

SECTION 4  
**PHYSICS OF RADIOTECHNOLOGY  
AND ION-PLASMA TECHNOLOGIES**

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**EFFECT OF CHROMIUM COATINGS ON THE MECHANICAL  
PROPERTIES OF Zr1Nb FUEL CLADDINGS IN LONGITUDINAL  
AND TRANSVERSE DIRECTIONS**

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This study was aimed at investigating the chromium coating effect on the mechanical properties of Zr1Nb fuel claddings in the case of a tensile fracture in longitudinal and transverse directions at temperatures of 20 and 350 °C. Tests were carried out using the samples of a similar shape, namely, with an equal test portion length that is important for comparison of results. The obtained results have shown that at a test temperature of 20 °C the mechanical properties of initial samples are higher in the longitudinal direction than in the transverse direction. The ductility is slightly higher at  $T = 350$  °C in the longitudinal direction, and  $\sigma_{0.2}$  and  $\sigma_b$  are practically equal. The preliminary deformation of ring samples increases their ductility from 23 to 34% independently on the test temperature. Deposition of the chromium coatings on the samples leads to the slight increase of mechanical properties in the transverse direction and decrease of  $\sigma_{0.2}$  with unchanged  $\sigma_b$  and  $\delta\%$  in the longitudinal direction at the room test temperature, and at 350 °C it practically do not change the properties of coated samples in both the directions except the ring sample ductility decrease.

**INTRODUCTION**

For Ukraine an urgent problem is to increase the operation life of existent WWER reactors and to provide the NPP safe operation. First of all it is to prevent possible hard loss-coolant accident effects. Fuel claddings from zirconium-based alloys, are a main cause of an explosive hydrogen concentration in such conditions due to the intense zirconium oxidation with a sharp temperature increase in the reactor [1]. The deposition of protective coatings having a high oxidation resistance onto the fuel elements is an expedient solution of this problem [2]. The results of experiments have shown that from a variety of protective coatings just chromium coatings possess desired properties (resistance to oxidation, corrosion and irradiation) [3–5]. Therefore, of special importance is to investigate the effect of chromium coatings on the mechanical properties of zirconium tubes at different test temperatures and to find out the causes of structural material degradation during the long reactor operation.

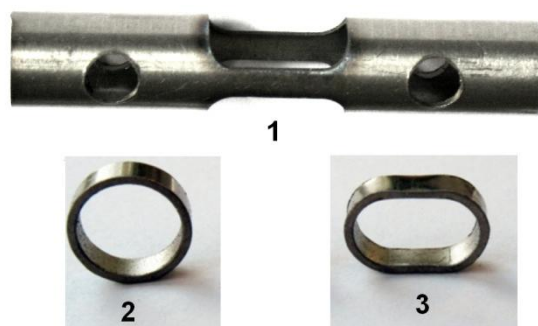
Mechanical properties of fuel claddings made from zirconium-based alloys were studied in many research works all over the world [6–12]. Over a long period of time the efforts were undertaken to improve the main peculiarities of applied methods, state standards (GOST), specifications and other special instructions, e.g. OI000.325-91 [6]. However, there is no authors' common approach to the zirconium tube sample preparation and to the test procedures.

The purpose of this study is to investigate the mechanical properties of Ukraine-produced Zr1Nb fuel claddings in the longitudinal and transverse directions at 20 and 350 °C (storage and operation temperatures) in the initial state and after deposition of a nanostructured

chromium coating. Investigations were carried out using the NSC KIPT experience gained when studying the strength properties of fuel claddings made from Zr-based alloys.

**EXPERIMENTAL PROCEDURES**

Investigations of mechanical properties have been carried out on samples cut, by the electric-spark technique, from the fuel cladding tube made of Ukraine-produced Zr1Nb alloy. The samples in the modified form [8] were tested in the longitudinal direction and the ring samples were tested in the transverse directions (Fig. 1).



*Fig. 1. Samples for mechanical tests:  
1 – modified sample; 2 – ring sample  
and 3 – ring sample deformed to the point “M”*

In the second case the ring samples were also used after the preliminary deformation to the point “M” (the tensile test diagram Fig. 2), which is characterized by the onset of elastic deformation in the elongated loop, i.e. transformed ring sample.

