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COMPARISON OF FRACTURE TOUGHNESS AND CHARPY IMPACT PROPERTIES RECOVERY BY THERMAL ANNEALING OF IRRADIATED REACTOR PRESSURE VESSEL STEELS*

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Comparison of Fracture Toughness and Charpy Impact Properties Recovery by Thermal Annealing of Irradiated Reactor Pressure Vessel Steels

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Abstract

The objective of this investigation was to study the effects of thermal annealing on the recovery of the transition region toughness of reactor pressure vessel steels. The toughness was measured by Charpy V-notch impact energy and fracture initiation toughness, K_{Jc}. The materials were A 533 grade B class 1 plate and a commercial reactor vessel submerged-arc weld irradiated at 288°C to neutron fluences of 1.0 to 2.5×10^{19} neutrons/cm^2 (>1 MeV). The irradiated materials were annealed at 343 and 454°C for 1 week. The recently developed Weibull statistic/master curve approach was applied to analyze fracture toughness properties of unirradiated, irradiated, and irradiated/annealed pressure vessel steels. The effects of irradiation or annealing were determined by the shift in temperature of the Charpy V-notch curve at 41 J and the fracture toughness curve at 100 MPa√m. After annealing at 454°C, the residual shifts in fracture toughness are approximately the same as the residual Charpy shifts. The differences observed in these residual shifts after annealing are approximately the same as differences in the radiation-induced shifts.

Key terms: annealing, fracture toughness, master curve, Charpy transition temperature

Introduction

Prevention of a reactor pressure vessel (RPV) failure in light-water moderated nuclear power reactors depends primarily on maintaining adequate levels of fracture toughness during plant operation under both normal and emergency conditions. The *American Society of Mechanical Engineers(ASME) Boiler and Pressure Vessel Code*[1] contains a fracture toughness (K_{Ic}) curve as a function of temperature (T) normalized to the reference nil-ductility temperature (RT_{NDT}), namely, T - RT_{NDT}. Since the reactor vessel is subjected to neutron irradiation, the material will be embrittled as manifested by a shift (ΔRT_{NDT}) in RT_{NDT}. Title 10, *Code of Federal Regulations*, Part 50 (10CFR50)[2] includes provisions for the determination of the upward temperature shift of RT_{NDT}, which are based on the assumption that it is the same as the Charpy impact curve shift at 41 J (ΔT_{41J}). However, the similarity of Charpy T_{41J} and fracture toughness transition curve shifts due to irradiation is sometimes challenged, and an interested reader can refer, e.g., to Refs. 3-5 for correlations that have been developed.

The fracture toughness requirements during a pressurized thermal shock (PTS) scenario are determined on the basis of an irradiated RT_{NDT} "screening criterion" called RT_{PTS} in 10CFR50. Some early nuclear RPVs may not meet this screening criterion as they near end-of-life. In particular, it is believed that, in the next decade or so, several vessels may exceed the RT_{PTS}. Thermal annealing may be needed to mitigate the effects of neutron irradiation on fracture toughness. A dozen or so RPVs have already been thermally annealed in Europe.[6,7] Development of a U.S. Nuclear Regulatory Commission (NRC) regulatory guide on recovery of properties by annealing is under way. The basis for proposed regression correlations given in the guide is data gathered from the Test Reactor Embrittlement Data Base and from various annealing reports.[8] These data deal with recovery of Charpy or hardness properties only. Thus, the proposed regulatory guide is based on the same philosophy as mentioned earlier, namely, the assumption that recovery of the fracture toughness by annealing is the same as the recovery of Charpy T_{41J}.

The objective of this paper is to compare the recovery of the fracture toughness and Charpy impact properties by thermal annealing of two irradiated RPV steels.

