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Effect of neutron flux on the characteristics of irradiation-induced nanofeatures and hardening in pressure vessel steels

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ABSTRACT

Effects of neutron flux (or dose rate) on the characteristics of irradiation-induced nanofeatures in neutron-irradiated pressure vessel steels were occasionally reported. Such effects let one expect that the mechanical properties mediated by the operation of the nanofeatures as dislocation obstacles would also depend on flux. However, there are controversial views on the presence of flux effects on mechanical properties. The present approach is based on the investigation of pairs of samples from the same batch of material for a number of RPV steels including different levels of Cu as well as base and weld materials. The samples of each pair were irradiated at about the same temperature but different fluxes up to about the same fluence, thus automatically revealing potential flux effects. Small-angle neutron scattering and Vickers hardness testing were applied to characterize the nm-scale solute clusters and the resulting irradiation hardening. A number of analytic models of cluster evolution, namely deterministic growth and coarsening, and hardening as well as combinations thereof were applied to interpret the trends extracted from the experimental results. It is found that there are indeed trends of the cluster size and volume fraction as functions of flux but no resolvable trend for hardening. The absence of a flux effect on hardening can be rationalized in terms of size and number density of solute clusters that enter the hardening expressions. Size and number density depend on flux in opposite directions and, therefore, partly cancel out. Flux effects for the whole set of experimental data including low- and high-Cu base metals and welds can be consistently described by a combination of deterministic growth and dispersed barrier hardening using a unique value of the obstacle strength of solute clusters.

1. Introduction

Reactor-immanent neutron irradiation of the reactor pressure vessel (RPV) steel gives rise to material damage and an advancing shift of the ductile–brittle transition temperature. The safety assessment procedure of RPVs is based on the testing of surveillance specimens irradiated at a higher flux (lead factors of 3–10 [1] or 1.5–12 [2]) and, therefore, within a shorter period of time than the RPV material to be evaluated. The required fluence is accumulated in advance and the behavior of the material during the next surveillance interval can be anticipated. According to best practice of safety evaluation, this procedure, although generally accepted, has to be permanently reconsidered on the basis of the latest insight including microstructural evidence.

From the viewpoint of long-term operation (LTO) and life extension of nuclear power plants (NPPs), the issue of flux effects becomes even more important. There is a number of LTO-specific issues:

- In some cases surveillance material was not foreseen or has run out and is not available for additional testing to a sufficient extent.
- Specific long-term irradiation effects that need complementary basic investigation have been reported [1,3–5].
- Parts of the RPV exposed to very low flux accumulate severe levels of fluence under LTO conditions.

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In order to reach levels of the neutron fluence representative of NPPs in LTO conditions within practical time scales, it is necessary to use accelerated tests based on irradiations in material test reactors (MTRs). The corresponding neutron fluxes strongly exceed the range of lead factors allowed in surveillance testing giving the issue of flux effects additional significance.

It is now state of the art to underpin empirically established irradiation effects on mechanical properties with both nanostructural observations and mechanistic understanding [1,6,7]. However, exhaustive studies of flux effects on the irradiation-induced nanofeatures (i.e. mainly defect-solute clusters, nanovoids and dislocation loops) in neutron-irradiated RPV steels are still missing. In some cases, a correlative approach was used to separate secondary flux effects from the primary effect of neutron fluence. According to this approach, flux and fluence were varied within certain ranges, microstructural data were accumulated, and the data were plotted either as a function of fluence in groups of different flux ranges [6–8], or as a function of flux-corrected fluence [9] with the flux correction based on rate-theory considerations [9]. Another more direct and efficient approach to separate flux effects from fluence effects on the characteristics of irradiation-induced nanofeatures is to investigate pairs of samples of the same heat of material irradiated at different fluxes up to the same fluence [3,10,11]. The present study is based on this kind of flux pairs. The primary objective is to add more flux pairs including a wider range of steel composition to the few cases that have been reported so far and to draw conclusions about flux effects from the broader data base.

In order to identify the dominant mode of how neutron flux exerts influence on the characteristics of the irradiation-induced nanofeatures, the study is underpinned with calculations based on specific models. These models aim to link neutron flux and mechanical properties via both the notion of irradiation-enhanced diffusion and the characteristics of irradiation-induced nanofeatures impeding dislocation glide.

