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EMBRITTLEMENT OF THE SHIPPINGPORT REACTOR SHIELD TANK*

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Abstract

The irradiation embrittlement of the Shippingport neutron shield tank material has been characterized. Irradiation increases the Charpy transition temperature (CTT) by ~25°C (45°F) and decreases the upper shelf energy. The shift in CTT is not as severe as that observed in the HFIR surveillance specimens. However, the actual value of CTT is higher than that for the HFIR data and the toughness at service temperature is low. The increase in yield stress is 51 MPa (7.4 ksi), which is comparable to the HFIR data. The results also indicate a low impact strength and higher transition temperature for the TL orientation than that for the LT orientation. Some effects of the location across the thickness of the wall are also observed for the LT specimens; CTT is slightly greater for the specimens from the inner region of the wall. The data agree well with results from high-flux test reactors. Annealing studies indicate complete recovery of embrittlement after a 2-h anneal at 400°C. The transition curve for the annealed inner wall specimens is virtually identical to that for the as-received outer wall. The results for weld specimens from the inner and outer walls are also presented.

1. Introduction

Surveillance specimens from the high flux isotope reactor (HFIR) at Oak Ridge National Laboratory showed an very high degree of embrittlement compared with data obtained on similar materials in materials testing reactors .[1-3] One explanation of the difference between the HFIR data and the test reactor data is that, for a given irradiation level, embrittlement is greater at lower flux. Since current NRC guidelines for the assessment of the embrittlement of the pressure vessel support structures of commercial light water reactors do not consider flux, the HFIR results raise the possibility that they may not be sufficiently conservative.

To help resolve this issue, a program was initiated to characterize the irradiation embrittlement of the neutron shield tank (NST) from the decommissioned Shippingport reactor. The Shippingport NST, which operated at 55°C (130°F), was fabricated from rolled A212 grade B firebox steel similar to that used for the HFIR vessel. The inner wall of the NST was exposed to a total maximum fluence of ~6 x 10^{17} n/cm^2 (E>1 MeV) over a life of 9.25 effective-full-power years. This corresponds to a fast flux of 2.1 x 10^9 n/cm^2·s. In comparison the HFIR surveillance specimens were exposed at the inner surface of the pressure vessel at a temperature of 50°C (122°F) over a period of ~17 effective-full-power years; the flux of fast neutrons was ~2 x 10^8 n/cm^2 s.[1]

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* RSR FIN Budget No. A2256; RSR Contact: E. Woolridge.

2. Material Characterization

The effort to obtain samples from the NST was sponsored jointly by the NRC and the DOE Plant Life Extension Program (PLEX) at Sandia National Laboratories. The actual sampling was performed by personnel from Battelle Pacific Northwest Laboratory. Eight ~155 mm (~6 in) disc samples of the base metal and three weld samples were obtained from the inner wall, along with the corresponding samples from the outer wall. The layout for the sample locations from the inner wall is shown in Fig. 1. The inner wall is constructed from four plates ~25.4-mm (1 in) thick, welded after assembly. Thus, specimen locations 13, 14, and 15 contain vertical welds; the other locations represent the base metal from the four plates. The total fluence varied among the various locations; the maximum vertical fluence occurred at an elevation of 211.07 m (692.5 ft), whereas the maximum azimuthal fluence occurred at the 20 and 200° positions. The estimated fluence was ~6×10^{17} n/cm^2 (E>1 MeV) for locations 3 and 9, and ~4×10^{17} n/cm^2 for locations 2 and 8.* The outer wall was constructed from two plates joined together by vertical welds at azimuthal positions 0 and 180°. A weld sample was obtained from the outer wall at location 1. No effect of irradiation is expected at the extremely low levels of irradiation for the outer wall; therefore, the outer wall samples were used to determine the unirradiated baseline data.

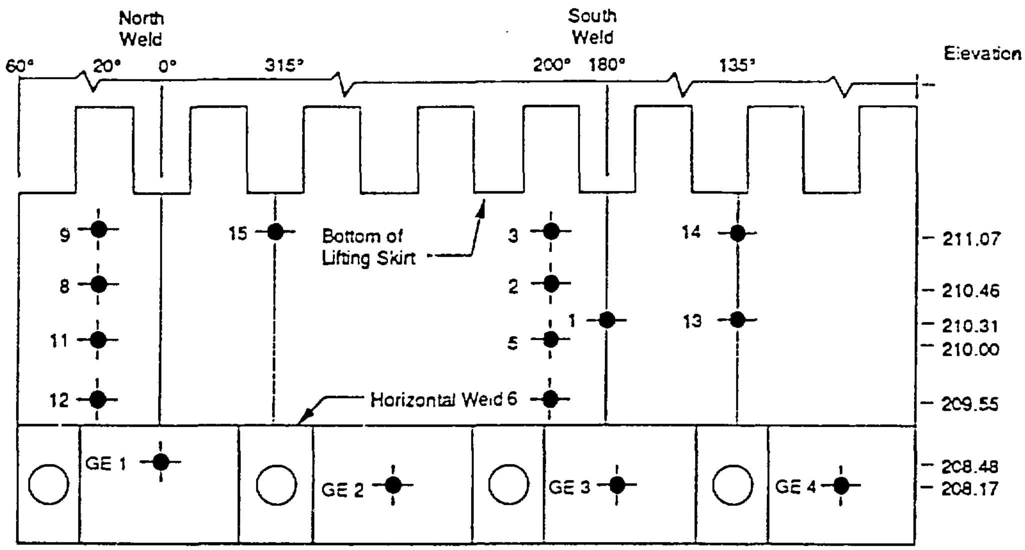

Figure 1. Layout of Sample Location in the Shippingport Neutron Shield Tank Inner Wall.

Metallurgical characterization and chemical analyses of samples taken from each of the plates strongly suggest that both the inner and outer walls of the NST were fabricated from a single heat. Typical chemical analyses for the plate and weld metal are given in Table 1. Metallographic examination of the NST material indicates that the rolling direction is horizontal. Micrographs of the grain structure along the rolling and transverse directions are shown in Fig. 2. The surfaces shown in the micrographs are designated by the direction

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* L. James, Bettis Atomic Power Laboratory, private communication (April 1989).

