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COMPARISON OF THE EMBRITTLEMENT METHODS FOR METAL OF REACTOR PRESSURE VESSELS

СРАВНЕНИЕ НА МЕТОДИТЕ ЗА ОКРЕХКОСТЯВАНЕ НА МЕТАЛА НА КОРПУСИТЕ НА РЕАКТОРИ

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Abstract: The nuclear materials of Reactor Pressure Vessel (RPV) in Nuclear Power Plant (NPP) are submitted to nigh temperature and pressure. A neutron induced embrittlement and thermal ageing of RPV materials are going. Scoping safety exploitation of NPP the embrittlement process must be study. The irradiation causes a shift of the initial critical brittleness temperature T_{K_0} to the higher values (ΔT_K). There are two ways for receiving the critical temperature shift – numerical and experimental. The scope of current work is to be made analysis of four numerical methods. The theoretical results are compared with data from surveillance specimens. It has a good correlation between calculated data (by Methods 2 and 3) and experimental one.
The study is intended to assist in the development of the process of embrittlement of the RPV materials.
Keywords: Reactor Pressure Vessel (RPV), Nuclear Power Plant (NPP), embrittlement process, irradiations, critical brittleness temperature

1. Introduction

Reactor pressure vessel (RPV) beltline materials are exposed to a high-energy neutron flux which results in embrittlement and hardening. The change of properties is increase in yield strength, increase in brittle-ductile transition temperature determined from Charpy V-notch impact or fracture toughness tests, decrease in upper shelf energy, tensile elongation and reduction of area. Scoping safety exploitation of NPP the embrittlement process must be study. The irradiation causes a shift of the initial critical brittleness temperature T_{K_0} to the higher values (ΔT_K). The assessment of the critical temperature of RPV materials and analysis of influences factors have important significance for safety work of NPP. There are two ways for receiving the critical temperature shift – by numerical and by experimental methods.

Numerical methods are developed based of the results and analysis from RPV surveillance specimens [1, 2, 3, 4]. Neutron embrittlement process of RPV materials depends on the metal chemical composition, the neutron dose and the operation temperature. The microstructural testing proves a formation of Cu-Ni-Si-Mn-P clusters [5]. There can be seen Cu atoms are at the center of clusters, Ni, Mn and Si atoms are around the Cu core and P atoms are at the periphery. RPV materials which is used in reactors type VVER 1000 are ferrit-perlit steels. Steel 15Kh2HMFAA and its welds are characterized by lower level of content of P and Cu and higher concentration of content of Ni in comparison to 15Kh2MFA steel and its welds.
The high-energy neutron flux causes a process of neutron and thermal embrittlement of RPV metal [2]. The thermal embrittlement depends on the composition of materials, operation time (worked off time) and temperature. The value of ΔT_K can be obtained by numerical methods using empirical formulae [1, 2, 3, 4], or experimentally by impact three point bending test [7, 8, 9] of unirradiated and irradiated surveillance Charpy specimens.

The scope of this work is to made a comparison between embrettlement methods.

The results shows the embrittlement process is intensive in case of calculations by Method 1, following from these by Methods 2, 4 (in this succession) for base metal. For weld metal the embrittlement process is more intensive in case of calculation by Method 1, following from the results according Methods 2, 3 and 4 (in this succession). The embrittlement rate is more intensive for weld metal in comparison with base metal, but in the beginning of the unit's exploitation the speed of ageing (by Method 2) for base metal is higher than weld metal.

The study is intended to assist in the development of the embrittlement process of the RPV materials.

2. Methodology

Four different computing methods are used for ΔT_K calculation [1, 2, 3, 4] and the results are compared with the data for ΔT_K which is received by experimental way.