The preliminary pre-strain diagram is shown in Fig. 2. The purpose of such deforming was to exclude a

possible coating cracking at the beginning of the form changing.

For correct comparison of the test results it is necessary to use all the samples with equal dimensions

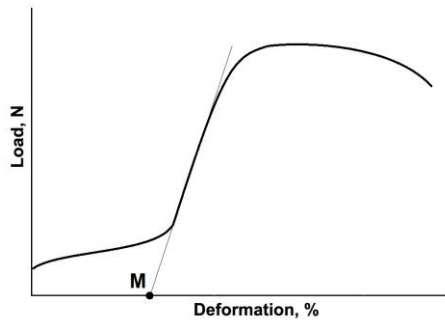


Fig. 2. Diagram of the preliminary deformation of the ring sample

The tests were carried out on the tensile machine Instron 5581 with a strain rate of 1 mm/min. The tensile stress diagrams in Fig. 3 show typical stress-strain curves.

Chromium coatings were formed using unfiltered cathodic arc evaporation (CAE) in a “Bulat” system, which is equipped with two Cr (99.9%) cathodes of 60 mm diameter [14]. The deposition chromium coating was performed on planetary rotating holder at a substrate bias voltage of -50 V and a substrate

and similar shape of their test portion, namely, with the test portion length  $L_0 = 8.0$  mm and width  $b_0 = 2.8$  mm (according to RMI-1-2001) [13].

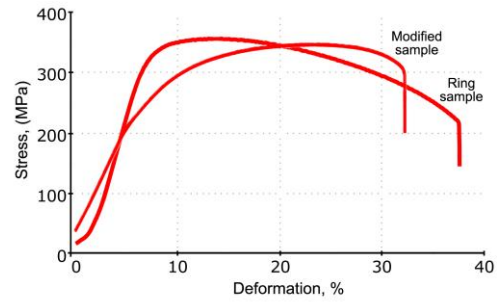


Fig. 3. Tensile stress-strain diagrams for Zr1Nb samples: modified sample and ring sample

temperature of  $\sim 400$  °C. The coating thickness was  $\sim 5$  and  $8$   $\mu\text{m}$ .

## EXPERIMENTAL RESULTS AND DISCUSSION

The mechanical properties of initial samples and preliminary deformed samples with coating and without coating are represented for two test temperatures in Fig. 4.

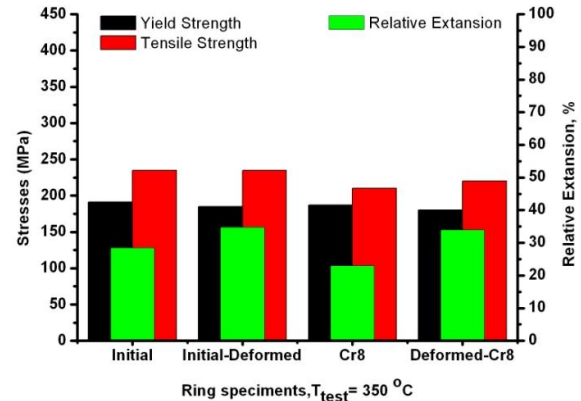
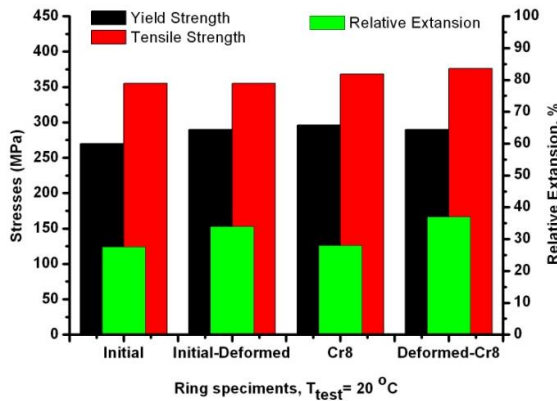


