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Radiation-Induced Embrittlement of WWER-440 Reactor Pressure Vessel Steel under Loading

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Радиационное охрупчивание корпусной стали реактора ВВЭР-440, находящейся под нагрузкой

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Приведены результаты определения эталонной температуры T_0 *и построена "Master curve" на основе экспериментов, выполненных для стали марки 15Х2МФА (основной металл корпуса реактора типа ВВЭР-440) в трех состояниях: необлученном, облученном и облученном под нагрузкой. Показано, что механическая нагрузка, имитирующая давление теплоносителя, ускоряет радиационное охрупчивание, причем вклад ее сравним с вкладом нейтронного облучения.*

Ключевые слова: нейтронное облучение, радиационное охрупчивание, корпусная сталь, "Master curve", механическая нагрузка, давление теплоносителя.

Introduction. At the time being, the radiation embrittlement of the reactor vessel materials is evaluated by the results of the Charpy-type surveillance specimens or fatigue pre-cracked compact tension specimens. The surveillance specimens are irradiated in the hermetically closed container assemblies without stress. The reactor vessel material is actually subjected to the pressure of the coolant.

For determining the effect of mechanical load under irradiation on the embrittlement rate of the reactor vessel materials two container assemblies were irradiated in commercial unit. They were completed with the specimens cut of four various grades of steel, having been utilized for manufacturing reactor pressure vessels in the former Soviet Union. Three kinds of specimens were prepared of each type of steel, namely:

- static tensile specimens;
- Charpy-V type impact specimens;
- *T-L*-oriented stressed and unstressed compact tension specimens.

The specimens of each steel were located in three layers. There were 2 tensile, 2 Charpy-V and 4 CT (2 stressed and 2 unstressed) specimens in every layer of each assembly. Thus, each assembly was completed with six stressed and six unstressed 1/2T compact tension specimens manufactured of each type of vessel steel.

Irradiation was performed on such a level of the reactor core that within one year of irradiation to accumulate the dose comparable with that accumulated by the reactor core in the center of the active zone during the design service life (40 years).

The primary material property, determining its susceptibility to spalling, is the ability of the material to resist the propagation of a crack. For reactor steels the ductile-brittle transition temperature shift in the irradiated state has to be defined as compared to the unirradiated one, therefore the application of large-size surveillance specimens is unacceptable.

Employing the Master Curve approach for ferritic steels enables to obtain credible values of the critical stress-intensity factor K_{Jc} in the wide temperature range testing small-size specimens [1]. According to the recent standardizing documents, Master Curve is constructed on the basis of the results of testing small-size specimens at a single temperature such that steady growth of a crack before the beginning of the fracture is practically precluded. This allows to simplify considerably the procedure of defining J-integral and use experimental basic approach of nonlinear fracture mechanics.

The paper presents the results of determining the reference temperature T_0 and constructing a Master Curve proceeding from the experiments, carried out for the steel 15Cr2MoVA (base metal of WWÉR-440 reactor vessel metal) in irradiated, unirradiated, and irradiated stressed states.

Material and Specimens. Chemical composition of the investigated steel is presented in Table 1. 1/2T CT specimens were studied. Fatigue cracks (a_0) on them were initiated in line with the requirements of the State Standard 25.506-85 [2].

Table 1

Chemical Composition of 15Cr2MoVA Steel

C	Si	Mn	Ni	Cr	S	P	Cu	V	Mo
0.14	0.2	0.36	0.11	2.0	0.012	0.007	0.09	0.2	0.6

The vessel metal of an operating WWÉR-type reactor is under the pressure of the coolant. In WWÉR-1000 it equals 16 MPa, that results in the stress of the vessel metal of about 173 MPa. For imitating such stresses mechanical load was created on CT specimens by means of the loading screw and metallic bellows. Stressed specimens were assembled into two chains, linked with two bellows, next to unstressed specimens, enabling the correct comparison of the data for both groups of specimens.

The design of the assemblies allowed water of the primary circuit to wash the specimens, so their irradiation temperature was equal to the temperature of the coolant, i.e., ~290°C.

Fast neutron fluences ($E > 0.5$ MeV) accumulated by the studied specimens in experimental assemblies are calculated by the Kurchatov Institute in the framework of the Project TACIS PCP-IV [3] and are illustrated in Table 2.