Table 1. Typical Chemical Compositions (wt.%) of the A212 Grade B Plate and Weld Metal from the Shippingport Neutron Shield Tank

Element	Plate	Weld
C	0.23	0.065
Mn	0.75	0.93
P	0.02	-
S	0.03	-
Si	0.27	0.73[1]
Cu	0.05	0.06[2]
Ni	0.04	0.02
Cr	0.04	0.03
O	0.01	-
N	0.004	-
Ti	<0.005	0.025
V	<0.005	0.019
Zr	<0.005	<0.005
Mo, Ca, Al	<0.01	<0.01
B, Se, Sn	<0.01	<0.01

[1]0.86 for inner-wall weld from location 14.

[2]0.07 for outer-wall weld and 0.04 for inner-wall weld from location 15.

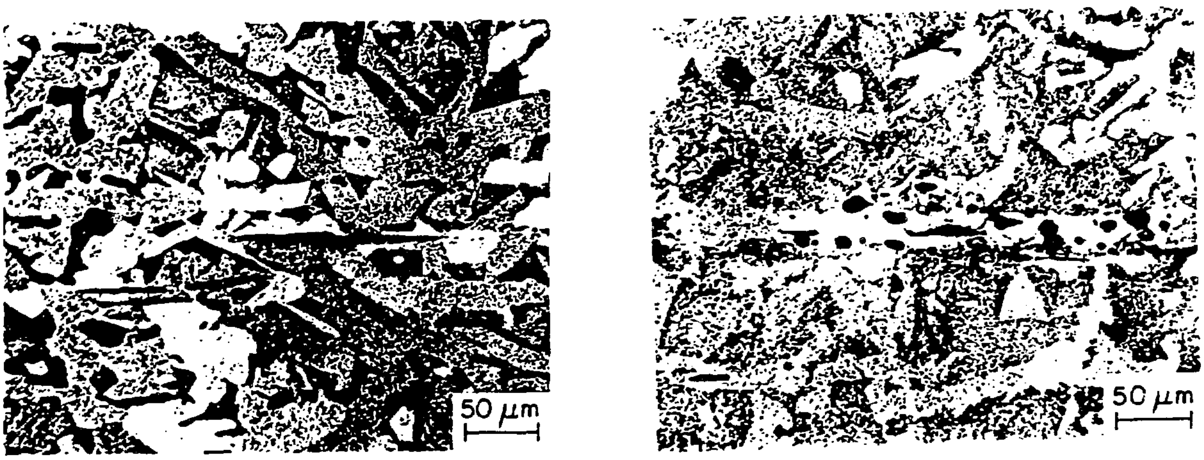
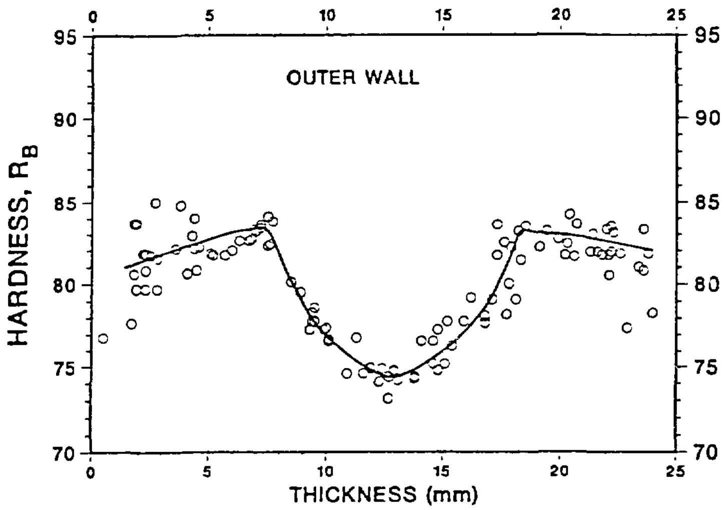


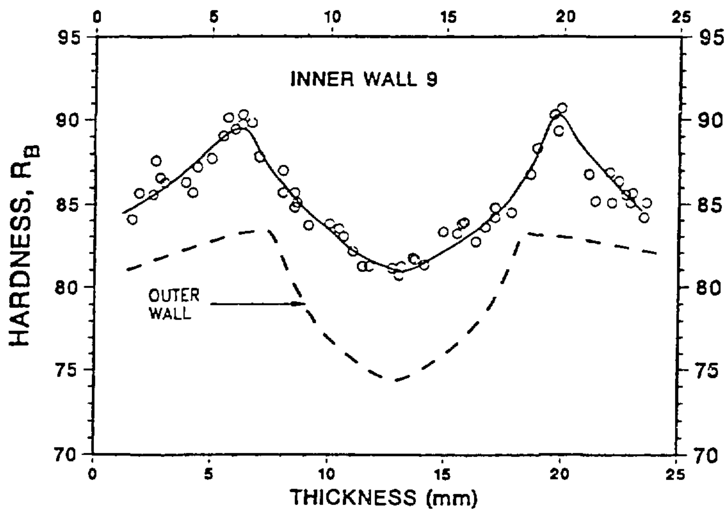
Figure 2. Micrographs of the NST Material Along the (a) Transverse and (b) Rolling Sections.

normal to that surface. The transverse section shows some elongated grains and all the inclusions are elongated in the rolling direction. The inclusions in the rolling section are globular or flat.

There are significant variations in hardness across the thickness of the wall. A typical hardness profile (Rockwell B) for the outer wall is shown in Fig. 3. (The depth is measured from the inner surface of the plate, i. e., the surface towards the reactor core.) The hardness



(a)



(b)

Figure 3. Hardness Profile across the Thickness of the NST Outer Wall and Location 9 of the NST Inner Wall.

values of the inner and outer regions of the plate are ~10% higher than those of the plate center. However, no measurable change in grain size was observed across the thickness of the wall; the average through-wall grain size was ~12 µm. Although irradiation increases the hardness of the material, the V-shape profile is maintained at most locations, Fig. 3.

