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Comparative Study for the Estimation of *T_o* Shift Due to Irradiation Embrittlement

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Abstract

Recently, an approach called the "Master Curve" method was proposed which has opened a new means to acquire a directly measured material-specific fracture toughness curve. For the entire application of the Master Curve method, several technical issues should be solved. One of them is to utilize existing Charpy impact test data in the evaluation of a fracture transition temperature shift due to irradiation damage. In the U.S. and most Western countries, the Charpy impact test data have been used to estimate the irradiation effects on fracture toughness changes of RPV materials. For the determination of the irradiation shift the indexing energy level of 41 joule is used irrespective of the material yield strength. The Russian Code also requires the Charpy impact test data to determine the extent of radiation embrittlement. Unlike the U.S. Code, however, the Russian approach uses the indexing energy level varying according to the material strength. The objective of this study is to determine a method by which the reference transition temperature shift (ΔT_o) due to irradiation can be estimated. By comparing the irradiation shift estimated according to the U.S. procedure (ΔT_{41J}) with that estimated according to the Russian procedure (ΔT_F), it was found that one-to-one relation exists between ΔT_o and ΔT_F .

1. Introduction

For the safe operation of nuclear power plant (NPP), the structural integrity of reactor pressure vessel (RPV) should be maintained during the license period. RPV steels should have enough material toughness to prevent brittle fracture under all possible operating conditions. During the operation RPV materials are continuously irradiated by neutron. The

irradiation causes degradation of RPV material and leads to a steady decrease in its fracture toughness. This phenomenon is known as radiation embrittlement. A measure of radiation embrittlement is the fracture toughness transition temperature shift. The Master Curve method introduced recently made it possible to construct a material-specific fracture toughness curve for the irradiated material directly from the fracture toughness tests.

For the entire application of Master Curve method, several technical issues should be solved. One of them is to utilize existing Charpy impact test data in the evaluation of a fracture toughness transition temperature shift due to irradiation damage. In the U.S. and most Western countries, the Charpy impact test data have been used to estimate the irradiation effects on the fracture toughness changes of RPV materials and the estimation procedure was specified in the U.S. Regulatory Guide 1.99, Rev. $2^{[1]}$. For the determination of the irradiation shift, the indexing energy level of 41J is used irrespective of material yield strength. But some experimental results indicated that the Charpy transition temperature shift at 41J, DT_{41J} , is less than the fracture toughness transition temperature shift. It means that the U.S. approach can lead to non-conservative assessment of irradiation effects. The Russian Code PNAE G-7-002-86 also requires the Charpy impact test data to determine the extent of radiation embrittlement^[2]. Unlike the U.S. Code, however, the Russian approach uses the indexing energy level is selected according to the code criteria when the material yield strength increases by irradiation.

In this study, the radiation response of several RPV materials estimated according to the U.S. procedure was compared with that estimated according to the Russian procedure. Charpy impact test data and static fracture toughness test data taken from the literature were used to estimate the irradiation shift for a number of RPV materials. Correlations between the Charpy transition temperature shift (ΔRT_{NDT}) and the fracture toughness transition temperature shift (ΔT_o) were examined.

2. Estimation of Reference Temperature T_o

The Master Curve method suggested by Wallin and co-workers in 1984 is a new understanding of the brittle fracture in the transition temperature region. At present it is considered as a substantial advance in describing the ductile-brittle transition behavior of RPV steels. The reference temperature T_o is used in the characterization of Master Curve and determined according to the following procedure.

Fracture toughness tests are carried out and the reference temperature T_o is determined according to the ASTM Standard E1921-97^[3]. It is recommended that the test temperature be close to that at which the $K_{Jc(med)}$ values will be about 100 MPa \sqrt{m} for the specimen size tested. Charpy impact test data can be used for the selection of the fracture toughness test temperature. The test temperature can be determined as follows:

$$T = T_{28J} - C \tag{1}$$

where T_{28J} is the temperature corresponding to the Charpy impact energy of 28J and the constant *C* is a function of specimen size. For the precracked Charpy specimens, *C* is -50°C. The elastic-plastic equivalent stress intensity factor (K_{Jc}) is derived from the *J*-integral

