EXTRAPOLATION OF FRACTURE TOUGHNESS DATA FOR HT9 IRRADIATED AT TEMPERATURES 360-390°C—R. J. Kurtz and D. S. Gelles (Pacific Northwest National Laboratory)*

OBJECTIVE

The objective of this task is to provide estimated HT9 cladding and duct fracture toughness values for test (or application) temperatures ranging from -10°C to 200°C, after irradiation at temperatures of 360-390°C. This is expected to be an extrapolation of the limited data presented by Huang[1, 2]. This extrapolation is based on currently accepted methods (ASTM 2003 Standard E 1921-02), and other relevant fracture toughness data on irradiated HT9 or similar alloys.

SUMMARY

Following irradiation in the AC01 test at 360°C to 5.5 x 10^{22} n/cm^2, two HT9 samples tested at 30°C were found to have fracture toughness levels of 28.2 and 31.9 MPa $m^{1/2}$, whereas a third identical specimen tested at 205°C gave 126 MPa $m^{1/2}$. Based on testing of notched tensile specimens from the same irradiation test, the low toughness was a result of brittle fracture. A similar low level of toughness has also been demonstrated in HT9 following irradiation at 250°C and therefore such behavior is reproducible.

Using ASTM Standard E1921-02, which characterizes the fracture toughness of ferritic steels that experience onset of cleavage cracking at instabilities, it has been shown that these data can be analyzed by a Master Curve approach, and that the trend of the fracture toughness over a wider range of temperatures can be estimated. Master Curve analysis shows that toughness will remain low over a wide range of temperatures near 30°C, but will degrade only slightly when temperatures drop below that value. Application of the ASTM Standard methodology did not permit a rigorous, statistically significant determination of the lower bound fracture toughness of HT9 due to the limited data available.

PROGRESS AND STATUS

Introduction

The FFTF project is planning on shipping irradiated sodium-bonded metal fuel pins with HT9 cladding material, to INEEL. These pins are to be shipped in the T-3 Cask, either as full fuel assemblies or loose pins packaged in a liner. The current NRC license does not address this type of fuel packaging. An addendum to the Safety Analysis Report (FFTF-14624), which addresses these changes, has been prepared for acceptance by the DOE/NRC and is to be in compliance with the requirements of 10CFR-71. The structural section of FFTF-14624, addresses the potential for “brittle fracture” of the fuel pins under hypothetical accident conditions (HAC). For cold HAC, a fuel pin temperature of 10°C was estimated. Fracture toughness data [1, 2] relevant to the brittle fracture of the fuel pins consist of 3 data points (one at 205°C and two at 30°C). The planned shipments may be below the temperature range of the test data. Fracture toughness values used in previous evaluations were based on straight-line extrapolation, which are conservative.

Approach

Since the generation of an irradiation effects data base for HT9 fracture toughness that ended about 1989, considerable progress has been made to understand the consequences of irradiation on fracture toughness and to generate a means for extrapolating the available data base. Much of that work is based on the behavior of pressure vessel steels, but it should be directly pertinent for HT9, a “super 12Cr steel” intended for high temperature applications. That body of work has resulted in an ASTM Standard for fracture toughness evaluation, designated E1921-02 and entitled “Determination of Reference

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Temperature T_0, for Ferritic Steels in the Transition Range" [3]. The standard takes note of the fact that at low temperatures, fracture toughness response replicates that of Charpy Impact behavior, and shows a transition from ductile-to-brittle behavior with decreasing temperature. The standard defines the parameters for a Master Curve for a given alloy in terms of a reference temperature, T_0. The Master Curve defines the temperature dependence for fracture toughness in the ductile-to-brittle transition range in the form:

$$K_{Jc(med)} = 30 + 70 \exp [0.019(T - T_0)], \text{ MPa m}^{1/2} \quad . . . . . (1)$$

where $K_{Jc(med)}$ is the median value for fracture toughness at a given temperature, T, in units of MPa $m^{1/2}$. All materials, irradiation conditions, test conditions and specimen geometries will behave similarly and can be fit with the adjustable parameter T_0. Therefore, data generated with different specimen geometries can be converged, and different material heat treatments and irradiation conditions may result in correspondingly different values for T_0. Using this approach, it should be possible to estimate HT9 fracture toughness values for test (or application) temperatures ranging from -10°C to 200°C, after irradiation at the 360-390°C temperature range given data obtained at room temperature and 205°C.

HT9 Data Base

An HT-9 fracture toughness data base has been generated primarily by F. H. Huang and co-workers as part of the Department of Energy National Cladding and Duct (NCD) and Fusion Reactor Materials (FRM) Programs with more recent contributions from UCSB [1, 2, 4-6]. The NCD program was intended to qualify HT9 as a duct and fuel cladding material for fast breeder reactor applications, and the FRM program followed with the intention of expanding the data base to lower irradiation temperatures and to include weldments. The standard specimen geometry used by Huang is shown in Figure 1, with B at 2.54 mm, W at 11.94 mm and diameter at 16 mm. Specimens were fatigue pre-cracked to a total crack length of about 6 mm leaving an uncracked ligament of about 6 mm. The thickness selected corresponds to that of a standard FFTF duct. Specimens had electrodes attached so that a crack length measurement could be obtained from each specimen tested using a potential drop technique.

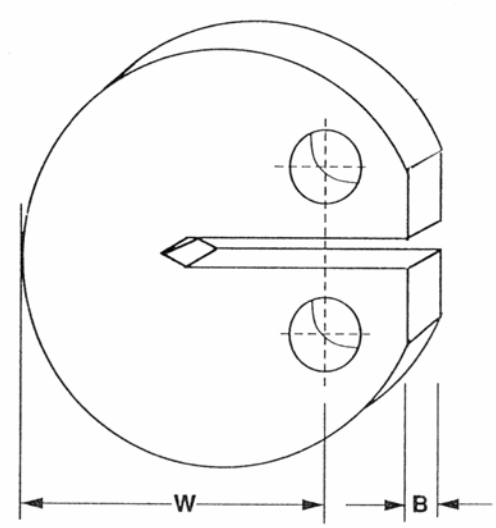

Figure 1. Specimen geometry used by Huang.

The Huang data base for HT9 base metal along with the UCSB bend bar data is shown in Figure 2. {Where K_C values were measured, they have been converted to J_C values using the relation $J_c=K_c^2/E$ where E is the elastic modulus.} The data base contains both unirradiated and irradiated specimens, the former shown with solid symbols and the latter with crossed or open symbols. From Figure 2, the behavior for unirradiated specimens shows low values of fracture toughness at very low temperatures due to brittle fracture, and a strong dip in toughness at ~300°C due to interstitial impurity hardening associated with dynamic strain aging.

