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EFFECTS OF THERMAL ANNEALING AND REIRRADIATION ON TOUGHNESS OF REACTOR PRESSURE VESSEL STEELS*

R. K. Nanstad, S. K. Iskander, and M. A. Sokolov
Oak Ridge National Laboratory
Oak Ridge, Tennessee

A. A. Chernobaeva, Y. A. Nikolaev, A. M. Kryukov, and Yu. N. Korolev
Russian Research Center-Kurchatov Institute
Moscow, Russia

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R. K. Nanstad, S. K. Iskander, and M. A. Sokolov
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Russian Research Center-Kurchatov Institute
Moscow, Russia

ABSTRACT

One of the options to mitigate the effects of irradiation on reactor pressure vessels (RPV) is to thermally anneal them to restore the toughness properties that have been degraded by neutron irradiation. This paper summarizes recent experimental results from work performed at the Oak Ridge National Laboratory (ORNL) to study the annealing response, or "recovery," of several irradiated RPV steels; it also includes recent results from both ORNL and the Russian Research Center-Kurchatov Institute (RRC-KI) on a cooperative program of irradiation, annealing and reirradiation of both U.S. and Russian RPV steels. The cooperative program was conducted under the auspices of Working Group 3, U.S./Russia Joint Coordinating Committee for Civilian Nuclear Reactor Safety (JCCCNRS). The materials investigated are an RPV plate and various submerged-arc welds, with tensile, Charpy impact toughness, and fracture toughness results variously determined. Experimental results are compared with applicable prediction guidelines, while observed differences in annealing responses and reirradiation rates are discussed.

DESCRIPTION OF MATERIALS AND IRRADIATION-ANNEALING CONDITIONS

The materials used for the studies described here are commonly used for the fabrication of RPVs in U.S. and Russian pressurized water reactors. Three submerged-arc welds (weld 73W and the two Midland Reactor welds) and one plate of A533 grade B class 1 steel (HSST Plate 02) are representative of materials used in U.S. reactors and have been used in a number of irradiation studies at ORNL [1,2,3]. The two Russian welds are representative of those used in VVER-440 and VVER-1000 type reactors and have been used in other studies by the RRC-KI. Table 1 provides the chemical compositions for all six of the materials. The U.S. steels are generally designated Mn-Mo-Ni steels, while the Russian welds are similar to 1.5 Cr-0.5 Mo steels. With regard to radiation sensitivity, the U.S. welds are characterized primarily by relatively high copper and low phosphorous contents, while the Russian welds have relatively low copper but high phosphorous (VVER-440) or high nickel (VVER-1000) contents. Because the VVER-1000 reactor is a newer design than the VVER-440, the welds were designed with reduced phosphorous and copper contents based on experience within Russia as well as that from Western countries. Unfortunately, the high nickel content in the VVER-1000 welds may result in relatively high radiation sensitivity in spite of the reduced phosphorous and copper contents. HSST Plate 02 is similar to the U.S. welds in most respects except for its relatively lower copper content. A summary of some basic mechanical properties for the U.S. materials is provided in Table 2 and shows that the materials exhibit a fairly wide range of Charpy V-notch (CVN) upper-shelf energies (USE) and transition temperatures. The U.S. materials have generally lower strength than the Russian welds. All the materials were postweld heat treated prior to testing.

In the ORNL experiments, the U.S. materials were all irradiated in material test reactors at a nominal irradiation temperature of 288°C (550°F) to neutron fluences ranging from about 0.5 to 2.0 × 10^{19} neutrons/cm^2 (>1 MeV); the Russian welds were irradiated under similar conditions except the VVER-440 weld was irradiated at a nominal irradiation temperature of about 270°C (518°F) because that is close to the temperature at which the VVER-440 RPV operates. For the RRC-KI experiments, two U.S. steels (HSST Plate 02 and HSSI Weld 73W) were irradiated in the Novovoronezh-5 commercial power plant at a nominal irradiation temperature of 290°C (554°F) to various fluences from about 2 to 18 × 10^{19} neutrons/cm^2 (>1 MeV). The preliminary fluences (>1 MeV) reported for the RRC-KI irradiations are estimated from the measured fluences (>0.5 MeV) using a factor provided by RRC-KI and based on previous experience. Thermal annealing experiments at ORNL have concentrated on two temperatures, 343 and 454°C (650 and 850°F). These two temperatures have been reported as the most likely "wet anneal" and "dry anneal" temperatures, respectively, for U.S. reactors [4]. Annealing times investigated by ORNL range from 24 to 336 hours, with 168 hours (one week) considered the "standard" annealing time for U.S. practice. The RRC-KI annealing experiments were conducted at 454°C (850°F) for 72 hours, except for the two reirradiation experiments wherein the specimens were annealed for 150 hours for the first anneal and 72 hours for the second anneal. The RRC-KI also conducted post-anneal reirradiation experiments for the two U.S. steels under the same conditions used for the initial irradiations.

