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ABSTRACT: When alloys based on the Fe-Ni-Cr system are subjected to irradiation at high temperatures a substantial amount of elemental segregation occurs. Two categories of segregation have been observed, one involving radiation-induced precipitation or changes in precipitate composition, and another involving the establishment of compositional gradients near microstructural sinks. While segregation into precipitate phases is known to strongly influence the development of void swelling, it now appears that segregation to void surfaces plays an equally important role in the development of voids. A general feature of both types of segregation is the major role of nickel as a primary segregant.

An investigation of the basic flux equations for diffusion of interstitials and vacancies in superimposed strain and composition gradients shows that several previously unconsidered drift terms arise in the description of the bias as a result of elemental segregation. These terms originate from the compositional dependence of point defect formation and migration energies and the compositional dependence of elastic properties and lattice parameter. When compounded with the operation of the inverse Kirkendall segregation mechanism, these terms strongly alter the bias for void nucleation and growth and provide an explanation for the marked variation of void nucleation and swelling with nickel concentration. The segregation-induced drift terms are comparable in magnitude to the strain-induced drift terms and are strongly dependent on nickel concentration. This is demonstrated for binary Fe-Ni alloys using published tracer diffusion and thermodynamic data.

KEY WORDS: radiation effects, void swelling, segregation, diffusion, vacancy, interstitial, alloys

Radiation-induced segregation (RIS) has been found to occur in many alloys during neutron or charged-particle irradiation. A survey of the progress in our understanding of this phenomenon can be found in the proceedings [1,2] of two recent conferences. Although many elements have been observed to participate, silicon and nickel are particulary effect-

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ive segregants in austenitic alloys. The significance of RIS to void formation and swelling has been recognized by many researchers, although the precise role of the various processes involved is still a matter of active research.

Okamoto et al. [3] have shown that RIS may be associated with point defect trapping by impurity or alloying elements, and that this trapping modifies the recombination of interstitials and vacancies. As a result, void swelling can be reduced by either vacancy or interstitial trapping, although the latter would require a very large binding energy between the interstitial and the solute atom.

The vacancy-solute binding can also lead to a change of the vacancy diffusivity and thereby affect the void nucleation rate, as shown recently by Garner and Wolfer [4]. While the role of silicon in suppressing swelling can be ascribed at least partially to vacancy-solute binding, the role of nickel cannot, however.

When radiation-induced segregation results in precipitation, void formation and swelling can be strongly affected. Mansur et al. [5,6] have proposed that voids associated with incoherent precipitates possess a larger collecting surface for point defect absorption. As a result, radiation-induced precipitation may enhance void growth. Another consequence of radiation-induced precipitation is connected with the associated change in the matrix composition of the alloy. Garner [7] has pointed out that radiation-induced precipitation reactions in the austenitic steels generally involve the removal of nickel and silicon from the matrix and that the sensitivity of swelling to many variables is mirrored in the nickel and silicon removal processes [8]. The reduction in the matrix concentrations of these elements then leads to an increase in void swelling as demonstrated in the experimental findings of Johnston et al. [9].

Finally, RIS can affect both void nucleation and growth by modifying the bias factors of sinks. Wolfer and Mansur [10] have investigated this possibility in detail for voids surrounded by a discrete segregation shell whose elastic properties and lattice parameter differ from those of the matrix. In another paper, Si-Ahmed and Wolfer [11] show that very small differences in these parameters can change void nucleation rates by orders of magnitude.

In this paper, we report on recent investigations on nickel segregation to void surfaces. The associated compositional gradients can extend thousands of angstroms, and the segregation results in a continuous radial change in the concentration of various elements. Although the discrete shell model of Wolfer and Mansur does not provide an adequate description for this case, the effect of compositional gradients on the void bias is similar to that of the discrete shell.

It is well known that many physical properties, such as thermal expansion, elastic moduli, lattice parameter, and excess free energy, exhibit a marked variation with composition in the Fe-Ni-Cr austenitic alloys. In particular, so-called anomalous variations occur in these properties in the composition range of the Invar alloys, which happens to coincide with the composition range of low-swelling alloys. Accordingly, we suggest in this paper that a possible connection exists between these anomalous variations, the segregation of nickel to microstructural sinks, and the associated modifications of the sink bias factors. An unambiguous correlation cannot yet be made because critical information is lacking on the basic physical properties in this class of alloys. It will become evident, however, that nickel segregation combined with known changes of basic lattice properties will result in a strong modification of the bias, and therefore strongly affect void swelling. In view of this connection, the proposed explanation for the dependence of swelling on the alloy composition has a mechanistic basis that can be incorporated in rate and nucleation theories for void formation, and therefore can be subject to a quantitative examination. In contrast, previous explanations of the compositional variation of swelling are not considered satisfactory when examined in quantitative terms.

Watkins et al. [12] have proposed that in fcc Fe-Ni-Cr alloys, the iron atoms may exist in two states, an anti-ferromagnetic and a ferromagnetic one. These are postulated to differ in their atomic volumes and in their contributions to the elastic moduli of the alloy. With increasing nickel content, the ratio of anti-ferromagnetic to ferromagnetic iron atoms decreases with a corresponding increase in the lattice parameter and a decrease in the elastic shear modulus. They speculate that the increase of the average atomic volume may imply a greater recombination of interstitials and vacancies.

According to recent results by Wolfer and Si-Ahmed [13] the recombination rate at reactor irradiation temperatures is simply proportional to the lattice parameter. Figure 1 shows the lattice parameter of Fe-Ni-Cr-phase alloys at room temperature [14]. It is seen that the maximum variation with composition is no more than about 2%. Such a small variation in the recombination rate would not be sufficient to account for the much larger variation observed in the swelling as a function of nickel content.

