Available online at www.sciencedirect.com
ScienceDirect
Procedia Structural Integrity 2 (2016) 622–625
Structural Integrity Procedia
www.elsevier.com/locate/procedia

21st European Conference on Fracture, ECF21, 20-24 June 2016, Catania, Italy

The effect of heat treatments on the constituent materials of a nuclear reactor pressure vessel in hydrogen environment

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Abstract

A nuclear reactor pressure vessel (NRPV) wall is formed by two layer of different materials: an inner layer of stainless steel (*cladding material*) and an outer layer of low carbon steel (*base material*) which is highly susceptible to corrosion related phenomena. A reduction of the mechanical properties of both materials forming the wall would appear due to the action of the harsh environment causing hydrogen embrittlement (HE) related phenomena. As a result of the manufacturing process, residual stresses and strains appear in the NRPV wall, thereby influencing the main stage in HE: hydrogen diffusion. A common engineering practice for reducing such states is to apply a *tempering heat treatment*. In this paper, a numerical analysis is carried out for revealing the influence of the heat treatment parameters (*tempering temperature* and *tempering time*) on the HE of a commonly used NRPV. To achieve this goal, a numerical model of hydrogen diffusion assisted by stress and strain was used considering diverse residual stress-strain states after tempering. This way, the obtained hydrogen accumulation during operation time of the NRPV provides insight into the better tempering conditions from the structural integrity point of view.

Keywords: nuclear reactor pressure vessel; hydrogen embrittlement; heat treatments; residual stresses and strains.

1. Introduction

A nuclear reactor pressure vessel (NRPV) wall is composed by a bi-material of low carbon steel (*base material*) and stainless steel (*cladding material*), the first one being highly susceptible to corrosion-related phenomena, such as

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10.1016/j.prostr.2016.06.080

hydrogen embrittlement (HE). During operation time, the stress state undergone by the NRPV can be estimated as the combined action of in-service stress caused by (i) the existing gradient of temperature in the NRPV wall and (ii) the remaining residual stress states after tempering heat treatment.

The aim of this paper is to analyze the effect of residual stresses and plastic strains generated after tempering on the HE susceptibility of a NRPV. To achieve this goal a model of hydrogen diffusion assisted by stress and strain previously developed by Toribio et al. (2010) is applied to determine the best conditions of the tempering process from the structural integrity point of view to avoid HE damage.

2. Stress-strain state in the vessel

To perform the analysis, a real cylindrical vessel of a nuclear reactor WWER-440 was considered according to the data previously given by Kostylev and Margolin (2000): inner radius (r_{in} = 1.77 m), width of the stainless steel first layer (w_A= 8 mm), and width of low carbon steel second layer (w_B= 142 mm), and consequently the outer radius must be r_{out} = 1.92 m. According to the results presented by Kostylev and Margolin (2000), the residual stress-strain state is uniformly distributed along the cladding width (w_A) whereas, in the case of second layer of low carbon steel (w_B), it is divided into two intervals: the first one is extended over a zone of width $2w_A$ with tensile stresses (zone B^+) while the second one ($w_B - 2w_A$) with compressive stresses is denoted as B^-. In addition, the in-service thermal-origin stress can be estimated by considering the constraint caused by the own vessel geometry and the different deformation of both layers due to different thermal expansion coefficients of each material. Such deformations are caused by the in-service gradient of temperature ($T_{in} \approx 300$ºC; T_{out} (environment) = 25ºC).

To go further in the analysis of HE, the values of the variables governing the hydrogen diffusion process, namely, the *hydrostatic stress* (σ) and the *equivalent plastic strain* (ε^P), were obtained from the results given by Kostylev and Margolin (2000) in terms of the components of the stress tensor. In Fig. 1 the distributions of both variables σ and ε^P through the vessel width w are shown as a function of the depth from the outer surface, defined as $x = r_{out} - r$, r being the common radial cylindrical coordinate. Thus, $x = 0$ represents the outer surface exposed to the environment whereas $x = w_A + w_B = 150$ mm represents the inner surface of the NRPV wall exposed to the hydrogenating environment.

To evaluate the influence of the tempering heat treatment on the stress-strain state of the NRPV wall, two different tempering temperatures (T_{temp}), 650 ºC and 670 ºC, and two different tempering times (t_{temp}), 1 and 100 hours, were considered. Although a slightly higher residual stress was obtained for T_{temp} = 650 ºC, obtained results showed that σ distributions are quite similar for the two T_{temp} considered. Regarding the distribution in Fig. 1a, three intervals can be observed: (i) positive gradient of σ through wall A –stainless steel–; (ii) slight gradient of σ through wall B^+; (iii) slight negative gradient of σ in the zone B^-. On the other hand, the lower the t_{temp}, the higher the residual stress σ, mainly in zone B^+ (Fig. 1b).