Materials and Irradiation

The materials were American Society for Testing and Materials (ASTM) A 533 grade B class 1 plate, designated Heavy-Section Steel Technology (HSST) Program Plate 02, and the submerged-arc weld from the Midland Unit 1 reactor vessel. This vessel was built for a pressurized-water reactor that was canceled prior to startup. The welds from that vessel have the Babcock and Wilcox designation WF-70. The WF-70 welds were fabricated using copper-coated wire and Linde 80 flux and are known to be low upper-shelf (LUS), high-copper welds. Twenty-four Charpy specimens and 34 compact specimens 12.7 and 25.4 mm thick [0.5T C(T) and 1T C(T), respectively] from a beltline portion of the reactor vessel weld were tested after irradiation at 288°C to a neutron fluence level of about 1.0×10^{19} neutrons/cm^2 (>1 MeV) at the University of Michigan Ford Reactor.[9] Two hundred and thirty Charpy specimens and 65 compact specimens in sizes ranging from 1/2T to 4T were tested to perform fracture toughness[10] and Charpy impact[11] characterization in the unirradiated condition. In this study, 12 of the Charpy specimens and 6 of the 1T compact specimens that were irradiated to 1.0×10^{19} neutrons/cm^2 (>1 MeV) were annealed at 454°C (850°F) for 1 week and 10 of the Charpy specimens were annealed at 343°C (650°F) for 1 week.

HSST Plate 02 was produced by Lukens Steel Company. Portions of this plate have been used in many investigations around the world. In the Fourth Heavy-Section Steel Irradiation (HSSI) Program Irradiation Series, up to 70 Charpy and 28 1T compact specimens were tested before and after irradiation at 288°C to neutron fluences from 1.1 to 2.4×10^{19} neutrons/cm^2 (>1 MeV) in the Oak Ridge Bulk Shielding Reactor.[12] Six 1T irradiated compact specimens, left from that program, were annealed at 454°C (850°F) for 1 week and five 1T irradiated compact specimens were annealed at 343°C (650°F) for 1 week. Test specimens in that irradiation series were prepared in the transverse (T-L) orientation. No Charpy specimens were available for an annealing study from that series. Twenty-two Charpy specimens of HSST Plate 02 in the longitudinal (L-T) orientation were irradiated together with the Midland weld to 1.0×10^{19} neutrons/cm^2 (>1 MeV) at the University of Michigan Ford Reactor. Twelve of them were annealed at 454°C (850°F) for 1 week with ten specimens annealed at 343°C (650°F) for 1 week. It had been shown previously that values of T_{41J} differ for different specimen orientations, but the irradiation-induced shift of transition temperature is the same for T-L- and L-T-oriented Charpy specimens.[13]

Results and Discussion

The Charpy V-notch impact data for each material condition were fit with a hyperbolic tangent function:

$$\mathbf{KCV} = \frac{USE + LSE}{2} + \frac{USE - LSE}{2} \cdot \tanh\left[\frac{T - T_{MT}}{C}\right], \qquad \mathbf{(1)}$$

where KCV is the absorbed energy; T is test temperature; USE and LSE are upper- and lower-shelf energy values, respectively; T_{MT} is the temperature at the middle of the transition range; and C is half of the transition zone width, reflecting the slope of the curve in the transition zone. The lower-shelf was fixed at 2.7 J (2 ft-lb).

The elastic-plastic fracture toughness data were analyzed by a procedure based on earlier work by Wallin[14] and developed in an ASTM draft standard by McCabe et al.,[15] applying statistical concepts developed by Weibull.[16] The analysis procedure is based on fitting fracture toughness data to a three-parameter Weibull distribution at the test temperature:

$$P_f = 1 - \exp\left(-\left[\frac{K_{Jc} - K_{min}}{K_o - K_{min}}\right]^b\right), \qquad (2)$$

where P_f is the cumulative fracture probability, K_{min} is a lower limiting value of K_{Jc}, K_o is a specimen thickness and temperature-dependent scale factor, and b is the Weibull slope. Wallin determined that the shape parameter (Weibull slope) for fracture mechanics based K_{Jc} values is either near to or equal to four when a value of K_{min} is about 20 MPa√m. Because parameters b and K_{min} in Eq. (2) have been shown to be essentially constant, then only the scale factor, K_o, needs to be determined. As a consequence, only a few replicate tests are needed to obtain this third parameter with good accuracy. This procedure employs the maximum likelihood concept[17] regarded as the most accurate method of obtaining median value of fracture toughness, $K_{Jc(med)}$, at a given temperature. Additionally, weakest-link theory is used to explain specimen size effects so that data equivalent to that for a 1T specimen size can be calculated from data measured with specimens of different sizes. Finally, the master curve concept for 1T size specimens was applied to median K_{Jc} to define the temperature dependence of K_{Jc} in the transition region as follows:

$$K_{Jc(med)} = 30 + 70 \exp[0.019(T - T_o)], \qquad (3)$$

where T_o is the reference temperature at $K_{Jc(med)}$ = 100 MPa√m. The T_o values obtained from data sets at two or more temperatures tend to be the same. Multiple values of T_o were averaged. Details of this analysis are published elsewhere.[5]