Computing methods

Methods 1, 2, 3, 4 are applied for ΔT_K calculation for base metal and weld metal for RPV type VVER 1000 (RPV*a* and RPV*b*). These methods includes the influence of neutron dose and chemical element (Ni, Si, Mn, P, Cu), important for the ageing process. Only one method (method 2 [2]) interprets difference between effects from neutron and thermal ageing. The following designation of parameters are accepted in formulae used for calculation:

- T_{K_0} is the initial critical temperature of metal, $[^0C]$;
- T_K is critical temperature of metal after reactor operation $[^0C]$;
- $\Delta T_K(T)$ is the shift of the critical brittleness temperature due to thermal ageing, $[^0C]$;
- $\Delta T_K(t)$ is the shift of the critical brittleness temperature due to thermal ageing, $[^0C]$;
- t is work off time [hours];
- $\Delta T_K(N)$ is the shift of the critical brittleness temperature due to cycle load, $[^0C]$;
- N is number of cycles;
- $\Delta T_K(F)$ is the shift of the critical brittleness temperature due to neutron fluence F, $[^0C]$;
- F is the neutron fluence with energy of neutrons greater than $0{,}5 MeV$ at RPV, $[\frac{n}{m^2}]$
- $F_0 = 10^{22} n/m^2$ is a standardized coefficient;
- A_F is the irradiation embrittlement coefficient, $[^0C]$;
- ω – double standard deviation of ΔT_K;
- $\Delta T_t^{\inf}$ - is the shift of the critical brittleness temperature when $t = \infty$;
- t_{OT}, t_T, b_T – constants of materials;
- Ni, Mn, Cu и P is the concentration of chemical elements in the composition of material, [weighing units];
- $D = 72.10^{22}, [\frac{n}{m^2}]$ is a standardized coefficient.

2.1.1 Method 1 [1]

This computing method is included in Russian document PNAEG 7-002-86 [1] for calculation of T_K of RPV base and weld metal during operation. It is derived upper 95 % boundary curve of experimental data, available at that time. Following relations are applicable for reactors VVER 1000:

$$T_K = T_{K_0} + \Delta T_K = T_{K_0} + \Delta T_K(T) + \Delta T_K(N) + \Delta T_K(F) \qquad (1)$$

It is accepted that $\Delta T_T = 0$ and $\Delta T_N = 0$ and in this case

$$T_K = T_{K_0} + \Delta T_K(F)$$

$$\Delta T_K(F) = A_F \cdot \left(\frac{F}{F_0}\right)^{\frac{1}{3}} \qquad (2)$$

$A_F = 23^0 C$ for base metal; $A_F = 20^0 C$ for weld metal

The values of $\Delta T_K(F)$ based on Method 1 includes a cumulative effect from both neutron and thermal embrittlement of RPV metals.

2.1.2 Method 2 [2]

The Method 2 is developed by CNII Prometey, RNC Kurchatovskij Institute, OKB Gidropress and it is recommended by International Atomic Energy Agency [2]. This method is based on the investigation of the influence of high Ni content on RPV metal. It was established a process of thermal ageing of RPV materials is going during unit operation. The following method for calculation T_K shift is proposed for reactors VVER 1000 [2].

$$T_K = T_{K_0} + \Delta T_K(F, t)$$
$$\Delta T_K(F,t) = \Delta T_K(F) + \Delta T_K(t) + \omega \qquad (3)$$

$$\Delta T_K(F) = A_F \left(\frac{F}{F_0}\right)^m \qquad (4)$$

<u>for base metal</u> $m = 0{,}8; A_F = 1{,}45^0 C$

<u>for weld metal</u> $m = 0{,}8; A_F = \alpha_1 . \exp(\alpha_2 . C_{eq}); [^0C]$

$C_{eq} = Ni + Mn - \alpha_3 .Si$ if $Ni + Mn - \alpha_3 .Si \geq 0$

or $C_{eq} = 0$ if $Ni + Mn - \alpha_3 .Si < 0$

$\alpha_1 = 0.703;\ \alpha_2 = 0.883;\ \alpha_3 = 3.885$

$$\Delta T_K(t) = \left(\Delta T_t^{\inf} + b_T \exp\left(\frac{t_T - t}{t_{OT}}\right)\right) \cdot th\left(\frac{t}{t_{OT}}\right) \qquad (5)$$

Material	$\Delta T_{\inf} [^0C]$	$b_T [^0C]$	$t_{OT} [hours]$
Base metal	18	26,2	32 700
Weld metal, Ni>1,3%	18	10,1	23 200
Weld metal, Ni<1,3%	18	26,2	32 700

Table 1. The values of $\Delta T_{\inf}, b_T, t_{OT}$ of RPV metal

Method 2 suggests a separated assessment of the shift of critical temperature ΔT_K due to neutron fluence (a neutron part) and due to thermal ageing (a thermal part). The values of fluence and the values of weight percents of the elements nickel Ni, manganese Mn, silicon Si are important factors for neutron embrittlement. The composition of the materials, temperature and worked off time are important factors for the thermal embrittlement.