Another explanation was advanced by Bates [15] based on the variation of the shear modulus with nickel composition. Figure 2 shows the results of Masumoto et al. [16] for

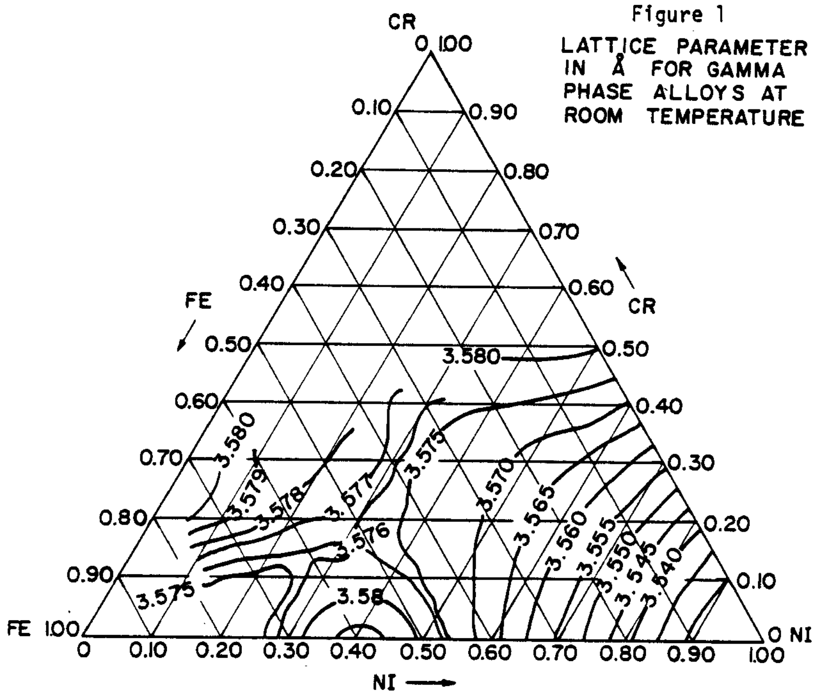


Figure 1

LATTICE PARAMETER IN Å FOR GAMMA PHASE ALLOYS AT ROOM TEMPERATURE

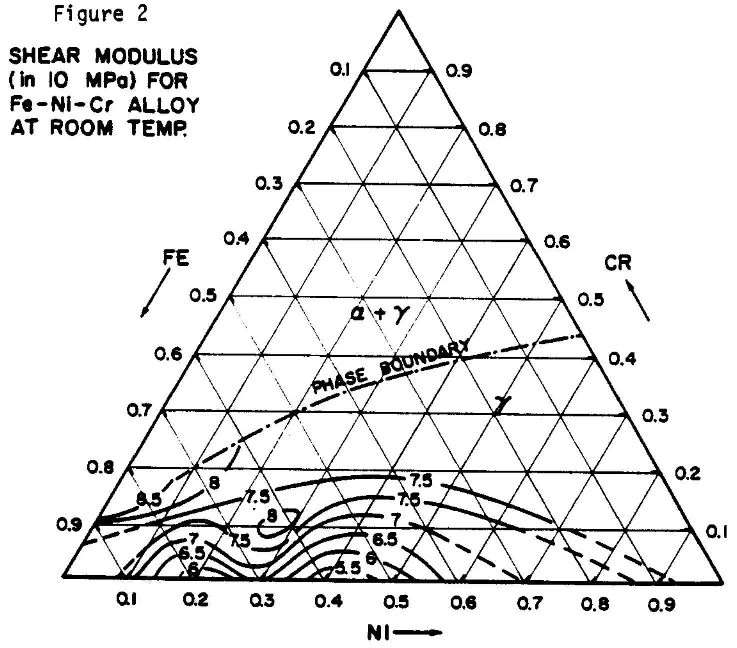


Figure 2

SHEAR MODULUS (in 10 MPa) FOR Fe-Ni-Cr ALLOY AT ROOM TEMP.

the shear modulus of Fe-Ni-Cr alloys at room temperature. Bates suggested that the arrival ratio of interstitials and vacancies at void embryos decreases with increasing shear modulus. Therefore, in this treatment the void nucleation rate would increase with the shear modulus.

This suggestion was based on a model proposed by Wolfer and Ashkin [17] for the capture efficiency of interstitials at a bare void. In this latter derivation, the image interaction of an interstitial with the void surface was not included. When this important interaction was included later by Wolfer and Yoo [18], it was found that the arrival ratio increased with increasing shear modulus. This would imply that the void nucleation rate increases sharply as the shear modulus drops to its minimum value for a binary Fe-Ni alloy with 40% nickel. In contrast, the data show that both void density and swelling exhibit minimums for this alloy composition [9].

It should further be noted that Bates did not consider the fact that the bias factor of dislocations also increases with the shear modulus, and that it is the net bias or bias difference between dislocations and voids which is the driving force for both void nucleation and growth.