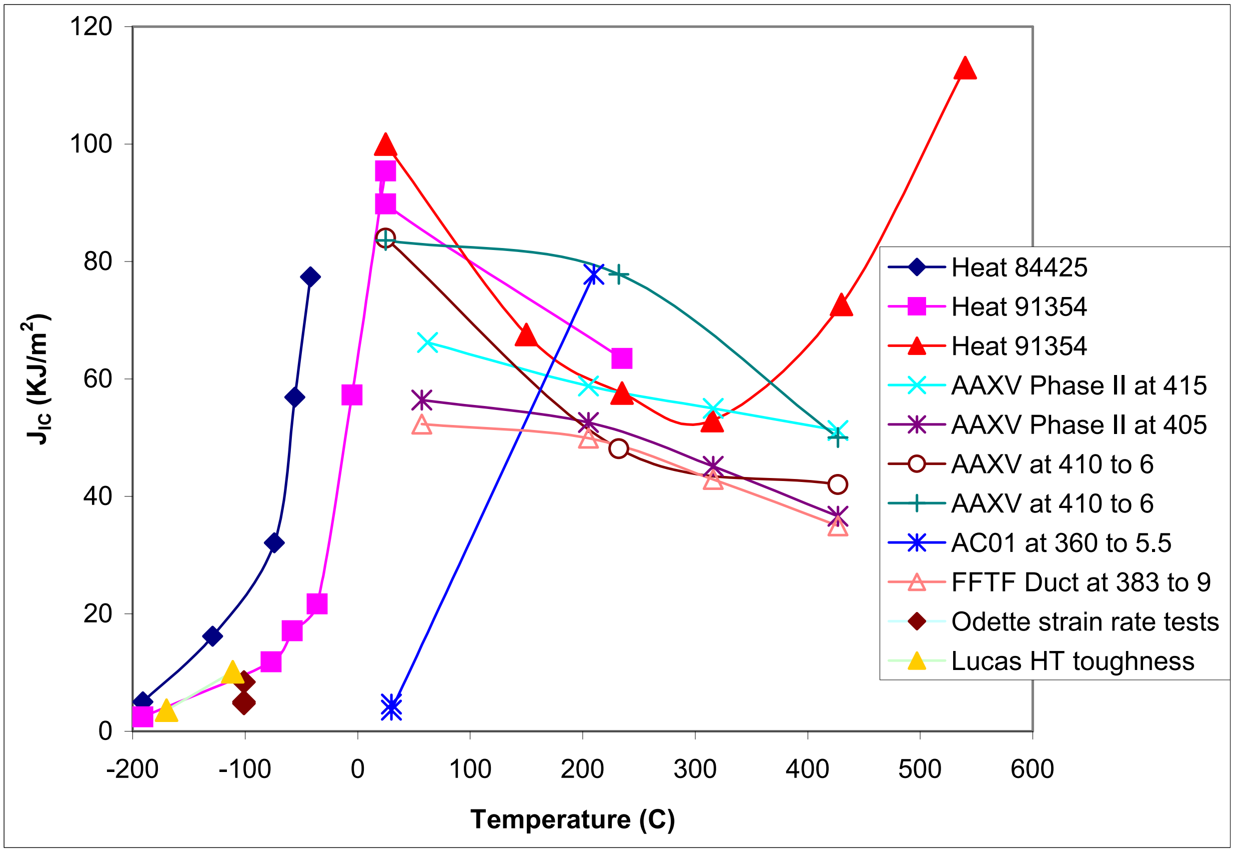

Figure 2. HT9 fracture toughness data base generated by Huang and co-workers. Unirradiated conditions are denoted by filled solid data points.

Irradiation at temperatures of 390°C and higher is found to cause only minor changes in toughness whereas irradiation at 360°C, based on the AC01 test gave two measurements on the order of 5 KJ/m^2 when tested at room temperature. Therefore, irradiation at temperatures below 390°C can result in large reductions in fracture toughness. Similar behavior was reported in support of the FRM program. [7] There it was found that following irradiation at 250°C to low dose, the fracture toughness at room temperature was also 5 KJ/m^2. However, those results were not included in Figure 2 because toughness levels reported for unirradiated conditions were approximately four times those of Huang, and the differences were attributed to a different, softer tempering treatment (780°C/2h verses 740-760°C/1h for Huang). Such high toughness levels should make it very difficult to obtain valid measurements on miniature specimens. Also, it can be noted that independent measurements showed toughness levels on the order of 100 KJ/m^2 for HT9 at room temperature after a tempering treatment of 780°C/2.5h, in agreement with the Huang data but in disagreement with the FRM data [8].

Fracture toughness measurements have also been made for HT9 weld metal and heat-affected-zone (HAZ) specimens. The results are shown in Figure 3, in comparison to the unirradiated base metal data. The temperature dependence at high temperatures was somewhat different, but toughness values remained high following irradiation at 390°C or higher.