Figures 1 and 2 present Charpy and fracture toughness curves, respectively, of the Midland beltline weld in the unirradiated, irradiated and irradiated/annealed conditions. The shift of the Charpy transition temperature, ΔT_{41J}, due to irradiation was 103°C compared to a 92°C shift of fracture toughness transition temperature, ΔT_o. Annealing at 343°C for 168 h resulted in full recovery of Charpy upper-shelf energy (USE) but in only 49% recovery of ΔT_{41J}. The residual shift (unrecovered after annealing) of the Charpy transition temperature (ΔT_{res}^{CV}) is 53°C. The USE after annealing at 454°C for 168 h increased to 106 J, which is 17 J higher than the USE in the unirradiated condition. The residual shift, ΔT_{res}^{CV}, is 24°C. The fracture toughness specimens were annealed at 454°C for 168 h. The residual shift of the fracture toughness transition temperature (ΔT_{res}^{K}), Fig. 2, is 13°C.

Figures 3 and 4 present Charpy and fracture toughness curves, respectively, of HSST Plate 02 in the unirradiated, irradiated, and irradiated/annealed conditions. Charpy specimens, available for annealing, had the L-T orientation and no baseline Charpy transition curve was developed for L-T oriented specimens in the as-irradiated condition. However, the irradiation-induced shift was determined for Plate 02 in the T-L orientation, as reported in Ref. 12. Based on those results, and the accepted equivalence with the shift for the L-T orientation,[13] ΔT_{41J} is estimated to be 55°C at

1×10^{19} neutrons/cm^2 (>1 MeV), as shown in Fig. 3. Annealing at 343°C for 168 h resulted in full recovery of the Charpy USE. The residual shift of the Charpy transition temperature after annealing at 343°C is 35°C. Similar to the Midland weld, annealing at 454°C for 168 h resulted in "over-recovery" of the USE; the USE of the irradiated and annealed plate rose 24 J above that the USE in unirradiated condition. The ΔT_{41J} recovered almost fully after annealing at 454°C/168 h since ΔT_{res}^{CV} was only 6°C. The residual shifts of ΔT_o, however, were 78 and 22°C after annealing at 343 and 454°C, respectively.

For both materials, full recovery of the Charpy USE was observed even at the lower annealing temperature, although recovery of the transition temperature was far from complete at 343°C. At the annealing temperature of 454°C, over-recovery of the USE was observed. The definition of over-recovery is that the USE after annealing of irradiated material is greater than the unirradiated level. In addition to irradiated specimens, unirradiated Charpy specimens of Plate 02 were heat treated at 454°C for 168 h (the same regimen as for annealing of irradiated specimens) and there was no change in USE compared with the unirradiated level. Annealing at 454°C for 168 h was also performed on Charpy specimens from the Midland nozzle course weld, irradiated to 1.0×10^{19} neutrons/cm^2 (>1 MeV), which are identical to the beltline weld metal except for copper content. The USE of irradiated/annealed nozzle course weld was 17 J higher than the unirradiated level, which was also observed for the beltline weld.

Such behavior is consistent with other annealing studies.[18,19] In Ref. 18, annealing of irradiated and unirradiated submerged-arc HSSI weld 73W at 454°C for 168 h resulted in the same increase of the USE compared with the unirradiated condition. It might be concluded, therefore, that the observed over-recovery of the USE of weld metal is independent of any irradiation effect since such heat treatment of unirradiated material also resulted in an increase of the USE. Over-recovery of the ductile fracture toughness as measured by J_{Ic} and tearing modulus was reported for the irradiated/annealed Linde 80 high-copper, low upper-shelf welds.[20,21] It is interesting to note that, in Ref. 21, welds that over-recovered the ductile fracture toughness did not show full recovery of the fracture toughness in the transition region, which corresponds well with the present Charpy data. The current data on the recovery of the Charpy USE in different materials are generally consistent, but the mechanism responsible for the over-recovery is not clear. Undoubtly, the more rapid and more extensive recovery of USE with annealing compared with transition temperature indicates different mechanisms for degradation of these properties upon irradiation. The effect of over-recovery of Charpy USE makes annealing an attractive measure for plant life extension, especially for so-called low upper-shelf materials. But the real value of this advantage can be judged only after an extensive study of behavior of materials after reirradiation.

