Factors Which Determine the Swelling Behavior of Austenitic Stainless Steels

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Once void nucleation subsides, the swelling rate of many austenitic alloys becomes rather insensitive to variables that control the transient regime of swelling. Models are presented which describe the roles of nickel, chromium and silicon in void nucleation. The relative insensitivity of "steady-state" swelling to temperature, displacement rate and composition is also discussed.

1. INTRODUCTION

Neutron-induced swelling of austenitic alloys can be characterized by a transient regime followed by a regime of essentially constant swelling rate. The latter regime is often misdesignated as "steady-state". In some alloys, the transient is rather abrupt, but in others the transient regime persists, yielding curvature to swelling levels of 20% or more[1]. Recently there has emerged a reasonably coherent description of the major compositional, fabricational and environmental factors which influence swelling. This paper reviews our conception of these factors, progressing from simple alloys with uncomplicated irradiation histories to more complicated alloys and histories.

2. SIMPLE FE-NI-CR TERNARY ALLOYS

Johnston and coworkers first demonstrated with self-ions the sensitivity of swelling to nickel and chromium levels[2,3]. As shown in Figure 1a the primary influence of nickel lies in the duration of the transient regime. While it appears that the post-transient swelling rate is influenced to a lesser degree, it has recently been shown that this is an illusion which arises from the previously unsuspected distortion of swelling when determined by step-height measurements[4]. It was also shown that self-ion irradiations are dominated by injected interstitials, which distort the temperature and compositional dependence of swelling relative to that of neutron irradiations[4,5].

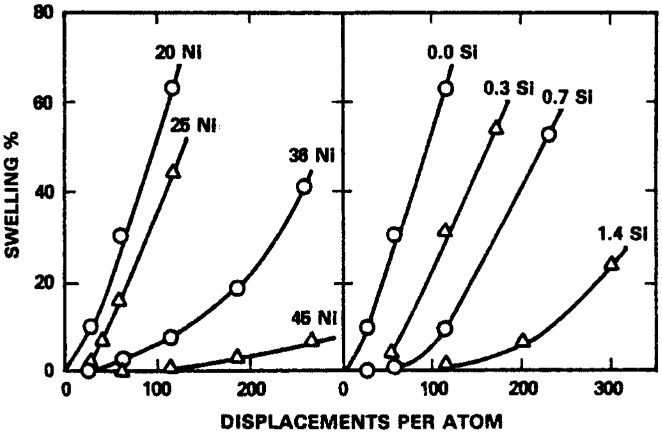

FIGURE 1

Ion-Induced Swelling of Fe-15Cr-XNi and Fe-15Cr-20Ni-XSi Alloys at 675°C.[2,3]

Neutron irradiations of Fe-Ni-Cr alloys in EBR-II have shown that nickel and chromium influence void nucleation and therefore the duration of the transient regime[6,7]. At a given temperature, however, the saturation void density is rather insensitive to composition. Unlike ion irradiations, neutron-induced post-transient swelling rates are ∿1%/dpa, relatively insensitive to both composition and temperature[8,9] as shown in Figure 2. For each alloy there is a break-away temperature above which the transient is temperature-sensitive (see Figure 3). The breakaway temperature itself is a function of nickel and chromium level. The longest transients occur at ~45% Ni for Fe-15Cr-XNi alloys and are even longer at lower chromium levels.

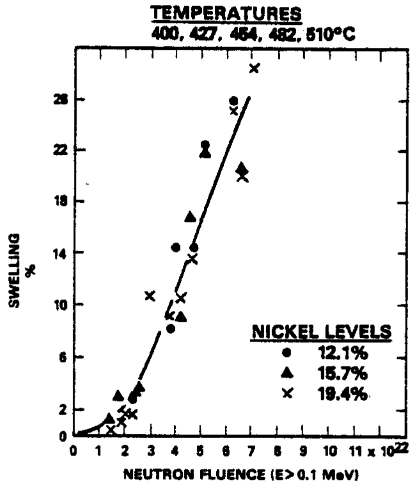

FIGURE 2

In Some Fe-15Cr-XNi Ternaries the Swelling in EBR-II is Relatively Independent of Nickel Level and Temperature Between 400 and 510°C.[8] (See Figure 3 for additional data at 25% Ni.)