In any case, homogeneous nucleation of voids without segregant shells was shown to occur with such low probability [11,18] that they cannot be invoked in any explanation of the composition dependence of swelling. In order for void nucleation to proceed at a rate commensurate with experimental findings, segregation to void embryos must precede their growth. As shown by Si-Ahmed, Wolfer and Mansur [10,11] this requires that segregation must result in a slight compositional change around the void embryo such that the shear modulus increases locally by a few percent and/or that the lattice parameter increases by a few tenths of a percent. Such minor segregation-induced changes are well within the range of the compositional variation of the shear modulus, Fig. 2, and that of the lattice parameter, Fig. 1. Other segregation-induced changes, to be discussed below, arise from the composition dependence of point defect diffusivities and the inverse Kirkendall effect. Prior to the discussion of these changes, however, it is useful to review the available data for clues to the nature of the influence of segregation on void nucleation and growth.

PUBLISHED EXPERIMENTAL EVIDENCE FOR NICKEL SEGREGATION TO VOIDS

Evidence has been accumulating which demonstrates radiation-induced segregation to void surfaces. Nickel segregation to voids in neutron-irradiated AISI 316 (with 12-13.5%

nickel and varying silicon levels) has been observed by Brager and Garner [19,20]. Porter and Wood [21] have found that in neutron-irradiated 304 stainless steel (9.3% nickel) voids are surrounded by an austenitic shell embedded in a ferritic matrix. While nickel was shown to segregate into precipitates it was inferred that nickel also segregated to voids. The combined segregation is thought to deplete the matrix to a nickle level below the limit for austenite stability, but the higher nickel content near the void surface resisted the austenite-to-ferrite transformation. Segregation of nickel to void surfaces was also found in titanium-modified AISI 316 by Brun et al. [22].

Whereas the above observations were made on neutron-irradiated steels, Terasawa et al. [23] found nickel, silicon and molybdenum segregation to void surfaces in carbon ion irradiations of AISI 316. Marwick et al. [24] discovered an enrichment of 54% nickel at voids in an alloy with an average nickel content of 34%. Hishinuma et al. [25,26] irradiated with 1.0 MeV electrons a series of alloys including both commercial steels and an experimental series Fe-17Cr-XNi, where X = 12, 35 and 50%. The researchers also observed that the void density obtained at 30 dpa for temperatures above 400°C decreased strongly with increasing nickel content.

Hishinuma et al. [15,26] further observed that voids in an Fe-17Cr-12Ni alloy exhibited a strain-field contrast in transmission electron microscopy, whereas those in Fe-17Cr-50Ni did not. The incubation times for void nucleation were very short for the low nickel alloy, and long for the high nickel alloy. Since the lattice parameter decreases with increasing nickel content particularly for high nickel alloys (see Fig. 1), the lack of a strain-field contrast around voids in the high nickel alloy was interpreted by Hishinuma et al. to indicate the absence of nickel segregation. Conversely, the strong strain-field contrast in the low nickel alloy was believed to be associated with substantial nickel segregation. It was therefore concluded by Hishinuma that the marked difference in swelling behavior was due to the difference in nickel segregation behavior at void embroys.

RECENT EXPERIMENTAL STUDIES ON NICKEL SEGREGATION

In order to obtain further insight on the nature of the segregation process, a series of microanalytical studies is being conducted at Hanford Engineering Development Laboratory using TEM/STEM methods, primarily energy dispersive X-ray microanalysis. The scanning transmission electron microscope with a field emission electron source operated at 100 kV allows X-ray microanalysis using probes as small as 1 to 2 nm. Beam broadening within the specimen controls the ultimate resolution, however. Measurements in a Vacuum Generators HB-501 STEM lead to an estimate of 5 nm spatial resolution in materials with low-to-medium atomic number [27].

While it is relatively easy to measure compositions by EDX microanalysis at different points on a thin specimen and to construct an apparent composition profile, it is not so easy to extract the actual composition profile around the void. This requires exact knowledge of the specimen thickness and void location in the foil as well as a sophisticated deconvolution of the data using the initial electron distribution in the probe. A discussion of the complexities involved is given in Ref. [27].

The degree of nickel segregation to voids has been found to be quite large even for relatively small voids. As shown in Fig. 3 the apparent nickel concentration at a precipitate-free void surface is on the order of 40%. (After deconvolution of the profile it is anticipated that the actual surface concentration will be much higher.) This void was found in neutron-irradiated Fe-15Cr-25Ni-0.3Si and was 35 nm in diameter and located at the center of a 55 nm thick foil. The probe diameter was less than 2 nm. Some silicon segregation at the void surface was also observed.

It is not necessary for silicon or other solutes to be present for nickel segregation to occur, however. Figure 4 shows the development of nickel segregation at void surfaces in neutron-irradiated Fe-15Cr-25Ni. The apparently lesser level of nickel segregation is primarily a reflection of the larger probe size (8 nm) employed.

A number of important observations relevant to the subject of this paper have been derived from these experimental studies.

a. The primary influence of matrix nickel content on swelling at low fluence is expressed in the void density rather than the size. The void density decreases strongly with increasing nickel content in Fe-15Cr-XNi alloys in the range of 15 to 35% nickel. However, with increasing fluence, differences in void number densities diminish.

b. When the matrix nickel content of a solute-bearing alloy is decreasing due to precipitation of nickel-rich phases, it is found that the matrix nickel content drops below a "critical" level prior to the nucleation of voids [28].

c. The segregation of nickel occurs in all nickel-bearing Fe-Cr-Ni alloys studied to date. This spans the range of 15-75% nickel and therefore covers the anomalous behavior range at about 35% nickel. Nickel segregation is not confined to voids alone, but also

