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EXPLORATORY STUDY OF IRRADIATION, ANNEALING, AND REIRRADIATION EFFECTS ON AMERICAN AND RUSSIAN REACTOR PRESSURE VESSEL STEELS

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DISCLAIMER

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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 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; so far, only Charpy impact and tensile specimens have been included. Oak Ridge National Laboratory (ORNL) conducted such a program (irradiation and annealing) 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/cm^2 (>1 MeV), while the RRC-KI experiments with the U.S. materials included irradiations from about 2 to 18×10^{19} n/cm^2 (>1 MeV). In both cases, irradiations were conducted at ~290°C and annealing treatments were conducted at ~454°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. Although 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, the desired material property is fracture toughness and uncertainties exist because of the paucity of fracture toughness data upon both annealing and reirradiation.

I. INTRODUCTION

One of the options to mitigate the effects of irradiation on reactor pressure vessels 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; so far, only

Charpy impact 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, 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.

II. MATERIALS

Three welds and one base metal were characterized in the present study. The chemical compositions of the materials are listed in Table 1. 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. 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 content (<0.20%), and high phosphorus content (~0.030%), 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 (<0.10%) and phosphorus (<0.012%) contents. The 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.

III. 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 impact testing machine of the Amsler firm. An International Standards Organization-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 long were tested, while compact specimens were used for fracture toughness characterization of irradiated and annealed material.

IV. ANALYSIS OF CHARPY DATA

Charpy absorbed energy data of the four materials studied in the unirradiated condition are presented in Fig. 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 instrumented strikers of 2- and 8-mm radii (including one with RRC-KI) and summarized the comparison results in Ref. 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 and 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 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 United States, while it is measured at 47 J in Russia.

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	N/A	-1	27	164	467	622
Weld 73W	-39	N/A	-27	-15	135	494	604
Weld 502	N/A	-3	2	20	130	437	546
Weld 260	N/A	-62	-51	-39	130	490	583