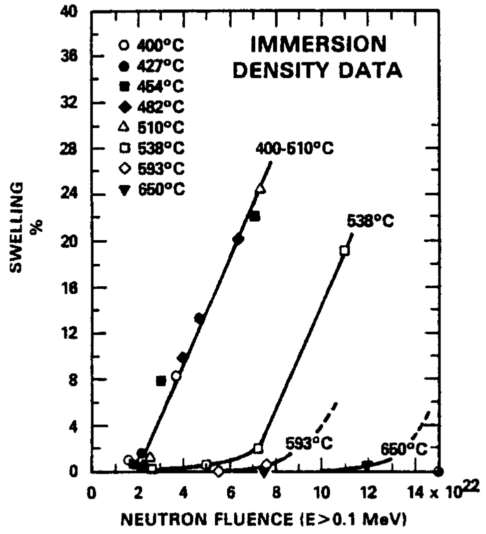

FIGURE 3

Swelling of Fe-15Cr-25Ni in EBR-II at 400-650°C.[4]

A model was proposed earlier to explain some of these observations[6,10]. While many properties of ternary alloys vary slightly with composition, the model concentrated on the compositional dependence of the void bias and the impact on the bias of the nickel segregation always observed at void surfaces. The model did not include composition-dependent dislocation biases since no data are available showing sensitivity of dislocation evolution to nickel and chromium levels. Moving dislocations also cannot segregate major elements as efficiently as can stationary voids.

Investigation of the diffusion of vacancies and interstitials in superimposed strain and composition gradients shows that drift terms arise in the void bias description as a result of segregation. These terms originate from the compositional dependence of elastic properties and lattice parameter. When compounded with forces arising from the inverse Kirkendall segregation mechanism, these terms substantially alter the void bias, providing a qualitative explanation for the dependence of swelling on nickel and chromium. At low chromium and high nickel levels, where voids have difficulty nucleating, loss of chromium and segregation of nickel at surfaces of the first voids not only stabilizes them against dissolution but changes the matrix composition, thus making it easier for other voids to nucleate.

Two features of this model are particularly attractive. First, it offers a rationale for the minimum in swelling observed at 35%-45% nickel[2,3]. A reversal in sign for the Kirkendall force is experienced by vacancies when the matrix nickel level is ~35%, as calculated for Fe-Ni alloys using published diffusion and thermodynamic data. Below 35% vacancies are attracted into regions of higher nickel concentration while the opposite occurs above 35%. Second, the dependence of shear modulus on nickel and chromium at void-relevant temperatures is consistent with the compositional dependence of swelling[7]. Segregation-induced increases in shear modulus at void surfaces reduce the attractive force on interstitials and thus help stabilize voids. For ternary alloys with 15% chromium, nickel segregation increases the modulus, but at 7.5% chromium there is no appreciable change in modulus with nickel segregation[11]. This extends the transient relative to alloys with 15% chromium.

This model implies a compositional sensitivity of both transient and post-transient swelling. Since only the former is observed this suggests that segregation effects are second order in magnitude (compared to temperature and displacement rate) for large voids and primarily influence void nucleation. However, there is another second order effect that has not been previously considered. Small increases (≤100%) in the effective vacancy diffusion coefficient D^v_{eff} have been shown to strongly decrease the void nucleation rate, particularly at higher temperatures[12]. In general, the addition of nickel to Fe-Ni[13], Fe-Ni-Cr[14] and Fe-Ni-Cr-Mo[15] alloys increases the diffusivity of all alloy components, while additions of chromium decrease diffusivities[14,15]. For example, D^v_{eff} increases ~100% in going from Fe-15Cr-20Ni to Fe-15Cr-45Ni[14]. At temperatures where void nucleation begins to become difficult, the combined influence of these two second order compositional effects explains much of the observed swelling behavior of Fe-Ni-Cr alloys.

3. SILICON-DOPED Fe-Ni-Cr ALLOYS

As shown in Figure 1b, silicon's primary influence on swelling is also to extend the transient but it is much more effective per atom than nickel, however. It suppresses nucleation at all void-relevant temperatures[16] but the suppression is only temporary. As shown in Figure 4, Singh and coworkers have observed that void densities eventually converge at a level which is relatively insensitive to silicon content[17].

