The main mechanisms of the degradation of material properties in the process of NPP equipment operation

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ABSTRACT

The paper presents the general analysis of possible degradation mechanisms of NPP equipment materials and their weldments, which are associated both with the influence of thermal-force loading in the process of operation, and neutron fluence, coolant and other factors. It is shown, that the main mechanisms of degradation are radiation and thermal embrittlement, fatigue and corrosion.

1. INTRODUCTION

At present for electricity production in Russia nuclear power plants (NPP) with three types of reactors are operating. They are VVER-440/230 and 213, VVERE-1000 and RBMK-1000. The operation period of the first units of VVER-440 (Novovoronezh NPP unit 3, start in 1971) and RBMK-1000 (Sosnovyj Bor unit 1, start in 1973) is approaching to 25 years, VVER-1000 (Novovoronezh NPP unit 5, start in 1980) is approaching to 15 years. The first NPP with VVER-210 reactor (Novovoronezh NPP unit 1, start in 1964 and unit 2, start in 1969), having operated for more, than 20 years, were removed from operation. The service life of the first in the world NPP (Obninsk) is more than 30 years.

The average age of the world's nuclear power plants (NPP) is increasing as new NPP constructions fail to be realized. In this connection at present it is necessary to put a greater attention to the variation of mechanical properties of active NPP during the operation. Unfortunately, such information is insufficient, but such information is necessary to solve the problem of NPP life extension.

2. STATUS

Ageing occurs in all components of equipment (reactor, steam generator, pipe etc) during nuclear power plant operation. Ageing has been defined as a continuous time-dependent degradation of materials due to actual operating conditions, which include normal operation, transient regimes and design basis accidents. The service conditions identified as contributing to ageing are:
- environmental factors - chemical and physical processes that affect material properties (neutron fluence, corrosion, various thermal effects etc);
- operating conditions - loading (cyclic, dynamic, thermoshock), wear, improper testing and maintenance etc.

The main ageing mechanisms, usually considered, are: radiation embrittlement, thermal embrittlement, creep, fatigue, environmentally assisted cracking (EAC), corrosion and erosion, wear-assisted cracking. Cumulative effects which occur in equipment owing to one or more of these factors determine its state at a given time.

For example, in various regions of pressure vessel the following mechanisms are acting simultaneously (but to a different degree): radiation and thermal embrittlement, corrosion attack and fatigue damage accumulation. Whereas corrosion depends, in the first place, on the production processing of the accepted pressure vessel design (with anticorrosive cladding on the inner surface or without it) and thermal embrittlement, on the composition of the selected base metal and welding materials, the variation of properties due to radiation influence and fatigue is determined, mainly, by the pressure vessel design. Figure 1 gives the scheme of neutron fluence and fatigue damage distribution along the VVER-440 pressure vessel height. This scheme demonstrates, in what reactor zones either of mechanisms of the variation of properties dominate in the process of operation. And proceeding from the stated above factors, it is possible to mark out two characteristic zones. One region of pressure vessel with stresses from inner pressure to 40% UTS and low thermal

Fig.1. Diagram for distribution of neutron fluence and fatigue damage over the height of VVER-440.

stresses in the wall is located opposite the core zone. This region of pressure vessel is affected by maximum neutron fluence, which can lead to a remarkable material embrittlement and, in the first place, weld metal. The other region of pressure vessel
(first of all, nozzle zone) is characterized by increased stresses, caused by their concentration. And here maximum fatigue damage accumulation is possible as well as brittle fracture resistance variation.

Thus, owing to a simultaneous influence of various embrittlement mechanisms on the pressure vessel in the process of operation, the ductile-brittle transition temperature (DBTT) is determined as follows, according to Norms [1]:

\[ T_k = T_{ko} + \Delta T_T + \Delta T_N + \Delta T_F, \]

where \( T_{ko} \) - the DBTT in as-produced condition;
\( \Delta T_T \) - the shear of DBTT due to thermal ageing;
\( \Delta T_N \) - the shear of DBTT due to cyclic damage;
\( \Delta T_F \) - the shear of DBTT due to neutron irradiation.

For such type of equipment, for example, as pipings corrosion and erosion are dominating mechanisms of fracture. Due to the statistic data from 31 to 41% of the total number of the damages of pipings during operation is associated with the nucleation of pitting, pits, corrosion-fatigue cracks and other defects of corrosion or erosion types.