The value of ΔT_{41J} of the Midland beltline weld after irradiation to a neutron fluence of 1.0×10^{19} neutrons/cm^2 (>1 MeV) was about 10°C higher that the ΔT_o. Annealing of the irradiated weld at 454°C for 168 h significantly recovered Charpy and fracture toughness transition temperatures. However, the residual or unrecovered shift of the Charpy 41-J transition temperature was also about 10°C higher than the residual shift of the fracture toughness transition curve.

Sets of compact specimens and Charpy specimens of Plate 02 were irradiated to different neutron fluences, so the comparison of Charpy and fracture toughness recovery cannot be performed as directly as in the case of the Midland weld. To overcome this, the Charpy T_{41J} and fracture toughness T_o shifts from analysis[5] of the Fourth HSSI Irradiation Series were fit to following equation:

$$\Delta T = A \times F^{1/2} , \qquad (4)$$

where ΔT is the shift of fracture toughness T_o and/or Charpy T_{41J}, A is a fitting parameter and F is neutron fluence ($\times 10^{19}$ neutrons/cm^2), see Fig. 5. The first observation is that the fracture toughness shifts due to irradiation are slightly higher than those for Charpy impact toughness. Annealing at 343°C for 1 week resulted in noticeable recovery of ΔT_{41J}, but ΔT_o did not show any recovery. This could be due to different responses of Charpy and fracture toughness properties to low-temperature annealing; and/or due to dependence of residual shift on neutron fluence. For a 454°C annealing temperature, the residual fracture toughness shift was 22°C compared with 6°C of the residual Charpy shift. The residual fracture toughness shift was higher than ΔT_{res}^{CV} by about the same amount as ΔT_o due to irradiation was higher than ΔT_{41J}.

The data obtained from the annealing investigation of the Midland beltline weld and the HSST Plate 02 appear to be consistent. The values of residual shift in fracture toughness are comparable to the residual Charpy transition temperature shifts at 41-J following annealing at 454°C and the degree of agreement is similar to that observed when the radiation-induced temperature shifts are compared.

Summary of Observations

Annealing of irradiated A 533 grade B class 1steel (HSST Plate 02) and Midland beltline weld (WF-70) at 288°C was performed at 343 and 454°C for 1 week to compare the recovery of the fracture toughness and Charpy impact properties. The Weibull statistic and master curve approach were applied to analyze fracture toughness properties of unirradiated, irradiated, and irradiated/annealed pressure vessel steels. The following are concluded:

1. Recovery of the Charpy USE appears to be more rapid and more extensive compared to the Charpy transition temperature, which indicates different mechanisms for degradation of these properties upon irradiation. At the annealing temperature of 454°C, the Charpy USE rises above the unirradiated level. This effect of over-recovery of Charpy USE makes annealing a very attractive measure

for plant life extension, especially for so-called low upper-shelf materials. But the real advantage to be gained can be judged only after extensive study of behavior of materials with reirradiation.

2. Shifts of the reference fracture toughness of the materials studied were slightly different from the Charpy 41-J transition temperature shifts. Annealing of A 533 grade B steel (Plate 02) at 343°C did not result in any apparent recovery of fracture toughness reference temperature, T_o, compared with noticeable recovery of the Charpy T_{41J}. The values of residual (unrecovered) shift in fracture toughness are comparable to the residual Charpy transition temperature shifts at 41 J following annealing at 454°C and the degree of agreement is similar to that observed between the radiation-induced ΔT_o and ΔT_{41J}.

