

Correlation Between Measured Mechanical and Microphysical Parameters for an Irradiated Pressure Vessel Material

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Abstract. Results of recent mechanical [1] and neutron scattering studies [2] have been combined in order to search for useful empirical correlations between radiation induced changes of mechanical and microphysical properties. The investigated RPV steel contained 0.17 wt% copper, it was irradiated in a test reactor to four different neutron fluences and subjected to various annealing schedules.

1. Introduction

Reasons for considering annealing procedures as applied to reactor pressure vessels have been given large space in the corresponding literature [3,4]. One of the aims of such programmes would be plant life extension. Along this line, work has also been carried out at PSI to obtain answers to the questions 'what effect would have various heat treatments on a typical medium Cu-content RPV steel', and 'would these effects be significant enough for the implementation of extended specific case studies in Switzerland' [5,6].

Investigations at PSI have been carried out along two lines: (i) by studying mechanical properties responding to irradiation and annealing [1,7,8] and (ii) by measuring the evolution with heat treatments of the microstructural parameters, such as the radius of irradiation induced precipitates [2,9,10]. The objective of this work is to combine the data resulting from these approaches, to study the trends in such empirical correlations, in order to possibly achieve a deeper understanding of steel embrittlement mechanisms.

In the following two sections the specific material investigated, irradiation and heat treatment procedures and the mechanical testing and neutron scattering results are summarized. In section 4 we discuss the correlation of defect size and density with Charpy parameters, and section 5 contains the conclusions.

2. Material, irradiation and heat treatments

In this section some relevant information on the material investigated, the specimens, irradiation conditions and annealing schedules will be given. Related investigations have been carried out in the frame of a programme which has been defined and discussed in refs. [5,6].

The type of RPV steel studied is a forging of 270 mm thickness, heat treated to 875°C in 7h, hold for 10h, water quenched, tempered to 635°C in 6h, hold for 11h, furnace cooled to 300°C and then air cooled. Alloy and impurity elements of interest in wt % are Si0.29, Mn1.47, Ni0.87, P0.02 and Cu0.17.

The standard loading of the irradiation capsule consisted of 30 Charpy, 18 precracked Charpy, 6 tensile specimens, two 3-point-bend-bars and a variable number of mini specimens to serve for various microphysical investigation techniques.

Irradiation conditions have been controlled by thermocouples and neutron monitors and were otherwise determined by several factors: (i) the position relative to the core, which together with the tailoring elements determined the overall neutron flux (125 x flux level of a typical PWR) and the neutron spectrum for $E > 1\text{MeV}$. (ii) The time at a given flux determined the fluence; the nominal values have been chosen to be 0.5, 1.7, 3.0 and $5.0 \times 10^{19} \text{cm}^{-2}$. (iii) The specific construction of the irradiation capsule which was part of a gas loop circulating a gas mixture of helium/nitrogen at normal pressure. Together with the reactor- γ -intensity heating up the steel specimens, this formed a device permitting to keep the specimen temperature at a level of 290°C , achieved by controlling the gas mixture. The temperature profile over the specimen height of 37cm has been achieved to stay within $\pm 5^\circ\text{C}$ in 80 % and $\pm 7^\circ\text{C}$ in 100 % of the irradiation time by choosing a specific gas gap geometry between the specimen holder and the capsule.

Heat treatments $A = 455^\circ\text{C}/168\text{h}$ and $A' = 455^\circ\text{C}/1.5\text{h}$ have been chosen after a detailed study of isothermal hardness recovery determinations [1]. With these schedules a number of individual specimens have been annealed in the hot cell. However, for purposes of obtaining full sets of specimens with IAR condition, i.e. after an intermediate A treatment (at half of the above mentioned fluence values) of the fully instrumented irradiation capsule in the hot cell, the capsule was reintroduced into the reactor for completion of the reirradiation (R) cycle (second half of the target fluence).

3. Mechanical testing, Neutron scattering

This section summarizes some of the results of systematic studies by means of impact testing on Charpy specimens [1,7] on the one hand and, essentially on the same specimen material, by means of small angle thermal neutron scattering [2,9] on the other hand.

Charpy specimens in the conditions U, UA, I, IA, IA' and IAR have been tested and the data have been analysed by using the usual $y = \tanh x$ function. Figure 1 shows the plots of the 41J-transition-temperature values vs fluence. The main conclusion that may be derived from this figure is the possibility of separating off an aging contribution from the recovery process, when considering the difference between the IA and IA' curves. For a detailed discussion of this point we refer to ref. [1]. However, a precise statement about embrittlement mechanisms cannot be drawn from such measurements. The only chance for reaching such objectives would be found in applications of microphysical techniques. In Table 1 are indicated the dosimetrically determined fluence values reached in the corresponding irradiation experiments as well as the measured temperature shifts $\Delta_{41J}T$ for the conditions I and IAR.

