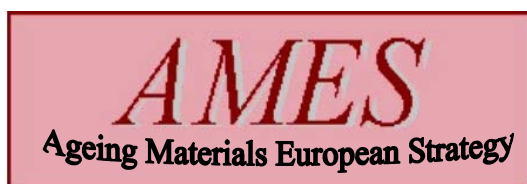




Annealing and re-embrittlement of reactor pressure vessel materials

State of the art report
ATHENA WP-4

AMES Report N. 19



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ANNEALING AND RE-EMBRITTEMENT OF RPV MATERIALS

State-of-the-Art Report

ATHENA – WP 4

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Foreground

The AMES Thematic Network on ageing, ATHENA, aimed, within the “enlarged” Europe, at reaching a consensus on important issues, identified by the AMES European Network Steering Committee, that have an impact on the life management of nuclear power plants. ATHENA created a structure enhancing the collaboration between European-funded R&D projects, national programs, and TACIS/PHARE programs. The objectives of ATHENA were to summarize the state-of-the-art and edit guidelines on relevant issues to PLiM, like the “Master Curve”, the effect of chemical composition on embrittlement rate in reactor pressure vessel steels, synergies between ageing mechanisms, validation of re-embrittlement models after annealing and other open issues in embrittlement of WWER type reactors,. The ATHENA Thematic Network, funded by EURATOM 5th Framework Programme, started in November 2001 and came to an end in October 2004.

This state-of-the-art report is the result of the tasks performed within the ATHENA work-package 4; in which all the exiting information concerning annealing and re-embrittlement of reactor pressure vessels has been compiled.

Index

1	INTRODUCTION	7
2	ANNEALING EFFECTS ON RADIATION EMBRITTLEMENT	11
2.1	Neutron irradiation damage in RPV steels	11
2.2	Importance of annealing temperature and time	16
2.3	Dependence of Tk recovery on neutron fluence and flux	23
2.4	The role of impurities in the annealing process	25
2.5	The risk of temper embrittlement during the thermal annealing	31
2.6	Effects of test methods	31
2.7	Mechanism of annealing	33
3	RE-EMBRITTLEMENT RATE AND MODELS	43
3.1	Evaluation of transition temperature of RPV base and weld metals after repeated irradiation, and annealing	43
3.2	Comparison of the experimental data with traditional models of re-embrittlement assessment	47
3.3	The assumed mechanism of RPV material re-embrittlement	52
3.4	Semi-mechanistic models for re-embrittlement assessment	58
4	TESTING AND SAMPLING	67
4.1	WWER-440 RPV material radiation embrittlement and annealing effectiveness assessment using boat samples	67
4.2	Investigation on decommissioned reactor pressure vessel	71
5	ANNEALING TECHNOLOGY	75
5.1	Russian Annealing Device	77
5.2	Annealing in SKODA	80
5.3	Annealing in Germany	84
5.4	Marble Hill Demonstration Project	85
6	REGULATORY REQUIREMENTS FOR RPV ANNEALING AND LIFETIME EVALUATION AND RELEVANT PRACTICE	93
6.1	Thermal Annealing Regulatory Rules and Guidance, ASME Code Requirements, and ASTM Guidance	93
6.2	Russian requirements to RPV annealing and lifetime evaluation	97
6.3	Licensing in Slovakia	101
6.4	Licensing the RPV Annealing in Finland	106
7	CONCLUSIONS AND RECOMMENDATIONS FOR FUTURE RESEARCH & DEVELOPMENTS ACTIVITIES IN THE FIELD OF ANNEALING AND RE-EMBRITTLEMENT OF RPVS	115

8	REFERENCES	117
8.1	Additional references:	123

List of Tables

Table 2.1	The Mn-Mo-Ni –weld wire/Linde 80 flux weld ferrite matrix composition in the different conditions, [22]	39
Table 3.1	Embrittlement mechanisms considered in semi-mechanistic model, [47]	62
Table 5.1	Annealing of WWER-440 type RPVs	76
Table 6.1	Material research programmes supporting the licensing of the annealing and operation of Loviisa-1 after the vessel anneal.	107
Table 6.2	The scope of the program by Moht Otjig	109

List of Figures

Fig. 2.1	Effect of test temperature on the energy absorbed on fracturing Charpy-V toughness specimens	12
Fig. 2.2	The effect of neutron irradiation on a) the Transition curve and b) the fracture toughness temperature	14
Fig. 2.3	Effect of neutron irradiation on upper shelf energy effects	14
Fig. 2.4	Annealing recovery of Yankee A302-B steel surveillance plate at BR3, [2]	17
Fig. 2.5	Example of an isothermal investigation on A533 type RPV steel, [4]	18
Fig. 2.6	The annealing recovery of a number of Western RPV steels and welds irradiated at temperature >270 °C and annealed for 168 hours, [5].	19
Fig. 2.7	Comparison of transition temperature, and upper shelf energy recovery for 73W weld annealed at 454 °C, [6].	20
Fig. 2.8	The annealing effectiveness for WWER-440 base metal and welds as a function of annealing temperature. $T_{ir} = 270$ °C, [7]	21
Fig. 2.9	WWER-440 materials annealing effectiveness as a function of annealing time (O- base metal, ●- weld), [7]	23
Fig. 2.10	Recovery of T_k due to the annealing at 420 °C with 150 hours of WWER-440 weld (P=0.028 %, Cu=0.18 %0) irradiated with the different fluences, [7]	24
Fig. 2.11	Dependence of residual embrittlement on phosphorus content in WWER-440 materials for different annealing temperatures, [7]	26
Fig. 2.12	Updated dependencies of residual after annealings at 460-475 °C T_k shifts of WWER-440 materials on phosphorus and copper contents.	28
Fig. 2.13	Notch ductility changes for various welds after irradiation (288 C) and post-irradiation annealing P=0.010-0.014 %; $F=1.4 \times 10^{19}$ cm ⁻² (E>1 MeV), [16]	29
Fig. 2.14	Charpy-V transition temperature changes for SA 533 B plates with 288 °C irradiation(left-hand bars) and with 399 °C post-irradiation annealing (right-hand bars). $F=2.5 \times 10^{23}$ m ⁻² (E>1 MeV), [17]	30

Fig. 2.15 The impact test results of irradiated with fluence $1 \times 10^{24} \text{ m}^{-2}$ and annealed at 500 °C WWER-440 base (a) and weld (b) metal with P~0.02%	32
Fig. 2.16 The comparison of recovery annealing results for different welds (Ni=0.59 %), [19]. Here, T is a tearing modulus	33
Fig. 2.17 Schematic dose dependence of yield strength increase and transition temperature shift for steel A302B, [30]	36
Fig. 2.18 Transition temperature shift with different nickel content but with 0.05 % copper and 0.010 % phosphorus irradiated to $1 \times 10^{24} \text{ m}^{-2}$, irradiation temperature 270 °C, [35]	38
Fig. 3.1 The transition temperature shift behavior of PWR welds with high copper content 0.35 % under primary irradiation, annealings and re-irradiations, [41]	43
Fig. 3.2 The variation of WWER-440 weld T_k value under primary and repeated irradiations	44
Fig. 3.3 Scheme of embrittlement of reactor pressure vessel under re-irradiation of sequentially irradiated and annealed materials	46
Fig. 3.4 Hardness, yield strength and transition temperature changes of irradiated, annealed and –re-irradiated WWER-440 weld metal, [20]	48
Fig. 3.5 Changes of transition temperature shift of WWER-440 welds under irradiation, annealing and re-irradiation	49
Fig. 3.6 Comparison between observed T_k shifts due to re-irradiation after annealing at 340 °C (a), 380 °C (b) and those predicted by conservative, lateral and vertical shift evaluation models	50
Fig. 3.7 Comparison between observed T_k shifts due to re-irradiation after annealing at 420-460 °C and those predicted by conservative, lateral and vertical shift evaluation models	51
Fig. 3.8 Weld metal transition temperature shift observed after re-irradiation to a fluence of $2.7 \times 10^{23} \text{ m}^{-2}$ (IAR ₂) or $1.8 \times 10^{23} \text{ m}^{-2}$ (IAR ₁) (> 1 MeV) vs. first exposure cycle. The left-hand and center bars indicate the total T_k shift with IAR treatment, [16]	53
Fig. 3.9 Effect of re-irradiation after various annealing on the transition temperature shift of weld EP-19 (Cu=0.40 %, Ni=0.59 %), [44]	53
Fig. 3.10 Distribution of copper in irradiated (a) by neutron fluence $9 \times 10^{23} \text{ m}^{-2}$ (E>0.5 MeV) and re-irradiated (b) after annealing at 475 °C by fluence $1.5 \times 10^{23} \text{ m}^{-2}$ WWER-440 weld, [45]	54
Fig. 3.11 Distribution of phosphorus in irradiated (a) by neutron fluence $9 \times 10^{23} \text{ m}^{-2}$ (E>0.5 MeV) and re-irradiated (b) after annealing at 475 °C by fluence $1.5 \times 10^{23} \text{ m}^{-2}$ WWER-440 weld, [45]	55
Fig. 3.12 The dependence of WWER -440 weld transition temperature shift under re-irradiation (after annealing at 460 °C) on copper content in the steel	56
Fig. 3.13 The dependence of WWER -440 weld transition temperature shift under re-irradiation (after annealing at 460 °C) on phosphorus content in the steel	56
Fig. 3.14 The dependence of transition temperature shift on re-irradiation fluence, [46]	58
Fig. 3.15 Schematic of embrittlement and re-embrittlement	61

Fig. 3.16 Comparison of experimental and predicted re-embrittlement shifts [34]	61
Fig. 3.17 Example of primary radiation embrittlement calculated with semi-mechanistic model, [47]	64
Fig. 3.18 Example of primary radiation embrittlement and re-embrittlement calculated with semi-mechanistic model, [47]	65
Fig. 3.19 The results using the semi-mechanistic model for the WWER-440 re-embrittlement assessment, [47]	66
Fig. 4.1 The scheme of cutting samples from Kozloduy unit 2 reactor pressure vessel, [48]	69
Fig. 4.2 Scheme of samples cutting to fabricate sub-size impact specimens (5×5 and 3×4 mm ²), [48]	69
Fig. 4.3 T _F as a function of neutron fluence (and time) for Kozloduy unit 2 RPV weld, [48]	70
Fig. 4.4 Transition temperature as a function of neutron fluence for “Kozloduy-1” weld metal 4, [52]	71
Fig. 4.5 Scheme of taking trepans of Novovoronezh unit 2 reactor pressure vessel, [49]	72
Fig. 4.6 The correlation between predicted and experimental values of irradiation embrittlement of Novovoronezh unit 2 metal, [49]	73
Fig. 4.7 Evaluation of irradiation embrittlement and also efficiency of Novovoronezh unit 2 weld annealing, [49]	74
Fig. 5.1 Annealing arrangements in Novovoronezh 3	78
Fig. 5.2 Schematic drawing (not to scale) of temperature monitoring and actual temperatures in Greifswald 1 [59]	79
Fig. 5.3 WWER 440 RPV temperature and stress levels with 1 and 3 metre annealing zone heating apparatus. Note the higher calculated stress peak for the larger annealing zone [57]	80
Fig. 5.4 Scheme of SKODA annealing device	81
Fig. 5.5 Photo of the SKODA annealing device before loading into the RPV	82
Fig. 5.6 Display of the SKODA monitoring and computing system	84
Fig. 5.7 Marble Hill ADP heating system	89
Fig. 5.8 Reactor vessel top cover with the heat exchanger	90
Fig. 5.9 Heat exchanger reactor assembly	91
Fig. 5.10 Marble Hill ADP, Ductwork Routing	92
Fig. 6.1 General principles of WWER RPV lifetime assessment	98
Fig. 6.2 Conservative approach to WWER RPV lifetime evaluation	99
Fig. 6.3 Complete set of NSSP chains for irradiation in the Unit 3 Bohunice	105
Fig. 6.4 Scheme of the location of surveillance chains in WWER-440/V-213 RPV	105
Fig. 6.5 Location of surveillance channels in WWER-440/V-213 RPVs	105
Fig. 6.6 Location of individual NSSP chains in active core of WWER-440/V-213 RPV	106
Fig. 6.7 Location of surveillance channels on the outer surface of core barrel	106

1 Introduction

Reactor pressure vessels are the most important components of any NPP as they contain practically full inventory of radioactive materials, their failure cannot be neutralised by any measure and their lifetime determine economical lifetime of NPP as they are practically non-replaceable. Lifetime of reactor pressure vessels is practically determined by their resistance against potential brittle/non-ductile failure. This resistance is then governed by the damaging effect of neutron embrittlement.

Particular sensitive locations in the RPV include the reactor core region of the RPV-the “belt-line region” that is the cylindrical portion of the RPV and which is subjected to neutron irradiation degradation of its mechanical properties. Welds and their heat affected zones in this region are important because welds are the usual location of cracks or defects.

For many years an important test to measure the fracture resistance of PV steels has been, and continues to be the empirical “Charpy V-notch specimen test”, where a relatively small notched specimen is struck by a swinging pendulum of constant energy thereby breaking the specimen. The amount of this energy absorbed during fracture is a measure of the amount of energy absorbed to cause the fracture. The larger the amount of absorbed energy for fracture, the more ductile the specimen is and this can be plotted against the test temperature to determine the brittle ductile transition temperature.

This “toughness” curve, at lower temperatures, shows the brittle regime. At higher temperatures the ‘toughness’ curve rises to a plateau (the ‘upper shelf’ value). At intermediate temperatures there is a transitional region between brittle and ductile failure.

The effect of neutron irradiation is to displace the Charpy toughness curve to higher temperatures, and by specifying the transition temperature at a given energy level, the “shift in transition temperature” for a given neutron fluence can be established.

Values of ‘shift’ can be plotted against neutron fluence to establish the trend curve of shift with fluence for those specimens. The second characteristic of the effect of irradiation is that the upper ‘shelf’ value of fracture resistance decreases with neutron fluence. As the procedures for fracture assessment and stress analysis of RPVs developed Linear Elastic Fracture Mechanics (LEFM) analyses techniques were introduced and the correlation between fracture tests and the Charpy tests were also developed.

The assessment of reactor pressure vessel integrity has been the subject of continuous development particularly when the resistance to brittle, non-ductile failure is evaluated. In last years, application of the “Master Curve” approach is becoming a more used method for direct determination of fracture toughness of RPV materials to be used in their integrity evaluation.

Irradiation embrittlement is pronounced in two consequences: firstly, it narrows the “pressure-temperature” operation window for normal operating conditions, and second, it limits RPV lifetime as the transition temperature of RPV materials cannot be higher than that determined from the pressurized thermal shock calculations.

Several mitigation measures can be applied to decrease radiation embrittlement of RPV beltline materials:

- use of a “low-leakage core” that could decrease neutron flux on RPV wall by 30 – 40 %
- use of “dummy elements” in reactor core periphery/corners that could decrease the original peak flux by a factor of 4.5 and the "new" peak flux by a factor of about 2.5

- recovery annealing as the most effective measure as it could practically restore initial mechanical properties of RPV materials

All three measures have been applied in different type of reactors:

- “low-leakage core” is used practically in all reactors throughout the world as it is the cheapest measure, even though with a limited efficiency,
- dummy elements were inserted practically only to WWER-440/V-230 type reactors where a substantial decrease of neutron flux was required. In most cases this insertion was connected with RPV annealing
- recovery annealing was applied

The first RPV annealings were done using primary coolant and nuclear heat (US Army SM-1A) or primary pump heat (Belgian BR-3). The annealing temperature in the former case was 293-300 °C (72-79 °C above the service temperature). The degree of recovery in this case was about 70%. In the BR-3 reactor the service temperature was 260 °C and the vessel was annealed at 343 °C. The recovery was estimated to be at least 50%. The planned annealing of the Yankee Rowe vessel at 343 °C (83 °C above the service temperature) was estimated to give a 45-55% recovery (Server & Biemiller, 1993). The “wet” annealing technique is easy to implement because usually only the fuel is needed to be removed from the RPV, but unfortunately it can be utilised only in reactors which have a low service temperature. RPVs are not designed to stand the pressure of water at higher temperatures and the critical point of water is reached already at 374 °C ($p_{crit} = 219$ bar). Due to very limited recovery, wet annealing with water is not a practical solution for power reactors and it needs to be frequently repeated.

Following the publication of the Westinghouse conceptual procedure for dry thermal annealing on embrittled RPV, the Russians (and recently, the Czechs) undertook the thermal annealing of several highly irradiated WWER-440 RPVs. To date, at least 15 vessel thermal annealings have been realized. The WWER

experience, along with the results of relevant laboratory scale research with western RPV material irradiated in materials test reactors and material removed from commercial RPV surveillance programmes, is consistent and indicates that an annealing temperature at least 150 °C more than the irradiation temperature is required for at least 100 to 168 hours to obtain a significant benefit. A good recovery of all of the mechanical properties was observed when the thermal annealing temperature was about 450°C for about 168 hours (1 week). And, the re-embrittlement rates upon subsequent re-irradiation were similar to the embrittlement rates observed prior to the thermal anneal. The dominant factors which influence the degree of recovery of the properties of the irradiated RPV steels are the annealing temperature relative to the irradiation (service) temperature, the time at the annealing temperature, the impurity and alloying element levels, and the type of product (plate, forging, weldment, etc.).

2 Annealing Effects on Radiation Embrittlement

2.1 Neutron irradiation damage in RPV steels

Neutron irradiation degrades the mechanical properties of RPV steels and the extent of the degradation is determined by a number of factors such as neutron fluence, irradiation temperature, neutron flux and the concentration of deleterious elements in the steel.

Particular sensitive locations in the RPV include the reactor core region of the RPV, the “belt-line region”, that is the cylindrical portion of the RPV and which is subjected to neutron irradiation degradation of its mechanical properties. Welds and their heat affected zones in this region are important because welds are the usual location of cracks or defects. For many years an important test to measure the fracture resistance of PV steels has been, and continues to be the empirical “Charpy V-notch specimen test”, where a relatively small notched specimen is struck by a swinging pendulum of constant energy thereby breaking the specimen. The amount of this energy absorbed during fracture is a measure of the amount of energy absorbed to cause the fracture. The larger the amount of absorbed energy for fracture then the more ductile the specimen and this can be plotted against the test temperature to determine the brittle ductile transition temperature.

The Charpy absorbed energy (impact energy) increases with increased test temperature as shown in Fig. 2.1. The “toughness” curve, at lower temperatures, shows the brittle regime. At higher temperatures the “toughness” curve rises to a plateau (the “upper shelf” value). At intermediate temperatures there is a transitional region between brittle and ductile failure.

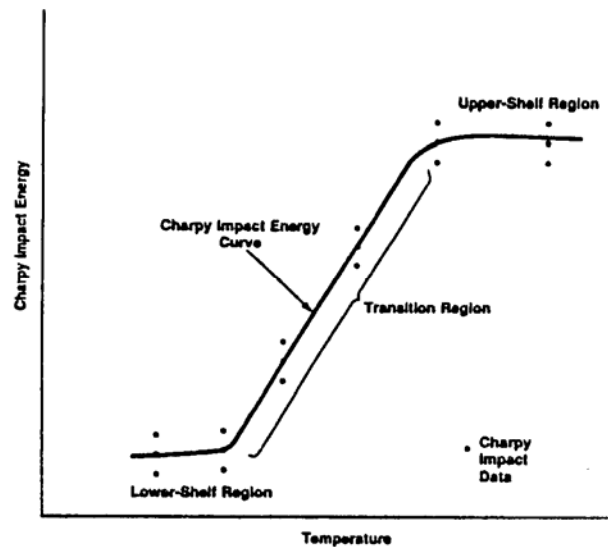


Fig. 2.1 Effect of test temperature on the energy absorbed on fracturing Charpy-V toughness specimens

The effect of neutron irradiation is to displace the Charpy toughness curve to higher temperatures, (see Fig. 2.2a) and by specifying the transition temperature at a given energy level, the 'shift in transition temperature' for a given neutron fluence can be established. Values of 'shift' can be plotted against neutron fluence to establish the trend curve of shift with fluence for those specimens. The second characteristic of the effect of irradiation and shown schematically in Fig. 2.2a is that the upper shelf value of fracture resistance decreases with neutron fluence. In this essay we only consider, generally, the increase in transition temperature on irradiation. The decrease in upper shelf was of concern in earlier PWRs which had been welded with Linde-90 fluxes which resulted in low upper shelf properties and did not allow the clear determination of 'shifts'. This problem was extensively studied and eventually resolved. As the procedures for fracture assessment and stress analysis of RPVs developed Linear Elastic Fracture Mechanics (LEFM) analyses techniques were introduced and the correlation between fracture tests and the Charpy tests were also developed. The Charpy transition shift corresponds to an equivalent decrease in the material's linear elastic fracture toughness in the region of the transition curve where the tests are valid (see Fig 2.2b). Also, as the Charpy upper

shelf decreases the Elasto-Plastic Fracture Mechanics fracture resistance (J-integral) decreases, see Fig.2.3a and 2.3b. These approaches, whilst providing a more specific analysis of the behaviour of known and assumed defects, had limitations with regard to the validity of test data to the behaviour of actual RPVs, particularly, in the case of LEFM techniques, with regard to the upper shelf ductile region. Developments to include ductility as well as brittle components in fracture mechanics assessments has led to further developments in mechanical property testing and fracture assessment.

The assessment of reactor pressure vessel integrity has been the subject of continuous development particularly when the resistance to brittle, non-ductile failure is evaluated.

There have been important developments based on the “Master Curve” approach. This approach has been developed on the basis of the general statistical model when the unconditional probability of cleavage fracture initiation as well as conditional cleavage crack propagation led to a three-parameter Weibull model. An average fracture toughness value is determined by performing tests at a single temperature and using Weibull statistics. The “Master Curve” methodology is based on observation that fracture toughness transition curves present a characteristic shape common to all ferritic steels. Such an approach can be relatively easily implemented. This approach, as well as the development of the test method, allows testing of relatively small scale specimens to obtain valid data of fracture toughness, K_{JC} , and consequently to determine a “Reference Temperature, T_0 ”. The reference temperature T_0 is defined as the temperature at which the fracture toughness curve reaches a value of $100 \text{ MPa}\cdot\text{m}^{1/2}$.

So with the Master Curve we can directly establish the material toughness properties, without needing to use RT_{NDT} . One same Master Curve may be used for all vessels steels, changing only its position on the temperature axis until it passes

through the average fracture toughness value, which is obtained from the tests performed on the material of interest.

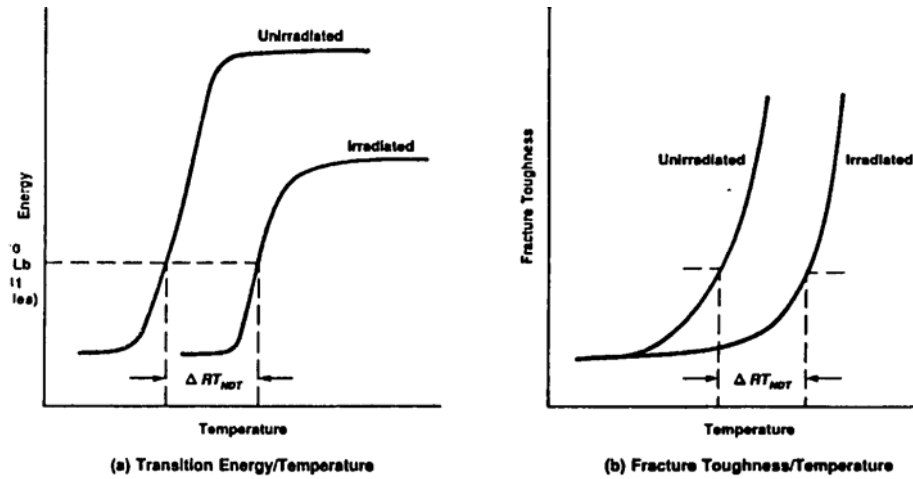


Fig. 2.2 The effect of neutron irradiation on a) the Transition curve and b) the fracture toughness temperature

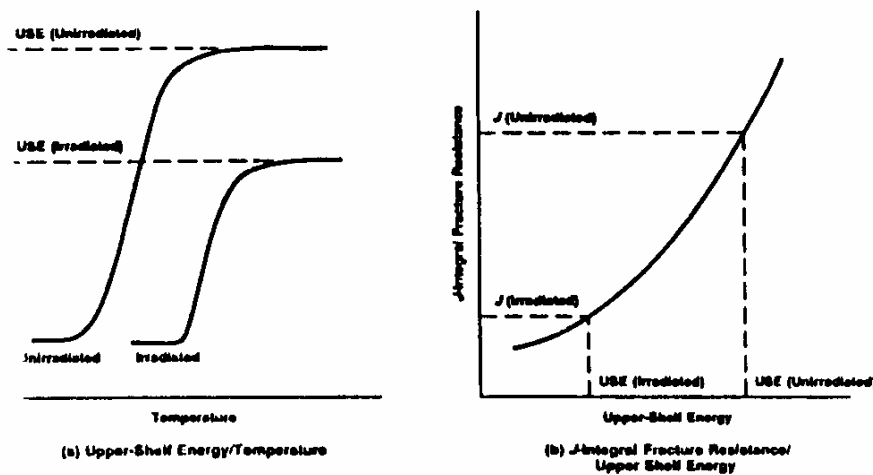


Fig. 2.3 Effect of neutron irradiation on upper shelf energy effects

The overall effect of fast neutron exposure is that ferritic steels experience an increase in hardness and tensile properties and a decrease in ductility and toughness, under certain conditions of radiation.

For example:

1. The effect of neutron fluence on radiation hardening and embrittlement has been reported to be significant at fluences above 10^{22} m⁻² ($E > 1$ MeV). Unless a steady state or saturation condition is reached, an increase in neutron fluence results in an increase in RT_{NDT} , yield strength and hardness, and a decrease in the Charpy toughness, also in the upper shelf temperature region. There are significant variations in the fluence and radiation damage around the circumference and in the longitudinal direction of RPVs.
2. Alloy composition, (especially when consideration is given to impurity copper and phosphorus and alloying element nickel) is known to have a strong effect on radiation sensitivity. Data have been generated on both commercial and model alloys to show the effects of alloy composition.
3. Radiation temperature has long been recognized to have an effect on the extent of the radiation damage. Data from the early nineteen-sixties demonstrated that the maximum embrittlement occurred during radiation at temperatures below 120°C. Recent studies have reported a decrease in radiation embrittlement at higher temperatures ($>310^{\circ}\text{C}$), which is attributed to the dynamic in-situ “annealing” of the damage.
4. Microstructural characteristics, such as grain size and metallurgical phases (lower or upper bainite, ferrite), can influence the severity of radiation damage associated with a given fluence.
5. The neutron flux energy spectrum contributions to the embrittlement behavior of ferritic steels are secondary effects. However, recent reactor experience has suggested that, under certain conditions, the flux spectrum may influence the degree of radiation embrittlement caused in ferritic steels.

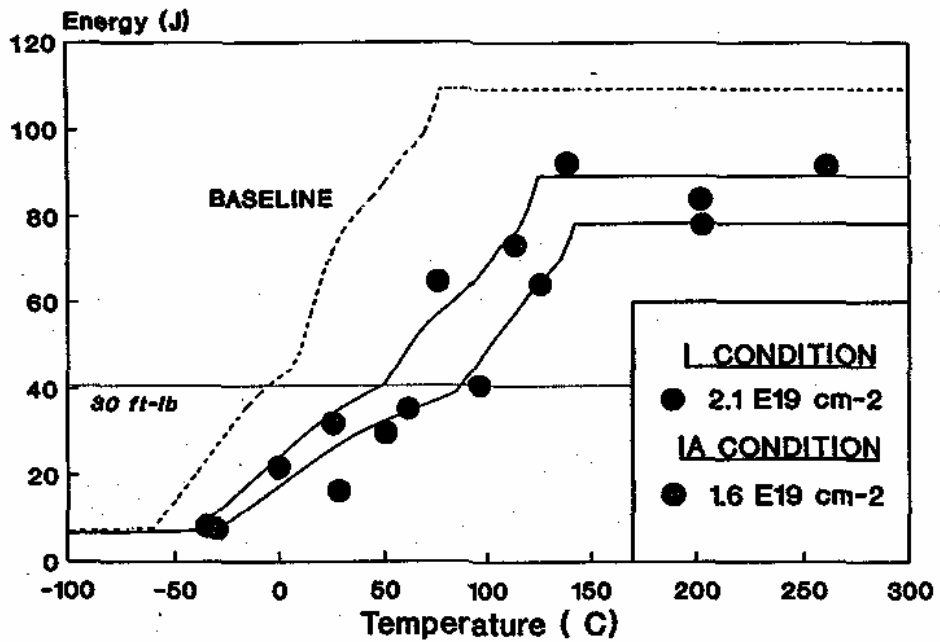
Once a RPV is degraded by radiation embrittlement (e.g. significant increase in Charpy ductile-brittle transition temperature or reduction of fracture toughness), thermal annealing of the RPV is the only way to recover the RPV material toughness properties. Thermal annealing is a method by which the RPV (with all internals removed) is heated up to some temperature by use of an external heat source (electrical heaters, hot air), held for a given period and slowly cooled.

2.2 Importance of annealing temperature and time

There have been many investigations, studies and reviews of the annealing of irradiation effects in both Eastern and Western steels. Annealing is carried out in order to recover the mechanical properties. Radiation effects are usually manifested as an increase in ductile-brittle transition temperature, a drop in the upper shelf fracture energy and decrease in fracture toughness. Heating the steel at a temperature higher than the irradiation temperature, reverses these changes and this action can lead towards some recovery of the unirradiated mechanical properties.

Before 1987 (first annealing of operating WWER-440 at Novovoronezh NPP), two pressure vessels had been annealed – an army reactor SM-1A in USA and BR-3 in Belgium. In both cases the so-called “wet” annealing method was applied, the annealing at that temperature ~ 340 °C was reached without external heating, but by increasing the coolant temperature achieved by the energy of the circulating pumps of the primary circuit. This procedure produced an insignificant recover of RPV materials properties, [1]. The temperature ~ 340 °C is the maximum possible in “wet” annealing. This is an essential obstacle preventing the achievement of maximum reduction of irradiation embrittlement. Advantages of the “wet” annealing are the possibility of heat treatment applied to the whole RPV and the absence of a special heating device.

The study of the "wet" annealing efficiency has been carried out by A. Fabry, [2]. The impact toughness before and after 2 weeks annealing at 343 °C is presented in Fig. 2.4. The transition temperature shift recovery is less than 50 %.



Neutron Fluence Rate: 6.0 E10 cm⁻².sec⁻¹
Irradiation Temperature: 274 C

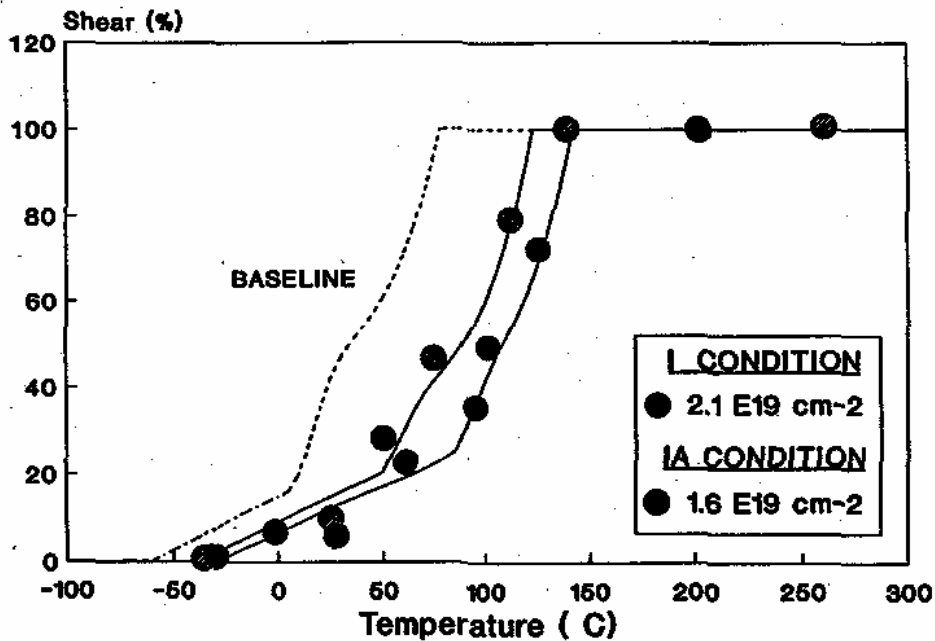


Fig. 2.4 Annealing recovery of Yankee A302-B steel surveillance plate at BR3, [2]

To achieve a temperature higher than 340 °C it is necessary to remove the core and all reactor internals, and also to use an external source of heating inside the RPV. This variant of annealing is called “dry”. In this scheme, restrictions on temperature can be defined by thermal stresses in RPV. The “dry” annealing permits the recovery heat treatment with a difference between annealing and irradiation temperatures up to 230 °C. It was recognized that the “dry” annealing option is a practical solution [3].