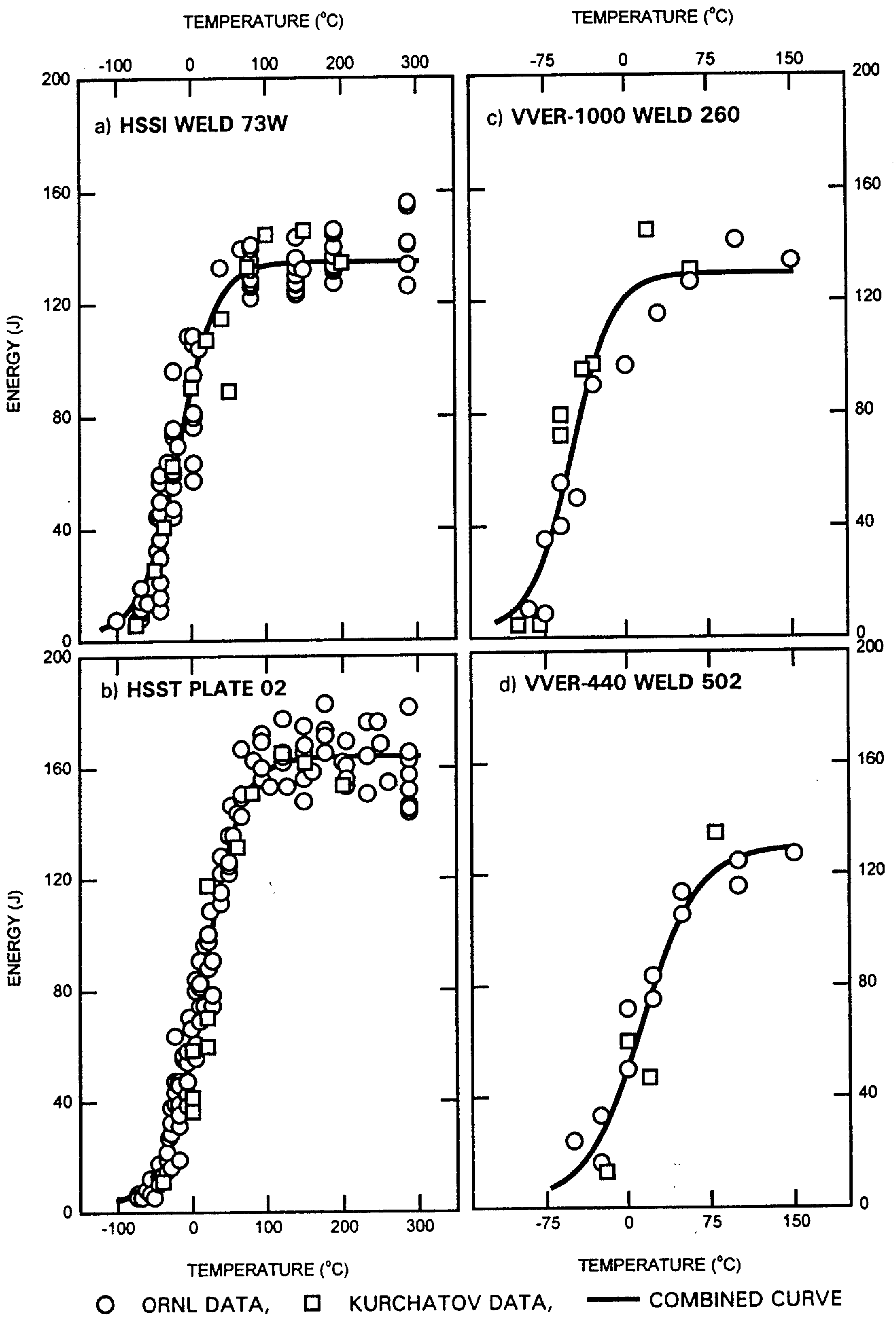

Fig. 1. Charpy data of materials studied in the unirradiated condition tested by ORNL and RRC-KI. Solid line is the best fit to combined data using a hyperbolic tangent function.

Indexing at 41 or 47 J reflects a difference of a historical matter 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. 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. The mechanical properties of materials studied in the unirradiated condition are presented in Table 2.

V. 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 the 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 measured 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 side 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.

VI. IRRADIATION AND ANNEALING RESULTS

Results from the primary irradiation experiments with HSST Plate 02 and HSSI weld 73W performed by ORNL were published previously.[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 by 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.[8] It introduces the reference fracture toughness temperature, T_{100}, as a temperature at which the median fracture toughness equals 100 MPa·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.

Irradiation and annealing results are summarized in Fig. 2 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 for the materials studied. For Plate 02 and weld 73W:

$$\Delta TT = A \cdot F^{(0.28 - 0.01 \log F)}, \qquad (2)$$

where ΔTT is the shift of Charpy energy at 41J or fracture toughness at 100 MPa·m$^{1/2}$; F is neutron fluence in units of $10^{19} \times$ n/cm² (>1 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 the U.S. Nuclear Regulatory Commission *Regulatory Guide 1.99* (RG 1.99), Rev. 2 (Ref. 9). For VVER-440 and VVER-1000 welds:

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

where F is neutron fluence in units of $10^{18} \times$ n/cm² (>0.5 MeV). This fitting function has the same form and the same constant in the exponent as the one in the Russian Calculation Standards (CS-86) (Ref. 10).

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. The 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

