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Influence of radiation-induced voids and bubbles on physical properties of austenitic structural alloys

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Abstract

Void swelling in austenitic stainless steels induces significant changes in their electrical resistivity and elastic moduli, as demonstrated in this study using a Russian stainless steel irradiated as fuel pin cladding in BN-600. Precipitation induced by irradiation also causes second-order changes in these properties, but can dominate the measurement for small swelling levels. When cavities are full of helium as expected under some fusion irradiation conditions, additional second-order changes are expected but they will be small enough to exclude from the analysis.
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1. Introduction

Void swelling is known to influence not only the dimensional stability of irradiated stainless steels, but also many of its basic physical properties. Recently, there has been a focus in the light water reactor community on measurement of void-induced changes in electrical resistivity, elastic moduli and ultrasonic velocity, and to use these measurements to measure swelling nondestructively [1–3].

Electrical resistivity changes were used earlier in the fusion materials program as a method to estimate the swelling-induced changes of thermal resistivity for high heat flux components [4]. It now appears that nondestructive applications of changes in this and other important physical properties may be useful for stainless steels chosen for use in the fusion energy program. As shown in this study, there are other microstructural contributions to changes in physical properties that must be taken into consideration.

2. Experimental details

Fuel element cladding tubes (6.9 mm initial outer diameter and thickness 0.4 mm) constructed from 20% cold-worked austenitic steel of nominal composition 0.1C–16Cr–15Ni–2Mo–lMn that had been irradiated in the BN-600 fast reactor were examined after defueling and cleaning. Specimens of 30 mm length were cut from various areas to obtain a variety of temperatures (430–590 °C), dose levels (50–90 dpa) and swelling values as high as 23%.

The electrical resistivity R was measured for each specimen at room temperature using a technique in which the potential difference between standard and test specimens was determined. The procedure of electrical resistivity measurements was described in detail in [5]. The relative measurement error did not exceed 1%.

To measure Young's modulus E and shear modulus G an ultrasonic resonant technique was utilized which is

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based on both excitation of ultrasonic vibrations and measurement of natural frequencies of longitudinal and shear vibrations in the test specimen [6]. The magnitudes of both moduli were calculated based on dynamic elasticity theory using measured values of resonant frequencies and specimen sizes. Measurement error of the moduli did not exceed 1%.

Void swelling S was determined from density measurements using a hydrostatic weighing technique

$$S = (\delta_0/\delta - 1) \cdot 100\%, \tag{1}$$

where δ_0 and δ are the density in the initial state and after irradiation, respectively, with measuring error of density was ±0.02 g/cm^3.

3. Results

Results of electrical resistivity measurements and determination of Young's modulus E and shear modulus G are shown in Figs. 1–3. The dependence of these physical properties on void swelling is clearly demonstrated. One can see also some deviations from expected behavior that may reflect experimental problems or may be connected with other microstructural changes. As radiation-induced structural changes are usually dependent on irradiation temperature the data were separated into two sets: 'high-temperature' (>520 °C) and 'low-temperature' (<520 °C).

4. Discussion

As was shown in [6] voids in the two-phase model can be considered as a second-phase with zero values of electrical conductivity and elastic modulus. Relative change can be defined in the following analytical expressions assuming an invariance of physical and mechanical properties of the matrix material,

$$\frac{\Delta R}{R_0} = \frac{5 \cdot S}{4 \cdot S + 6}, \tag{2}$$

$$\frac{\Delta E}{E_0} = \frac{1}{(1+S)^2}, \tag{3}$$

$$\frac{\Delta G}{G_0} = \frac{1}{(1+S)^2}. \tag{4}$$

ΔR, ΔE and ΔG are absolute changes in resistivity and moduli, respectively, and R_0, E_0, and G_0 are the values in the initial state, and swelling S is expressed as a fraction.

Theoretical dependencies of electrical resistivity and elasticity moduli on swelling calculated using Eqs. (2)–(4) are shown by lines in Figs. 1–3. Swelling is confirmed to be the dominant factor in producing changes in these

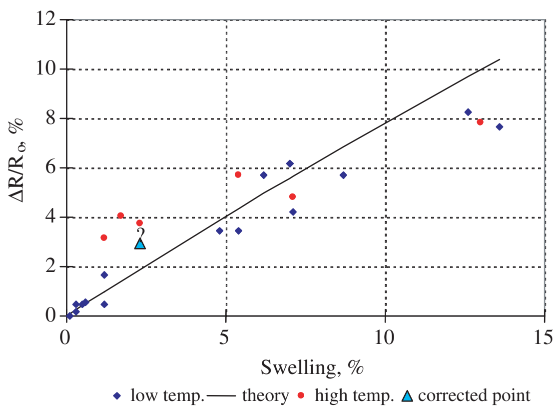

Fig. 1. Dependence of relative change in electrical resistivity on void swelling. One point at 2.2% swelling shows the downward correction that would arise when partially accounting for nickel segregation into precipitates.

