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Mechanisms of radiation embrittlement of VVER-1000 RPV steel at irradiation temperatures of (50–400)°C

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H I G H L I G H T S

- Structural elements in RPV steel are studied at different irradiation temperatures.
- Highest number density dislocation loops are formed at low temperature irradiation.
- Carbide transformations are observed only at 400 °C irradiation.
- Embrittlement mechanisms at different irradiation temperature are discussed.

A R T I C L E I N F O

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A B S T R A C T

This work summarizes and analyzes our recent research results on the effect of irradiation temperature within the range of (50–400)°C on microstructure and properties of 15Kh2NMFAA class 1 steel (VVER-1000 reactor pressure vessel (RPV) base metal). The paper considers the influence of accelerated irradiation with different temperature up to different fluences on the carbide and irradiation-induced phases, radiation defects, yield strength changes and critical brittleness temperature shift (ΔT_K) as well as on changes of the fraction of brittle intergranular fracture and segregation processes in the steel. Low temperature irradiation resulted solely in formation of radiation defects – dislocation loops of high number density, the latter increased with increase in irradiation temperature while their size decreased. In this regard high embrittlement rate observed at low temperature irradiation is only due to the hardening mechanism of radiation embrittlement. Accelerated irradiation at VVER-1000 RPV operating temperature (~300 °C) caused formation of radiation-induced precipitates and dislocation loops, as well as some increase in phosphorus grain boundary segregation. The observed ΔT_K shift being within the regulatory curve for VVER-1000 RPV base metal is due to both hardening and non-hardening mechanisms of radiation embrittlement. Irradiation at elevated temperature caused more intense phosphorus grain boundary segregation, but no formation of radiation-induced precipitates or dislocation loops in contrast to irradiation at 300 °C. Carbide transformations observed only after irradiation at 400 °C caused increase in yield strength and, along with a contribution of the non-hardening mechanism, resulted in the lowest ΔT_K shift in the studied range of irradiation temperature and fluence.

1. Introduction

Influence of the reactor pressure vessel (RPV) operating temperature and irradiation during operation causes degradation of mechanical properties of RPV steel resulting in a shift of ductile-to-brittle transition temperature (DBTT) (critical brittleness temperature shift ΔT_K).

Numerous studies [1,2] have shown the ΔT_K shift to be due to two mechanisms, which relative contribution can change during irradiation. These are the hardening and the non-hardening mechanisms. The hardening mechanism of radiation embrittlement is associated with an increase in yield strength due to formation of radiation-induced microstructural elements or phase inclusions being the dislocations pinning points. The non-

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hardening radiation embrittlement mechanism is associated with the lowering of grain boundary cohesion due to segregation processes at the grain boundaries, assisting initiation and propagation of brittle cracks in the steel. Under conditions specific for VVER-1000-type RPV operation (operating temperature ~ 300 °C) both the hardening and the non-hardening mechanisms of radiation embrittlement develop significantly due to formation of dislocation loops and radiation-induced precipitates and due to grain boundary segregations of impurities and alloying elements, correspondingly [3–6].

The irradiation temperature has a significant influence on the processes in steel providing the listed above hardening and non-hardening embrittlement mechanisms; it also can change the ratio between them.

The whole irradiation temperature range (T_{irr}) taking into account the melting temperature (T_m) can be divided into four ranges. Irradiation in each temperature range demonstrates specific features of radiation defects formation and microstructure evolution:

- The cryogenic irradiation: ($T_{irr} < 0.06 \cdot T_m$) – irradiation temperature range implying almost no mobility of interstitial atoms and vacancies
- Low temperature irradiation: ($0.06 \cdot T_m < T_{irr} < 0.3 \cdot T_m$) – irradiation temperature range in which interstitial atoms are mobile while vacancies do not have diffusion mobility
- Intermediate temperature irradiation: ($0.3 \cdot T_m < T_{irr} < 0.6 \cdot T_m$) – irradiation temperature range in which both vacancies and interstitial atoms exhibit mobility, but mobility of vacancies is not high enough to provide a sufficient level of recombination and thus the concentration of point defects exceeds the value for thermal equilibrium.
- High temperature irradiation: ($T_{irr} > 0.6 \cdot T_m$) – irradiation temperature range in which the mobility of point defects is so high that their intensive spontaneous recombination leads to saturation of point defects concentration almost at the thermal equilibrium value.

