

Paper ID 5476

To be published in *Effects of Radiation on Materials, 19th International Symposium, ASTM STP 1366*, held in Seattle, Washington, June 16-18, 1998

Irradiation, Annealing, and Reirradiation Effects on American and Russian Reactor Pressure Vessel Steels

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The logo consists of the lowercase letters 'oml' in a bold, stylized, sans-serif font. The 'o' and 'm' are connected at the top, and the 'l' is a simple vertical bar.

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Reference: Sokolov, M. A., Chernobaeva, A. A., Nanstad, R. K., Nikolaev, Y. A., and Korolev, Y. N., "Irradiation, Annealing, and Reirradiation Effects on American and Russian Reactor Pressure Vessel Steels," *Effects of Radiation on Materials, 19th International Symposium, ASTM STP 1366*, M. L. Hamilton, A. S. Kumar, S. T. Rosinski, and M. L. Grossbeck, Eds., American Society for Testing and Materials, West Conshohocken, Pennsylvania, 1999.

Abstract: One of the options to mitigate the effects of irradiation on reactor pressure vessels (RPVs) is to thermally anneal them to restore the toughness properties that have been degraded by neutron irradiation. Even though a postirradiation anneal may be deemed successful, a critical aspect of continued RPV operation is the rate of embrittlement upon reirradiation. There are insufficient data available to allow for verification of available models of reirradiation embrittlement or for the development of a reliable predictive methodology. This is especially true in the case of fracture toughness data. Under the U.S.-Russia Joint Coordinating Committee for Civilian Nuclear Reactor Safety (JCCCNRS), Working Group 3 on Radiation Embrittlement, Structural Integrity, and Life Extension of Reactor Vessels and Supports agreed to conduct a comparative study of annealing and reirradiation effects on RPV steels. The Working Group agreed that each side would irradiate, anneal, reirradiate (if feasible), and test two materials of the other. Charpy V-notch (CVN) and tensile specimens were included. Oak Ridge National Laboratory (ORNL) conducted such a program (irradiation and annealing, including static fracture toughness) with two weld metals representative of VVER-440 and VVER-1000 RPVs, while the Russian Research Center-Kurchatov Institute (RRC-KI) conducted a program (irradiation, annealing, reirradiation, and reannealing) with Heavy-Section Steel Technology (HSST) Program Plate 02 and Heavy-Section Steel Irradiation (HSSI) Program Weld 73W. The results for each material from each laboratory are compared with those from the other laboratory. The ORNL experiments with the VVER welds included irradiation to about 1×10^{19} n/cm² (>1 MeV), while the RRC-KI experiments with the U.S. materials included irradiations from about 2 to 18×10^{19} n/cm² (>1 MeV). In both cases, irradiations were conducted at ~290°C and annealing treatments were

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conducted at $\sim 454^{\circ}\text{C}$. The ORNL and RRC-KI experiments have shown generally good agreement for both the Russian and U.S. steels. While recoveries of the Charpy 41-J transition temperatures were substantial in all cases, significantly less recovery of the lateral expansion and shear fracture in some cases (no recovery in one case) deserves further attention. The RRC-KI results for the U.S. steels showed reirradiation embrittlement rates which are conservative relative to the lateral shift prediction based on Charpy impact energy.

Keywords: Charpy, embrittlement, annealing, reirradiation, reactor pressure vessel

Introduction

One of the options to mitigate the effects of irradiation on RPVs is to thermally anneal them to restore the toughness properties that have been degraded by neutron irradiation. Even though a postirradiation anneal may be deemed successful, a critical aspect of continued RPV operation is the rate of embrittlement upon reirradiation. There are insufficient data available to allow for verification of available models of reirradiation embrittlement or for the development of a reliable predictive methodology. This is especially true in the case of fracture toughness data. Under the U.S.-Russia JCCCNRS, Working Group 3 on Radiation Embrittlement, Structural Integrity, and Life Extension of Reactor Vessels and Supports agreed to conduct a comparative study of annealing and reirradiation effects on RPV steels. The Working Group agreed that each side would irradiate, anneal, reirradiate (if feasible), and test two materials of the other. CVN and tensile specimens have been included. ORNL conducted such a program (irradiation and annealing) with two weld metals representative of VVER-440 and VVER-1000 RPVs including static three-point bend tests of precracked CVN (PCVN) specimens, while the RRC-KI conducted a program (irradiation, annealing, reirradiation, and reannealing) with HSST Plate 02 and HSSI Weld 73W. The results for each material from each laboratory are compared with those from the other laboratory.

Materials

Three welds and one base metal were characterized in the present study. The chemical compositions and mechanical properties in the unirradiated condition are listed in Tables 1 and 2, respectively. The base metal was American Society for Testing and Materials (ASTM) A 533 grade B class 1 plate, designated HSST Plate 02, portions of which have been used in many investigations around the world. The plate was edge-quenched in water after austenitizing at 871°C , then tempered at 663°C for 4 h and furnace cooled. The final stress relief was performed at 621°C for 40 h [1]. One of the welds, designated HSSI Weld 73W, was specially produced for the HSSI Program by Combustion Engineering, Inc., using the submerged-arc process with Linde 0124 flux. The weld was postweld heat treated at 607°C for 40 h [2,3]. The weld is known to be a high-copper (radiation-sensitive) weld.

The two other welds were specially produced by the Izhora Plant to exemplify radiation-sensitive welds of two Russian-made commercial reactors, VVER-440 and

VVER-1000. Both welds were made using the submerged-arc process. The VVER-440 weld, designated Weld 502, was made by weld wire Sv-10KhMFT with flux AN-42M. It is characterized by low nickel and high phosphorus contents (<0.20 and ~0.030%, respectively), typical for older VVER-440 vessel welds. The weld was heat treated at 675°C for 34 h. The VVER-1000 weld, designated Weld 260, was made by weld wire Sv-12Kh2N2MAA with flux FTs-16. The VVER-1000 materials, including the Weld 260, have low copper and phosphorus contents (<0.10 and <0.012%, respectively). Weld 260 is also characterized by high nickel content (1.64%), which is typical for old VVER-1000 vessel welds. The weld was heat treated at 620°C for 17.5 h, furnace cooled, then heat treated at 640°C for 9.5 h.

Test Procedures

The test program concentrated on comparison of changes in Charpy impact properties due to irradiation and annealing. RRC-KI performed tests using the RKP-300 pendulum-type Amsler impact testing machine. A ISO-type instrumented striker with a 2-mm radius was used for tests at RRC-KI. Charpy tests at ORNL were conducted in accordance with ASTM Standard E-23, using an 8-mm radius instrumented striker.

Additionally, ORNL performed a limited number of tensile and static fracture toughness tests. Round tensile specimens with a gage section 4.52 mm in diameter and 31.75 mm in length were tested, while compact and PCVN specimens were used for fracture toughness characterization of irradiated and annealed material.

Analysis of Charpy Data

Charpy absorbed energy data of the four materials studied in the unirradiated condition are presented in Figure 1(a) through (d). Charpy data from both laboratories are in good agreement, despite the difference in the strikers used.