The example of isothermal investigation, using microhardness as an indicator of response of a mock-up RPV forging, containing 0.14 wt. % copper as a deliberate impurity condition, is shown in Fig. 2.5 [4]. It was concluded that if the temperature is high enough, then times excess of 168 hours are not likely to be necessary to obtain good recovery of mechanical properties.

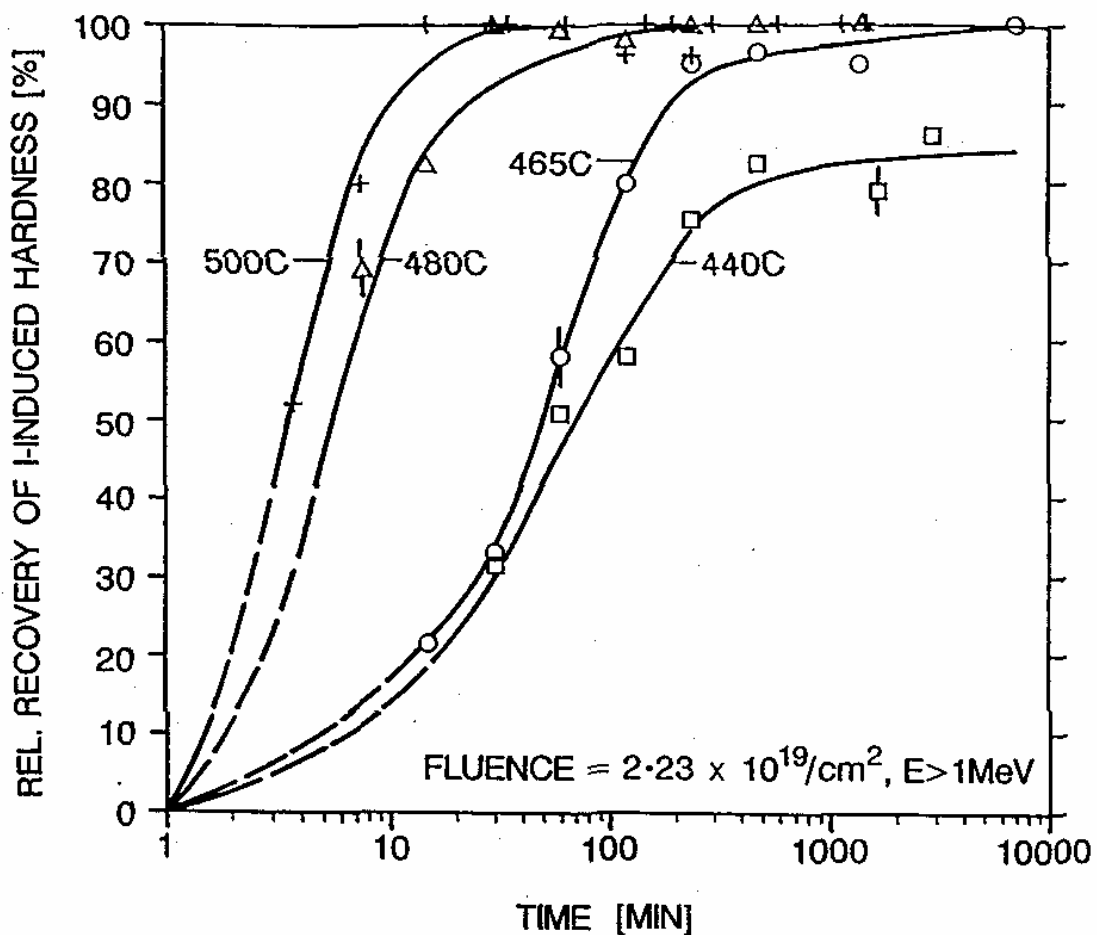


Fig. 2.5 Example of an isothermal investigation on A533 type RPV steel, [4]

The basic result of comprehensive investigation of PWR materials is presented in [5]. The annealing recovery of a number of Western RPV steels and welds irradiated at temperature higher than 270 °C and annealed for 168 hours is shown in Fig. 2.6.

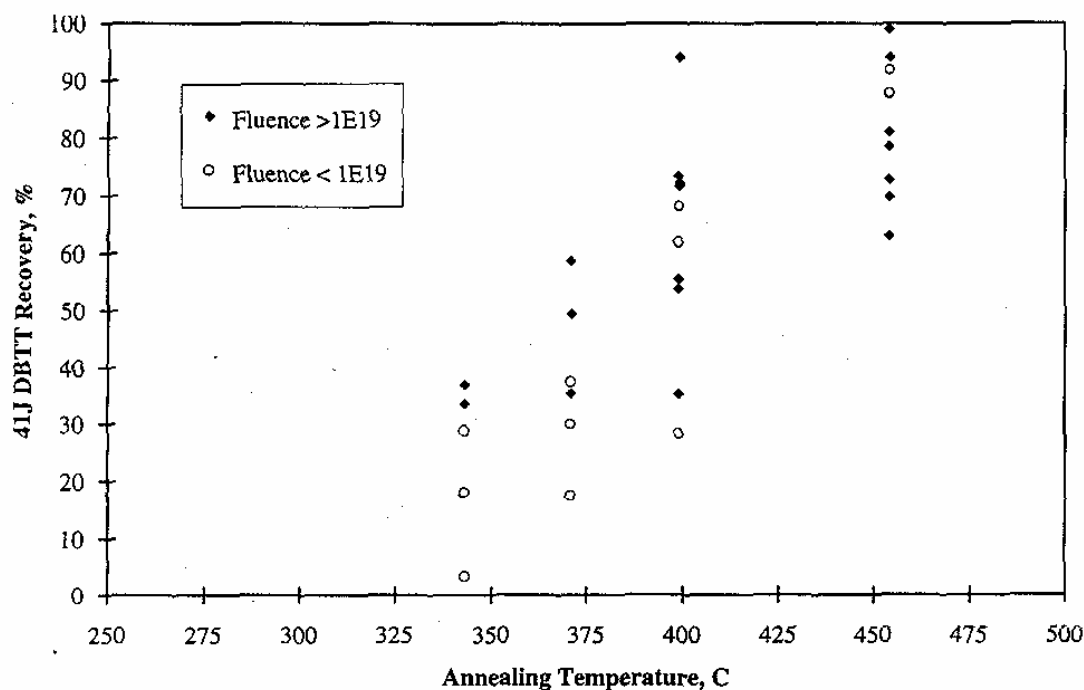
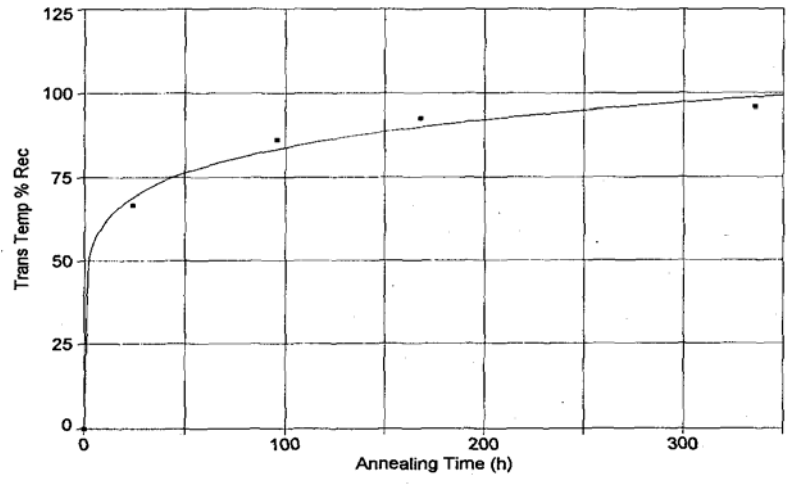


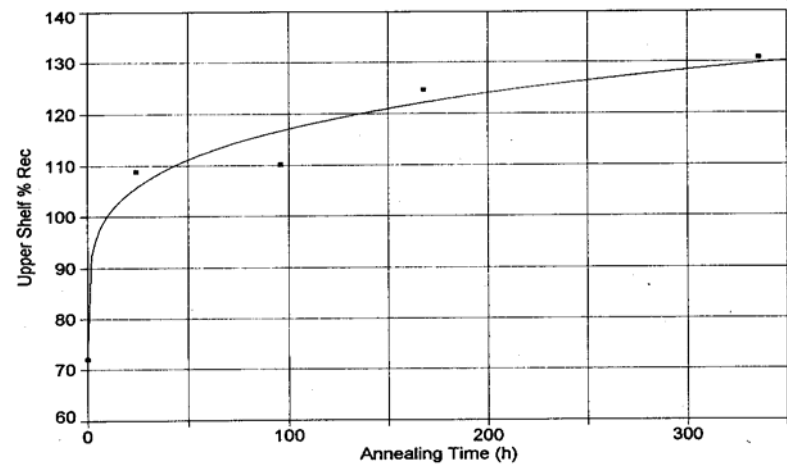
Fig. 2.6 The annealing recovery of a number of Western RPV steels and welds irradiated at temperature >270 °C and annealed for 168 hours, [5].

The detailed study of mechanical properties recovery dependence on annealing time has been carried out in ORNL, [6], Fig. 2.7. It was shown that for the specially fabricated (high copper, low upper shelf energy, weld 73W) the upper shelf recovered 100 % after 24 hours annealing and more rapidly than transition temperature. Annealing for 168 hours recovered 90 % of transition temperature shift. Annealing for another 168 hours only resulted in extra 5 % recovery.

a)



b)



c)

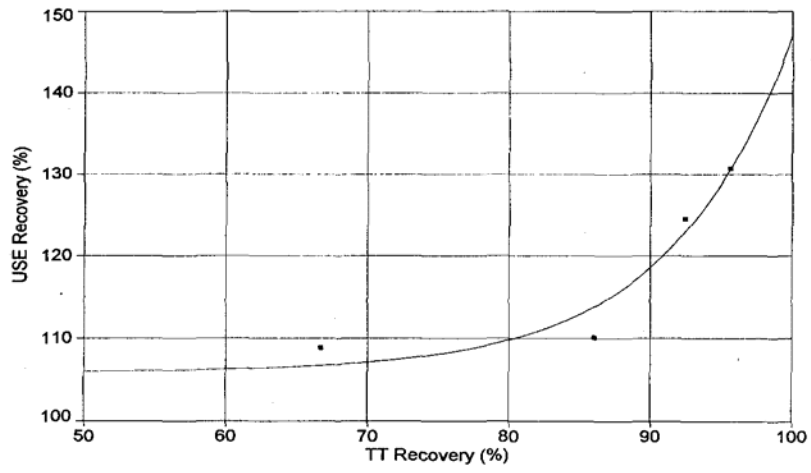


Fig. 2.7 Comparison of transition temperature, and upper shelf energy recovery for 73W weld annealed at 454 °C, [6].

To investigate these factors in the WWER RPV material property recovery complex investigations were carried out with the specimens irradiated in NPP reactors in locations of surveillance specimens [7]. The data obtained provided evidence of partial recovery of T_k at temperature difference between annealing and irradiation of 70 °C. With an increase in annealing temperature the degree of T_k recovery increases too. The considerable amount of WWER-440 base and weld metals with a wide range of impurity contents were irradiated with different fluences and for these reasons had different T_k shift after irradiation. The value

$$\eta = \frac{(T_F - T_a)}{(T_F - T_{k0})} \cdot 100 \quad (2.1)$$

was used for T_k recovery efficiency assessment at different annealing temperatures. Here T_{k0} , T_F and T_a are the values of T_k for unirradiated, irradiated and annealed material, respectively. The results are presented in Fig. 2.8.

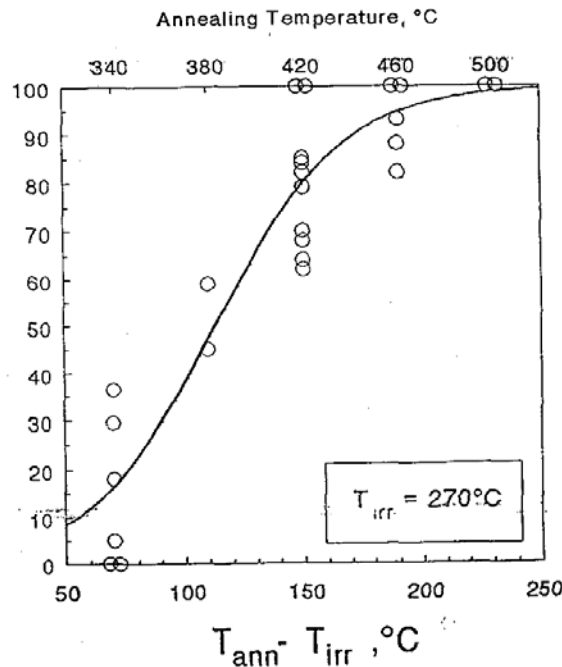


Fig. 2.8 The annealing effectiveness for WWER-440 base metal and welds as a function of annealing temperature. $T_{irr} = 270^\circ\text{C}$, [7]

Rather big scatter of the data in Fig. 2.8 should be emphasized. Detailed study of factors in recovery of properties has provided evidence of the dependency of recovery efficiency not only on annealing temperature, but on the impurity content (phosphorus and copper) of the material. In particular, the value of η depends on neutron fluence. Nevertheless this relation shows a common tendency to increase the degree of recovery of T_k shift in irradiated WWER material with an increase in annealing temperature. It agrees with PWR material recovery results presented in Fig. 2.6.

The following common conclusion is to be made about the role of annealing temperature for PWR and WWER-440 material recovery. For temperatures in the interval 340 to 420 °C the dependence of T_k recovery is close to linear; at temperatures 450 - 470 °C the recovery is about 80 % or more, which corresponds to a residual embrittlement after annealing not bigger than 20 - 30 °C. At an annealing temperature of 340 °C only an insignificant recovery in T_k for irradiated materials was observed, 20 % on average.

On the basis of experimental results above it was concluded that “wet” annealing at the temperature of 340 °C is not effective for the WWER-440 RPV irradiated material recovery.

The recovery of upper shelf energy for WWER materials also occurs more efficiently during annealing than the recovery of T_k [7]. Practically always the upper shelf reaches the value corresponding to the unirradiated state.

Experimental validation of WWER material annealing time has been determined using specimens made of 15Kh2MFA base and weld metal, irradiated in Armenia NPP to a fluence of $\sim 1 \times 10^{24} \text{ m}^{-2}$ ($E > 0.5 \text{ MeV}$). Results are presented in Fig. 2.9. It is seen that the greatest recovery of T_k occurs at first 10 hours of annealing. The prolongation of annealing leads to a retardation of the process and after that to a saturation of T_k recovery. The recovery occurs faster at higher temperature.

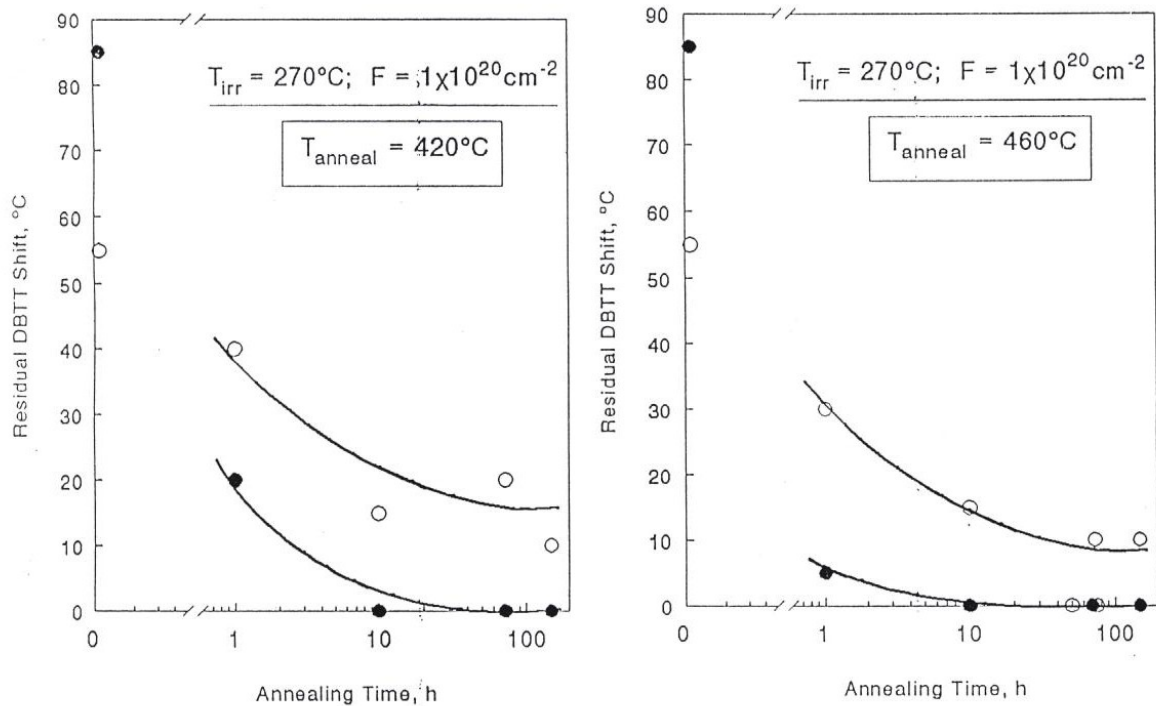


Fig. 2.9 WWER-440 materials annealing effectiveness as a function of annealing time (○- base metal, ●- weld), [7]

2.3 Dependence of T_k recovery on neutron fluence and flux

There is no general agreement reported about the influence of neutron fluence on the recovery of irradiated steel properties due to annealing. For instance, the data presented in [8] demonstrate a decrease of recovery for 15Kh2MFA steel irradiated at 60°C and annealed at 400°C with increase in neutron fluence. Recovery of T_k also decreases with an increase in neutron fluence if the irradiation temperature is $100 - 130^{\circ}\text{C}$ and annealing temperature is 300°C . At irradiation temperature of 288°C and annealing at 343°C , T_k recovery depends only slightly on neutron fluence and increases with an increase in fluence at annealing temperature of 399°C in A302-B steel, [9].

The complexity of experiments aimed at clarifying the effect of neutron fluence influence on the recovery of mechanical properties in irradiated RPV steels is defined by the variety of factors influencing steel embrittlement, and therefore property recovery. The experiments should be carried out on the same material, and also at

the same irradiation temperature. Reliable data on neutron fluence influence can be obtained by testing of surveillance specimens.

In [7] the dependence of T_k recovery from fluence for WWER-440 materials was obtained on the basis of testing 4 surveillance sets irradiated of Armenia NPP and annealed at 420 °C. After annealing for 150 hours the ΔT_k returned to the same absolute value ~ 30 °C for all 4 groups tested specimens, Fig. 2.10. So the annealing at 420 °C for 150 hours causes the same residual metal embrittlement. In other words, the T_k of metal annealed after irradiation does not depend on neutron fluence. Therefore the degree of T_k recovery increases with increase in fast neutron fluence.

A similar result has been obtained in [10] for NiCrMo weld metal irradiated at 315 °C and annealed at 450 °C for 60 hours.

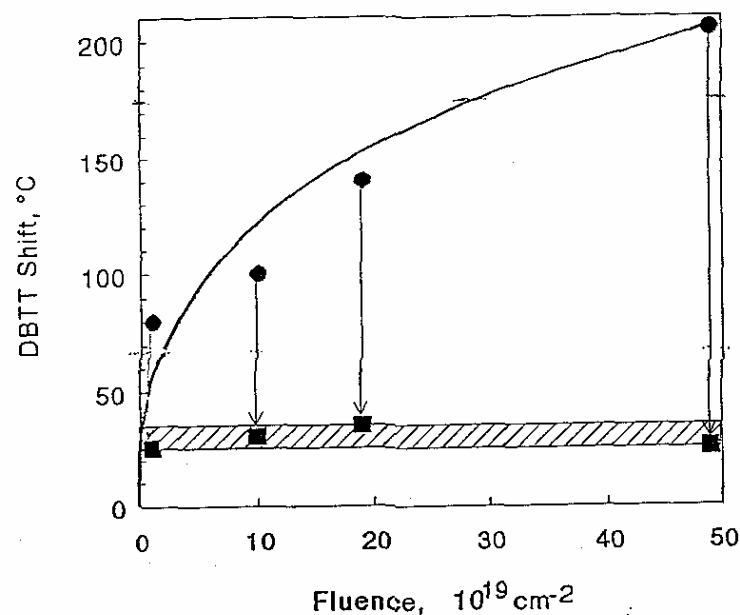


Fig. 2.10 Recovery of T_k due to the annealing at 420 °C with 150 hours of WWER-440 weld (P=0.028 %, Cu=0.18 %) irradiated with the different fluences, [7]

As regards a neutron flux influence the special assessment of effect has not been done. But indirect evidence of limited effect could be found in Fig. 2.10, where the point with smallest fluence was obtained by irradiation with flux ~10 times less than others. The results presented in Chapter 5 demonstrated good agreement between predicted results with the results obtained from sampling of operating RPVs.

2.4 The role of impurities in the annealing process

The independence of residual embrittlement (ΔT_{res}) after annealing on the neutron fluence established in 2.2, suggests that residual irradiation embrittlement is an individual peculiarity of the material, dependent on its chemical composition. The sensitivity of RPV steels to neutron irradiation due to phosphorus, copper and nickel contents in steel is clearly demonstrated [8, 11-15]. It would be reasonable to assume that the residual (after annealing) shift of T_k also depends on these chemical elements content.

The influence of impurity content on WWER-440 materials' ability to recover T_k has been studied in 15Kh2MFA base and weld metal with different phosphorus and copper contents, [7]. The phosphorus content varied over the range 0.006 % to 0.055 % and copper from 0.03 % to 0.18 %. All materials were industrially prepared with phosphorus and copper content according to the standard. The specimens were irradiated in surveillance channels at a temperature 270 °C.

Independence of ΔT_{res} value on neutron fluence has allowed using it as a characteristic of annealing efficiency. The conclusion was that the major impact on residual embrittlement is phosphorus content of the steel, if the irradiation and annealing conditions are similar. The higher the content, the larger the ΔT_{res} value:

$$\Delta T_{res} = a_1 \cdot (\%P) + a_2 \pm \delta \quad (2.2)$$

As an upper limit the relation:

$$\Delta T_{res} \leq 1.4 \times 10^3 \cdot (\% P) \tag{2.3}$$

was recommended for RPV material assessment.

This relation was used for the Novovoronezh Unit 3 NPP RPV weld residual embrittlement assessment. That RPV has been annealed at a temperature 430 °C in 1987. It is necessary to note that at the time when experiments above have been carried out the variation in copper content in the materials tested was limited with lower phosphorus content, so the impact of copper practically was not assessed.

In [7] the dependencies of residual embrittlement on phosphorus content in WWER metal at annealing temperatures of 340 and 460 °C are also presented. The materials annealed at 340 °C have been irradiated to the same neutron fluence $1 \times 10^{24} \text{ m}^{-2}$. For the materials annealed at 460 °C the independence of ΔT_{res} on neutron fluence has been accepted. In the Fig. 2.11 the dependencies of ΔT_{res} for the WWER-440 materials annealed at 340, 420 and 460 °C on phosphorus content in steel are presented.

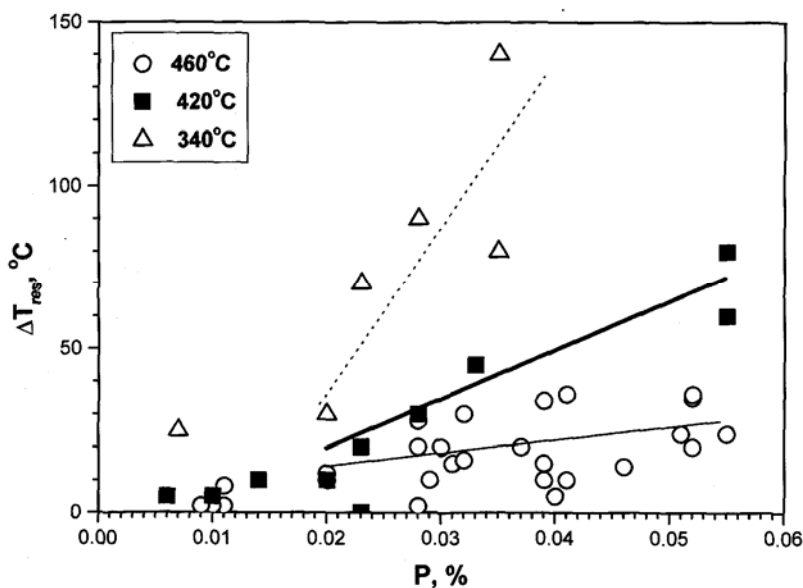


Fig. 2.11 Dependence of residual embrittlement on phosphorus content in WWER-440 materials for different annealing temperatures, [7]

The data presented show that for the same irradiation conditions the value of residual embrittlement is proportional to the phosphorus content. If the phosphorus content is the same, the shift of ΔT_{res} is lower when the annealing temperature is higher. It has to be emphasized the results published in [7] were obtained in the late 80-ties/early 90-ties. Since then a substantial amount of information has been obtained. The updated dependencies of ΔT_{res} on phosphorus and copper content for WWER-440 materials are presented in Fig. 2.12. In spite of big scatter in Fig. 2.12, it is seen that for steels with low phosphorus and copper ΔT_{res} is not more than 10 °C. If phosphorus content is more than at least ~0.03 % or Cu content is more than 0.13 %, ΔT_{res} could reach 30 – 40 °C.

Irradiation embrittlement in PWR reactors is mainly due to high copper content. Because older RPVs are usually constructed of hot-rolled plates, they also have axial joints and, consequently weld metal in the entire reactor core area. The majority of test results come from ~400 °C annealing, which seems to be inadequate for high Cu welds. Fig. 2.13 and Fig. 2.14 show the recovery in the T_k achieved with 399 and 454 °C post-irradiation annealing, [16-17]. The degree of recovery depends strongly on the copper content. The nickel and phosphorus have only slightly reduced the recovery. The influence of impurities is similar in both base and weld metals.

The WWER and PWR results do not contradict each other. In WWER-440 steels the variation of phosphorus is wider than copper, in PWR quite the contrary. The overall conclusion is the following: for material with low level of P and Cu annealing at temperatures of 450-470 °C leads to nearly full recovery of T_k . If phosphorus content is more than 0.02 % and copper content is more than 0.2 %, the residual shift of T_k could be up to 30-40 °C.

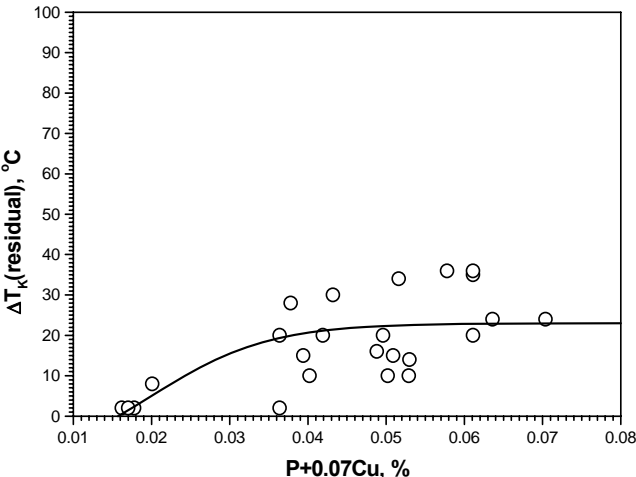
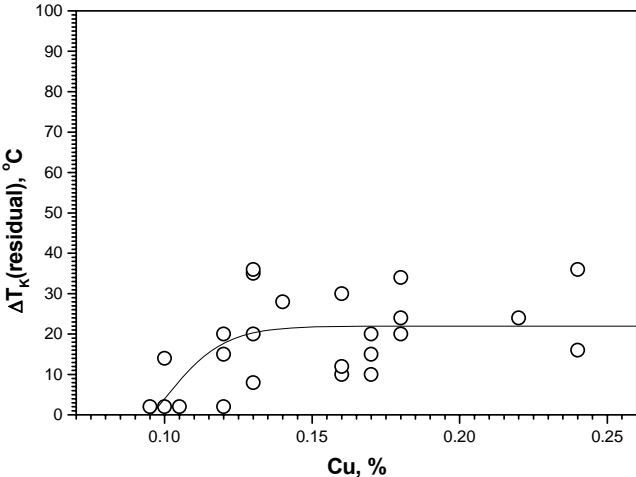
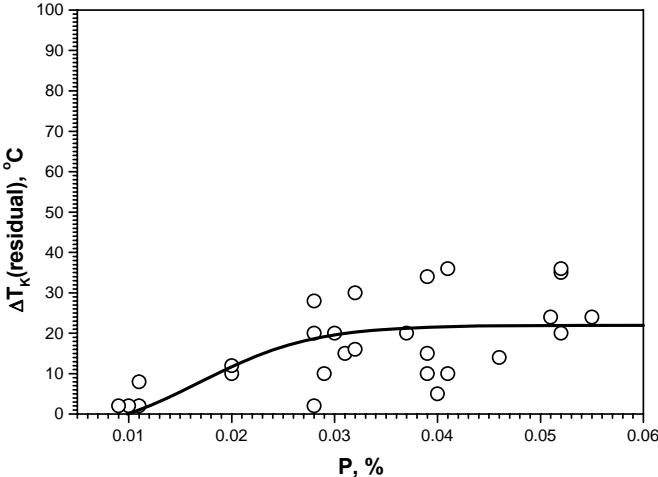


Fig. 2.12 Updated dependencies of residual after annealings at 460-475 °C T_k shifts of WWER-440 materials on phosphorus and copper contents.

