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Prediction of the effects of thermal ageing on the embrittlement of reactor pressure vessel steels

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ABSTRACT

A new method has been proposed for prediction of the effects of thermal ageing on the embrittlement of reactor pressure vessel (RPV) steels. The method is based on the test results for materials in two conditions, namely, aged at temperatures of temper embrittlement and annealed after irradiation. The prediction is based on the McLean's equation and the dependencies describing thermally activated and radiation-enhanced phosphorus diffusion. Experimental studies have been carried out for estimation of thermal ageing of the WWER-1000 RPV 2Cr–Ni–Mo–V steel. The ductile to brittle transition temperature shift ΔT_k due to phosphorus segregation has been estimated on the basis of experimental data processed by the proposed method for the time $t = 5 \times 10^5$ h (more than 60 years of operation) for the base and weld metals of the WWER-1000 RPV.

1. Introduction

Thermal ageing of materials used in WWER reactor pressure vessels (RPVs) has been studied well enough by now and covered adequately in the relevant literature [1–10].

It was shown that 2.5Cr–Mo–V steel and its welds (used for WWER-440 RPVs) are practically not subjected to thermal aging, at least at temperatures not exceeding 350 °C [2–7].

Unlike the above composition, 2Cr–Ni–Mo–V steel (used for WWER-1000 RPVs) is subjected to thermal ageing. The thermal ageing mechanism involves precipitation of carbides and is followed by their coalescence [8–10]. Carbide precipitation results in the material hardening and, as a consequence, in its embrittlement. Carbide coalescence decreases the material hardening and recovers brittle fracture resistance. This process is typical for the operating temperature T_{oper} of WWER-1000 RPVs and brings about a nonmonotonic dependence $\Delta T_k(t)$ where ΔT_k is the shift of the ductile to brittle transition temperature T_k induced by thermal ageing, t is the exposure time at the ageing temperature. For $t \to \infty$, $\Delta T_k \to 0$; the maximum value of $\Delta T_k(t)$ does not exceed 30 °C [8–10]. T_k is determined on the basis of impact test results and corresponds to an absorbed energy of 47 J.

The weld metal of WWER-1000 RPVs is not subjected to thermal ageing [7]. No thermal ageing is normally attributed to low carbon content in the weld metal ($C_C \leqslant 0.07\%$ where C_C is the carbon content in mass%) and, as a consequence, precipitation of carbides during operation. At the same time, it was shown that there is a nonzero dependence $\Delta T_k(t)$ for the weld metal of 2 Cr–Ni–Mo–V steel [11]: indeed, for $t > 10^5$ h, $\Delta T_k \approx 20$ °C, while the max. $\Delta T_k(t) \approx 30$ °C. It results from the test data obtained on thermal sets of surveillance specimens (SS) used in WWER-1000 RPVs. This result seems to suggest the precipitation of phases other than carbides during operation and absence of their complete coalescence. It seems to be connected with phosphorus segregation.

Such a conclusion results from the following circumstances. The materials of some RPV parts (for example, the steel and weld metal of a shell with nozzles) that are not subjected to irradiation have a higher phosphorus content (as per the applicable technical specifications) than irradiated RPV parts (the steel and weld metal of the core and support shells). For in-service RPVs the phosphorus content of material used in the shell nozzles may reach $C_P = 0.020\%$ (C_P is the phosphorus content in mass%).

Phosphorus is known to result in thermal embrittlement of a metal that is caused by phosphorus segregation at different interfaces and grain boundaries [1], while nickel accelerates the process of phosphorus segregation [1].

Estimations based on the kinetics of phosphorus segregation at the grain boundaries and the correlation of segregation with the transition temperature for 2Cr–Ni–Mo–V steel (with $C_P = 0.020\%$ and $C_{Ni} \approx 1.0\%$, where C_{Ni} is the nickel content in mass%) and for its weld metal (with $C_P = 0.010\%$ and $C_{Ni} \approx 1.90\%$) have shown that ΔT_k may exceed 40 °C at $T = 320$ °C and $t = 5 \times 10^5$ h [1,12].

It should be noted that at the operating temperature $T_{oper} = (290–320)$ °C "carbide" and "phosphorus" embrittlement do not add because these processes are time shifted. Thus the maximum value of ΔT_k for "carbide" embrittlement corresponds

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to the time when "phosphorus" embrittlement is weak. ΔT_k due to "carbide" ageing decreases with increasing the time t, while "phosphorus" embrittlement continues to grow as schematically shown in Fig. 1.

Given that the WWER-1000 life extension is currently considered to be up to 60 years ($\approx 5 \times 10^5$ h) and the new RPVs are designed to operate for more than 60 years, the question arises whether the estimations obtained are adequate [1,12].

If the estimations [1,12] are not adequate, the predicted ΔT_k has to be revised for unirradiated RPV components. Conversely, if it is proved that the maximum value of ΔT_k caused by "carbide" and "phosphorus" embrittlement does not exceed 30 °C during the total RPV operation period, the revision of the normative assessment of ΔT_k for unirradiated RPV components will not be required.

The estimations of $\Delta T_k(t, C_P, C_{Ni})$ [1] were verified for experimental data at the ageing temperatures $T > 350$ °C. The calculation of ΔT_k was based on the assumption that for material in the initial condition there are no grain-boundary segregations. In other words, the C-shaped thermal ageing curves were determined for the case of very fast metal cooling from the tempering temperature to a given temperature followed by isothermal exposure [1]. Actually, the RPV shells cool down very slowly after tempering, and, therefore, there are grain-boundary segregations already before operation [13,14]. Hence, the approach suggested [1] may provide rather a conservative estimation of ΔT_k for the real RPV shells. To decrease the conservatism of ΔT_k assessment it is necessary to consider a ΔT_k decrease with increasing the ductile to brittle transition temperature in the initial condition (T_{k0}) due to phosphorus segregations arising during cooling after tempering. A decrease in ΔT_k with increasing T_{k0} is taken into account in our calculations below.