ORNL has been involved in a number of cross-comparison Charpy testing exercises with 2- and 8-mm radii instrumented strikers (including one with RRC-KI) and summarized the comparison results [4]. The overall results have showed generally good agreement in absorbed energy measurements by the two strikers in the energy range below 200 J. The current data are consistent with that observation. Therefore, the unirradiated data from the two laboratories for each material in this study were combined and these results [shown in Fig. 1(a) through (d)] were used as the reference Charpy data set for each material. Charpy data for each material/condition were fit to the hyperbolic tangent function:

$$E = \frac{USE + LSE}{2} + \frac{USE - LSE}{2} \times \tanh\left(\frac{T - T_{MT}}{C}\right), \quad (1)$$

where E is absorbed energy, lateral expansion, or shear fracture; US and LS are upper- and lower-shelf values of E ; T_{MT} is the temperature at the middle of the transition range; and C is the slope of the curve in the transition range at T_{MT} . Fitting was performed with

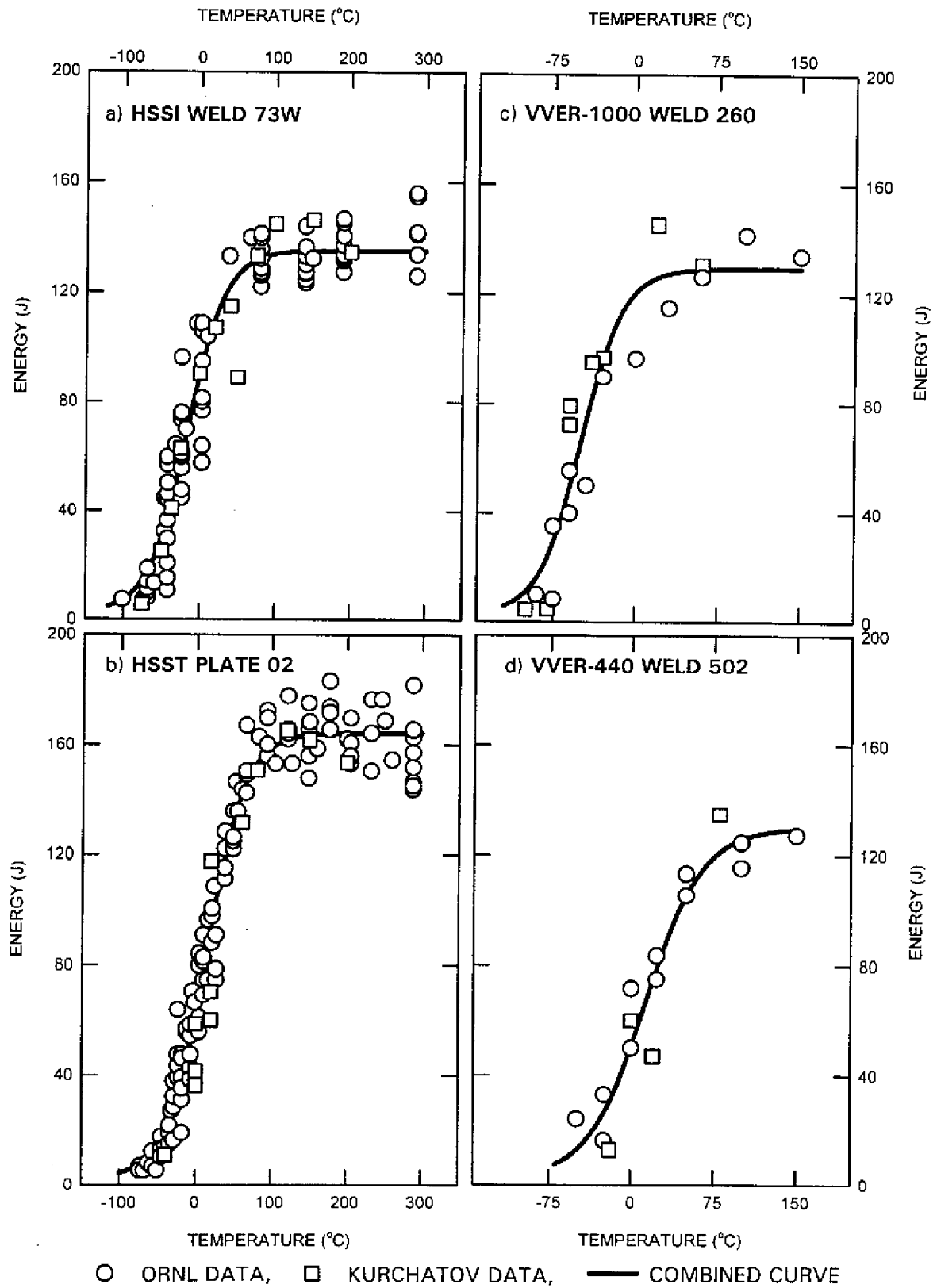


Fig. 1. Charpy data from materials studied in the unirradiated condition tested at ORNL and RRC-KI. Solid line is the best fit to combined data using a hyperbolic tangent function.

LSE fixed at 2.7 J for energy, 0.061 mm for lateral expansion, and 0% for shear, respectively [2]. Additionally, the upper-shelf value for shear was fixed at 100%.

The transition temperature is the main parameter derived from the Charpy tests. It defines a temperature which corresponds to a certain level of energy, lateral expansion, or shear. This level is accepted to be 0.9 mm for lateral expansion and 50% for shear according to the current practices in both countries. However, the radiation-induced transition temperature shift is measured at the level of 41 J in the U.S., while it is measured at 47 J in Russia. Indexing at 41 or 47 J reflects a difference of a historical practice rather than a technical one. The 41-J level corresponds to 30 ft-lb, while 47 J corresponds to 6 kgf·m/cm², commonly used indices in the United States and Russia, respectively. But practically, they are from about the same neighborhood in the transition range for RPV materials. In order to provide data according to the relevant national practice, the shifts of transition temperature due to irradiation or annealing will be reported as measured at 47 J for Russian welds, and at 41 J for the U.S. steels, regardless of the test laboratory.

Irradiation Conditions

The ORNL experiments with the VVER welds included irradiation to about 1×10^{19} n/cm² (>1 MeV) in the University of Michigan Ford Reactor, while the RRC-KI experiments with the U.S. materials included irradiations from about 2 to 18×10^{19} n/cm² (>1 MeV) in Unit 5 of the Novo-Voronezh Nuclear Power Plant (NVNPP-5), a VVER-1000 type reactor. In both cases, irradiations were conducted at ~290°C. Additionally, Charpy and tensile specimens of Weld 502 were irradiated by ORNL at 275°C, a temperature which is closer to the operating temperature (270°C) of VVER-440 reactor vessels.

Besides the differences between U.S. and Russian practices in indexing absorbed energy for determination of the transition temperature, there is also a difference in the measures of neutron fluence used. In the United States, neutron fluence for RPVs is measured for neutrons having energies greater than 1 MeV, while in Russia it is for neutrons having energies greater than 0.5 MeV. Thus, the values of neutron fluence for U.S. and Russian materials in this paper will be reported for neutrons with energies greater than 1.0 and 0.5 MeV, respectively, regardless of the source of irradiation. However, because the dosimetric procedures for each source were performed in the standard manner, estimates for adjustment must be determined. To determine the neutron irradiation exposure parameters, each country uses a neutron spectrum adjustment procedure that combines transport calculations of the neutron field and measurements using neutron monitors from irradiation capsules. The values of neutron fluence with >0.5 MeV for Russian welds irradiated by ORNL as well as the values of neutron fluence with >1 MeV for American steels irradiated by the RRC-KI given in this paper should be considered as the best currently available estimates. ORNL made this estimate by using a ratio, of flux of neutrons with >0.5 MeV to those with >1 MeV, equal to 1.76 for the capsule irradiated in the Ford Reactor, while RRC-KI used a ratio of 2.11 for their capsules irradiated in NVNPP-5.

