Fracture Mechanics Concepts for Evaluation of Irradiated Reactor Pressure Vessel Material Behavior

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Abstract

Within the German research program Integrity of Components (FKS) irradiation experiments were performed with ferritic reactor pressure vessel (RPV) steels and welds. The materials cover a wide range of chemical composition and initial toughness to achieve different susceptibility to neutron irradiation. Different neutron flux was applied and the neutron exposure extended up to $8 \cdot 10^{19} \text{ cm}^{-2}$. The change in material properties was determined by means of tensile, Charpy impact, drop-weight and fracture mechanics tests, including crack arrest. The results have provided more insight into the acting embrittlement mechanisms and shown that the fracture mechanics concept of the Code provides in general an upper bound for the material which can be applied in the safety analysis of the RPV.

1 Introduction

For reactor pressure vessels fine grained ferritic steels are used in form of forged rings or rolled plates. They are joint together by circumferential and/or longitudinal welds. During service the materials in the core belt-line region are exposed to conditions which cause a change in material properties and thus a reduction of the safety margin. The main effect is the neutron irradiation, which leads to a strengthening of the material and to a reduction in toughness.

In the frame of the overall aim of the research program "Integrity of Components" (FKS) the focal point of the irradiation program was to investigate the parameters which are responsible for the change in material properties and to describe the consequences quantitatively as far as possible.

On the basis of the material properties which are determined during the acceptance tests of the component the K_{Ic} curve and the K_{IR} curve, resp., of a particular material in the unirradiated and in the irradiated condition can be established by means of the reference temperature RT_{NDT} .

Those boundary curves are being compared with the load pathes during service and under emergency cooling conditions, i.e. the stress intensity as the result of the loading conditions and the postulated flaw size. In case of irradiation the change in material properties shifts the curves which describe brittle fracture (K_{Ic} , K_{IR}) to higher temperatures as proposed by the fracture mechanics concept of the Code as presented in KTA 3203, U.S. Reg. Guide 1.99, 10 CFR 50.61 so that the safety margin to the load pathes in terms of temperature is reduced.

2 Determination of the Change in Material Properties

The tests which are necessary to determine the change in material properties comprise Charpy impact, drop-weight, tensile and fracture mechanics tests.

The RT_{NDT} is determined from the Charpy impact test and the drop-weight test in the following manner:

$$T_1 = T_{NDT} \quad \text{from drop-weight test} \quad (1)$$

$$T_2 = T_{68} - 33 \text{ K} \quad \text{energy criterion} \quad (2)$$

$$T_3 = T_{0.9} - 33 \text{ K} \quad \text{lateral expansion criterion} \quad (3)$$

The reference temperature results in

$$RT_{NDT} = (T_1, T_2, T_3)_{\max}. \quad (4)$$

Using that reference temperature a conservatively enveloping K_{Ic} curve for static crack initiation can be determined for the initial (unirradiated) material state as proposed in the Standards KTA 3201.2 and ASME BPVC, Section III, App. G.

It is assumed that the shape of the curve is maintained also in the irradiated condition and that for all material states the curve is only shifted on the temperature axis corresponding to the reference temperature RT_{NDT} or the adjusted reference temperature.

From the Charpy impact tests a complete energy temperature curve is established from which the temperature in the transition region at the level of $A_v = 41 \text{ J}$ is derived. The K_{Ic} curve of the initial material state is adjusted by the shift ΔT_{41} , e.g. the reference curve is shifted to higher temperatures by the amount of ΔT_{41} .

Besides the effect of neutron irradiation on transition temperature shift the drop in upper shelf energy (USE) has to be regarded. The upper shelf energy is taken alternatively to elastic-plastic fracture mechanics data to evaluate safety against ductile crack initiation. Corresponding to international Codes and experience, catastrophic failure of a component in the regime of elastic-plastic material behavior can be excluded if an upper shelf energy of 68 J can be assured.