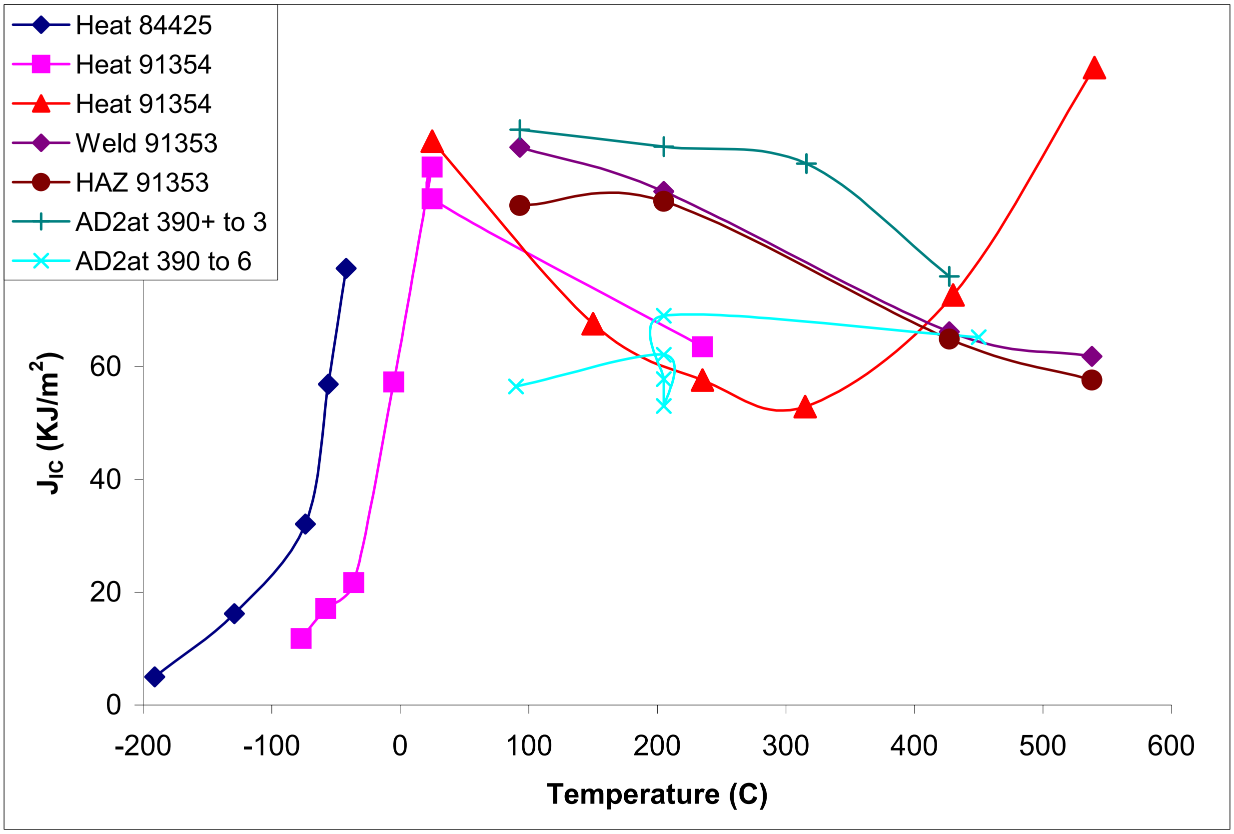

Figure 3. Fracture toughness response in HT9 weld specimens.

Therefore, some fracture toughness data exists for HT9, including testing following irradiation. Measurements of fracture toughness on the order of 5 KJ/m^2 do occur in unirradiated samples tested at temperatures below 0°C, as well as in specimens irradiated at 250 and 360°C when tested at room temperature. The fracture appearance for unirradiated samples with low fracture toughness was brittle, but fractography was not performed on irradiated samples showing low fracture toughness. However, a notched tensile specimen irradiated under identical conditions at 360°C and tested at room temperature was shown to have a brittle appearance [1]. Therefore, it is likely that the fracture toughness levels on the order of 5 KJ/m^2 for irradiated specimens are a result of an irradiation induced ductile-to-brittle transition.

Analysis

The ASTM Standard, E1921-02, covers the determination of a reference temperature, T_0, that characterizes the fracture toughness of ferritic steels, which experience the onset of cleavage cracking at elastic, or elastic-plastic instabilities, or both. The specific types of steels covered are those with yield strengths ranging from 275 to 825 MPa. The temperature dependence of fracture toughness is assumed to conform to a standard shape known as the Master Curve (Equation 1). The Master Curve is indexed to a reference temperature, T_0. Variations in material properties such as heat treatment and irradiation history are characterized by temperature shifts. The standard places requirements on specimen size and the number of replicate tests needed to reliably determine T_0 and, therefore, the temperature dependence of the fracture toughness for a given metallurgical condition. The specimen remaining ligament must be sufficiently large to ensure that a condition of high crack-front constraint exists at fracture in order to obtain valid fracture toughness measurements. The maximum K_{Jc} capacity of a specimen is given by:

$$K_{limit} = [(Eb_0\sigma_{ys})/30(1-\nu^2)]^{1/2} \quad (2)$$

where E is Young's modulus, b_0 is the remaining ligament, σ_{ys} is the material yield strength at the test temperature, and is ν Poisson's ratio. A minimum of six tests is needed at a single test temperature in order to satisfy statistical requirements of the standard. More than six tests may be needed if the testing is performed over a range of temperatures. All of the results are normalized to a standard specimen thickness of 25.4 mm. The standard includes a size effect adjustment, as follows:

$$K_{Jc(x)} = K_{min} + [K_{Jc(o)} - K_{min}](B_o/B_x)^{1/4} \quad (3)$$

where:

$K_{Jc(o)} = K_{Jc}$ for a specimen size B_o,
B_o = gross thickness of test specimens (side grooves ignored),
B_x = gross thickness of prediction (side grooves ignored), and
K_{min} = 20 MPa $m^{1/2}$.

However, the application under consideration in this study is for cladding and duct where thicknesses do not exceed 2.54 mm, and the data available that has been generated by Huang is for a specimen thickness of 2.54 mm. Note that greater thickness results in higher values of T_0 and therefore lower values of toughness at a given temperature.

The relevant fracture toughness data used for estimating T_0 is presented in Table 1. Note that none of the data sets satisfy statistical requirements for the number of replicate tests since multiple test temperatures were used. Also note that tests performed at –191°C (italicized values in Table 1) were excluded from the analysis since these tests fell outside the acceptable temperature range for T_0 determination. Values of E, σ_{ys}, and ν used to compute the K_{Jc} measurement capacity of the disc compact tension specimen utilized by Huang were obtained from Spatig et al., [9] an IEA data base report, [10] and Zinkle, et al [11]. The temperature dependence of the elastic properties are plotted in Figure 4 and the yield strength in Figure 5. Calculation of K_{limit} values shows that none of the specimens violated dimensional requirements, but it should be noted that at high fracture toughness the values of K_{Jc} that meet the requirements of Eq. 2 may not always provide a unique description of the crack-front stress-strain fields due to some loss of constraint caused by excessive plastic flow. This condition may be more prevalent in materials with low strain hardening characteristics.

Values for T_0 have been estimated for four sets of HT9 data, three unirradiated conditions and one condition following irradiation. These analyses are shown in Figures 6 through 9. Figure 6 shows Master Curve analysis for Heat 84425 from [4] which gives T_0 at –25.5°C. Figure 7 shows Master Curve analysis for a different heat of HT9, 91353, from [4] which gives T_0 at 34.1°C. Figure 8 shows Master Curve analysis for heat 84425 tested at a higher strain rate of 3.2×10^{-2} from [4] (and not plotted in Figure 2) which gives T_0 at 33°C. Applying the same criterion to the AC01 test data, Figure 9 shows Master Curve analysis for heat 91354 irradiated at 360°C [1-2] which gives T_0 at 238°C. It should be noted that the confidence bounds presented in Figures 6 through 9 are tied directly to the T_0 values obtained from application of the ASTM analysis methodology, but none of the data sets satisfy ASTM statistical requirements so these curves may not bound the data.