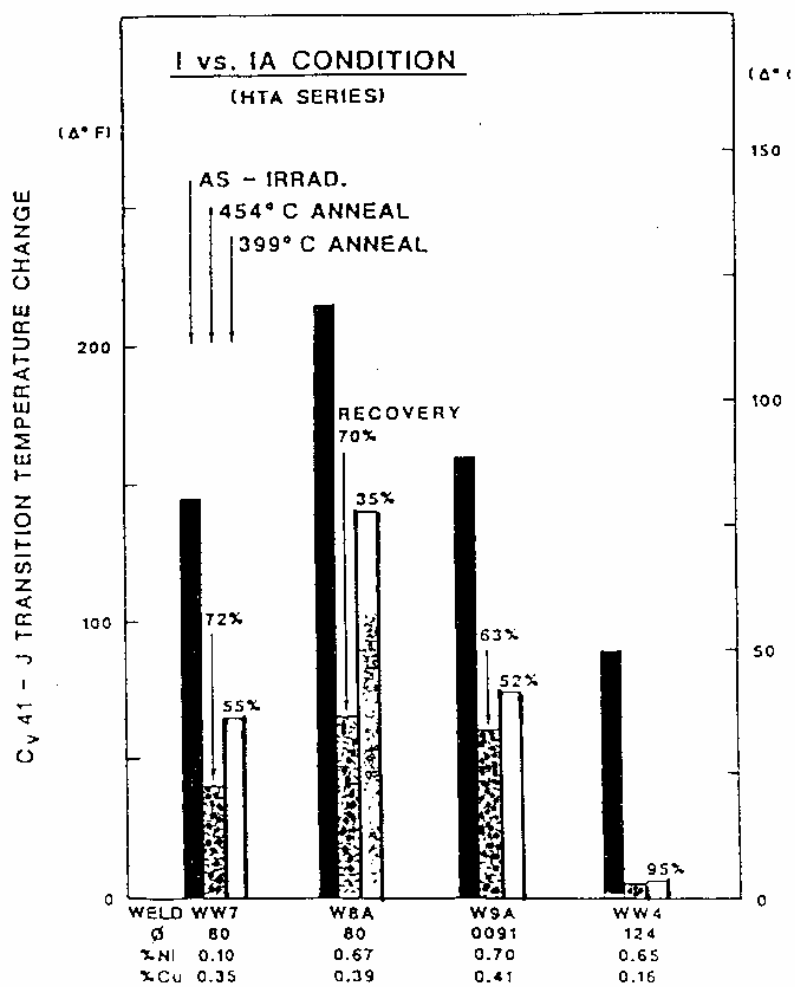


Fig. 2.13 Notch ductility changes for various welds after irradiation (288 C) and post-irradiation annealing P=0.010-0.014 %; F=1.4×10¹⁹ cm⁻² (E>1 MeV), [16]

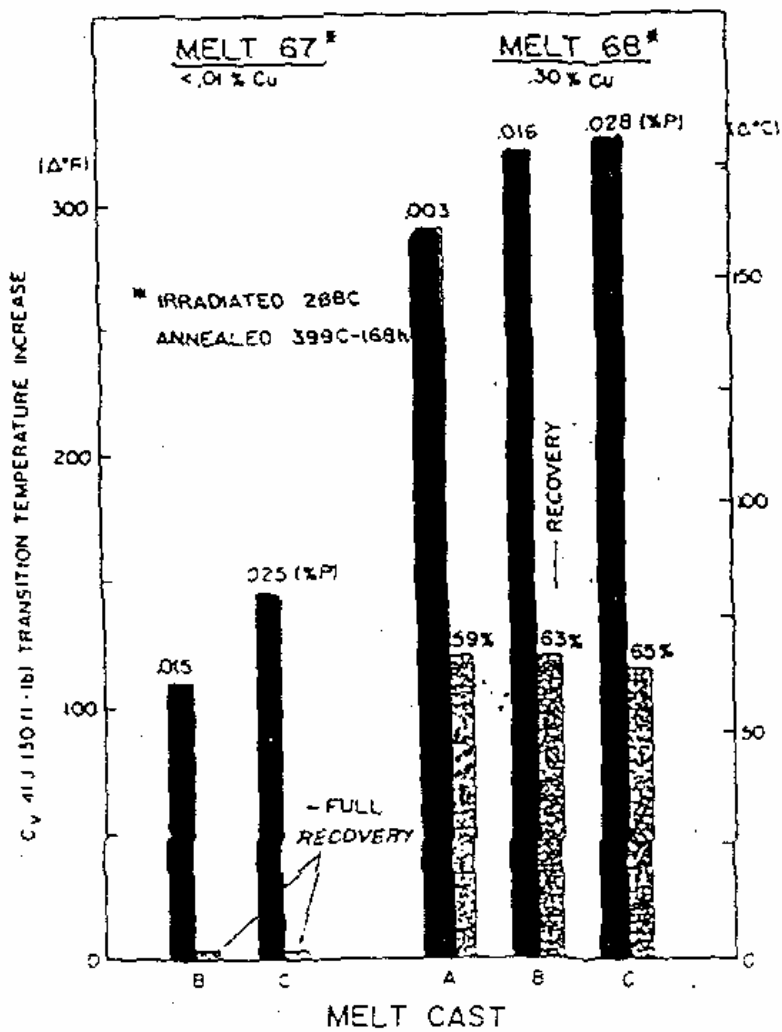


Fig. 2.14 Charpy-V transition temperature changes for SA 533 B plates with 288 °C irradiation(left-hand bars) and with 399 °C post-irradiation annealing (right-hand bars). $F=2.5 \times 10^{23} \text{ m}^{-2}$ ($E>1 \text{ MeV}$), [17]

2.5 The risk of temper embrittlement during the thermal annealing

To justify RPV annealing schedules, it had to be demonstrated that thermal ageing did not occur after annealing in RPV materials. To solve this problem the sets of specimens of WWER-440 base metal (P=0.020 % and Cu=0.11 %) and weld (P=0.023 % and Cu=0.12 %) were annealed for 1,500 and 3,000 hours after irradiation at a neutron fluence of $1 \times 10^{24} \text{ m}^{-2}$ ($T_{\text{irr}} = 270 \text{ }^{\circ}\text{C}$). Annealing temperature was $500 \text{ }^{\circ}\text{C}$, which is the maximum temperature for RPV annealing, in order to obtain conservative data. The results of base and weld metal testing have not shown any change in T_k , Fig. 2.15. Consequently during the annealing at $500 \text{ }^{\circ}\text{C}$ the full recovery of T_k had occurred, and further treatment did not cause any thermal ageing in tested steels. It was concluded that if thermal annealing can be carried out repeatedly, it would not lead to additional increase in T_k of WWER-440 steel at least up to a total heat treatment of 1,000 hours.

2.6 Effects of test methods

For the PWR materials the comparison of properties recovery measured by different test method was made in [18-19]. It was mentioned that the degree of recovery measured by ductile fracture toughness and tensile testing seemed to be smaller than the other properties, Fig. 2.16.

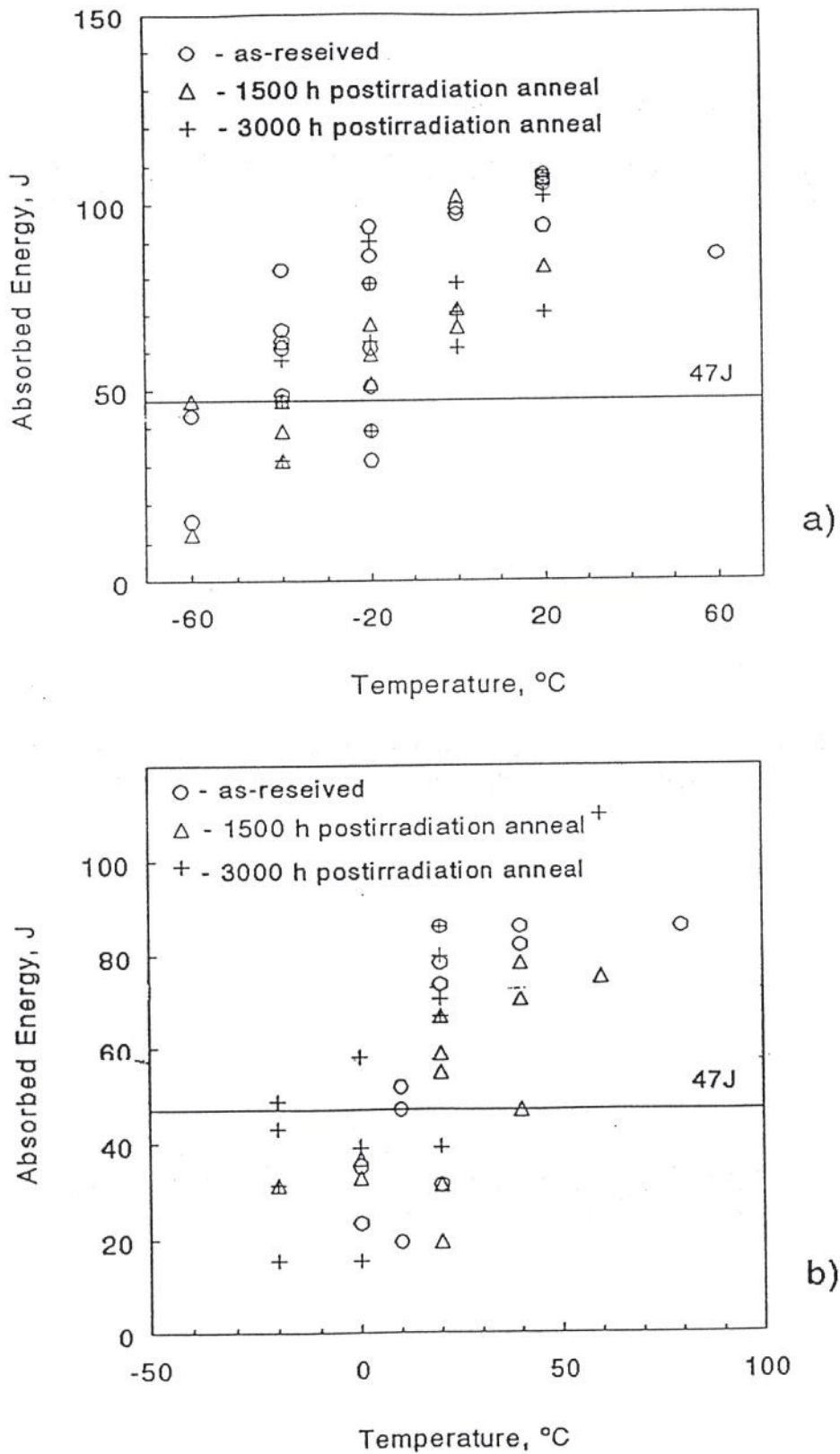


Fig. 2.15 The impact test results of irradiated with fluence $1 \times 10^{24} \text{ m}^{-2}$ and annealed at $500 \text{ }^\circ\text{C}$ WWER-440 base (a) and weld (b) metal with $P \sim 0.02\%$

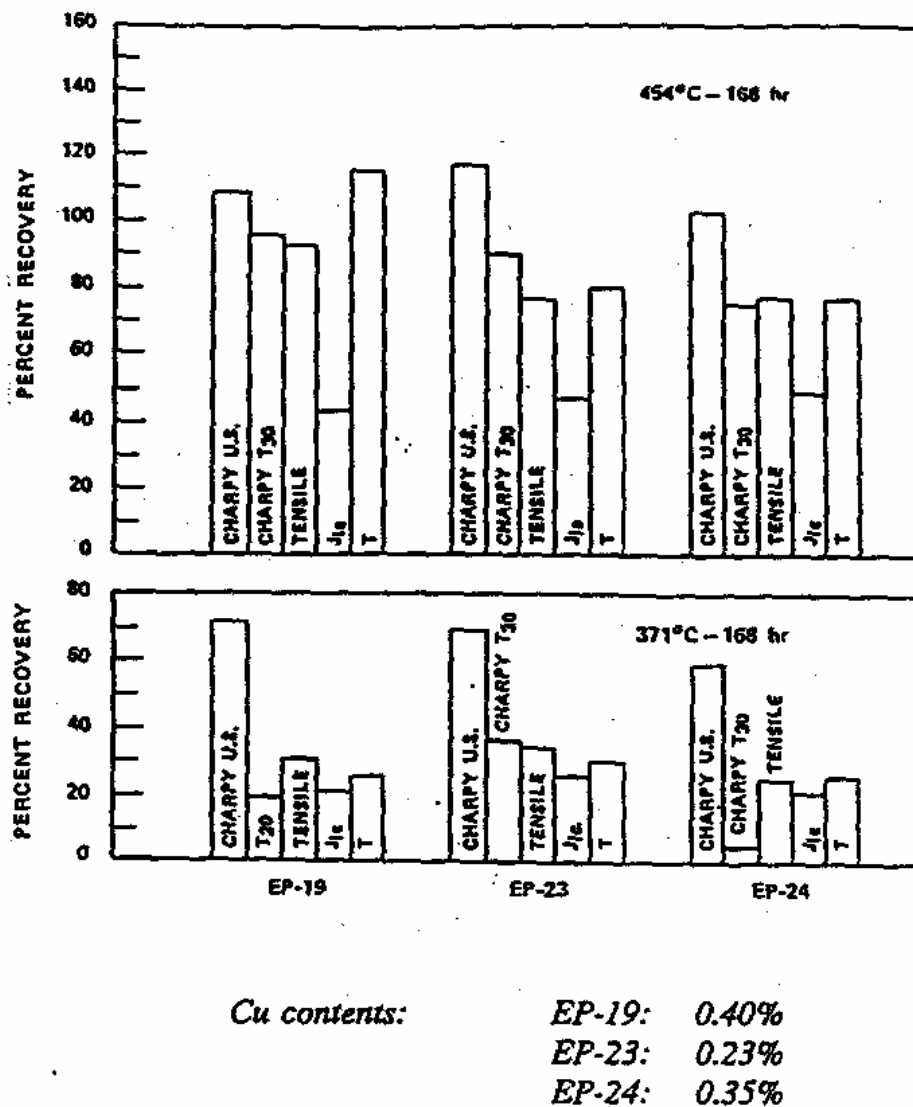


Fig. 2.16 The comparison of recovery annealing results for different welds (Ni=0.59%), [19]. Here, T is a tearing modulus

2.7 Mechanism of annealing

2.7.1 Radiation damage of reactor pressure vessel materials

The reason for material degradation, and in particular RPV materials, under irradiation is radiation damage of the atomic structure. Initial structure defects are vacancies and interstitial atoms. Further change of microstructure is a result of different diffusion processes. In RPV materials this change goes in two basic directions. Firstly, there is generation and growth of point defects complexes and at definite sizes they converge into dislocation loops and pores (these components in literature are termed as «matrix» defects). Secondly, there are accelerated processes

of reallocating of chemical elements in them. These processes are diffusion of copper and other elements (Ni, Mn, Si) from solid solution and segregation of some elements (first of all, phosphorous) on the defect areas. As a result there is formation of zones enriched with these elements, finally generating precipitates.

At a mechanistic level the mechanisms of radiation damage can be sub-divided in hardening and non-hardening. It is supposed, that «matrix» defects and precipitates, as “ostacles” for moving dislocations, are responsible for hardening mechanisms, and phosphorous segregation intra and on boundaries (decreasing their strength) is responsible also for non-hardening mechanisms. It is usually considered as indirect evidence of hardening mechanism a correlation (as a rule, linear) between T_k shift and yield strength increase. Absence of such correlation is considered as evidence of non-hardening mechanism.

Other radiation damage mechanisms can be considered; for example by the influence of impurity elements, i.e. mechanisms that are associated to copper and phosphorous and, -in case of nickel-steels- to nickel. Mechanisms connected with formation of «matrix» defects are considered additive and mechanisms of combined effects are expected.

The analysis of microstructure of RPV irradiated materials allows revealing some components that cause radiation damage. Primarily, these components are finely dispersed (1-3 nm) precipitates (or impurity clusters) of high-density (up to 10^{24} m^{-3}), [22]. Chemical analysis of the segregation reveals a special role of copper in their formation. It is supposed that the impurity clusters contribute considerably to hardening of RPV materials at rather low doses, such as $(3-5) \times 10^{23} \text{ m}^{-2}$. In this case their volumetric share reaches saturation by depletion of solid solution. At higher doses some decrease of their contribution to hardening is possible, because of growth process. In some works, [22-25], studies of morphology and chemical composition of these clusters in different RPV materials were carried out using atomic probe field

ion microscopy (APFIM), small angle neutron scattering (SANS) and electron microscopy. It was established that copper atoms are in the center of impurity clusters; other elements (Ni, Mn, Si) are uniformly distributed or located at the periphery. As a rule, in materials with sufficient phosphorous contents, the clusters are decorated by this element. The elements content in impurity clusters are considerably higher than in the ferrite matrix (mostly with copper and phosphorous). Iron atoms ensure the concentration element balance. Concerning kinetics of formation of impurity clusters there are different hypothesis. According to recent observations, all nucleating centers of impurity clusters occur at the initial stage of irradiation (up to fluence $\sim 10^{22} \text{ m}^{-2}$), their later density remains constant and mean size grows up to saturation. The Odette model was designed on the base of this kinetics [26], describing the depletion of solid solution of copper atoms. Other conclusion have been drawn using recent studies made by the APFIM [22] method: the impurity clusters occur in short time, presumably, in atomic collisions cascades, and do not change during further irradiation, however, their density continuously increases with radiation dose up to saturation. It should be noted that in any case the dose dependence of hardening caused by impurity clusters, will be of the same character; more volumetric share of impurity clusters implies more hardening up to saturation at copper contents comparable to its limit solubility in ferrite matrix at the temperature of irradiation.

To observe directly such dependence experimentally is rather difficult, as during irradiation there are several mechanisms that affect hardening simultaneously. However, in some studies concerning irradiation of Fe-Cu binary alloys and commercial RPV steels, the results of which are reported in the review [27], it was observed dose dependence of hardening, Fig. 2.17, which can be assumed to be a proof of described kinetics of impurity clusters effect.

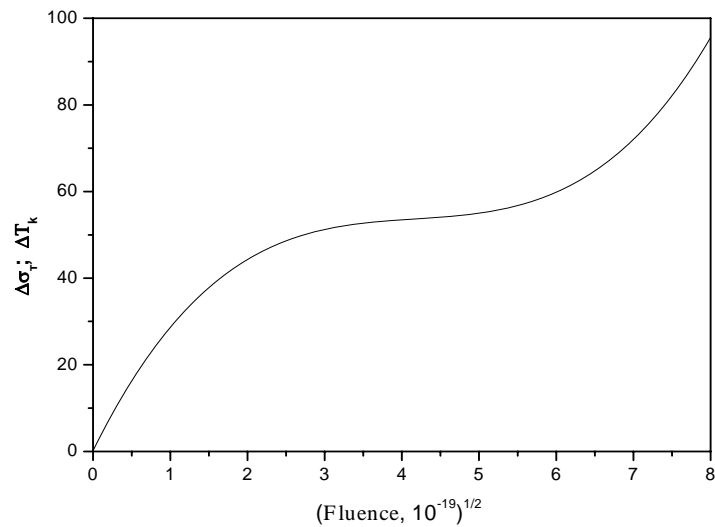


Fig. 2.17 Schematic dose dependence of yield strength increase and transition temperature shift for steel A302B, [30]

It is supposed, that the initial increase of the curve and saturation are attributed to impurity clusters influence, a segment of secondary increase is attributed to other mechanisms.

According to experimental data [8], ΔT_k dependence on copper content from 0.1 up to 0.35 atomic % is practically linear at any fluence. Out of this content interval, there are indications of the existence of lower threshold for copper influence and upper threshold of saturation of such influence. The upper threshold is connected to limit solubility of copper in solid solution at the temperature of the final heat treatment condition (for RPV materials of WWER-440 it is ~ 665 °C).

In literature, «matrix» defects are divided into unstable and stable. Vacancy and impurity complexes, which are formed in atomic collisions cascades, are of unstable type. The lifetime of these complexes is limited, therefore they saturate fast and can contribute to material hardening considerably only at early stages of irradiation. It is reasonable to presume, that after vacancies decrease there is occurrence of impurity aggregations, which are the nucleating centers of copper enriched clusters mentioned above.

Vacancy and interstitial atom clusters that are capable to accumulate and grow steadily during irradiation are referred to as stable «matrix» defects. After irradiation times longer than safe life of reactor pressure vessel, they can converge, finally, into classic dislocation loops and pores. Apparently, these stable «matrix» defects contribute mainly to hardening kinetics of RPV materials at rather high radiation doses.

The relation for estimation of yield strength increase of the material caused by influence of defect clusters of j type (particularly, loops and pores) was proposed in [28]:

$$(\Delta\sigma_T)_j = \beta_j \cdot G \cdot b \cdot (2N_j \cdot d_j)^{1/2} \quad (2.4)$$

where G is shear modulus, b is Burgers' vector, N_j and d_j are volume concentration and mean diameter of clusters, respectively, and β_j is hardening factor.

Interpretation of phosphorous influence is more complex. It is agreed that phosphorous contributes in hardening mechanism, however, some test results cause reasonable arguments concerning the possibility of additional non-hardening mechanisms. It is proposed to separate the mechanisms of phosphorous influence into mechanisms of independent influence (formation of phosphorus enriched zones, phosphides, dislocation atmospheres, grain boundary segregation) and mechanisms of synergetic influence (by kinetics of copper precipitation and «matrix» defects). The indicated mechanisms of phosphorous influence on residual embrittlement for RPV materials are mentioned in [29-33]. On the basis of available information the only key mechanism caused by phosphorous influence, at the present time cannot be defined precisely. Therefore it is necessary to consider different mechanisms; in particular referring to the fact that in WWER-440 RPV materials the main mechanism of phosphorous influence is of the hardening type. At the same time the physical process depends on copper content in material. At high copper content in solid solution, during irradiation, the phosphorous contribution to hardening is defined by

its segregation on a surface of these clusters resulting in strengthening of their power as «obstacles" for dislocation moving. At low copper content the phosphorous contribution to hardening is connected, probably, with formation of phosphorous-containing impurity clusters and/or phosphides.

Discussion of the role of nickel in radiation damage, in a comparative exercise such as this, is difficult because of a shortage of surveillance data and comparative irradiations. There are test reactor data, [34], which show that above certain nickel content there is a marked sensitization. The role of nickel in irradiation effects is shown in Fig. 2.18, [35].

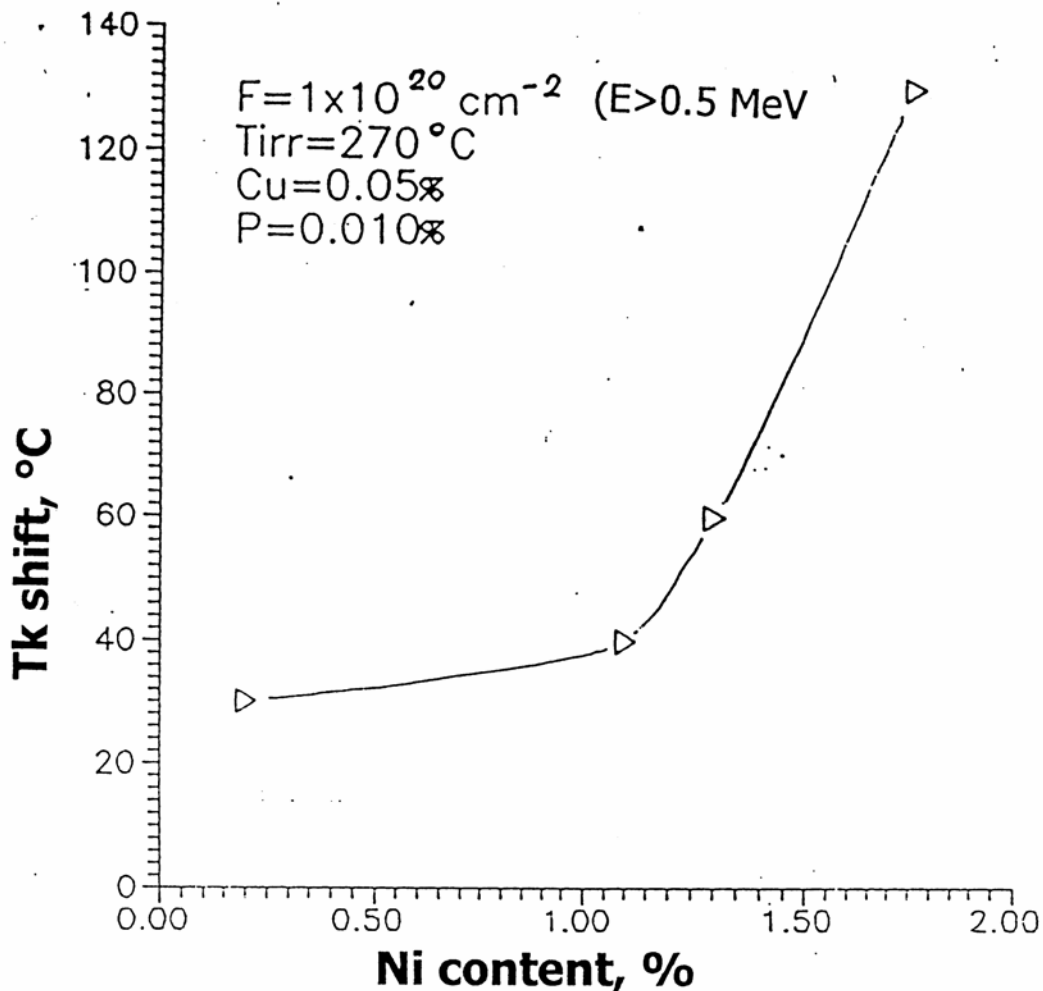


Fig. 2.18 Transition temperature shift with different nickel content but with 0.05 % copper and 0.010 % phosphorus irradiated to $1 \times 10^{24} \text{ m}^{-2}$, irradiation temperature 270 °C, [35]

2.7.2 Radiation defect elimination by annealing

The dramatic improvement in T_k and recovery of upper shelf energy which occurs as a result of post-irradiation annealing must be related to the fine-scale changes in microstructure. It was proposed that the annealing process promotes the coarsening of the irradiation induced copper clusters (precipitates). This coarsening process is diffusion controlled and is accompanied by the dissolution of other copper rich features in the matrix. In [36] a model was proposed based on copper precipitate coarsening that predicts the observed recovery in yield stress and T_k shift for annealing temperatures between 350 °C and 450 °C. The model also accounts for the low re-embrittlement rates in annealed materials. The formation of discrete copper precipitates has been previously observed in model alloys aged at 550 °C, [37-39], but these features have not been documented in post irradiated annealed RPV materials. Thus, it is likely that the copper rich clusters will develop into discrete stable copper precipitates which should be amenable to detailed microstructural and micromechanical characterization. Annealing at temperatures 450-470 °C should also result in the elimination of most matrix damage due to irradiation.

Important results were obtained in [22] where microstructural changes due to annealing were characterized using the APFIM method. A typical Mn-Mo-Ni-weldwire/Linde 80 flux weld irradiated by fluence $3.5 \times 10^{23} \text{ m}^{-2}$ with the nominal copper level 0.3 wt % and 0.016 wt % of phosphorus was annealed at 454 °C for 168 hours and at 610 °C for 29 hours. The ferrite matrix composition of the annealed weld is presented in Table 2.1.

Table 2.1 The Mn-Mo-Ni –weld wire/Linde 80 flux weld ferrite matrix composition in the different conditions, [22]

	At. %							
	Cu	Ni	Mn	Si	P	C	Mo	Cr
As-stress-relieved	0.14±0.03	0.45	1.2	1.05	0.03±0.02	0.005	0.18	0.03
Irr., $3.5 \times 10^{23} \text{ m}^{-2}$	0.05±	0.57	1.08	1.22	0.013±	0.005	0.23	0.08
Irr + anneal., 454 °C	0.04±	0.51	1.10	1.10	0.03±	0.04	0.18	0.05
Irr + anneal., 610 °C	0.17±	0.58	0.85	0.90	0.01±	0.01	0.17	0.05

The results show that there are no significant changes in the matrix composition due to annealing at 454 °C. The copper content is still depleted to about 0.04 % similar to that observed after neutron irradiation. Only phosphorus and carbon are detected at higher levels (2.3 and 8 times, respectively) after the annealing treatment. However, none of the neutron-induced solute clusters observed in irradiated condition were detected in the annealed material. This implies that they either all dissolved during the annealing, or their number density decreased so much that they were not detected with the atom probe technique. In order to answer the question of how the copper had been redistributed, several additional specimens were observed using atom probe field ion microscope (APFIM). This permits the observation of the atomic plane by atomic plane evaporation of a large volume of the material. The volume examined was about a hundred times more than the volume sampled with the atom probe.

One of the APFIM experiment revealed the presence of a small dark-contrast area, which is characteristic of a copper precipitate. The size of this precipitate was estimated to be on the order of 5 nm. The selected area analysis of the remaining portion of precipitate gave the composition profile which indicated that the copper content is at least 60 %.

In addition, an enrichment of Ni and Mn solutes was observed at the interface. This could be explained by the rejection of some solute atoms in the core of the particle towards the interface during the growth process of a nearly pure copper precipitate. The observation of this large copper precipitate explains why the matrix is still copper-depleted despite the disappearance of the small neutron-induced clusters. The large amount of copper detected in this particle is in good agreement with the fact that the particle number density appears to be very low. Assuming that these large copper precipitates (5 nm in diameter) are from 60 to 100 % pure copper, the results of this study indicate that the precipitate density would be in the range of

1.5 to $2.5 \times 10^{22} \text{ m}^{-3}$. Moreover, the detected copper level in the matrix is exactly equal to the copper solubility limit for the temperature of 450 °C, [40].

After annealing at 610 °C for 29 hours the ferrite matrix composition is similar to the stress-relieved condition, Table 2.1. The copper content has increased from 0.05 % to 0.17 % which suggests that all the intragranular neutron-induced copper clusters have dissolved.

So, it was concluded in [22] that during the annealing at 454 °C the neutron-induced copper clusters evolve by an Ostwald ripening process. The smallest clusters dissolve and the growth of larger copper precipitates takes place. The intragranular neutron-induced phosphorus atmospheres observed in irradiated material seems to be dissolved due to annealing – the phosphorus matrix content after annealing is the same as in unirradiated condition, Table 2.1.

The dissolution of copper clusters and phosphorus atmospheres during annealing is in agreement with described above high recovery of mechanical properties. The low density of copper precipitates in the ferrite matrix after annealing has little influence on the mechanical properties of the recovered material.

It also can explain the difference in residual T_k shift dependence of PWR and WWER materials on phosphorus and copper. In PWR the range of copper is much larger than in WWER studied steels (from 0.10 up to 0.35 % and 0.18 %, respectively). The dependence of ΔT_{res} on copper content was detected for PWR metal.

In turn the phosphorus variation in WWER is 2.5 times larger than for PWR and the dependence of ΔT_{res} on phosphorus was observed for WWER steels.

Finally, if the copper and phosphorus contents in both types of steel are low, the nearly full recover of T_k could be obtained after annealing at 455-470 °C. If phosphorus and/or copper contents are relatively high the residual after annealing shift T_k irradiated by fluence $3.5 \times 10^{23} \text{ m}^{-2}$ for both materials could reach 30-40 °C.

3 Re-embrittlement rate and models

3.1 Evaluation of transition temperature of RPV base and weld metals after repeated irradiation, and annealing

The efficiency of recovery annealing and, consequently, the lifetime of operating after annealing RPV are defined by two factors: firstly, by the degree of T_k shift recovery or residual irradiation embrittlement value and, secondly, by the rate of irradiation embrittlement during re-irradiation. Hence the need to understand regularities in irradiation embrittlement of alternately irradiation and annealing, taking into consideration the possible repeated annealings of RPV.

There are several stages of solving this problem. On the first stage (during the 80's) a limited number of experiments were carried out.

The transition temperature shift behavior of PWR welds with high copper content 0.35 % under irradiation, annealing and re-irradiation is presented in Fig. 3.1, [41].

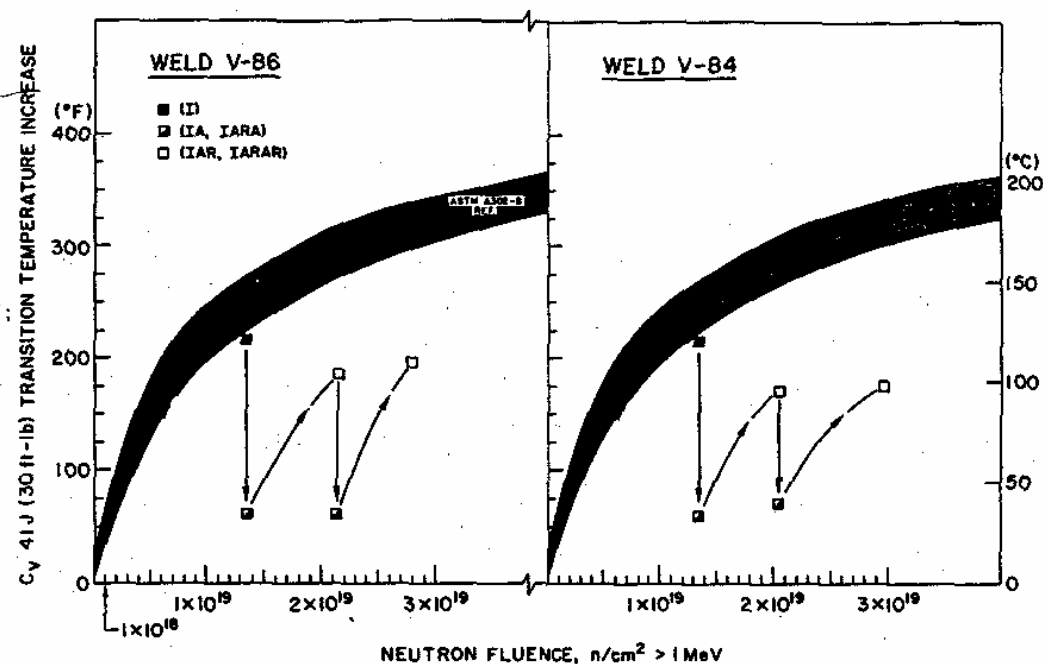


Fig. 3.1 The transition temperature shift behavior of PWR welds with high copper content 0.35 % under primary irradiation, annealings and re-irradiations, [41]

In another experiment after irradiation in Armenia NPP during one campaign, some weld surveillance specimens (0.028 % phosphorus and 0.18 % copper) were annealed at 420 °C for 150 hours, and half of them were loaded back into reactor for re-irradiation. After re-irradiation a part of the specimens was repeatedly annealed. The variation of T_k value in this test program is shown in Fig. 3.2. Weld metal having $\Delta T_F = 100$ °C caused by primary irradiation decreased to 30 °C, then increased by 90 °C after secondary irradiation to the same fluence. The efficiency of the secondary annealing turned out to be the same as that of the first - $\Delta T_F = 30$ °C.