To describe the temperature margin in relation to the experimentally determined fracture toughness data often the so called index temperature $RT_{K_{Ic}}$ was widely used [1].

In that case the K_{Ic} curve of the Code is drawn as an envelope to the measured data from which the index temperature can be determined at a K_{Ic} value of $59.2 \text{ MPa}\sqrt{\text{m}}$.

Corresponding to that procedure a curve for dynamic crack initiation and crack arrest can also be determined on the basis of experimental data.

The temperature margin between experimental data and the K_{Ic} curve adjusted to RT_{NDT} can be expressed in the form of

$$\Delta T = RT_{NDT} - RT_{K_{Ic}}. \quad (5)$$

The adjustment of the fracture toughness curves for irradiated materials can be performed corresponding to the Code on the basis of the ΔT_{41} shift.

The adjusted reference temperature $RT_{NDT}(\text{adj.})$ will be obtained by the equation.

$$RT_{NDT}(\text{adj.}) = RT_{NDT} + \Delta T_{41} \quad (6)$$

3 Materials

Materials with high toughness are required to withstand service and emergency conditions. Therefore reliable control of material manufacturing, especially during welding is necessary which was the aim of the development of reactor pressure vessel (RPV) steels [1].

Regarding the toughness reduction during service - especially as a result of neutron irradiation and postulated flaws - requirements for the materials in the initial state were established which guarantee also in the end-of-life condition (EOL) sufficient safety margin against brittle crack initiation.

For the research program chemical composition, heat treatment as well as welding parameters were selected to achieve a wide variety of material toughness covering the range from optimized heats to rejected parts, so that the material concepts could be verified also taking neutron irradiation into consideration. In addition, materials beyond the specified limits were investigated to cover the degrading influences of manufacturing and the change in properties due to long range service conditions.

4 Effect of Neutron Irradiation on Material Behavior

Neutron irradiation can cause damage in the microstructure such as fine disperse precipitations and voids which lead to an increase in strength and decrease in toughness. The consequences for the parameters responsible for brittle crack initiation are the shift of the Charpy energy curve, the Nil-Ductility Transition temperature derived from drop-weight test and the shift of the K_{Ic} curve to higher temperatures, fig. 2a and 2b, [1]. The low K_{Ic} values cannot always be enveloped due to the limitations of the curve. However, it is more important to envelope data in the rising branch of the curve.

From data of irradiated materials curves were developed which show a correlation between ΔT_{41} and the fluence for materials of different chemical composition.

According to a statistical evaluation of the U.S. surveillance data a significant contribution to irradiation embrittlement of the combination of the elements Cu and Ni in steel was recognized which is incorporated in U.S. Reg. Guide 1.99 Rev. 2.

On this basis the shift in transition temperature can be calculated by using a chemistry factor CF and a fluence factor FF.

In the German rule KTA 3203 the shift of the transition temperature is predominantly determined by Cu. The influence of phosphorus ($P > 0,012\%$) is considered by a Cu equivalent.

While the design curves of Reg. Guide 1.99, Rev. 2 represent mean curves for the temperature shift, the curves in KTA 3203 provide an upper bound for a given chemical composition.

Figure 2a: Effect of the neutron irradiation on Charpy energy and NDT temperature for KS 01 base material

Figure 2b: Effect of the neutron irradiation on fracture toughness of KS 01 base material

To check the applicability of U.S. Reg. Guide 1.99 for the FKS materials the shift in transition temperature ΔT_{41} (Reg. Guide) was calculated on the basis of the fluence of the particular specimen set and the chemical composition (Cu, Ni) and then compared with the measured value ΔT_{41} (experimental). The deviation of the calculated value from the experimental value is called "residual shift" (RS) and is represented in the form

$$RS = \Delta T_{41} (\text{experimental}) - \Delta T_{41} (\text{Reg. Guide}). \quad (7)$$

If the predicted value is equal to the measured value $RS = 0$. RS is negative if the prediction is conservative and positive for a non conservative prediction.

