STRUCTURAL FEATURES AND OPERATIONAL CHARACTERISTICS OF STEEL T91

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The microstructure and radiation resistance of T91 martensitic steel were studied after thermomechanical treatment. The physical and technological foundations of the process of creating of a nanostructured state in T91 reactor steel have been developed. This structure was received by severe plastic deformation of T91 steel by the multiple "upsetting-extrusion" method (developed at the NSC KIPT) in two temperature ranges of deformation: in the region of austenite existing and with a successive decrease in the deformation temperature and an increase in cycles of "upsetting-extrusion" in the field of ferrite existence. For the further heat treatment the particular temperature range and deformation modes were chosen to obtain optimal structure. Also, the optimum temperature of tempering to receive the uniform structure was established. It was found that the average grain size of T91 steel decreases from 20 μm in the initial state to ~ 140 nm after 5 cycles of "upsetting-extrusion" in the ferrite interval and to ~ 100 nm after 3 cycles of deformation in the austenitic region. It was determined that with an increase in the number of cycles and a decrease in the deformation temperature, a rise in the degree of uniformity of grain size distribution occurs. In this case, the microhardness increases from 2090 MPa to 2850 MPa after 5 cycles of "upsetting-extrusion" in the ferritic interval. In the austenitic region, the microhardness values increase from 3400 to 3876 MPa. The swelling of T91 steel in two structural states, martensitic and ferritic, was determined. Thus, steel swelling at a high dose of irradiation with argon ions with an energy of 1.4 MeV (120 displacements per atom, irradiation temperature 460 ° C) is $\Delta V / V = 0.26\%$ in the initial state (martensitic structure) and 0.65% for samples with a ferritic structure.
KEYWORDS: martensitic steel, thermomechanical treatment, microhardness, nanostructure, severe plastic deformation, radiation resistance.

The creation of GenIV fission and fusion reactors poses new problems in materials science in terms of higher operating temperatures, increased neutron doses, as well as ensuring the safe and efficient operation of reactors. The problem of conventional structural materials, in particular austenitic steels, lies in the fact that they will not be able to be operated under more severe irradiation conditions due to their low radiation resistance. Ferritic and / or martensitic steels are considered as potential structural materials for their good resistance to irradiation, less swelling, better ductility, etc. [1, 2]. In particular, T91 martensitic steel is considered as a promising material for future new generation nuclear reactors [3]. However, the steels of these classes have a significant drawback - heat resistance, which limits the maximum working temperature to ~ 500 °C. The main way of solving this problem is thermomechanical treatment, which allows not only to qualitatively improve the mechanical characteristics, but also to increase the radiation resistance of the material.

The standard technological scheme for the heat treatment of T91 martensitic steel consists in normalization at temperatures of 1040–1080 °C followed by air cooling [4]. In this case, the microstructure of quenched martensite is formed, which leads to high strength but low ductility of steel. To increase ductility, steel is tempered at temperatures of 730–780 °C. As a result, there is a formation of a coarse-grained microstructure with inhomogeneity in grain size. Although ductility increases considerably, the strength characteristics are reduced. A significant improvement in mechanical properties can be expected when an ultrafine-grained or nanostructured state is produced in steel, which usually contributes to an increase in the radiation tolerance of the material [5, 6]. The creation of such a microstructure is possible using thermomechanical treatment using severe plastic deformation (SPD), carried out by various methods [7 - 9].

A distinctive feature of T91 steel in comparison with other steels of the ferritic-martensitic class is the high sensitivity of its structure to the composition and type of heat treatment, and, as a result, a large data scattering of the swelling values for the same exposure conditions [10-12]. Optimization of the factors of the structural-phase state of T91 steel and study of its swelling in various structural states is needed.