Table 2

Calculated Fluences Defined for Middle Areas of Each Assembly Layer

Steel grade	Layer number	Neutron fluence ($E > 0.5$ MeV), n/cm^2	
		Assembly No. 1	Assembly No. 2
Base metal 15Cr2MoVA	7	$9.15 \cdot 10^{19}$	$9.24 \cdot 10^{19}$
	8	$1.04 \cdot 10^{20}$	$1.05 \cdot 10^{20}$
	9	$1.16 \cdot 10^{20}$	$1.17 \cdot 10^{20}$

Methods of Testing. Static tensile testing was conducted in accordance with the State Standard 1497-73 [4]. Remote-controlled tensile-testing machine installed in the "hot" chamber was applied as experimental equipment. The error of the breaking stress determining was ±25 MPa. The active grip moved with the speed of 1 mm/min. Tests were performed at room temperature and at 350°C. Keeping temperature was with the accuracy ±2°C. The maximum error of determining strength characteristics is 5% and that of plasticity – 2%. The results of static tensile tests are shown in Table 3.

Table 3

Radiation-Induced Changes of Mechanical Characteristics of Base Metal 15Cr2MoVA Averaged over Specimens of Both Assemblies

T_{test}, °C	$R_{p0.2}$, MPa	R_m, MPa	A_0, %	A_m, %	Z, %
20	333	441	17.0	7.3	77
	601	670	5.1	2.2	44
Change, %	80	51	−70	−70	−43
350	316	387	12.3	3.6	80
	449	506	5.0	2.5	43
Change, %	42	31	−59	−31	−46

Note. Over the line are given results for unirradiated specimens and under the line – for irradiated specimens.

Fracture toughness tests were carried out in line with ASTM Standard 1921-97 [1] on the testing electromechanically-driven machine Instron-8500 with the critical load 100 kN. The machine is remote-controlled and it is installed in the "hot" chamber of the Institute for Nuclear Research of the National Academy of Sciences of Ukraine in the framework of the program TACIS PCP-IV. The procedure of measuring temperature, load and crack opening satisfied the requirements [1] that provided determination of the above characteristics with high accuracy and obtaining reliable $P-V$ diagrams.

Since crack opening was measured on the front surface of a specimen, according to p. 7.1 [1] the correction for displacement of the loaded line was applied by multiplying the measured values by 0.73.

Fracture toughness testing temperature was found as

$$T = T_{28\mathrm{J}} + C, \tag{1}$$

where $C = -28$°C for 1/2T specimens [1]. The temperature $T_{28\mathrm{J}}$ was defined by the results of Charpy impact testing ($10 \times 10 \times 55$ mm) on the pendulum hammer KMD-30D with impact accumulated energy 300 J in the temperature range (-80°C)–($+100$°C) in line with the standard [5] (Fig. 1). The processing of impact toughness data (E_{ab}) was conducted according to [6] by means of the approximation function of hyperbolic tangent, the equation of which looks like

$$E_{ab}(T) = A + B \tanh\left(\frac{T - T_{k0}}{C}\right), \tag{2}$$

where E_{ab} is the absorbed energy (J), A is the average value of E_{ab} between the maximum ($E_{\max}$) and minimum ($E_{\min}$) values, $B = (E_{\max} - E_{\min})/2$, T_{k0} is the temperature corresponding to the value A, and C is the empiric constant. Parameters A, B, C, and T_{k0} are defined by the processing of the experimental points by the methods of least squares.

Ductile-brittle transition temperature for unirradiated specimens (T_{k0}) is -62°C and for irradiated ones it is $T_{kF} = -7$°C.

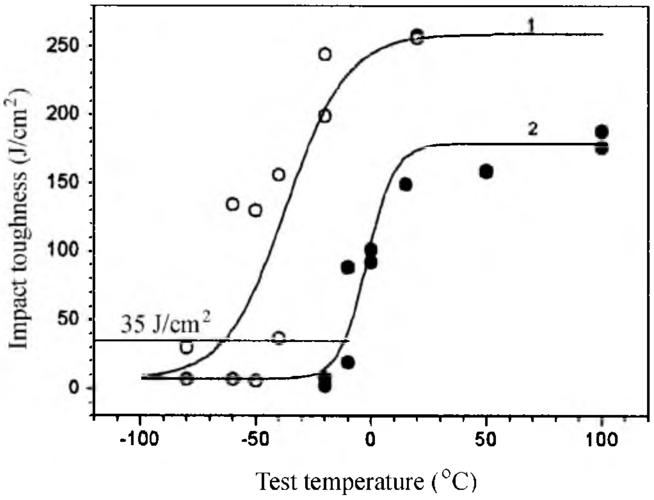


Fig. 1. The temperature dependence of absorbed energy of base metal 15Cr2MoVA.

