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Severe embrittlement of neutron irradiated austenitic steels arising from high void swelling

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ABSTRACT

Data are presented from BOR-60 irradiations showing that significant radiation-induced swelling causes severe embrittlement in austenitic stainless steels, reducing the service life of structural components and introducing limitations on low temperature handling especially. It is shown that the degradation is actually a form of quasi-embrittlement arising from intense flow localization with high levels of localized ductility involving micropore coalescence and void-to-void cracking. Voids initially serve as hardening components whose effect is overwhelmed by the void-induced reduction in shear and Young's moduli at high swelling levels. Thus the alloy appears to soften even as the ductility plunges toward zero on a macroscopic level although a large amount of deformation occurs microscopically at the failure site. Thus the failure is better characterized as "quasi-embrittlement" which is a suppression of uniform deformation. This case should be differentiated from that of real embrittlement which involves the complete suppression of the material's capability for plastic deformation.

1. Introduction

It is known that a loss of ductility is expected in austenitic steels when void swelling arises in fast reactor, fusion and light water reactor environments [1]. The only data available at high swelling levels, however, has been drawn from examination of fuel pin claddings and fuel assembly wrappers from fast reactors such as BOR-60, BN-600 and various western reactors [1–15]. In this paper we concentrate on swelling data derived from fuel pin claddings and fuel assembly wrappers from BOR-60 to demonstrate the parametric dependencies of swelling-induced embrittlement in order to anticipate its behavior in fusion environments.

2. Experimental details

Data were developed from conventional flat tensile specimens with gauge sections of 22.5 × 5.5 × 1 mm cut from a hexagonal wrapper and from 2 mm wide ring-pull specimens cut from fuel cladding. Swelling values were determined using an immersion density technique in CCl_4 at room temperature with an accuracy of ±0.2%.

The wrapper was constructed from a titanium-stabilized Russian stainless steel designated Kh18H10T with nominal composition Fe–18Cr–10Ni–0.5Ti. This steel is used in Russian reactors for nuclear applications where AISI 304 would be used in Western reactors. The closest Western analog of the Russian steel is AISI 321 stainless steel. The duct wall was 1 mm thick. The duct was fabricated and then annealed at 800 °C for 2 h, followed by annealing at 600 °C for 1 h.

After removal of fuel and fission products from annealed Fe–16Cr–15Ni–3Mo–Nb cladding of fuel pins irradiated in BOR-60, ring-pull tests were conducted at room temperature using semicircular mandrels inserted into the rings.

3. Experimental observations

3.1. Wrapper specimens

It was observed that the maximum embrittlement zone coincides with the maximum swelling zone in claddings and wrappers rather than with the location of the maximum fluence. This effect is particularly pronounced in fast reactors with lower inlet temperatures such as BOR-60 at 330 °C. As shown in Fig. 1 brittle fracture (defined as strength reduction with zero plasticity) of a Fe–18Cr–10Ni–Ti stainless wrapper at 72 dpa maximum was observed at positions 25–125 mm higher than the core center-plane where the peak flux occurs. As expected, there appears that there is some decrease of strength with increasing irradiation temperature, but as shown in Fig. 1 the primary strength reduction for specimens tested at the irradiation temperature arises from the magnitude of swelling.

As also shown in Fig. 1 testing at temperatures lower than the irradiation temperature (e.g. 20 °C) demonstrates the same dependence on swelling and irradiation temperature but the strength