EXPERIMENTAL RESULTS

HSSI Weld 73W was irradiated to a fluence of about 1.5 × 10^{19} neutrons/cm^2 (>1 MeV) and exhibited a CVN 41-J temperature shift of 82°C. Figure 1 provides a summary of the results of thermal annealing of Weld 73W at 454°C for various times. The percent recovery of the transition temperature (ΔTT) and USE (ΔUSE) are referenced to the shift or drop, respectively, due to irradiation, as shown below:

$$\% \text{ Recovery } TT_{ia} = \frac{TT_i - TT_a}{TT_i - TT_{unirr}} \cdot 100 \ ,$$

$$\% \text{ Recovery } USE_{ia} = \frac{USE_a - USE_i}{USE_{unirr} - USE_i} \cdot 100 \ ,$$

where TT is the transition temperature at the 41-J energy level, USE is the average upper-shelf energy, and the subscripts "i, a, ia, and unirr" refer to "irradiated, annealed, irradiated and annealed, and unirradiated," respectively. A 100% recovery indicates that the property after annealing has fully recovered its unirradiated value. Although the 41-J TT recovery does reach nearly 100%, it appears to occur at a slower rate than that of the CVN USE. However, whereas the TT recovery increase is insignificant after 96 hours of annealing, the USE recovery continues to increase even up to 336 hours and reaches recovery levels of about 200%. Figure 2 provides a summary of the annealing experiments conducted by ORNL on the four U.S. steels (more detailed discussions are available in references 5 and 6). The results of annealing at 343°C for 168 hours resulted in TT recoveries from 10 to 50%, while annealing at 454°C for 168 hours resulted in TT recoveries from about 75 to 100%. In every case at 454°C, the USE recovery exceeded 100% (over recovery). Models have been developed by Eason et al. [7] for prediction of transition temperature and upper-shelf energy recovery as a consequence of thermal annealing. Figure 3 shows a comparison of the ORNL annealing results with the predictions of the Eason et al. model. Most of the experimental results are within ±2 standard errors of the predicted results; for the few values which fall outside of the ±2 standard error bounds, the models provide conservative predictions. Regarding the over recovery of upper-shelf energy, Figure 4 shows a plot of published data (about 105 data) comparing the % recoveries of USE and 41-J TT. It is seen that much greater recoveries occur for the USE than for the 41-J TT; there are very few data for which the percent recovery of the USE is lower than that of the 41-J TT.

Regarding comparisons of annealing effects on fracture toughness with those on CVN toughness, Figure 5 provides one example of results for the low upper-shelf energy submerged-arc weld from the beltline region of the Midland Reactor. After annealing at 454°C for 168 hours, the residual transition temperature shifts of 24°C and 13°C for the CVN 41-J temperature and fracture toughness 100 MPa√m temperature represent recoveries of 77 and 86%, respectively. Additional comparisons are shown in Figure 6(a) for HSST Plate 02. For annealing at 343°C, the CVN 41-J TT recovered about 50%, whereas, at a similar fluence, the fracture toughness TT showed no recovery. For annealing at 454°C, both CVN and fracture toughness TTs recovered substantially, although that for fracture toughness was somewhat less. There are very sparse data available for such comparisons.

The results of the Working Group 3, JCCCNRS cooperative experiments are shown in Figures 6 and 7. As mentioned earlier, the neutron fluences (>1 MeV) for the RRC-KI experiments are estimated from the measured fluences (> 0.5 MeV) and are considered preliminary at this time. In general, the irradiation and annealing experiments conducted by RRC-KI with HSST Plate 02 and HSSI Weld 73W show reasonable agreement with those by ORNL. A significant difference in the experiments is that those conducted by RRC-KI involved very high neutron fluences. These results are extremely important 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 CVN toughness after annealing at 454°C. Moreover, the results of reirradiation experiments show that the so-called "lateral shift" method for prediction of reirradiation embrittlement is applicable for these steels. The dashed lines on Figures 6 (a) and (b) represent the predicted reirradiation response while the filled triangles (▲) show the measured reirradiation data. In Figure 7, the annealing results indicate a potentially increasing post-annealing residual transition temperature shift with increasing fluence for the VVER-440 weld, but the data are too sparse to draw a conclusion.

SUMMARY OF OBSERVATIONS

1. Annealing has resulted in various degrees of recovery of the transition temperature and upper-shelf energy that depend strongly upon the annealing temperature and to a somewhat lesser degree upon the annealing time.

2. Recovery at the lower annealing temperature investigated, 343°C (for 168 hours), resulted in recovering most of the upper-shelf energy, but the recovery of the 41-J transition temperature varied from insignificant to about 50%, depending upon the material.

3. The post-annealed recovery of the fracture toughness transition temperature appears to be somewhat less than that for the Charpy impact toughness, but the available data are too sparse to allow for a conclusion.

4. Irradiation and annealing experiments of both U.S. and Russian steels by ORNL and RRC-KI show generally good agreement.

5. For the RRC-KI experiments with HSST Plate 02 and HSSI Weld 73W in a Russian commercial nuclear plant, (a) neither material exhibits unexpected radiation sensitivity at very high fluences and, further, both highly irradiated materials show nearly full recovery of CVN toughness after annealing at 454°C; (b) the reirradiation embrittlement rates for both steels are conservative relative to the "lateral shift" method for reembrittlement prediction.