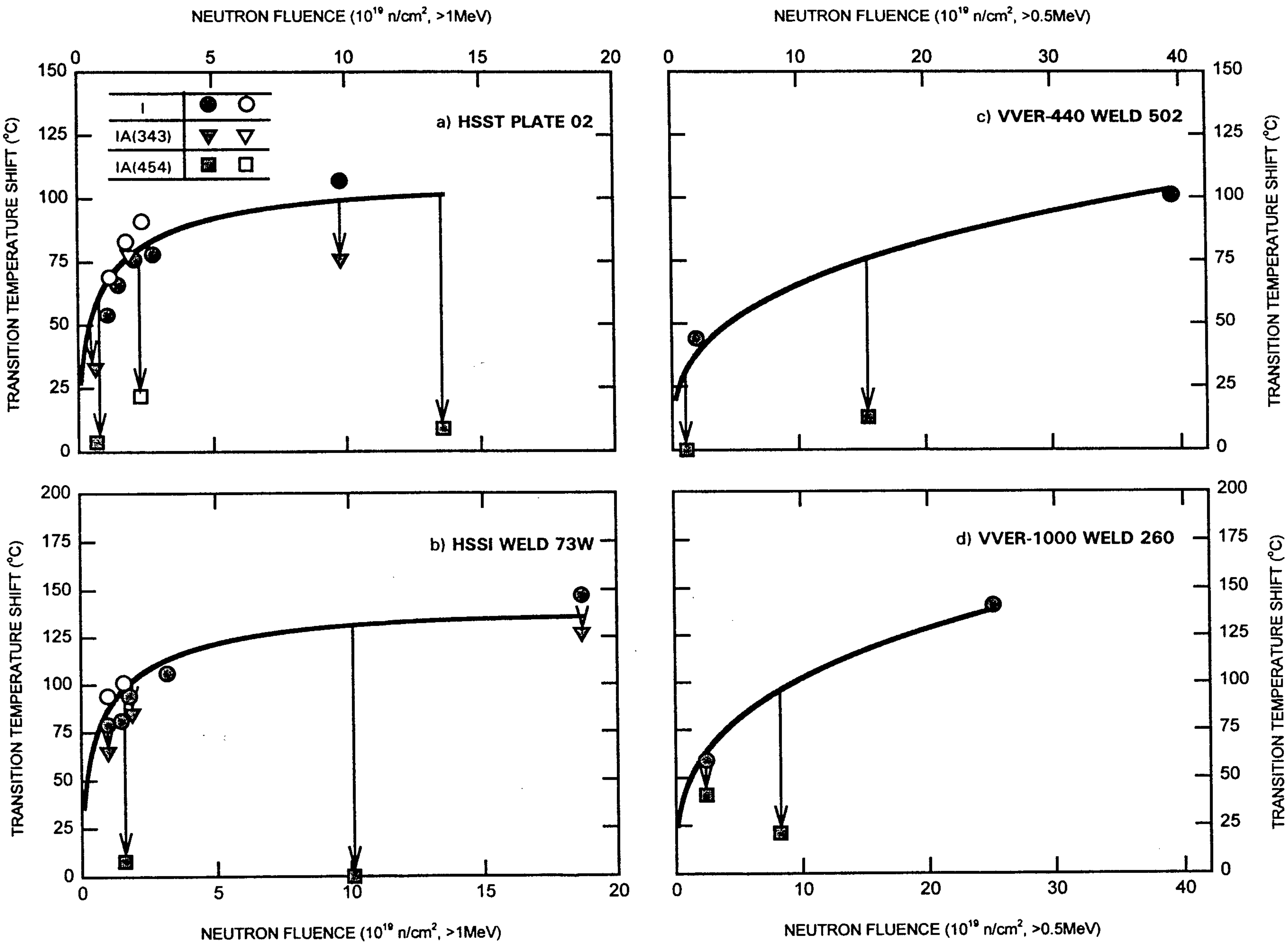

Fig. 2. Shift of the transition temperature due to irradiation and annealing. The legends in (a) apply to all four plots. Filled symbols are Charpy shift and open symbols are K_{Jc} shift.

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 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.

Table 3. Fitting coefficient, A, compared with chemistry factor (CF) from *Regulatory Guide 1.99* for U.S. materials and embrittlement coefficient, A_F, from CS-86 for Russian welds

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

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 reported previously.[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.

Two annealing temperatures were involved in the present study. An annealing temperature of 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) tends to increase with increase in neutron fluence for the 343°C annealing temperature.

It was noticed that fracture toughness shifts [see 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, but there are some other data (see Ref. 11, for example) which have shown a similar relationship between recovery of fracture toughness and Charpy shifts. However, the available data are very sparse for such comparisons.

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 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. For example, Figs. 3 and 4 present absorbed energy, lateral expansion, and percent shear data of specimens tested after annealing at 454°C for welds 73W 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