measured at the instant of cleavage fracture (J_C) using

$$K_{J_c} = \sqrt{J_c \cdot E}$$
 MPa \sqrt{m} (2)

where *E* is the elastic modulus in plane stress condition. The K_{Jc} values derived from the fracture toughness tests are used to evaluate the Weibull fitting parameter, K_o . Wallin showed that the cleavage fracture toughness distribution for the given steel at a single temperature in the transition region can be characterized by the following three-parameter Weibull distribution^[4]:

$$p_f = 1 - \exp\left[-\left(\frac{K_{Jc} - K_{\min}}{K_0 - K_{\min}}\right)^b\right]$$
(3)

where P_f is the cumulative failure probability, K_{Jc} is the fracture toughness, K_{min} is the minimum K_{Jc} value, and *b* is the Weibull slope. K_o is equal to the K_{Jc} value corresponding to 63.2% cumulative failure probability. In Master Curve method, *b* and K_{min} values are fixed as 4 and 20 MPa \sqrt{m} , respectively. A maximum likelihood estimate for K_o is given by

$$K_{0} = \left[\sum_{i=1}^{N} \frac{\left(K_{Jc(i)} - 20\right)}{r - 0.3068}\right]^{1/4} + 20 \qquad \text{MPa}\sqrt{\text{m}}$$
(4)

where *N* denotes the total number of valid K_{Jc} and invalid K_J values and *r* the number of valid data. For the use of Eq. 3, six or more valid K_{Jc} data should be acquired. The invalid K_J data are censored using the following equation and the $K_{Jc(limit)}$ toughness value is assigned.

$$K_{Jc(limit)} = \left(\frac{Eb_0 \mathbf{s}_{ys}}{30}\right)^{1/2} \qquad \text{Ma}\sqrt{m}$$
(5)

where \mathbf{s}_{ys} is the yield strength, and b_0 is the initial ligament length. Then, the estimated median K_{Jc} value of the population is obtained from K_o using the following equation.

$$K_{J_{c(med)}} = (K_0 - 20) \cdot [\ln(2)]^{1/4} + 20$$
 MPa \sqrt{m} (6)

Since the Master Curve shape was determined using the data adjusted to 1T specimen, the $K_{Jc(med)}$ value should be converted to its 1T equivalent one. The statistical dependence of fracture toughness data on specimen size can be predicted using the weakest-link theory. The following equation can be used to adjust $K_{Jc(med)}$ or individual K_{Jc} values.

$$K_{Jc(med)1T} = 20 + \left[K_{Jc(med)} - 20\right] \left(\frac{B_o}{B_{1T}}\right)^{1/4}$$
 MPa \sqrt{m} (7)

where $K_{Jc(med)IT}$ is the 1*T* equivalent value of $K_{Jc(med)}$, B_o is the specimen thickness tested, and B_{IT} is the 1*T* specimen size (25mm). Finally, the reference temperature T_o which can position the Master Curve on the temperature axis is determined as follows:

$$T_0 = T - \frac{1}{0.019} \ln \left[\frac{K_{Jc(med)} - 30}{70} \right]$$
 °C and MPa \sqrt{m} (8)

where *T* is the test temperature. If multiple values of T_o are obtained from different test temperatures, the average T_o value is used. The Master Curve of $K_{Jc(med)}$ for 1*T* specimens in the transition region has the final form expressed by

$$K_{Jc(med)} = 30 + 70 \exp[0.019(T - T_o)]$$
 MPa \sqrt{m} (9)

3. Determination of irradiation shift: the U.S. approach and Russian approach

In this chapter the irradiation-induced transition temperature shift estimated according to the U.S. Regulatory Guide 1.99, Rev. 2 is compared with that estimated according to the Russian Code PNAE G-7-002-86. The irradiation shifts are compared using the experimental data and the correlations between DRT_{NDT} and DT_o are examined.

3.1 Materials used to compare procedures

A total of eleven test data sets for the irradiated RPV materials were collected from the literature. It includes plate and weld data. They cover the material yield strength ranging from 437MPa to 711MPa.