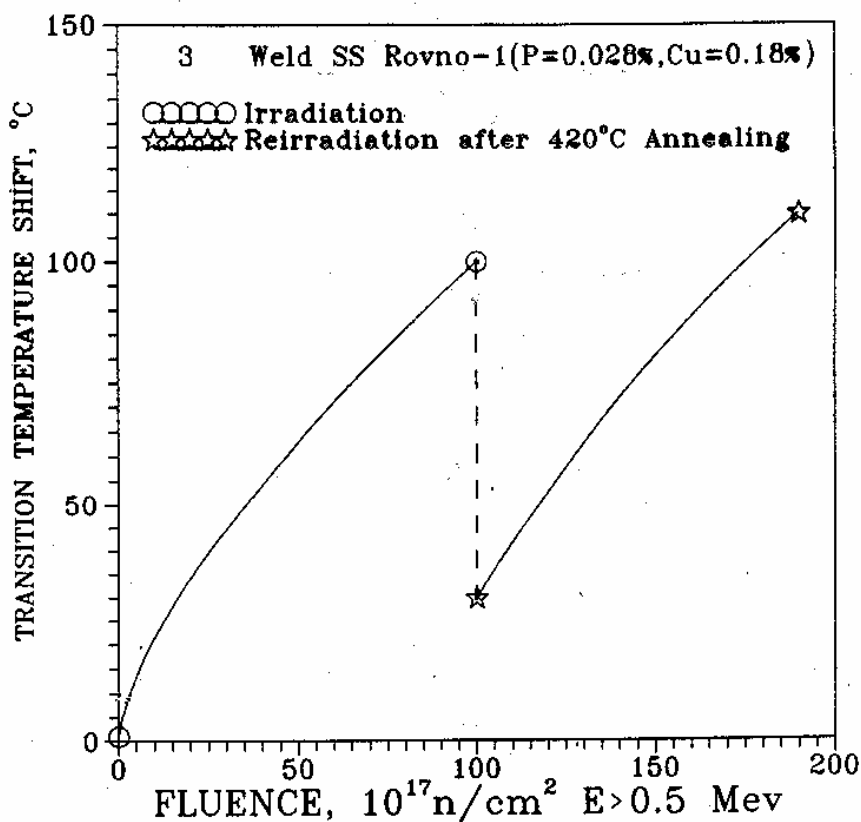


Fig. 3.2 The variation of WWER-440 weld T_k value under primary and repeated irradiations

From the data above the following conclusion was drawn. The rate of irradiation embrittlement during the re-irradiation is, at least, not higher than during the primary irradiation.

Taking into account both the limited quantity of experimental information and degree of conservatism in the evaluation of irradiation embrittlement, the so-called “conservative” scheme of T_k assessment was suggested for the prediction of WWER-440 RPV lifetime after annealing.

In accordance with this scheme, the rate of irradiation embrittlement during irradiation is the same as during the primary irradiation. So T_r during the re-irradiation is calculated by the formula:

$$T_r = T_{k0} + \Delta T_{res} + A_F \cdot F_r^{1/3} \quad (3.1)$$

where A_F is the coefficient of irradiation embrittlement and F_r is the neutron fluence during re-irradiation.

This formula has been applied in the assessment of irradiation lifetime for WWER-440 pressure vessels annealed from 1987 until 1992, [7].

However, this approach contradicts the mechanistic concepts concerning the nature of irradiation embrittlement of materials because it does not follow adequately the processes of radiation damage and annealing of RPV materials. For these reasons, experiments aimed at evaluating the factors in material behavior during re-irradiation were continued.

Fig. 3.3 presents the 3 traditional ways for irradiation embrittlement calculation used until recent different approaches to determine the irradiation embrittlement during re-irradiation.

For WWER materials the initial irradiation embrittlement is described by formula:

$$\Delta T_F = A_F \cdot F_r^{1/3} \quad (3.2)$$

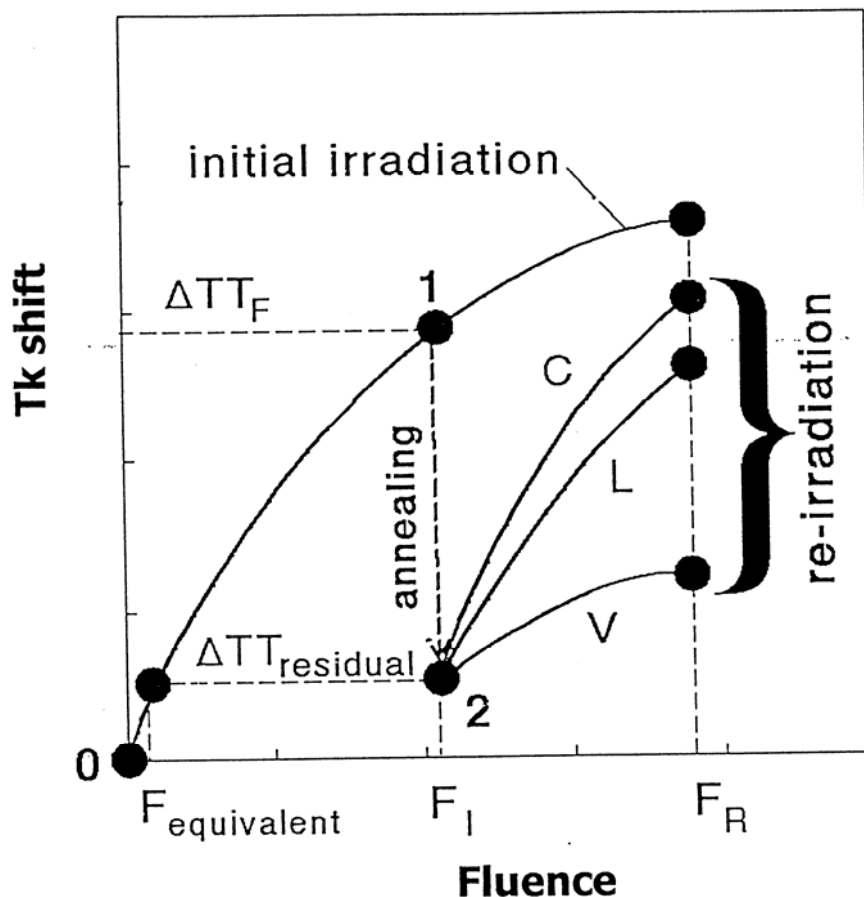


Fig. 3.3 Scheme of embrittlement of reactor pressure vessel under re-irradiation of sequentially irradiated and annealed materials

The curve C (conservative shift) corresponds to a conservative evaluation of irradiation embrittlement during re-irradiation, which was adopted for WWER-440 until 1992. The T_k shift during re-irradiation is defined by (3.1).

The curve L (lateral shift) demonstrates the scheme, in which irradiation embrittlement during re-irradiation is defined by a lateral shift of the curve, describing the primary dependence of ΔT_F on neutron fluence. Here ΔT_r is defined by:

$$\Delta T_r^l = A_F \cdot (F_{equiv} + F_r)^{1/3} \tag{3.3}$$

Where $F_{equiv} = (\Delta T_{res} / A_F)^3$ and $F_r = F_2 - F_1$.

The curve V (vertical shift) shows the results of repeated irradiation embrittlement for vertical shift of the primary dependence. So:

$$\Delta T_r^V = \Delta T_{res} + A_F \cdot (F_2^{1/3} - F_1^{1/3}) \quad (3.4)$$

The comprehensive experimental programs were developed to discover a suitable scheme, adequately taking into account the changes in material during re-irradiation. The results are described in [7, 20, 42-43]. The basic results of [20] are presented in Fig. 3.4.

3.2 Comparison of the experimental data with traditional models of re-embrittlement assessment

Comparisons of experimental data with conservative, lateral and vertical shift models were performed on the base of WWER material investigation results. Seven WWER-440 welds with different phosphorus (from 0.010 % up to 0.035 %) and copper (from 0.11 % up to 0.18 %) were irradiated in surveillance channels with irradiation temperature 270 °C, annealed at 340, 380, 420 and 460 °C. After annealing part of the specimens were re-irradiated. Some results are presented in Fig. 3.5.

Fig. 3.6 and Fig. 3.7 show the comparison between predicted and observed data at different annealing temperatures. The quality and accuracy of experimental data fitting have been characterized using the correlation coefficient (R_{xy}), variance (δ) and determination coefficient (R^2).

Evidently for WWER-440 materials, at annealing temperature 340 °C the most appropriate approach for prediction of the secondary irradiation embrittlement is of the vertical shift, although this overestimates ΔT during re-irradiation. Besides, the determination coefficient, characterizing the model quality, is too low in this case. At temperature 380 °C the amount of data is not enough to make a conclusion.

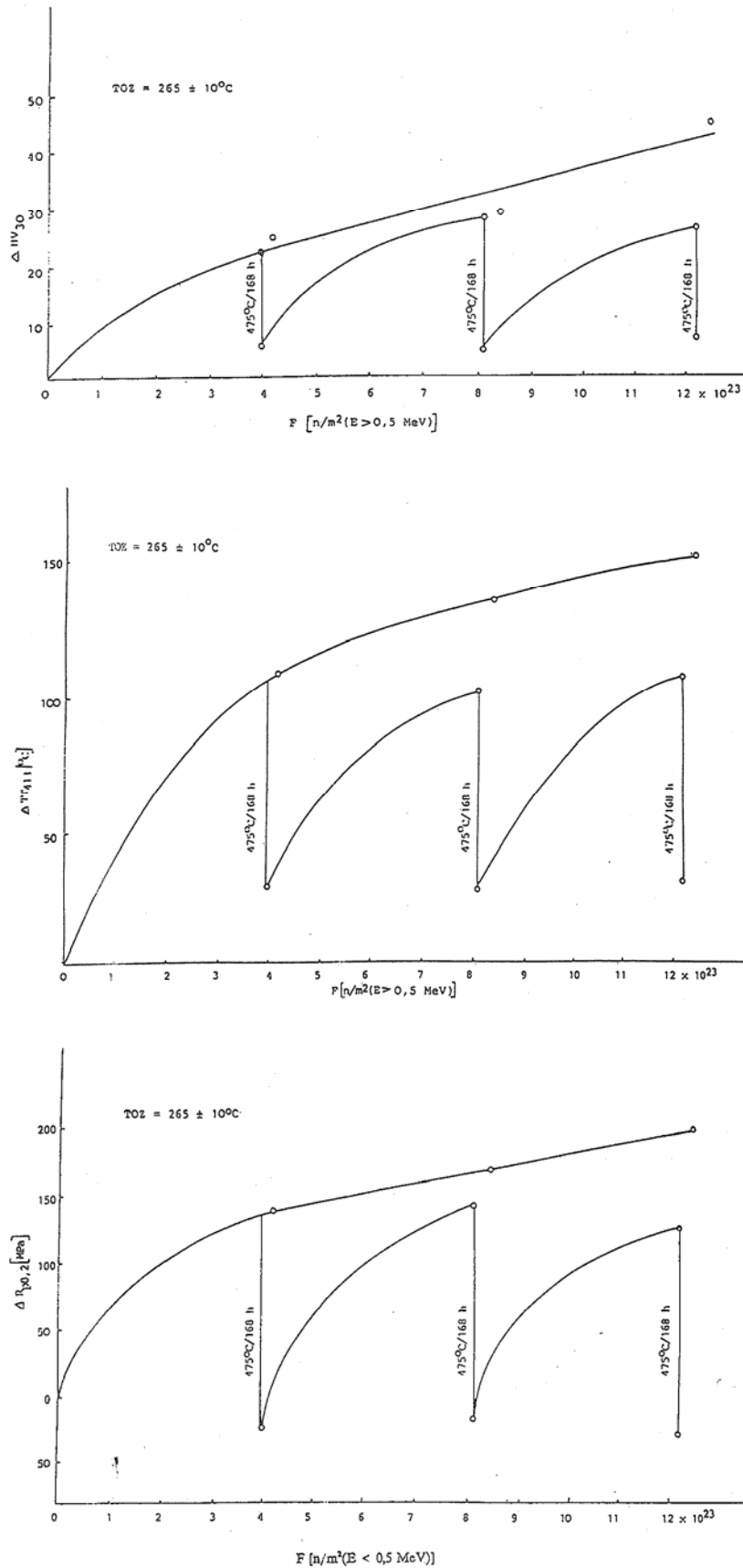


Fig. 3.4 Hardness, yield strength and transition temperature changes of irradiated, annealed and – re-irradiated WWER-440 weld metal, [20]

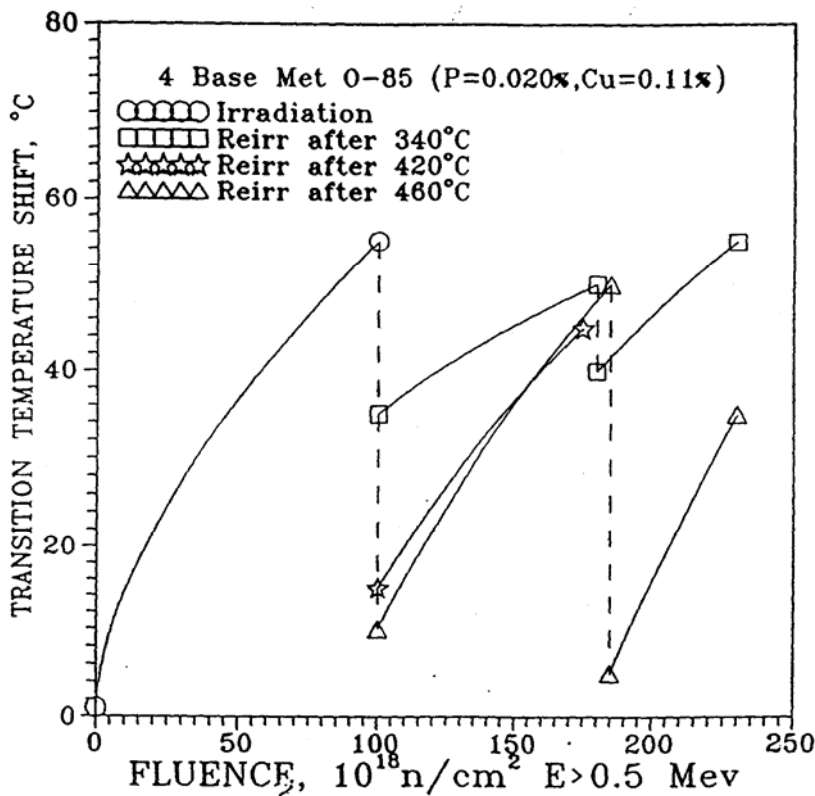
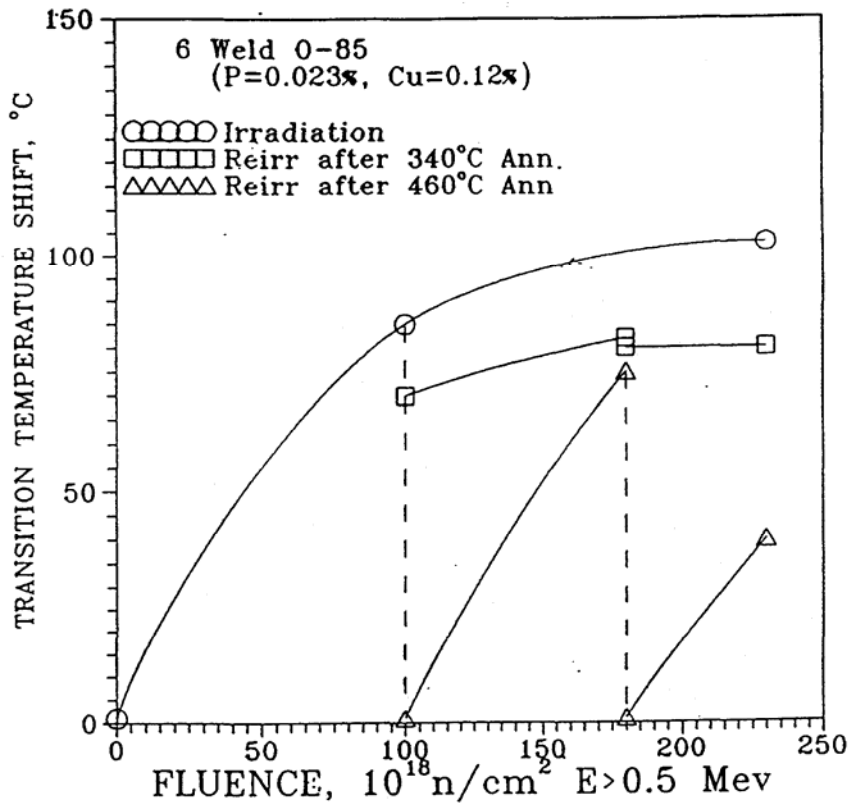
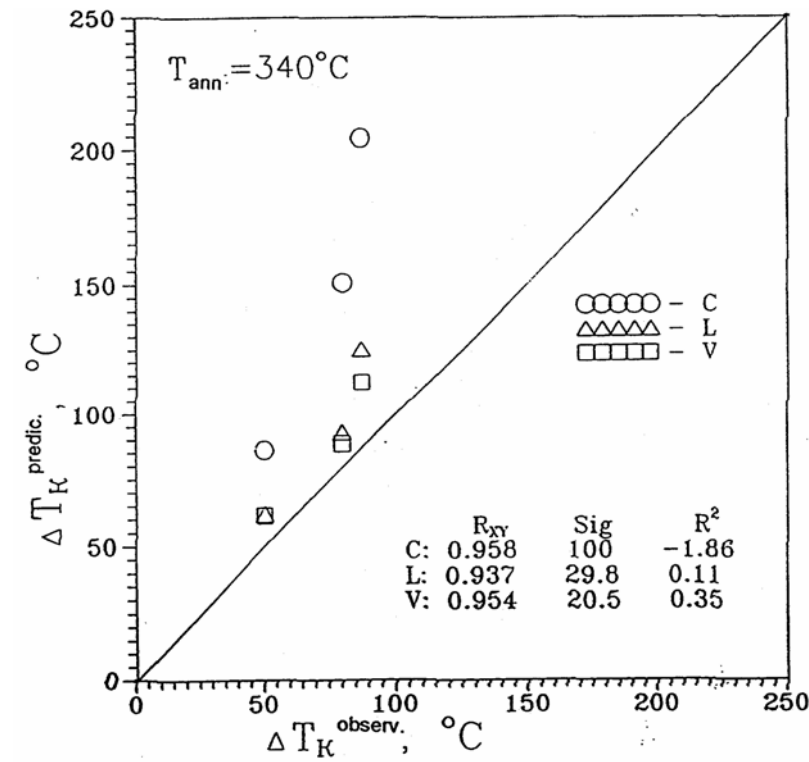
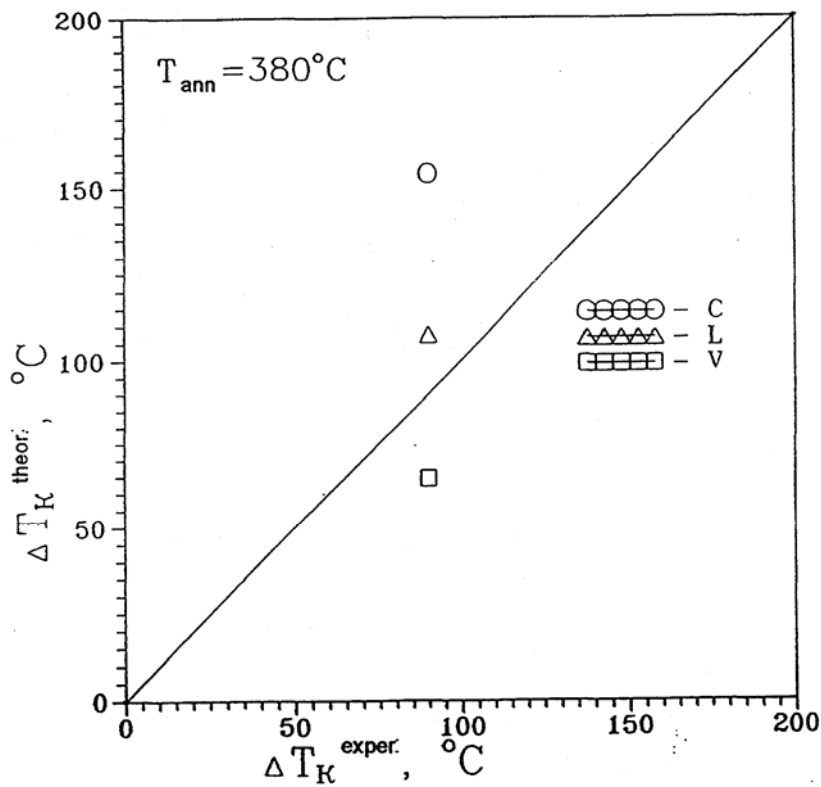


Fig. 3.5 Changes of transition temperature shift of WWER-440 welds under irradiation, annealing and re-irradiation



a)



b)

Fig. 3.6 Comparison between observed T_k shifts due to re-irradiation after annealing at 340°C (a), 380°C (b) and those predicted by conservative, lateral and vertical shift evaluation models

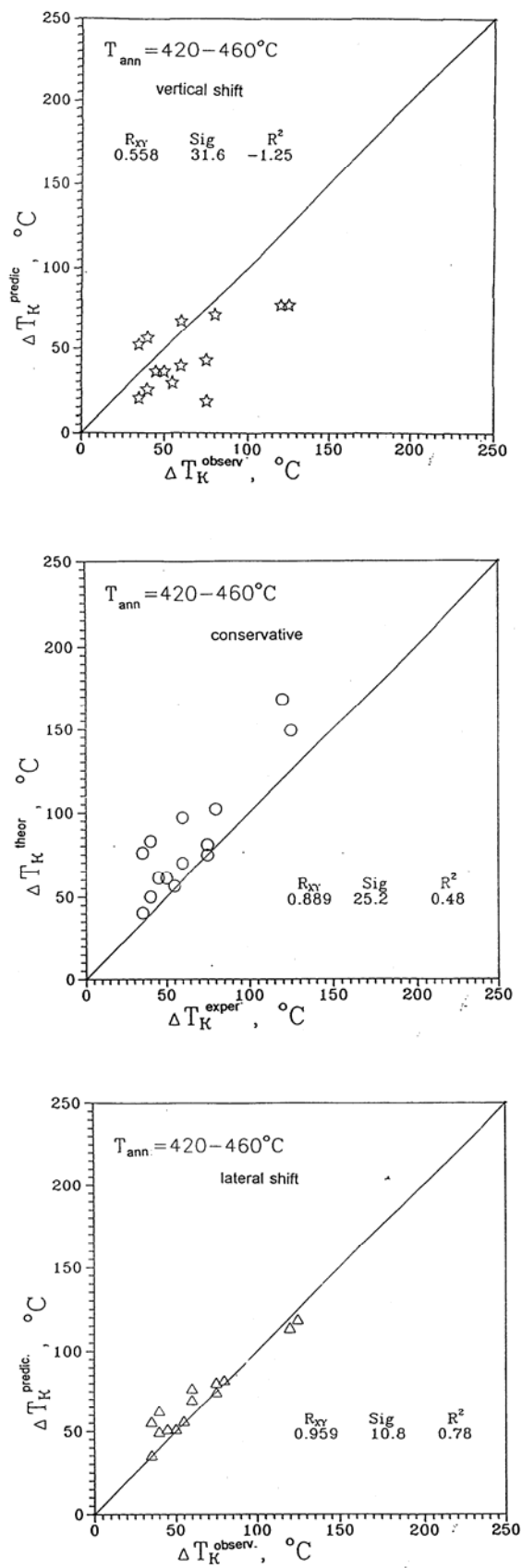


Fig. 3.7 Comparison between observed T_k shifts due to re-irradiation after annealing at 420-460 °C and those predicted by conservative, lateral and vertical shift evaluation models

At annealing temperature higher than 420 °C the maximum values of correlation coefficient are attained for the lateral shift: 0.77 at 420 °C and 0.92 at 460 °C.

Thus, as for the traditional models, the best method for the prediction of WWER - 440 weld re-irradiation after annealing at 420-460 °C, is the evaluation corresponding to the horizontal shift of initial dependence of irradiation embrittlement, that is eq.3.3 which can be transformed to (3.5) after substitution of F_{equiv} .

$$\Delta T_r^l = \left(\Delta T_{res}^3 + A_F^3 \cdot F_r \right)^{1/3} \quad (3.5)$$

The results of PWR re-embrittlement studies also support the lateral shift model assessment [41, 43]. However the comparison of WWER results above with the data from [16], Fig. 3.8 reveals a difference in the WWER and PWR material embrittlement rate under re-irradiation. In the view of above for WWER the higher is the annealing temperature the higher is the re-embrittlement rate, Fig. 3.9. For PWR steels, on the contrary, the re-embrittlement rate after annealing at 399 °C is higher then at 454 °C. The interpretation of this will be given by the authors of this survey in § 3.3.

3.3 The assumed mechanism of RPV material re-embrittlement

The results of the studies carried out to the present time, including the evaluation of T_k using boat samples taken from operating WWER-440 RPVs after annealing, have shown that the material behavior under re-irradiation is more complex than expected. The reason of difference in material behavior under primary irradiation and re-irradiation is treated in the following.

The microstructure investigation results demonstrate that copper clusters that are formed in material under primary irradiation are not recovered during annealing to a structure of solid solution as in the unirradiated state.

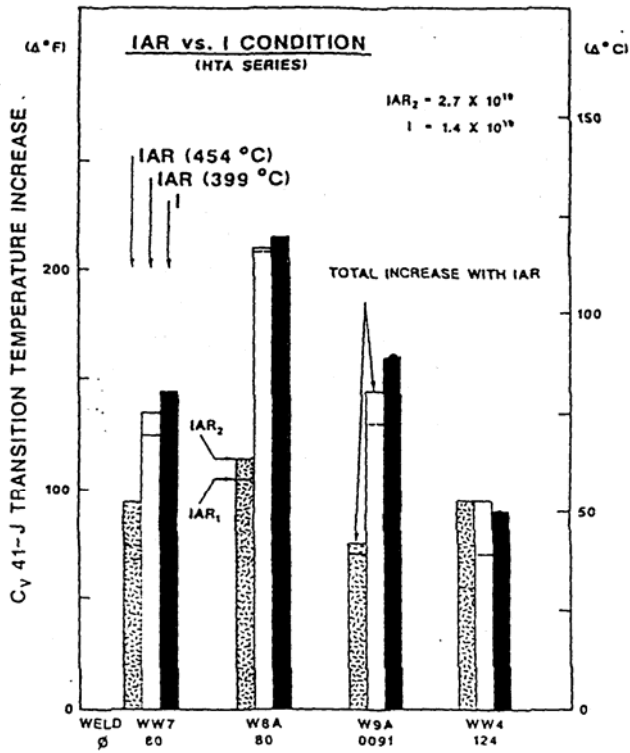


Fig. 3.8 Weld metal transition temperature shift observed after re-irradiation to a fluence of $2.7 \times 10^{23} \text{ m}^{-2}$ (IAR₂) or $1.8 \times 10^{23} \text{ m}^{-2}$ (IAR₁) (> 1 MeV) vs. first exposure cycle. The left-hand and center bars indicate the total T_k shift with IAR treatment, [16]

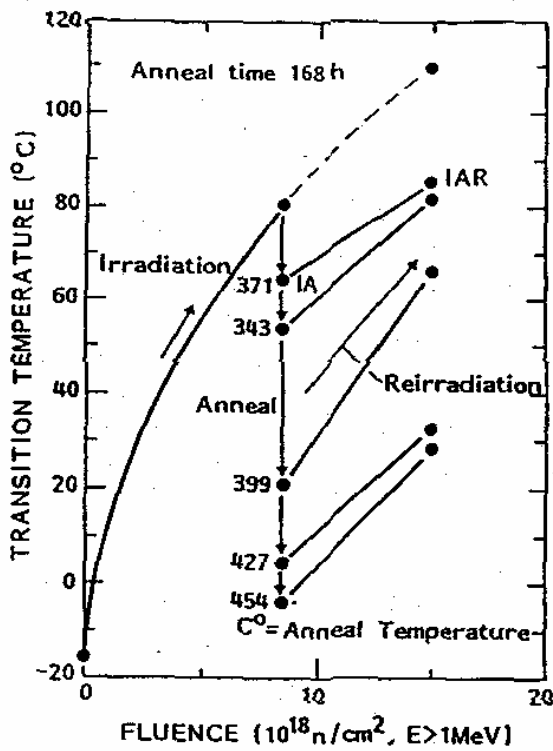


Fig. 3.9 Effect of re-irradiation after various annealing on the transition temperature shift of weld EP-19 (Cu=0.40 %, Ni=0.59 %), [44]

Annealing of the material at 455-470 °C causes changes in the system of copper clusters, namely the growth of their size and the decrease of their density. This low density of copper precipitates has little influence on the mechanical properties of the recovered material. So, if annealing leads to a low density of small, nearly pure copper precipitates [22] and low matrix copper content, further neutron irradiation of this neutron irradiated and annealed material should not produce less transition temperature shifts as under the primary irradiation. As follows from the APFIM results [22] the phosphorus content in the matrix after annealing is recovered approximately to the level of unirradiated material. It means the phosphorus influence for the material embrittlement under the re-irradiation is substantial.

The results of the investigation of irradiated and re-irradiated WWER-440 weld with 0.03 % phosphorus and 0.17 % copper are presented in Fig. 3.10 and Fig. 3.11 [46].