The excellent swelling resistance of ferritic-martensitic alloys compared to austenitic stainless steels is well proven. The mechanisms of this phenomenon (higher self-diffusion rate, low preference factor for high dislocation density, low rate of helium generation, strong defect capture, microstructural lamellas and subgrains, boundary effects, etc.) are well described in the literature [13]. However, a comparison of the swelling of 9-12 wt. % Cr ferritic-martensitic steels shows a significant data scattering [14]. The reason for this scatter may be a difference in the structure and composition of the studied steels.

The aim of the study was to optimize the factors of the structural-phase state (grain size, parameters of secondary phase precipitation, degree of decomposition of the solid solution) to increase operational characteristics and to compare the level of swelling for two different structure states.

To achieve the goal of the study, the following tasks were set:
- to develop a technology for creating an ultrafine-grained structure, which can be used on an industrial scale;
- determine the optimal parameters of deformation and heat treatment to obtain a uniform and stable microstructure;
- to examine the effect of thermomechanical treatment and the type of obtained structures on the radiation tolerance of T91 steel.

MATERIAL AND METHODS

T91 steel manufactured by INDUSTELL, Belgium (melting: 504/3, heat: 82566-4) was supplied in the form of a 40 mm thick plate obtained by hot rolling followed by heat treatment. The heat treatment includes normalization at 1040 °C for 30 minutes, followed by air cooling and then tempering at 730 °C for 60 minutes with air cooling to room temperature (the so-called N&T state). The chemical composition of T91 steel is shown in Table 1.

Table 1.
The chemical composition of T91, (wt.%)

Fe	Cr	Mo	Mn	Si	V	Ni	Nb	Cu
bulk	8.76	0.862	0.597	0.317	0.186	0.099	0.073	0.054
Al	C	N	P	S	Sn	B	Co	As
0.021	0.088	0.003	0.019	0.0006	0.005	0.0001	0.019	0.007

In the present study the multi-cycle "upsetting-extrusion" method was chosen to carry out the SPD. The method was developed in NSC KIPT and it has proved itself both in laboratory research and in the industrial production of a number of materials with high characteristics [15, 16]. This method consists in multiple reiterations of operations of upsetting and extrusion on a hydraulic press ДБ 2432 with a force of 160 tf. The method is described in more detail in [17].

Deformation was carried out in two temperature regions: austenitic-ferritic at a temperature 870 °C and ferritic at temperatures 750 - 575 °C. Samples with a 20 mm diameter and a 60 mm height were subjected to upsetting to a diameter 30 mm and then again extruded to a diameter of 20 mm.

"Upsetting-extrusion" cycles in the ferritic region were carried out with a gradual decrease in the deformation temperature from 750 °C on the first cycle to 575 °C on the last, fifth cycle. In general, the temperature regime of the "upsetting-extrusion" cycles was as follows: 750-700-650-635-575 ° C.

To determine the stability of microstructures formed as a result of SPD, heat treatments were carried out in the temperature range 550–750 ° C for 1–50 hours.

Understanding the evolution of the microstructure during irradiation and its effect on the decline of mechanical properties is of a paramount importance. Neutron irradiation has several disadvantages, including high cost, residual activity, and prolonged irradiation of samples. Furthermore, many nuclear facilities, such as FFTF, RAPSODIE, DFR, PFR, Superfenix, EBR-II, BR-10, BN-350, etc. - are shut down.

The applying of heavy ion implantation to simulate neutron irradiation in a reactor is common practice by reason of the high rate of achievement of relatively high levels of damage and the absence of induced radioactivity.