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Table 1. Average chemical composition of materials used in the annealing and reirradiation studies

Material	Composition (wt %)									
	C	Mn	P	S	Si	Cr	Ni	Mo	Cu	V
HSSI weld 73W	0.098	1.56	0.005	0.005	0.45	0.25	0.60	0.58	0.31	0.003
Midland beltline weld	0.084	1.61	0.017	0.007	0.62	0.10	0.57	0.41	0.21-0.34	0.004
Midland nozzle weld	0.088	1.57	0.015	0.010	0.56	0.11	0.58	0.39	0.37-0.46	0.008
HSST Plate 02	0.23	1.55	0.009	0.014	0.20	0.04	0.67	0.53	0.14	0.003
Weld 502-1 (VVER-440)	0.03	1.16	0.030	0.013	0.57	1.60	0.12	0.48	0.15	0.24
Weld 260-11 (VVER-1000)	0.08	0.85	0.009	0.015	0.30	1.83	1.59	0.61	0.07	---

Table 2. Mechanical properties of U.S. materials

Material	CVN impact USE (J)	CVN 41-J transition temperature (°C)	Room temperature tensile strength (MPa)	
			Yield	Ultimate
HSSI weld 73W (undersize)	118	–38	495	603
HSSI weld 73W (full size)	135	–40	495	603
Midland beltline weld	89	–9	407	586
Midland nozzle weld	88	–1	505	655
HSST Plate 02	164	–16	471	619

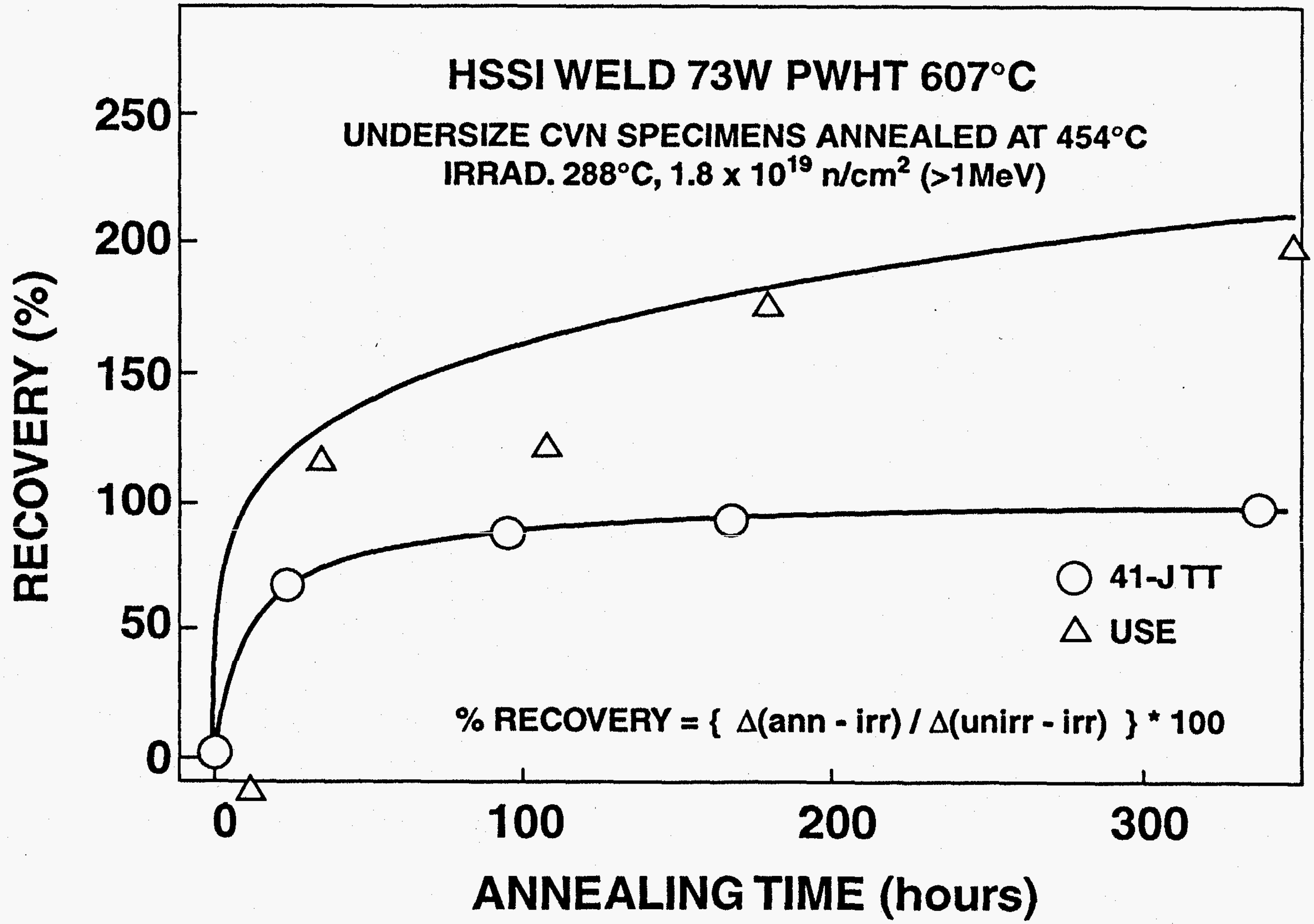


Figure 1. Upon thermal annealing at 454°C, HSSI Weld 73W exhibited rapid recovery of Charpy impact toughness. Diminishing returns of the transition temperature were observed after 96 hours, but the upper-shelf energy continued to increase even up to 336 hours.