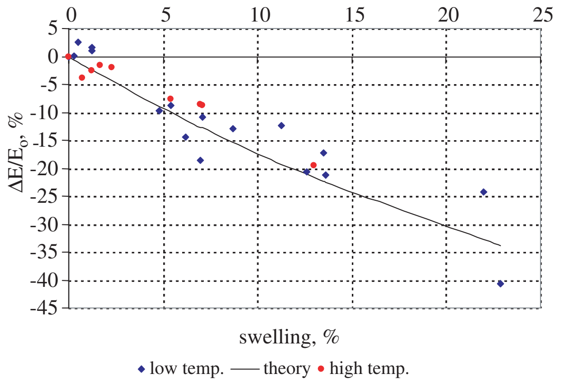

Fig. 2. Dependence of relative change in Young's moduli on void swelling.

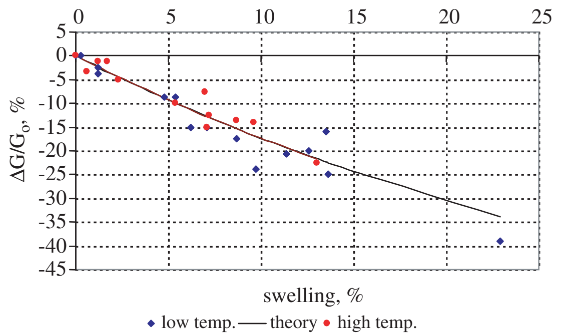

Fig. 3. Dependence of relative change in shear moduli on void swelling.

physical properties. At the same time, other microstructural changes contribute to measured changes in electrical resistivity and elastic moduli, but in a second order manner. Electrical resistivity in particular is affected. Note that Fig. 1 shows that experimentally determined magnitudes are higher than the void-based prediction at lower swelling and higher irradiation temperatures. Thus at low swelling levels the precipitate contributions dominate the measurement. Also note that the measured values of resistivity are lower than the prediction at higher swelling.

It is known well that precipitates usually have different densities, ultrasonic velocities and electrical resistivity, but precipitates also change the property of the matrix from which they form, especially by removal of elements that have a strong effect on the electrical resistivity. Let's consider how it is possible to change the electrical resistivity. The main radiation-induced changes in steel are changes in dislocation structure, second-phase precipitates, and change in composition and substitution of neighboring atoms.

When the dislocation density is changed no more than usually observed during irradiation it does not significantly affect electrical resistivity, so this factor can be neglected. Second phases at small volumes can significantly affect electrical resistivity when their specific electrical resistivity is different from the matrix. Phases known to precipitate in this steel under irradiation are (Fe, Cr)$_{23}$C$_6$, a phase enriched by Cr, and G-phase which is enriched in Ni at the expense of matrix reduction in this element.

There are no data on specific electrical resistivity of these phases in the literature, but data do exist on various Fe–Cr–Ni alloys in [7] where one can select alloys of Fe–Cr and Fe–Ni close to the considered phases. Based on these reference data it appears that Fe–Cr alloys have electrical resistivities that are relatively independent of Cr content so we can deduce that influence of the (Fe, Cr)$_{23}$C$_6$ phase on electrical resistivity of the matrix can be neglected. However, this does not address the removal of carbon from the matrix, which should have some influence to reduce the matrix resistivity. Nor does it address the resistivity contribution of the carbides.

Changing of Ni content in Fe–Ni alloys has a strong influence on electrical resistivity. G-phase is known to contain 42–57% Ni [8] and according to the data on physical properties of high-content Ni alloys [7] the specific electrical resistivity of G-phase can be higher than that of the matrix by ~50 %, based only on this consideration. This estimate ignores, however, the effects of concentrating Si and Ti in the precipitate and their concurrent reduction in the matrix.