Samples of steel T91 in the initial state (martensitic structure) and after severe plastic deformation (ferritic structure) were irradiated with argon ions with energy 1.4 MeV to a dose 120 dpa at a temperature 460 °C. This irradiation temperature was chosen due to the fact that the peak of temperature swelling for steel T91 falls on ~ 460 °C. All irradiations were carried out using the acceleration-measuring system "ESU-2" containing the Van de Graaff accelerator. The depth distribution of Ar atoms concentration and damage was calculated by SRIM 2008 [18] (Fig. 1). Damage calculations are based on the Kinchin-Pease model, with a displacement energy of 40 eV for Fe and Cr, as recommended in ASTM E521-96 (2009) [19].
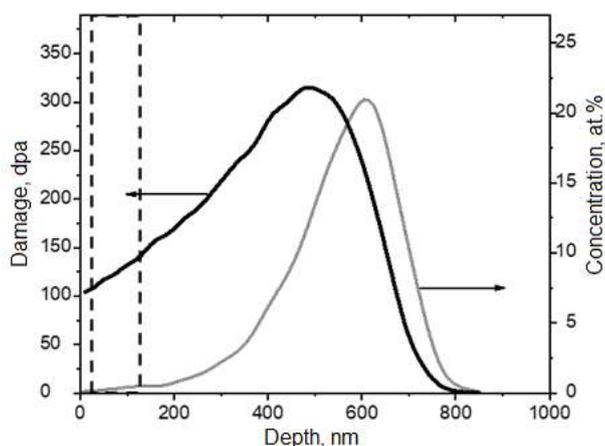

Figure 1. Calculated profiles of damages and concentrations of 1.4 MeV Ar ions implanted in T91 steel to a dose of 5×10^{17} cm^{-2}. The area of study is marked with a dash.

The steel structure was studied by metallography and electron microscopy using electron microscopes JEM-100CX and JEM-2100. Microhardness was measured on a semi-automatic microindenter LM 700AT at a load 200 g.

RESULTS AND DISSCUSSION

The microstructure of T91 steel in initial state is shown in Fig. 2. The structural state of tempered martensite is characterized by the presence of prior austenite grain boundaries. These boundaries are decorated with $M_{23}C_6$ carbide precipitates. The prior austenite grain size is about 20 μm, the size (thickness) of $M_{23}C_6$ precipitates varies from about 100 to 200 nm. Inside the prior austenite grains, martensitic lamellas with a transverse size of 0.25-0.5 μm are observed.
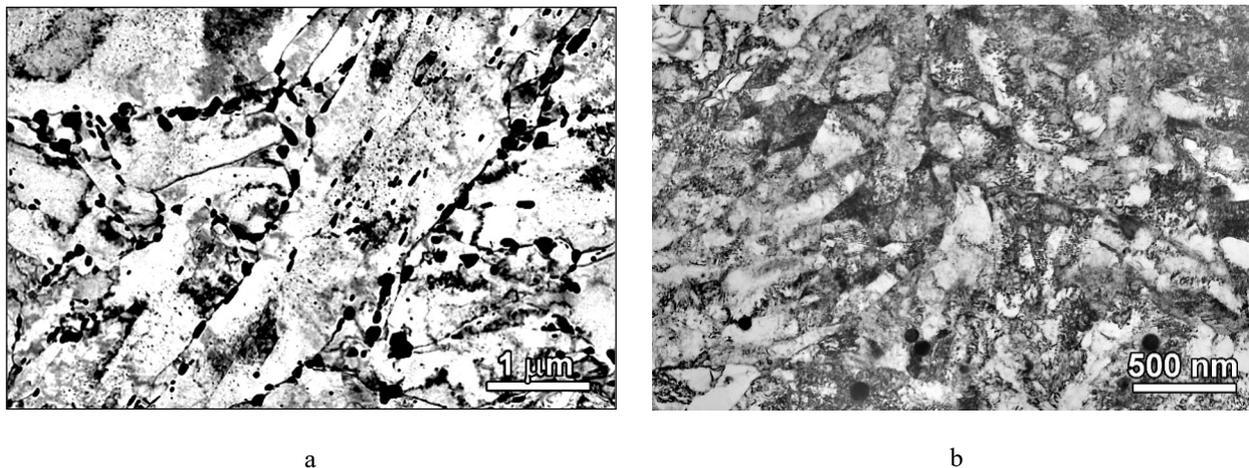

a   b

Figure 2. The microstructure of steel T91: a – in initial state, b – after 3 cycles of "upsetting-extrusion" at 870 ºC

The finely dispersed microstructure of steel after 3 cycles of "upsetting-extrusion" at a temperature 870 °C belongs to the martensitic-ferritic type (Fig. 2b). From a comparison of Fig. 2a and b, it can be seen that the SPD led to a significant refinement of the martensitic structure.

