THE EFFECT OF VOID SWELLING ON ELECTRICAL RESISTANCE AND ELASTIC MODULI IN AUSTENITIC STEELS - A. V. Kozlov, E. N. Shcherbakov, S. A. Averin (Research & Development Institute of Power Engineering Zarechny**,** Russia) and F. A. Garner (Pacific Northwest National Laboratory)*

OBJECTIVE

The objective of this effort is to determine for austenitic steels the impact of void swelling and precipitation on physical properties such as elastic moduli and thermal or electrical resistivity. The results of this study will allow the development of nondestructive diagnostic procedures for materials irradiated in either fission or fusion devices.

SUMMARY

Measurements are presented of electrical resistance and elastic moduli (Young's modulus and shear modulus) of stabilized austenitic fuel pin cladding after irradiation in the BN-600 reactor. Additional data are presented on changes in electrical resistivity of another stabilized austenitic steel irradiated in the BN-350. The elastic moduli are reduced and the electrical resistance is increased as the neutron dose increases. These changes are correlated with void swelling measured on the same specimens.

Dependencies of these changes in physical properties on neutron irradiation dose, temperature and swelling level are plotted and it is shown that to the first order, the property changes are dependent on the swelling level in agreement with earlier U.S. and Russian data, and also in agreement with various theoretical predictions. It is also observed, however, that changes in electrical resistance and elastic moduli frequently differ slightly for specimens with equal swelling, but which were obtained at different combinations of temperature and dose. These second-order differences appear to arise from contributions of other radiation-induced structural changes, especially in precipitation, which depends strongly on irradiation temperature in stabilized steels.

PROGRESS AND STATUS

Introduction

Nuclear power plants, both fission and perhaps fusion driven, can operate from 30 to 60 years under some circumstances, allowing near-core structural components to achieve radiation doses ranging from 10 to 120 dpa at irradiation temperatures varying from 280 to 400°C. Void swelling in this temperature range is a potential concern for further operation of such reactors [1,2]. Furthermore, much time is needed to accumulate high damage doses in existing facilities, and therefore experimental examination possibilities are somewhat limited, especially since it usually requires the removal of structural elements from the reactor that still serve as necessary operational components.

Another way to determine the state of swelling within a given plant component is to *in-situ* measure non-destructively properties affected by swelling. Such an approach requires a demonstration that swelling-induced changes induced in physical properties can indeed be measured and correlated with the swelling level. This paper demonstrates such swelling-sensitive dependencies, focusing on electrical resistance and elastic moduli of two stabilized austenitic steels after irradiation to high doses in several fast reactors.

Material and examination procedure

Two sets of specimens were examined. First, specimens of 20 mm length were cut from 1 mm diameter spacing wire irradiated in the BN-350 fast reactor. This wire is made of 0.1C-16Cr-15Ni-3Mo-1Nb austenitic steel and is wrapped in a spiral around fuel pins to provide spacing between adjacent pins. Average temperatures and irradiation doses of these specimens are listed in Table 1.

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The second set contains cladding tube specimens removed from two fuel elements designated "central" and "peripheral" according to their different location in the BN-600 fast reactor core. The cladding was made of 0.1C-16Cr-15Ni-3Mo-1Mn steel in the 20% cold-worked condition. Average temperatures and irradiation doses of these specimens are listed in Table 2. Tested specimens are 30 mm long, 6.9 mm outer diameter and 0.4 mm thickness. The fuel was removed before testing and the tube cleaned to remove contamination deposits. Table 3 contains the detailed composition of the two steels.

Electrical resistance *R* was determined by comparison of voltage reduction in a reference specimen and the measured specimen. The procedure of electrical resistance measurement is described in detail in Ref. [3]. The relative error in this measurement did not exceed 1%.

For measurement of elasticity characteristics (Young's modulus *E* and shear modulus *G*) an ultrasonic resonant method was employed. The method is based on excitation of ultrasonic oscillations, with measurement of natural frequencies of longitudinal and shear oscillations in the specimen [3]. Using dynamic elasticity theory, values of both Young's modulus *E* and shear modulus *G* were calculated using measured values of resonant frequencies. The error in the measurement did not exceed 1%.

