ORNL/LTR-2011/351

OAK RIDGE NATIONAL LABORATORY MANAGED BY UT-BATTELLE FOR THE DEPARTMENT OF ENERGY

# Reactor Pressure Vessel Task of Light Water Reactor Sustainability Program: Initial Assessment of Thermal Annealing Needs and Challenges

# September 2011

Prepared by

R.K. Nanstad, Oak Ridge National Laboratory

and

W. L. Server, ATI Consulting



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Light Water Reactor Sustainability

# **Reactor Pressure Vessel Task of Light Water Reactor Sustainability Program: Initial Assessment of Thermal Annealing Needs and Challenges**

**R.K. Nanstad** Materials Science and Technology Division Oak Ridge National Laboratory

and

#### W. L. Server ATI Consulting

Date Published: September 2011

Prepared under the direction of the U.S. Department of Energy Office of Nuclear Energy Light Water Reactor Sustainability Materials Aging and Degradation Pathway

Prepared by OAK RIDGE NATIONAL LABORATORY Oak Ridge, Tennessee 37831-6283 managed by UT-BATTELLE, LLC for the U.S. DEPARTMENT OF ENERGY under contract DE-AC05-00OR22725 This page intentionally left blank

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## ACKNOWLEDGMENTS

This research was sponsored by the U.S. Department of Energy, Office of Nuclear Energy, for the Light Water Reactor Sustainability Research and Development effort. The authors extend their appreciation to Dr. Jeremy Busby for programmatic support.

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#### 1. INTRODUCTION

The most life-limiting structural component in light-water reactors (LWR) is the reactor pressure vessel (RPV) because replacement of the RPV is not considered a viable option at this time. LWR licenses are now being extended from 40y to 60y by the U.S. Nuclear Regulatory Commission (NRC) with intentions to extend licenses to 80y and beyond. The RPV materials exhibit varying degrees of sensitivity to irradiation-induced embrittlement (decreased toughness), as shown in Fig. 1.1, and extending operation from 40y to 80y implies a doubling of the neutron exposure for the RPV. Thus, for the RPVs of pressurized water reactors (PWRs) expected to experience neutron fluences from 1 to  $5 \times 10^{19}$  n/cm<sup>2</sup> (>1 MeV) after 40y, the exposures will be 2 to  $10 \times 10^{19}$  n/cm<sup>2</sup> after 80y. Additionally, because the recent pressurized thermal shock (PTS) re-evaluation project has resulted in lower average failure probabilities for PWRs [1], many plants will increase their operating power level which will further increase the neutron flux and the resultant fluence [2]. Even for normal start-up and cool-down transients, the coolant-pressure-temperature (P-T) curve must be below the corresponding stress (from pressure) that could cause fracture for an assumed very large crack size. Fig. 1.2 shows a schematic depiction of a P-T operating envelope progressively decreased by irradiation embrittlement ( $\Delta RT_{NDT}$ ) of a sensitive RPV steel [3]. Since the power reactor surveillance database contains only sparse data higher than  $3 \times 10^{19}$  n/cm<sup>2</sup>, the existing embrittlement models, ([for example, the Eason, Odette, Nanstad, Yamamoto (EONY) model in reference 4], are inadequate as predictive tools to those high fluence levels. To obtain data at the high fluences for life extension will require either very long term surveillance data (for which material is now in short supply), or through the use of test reactor experiments which use high neutron fluxes.



Figure 1.1. Irradiation-induced embrittlement of RPV materials is dependent on many factors, with chemical composition being the dominant factor.



Figure 1.2. Effects of radiation embrittlement on restriction of the operating envelope for an RPV with a radiation-sensitive steel.

Our current understanding of radiation damage mechanisms suggests that it is not appropriate to use highly accelerated test reactor data directly to predict high fluence behavior for RPV operating conditions. Moreover, there is now experimental evidence that phases rich in Ni and Mn do form in irradiated low Cu steels [5]. Because these phases may require a small degree of Cu precipitation to catalyze their nucleation, they may not contribute to hardening and embrittlement until relatively high fluences. The delayed embrittlement caused by these so-called "late-blooming phases" (LBP) may produce an effect that could have serious implications to RPV life extension. As discussed in [2,5], it is important to understand and quantify the composition-flux-fluence-temperature regime in which they evolve, and develop a better quantitative description of their contribution to embrittlement. The potential for late blooming phases emerging in some composition-fluence-temperature-flux regimes could result in severe underestimates of shifts based on current models by up to 50°C or more [2,5]. The mechanisms that cause irradiation-induced embrittlement of RPV steels are discussed in [2,4,5] and will not be discussed further here, except in the context of the mechanisms that take place during the thermal annealing process which are inextricably linked to those which cause the embrittlement.