The microstructure that formed after treatment in the ferritic range with a gradual decrease in temperature and the ferrite grains size distribution are shown in Fig. 3. The average grain size was 145 nm.
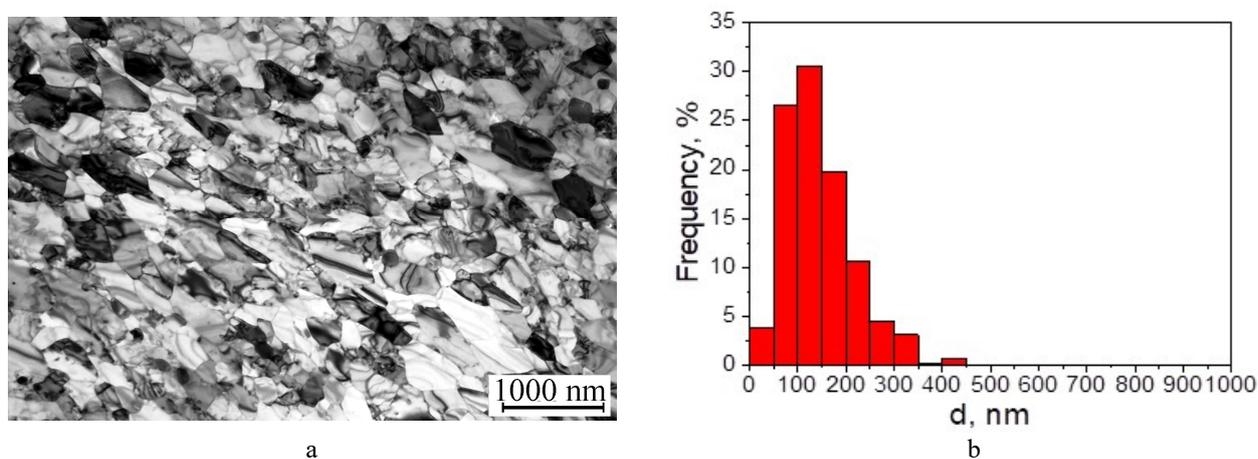

a   b

Figure 3. The microstructure of steel after 5 cycles of "upsetting-extrusion" in the ferritic range (a) and grain size distribution (b)

The results of the microhardness measuring of the samples after heat treatment in the temperature range of 550-750 °C for 1-50 hours showed that high mechanical characteristics are maintained during exposure at a temperature of 550 °C. It was found, that, the temperature of 550 °C is the optimal regime for tempering, which allow to stabilize the structure, since at higher temperatures there is recrystallization and, as a result, a decrease in hardness. Therefore, for further studies, a sample that underwent heat treatment for 25 hours at a temperature of 550 °C was selected. The average grain size for sample with a ferritic structure was 210 nm, and the microhardness was at the level of 2948 MPa. The microstructure of the sample is shown in Fig. 4.