Influence of precipitate phases on electrical resistivity can be included in the two-phase model using the approach of Ref. [9], employing the electrical resistivity of the G-phase precipitate and steel in its initial state. We obtain the following expression for electrical resistivity.

$$\frac{\Delta R}{R_0} = \frac{5S}{4S+6} + c \times \frac{\Delta\rho}{\rho_0} \times \left(1 + \frac{5S}{4S+6}\right) \times \frac{3}{3 + 2 \times \Delta\rho/\rho_0 \times (1-c)}, \quad (5)$$

where ρ_0 is the specific electrical resistivity of matrix, $\Delta\rho$ is the difference between electrical resistivity of the phase and matrix, and c is the volume ratio of the phase/matrix.

To estimate the magnitude of the influence of G-phase on electrical resistivity, microscopy examination was conducted on a specimen having 2.3% swelling, as shown in Fig. 4. The volume ratio of G-phase was determined to be 2.2% in this specimen.

Calculations using Eq. (5), in which the corresponding value of the unirradiated specimen was assumed to be the electrical resistivity of matrix, gives an addition of ~1% to the electrical resistivity. Therefore, in order to use Eq. (2) for estimating swelling from electrical resistivity measurements, it is necessary to adjust the experimental results to account for G-phase formation. We have to shift the experimental value 1% downwards in our case for the test specimen, as shown by the corrected datum shown in Fig. 1. We obtain movement toward the curve of ~1% (instead of 2%). This difference is in agreement with accuracy of determination of electrical resistivity.

What changes in this analysis might we anticipate for application of these fast reactor results to fusion-relevant conditions? In this discussion, we ignore the possible effect of helium to change the amount of swelling. We focus only on the possibility that, for a given swelling level, fusion-produced property changes arising from voids or bubbles will be different from that

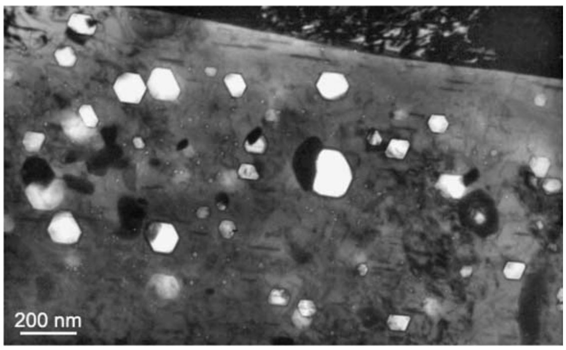

Fig. 4. G-phase precipitates and voids observed in an irradiated specimen of 20% cold-worked 0.1C–16Cr–15Ni–2Mo–1Mn steel with swelling of 2.3%.

produced in low helium generating spectra found in fast reactors.

The first order effect, if any, must arise from some aspect of the very high helium generation rate characteristic of fusion spectra. It is well known that high helium generation rates tend to 'homogenize' swelling, distributing the swelling into a higher density of smaller cavities, whether they are voids or bubbles. However, this aspect of high helium generation rates can be ignored, since it is only the total void volume and not its distribution that determines the void-induced change in the physical properties [7].

More appropriately, what is the consequence of helium, possibly at high equilibrium values, on void contributions to changes in physical properties? Helium, especially at high pressures, might be expected to decrease the effectiveness of voids to reduce the elastic moduli, and possibly to increase the electrical and thermal resistivity.

Wolfer has examined this possibility for elastic moduli [10]. Gas content can contribute to the bulk moduli but not the shear moduli. Considering the theoretical maximum helium content, the cavity modulus change would be reduced much less than 10%, with most cases resulting in even smaller gas-induced moduli change.

Since helium is effectively a nonconductor there will be no measurable change in a cavities' ability to resist the flow of electrons. Similarly, the large mass difference between helium and the Fe, Cr, Ni of the matrix will make heat transfer very inefficient, yielding no improvement in the thermal conductivity.

5. Conclusions

Void swelling is the dominant contributor to changes in electrical resistivity and elastic moduli of austenitic steels under high-dose neutron irradiation in fast reactors. Second-order contributions appear to arise from radiation-induced precipitate formation, both directly and indirectly via the effect of precipitation to change the composition of the alloy matrix. At low swelling levels the precipitate contribution can dominate the measurement.

It is anticipated that helium-pressurization of cavities in fusion neutron spectra will not appreciably change the influence of cavities on these physical properties, allowing the results of this fast reactor study to be applied without correction to fusion conditions.

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