Various options are possible to mitigate the effects of irradiation embrittlement on the RPV: (1) fuel management schemes can be used to reduce the neutron flux, which reduces the fluence and, therefore, the embrittlement; (2) shielding of critical areas with, e.g., stainless steel can reduce the flux; (3) the emergency core cooling system (ECCS) water can be heated to reduce the thermal shock effects during a PTS event: (4) the RPV could be replaced; (5) various analytical methods, such as the alternative PTS rule in *Title 10, U.S. Code of Federal Regulations, Part 50.61a* (10CFR50.61a) [6] can potentially be used to allow for operation with RT<sub>PTS</sub> values above the screening criteria; (6) mechanically prestressing the beltline region of the RPV by compressive loading with structural bands [7]; (7) if a weld is the critical area for embrittlement, replace the weld with more resistant material [8]; (8) thermal annealing to recover fracture toughness of the RPV materials. Fuel

management can have only a slight effect on reducing neutron flux, while shielding, although somewhat effective, is expensive. Conditioning of the ECCS water relates only to thermal shock situations, and replacement of the RPV is not considered practicable at this time. The analytical options, although effective, would likely offer relatively short term relief, while the prestressing and weld replacement concepts have not been thoroughly evaluated. Many of these options are discussed by Planman, Pelli and Torronen [9].

Recovery of the material toughness through thermal annealing is one method of increasing safety margins of the RPV. Thermal annealing involves heating the RPV beltline materials to temperatures ~ 50 to 200°C above the normal operating temperature for about one week, with the amount of recovery increasing with increasing annealing temperature. Two different procedures can be used to perform the thermal anneal, a wet anneal or a dry anneal. A wet anneal is performed with cooling water remaining in the RPV and is limited to the RPV design temperature of 343°C. A dry anneal requires removal of the cooling water and internal components and would normally be performed at temperatures in the range of 430-500°C. If thermal annealing is considered, then the post-annealing reirradiation response of the steel must also be evaluated. 10CFR50 specifies thermal annealing as a method for recovering the fracture toughness and refers to *Regulatory Guide 1.162* (RG 1.162) [10]. RG 1.162 also provides guidance for determining the amount of recovery, the re-embrittlement trend (assumed to occur at the same rate as in the irradiated case) and for establishing post-anneal material properties.

This report provides an introduction to the subject of thermal annealing of reactor pressure vessels and materials, including a summary of experience with actual thermal annealing of power reactors. The report is prepared in satisfaction of Milestone L-110R040602 Level M3 #M3L110R04060203 "Complete initial assessment of thermal annealing needs and challenges."

## 2. SUMMARY OF THERMAL ANNEALING EXPERIENCE

### 2.1 THERMAL ANNEALING OF IRRADIATED MATERIALS

Thermal annealing has been recognized as a method to mitigate the embrittlement and recover the fracture toughness of the RPV materials for decades. Thermal annealing is not a traditional "annealing" heat treatment; a much lower temperature (typically  $< 500^{\circ}$ C) and about a one week time period (168 h) are typically applied. Annealing recovery is a function of irradiation temperature and flux, copper and other elemental contents, annealing temperature and time, with annealing temperature being the dominant factor in recovery of fracture toughness.

A key aspect of thermal annealing is the rate of reembrittlement that occurs following the annealing treatment. Fig. 2.1 shows schematic depictions of two different procedures to predict the post-annealing reembrittlement of the material. The lateral method assumes the reembrittlement will occur at the same rate that occurs at the beginning of the original irradiation embrittlement, while the vertical shift method assumes the reembrittlement will occur at the rate of the original embrittlement from the point of annealing.

There are many examples of thermal annealing results on U.S. PWR steels, with one example for a high-copper weld from the Midland Unit 1 reactor shown in Figs. 2.2 and 2.3 [11]; this reactor did not operate and material was removed from the RPV for various fracture mechanics and irradiation effects studies by the NRC-sponsored Heavy-Section Steel Irradiation (HSSI) Program [12, 13]. Fig. 2.2 shows the beneficial effect of the one week high temperature annealing at 454°C (850°F) compared with that for a one week anneal at 343°C (650°F). The higher temperature anneal

resulted in a Charpy 41-J transition temperature recovery of about 80%, compared with about 50% for the lower temperature anneal. For these experiments, the materials were irradiated in a test reactor at a flux of about  $8 \times 10^{11}$  n/cm<sup>2</sup>/s (>1 MeV). Fig. 2.3 shows that the high temperature anneal provided a fracture toughness recovery of over 90%, somewhat greater than the 80% recovery of the Charpy impact results.