To compare the degree of swelling, austenitic steel SS316 was chosen which is now used as a structural material of the current generation of reactors. Herewith, irradiation of SS316 steel was carried out under the following conditions: dose - 80 dpa, temperature - 615 °C. The microstructure of irradiated steels is shown in Fig. 5. For three types of samples, namely, T91 steel with martensitic and ferritic structures and steel SS316 with an austenitic structure, the formation of a void structure is observed.
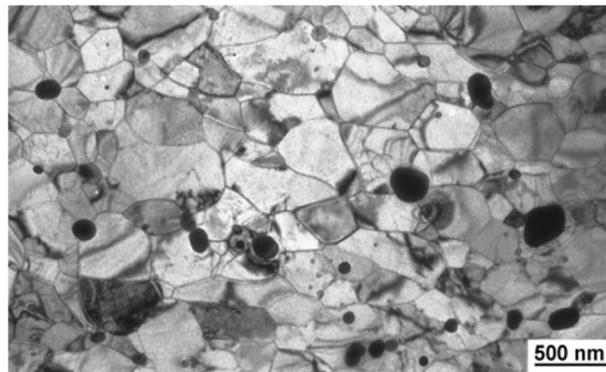
Figure 4. The microstructure of the sample of steel T91 after heat treatment at 550°C for 25 h after the SPD in a ferritic range
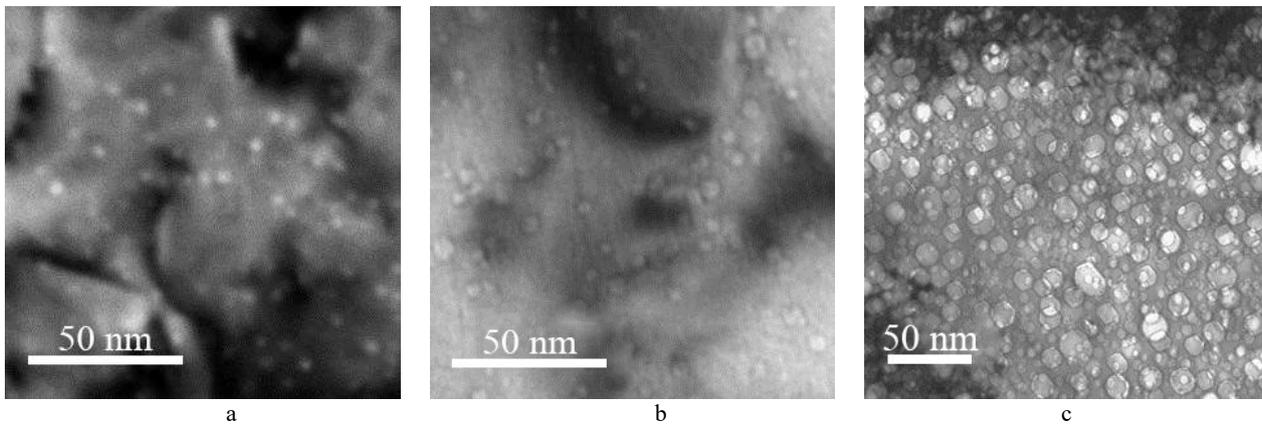

a   b   c

Figure 5. The microstructure of steel T91 for samples with martensitic (a) and ferritic (b) structures, irradiated to a dose 120 dpa (E = 1,4 MeV Ar$^+$) at 460°C and austenitic steel SS316, irradiated to a dose 80 dpa at 615°C (c)

Using experimentally determined parameters of a void structure, the swelling value S was calculated by the formula (1):

$$S = \frac{\frac{\pi}{6 \cdot A \cdot h} \sum_{i=1}^{N} d_i^3}{1 - \frac{\pi}{6 \cdot A \cdot h} \sum_{i=1}^{N} d_i^3} \cdot 100\%, \qquad (1)$$

where d_i-diameter of i-th void; N- the number of voids on the image; A- area of the image with which the calculations were carried out; h- sample foil thickness.

The parameters of void structure and the magnitude of the swelling are shown in Table 2.

Table 2.

Diameter d, number density ρ of voids and swelling values S

Sample	d, nm	ρ, 1/m^3	S, %
T91 Martensitic	2,9	1×10^{23}	0,26
T91 Ferritic	6,6	5,8×10^{22}	0,65
SS316 Austenite	18	9,3×10^{21}	15,6

A comparison of the steel with a martensitic and ferritic structure (Fig. 5 a, b) showed that the level of swelling of the sample in the martensitic state is 2.5 times lower than in the ferritic state. It should be noted that the swelling of the ferritic structure (BCC) is 24 times less than the austenitic structure (FCC). Such a large difference in the levels of swelling is associated with the difference in the types of crystal lattices of these materials, and as a result, with different mechanisms of the development of radiation damage.

