Effect of Radiation on Fracture Toughness and Microstructure of a High-Cu RPV Weld

Mikhail A. Sokolov, Randy K. Nanstad and Michael K. Miller

Oak Ridge National Laboratory, P.O. Box 2008, Oak Ridge, TN, 37831, sokolovm@ornl.gov

ABSTRACT - *A radiation-sensitive reactor pressure vessel (RPV) weld with intentionally enhanced copper, nickel, and manganese content, designated KS-01, is characterized in terms of static initiation (K_{Ic}, K_{Jc}) and Charpy impact toughness in the unirradiated and irradiated conditions. After neutron irradiation to a fluence of 0.8 x 10^{19} neutron/cm^2 (E > 1 MeV) at a temperature of 288°C, this weld exhibited a large Charpy T_{41J} shift of 169K, a large shift of the fracture toughness transition temperature of 165K, a decrease in upper shelf energy (USE) from 118 J to ~78 J, and an increase in the yield strength from 600 to 826 MPa. Atom probe tomography revealed a high number density (~3× 10^{24} m^{-3}) of Cu-, Mn-, Ni-, Si- and P-enriched precipitates and a lower number density (~1 × 10^{23} m^{-3}) of P clusters.*

I.INTRODUCTION

The purpose of this pilot study was to perform fracture toughness characterization of a metallurgically modified RPV steel with a degree of embrittlement at low fluence corresponding to the amount of embrittlement normally expected for a RPV steel at the pressurized thermal shock screening criterion condition, to investigate the ability of highly embrittled material to maintain the shape of the unirradiated transition fracture toughness curve, as well as to examine the ability of the Charpy 41-J shift to predict the fracture toughness shift at such high level of embrittlement.

The pressurized thermal shock screening criterion for circumferential weld metals, RT_{PTS}, is prescribed in the Code of Federal Regulations, Title 10, Part 50 (10CFR50) as 300°F (149°C) [1]. This criterion was converted in terms of the master curve transition temperature to T_{oPTS} = 130°C using the American Society of Mechanical Engineers Code Case N-629 which allows RT_{NDT} to be replaced by $RT_{To} = T_o +$ 35°C (19°C).

The shape of the master curve was established with unirradiated and some irradiated fracture toughness data [2] as an empirical fit to these data size adjusted to 1T. The majority of fracture toughness data support this empirical approximation. However, the physical basis for the universal shape of the transition fracture toughness region remains open for discussion. That raises concern regarding the ability of a highly-embrittled material to maintain the same shape of the transition fracture toughness as in the unirradiated condition, and the necessity to perform experimental validation of this assumption. Some limited irradiated fracture toughness data [3-5] also suggest a potential change in the shape of the fracture toughness transition as result of irradiation.

For this study, MPA-Stuttgart, Germany provided a weld, designated KS-01, with intentionally increased copper, nickel, and manganese contents, as can be seen in Table 1 provided by MPA. Chromium content is also large relative to a typical US-made RPV weld but it is more common for German-made welds.

TABLE I. Chemical composition of KS-01 weld, wt%.

C	Ni	Mn	Mo	Cr	Cu	Si	P
.06	1.23	1.64	.70	.47	.37	.18	.017

Irradiation was performed in the HSSI facility at the University of Michigan Ford Reactor in reusable capsules at 288°C. Based on MPA data, the neutron fluence of 0.8×10^{19} neutron/cm^2 (E > 1 MeV) was selected as target fluence for this irradiation experiment. It was also anticipated that irradiation to this fluence would induce a Charpy ductile-to-brittle transition temperature (DBTT) shift of about 170°C and Charpy upper shelf energy (USE) would not decrease below 68 J (50 ft-lb), in accordance with the requirements of 10CFR50, Appendix G.

II. TENSILE PROPERTIES

Round tensile specimens with a gage section 5.08 mm in diameter and 31.75 mm in length were tested in the unirradiated and irradiated conditions. Four tensile specimens were tested in the unirradiated condition. Two specimens were tested at room temperature and two at -60°C. MPA tested unirradiated tensile specimens in the range from -100°C to 275°C. Tensile properties of unirradiated KS-01 weld measured at ORNL and MPA are in very good agreement. On average, room temperature yield strength is 600 MPa and ultimate strength is 683 MPa. This material has a relatively high strength in the unirradiated condition compared to a typical RPV weld. It was initially considered as an advantage since high yield strength increases the validity limit for fracture toughness measurement according to ASTM standard E 1921 [6]. After irradiation, a total of seven specimens was tested in the temperature range from -100°C to 250°C. The yield strength at room temperature increased by 226 MPa and ultimate strength increased by 203 MPa compared to the unirradiated condition, see Fig. 1.