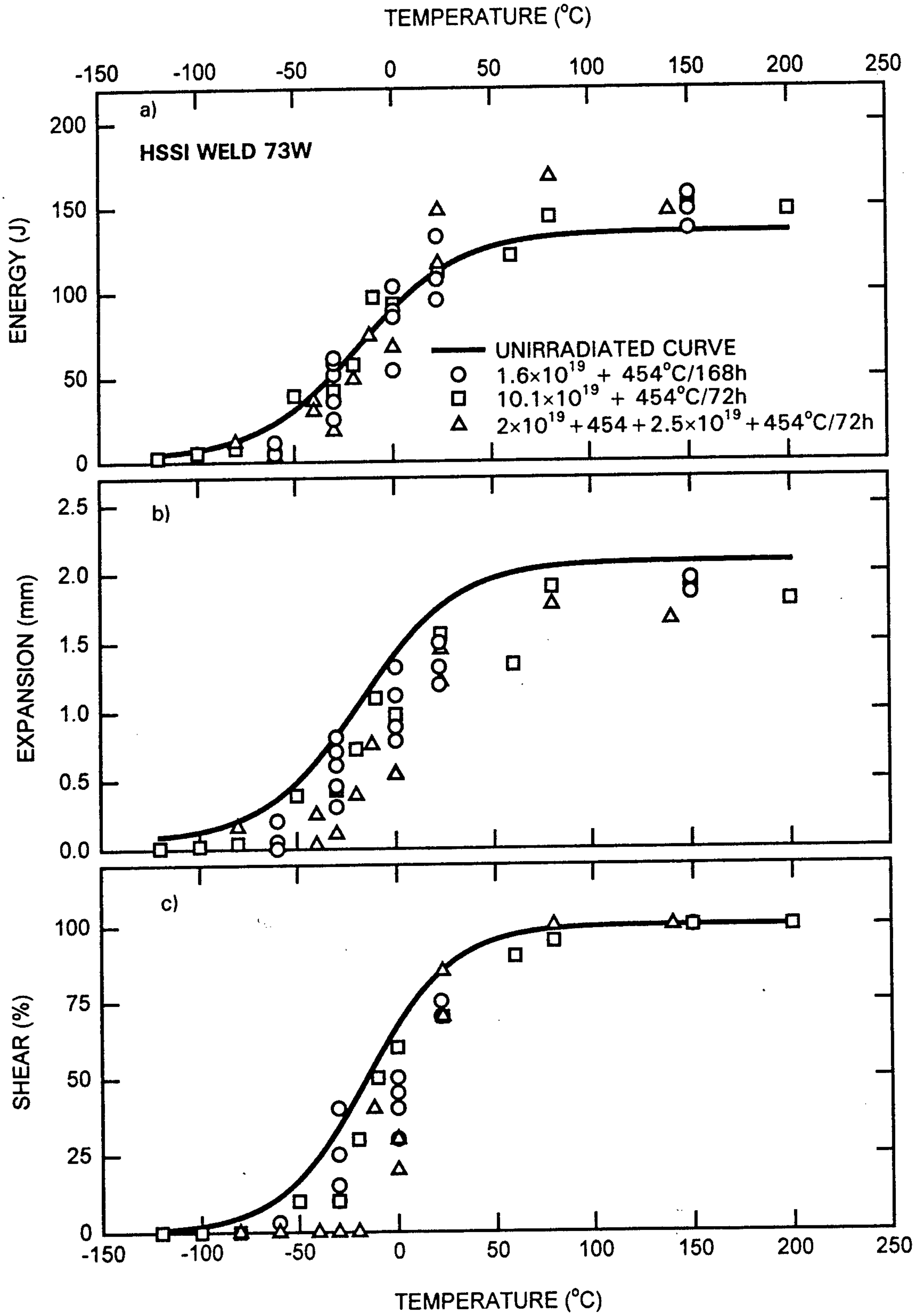

Fig. 3. Charpy data of Weld 73W in the annealed conditions compared with the combined unirradiated curves. The legends in (a) apply to all three plots.