Table 1 - Chemical Composition of Materials Used in the Annealing and Reirradiation Studies

Material	Composition (wt %)								
	C	Mn	P	S	Cr	Ni	Mo	Cu	V
HSST Plate 02	0.23	1.55	0.009	0.014	0.04	0.67	0.53	0.14	0.003
HSSI Weld 73W	0.098	1.56	0.005	0.005	0.25	0.60	0.58	0.31	0.003
Weld 502 (VVER-440)	0.03	1.16	0.030	0.013	1.60	0.12	0.48	0.15	0.24
Weld 260 (VVER-1000)	0.08	0.85	0.009	0.015	1.83	1.64	0.61	0.07	---

Table 2 - Mechanical Properties of Materials Studied in the Unirradiated Condition

Material	Transition temperature (°C)				USE (J)	Room temperature tensile strength (MPa)	
	at 41 J	at 47 J	at 0.9 mm	at 50%		Yield	Ultimate
Plate 02	-14	NA	-1	27	164	467	622
Weld 73W	-39	NA	-27	-15	135	494	604
Weld 502 (VVER-440)	NA	-3	2	20	130	437	546
Weld 260 (VVER-1000)	NA	-62	-51	-39	130	490	583

Table 3 - Fitting Coefficient (A) Compared with Chemistry Factor (CF) from RG 1.99 for U.S. Materials and Embrittlement Coefficient (A_F) from CS-86 for Russian Welds

Material	Composition (wt %)			(°C)		
	P	Cu	Ni	A	CF	A_F
Plate 02	0.009	0.14	0.67	65.6	56	NA
Weld 73W	0.005	0.31	0.60	86.7	110	NA
Weld 502 (VVER-440)	0.030	0.15	0.12	16.6	NA	32.4
Weld 260 (VVER-1000)	0.009	0.07	1.64	22.0	NA	20

Irradiation and Annealing Results

Results from the primary irradiation experiments with HSST Plate 02 and HSSI Weld 73W performed by ORNL were previously published [3,5,6]. Fracture toughness characterization of these steels has been also performed in these studies. Recently, cleavage fracture toughness data (K_{Jc}) for these steels were reevaluated using the master curve analysis procedure [7]. The procedure applies a statistical distribution function to characterize scatter of fracture toughness in the transition range and weakest-link theory to account for size effects, see Ref. [7] for details. It introduces the reference fracture toughness temperature, T_{100} , as a temperature at which the median fracture toughness equals $100 \text{ MPa}\cdot\text{m}^{1/2}$. The shift of T_{100} is used in the present work to characterize effects of irradiation and/or annealing on cleavage fracture toughness. In the present study, PCVN specimens of VVER-440 Weld 502 were tested in three-point bending by ORNL to provide fracture toughness characterization of this weld in the unirradiated, irradiated, and irradiated/annealed conditions.

Irradiation and annealing results are summarized in Fig. 2(a) through (d) for both the U.S. and Russian steels. The materials studied were irradiated to a wide range of neutron fluences. In general, the irradiation and annealing experiments conducted by RRC-KI and ORNL show reasonable agreement. Two different fitting functions were used to represent the embrittlement path (I) for the materials studied. For Plate 02 and Weld 73W it is

$$\Delta TT = A \cdot F^{(0.28 - 0.1 \log F)}, \quad (2)$$

where ΔTT is the shift of Charpy energy at 41 J or fracture toughness at $100 \text{ MPa}\cdot\text{m}^{1/2}$, F is neutron fluence in units of 10^{19} n/cm^2 ($>1 \text{ MeV}$); and A is a fitting parameter. This fitting function has the same form and the same constants in the exponent as the one in U.S. Nuclear Regulatory Commission (NRC) *Regulatory Guide 1.99* (RG 1.99), Revision 2 [8]. For VVER-440 and VVER-1000 welds it is

$$\Delta TT = A \cdot F^{1/3}, \quad (3)$$

where F is neutron fluence in units of 10^{18} n/cm^2 ($>0.5 \text{ MeV}$). This fitting function has the same form and the same constant in the exponent as the one in the Russian Standards (CS-86) [9].

The fitting parameters are compared with chemistry factors (CF) from RG 1.99 for Plate 02 and Weld 73W and embrittlement coefficients (A_F) from CS-86 for VVER-1000 Weld 260 in Table 3. Weld 502 represents the materials for the older generation of VVER-440 reactors which operate at 270°C and CS-86 provides the embrittlement coefficient calculation procedure for this irradiation temperature as well as for 250°C , but not for an irradiation temperature of 290°C . However, ORNL performed irradiation of this weld at 275°C in addition to irradiation at 290°C . This allowed for a possibility to

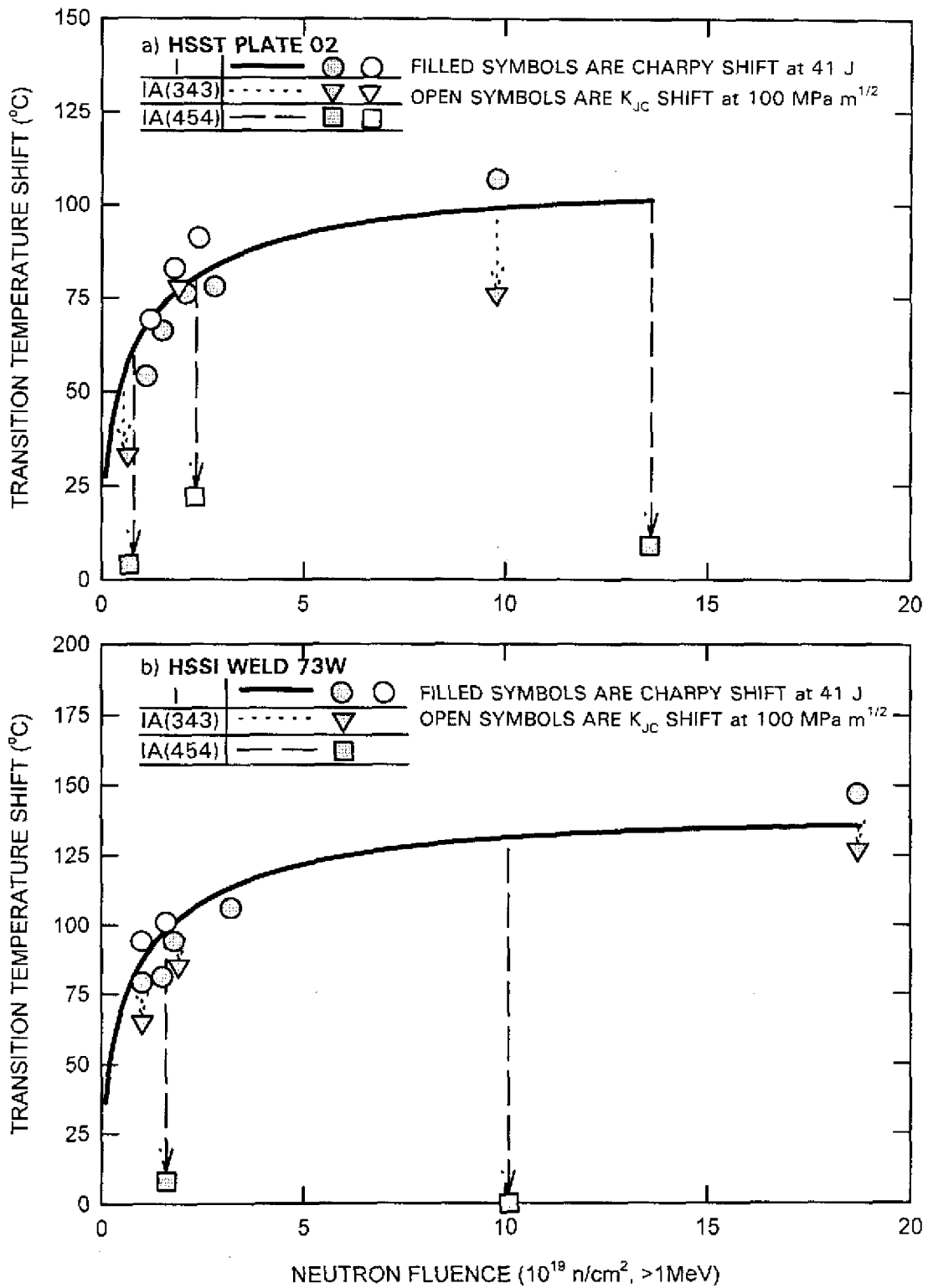


Fig. 2. Shift of the transition temperature due to irradiation and annealing.
 (a) HSST Plate 02, (b) HSSI Weld 73W.