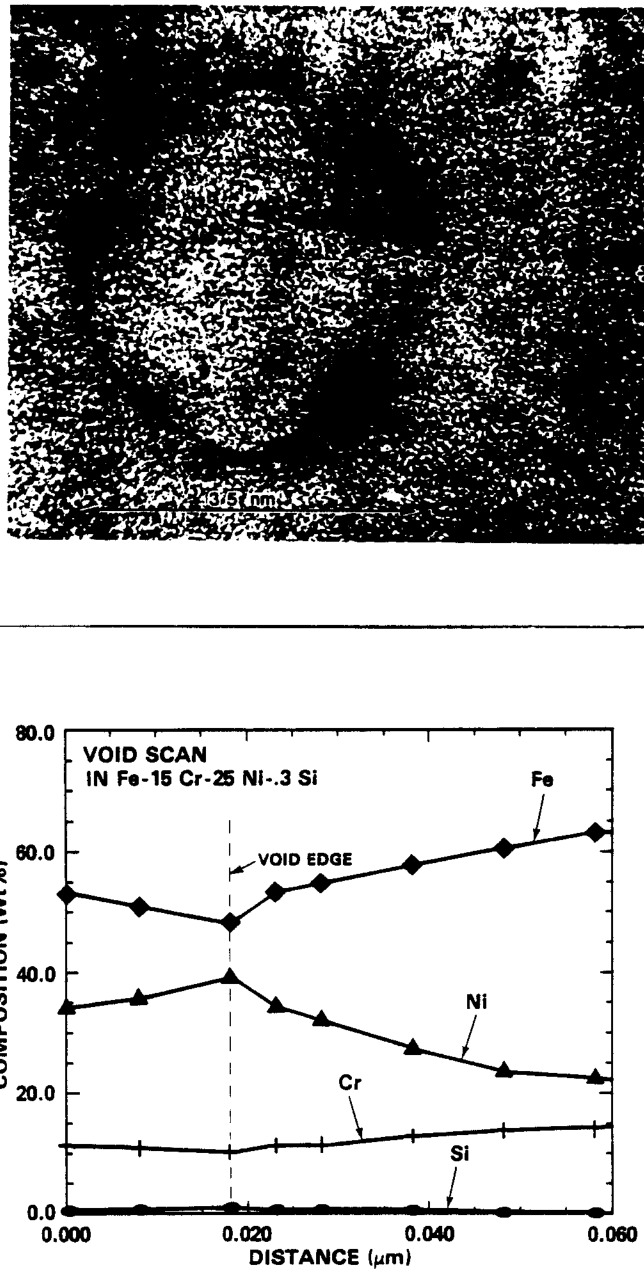


Fig. 3. STEM image of void and composition profiles obtained by EDX microanalysis in neutron irradiated Fe-15Cr-25Ni-0.3Si alloy irradiated in EBR-II at 510°C. Analysis was performed with a 2 nm diameter electron probe in a field ion gun/scanning transmission electron microscope.

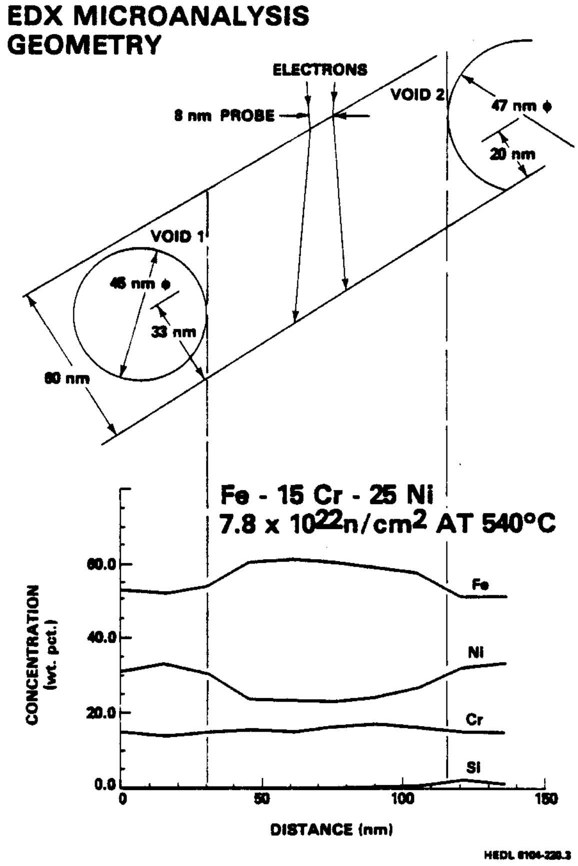


Fig. 4. Microanalysis geometry and composition profiles derived from Fe-15Cr-25Ni irradiated in EBR-II at 540°C. Analysis was performed in a 100 keV TEM/STEM.

occurs to grain boundaries and dislocation loops. When other solutes are present in the alloy, many of these also segregate to microstructural sinks. A summary of the segregation behavior observed in these studies is presented elsewhere [28].

ORIGIN OF SEGREGATION-INDUCED BIAS MODIFICATIONS

The physical basis for the effect of segregation on the bias is to be found in the drift terms contained in the equations for the flux of vacancies and interstitials. In alloys with uniform alloy composition and uniform properties, the flux of point defects to any sink is given by

$$J_o = -\nabla(DC) - \frac{DC}{kT} \nabla U^{\sigma} \qquad (1)$$

where D is the diffusion coefficient for migration, C the concentration of point defects, and U^{σ} is the mechanical interaction with the stress field of the sink to which the point defects are diffusing. As usual, k is the Boltzmann constant, and T is the absolute temperature.

The second term is the only drift term that must be considered when bias factors are computed for uniform materials. During the initial stages of high-temperature irradiations, when RIS has not yet occurred, it may be assumed that Eq. (1) is valid.

