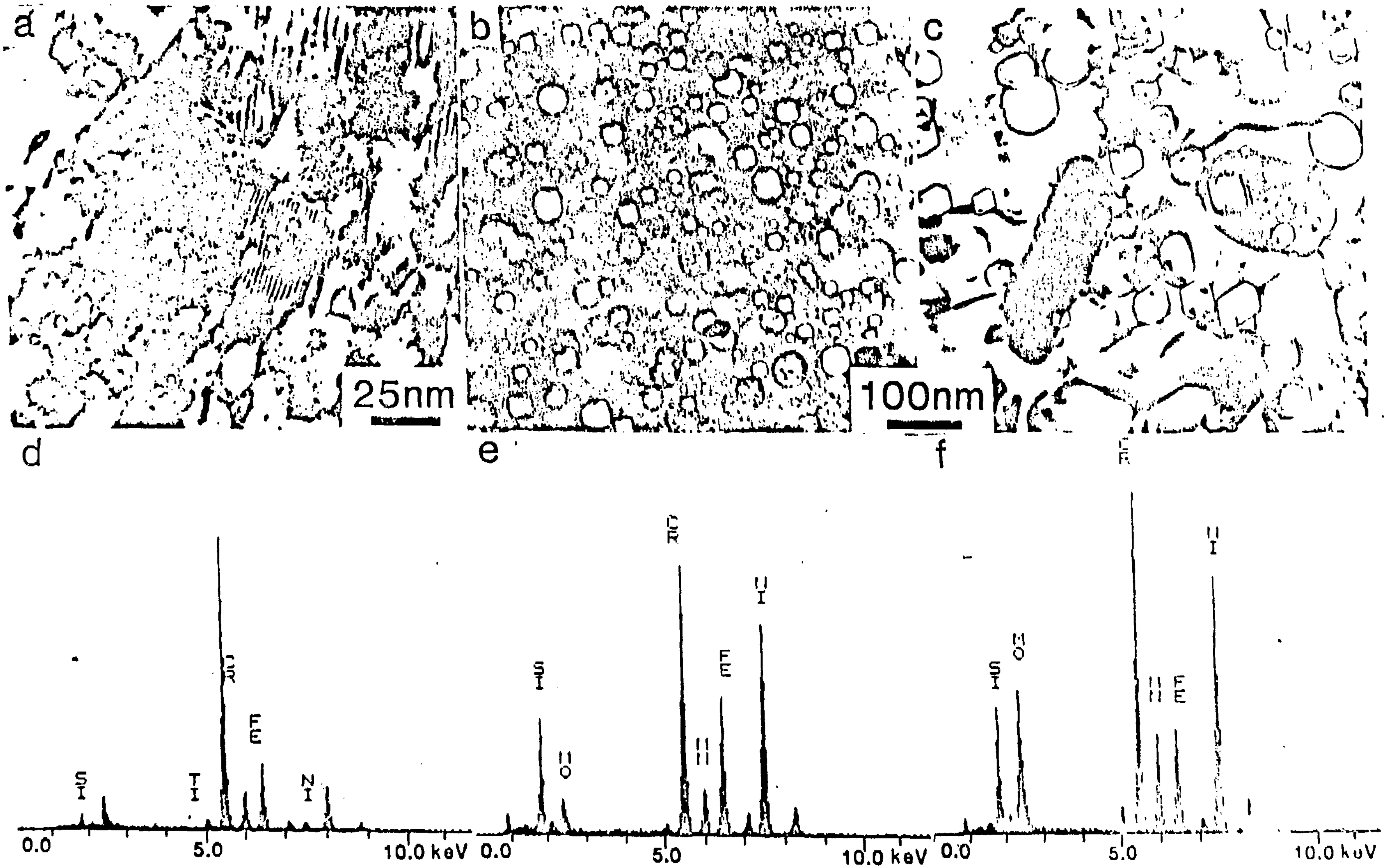

Fig. 3. Precipitates and X-ray spectrum from extracted particles in SA 316 irradiated in HFIR. (a,d) 400°C, 33.4 dpa; (b,d) 500°C, 34.0 dpa; and (c,e) 600°C, 36.1 dpa.