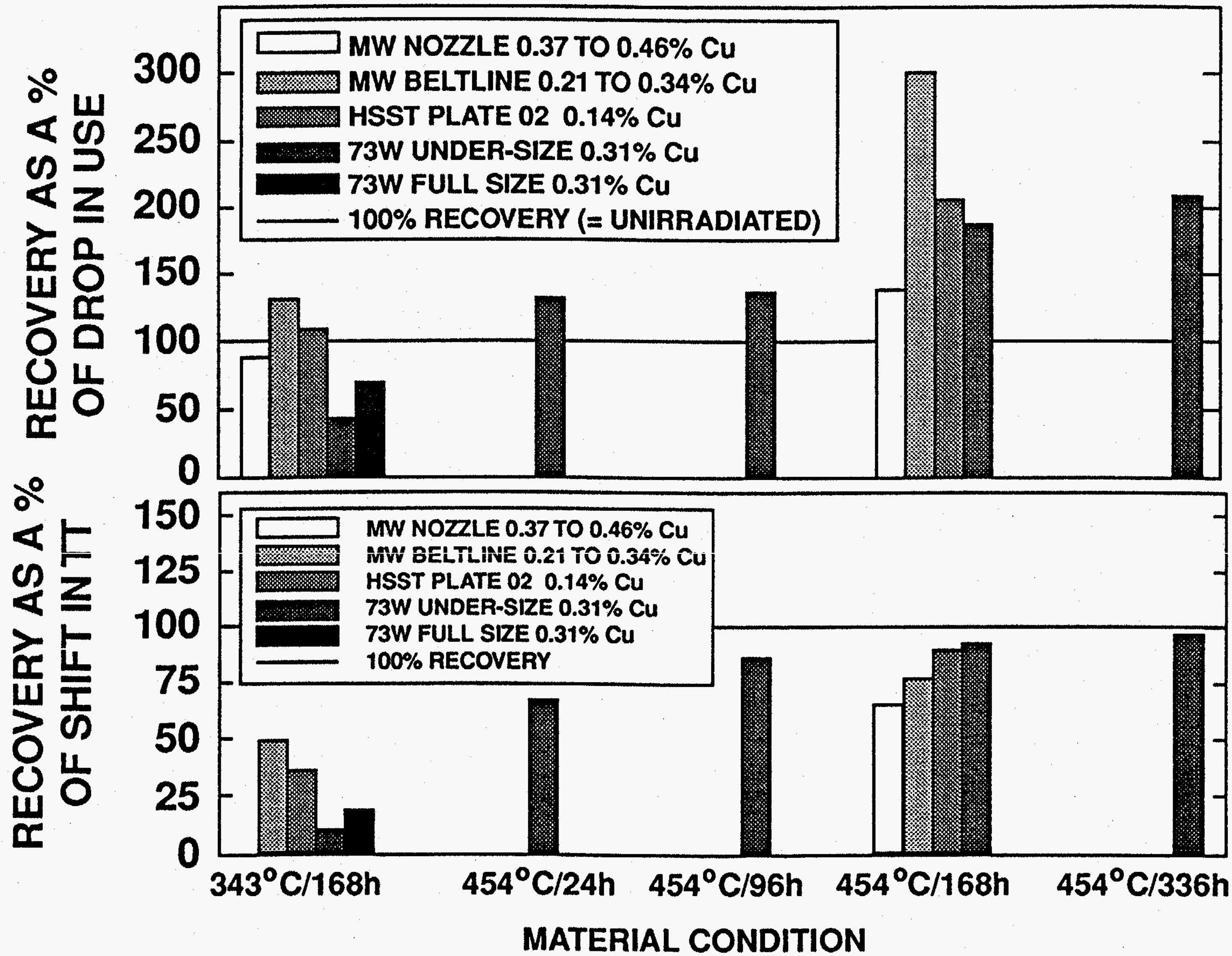


Figure 2. Percent recovery of Charpy upper-shelf energy and 41-J transition temperature for three submerged-arc welds and one plate following thermal annealing at various temperatures and times.