Table 1. Average temperatures and neutron irradiation dose of 0.1C-16Cr-15Ni-3Mo-1Nb steel

T_{irr}, °C	595	580	560	530	495	440	335
D, dpa	40	46	45.5	43	35	32	28

Table 2. Average temperatures and neutron irradiation dose of 0.1C-16Cr-15Ni-3Mo-1Mn steel

Central fuel element cladding									
T_{irr}, °C	370	431	439	488	500	522	561	570	586
D, dpa	3	53	56	69	71	72	70	68	63
Peripheral fuel element cladding									
T_{irr}, °C	370	427	435	510	518	567	586		
D, dpa	3	52	55	69	69	65	63		

Table 3a. Chemical composition of fuel pin cladding of 0.1C 16Cr-15Ni-2Mo-1Mn steel

Element	C	Cr	Ni	Mo	Mn	Si	Ti	V	B	N	Co	P	S
wt %	0.06-0.07	15.9-16.7	14.0-14.7	2.09-2.24	1.4 1.6	0.39-0.50	0.30-0.41	0.10-0.24	0.001-0.003	0.009-0.015	0.004-0.010	0.006-0.018	0.003-0.007

Table 3b. Chemical composition of spacing wire of 0.1C-16Cr-15Ni-3Mo-1Nb steel

Element	C	Cr	Ni	Mo	Mn	Nb	Si	Ti	B	N	P	S
wt %	0.04-0.06	15.0-16.5	15.0-16.0	2.7 - 3.2	0.4 - 0.8	0.6 0.7	0.3 - 0.6	0.05	0.002	0.03	0.02	0.010

The void swelling *S* in % was determined by hydrostatic weighing to determine the density by

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

where δ_0 and δ are the density in the initial state and after irradiation, respectively. The error in the measurement did not exceed 0.02 g/cm^3.

Values of both electric resistance and swelling were measured on the spacing wire specimens. Similar measurements were made on the tube specimens, but elastic moduli also were determined. All measurements were performed in hot cells.

Results

Figure 1 presents the temperature dependence of swelling and the relative change in electrical resistance of the 0.1C-16Cr-15Ni-3Mo-1Nb spacing wire irradiated in the BN-350 fast reactor. The correlation of the two dependencies is quite obvious. Peak swelling was 9.5% in this specimen set, achieved at a dose of 43 dpa and an irradiation temperature of 540°C. The resultant effect on electrical resistance was an increase of 8.2%.