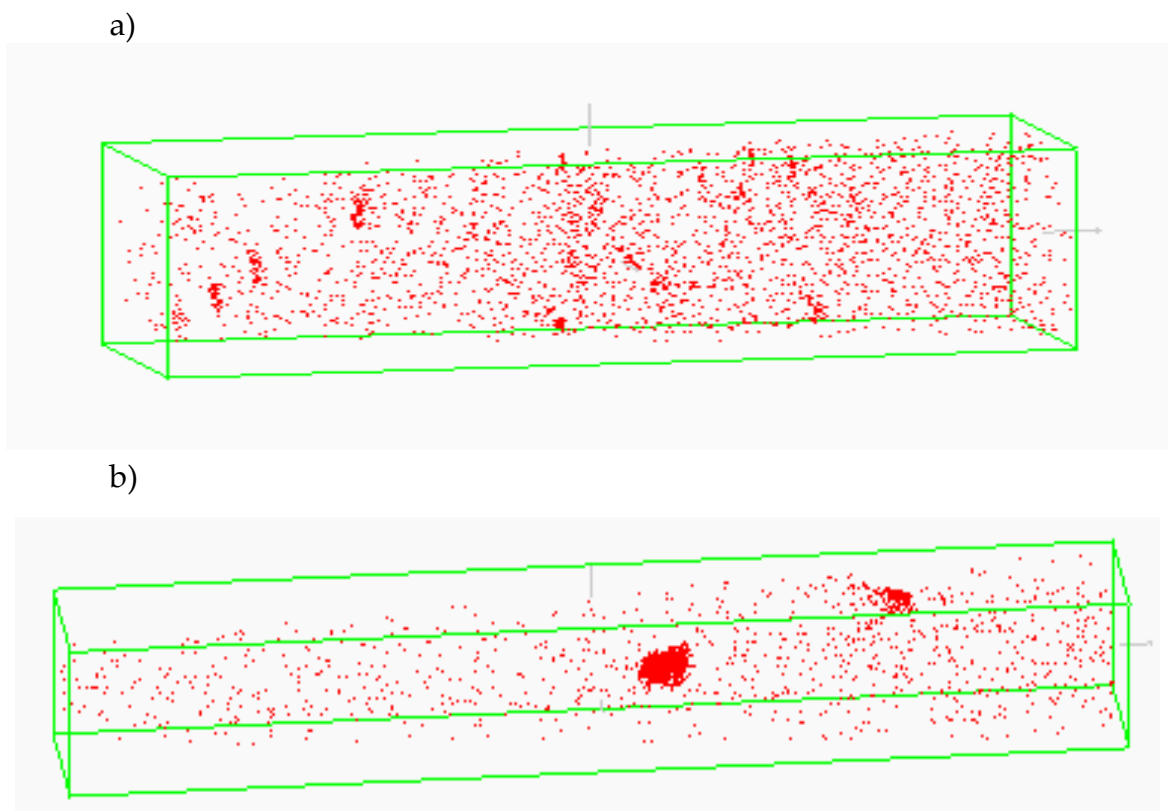
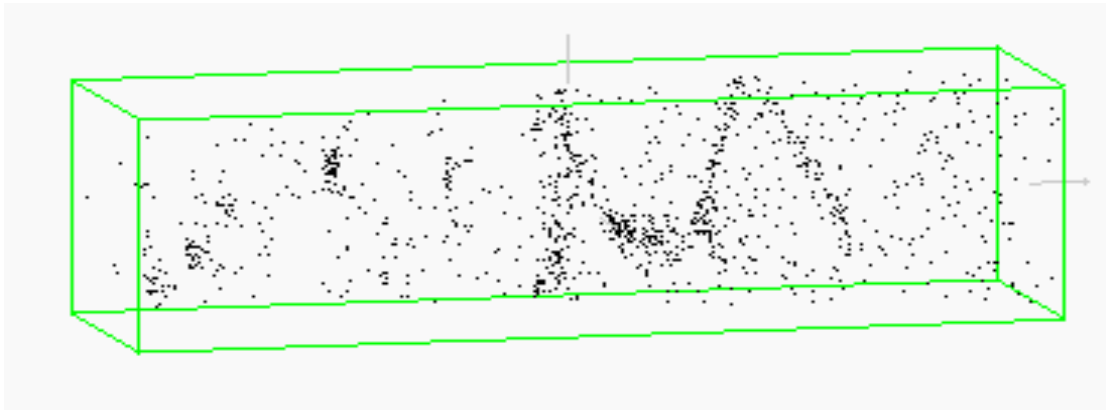


Fig. 3.10 Distribution of copper in irradiated (a) by neutron fluence $9 \times 10^{23} \text{ m}^{-2}$ ($E > 0.5 \text{ MeV}$) and re-irradiated (b) after annealing at 475 °C by fluence $1.5 \times 10^{23} \text{ m}^{-2}$ WWER-440 weld, [45]

a)



b)

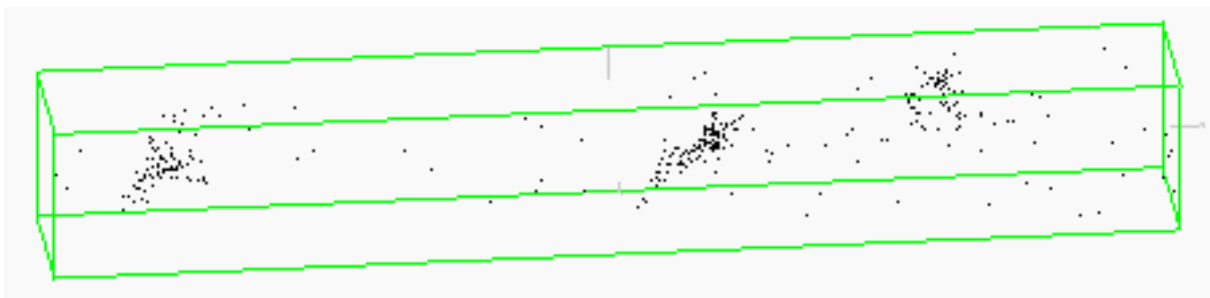


Fig. 3.11 Distribution of phosphorus in irradiated (a) by neutron fluence $9 \times 10^{23} \text{ m}^{-2}$ ($E > 0.5 \text{ MeV}$) and re-irradiated (b) after annealing at $475 \text{ }^\circ\text{C}$ by fluence $1.5 \times 10^{23} \text{ m}^{-2}$ WWER-440 weld, [45]

Copper precipitates as result of annealing are shown in Fig. 3.10(b). The structure of phosphorus distribution looks to be a similar for both irradiated and re-irradiated materials. Some difference for Fig. 3.11(a) and (b) could be due to the difference 6 times in neutron fluence values for primary irradiated and re-irradiated specimens.

The above microstructural results are in good agreement with the WWER-440 mechanical testing results, presented in Fig. 3.12 and Fig. 3.13.

It is seen from Fig. 3.12 and Fig. 3.13 that for WWER-welds there is a very weak dependence of the re-embrittlement on copper and a strong dependence on phosphorus.

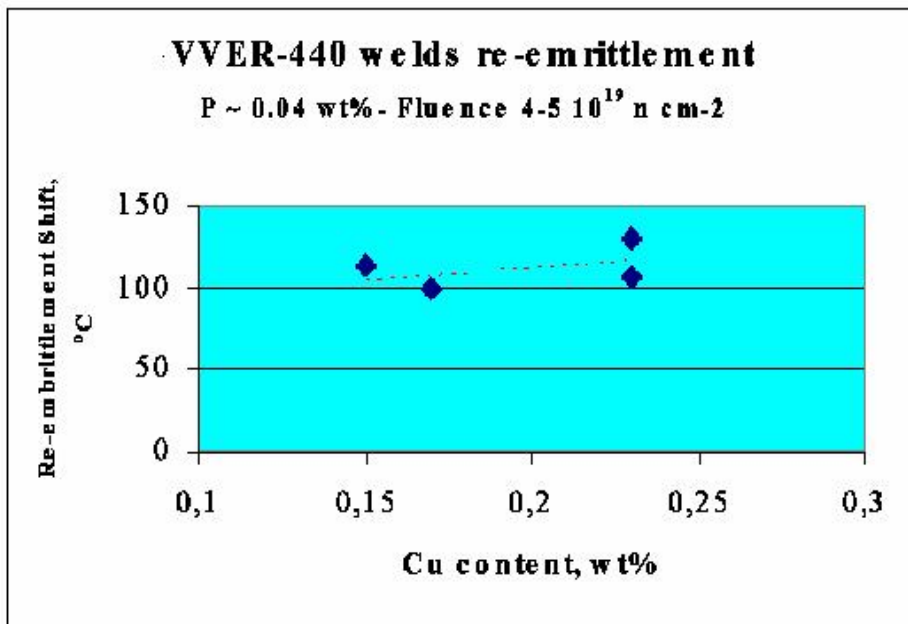


Fig. 3.12 The dependence of WVER -440 weld transition temperature shift under re-irradiation (after annealing at 460 °C) on copper content in the steel

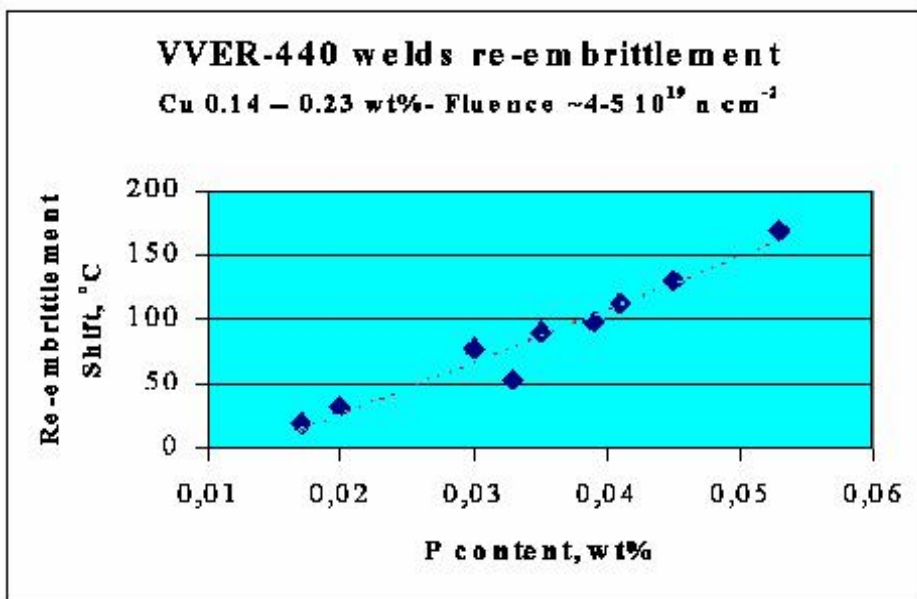


Fig. 3.13 The dependence of WVER -440 weld transition temperature shift under re-irradiation (after annealing at 460 °C) on phosphorus content in the steel

Turning back to the difference observed above in re-embrittlement rates for WVER and PWR materials it is necessary to note the important distinction in phosphorus and copper content levels between “old” WVER and PWR.

For PWR welds the main damaging element is copper (up to 0.4 mass. %). Maximum phosphorus content is ~0.02 mass. %. It seems that the higher the annealing temperature the more copper turns into precipitate and less copper remains in matrix. Matrix copper influences irradiation embrittlement. If the phosphorus level is less than 0.02 mass % it does not influence so much to the embrittlement. It means in both cases (primary irradiation and re-irradiation) the copper matrix level controls the embrittlement rate. For re-irradiation the matrix copper level depends on the annealing temperature. So, the higher the annealing temperature the lower is the re-embrittlement rate for PWR welds with high copper content.

In the case of WWER “old” welds, the contribution of copper in embrittlement is relatively small due to the low copper content (less than 0.2 mass %). The phosphorus level is high (up to 0.045 mass %) and according to the microstructural and mechanical testing results above, both embrittlement and re-embrittlement depend on phosphorus.

The higher the annealing temperature the more phosphorus turns back to the matrix and during the re-irradiation segregates to the defects damaging the material. If the annealing temperature is relatively low (~340 C) to eliminate the radiation defects created with the phosphorus participation, and neutron fluence had been enough to the copper precipitation saturation, then the re-embrittlement rate is to be minimal. It means that for WWER welds with high phosphorus, the higher is the annealing temperature the higher is the re-embrittlement rate.

3.4 Semi-mechanistic models for re-embrittlement assessment

The traditional models above are not able to describe the peculiar behavior of re-embrittlement after annealing of high phosphorus steels, like some WWER-440 high P welds, Fig. 3.14, [46]. What is in fact observed in the mentioned steels is that the re-embrittlement kinetic after annealing is different from the kinetics of primary embrittlement. After annealing the embrittlement seems to start with a certain delay and afterwards increases rapidly.

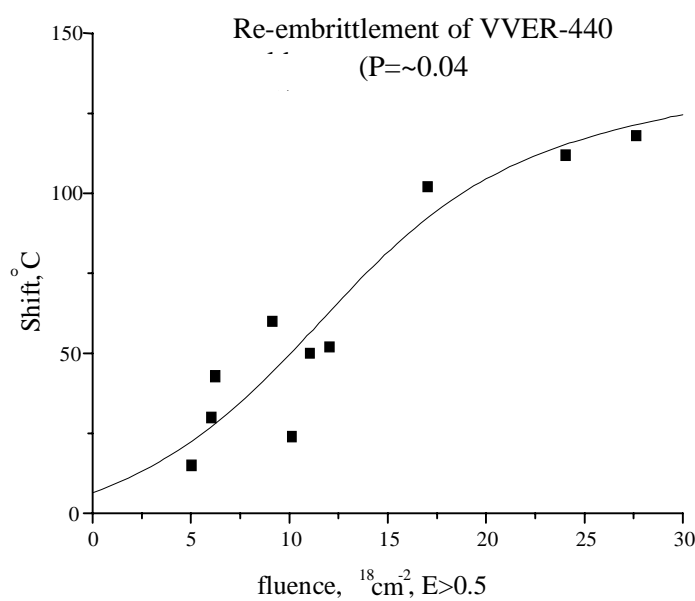


Fig. 3.14 The dependence of transition temperature shift on re-irradiation fluence, [46]

The first attempt to deviate from traditional models was described in [42], the so called “Kohopää model”.

In this approach the solute P is considered the governing active element in the post annealing re-embrittlement. It is supposed that the Cu-precipitates are growing and not dissolving during annealing. Accordingly the volume of dissolved Cu is much smaller in the post-annealing re-irradiation than in the 1st irradiation cycle.

When assuming that P is mostly dissolved during thermal annealing, and that Cu remains bound in precipitates, the re-embrittlement of Cr-Mo-V steels is mainly governed by P rich features. According to these assumptions the re-embrittlement (ΔT_{re}) may be expressed in a general form by the following equation:

$$\Delta T_{re} = \Delta T_{res} + \Delta T_{dam} + \Delta T_p \quad (3.6)$$

Where ΔT_{res} is the post annealing residual embrittlement and ΔT_{dam} and ΔT_p are transition temperature shifts due to matrix damage and P precipitation respectively. This approach is valid only when the fluence during the previous irradiation cycle is high enough, the irradiation temperature is around 260-300 °C and the irradiation time is long enough for formation of fully evolved Cu-rich precipitates. The residual transition temperature with an annealing temperature of 475 °C is estimated at +20 °C according to findings in this study and to findings by Amayev et al., Brumovsky et al., and Valo et al.

The damage factor ΔT_{dam} was estimated from the shift in transition temperature of weld 1 in IAI₁ condition. Since the P-content is varying it is possible to exclude the effect of P and the residual shift is considered as above. Fisher et al [29] have determined that the matrix damage has square root dependence. Brumovsky and Pav /9/ studied some relatively clean Cr-Mo-V steels and concluded that the fluence exponent value varies between 0.55 and 0.62. Also the reported results of Ozwald and Trampus /10/ indicate a square root of fluence dependence for Cr-Mo-V steels with relatively low Cu- and P-contents. Accordingly the damage contribution can be mathematically solved and expressed as equation (3.7):

$$\Delta T_{dam} = 2.4 \cdot (F_{re})^{0.5} \quad (3.7)$$

where F_{re} is the post annealing fluence expressed in 10^{22} m^{-2} ($E > 1 \text{ MeV}$).

The contribution of the P-factor can be described by the expression:

$$\Delta T_p = A \cdot P \cdot (F_{re})^n \quad (3.8)$$

Where A is a constant that depends on the irradiation and the annealing conditions, F_{re} is the post annealing fluence expressed in 10^{22} m^{-2} ($E > 1 \text{ MeV}$), n is the fluence exponent for the P contribution and P is the phosphorus content of the material. Based on the results from the re-embrittlement studies on weld 1 in the IAI conditions, the exponent n seems to be best described by a value of 0.1 and the average value for the factor A is 1,400. The re-embrittlement rate may accordingly be expressed by the following equation:

$$\Delta T_{re} = \Delta T_{res} + 2.4 \cdot (F_{re})^{0.5} + 1400 \cdot P \cdot (F_{re})^{0.1} \quad (3.9)$$

The re-embrittlement behaviour is schematically shown in Fig. 3.15. In Fig. 3.16 the results of the re-embrittlement shifts based on the transition temperature criteria ΔT_{47J} and ΔT_0 is compared with the corresponding dependence equation (3.9). It can be seen, that the model function shows a rather good average dependence of the re-embrittlement behaviour of the welds. The amount of data included in this study is, however, too limited to allow satisfactory checking the model for re-embrittlement. Data in the open literature concerning re-embrittlement behaviour of the Cr-Mo-V do not contain enough details for effectively checking this model.

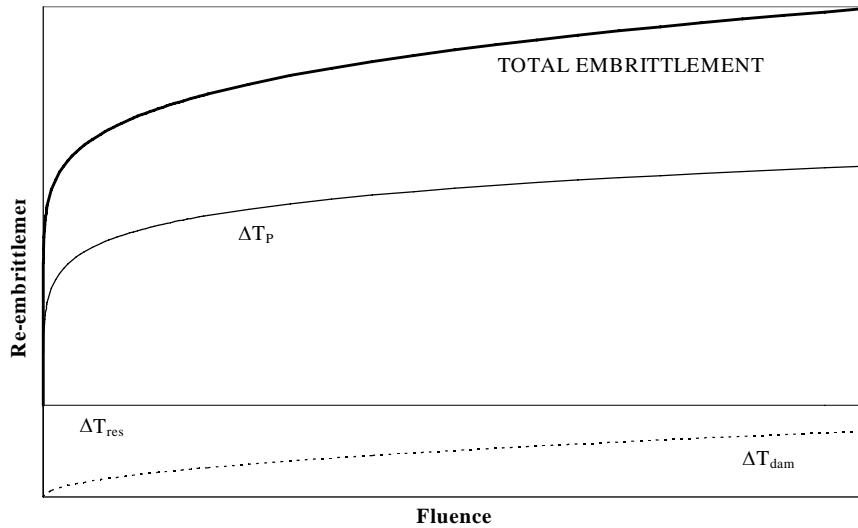


Fig. 3.15 Schematic of embrittlement and re-embrittlement

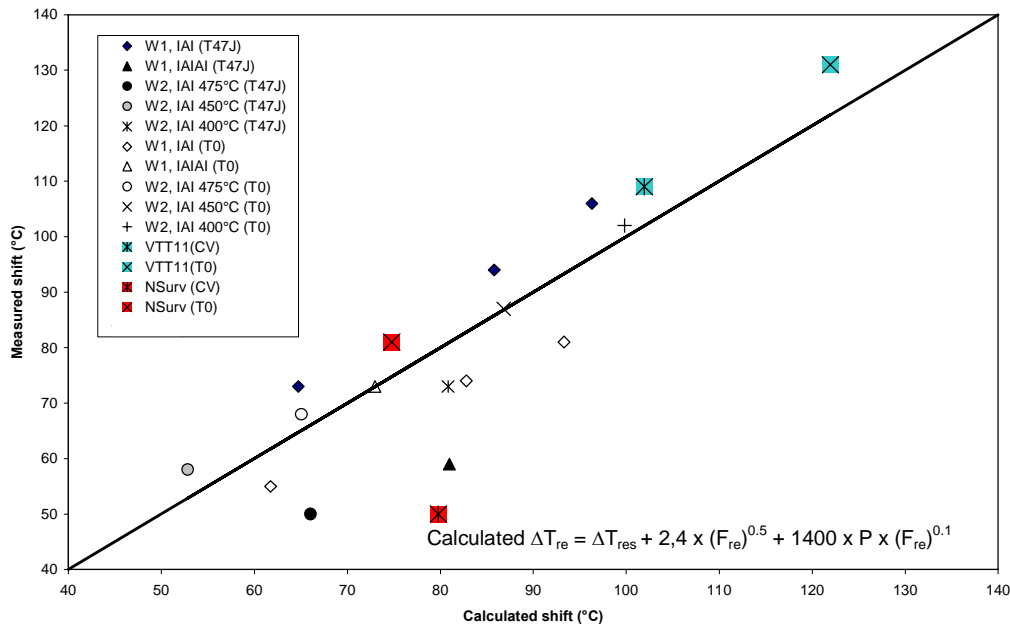


Fig. 3.16 Comparison of experimental and predicted re-embrittlement shifts [34]

A so called semi-mechanistic model for re-embrittlement assessment was proposed in [47]. In the framework of this model it was postulated that the basic mechanism of radiation embrittlement (primary) of steels and welds is based on three major contributions to damage: direct matrix damage, precipitation (mainly copper) and element segregation (mainly phosphorus), table 3.1. The semi-mechanistic model is based on these key mechanisms.

The advantages of the semi-mechanistic model, when compared to non-mechanistic models, are that it allows improved fitting of data and permits the visualization of the relative contribution of the various damage components.

Table 3.1 Embrittlement mechanisms considered in semi-mechanistic model, [47]

Embrittlement mechanism	Remarks
Direct matrix damage	Due to neutron bombardment
Precipitation hardening the matrix	Cu is the leading element
Segregation	P is a recognized segregating element

The effect of the various embrittlement parameters is considered to be additive to the total damage expressed in terms of ΔT_{shift} . Matrix damage contribution is then described simply as follows:

$$\Delta T_{shift_matrix} = [a \cdot \Phi^n] \tag{3.10}$$

where: ΔT_{shift_matrix} is the transition temperature shift component, Φ is the neutron fluence, a is model fitting parameter and n is the exponent (normally $\frac{1}{2}$). The parameter a is a constant for a given material and a given irradiation temperature; it decreases with increasing irradiation temperature.

In addition to direct matrix damage, during primary embrittlement, copper, together with other elements, is known to induce precipitation mechanism of nano-precipitates also inducing matrix hardening and embrittlement. Such precipitation mechanism continues until saturation depending on available amount of precipitants, Cu concentration in particular. The contribution to the total transition temperature shift can be described as:

$$\Delta T_{shift_Cu_precipitation} = b1 \cdot \left[1 - e^{-\Phi/\Phi_{sat}} \right] \tag{3.11}$$

Where $\Delta T_{\text{shift Cu precipitation}}$ is the transition temperature shift component, $b1$ is a model fitting parameter, representing the maximum saturation value of the shift due to precipitation and Φ_{sat} is a model fitting parameter, representing the fluence at which saturation effects begin

Subsequently other segregants can be formed both proportionally to the matrix damage and attracted into the Cu precipitates. Diffusion of segregants plays also a role. A simple model to describe generally this additional contribution is proposed in the following. The proposed model is based on a 'logistic' shape type of function describing a process of gradual increase then a rapid saturation of the process:

$$\Delta T_{\text{shift}_P\text{ segregation}} = c1 \cdot [1/2 + 1/2 \cdot \text{Tanh}(\frac{\Phi - \Phi_{\text{start}}}{c2})] \quad (3.12)$$

Where $\Delta T_{\text{shift}_P\text{ segregation}}$ is the transition temperature shift component, $c1$ is a model fitting parameter, representing the maximum saturation value of the shift due to segregation, Φ_{start} is a model parameter, representing the fluence at which segregation starts $c2$ is a model parameter, representing the velocity of increase of the effect towards saturation

Based on the above mentioned partial effects, the total effect in term of transition temperature shift is:

$$\Delta T_{\text{shift}} = a \cdot \Phi^n + b1 \cdot \left[1 - e^{(-\Phi / \Phi_{\text{sat}})} \right] + c1 \cdot [1/2 + 1/2 \cdot \text{Tanh}(\frac{\Phi - \Phi_{\text{start}}}{c2})] \quad (3.13)$$

An example of primary radiation embrittlement calculated with the proposed model is given in Fig. 3.17. The relative contribution of the various damage components is also visualized in this.

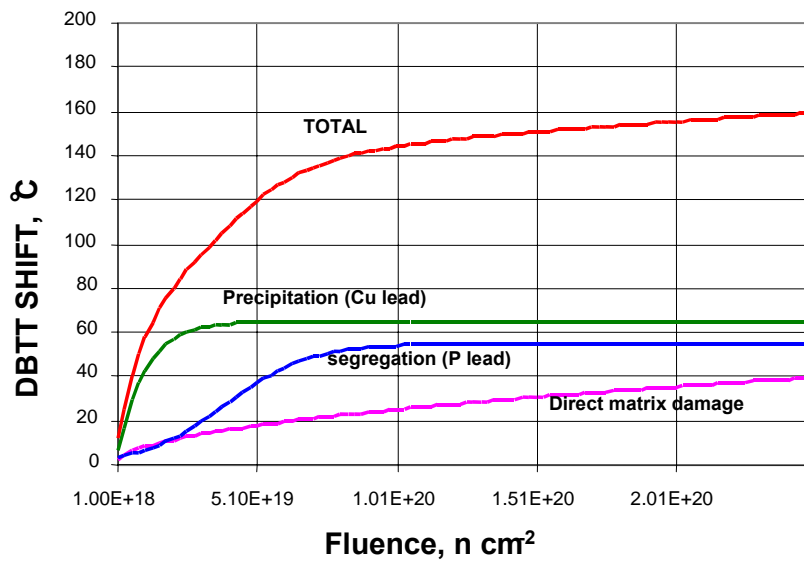


Fig. 3.17 Example of primary radiation embrittlement calculated with semi-mechanistic model, [47]

In total, a maximum of 6 parameters are required for the proposed model: a , $b1$, Φ_{sat} , $c1$, $c2$, Φ_{start} . Some parameters are of secondary importance (Φ_{sat} , Φ_{start} and $c2$) and/or can be derived and fixed in first instance depending on the general behaviour of the data set to be analyzed, thus making the fitting easier. The most important parameters are: a , $b1$ and $c1$. Parameter $b1$ if the model is correct will be depending mainly on Cu content while $b2$ mainly on P content.

Such behaviour is supported by microstructural investigations indicating that during annealing P does massively re-solute back and is almost fully available for the re-embrittlement (some will not re-solute back and a fraction might reach grain boundary). Copper does not re-solute back the same way and would thus contribute marginally to re-embrittlement. The hypothesis that phosphorus is leading re-embrittlement after annealing is also supported by available data on WWER-440. In fact, the transition temperature shift is mainly strongly correlated with phosphorus content and not with copper content.

Using the semi-mechanistic model it is possible to predict the behavior difference of the re-embrittlement in comparison with the primary embrittlement. In fact assuming that phosphorus is the leading element for re-embrittlement and that copper has marginal effect, the copper is suppressed during the re-embrittlement after annealing. The behaviour obtained, Fig. 3.18, reproduces qualitatively well the behaviour shown by WWER-440 high P welds.

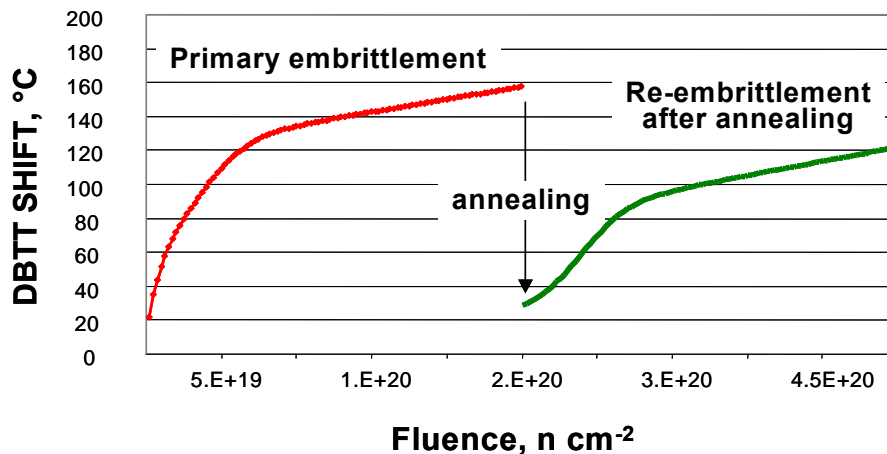


Fig. 3.18 Example of primary radiation embrittlement and re-embrittlement calculated with semi-mechanistic model, [47]

The complete data set of available experimental data was separated into three groups with different level of phosphorous. In statistical analysis several variants were counted with different value of n parameter in «matrix» term. Limitation on “matrix” damage contribution in ΔT_k was imposed for correspondence with available experimental data on clean steels. Limit value was estimated on a base of experimental data at fluence of $\sim 5 \times 10^{23} \text{m}^{-2}$, obtained on the material with low P and Cu content (~ 40 °C). To illustrate this, the results are given in Fig. 3.19.

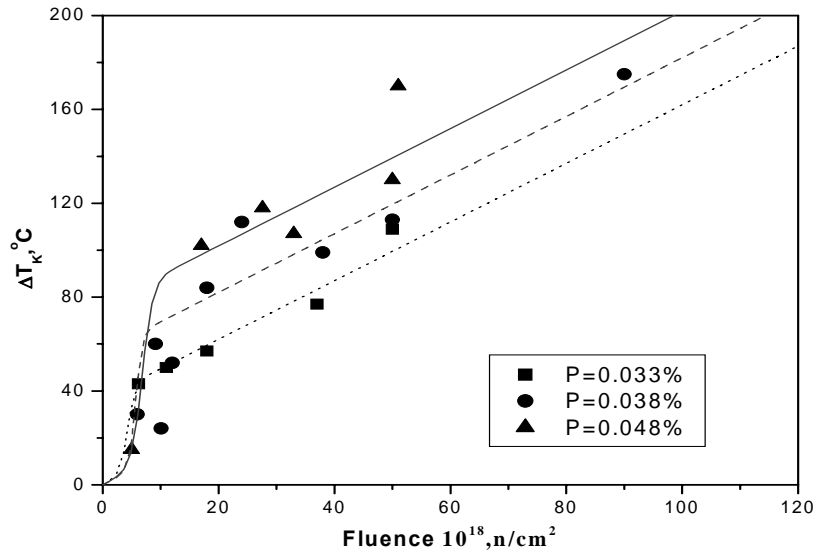


Fig. 3.19 The results using the semi-mechanistic model for the WWER-440 re-embrittlement assessment, [47]

Correlation for the variant with the dose factor traditionally used for «matrix» term ($\sim F^{1/2}$) and the same “matrix” contribution limit gives greater dispersion and is as follows:

$$\Delta T_k = 5.675 \cdot F^{1/2} + 1860 \cdot P \cdot \left\{ 0.5 + 0.5 \cdot \tanh \left[\frac{(F - 13.433)}{10.12} \right] \right\}; (\sigma=23 \text{ } ^\circ\text{C}) \quad (3.15)$$

The advantages of the proposed semi-mechanistic model, when compared to non-mechanistic models, is that this model explains the peculiar re-embrittlement kinetic at low fluence after annealing and the reasons for such differences when compared to primary embrittlement. This is very important and crucial because it allows the correct interpretation of re-embrittlement development avoiding too optimistic conclusions from the results obtained from surveillance in the first years after vessel annealing while avoiding too pessimistic conclusions based on the surveillance results obtained in the subsequent phase.

4 Testing and Sampling

To prove the effectiveness of annealing for the real pressure vessel of some old plants, where no surveillance specimens to monitor the irradiation embrittlement are present in the reactor pressure vessel, the extraction of so-called “boat samples” or templates was proposed. Up to now the sampling is used only for uncladded RPV.

Since 1991 boat samples were taken from all uncladded WWER (several times for some of them) to verify the material status (comparison of experimental data to the predicted values determined by the Russian standards) and the effectiveness of the annealing technology applied for the recovery of RPV, [48-52]. This procedure is widely used for the WWER-440/230 RPV. For these RPVs, besides the lack of surveillance programs and the archive material to perform supplementary evaluation is also not available. The evaluation of the vessel material status in terms of T_k was based on the empirical relationship using the assumed chemical composition. Later the chemical composition has been verified by the analysis of scraps taken from the vessel surface. Sub-size Charpy specimens for the mechanical testing have been developed.

4.1 WWER-440 RPV material radiation embrittlement and annealing effectiveness assessment using boat samples

The scheme of the boat samples cutting from Kozloduy unit 2 RPV is presented in Fig. 4.1, [48]. A total of 15 samples were cut out (four for base metal and eight of weld metal). Seven samples were taken before annealing and five after.

The samples were to be used for impact testing to determine the T_k values and for tensile testing to provide strength and plastic characteristic before and after annealing. A special thermal treatment was used to simulate a state of the weld similar to that of the initial (unirradiated) state. Fabrication of sub-size specimens

was performed in several stages. Workpieces and then plates were manufactured from the samples, and then specimens were produced from the plates. Fig. 4.2 shows the scheme of samples cutting to fabricate both types of specimen (5x5 for weld and 3x4 mm² for base metals) for impact testing.

The main components and impurities were determined by spectrometric analysis for every sample tested. In the case of weld metal, the weld and crown were determined separately. Also hardness measurements were performed with a diamond cone using a standard TK-2M hardness meter.

Tensile testing data indicated that annealing causes a reduction in yield strength, on average 120 MPa at room temperature and 140 MPa at 270 °C. The effect of annealing on the reduction of ultimate tensile strength is less pronounced (approximately by 100-120 MPa). The plastic properties were high before and after annealing.