The test data for A533 Grade B Class 1 plates and welds were taken from the Fourth Irradiation Series in the Heavy-Section Steel Irradiation (HSSI) Program^[5]. These materials were irradiated in the Bulk Shielding Reactor at Oak Ridge National Laboratory at 288°C to the target neutron fluence of 2×10^{19} n/cm² (E > 1 MeV). The experimental data obtained from 1T C(T) specimens were used to estimate the static fracture toughness of these materials. Four RPV materials were lab-melt plates^[6]. One has the composition of A302-B steel and

Four RPV materials were lab-melt plates^{10]}. One has the composition of A302-B steel and others meet the composition requirements for A533-B steel. At 288°C these materials were irradiated to the neutron fluence of 1.6×10^{19} n/cm² (E > 1 MeV) in the UBR reactor at BMRC (Buffalo Materials Research Center at the State University of New York at Buffalo). The 0.5*T* C(T) specimens were used for the static fracture toughness tests of these materials.

The test data of two materials were taken from the Fifth Irradiation Series in the HSSI Program^[7]. These materials were submerged-arc welds with relatively high copper contents. These materials were irradiated at 288°C to the neutron fluence of 1.51×10^{19} n/cm² (E > 1 MeV). The 1*T* C(T) specimens were used to estimate the static fracture toughness tests of these materials.

3.2 The U.S. Code and Russian Code

In the U.S. and most Western countries, the irradiation embrittlement of the RPV steel is estimated according to the U.S. Rgulatory Guide 1.99, Rev. 2 as a part of the surveillance program. For this, Charpy impact test results are used. The magnitude of irradiation embrittlement is defined as a change in the transition temperature at the energy level of 41J irrespective of the material yield strength. And it is assumed that this transition temperature can describe the irradiation shift of the actual fracture toughness, i.e., the shift of the Charpy transition temperature has the same value as the shift of the transition fracture toughness curve.

In Russia and some Eastern European countries, a different approach following the Russian Code PNAE G-7-002-86 is used. The irradiation shift of the RPV steel is also assessed using the Charpy impact test data. Unlike the U.S. Code, however, this approach uses the indexing energy level varying according to the material strength. The yield strength range and the corresponding indexing Charpy energy level are shown in Table 1. Using the varying indexing energy level, the irradiation-induced transition temperature shift is evaluated according to the following procedure:

- a) determine the yield strength of unirradiated material at room temperature,
- b) select the indexing energy level belonging to the appropriate yield strength range from Table 1,
- c) determine the temperature corresponding to the selected indexing energy level on the Charpy transition curve,
- d) determine the yield strength of irradiated material at room temperature,
- e) repeat b) and c) for irradiated material, and finally
- f) calculate the irradiation shift (ΔT_F) from the difference between the index temperatures for unirradiated and irradiated material.

This approach implies that the indexing energy levels for unirradiated and irradiated material are identical when the material hardening due to irradiation is not large. But they are different and the gap is increased according as the radiation-induced material hardening is deepened. It is explained schematically in Fig. 1.

Yield strength at room temperature, MPa	Absorbed energy, J
up to 304	23
304 - 402	31
402 - 549	39
549 - 687	47

Table 1 Indexing Charpy energy levels varying with yield strength (Russian Code)



Fig. 1 Schematic procedure to determine the irradiation shifts for (a) moderately and (b) excessively hardened material due to irradiation (Russian Code)

4. Results and discussion

To compare the U.S. and Russian regulatory approaches, the irradiation shifts were determined from the Charpy impact test results. The difference between transition temperature shifts is shown in Fig. 2. It can be noticed that in most cases the irradiation shift determined according to the Russian Code is larger than that determined according to the U.S. Code. As mentioned above the U.S. approach is based on the assumption that there exists one-to-one relation between the irradiation shifts of the 41J Charpy energy and the K_{IC} curve. Unfortunately, the experimental verification of this assumption has not been completed. Some experimental results indicate that the shift in the 41J index temperature (DT_{41J}) is less than that in the transition fracture toughness curve (DT_o or DT_{100})^[8].

A similar result was obtained by Wallin et al. through the IAEA coordinated program on optimizing of RPV surveillance programs^[9]. Based on the evaluation of the experimental results it was demonstrated that the static fracture toughness transition temperature shift might be considerably larger than the Charpy transition temperature shift. The largest difference between the dynamic Charpy energy and static fracture shifts was observed in modern "pure" RPV steels with irradiation insensitive upper shelf energy and a low transition temperature in the unirradiated condition. They concluded that for such materials the use of the Charpy impact test data in monitoring the fracture toughness irradiation response can produce non-conservative results, so it can be dangerous.