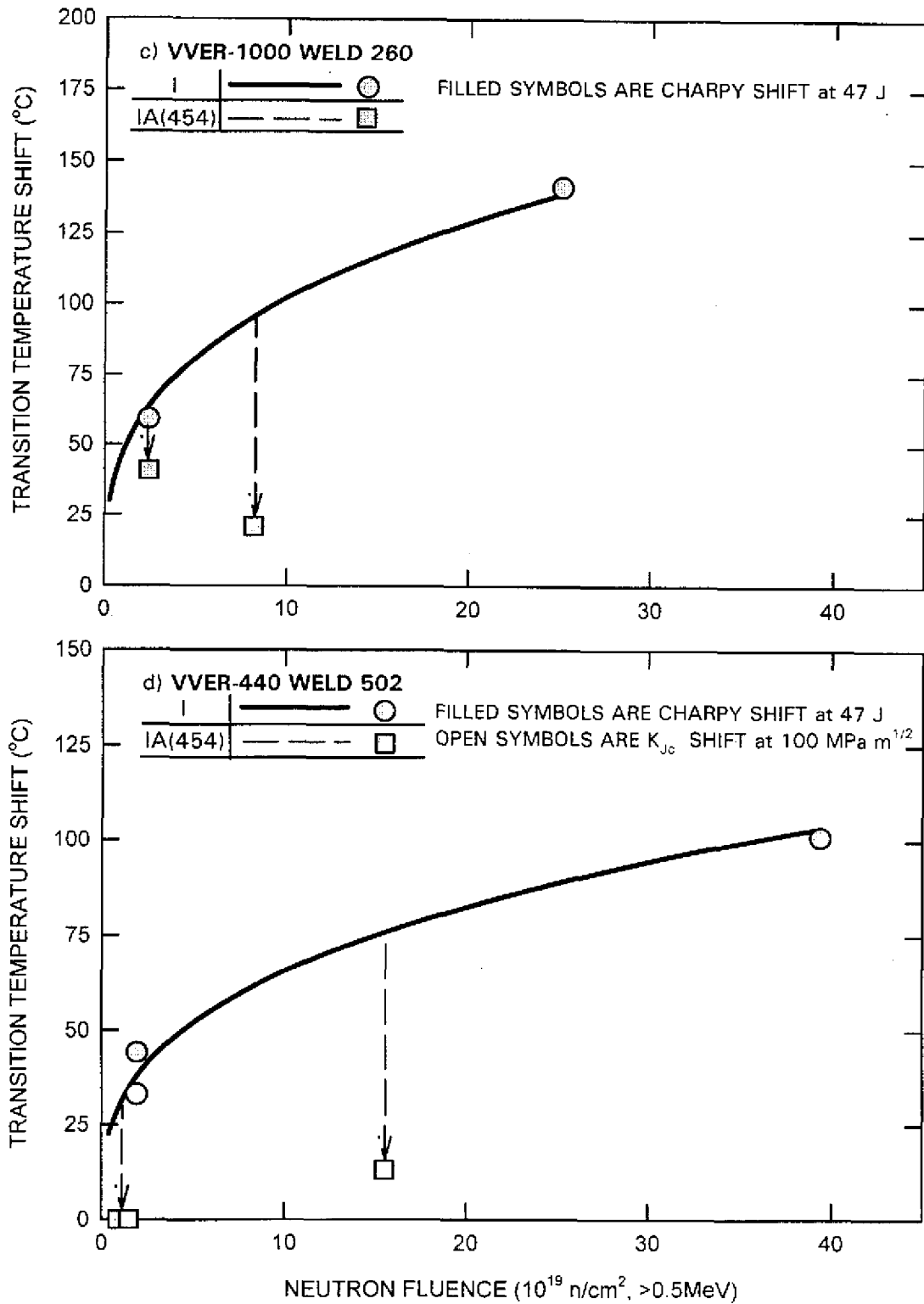


Fig. 2. Shift of the transition temperature due to irradiation and annealing.
(c) VVER-1000 Weld 260, (d) VVER-440 Weld 502.

compare A_F predicted from CS-86 to the one determined from the Charpy data irradiated at 275°C. These values of A_F for the Weld 502 are also presented in Table 3. Figure 3 shows the Charpy energy data for Weld 502 irradiated at two temperatures.

The data in Table 3 show that the CS-86 provides a very conservative value of A_F for the VVER-440 Weld 502 compared to the fitting coefficient from the present study. Contrary to that, however, the high-nickel VVER-1000 weld exhibited an embrittlement coefficient higher than that from the CS-86. That raises concerns regarding the ability of the current Russian practice to adequately predict the radiation embrittlement of high-nickel welds in VVER-1000 reactor vessels. For HSST Plate 02 and HSSI Weld 73W, the fitting coefficients are within 2 standard deviations of the mean CF parameters from RG 1.99.

A significant difference in the experiments is that those conducted by RRC-KI involved much higher neutron fluences than those conducted by ORNL. These results are extremely important for Plate 02 and Weld 73W in that they show neither material exhibits unexpected radiation sensitivity at very high fluences and, further, that both highly irradiated materials show nearly full recovery of Charpy toughness after annealing at 454°C.

ORNL also performed tensile tests of VVER-440 and VVER-1000 welds in the irradiated and annealed conditions. Tensile properties of Plate 02 and Weld 73W in the irradiated condition have been previously reported [2,5]. The radiation-induced changes in yield strength for the materials studied were compared to the shifts of Charpy transition temperature. The data are in very good agreement with the large Power Reactor Embrittlement Data Base (PR-EDB) maintained at ORNL for the NRC (Fig. 4).

Two annealing temperatures were involved in the present study. Annealing temperature 343°C was applied to Plate 02 and Weld 73W only, while all four materials were annealed at 454°C. In general, annealing at 454°C provides more complete recovery of the transition temperature than that at 343°C. The residual shift of transition temperature (the difference between transition temperatures of a material after annealing and in the unirradiated condition) increases with increase in neutron fluence for the 343°C annealing temperature.

It was noticed that fracture toughness shifts [Fig. 2(a) and (b)] are slightly higher than the Charpy shifts for Plate 02 and Weld 73W. Additionally, recovery of the fracture toughness shift seems to be less than the Charpy shift recovery at both annealing temperatures for Plate 02. The difference is relatively small. Contrarily, fracture toughness shifts for VVER-440 Weld 502 were slightly less than Charpy shifts and full recovery after annealing at 454°C was observed for both transition temperatures. Observed differences between Charpy and fracture toughness shifts are within the scatter reported in Refs. [10,11].