The analysis of the correlation between standard and sub-size Charpy specimens has been performed by Russian organizations in the beginning of the 90', [53-54]. They have accomplished the experiments devoted to determine those dependencies and their relation to the irradiation. Based on these results the following equations have been developed:

$$T_{k_{10 \times 10 \times 55}} = T_{k_{3 \times 4 \times 27}} + 65 \text{ K}$$

$$T_{k_{10 \times 10 \times 55}} = T_{k_{3 \times 4 \times 27}} + 50 \text{ K}$$

As for the Kozloduy unit 1 RPV boat samples investigation, instrumented impact tests were carried out on sub-size specimens of base and weld metal. The correlations above were used to determine the T_k values corresponding to standard specimen tests: base metal before annealing 40 °C, after annealing 16 °C, weld metal before annealing 212 °C, after annealing 70 °C. The estimated value of T_k corresponding to the initial, unirradiated state for the weld was 50 °C.

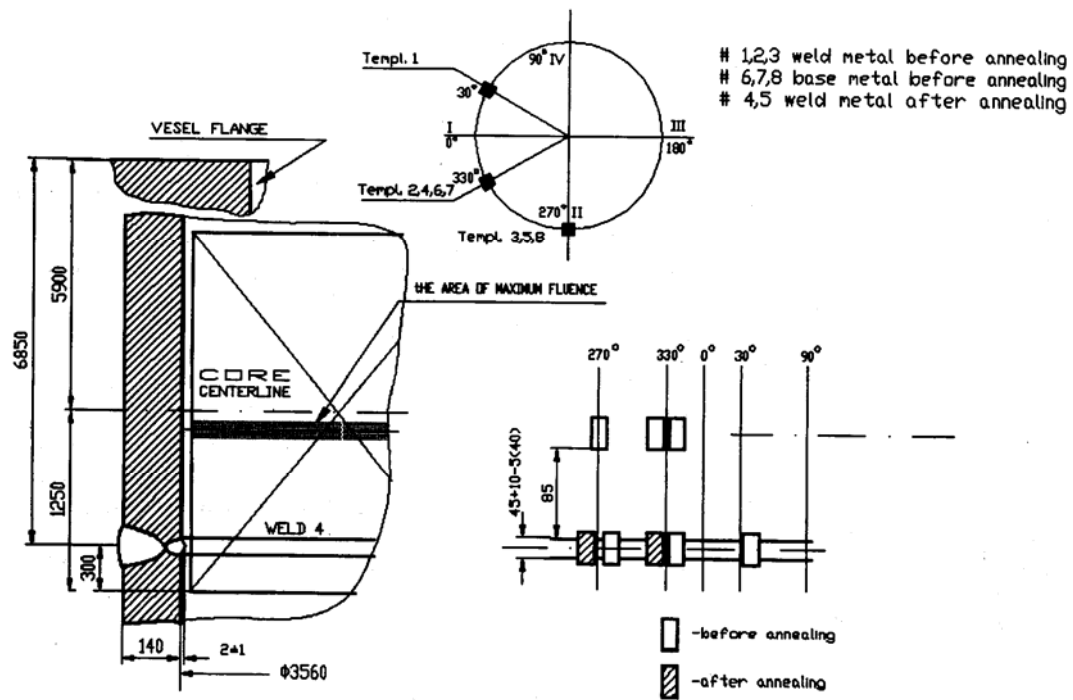


Fig. 4.1 The scheme of cutting samples from Kozloduy unit 2 reactor pressure vessel, [48]

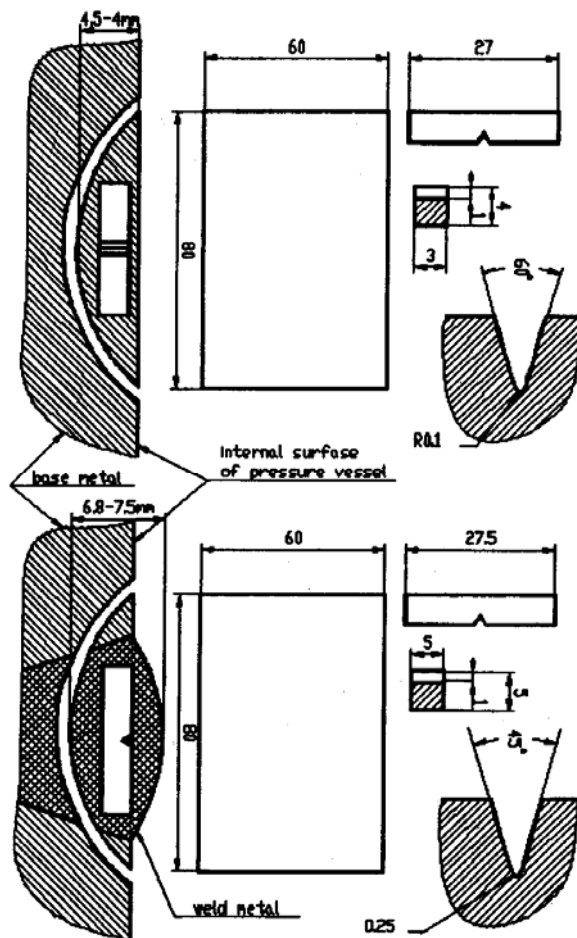


Fig. 4.2 Scheme of samples cutting to fabricate sub-size impact specimens (5×5 and 3×4 mm²), [48]

The experimental results were compared with a prediction of the extent of radiation-induced embrittlement of Kozloduy unit 2 RPV materials, Fig. 4.3, [48].

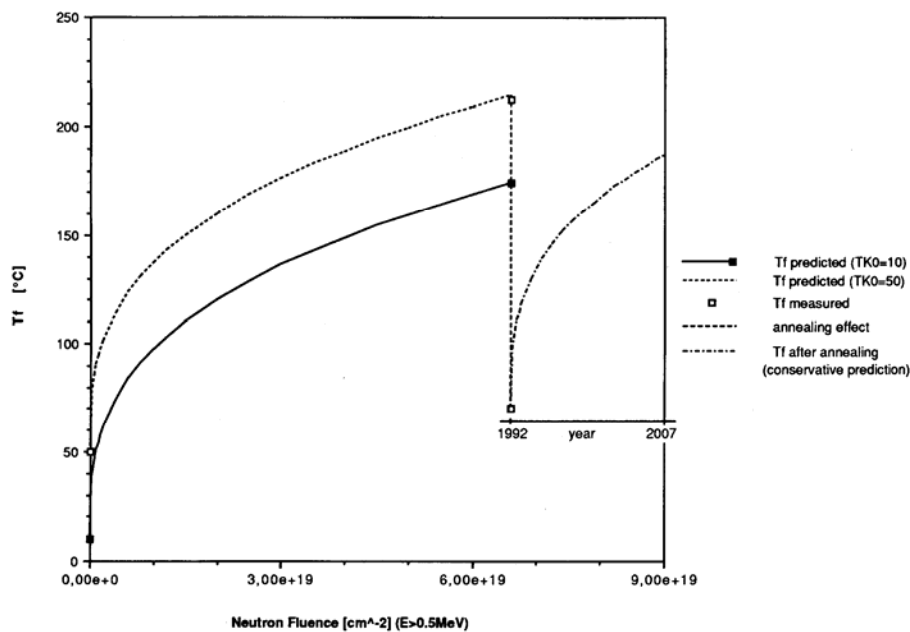


Fig. 4.3 T_F as a function of neutron fluence (and time) for Kozloduy unit 2 RPV weld, [48]

It was confirmed that radiation-induced embrittlement of the base metal does not impose any limitation on the radiation-influenced lifetime of the pressure vessel. The predicted increase in T_k of weld metal as a result of irradiation (about 165 °C) is practically equal to the experimental result (162 °C). However, the value of T_k before annealing (212 °C) obtained from test is about 40 °C higher than the estimated value, i.e. the calculation does not produce a conservative estimate. This was explained by an underestimated value of T_{k0} (10 °C), which had been calculated based on a chemical analysis of the weld metal performed by the manufacturer. The results of boat samples investigations, however, gave an estimated value of T_{k0} = 50 C.

The effectiveness of annealing as a means of restoring the mechanical properties of irradiated WWER-440 reactor pressure vessels was confirmed. The T_k of the weld metal was reduced by no less than 85 % of its radiation-induced shift.

A similar investigation have been carried out for Kozloduy unit 1 reactor pressure vessel, [52], Novovoronezh units 3 and 4,[55] and Kola NPP, [56].

In some cases some material of samples could be used for the additional accelerated irradiation to provide the information about the re-embrittlement rate during the vessel operation after annealing. Sub-size specimens of some RPVs have been irradiated in a commercial operating WWER, which has the channels for surveillance specimens. The experimental re-irradiation results and comparison with the traditional predicted models for Kozloduy unit 1 are presented in Fig. 4.4. In all re-irradiated boat samples the T_k shift is not larger than the prediction by the lateral shift model.

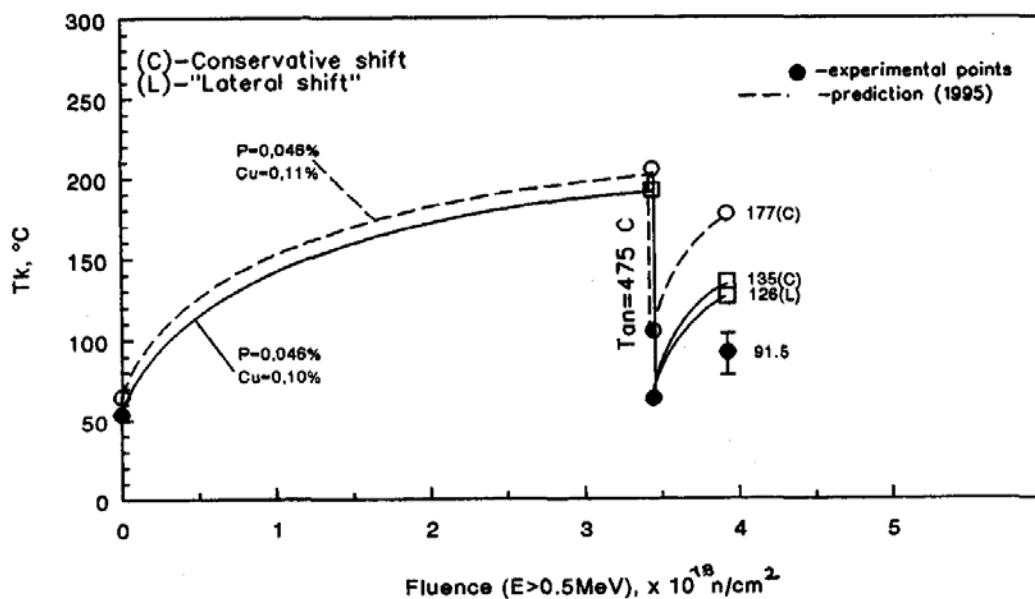


Fig. 4.4 Transition temperature as a function of neutron fluence for "Kozloduy-1" weld metal 4, [52]

4.2 Investigation on decommissioned reactor pressure vessel

Within the project TACIS 91/1/1 Reactor Pressure Vessel Embrittlement, eight trepans (diameter 110 mm, full vessel thickness of 120 mm) have been cut out of the Novovoronezh unit 2 reactor pressure vessel for investigations [49]. This unit was in operation since 1969 up to 1989.

The main objectives of the investigations were:

- to estimate the adequacy of Russian methods for determination of the condition of WWER-440 RPV materials and
- to estimate the effectiveness of Russian annealing technology applied for the recovery of RPV mechanical properties.

The scheme of taking trepans is presented in Fig. 4.5, [49].

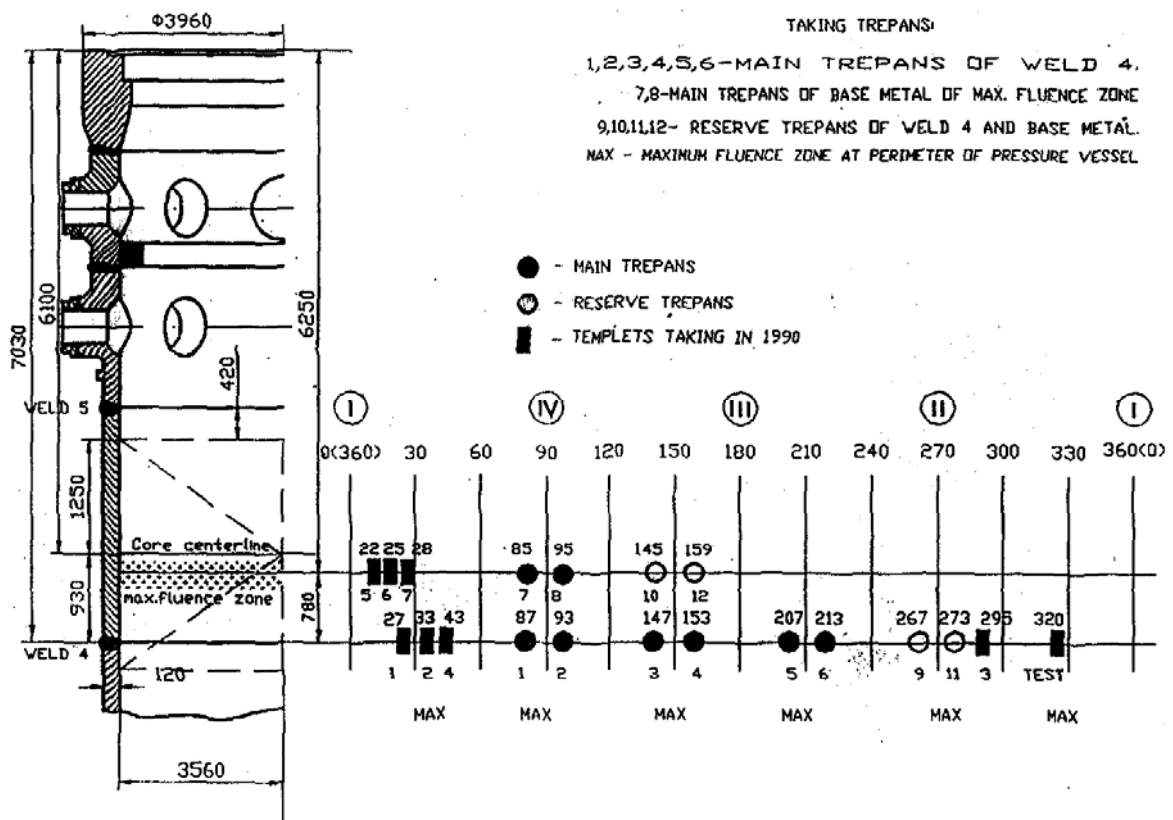


Fig. 4.5 Scheme of taking trepans of Novovoronezh unit 2 reactor pressure vessel, [49]

A good correlation was found between transition temperature shift predicted on the base of Russian standard $\Delta T_k=800 (P +0.07 Cu) F^{1/3}$ (with correction for 250 °C irradiation temperature) and experimental determined in the project, Fig. 4.6.

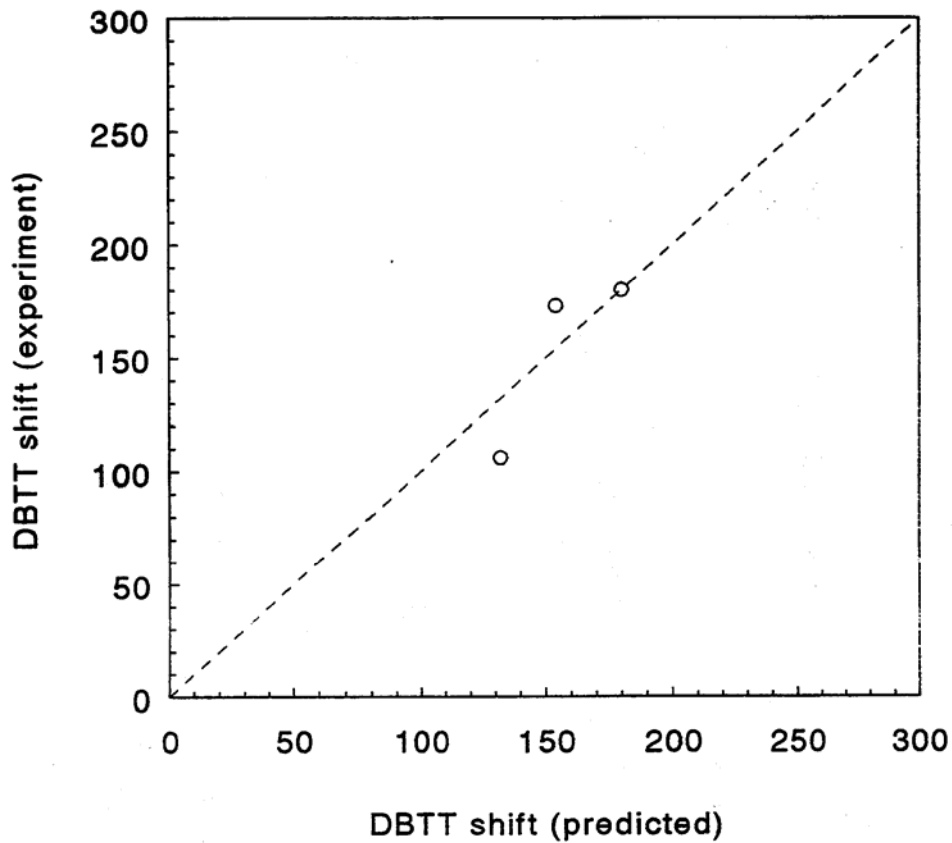


Fig. 4.6 The correlation between predicted and experimental values of irradiation embrittlement of Novovoronezh unit 2 metal, [49]

It was concluded that annealing at 475 °C leads to significant recovery of T_k values in the weld metal. The percentage of recovery is higher for the most irradiated material, showing the maximum values of T_k shift, Fig. 4.7. The material from the inside layer is recovered to 90 %. Almost the same recovery is reached also for the intermediate layers. Slightly lower recovery is observed for T_k values of the outside layer. This is caused by the fact that the value of absolute shift of T_k caused by irradiation is smaller for the outside layer, characterized by a neutron fluence which is 4 times lower compared to the one determined inside.

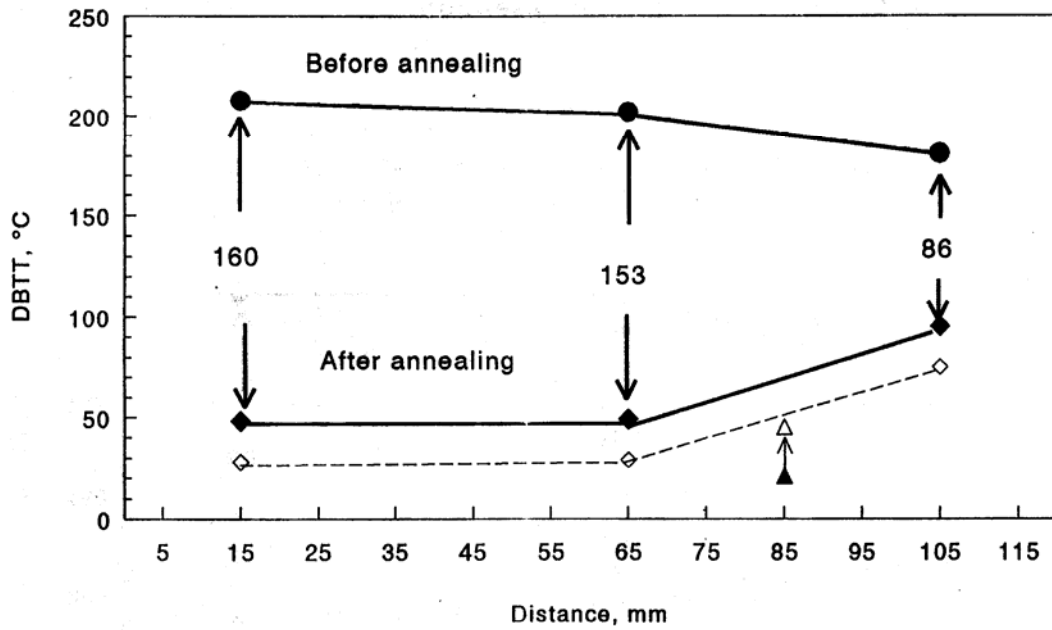


Fig. 4.7 Evaluation of irradiation embrittlement and also efficiency of Novovoronez unit 2 weld annealing, [49]

In conclusion, the investigations of metal from operating WWER-440 confirm that annealing at 460-475 °C is an effective instrument to decrease the RPV irradiation embrittlement and extend the RPV service life.

5 Annealing Technology

From the point of view of the execution itself, there are three possible methods of annealing:

- “wet” annealing without removal of the core
- “wet” annealing with the removal of the core
- “dry” annealing with the removal of the core

If the recovery annealing requires a temperature over 343°C which is the case of most operated PWR, only the dry method with the core removed (the execution of which is the most complicated), can be used for this purpose.

- First “wet” anneal on U.S. Army SM-1A vessel in Alaska in 1967
- Non-commercial vessel anneals in the late twentieth century
- “Dry” annealing demonstration on cancelled Marble Hill vessel in 1996
- Nozzle-supported four-loop Westinghouse design vessel
- Successful demonstration in terms of temperature control and predictive aspects during the annealing
- Yankee Rowe was planning to conduct a “wet” anneal
- Operated at lower temperature (260°C) than other PWRs
- Plant was shut down before annealing due to political issues
- Two “dry” annealing demonstrations initiated by industry and DOE in early 1990s
- Marble Hill – indirect gas-fired can process (demonstration successful)
- Midland – electric resistance heating designed by Russians (cancelled)
- Palisades was planning to “dry” anneal in 1998
- Precipitated ASME Code Case N-557, NRC Annealing Rule 10CFR50.66, Regulatory Guide 1.162, and revised ASTM E 509
- Annealing plans canceled once fluence re-evaluation allowed meeting end-of-life operating license.

Up to spring 1994, in total 14 annealing for commercial power reactors have been carried out (Table 5.1). They are all WWER-440 typer reactors. Additionally there was a project for a prototype annealing of the decommissioned Novovoronezh 1 (WWER-210). Practically all “old” RPVs of WWER 440/V-230 type power reactors were annealed with the “dry” method using electric furnace technology:

- **MOKHT-OTJIG RM**, the Russian consortium successfully annealed 12 RPVs
- **SKODA**, the Czech and Slovak consortium successfully annealed 3 RPVs

Table 5.1 Annealing of WWER-440 type RPVs

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-annealing)	1991	475 °C / 150 h	No
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

5.1 Russian Annealing Device

The Russian apparatus (Cole & Friedrichs, 1991 [57]) has 54 heater panels in three elevations and they form 9 individually controlled groups, Fig. 5.1. The heater panels are about 20 cm away from the vessel surface. The temperature is monitored by 18 thermocouples in all, on the inner surface of the reactor vessel. The temperature is measured also at several points outside the vessel. The maximum output of the heater is 800 kW. During the heat-up about 225 kW is actually used and only about 150 kW is needed to keep the vessel at the holding temperature. The specified maximum heat-up rate was 20 °C/h and cool-down rate 30 °C/h, in order to limit thermal stresses. The temperature variations in the inner surface of the annealing zone had to be within 50 °C. The maximum temperatures at different points are given in Fig. 5.2. The maximum temperature on the outer surface remained about 20 °C below the temperature of the inner surface. The actual vessel wall temperatures, as well as maximum calculated stress along the vessel height during the annealing, are given in Fig. 5.3. An increase in the size of the heating zone from 1 metre to 3 would raise peak stresses from 130 MPa to 300 Mpa. In another reference (Aleksandrov et al., 1987 [58]) the maximum thermal stresses, using a 1 metre nominal annealing zone and a 20 °C/h rate, were 200 MPa after 32 h heating, when the holding temperature was approximately reached. Temperature changes distributed over very narrow and step vertical areas also cause high thermal stresses.

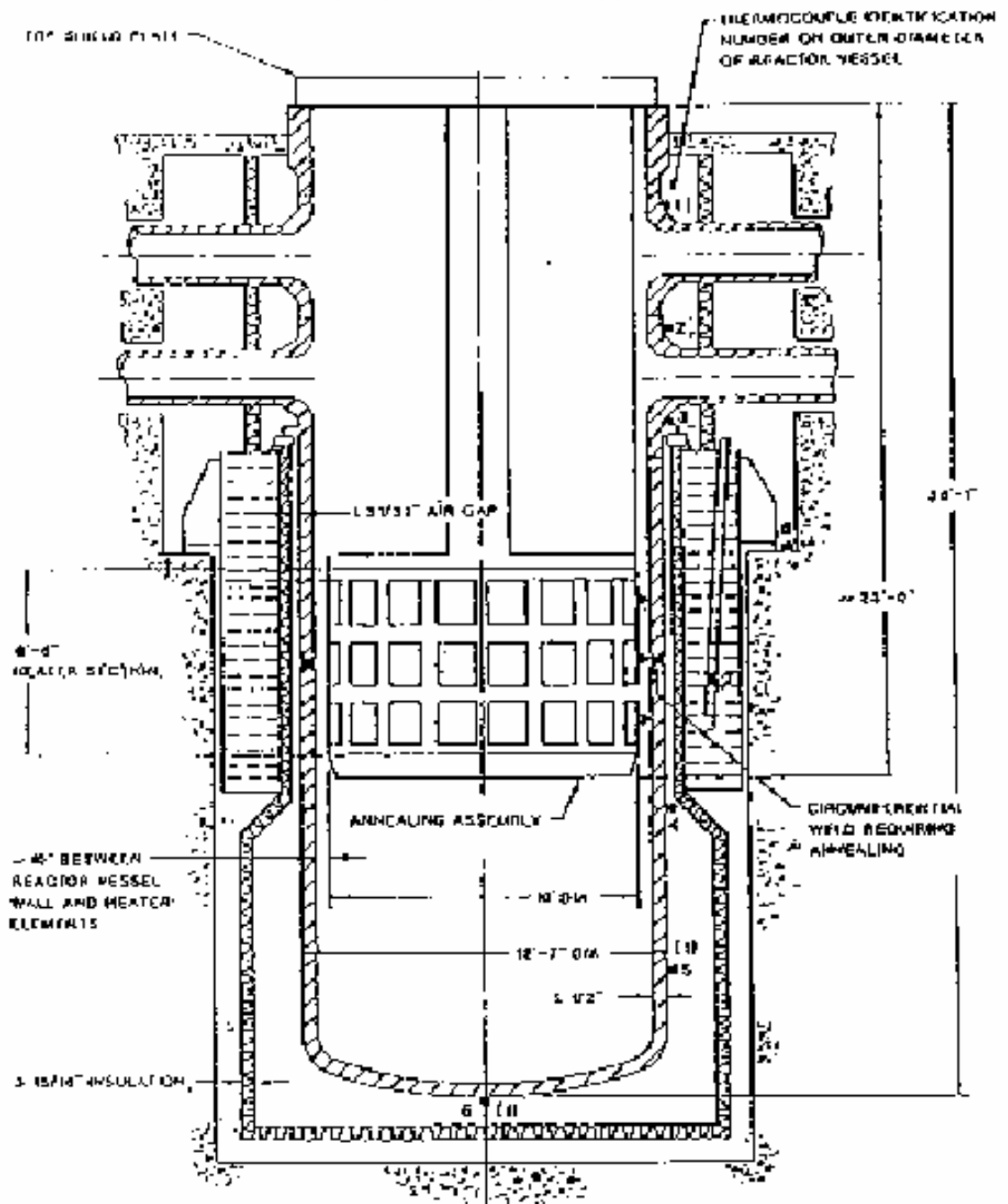


Fig. 5.1 Annealing arrangements in Novovoronezh 3

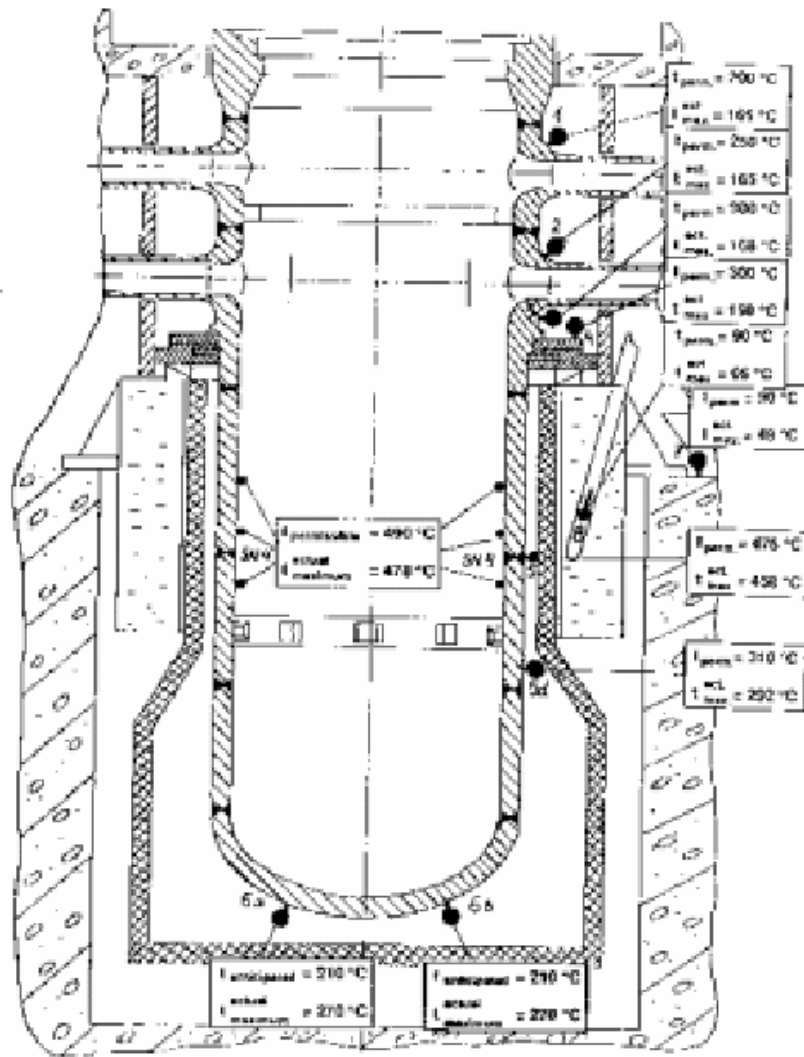


Fig. 5.2 Schematic drawing (not to scale) of temperature monitoring and actual temperatures in Greifswald 1 [59]

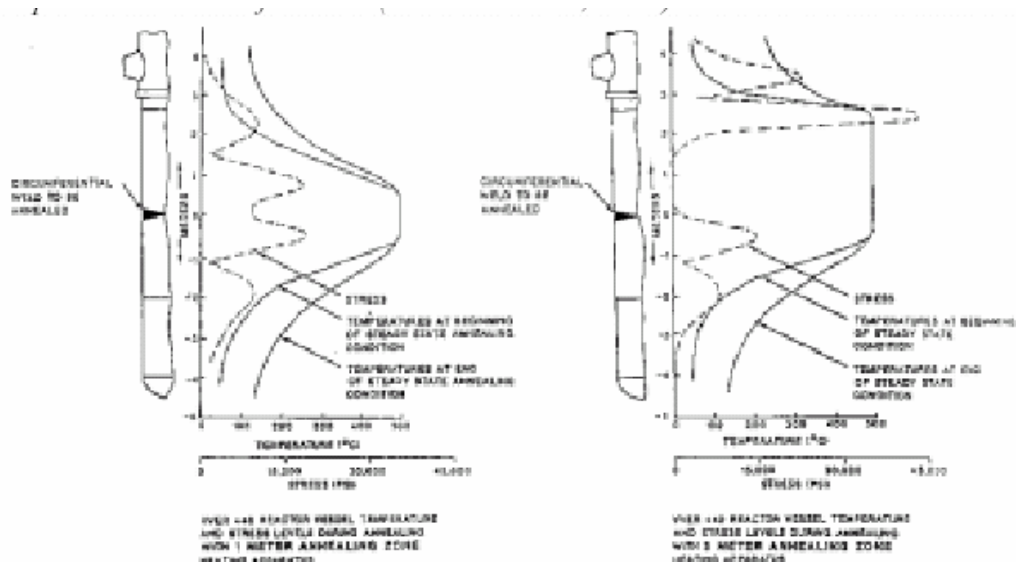


Fig. 5.3 WWER 440 RPV temperature and stress levels with 1 and 3 metre annealing zone heating apparatus. Note the higher calculated stress peak for the larger annealing zone [57]

5.2 Annealing in SKODA

5.2.1 Preparation of the project in SKODA

Annealing process for RPV VVER 440 type reactor is focused on the critical weld No. 4. Annealing projects are carried out in accordance with the ŠKODA Quality Assurance Program and ISO 9001 standard. The annealing program takes approximately one month to perform at the site of the reactor to be serviced. However, due to the safety and documentation requirements of the nuclear industry, preparatory works at the supplier's facility take approximately 12 months and include:

- determination of annealing temperature and holding time
- thermo-technical and strength calculations which model the pressure vessel condition during annealing under normal annealing furnace operation, as well as under an emergency disconnection of furnace heaters (in case of power failure or system fault, the RPV material condition must be known should the complete annealing time not be reached)

- preparation of the Quality Assurance Program and the Annealing Program specific to the plant in question
- maintenance, upgrades and repairs of the annealing equipment itself
- preparation of the temperature measuring system and on-line temperature and strength calculation system in the pressure vessel (verification and validation)

Annealing parameters, essentially the annealing temperature and holding time, are determined by the analysis of material specimens. Typically, the annealing temperature is 475 °C, holding time 100 - 170 hours, the heating rate is 20 °C per hour, and the cooling rate is between 20 to 30 °C per hour. For materials outside the pressure vessel, temperatures limits for normal reactor operation are observed to avoid any tertiary effects of the annealing program on the remainder of the primary circuit components.