Fig. 2.1. Schematic depictions of two methods, (a) lateral shift, and (b) vertical shift, for predicting post-annealing reembrittlement of RPV materials.



Figure 2.2. Effects of thermal annealing at 343 and 454°C on the high copper Midland Unit 1 RPV beltline weld.



Figure 2.3. Effect of thermal annealing at 454°C/168h on the fracture toughness of the high-copper Midland Unit 1 RPV beltline weld.

As with irradiation effects mechanisms that cause RPV material embrittlement, the mechanisms of thermal annealing that result in recovery of the material toughness are quite complicated. For a copper-bearing material such as the example shown for the Midland weld, irradiation of the material caused the formation of copper-rich precipitates (CRPs), which are about 1-2nm in diameter, and various forms of matrix damage (e.g., dislocation loops). The high-temperature thermal annealing treatment dissolves most of the small irradiation-induced defect clusters and complexes, but the dissolution of the CRPs actually results in an increase of their size to 3-5nm, slightly reduces the volume fraction and significantly reduces the number density. An example of this is shown in Fig. 2.4, results from small-angle neutron scattering (SANS) studies of the Midland beltline weld [14]. The results of these dissolution mechanisms is a significant decrease in hardening (i.e., yield strength), and recovery of toughness (i.e., decreases in Charpy impact and fracture toughness transition temperatures).



Figure 2.4. Evolution of scattering centers in the irradiated Midland beltline weld after annealing at 343, 399, and 454°C for 168h.

The available data on thermal annealing of western steels is relatively robust and well documented. However, there are only limited results available regarding the re-embrittlement rate, especially of U.S. RPV steels. The HSSI Program, in a cooperative study with Paul Scherrer Institute (PSI) in Switzerland, performed thermal annealing, reirradiation, and re-annealing (IARA) experiments with a plate of A533 grade B class 1 RPV steel designated JRQ. The JRQ heat is essentially a reference heat of RPV steel for the IAEA and has been used for many different irradiation studies. The JRQ plate has a copper content of 0.16 wt% and nickel content of 0.60 wt%. Fig. 2.5 shows Charpy impact results for JRQ steel following initial irradiation to  $0.85 \times 10^{19}$  n/cm<sup>2</sup>, thermal annealing at 460°C/18h, reirradiation to  $0.85 \times 10^{19}$ , and re-annealing at 460°C/168h [15]. These results show that re-annealing after two irradiation series provides almost full recovery of Charpy impact toughness. It is interesting that the Charpy upper-shelf energy is increased following the re-annealing procedure; this also was observed after the first thermal annealing procedure.



Figure 2.5. Re-annealing of JRQ steel following a re-irradiation cycle results in nearly full Charpy toughness recovery.

Another example for a high-copper weld is for HSSI Weld 73W, a submerged-arc weld that used Linde91 welding flux and has 0.31 wt% copper, 0.60 wt% nickel, and 0.005 wt% phosphorus [15]. HSSI Weld 73W was irradiated to  $1.8 \times 10^{19} \text{ n/cm}^2$  (>1 MeV) at 288°C, annealed at 454°C/168h, then reirradiated (IAR) to total fluences of 2.3, 2.7, 4.8, and  $7.1 \times 10^{19} \text{ n/cm}^2$ , with the highest fluence set then re-annealed at 454°C/168h (IARA). Fig. 2.6 provides a summary of the CVN impact results and shows that the re-irradiation rate after annealing is much smaller than predicted by the Lateral Shift method but slightly higher than by the Vertical Shift method [16, 17]. Other observations from these series of tests were that: (1) the upper shelf energies after annealing for all re-irradiation conditions were significantly higher than that for the unirradiated condition, and (2) CVN impact energy *vs* temperature curves for IAR tests at 4.8 and  $7.1'10^{19} \text{ n/cm}^2$  overlap, indicating the possibility that irradiation embrittlement due to CRPs has reached a plateau due to decreased availability of radiation-sensitive constituents (e.g. matrix Cu).



Figure 2.6. Results of Charpy impact tests for HSSI Weld 73W following various indicated stages of irradiation, annealing, reirradiation, and reannealing. Irradiation and reirradiation were performed at 288°C and each annealing step at 454°C/168h.