2.1.3 Method 3 [3]

This method is developed trough many testing of surveillance specimens, providing by RNC Kurchatovskij Institute [3]. For first time is taken into account the influence of the elements Ni, Mn, Si on the embrittlement rate. The method is published before establishment of the presence thermal effect. Method 3 suggests $\Delta T_K(F)$ includes a total effect of neutron and thermal embrittlement. The method is not accepted as a standard; calculation of the shift of the critical temperature ΔT_K is referring for weld metal only.

$$T_K = T_{K_0} + \Delta T_K(F) \qquad (6)$$

$$\Delta T_K(F) = 33{,}5.Ni^{1,35}.Mn^{0,7}.(0{,}64 - Si).\left(\frac{F}{F_0}\right)^{\frac{1}{3}} \qquad (7)$$

2.1.4 Method 4 [4]

This method is developed by OA Shopen National Science Center „Kharkov Institute of Physics and Technology”, Kharkov, Ukraine (1997) for prediction of embrittlemen rate of RPV metals [4]. The Method is not standardized. There is taken into account the influence of the elements nickel Ni, cuprum Cu and phosphor P on the embrittlement rate. Method 4 suggests $\Delta T_K(F)$ includes both an neutron and a thermal part of the embrittlement.

$$T_K = T_{K_0} + \Delta T_K(F) \qquad (8)$$

$$\Delta T_K(F) = \frac{\left[220.(Ni)^{0,5}.Cu + 3400.P + 39.Ni\right]\left(\frac{F}{D}\right)^3}{1 + \left(\frac{F}{D}\right)^3} + 8 \qquad (9)$$

2.1.5 Method 5 (Experimental Method)

The testing of surveillance specimens (SS) was completed, in the frame of other project. There was applying the three point impact bending testing of Charpy [7, 8, 9]. The obtained results for the shift of the critical temperature ΔT_K are used for the current investigation [10÷13].

2.2 Input parameters

The calculation of the neutron and thermal embrittlement rate was performed for base metal BM (15Kh2HMFAA steel) and weld metal WM (SV 12Ch3NMFA-A) of two RPV type VVER1000 (*a* and *b*).

The values of the neutron fluence F used for calculations are in the range $1.10^{18}\frac{n}{m^2} \div 20.10^{18}\frac{n}{m^2}$. This values of the fluence F and the operation time t corresponds to the mean values for one fuel cycle of reactor type VVER 1000, as follow:

For base metal: $\overline{F} = 0{,}8.10^{18}\frac{n}{m^2}$ (for one fuel cycle);

For weld metal: $\overline{F} = 0{,}79.10^{18}\frac{n}{m^2}$;

Time $\bar{t} = 7200 hours$;

The neutron energy of the fluence F is accepted $E > 0{,}5MeV$.

3. Discussion

3.1 Methods 1, 5

The rate curves of the shift of the critical temperature $\Delta T_K(F)$ by Method 1 for base metal (BM) and weld metal (WM) are shown on Figure 1. The mean prognostic curve based on experimental data are presented on the same figure.

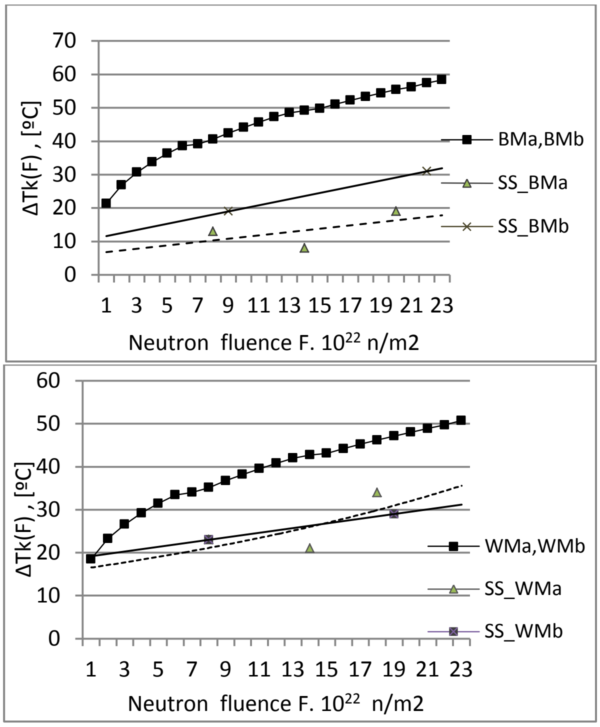


Fig. 1. Dependency of $\Delta T_K(F)$ on fluence F for a) BM and b) WM, calculated by Method 1. The mean lines from experimental data .