According to the present data, the annealing temperature of 454°C is high enough to provide nearly complete recoveries of the transition temperatures even after irradiation to very high fluences. Moreover, RRC-KI has reirradiated some Charpy specimens of Plate 02 and Weld 73W which were annealed at 454°C and then performed additional annealing of reirradiated specimens at 454°C (see the following section). Altogether, the data indicate very positive recovery of transition temperature after annealing at 454°C and that residual shift does not accumulate with neutron fluence or with cycling of irradiation

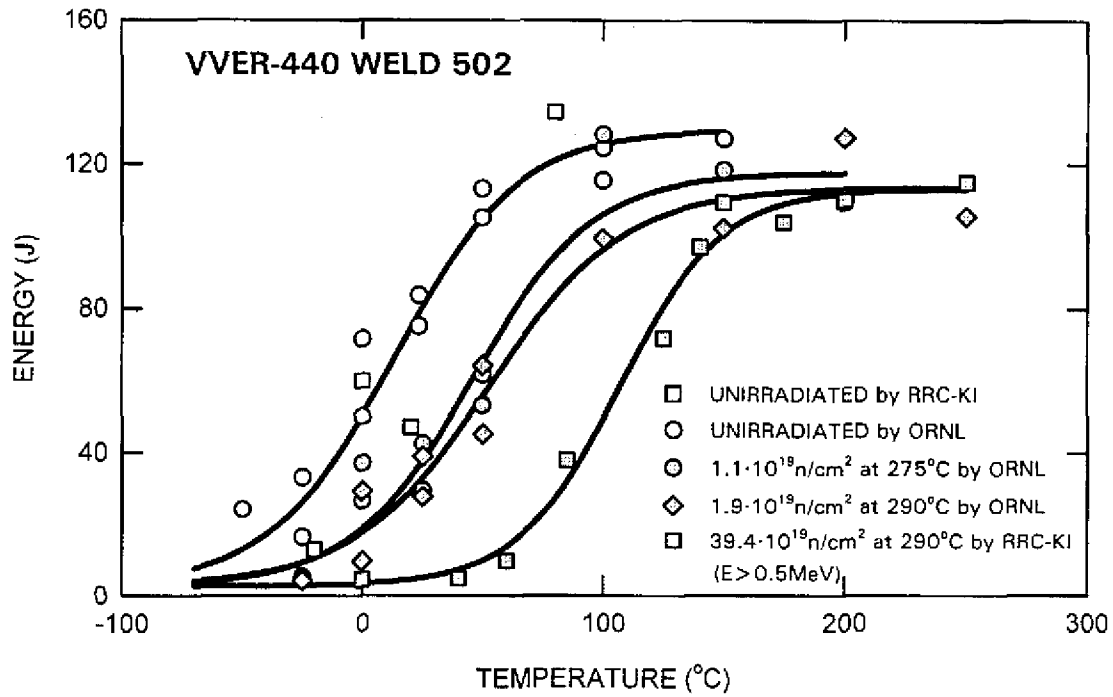


Fig. 3. ORNL and RRC-KI Charpy data of VVER-440 Weld 502 in the unirradiated condition and after irradiation at 275 and 290°C.

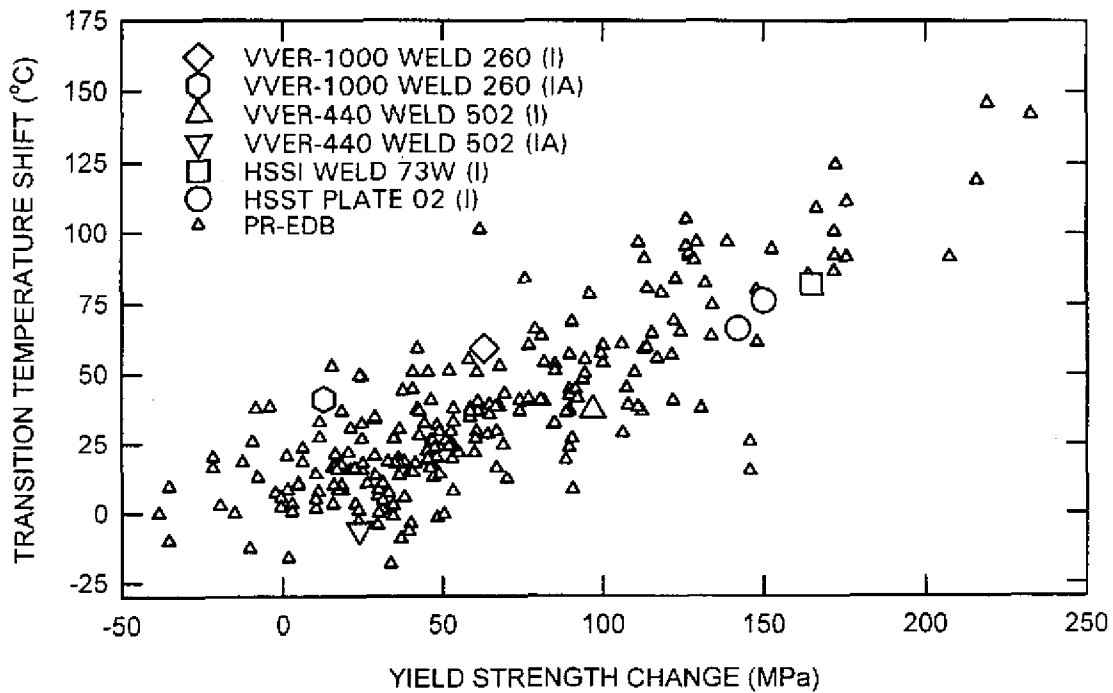


Fig. 4. Transition temperature shift versus change in yield strength for the materials studied by ORNL in comparison with PR-EDB data.

and annealing at 454°C. This observation is in a good agreement with earlier work on VVER-440 materials [12].

Recovery of the Charpy energy transition temperature was the primary objective in the present study. However, it was observed that other Charpy specimen measurements react differently on annealing at 454°C. Figures 5 through 8 present absorbed energy, lateral expansion, and percent shear data of specimens tested after annealing at 454°C for Plate 02 and Welds 73W, 502, and 260, respectively. Specimens were irradiated to different fluences, including those annealed after irradiation-annealing-reirradiation cycle. The data are plotted against the unirradiated curves to observe the degree of recovery after annealing. For each material in general, the annealed data tend to group into one data set regardless of irradiation and annealing history. Among the three Charpy toughness parameters, absorbed energy appears to recover the most, lateral expansion the least, and percent shear is somewhat in between in the transition region.

On the upper-shelf, energy values of all four materials rise above the unirradiated level after annealing, similarly to results reported in Refs. [10,13], while none of the materials exhibited complete recovery of lateral expansion. A limited number of unirradiated specimens of Plate 02 and Weld 502 were annealed at 454°C. Plate 02 specimens were tested on the upper-shelf only, while Weld 502 specimens were tested over a wide temperature range. For unirradiated/annealed specimens of Plate 02, the upper-shelf values of absorbed energy and lateral expansion are within scatter of the unirradiated values. Unirradiated/annealed specimens of Weld 502 showed slightly different behavior; the upper-shelf energy increased to the same level shown by the irradiated/annealed specimens, while lateral expansion did not change as a result of annealing.

The need for direct fracture toughness measurements is critical, especially in light of the present results where different Charpy parameters showed mixed response to annealing.