The paper considers the main mechanisms of NPP equipment material properties degradation in the process of operation. The authors do not set a task to discuss all the problems, associated with either mechanism, because these problems cannot be presented completely even in one invited paper. The Workshop "Ageing of NPP Component Materials", held in St. Petersburg from 28 February to 2 March 1995 was devoted to these problems.

3. RADIATION EMBRITTLEMENT

As it is shown in [2-4] radiation embrittlement has a dominating influence on the degradation of pressure vessel material (base metal and weld) properties. In estimations of RPV brittle fracture resistance to the end of operation DBTT shear \( T_k \) due to neutron irradiation \( \Delta T_F \) has maximum value and is defined on the formula \( \Delta T_F = A_F (P_n/F_n)^{1/3} \), where \( A_F \) is the coefficient of irradiation embrittlement; \( F_n \) - neutron transfer with \( E > 0.5 \text{ MeV}, \text{neutr}/\text{m}^2; F_n = 10^{22} \text{ neutr}/\text{m}^2 \). The formula is true for the equipment at \( 10^{22} < F_n < 3 \times 10^{24} \text{ neutr}/\text{m}^2 \).

The coefficient of radiation embrittlement depends on irradiation temperature, material alloying composition, content of harmful impurities (P and Cu). According to Strength Calculation Norms [5] the \( A_F \) values are defined from the correlations \( A_F = 800 (P + 0.07 \text{ Cu}) \) at the irradiation temperature \( 270^\circ \text{C} \), \( A_F = 800(P + 0.07 \text{ Cu}) + 8 \) at the irradiation temperature \( 250^\circ \text{C} \), where P and Cu - the content of phosphorus and copper in %.

The service life of reactors is determined by brittle fracture resistance, which is limited by a circumferential weld located in the core area. The weld is fabricated by submerged arc welding with the use of Cb-10XMFT wire and AH-42 flux. In this case weld metal is less alloyed as compared with base metal and therefore has a lower radiation resistance and in the process of operation is embrittled to a greater degree. The DBTT of this weld equals to 150-200\(^\circ\text{C}\) after 10-15 years of operation, and it makes impossible further operation of reactor because of pressure vessel brittle fracture danger (Fig.2). As this operation period is remarkably less, than the design service life (30-40 years), special measures were necessary, especially
annealing of this circumferential weld. This procedure was successfully carried out on 15 reactors in Russia, Armenia, Bulgaria, Germany and Slovakia [6].

Fig.2. Assessment of radiation service life of circumferential weld of VVER-440.

For advanced reactors of this type, the steel making practice was improved, which provided minimum content of detrimental impurities, including phosphorus, sulphur and non-ferrous metals. Besides, new welding materials were developed (Cb-10XMFTY wire and AH-42 flux), permitting to produce weld metal with less than 0.012% phosphorus and less than 0.10% copper. The application of new pure steel and welding materials improved remarkably the radiation resistance of advanced reactors.

To increase the radiation service life of reactor one may fabricate a longer core shell. In this case circumferential weld is to be removed from the maximum neutron fluence area and the metallurgy is to be changed. The maximum ingot weight for VVER reactor shells was equal to 205 t. The ingots of 140 t weight were subjected to duplex process in acid open-hearth furnace (liquid casting was produced in basic open-hearth furnace) or in electric arc furnace with basic lining. The ingots of the weight more than 140t were produced by the method of successive discharge in steel ladle of the metal, produced in basic open-hearth furnace and refined on the installation of out-of-furnace refining and vacuum processing (ASEA-SKF), and the metal, produced in basic-lined electric furnace and subjected to ladle treatment [7].

The problems of radiation embrittlement were discussed in details at the seminars, held by IAEA in Zavazna Poruba, Slovak Republic 29-31 March 1994 "International Workshop on WWER-440 Reactor Pressure Vessel Embrittlement and Annealing" and in Zurich, Switzerland 29 November - 1 December 1993 International Conference and Exibition PLEX-93 (Plant Life Extension).