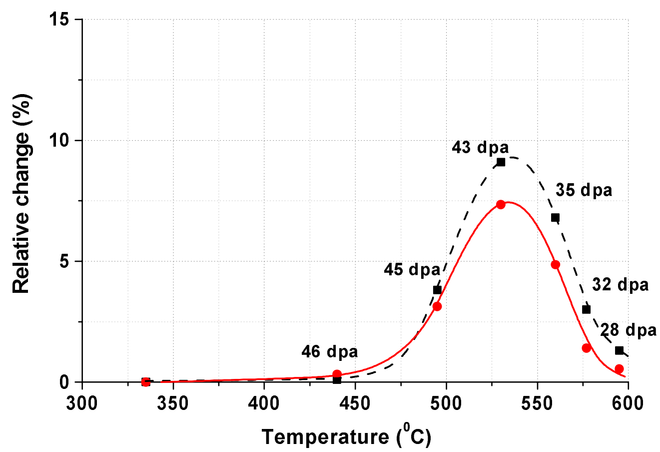


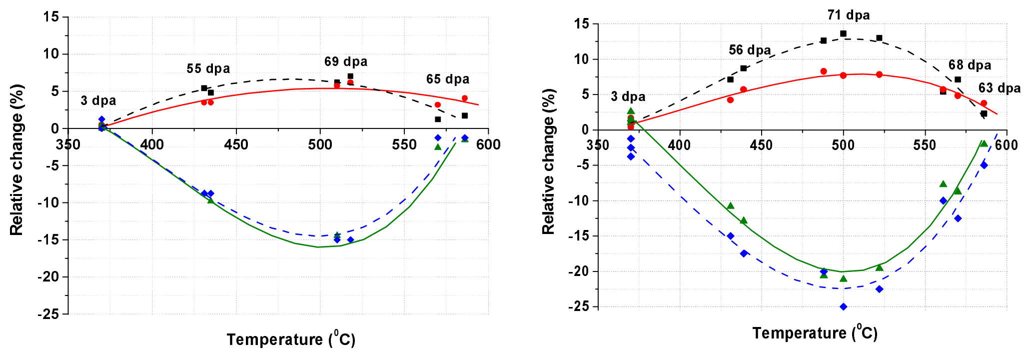
Figure 1. Dependence of swelling (■ - S) and relative changes in electrical resistance (● - $\Delta R/R_0$) on irradiation temperature for 0.1C-16Cr-15Ni-3Mo-1Nb austenitic steel irradiated in the BN-350 reactor.

Figure 2 shows the measurement results for cladding specimens of 0.1C-16Cr-15Ni-2Mo-1Mn irradiated in the BN-600 reactor. The peak swelling of peripheral fuel element specimens was 7%, the resultant electrical resistance was increased 6.2%, while Young's modulus and shear modulus were each reduced 15%. The central fuel element cladding had a value of peak swelling of 13.6%, an increase in electrical resistance of 8.3%, and reductions of Young's modulus and shear modulus of 21.1% and 25%, respectively.

Changes in electrical resistance, Young's modulus and shear modulus obviously correlate with swelling for both sets of cladding. Moreover, the relative changes in elastic moduli are approximately two times larger than the concurrent change in electrical resistance.

Discussion

Changes in physical and mechanical properties of irradiated metallic materials are caused by changes in their microstructure and elemental composition of the alloy matrix, [4]. Formation of voids and second-phase precipitates, along with a modification of the dislocation microstructure are the major microstructural factors affecting moduli and resistivity. Additionally, second order contributions may arise from point defect populations and their impact on the distribution of alloying elements, which can



a b

Figure 2. Dependence of swelling (■ - S) and relative changes in electrical resistance(● - $\Delta R/R_0$), Young's modulus (▲ - $\Delta E/E_0$) and shear modulus (♦ $\Delta G/G_0$) on irradiation temperature for 0.1C-16Cr-15Ni-3Mo-1Mn steel (20% c.w.) irradiated in the BN-600 reactor; a – peripheral fuel element, b - central fuel element.

serve as scattering centers for moving electrons in the alloy matrix.

Changes in electrical resistance

Voids can be viewed as second phase particles with zero conductivity. The aggregate electrical resistance of a two-phase system (a system of a single-phase matrix with unconnected second phase particles) is determined by the equation described in [5].

$$\gamma = \gamma_0 \cdot \left(1 + \frac{c}{(1-c)/3 + \gamma_0/(\gamma_1 - \gamma_0)}\right) \tag{2}$$

where γ is specific electric conductivity of an alloy, γ_0 and γ_1 are electric conductivities of the matrix and second phase, respectively, and *c* and *(1-c)* are volume contents of the second phase and matrix phases. For the case of voids $\gamma_1 = 0$, the volume content *c* is the porosity *P.* Taking into account that $\rho = 1/\gamma$, the aggregate resistance has the following description,

$$\rho = \rho_m \cdot \frac{(2+P)}{2 \cdot (1-P)} \tag{3}$$

where ρ_m is specific resistance of the matrix phase. Expressing porosity *P* through the fractional swelling S_f the following equation can be written.

$$\frac{\rho}{\rho_m} = \frac{3 \cdot S_f + 2}{2} \tag{4}$$

To express relative change in electric resistance, it is necessary to take into account an increasing of cross-section area *F* of an irradiated specimen in comparison with the cross-section of an unirradiated specimen F_0 arising from swelling.