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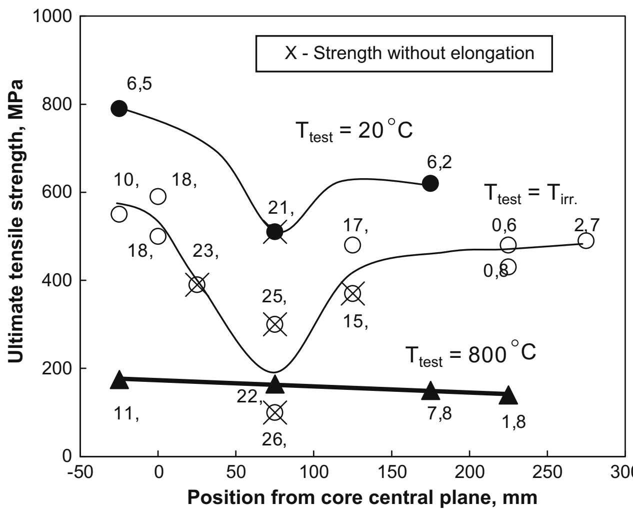

Fig. 1. Ultimate tensile strength of Fe–18Cr–10Ni–Ti stainless steel wrapper specimens irradiated in the BOR-60 to a maximum dose of 72 dpa. Three tensile test temperatures are shown: ● – 20, ○ – 450–550, ▲ – 800 °C. Swelling values of each specimen are given near the points in units of %.

and plasticity values are higher. As expected, the strengths for tests conducted at 800 °C is uniformly much lower than that observed at lower temperatures, but there is an absence of any relationship between strength and swelling, with uniform elongation reaching saturation at ~1.2% for ⩾10% swelling.

As shown in Fig. 2, plotting the strength against swelling for a number of Fe–18Cr–10Ni–Ti wrappers [3,7,9,12,14] tested at temperatures at or below the irradiation temperature reveals that there is a critical value of swelling (15–20%) where the plasticity is essentially zero.

The fracture surfaces of wrapper specimens with high swelling values exhibit characteristic features and well-defined zones. Secondary cracks were found along the grain boundaries (Fig. 3) and within the grains [14]. A transgranular cup-cone morphology was observed on the fracture surface where failure proceeded by micropore coalescence arising from stress concentration between deforming voids (Fig. 4). Similar fracture morphology has been observed in other studies on different stainless steels [1,15].

A short inter-particle distance (particles being defined as voids and precipitates in Ref. [3]) contributes to a quasi-brittle fracture mode such that as swelling approaches 20% the fracture surface rotates to become perpendicular to the strain axis as shown in Fig. 5. In this case the stress-strain characteristics correspond to that of an entirely brittle material that is not capable of strain hardening.

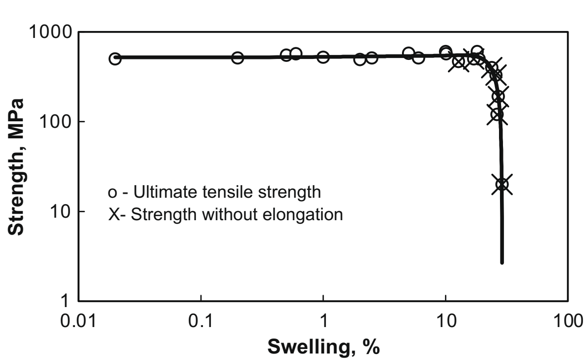

Fig. 2. Strength characteristics as a function of swelling of Fe–18Cr–10Ni–Ti steel at irradiation temperatures of 400–500 °C.