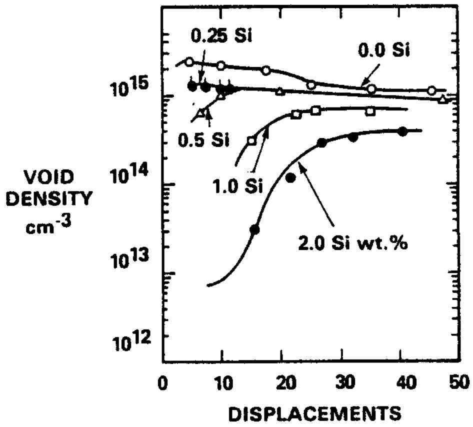

FIGURE 4

Silicon Effect on Electron-Induced Void Density in Fe-15Cr-13Ni-0.9Mn at 700°C.[17]

The model[12] developed to explain silicon's role in pure metals is based on its higher diffusivity compared to that of the host metal and thus its effect on increasing D^v_{eff}. In addition, the diffusivities of all components of Fe-Ni-Cr alloys increase substantially upon small additions of silicon[14,18,19] as shown in Figure 5. Rothman[14] found that the diffusivities in Fe-15Cr-20Ni increased roughly 45% when 1.4 wt.% silicon was added. D^v_{eff} increases even more than 45% when silicon's diffusivity contribution is included. The enhanced diffusivity of silicon-doped alloys strongly decreases void nucleation, particularly at higher temperatures.

Silicon is also a smaller atom in Fe-Ni-Cr alloys[20,21] and is proposed in this model to form interstitial-solute pairs. Dimitrov and coworkers[22] have recently demonstrated strong interstitial-silicon trapping in neutron-irradiated Fe-16Cr-25Ni. Silicon will thus not only drift down interstitial gradients but also

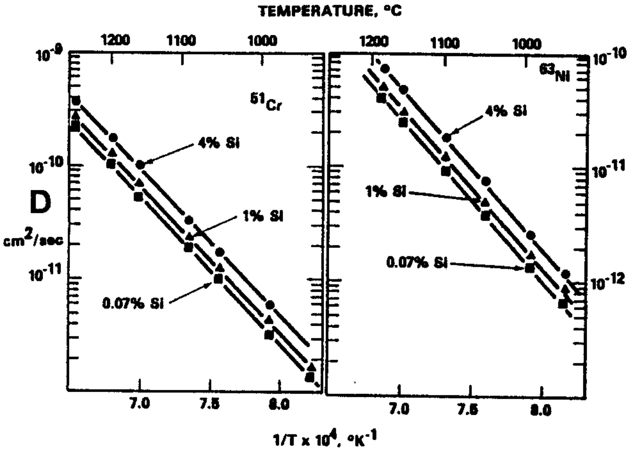

FIGURE 5

Increase in Tracer Diffusivities of Cr and Ni with Silicon Content in Fe-16Cr-14Ni.[18]

up vacancy gradients, both of which exist at microstructural sinks. The combined fast-diffusion/interstitial binding allows silicon segregation to proceed at sinks biased toward interstitials but dispenses with the requirement of unrealistically large binding energies needed for solute binding alone to suppress swelling. Nickel segregates to the same sinks by the inverse Kirkendall mechanism.

Addition of silicon inevitably causes irradiation-induced segregation of nickel and silicon into precipitates. The removal of sufficient amounts of these nucleation-controlling elements largely determines the duration of the transient. For each silicon-doped alloy there appears to be threshold levels of nickel and silicon that must be reached in the matrix before nucleation proceeds[23,24].

4. MORE COMPLEX Fe-Ni-Cr ALLOYS

Addition of C, P, Mo, Mn, or Ti to Fe-Ni-Cr-Si alloys allows other phases to develop during irradiation[25]. Most, but not all, of these phases concentrate nickel and silicon, and each responds to fabricational and environmental variables in a different manner[25-29].

The combined sensitivity of the microstructural and microchemical evolutions to the many operating variables has been judged to be too complicated to model. A correlation has been established, however, between the history-sensitive phase evolution and the history dependence of transient swelling. The sensitivities of swelling to neutron flux and fluence, temperature history, stress and preirradiation thermal-mechanical treatment are mirrored in the sensitivities of the nickel-silicon removal process[26]. Details of phase identity do not appear to be as significant as are their consequences on matrix composition.

It has been shown that the eventual swelling rate of several 300 series stainless steels is ∿1%/dpa over a wide range of temperature[1,30]. While the transient regime may be sensitive to temperature for some heats of steel, others are relatively insensitive to temperature. At temperatures outside the temperature-insensitive range the transients are longer but eventually the same swelling rate is reached . Applied stresses and some temperature changes can accelerate phase formation and lead to an abrupt truncation of the transient, however. Under these conditions rates of ∿1%/dpa can be reached much quicker than is typical for isothermal or stress-free histories[23,28,29].

