

Background

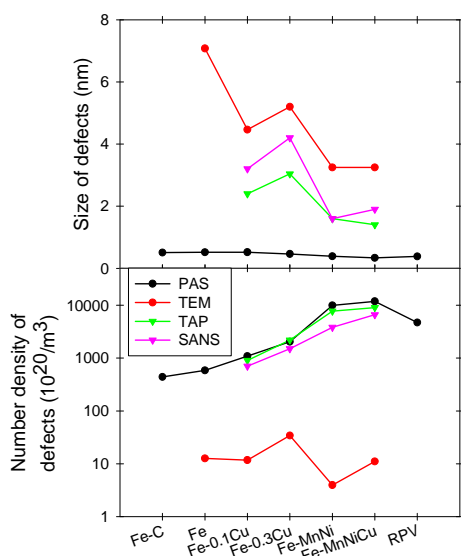
For the safe operation of the NPPS (Nuclear Power Plants), the materials used for the reactors Reactor Pressure Vessel (RPV) steels need to be tested on regular basis as they are prone to embrittlement due to neutron irradiation. The study of defect formation under neutron irradiation and their stability is of paramount importance for the understanding of radiation damage of these materials. To extend the lifetime of the NPPS, there is an urgent need for predictive models based simultaneously on experiments and computer simulation. Nowadays, the progress achieved in advanced experimental techniques such as e.g. TEM (Transmission Electron Microscopy), PAS (Positron Annihilation Spectroscopy), TAP (Tomographic Atom Probe) and SANS (Small Angle Neutron Scattering) enables a detailed investigation of the nano-features induced by irradiation in RPV steels, in addition to the conventional mechanical tests in order to correlate the microstructure changes with the mechanical performance of the RPV steels. This work has been performed within the FP6-PERFECT project aiming at building predictive theoretical tools for the behaviour of reactor components.

Objectives

The main aim of this work is to analyse in a quantitative manner, the nano/microscopic features induced by irradiation under very controlled conditions in well-defined materials. Thus, starting from electrolytic iron, seven different materials were made, covering different model alloys in growing chemical complexity, i.e. pure Fe, Fe-C, Fe-0.1%Cu, Fe-0.3%Cu, Fe-Mn-Ni, Fe-Mn-Ni-Cu and French RPV-steel. They were prepared using argon-arc melting and zone refinement methods. After, an appropriate heat treatment, the materials have been irradiated in the materials testing reactor BR2 at SCK•CEN to a dose of about 0.2 dpa (corresponding to about 60 years of effective operation of a PWR).

Tensile properties have been performed, at room temperature, with a strain rate of about $2 \cdot 10^{-4}$ /s. The positron lifetime measurements were performed using the same procedure described elsewhere [3]. Transmission Electron Microscope using a JEOL, model JEM-2010 of 200 keV and LaB₆ filament, has been carried out at CIEMAT (Spain). Data from TAP analysis are used to reconstruct the atomic-scale chemical microstructure of a material in three dimensions at CNRS-Rouen (France). This method allows the detection of clustering of few atoms although only small volumes are accessible (few thousand of nm³). Thus features with a number density of which is less than 10^{22} m^{-3} can not be detected.

The SANS experiments (using the same samples as PAS) were performed by SCK•CEN at the SINQ facility of PSI as well as at LBL of Saclay, at a wavelength of 0.50 nm with a sample-detector distance of 2 m. The range of the magnitude of the scattering vector, Q , from 0.3 nm^{-1} to 2.7 nm^{-1} was covered. The samples were placed in a saturation magnetic field. For the analysis including corrections related to both sample holder and detector, absolute calibrations as well as separation of magnetic and nuclear contributions of SANS data were performed. In this report, the results obtained from the PAS-, TEM-, TAP- and SANS- investigations [1] are compared and discussed, in relation with the irradiation induced hardening data obtained from the same samples [2].



The number density and mean size of the defects for all materials, using four different techniques.

Principal results

In this report and for brevity only the results obtained after irradiation to 0.1 dpa are described. Using all the above mentioned techniques, the number density and the mean size of the observed/detected defects have been determined and are reported in the figure. As it has been demonstrated earlier, the vacancies are clustering together shown with some alloying elements [2]. Nevertheless, the size, obtained from the lifetime measurement with PAS depends on the amount of vacancies only. These "mixed" clusters should therefore be much bigger than estimated from PAS-results. With positrons it is however possible to see all kind of defects containing vacancies. Even a few vacancies pinned by foreign elements are detectable.