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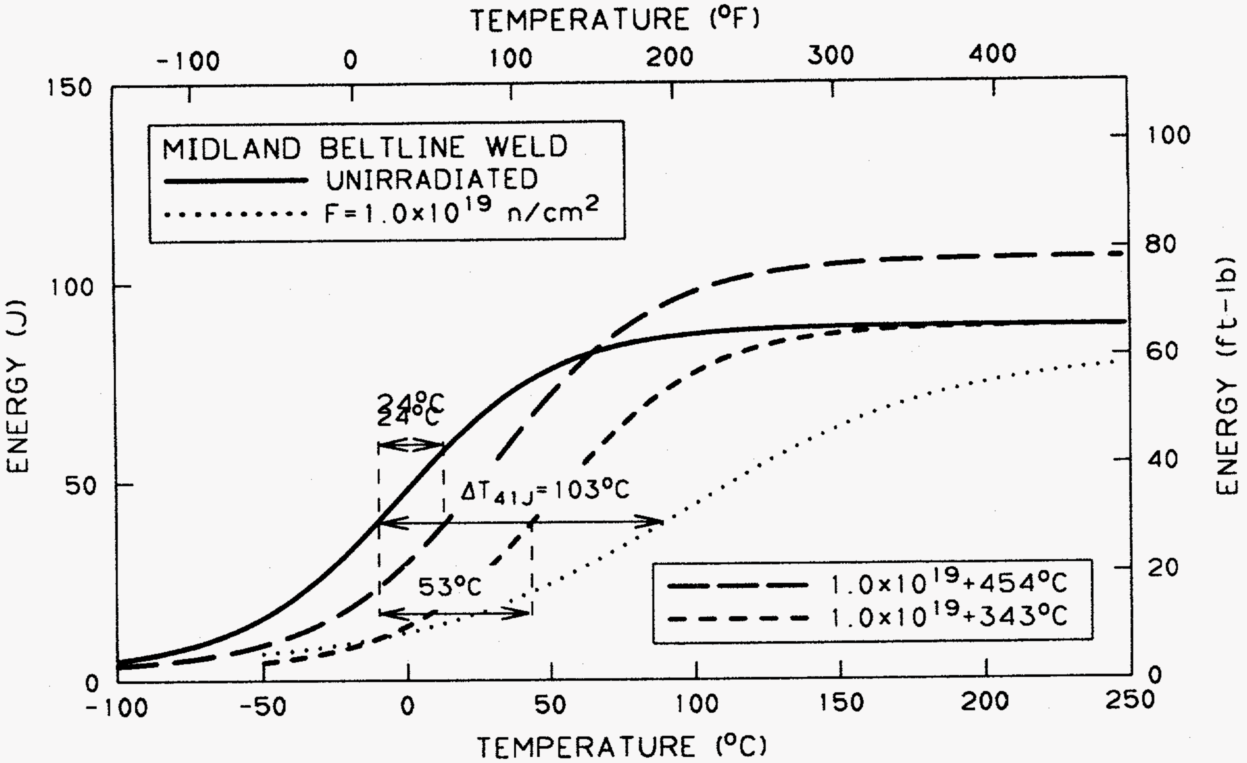

Fig. 1. Charpy impact curves of Midland beltline weld (WF-70) in unirradiated, irradiated and irradiated/annealed conditions.