Due to the different mobilities of alloying elements and impurities, radiation-induced segregation will occur in the vicinity of sinks. Thus, as composition gradients develop, the point defect fluxes will change, and new drift terms are required in Eq. (1).

For the case of a substitutional binary alloy consisting of A and B atoms, Wolfer [29] has recently shown that the Eq. (1) must be replaced by the following two equations. For the vacancy flux, it is found that

$$J_V = -\nabla(D_V C_V) - \frac{D_V C_V}{kT} \nabla G_V^s + (D_{AV} - D_{BV}) \alpha C_V \nabla x_A \tag{2}$$

and the interstitial flux is given by

$$J_I = -\nabla(D_I C_I) - \frac{D_I C_I}{kT} \nabla G_I^s - (D_{AI} - D_{BI}) \alpha C_I \nabla x_A \ . \tag{3}$$

Here, the subscripts "V" and "I" indicate quantities for vacancies and interstitials, respectively, $x_A = 1 - x_B$ is the local atomic fraction of A atoms, and

$$\alpha = 1 + d \ln \gamma_A / d \ln x_A \tag{4}$$

is the thermodynamic factor which differs from one when the solid solution alloy is not ideal, i.e. when the activity coefficient γ_A deviates from unity.

The diffusivities for the A atoms are denoted by D_{AV} and D_{AI} for migration via vacancies and interstitials, respectively, and analogous definitions are employed for the B atoms. The diffusion coefficients for vacancy and interstitial migration are then given by

$$D_V = x_A D_{AV} + x_B D_{BV} \tag{5}$$

and

$$D_I = x_A D_{AI} + x_B D_{BI} \ . \tag{6}$$

A determination of the diffusivities D_{AV} and D_{BV} is possible when tracer diffusion coefficients are known as a function of the alloy composition. If, for example, D_A^* is the tracer diffusion coefficient in the alloy with the composition x_A and a vacancy fraction of $x_V = C_V/N$ (N is the number of atoms per unit volume), then

$$D_{AV} = D_A^*/(x_V f_V) = D_A^{o*}/(x_V^{eq} f_V) \tag{7}$$

where f_V is the correlation factor for diffusion by the vacancy mechanism. For fcc crystals, $f_V = 0.78145$. In general, only the tracer diffusion coefficient D_A^{o*} for thermal equilibrium condition is known when the vacancy fraction is equal to x_V^{eq}. Although a relationship similar to Eq. (7) can in principle be written down for the diffusivity D_{AI}, it is of no practical significance as $x_I^{eq} \cong 0$. Furthermore, the measurement of a tracer diffusion coefficient for the interstitialcy mechanism is possible only under radiation conditions, and would be obscured by the additional diffusion arising from the vacancy mechanism.

In order to cast the last term in Eqs. (2) and (3) into a form similar to the other drift terms, we define the Kirkendall forces as

$$F_{KV} = -\frac{(D_{AV} - D_{BV})}{(x_A D_{AV} + x_B D_{BV})} \alpha kT\nabla x_A = -K_V kT\nabla x_A \tag{8}$$

and

$$F_{KI} = \frac{(D_{AI} - D_{BI})}{(x_A D_{AI} + x_B D_{BI})} \alpha kT\nabla x_A = +K_I kT\nabla x_A \tag{9}$$

which can now be compared with the other drift forces ∇G_V^s and ∇G_I^s.

These other drift forces arise from the spatial and compositional dependence of the point defect energy in its saddle point, and they will henceforth be called intrinsic drift forces. The Gibbs free energy G_V^s is composed of the following terms:

$$G_V^s = G_V^f + G_V^m + U_V^s \ . \tag{10}$$

Here, G_V^f and G_V^m are the Gibbs free energies for vacancy formation and migration, respectively, and U_V^s is a mechanical interaction energy to be discussed below.

Both G_V^f and G_V^m are functions of the local alloy composition, and their dependence on x_A can be deduced from measurements of the self-diffusion coefficient as a function of alloy composition. If D_{SD} denotes this coefficient, then the intrinsic drift force on vacancies in the absence of U_V^s is given by

$$\nabla(G_V^f + G_V^m) = -kT\nabla[\ln D_{SD}] \ . \tag{11}$$

As stated earlier, the measurement of a tracer diffusion coefficient for interstitials is impossible. Therefore the definition of the intrinsic drift force on interstitials requires a different approach. In this case, we must utilize the fact that the formation energy of an interstitial is mainly determined by its strain or bulk relaxation energy [10,29]

$$U_I^{BR} = \frac{2\kappa\mu}{3\kappa + 4\mu} \frac{v_I^2}{\Omega} \tag{12}$$

where μ and κ are the shear and bulk modulus, respectively, and v_I is the interstitial relaxation volume. The energy U_I^{BR} is for an interstitial in the bulk, i.e. far from a free surface, and is henceforth referred to as the bulk relaxation energy. If one uses the value of the relaxation volume v_I for the saddle-point configuration, then $U_I^{BR} \equiv G_I^f + G_I^m$.

In a composition gradient, the elastic moduli κ and μ become functions of the point defect's position and Eq. (12) is no longer valid in a strict sense. However, since most of the strain energy of a point defect is contributed from the lattice distortions in its immediate surrounding, Eq. (12) remains an excellent approximation when the elastic moduli vary little over distances on the order of a few lattice parameters. In fact, it has been shown [29] that when position-dependent elastic moduli $\kappa(\vec{r})$ and $\mu(\vec{r})$ are used in Eq. (12), then ∇U_I^{BR} represents an exact upper bound to the actual force $\nabla(G_I^f + G_I^m)$; here $\vec{r}$ denotes the position of the interstitial.