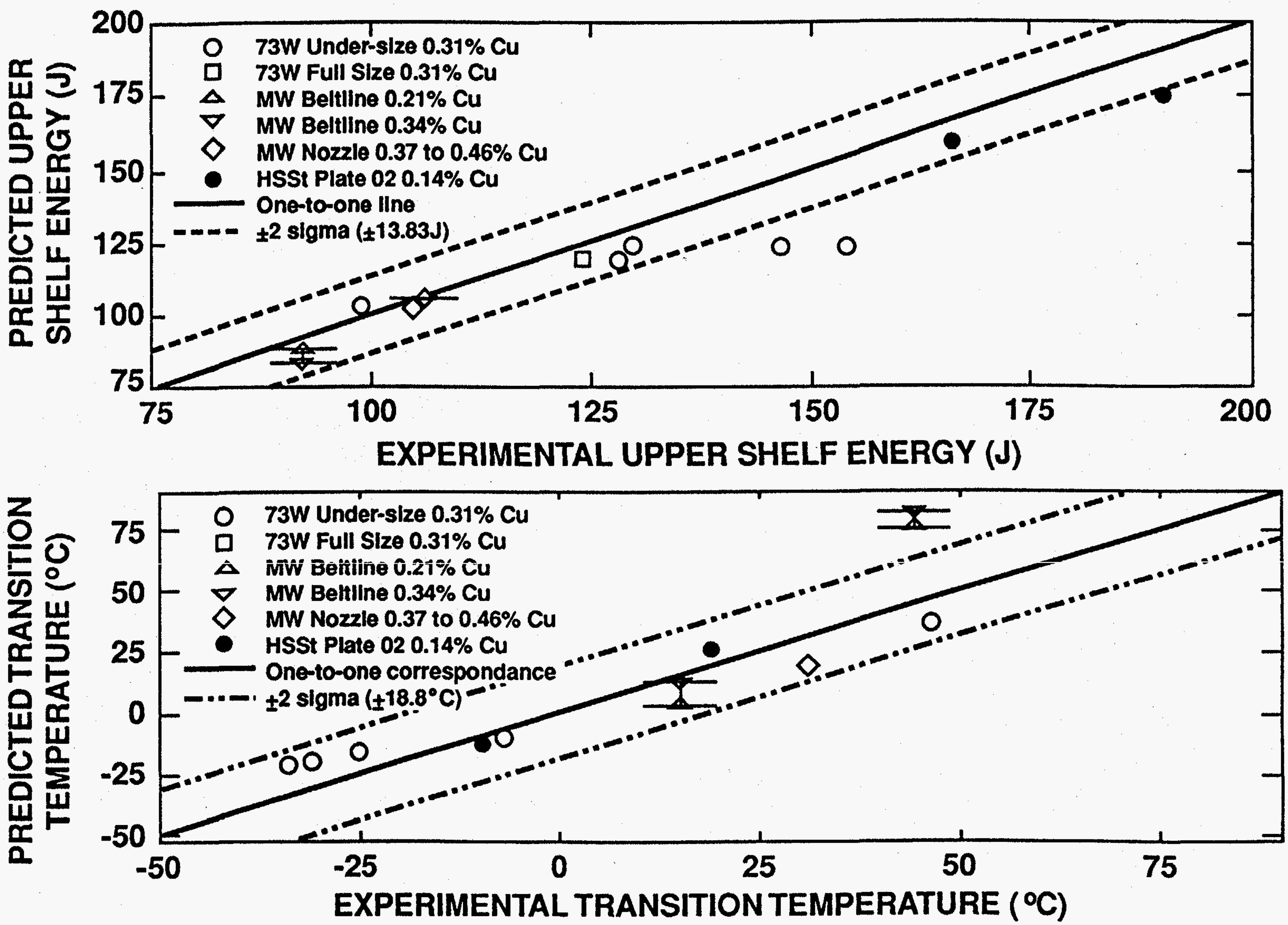


Figure 3. Comparison of annealing recovery measurements with the Eason et al., prediction models. For the materials and conditions investigated, the annealing recovery model has generally given, within ±2σ, good to conservative predictions of transition temperature and upper-shelf energy.