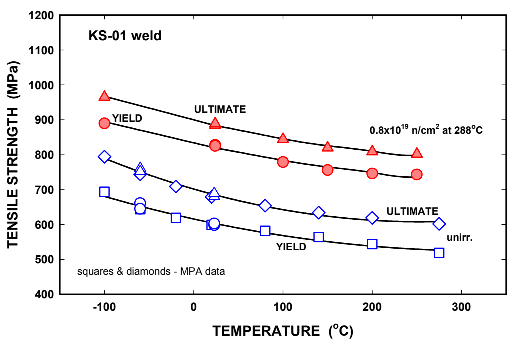

Fig. 1. Temperature dependence of yield and ultimate strengths of KS-01 weld before and after irradiation.

III. CHARPY IMPACT PROPERTIES

Charpy specimens were examined in the T-L orientation. Fifteen CVN specimens were tested in the unirradiated condition by MPA and ten specimens by ORNL. As in the case with tensile properties, CVN data from both laboratories are in very good agreement. In the unirradiated condition, Charpy transition temperature, T_{41J}, is –10°C and the upper shelf energy is 118 J. As expected, KS-01 weld exhibited a large shift of transition temperature, 169°C, after irradiation at 288°C to 0.8×10^{19} neutron/cm^2 (E > 1 MeV). The upper shelf energy reduced to 78 J. Charpy impact properties of KS-01 weld before and after irradiation are presented in Fig. 2. In addition to a large shift of transition temperature and drop in the upper shelf energy, this weld also exhibited change in the slope of the transition region typical for irradiated RPV materials. For example, the shift of T_{41J} was 169°C compared to 217°C shift of T_{68J}.

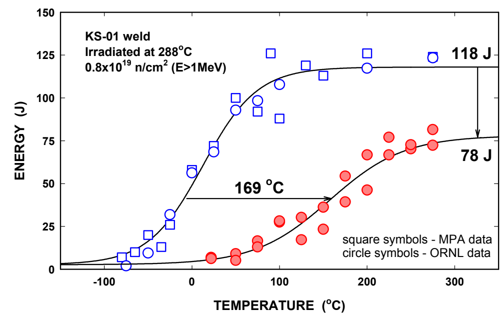

Fig. 2. Charpy impact energy versus test temperature for KS-01 weld in the unirradiated condition (open points) and following irradiation (filled points) at 288°C to an average fast fluence of 0.8×10^{19} neutron/cm^2 (E>1 MeV).

IV. FRACTURE TOUGHNESS RESULTS

Fracture toughness data have been generated using compact specimens, C(T), of 0.5T and 1T sizes, although a limited number of precracked Charpy V-notch (PCVN) specimens has also been tested in the unirradiated condition. Only the 0.5T C(T) specimens were 20% side-grooved (10% from each side) after fatigue precracking. An outboard clip gage was used to measure load-line displacement on C(T) specimens and an LVDT gage was used to measure load-line displacement on PCVN specimens. The unloading compliance method was used to obtain J-integral versus crack extension data. From J-integral versus crack extension data, a J-integral at the point of cleavage instability, J_c, was determined and a critical value of stress intensity, K_{Jc}, was calculated from:

$$K_{Jc} = \sqrt{J_c \frac{E}{1-\nu^2}} \qquad (1)$$

where E is Young's modulus and ν=0.3 is Poisson's ratio. All K_{Jc} data were converted to 1T equivalence, $K_{Jc}(1T)$, using the size adjustment procedure of E1921 [2,6]:

$$K_{Jc(1T)} = 20 + \left[K_{Jc(x)} - 20\right]\cdot\left(\frac{B_x}{B_{1T}}\right)^{1/4} \qquad (2)$$

where $K_{Jc(x)}$ = measured K_{Jc} value,
B_x = gross thickness of test specimen,
B_{1T} = gross thickness of 1T C(T) specimen.

Two validity criteria were used to qualify K_{Jc} data. A K_{Jc} datum would be considered invalid if this value exceeded the $K_{Jc(limit)}$ requirement of E 1921 [6]:

$$K_{Jc(\text{limit})} = \sqrt{\frac{b_o\, \sigma_{YS}}{30} \cdot \frac{E}{1-\nu^2}} \qquad (3)$$

where b_o is the specimen remaining ligament.