From the U.S. data base a scatter band of ± 29 K was recognized for base materials and ± 36 K for weld metals [2]. In this temperature range fall 95% (2 Sigma) of all available data. Up to a fluence of $2 \cdot 10^{19} \text{ cm}^{-2}$ all values for the FKS base materials fall into this scatter band, fig. 3. At higher fluence the prediction is not conservative for most of the data and some data fall even out of the scatter band. The weld metals show a similar behavior. A reason for this

Figure 3: Deviation of the experimentally determined shift in transition temperature of the FKS-base materials from the prediction according to the U.S. Reg. Guide 1.99

behavior could be that the data base of Reg.Guide 1.99 represents mainly fluences between 2 and $4 \cdot 10^{19} \text{ cm}^{-2}$ and for the higher fluence range above $6 \cdot 10^{19} \text{ cm}^{-2}$ only a few data exist.

5 Fracture Mechanics Behavior of Irradiated Materials

5.1 Linear-Elastic Fracture Mechanics

The comparison of the fracture toughness data with the K_{Ic} of the Code $K_I = f(T)$ confirms that the K_{Ic} data of all FKS materials in the unirradiated conditions are enveloped by the fracture toughness curve adjusted to the individual reference temperature at least in the upper branch of the curve, fig. 4. The reference fracture toughness K_{IR} which takes into account dynamic crack initiation and crack arrest data reveals the existing margin in temperature

Figure 4: Comparison of the experimentally determined fracture toughness data of the FKS base materials and weld metals with the fracture toughness curve according to the German Standard KTA 3201.2 in the unirradiated condition

compared with the data for static crack initiation. In general the K_{Ic} data of the base materials have a greater distance to the K_{Ic} curve than those of the weld metals. The base material KS 01 GW shows the largest temperature margin of 126 K and the optimized weld metal KS 12 SG shows the smallest of 6 K. These differences make evident that in the unirradiated condition the fracture mechanics concept of the Code is conservative in case of considering only the inclining branch of the curve, however, it includes additional material specific safety margins which cannot be quantified on the basis of the RT_{NDT} . In particular the small difference between the index temperature $RT_{K_{Ic}}$ which is derived from measured values and the reference temperature RT_{NDT} of weld metal KS 12 SG does not cause any safety problems because the reference temperature for this material is far below 0°C and the irradiation induced shift is small because of the low end-of-life fluence and the low susceptibility of the material and does not shift $RT_{K_{Ic}}$ above 0°C . Therefore, a large distance in temperature exists to the relevant load paths even at end-of-life fluence.

The experimental data confirm that the fracture mechanics concept on the basis of the reference temperature is not very material specific but leads in general to a conservative bound and is thus applicable in the unirradiated conditions to base materials and to weld metals, although the original data base for the ASME and the KTA curve includes experimental data of base materials only.

At low temperatures where the curve asymptotically approaches the horizontal course at a K_{Ic} value of about $36.5 \text{ MPa} \cdot \sqrt{\text{m}}$, the K_{Ic} curve of the Code does not cover all experimental data,

fig. 4. A significant number of data fall below the curve which, however, is not significant for the safety aspect considering postulated pressurized thermal shock events. More decisive for the fracture analysis in the linear-elastic regime is the upper branch of the K_{Ic} curve at higher temperatures. Because of the specimens with limited size (maximum $B = 100$ mm) investigated in the FKS program that range of the curve could not be verified sufficiently. Applying the fracture mechanics concept also to irradiated materials by adjusting the RT_{NDT} , all the experimental results of static crack initiation for base materials in the upper branch are enveloped by the individual K_{Ic} curve of the Code, fig. 5. Thus for irradiated base materials the validity of the fracture mechanics concept could be confirmed.