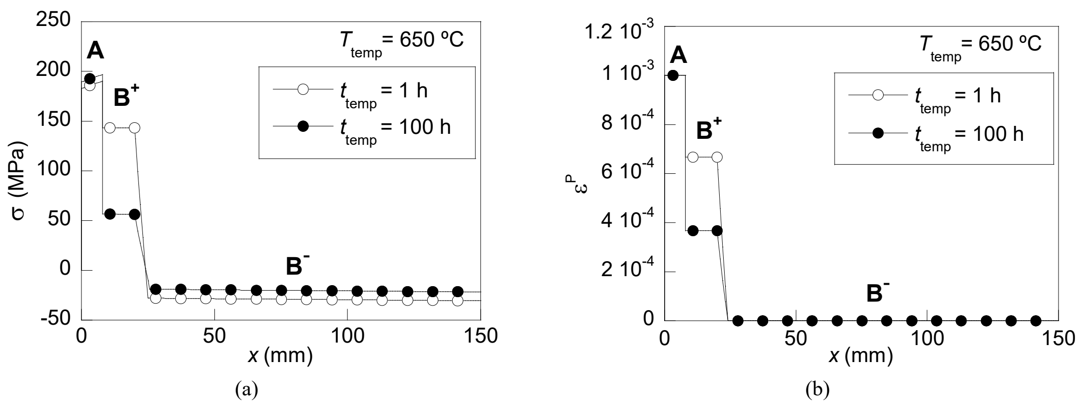
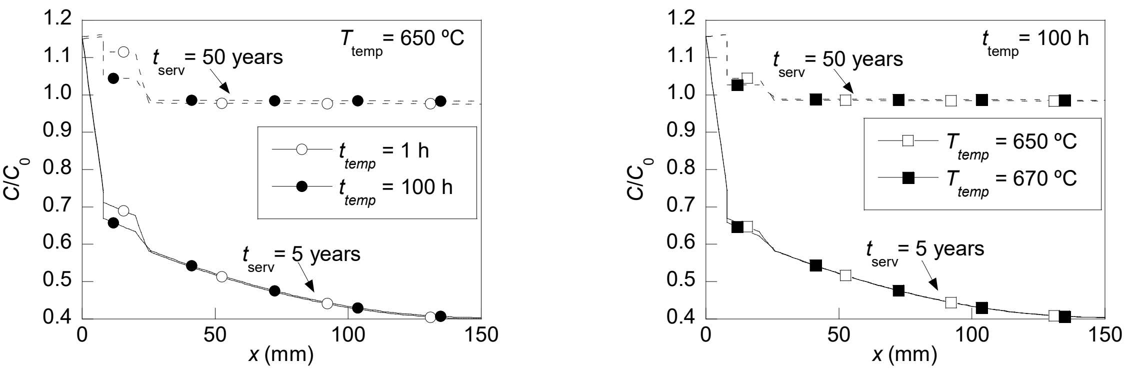


Fig. 1. Distribution of total residual hydrostatic stress σ (a) and equivalent plastic strain ε^P (b) through the vessel width, for different tempering times (t_{temp} = 1 and 100 hours) for a given tempering temperature (T_{temp}= 650 ºC).

3. Hydrogen Embrittlement

Numerical modeling of hydrogen diffusion assisted by stress and strain in the NRPV wall WWER-440 was based on a numerical model developed by Toribio et al. (2010). A 1D approach of such a model is adequate in this case, as explained by Toribio et al. (2011). On the basis of this model, hydrogen diffusion is governed by: (i) negative gradient of hydrogen concentration, (ii) positive gradient of hydrostatic stress and (iii) positive gradient of hydrogen solubility depending on plastic strain. Simulations were carried out with the following values of the parameters: partial molar volume of hydrogen (2 cm^3/mol, according to Hirth (1980)), hydrogen diffusion coefficient at zone A, $D_A = 6 \cdot 10^{-12}$ m^2/s (Mine et al. (2009)), and for the zone B, $D_B = 1 \cdot 10^{-10}$ m^2/s (Nagao et al. (2000)). Finally a service temperature T = 300 ºC was used.