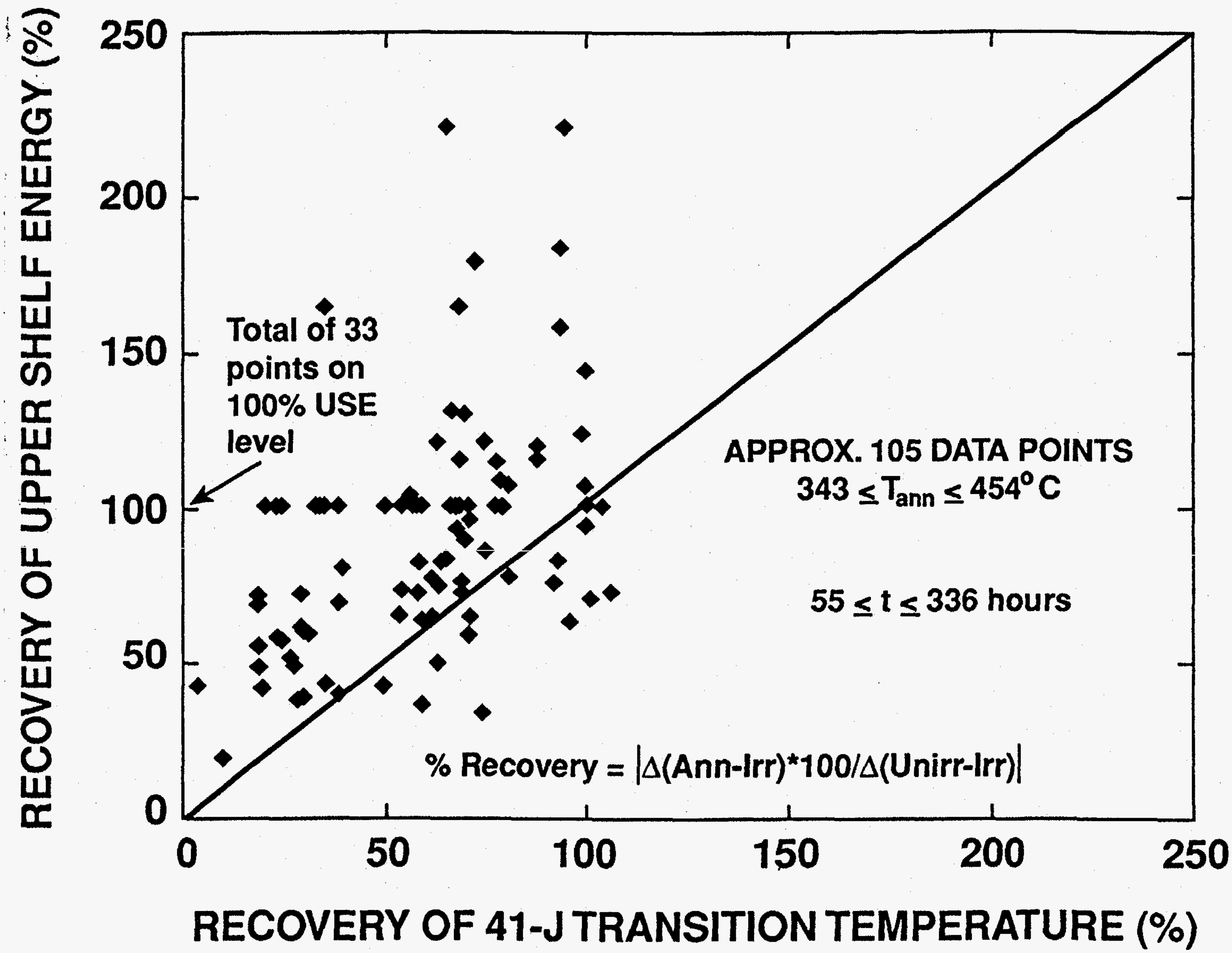


Figure 4. Comparison of Charpy upper-shelf energy and transition temperature recoveries following thermal annealing from published data.