In [14], data on the swelling of ferritic-martensitic steels irradiated with neutrons up to a dose of 208 dpa in a temperature range from 400 to 443 °C were compared (Fig. 6). Let's note several key features. There is a general tendency to swelling at a rate of ∼ 0.01% / dpa, although the data are very scattered. In general, there is a large difference in swelling between alloys (HT9 and T91), though T91 tends to swell more for a given level of damage. Finally, the swelling of HT9 steel of the same heat of 9607R2, but of different heat treatment, differs by an order of

magnitude (see Fig. 6). Our data - indicated by a triangle and a star in a dashed rectangle - show that the scatter may be due to the different microstructure states that form before irradiation (ferrite (★) or martensite (▲)).

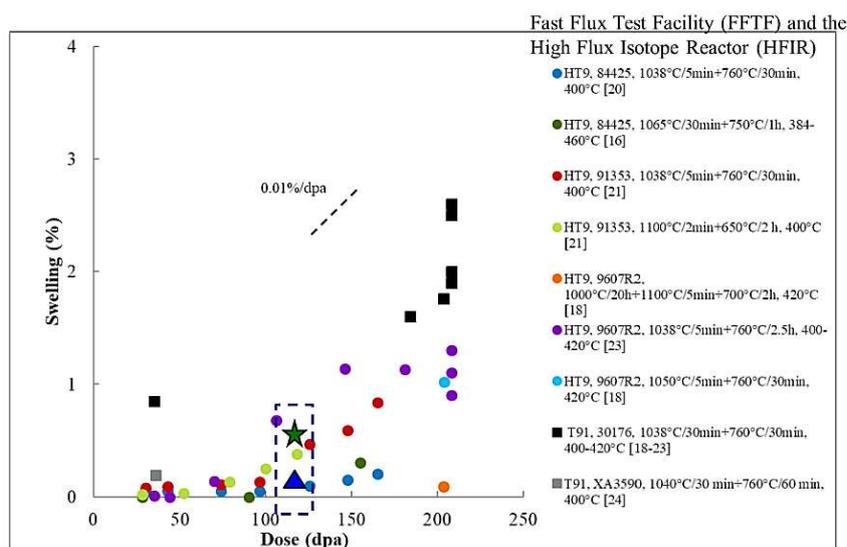Further studies are needed to study the level of swelling at even higher radiation doses.

Figure 6. Generalized data for steels HT9 and T91 irradiated in a reactor to a dose 208 dpa at temperatures from 400 to 443ºC [14]

CONCLUSIONS

The effect of thermomechanical treatment on the type of obtained structures of T91 steel is investigated. It was shown that the receiving of a nanostructure can be achieved by severe plastic deformation method. The factors of the structural-phase state of T91 martensitic steel have been optimized during thermomechanical treatment by the method of multiple "upsetting-extrusion". The optimal parameters of deformation and temperature regimes for obtaining a uniform ultrafine-grained structure are determined.

Severe plastic deformation combined with heat treatment allows to reduce the average grain size. After 5 cycles of multiple "upsetting-extrusion" in the ferritic range, the minimum average grain size of ~ 140 nm was achieved. The average grain size of the initial steel (martensitic structure) is 20 μm.

A study of the development of void structure after irradiation of steel T91 in various structural states was carried out. It was shown that after irradiation with Ar ions with an energy 1.4 MeV to a dose 120 dpa at T = 460 °C, the swelling in the ultrafine-grained ferritic structure is 0.65% compared with 0.26% for the initial martensitic structure, which confirms the influence of structural-phase state of steel T91 on its swelling.

The swelling of steel with an austenitic structure is ~ 15.6%, which is 24 times more than the level of swelling of steel with a ferritic structure.