$$F = F_0 \cdot \left(1 + \frac{2}{3} \cdot S_f\right) \tag{5}$$

Using equations (4) and (5) and expressing the electrical resistance *R* in terms of its specific electrical resistance and the specimen size, the final equation is attained.

$$\frac{\Delta R}{R_o} = \frac{\rho / \rho_o - F / F_o}{F / F_o} = \frac{5 \cdot S_f}{4 \cdot S_f + 6} \qquad (6)$$

Figure 3a shows the relationship between changes in electrical resistance and swelling for specimens of 0.1C-16Cr-15Ni-3Mo-1Nb steel irradiated in BN-350, and also shows the predicted dependence on swelling describing by equation (6). The similar dependencies for specimens of 0.1C-16Cr-15Ni-3Mo-1Mn steel irradiated in the BN-600 are shown in Figure 3b. One can see that dependence of relative changes in electrical resistance on swelling for different steel and initial thermal treatment and irradiated in different reactors is in good agreement with equation (6). At the same time it should be noted that are some differences.

Specimens of 0.1C-16Cr-15Ni-3Mo-1Mn steel irradiated above 570°C in general have higher electrical resistance compared with the value predicted by equation (6). Formation of rather large G-phase precipitates at these higher temperatures is most likely the cause of this divergence [5]. Precipitates of this phase are small and their volume fraction is much lower at temperatures near 400°C, as shown in Figure 4a. The size of G-phase precipitates and their volume fraction increases as temperature increases, as seen in Figure 4b. G-phase is known to contain 42-57% of nickel and to concentrate other elements such as silicon, removing these elements from the alloy matrix [7]. According to data on physical properties of Ni-based alloys [8] the electrical resistance of such precipitates may be on 30 – 50% higher than that of the original matrix. Of course there are concurrent changes in the matrix composition and the aggregate change will reflect the sum of these two contributions.

Considerable changes in relative distribution of various elements are also observed at high swelling levels, especially at void surfaces, a process which may also result in changes in aggregate electrical resistance.

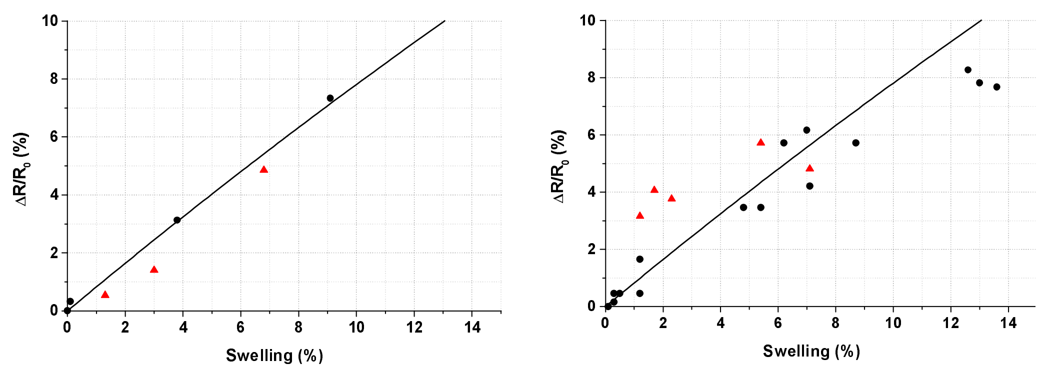


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Figure 3. Dependence of relative changes in electrical resistance on swelling of 0.1C-16Cr-15Ni-3Mo-1Nb steel irradiated in the BN-350 reactor (a) and 0.1C-16Cr-15Ni-2Mo-1Mn (20% c.w.) steel irradiated in the BN-600 reactor (b): ● - low-temperature range T_{irr}<530°C, ▲ - high-temperature range T_{irr}> 530°C; —— - dependence calculated by the equation (6).