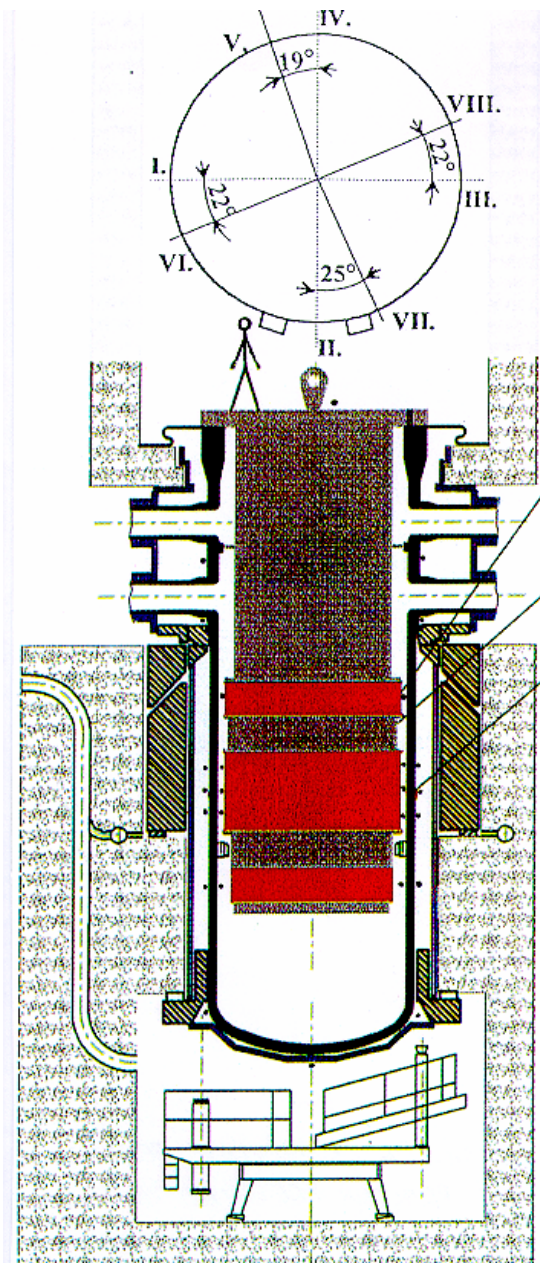


Fig. 5.4 Scheme of SKODA annealing device



Fig. 5.5 Photo of the SKODA annealing device before loading into the RPV

5.2.2 Skoda Annealing Device -Technical Parameters

The annealing equipment is a ring-shaped furnace with heating elements on its external surface. Annealing equipment basic parameters are a maximum diameter of 4.27 m, a height of 10.6 m and a total weight of 64.8 tons. Installed power output of heating elements is 975 kW, while approximately 200-400 kW is sufficient for the annealing. Heating elements are connected to five adjustable heating sections. The equipment also consists of control boxes, a transformer, a power supply cable network, and a control system. Power supply is drawn from the main circulation pumps feed system. The control system works in a semi-automatic mode where surface temperatures are determined in individual heating sections and these are automatically maintained by the control system. The same is applicable for heating and cooling rates. Control correction can also be made manually at any time.

5.2.3 Monitoring Systems for the Annealing Furnace

The temperature measuring system of the pressure vessel surface consists of two parts:

- the pressure vessel internal surface temperature measuring system is part of the furnace, thermocouples are placed in contact with the pressure vessel surface by disengaging mechanisms, measurements are taken at five locations on the pressure vessel axis at two surface lines circumferentially turned through an angle of 180°
- pressure vessel external surface temperature measurements are taken at four surface lines circumferentially turned through an angle 90°, thermocouples are attached to the surface by special mechanisms or magnets.

There are a total of 39 type K thermocouples located on the pressure vessel. The furnace temperature sensor data are corrected and processed by the information system and by the furnace control system. Correction factors are established during model experiments prior to the actual annealing program at the customer site.

The information system displays measured temperature values and heating section power output data on the display in numerical and graphic form. Simultaneously, the measured temperature data are furnished to the pressure vessel temperature and stress on-line calculation system.

The on-line calculation system of temperatures and tensions is provided by specially prepared calculation software, verified and validated according to the quality assurance system requirements. The system calculates temperature distribution in the pressure vessel in twenty minute intervals. For the calculated temperature fields, the fields of deformations and stresses are determined in sixty minute intervals.

At the same time the system indicates if the reactor pressure vessel base material yield point is exceeded at any point during annealing. This relatively advanced system has been developed over a period of time coincident with the rapid expansion of powerful new micro-computers which were a prerequisite thereof (previously, only two-dimensional calculations were performed and at the present time three-dimensional calculations are available).

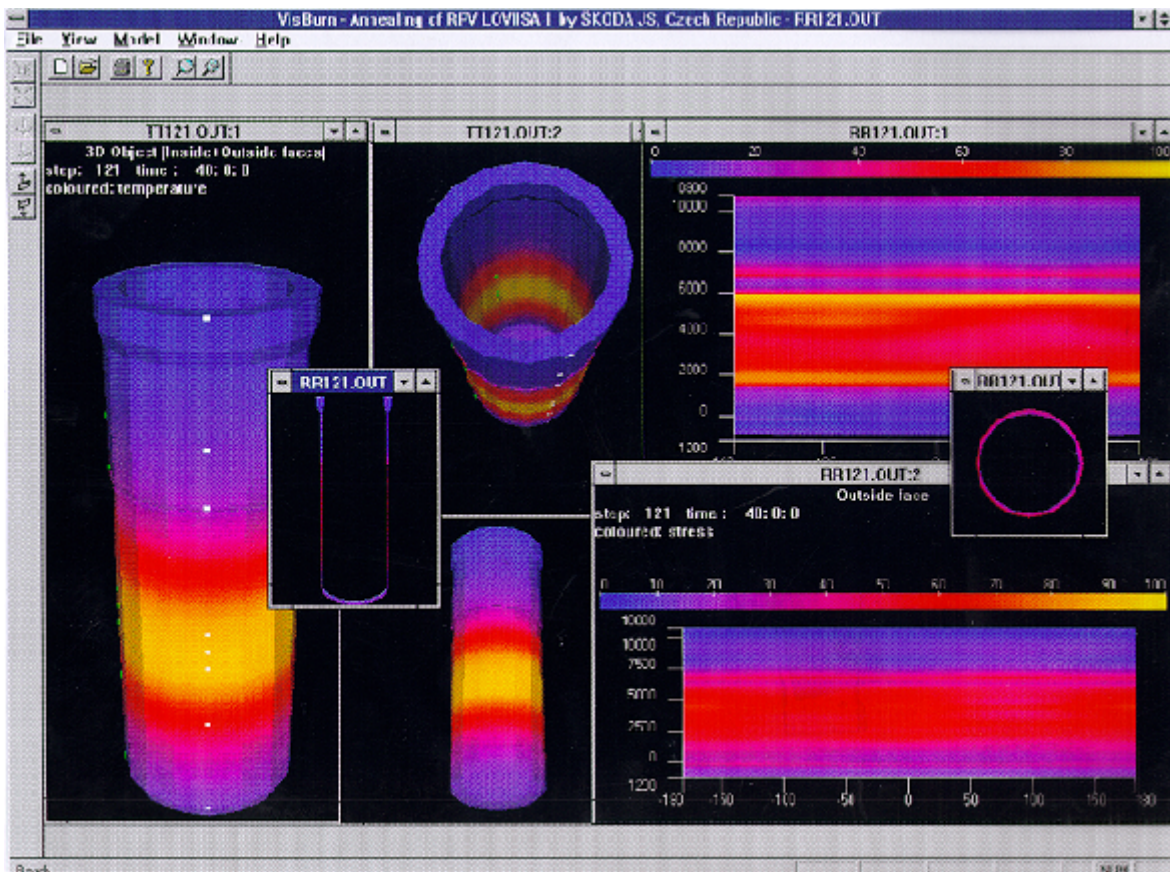


Fig. 5.6 Display of the SKODA monitoring and computing system

5.3 Annealing in Germany

In Germany KWU has designed and constructed an electric radiant heating annealing device for a 600 MW reactor (Kussmaul, 1983, [60]). It has, however, not been in use.

5.4 Marble Hill Demonstration Project

Currently there are no active programs in the U.S. concerning actual RPV thermal annealing. Some basic research activities are ongoing involving the basic understanding of radiation embrittlement mechanisms, and these studies utilize either isochronal or isothermal annealing in conjunction with measurement of property changes (such as positron annihilation or microhardness changes).

There have been programs in the U.S. concerning RPV annealing that date back to the mid 1960s. These early studies (primarily conducted at the Naval Research Laboratory, NRL) were focused on the annealing recovery for the steels in the Army SM-1A RPV which was located in Alaska. This RPV was successfully thermal annealed using a low temperature wet procedure in 1967.

Later interest in annealing occurred in the 1980s when pressurized thermal shock was identified as a significant event and a few plants were looking at mitigative measures for recovering the toughness of highly embrittled RPVs. Research activities were funded by both the U.S. utilities and NRC Research to better understand the degree of recovery and re-embrittlement behavior of typical RPV steels. Several programs were conducted at NRL, Westinghouse Electric Company, Oak Ridge National Laboratory (ORNL), and Idaho National Engineering Laboratory (INEL). The NRL and ORNL studies were focused on measuring changes in Charpy V-notch toughness properties at different annealing temperatures and times for a variety of RPV steels. The Westinghouse studies used three different simulated welds to evaluate the Charpy V-notch and upper shelf fracture toughness after annealing at different annealing temperatures. The INEL project summarized the available toughness data relative to current RPV steels and suggested a different high temperature dry annealing method utilizing a gas-fired radiant heater instead of more conventional resistance heating. This indirect heating method was later used for a demonstration project for the Marble Hill RPV.

The Yankee Rowe RPV was planned for a wet thermal anneal in the early 1990s. The Yankee Rowe plant operated at a slightly lower temperature than other U.S. reactor pressure vessels (approximately 260 °C versus 290 °C), thus allowing the potential for more recovery for a thermal anneal at 343°C. However, the Yankee Rowe plant was shutdown prior to performing a thermal anneal due to political issues and the relatively small out put of the plant.

In the mid-1990s, the Palisades RPV was considering a dry thermal anneal. Plans for annealing the Palisades reactor pressure vessel were canceled in the late 1990s when fluence re-evaluation calculations showed that the plant could nearly meet projected end-of-life PTS Regulatory requirements in 10 CFR 50.61. However, in preparation for the planned annealing of the Palisades RPV, several NRC Regulatory rules and guides, American Society of Mechanical Engineers (ASME) Boiler and Pressure Vessel Code requirements, and American Society for Testing and Materials (ASTM) standards were produced: NRC Thermal Annealing Rule (10 CFR 50.66, "Requirements for thermal annealing of the reactor pressure vessel"), NRC Regulatory Guide 1.162, "Format and Content of Report for Thermal Annealing of Reactor Pressure Vessels," February 1996, ASME Code Case N-557, "In-Place Dry Annealing of a PWR Nuclear Reactor Vessel," approved in March 1996, and revision to ASTM E 509, "Standard Guide for In-Service Annealing of Light-Water Moderated Nuclear Reactor Vessels" published in 1997 (ASTM E 509-97) and recently updated in 2003 (ASTM E 509-03).

A demonstration project was conducted at the canceled Marble Hill plant as a demonstration of the engineering feasibility of performing a thermal annealing treatment on a U.S.-designed reactor pressure vessel. The Marble Hill plant was partially completed with the vessel in place and provided a unique opportunity to test the logistics of performing a dry anneal on a large commercial vessel. The Marble Hill demonstration was completed in 1997. A similar demonstration project was planned utilizing resistance heating as used in the annealing of Russian design RPVs.

The Midland annealing demonstration project was never completed. These efforts were funded by the Department of Energy (DOE), EPRI, and Consumers Power Company (the utility that ran the Palisades plant).

The Marble Hill RPV was Westinghouse design four-loop pressurized water reactor with nozzle supports, similar to the Palisades RPV. The heating and cooling arrangement used an indirect gas-fired method through a heat exchanger as illustrated in the Figures. The heat exchanger was designed for potential reuse and easy cleanup after the annealing procedure. The results were successful in showing that annealing could be performed at a nominal temperature of 454 ± 14 °C at the inside surface of the reactor vessel for a time period one week. Analytical models of the Marble Hill vessel and the reactor coolant system were shown to be correct based on measured temperatures and strains in the actual vessel during the annealing process. Documentation of critical vessel dimensions both before and after the annealing procedure confirmed that all vessel interfaces and dimensions were maintained within acceptable tolerances.

A demonstration project was conducted at the canceled Marble Hill plant as a demonstration of the engineering feasibility of performing a thermal annealing treatment on a U.S.-designed reactor pressure vessel. The Marble Hill plant was partially completed with the vessel in place and provided a unique opportunity to test the logistics of performing a dry anneal on a large commercial vessel. The Marble Hill demonstration results are documented in the proceedings of an EPRI Reactor Pressure Vessel Thermal Annealing Demonstration Workshop held in Santa Fe, New Mexico, USA, in 1998 and in EPRI TR-108316. The status of the uncompleted Midland annealing demonstration project is also described in the EPRI Workshop proceedings.

The Marble Hill vessel was a Westinghouse design four-loop pressurized water reactor with nozzle supports. The heating and cooling arrangement used an indirect gas-fired method through a heat exchanger. The heat exchanger was designed for potential reuse and easy cleanup after the annealing procedure. The results were successful in showing that annealing could be performed at a nominal temperature of 454 ± 14 °C at the inside surface of the reactor vessel for a time period of one week. Analytical models of the Marble Hill vessel and the reactor coolant system were shown to be correct based on measured temperatures and strains in the actual vessel during the annealing process. Documentation of critical vessel dimensions both before and after the annealing procedure confirmed that all vessel interfaces and dimensions were maintained within acceptable tolerances.

Scheme of the overall system, Fig. 5.7 and Fig. 5.8, heating system (Fig. 5.9) and the ducts disposition, Fig. 5.10, are shown in the following.

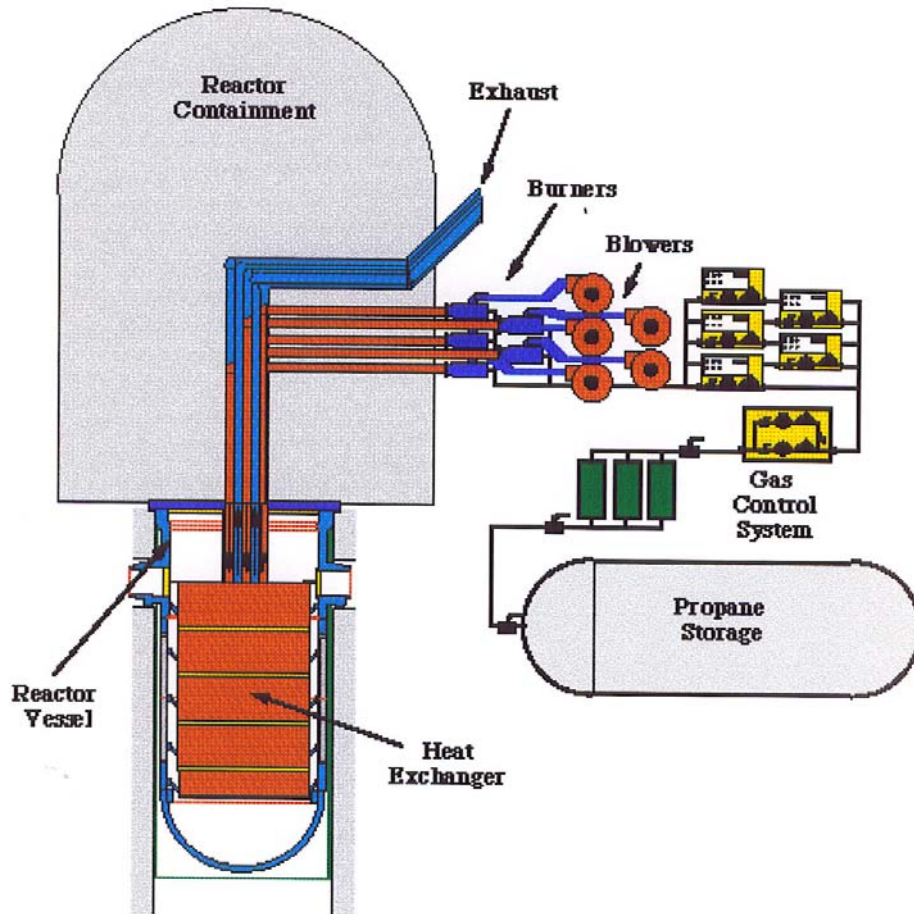
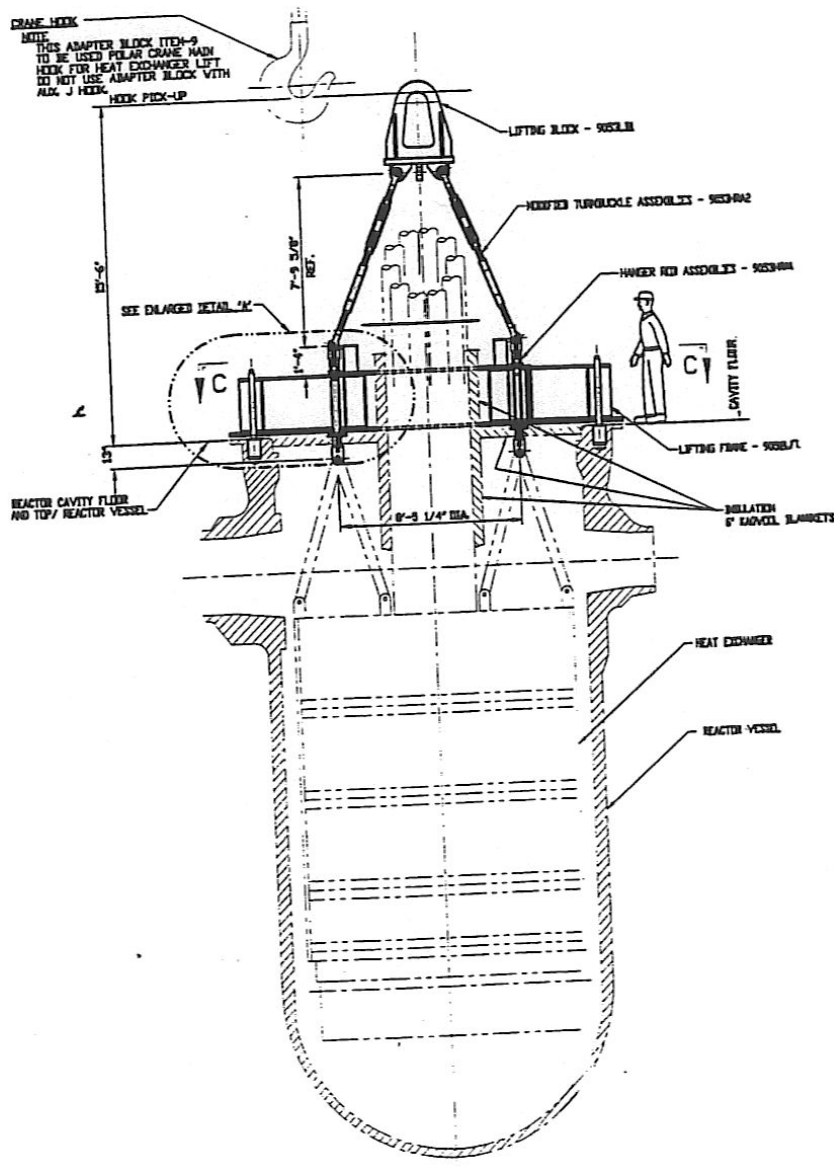


Fig. 5.7 Marble Hill ADP heating system

Figure 5-9

Reactor Vessel Top Cover (with Heat Exchanger)
on Reactor Vessel Flange



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5-26

Fig. 5.8 Reactor vessel top cover with the heat exchanger

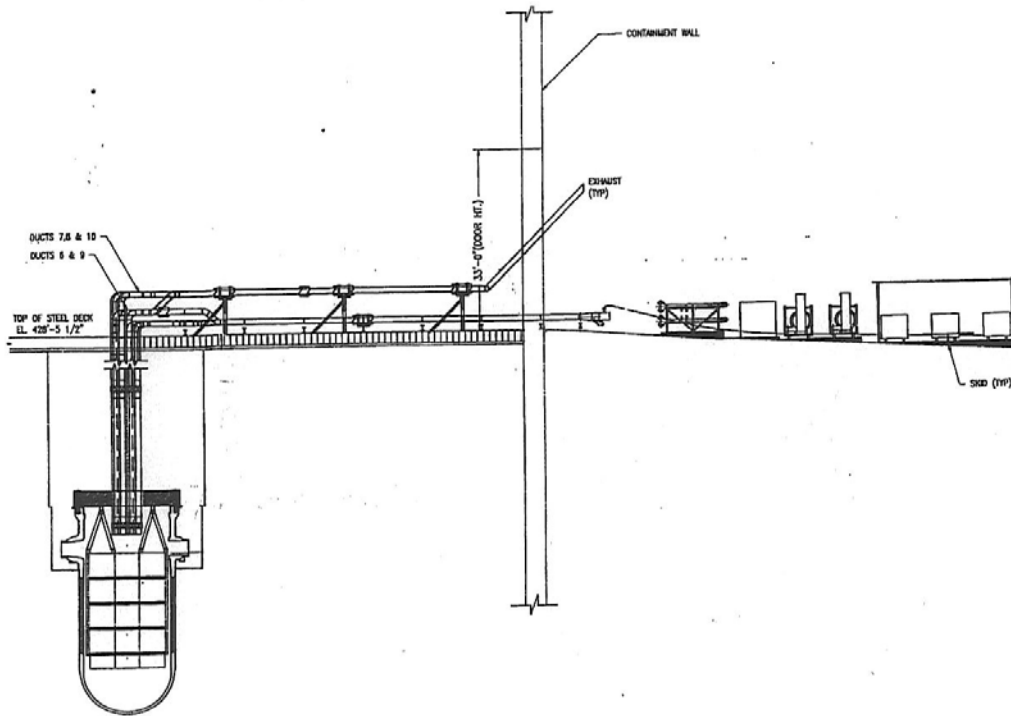


Figure 5-13
Heat Exchanger Reactor
Assembly

Fig. 5.9 Heat exchanger reactor assembly

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S-34



Ductwork Routing

Figure 5-17

Fig. 5.10 Marble Hill ADP, Ductwork Routing

6 Regulatory Requirements for RPV Annealing and Lifetime Evaluation and Relevant Practice

6.1 Thermal Annealing Regulatory Rules and Guidance, ASME Code Requirements, and ASTM Guidance

6.1.1 Introduction

Thermal annealing, as used for recovering fracture toughness properties after irradiation exposure, is a low temperature heat treatment (< 500 °C for about one week) that is not really an “anneal.” For pressurized water reactors (PWRs), there have been two types of thermal annealing treatments: (1) “wet” using the coolant water as the heat transfer medium and staying within the design restrictions which limit the temperature of annealing to 343 °C; and (2) “dry” using an air medium which allows a higher temperature for the thermal anneal (430 – 500 °C). For dry thermal annealing, two different heating methods have been tested: electric resistance heaters and indirect gas-fired heat exchanger.

In the U.S., there was a successful wet thermal anneal on the Army SM-1A reactor vessel in Alaska in 1967. A dry thermal anneal demonstration on a cancelled reactor pressure vessel (Marble Hill) using the indirect gas-fired method was successfully completed in 1996. Another dry thermal annealing demonstration project was planned, but never completed, using the electric resistance heat method on the cancelled Midland reactor vessel. There have been other successful dry thermal anneals conducted on Russian-designed vessels in Europe using the electric resistance heat method. Other thermal anneals on non-commercial vessels may have been conducted in the U.S. and elsewhere.

There has been a series of reactor pressure vessels that have seriously considered thermal annealing as a mitigative measure for recovering embrittled mechanical properties. The Yankee Rowe vessel was being considered for a wet thermal anneal

in the early 1990s. The Yankee Rowe plant operated at a slightly lower temperature than other U.S. reactor pressure vessels (approximately 260 °C versus 290 °C), thus allowing the potential for more recovery for a thermal anneal at 343 °C. However, the Yankee Rowe plant was shutdown prior to performing a thermal anneal due to political issues and the relatively small out put of the plant.

In the mid-1990s, the Palisades reactor pressure vessel was being considered for a dry thermal anneal. This planned thermal anneal prompted the development of several Nuclear Regulatory Commission (NRC), American Society of Mechanical Engineers (ASME) Boiler and Pressure Vessel Code, and American Society for Testing and Materials (ASTM) documents:

- NRC Thermal Annealing Rule (10 CFR 50.66, "Requirements for thermal annealing of the reactor pressure vessel")
- NRC Regulatory Guide 1.162, "Format and Content of Report for Thermal Annealing of Reactor Pressure Vessels," February 1996
- ASME Code Case N-557, "In-Place Dry Annealing of a PWR Nuclear Reactor Vessel," approved in March 1996
- Revision to ASTM E 509, "Standard Guide for In-Service Annealing of Light-Water Moderated Nuclear Reactor Vessels" published in 1997 (ASTM E 509-97) and recently revised in 2003 (ASTM E 509-03).

Plans for annealing the Palisades reactor pressure vessel were canceled in the late 1990s when fluence re-evaluation calculations showed that the plant could nearly meet projected end-of-life pressurized thermal shock (PTS) Regulatory requirements (10 CFR 50.61).

The following sections provide further details on the documents developed for thermal annealing and the Marble Hill annealing demonstration.

6.1.2 NRC Thermal Annealing Rule (10 CFR 50.66) and Regulatory Guide 1.162

Relative to material toughness performance, the Thermal Annealing Rule and Regulatory Guide 1.162 rely on a thermal annealing recovery correlation developed in NUREG/CR-6327. The development of a re-embrittlement projected curve is also needed and will require a supplemental surveillance program. Other aspects of the annealing heat treatment are also presented that require a detailed report and conformance to the basic rules related to original design and assurance of structural integrity of the RPV. The Thermal Annealing Report must contain the following sections:

- Thermal Annealing Operating Plan – detailed thermal and structural analyses must be provided that show that the effects of mechanical and thermal stresses are insignificant for the vessel and all surrounding and attached structures/components; a complete description of the heat treatment method must be included as well as the selected boundary conditions for the temperatures and times for heat-up, steady-state hold, and cool-down.
- Requalification, Inspection, and Test Program – this program is an inspection and test program to requalify the vessel after the annealing operation; it must be shown that the thermal annealing operation did not adversely degrade the reactor pressure vessel, attached piping and appurtenances, or adjacent concrete structures.
- Fracture Toughness Recovery and Reembrittlement Trend Assurance Program – this is the program that defines the level of recovery and establishes the estimated reembrittlement rate to be validated using a supplemental surveillance program.
- Identification of Changes Requiring a License Amendment – any changes to the current operating license or to the Technical Specifications must be identified.

Final conformance of the annealing operation with the Thermal Annealing Operating Plan must be made. Regulatory Guide 1.162 provides the overall format and content for meeting all aspects of the Thermal Annealing Rule.

6.1.3 ASME Code Case N-557

This ASME Code Case was developed to define the operating limits for an in-place dry anneal. The key restrictions identified are a maximum temperature of 505°C and a hold of no longer than 300 hours above 482 °C or more than 1000 h above 454 °C. Temperatures during the annealing must be monitored using thermocouples and a gradient must be established from the edges of the heated region to avoid high thermal stresses. Analyses must be performed to show that no harmful permanent set or ductile flaw growth can occur during the annealing procedure for the pressure boundary components and their supports. EPRI TR-106967 provides the technical basis for this Code Case.

6.1.4 ASTM E 509

ASTM E 509 provides general guidance on the overall annealing process and specific guidance relative to demonstration of annealing recovery and development of a supplemental surveillance program to demonstrate reembrittlement behaviour. The primary factors to be considered in developing an effective annealing program include the determination of the feasibility of annealing the specific reactor vessel; the availability of the required information on vessel mechanical and fracture properties prior to annealing; evaluation of the particular vessel materials, design, and operation to determine the annealing time and temperature; and, the procedure to be used for verification of the degree of recovery and the trend for reembrittlement. Guidelines are provided to determine the post-anneal reference nil-ductility transition temperature (RT_{NDT}), the Charpy V-notch upper shelf energy level, fracture toughness properties, and the predicted reembrittlement trend for these properties for reactor vessel beltline materials.

The guide emphasizes the need to plan well ahead in anticipation of annealing if an optimum amount of post-anneal reembrittlement data is to be available for use in assessing the ability of a nuclear reactor vessel to operate for the duration of its present license, or qualify for a license extension, or both.

6.2 Russian requirements to RPV annealing and lifetime evaluation

There are no clear and specific regulatory requirements for the annealing procedure of WWER RPVs. Practically, each annealing is approved by a special procedure, including QA manual, description of the annealing procedure, measurements and results evaluation during annealing and final evaluation of the annealing recovery and prediction of the re-embrittlement rate. The procedure and residual lifetime evaluation should be based on experimental data from irradiation of similar RPV materials and analysis based on transition temperature shifts prediction based on Russian Code and lateral shift model.

6.2.1 Licencing in Russia (and Bulgaria)

Licencing in Russia (and in Bulgaria) is performed using similar procedures as the same annealing company (MOHT OTJIG) and evaluating organisations (Kurchatov Institute and OKB Hidropress) took place in the annealing in these countries.

Safe operation and extension of operation, as well as issues of license renewal for operation, are regulated in Russia by regulatory documents.

RPV lifetime is a subject of thorough consideration of Gosatomnadzor in frame of expertise of safety justification within the license renewal.