The embrittlement rate of BM is higher than WM rate due to the different chemical coefficients A_F. The trend curves $\Delta T_K(F)$ based on the calculated data for base and weld metal are disposed vastly above the experimental curves. In the case Method 1 gives very conservative values of $\Delta T_K(F)$ assessment of embrittlement rate up to fluence $20.10^{22}\frac{n}{m^2}$.

3.2 Methods 2, 5

The shifts of critical temperature $\Delta T_K(F)$ due to the neutron fluence, by Method 2 for both base and weld metal are presented on Figure 2.

The values of $\Delta T_K(F)$ for both base and weld metal exponentially increases with the growth of the fluence F. The trend curves $\Delta T_K(F)$ for base metal (BMa and BMb) coincides, because of A_F doesn't depend the metal chemical composition for base metal. It can be seen the difference of the speed of neutron embrittlement rate for weld metal (WMa and WMb). The embrittlement rate is going faster for WMa. For RPVb - the trend curves $\Delta T_K(F)$ for base metal BMb and weld metal WMb coincides almost.

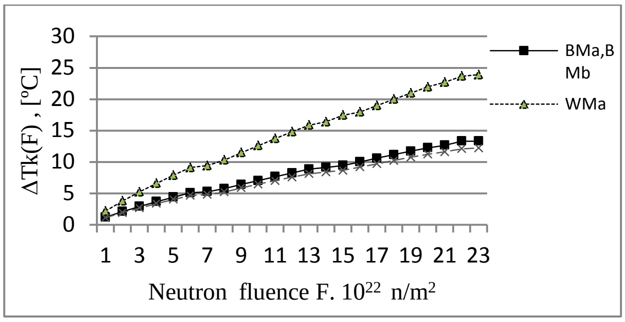


Fig. 2 . Dependency of $\Delta T_K(F)$ on fluence F for BM and WM, calculated by Method 2, or a neutron inducted part of $\Delta T_K(F,t)$

The shift of the critical temperature due to thermal embrittlement $\Delta T_K(t)$ by Method 2 for base metal and weld metal (BMa, BMb, WMa, WMb) are presented on Figure 3.

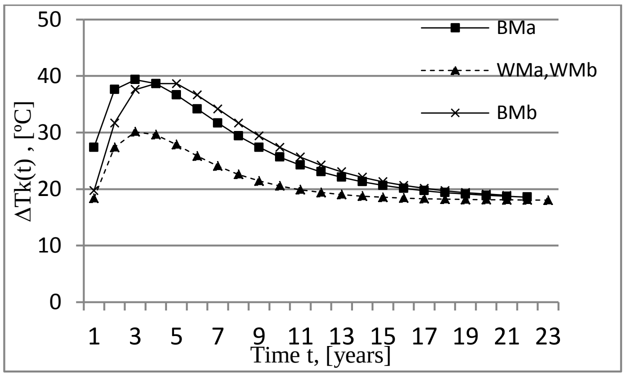


Fig. 3. Dependency of $\Delta T_K(t)$ on time t (or thermal inducted part of ΔT_K) for BM and WM, calculated by Method 2.

The thermal inducted $\Delta T_K(t)$ trend curves for base metal (BMa, BMb) prevails the trend curve for weld metal (WMa, WMb). It can be seen a pick of values of the thermal embrittlement in a period years $2-5$, after the thermal embrittlement rate is quickly reducing. After first years $10-11$ the function $\Delta T_K(t)$ has almost constant values during period of exploatation of NPP, for both base and weld metal.

The sum of the shift of critical temperature due to neutron and due to thermal embrittlement:

$$\Delta T_K(F,t) = \Delta T_K(F) + \Delta T_K(t)$$

for both base and weld metal are presented on Figure 4.