For both vacancies and interstitials, the mechanical interaction U^s is composed of three contributions. First, it contains the interaction U^σ with the stress field of the sink. This contribution remains independent of the radiation-induced segregation unless precipitates form. The second contribution is the image interaction U^{Im} in the case of voids, and it simply represents a correction to the bulk relaxation energy U^{BR} when the point defect approaches the free void surface. Again, this interaction U^{Im} is affected very little by the segregation provided the latter does not result in the formation of a precipitate shell. The final contribution arises from coherency strains, when segregation leads to a spatially varying lattice parameter $a_0(\vec{r})$. If $\bar{a}_0$ is the average lattice parameter, and

$$\eta(\vec{r}) = [a_0(\vec{r}) - \bar{a}_0]/\bar{a}_0 \tag{13}$$

the relative variation, then the coherency strain interaction can be shown to be given by [29]

$$U^c = \frac{3\kappa 4\mu}{3\kappa + 4\mu} v\eta(\vec{r}) \ . \tag{14}$$

The physical origin of the segregation-induced bias changes can now be summarized as follows.

Whereas the bias factors of sinks in materials without segregation is due to the action of the interaction force ∇U^{σ} in the case of dislocations, and due to the force $\nabla(U^{\sigma} + U^{Im})$ in the case of voids, additional forces emerge as a result of segregation. These are:

a. the intrinsic force due to the composition dependence of both the point defect formation and migration energy; both can be expressed as a strain energy U^{BR} in the case of interstitials; this force is a function of the local elastic moduli;

b. the lattice-mismatch force ∇U^{c} arising from the coherency strain field when the lattice parameter changes with composition;

c. the Kirkendall force $\vec{F}_K = \pm K kT \nabla x_A$, arising from the differences in atomic diffusivities.

COMPARISON OF THE VARIOUS DRIFT TERMS

The various drift forces require for their evaluation basic material parameters such as elastic moduli, lattice parameter, atomic diffusivities, and the activity coefficients. These are required both as a function of composition and temperature. Unfortunately, such detailed information is presently not available, and an accurate determination of the drift forces cannot be made. However, it is possible to assess the order of magnitude of these forces with the available data.

For an estimate of the intrinsic and the Kirkendall force on vacancies we consider the measurements of Million et al. [30] on tracer diffusion coefficients and the activity coefficient γ_{Ni} in binary austenitic Fe-Ni alloys. For the temperature range between 985°C and 1305°C, they found the following relationships for the tracer diffusion coefficients:

$$\log_{10} D^{o*}_{Ni} = 0.537 - \frac{15152}{T} - x_{Fe}\ (0.488 - \frac{340}{T}) \tag{15}$$

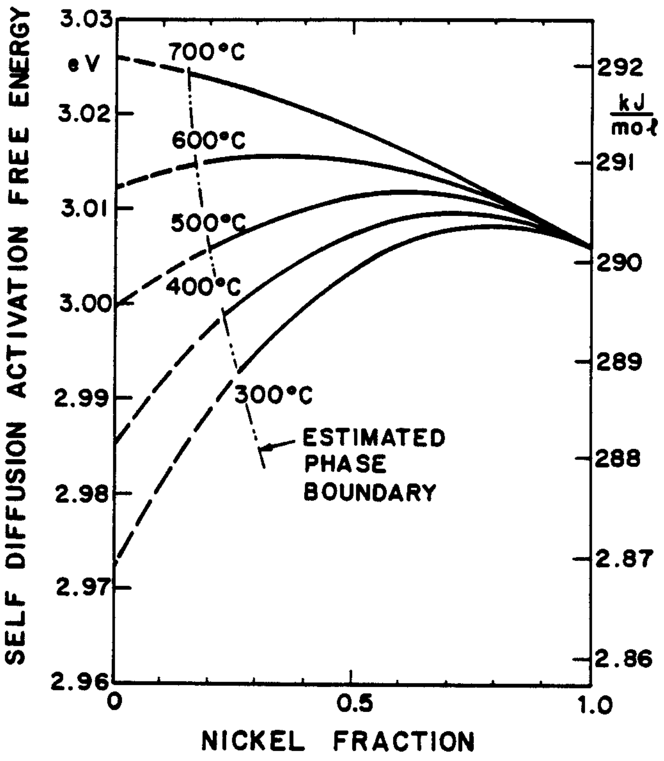
$$\log_{10} D^{o*}_{Fe} = -0.142 - \frac{14561}{T} + x_{Ni}\ (1.106 - \frac{993}{T}) \tag{16}$$

Here, T is the absolute temperature, and the diffusion coefficients are given in units of cm^2/s. Using the definition

$$D_{SD} = \frac{1}{f_V} [x_{Ni} D^{o*}_{Ni} + x_{Fe} D^{o*}_{Fe}] \tag{17}$$

for the self-diffusion coefficient we can compute the intrinsic force or the effective activation energy $-kT \ln D_{SD}$ for self-diffusion. Figure 5 shows the results as a function

Fig. 5. Activation free energy for self-diffusion in binary Fe-Ni alloys.



of the nickel composition and temperature. These results were normalized to give the same value of the activation energy at $x_{Ni} = 1$. Note that the intrinsic force is equal to the slope of these curves multiplied by the nickel gradient. For a segregation gradient spanning a 20% difference in nickel composition, we find that the potential of the intrinsic force on the vacancies is of the order of 0.01 eV or less, and it can be positive or negative depending on temperature and nickel composition. Note that a decrease in the force potential with increasing nickel means that the vacancy is attracted towards higher nickel concentration.