c

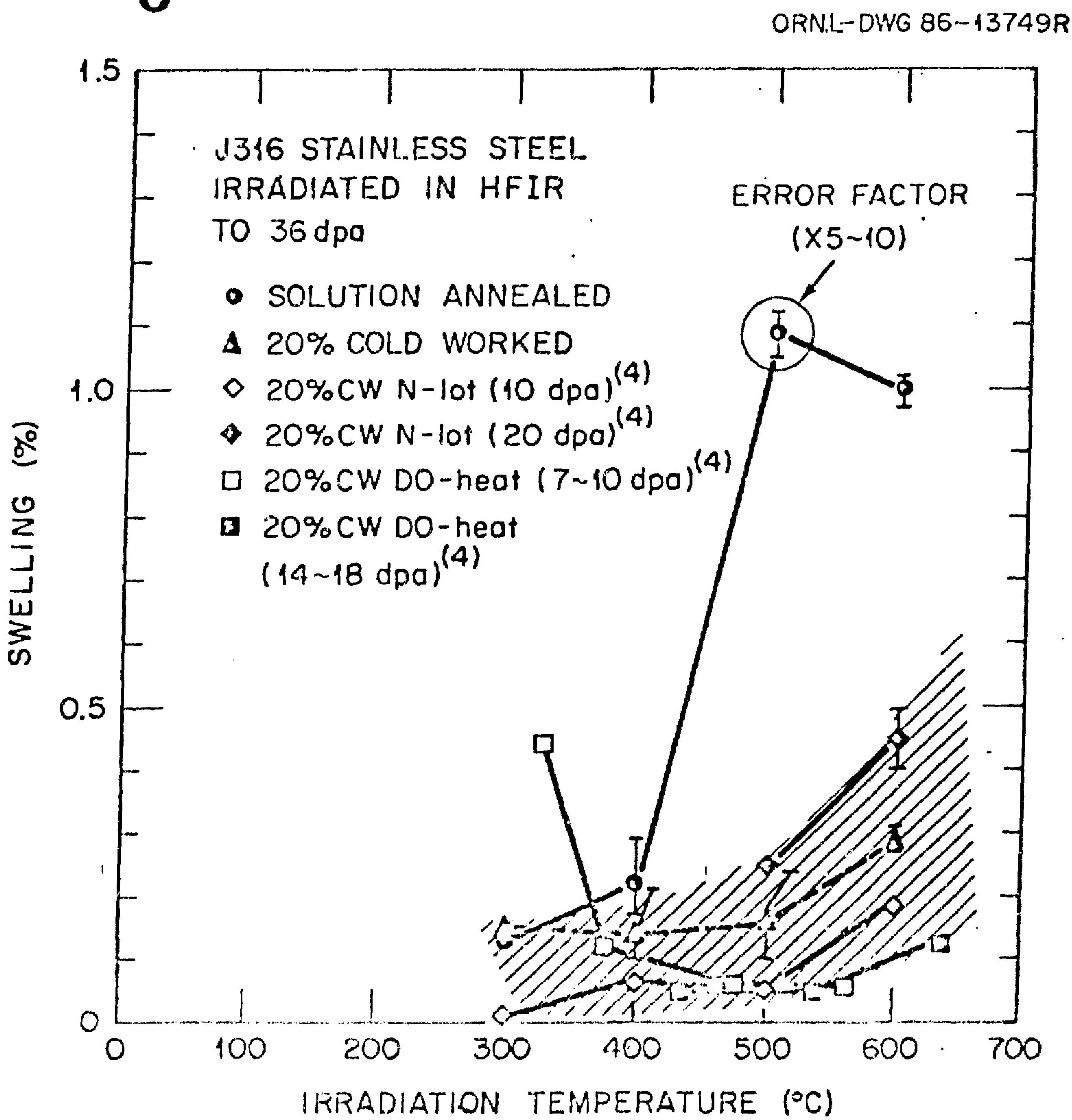

Fig. 4. Cavity swelling plotted as functions of irradiation temperature for SA and 20% CW type 316 stainless steel irradiated in HFIR at 300 to 600°C up to 36 dpa and to 2327 appm He. Data indicated with ref. 4 were reported by Maziasz and Braski.