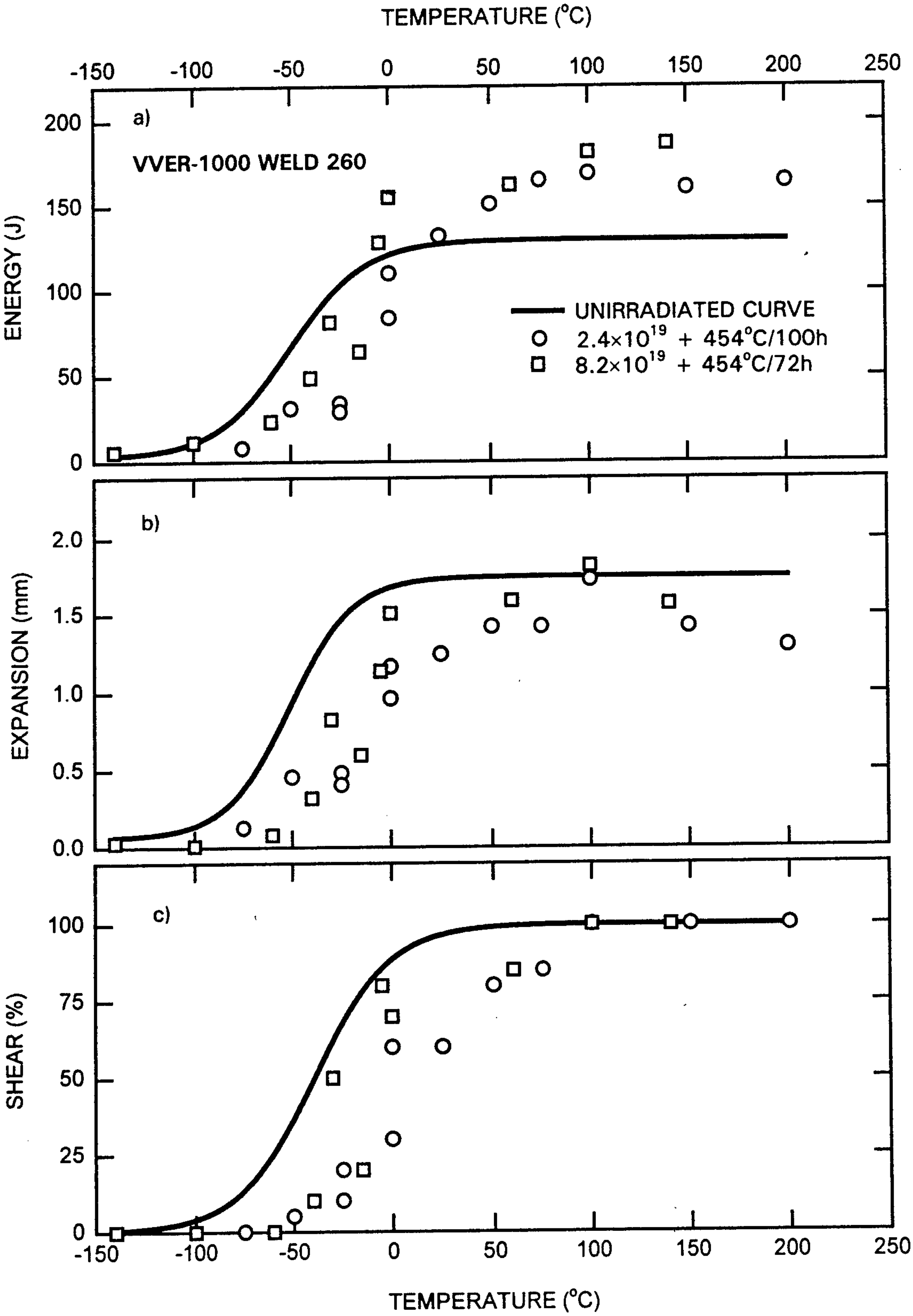

Fig. 4. Charpy data of Weld 260 in the annealed conditions compare with the combined unirradiated curves. The legends in (a) apply to all three plots.

transition region. Such behavior was observed for all four materials.

On the upper-shelf, energy values of all four materials rise above the unirradiated level after annealing, similarly to results reported in Refs. 11 and 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.

VII. REIRRADIATION RESULTS

RRC-KI also performed a 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. 5. 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/cm^2 (>1 MeV), then annealed at 454°C for 72 h (dashed line in Fig. 5). After annealing, specimens were loaded back into NVNPP-5 reactor for reirradiation. The total neutron fluence was about 4.7×10^{19} n/cm^2 (>1 MeV) for Plate 02 and about 5.0×10^{19} n/cm^2 for weld 73W. The measured shifts of transition temperature following reirradiation (IAR) were compared to those predicted by the NRC *Regulatory Guide 1.162* (RG 1.162).[14] According to RG 1.162, the reembrittlement path can be predicted by the scheme of lateral shift (dotted lines in Fig. 5). As an alternative, the vertical shift method for prediction of reembrittlement rate is also presented in Fig. 5 by dashed-dotted lines. 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 scheme provides conservative predictions of the transition temperature shifts after reirradiation, while prediction by the vertical shift scheme significantly underestimates the experimental result. Annealing of the reirradiated materials showed the same degree of transition temperature recoveries as those achieved for the first annealings.

VIII. 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 to 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 have shown generally good agreement for both the Russian and U.S. steels.