Fig. 4. Test results of the ring samples: a –  $T = 20$  °C; b –  $T = 350$  °C

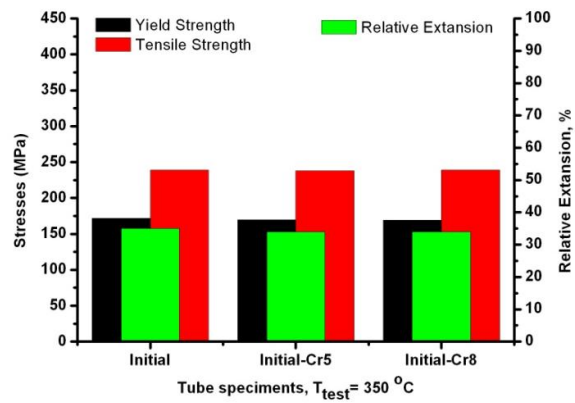
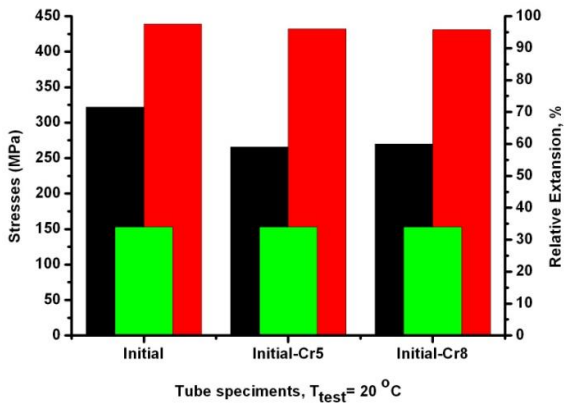


Fig. 5. Test results of the modified samples: a –  $T = 20$  °C; b –  $T = 350$  °C

At a test temperature  $T = 20$  °C the tensile yield stress and the ultimate tensile strength of initial ring

samples are  $\sigma_{0.2} = 270$  MPa and  $\sigma_b = 355$  MPa, respectively, and the specific elongation  $\delta = 28\%$ .

Owing to the chromium coating  $\sigma_{0.2}$  is increased by 8.5% and  $\sigma_b$  by 3.5% and  $\delta$  is not changed. The preliminary deformation increases the initial sample ductility by 23% and that of the coated sample by 1/3. At a test temperature  $T = 350$  °C the values of  $\sigma_{0.2}$  and  $\sigma_b$  of the initial samples are lower:  $\sigma_{0.2} = 190$  MPa and  $\sigma_b = 235$  MPa, and the ductility is not changed.

The coating deposition cause decrease  $\sigma_b$  by 10% and the specific elongation by 19%. The preliminary deformation leads to the increase of  $\delta$  by 23% for the initial sample and by 48% for the coated sample.

The results of modified sample tests in the longitudinal direction are given in Fig. 5. At a test temperature of 20 °C the tensile yield stress of initial samples is  $\sigma_{0.2} = 320$  MPa and after coating deposition it decreases by 15%, the ultimate tensile strength  $\sigma_b = 439$  MPa is almost equal for all the samples, as well as, the specific elongation  $\delta = 34\%$ . At a test temperature  $T = 350$  °C the strength and ductility of the coated samples do not change:  $\sigma_{0.2} = 172$  MPa,  $\sigma_b = 239$  MPa and  $\delta = 34\%$ .

It may be noted that all the values of  $\sigma_{0.2}$ ,  $\sigma_b$ , and  $\delta\%$  in the longitudinal direction for the initial samples at 20 °C are slightly higher than for the ring samples, and at 350 °C the increase is observed only for  $\delta\%$ .

At the room temperature the coating deposition keeps the advantage of the tube mechanical characteristics in the longitudinal direction, without changing  $\sigma_b$  and  $\delta\%$  but with decreasing  $\sigma_{0.2}$ . The tensile yield stress decrease by 15% at the room test temperature can be caused by the presence of insignificant defects on the tube wall face after electric-arc cutting its part to make modified samples. It is well-known [15] that even a slight change in the material surface undulation decreases its strength properties. In the stressed chromium coating at a test temperature of 20 °C in the initial strain stage (to the yield stress) the crack nucleation can occur on the substrate defects that decreases the yield stress but does not change the ultimate strength. It would be more correct to perform the tensile tests using one-piece uncut tubes with a coating as the authors of [12] have done.

The ring and deformed samples with coatings show at 20 °C an insignificant increase of all the values of mechanical properties in the transverse direction.