Atom probe tomography was performed for the annealing and reirradiation experiments with Weld 73W as well [18], and are summarized as follows:

- 1. In the irradiated condition (1.8×10<sup>19</sup> n/cm2), a high number density of ultrafine Cu-Mn-, Ni- and Si-enriched precipitates were observed.
- 2. In the irradiated/annealed condition (1.8×10<sup>19</sup> n/cm2 + 454°C/168h), the number density of Cu-enriched precipitates had decreased more than an order of magnitude, and their size (radius) had increased by about 60%.
- 3. In the irradiated/annealed/reirradiated condition  $(1.8 \times 10^{19} \text{ n/cm2} + 454^{\circ}\text{C}/168\text{h} + 0.8 \times 10^{19} \text{ n/cm2})$ , the size (radius) of the copper-enriched precipitates were about 40% larger than in the IA condition. Additionally, solute segregation of Cu, Ni, Mn, Si, and P to dislocations was observed.
- 4. These observations support many other studies that show synergism of copper, nickel, manganese, silicon, and phosphorus in increasing radiation sensitivity of RPV steels.

An issue of investigation regarding effects of thermal annealing and reirradiation on the microstructural changes in RPV steels involves the potential for temper embrittlement resulting in significant intergranular fracture (IGF). A study by McElroy, et. al. [19] clearly demonstrated the embrittlement susceptibility of grain-coarsened RPV steels, with the notable observation that the study was performed to intentionally heat treat eleven different steels with temper embrittlement procedures to assess susceptibility to the phenomenon. It is important to note that very coarse grains

(e.g., ASTM 0 to 1) were obtained and, as stated by Nanstad, et. al. [20], such coarse grains may not truly represent the coarse grain microstructure in RPV weld heat-affected zones. Given the concerns about thermal annealing effects, Nanstad, et. al. performed similar studies with five commercial RPV steels and obtained similar results with two of the steels, thus demonstrating the susceptibility of these U.S. commercial steels to temper embrittlement, providing motivation for further study with more prototypical microstructures. Specialized procedures were used to produce a microstructure representative of that in a prototypic submerged-arc weldment heat-affected-zone of an RPV steel (ASTM A302 grade B, modified) with a relatively low phosphorus content (0.007 wt%). The thermal history used was prototypic of submerged-arc welds, except that a rapid cool following the post-weld heat treatment was imposed [20]. The heat treated specimens were irradiated at 288°C (550°F) to  $1.0 \times 10^{19}$  n/cm<sup>2</sup> (>1MeV), with Charpy impact testing of the irradiated specimens exhibiting only a 41-J transition temperature shift of 22°C. Thermal annealing at 460°C/168h produced no recovery of the toughness. In the irradiated condition, the specimens exhibited about 10 to 20% IGF, but specimens exhibited more than 75% IGF following thermal annealing, as shown in Figs. 2.7 (a) and (b). It was recommended in [20] that a subsequent study be performed with the same material given a prototypic slow-cool following the PWHT, but the HSSI Program was terminated before those experiments could be completed.



Figure 2.7. Scanning electron fractographs of Gleeble austenitized/PWHT modified A302B steel charpy impact specimens following (a) irradiation at 288C to 11019 n/cm2, and (b) the same irradiation exposure followed by thermal annealing at 460C for 168h. In (b), the notch of the specimen appears at the right side of the fractograph.

#### 2.2. THERMAL ANNEALING OF COMMERCIAL POWER REACTORS

Title 10CFR 50.66 [6] permits thermal annealing of light-water reactors, but requires a plan conducting the thermal annealing be submitted at least three years before the fracture toughness criteria are predicted to be exceeded. It refers to RG 1.162 [10] which describes the format and content of an acceptable Thermal Annealing Report and addresses the metallurgical and engineering issues that need to be addressed in an application to perform a thermal annealing. As mentioned in the Introduction, RG 1.162 [10] provides guidelines for determining the percent recovery and the re-embrittlement trend and for establishing the post-anneal reference temperature and Charpy upper-shelf energy values. The re-embrittlement trend in RG 1.162 is based on the "lateral" shift procedure in which re-embrittlement of the steel is assumed to occur at the same rate as in the irradiated case, as shown in Fig. 2.1.

As mentioned in the Introduction, two different procedures can be used to perform the thermal anneal

of an RPV, a wet anneal or a dry anneal. A wet anneal is performed with cooling water remaining in the RPV and is limited to the RPV design temperature of 343°C. Annealing at this temperature results in relatively low amounts of recovery of the irradiation-induced transition shift, e.g., 10 to 30%. A dry anneal requires removal of the cooling water and internal components and would normally be performed at temperatures in the range of 430-500°C. Different heating methods are possible for dry annealing: electric resistance heaters, indirect gas-fired heat exchanger, etc. A dry anneal at such temperatures typically results in recoveries of at least 80%.