From the data of Fig. 1 the fracture toughness test temperature $T_{28\mathrm{J}} = T_{35\mathrm{J/cm}^2}$ for unirradiated specimens was -62°C and for irradiated ones it was -11.5°C. Thus, according to the standard [1] and on the basis of Eq. (1) fracture toughness tests have to be performed at the temperature $T = -90$°C and $T = -40$°C for unirradiated and irradiated specimens, respectively.

Obtained Results. As a result of testing seven unirradiated specimens at -90°C $P-V$ diagrams, characteristic for cleavage cracking, have been got. By the results of processing these diagrams the values of J-integral were defined as the sum of elastic and plastic components. The values of the stress intensity factor (K_{Jc}) were calculated from the J_c values obtained for each individual specimen. The values of J_c and K_{Jc} for unirradiated specimens of steel 15Cr2MoVA are illustrated in Table 4.

$P-V$ diagram and the fracture surface in first irradiated specimen at the testing temperature $T_{test} = -40$°C testify the considerable stable growth of a crack. The value of K_{Jc} for this specimen is equal to 129.95 $\mathrm{MPa\sqrt{m}}$ (Table 4). According to the recommendations of CSRIEM "Prometei" on the assessment of fracture toughness in WWÉR-440, WWÉR-1000 reactor vessel materials, the testing temperature was lowered by 20°C, but at $T_{test} = -60$°C the stable crack growth was also more intensive. At $T_{test} = -80$°C cleavage cracking took place in the irradiated specimens. Therefore all the rest irradiated (stressed and unstressed) specimens were tested at $T_{test} = -80$°C. The results of testing all the specimens are shown in Table 4. None of the K_{Jc} values should be qualified as all the values do not exceed $K_{Jc(\mathrm{limit})}$ value for the above testing temperature.

Master Curve Approach Application. Master Curve approach is used for construction temperature dependence of fracture toughness on the basis of test results of limited quantity of specimens [7, 8]. The position of the curve $K_{Jc}(T)$ on the temperature coordinate is established from experimental determination of the temperature T_0 at which the median value $K_{Jc(med)}$ for 1T specimens is 100 $\mathrm{MPa\sqrt{m}}$. Ferritic steels are known to be very heterogeneous due to the orientation of individual grains as well as to heterogeneities of the grain boundaries. Carbides and different non-metallic inclusions on the grain boundaries may be nucleus of microcracks. Their random location relative to the crack front in the material conditions the large spread in values of fracture toughness testing small-size specimens.

As it is shown by Weibull [9] the curve shape for ferritic steels with yield strength from 275 to 825 MPa is determined by the index of the exponential curve $b = 4$ and $K_{\min} = 20\ \mathrm{MPa\sqrt{m}}$. These data are received on the basis of the statistic analysis of a huge number of experimental data, obtained by various researchers all over the world on the specimens of different sizes.

According to the requirements [1], whose adequacy has been confirmed in [10], the values obtained on specimens of size 0.5T were recalculated into the values equivalent to those obtained on specimens of size 1T by the formula

$$K_{Jc(1\mathrm{T})} = 20 + [K_{Jc(1/2\mathrm{T})} - 20]\left(\frac{B_{1/2\mathrm{T}}}{B_{1\mathrm{T}}}\right)^{1/4} \qquad (3)$$