Discussion

The ASTM Standard E1921-02 for Master Curve analysis of ductile-to-brittle fracture toughness behavior was developed after an irradiation effects data base for HT9 was generated. The Standard recommends that tests be replicated six times in order to estimate a median K_{Jc} because extensive scatter among replicate tests is expected. The recommended test temperature for those tests is one giving a toughness of ~100 MPa $m^{1/2}$, but testing over a range of temperatures is allowed, given the understanding that the uncertainty in T_0 determination increases as the toughness level decreases (i.e., more specimens are

Table 1. Fracture Toughness Data for Unirradiated and Irradiated HT9 Used to Determine T_0

T, °C	E, MPa	σ_{ys}, MPa	J_c, KJ/m^2	K_{Jc}, MPa m$^{1/2}$	$K_{Jc(1T)}$, MPa m$^{1/2}$	K_{limit}, MPa m$^{1/2}$
Heat 84425						
-191	*228*	*1099*	*5*	*35*	*29*	*234*
-129	225	806	16.2	63	44	199
-74	222	643	32.1	88	58	177
-56	221	608	56.9	117	75	171
-42	220	589	77.4	136	85	168
Heat 91353						
-191	*228*	*1099*	*2.43*	*25*	*23*	*234*
-77	222	650	11.8	53	39	178
-58	221	612	17.1	64	45	172
-36	220	582	21.7	72	49	167
-5	218	562	57.3	117	74	164
25	216	556	95.4	150	93	162
25	216	556	89.8	146	91	162
Heat 84425 (high strain rate)						
-191	*228*	*1099*	*2.5*	*25*	*23*	*234*
-74	222	643	11	52	38	177
-58	221	612	17	64	45	172
-36	220	582	22	73	50	167
-6	218	562	57	116	74	164
25	216	556	97	151	94	162
Heat 91354						
30	216	1095	-	32	27	227
30	261	1095	-	28	25	250
210	206	568	-	126	80	160

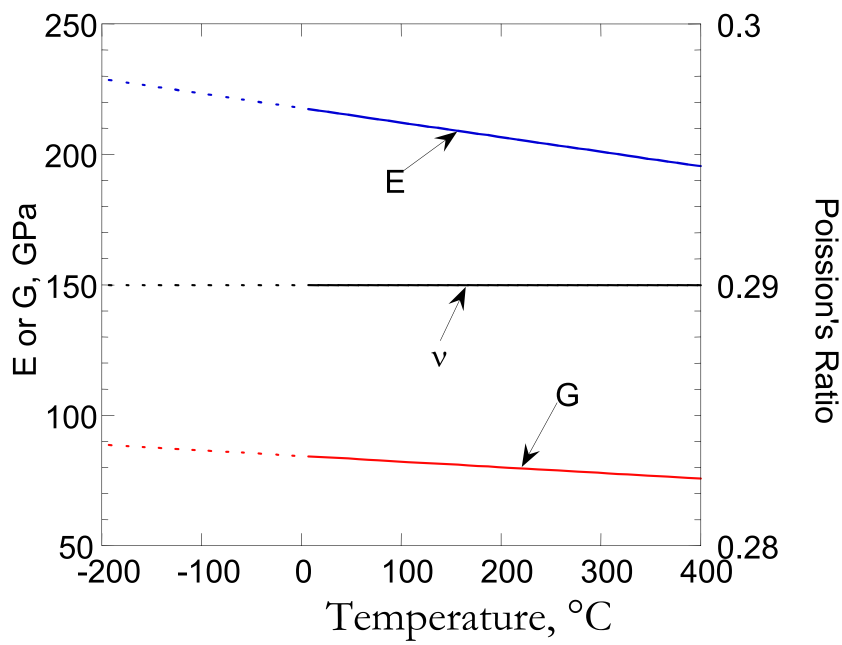

Figure 4. Temperature dependence of Young's modulus (E), shear modulus (G) and Poisson's ratio (ν).

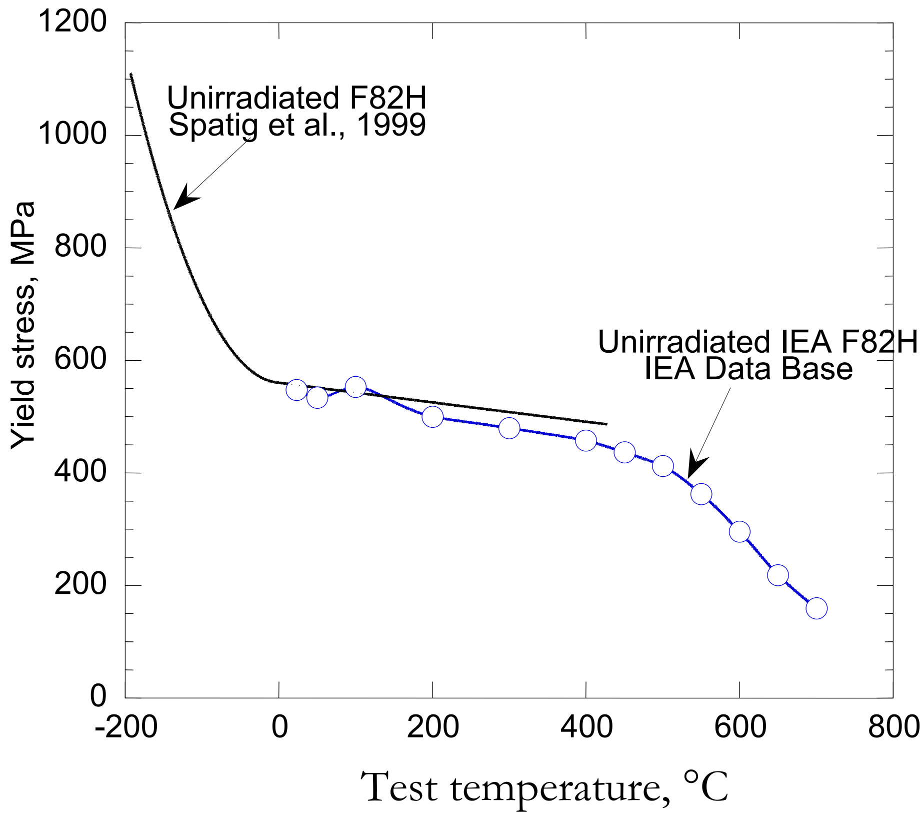

Figure 5. Temperature dependence of the 0.2% offset yield strength for F82H.

required to reliably compute T_0). Therefore, insufficient data has been obtained by Huang to satisfy ASTM Standard criterion for HT9 duct irradiated at 360°C to 5.5 x 10^{22} n/cm^2. Nonetheless, extrapolation of available data from the AC01 test may be justifiable in the spirit of the Standard, and a Master Curve should be a better estimate of fracture toughness over the temperature range -10°C to 200°C than a straight line extrapolation of the data points, shown in Figure 2, would provide. The Master Curves presented in Figures 6 through 9 show the trend of the HT9 fracture toughness as a function of temperature, but they cannot give precise lower bound fracture toughness values due to the data base limitations mentioned above.

Fracture toughness behavior for HT9 duct and cladding can be estimated based on the ASTM standard using Equation 3. As the cladding to be shipped has a thickness of 0.559 mm (0.022"), corrections for K_{Jc} can be obtained and a value for T_0 estimated. Results are shown in Figures 10 for duct and in Figure 11 for cladding with T_0 estimated at 203 and 178°C, respectively. Comparison of Figures 10 and 11 reveals that below room temperature, the curves are effectively identical, but the data points at 30°C are shifted up, from 28 and 32 to 32 and 37 or about 15%.

However, it is not clear that such an approach is valid for very thin samples. Therefore, it is perhaps best to expect that toughness in thin sections should increase but the actual magnitude estimated by the standard for going from duct to cladding geometries of 15% may be low.