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REFERENCES

[1] R. Klueh and D. Harries, *High-Chromium Ferritic and Martensitic Steels for Nuclear Applications,* ed. (ASTM International, West Conshohocken, 2001), pp. 221, https://doi.org/10.1520/MONO3-EB.
[2] R. Klueh, Philosophical Magazine. **98**(2012), 2618-2636 (2008), https://doi.org/10.1080/14786435.2018.1497307.
[3] S. Baindur, Bulletin of the Canadian Nuclear Society, **29**(1), 32-38 (2008).
[4] https://www.astm.org/Standards/A213.htm.
[5] M. Song, C. Sun, Z.Fan, Y. Chen, R.Zhu, K.Y. Yu, K.T. Hartwig, H.Wang, and X.Zhang, Acta Materialia, **112**, 361-377 (2016), https://doi.org/10.1016/j.actamat.2016.04.031.
[6] R.A. Andrievskii, The Physics of Metals and Metallography. **110**(3), 229–240 (2010), https://doi.org/10.1134/S0031918X10090061.
[7] M.V. Karavaeva, M.A. Nikitina, A.V. Ganeev, R.K. Islamgaliev, IOP Conf. Series: Materials Science and Engineering. **179**, 012037 (2017), https://doi.org/10.1088/1757-899X/179/1/012037.

[8] D.C. Foley, K.T. Hartwig, S.A. Maloy, P. Hosemann, X. Zhang, Journal of Nuclear Materials. **389**, 221-224 (2009), http://doi.org/10.1016/j.jnucmat.2009.02.005.

[9] Z.Q. Fan, T. Hao, S.X. Zhao, G.N. Luo, C.S. Liu, Q.F. Fang, Journal of Nuclear Materials. **434**, 417–421 (2013), https://doi.org/10.1016/j.jnucmat.2012.12.009.

[10] D.S. Gelles, Journal of Nuclear Materials. **233-237**, 293-298 (1996), https://doi.org/10.1016/S0022-3115(96)00222-X.

[11] J.J. Kai, R.L. Klueh, Journal of Nuclear Materials. **230**, 116-123 (1996), https://dx.doi.org/10.1016/0022-3115(96)00165-1.

[12] J. Van Den Bosch, O. Anderoglu, R. Dickerson, M. Hartl, et al., Journal of Nuclear Materials. **440**, 91-97 (2013), https://dx.doi.org/10.1016/j.jnucmat.2013.04.025.

[13] M.B. Toloczko, F.A. Garner, C.R. Eiholzer, Journal of Nuclear Materials. **215**, 604-607 (1994), https://doi.org/10.1016/0022-3115(94)90131-7.

[14] E. Getto, K. Sun, A.M. Monterrosa, et al., Journal of Nuclear Materials. **480**, 159-176 (2016), https://doi.org/10.1016/j.jnucmat.2016.08.015.

[15] O.V. Chorniy, Ya.D. Starodybov, O.I. Volchok, G.E. Storozhilov, Patent Ukraine No. 42487 A (15 January 2001). (in Ukrainean)

[16] I.I. Papirov, G.F. Tikhinskiy, The Physics of Metals and Metallography. **29**(5), 1057-1060 (1970). (in Russian)

[17] V.M. Azhazha, O.V. Chorniy, G.E. Storozhilov, N.F. Andrievskaya, T.U. Rydicheva, Problems of Atomic Science and Technology, Series: "Vacuum, Pure Materials, Superconductors", **6**(14), 136 -139 (2004). (in Russian)

[18] J.F. Ziegler, Version – SRIM-2008.04, www.srim.org.

[19] ASTM E521-96, *Standard Practice for Neutron Radiation Damage Simulation by Charged-Particle Irradiation*, https://www.astm.org/DATABASE.CART/HISTORICAL/E521-96.htm.