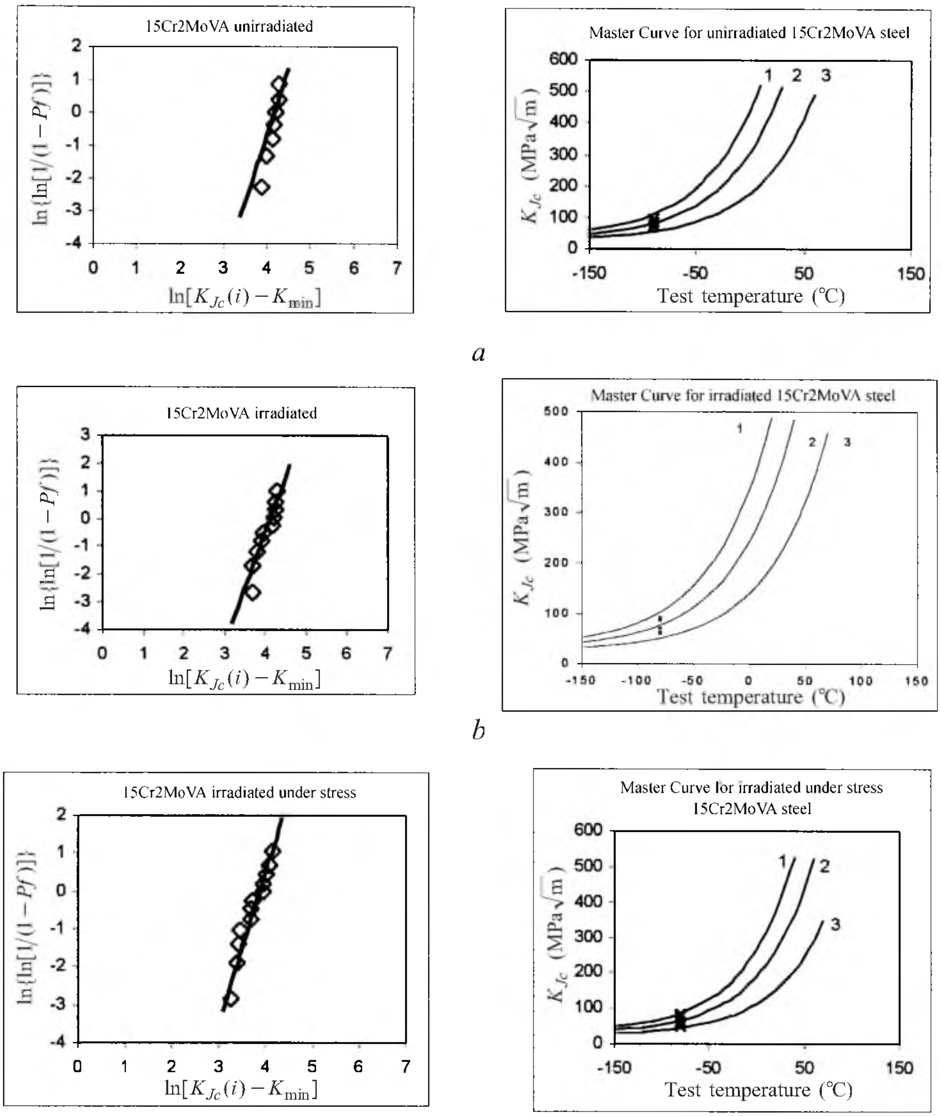
Table 4

Results of Fracture Toughness Testing of Base Metal 15Cr2MoVA

No.	State	T_{test}, °C	$K_{Jc(1/2T)}$, MPa$\sqrt{\text{m}}$	$K_{Jc(1T)}$, MPa$\sqrt{\text{m}}$	K_0, MPa$\sqrt{\text{m}}$	$K_{Jc(med)}$, MPa$\sqrt{\text{m}}$	T_0, °C
1	Unirradiated	−90	77.57	68.42	85.58	79.83	−72.1
2		−90	85.53	75.11			
3		−90	93.65	81.94			
4		−90	98.09	85.67			
5		−90	98.93	86.38			
6		−90	106.12	92.44			
7		−90	107.09	93.24			
1	Irradiated unstressed	−80	67.24	59.73	81.98	76.56	−58.5
2		−80	67.92	60.30			
3		−80	73.98	64.81			
4		−80	80.02	70.47			
5		−80	86.28	71.84			
6		−80	98.42	85.95			
7		−80	103.83	88.92			
8		−80	101.95	90.50			
9		−80	104.93	91.43			
10		−80	106.48	92.73			
11		−40	129.95	112.46			
12		−60	144.78	124.94			
1	Irradiated stressed	−80	51.51	46.50	68.60	64.35	−42.5
2		−80	55.17	49.58			
3		−80	56.45	50.66			
4		−80	58.30	52.21			
5		−80	67.64	60.07			
6		−80	68.15	60.50			
7		−80	69.26	61.42			
8		−80	82.22	72.33			
9		−80	82.24	72.34			
10		−80	86.94	76.30			
11		−80	91.83	80.41			
12		−80	94.55	82.70			

and then we calculated the scale parameter

$$K_0 = \left[\sum_{i=1}^{N} \frac{(K_{Jc(1\text{T})})^4}{N - 0.3068} \right]^{1/4} + 20, \tag{4}$$



a

b

c

Fig. 2. Weibull's plots and Master Curves for three states of specimens: unirradiated (*a*), irradiated without stress (*b*) and irradiated under mechanical load (*c*): (*1*, *3*) tolerance bounds 95% and 5%, respectively; (*2*) Master Curves.