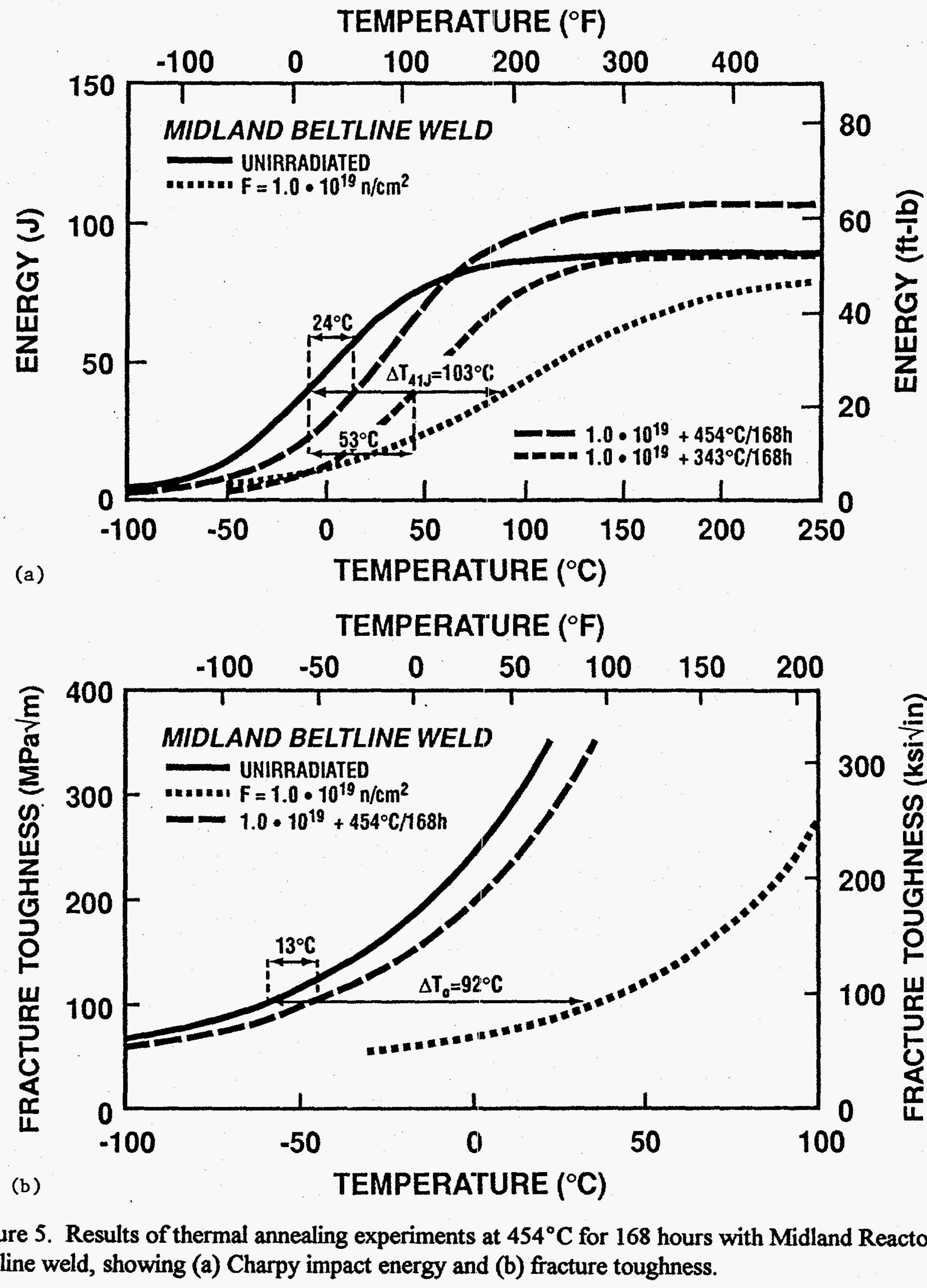
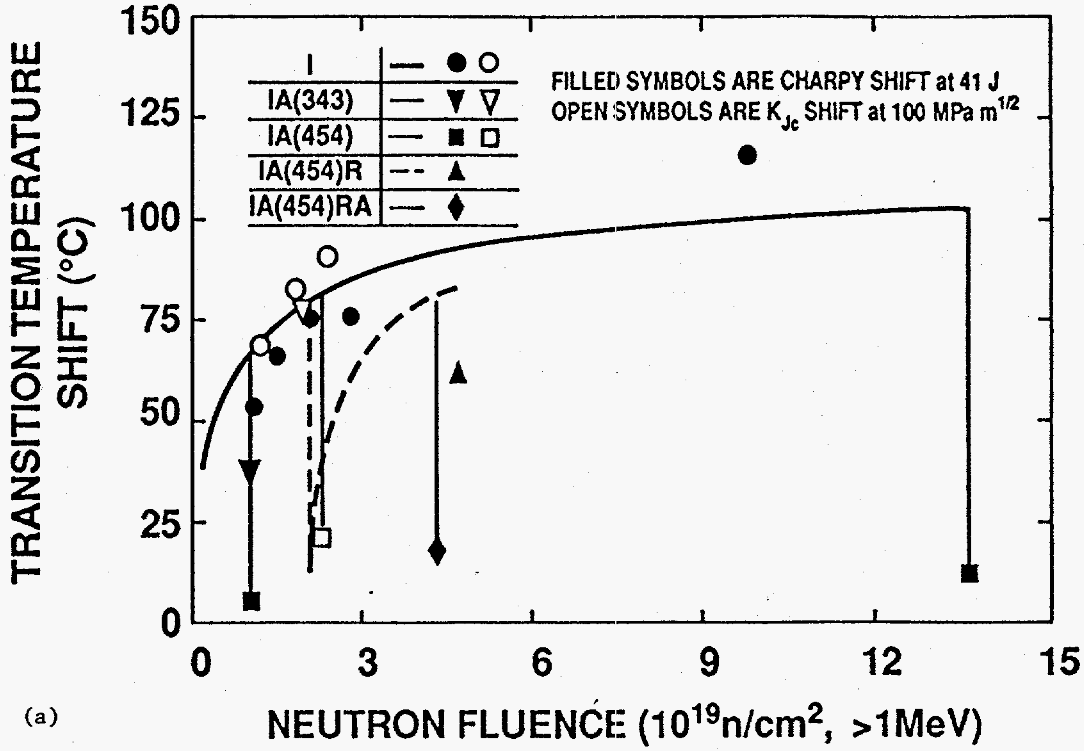
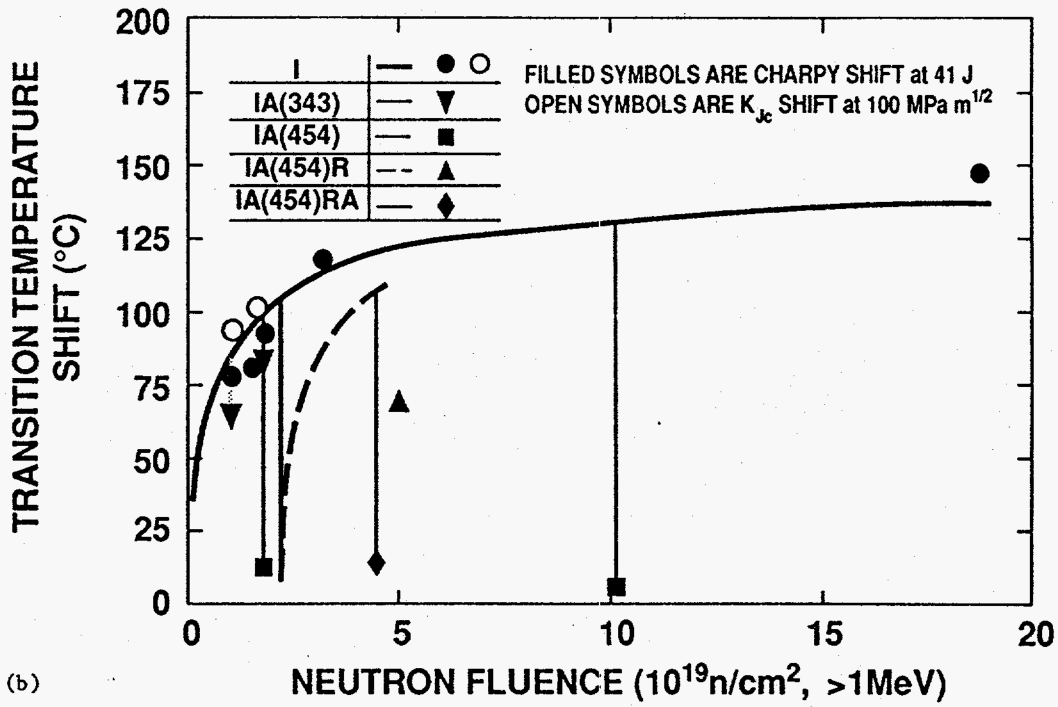