The chromium coating deposition did not lead to appreciable changes in  $\sigma_{0.2}$ ,  $\sigma_b$ , and  $\delta\%$  values of the modified samples but decreased significantly  $\delta\%$  in the initial ring samples the transverse direction at a test temperature of 350 °C. As is seen from Fig. 4 the preliminary deformation leads to the significant increase in the tensile ductility of both, coated and uncoated, ring samples compared to the initial one. When the results obtained are compared it is necessary to take into account the difference in the initial state of the fuel cladding material before and after the nanostructured chromium coating deposition. In the first case the strain and degradation processes over a long operation period go by the classic dislocation mechanisms, and in the second case the fuel cladding material, after the nanostructured coating deposition, presents a multilevel system in which the plastic flow and plastic collapse is developed at nano-, micro-, meso-, and macrolevels

[16]. In a natural crystal the surface layer acts as a “pump” injecting dislocations into the loaded material and thus accelerating its damage. In the nanostructured surface layer a process occurs which slows down the deformation defect accumulation in the material and thus increases its durability [17].

Experiments with the surface layer nanostructuring resulted in finding a new effect, namely, “checkerboard” stress distribution on the “coating-crystalline substrate” interface in the loaded material [18]. On basis of this effect a new method of structural material hardening has been developed which provides improvement of all the mechanical characteristics. On the coating-substrate interface a “checkerboard” with a minimal check size should be formed. Thus it is possible to provide a quasi-uniform stress distribution and to prevent the occurrence of stress macroconcentrators which nucleate a neck, crack and material destruction [19].

A joint consideration of the test results obtained for the ring and deformed samples with chromium coatings has shown that the application of the preliminary ring sample deformation before the coating deposition leads to the increase in the ductility (specific elongation  $\delta\%$ ) from 23 to 34%. This can evidence on the fact that due to the deformation of the ring sample its coating is cracking in the part of initial deformation when the ring shape transforms into an “elongated loop” (see Fig. 2 to the point “M”), and the application of preliminary deformation can exclude such probability. This assumption is confirmed by the presence of high-amplitude acoustic signals in the elastic strain region of Zr1Nb ring samples with TiN and ZrN coatings that has been observed in [10].

The mechanical properties of chromium coated fuel claddings, depending on the operating time, i.e. their integrity in the range of load stresses below the tensile yield stress, can be studied using the internal friction procedure as the most sensitive method for consideration of physical processes at the atomic bond level and minimum stress-strain levels.

## CONCLUSIONS

For the first time the effect of protective chromium coatings on the mechanical properties of Ukraine-produced Zr1Nb fuel claddings in the longitudinal and transverse directions has been investigated. The tests were carried out on the samples having equal dimensions and similar shape of their test portion at temperatures of 20 and 350 °C.

The experimental results obtained for the initial samples have shown that at a test temperature of 20 °C the values of  $\sigma_{0.2}$ ,  $\sigma_b$ , and  $\delta\%$  in the longitudinal direction are significantly higher than in the transverse direction, and at 350 °C only the specific elongation  $\delta\%$  is higher.

The coating deposition at the room test temperature leads to the insignificant increase of all the mechanical properties in the transverse direction and does not change  $\sigma_b$  and  $\delta\%$  but decreases  $\sigma_{0.2}$  in the longitudinal direction.

At a test temperature  $T = 350$  °C the coatings practically has no effect on the mechanical properties

along the fuel cladding but decrease significantly the ductility  $\delta\%$  of initial ring samples.

The preliminary deformation of ring samples increases their ductility from 23 to 34% independently on the test temperature.

The present results indicate that the deposition of vacuum-arc chromium coatings of 5...8  $\mu\text{m}$  thick is an effective method of Zr1Nb fuel cladding hardening.