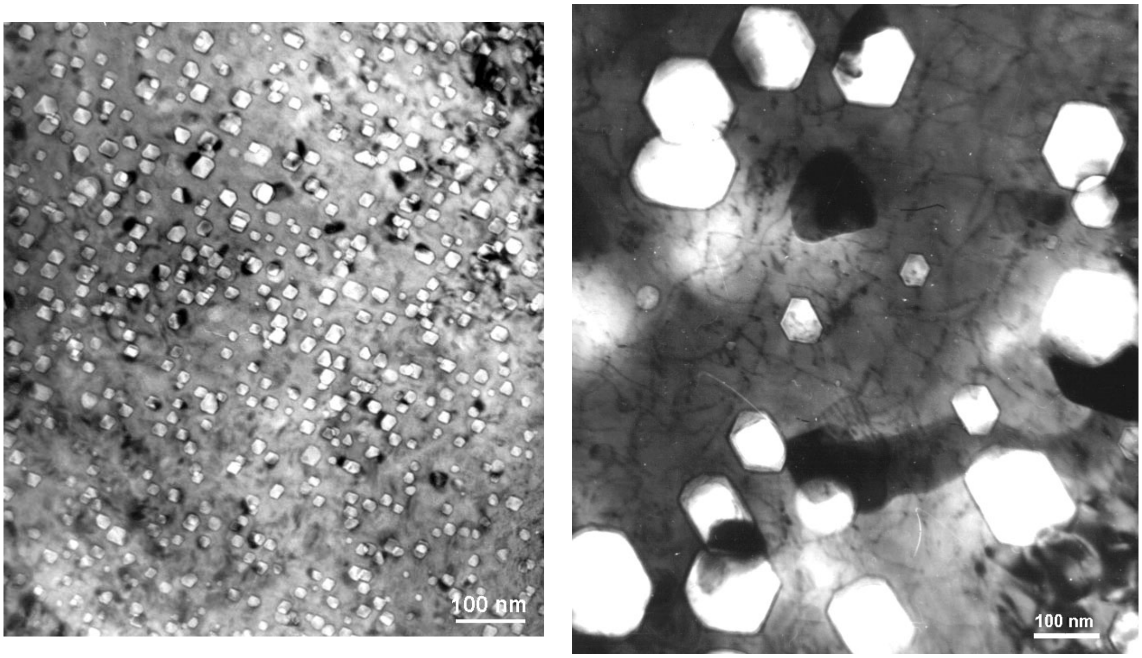

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Figure 4. G-phase precipitates in 0.1C-16Cr-15Ni-2Mo-1Mn steel irradiated in the BN-600: a – $T_{irr.}$= 420°C, D = 49 dpa; b – $T_{irr.}$= 580°C, D= 65 dpa.

Changes in elastic moduli

Similarly, in order to examine Young's modulus E of metal materials containing voids we apply the same two-phase material approach. Voids are considered to be particles of second phase with $E=0$. The modified equation for Young's module is [8].

$$E_i = E_m \cdot (1 - P) \quad (7)$$

where E_i is Young's modulus of an irradiated material containing voids, and E_m is Young's modulus of the unvoided matrix.

Young's modulus is calculated by equation [4] during determination of elastic characteristics by the ultrasonic resonant method.

$$E = 4 \cdot \delta \cdot l^2 \cdot f^2 \quad (8)$$

where δ is the density, l is the specimen length, f is the value of the first harmonic of resonant frequencies of longitudinal oscillations in the specimen.

The value δ was determined by hydrostatic weighing and δ is the average density including voids. At the same time density is included into formula (8) describing the propagation of sound speed in the matrix. Therefore, formula (8) for the case of porous materials expresses the effective value of Young's modulus, which is different from the value in formula (7).

$$E_i = E \cdot \frac{\delta_m}{\delta} = E \cdot (1 + S_f) \quad (9)$$

Using (7), (9) and the relationship of swelling to porosity, equation (10) can be written as follows.

$$\frac{\Delta E}{E_0} = \frac{E_m}{E_0} \cdot \frac{1}{(1+S_f)^2} - 1 \qquad (10)$$

Equation (10) transforms into equation (11) without taking into account changes in matrix Young's modulus.

$$\frac{\Delta E}{E_0} = \frac{1}{(1+S_f)^2} - 1 \qquad (11)$$

The same procedure may be used to assess void-induced changes in the shear modulus. Note that at low swelling levels this equation is approximated by a linear decrease in modulus of 2% per 1% swelling.