The thermal inducted embrittlement process prevails for a period of the first years 10 of NPP's work, according Figure 3, after that prevails the neutron inducted embrittlement process, Figure 4. In the begging of NPP exploitation the embrittlement rate of base metal BMa, BMb prevails that of the weld metal WMa, WMb. After the values of $\Delta T_K(F,t)$ for base metal are reducing slowly, the values of $\Delta T_K(F,t)$ for weld metal growth. The values of $\Delta T_K(F,t)$ for WMa quickly growths too. The speed of the neutron embrittlement rate for WMa is higher than BMa, BMb and WMb.

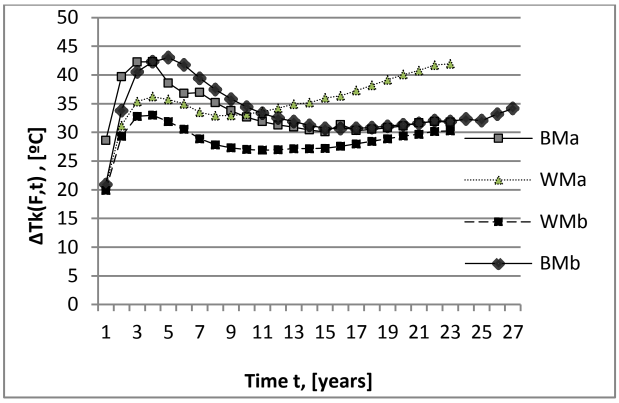


Fig. 4 - Dependency of $\Delta T_K(F,t)$ on worked off time t for BM and WM, calculated by method 2.

A comparison between the values of the shift of the critical temperature $\Delta T_K(F,t)$ based on calculation by Method 2 and experimental data from surveillance specimens SS is made. The results are shown on the Figure 5a (BM) and Figure 5b (WM).

For base metal – calculated results by Method 2 are higher than the experimental one. The trend curves $\Delta T_K(F,t)$ is prevailing the values from experimental SS BMa data, in opposite to the case of base metal BMb.

For weld metal WMa - the trend curve $\Delta T_K(F,t)$, based on the calculations by Method 2 is high and quickly growths. The trend curve $\Delta T_K(F,t)$ for WMb coincides (almost) with the mean lines from experimental data SS_WMb.

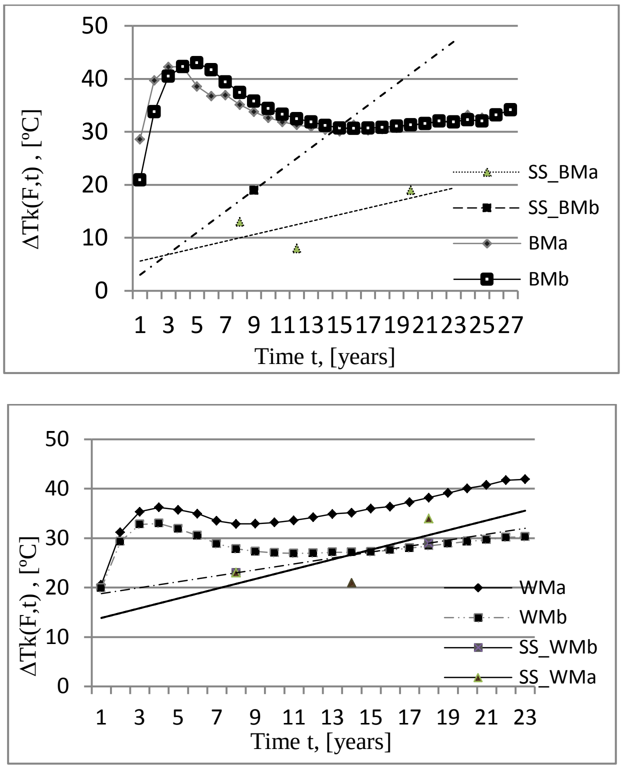


Fig. 5 - Dependency of $\Delta T_K(F,t)$ on worked time t for a) BM and b) WM, calculated by Method 2.
The mean lines from experimental data from surveillance specimens SS for a)BM and b)WM.