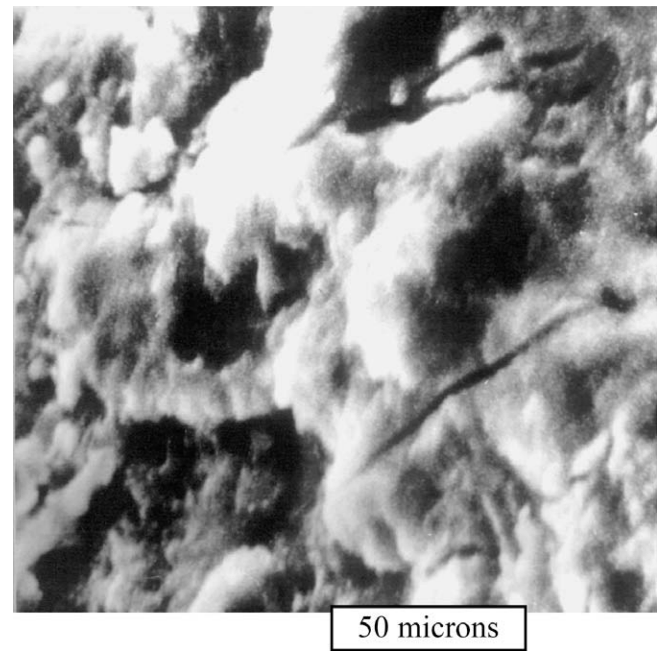

Fig. 3. Microcracks on the BOR-60 wrapper fracture surface at the maximum swelling position of 21.7%.

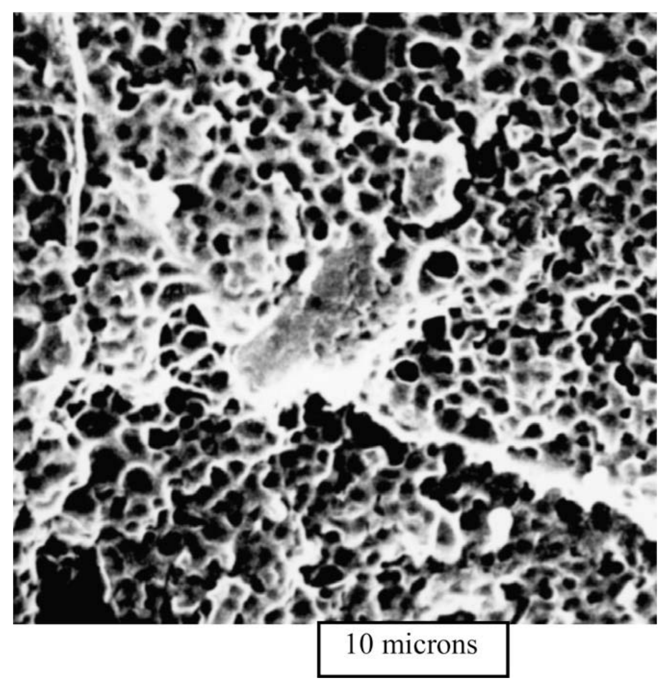

Fig. 4. Nature of the fracture surface in the Fe–18Cr–10Ni–Ti stainless steel specimen at a swelling level of 30%.

Note that the ratio of yield strength and ultimate strength approaches unity when swelling is about 5%.

3.2. Cladding specimens

When using flat tensile specimens the fracture process in highly swollen steels occurs through crack initiation and growth with cracks often originating within the specimen. In ring-pull specimens tested using semi-circular mandrels, the crack leading to

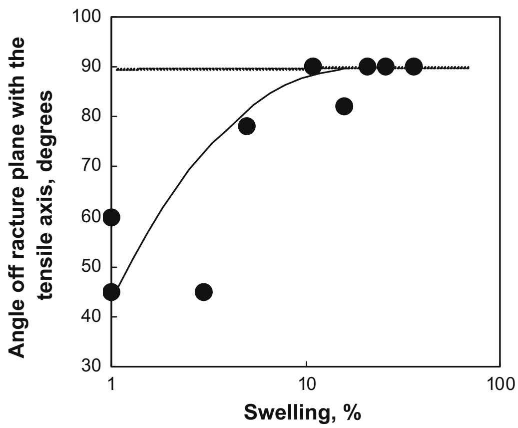

Fig. 5. Angle of the fracture surface with the axis of specimen elongation against swelling of Fe–18Cr–10Ni–Ti stainless steel over the test temperature range of 20–500 °C.

failure is almost always observed to initiate on the inside of the cladding. As shown in Fig. 6 comparing tests on flat and ring specimens it is seen that swelling also reduces the strength of rings but the critical swelling level is lower (5–10%) and the rate of decrease is not as steep as observed in the flat specimens.