Figure 5: Comparison of the experimentally determined fracture toughness data of the FKS base materials with the fracture toughness curve of the German Standard KTA 3201.2 in the unirradiated and irradiated condition

Figure 6: Comparison of the experimentally determined fracture toughness data of the FKS weld metals with the fracture toughness curve of German Standard KTA 3201.2 in the unirradiated and irradiated condition

Also for irradiated weld metals this concept could be proved except for the weld metal S3 NiMo 1/0P 41TT which is used for the optimized base material 20 MnMoNi 5 5, fig. 6.

This weld metal, however, is characterized by a low reference temperature of about $-50\text{ }^{\circ}\text{C}$ in the unirradiated condition and a low irradiation response for the anticipated fluence.

5.2 Elastic-Plastic Fracture Mechanics

For crack initiation in a temperature range where material show elastic-plastic behavior the upper shelf Charpy energy is a decisive parameter.

Due to irradiation the upper shelf energy is decreasing. All of the investigated materials with one exception do not fall below 68 J even for the high fluences applied in the FKS. The exception refers to the weld metal KS 01 SG which is extremely susceptible to irradiation because of the high Cu content of 0,37 %. The upper shelf energy dropped down to values of about 40 J and at the high fluence of $8 \cdot 10^{19}\text{ cm}^{-2}$ the upper shelf could not even be reached in the temperature range up to $275\text{ }^{\circ}\text{C}$ with this material. The other weld metals and the base materials which show a high upper shelf energy in the initial unirradiated condition particularly the materials with low sulphur content did not drop below 100 J even at the highest fluence, fig. 7.

Figure 7: Upper shelf Charpy energy of the FKS base materials with reduced sulfur content and the FKS weld metals as a function of fluence

Figure 8: Static fracture toughness data (initiation) and crack arrest data in comparison with the corresponding design curves of the German Standard KTA 3201.2

5.3 Crack Arrest Behavior

With base material specimens KS 05 GW and KS 12 GW crack arrest properties were determined in the unirradiated and in the irradiated condition at a fluence of about $2 \cdot 10^{19} \text{ cm}^{-2}$.

In case of the KS 05 GW the arrest values in the upper branch of the curve are remarkably shifted to higher temperatures against the initiation values. For the material KS 12 GW the arrest values fall into the scatter band of the initiation values. To summarize the results in one diagram the fracture toughness and the arrest data are normalized to the RT_{NDT} temperature, fig. 8. This presentation shows that the experimentally determined K_{Ic} and K_{Ia} values of the investigated materials can be enveloped by the Code curves based on the reference temperature RT_{NDT} .

6 Summary

Materials of the reactor pressure vessel (RPV) of light water reactors must have toughness properties which rule out brittle fracture under all postulated service and emergency conditions. The safety analysis is based on fracture mechanics properties which are derived from Charpy impact and drop-weight tests and provide a conservative bound.

During service the materials in the core belt-line region suffer from neutron irradiation which leads to a reduction in the initial safety margin against brittle fracture.

With the fracture mechanics concept on the basis of the reference temperature the Code provides a safe tool to establish an upper limit for brittle crack initiation for the component at the begin of life and after in-service neutron irradiation. The investigations of the FKS program have validated the basis of the fracture mechanics concept as presented in the Code.

A quantitative evaluation of safety margins additional to those contained in the Code on the basis of the reference temperature alone, however, is only possible on the basis of sufficient experimental data.

For irradiated weld metals the additional safety margin was found to be less than for base materials. The experimentally determined initiation values of the optimized weld metal used for the material 20 MnMoNi 5 5 are beyond the fracture toughness curve related to the adjusted reference temperature. However, the reference-temperature RT_{NDT} of this material is at such a low absolute value that, together with the low shift in transition temperature caused by irradiation of the material there is no safety concern.

With the exception of the weld metal with the high copper content of 0,37% the minimum Charpy upper shelf energy of none of the investigated materials has fallen below 68 J even at high neutron fluences in the order of $8 \cdot 10^{19} \text{ cm}^{-2}$.

Literature

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