СТРУКТУРНІ ОСОБЛИВОСТІ ТА ЕКСПЛУАТАЦІЙНІ ХАРАКТЕРИСТИКИ СТАЛІ Т91

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Досліджено мікроструктуру і радіаційну стійкість мартенситної сталі Т91 після термомеханічної обробки. Розроблено фізико-технологічні основи процесу створення наноструктурного стану в реакторній сталі Т91. Ця структура була отримана в результаті інтенсивної пластичної деформації сталі Т91 методом багаторазового «осадження-вичавлювання», розробленим в ННЦ ХФТІ, в двох температурних діапазонах: в області існування аустеніту і з послідовним зниженням температури деформації і збільшенням циклів «осадження-вичавлювання» в області існування фериту. Для подальшої термообробки були обрані особливі температурний діапазон і режими деформації для отримання оптимальної структури. Також була знайдена оптимальна температура відгартовування для отримання однорідної структури. Було виявлено, що середній розмір зерна сталі Т91 зменшується з 20 мкм в вихідному стані до ~ 140 нм після 5 циклів «осадження-вичавлювання» в феритному інтервалі і до ~ 100 нм після 3 циклів деформації в аустенітній області. Було встановлено, що зі збільшенням кількості циклів і зниженням температури деформації відбувається підвищення ступеню однорідності розподілу зерен за розмірами. В цьому випадку мікротвердість збільшується з 2090 МПа до 2850 МПа після 5 циклів «осадження-вичавлювання» в феритному інтервалі. В аустенітній області значення мікротвердості збільшуються з 3400 до 3876 МПа. Визначено розпухання сталі Т91 в двох структурних станах - мартенситному та феритному. Так, розпухання сталі при високій дозі опромінення іонами аргону з енергією 1,4 МеВ (120 зсувів на атом, температура опромінення 460ºC) становить $\Delta V / V = 0.26\%$ в вихідному стані (мартенситна структура) і 0,65% для зразків з феритною структурою.

КЛЮЧОВІ СЛОВА: мартенситна сталь, термомеханічна обробка, мікротвердість, наноструктура, інтенсивна пластична деформація, радіаційна стійкість.

СТРУКТУРНЫЕ ОСОБЕННОСТИ И ЭКСПЛУАТАЦИОННЫЕ ХАРАКТЕРИСТИКИ СТАЛИ Т91

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Исследована микроструктура и радиационная стойкость мартенситной стали Т91 после термомеханической обработки. Разработаны физико-технологические основы процесса создания наноструктурного состояния в реакторной стали Т91. Эта структура была получена в результате интенсивной пластической деформации стали Т91 методом многократной «осадки-выдавливания», разработанным в ННЦ ХФТИ, в двух температурных диапазонах: в области существования аустенита и с последовательным снижением температуры деформации и увеличением циклов «осадки-выдавливания» в области существования феррита. Для дальнейшей термообработки были выбраны особые температурный диапазон и режимы деформации для получения оптимальной структуры. Также была найдена оптимальная температура отпуска для получения однородной структуры. Было обнаружено, что средний размер зерна стали Т91 уменьшается с 20 мкм в исходном состоянии до ~ 140 нм после 5 циклов «осадки-выдавливания» в ферритном интервале и до ~ 100 нм после 3 циклов деформации в аустенитной области. Было установлено, что с увеличением числа циклов и уменьшением температуры деформации происходит повышение степени однородности распределения зерен по размерам. В этом случае микротвердость увеличивается с 2090 МПа до 2850 МПа после 5 циклов «осадки-выдавливания» в ферритном интервале. В аустенитной области значения микротвердости увеличиваются с 3400 до 3876 МПа. Определено распухание стали Т91 в двух структурных состояниях - мартенситном и ферритном. Так, распухание стали при высокой дозе облучения ионами аргона с энергией 1,4 МэВ (120 смещений на атом, температура облучения 460ºC) составляет $\Delta V/V = 0.26\%$ в исходном состоянии (мартенситная структура) и 0,65% для образцов с ферритной структурой.

КЛЮЧЕВЫЕ СЛОВА: мартенситная сталь, термомеханическая обработка, микротвердость, наноструктура, интенсивная пластическая деформация, радиационная стойкость.