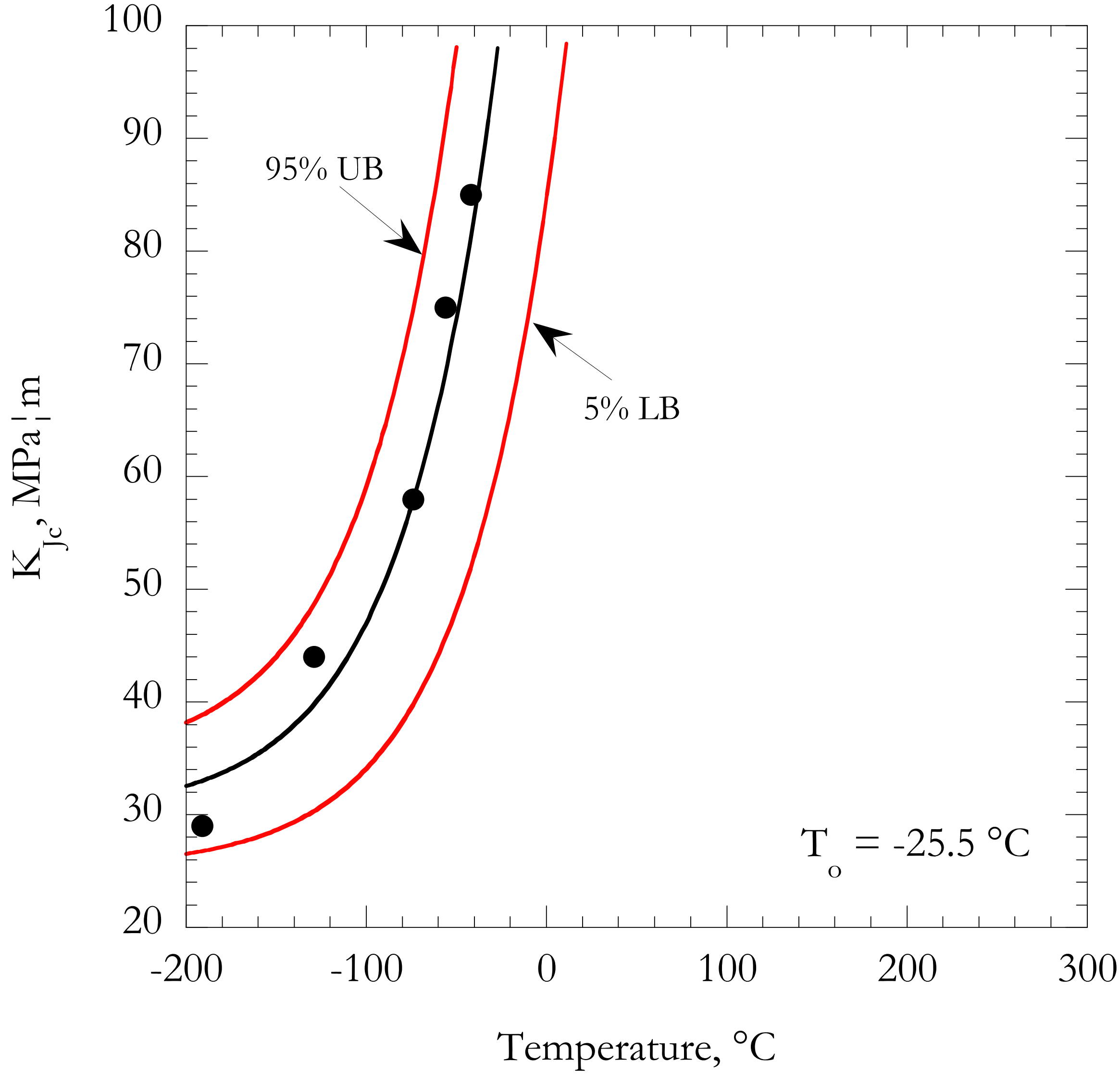

Figure 6. Heat 84425 Master Curve with thickness adjustment to the ASTM Standard of 25.4 mm. This material is unirradiated. The calculated reference temperature, T_0, is –25.5°C. The predicted median curve is shown in black with the measured values plotted as filled circles. Upper (95%) and lower (5%) bounds on the median curve are shown in red.

A concern can be raised that the fluence obtained in the AC01 experiment of 5.5 x 10^{22} n/cm^2 (E>0.1MeV) was insufficient to reach saturation. If this were the case, then irradiation to higher fluence might result in further degradation of fracture toughness properties. This concern can be countered in two ways. Sufficient Charpy impact data exists to show that impact energy changes saturate by 3 x 10^{22} n/cm^2 [12]. As behavior is qualitatively similar between Charpy impact and fracture toughness, it is therefore likely that saturation in toughness degradation has been achieved by 5.5 x 10^{22} n/cm^2. However, the Master Curve approach emphasizes that once a material has been embrittled so that response is brittle, and then fracture toughness will degrade little more.