and the mean value of the sample:

$$K_{Jc(med)} = (K_0 - 20)[\ln 2]^{1/4} + 20. \tag{5}$$

The magnitude of the reference temperature (T_0) was determined using the following expression:

$$T_0 = T_{test} - \frac{1}{0.019}\ln\left(\frac{K_{Jc(med)} - 30}{70}\right). \tag{6}$$

The values of the parameters calculated according to formulas (3)–(5) for three states of steel 15Kh2MFA are listed in Table 4. Master Curves (Fig. 2) were constructed by the equation [1]:

$$K_{Jc(med)} = 30 + 70\exp[0.019(T - T_0)]. \tag{7}$$

For visual representation of the results of testing, Weibull's model was used according to which the probability of fracture is evaluated by the formula

$$P_f = 1 - \exp\{-[(K_{Jc} - K_{\min})/(K_0 - K_{\min})]^b\}, \tag{8}$$

where $K_{\min} = 20\ \mathrm{MPa}\sqrt{\mathrm{m}}$ and $b = 4$.

Figure 2 demonstrates that the data of K_{Jc} lie within ±5% confidence interval of Master Curve approach.

Mean-square deviations T_0 estimated according to [1] are equal ±7°C for unirradiated specimens and ±6°C for irradiated ones in both states.

Figure 3 illustrates Master Curves for three states of steel 15Cr2MoVA: unirradiated (*1*), after irradiation unloaded (*2*), and irradiated loaded (*3*). Reference temperature T_0 for irradiated unstressed specimens exceeds its value for unirradiated specimens. Total effect of neutron irradiation and stress results in the increase of the reference temperature in this case by 30°C.

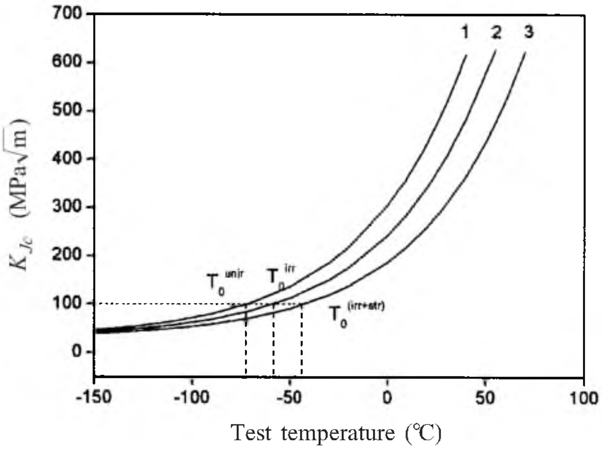


Fig. 3. Master Curves for three states of specimens: (*1*) unirradiated, (*2*) irradiated, and (*3*) irradiated under mechanical load.

Conclusions. The effect of neutron irradiation on radiation embrittlement of Ni-free vessel steel 15Cr2MoVA is studied in two states: under mechanical load and without it.

The effect of stressed state due to mechanical load, imitating the pressure of the coolant, on the ductile-brittle transition temperature of the fatigue pre-cracked specimens of steel is discovered. The above effect is revealed in the reference temperature T_0 for stressed irradiated specimens being higher than for irradiated unstressed ones. The effect of the stressed state caused by mechanical load, imitating the pressure of the coolant, on the ductile-brittle transition temperature is qualitatively comparable with the effect of neutron irradiation. From the physical point of view, this phenomenon is conditioned by the higher rate of the formation of radiation defects due to the total effects of neutron irradiation and stress.

Резюме

Наведено результати визначення еталонної температури T_0 та побудовано "Master curve" на основі експериментів, проведених для сталі марки 15Х2МФА (основний метал корпусу реактора типу ВВЕР-440) в неопроміненому, опроміненому й опроміненому під навантаженням стані. Показано, що механічне навантаження, яке імітує тиск теплоносія, прискорює радіаційне окрихчення, при цьому його внесок можна порівняти з внеском нейтронного опромінення.

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