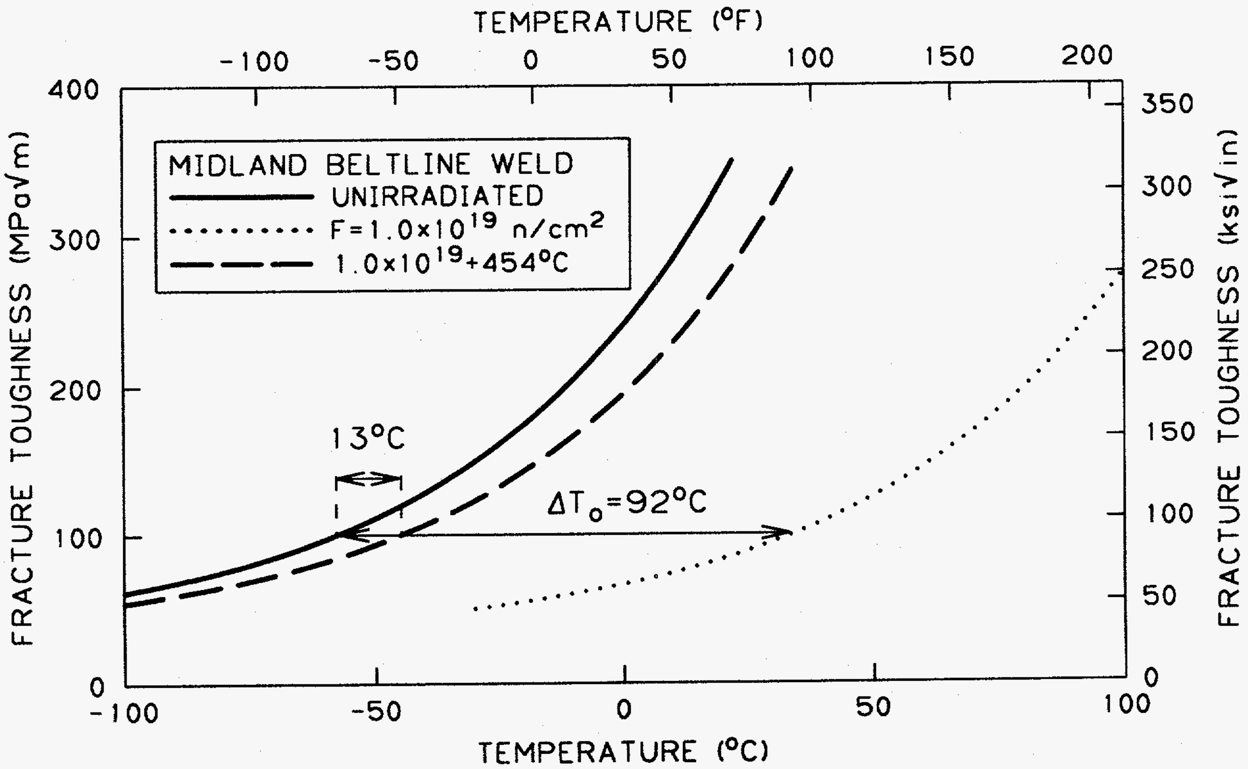

Fig. 2. Fracture toughness master curves of Midland beltline weld in unirradiated, irradiated and irradiated/annealed conditions.