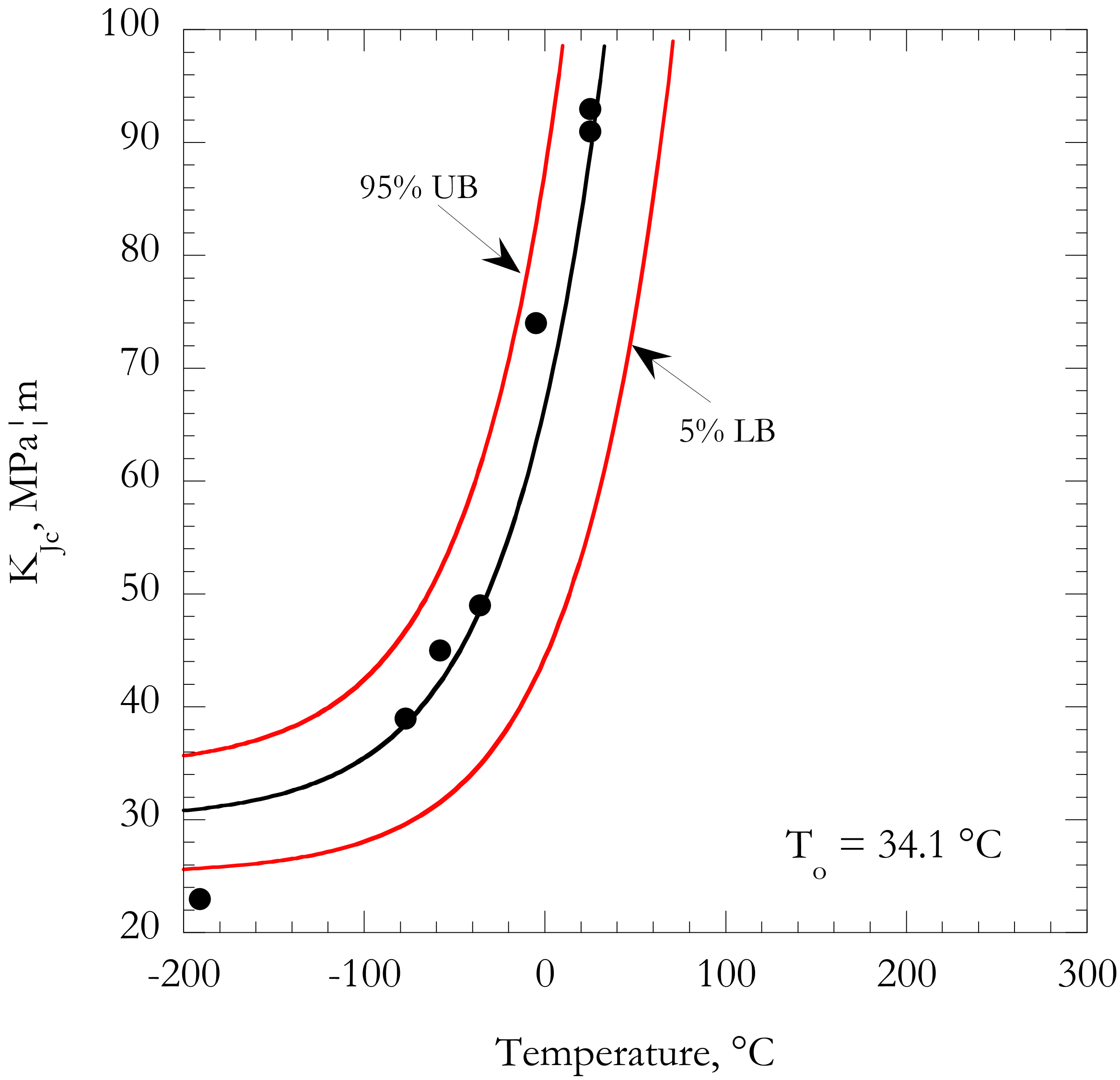

Figure 7. Heat 91353 Master Curve with thickness adjustment to the ASTM Standard of 25.4 mm. This material is unirradiated. The calculated reference temperature, T_0, is 34.1°C. The predicted median curve is shown in black with the measured values plotted as filled circles. Upper (95%) and lower (5%) bounds on the median curve are shown in red.

It is worthwhile to emphasize the consequences of Master Curve analysis. Extrapolation of toughness data due to brittle fracture both above and below the test temperature of 30°C results in only minor further changes in toughness. For example, from Figure 10 for the case of an HT9 duct irradiated at 360°C to 5.5 x 10^{22} n/cm^2, the median fracture toughness expected at 30°C is 33 MPa $m^{1/2}$. The temperature at which the median value drops to 30 MPa $m^{1/2}$ is –200°C, but at 100°C, the median toughness is only 40 MPa $m^{1/2}$. Therefore, toughness degradation is insensitive to temperature fluctuations anticipated during shipment. However, in comparison, linear extrapolation of the AC01 data gives misleading predictions.

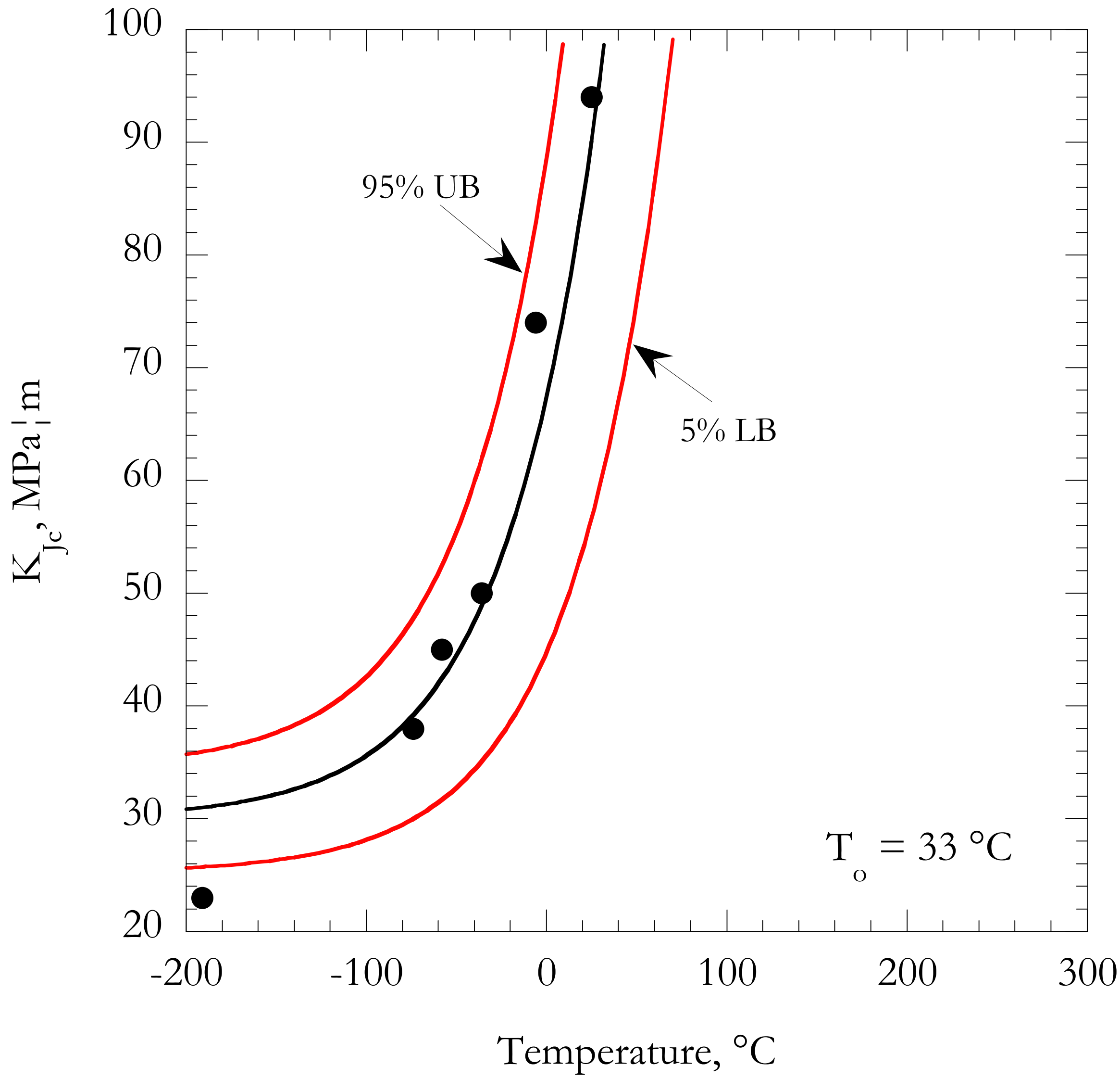

Figure 8. Heat 84425 (tested at high strain rate) Master Curve with thickness adjustment to the ASTM Standard of 25.4 mm. This material is unirradiated. The calculated reference temperature, T_0, is 33°C. The predicted median curve is shown in black with the measured values plotted as filled circles. Upper (95%) and lower (5%) bounds on the median curve are shown in red.