Figure 5. Results of thermal annealing experiments at 454°C for 168 hours with Midland Reactor beltline weld, showing (a) Charpy impact energy and (b) fracture toughness.



(a)



(b)

Figure 6. Results of irradiation, annealing, and reirradiation experiments with (a) HSST Plate 02 and (b) HSSI Weld 73W, by Oak Ridge National Laboratory and Russian Research Center-Kurchatov Institute.

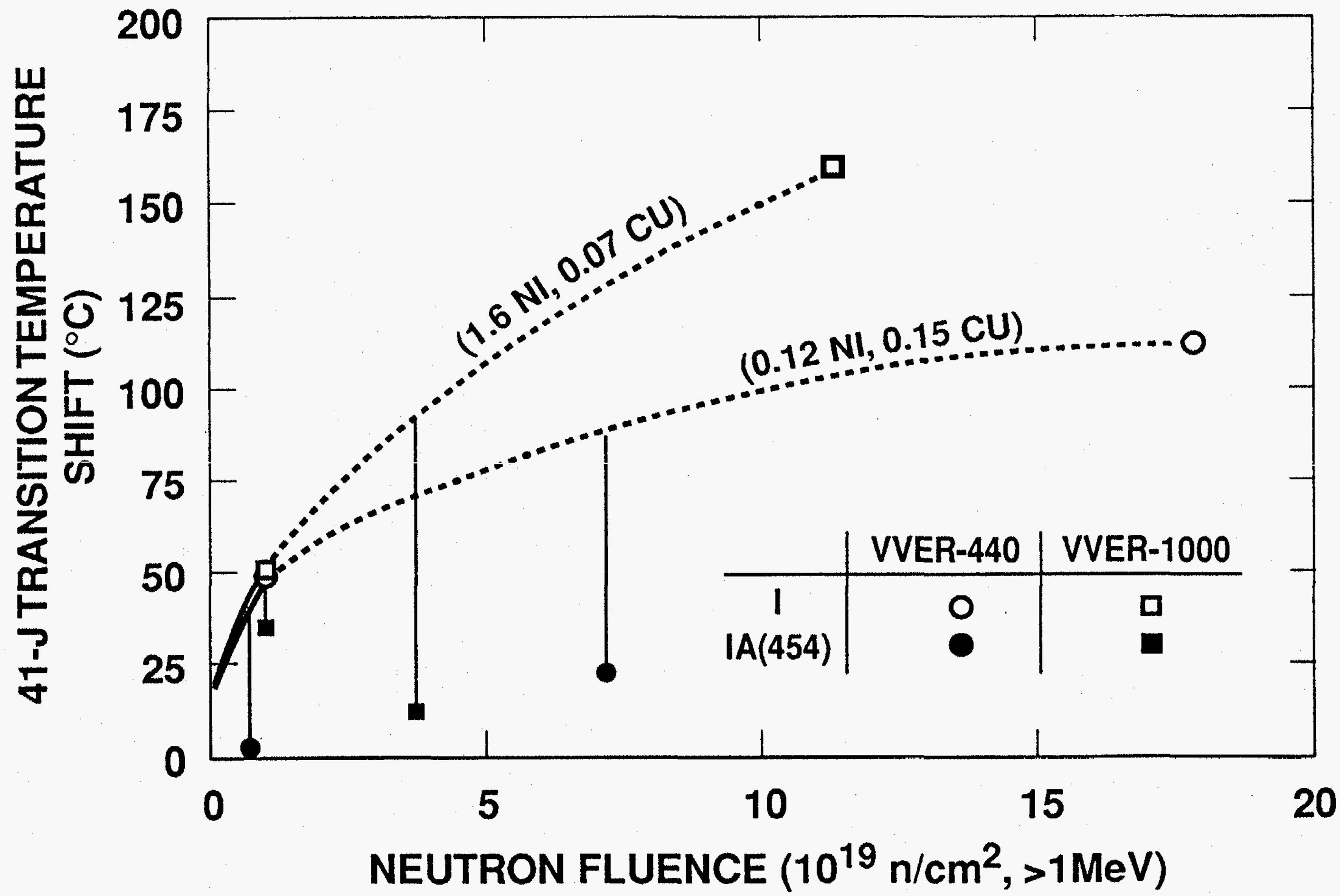


Figure 7. Results of irradiation and annealing experiments with Russian VVER-440 and VVER-1000 welds by Oak Ridge National Laboratory and Russian Research Center-Kurchatov Institute.