3.3. Methods 3, 5

A comparison between the calculated data by Method 3 and the experimental data for the shift of critical temperature $\Delta T_K(F)$ is made, (Figure 6).

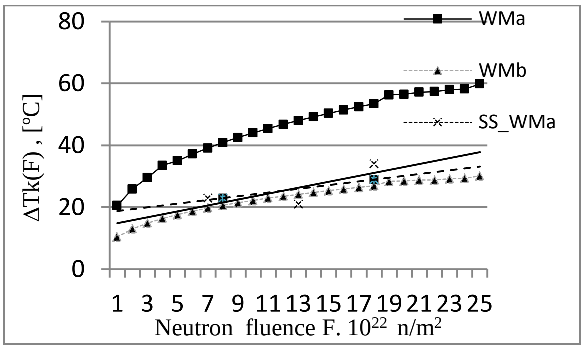


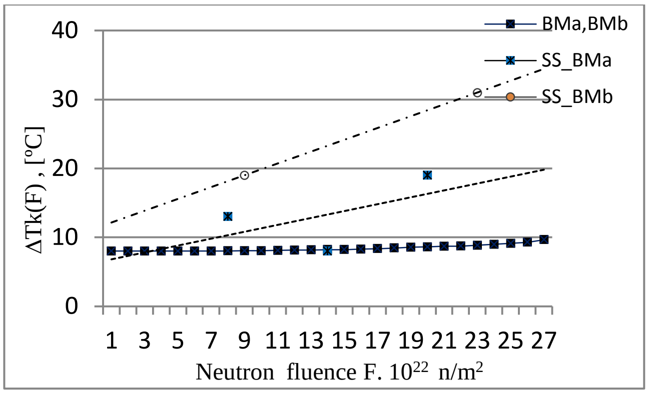
Fig. 6. Dependency of $\Delta T_K(F)$ on fluence F for WM, calculated by Method 3.
Experimental data from surveillance specimens SS, mean curves.

It can be seen on Figure 6 the values of the shift of $\Delta T_K(F)$ for WMa is very high and prevails that of the surveillance specimens. The trend curve of the shift of $\Delta T_K(F)$ for WMb coincides (is very close) with mean curve from the experimental data SS_WMb. The results shows a good correlation of the calculated and the experimental data for WMb. But for WMa calculated values are higher than experimental one.

3.4 Methods 4,5

The function $\Delta T_K(F)$ on fluence F, calculated by method 4 is presented on Figure 7, as well experimental data from surveillance specimens SS; for a) the base metal and for b) the weld metal.

The results from Figure 7 shows the trends curve of the shift $\Delta T_K(F)$, calculated by Method 4, coincides for BMa and BMb; as well for WMa and WMb. The assessment of the shift $\Delta T_K(F)$ from experimental data for base metal (SS_BMa, SS_BMb) and weld metal (SS_WMa, SS_WMb) are higher than calculated data.



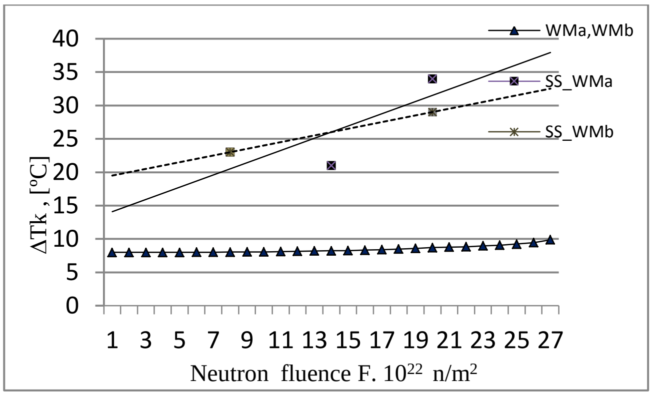


Fig. 7. Dependency of $\Delta T_K(F)$ on fluence F, calculated by method 4 for a)BM and b)WM. Experimental data from surveillance specimens SS and mean curves.

3.5 Methods 1-5

The prognostic curve for the shift of critical temperature $\Delta T_K(F)$ of the embrittlement during unit operation, calculated according different methods 1-4 are compared on Figure 8 for a)base metal and for b) weld metal. The result from the surveillance specimens SS, according Method 5 are shown on the same figure.