Figure 5 presents the dependence on swelling of experimentally derived values of relative change in Young's modulus. Predictions based on equation (11) are also shown and are in relatively good agreement with experimental data. There are some differences in prediction and measurement, however, especially at low swelling levels, where other changes such as segregation and precipitation overwhelm the void contributions to changes in modulus. It should also be noted that small changes in density usually result from precipitation.

Finally, the results presented above can be compared with those of previous studies conducted on irradiated metals. There were two previous experimental studies conducted in the U.S. fast reactor program in the early 1970s showing that the shear and Young's modulus decreased ~2% for each percent of swelling (10,11). These results are in good agreement with our results, where at low swelling levels the contribution is essentially linear with swelling content at ~2% reduction per percent swelling.

Various experimental studies conducted in the U.S. fusion materials program showed that the electrical resistivity of copper alloys increased ~1% for each percent of swelling (12-18). Once again these results are consistent with our results and theoretical prediction.

In particular it was shown by Garner and coworkers (13-18) that it was possible to separate the effects of voids and transmutant elements strongly formed in pure copper and various copper alloys during high fluence irradiation. Such separation of contributions arising from voids and other microstructural

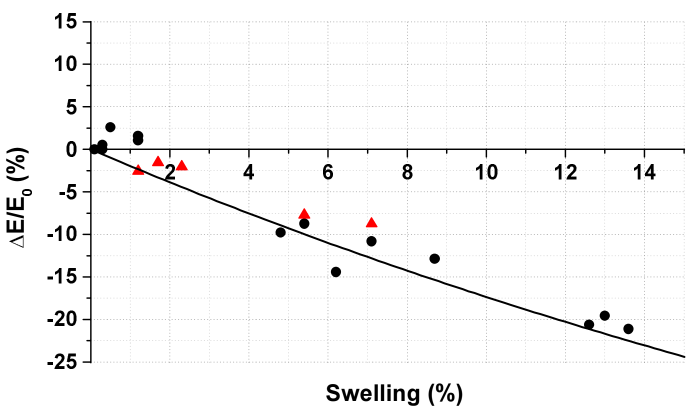

Figure 5. Dependence of relative changes in Young's modulus on swelling of 0.1C-16Cr-15Ni-2Mo-1Mn steel irradiated in the BN-600: ● - low-temperature range $T_{irr.}$<530°C, s - high-temperature range T_{irr}> 530°C, ——— - dependence calculated by the equation (11).

or microchemical features lies at the heart of the technical challenge required to measure swelling in-situ during reactor downtimes. When the steel is not stabilized, however, precipitation-induced contributions will be smaller. Fortunately, the AISI 304 and 316 steels used in the U.S.A. are not stabilized, and therefore are much less prone to form precipitates, especially at the temperatures <400°C experienced in light-water cooled power reactors.

Conclusions

- High dose neutron irradiation at elevated temperatures causes swelling of austenitic steels, which is a dominating effect in changing not only the volume of the metal, but also causing significant and measurable changes in both electrical resistance and elastic moduli.
- Measured changes in electrical resistance are reasonably well described by an equation, which includes only the swelling contribution, and is relatively independent of steel composition, starting condition and irradiation conditions.
- Measured changes in elastic moduli are also well described by an equation in which swelling is the single parameter.
- The changes induced in elastic moduli by a given amount of swelling are approximately twice that induced in the electrical resistance.
- Deviations from void-based predictions are the result of other microstructural components, especially radiation-induced precipitates, and can lead to both over-predictions or under-predictions, depending on the property being measured, the steel composition, and especially the irradiation temperature.
- It appears to be feasible to use the changes induced by voids in these physical properties to nondestructively measure void swelling in-situ in a reactor.
- It also appears that the different responses of the physical properties may allow separation of the void and precipitate contributions. Additional microstructural and microchemical analysis is required on a variety of steels and irradiation conditions in order to facilitate this separation.

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