Thermal annealing of operating nuclear reactors has been performed at least 16 times, once in the USA, once in Belgium and 14 times at Russian-designed VVER-440 plants. The first annealing operation was performed on the U.S. Army SM-1A (SM designates Stationary Medium Power, and 1A indicates the first field plant of this type), nuclear reactor in Fort Greely, Alaska in August 1967. The SM-1A nuclear power plant was developed as a result of studies made in 1952 by the U.S. Army Corps of Engineers [21]; these studies included a design study for a nuclear power plant to meet military specifications that was performed at the Oak Ridge National Laboratory (ORNL). As a result of that study, the Army recommended that a prototype nuclear power plant be constructed in the United States and the Army Nuclear Power Program was established in 1954 to coordinate Atomic Energy Commission (AEC) and Department of Defense (DOD) activities. Early in 1954, proposals from 33 major industrial firms were invited for design, construction, and test operations of a prototype nuclear power plant. Out of 18 received proposals, the contract was awarded to the American Locomotive Co. (ALCO) in December 1954. The site for this first plant was located at Fort Belvoir, Virginia because it was within the U.S. Army Engineer Research and Development Laboratories area. The design of the plant, designated SM-1, was conceived by ORNL and detailed by ALCO. The design concept was that of a pressurized water reactor (PWR) operating at a thermal power of 10MW, with net electrical power of 2MW. The SM-1 went critical in April 1957.

Based on the SM-1 concept, design studies were made in 1956 for eventual installation of a similar plant at Fort Greely, Alaska, to accommodate the Army's need for electrical power and steam heating in an expanding remote area where delivery of more common fuels were difficult and expensive [21]. Thus, the contract for construction of the SM-1A was awarded to Peter Keiwit Sons' Construction Co., with ALCO Products, Inc., as nuclear subcontractor [21]. The SM-1A went critical in March 1962, was a PWR operating at 1200 psia, with coolant inlet and outlet temperatures of 221°C (430) and 232°C (450F), respectively; the reactor produced 20MW thermal power with gross ouput of 2.5MW electric power and 36,000 lbs/h of post heating steam [21]. It used 93% enriched U235 fuel with stainless steel cladding, and the RPV was constructed of A350-LF1 (modified), a low-alloy steel with nickel content of about 1.7 wt% [21]. Significantly, the SM-1A RPV operated at 221°C (430°F), a very low temperature compared with typical commercial PWRs that operate at about 288°C (550°F). Because the A350-LF1 steel has relatively low resistance to irradiation embrittlement due to its nickel content, such a low irradiation temperature exacerbated that low resistance and resulted in quite rapid embrittlement of the RPV steel [22]. Unfortunately, the reactor's compact design did not allow for surveillance capsules to be located in the beltline region, but test reactor irradiations of a duplicate forging showed a Charpy impact 41-J transition temperature shift of 211°C (380°F) at  $2.6 \times 10^{19}$  n/cm<sup>2</sup> (>1 MeV) [23], which resulted in a transition temperature for the RPV of 149°C (300°F), the upper limit designated by the Army for the RPV. Thus, a thermal annealing procedure was planned and executed on the SM-1A RPV, with the use of primary cooling and nuclear heat only as the means of heating the RPV to 307°C (585°F) for one week (168h). The actual anneal was performed during August of 1967 with conditions of 28h at 293°C (560°F) and 144h at 300°C(572°F). The resultant embrittlement recovery for the A350-LF1 forging specimens was 70 to 80% as measured by the Charpy 41-J transition temperature. Fig. 2.8 shows NRL results for the duplicate forging irradiated and annealed results under similar conditions as imposed on the SM-

1A RPV, with a transition temperature recovery of about 70% [23, 24]. The irradiation experiments were performed at the Low-Intensity Test Reactor (LITR) at ORNL. Fig. 2.9 shows results of irradiation, annealing, and reirradiation for the same steel, indicating that subsequent post-annealing reirradiation was predicted to follow the lateral shift response as shown in Fig. 2.1 [23, 24, 25]. The SM-1A reactor was shut down in 1972, but provided a significant wealth of information regarding irradiation effects and irradiation-induced embrittlement mitigation by thermal annealing.



Figure 2.8. Charpy impact transition temperature results for the SM-1A duplicate ring forging specimens irradiated in the LITR and thermally annealed as indicated.



