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On flux effects in a low alloy steel from a Swedish reactor pressure vessel

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HIGHLIGHTS

- Hardness testing is combined with post irradiation annealing at 330, 360 and 390 °C.
- Unstable matrix defects is studied in a reactor pressure vessel steel.
- Comparison between surveillance material and accelerated irradiation.
- No evidence of unstable matrix defects, i.e. not present in studied material.
- Difference in hardness recovery between irradiation conditions found at 390 °C.

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ABSTRACT

This study aims to investigate the presence of Unstable Matrix Defects in irradiated pressure vessel steel from weldments of the Swedish PWR Ringhals 4 (R4). Hardness tests have been performed on low flux (surveillance material) and high flux (Halden reactor) irradiated material samples in combination with heat treatments at temperatures of 330, 360 and 390 °C in order to reveal eventual recovery of any hardening features induced by irradiation. The experiments carried out in this study could not reveal any hardness recovery related to Unstable Matrix Defects at relevant temperatures. However, a difference in hardness recovery was found between the low and the high flux samples at heat treatments at higher temperatures than expected for the annihilation of Unstable Matrix Defects—the observed recovery is here attributed to differences of the solute clusters formed by the high and low flux irradiations.

1. Introduction

Mechanical properties of structural materials have been shown to degrade due to microstructural changes induced by irradiation; these phenomena are specifically evident in the components of nuclear power plants subjected to irradiation by neutrons from the nuclear core. The main effect on the structural materials that make up the Reactor Pressure Vessel (RPV), i.e. both the base material and especially the weld metal, is an evident hardening and embrittlement, implying a weakening of the structural integrity of the RPV. It has been realized for many years that small clusters, commonly called solute clusters or agglomerates are formed within the microstructure due to neutron irradiation. These agglomerates impede on the dislocation motion during plastic deformation of the material, which manifests as an increase of the tensile yield strength, but also as an increase of the ductile-to-brittle transition temperature (DBTT), and a lowering of the other fracture mechanical properties such as the fracture toughness and the upper shelf energy (USE) [1], [2], [3]. The weld metal investigated in this study has a characteristic high Mn-Ni – low Cu composition, leading to the formation of agglomerates mainly composed of Mn and Ni during irradiation. Among the agglomerates of alloying elements that emerge during irradiation, it has been proposed that there may emerge a new type of microstructural feature called Unstable Matrix Defects. Unlike the agglomerates, these defects are proposed to be unstable at the irradiation temperature, making the number density heavily dependent on the incident flux of neutrons from the core [4]. The purpose of this study has been to investigate the presence of Unstable Matrix Defects in irradiated pressure vessel steel from weld metal of the Swedish PWR Ringhals 4 (R4) by combining hardness testing and Post-Irradiation Annealing (PIA).

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The features that emerge following irradiation of low alloyed steels are commonly characterized into groups of agglomerates and matrix features. The group of matrix features has been proposed by Odette et al. [4] to be further divided into Stable Matrix Features (e.g. dislocation loops, nano-voids, etc.) and Unstable Matrix Defects (UMD). Both the agglomerates and the stable matrix features has been characterized to develop in rough proportion to the accumulated neutron fluence [2], while the latter has been found to develop as a function of several factors such as temperature, flux and fluence [4], [5]. The UMD are deemed unstable due to how these features are prone to annealing in-situ at RPV operating temperatures unlike other identified embrittlement mechanisms which appear thermally stable at reactor operating temperatures. Odette et al. proposed that the presence of UMD in the material would have dual properties. Firstly, by acting as a hardening feature, i.e. as a dislocation obstacle, impeding the dislocation motion, but not to the extent as of the agglomerates [2], [4], and secondly, as a sink to the radiation enhanced diffusion that is driving the formation of other features during irradiation such as the agglomerates, stable matrix features and a general diffusion of alloying elements towards e.g. grain boundaries and dislocations [2], [4], [5]. The precise constitution of the proposed UMD is not yet known, it is however theorized that the main population of UMD could exist as both interstitial and vacancy clusters complexed with segregated solutes such as C, N and/or Ni. Of the two proposed configurations, the vacancy clusters are characterized by being the more unstable – dissolving at a faster rate, thus being more probable of the two in relation to the experimental results of Odette et al. [2], [4]. The annihilation of proposed vacancy clusters has been theorized to occur by vacancy emission with recovery times strongly dependent on temperature, cluster size and free surface energy. Odette et al. suggested equation (1) as a simplified description of the number density of the UMD resulting from neutron irradiation

$$N(\phi,\phi t) = \phi\tau\sigma_{\text{UMD}}N_{\text{a}}\left[1-\exp\left(\frac{-\phi t}{\phi\tau}\right)\right] \quad (1)$$

where ϕt is the neutron fluence, ϕ is the neutron flux, τ is the characteristic annihilation time of the UMD, σ_{UMD} is the formation cross-section of the UMD, and N_{a} is the atom density of the lattice ($8.55\cdot10^{28}$ 1/m^3 for iron). This implies that the number density of the UMD will form with a build-up phase and then saturate at a steady-state level, where the steady-state number density is linearly proportional to the neutron flux; for clarity in the conceptual formation of the UMD, see Fig. 1.