3. Irradiation Embrittlement

The irradiation embrittlement has been characterized by Charpy impact and tensile tests. Specimens were obtained in the LT and TL orientations* from three regions, i.e., inner, center, and outer 10-mm-wide regions, across the thickness of the NST wall. A typical layout for the base metal specimens from the inner and outer walls is shown in Fig. 4. Material from

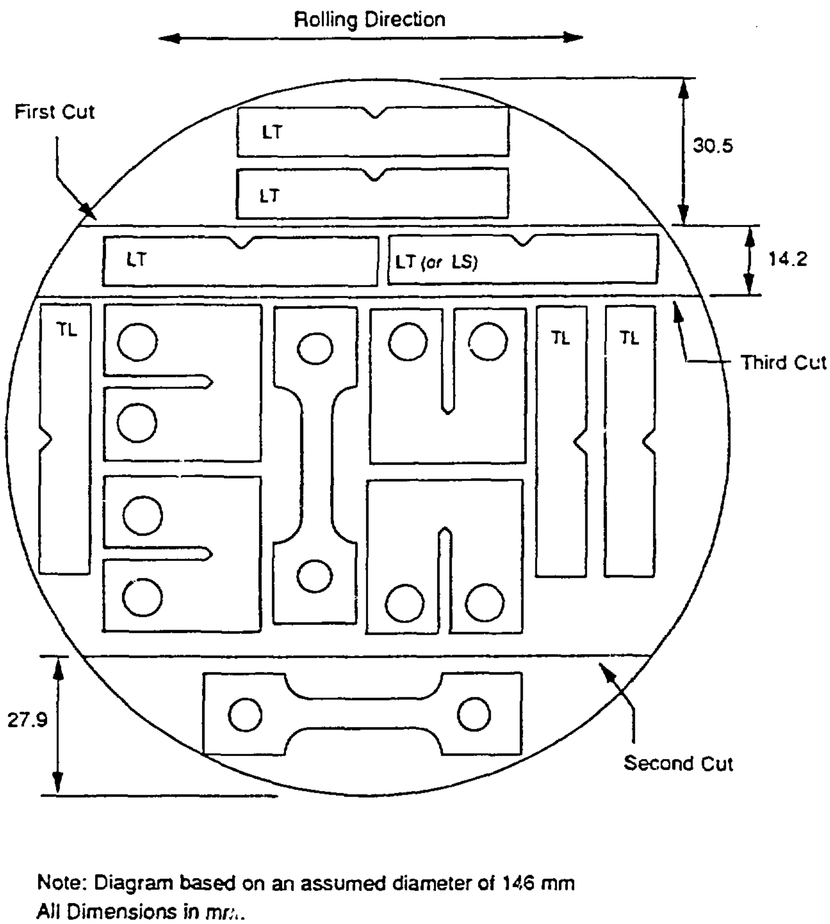


Figure 4. Cutting Diagram for Base Metal Samples from the NST Inner and Outer Walls.

* The first digit designates the direction normal to the plane of the crack; the second digit represents the direction of crack propagation. L = longitudinal or rolling direction and T = transverse direction.

the outer wall, which was protected by ~0.9 m (3 ft) of water and hence had a six-order-of-magnitude lower fluence than the inner wall, was used to obtain baseline data for unirradiated material.

Charpy impact tests were conducted on standard Charpy V-notch specimens machined according to ASTM specification E 23. A Dynatup Model 8000A drop weight impact machine with an instrumented tup and data readout system was used for the tests. Tensile tests were performed on dog bone specimens, with a cross section of 4 x 5 mm and a gauge length of 20 mm.

3.1 Base Metal

Charpy transition curves for the LT and TL specimens from different regions of the NST outer wall are shown in Fig. 5. The results indicate that the variation in the transition curves with vertical and azimuthal position is very slight. However, the TL orientation is weaker than the LT orientation. The Charpy transition temperature (CTT) at the 20.3-J (15 ft·lb) level is higher, and the upper-shelf energy (USE) is lower for the TL specimens. The CTT and USE, respectively, are 16°C (61°F) and 98 J/cm^2 (~58 ft·lb) for LT specimens and 19°C (66°F) and 56 J/cm^2 (~33 ft·lb) for TL specimens. The differences in impact strength are attributed primarily to differences in the structure of the material. The plane of the crack for TL orientation, i.e., transverse section shown in Fig. 2, contains elongated inclusions.

The results also indicate some effect of position through the thickness of the wall; impact energies for specimens from the inner and outer regions of the wall are comparable, whereas those for the center specimens are slightly higher, Fig. 5b. The CTT and USE for the center specimens are 9°C (48°F)and 104 J/cm^2 (~61 ft·lb), respectively. This correlates with the differences in the hardness of the material. The hardness of the center region is R_B ~75 (142 DPH) and that of the inner or outer regions is R_B ~83 (159 DPH).

The transition curves for the LT and TL specimens from different positions on the NST inner wall, are shown in Fig. 6. The irradiated inner wall specimens show a higher CTT and lower USE relative to those from the unirradiated outer wall. The transition curves are almost independent of vertical position, which suggests that a factor of two in the variation of fluence has relatively little effect. For example, the impact energies for specimens from locations 2 and 8 (~4 x 10^{17} n/cm^2 fluence) are comparable to those for specimens from locations 3 and 9 (~6 x 10^{17} n/cm^2 fluence). For LT specimens, some differences are observed for those from the inner and outer regions of the wall; the shift in CTT is slightly greater for the inner region. The values of CTT are 37°C (99°F) for the outer region and 41°C (106°F) for the inner region, a shift of ~21 and 25°C (~38 and 45°F) for the outer and inner regions, respectively. The USE can not be established from the data in Figs. 6a and 6b. However, the specimens tested at 55°C show 100% shear fracture; thus, the impact energies for these specimens are representative of USE.