6.2.2 Russian regulatory guides

Basic Russian regulatory document, guiding RPV integrity evaluation, is “Strength Calculation Standards for Components and Pipelines of Nuclear Power Installations. PNAE G-7-002-86”. It defines:

- neutron dosimetry requirements
- forecast critical property degradation depending on neutron irradiation
- brittle fracture calculation criteria

Approach to prediction of RPV lifetime was formulated in detail in Appendix 2 of the Safety Guide RB-007-99 “Expert evaluation of residual lifetime of WWER pressure vessel”. It is based on:

- concept, that limiting factor of RPV lifetime is radiation embrittlement
- criterion of reaching the maximum allowable value of critical embrittlement temperature (so called T_k concept)
- fact, that T_k growth depends on fast neutron fluence only

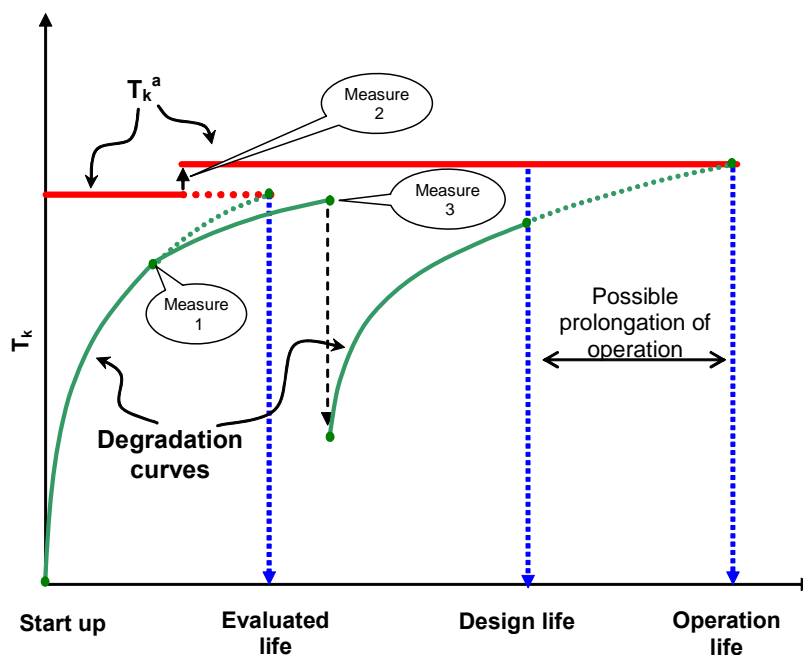


Fig. 6.1 General principles of WWER RPV lifetime assessment

6.2.3 Subjects of Gosatomnadzor's attention:

Approaches, developed and applied during expertise of safety justifications, are based on a system of regulatory documents of Gosatomnadzor

- validity and reliability of all input data, used for evaluation of WWER RPV lifetime
- effectiveness of measures, directed to the lifetime extension (management)
- evaluation of completeness and quality of programs of RPV surveillance
- safety margins and their conservatism.

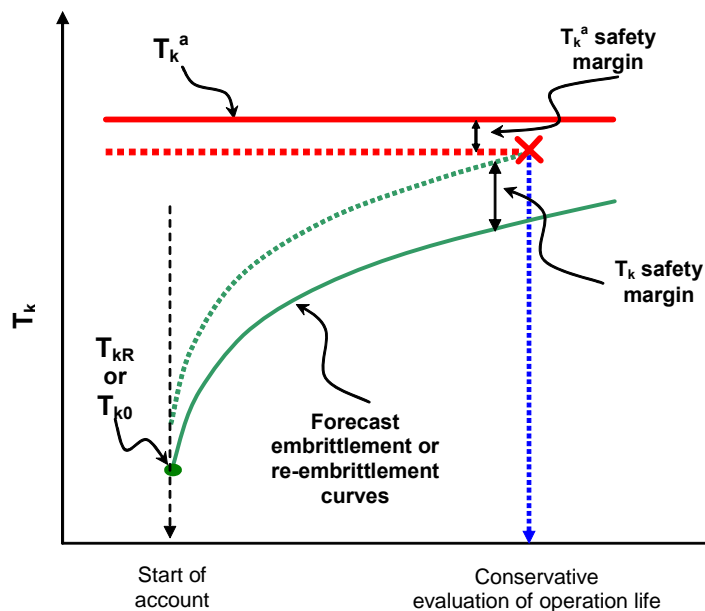


Fig. 6.2 Conservative approach to WWER RPV lifetime evaluation

Embrittlement of RPV is factor limiting safe operation of NPP Unit as a whole. Basic issues, which are considered by experts in analysis of RPV embrittlement and lifetime justification, are following:

- what place of RPV is critical
- reliability of definition of initial properties
- reliability and conservatism of forecast degradation curve

- reliability of definition of neutron fluence and fluence rate
- reliability of surveillance specimen programs
- reliability and effectiveness of measures directed to reducing the brittle fracture possibility
- account and validation of conservatism (safety margins). Measures used by

Utility for RPV lifetime justification:

- thermal annealing of weld #4
- cut out templates from inner surface of RPV
- re-irradiation of sub-size specimens
- reconstruction of specimens
- use of “lateral” shift model for Tk degradation curve after annealing
- installation of burnup fuel on periphery of core.

Additional issues for expert’s analysis: reliability of recovery of initial properties (Tk) by annealing

- reliability of sub-size specimen test results
- reliability of correlation of standard and sub-size Charpy specimens
- validity of a forecast re-irradiation curve after the annealing
- completeness, sufficiency and reliability of results provided by surveillance program on the base of re-irradiation of sub-size specimens
- evaluation of residual embrittlement after annealing and conservatism of forecast dependencereliability of evaluation of fast neutron fluence resulted from calculations, which were validated by dosimetry measurements of ^{54}Mn in templates. The problem items, first of all, concerning the evaluation of uncertainty of research results are to be regulated by the relevant regulatory documents.

With the purpose of regulation of neutron dosimetry of surveillance specimens and of improvement of procedures of handling the surveillance specimen test results

the development of regulatory documents is necessary. Degree of embrittlement of WWER pressure vessel metal is a basic issue of justification of NPP Unit service life. While embrittlement process managing, it is possible to manage the service life of RPV and overall WWER. In case of extension of operation (renewal of license) the special attention should be drawn to reliability of prediction of RPV embrittlement. Supplement of experimental database on WWER-440 pressure vessel embrittlement by reliable results of measurements with as templates (Russian WWER-440), so as trepans (NPP Greifswald) is required. It may allow to decrease conservatism in embrittlement evaluations and to increase reliance in justification of lifetime extension.

6.3 Licensing in Slovakia

The New Surveillance Specimen Programme (NSSP) for WWER-440/V-230 RPVs (without any surveillance programmes) was created as a part of the IAEA international project "Round-Robin Exercise for Irradiation, Embrittlement, Annealing and Re-embrittlement of WWER 440/230 Weld Metal". NSSP main stages

- Co-ordination and NSSP Management,
- Project Preparation and Construction of NSSP Chains,
- Assessment of Initial RPV Steel Material Properties properties,
- Manufacturing of five NSSP samples sets (Chains),
- Annealing of Irradiated N2, N3, N4 and N5 Chains after one year of irradiation; N4 and N5 after re-irradiation, Evaluation of irradiation conditions: Irradiation temperature,
 - Fast neutron fluence.
- Evaluation of irradiated samples properties:
 - by non-standard special punch test samples,
 - by standard mechanical testing.
- Analysis of NSSP specimen's results:

- Residual lifetime evaluation,
- Tk trends (shift of transition temperature),
- Data storage to IAEA database.
- Transport of irradiated chains and storage of rests after evaluation.

The NSSP results serve as powerful tool for the estimation of RPV integrity assessment reliability - by analysis and results comparison, from seven independent laboratories* from values of unexposed reference material, irradiated material, residual embrittlement after annealing, and from rate of re-embrittlement.

Mandatory part of NSSP experimental program is the evaluation of:

- (R) reference material,
- (I) irradiation embrittlement in typical operating conditions of WWER 440 reactor, (N1)
- (IA) irradiation embrittlement recovery after annealing, (N2)
- (IAI) re-embrittlement after annealing. (N3)

Extended (optional) part NSSP consists of:

- (IAIA) recovery efficiency after double annealing, (N4)
- (IAIAI) analysis of re-embrittlement rate after double annealing (N5)

6.3.2 Material manufactured for NSSP programme

Weld model No.502 manufactured in Izhora plant by technology used in RPVs WWER 440/230 manufacturing:

- Forged RPV ring -3.3 m length, 3.8 m external diameter, 0.165 m wall thickness,

- Material: CrMoV steel 15Kh2MFA used for RPVs WWER 440/230;
- Chemical composition and material properties represent weld No.4 of RPVs 440/230 type. Templates and samples were manufactured from 6 layers of the weld in the following way:

- Layer 1 & 2 for Charpy specimen,
- Layer 3 & 4 for COD specimen,
- Charpy mini-specimen (Layer 1 & 2) - from broken halves of unirradiated Charpy samples,
- Special tensile specimen Layer 5 (Gagarinsky type) 5 mm diameter, active length 15 mm NSSP Implementation

Justification:

Irradiation embrittlement is the main degradation process for RPV material damage and the limiting factor for NPP safety operation. For older Bohunice Units 1 & 2 the surveillance specimen program was not implemented due to absence of irradiation channels in the design of WWERs 440/230. Current status of RPV material degradation is defined using computational methodology, which is not satisfactory nowadays. The V1 NPP RPV material samples are exposed to fast neutron irradiation in channels of V2 NPP reactor (Unit 3) and are annealed in annealing oven according to schedule of NSSP contract between Bohunice NPP and Nuclear Power Plant Research Institute (VÚJE). Because WWERs 230 & 213 operating parameters are similar, samples degradation processes by operational environment will be similar too.

6.3.4 Irradiation & annealing conditions

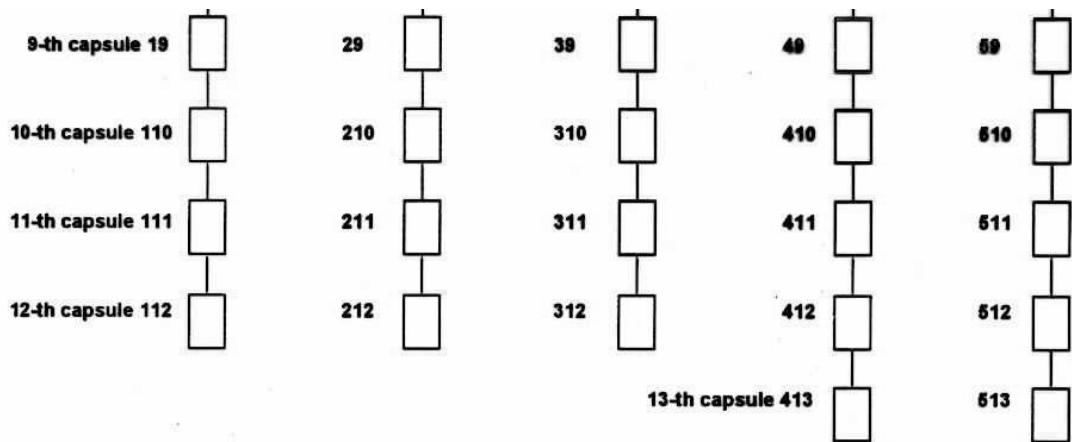
Irradiation conditions Samples are irradiated in the RPV WWER 440/213 irradiation channels used for the surveillance specimen programme. Irradiation temperature is 268 ± 5 °C. This important parameter is measured by the melting monitors. Fe, Nb and Cu activation detectors are used for the measurement of neutron

fluence. Annealing parameters: 475 ± 10 °C annealing temperature, 100 h annealing time, 20 °C/h heating rate, 30 °C/h cooling rate

6.3.5 NSSP Samples sets

Surveillance specimens are placed in special capsules Fig. 6.3, which are coupled into chains and placed in (V2 NPP Unit 3) RPV WWER 440/213 irradiation channels, Fig. 6.5, Fig. 6.6 and Fig. 6.7. The N1 set is made by capsules coupled together with bellows into a chain of the same type as used in Extended SSP. A bellow is used for the exactly defined position of the capsules to the reactor core, since this condition is important for the fast neutron fluence monitoring precision along the reactor core.

N2, N3, N4, N5 sets are assembled with capsules joined with circular chain links. This construction is used to avoid the problems with the manipulation of irradiated chains by annealing operation.

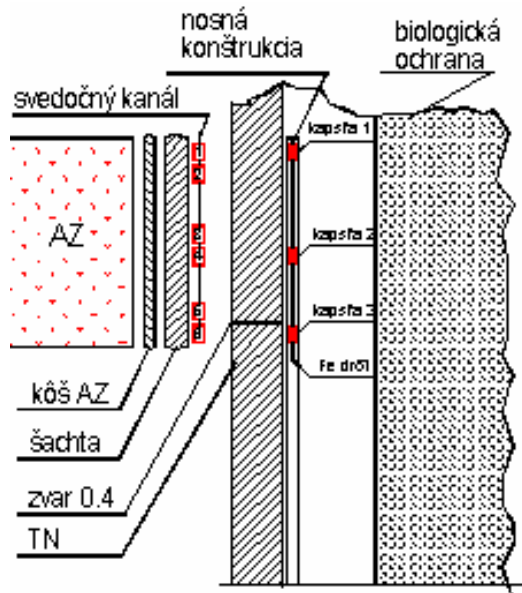


- | | |
|-------------------------------------|---|
| 0. capsule: 12 samples p-CV rec.+MN | 7. capsule: 3 samples p-CV+MN |
| 1. capsule: 3 samples CV+MN | 8. capsule: 3 samples p-CV+MN |
| 2. capsule: 3 samples CV+MN | 9. capsule: 3 samples p-CV+MN |
| 3. capsule: 3 samples CV+MN | 10. capsule: 3 samples p-CV+MN |
| 4. capsule: 3 samples CV+MN | 11. capsule: 16 samples s-CV+MN |
| 5. capsule: 3 samples CV+MN | 12. capsule: 6 samples T + 90 pcs PT (30pcs BM+30pcs HAZ+30pcs WM)+MN |
| 6. capsule: 3 samples p-CV+MN | 13. capsule: 12 samples CV rec.+MN |

CV, s-CV : Charpy test (s-small)
 p-CV : precracked Charpy
 T : tensile test
 PT : Punch test
 MN : neutron fluence monitor

CV rec. : Charpy test on the reconstituted specimens
 p-CV rec. : precracked Charpy test on the reconstituted specimens
 BM : base metal
 HAZ : heat affected zone
 WM : weld metal

Fig. 6.3 Complete set of NSSP chains for irradiation in the Unit 3



Bohunice

Fig. 6.4 Scheme of the location of surveillance chains in WWER-440/V-213 RPV

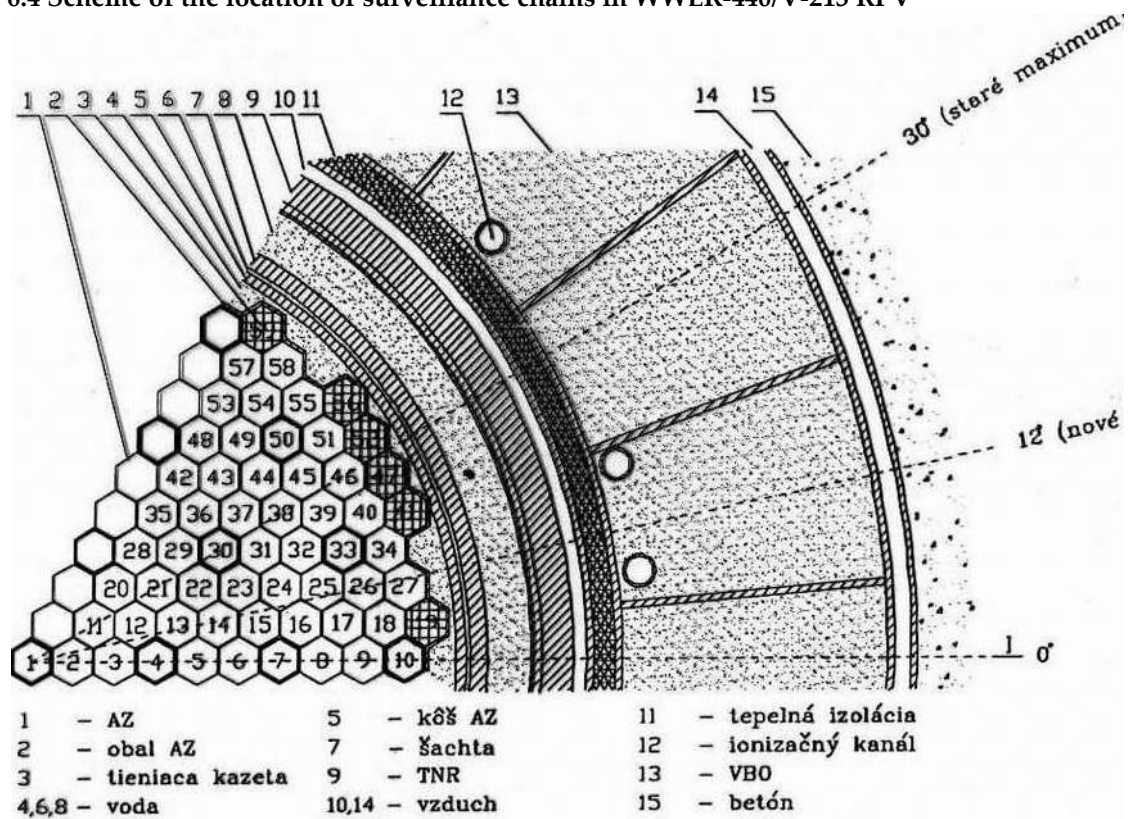


Fig. 6.5 Location of surveillance channels in WWER-440/V-213 RPVs

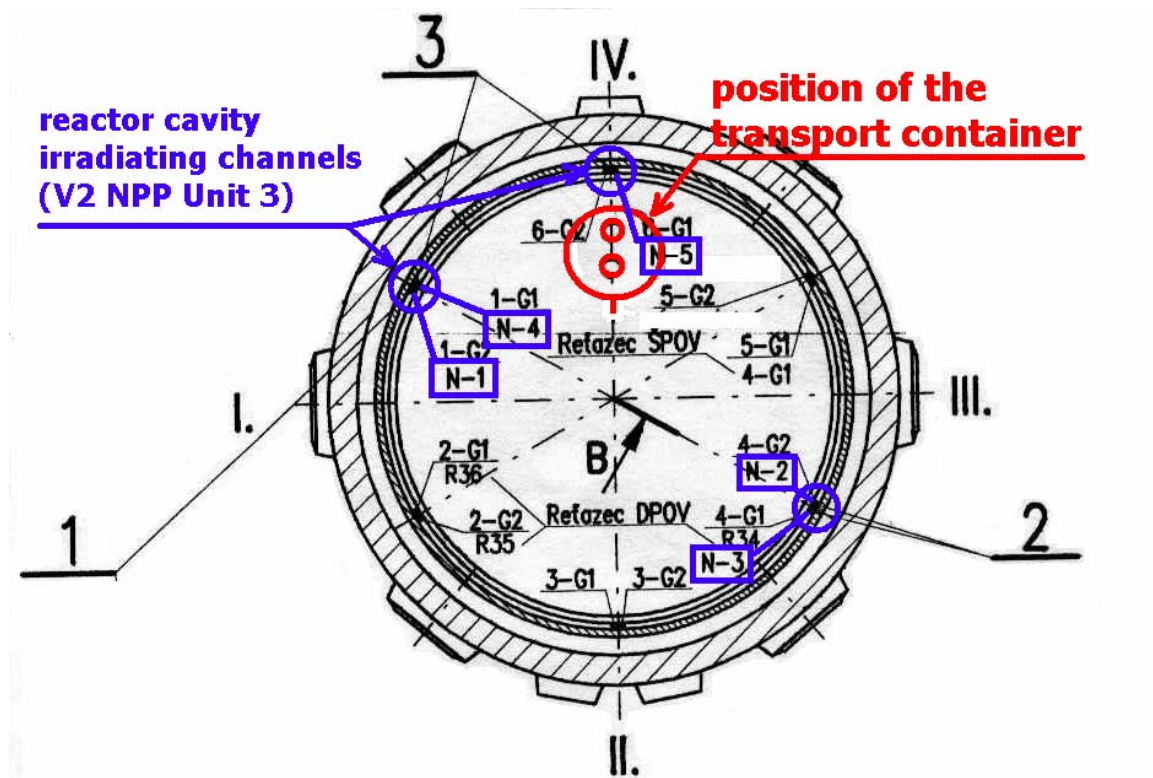


Fig. 6.6 Location of individual NSSP chains in active core of WWER-440/V-213 RPV

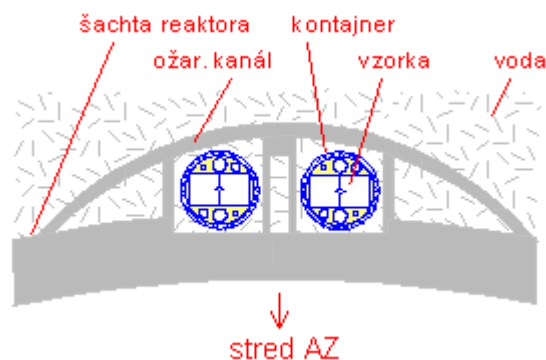


Fig. 6.7 Location of surveillance channels on the outer surface of core barrel

6.4 Licensing the RPV Annealing in Finland

Licensing of the RPV annealing in Finland includes material investigations as well as thermal and stress field analyses. Thermal and stress calculations were performed independently by both Škoda Nuclear Machinery and IVO Power Engineering. Material investigations were carried out by the Finnish research center “VTT Manufacturing Technology” and by the Russian “Moht Otjig RM” company.

Preparation and licensing work for the annealing started in Finland in a smaller scale already in 1980 when an embrittlement faster than expected was discovered. The final and large effort in the licensing procedure started in 1994 when the requirement on annealing was announced by the Finnish Nuclear Authorities (STUK).

6.4.1 Materials testing

Since the amount of irradiated material was limited, the research programmes were carefully planned and balanced. The material shortage problem was solved in two different ways; firstly by using a reconstitution technique to make new test specimens from halves of already tested surveillance specimens. This method was developed by VTT Manufacturing Technology and was used in annealing and re-embrittlement studies. Additionally a so called "tailored" weld material, which has a similar chemical composition, initial transition temperature and mechanical properties as the beltline weld of the Loviisa 1 RPV, was purchased. The material research programmes supporting the licensing of the annealing and post annealing operation of Loviisa units are described in Table 6.1. The irradiation of the test specimens of the Finnish program was carried out in the surveillance position of the RPV in Loviisa.

Table 6.1 Material research programmes supporting the licensing of the annealing and operation of Loviisa-1 after the vessel anneal.

Programme	Material	Irradiation facility	Material conditions	Specimen types	Duration*
Extended surveillance	Lo-1 surv	Lo-surv	R, I, IA, IAI	CH-V, KLST, -5 to +5 K _{JC}	
Extended surveillance	Lo-2 surv	Lo-surv	R, I, IA, IAI	CH-V, KLST, -5 to +5 K _{JC}	
Accelerated licensing	Tailored weld 501	MTR**	R, I, IA, IAI, IAIA, IAIAI	CH-V, K _{JC}	-1 to +1
New surveillance program	Tailored weld 501	Lo-surv	R, I, IA, IAI, IAIA, IAIAI	CH-V, K _{JC}	0 to +15

R Unirradiated reference condition

I Irradiated condition

A Annealing

CH-V ISO Charpy-V

KLST Subsize Charpy-V

K_{JC} Cleavage fracture initiation toughness measured with ISO CH-V size specimens

* approximate duration of the programme in relation to vessel annealing

** In Dimitrovgrad (RIAR) by MOKT OTJIG

Mini test specimens (KLST) were used in “sensitivity studies” to look at the influence of annealing parameters on the recovery and re-embrittlement of the material. The use of mini specimens is very feasible when the availability of testing material is small. Re-embrittlement conditions of the same material will be checked later when the specimens will be taken out after the second period of irradiation in the RPV in Loviisa. The first available results of the annealing and re-embrittlement studies in Finland indicate that:

- The residual transition temperature shift after annealing ΔT_{res} remains practically constant, when the annealing temperature is varied between 420 °C – 560 °C and the annealing time between 100 -500 h.
- The measured values of ΔT_{res} for RPV materials is 10-40 °C depending on material, testing method, fluence of the pre annealing irradiation and annealing parameters.
- The annealing studies did not reveal any indication of temper embrittlement in any of the pressure vessel materials.
- The so called lateral shift approach gives a more accurate prediction for re-embrittlement rate than the so called conservative shift approach.

The licensing program carried out by MOKHT OTJIG in Russia included irradiation of test specimens in a MTR (Material Testing Reactor) in Dimitrovgrad (RIAR). The scope of the test program is shown in Table 6.2.

Table 6.2 The scope of the program by Moht Otjig

Material	R	I	IA	IAI₁	IAI₂	IAIA	IAIAI
Weld, K _{Ia}	20						
Weld, CV	15	15	15	12	12	12	12
Weld, K _{jc}	15	15	15	12	12	12	12
Cl 1st layer, K _{jc}	8	8	8	8	8	8	8
2nd layer, K _{jc}	8				8		8
HAZ Instr. CV	12	12	12	12	12	12	12
Total	<u>78</u>	<u>50</u>	<u>50</u>	<u>44</u>	<u>52</u>	<u>44</u>	<u>52</u>

I = Irradiated	IAIAI = As above and re-irradiated
IA = Irradiated and annealed	K _{jc} = Charpy sized K _{jc} -specimen
IAI = As above and re-irradiated	CV = Charpy V specimen
IAIA = As above and re-annealed	Cl = Cladding

The program also included testing of cladding specimens and a simulated HAZ (Heat Affected Zone). The simulation of the HAZ was carried out by ZNIIKM Prometey in St. Petersburg. The aim was to simulate the heat cycles of the worst region of the HAZ and testing the heat treated material volume in several different material conditions. The material conditions that were investigated are shown in the Table 6.2.

The main testing methods were static fracture toughness (K_{Ia}) and dynamic Charpy-V testing. In the reference condition the crack arrest properties K_{Ia} of the weld were also determined (by ZNIIKM Prometey). As can be seen from the above testing conditions the licensing program included already at this stage a re-anneal as well as re-irradiation of the material after that (IAIAI).

The results from the cladding testing (1st layer; Sv-07Kh25H13) showed that for all conditions ending with irradiation (I, IAI, and IAIAI) an embrittlement effect

could be seen in K_{Jc} at lower testing temperatures only. At higher temperatures practically no embrittlement took place in the different material conditions. The material always recovers very well in the annealing treatments almost to the original level. Only a few conditions were tested for the 2nd layer, the surface layer, of the cladding (Sv-08Kh19H10G2B). The aim was to confirm that the annealing-irradiation cycles do not cause embrittlement in general. The results were difficult to interpret since the position of the fatigue crack tip was generally in a mixed layer. The variation in test results was so large that it was difficult to see any effects of different material conditions. This part of the investigations was also carried out by ZNIIM Prometey. The conclusion of this rather academical part of the studies showed that the cladding retains its toughness relatively well in the above mentioned conditions.

These tests were carried out by RRCKI (Russian Research Center Kurchatov Institute) in Moscow. The measured results are shown as circle for T_{ko} shift and triangle for T_o shift. The 1st irradiation and annealing cycle is perfect when compared to estimations according to Russian standards ($A_f = 39.9$). The recovery is almost complete. The 2nd cycle includes 2 fluence steps and 2 P-contents. The 3rd cycle is also shown and is also quite good. The results show the following:

- the lateral shift is good and conservative enough for estimating the re-embrittlement of the material.
- the residual shift is less than 20 °C in this case.
- the shift in T_o and T_{ko} varies in different cycles ($K_{Jc}/100\text{MPa}\cdot\text{m}^{0.5}$ and $CV/47J$).
- the shift in transition temperature decreases with repeated annealings
- the shift in transition temperatures are much higher than the so called vertical shift would predict.

6.4.2 Thermal and strength calculations

Evaluation of thermal stresses was one of the main tasks in assessing the safety and reliability of the RPV annealing. In this work, the boundary conditions were

considered to have an important role. Several calculations were performed with different assumptions and boundary conditions. The flow routes and rates of the ventilation air around the RPV were studied experimentally at the site and also theoretically. All heat transfer mechanisms, convection, conduction and radiation, were taken into consideration in the calculations.

Temperature distribution in the RPV wall and **air flow distribution** in the gaps surrounding the RPV were first calculated with the widely used general flow analysis code PHOENICS, based on finite volume method. In the axisymmetric calculation model the RPV wall and the surrounding gap were included. The influence of the location of an optional sealing was studied by steady state calculations. These calculations were performed by Škoda Nuclear Machinery.

The actual **time dependent temperature field calculations** were performed by more detailed finite element models, utilising heat transfer coefficients calculated from the results of the PHOENICS calculations. These calculations were performed simultaneously by two organisations and codes: IVO Power Engineering using the ABAQUS code and Škoda Nuclear Machinery using the TEPLO code. The axisymmetric calculation model was quite comprehensive including also supporting structure, mirror insulation, dry protection, nozzle area shielding, bottom shielding and concrete structures surrounding the RPV. To find out the sensitivity, different calculations with different boundary conditions were performed. In these calculations, the existence and location of an optional thermal insulation was studied. In addition to that, the influence of the ventilation system and flow routes and rates were studied. Also many malfunction situations like partial interruption in electric supply were studied. A relatively good consistency was found between the results obtained by the two different organisations.

The **stress calculations** were also performed simultaneously by the two organisations, IVO Power Engineering using the ABAQUS code and Škoda Nuclear

Machinery using the COSMOS code. The calculation models were axisymmetric, except in one case where the influence of non-uniform circumferential temperature distribution was studied. The calculations showed that the reduced stress (von Mises) of the base material did not exceed the yield limit in any of the calculation cases. The maximum stress values were obtained under the cladding. Stresses at the outer surface of the RPV were much lower. The yield limit was occasionally exceeded in the cladding. However, in most of the calculated cases the maximum stresses were located outside the weld area. It was found out that the differences in the results between the two and three dimensional calculations were insignificant meaning that there is no need for additional three dimensional calculations. A relatively good consistency was found between the analyses performed by the two different organisations.

The temperature field calculations showed that the efficiency of heat transfer in the cylindrical gaps between the RPV and the mirror insulation and also behind the insulation has remarkable influence on the temperature field in the RPV wall and in the surrounding structures. The influence of an optional heat insulation considered to be installed in alternative locations was also theoretically studied. During the calculations it became clear that it is very important to know all the flow routes and rates as accurately as reasonably possible. The potential undesirable leak routes should be sealed, whenever possible.

In case of the supporting structure and the concrete structures surrounding the RPV, the calculated temperature values were compared to the design ones. In all cases, the calculated temperatures were less than or about equal to the design values.

It was found out that to achieve the proper calculated axial temperature distribution in the RPV wall it was important to model the heating sections accurately, in other words, to simulate the controlling process accurately enough and pay attention to the boundary conditions.

Also some calculations concerning unexpected events were performed. For instance, the influence of potential malfunctions of the annealing device was studied. Detailed strength calculations were performed for the cladding and the radial supports of the RPV. The role of creep of the base material of the RPV was also checked and found insignificant.

7 Conclusions and Recommendations for Future Research & Developments Activities in the Field of Annealing and Re-Embrittlement of RPVs

Large amount of work was performed to define and explain the effect of annealing for recovery of radiation damage in RPV steels, both for PWR and WWER-440 type of reactors, results showed that only dry annealing can be effectively applied for substantial recovery of RPV material properties. This work served as a basis for definition of thermal regimes (475 °C – 100 to 150 h) for successful annealing of 13 RPVs of WWER-440/V-230 type.

Additionally, many projects have been concentrated on the evaluation and explanation of re-embrittlement of RPV materials irradiated after annealing and models for evaluation of re-embrittlement rate in WWER-440 materials have been proposed and tested with limited experimental data. However more general validation of re-embrittlement models based on studies of mechanical properties as well as on changes in material substructures is still needed.

RPV materials behaviour during re-irradiation after annealing should be adequately monitored, e.g. by surveillance specimen programmes with actual or surrogate materials, and/or by sampling from inner RPV surface in uncladded vessels. Also the use of fracture toughness data for RPV lifetime evaluation is recommended in addition to traditional approaches.

In order to increase accuracy in prediction of RPV lifetime after annealing and decrease extra-conservatism, the future work should be concentrated on:

- Better understanding of re-embrittlement mechanisms (hardening and non-hardening components),
- Development of improved model for RPV materials re-embrittlement assessment also considering behaviour of upper shelf both in Charpy impact and in static fracture toughness tests,
- Acquisition of more experimental data focused on further validation of improved models and
- Preparation of regulatory documents for conducting and evaluation of WWER RPV annealing.

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Abstract

Annealing of a reactor pressure vessel embrittled by neutron irradiation constitutes the only known technique to restore the initial material properties, to an extent that depends on the annealing conditions and on the materials. This technique is used in WWER-440 type reactor pressure vessels. A very important related issue is the one of re-embrittlement behaviour of the material after the annealing. In this respect, there is an obvious link with radiation embrittlement understanding. This report compiles the vast amount of information on annealing and re-embrittlement, which is available in the European countries where such annealing operations have been performed. In addition this topic was also investigated in various TACIS-PHARE projects, and the conclusions are included here as well. To complete the state-of-the-art, the results from a number of annealing experiments carried out in US on “Western” type RPV steels have also been considered.

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