Figure 2.9. Projected reirradiation behavior of the SM-1A RPV following the in-place annealing operation.

Following the experience with thermal annealing in the Army Nuclear Power Program, the Belgian Reactor 3 (BR3) was the first commercial power reactor to be annealed, with a wet anneal being performed in 1984 using primary pump heat [26]. The BR3 was the first PWR in Western Europe and went critical in August 1962, with the annealing procedure performed 22y later. The BR-3 operated at 260°C (500°F) and the thermal annealing operation was performed at 343°C (650°F), the RPV design temperature, with the Charpy impact transition temperature recovery estimated to be 50% [26].

Following the annealing of the BR3 reactor, many thermal annealing treatments were performed on Russian-designed VVER-440 reactors. Table 2.1 shows the name of each reactor, the year of the annealing, the annealing temperature and time, and whether the RPV was clad with austenitic steel on the inside surface [26]. As shown in the table, Novovoronezh-3 was given two annealing treatments, the first in 1987 and the second one in 1991. The last one in the table, Loviisa-1, is located in Finland. The second annealing process at the Novovoronezh-3in Russia was witnessed by a U.S. delegation composed of U.S. Nuclear Regulatory Commission staff, industry, and research groups (e.g. ORNL). Thus, there is experience with annealing and reoperation of nuclear plants to give credence to application of the technology to U.S. plants [27].

For some of the VVER-440 RPVs given thermal annealing treatments, e.g., Kozloduy 2, RPV material was machined from the inside surface using a special extraction procedure to remove so-called "boat samples." These samples were used to verify the status of the RPV material in the thermally annealed condition in cases where surveillance samples were not available. The boat samples were used to obtain chemical composition and to machine sub-size Charpy impact specimens for transition temperature determination [28].

Reactor		Year	Temperature/time (°C/h)	SS clad
Novovoronezh 3		1987	430 ± 20 °C / 150 h	No
Armenia 1		1988	450 + 50 °C / 150 h	No
Greifswald (Nord 1)		1988	475 – 10 °C / 150 h	No
Kola 1		1989	475 °C / 150 h	No
Kola 2		1989	475 °C / 150 h	No
Kozloduy 1		1989	475 °C / 150 h	No
Kozloduy 3		1989	475 °C / 150 h	Yes
Greifswald 2 (Nord 2)		1990	475 – 10 °C / 150 h	No
Greifswald 3 (Nord 3)		1990	475 °C / 150 h	Yes
Novovoronezh 3	(re-	1991	475 °C / 150 h	No
annealing)				
Kozloduy 2		1992	475 °C / 150 h	No
J. Bohunice V-1/2		1993	475 – 503 °C/ 160 h	Yes
J. Bohunice V-1/1		1993	475 –496 °C / 168 h	Yes
Loviisa 1		1996	475 °C / 100 h	yes

Table 2.1 Thermal annealing performance of VVER-440 type RPVs.

In fact, an Annealing Demonstration Project (ADP) funded jointly by the U.S. Department of Energy and the nuclear industry was performed at the uncompleted Marble Hill nuclear plant in Indiana in 1996/1997 and an independent evaluation concluded that "Successful completion of the ADP has demonstrated that functional requirements for in-place annealing of a U.S. RPV can be met using existing equipment and procedures." [29, 30] The Marble Hill RPV was a four-loop PWR with nozzle supports and designed by Westinghouse. The Marble Hill plant was a partially completed plant but the vessel was in place which allowed for a prototypic annealing demonstration to be executed. The objectives of the demonstration were:

- 1. Demonstrate engineering feasibility of annealing system,
- 2. Determine component thermal/stress response,
- 3. Determine RPV dimensional stability following anneal,
- 4. Biological shield wall temperature,
- 5. Magnitude of thermally induced stresses in nozzle region (satisfy ASME Code Case N-557),
- 6. Benchmark 3-D stress analysis models,
- 7. Provide information to resolve regulatory concerns,
- 8. Provide realistic cost data regarding equipment costs, modeling, etc.

The method used was a dry annealing procedure with an indirect gas-fired method through a heat exchanger. The RPV was instrumented with strain gages and thermocouples to assess strain levels and temperatures over the entire RPV, including nozzles, during and after the annealing operation. Overall, the results were successful in showing that annealing could be performed with reasonable assurance of low thermally-induced strains in the RPV and an adequately uniform temperature distribution [29, 30].