As mentioned earlier, this weld had a relatively high yield strength (σ_{YS}) even in the unirradiated condition. Irradiation increased it even higher. As a result, none of the specimens tested within this study violated this validity requirement. The second validity requirement limits the amount of stable crack growth prior to cleavage instability. A K_{Jc} datum was considered invalid if the test terminated in cleavage after slow-stable crack growth of more than $0.05(W-a_o)$ or 1 mm, whichever is smaller. Several specimens in this study violated this requirement and those K_{Jc} values were replaced by the highest valid K_{Jc} value in the data set for any given specimen size.

The reference fracture toughness transition temperature, T_o, was determined using the multi-temperature equation from E 1921 [2,6]:

$$\sum_{i=1}^{N} \delta_i \frac{\exp[0.019(T_i - T_o)]}{11+77\exp[0.019(T_i - T_o)]} - \sum_{i=1}^{N} \frac{(K_{Jc(i)} - 20)^4 \cdot \exp[0.019(T_i - T_o)]}{\{11+77\exp[0.019(T_i - T_o)]\}^5} = 0 \qquad (4)$$

where $\delta_i = 1.0$ if the datum is valid, or zero if the datum is a dummy substitute value, and T_i = test temperature corresponding to $K_{Jc(i)}$.

In the unirradiated condition, two of the specimens tested at room temperature cleaved after more than 1 mm stable crack growth and the resulting data were used as invalid data points for T_o and $K_{Jc(med)}$ determination. One of the PCVN specimens cleaved with a K_{Jc} value just slightly lower than the $K_{Jc(limit)}$ for this specimen size/temperature. The final value of the reference fracture toughness transition temperature for KS-01 weld in the unirradiated condition is –26°C.

Irradiated fracture toughness specimens were tested from 100°C to 200°C, at 25°C intervals to characterize the transition fracture toughness. After initial calculation, it was determined that data at 200°C should be excluded from analysis since they were outside the $T_o \pm 50$°C range. The final calculation gave a value of T_o equal to 139°C. Thus, the shift of the transition fracture toughness temperature after irradiation of KS-01 weld at 288°C to 0.8×10^{19} neutron/cm^2 is 165°C, which is in remarkable agreement with the Charpy T_{41J} shift of 169°C. The fracture toughness data of KS-01 weld size-adjusted to 1T equivalence before and after irradiation are summarized in Figure 3. Master curves are drawn as solid lines and 5% and 95% tolerance bounds are drawn as dashed lines. Master curves and tolerance bounds are drawn only within $T_o \pm 50$°C range. As in the case with unirradiated data, 5% and 95% tolerance bounds appear to provide a good description of irradiated fracture toughness data within the temperature range of $T_o \pm 50$°C (89 to 189°C). However, the repeatable number of low toughness cleavage fractures at 200°C indicates a potential for deviation of the transition fracture toughness from the master curve shape for a highly embrittled RPV weld.

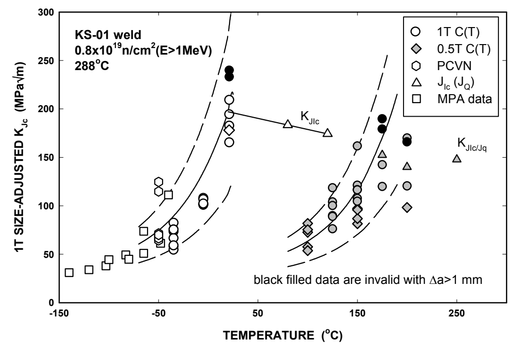

Fig. 3. 1T size-adjusted fracture toughness data of KS-01 weld before and after irradiation.

It appears that irradiation-induced changes in mechanical and fracture toughness properties of KS-01 weld are in agreement with general trends for RPV steels [5] as can be seen in Figs. 4 and 5.

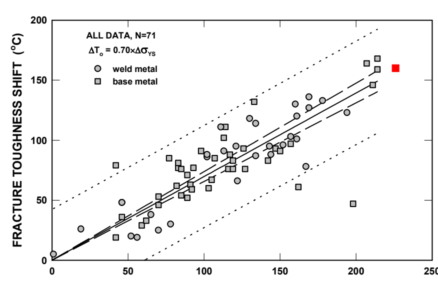

Fig. 4. Relationship between fracture toughness T_o shift and yield strength increase for RPV steels from Ref. [5] and position of KS-01 weld from the present study.