Reirradiation Results

RRC-KI also performed reirradiation of Charpy specimens of Plate 02 and Weld 73W which had been irradiated and annealed at 454°C. Moreover, some of the reirradiated specimens were subjected to additional post-reirradiation annealing at 454°C. The data are summarized in Fig. 9. The solid line represents the embrittlement path (I) as shown earlier in Fig. 2. Specimens of both materials were initially irradiated to about 2.1×10^{19} n/cm² (>1 MeV), then annealed at 454°C for 72 h (dashed line in Fig. 9). After annealing (IA), specimens were loaded back into NVNPP-5 reactor for reirradiation. The total neutron fluence was about 4.7×10^{19} n/cm² (>1 MeV) for Plate 02 and about 5.0×10^{19} n/cm² for Weld 73W. The measured shifts of transition temperature following reirradiation (IAR) were compared to those predicted by NRC *Regulatory Guide 1.162* (RG 1.162) [14]. According to this guide, the reembrittlement path can be predicted by the scheme of lateral shift (dotted lines in Fig. 9). The present data show that experimentally measured shifts of transition temperature after reirradiation are well below primary embrittlement path proving benefits of annealing for RPV steels. Even the lateral shift approach provides conservative predictions of the transition temperature shifts after

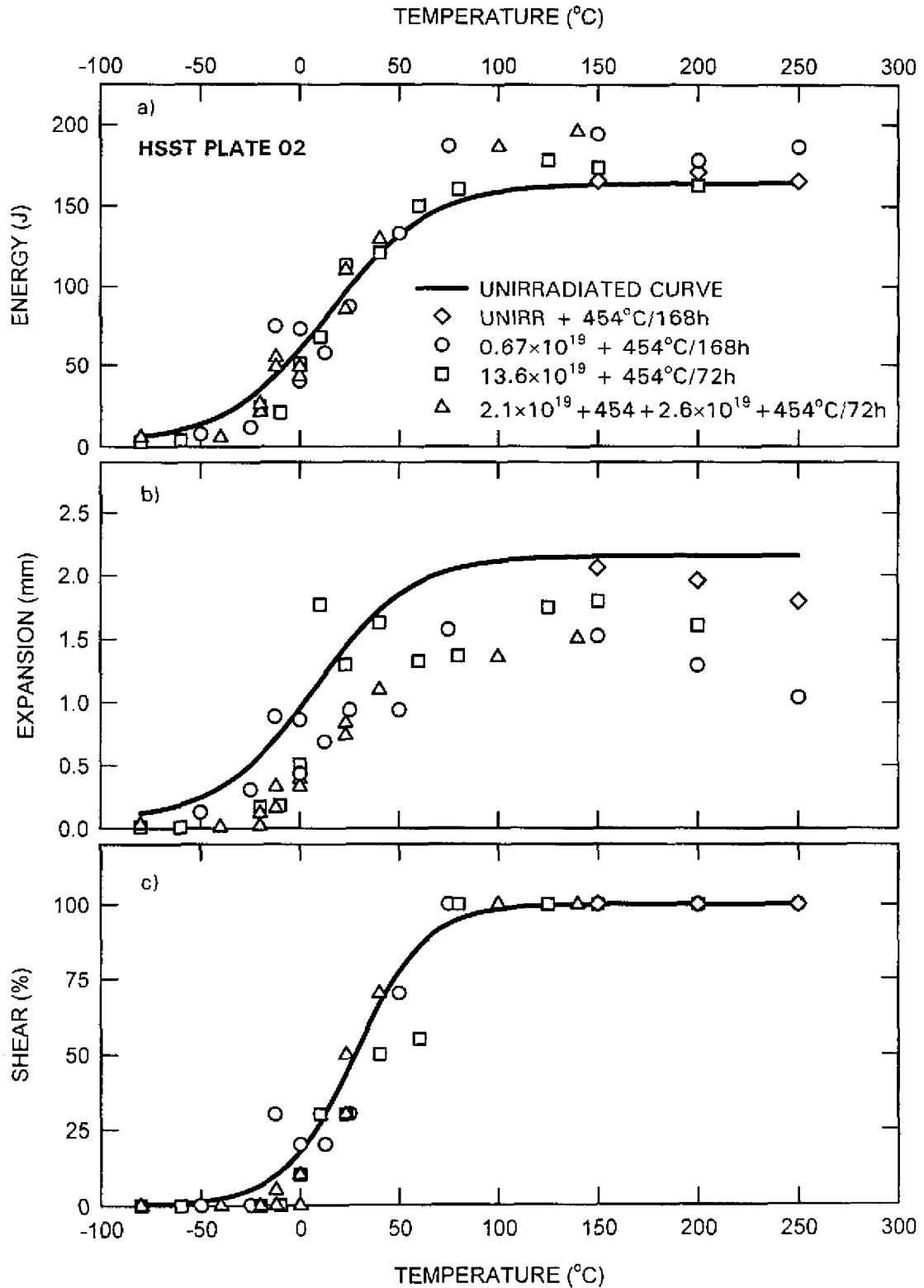


Fig. 5. Charpy data of Plate 02 in the annealed condition compared with the combined unirradiated curves. The legends in (a) apply to all three plots.

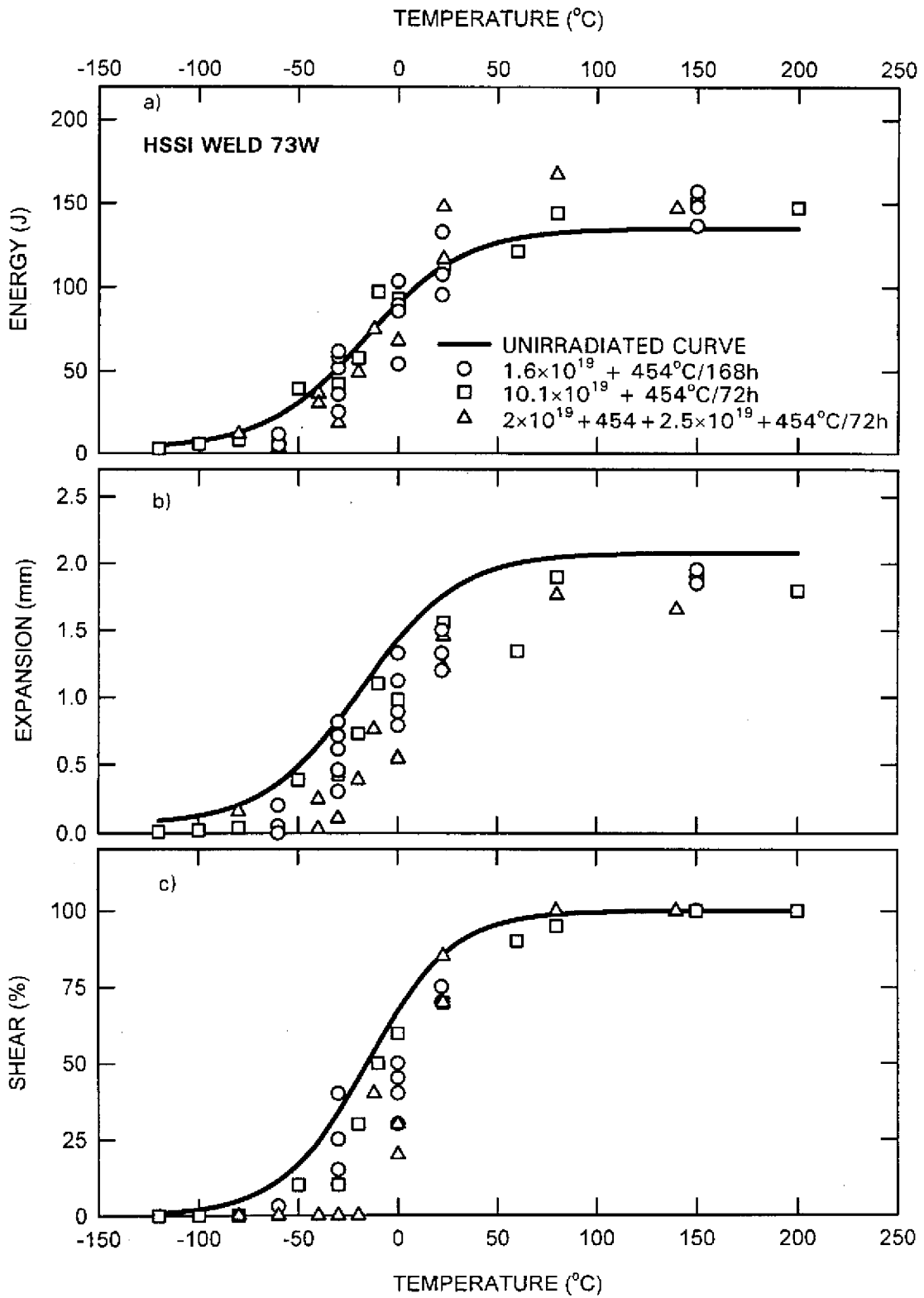


Fig. 6. Charpy data of Weld 73W in the annealed conditions compared with the combined unirradiated curves. The legends in (a) apply to all three plots.