For base metal BM (Figure 8a): The values of the shift of the critical temperature $\Delta T_K(F)$ are biggest by Method 1, followed from these by methods 2 and 4 (in this order). For the first 10 years of NPP's work it can be seen a good correlation between 1) the data for the shift $\Delta T_K(F)$ for BMa, BMb, calculated according method 4 and 2) the experimental data SS_BMa. It is observing the trend curve $\Delta T_K(F)$ for BMa and BMb, based on calculations by Method 2, is closely to the mean curve for SS_BMb.

For weld metal (Figure 8b): The embrittlement rate of the curve $\Delta T_K(F)$ is most intensive for WMa (by Method 3). There is a very important exception – $\Delta T_K(F)$ calculated by Method 3 for WMa has very high values, prevailing even $\Delta T_K(F)$, calculated by Method 1. The embrittlement rate is intensive for WMa, WMb (calculated by Method 1), followed by the embrittlement rate for WMa, WMb (by Method 2), followed - for WMb (by Method 3) and finally – the embrittlement rate is less for WMa, WMb (by Method 4).
From Figures 8a, 8b it can be seen the different speed of the embrittlement process for base metal and weld metal. Calculated and experimental results defines the embrittlement rate is more intensive for weld metal in comparison with base metal.

Comparatively the other methods, Method 1 defines quickly growth of the rate of the embrittlement process, (Fig. 8a, 8b).

It can be seen the values of the calculated data of the shift $\Delta T_K(F)$ for WMb (by both Methods 2 and 3), are placed closely to the experimental data.

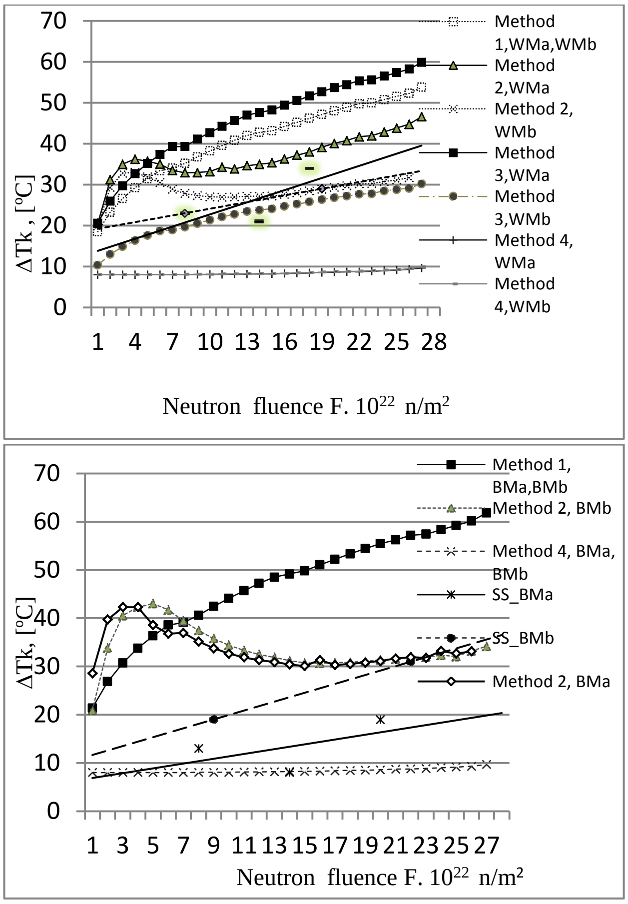


Fig.8. The prognostic curves for the shift $\Delta T_K(F)$, calculated according methods 1-4 for a) BM and for b) WM. Experimental data from surveillance specimens SS and mean curves.

4. Conclusions

There are presented 4 methods for calculation of the shift of the critical brittleness temperature $\Delta T_K(F)$. The trend curves of the calculated $\Delta T_K(F)$ were compared with the experimental data from surveillance specimens (Method 5).

Method 1 [1], according PNAEG 7-002-86, interprets important factors for the embrittlement process – the values of neutron fluence F and type of metal (base or weld). The shift of the critical temperature $\Delta T_K(F)$ calculated by Method 1 prevails results, calculated by the other methods, as well as the experimental data. Method 1 is quite conservative for calculations of the embrittlement rate due to irradiation of the metals. The reason is the approach for assessment of values of A_F doesn't depend from chemical content of the metal.

