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## Neutron Embrittlement of WWER Reactors: EC-Supported Projects

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# Радиационное охрупчивание корпусов реакторов ВВЭР: проекты Европейской комиссии

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Представлены результаты исследований по определению ресурса корпусных сталей АЭС с учетом радиационного охрупчивания. Проанализированы данные по изучению прочности корпусов реакторов BBЭР-440 с высоким и умеренным содержанием вредных примесей (медь и фосфор) в стали. Для сварных соединений отмечены недостатки процедуры испытаний образцов-свидетелей, технологии их изготовления, нейтронной дозиметрии. С целью обеспечения надежной эксплуатации корпусов реакторов разработаны новые проекты, направленные на изучение влияния радиационного охрупчивания сталей на структурную целостность и усовершенствование экспериментальных методов оценки прочности материалов.

*Ключевые слова*: радиационное охрупчивание, целостность корпусной реакторной стали, сварной шов в активной зоне реактора, программа образцовсидетелей.

**Introduction**. Neutron embrittlement of the WWER-440 NPP core weld was recognized to be much higher than expected when the first surveillance specimens of the Loviisa 1 reactor were tested in 1980 after 3 years of plant operation [1]. The shift in the transition temperature of the weld metal of the surveillance program was almost 3 times higher than that defined in design specification and other Russian documentation. Due to the cubic root shape of the embrittlement function, the embrittlement of the core weld in Loviisa 1 in 3 years of operation reached the level that was expected only at the EOL (End of Life = 40 years for the RPV). Several mitigations were implemented in Loviisa NPP in order to ensure safe operation. Measures like reduction of the core, implementation of the

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low leakage core management, heating of the ECCS (Emergency Core Cooling System) water, adjusting the capacity of high pressure emergency pumps, etc. were taken to reduce the embrittlement rate and to make a hypothetical PTS (Pressurized Thermal Shock) softer in order to ensure acceptable safety margins. Similar measures were implemented also in other operating WWER-440 plants. In 1987, the first annealing of the RPV core weld was implemented in Russia at Novovoronezh Unit 3. Later on annealing was performed in all WWER-440 units of the old plant of type 230 and in 1996 the first WWER-440 of the new plant of type 213 was annealed at Loviisa 1 NPP.

In WWER-1000 NPP units with a high Ni content in the RPV core welds, neutron embrittlement also exhibited a higher rate than initially expected and specified. However, the surpassing is much smaller in relation to what was than observed in the WWER-440 NPP. The neutron dose rate is also much lower, and accordingly, the embrittlement is not so pronounced in the WWER-1000 plants. Some shortcomings have been observed in the surveillance capsules, which still need to be properly addressed. The current temperature monitoring in the surveillance capsules is not reliable, and there is a big gradient in the neutron flux within one set of surveillance specimens. As a result, mitigation measures have already been implemented in some of the WWER-1000 NPPs. The temperature of the ECCS has been increased in order to soften possible PTS transients. Furthermore, the surveillance program was upgraded and improved in some plants in order to reduce the neutron fluence gradient in surveillance capsules.

Since 1991, the European Commission has financed a number of TACIS, PHARE, and EURATOM R&D projects in order to support and ensure safe operation of WWER plants [2–18]. In these projects and actions, main attention and support were focused on the problem of WWER-440/230 and WWER-1000 RPV embrittlement and integrity assessment. This paper summarizes the results already obtained in the projects financed by the European Commission and introduces the necessary actions and tasks that should still be considered in order to cope with remaining open issues.

WWER-440/230 RPV Issues. Historically, concern was first with the WWER-440/230 units. The lack of representative material data from the manufacturing stage, as well as the absence of a surveillance program, has led to a systematic direct characterization of non-clad operating units. High content of phosphorus along with moderate or even high content of copper in the welds lead to high embrittlement rates and to early development and implementation of thermal annealing technology. In the TACIS and PHARE projects mentioned, main efforts were concentrated on direct characterization of the core weld material by sampling RPVs. Thin, so-called "boat samples" were cut out from the core region of Novovoronezh 3 and 4 and Kozloduy 1 and 2 RPVs [2, 5, 10, 11, 12]. Tensile and impact toughness (for mini specimens) as well as chemical analysis were the main characteristics studied. Neutron fluence was evaluated based on activity measurements of cut out samples. From the shut down reactor Novovoronezh 2, trepans were removed and through-wall material characteristics were studied. This unit is a predecessor to the WWER-440 NPP with somewhat different operation characteristics, so the results from these studies cannot be applied directly to operating WWER-440 plants. Based on the test results from

mini specimens, the fracture toughness of the studied material was evaluated through correlation procedures. The reliability of the correlation methods was not very good due to a large scatter of test results. Some improvements would still be recommended in order to assure a proper use and implementation of the method.