We have attempted to estimate the difference in toughness between HT9 ducts and HT9 cladding based on the ASTM Standard E 1921-02. An improvement on the order of 15% is predicted at 30°C, with less improvement at lower temperatures. However, we suspect that these procedures do not apply to such thin material, and recommend further analysis be performed.

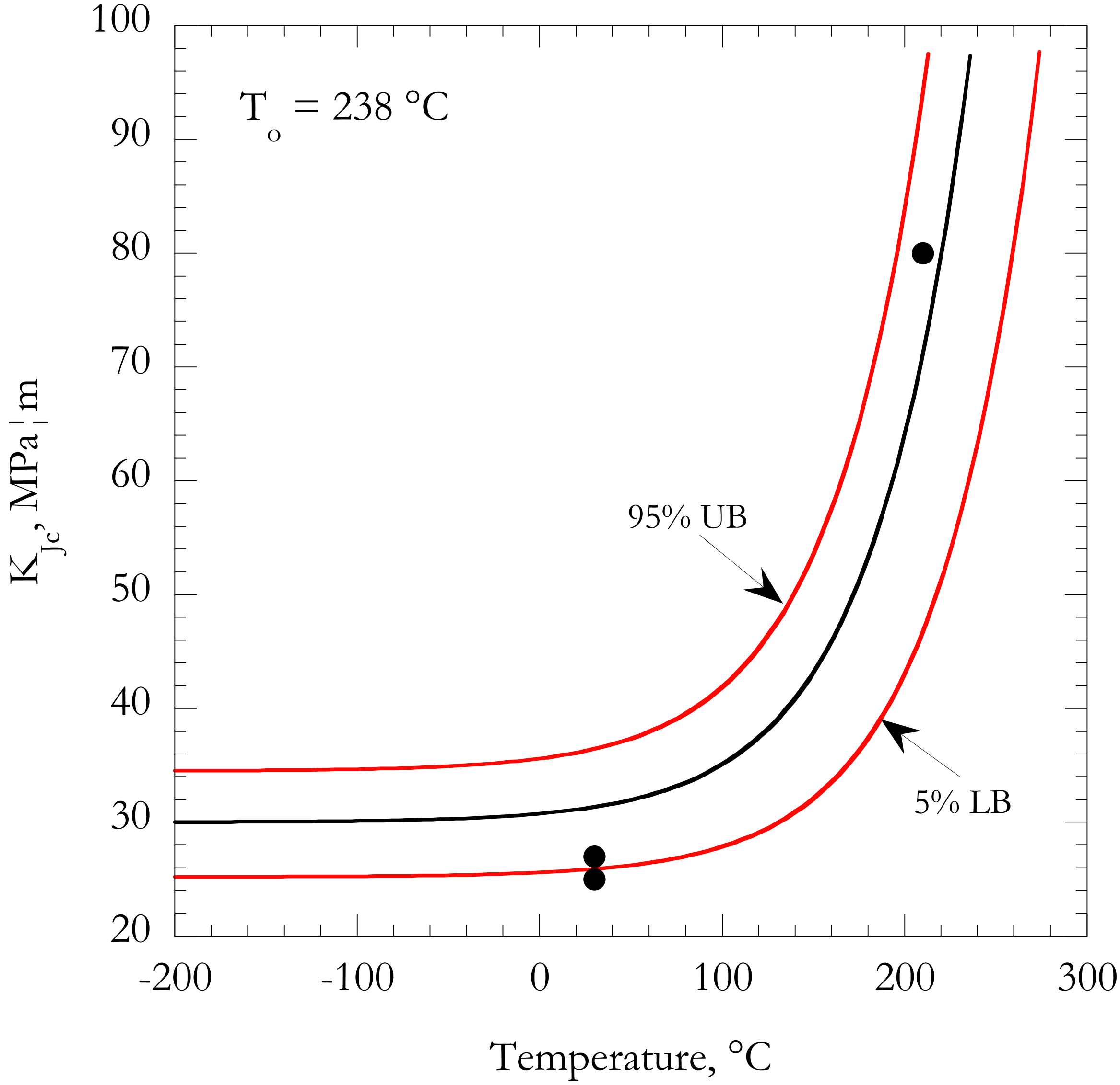

Figure 9. Heat 91354 Master Curve with thickness adjustment to the ASTM Standard of 25.4 mm. This heat was irradiated at 360 °C to 5.5 x 10^{22} n/cm^2 (E>0.1MeV). The calculated reference temperature, T_0, is 238°C. The predicted median curve is shown in black with the measured values plotted as filled circles. Upper (95%) and lower (5%) bounds on the median curve are shown in red.

Conclusions

Following irradiation in the AC01 test at 360°C to 5.5 x 10^{22} n/cm^2, two HT9 samples tested at 30°C were measured to have fracture toughness levels of 28.2 and 31.9 MPa $m^{1/2}$, respectively, whereas a third identical specimen tested at 205°C gave 126 MPa $m^{1/2}$. Based on testing of notched tensile specimens from the same irradiation test, the low toughness was a result of brittle fracture. A similar low level of toughness has also been demonstrated in HT9 following irradiation at 250°C and therefore such behavior is reproducible.