The factors, important for the embrittlement, according Method [2] are neutron fluence F, weight percents of the elements Ni, Mn, Si and worked off time t of the nuclear materials. Method [2] defines a different parts of the neutron and thermal embrittlement. At the beginning of the exploitation of nuclear power plant the thermal embrittlement of BM and WM prevails, after that the thermal embrittlement decreases. After a period from years $10-11$ thermal inducted part of embrittlement process has almost constant values. The function $\Delta T_K(F)$ - or neutron inducted part - growths exponential all the time during the exploitation of the NPP's unit. In generally it can be say the thermal inducted embrittlement process prevails for a period of the first 10 years of NPP work, after that prevails the neutron inducted embrittlement.

Method 3 defines the influence of neutron fluence F and weight percents of the elements Ni, Mn, Si for the embrittlement rate. Method 3 gives comparatively low speed of the embrittlement process. The assessment of the embrittlement rate, calculated by Method 4, depends on the influence of neutron fluence F and weight percents of the elements Ni, Cu and P. Method 4 defines a lowest speed of the embrittlement process, in comparison with other methods. As compare the trend curves of $\Delta T_K(F)$ based on all methods for base metal, and the mean curves from experimental data, it can be seen the function $\Delta T_K(F)$ has biggest value and growth in case of calculation by Method 1, following from the results according Methods 2, 4 (in this succession). For weld metal the function $\Delta T_K(F)$ has biggest value and growth in case of calculation by Method 1, following from the results according Methods 2, 3 and 4 (in this succession).

The embrittlement rate is more intensive for weld metal in comparison with base metal, but in the beginning of the unit's exploitation the speed of ageing (by Method 2) for base metal is higher than of weld metal.

5. References

[1] PNAEG 7-002-86 Strain norms for equipment, type VVER, Moskow,1986;www.serconsrus.ru/images/seism/PNAEG_7002_86.pdf; Appendix 2

[2] Unified procedure for integrity and lifetime assessment of components and piping in WWER NPPs during operation Verlife, IEAE, 2010; Nulife 2014, Project RER 4030; http://www.iaea.org/inis/collection/NCLCollectionStore/_Public/43/070/43070847.pdf#search=%22Verlife%20procedures%22;

[3] Radiation embrittlement of VVER-1000 RPV steels, Nikolaev Yuri A.,18th International Conference on Structural Mechanics in Reactor Technology, China, 2005.

[4] Investigation of the kinetic of degradation of properties of welds and development of models for lifetime assessment of welds of RPV, type WWER in Ukraine, Mahnenko W.I., Kasatkin O.G., Welding on the Threshold of XXI century, Bulgaria, 1999.

[5] Understanding on the Mechanisms Irradiation Embrittlement of RPV steels and developement of Embrittlement Correlation Method, IAEA Training Workshop on Assessment of degradation Mechanisms of Primary Components in Water Cooled Nuclear Reactors, Akiyoshi Nomoto, Madrid, Spain, 2014.

[6] Report for definition of mechanical characteristic and structural condition of set with surveillance specimens, BAN, Bulgaria, 1995; 1998.

[7] GOST 6996-66: Welds. Methods for definition of mechanical properties; ISO/TC 164/SC 4/N 191; gostexpert.ru/gost/getDoc/9083

[8] ISO/CD 14556 Steel –Charpy V Pendulum impact test – Instrumented test method; www.freestd.us › Standards Wordwide

[9] ASTM E 23-92 Standard Test Methods for Notched Bar Impact Testing of Metallic Materials

[10] Methodology for determination of the RPV surfaces neutron flux (outside), type WWER 1000; RB-018-01, GosAtomNadzor, Moscow, 2001

[11] Methodology for calculations of the neutron flux, with energy over 0,5 MeV on the position both of the detectors and the surveillance specimens

[12] Calculations and Dosimetry Methods for determining Pressure Vessel Neutron Fluence, Draft Regulatory Guide DG-1025; U.S. Nuclear Regulatory Commission, 1993
[13] Computing Radiation Dose to Reactor Pressure Vessel and Internals, state-of-the-Art Report, NEA/NSC/DOC (95)5, Nuclear Energy Agency, 1997