The significance of flux effects on mechanical properties is a matter of debate [9,12–17]. This debate will not be addressed directly in the present study. However, it is important to point out that, even in the absence of flux effects on mechanical properties, flux effects on the characteristics of the irradiation-induced nanofeatures are still an essential piece of information. This is particularly true under the aspect of mechanistic understanding of neutron embrittlement of RPV steels and also within the multiscale modeling approach, where the evolution of nanofeatures during long-term irradiation poses an essential link within the chain from atomistic modeling towards 'prediction' of mechanical property changes. From this perspective, it would be meaningful to raise a question of the kind, why there were no flux effects on certain mechanical properties, although the characteristics of irradiation-induced nanofeatures do depend on flux.

The present investigation is based on the characterization of irradiation-induced nanofeatures by means of small-angle neutron scattering (SANS). Indeed, SANS provides statistically reliable and macroscopically representative measures of both size and concentration of the nm-sized irradiation-induced defect-solute clusters. The effect of such clusters on hardening was shown to prevail over the effects of other kinds of irradiation-induced dislocation obstacles such as dislocation loops and nanovoids in particular cases [18]. The volume fraction of clusters that can be resolved by SANS is known to correlate well with the irradiation-induced increase of hardness and yield stress as well as the shift of the ductile–brittle transition temperature [19–23]. The SANS results will be compared with the irradiation-induced increase of Vickers hardness measured using the same pairs of samples and probing a comparable sample volume.

2. Experiments

2.1. Materials

The RPV base materials of types ASTM A508 and 22NiMoCr3-7 were provided by SCK·CEN Mol and AREVA NP GmbH Erlangen, respectively. The RPV weld materials Molytherm and NiCrMo1 UP(modified)/LW320, LW330 (in short NiCrMo1) were delivered by AREVA NP GmbH Erlangen [5] and the Russian-type VVER-440 weld material 10KhMFT was delivered by VTT Espoo [24]. The analyses of these materials are summarized in Table 1.

2.2. Irradiation conditions

The specimens of materials 22NiMoCr3-7 (Klöckner production) and Molytherm (Sulzer production) were irradiated in a commercial pressurized water reactor in standard capsules inserted in the RPV at a position of highest flux with a very low axial flux gradient near core midplane, and in so-called gradient capsules at a position with lower flux due to the axial flux gradient near the core edge [5]. The samples of ASTM A508, NiCrMo1 and 10KhMFT were irradiated within surveillance programs (lower flux) and in MTRs within complementary irradiation programs (higher flux). The essential point for the present investigation is the fact that a flux pair of samples irradiated at lower and higher flux up to the same fluence exists for each of the five materials. The irradiation conditions are summarized in Table 2. The sample code is composed of an indicator for base (BM) or weld (WM) material, the Cu content in wt%, the average fluence for each flux pair and an indicator for lower (L) or higher (H) flux for the sake of easy reference.

2.3. Methods

The SANS experiments were performed for each sample listed in Table 2 and each of the respective unirradiated reference conditions at the beamlines D11 of ILL Grenoble, PAXE of LLB Saclay, and 'Yellow Submarine' of BNC Budapest. The wavelength of the neutron beams was 0.5 nm. In order to separate the magnetic and nuclear scattering contributions, a magnetic field of 1.4 T was applied to the samples perpendicular to the neutron beam direction during the measurements. More details on the experimental setups are given in Refs. [25–27].

The software packages GRASP [28] and BerSANS-PC [29] were used for raw-data treatment including corrections, absolute calibration and separation of the magnetic contribution from the total intensities. Reproducibility and absolute calibration were cross-checked by means of a repeated SANS measurement using different samples from the same material that exhibited an unexpectedly (at the time of the first measurement) strong flux effect. The comparison of the two independent sets of measurement is given in Fig. 1. Both the irradiation effect itself and the flux effect on the irradiation-induced increase of the scattering cross sections are convincingly confirmed.

Transformation into the cluster size domain was performed starting from the difference scattering curves with the unirradiated condition taken as reference using a self-written version of a Monte Carlo fitting routine reported in Ref. [30]. For this algorithm, no preselection of the shape or type of distribution is required. The stability of the inverse transformation is estimated by varying the starting conditions of the fit. This will be indicated below by a scatter band in the calculated cluster size distributions.

Vickers hardness was measured on the SANS samples after completion of the SANS experiments using a load of 98.1 N. Average values and standard deviations of the Vickers hardness number (VHN, here denoted as HV10) were determined from at least 10