Annealing and re-embrittlement characteristics were one of the main objectives of the aforementioned projects. There were also efforts to determine the condition of the original material by conducting selected heat treatments of the material. No reliable and validated heat treatment procedures were found for that purpose. Some of cut out mini specimens were annealed and placed back in a host reactor for re-irradiation [5]. The results from re-embrittlement studies were quite incoherent. It was concluded, however, that the "lateral model" for re-irradiation kinetics could be acceptable for a limited application. Further studies would urgently be needed in order to ensure reliable re-embrittlement behavior of the core weld as long as annealed WWER-440 NPPs are in operation. In [8] preliminary plans for cutting of material blocks from the decommissioned RPVs in Greifswald in Germany including a comprehensive test plan have been elaborated. A significant effort has been dedicated to support the justification of further operation of those plants after annealing, but it is considered that the limited amount of representative material will be an obstacle for further justification of their safe operation.

Some of the Russian WWER-440/230 plants have reached or are approaching their EOL (30 years). The policy of REA is to extend the operational life of these plants (PLEX). The Russian nuclear authority has approved the extension of the operation of Novovoronezh Unit 3 for 5 years. The REA application was for 15 years. At the moment, the EC gives no support to these old plants of the so-called "first generation."

WWER-440/213 RPV Issues. WWER-440/213 RPVs have been considered as well designed and characterized and less sensitive to neutron irradiation effects mainly due to the use of materials with smaller amounts of detrimental components such as copper and phosphorus. Furthermore, these plants are provided with excellent surveillance programs including archive material with test specimens for direct fracture toughness determination. A shortcoming in the surveillance program is the relatively high lead factor, which is about 17 for weld specimens. The high EOL neutron dose and a non-conservative anticipation factor of the surveillance make the case quite specific. In-depth justification for long-term operation will be a concern for the future. Further validation of the surveillance results and possible reduction of the related uncertainties are still questions to be clarified, since they are partly subjected to discussions. According to current understanding, the standard reference dependencies specified for irradiation embrittlement in the Russian Guide [19] are not conservative. Important issues are, therefore, to develop new reference curves for irradiation embrittlement. Pre-fatigue cracks of fracture toughness specimens of the surveillance programs were produced quite a long time ago during the construction stage of NPPs. Testing of those specimens revealed that pre-fatigue cracks were not always properly made, and therefore, it is necessary to process the test results more globally and make an adjusted analysis of them. It is also necessary to make reconstruction of test specimens in order to evaluate the quality of the test results.

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The irradiation temperature of the surveillance specimens is an important parameter when evaluating test results. If the temperature of the surveillance specimens during irradiation is higher than the temperature of the down-comer water, the test results will be optimistic and non-conservative. The original temperature measurement methods are not reliable, and a more sophisticated monitoring was seen as an urgent issue. A specific shared cost action, the COBRA Project [14], was dedicated to the direct online measurement of the surveillance specimen irradiation temperature by using thermocouples. Thermocouples were installed in an ordinary surveillance capsule in Kola NPP Unit 3, which is operating with full core loading of fuel. The irradiation and monitoring took place during one year of operation from September 2001 to August 2002. The measurements showed that the temperature of the surveillance specimens during operation is close to the down-comer temperature ( $\Delta T \sim 5^{\circ}$ C). The standard requirement is  $\Delta T < 15^{\circ}$ C. Similar direct online temperature measurement was carried out in Loviisa NPP 20 years ago. This unit was operating with a reduced core loading and, accordingly, with less  $\gamma$ -heating. The results in Loviisa NPP showed the same temperature difference as was found in Kola Unit 3. Thermocouple temperature monitoring in Bohunice in Slovakia also confirmed similar irradiation temperature with full core loading of fuel.

**WWER-1000 RPV Issues**. WWER-1000 units are the most recently developed ones. They are contributing quite significantly to the current electricity production, especially in Ukraine. Two TACIS projects assessing the influence of neutron embrittlement on the RPV integrity were implemented in the -94 and -95 plans [2, 3]. For a few selected scenarios, fracture mechanics calculations were carried out in order to assess the stability of postulated cracks in the core weld. The results are shown in Fig.1 with the  $K_{Ic}$  and the calculated  $K_{I}$  blow-down curves in a SBLOCA (Small Break Loss of Coolant Accident). It can be seen in the figure, that a 20.6 mm deep circumferential crack would be initiated in this scenario without heating of the ECCS ("w" in Fig. 1). With a heated ECCS ("hw" in Fig. 1), crack initiation can be avoided.