4. FATIGUE

In process of operation, reactor pressure vessel material undergoes the influence of cyclic loads, which should be taken into account in estimations of brittle strength on the base of the variation of the ductile-brittle transition temperature $\Delta T_N$. By this, elastic-plastic strain may appear in concentration zones, inspite of a comparatively low level of nominal stresses. The accumulation of fatigue damages may effect brittle fracture resistance [8].
The $\Delta T_N$ definition was carried out as applied to VVER-440 and VVER-1000 reactor base metal. The method of $\Delta T_N$ definition is as follows. Plane specimens of 10 mm thickness were manufactured. With the use of the testing machine UE-50, the specimens were fatigue damaged (to the values 0.1 and 0.01) by means of loading in the rigid regime with the given number of cycles to failure (according to [1] the damage value is taken to be equal to the ratio of number of loading cycles to the number of cycles to the damage nucleation at a given amplitude of strain). The loading level (strain amplitude value) for the type 15X2MFA steel was taken to be $1.2\varepsilon_a$. The value was taken on the basis of generalized low cycle fatigue data at the lifetime level before crack initiation, which is equal to $3 \times 10^4$ cycles (which is by ten times greater, than the calculated number of loading cycles). The strain control was performed in the longitudinal direction with the use of strainmeter on the base of 25 mm. The number of loading cycles was 3000 and 300 (Fig.3).

![Graph of KCV vs Temperature for Base Metal and Weld](image)

Fig.3. Cyclic damage effect on impact strength for 15X2MFA steel and its weldments at $\varepsilon_a = 1.2\varepsilon$ ($T = 20^\circ C$, $N = N_0$, $N_c = 3 \times 10^4$ cycles).

After low cycle fatigue loading impact specimens were cut out, having cross section 4x5 mm (15X2MFA steel) and 10x10 mm (15X2MFA and 15X2HMFA steels) from tested samples (cross section 10 x 30 mm) and the temperature dependence of impact strength was determined. Not less than 9 specimens were tested for each damage level. The analogous tests were performed on specimens, cut out from the same samples, and the temperature dependence impact strength was determined for the material in as-produced condition. The test results are presented in Fig.4. As the results show that a considerable effect of a previous cyclic loading (close to operation conditions of reactor pressure vessels in zones of stress concentration) on the value and character of temperature dependence of impact strength is not observed [9].

Analogous experiments were carried out in ChSSR as applied to the types 15X2MFA and 15X2HMFA steels. Plane specimens of 12.5 mm thickness were subjected to elastic-plastic deformation at the temperature $20^\circ C$ and stress levels, equal to 1.1 and 0.95 YS. The number of loading cycles makes up 0.5; 0.75 and 0.98 from the number of cycles to failure. Then notches were made on specimens of a semi-elliptical shape (depth - 0.25-0.55 of thickness). The specimen tests at $20^\circ C$ did not
reveal differences in fracture stresses in comparison with specimens, not subjected to such cyclic loading.

![Graph](image)

Fig.4. Variation of impact strength as temperature function of 15X2MFA steel affected by low cycle fatigue and neutron irradiation.

The loading of plane specimens, made from the type 15X2HMFA steel, was carried out in three regimes: a - with a pulsating cycle with the cycle asymmetry \( R = 0.05 \) with maximum load, having created the strain maximum load, having created the strain \( \varepsilon_a = 2.3\% \) at the first cycle to the total plastic strain accumulation 2.5% (about 1000 cycles); b - with a pulsating cycle with the cycle asymmetry \( R = 0.05 \), having created the loads \( 3 \times 10^8 \) cycles; c - loading of plane specimens by tension to the residual (plastic) strain 2.5%. Impact test Charpy specimens were cut from plane specimens and the ductile-brittle transition temperature \( T_{ko} \) was estimated as well as its shift due to the cyclic loading \( \Delta T_N \).

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<td></td>
<td>( T_{ko} )</td>
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<tr>
<td>1</td>
<td>-10</td>
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<td>2</td>
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On the base of the presented data, it is possible to conclude, that the shift value of the ductile-brittle transition temperature (DBTT) from the preliminary cyclic loading \( \Delta T_N \) is equal to 0.