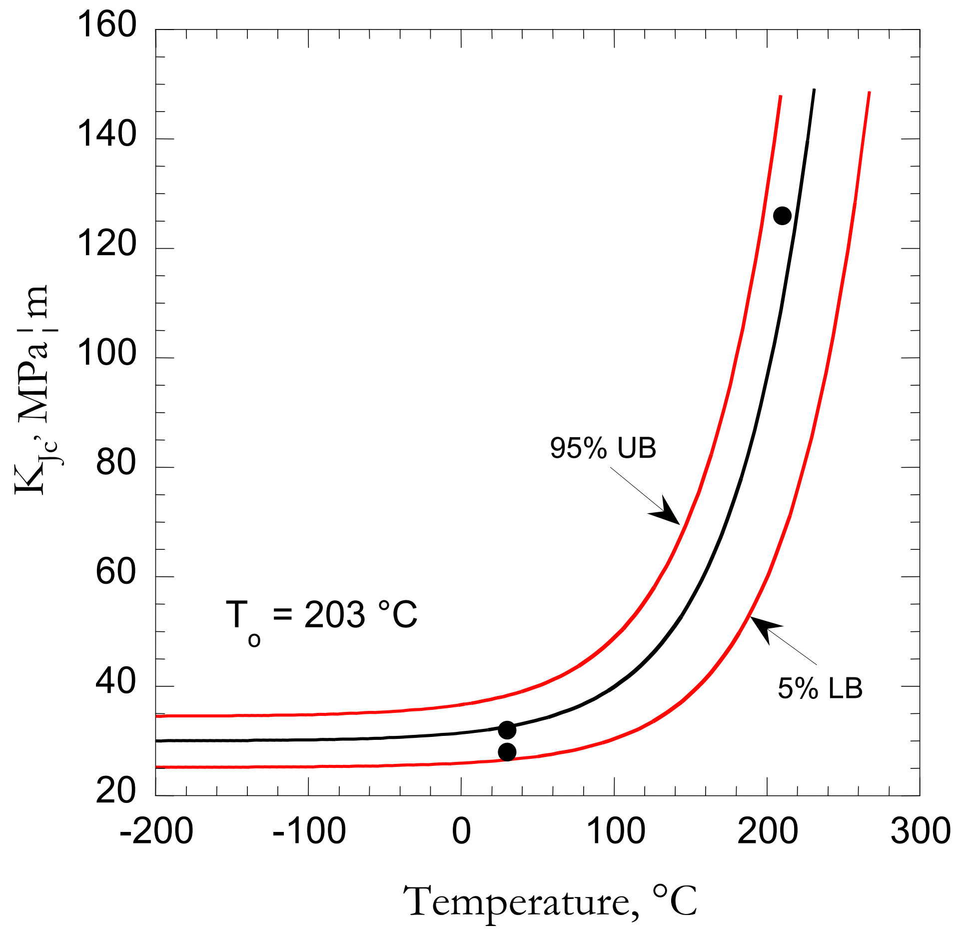

Figure 10. Heat 91354 Master Curve without thickness adjustment to the ASTM Standard of 25.4 mm. This heat was irradiated at 360 °C to 5.5 x 10^{22} n/cm^2 (E>0.1MeV). The calculated reference temperature, T_0, is 203°C. The predicted median curve is shown in black with the measured values plotted as filled circles. Upper (95%) and lower (5%) bounds on the median curve are shown in red. Note the curves shown in this figure are not strictly in accordance with the ASTM Master Curve analysis since this analysis applies only to specimens 25.4 mm thickness.

Using ASTM Standard E1921-02 which characterizes the fracture toughness of ferritic steels that experience onset of cleavage cracking at instabilities, it has been shown that these data can be analyzed by a Master Curve approach, and that the trend of the fracture toughness over a wider range of temperatures can be estimated. Master Curve analysis shows that toughness will remain low over a wide range of temperatures near 30°C, but will degrade only slightly when temperatures drop to –10°C. Application of the ASTM Standard methodology did not permit a rigorous, statistically significant determination of the lower bound fracture toughness of HT9 due to the limited data available.

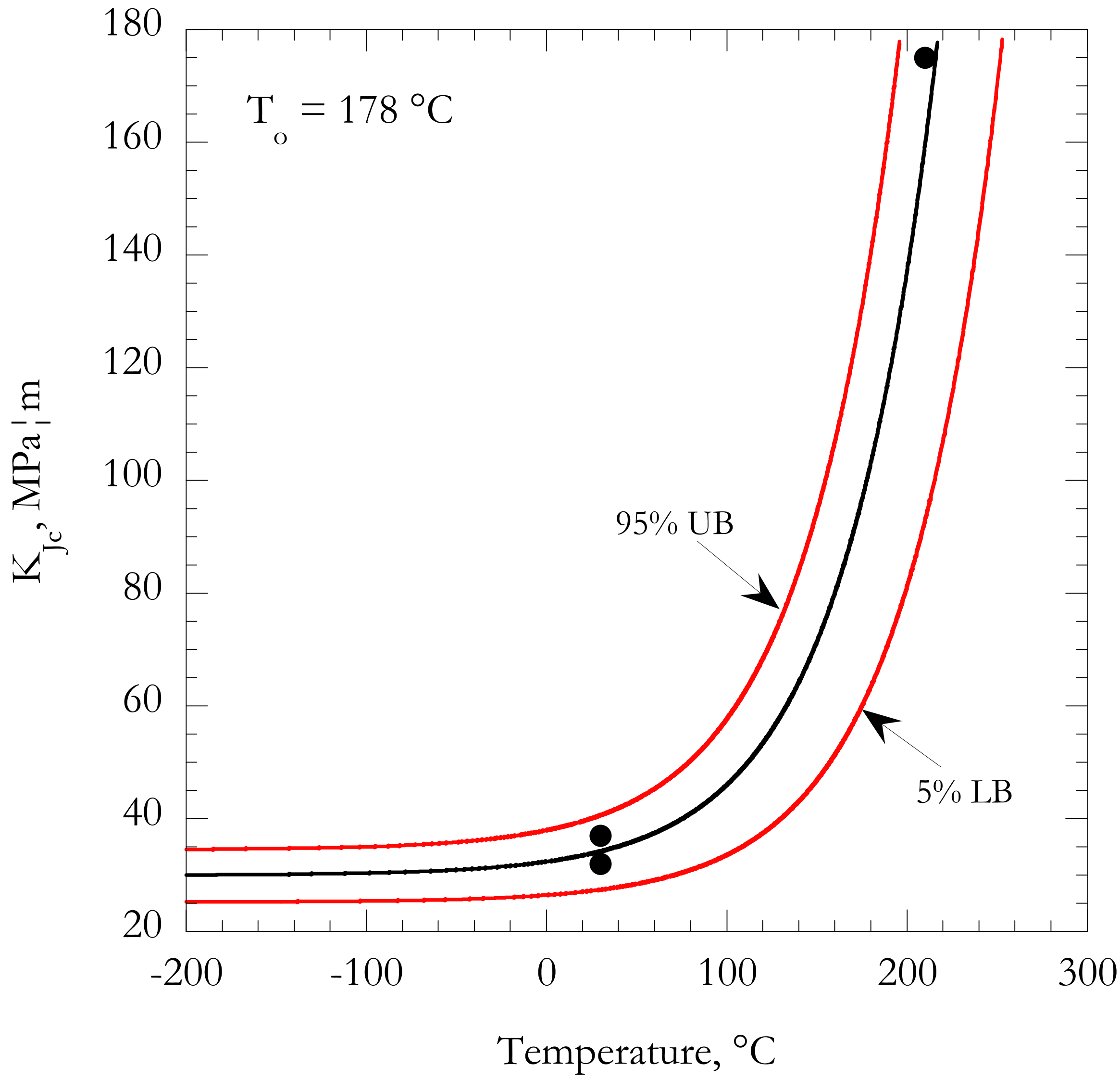

Figure 11. Heat 91354 Master Curve with thickness adjustment from 2.54 mm to 0.559 mm to estimate the fracture toughness of HT-9 fuel cladding. This heat was irradiated at 360°C to 5.5 x 10^{22} n/cm^2 (E>0.1MeV). The calculated reference temperature, T_0, is 178°C. The predicted median curve is shown in black with the measured values plotted as filled circles. Upper (95%) and lower (5%) bounds on the median curve are shown in red. Note the curves shown in this figure are not strictly in accordance with the ASTM Master Curve analysis since this analysis applies only to specimens 25.4 mm thickness.

Future Work

The effort will be continued as opportunities become available.

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