Fig. 2a plots the radial distribution of normalized hydrogen concentration (C/C_0) for different times (t_{serv}) during life in-service of the nuclear reactor obtained with a tempering treatment of T_{temp}= 650 ºC and different tempering times (t_{temp}= 1 h; t_{temp}= 100 h). The hydrogen content of the NRPV progressively increases with service time (Fig. 2a). The main difference in C distribution is localized over the zone B^+, where the previous trend is once again observed: the lower the tempering times, the higher the hydrogen concentration, which suggests a direct influence of the residual stress generated after tempering. In addition, the difference of hydrogen concentration in B^+ for different values of tempering time (t_{temp}) increases with t_{serv}. To complete the analysis, radial C distributions obtained for two tempering temperatures (T_{temp}= 650 ºC and T_{temp}= 670 ºC) for a given tempering time (t_{temp}=100 h) at diverse service times were compared in Fig. 2b. The influence of T_{temp} on C distribution is less accused than the effect produced by t_{temp} (cf. Fig. 2a). Thus the tempering parameters modify residual stress states and, consequently, affect the C in the NRPV wall, the effect of T_{temp} being more intense than the effect of t_{temp}. In both cases these effects are localized at zone B^+. That is especially important because the material of such a zone (low carbon steel) exhibit a higher susceptibility to HE phenomena.



(a) (b)

Fig. 2. Radial distribution of hydrogen concentration, normalized with the hydrogen concentration in a material free of stress and strain (C_0), for (a) a fixed T_{temp} (650 ºC) and two different t_{temp} at diverse instants of reactor life in-service: 5 years (continuous line) and 50 years (dashed line); (b) a fixed t_{temp} (100 hours) and two different T_{temp} at diverse instants of reactor life in service: 5 years (continuous line) and 50 years (dashed line).

4. Discussion

Main differences in C radial distributions (Figs. 2a and 2b) are placed at the zone of the NRPV wall where tensile residual stress were generated (zone B^+). Consequently, the time evolution of C was analyzed at one point representative of such a zone (Fig. 3), placed close to the interface of zone A (stainless steel) and zone B (low carbon steel) for different t_{temp} with a fixed T_{temp}. There, C is progressively increased until a maximum asymptotic value (equilibrium state), which is reached for long exposures times to the harsh environment. However, as shown in Fig. 2a, the amount of hydrogen in the NRPV increases as t_{temp} decreases, those increments being more noticeable as t_{serv} becomes higher. So, it can be considered that in those cases (low t_{temp}), HE process will be enhanced and therefore it is highly recommended to carry out the tempering treatment using long t_{temp}.

Fig. 3 reflects the delaying effect of NRPV zone A on hydrogen accumulation acting as a barrier against hydrogen diffusion towards the zone B (more susceptible to HE). Taking into account that a nuclear reactor is stopped every one or two years for maintenance purposes, the analysis of HE should be restricted to such a period of time. During this stop the hydrogenating source disappears and, therefore, the hydrogen accumulated after the maintenance stop tends to diffuse towards outer points of the NRPV. However, it is impossible to ensure that the material vessel is completely free of hydrogen. So, in the next working cycle the amount of hydrogen would reach higher values than the values obtained during the first cycle.

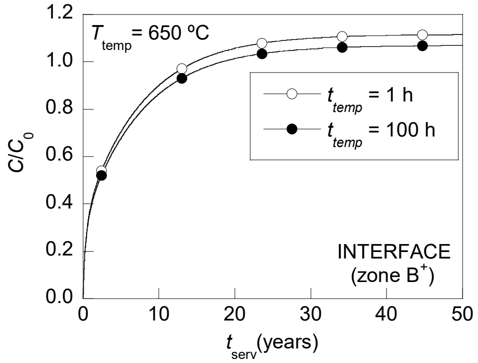


Fig. 3. Time evolution of hydrogen concentration in a point placed at zone B^+ near to the interface for two different t_{temp} (1 and 100 hours).

5. Conclusions

In-service stresses (generated by the gradient of temperatures between inner and outer points of the nuclear reactor pressure vessel) play a relevant role in hydrogen embrittlement phenomena due to the fact that they present a gradient of stresses which enhances hydrogen diffusion towards the inner points of the vessel.

The tempering conditions (*temperature* and *time*) are key issues in the stress reduction process developed during tempering: the higher the temperature or the time, the lower the hydrogen concentration and thus the hydrogen embrittlement susceptibility of the nuclear reactor pressure vessel, the effect of tempering time being more important.

Ackowledgements

The authors wish to acknowledge the financial support provided by the following Spanish Institutions: Ministry for Science and Technology (MCYT; Grant MAT2002-01831), Ministry for Education and Science (MEC; Grant BIA2005-08965), Ministry for Science and Innovation (MICINN; Grant BIA2008-06810), Ministry for Economy and Competitiveness (MINECO; Grant BIA2011-27870) and Junta de Castilla y León (JCyL; Grants SA067A05, SA111A07 and SA039A08).

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