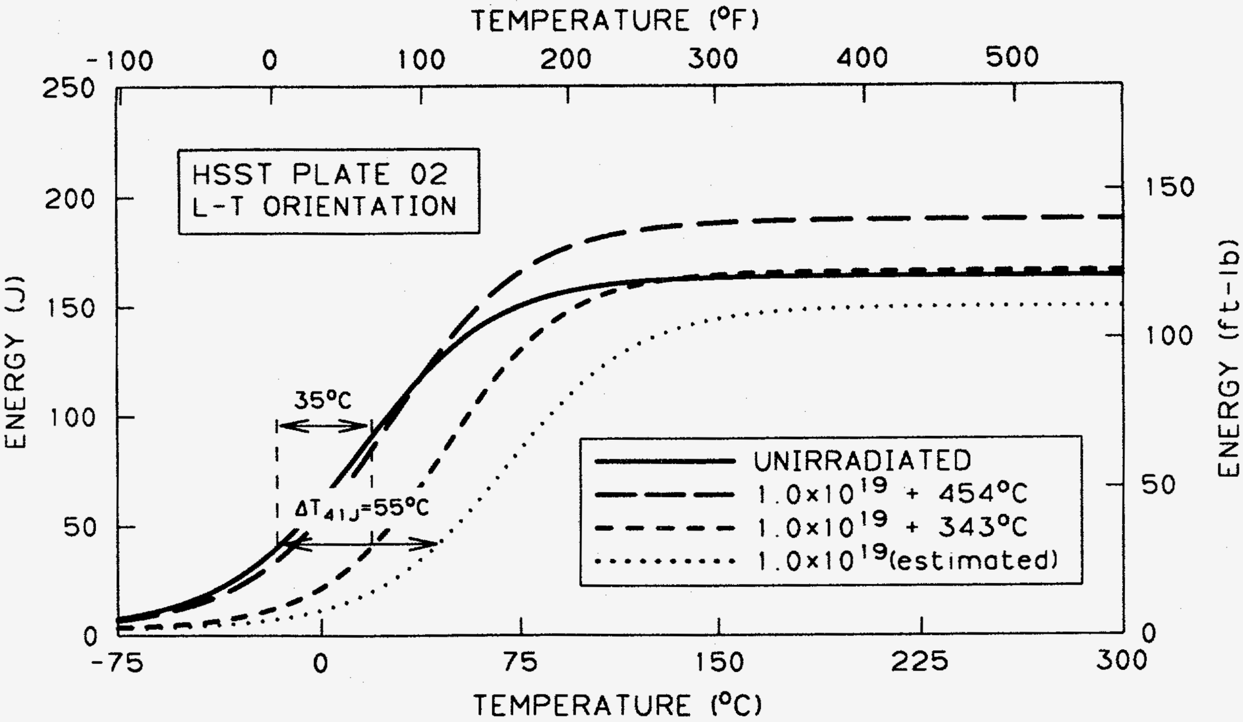

Fig. 3. Charpy impact curves of HSST Plate 02 (L-T orientation) in unirradiated and irradiated/annealed conditions. Irradiated curve was estimated from T-L data (see text).

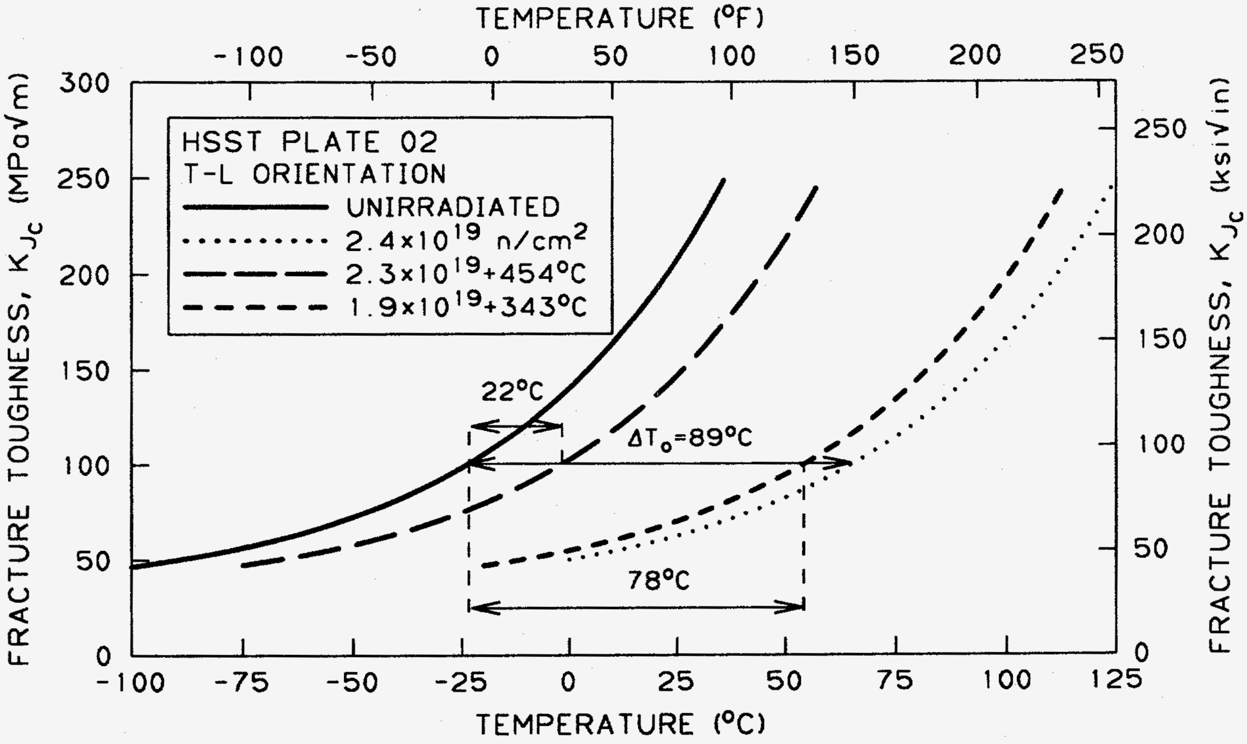

Fig. 4. Fracture toughness master curves of HSST Plate 02 (T-L) in unirradiated, irradiated and irradiated/annealed conditions.