Fig. 1. Calculated stress intensity factor for circumferential (c) cracks in weld 4 by the western approach. Crack depth is 20.6 mm and the crack aspect ratios are 1/3 (c2) and 2/3 (c1). The fracture toughness curve is also shown. The results are shown with (hw) and without (w) pre-heating of the ECCS [3].

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The shortcomings regarding the surveillance program of the WWER-1000 NPP was additionally recognized as a problem when collecting material properties and, especially, when determining the fracture toughness of the core weld. Accordingly, specified upper bound properties were used in the above integrity analyses. The shortcomings of the surveillance program are related to the positioning of the specimen capsules on the top of the core barrel with a significant neutron flux gradient between the specimens. Only about half of the specimens (6) in one set can be used for determining a toughness curve. The standard requirement is 12 specimens with a fluence deviation of <15%. The temperature of the specimens was also doubtful due to poor monitoring and positioning above the core.

The problem regarding the neutron flux and temperature of surveillance specimens was dealt with in the projects TACIS R2.06/96 and SRR2/95/6 and 7/, which have been completed recently. In the framework of the TACIS R2.06/96 project, two experimental surveillance sets were designed and manufactured including advanced temperature monitors (melting alloys) and variety of dosimeters (iron, nickel, cobalt, titanium, copper, and niobium) from different manufacturers. They were irradiated during one operating cycle in Balakovo unit 1 in available standard surveillance locations.

The test results showed that the irradiation temperature of the surveillance specimens did not exceed 300°C during one operation cycle. This result is extremely important, since it confirms that the temperature of the surveillance specimens is close to the temperature of the RPV down-comer during plant operation. The difference is less than 10°C, which allows assessment of the results without the application of a temperature correction factor according to the Russian standards.

The activity measurements showed good consistency of the results but some differences were observed on RRCKI dosimeters, especially on copper. The activity measurements on ring pieces cut from specimen capsules confirmed the existence of significant fluence gradients. It is recommended to measure the activity of each capsule in order to confirm the exact positioning direction. It was also recommended for future fluence evaluations to use cycle-by-cycle power distribution from the closest fuel elements instead of the average one as being used so far. Furthermore, it was recommended to include Nb dosimeters in neutron dosimetry due to the representative threshold energy level of 1 MeV. In the new, advanced WWER-1000 plants, surveillance specimens will be located in boxes at the inner wall of the RPV. This solution will definitely exclude all the above shortcomings.

It has been recently recognized that the embrittlement of the core weld of WWER-1000 RPV is higher than expected just as was recognized earlier for the WWER-440 plant. This was especially observed for welds with the nickel content above 1.5%, which is still well below the specified upper value of 1.9%. The majority of the WWER RPV welds have nickel content above 1.5%, so the problem can be considered quite generic. In the TACIS project R2.06/96, this was confirmed when making reconstitution of toughness specimens from broken surveillance specimens from Kalinin unit 1 with a welding machine provided in the project. As a conclusion it was noted that the present Russian trend curves are non-conservative and need to be upgraded, especially for welds with a high nickel

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content. However, it was addressed that since the initial toughness properties of the welds are below the specified initial upper bound in general, the absolute transition temperature of the core weld of the WWER-1000 RPVs will stay below the "specified" value at EOL.

**New Projects**. Two new TAREG projects have been under preparation under TACIS year 2000 budget (TAREG 2.01/00 and 2.02/00). They will be implemented simultaneously in Russia and Ukraine in a very close cooperation, since the results of the latter project shall be integrated in the final assessment of the former one.

The aim of the 2.01 project is to generate the conditions for an extensive understanding of the situation regarding the RPV integrity assessment, with a particular concern on the material embrittlement aspects. This project includes validation of the global program on the basis of a consistent state-of-the-art evaluation of the current knowledge, including a comprehensive identification of the most critical and urgent open safety issues in the topic. This task will be carried out within an international group (Senior Advisory Group) specifically set up for the purpose. Furthermore, this project will define the conditions for improving the results of the WWER-1000 & 440/213 RPV surveillance projects, the corresponding experimental program being implemented in the twin project and made available later. The evaluation of these results and their consistency with others shall be performed with the aim to conclude on specific aspects such as validation or re-assessment of the neutron embrittlement prediction laws, the 'quality" of the surveillance programs, further assessment of spectrum and flux effects on neutron embrittlement, and direct measurement of fracture toughness in comparison with the application of the codified Charpy-V/ $K_{Ic}$  correlation. This project also includes the preparation of the technical syntheses needed for performing EOL RPV integrity assessments with the aim to assess the most sensitive events. The Institute for Energy of the Joint Research Center from the European Commission is appointed to be the Main contractor for this project.