In addition to the Marble Hill demonstration, another demonstration was planned to be conducted with the Midland RPV, a skirt-supported Babcock & Wilcox-design vessel. The plan included use of

the electric resistance heating method developed and used a number of times for annealing of VVER-440 reactors by a Russian firm. The electric resistance heater had been fabricated and tested and the project was approximately 50 percent complete when DOE funding was eliminated and the demonstration was never completed.

The Yankee Rowe reactor was planning a thermal annealing procedure using the wet anneal process at 343°C (650°F) to recover determined RPV embrittlement but the plant was shut down before the procedure could be implemented [31].

The Palisades Reactor planned to anneal in 1998 to recover properties and continue operation to at least 2011, based on surveillance and fluence data. The Palisades plant had developed a supplemental surveillance program to assess material recovery and re-embrittlement trends for all beltline welds and the surveillance plate material, but the annealing was canceled due to revised fluence estimates that showed longer operation of the reactor could be accommodated.

## 2.3 U.S. REGULATIONS AND GUIDANCE RELATED TO THERMAL ANNEALING

As mentioned in the Introduction, 10CFR50.66 specifies thermal annealing (the Annealing Rule) as a method for recovering the fracture toughness and refers to Regulatory Guide 1.162 (RG 1.162) [10]. RG 1.162 also provides guidance for determining the amount of recovery, the reembrittlement trend (assumed to occur at the same rate as in the irradiated case) and for establishing post-anneal material properties. The Guide also describes the format and content of an acceptable Thermal Annealing Report. Also, the Annealing Rule and RG1.162 refer to NUREG/CR-6327 [32] for guidance regarding a predictive model for annealing recovery utilizing microhardness and CVN data to cover a broad range of annealing conditions. The model incorporates annealing time, annealing temperature, and the neutron flux on the portion of the RPV of interest. For annealing temperatures below 427°C (800°F), the model in NUREG/CR-6327 indicates a significant effect of neutron flux on recovery, with the amount of recovery decreasing with decreasing temperature as shown in Fig. 2.10. For a prototypic dry thermal annealing operation, the target annealing temperature is about 454°C (850°F), making neutron flux insignificant according to the predictive model. Further inspections of the curves in Fig. 2.10 indicate that the predicted recovery using a wet anneal, at 343°C (650°F), is very low in the flux range (i.e., 10<sup>10</sup> n/cm<sup>2</sup>) typical of PWRs. There are also data from a project on VVER-440 steels that indicated no flux effects (test reactor vs surveillance) for annealing near 460°C [33].

ASTM Standard Guide E509 also provides expanded guidance on thermal annealing and associated supplemental material surveillance programs. This standard guide was revised in 1997, in 2003, and reapproved in 2008 [34]. In 2002, ASTM Standard Practice E185 was split into two new practices, E185-02 on Design of Surveillance Programs [35], and E2215-02 on Testing of Surveillance Capsules [36]. Both of these standard practices emphasize the use of fracture toughness testing using the Master Curve approach of ASTM E1921 [37]. Other relevant standards are ASTM E636 on Supplemental Test Techniques [38] and ASTM E1253 on Charpy Specimen Reconstitution [39]. There is also an ASME Code Case, N-557, "In-Place Dry Annealing of a PWR Nuclear Reactor Vessel (Section XI, Division 1)." [40] This Code Case provides Code guidance for assuring design conformance after performing a thermal anneal heat treatment:

- Limits the magnitude of thermally induced stresses in nozzle region,
- Effectively limits the maximum temperature of annealing to 505°C, and
- Was passed in 1995 in anticipation of a Palisades NPP thermal anneal.

The technical basis for Code Case N-557 was published by EPRI in TR-106967 [41].



Figure 2.10. The thermal annealing predictive model in NUREG-CR-6327 includes a strong effect of neutron flux for annealing temperatures below 427°C (800°F).

## 3. ISSUES AND RECOMMENDATIONS FOR THERMAL ANNEALING

Annealing of LWR RPVs is a technically viable and, at least, a partially demonstrated technology. However, additional efforts will be required to gain acceptance within the nuclear power industry. A number of issues have been identified regarding the potential use of thermal annealing to recover the fracture toughness of U.S. RPVs that may experience irradiation-induced embrittlement significant enough to threaten structural integrity.