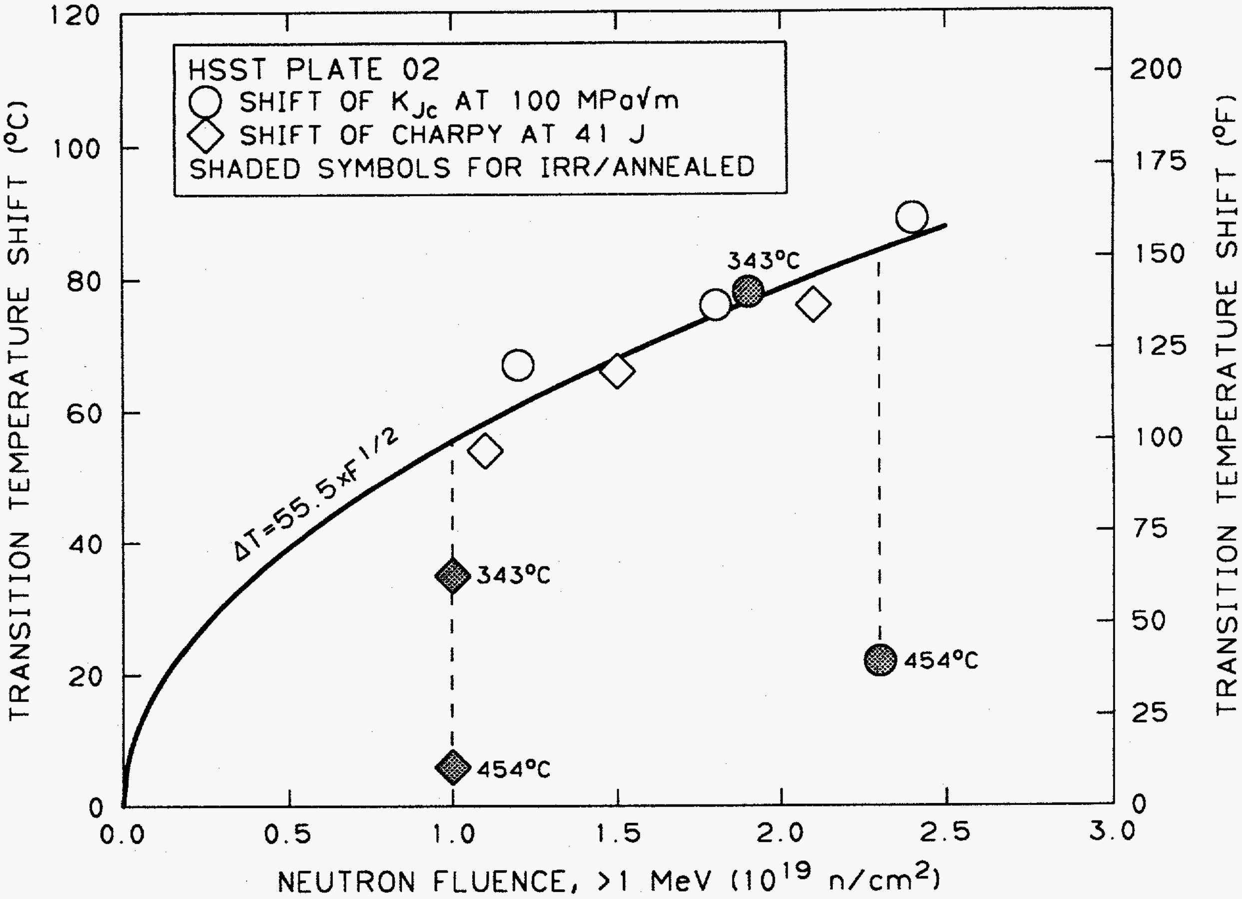

Fig.5. Comparison of Charpy 41J and fracture toughness 100MPa√m transition temperature shifts after irradiation and annealing of HSST Plate 02. Annealing temperature is indicated besides symbol.