The ring specimens are subjected to a number of processes that may promote cracking from the inside surface. First, it is known that a change of the surface state arising from a brittle coating can affect the stress-strain properties of the cladding [9]. Such coatings can arise from oxidation and fission product attack, as well as gas injection from neutron collisions with helium cover gas and fission gases. Second, as the test proceeds and the side walls are straightened, the inner surface is preferentially strained with estimates of the strain approaching 3–6%. Third, due to the large temperature gradient across the fuel cladding, the swelling is usually larger near the inner surface. Both the temperature and swelling gradients can result in tensile stresses in the inner layer leading to crack initiation as a rule along the grain boundaries on the inner surface of the fuel rod cladding [11,13,15].

4. Discussion

There are two major questions to address that arise from these studies. First, how general are the conclusions drawn from the current study concerning the behavior of steels in different chemical and thermal-mechanical conditions, especially when irradiated in different reactors at different temperatures and dpa rates? Is it certain that swelling is always the primary determinant of the embrittlement?

We have compiled data in Table 1 from a number of Russian studies [2,3,5–14] in the BOR-60 and higher-flux BN-350 fast reactors concerning swelling and stress-strain properties of various austenitic steels. In spite of differences in doses, temperatures and dpa rates the critical values of swelling for ring-shaped specimens cut from claddings made of different steels are nearly the same and within the range of 5–10%. This suggests that swelling is the dominant process to initiate cracking in both flat and ring specimens, but ring specimens experience additional conditions not occurring in flat specimens. To the first order the difference is proposed to arise from the side-wall straightening characteristic of ring-pull tests.

Note that one third of the critical swelling value (5–10%)/3 produces linear strains of ~ 2–3% for ring-shaped specimens and (15–20%)/3 = ~5–7% for flat specimens. The difference between the two sets of strain ranges is 3–4% which is comparable to the estimated side-wall straightening strains of 3–6%.

The second question involves the origin of the reduced strength and concurrent reduction in ductility. Although voids initially serve to harden the microstructure [16], large swelling levels allow previously second-order void effects to become dominant [17]. The most consequential of these second-order effects is the strong decrease of elastic moduli at high swelling levels. All of the elastic moduli are well-known to decrease initially at ~2% per each percent of void swelling [18–22]. At >15% swelling this leads to significant reduction in strength.

As a consequence the slope of the elastic region (Young's modulus) of the stress-strain curve decreases, and more even more importantly the barrier strengths of all sinks decrease as the shear modulus likewise decrease. Therefore the yield and ultimate strengths decrease with increasing swelling, even though the