5. THERMAL AGEING

Physical and mechanical properties of most materials will change after long-term operation. The degree of variation of mechanical properties depends on material composition, temperature and a prolonged exposure at the given temperature. According to the requirements of the certification of material the evaluation of the level of properties both in as-produced condition and after exposure for 5000 and 10000 hours at the maximum operating temperatures is carried out. In most cases such exposure does not result in the variation of material properties (Fig.5 and 6).
Fig. 5. Ageing effect on mechanical properties of 08X18N10T steel at various temperatures: o - 290°C, o - 340°C, o - 425°C.

UTS; YS, MPa

Fig. 6. Dependence of mechanical properties of 15X2MFA steel and its weld, produced by SAW, on thermal ageing at 350°C.
The metal state control in the process of operation is carried out by non-destructive and destructive methods [10]. The latter is performed both by testing surveillance specimens (equipment of group A and B) and after specimen cutting from pipings in each 100000 hours of operation for NPP with fast reactors. With the use of surveillance specimens it is possible to control: variation of mechanical properties (YS, UTS, A, Z), brittle fracture resistance (DBTT, fracture toughness or critical crack opening), characteristics of overall and local corrosion (including pitting, stress corrosion cracking, intercrystalline corrosion).

The piping investigations after 100000 hours operation at Sosnovyi Bor NPP (1-4 units), Kursk NPP (1 and 2 units) and Chernobyl NPP (1 and 2 units) - the metal is the type 22K low carbon steel and the type 08X18H12T Cr-Ni steel - showed, that both for base and weld metal the variation of properties was not observed in comparison with the properties at the beginning of operation. Earlier it was shown in [11,12]. Unfortunately, practically there are no data describing the influence of a prolonged influence of operating conditions on NPP equipment metal. However, these results are very important to predict NPP equipment service life. It is shown in [13] that for 15X2HMFA steel two kinds of thermal ageing process revealing are possible. One of them is associated with the precipitation and coagulation of carbides and is characterized by the maximum evidence (to 300°C) on the time dependence of DBTT at 300-350°C (Fig.7). The second kind of embrittlement during longer exposures may take place even at the temperature of the VVER pressure vessel operation.

Fig.7. Thermal ageing effect at 350°C on DBTT of the 15X2NMFA steel (o - base metal, ● - weld metal, Δ - HAZ).

Earlier in [14] it was shown, that after the operation period 10^4 hours of piping weldments from 08X18H12T steel the strength characteristics vary insignificantly. The same phenomenon is observed for fracture toughness, and it is demonstrated by the results, presented in Fig.8.

Fig.8. Temperature dependence of Jic for welded joint of Cr-Ni steel:
● - base metal, τ=0;
X - base metal, τ=10^4hrs,
o,□ - weld, τ=0;
+ - weld, τ=10^4hrs.
6. CORROSION

A reliable operation of nuclear power plants with water coolant depends greatly on physical and chemical processes, determining the generation and transport of corrosion products. Concerning the regulation of these processes, a great effect is provided by the optimum selection of NPP water regime. The variations of oxygen content, pH, environment electroconductivity and the introduction of additions can prevent the formation of deposits at heat transferring and heat releasing surfaces, can reduce the content of corrosion products in coolant and decrease harmful effect of radiolytic decay and prevent steel brittle fracture.

In a long period of operation (for more than 25 years) of NPP equipment with water coolant, damages of corrosion type took place in pressure vessels, collectors and pippings of the primary circuit, as well as in heat exchanging tubes. It is known, that pressure vessels of VVER-440 reactors (modification 230) were partially fabricated without anticorrosive cladding. A great number of corrosion damages of pitting type was typical of them. The density of distribution of corrosion damages, determined by the number of pittings on 1 m² of inner surface, is different in various zones. The maximum density of pittings is observed in three zones [15]: flange joint, shell of core zone and bottom. The causes of damages in these zones are different. In [15] it was studied both general corrosion and crevice and contact corrosion of base and weld metal [16-18]. It was found, that boric acid presence in reactor water increased only corrosion of pearlitic steel during operation. At the reactor operating temperature boric acid introduction at the initial stage of testing decreases the material corrosion properties. After 2000 hours material corrosion rate independent of water chemistry becomes stable at the level of 5-10 mg/m²h [17]. Such behaviour of materials in high temperature water is explained by a dense magnetic film at the metal surface, preventing corrosion to proceed. The results showed, that the reactor steel and its weldments might be considered as corrosion resistant materials.