The mean sizes observed by TAP are a bit smaller than those deduced from SANS analysis, while the number density is a little lower with the latter. This can be explained by the fact that very small defects were not detected by SANS. The results of TEM do not correspond to the same kind of defects. While the other techniques detect precipitates and vacancy-clusters, TEM is able to evidence interstitial- and vacancy-loops, but it is not capable of detecting defects/loops with sizes less than 2 nm.

To estimate the irradiation induced hardening of the same materials using the data obtained from the microstructural analysis above, a simple approach such as the Orowan one can be used, as expressed by the equation below:

$$\Delta\sigma = \alpha M G b \sqrt{N d} \quad [1]$$

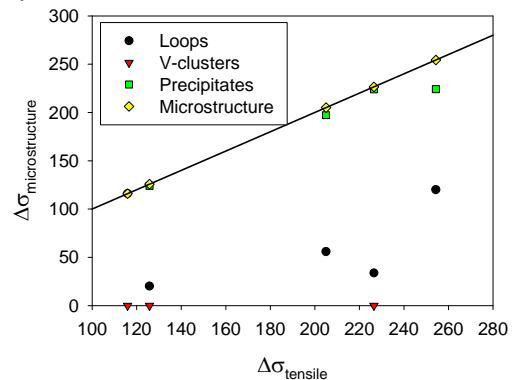
Here M is the Taylor factor (equal to 3.06), G is the shear modulus (71.8 GPa) and b is the Burgers vector (0.249 nm). N and d are respectively the number density of defects and their mean size, that are obtained experimentally. The obstacle strength is given by the constant α . This value can vary between zero and one, and it is strongly dependent on the type of defect.

In all the alloys, the hardening-contribution due to the loops has been calculated using the TEM-results in equation 1. The different kind of defect in these alloys, observed by PAS has been explained in details in [2]. For pure iron, next to loops, only nanovoids have been observed. The contribution to the hardening of the voids has been calculated taking into account the results of PAS. The total hardening is given by the root of the square sum of the two contributions. For the Fe-Cu alloys, it has been demonstrated that the vacancies and copper are clustering together [4]. The size of these vacancy-copper clusters is given by the TAP-results and the number density is given by the PAS-results. For the total hardening, the root of the square sum of the two contributions is used once more. For the alloys containing nickel and manganese some precipitates are formed, but next to this also some of the vacancies are trapped by foreign elements, outside the clusters. These very small defects have also a low contribution to the hardening. For the precipitates the TAP-results are used, while for the very small alloying-element-vacancy defects the number density is given by the difference between the PAS and the TAP, and the mean size is given by PAS. The contribution of these very weak obstacles is a simple additional hardening to the root of the square root of the other two contributions.

The hardening-contribution of the different defects are given on the figure on the right and calculated by following equations:

$$\begin{cases} \Delta\sigma_{Fe} = \sqrt{\Delta\sigma_{loops}^2 + \Delta\sigma_{voids}^2} \\ \Delta\sigma_{Fe-Cu} = \sqrt{\Delta\sigma_{loops}^2 + \Delta\sigma_{Cu-V-ppt}^2} \\ \Delta\sigma_{Fe-(Cu)MnNi} = \sqrt{\Delta\sigma_{loops}^2 + \Delta\sigma_{ppt}^2} + \Delta\sigma_{x-V_{1,2}} \end{cases} \quad [2]$$

This analysis yields to the fact that the SIA-loops are the strongest obstacles inside the investigated alloys, but their influence on the hardening is limited due to their low density. Due to the small size of the precipitates, they behave as rather weak obstacles. Nevertheless they are the main hardening features. The vacancy-type of clusters has a negligible contribution to the hardening, due to their small size.



The hardening-contribution of the different types of defects in function of the hardening measured by the tensile tests.

Future work

It can be concluded that the combination of four techniques leads to a complete description of the defects inside materials. The TEM-results informs about the SIA-loops created during irradiation. A combination of PAS and TAP leads to a full understanding of the irradiation-induced voids and precipitates in the alloys. While SANS is used as confirmation technique for TEM and PAS findings. Within future work the values obtained by the advanced experimental techniques will be used as input for a more realistic theoretical model that is being developed within PERFECT-framework in order to validate the model and extract the exact pinning force associated with each type of nano-feature observed.

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