5. DISCUSSION

The influence of environmental, fabricational and compositional variables seems to reside in their effect on the transient regime. In addition, their action primarily involves void nucleation and is largely chemical in nature. When radiation-induced segregation is sluggish the transient regime exhibits much curvature. The eventual swelling rate appears not to be very sensitive to composition or environmental variables such as displacement rate[31,32]. This implies that the temperature, composition and displacement rate sensitivities of microstructure are not as instrumental in determining the swelling rate as previously envisioned. A rationale for this insensitivity can be demonstrated using rate theory.

The swelling rate contains bias-driven and void annealing contributions[33]. The latter can be ignored if the temperature is below $0.6T_m$ or if the voids are relatively large or gas-pressurized. Thus for a system containing primarily large voids and dislocations,

$$\frac{d}{dt}\ \frac{\Delta V}{V} = \frac{S_o S_d}{(S_o + S_d)^2} \cdot \frac{Z_i^d Z_v^o - Z_v^d Z_i^o}{\overline{Z}_v} \cdot F(P,T) \quad (1)$$

The first right-hand term contains only void and dislocation sink strengths. While this term depends strongly on temperature for a microstructure dominated by one sink type, high fluence microstructures in austenitic alloys are characterized by a balance of sink strengths. Since both S_o and S_d possess similar temperature dependencies, $S_o(T) \approx S_d(T)$ over a large temperature range. If this condition is assumed, the first term equals 0.25 independent of temperature.

The second term is the system net bias and depends only weakly on temperature. The last term, however, potentially contains the major influence of temperature and displacement rate:

$$F(P,T) = \frac{S^2}{2\lambda}\left[\sqrt{\alpha^2 + \beta} - \alpha\right] \quad (2)$$

where

$\lambda = 4\pi R_c/\Omega = 32\ \pi a_o^2$, $\alpha = 1 + \lambda \overline{C}_v^{eq}/(S\overline{Z}_i)$

and $\beta = 4\lambda D_v P/S^2 \overline{Z}_i \overline{Z}_v$.

S is the total sink strength, R_c the recombination radius $2a_o$, a_o the lattice parameter, $\overline{C}_v^{eq}$ the average vacancy concentration in equilibrium with all sinks, Z_i and Z_v are bias factors for interstitials and vacancies, $\overline{Z}$ the average bias factor for the system, Ω the atomic volume, D_v the vacancy diffusivity and P the defect production rate. If we divide Eqn. (1) by P, we obtain the fractional swelling rate per dpa. Since the first two terms of Eqn. (1) are relatively insensitive to temperature, only F/P is plotted in Figure 6 to show the sensitivity of swelling to temperature and sink strength.

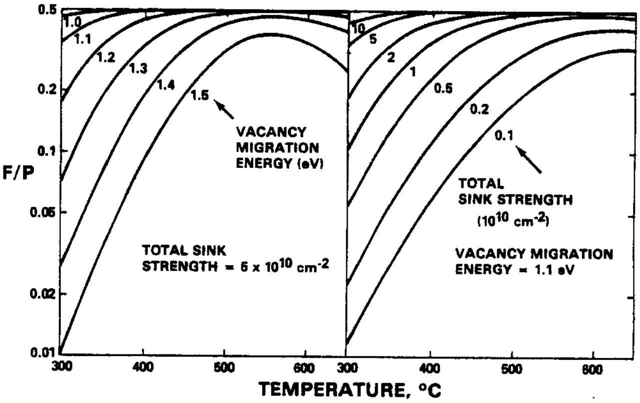

FIGURE 6

Sensitivity of Swelling Rate Parameter F/P to Temperature, Sink Strength and E_v^m.

The activation energy for self-diffusion in nickel is known to be 2.9 eV, but the formation and migration energies that contribute to the total are not as well characterized. The value of E_v^m (and hence also E_v^f) has been revised recently[34,35]. Whereas the accepted migration energy was 1.4 eV, it now appears to be 1.1 ± 0.1 eV. Fig. 6 shows that a strong dependence on temperature occurs when E_v^m is 1.4 eV but not for 1.1 eV. Low values of sink strength also yield predictions of strong temperature dependency. In Fe-Ni-Cr alloys, however, the high fluence sink strengths ($>5 \times 10^{10}$ cm^{-2}) are sufficiently large that the swelling rate is insensitive to temperature and second-order composition effects on diffusivity.