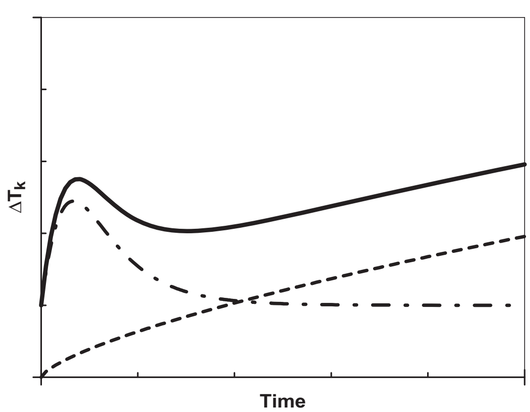

Fig. 1. The scheme for dependence of ΔT_k on time: -·-·-·- ΔT_k. induced by "carbide" ageing; - - - - ΔT_k. induced by "phosphorus" ageing; ——– ΔT_k. induced by the joint action of "carbide" and "phosphorus" ageing.

Another aspect used in the estimation of ΔT_k [1] to be substantiated properly is the value of equilibrium phosphorus segregation (at the grain boundaries or interfaces) at the RPV operating temperature [1]. It is clear that this value cannot be verified experimentally, because at low temperatures corresponding to the RPV operating temperatures (290–320 °C) the time required for reaching the segregation level close to the equilibrium segregation is at least much more than the RPV operation life.

In view of the above, the present investigation is aimed at the development of a new method of $\Delta T_k(t)$ prediction based on the results of testing the material in two conditions: aged at temperatures of temper embrittlement and annealed after irradiation.

2. New method of maximum embrittlement estimation under thermal ageing

2.1. Basic considerations

The main idea of the new method consists in accelerating segregation processes by neutron irradiation at a temperature corresponding to that of RPV operation. Unlike the acceleration of segregation processes due to increasing temperature, neutron irradiation does not practically affect the value of equilibrium phosphorus segregation [15,16]. At the same time, the irradiation intensifies the formation of segregated phosphorus through radiation-enhanced diffusion [17–31].

There are sufficiently representative experimental data obtained by Nikolaev and presented in [19] that demonstrate the enhanced of phosphorus diffusion due to neutron irradiation. Fig. 2 shows experimental data demonstrating the effect of the phosphorus content of 2Cr–Ni–Mo–V steel on ΔT_k and $\Delta\sigma_y$ after neutron irradiation at T^{irr} = 50–80 °C [19]. A strong phosphorus influence on ΔT_k is observed, while $\Delta\sigma_y$ does not change with an increase in phosphorus content. The fact of a strong phosphorus influence on ΔT_k at T^{irr} = 50–80 °C cannot be explained from the point of view of the classical diffusion processes. Hence, phosphorus is translated to interphase boundaries and (or) grain boundaries by some other mechanism that differs from the classical thermoactivated diffusion process. In this case the phosphorus effect is related to directly not only to time, but to a neutron dose or to neutron fluence.

Prior to analyzing this method in more detail, let us consider briefly the major mechanisms responsible for metal embrittlement under neutron irradiation, more specifically, the hardening mech-

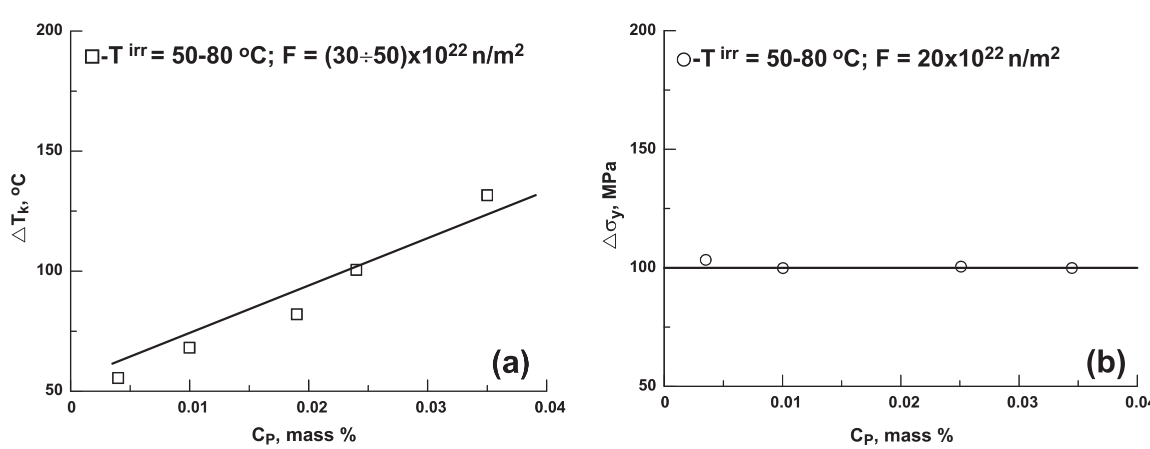

Fig. 2. Effect of the phosphorus content of 2Cr–Ni–Mo–V steel on ΔT_k (a) and $\Delta\sigma_y$ (b) after neutron irradiation [19].