The 2.02 project shall be seen as an experimental "support project". It includes the performance of in-depth analyses, as well as complementary investigations and tests, which are being considered as necessary for upgrading the available surveillance results. A significant number of reconstituted standard and pre-cracked Charpy-V surveillance specimens will be prepared according to the needs defined in TAREG 2.01/00 project. Impact tests and fracture toughness measurements according to the "Master Curve" approach are also performed in the framework of this project. Specific consideration is given to the implementation of the reconstitution technique in Ukraine and qualification of Ukrainian specialists for the corresponding techniques. Further tests for justifying advanced methods for the evaluation of the fracture toughness are also proposed. They are partly dedicated to further validation of the "local approach," but they also provide for complementary assessment of the shape of the fracture toughness temperature dependence curve. No additional reference irradiation is proposed at that stage of programming, since it has been considered more efficient to rely on upgraded surveillance results. The detailed program will not be in force until approved by the Senior Advisory Group. The results of this project will later be included in the final stage of the 2.01 project. A tender is foreseen, which is intended to identify the most appropriate industrial Western main Contractor.

**Conclusions**. The EC and other sources have been used since 1991 for improving knowledge in neutron embrittlement of WWER RPVs. Most of the above mentioned and described support projects have been successful and have contributed to ensuring RPV integrity and safe operation of the plants. It is clear that there is still a need for further clarification of material issues relating to the RPV integrity. For this purpose, two new TAREG projects are being launched. The beneficiaries REA and Energoatom have already approved the 2.01 project and the administrative arrangement between AIDCO and IE/JRC is foreseen very soon. The 2.02 project is expected to be launched within one year.

## Резюме

Представлено результати досліджень щодо визначення ресурсу корпусних сталей AEC з урахуванням радіаційного окрихчення. Проаналізовано дані про міцність корпусів реакторів BBEP-440 із високим і помірним вмістом шкідливих домішок (мідь і фарфор) у сталі. Для зварних з'єднань відмічено недоліки процедури випробувань зразків-свідків, технології їх виготовлення, нейтронної дозиметрії. Із метою забезпечення надійної експлуатації корпусів реакторів розроблено нові проекти, що направлені на вивчення впливу радіаційного окрихчування сталей на структурну цілісність та удосконалення експериментальних методів оцінки міцності матеріалів.

- 1. R. Ahlstrand, B. Bars, K. Törrönen, and M. Valo, "Surveillance programs and irradiation embrittlement research of the Loviisa NPP," in: *ASTM STP* 909 (1986).
- 2. TACIS R 1.1/91, Reactor Vessel Embrittlement (WWER-440) (1991).
- 3. TACIS U 1.02/92 A2, Evaluation of Reactor Pressure Vessel Integrity of South Ukraine 1-3 Including Embrittlement Aspects (WWER-1000) (1992).
- 4. TACIS R 2.09/94, Integrity Assessment of WWER-1000 RPVs Including Embrittlement Aspects (WWER-1000) (1994).
- 5. TACIS R 2.02/95, Reactor Pressure Vessel Integrity Assessment (WWER-440) (1995).
- 6. TACIS SRR 2/95, Development of Advanced Method for the Evaluation of Irradiation Embrittlement of WWER-1000/320-type RPVs (WWER-1000) (1995).
- 7. TACIS R 2.06/96, Surveillance Program for WWER-1000 RPV (1996).
- 8. TACIS R 6.01/96, Aging of RPV. Embrittlement Analysis at the WWER-440 Greafswald RPVs (1996).
- 9. TACIS R/TSO/WWER01C, Licensing Related Assessment for Design and Operational Safety of WWER-Subtask C Reactor Embrittlement (2001).
- 10. PH/91, *RPV Embrittlement of Units 1-4 and RPV Sampling on Unit 2* (Kozloduy) (1991).
- 11. PH/92, Kozloduy Unit 1 RPV Assessment, PTS on Unit 1 and Spectroscopy Analysis on Unit 1 (1992).

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- 12. PH/93, Kozloduy RPV 1 Assessment: Testing (1993).
- 13. PH/2.01/95, Integrity Assessment of WWER-440/213 RPVs, Implementation of Dynamic Fracture Mechanics Properties and Criteria (WWER-440) (1995).
- 14. COBRA, WWER-440/213 Surveillance Specimen Temperature Measurements.
- 15. FRAME, Validation of Fracture Toughness Trend Curve for RPV Materials.
- 16. GRETE, Round Robin Exercise on Nondestructive Techniques to Assess and/or Monitor Material Properties Degradation.
- 17. MODEL ALLOYS, Phosphorus, Copper, Nickel Effects on Embrittlement.
- 18. PISA, Phosphorus Role on Embrittlement Effects and Development of a Physically Based Model.
- Regulatory Document, PNAÉ G-7-002-86, Strength Analaysis Standards for Equipment and Piping at NPPs [in Russian], Moscow, Énergoatomizdat, (1989).

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