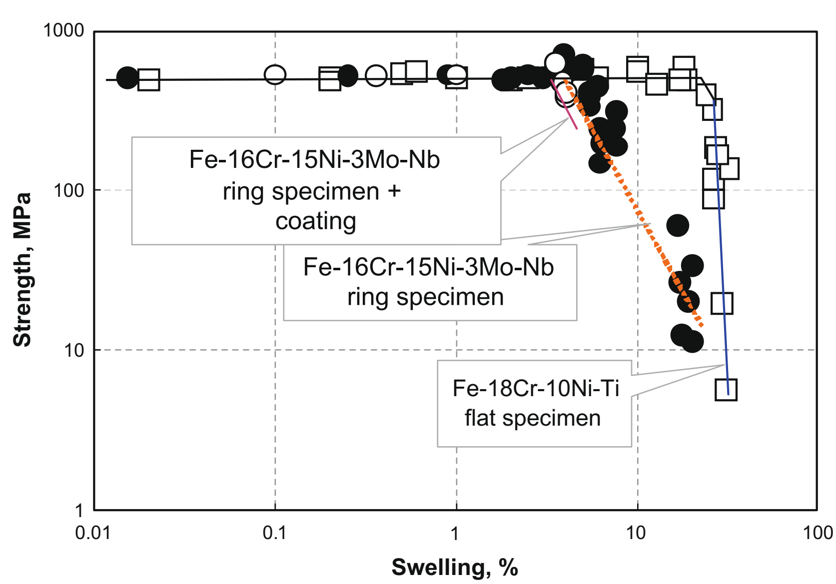

Fig. 6. Ultimate strength of austenitic steels irradiated in BOR-60 at a temperature close to the maximum swelling temperature: Fe–16Cr–15Ni–3Mo–Nb stainless steel – ring-shaped specimens with coating (○), ring-shaped specimens without coating (●); Fe–18Cr–10Ni–Ti stainless steel – flat specimens (□).

Table 1
Relationship between the temperature extremums of the stress-strain properties and swelling of austenitic steel specimens cut out from claddings and wrappers of fast reactors BOR-60 and BN-600 [2,3,5–14].

Steel grade	Thermal treatment	Temperature of the maximum swelling, °C	Temperature of the minimum strength, °C	Temperature of the minimum plasticity, °C	Dose of the drastic strength decrease, dpa	Swelling corresponding to the strength decrease, %
Fe–16Cr–15Ni–3Mo–Nb	Annealing	500–520	500–550	400–650	35–40	8–10
Fe–16Cr–15Ni–3Mo–Nb–B	Annealing	500–520	500–530	450–600	55–60	8–10
Fe–16Cr–15Ni–3Mo–Nb–B + rare earths	Annealing	500–520	500–530	500–530	65–70	8–10
Fe–16Cr–15Ni–3Mo–Nb	Annealing	480	440–550	400–590	30–40	6–10
Fe–16Cr–15Ni–3Mo–Nb	Cold working	445	430–570	430–570	55–60	5–8
Fe–16Cr–15Ni–3Mo–Nb	Cold working	450	450–480	450–480	50	5–7
Fe–16Cr–15Ni–3Mo–Nb–B	Cold working	450	500	450–480	60–65	5–9
Fe–16Cr–15Ni–2Mo–2Mn–Ti–V–B	Cold working	480	440–500	400–500	60–70	5–10
Fe–18Cr–10Ni–Ti	Annealing	500–520	480–530	450–550	35–40	15–20

elongation strongly decreases. Similar behavior has also been observed in pure copper [23].

On a macroscopic level the elongation plunges toward zero although a large amount of deformation occurs locally at the failure site. Therefore we should characterize the failure as occurring by "quasi-embrittlement" which is a suppression of uniform deformation, and this mechanism should be differentiated from that of real embrittlement which involves the complete suppression of the material's capability for plastic deformation.

There is another late-term form of embrittlement that arises at the highest swelling levels. As shown by Hamilton and coworkers segregation of nickel to void surfaces causes martensitic instability in the matrix, especially at the crack tip. This in turn produces a tearing modulus of zero and transgranular failure with the failure surface completely coated with alpha martensite [24]. Others have noted this instability [15].

In this case there is total suppression of capability for plastic deformation. One of the consequences of this instability is that the failure surface becomes perpendicular to the strain direction as was also observed in this study. When this mechanism comes into play, however, the microstructure has already evolved to the point where failure is guaranteed.

5. Conclusions

While voids initially serve to harden the matrix of austenitic steels, increasing levels of swelling cause strong decreases in the elastic moduli, giving rise to a softening of strength in uniaxial tensile tests even as the plastic deformation plunges toward zero as a result of flow localization. On the failure surface there is a large amount of deformation which proceeds via micropore coalescence upon reaching some critical swelling level. This in turn generates internal cracks that eventually lead to failure. In some situations such as in ring-pull specimens there are nonuniform strains developing during the test such that the critical swelling level for zero plasticity is lower than that of uniaxial tensile tests

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