- 1. There are no applicable annealing data for irradiation effects at the high dose levels that RPVs will experience with 80y of operation, e.g., a fluence of  $\sim 1 \times 10^{20}$  n/cm<sup>2</sup>. There is a reasonable amount of annealing data for U.S. RPV steels applicable to 40y of operation, as evidenced by the existence of RG 1.162, but not 80y.
- 2. Significantly, because a crucial aspect of an annealing operation is the behavior of the RPV during reirradiation, the amount of post-annealing reirradiation data is sparse for the 40y scenario and non-existent for 80y. Thus, the uncertainties associated with the reirradiation response are high. Moreover, the development if improved models for the reirradiation condition are needed and the use of fracture toughness data instead of Charpy impact data are preferable.
- 3. The microstructural processes involved in damage recovery are reasonably understood, but those for the reirradiation response are not as well understood and have had only cursory examination to date. Understanding the underlying physical mechanisms involved in post-irradiation annealing and, especially, re-irradiation embrittlement will be key to reducing

uncertainties regarding fracture toughness recovery and reembrittlement.

- 4. Based on NRC Regulations and guidance, e.g., RG 1.162, there is evidence of a flux effect (dose rate effect) on annealing recovery at low annealing temperatures (less than 427°C). If consideration is given by the U.S. nuclear industry to thermal annealing in that temperature range, substantial additional information is required regarding such effects on the annealing recovery as well as the reirradiation rate. This is also noted in ASTM E509.
- 5. Although significant intergranular fracture (IGF) has not been observed in irradiated U.S. RPV steels, these steels have been demonstrated to be sensitive to temper embrittlement under certain circumstances and IGF has been observed in the heat-affected-zone region of some steels in the post-annealed condition after irradiation to a fluence about  $1 \times 10^{19}$  n/cm<sup>2</sup>, giving some concern regarding behavior after irradiation to  $1 \times 10^{20}$ .
- 6. Engineering considerations for thermal annealing may be the least problematic aspect of the technology given that many previous procedures have been applied to commercial reactors. Although those annealing operations were performed in other countries, the U.S. does have the benefit of the joint DOE/Industry Annealing Demonstration Project. Nonetheless, various engineering considerations must be addressed, e.g., potential degrading effects of the high temperature exposure on other parts of the structure, etc., and such issues will likely differ with different reactor designs. It is important to note that, based on guidance in ASTM E 509 and the applicable ASME Code Case, the annealing operation must be performed to minimize thermally induced stresses in nozzle region, which effectively limits the maximum temperature of annealing to 505°C.
- 7. In addition to the research needs discussed, development of a surveillance program for the post-annealed operation is needed. This issue could be problematic as the availability of materials could be very limited or even nonexistent.

## 4. SUMMARY AND CONCLUSIONS

Of the many significant issues discussed for RPVs, the issue considered to have the most impact on the current regulatory process is that associated with effects of neutron irradiation on RPV steels at high fluence, for long irradiation times, and as affected by neutron flux. It is clear that embrittlement of RPV steels is a critical issue that may limit LWR plant life extension. The primary objective of the LWRSP RPV task is to develop robust predictions of transition temperature shifts (TTS) at high fluence ( $\phi$ t) to at least 10<sup>20</sup> n/cm<sup>2</sup> (>1 MeV) pertinent to plant operation of some pressurized water reactors (PWR) for 80 full power years. The RPV task is a participant in the UCSB ATR-2 irradiation experiment that is underway at the ATR in INL under a NEUP from DOE. We have worked with various organizations to obtain commercial surveillance materials that are now included in the ATR-2 experiment. In addition to the vast amount of post-irradiation testing that will be done for the ATR-2 experiment, some selected thermal annealing experiments will also be performed. It is possible that some amount of annealed material from the ATR-2 experiment could be included in a subsequent reirradiation experiment. Additionally, thermal annealing of other previously irradiated materials, such as those from the International Atomic Energy Agency program on attenuation (this program includes specimens irradiated to relatively high fluence), could be very useful in such studies. This concept also applies to the potential use of irradiated surveillance specimens from commercial reactors.

Despite the technical success of the Marble Hill demonstration and the existence of RG 1.162, as discussed in this brief report, thermal annealing may not be deployed for plants operating to 40 or 60 years based on the current understanding of RPV degradation (and resulting reduced uncertainty in safety margins) as well as the potential liability of permanently damaging a reactor vessel. However, in some cases, thermal annealing may be required to extend plant life to 80 years.

Additional research is needed to overcome the technical needs described above and reduce the uncertainties, to reduce the liabilities and make this technique more acceptable for industry.

Thermal annealing has already been identified as a research task within the LWRSP Materials Aging and Degradation Pathway and this report provides a brief summary of the background and technical issues associated with the technology of thermal annealing of reactor pressure vessels. Under LWRSP, the effects of thermal annealing on higher fluence RPV materials and the effects of reirradiation will be examined to provide resolution to the identified issues.

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