## REFERENCES

1. Z. Duan, H. Yang, Y. Satoh, K. Murakami, S. Kano, Z. Zhao, J. Shen, H. Abe. Current status of materials development of nuclear fuel cladding tubes for light water reactors // *Nucl. Eng. Des.* 2017, v. 316, p. 131-150.
2. C. Tang, M. Stueber, H.J. Seifert, M. Steinbrueck. Protective coatings of zirconium-based alloys as accident-tolerant fuel (ATF) claddings // *Corrosion Rev.* 2017, v. 35 p. 141-165.
3. K. Terrani. Accident tolerant fuel cladding development: promise, status, and challenges // *Journal of Nuclear Materials.* 2018, v. 501, p. 13-30.
4. J. Bischoff, C. Delafoy, C. Vauglin, P. Barberis, C. Roubeyrie, D. Perche, D. Duthoo, F. Schuster, J.-C. Brachet, E.W. Schweitzer, K. Nimishakavi. AREVA NP's enhanced accident-tolerant fuel developments: focus on Cr-coated M5 cladding // *Nucl. Eng. Technol.* 2018, v. 50 (2), p. 223-228.
5. A.S. Kuprin, V.A. Belous, V.N. Voyevodin, R.L. Vasilenko, V.D. Ovcharenko, G.D. Tolstolutskaia, I.E. Kopanets, I.V. Kolodiy. Irradiation resistance of vacuum arc chromium coatings for zirconium alloy fuel claddings // *Journal of Nuclear Materials.* 2018, v. 510, p. 163-167.
6. Special Instruction OI000.325-91. *Testing technique for determining mechanical property characteristics of ring samples from tubes by Specifications 95405-89 unde restraining.*
7. O.Ju. Makarov, V.I. Prokhorov, A.V. Goryatchew, et al. Improved measurement techniques for Zr1Nb alloy claddings mechanical properties while simple ring samples testing // *Abstracts for VI Russian Conference for Reactor Material Science, Dimitrovgrad, September 11–15, 2000.*
8. L.S. Ozhigov, A.S. Mitrofanov, V.I. Savchenko, P.N. V'yugov, Y.A. Krainyuk. Determining the ductility of small-diameter metal pipe in the tangential direction // *Zavodskaya laboratoriya. Diagnostika materialov.* 2014, N 3, v. 20, p. 60-62 (in Russian).
9. V.M. Azhazha, L.S. Ozhigov, S.D. Lavrinenko, P.N. V'yugov, V.S. Vakhrushina, V.I. Savchenko, N.N. Pilipenko, N.P. V'yugov, A.G. Rudenko, V.N. Pelykh, I.G. Tantsyura. Investigation on the mechanical properties of fuel element tubes from Ukraine-produced Zr1Nb alloy in the longitudinal and transverse directions at 20 to 700 °C temperatures // *Proceeds of XIX International Conference on Physics of Radiation Phenomena and Radiation Materials Science.* Ukraine, Alushta, 2010, p. 190-191.
10. P.I. Stoev, V.A. Belous, V.N. Voyevodin, A.S. Kuprin, S.A. Leonov, V.D. Ovcharenko, M.A. Tikhonovsky, V.M. Horoshih. Mechanical properties and acoustic parameters of tubes from zirconium Zr1Nb alloy with a protective coatings // *Problems of Atomic Science and Technology. Series "Physics of Radiation Effect and Radiation Materials Science"*. 2015, N 5(99), p. 87-97.
11. L.S. Ozhigov, V.I. Savchenko, A.S. Mitrofanov, Y.A. Krainyuk, S.V. Shramchenko, P.N. V'yugov, A.P. Redkina. Peculiarities of the investigation into the mechanical properties of fuel claddings from Zr1Nb alloy in the longitudinal and transverse directions // *East European Journal of Physics.* 2016, v. 3, N 3, p. 88-91.
12. V.A. Belous, P.N. V'yugov, A.S. Kuprin, S.A. Leonov, G.I. Nosov, V.D. Ovcharenko, L.S. Ozhigov, A.G. Rudenko, V.I. Savchenko, G.N. Tolmachova, V.M. Khoroshikh. Mechanical characteristics of Zr1Nb alloy tube after deposition of ion-plasma coatings // *Problems of Atomic Science and Technology. Series "Physics of Radiation Effect and Radiation Materials Science"*. 2013, N 2(84), p. 140-143.
13. *Methods of determining the mechanical properties of tubes from zirconium alloy under tensile testing in the transverse direction.* RMI24-1-2001.
14. A.S. Kuprin, V.A. Belous, V.V. Bryk, R.L. Vasilenko, V.N. Voyevodin, V.D. Ovcharenko, G.N. Tolmachova, I.V. Kolodiy, V.M. Lunyov, I.O. Klimenko. Vacuum-arc chromium coatings for Zr-1Nb alloy protection against high-temperature oxidation in air // *Problems of Atomic Science and Technology. Series "Physics of Radiation Effect and Radiation Materials Science"*. 2015, N 2(96), p. 111-118.
15. S.S. D'yachenko, I.V. Ponomarenko, S.N. Dub. Role of steel object surface condition on the behavior during deformation // *Met. Sci. Heat Treat.* 2015, v. 57, N 5, p. 245-253.
16. L.S. Ozhigov, V.A. Belous, V.I. Savchenko, G.I. Nosov, V.D. Ovcharenko, G.N. Tolmachova, A.S. Kuprin, V.S. Goltvyanitsa. Role of surface layer nanostructuring in improving mechanical and corrosion properties of reactor materials // *Problems of Atomic Science and Technology. Series "Physics of Radiation Effect and Radiation Materials Science"*. 2017, N 2(108), p. 168-172.
17. V.E. Panin, V.P. Sergeev, A.V. Panin, Y. Pochivalov. Surface layer nanostructuring and nanostructure coating deposition as an effective method of hardening advanced structural and instrumental methods // *The Physics of Metals and Metallography.* 2007, v. 104, N 6, p. 650-660.
18. V.E. Panin, V.E. Egorushkin. Deformable solid as a nonlinear hierarchically organized system // *Physical Mesomechanics.* 2011, v. 14, issue 5-6, p. 207-223.
19. V.E. Panin, V.P. Sergeev, A.V. Panin. *Nanostructuring of the surface layers of construction materials and nanostructured coating deposition.* Tomsk: Tomsk Polytechnic University, 2013, p. 254.