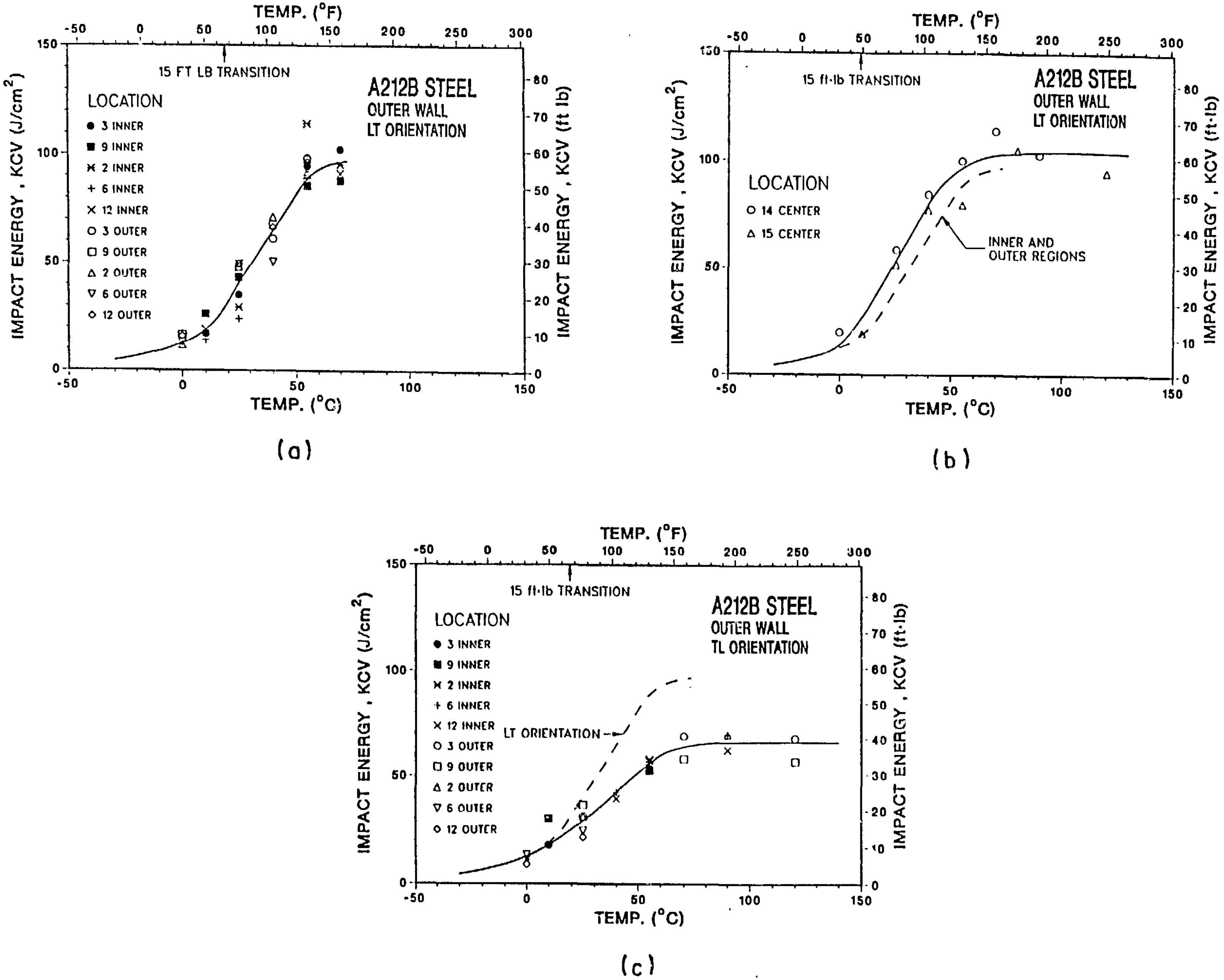


Figure 5. Charpy Impact Test Data for the NST Outer Wall. LT specimens from (a) inner and outer regions and (b) center region. TL specimens from (c) inner and outer regions.

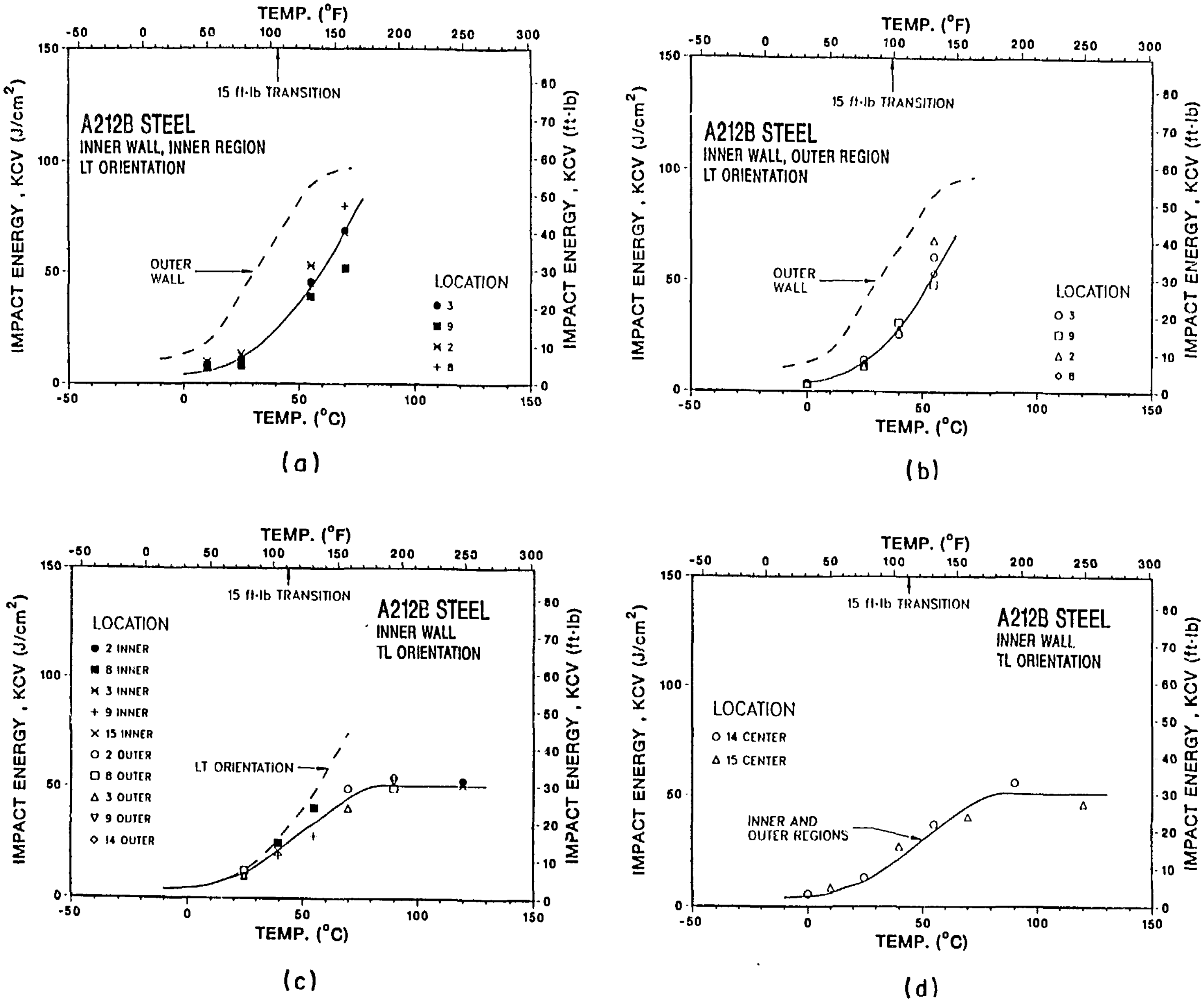


Figure 6. Charpy Impact Test Data for the NST Inner Wall. LT specimens from (a) inner and (b) outer region. TL specimens from (c) inner and outer regions and (d) center region.

The shift in CTT of the inner-wall TL specimens is also 25°C (45°F), similar to that for the LT specimens. However, the actual value of the CTT is higher, 44°C(111°F). The effects of material hardness are minimal for the TL specimens. The impact energies for the center specimens are comparable to specimens from the inner and outer region, although the hardness is significantly different, e.g., R_B ~82 and 88 (156 and 176 DPH) for the center and inner or outer regions, respectively.

Tensile tests on LT specimens from several locations of the NST inner and outer walls were conducted at room temperature and at 55°C (131°F). The results indicate a higher yield stress for the inner wall relative to the outer wall, whereas the ultimate stress is approximately the same. The increase in yield stress is ~51 MPa (7.4 ksi) at room temperature and ~40 MPa (5.8 ksi) at 55°C. The tensile strength for locations 2 and 8 (~4 x 10^{17} n/cm^2 fluence) is comparable to that for locations 3 and 9 (~6 x 10^{17} n/cm^2 fluence).

Hardness influences the tensile properties at both test temperatures, i.e., the yield stress for the specimen from the center of the wall is always lower than that for the specimens from inner or outer regions. The difference is ~30 MPa (4.4 ksi) at room temperature and ~15 MPa (2.2 ksi) at 55°C. However, the increase in yield stress due to irradiation is the same for specimens from the center and from the inner or outer regions of the walls.

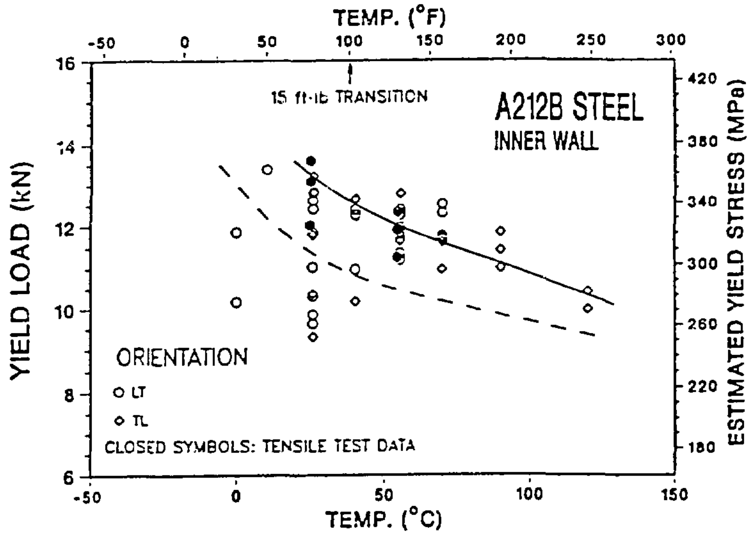
The tensile properties of the NST material were also estimated from the Charpy impact data. For dynamic loading, the yield stress is estimated from the expression

$$\sigma_y = AP_yB/Wb^2, \quad (1)$$

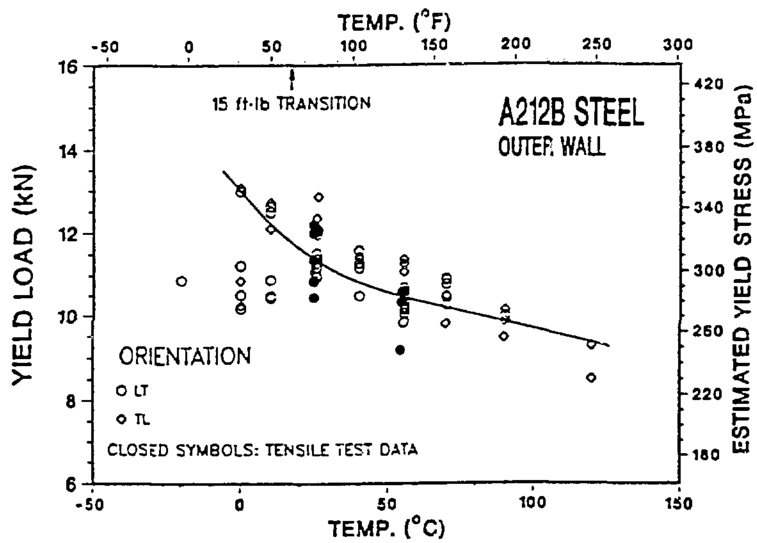
taken from Ref. 4, where P_y is the yield load obtained from the load-time traces of the instrumented Charpy tests, W is the specimen width, B is the specimen thickness, b is the uncracked ligament, and A is a constant. The constant A was obtained by comparing the tensile and Charpy data for LT specimens tested at room temperature and at 55°C. The best value of the constant was 1.73. The yield loads and the estimated yield stresses for the outer and inner walls are shown in Fig. 7. The measured yield stresses are compared with the estimated values. The results show the expected decrease in yield stress with an increase in test temperature. In the lower-shelf temperature regime, cleavage occurs before general yielding; therefore, the yield loads are very low. For the Charpy impact tests, cleavage fracture occurs when the yield loads are ~13 kN. Irradiation increases the yield stress at all test temperatures and, therefore, the CTT shifts to higher temperatures. Irradiation hardening is greater at room temperature than at higher temperatures.

3.2 Weld Metal

Weld samples were obtained at one position on the outer wall and three positions on the inner wall, i.e., locations 14 and 15 with ~3 x 10^{17} n/cm^2 fluence and location 13 with ~2 x 10^{17} n/cm^2 fluence. All the welds were transverse to the plate rolling direction. Charpy impact test specimens were machined perpendicular to the weld and from the inner and outer regions across the thickness of the plate. The impact strength of the outer-wall weld is significantly higher than that of the base metal, Fig. 8a. The 41-J (30 ft·lb) CTT and USE of the weld are -2°C (28°F) and 182 J/cm^2 (107 ft·lb), respectively. Location through the thickness of the weld has little or no effect on the transition curve.



(a)



(b)

Figure 7. Yield Stress Estimated from the Charpy Impact Data for the NST (a) Inner and (b) Outer Walls.

The inner wall specimens show significant effect of position around the wall as well as through the thickness of the wall. The impact energies of weld specimens from the outer region of positions 13, 14, and 15 are comparable but slightly higher than those of the outer-wall weld specimens, Fig. 8b. The transition curves for the inner region of the inner-wall welds are shown in Figs. 8c and 8d. The 41-J (30 ft·lb) CTT is 8°C (46°F) for welds at locations 13 and 15 and 23°C (73°F) for the weld at location 14, i.e., a shift of 10 and 25°C (18 and 45°F), respectively.

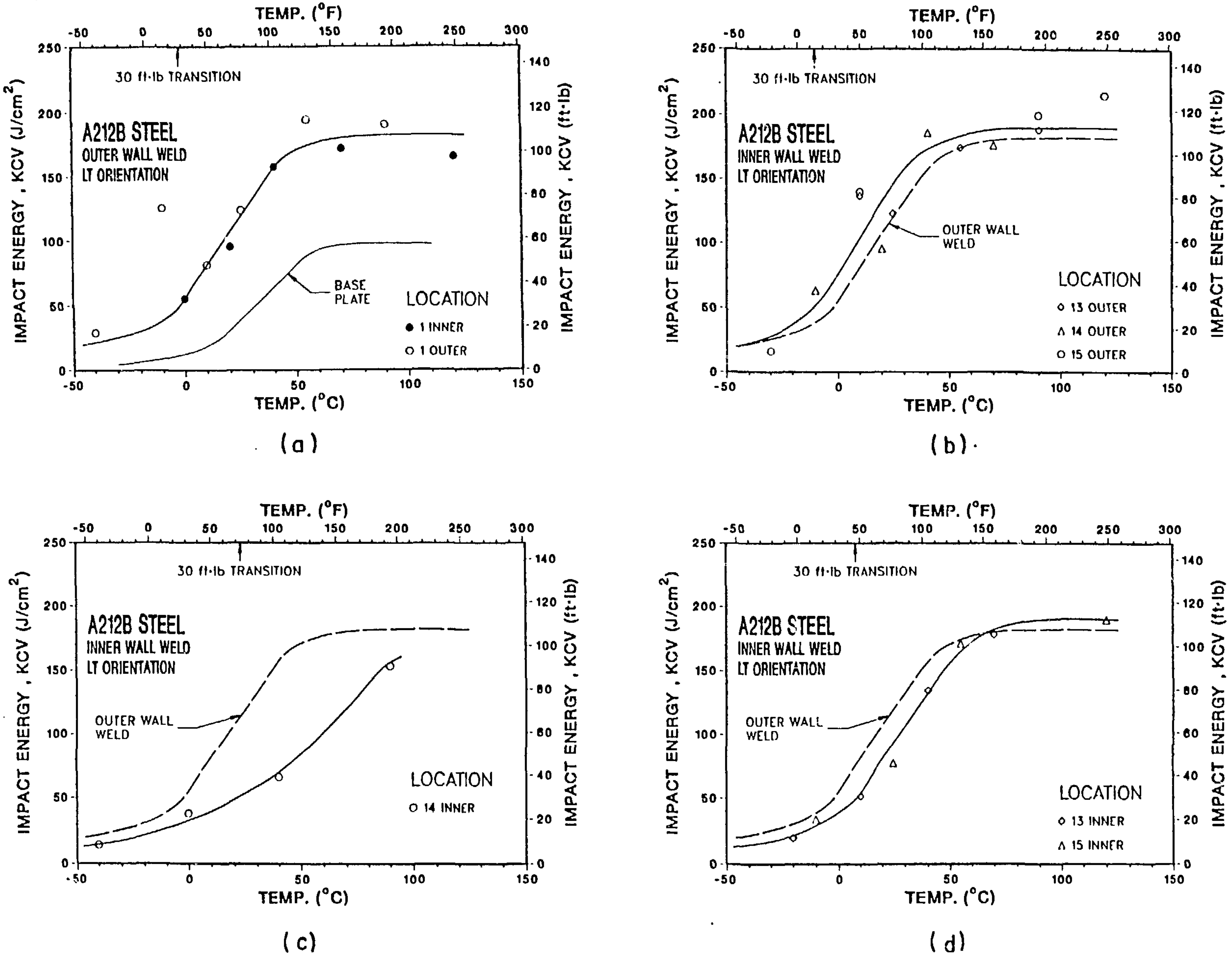


Figure 8. Charpy Impact Test Data for Weld Metal Specimens from (a) NST Outer Wall, (b) Inner Wall Outer Region, and (c) and (d) Inner Wall Inner Region.

for the two data sets. The results for the outer weld may not be representative of the unirradiated inner-wall welds. The chemical composition of weld metals from different locations indicates only minor variations in silicon and copper content, Table 1. Additional tests and metallographic characterization of the welds are being performed to better establish the transition curves. Annealing studies, to obtain baseline data and determine the embrittlement behavior of the welds, are also planned.

4. Recovery Annealing

Annealing studies were conducted on material from the NST inner and outer walls to study the recovery behavior of embrittled material. Specimens were annealed at 400°C for up to 154 h and the annealing behavior was characterized by hardness measurements. The results indicate that the hardness of irradiated material from the inner shell decreases after annealing, whereas the hardness of the outer-shell material increases. Annealing for 1 h at 400°C (752°F) was sufficient for recovery; only a very slight increase in hardness of both the inner- and outer-shell materials occurs after annealing for longer times. The changes in hardness of material from inner and outer shells, shown in Fig. 9, reflect the differences in the fluence and flux levels of the different locations, i.e., the decrease for locations 3 and 9 is greater than for location 8.

Charpy impact data for TL specimens from the NST inner and outer walls, annealed for 2 h at 400°C, are shown in Fig. 10. The results indicate a complete recovery from irradiation embrittlement, i.e., the transition curve of the annealed specimens from the inner wall is identical to that of the outer wall. Annealing has little or no effect on the transition curve of the outer wall, Fig. 10.

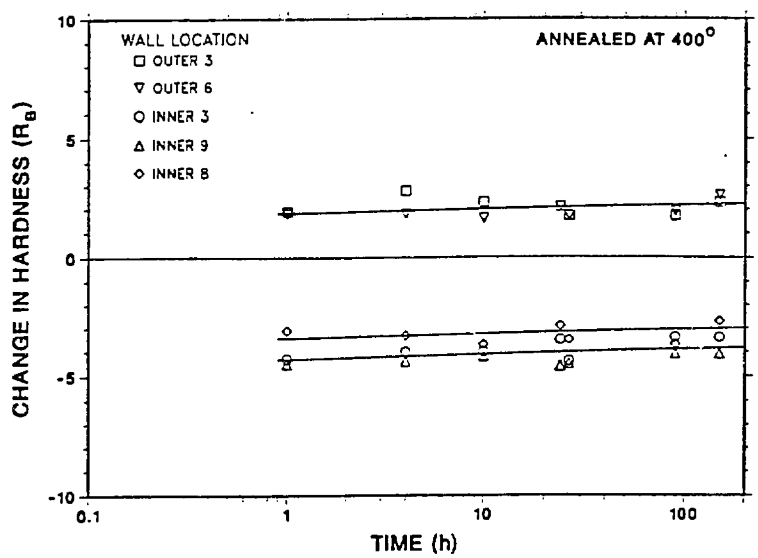
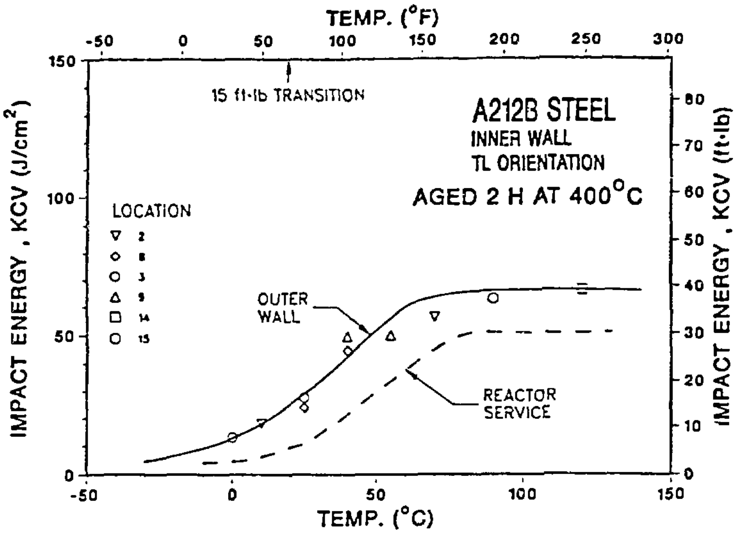
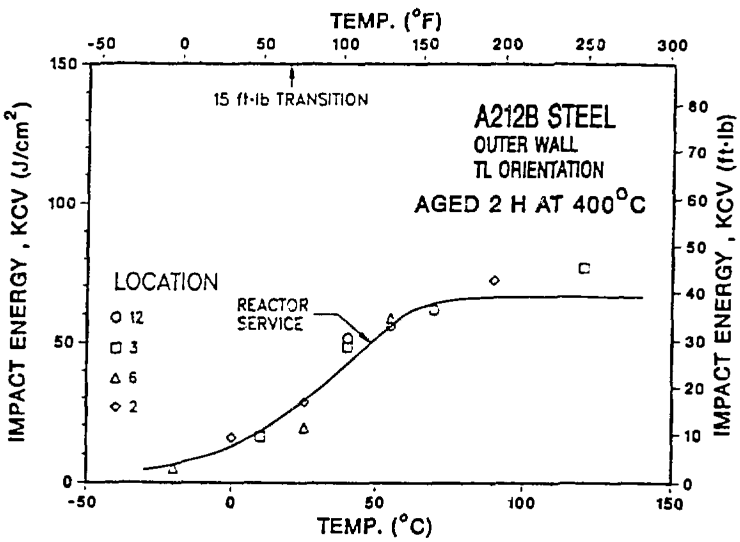


Figure 9. Change in Hardness of the Neutron Shield Material from Inner and Outer Walls after Annealing at 400°C.



(a)

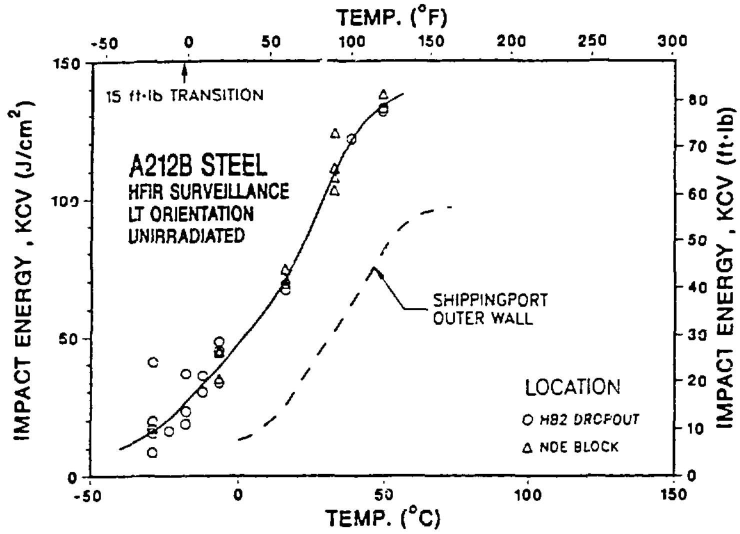


(b)

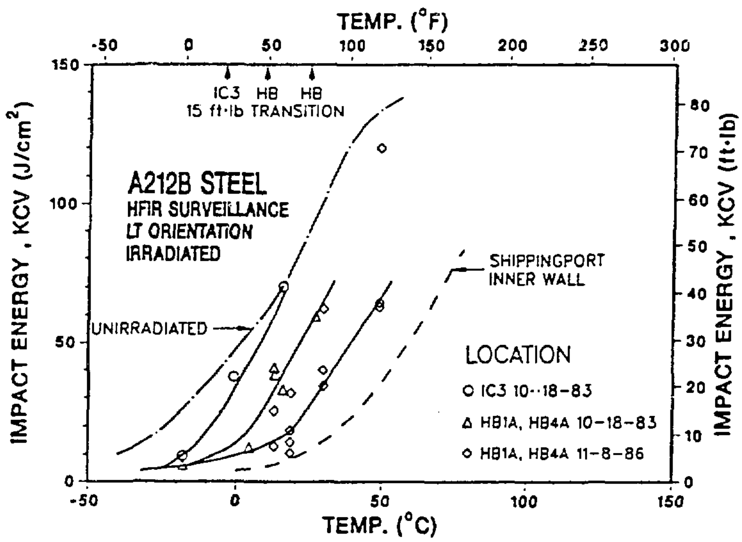
Figure 10. Charpy Impact Test Data for Annealed TL Specimens from (a) Inner Wall and (b) Outer Wall.

5. Discussion

Charpy impact and tensile data for the Shippingport NST indicate that the shift in CTT is not as severe as would be expected on the basis of the changes seen in HFIR surveillance samples. In Fig. 11, Charpy transition curves for the outer and inner walls of the NST are compared with the results for unirradiated and irradiated HFIR surveillance samples. Although the shift is smaller, the actual CTT of the NST material is significantly higher than that of the HFIR material. The impact energy of the NST inner wall is very low, ~40 J/cm^2 (24 ft·lb), at the service temperature of 55°C (131°F).



(a)



(b)

Figure 11. Comparison of Charpy Impact Data for (a) Unirradiated and (b) Irradiated Shippingport NST and HFIR Surveillance Samples.

Except for minor differences in copper and nickel content, the chemical composition of the two materials is comparable. The concentrations of copper and nickel are 0.15 and 0.20 wt.%, respectively, for HFIR material and 0.05 and 0.04 wt.% for NST. Although the HFIR material is tougher than the NST material, the tensile strength of the HFIR material is greater than that of the NST material. The yield strength and hardness, respectively, are 355 ± 11 MPa (~51 ± 2 ksi) and 170 DPH for the HFIR material[2] and 305 ± 19 MPa (44 ± 3 ksi) and 159 DPH for

NST. The difference in the transition curves of the two unirradiated materials is most likely due to microstructural factors, such as the amount and distribution of inclusions.

When the shifts in CTT of the NST material are compared with the results obtained from the HFIR material irradiated in the ORR and other A212B steels irradiated in high-flux test reactors, Fig. 12, the results from the Shippingport NST are consistent with the test reactor data. The results for the NST material also agree very well with correlations for the shift in CTT, the increase in tensile yield stress, and the increase in hardness that were developed for pressure vessel steels.[5-7] The shift in transition temperature, ΔT, with an increase in tensile yield stress, $\Delta\sigma_y$, is expressed as

$$\Delta T\ (°C) = C\Delta\sigma_y\ (MPa), \quad (2)$$

where C ~0.5°C/MPa for plate material, and 0.65°C/MPa for welds. The change in yield stress with hardness, ΔDPH, is given by the relation

$$\Delta\sigma_y\ (MPa) = 3.5\Delta DPH. \quad (3)$$

The shift in the CTT of both LT and TL specimens is 25°C and the increase in yield stress is 51 MPa. The increase in hardness due to irradiation is difficult to obtain because of the variation in hardness across the thickness of the wall. However, annealing studies indicate that the irradiation-induced hardness change, represented by the decrease in hardness after annealing at 400°C for 2 h, is ~4.3 R_B (14 DPH) for locations 3 and 9. This is consistent with Eq. (3). The change in hardness is slightly lower, i.e., ~3.5 R_B (10 DPH), for location 8.

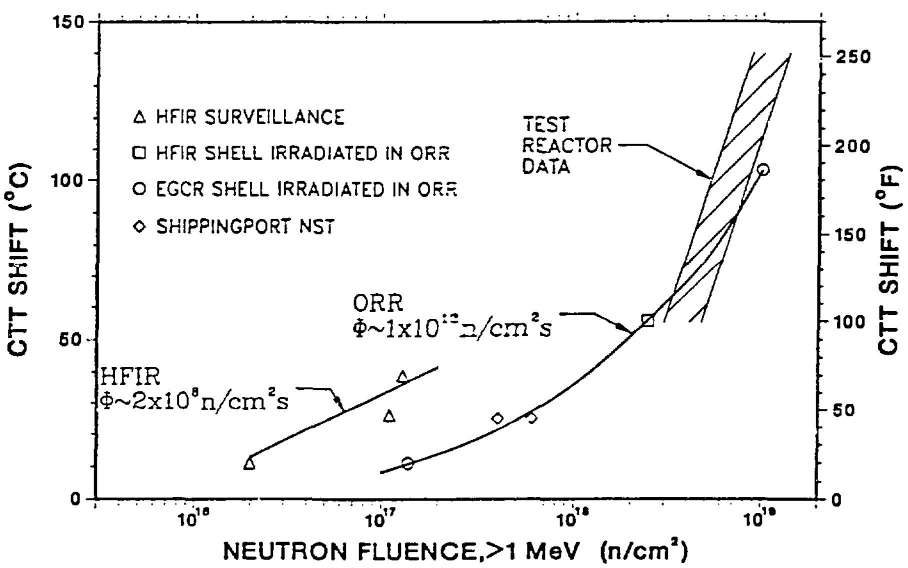


Figure 12. Comparison of Transition Temperature Shifts for Shippingport NST with HFIR Surveillance Results and A212B data from High-Flux Test Reactors.

The results from the present study suggest that the unexpectedly high embrittlement of the HFIR surveillance samples may be due to factors other than flux effects. The minor differences in copper and nickel content between the two materials are not expected to have any effect at the low irradiation temperatures of Shippingport NST and the HFIR. Analytical studies are in progress at ANL to investigate radiation damage measures to determine whether a unified picture of embrittlement in HFIR, ORR, and Shippingport can be achieved. High-flux irradiation experiments will be performed for materials from the Shippingport NST and the HFIR vessel to confirm the analysis and evaluate the possible effects of metallurgical differences between the two materials.

6. Conclusions

Characterization of material from the Shippingport NTS indicates that the embrittlement of A212 grade B steel in a low-temperature, low-flux environment is consistent with that expected from the current NRC regulatory guidelines. However, the shifts in CTT are not as great as those of the HFIR surveillance samples. The reasons for this difference are not well understood.

The data on the weld samples from the NST indicate that the weld metal is tougher than the plate material. The results, however, show significant scatter, and irradiation embrittlement could not be characterized. Annealing studies to investigate the recovery behavior of the weld metal and to obtain additional information on the shift in CTT are planned.

Acknowledgments

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