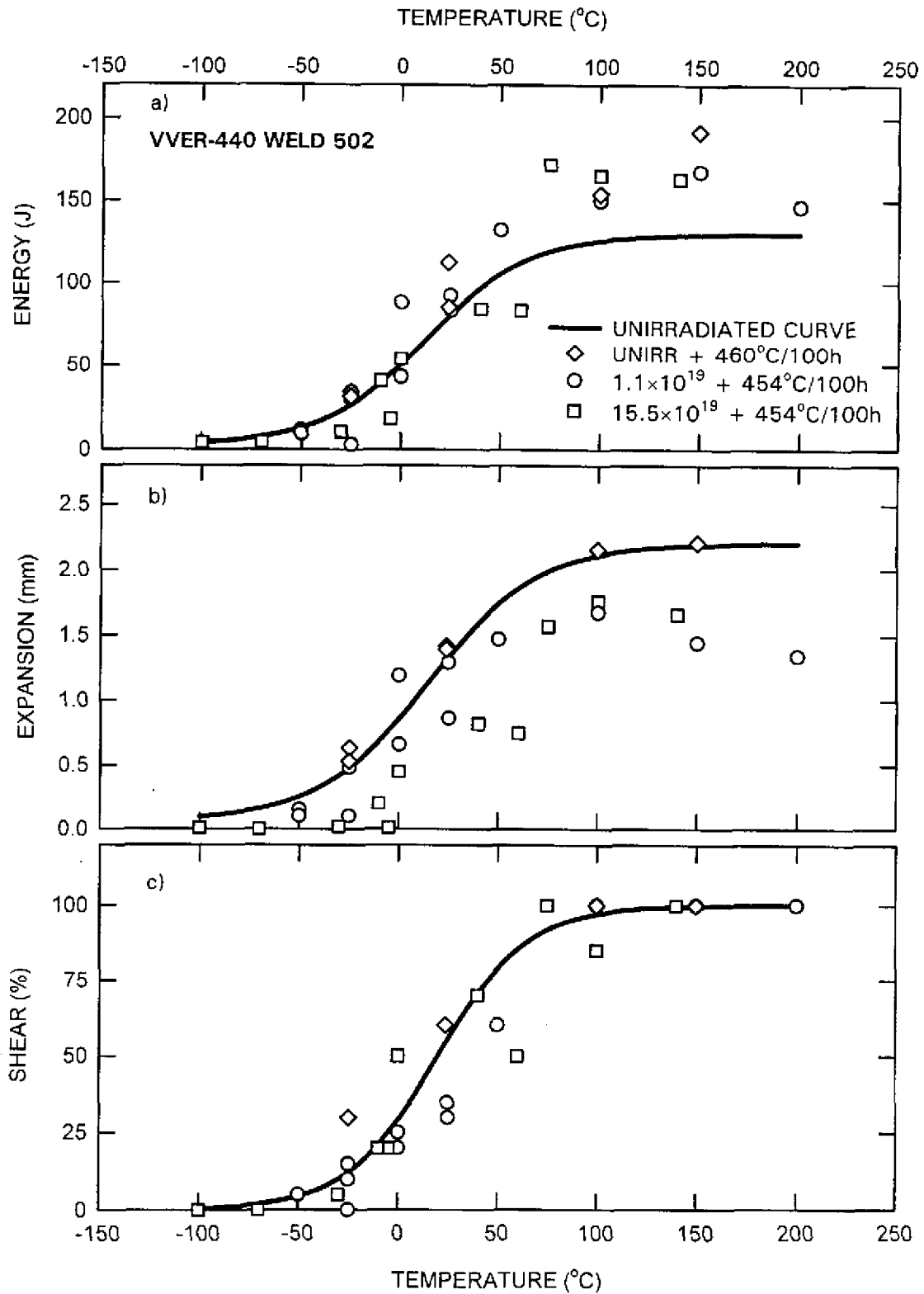


Fig. 7. Charpy data of Weld 502 in the annealed conditions compared with the combined unirradiated curves. The legends in (a) apply to all three plots.

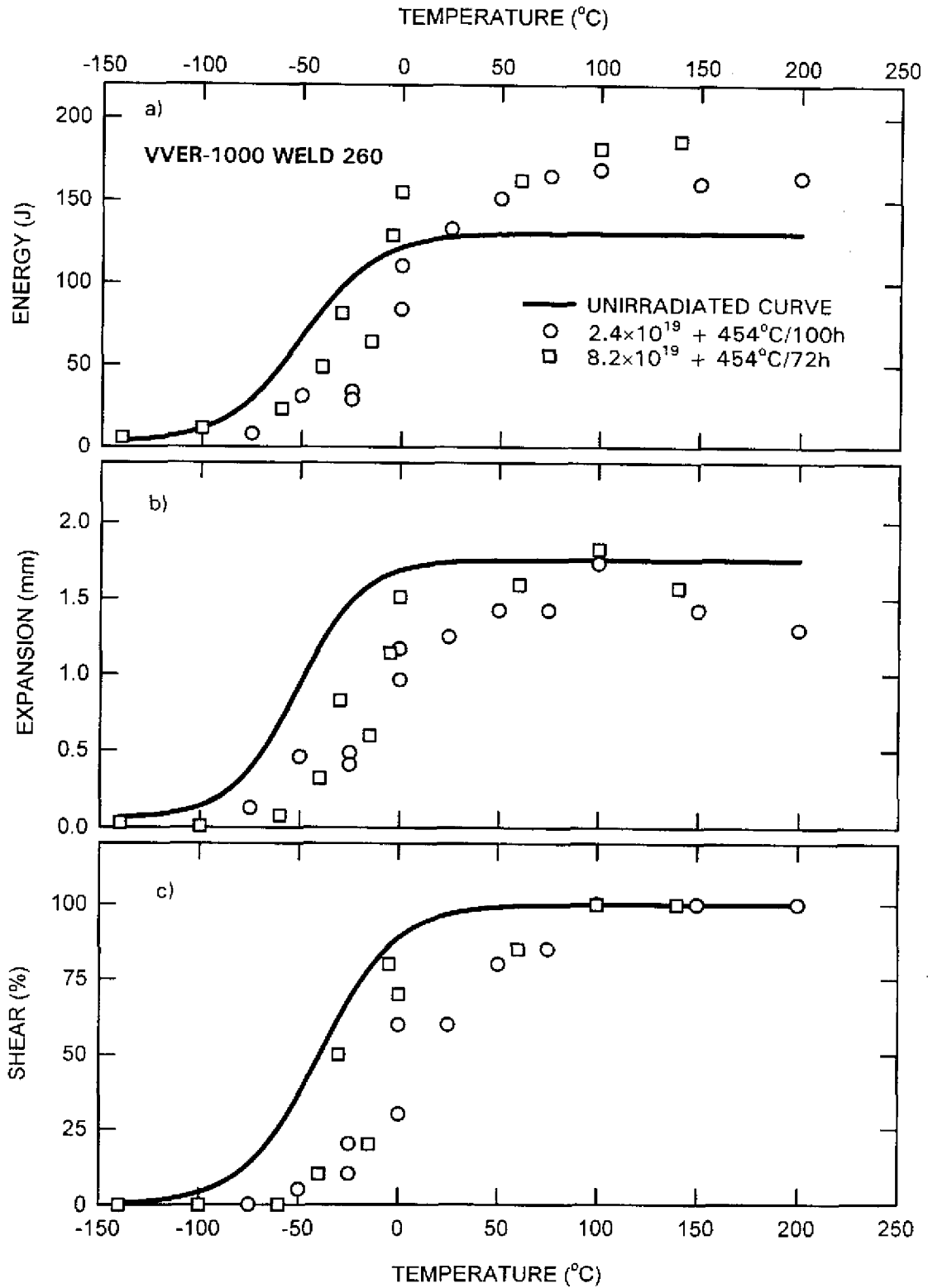


Fig. 8. Charpy data of Weld 260 in the annealed conditions compare with the combined unirradiated curves. The legends in (a) apply to all three plots.