Small angle neutron scattering is a powerful mean to observe defects with well defined contours in the diameter range of 5 - 500 Å and to determine their density and size distribution, eventually giving information on composition and shape as well. In the present context voids, various point defect agglomerates and precipitating second phase particles may be produced as a result of fast neutron bombardment. These will appear at the origin of additional diffracted intensity as compared to the unirradiated, otherwise identical sample. For the present work the experimental facility [11] at the FRG-1 reactor in Geesthacht was used, the selection of 'cold' neutrons ($E < 5.2\text{meV}$) from the thermal neutron beam and the detectors measuring the elastically scattered intensity from the magnetized sample are the same as in refs. [2,9,10].

The physical-metallurgical information is summarized in the pair of scattering curves as shown in Figure 2. Such curves are obtained for each sample condition, showing the radiation induced scattering for two scattering geometries with respect to the direction of magnetization in the sample. If the inhomogeneities embedded in the field of homogeneous magnetization have the same size as the underlying spatial inhomogeneities, called 'particles', what should reasonably be the case for voids or non-magnetic precipitates as well, the two cross section curves in the figure are only shifted from each other, and the shift characterizes the particle composition. The size distribution of the particles can be deduced from the curves by a straightforward procedure [9], the accuracy of which improves with better statistics (more measuring time) and broader angular range ($\kappa = (4\pi/\lambda)\sin(\theta/2)$, where θ is the scattering angle). In Table 1 and in the following section we limit the discussion to the average particle radii \bar{R} and the total volume fractions f of the particles (volume of one particle times their

number density) for the various sample conditions. On the base of the measured shift of the curves the particles are identified as copper containing precipitates.

4. Correlations between Charpy shift, defect size and density

Empirical correlations may be useful for extrapolation purposes into domains where no measurements exist, or they may inspire further research and give useful indications (in this context) for modelling embrittlement mechanisms. However, it should be stated clearly that such correlations hold true only for the specific material under investigation.

In this sense Table 1 was used to plot in Figure 3 the average radius \bar{R} and the volume fraction f vs the Charpy shift $\Delta_{41J}T$ which is used here as a measure of the degree of embrittlement. By taking $x_I = \{\text{flux, fluence, spectrum, irradiation temperature}\}$ and $x_A = \{\text{annealing temperature, time}\}$ as independent variables, we may then describe in a phenomenological way, say, $\bar{R} = F_{mat}(x_I, x_A)$, $mat = \{\text{material dependent parameters}\}$. The Charpy shift is now described as the functional $\Delta T = G_{mat}(x_I, x_A, \bar{R})$. Depending on whether $x_A = 0$ or $x_A \neq 0$ we find, that

$$\Delta T_{(I)} = G_{mat}(x_I, 0, \bar{R}|_{x_A=0}) \quad \text{and} \quad \Delta T_{(IAR)} = G_{mat}(x_I, x_A, \bar{R}|_{x_A \neq 0})$$

represent two curves in the $f, \bar{R} - \Delta T$ plane (the projections along the x_I -axis of curves with $x_A = 0$ and with $x_A \neq 0$). In order to draw conclusions on the base of these curves, one has to be aware of the fact that 2 points on the curves having the same value $f = f_0$ do not belong to the same fluence, so that the corresponding 'samples' cannot be used as qualifiers for two different heat treatments related to the same irradiation dose. On the other hand, interrelation of the parameters may lead to useful observations; in the above example a measured value f_0 for an IAR specimen is much more 'serious', from the point of view of embrittlement, than the same f_0 measured for an I-specimen, since it implies a much larger ΔT .

There are some further observations which can be taken from Figure 3:

- There seems to be no (or only weak) correlation between ΔT and \bar{R} . Hence, measuring only \bar{R} in company with Charpy parameters would not be sufficient to predict the degree of embrittlement.
- In the case of the I-condition: for a fixed \bar{R} (which is more or less fulfilled in this case), low values of f , i.e. a low number density of defects, produces less embrittlement and vice versa. Such a monotonic, approximately linear correlation was to be expected for smaller doses. However, there is a transition to a saturation value (approximately the nominal Cu-content of the material investigated) where any correlation between ΔT and f seems to be lost: this substantiates the assumption that inhomogeneities causing embrittlement consist mainly of precipitating copper.
- In the case of the IAR-condition: at least in the domain of measurement there seems to exist a linear correlation $f - \Delta T$ which however differs from the I-condition. This correlation may, of course, also saturate for higher shift (fluence) values.
- For a given value of $\Delta_{41J}T$ (and constant \bar{R}) the number density of defect agglomerates in the I case is much higher than in the IAR case.

The question of how these observations have to be validated in the light of existing models of interaction between defect agglomerates and dislocations [12], may be answered via the correlation $f, \bar{R} - \sigma_y - \Delta T$ ($\sigma_y = \text{yield strength}$). The latter correlation $\sigma_y - \Delta T$ is linear [8], which together with the Russel-Brown theory $\bar{R} - \sigma_y$, indicates a qualitative agreement with the trends as found in Figure 3.

5. Conclusions

Unless for a given material a maximum set of correlations between all 'independent' parameters is being considered and worked out, it will be difficult to draw generally valid, material independent conclusions on embrittlement mechanisms. The attempt was made in this work to combine results of mechanical testing and of microphysical studies on a specific RPV steel. We note the following points: