

DG03/5

RECENT EVALUATION OF "WET" THERMAL ANNEALING TO RESOLVE REACTOR PRESSURE VESSEL EMBRITTLEMENT

W.L. Server¹ and E.C. Biemiller²

¹ATI Consulting, San Ramon, CA, USA

²Yankee Atomic Electric Company, Bolton, MA, USA

INTRODUCTION

Prior to the decision to close the Yankee Rowe plant in 1992, a great deal of effort was expended in trying to resolve the degree of neutron embrittlement that the reactor pressure vessel had experienced after 30 years of operation. One mitigative measure that was examined in detail was the possibility of performing a relatively low temperature thermal anneal (at approximately 650°F) to partially restore the original design level of mechanical properties of the reactor pressure vessel beltline region which were lost due to the neutron radiation exposure. This low temperature anneal was to involve heating of the primary coolant water using pump heat in a similar manner as that used to anneal the Belgian BR-3 reactor pressure vessel in the early 1980s (Fabry et al., 1980). This "wet" anneal was successful in recovering mechanical properties for the BR-3 vessel, but the extent of the recovery, as well as the rate of re-embrittlement after the anneal, were issues that were difficult to quantify since the exact reactor pressure vessel steels were not available for experimental verification.

For the case of Yankee Rowe, material was available from past surveillance programs for at least one of the materials in the vessel, as well as materials obtained from various sources which could act as bounding surrogates. An irradiation / annealing / reirradiation program was developed to better quantify the degree of recovery and re-embrittlement for these materials, but this program was halted before significant test results were obtained. Prior to the initiation of the testing program, a review of past annealing data was performed and the data were scrutinized for direct relevance to the annealing response of the Yankee Rowe vessel. This paper discusses the results derived from this review.

MATERIAL CONSIDERATIONS

The Yankee Rowe reactor pressure vessel was fabricated in the late 1950s for Westinghouse by Babcock and Wilcox Company and contained two distinct types of base metal: A302B steel in the upper shell course which had very little nickel and A302B-modified steel in the lower shell course which had nickel as a distinct alloying element (approximately 0.63 wt%). Note that A302B-modified is the precursor to the more prevalent vessel plate material A533B-1. The plate materials have copper contents of

about 0.20 wt%. Both the axial and circumferential welds were made using a Linde 80 flux with an electrode that was copper coated. The exact chemistry for the weld was not known, and a plan to remove some of the actual vessel weld metal for chemical analysis was planned prior to the decision to close the Yankee Rowe plant.

The A302B steel was contained in the early Yankee Rowe surveillance program^[1]. This same heat of A302B steel also was contained in the surveillance program supplied by Westinghouse (another Babcock and Wilcox-fabricated vessel) for the BR-3 vessel in Belgium. Therefore, surveillance data exist for this material from two different sources. Since there was no unirradiated archive material for either plate material nor surveillance results for the lower plate steel, surrogate materials were carefully selected to best match or bound the actual Yankee Rowe vessel steels. Heat treatment and microstructure were examined as key aspects to embrittlement response. The most limiting material was predicted to be the lower plate steel based upon conservative estimates made by the NRC.

THERMAL ANNEALING DATA REVIEW

The first attempt at assessing the degree of recovery for the Yankee Rowe vessel steels utilized the correlation developed using a term called the Annealing Effect Parameter (AEP)^[2]. This approach proved to be inadequate due to the lack of data in the area of interest for the correlation (i.e., low values of AEP). Additionally, the correlation ignored data with high degrees of initial embrittlement which is generally inconsistent with the Yankee Rowe materials. Available data and analyses from the published literature^[1-7] were examined and a key source of data was acquired from the Belgians from extensive work performed in support of the anneal of the BR-3 vessel^[8,9].

Tables 1 and 2 list the most pertinent data for base metal and welds for a low temperature anneal of approximately 650°F for two weeks. The data were segregated in terms of: (1) the difference in annealing and normal operating temperature (i.e., annealing at 650°F and operation at about 500°F equals a difference of approximately 150°F), (2) the time of annealing, (3) the degree of embrittlement (shift) prior to annealing, and (4) comparable material. The most applicable data are listed first, and the average results for base metal shows recovery of around 45%; weld metal recovery for the most applicable data is on average above 50%. The data identified as "Other Related Data" are presented to validate the case for the degree of recovery expected even though the comparison conditions are not consistent. The arrows in the comments column indicate the direction expected for recovery based upon these inconsistencies. Examining these results reveals that there is nothing significant to dispute for anticipated recovery of at least 40% in both base and weld metal. Included in Table 1 are results derived from base metal experiments funded by Yankee Atomic in support of the Yankee Rowe vessel. The results for the A302B-modified surrogate plate for the lower shell course (YA-1C) were generated by Babcock and Wilcox Nuclear Services, the results for the actual upper shell course plate of A302B were developed by SCK-CEN in Belgium, and the Trino A302B results were derived from tests performed by Westinghouse. Note that the results in Table 1 (near the bottom) were obtained from a reanalysis of the Yankee Rowe A302B steel results in Reference 1, but uncertainties in fluence and limited anneal test results placed this data in the category of related data. The raw

Charpy results in Reference 9 (BR-3) were evaluated using a hyperbolic tangent curve fit to the Charpy V-notch energy data.

In terms of re-embrittlement response after annealing, data from Reference 9 were evaluated in detail and are listed in Table 3. The most applicable data represent those in which the experimental conditions were closer to the Yankee Rowe vessel in terms of operating temperature. The predicted shift is based upon the lateral shift method of estimating embrittlement after annealing^[10,11]. The results for these low temperature anneals indicate that the lateral shift method is fairly good in terms of estimating the re-embrittlement response. On average, the measured shift was 7-8°F higher than the predicted shift.

YANKEE ROWE VESSEL RE-EMBRITTEMENT ESTIMATES

Utilizing the 40% recovery estimate identified from the review of the literature and the limited results from additional tests, estimates for the recovery of the Yankee Rowe vessel were made using the lateral shift re-embrittlement method. An anneal for two weeks at a temperature near but not exceeding 650°F was assumed. In order to make these estimates for re-embrittlement, it was necessary to define embrittlement trend curves for each of the materials. Footnote A in Table 4 identifies the trend curves that were assumed for what is termed "best estimate." The exact trend curves for weld metal and the lower plate are less certain (but certainly realistic) since these materials were not evaluated directly with actual irradiated material specimens. Shift value estimates after one and three additional fuel cycles are listed and compared with the Pressurized Thermal Shock (PTS) screening criteria^[12] used in the U.S. The actual screening criteria of 270°F and 300°F were adjusted for the initial RT_{NDT} for each material and also for a one and two sigma margin (uncertainty) term equivalent to that used in Regulatory Guide 1.99, Revision 2^[13].

These projections suggest that at least one fuel cycle is possible and that up to three cycles may be achievable depending upon the magnitude of the margin term. It is felt that at least a one sigma margin term is appropriate to account for many of the uncertainties. Whether a larger margin term is needed depends upon Regulatory negotiation. The use of worst case trend curves would change the weld metal predictions significantly, but the base metal results are not much different than those listed in Table 4. In either case, it is the lower plate A302B-modified steel that is most limiting for the Yankee Rowe vessel after a thermal anneal heat treatment.

CONCLUSIONS

The results from the critical review of the past annealing data indicated that a "wet" anneal of the Yankee Rowe vessel may have been successful in reducing the degree of embrittlement to the point that the vessel could have continued to operate safely for at least one fuel cycle (and possibly more depending upon the degree of conservatism needed to satisfy regulators). The key elements for making this conclusion were: (1) there was approximately a 150°F difference between the annealing and operating temperatures for the vessel (a key parameter), (2) there was a relatively high degree of damage in the vessel prior to annealing, and (3) the Yankee Rowe vessel materials were thought to have moderate to good recovery potential (based upon limited experimental

data). For the materials in the Yankee Rowe vessel, a thermal anneal recovery of at least 40% was expected, and the lateral shift methodology for re-embrittlement after annealing was considered appropriate while maintaining at least a one sigma uncertainty similar to Regulatory Guide 1.99, Revision 2.

REFERENCES

1. Serpan, C. Z. and Hawthorne, J. R. 1967. Yankee Reactor Pressure Vessel Surveillance: Notch Ductility Performance of Vessel Steel and Maximum Service Fluence Determined from Experience During Cores II, III, and IV. Naval Research Laboratory. NRL Report 6616.
2. Macdonald, B. 1985. Post Irradiation Annealing Recovery of Commercial Pressure Vessel Steels. Effects of Radiation on Materials: Twelfth International Symposium. American Society for Testing and Materials. ASTM STP 870: pp. 972-978.
3. Potapovs, V. et al. 1968. Notch Ductility Properties of SM-1A Reactor Pressure Vessel Following the In-Place Annealing Operation. Nuclear Applications. Vol. 5, No. 6: pp. 389-409.
4. Hall, J. F. and Seman, D. J. 1973. The Effect of Post-Irradiation Annealing and Re-Irradiation on the Fracture Properties of A302B Pressure Vessel Steel. Westinghouse. WAPD-TM-1095.
5. Hawthorne, J. R. 1979. Survey of Postirradiation Heat Treatment as a Means to Mitigate Radiation Embrittlement of Reactor Vessel Steels, Naval Research Laboratory/U.S. Nuclear Regulatory Commission. NUREG/CR 0486, NRL Report 8287.
6. Mager, T. R. 1982. Feasibility of and Methodology for Thermal Annealing an Embrittled Reactor Vessel. Electric Power Research Institute. EPRI NP-2712.
7. Server, W. L. 1985. Review of In-Service Thermal Annealing of Nuclear Reactor Pressure Vessels. Effects of Radiation on Materials: Twelfth International Symposium. American Society for Testing and Materials. ASTM STP 870: pp. 979-1008.
8. Fabry, A. et al. 1985. Annealing of the BR3 Reactor Pressure Vessel. Twelfth Water Reactor Safety Research Information Meeting. U.S. Nuclear Regulatory Commission. NUREG/CP-0058, Vol. 4: pp. 144-175.
9. Hawthorne, J. R. and Hiser, A. L. 1988. Irradiation and Irradiation-Anneal-Reirradiation Studies of RPV Steels and Welds in Support of the BR-3 Reactor. Materials Engineering Associates. MEA-2218 (proprietary).
10. Server, W. L. and Taboada, A. 1985. An Approach for Estimating Post-Anneal Reirradiation Embrittlement of Reactor Vessel Steels. Proceedings of the Second International Symposium on Environmental Degradation of Materials in Nuclear Power Systems--Water Reactors, Monterey.
11. American Society for Testing and Materials. 1992. Standard Guide for In-Service Annealing of Water Cooled Nuclear Reactor Vessels. ASTM E509-92.
12. Office of the Federal Registrar. 1991. Code of Federal Regulations. 10 - Energy, Part 50.61.
13. U.S. Nuclear Regulatory Commission. 1988. Radiation Embrittlement of Reactor Vessel Materials. Regulatory Guide 1.99, Revision 2.

TABLE 1 - PERTINENT ANNEALING RECOVERY DATA FOR BASE METAL (A11 One Week Except as Noted)

<u>Material</u>	<u>T(irr), °F</u>	<u>T(ann.), °F</u>	<u>Delta T, °F</u>	<u>Initial Shift °F</u>	<u>Post-Anneal Residual Shift, °F</u>	<u>Recov., %</u>	<u>Comments</u>
<u>MOST APPLICABLE DATA</u>							
A302B-mod (PT-A/BR3)	500	650	150	240	134	44	Surrogate plate for BR3 vessel
A302B-Yankee (UPPER PLATE)	525	650 ^(A)	125	197 ^(B)	106	46	High recovery even though Delta T is low (↓), note 2 week anneal (↑)
A302B-mod (YA-1C)	500	650 ^(A)	150	243	134 ^(C)	45	Surrogate for Yankee Lower Plate, 2 week anneal (↑)
<u>OTHER RELATED DATA</u>							
SM-1A plate	430	572	142	115	45	61	Low T (irr) (↑) / low initial shift (↓), high recovery
SM-1A forging	430	572	142	195	30	85	Low T (irr) (↑) , high recovery
SM-1A forging	430	572	142	310	100	68	
A302B-Bettis, Matl. S	430	625 ^(D)	195	330	130	61	Low T (irr) (↑) / lower T (anneal) (↓) / high Delta T (↑), 9 day anneal (↑), high recovery
A302B-Bettis, Matl. U	430	625 ^(D)	195	330	155	53	
A302B-Bettis, Matl. S	430	650 ^(D)	220	320	70	78	Low T (irr) (↑) / high Delta T (↑), 9 day anneal (↑), high recov.
A302B-Bettis, Matl. U	430	650 ^(D)	220	305	90	71	
SA533B-1 (N27)	440	650	210	286	83	71	Low T (irr) (↑) / high Delta T (↑), high recovery
A302B-Yankee (UPPER PLATE)	500	640	140	296	187 ^(C)	37	Low Delta T (↓), high fluence (?), moderate recovery
A302B-Trino	500	650	150	144 ^(C)	94 ^(C)	35	Low ini. shift (↓), mod. recovery
A302B-Trino	500	650 ^(A)	150	144 ^(C)	79 ^(C)	45	

(A) Two-week anneal.

(B) Based on upper plate (525°F) trend curve for a fluence of 2.3×10^{19} using an irr. temp. correction of 1°F/°F.(C) Based upon limited number of Charpy specimens considering T_{30} only.

(D) Nine day anneal.

TABLE 2 - PERTINENT ANNEALING RECOVERY DATA FOR WELD METAL (A11 One Week)

<u>Material</u>	<u>T(irr), °F</u>	<u>T(ann.), °F</u>	<u>Delta T, °F</u>	<u>Initial Shift °F</u>	<u>Post-Anneal Residual Shift, °F</u>	<u>Recov., %</u>	<u>Comments</u>
<u>MOST APPLICABLE DATA</u>							
Linde 80 (W1/BR3)	500	650	150	290	123	58	Surrogate range for BR3 weld
Linde 80 (W-D/BR3)	500	650	150	31	172	45	Surrogate range for BR3 weld
Linde 80 (W-E/BR3)	500	650	150	276	131	53	Surrogate range for BR3 weld
<u>OTHER RELATED DATA</u>							
Non-Linde 80 (W)	550	700	150	220 ^(A)	133 ^(A)	40	High annealing T (↑), high recovery
Non-Linde 80 (W)	550	700	150	256 ^(A)	121 ^(A)	53	
Non-Linde 80 (EP24)	550	700	150	108 ^(A)	76 ^(A)	30	High annealing T (↑), low initial shift (↓), moderate recovery
Non-Linde 80 (EP24)	550	700	150	167 ^(A)	76 ^(A)	35	
Linde 80 (EP19)	550	700	150	175 ^(A)	144 ^(A)	18	High annealing T (↑), low initial shift (↓), low recovery
Linde 80 (EP19)	550	700	150	227 ^(A)	94 ^(A)	59	
Linde 80 (EP23)	550	700	150	130 ^(A)	81 ^(A)	38	High annealing T (↑), low initial shift (↓), moderate recovery
Linde 80 (EP23)	550	700	150	178 ^(A)	90 ^(A)	49	
Linde 80 (W3/BR3)	550	650	100	276	186	33	Low delta T (↓), moderate recovery
Linde 80 (W2/BR3)	550	650	100	228	168	26	Low delta T (↓), low recovery
Linde 80 (W1/BR3)	550	650	100	237	201	15	Low delta T (↓), low recovery

(A) Based upon limited number of Charpy specimens considering T_{30} only.

TABLE 3 - DATA FROM BR3 RE-EMBRITTEMENT RESULTS

Material	Irradiation Temp. (°F)	Initial Fluence (x 10 ¹⁹ n/cm ²)	Initial Shift °F	Residual Post-Anneal Shift, °F ^(A)	Additional Fluence (x 10 ¹⁹ n/cm ²)	Re-Embrittlement Trend	
						Predicted Lateral Shift (°F)	Measured Shift (°F)
<u>MOST APPLICABLE DATA</u>							
W1	500	3.50	293	176	0.58	225	241
W2	500	3.36	283	170	0.58	218	228
W2	500	3.50	285	171	0.58	219	228
W3	500	3.50	406	244	0.58	311	274
W-D	500	3.24	321	193	0.54	245	279
W-E	500	3.36	285	171	0.58	220	224
PT-A	500	3.24	247	148	0.54	189	205
<u>OTHER RELATED DATA</u>							
W-D	<500 ^(B)	2.90	314	188	1.80	298	367
PT-A	<500 ^(B)	2.90	242	145	1.80	230	255

^(A) Based upon a 40% recovery for these 500°F irradiation and 650°F anneal data sets (150°F differential driving force)
^(B) For the re-irradiation portion, the temperature was nearer 480°F.

TABLE 4 - ESTIMATES FOR RE-EMBRITTEMENT FOR YANKEE ROWE VESSEL (Best Estimate Trend Curves)

Material	Initial Shift, Delta T _{IRR} (°F) ^A	Shift Post Anneal, Delta T _{ANNEAL} (°F) ^B	Shift After 1 Cycle, Delta T _{RF} (°F) ^C	Shift After 3 Cycles, Delta T _{RF} (°F) ^C	Screening Criteria, Delta T _{SC} (°F) ^D
Circ. weld	287	172	195	225	<u>262</u> (300-10-28) / <u>234</u> (300-10-56)
Axial welds	252	151	169	194	<u>232</u> (270-10-28) / <u>204</u> (270-10-56)
Upper plate (A302B)	212	127	143	164	<u>223</u> (270-30-17) / <u>206</u> (270-30-34)
Lower plate (A302B-mod)	278	167	193	222	<u>223</u> (270-30-17) / <u>206</u> (270-30-34)

^(A) Trend Curves Assume:

$$\begin{aligned} \text{Welds: } \Delta RT_{\text{NDT}} &= 193 f^{(0.25 - 0.1 \text{ LOG } f)} + 50 && (.25 \text{ Cu}/.70 \text{ Ni}) \\ \text{Upper plate: } \Delta RT_{\text{NDT}} &= 171 f^{(0.28 - 0.1 \text{ LOG } f)} \\ \text{Lower plate: } \Delta RT_{\text{NDT}} &= 151 f^{(0.28 - 0.1 \text{ LOG } f)} + 43 + 50 \end{aligned}$$

Plate trend curves used all available data points (fluences <6 x 10¹⁹ n/cm²) for upper plate, and also for determining the lower plate sensitivity factor.

Maximum fluences at Planned Shutdown:

$$\begin{aligned} \text{Circ. welds: } f &= 2.33 \times 10^{19} \text{ n/cm}^2 \\ \text{Axial welds: } f &= 1.19 \times 10^{19} \text{ n/cm}^2 \\ \text{Upper plate: } f &= 2.42 \times 10^{19} \text{ n/cm}^2 \\ \text{Lower plate: } f &= 2.33 \times 10^{19} \text{ n/cm}^2 \end{aligned}$$

^(B) Using 40% recovery for a 650°F/2 week anneal

^(C) Using lateral shift method and per cycle fluences for additional cycles:

$$\begin{aligned} \text{Circ. welds: } &0.152 \times 10^{19} \text{ n/cm}^2 \\ \text{Axial welds: } &0.078 \times 10^{19} \text{ n/cm}^2 \\ \text{Upper plate: } &0.158 \times 10^{19} \text{ n/cm}^2 \\ \text{Lower plate: } &0.152 \times 10^{19} \text{ n/cm}^2 \end{aligned}$$

^(D) PTS screening limits adjusted for initial RT_{NDT} and a 1 sigma and 2 sigma margin adjustment, respectively (for plates sigma = 17°F and for welds sigma = 28°F per Regulatory Guide 1.99, Revision 2).