One consequence of this treatment is that we expect a similar relative insensitivity of the eventual swelling rate to both displacement rate and helium/dpa ratio as was observed in ref. 31 and 36. This treatment decouples many of the sensitivities of post-transient swelling from the sensitivities of composition and microstructure, providing both void and dislocation sink strengths are sufficiently high.

REFERENCES

1. F.A. Garner, Recent Insights on the Swelling of Irradiated Austenitic Alloys, Proc. 3rd Topical Mtg. Fusion Reactor Materials.
2. W.G. Johnston et al., J. Nucl. Mater., 54 (1974) 24.
3. J.F. Bates and W.G. Johnston, Proc. AIME Conf. on Radiation Effects In Breeder Reactor Structural Materials, June 1977, Scottsdale, AZ, 625.
4. F.A. Garner, J. Nucl. Mater., 117, (1983) 177.
5. D.L. Plumpton, H.H. Attaya and W.G. Wolfer, Proc. 3rd Top. Mtg. on Fusion Reactor Matls.
6. W.G. Wolfer, F.A. Garner, and L.E. Thomas, ASTM STP 782, (1982) 1023.
7. H.R. Brager and F.A. Garner, DAFS Quarterly Report DOE/ER-0046/11, (Nov. 1982) 221.
8. F.A. Garner, DAFS Quarterly Report DOE/ER-0046/14, (Sept. 1983) 133.
9. H.R. Brager and F.A. Garner, DAFS Quarterly Report DOE/ER-0046/14, (Sept. 1983) 152.
10. W.G. Wolfer, J. Nucl. Mater., 114, No. 2&3, (1983) 292.
11. J.F. Bates, J. Nucl. Mater., 87 (1979) 409.
12. F.A. Garner and W.G. Wolfer, J. Nucl. Mater., 102 (1981) 143.
13. B. Million et al., Mat. Sci. and Engr., 50 (1981) 43.
14. S.J. Rothman, L.J. Nowicki and G.E. Murch, J. Phys. F, 10 (1980) 383.
15. L.V. Pavlinov, Fiz. Metal. Metalloved. 41, No. 2, (1976) 344.
16. H.R. Brager and F.A. Garner, Proc. AIME Symp. on Phase Stability During Irradiation, Pittsburgh, PA, October 1980, 219.
17. B.N. Singh et al., J. Nucl. Mater., 103 & 104 (1981) 1041.
18. W. Assassa and P. Guiraldenq, Metal Science (1978) 123.
19. G.B. Fedorov, A.N. Semenikhin, Metallurgy and Metallography of Pure Metals Moscow, (1960) Vol II, 252.
20. J.L. Straalsund and J.F. Bates, Met. Trans., 5 (1974) 493.
21. J.F. Bates, Partial Molar Volumes and Size Factor Data for the Fe-Ni-Cr-Si System, HEDL-SA-2310 (1980).
22. C. Dimitrov, M. Tenti and O. Dimitrov, J. Phys. F, 11 (1981) 753.
23. F.A. Garner, Ref. 16, 165.
24. L.E. Thomas, 40th Ann. Proc. Electron Microscopy Soc. Amer., Wash. D.C., G.W. Bailey (ed.) 1982, 597.
25. W.S.J. Yang, H.R. Brager and F.A. Garner, ref. 16, 257.
26. F.A. Garner and D.L. Porter, ref. 6, 295.
27. H.R. Brager and F.A. Garner, ASTM STP 683, (1978) 207.
28. F.A. Garner, DAFS Quarterly Report DOE/ER-0046/5 (May 1981) 198.
29. W.S.J. Yang and F.A. Garner, ref. 6, 186.
30. F.A. Garner, DAFS Quarterly Report DOE/ER-0046/12 (Feb. 1983) 178.
31. J.L. Seran and J.M. Dupouy, ref. 6, 5.
32. J.L. Seran and J.M. Dupouy, Proc. Conf. on Dimensional Stability and Mechanical Behavior of Irradiated Metals and Alloys, Brighton, England (April 1983) Vol. I, 25.
33. W.G. Wolfer, L.K. Mansur and J.A. Sprague, (ref. 3), 841.
34. S.K. Khanna and K. Sonnenberg, Radiation Effects 59 (1981) 91.
35. L.C. Smedskjaer et al., J. Phys. F, 11 (1981) 2221.
36. H.R. Brager and F.A. Garner, J. Nucl. Mater., 117 (1983) 159.