The Charpy transition temperature shift derived according to the U.S. regulatory approach was compared with the measured irradiation shift of the transition fracture toughness curve for materials used in this study. The correlation is shown in Fig. 3. In most cases the T_{41J} shift underestimates the shift in the T_o reference temperature. This is in line with the results of other researchers mentioned above. And this effect depends on the degree of irradiation.



Fig. 2 Difference between transition temperature shifts (Russian vs. the U.S. regulatory approach)



Fig. 3 Correlation between DT_o and DT_{41J} (the U.S. guide)

A problem in the U.S. approach is that it does not take into account the change of the material yield strength due to irradiation. There are some experimental results showing the influence of material strength on the loading rate sensitivity. Barsom observed that in the fracture toughness-temperature graph the dynamic fracture toughness curve is always located in the higher temperature region than the static curve and the difference between them depends on the material yield strength^[10]. The biggest difference is observed for low strength steels and it decreases for higher strength steels. It is also proved that the strain rate sensitivity of the yield strength decreases with the increase of the material strength^[11]. From the Charpy and the static fracture toughness transition temperature shift database of the U.S., O' Donnell and Crombie made a conclusion that the DT_{41J} irradiation shift underestimates DT_o because of the yield strength effect^[12].

In Fig. 4, the same comparison was made for the Charpy transition temperature shift, DT_F ,

defined according to the Russian code. The relation between DT_F and DT_o is close to the oneto-one line. There is a good agreement between two values. It means that the Charpy energy irradiation shift can describe properly the radiation-induced changes in the fracture toughness properties when the material strength is taken into account.

In the case of static fracture toughness test the absorbed fracture energy corresponds to the brittle failure at the transition temperature. The absorbed energy value obtained from Charpy impact test includes both brittle and ductile part of fracture energy at the transition temperature. Neutron irradiation decreases the material resistance against ductile fracture as well as brittle fracture. And the ductile to brittle ratio of the absorbed energy is reduced at the transition temperature. To compensate the decrease of the ductile portion in the fracture energy due to irradiation, it is necessary to increase the indexing energy level of the Charpy transition curve.

Instead of the yield strength, Wallin suggested a simple method to determine the indexing energy level based on the change of the upper shelf energy as follows:

Indexing energy level of Charpy transition curve =
$$\frac{(USE)_{Unirradiated material}}{(USE)_{Irradiated material}} \cdot 28J$$
(10)

where $(USE)_{unirradiated material}$ and $(USE)_{irradiated material}$ denote the upper shelf energies for unirradiated material and irradiated material, respectively. Fig. 5 represents the relation between the values of DT_o and DT_{28J} . For the determination of the Charpy energy shift, a fixed 28J energy level was used for both unirradiated and irradiated steels. A poor correlation is observed. After Wallin's energy adjustment, one-to-one relation between two shifts was obtained as shown in Fig. 6. This showed almost the same results as the approach based on the Russian Code.

From these results, it can be said that the indexing energy level of the Charpy transition curve should be selected by taking the variation of the material yield strength into account. After the correction, the irradiation shift of the transition fracture toughness curve has one-to-one relation with that of the Charpy transition curve.



Fig. 4 Correlation between DT_o and DT_F (Russian Code)



Fig. 5 Correlation between DT_o and DT_{28J}



Fig. 6 Correlation between transition temperature shifts (Wallin's approach)

5. Conclusions

In this study the Russian and the U.S. regulatory approaches are compared in the point of view of the evaluation of irradiation-induced changes in the fracture toughness properties and the following conclusions are drawn.

- The irradiation shift derived from the Charpy impact test data according to the U.S. Regulatory Guide 1.99, Rev. 2 tends to underestimate the irradiation shift of the transition fracture toughness curve.
- The irradiation shift derived from the Charpy impact test data according to the Russian Code PNAE G-7-002-86 has one-to-one relation with the irradiation shift of the transition fracture toughness curve.

• The Charpy transition temperature shift due to irradiation can be used to describe the irradiation shift of the transition fracture toughness curve if the change of the material yield strength is taken into account.

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