The melting temperature T_m is about 1500 °C for low alloy steels. Irradiation at temperatures of about (50–140)°C (up to $0.3 \cdot T_m$ ~ 260°C) is the low temperature irradiation for low alloy steel, while irradiation at temperatures of (300–400)°C (from 260 °C up to ~ 790 °C) is intermediate temperature irradiation.

Paper [7] considers the basic features of fast neutron irradiation effect on metals at different irradiation temperatures. Let us discuss the difference of radiation defects formation and evolution of steel microstructure after low temperature and medium temperature irradiation.

1.1. Low temperature irradiation

If the irradiation temperature provides the interstitial atoms mobility in the metal, but not the mass mobility of vacancies, interstitial atoms continue to migrate when the cascade damage regions formed under irradiation are thermodynamically stabilized. At that a part of interstitial atoms is absorbed by radiation defects, where they recombine with vacancies; another part of them recombines with single vacancies in the matrix crystal lattice, and the third part of interstitial atoms goes to other sinks (grain or interphase boundaries, dislocations). At the same time, new interstitial atoms appear in the adjacent cascades. The interstitial atoms entering the radiation defects and recombining there with vacancies lead to a gradual reduction in recombination rate, depletion and collapse of the defects. At that the concentration of vacancies in radiation damage area gradually decreases down to the level of vacancies concentration in the matrix [7]. Increasing the irradiation dose at low temperature irradiation leads to formation of interstitial dislocation loops.

1.2. Intermediate temperature irradiation

When irradiation temperature increases single vacancies emitted by cascade regions at the stage of thermodynamic stabilization are the first to get mobility. The single vacancies diffuse inside the matrix and partially recombine with single interstitial atoms, or go to the sinks, or agglomerate into complexes (clusters). Since the vacancies concentration under neutron irradiation is much higher than the one at equilibrium, migration of substitutional impurities in alloys becomes more intensive [6]. This leads to acceleration of diffusion processes and changes the ratio between diffusion flows of different elements, which enables a wide range of segregation processes, including formation of radiation-induced phases.

Thus, decreasing irradiation temperature of RPV steel should reduce the rate of migration of point defects and atoms, that is, slow down the diffusion processes. On the one hand, it can decrease the intensity of radiation-induced phase transformation and decrease the rate of segregation processes in the alloy. On the other hand, the slowdown of point defects migration due to lower irradiation temperature should decrease the probability of vacancy - interstitial atom annihilation, which increases efficiency of radiation damage, i.e. increases the rate of radiation defects accumulation [8].

Increasing irradiation temperature of RPV steel should increase the rate of atoms and point defects migration, which implies acceleration of impurity elements diffusion. On the one hand, it should accelerate grain and interphase boundary segregation rate of impurities in the metal, as well as intensify formation of radiation-induced phases. Moreover, increase in the mobility of atoms in solid solution with increase in operating temperature might change the size and number density of the initial hardening phases (the carbides), which are stable at lower irradiation temperatures [9]. On the other hand, acceleration of lattice defects migration due to higher irradiation temperature increases the probability of their partial recombination with each other, thereby reducing the efficiency of radiation damage. This should reduce the number density of radiation-induced defects that contribute significantly to radiation hardening at VVER-type RPV operating temperature and, consequently, to radiation embrittlement as the irradiation temperature increases. At that number densities of both radiation-induced precipitates and radiation defects should decrease. For example, the rate of radiation-induced Cu-rich precipitates accumulation was shown to reduce in A533 steel as the temperature increased within the range of (270–310)°C [10].

The objective of this study is to clarify the mechanisms and peculiarities of radiation embrittlement at different irradiation temperatures for VVER-1000 RPV steels.

2. Materials and research methods

The paper considers the influence of the irradiation temperature on the microstructural-phase state and properties of three base metal (BM) RPV materials of 15Kh2NMFAA class1 steel. Table 1 shows the chemical composition of the studied RPV material.

The material was investigated in different states: initial, after accelerated irradiation in the IR-8 research reactor with different fluxes and up to different fluences. Table 2 shows the states of the material studied in this paper.

The samples were irradiated in the IR-8 research reactor with fast neutrons flux, exceeding the one typical for VVER-1000 reactors operating conditions. No flux effect (reduction of radiation