3. While transition temperature recoveries following annealing of the Charpy energy transition temperatures were substantial in all cases, significantly less recoveries 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 02 nor HSSI weld 73W exhibited unexpected radiation sensitivity at very high neutron fluences, i.e., greater than 1×10^{20} n/cm^2 (>1 MeV); furthermore, both highly irradiated materials exhibit nearly full recovery of Charpy V-notch 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.

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

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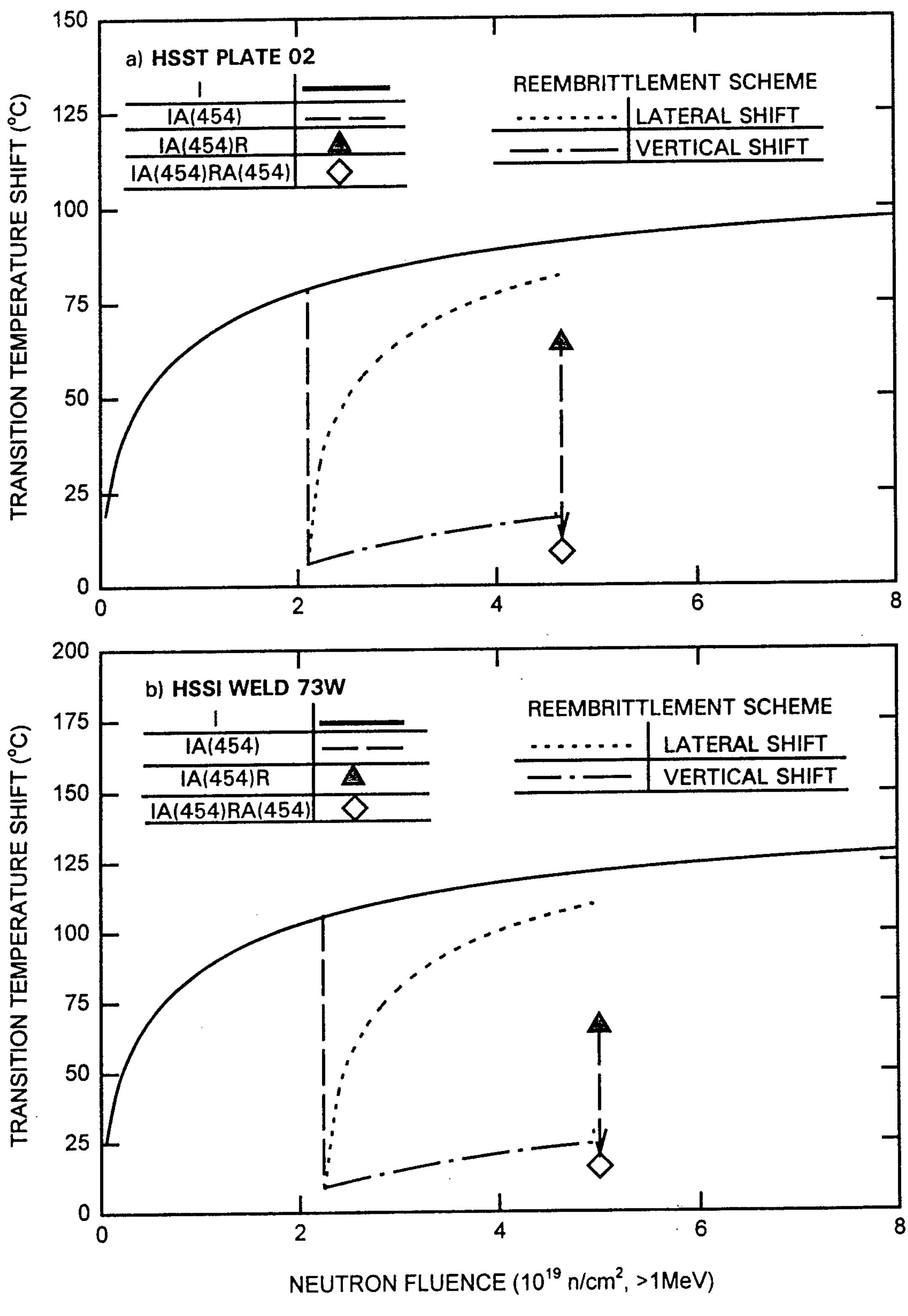

Fig. 5. Reirradiation data of (a) HSST Plate 02 and (b) HSSI Weld 73W.

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