Equations (15) to (17) can also be used to evaluate the Kirkendall force coefficient K_V defined by the relationship (8). For the thermodynamic factor we use extrapolations of values measured in the temperature range from 950°C to 1250°C [30]. Figure 6 shows the measured results as solid lines and our extrapolation to lower temperatures as dashed lines. The Kirkendall force coefficient K_V for vacancies is shown in Fig. 7. It is seen that K_V is negative for alloy composition with less than 35% nickel, and positive for high nickel compositions. A negative coefficient K_V implies that the vacancies are attracted to regions of higher nickel concentration whereas positive values imply the reverse. In order

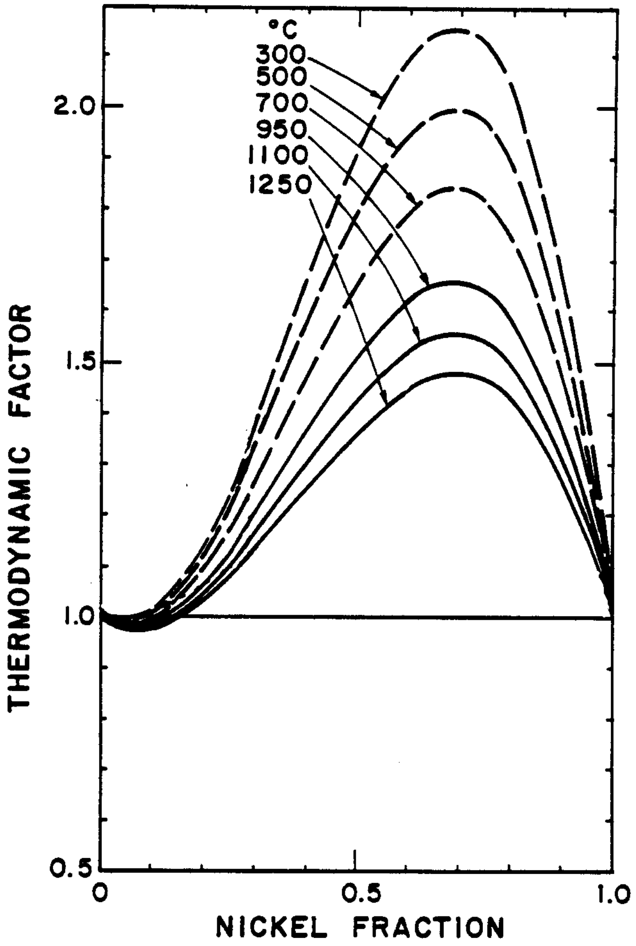


Fig. 6. Thermodynamic factor for binary Fe-Ni alloys. Solid lines are based on measurements and dashed lines are based on extrapolations.

to compare the Kirkendall force with other drift forces we integrate the force over a nickel variation of 20%. The resulting potential difference is then found to be on the order of 0.4 kT or less, depending on temperature and alloy composition. Therefore, the Kirkendall force corresponds to an interaction potential on the order of 0.02 eV or less.

In order to obtain an estimate for the Kirkendall force K_I on interstitials, we suppose that the interstitial formation energy in nickel and in fcc iron differs by

$$G^S_{Ni,I} - G^S_{Fe,I} = \pm 0.05 \text{ eV}$$

and that the energies $G^S_{Ni,I}$ and $G^S_{Fe,I}$ are the activation energies for the tracer diffusion coefficients of nickel and iron migrating as part of a dumbbell interstitial. It turns out that as long as these energies are larger than about 1 eV, only their difference affects the Kirkendall force K_I. The computed values of K_I are shown in Fig. 8 for the case that the difference between $G^S_{Ni,I}$ and $G^S_{Fe,I}$ is negative. In this case, the Kirkendall force

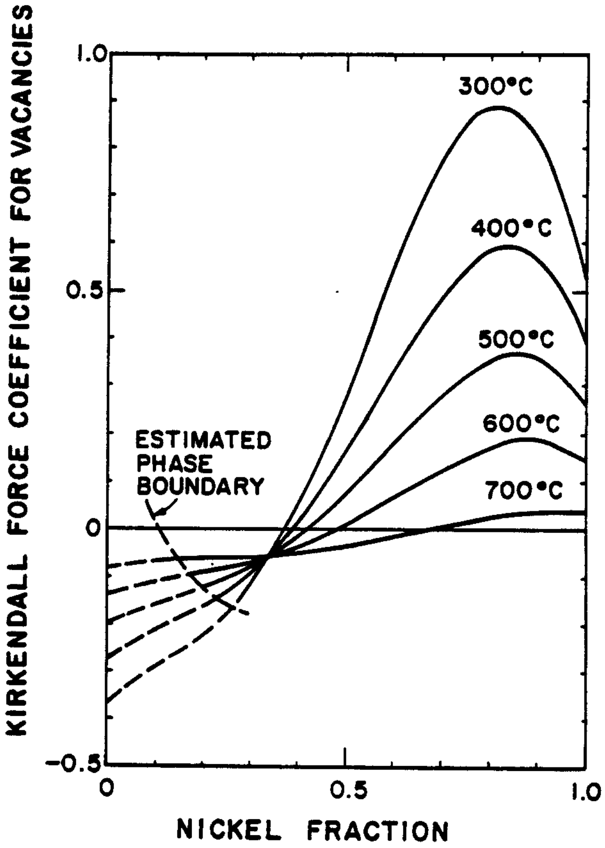


Fig. 7. Coefficient of the Kirkendall force on vacancies in binary Fe-Ni alloys.

makes the interstitials migrate preferentially towards higher nickel concentrations. On the other hand, if the difference in the activation energies is positive, then the interstitials are attracted toward regions of lower nickel concentration.