## **ВЛИЯНИЕ ХРОМОВОГО ПОКРЫТИЯ НА МЕХАНИЧЕСКИЕ СВОЙСТВА ОБОЛОЧЕК ТВЭЛОВ ИЗ СПЛАВА Zr1Nb В ПРОДОЛЬНОМ И ПОПЕРЕЧНОМ НАПРАВЛЕНИЯХ**

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Исследовано влияние хромового покрытия на механические свойства оболочек твэлов из сплава Zr1Nb при разрыве в продольном и поперечном направлениях при 20 и 350 °С. Испытания проводились на образцах подобной формы, а именно, с одинаковой протяженностью рабочей части, что существенно при сравнении результатов. Показано, что при температуре испытаний 20 °С механические свойства исходных образцов в продольном направлении выше, чем в поперечном. Пластичность незначительно выше при  $T = 350$  °С в продольном направлении, а  $\sigma_{0,2}$  и  $\sigma_b$  практически одинаковые. Осаждение хромового покрытия на образцы приводит к некоторому росту механических свойств в поперечном и снижению  $\sigma_{0,2}$  при неизменности  $\sigma_b$ ,  $\delta\%$  в продольном направлениях при комнатной температуре разрыва, а при 350 °С практически не изменяет свойства в обоих направлениях.

## **ВПЛИВ ХРОМОВОГО ПОКРИТТЯ НА МЕХАНІЧНІ ВЛАСТИВОСТІ ОБОЛОНОК ТВЕЛІВ ІЗ СПЛАВУ Zr1Nb У ПОДОВЖНЬОМУ ТА ПОПЕРЕЧНОМУ НАПРЯМКАХ**

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Досліджено вплив хромового покриття на механічні властивості оболонок твелів із сплаву Zr1Nb при розриві в подовжньому та поперечному напрямках при 20 і 350 °С. Випробування проводилися на зразках подібної форми, а саме, з однаковою довжиною робочої частини, що суттєво при порівнянні результатів. Показано, що при температурі випробувань 20 °С механічні властивості вихідних зразків у подовжньому напрямку вищі, ніж у поперечному. Пластичність незначно вища при  $T = 350$  °С у подовжньому напрямку, а  $\sigma_{0,2}$  і  $\sigma_b$  практично однакові. Осадження хромового покриття на зразки призводить до певного зростання механічних властивостей в поперечному і зниження  $\sigma_{0,2}$  при незмінності  $\sigma_b$ ,  $\delta\%$  у подовжньому напрямках при кімнатній температурі розриву, а при 350 °С практично не змінює властивості в обох напрямках.