In PIA studies presented by Odette et al. [4], [5], heat treatments are combined with micro-hardness testing in which a significant decrease of the hardness could be observed in a timeframe of $1.8\cdot10^4$ s (5 h) at a temperature of 343 °C, and $3.25\cdot10^5$ s (90 h) at 290 °C, proposedly connected to the annihilation of UMD within the material. These studies led to the theory that the UMD would mainly consist of small, sub-nm vacancy clusters that annihilate by vacancy emission at reactor relevant temperatures. Experimental results from Odette et al. [4], [5], on the contribution from UMD hardening can be found in Table 1 for different flux and fluence levels.

Another study of the UMD, or the neutron flux effect on irradiation hardening in general, is presented in two papers by Chaouadi and Gérard in 2011 [6] and 2013 [7]. The study was conducted on a large set of materials where in the 2013 paper, eight different RPV materials were covered in the experimental effort, i.e. a large combination of different RPV-relevant chemical compositions was explored. The experiments consisted of samples irradiated at fluxes between 0.4 and $5.6\cdot10^{14}$ n/s cm^2 and to fluences of

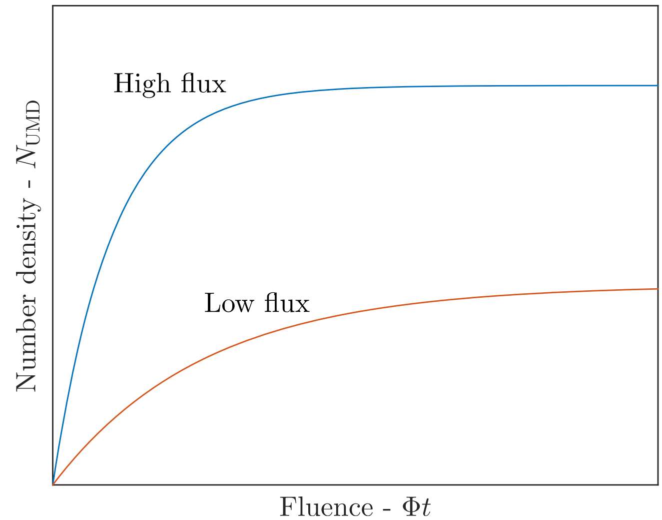

Fig. 1. Conceptual development of the number density of Unstable Matrix Defects at high and low flux levels.

$0.7–9.4\cdot10^{19}$ n/cm^2, which later was exposed to heat treatments at temperatures of 345 °C and 355 °C in combination with tensile testing. The studies concluded that no flux effect on the irradiation hardening of the tested materials could be proved to be present by the experiments, which according to the UMD theory proposed by Odette et al. should have been present [4].

Although surveillance specimens are mainly used for assessment of the structural integrity of the RPV, materials are also irradiated in test reactors to increase the amount of available material. A further understanding of the possible role of UMD is important, as the presence of these defects could affect results from mechanical test results on materials irradiated using different flux.

In the extensive study collaborated by IAEA [8], it was concluded that the sole presence of Ni in RPV-steels does not necessarily make the changes of mechanical properties of the material especially radiation sensitive. However, in synergy with alloying elements such as Mn and Cu, the changes of mechanical properties by the conditions in a nuclear reactor can be quite severe with increasing aging.

Prior studies of the low Cu – high Mn-Ni surveillance material of R4 have shown vast changes in the mechanical properties of the material due to irradiation, DBTT-changes (T_{41J}) of 162 °C, and increases in tensile yield strength of up to 215 MPa at a neutron fluence of $6.0\cdot10^{19}$ n/cm^2 [9]. It has also been shown that large number densities of solute clusters consisting of Mn, Ni, Cu, and Si form under irradiation along with segregation of P to dislocations and grain boundaries [10], [11], both being responsible for irradiation induced changes of material properties. In the study by Styman et al. in Ref. [11] it is shown how post-irradiation annealing treatments of the irradiated surveillance material from Ringhals

Table 1
Experimental results of UMD hardening from Odette et al.

Reactor	Flux ϕ[n/s cm^2] $\times 10^{12}$	Fluence ϕt[n/cm^2] $\times 10^{19}$	DPH$_{\text{UMD}}$[kg/mm^2][a]
Pluto	4	0.22	10
Pluto	46	0.1	8
Pluto	46	0.5	19
BR2	1.4	2.1	12
BR2	100	6.6	30
BR2	100	12.7	32

[a] DPH – Diamond Pyramid Hardness.