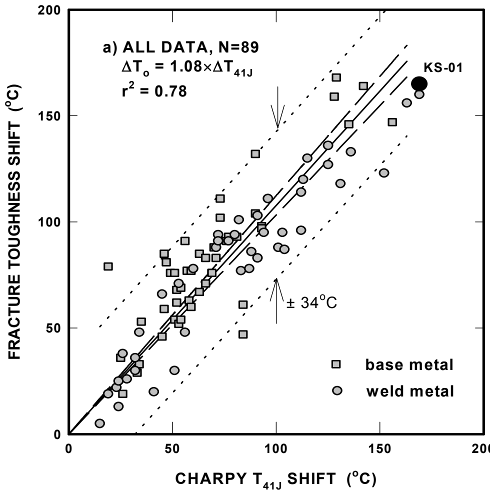

Fig. 5. Relationship between fracture toughness T_o and Charpy T_{41J} shifts from Ref. [5] and position of KS-01 weld from the present study.

V. ATOM PROBE CHARACTERIZATION

The microstructure of the weld was characterized with the Oak Ridge National Laboratory's energy-compensated optical position-sensitive atom probe. The analyses were acquired with a specimen temperature of 50K, a pulse fraction of 20% of the standing voltage, and a pulse repetition rate of 1.5 kHz. The presence of clusters was determined with the maximum separation method [8]. This method is based on the premise that the distance between solute atoms in a solute-enriched cluster or precipitate is significantly smaller than that in the surrounding matrix. Therefore, the atoms that belong to a solute-enriched cluster may be distinguished from those in the matrix based on a maximum separation distance, dmax.

Atom probe tomography revealed a high number density (~3×10^{24} m^{-3}) of Cu-, Mn-, Ni-, Si- and P-enriched precipitates in the neutron irradiated weld, as shown in the atom maps in Fig. 6. A lower number density (~1×10^{23} m^{-3}) of P clusters was also observed. Compositions of individual precipitates were determined by the maximum separation envelope method [8]. A significant variation in the solute concentrations was observed. The average composition of these precipitates was Fe- 17.0 ± 9.7 at. % Cu, 31.9 ± 13.8 at. % Ni, 31.7 ± 11.8 at. % Mn, 1.7 ± 1.7 at. % Si, 0.10 ± 0.10 % Cr, 0.20 ± 0.2 % P, and 0.17 ± 0.17 % Mo. These values represent significant enrichments of Cu (107x), Ni (27x), Mn (27x), Si (5.4x), and P (5.9x), and depletions of Cr (0.2x), Mo (0.6x) and Fe (0.2x), compared to the matrix composition.

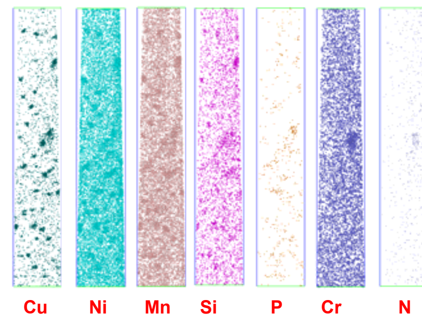

Fig. 6. Atom maps showing the solute distribution in neutron irradiated KS-01 weld. A high number density of Cu-, Mn- Ni-, Si- and P-enriched precipitates is evident.

VI. SUMMARY

Specimens of radiation-sensitive KS-01 weld with intentionally high contents of several elements, including copper, nickel, and manganese, were examined before and after irradiation at 288°C to an average fluence of 0.8×10^{19} neutron/cm^2 (E > 1MeV). The main observations from this study are as follows:

1. The high-strength, radiation-sensitive KS-01 weld exhibited a large Charpy T_{41J} shift of 169°C and an upper-shelf energy drop from 118J to 78J as a result of irradiation. Irradiation also altered the shape of the Charpy transition curve such that the Charpy T_{68J} shift, 217°C, was much larger than the T_{41J} shift.
2. Yield strength increased from 600 MPa to 826 MPa as a result of irradiation.
3. Master curve analysis indicated a fracture toughness T_o shift of 165°C which is in remarkable agreement with the Charpy T_{41J} shift.
4. Despite a high level of embrittlement and hardening, relationships between embrittlement and hardening of KS-01 weld follow the general trend for RPV steels.
5. In general, irradiated fracture toughness data follow the master curve shape. However, low toughness brittle fractures occurred at temperatures further above T_o (T_o + 61 °C) than expected with a leveling of the K_{Jc} data from the master curve shape concept.

6. The high copper, manganese and nickel levels in the weld produced a high supersaturation of copper, and hence a high number density of copper-, nickel-, manganese-, silicon- and phosphorus-enriched precipitates. The high phosphorus level in the weld also produced a distribution of phosphorus clusters.

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