Later, such considerable corrosion damage of pressure vessel inner surface made it necessary to provide the protection of this surface with anticorrosive cladding and this factor was taken into account by the fabrication of commercial VVER-440 and VVER-1000 reactor pressure vessels. The inner surface of reactor vessels is inspected by dye non-destructive method in 100% volume. The detected indications are often identified as rounded or linear stretched flaws [19]. Depending on their depths, the flaws may be grounded (depths up to 1-3 mm) or repaired (depths - more than 3 mm). Surface flaw detection by dye non-destructive inspection helps to carry out reactor hydrotest during its assembling. During operation these flaws propagate before pearlitic material.

The investigations of water environment effect on crack propagation in pressure vessel materials began, when in western countries cracks were revealed in anticorrosive cladding from austenitic metal. These cracks penetrated in pearlitic metal, which was under the cladding. Some investigations [20,21] were devoted to fatigue crack growth in cladding metal under the attack of water environment, as well as to the peculiarities of a crack propagation when it goes out of anticorrosive austenitic cladding into pearlitic material. In the presence of fatigue crack growth resistance in bimetallic specimens (pearlitic steel and austenitic cladding) we observed crack growth retarding in the vicinity of the fusion line austenite-pearlite, however crack propagation in cladding metal was sufficiently fast. Crack retarding was especially evident by testing in air, and to a smaller degree - in water.

For this heterogeneous system of materials fatigue crack kinetics has been estimated in water environments of various parameters. In Fig.9 fatigue crack
growth is plotted versus various $\Delta K$ values and a good correlation with microhardness variation in the vicinity of fusion line is seen. With sharp microhardness increase (in martensitic structure) fatigue crack growth retardation is observed, which influenced environment temperature and $\Delta K$ value. To clear up the reasons of this effect a transition zone of microstructure was studied on fracture test specimens. Visually detected pearlite-austenite fusion line was a zone of 0.025-0.080 mm width, a special microstructure, containing structure, bordering with a decarburized layer of base metal, and a lamellar structure, passing into austenite. The presence of bifurcation in the relation of $da/dN = C(\Delta K)^m$ points out to the change of fatigue fracture mode. The examination of fatigue test results and microstructure along crack propagation path showed that in these experiments the fatigue bifurcation point corresponded to the position of metal fraction with maximal microstructure hardness on the fusion line of investigated steels.

Fig.9. The variation of crack growth rate and microhardness in the vicinity of bimetal (15X2MFA steel + anticorrosive cladding) fusion line.

Sulphur content (in the form of MnS) in RPV material effects to a great degree corrosion strength of base metal and weldments. Strain induced cracking (SICC) is determined by anodic solution process and the kinetics of repassivation at the crack tip (cracking is transgranular, especially near MnS-inclusions and influence of hydrogen can be observed). The damage mechanism seems to be to some extent comparable to SICC, observed in low carbon steel piping of boiling water reactors in western countries; there, of course, the oxygen content plays a significant additional role [22]. The preconditions for this SICC are a simultaneous action of low alloy steel, low strain rates with stresses being sufficient to cause plastic strains, high oxygen content and temperatures between approximately 150 and 300°C. In all cases failures are associated with additional loadings (e.g. pipewhip restraints which inhibit strain, welding flaws leading to notch effects) or stress concentrations in connection with pitting.

7. CONCLUSION

The present report gives the main mechanisms, which might lead to the variation of material and weldment properties in the process of NPP operation. Of course, the degree of the influence of the stated mechanisms on the degradation of equipment
component material properties is different. Therefore, for each type of equipment it is necessary to determine, in the first place, the dominating factor or mechanism. And it will permit to manage the ageing process, to take measures in time, which would mitigate this process and thus provide a necessary safety and extend the service life of a reliable operation of equipment.

Therefore, ageing effects have to be considered for plant life management. Adequate and reliable monitoring of ageing effects and assessments allowing to make a correct decision in time are required to avoid a reduction of functional capability or even a complete loss of function of plant key components.

To ensure the safe and economic operation of NPP and to maintain required safety and reliability throughout its service life, ageing-related degradation has to be effectively managed. Ageing management (monitoring, assessment and mitigation methods) will provide essential bases for plant lifetime assurance and extension.

REFERENCES


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