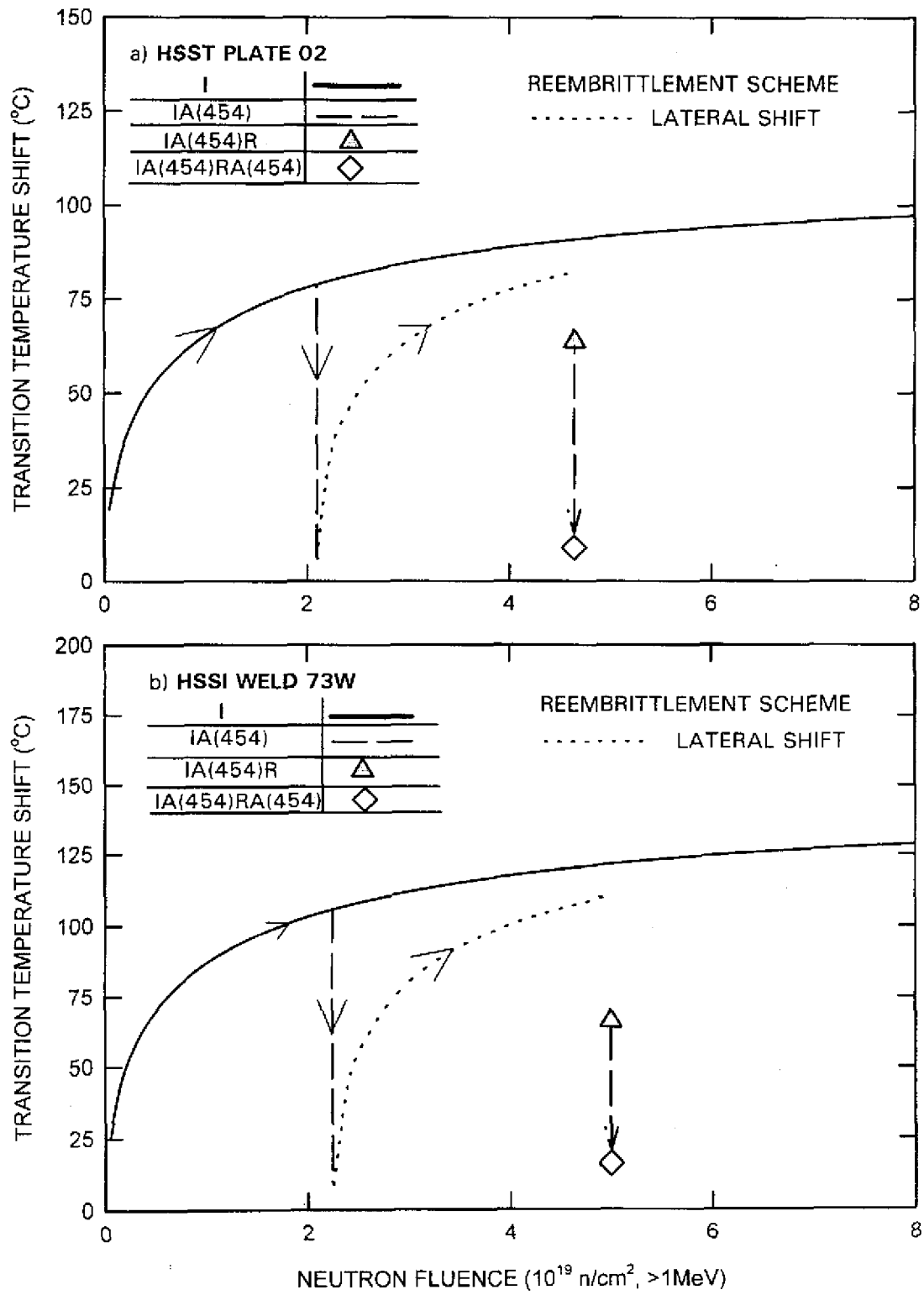


Fig. 9. Reirradiation data of (a) HSST Plate 02 and (b) HSSI Weld 73W.

reirradiation. Reirradiation data for Weld 73W are in agreement with a prediction made in Ref. [15] based on a small-angle neutron scattering (SANS) study of this weld in the irradiated and irradiated/annealed conditions. SANS measurements showed a large number of very fine copper-rich precipitates in this weld. A simple correlation between Charpy shifts and square root of volume fraction of these precipitates was shown in that study. Annealing at 454°C results in a significant drop of number density and volume fraction of precipitates, which corresponds well with recovery of Charpy transition temperature. However, it was found that, in addition to a decrease in number density and volume fraction, annealing results in a radius increase of these precipitates. Thus, annealing coarsens the copper-rich precipitates. As a result of these observations, it was concluded that the irradiated/annealed weld had less copper in solution than before irradiation; therefore, the reembrittlement rate of this weld should be less than it was in the primary irradiation cycle. From a radiation sensitivity point of view, the irradiated/annealed weld is microstructurally different than it was before irradiation. The present results support these conclusions. Annealing of the reirradiated materials showed the same degree of transition temperature recoveries as those achieved for the first annealings [16].

Observations and Conclusions

1. Two U.S. steels, one plate of A 533 grade B class 1 and one submerged-arc weld (with Linde 0124 flux), and two Russian welds, one representative of older VVER-440 RPVs and the other representative of older VVER-1000 RPVs, were irradiated at a wide range of neutron fluences. All four materials were annealed following irradiation, while the U.S. steels were also reirradiated and annealed following reirradiation. The experiments with the U.S. steels were conducted in Russia and those for the Russian steels in the United States.

2. The ORNL and RRC-KI experiments showed generally good agreement for both the Russian and U.S. steels.

3. While transition temperature recovery following annealing of the Charpy 41-J transition temperature was substantial in all cases, significantly less recovery of the Charpy lateral expansion and percent shear fracture were observed. On the upper shelf, energy values of all four materials recovered to values above those of the unirradiated condition, while none of the materials exhibited complete recovery of lateral expansion.

4. Neither HSST Plate nor HSSI Weld 73W exhibited unexpected radiation sensitivity at very high neutron fluences; i.e., greater than 1×10^{20} n/cm² (>1 MeV); furthermore, both highly irradiated materials exhibit nearly full recovery of CVN transition temperature after thermal annealing at 454°C.

5. The reirradiation embrittlement rates for both HSST Plate 02 and HSSI Weld 73W are conservative relative to the "lateral shift" method for reembrittlement prediction.

Acknowledgments

This research is sponsored by the Division of Engineering Technology, Office of Nuclear Regulatory Research, U.S. Nuclear Regulatory Commission, under Interagency Agreement DOE 1886-N695-3W with the U.S. Department of Energy under Contract DE-AC05-96OR22464 with Lockheed Martin Energy Research Corporation.

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