The magnitude of the Kirkendall force on interstitials is of the same order as the Kirkendall force on vacancies.

The remaining interaction energies U^{σ}, U^{I}, U^{BR}, and U^{c} have been evaluated in previous publications [10,17,18], and typical values are listed in Table 1 together with the above estimates for the potentials of the intrinsic and Kirkendall forces.

CONCLUSIONS

When the various potentials are compared in relationship to U^{σ} we arrive at the following conclusions.

a. Radiation-induced segregation introduces new drift forces which are of comparable magnitude to the strain-induced drift force present without segregation. As a result, segregation can profoundly change the bias of sinks for preferential absorption of

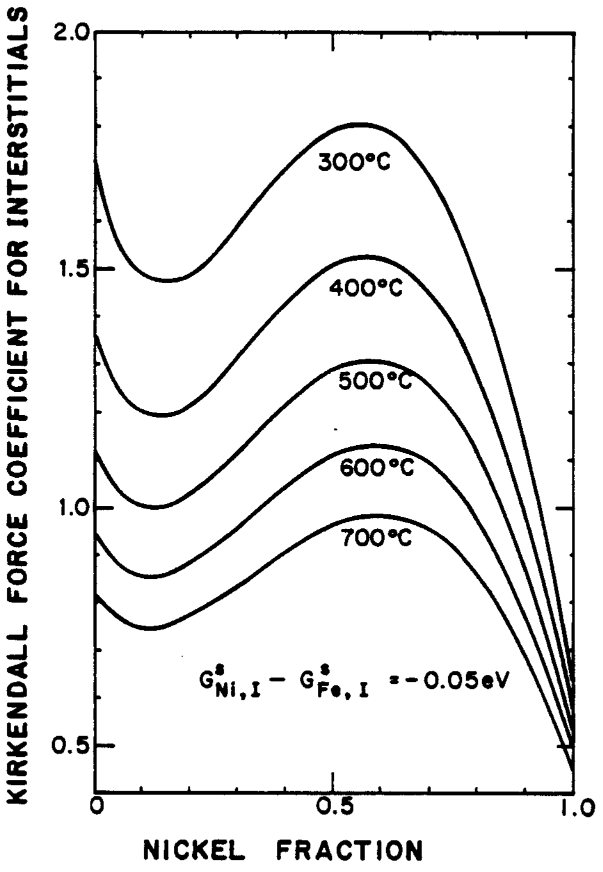


Fig. 8. Possible variation of the coefficient for the Kirkendall force on interstitials as a function of the composition of binary Fe-Ni alloys.

interstitial or vacancies.

b. The Kirkendall force on interstitials is small in comparison to the other forces. However, the forces on the interstitial introduced by variations in elastic moduli and lattice parameter are significant.

c. On the other hand, the Kirkendall and the intrinsic force on the vacancy are comparable to both the stress-induced force and the force arising from coherency strains, whereas the force originating from the gradient of the elastic moduli is of secondary importance.

d. In general, RIS affects the total force on interstitials to a much larger degree than the total force on vacancies. Therefore, the sink bias factors for interstitials are expected to be changed more than those for vacancies. Nevertheless, since it is the net bias, i.e. the relative difference of bias factors, which determines void nucleation and growth, the effect of RIS on the vacancy bias factors may not be negligible.

Table 1. Typical Values for the Potentials of the Drift Forces in Fe-Ni Alloys

Potential Energy in eV (and 10^{-21} J)	Interstitial	Vacancy
U^{σ} at a distance of 10 b* from an edge dislocation and above (upper sign) or below (lower sign) the glide plane	±0.25(±40)	∓0.05(∓8)
U^{Im} at a distance from the void surface		
of 2b*	-0.25(-40)	-0.025(-4)
and 5b	-0.005(-0.8)	-0.0005(-0.08)
Change in bulk relaxation energy, ΔU^{BR} for ±3% shear shear modulus change	±0.24(±39)	±0.006(±1)
U^{c} for a lattice parameter change of ±0.4%	±0.24(±39)	∓0.03(∓5)
Change of potential for the intrinsic force over a 20% nickel variation	---	$\lesssim$ 0.01(1.6)
Change of potential for the Kirkendall force over a 20% nickel variation	$\lesssim$ 0.02(3.2)	$\lesssim$ 0.02(3.2)

*b = Burgers vector

e. The reversal of sign for the Kirkendall force at 35% nickel may provide an explanation for the strong dependence of swelling on nickel content below 35% Ni in Fe-Ni-Cr alloys.

f. Since void nucleation is quite sensitive to the void bias, it is anticipated that the primary effect of nickel segregation will be exerted on void nucleation rather than growth.

Segregation, and in particular substantial nickel enrichment near sinks, appears to be a general phenomenon in nickel-containing austenitic alloys. Significant bias modifications must accompany this segregation, and the degree and nature of the bias change is expected to be a strong function of the alloy composition and the temperature. However, fundamental properties such as elastic moduli, lattice parameter, atomic diffusivities, and thermodynamic properties as a function of alloy composition and temperature are not sufficiently well known to establish a completely quantitative relationship between alloy composition and void swelling resistance.

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