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Сучасні технології виробництва труб-оболонок тепловиділяючих елементів (ТВЕЛ) з сплавів цирконію та стан виробництва в Україні

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Modern technologies for the production of fuel element cladding tubes (FEEL) from zirconium alloys and the state of production in Ukraine

Анотація. *У теперішній час атомна енергетика зберігає свої позиції як один з основних світових джерел енергії. В Україні передбачається подальший розвиток атомної енергетики тому одним з важливих напрямків є створення власного ядерно паливного циклу(ЯПЦ). Складовою частиною ЯПЦ є виробництво труб-оболонок тепловиділяючих елементів з сплаву цирконію Zr1Nb. Метою роботи є розробка технології, виготовлення дослідних партій труб із сплаву цирконію Zr1Nb в Україні та дослідження їх якості. Методика. Виготовлення труб в виробничих умовах. Дослідження якості труб з використанням оптичної мікроскопії, макроструктурного аналізу, механічних випробувань при кімнатної та підвищеної температурах, оцінка орієнтації гідрідів, корозійні випробування, дослідження тривалої міцності, повзучості. Результати. Вперше в Україні на базі Державного підприємства «Науково-дослідний та конструкторсько-технологічний інститут трубної промисловості ім. Я.Ю. Осади» виконані роботи по розробці технології та виготовленню дослідних партій труб-оболонок тепловиділяючих елементів(ТВЕЛ) з цирконієвого сплаву Zr1Nb розміром Ø9.13× вн. 7.72 мм. Оцінена якість труб згідно з вимогами стандартів ASTM. Комплексні дослідження структури, властивостей показали відповідність вимогам стандартів ASTM. Наукова новизна. Вперше в Україні розроблена технологія та в промислових умовах виготовлені труби–оболонки тепловиділяючих елементів з цирконієвого сплаву Zr1Nb розміром Ø9.13×вн.7.72мм. Показано, що якість труб відповідає вимогам стандартів на труби. Практична значимість. На основі виконаних досліджень можлива організація промислового виробництва в умовах трубних заводів України.*

Ключові слова: *атомна енергетика, труба-оболонка тепловиділяючий елемент, сплав цирконію, технологія, якість.*

Abstract. *At present, nuclear energy retains its position as one of the main world energy sources. In Ukraine, further development of nuclear energy is planned, therefore, one of the important directions is the creation of its own nuclear fuel cycle (NFC). An integral part of the NFC is the production of fuel element cladding tubes from zirconium alloy Zr1Nb. The purpose of the work is the development of technology, the manufacture of pilot batches of tubes from zirconium alloy Zr1Nb in Ukraine and the study of their quality. Methodology. Production of tubes in production conditions. Research of the quality of tubes using optical microscopy, macrostructural analysis, mechanical tests at room and elevated temperatures, assessment of hydride orientation , corrosion tests, studies of long-term strength, creep. Results. For the first time in Ukraine, on the basis of the State Enterprise "Scientific and Design and Technological Institute of Pipe Industry named after Ya.Yu. Osada", work was carried out on the development of technology and the manufacture of pilot batches of tubes-shells of fuel elements (TVEL) from zirconium alloy Zr1Nb with a size of Ø9.13× in. 7.72 mm. The quality of the tubes was assessed in accordance with the requirements of ASTM standards. Comprehensive studies of the structure and properties showed compliance with the requirements of ASTM standards. Scientific novelty. For the first time in Ukraine, a technology has been developed and tubes-shells of heat-generating elements made of zirconium alloy Zr1Nb with a size of Ø9.13×in.7.72mm have been manufactured in industrial conditions. It has been shown that the quality of the tubes meets the requirements of the standards for tubes. Practical significance. Based on the research performed, it is possible to organize industrial production in the conditions of Ukrainian tube plants.*

Keywords: *nuclear power, shell-and-tube fuel element, zirconium alloy, technology, quality.*

Introduction. Nuclear energy accounts for 1/6 of the world's fuel and energy balance and 43% for Western Europe. The growth of nuclear power plant capacities is predicted , primarily in the countries of Asia and the Asia-Pacific region (China, South Korea, India, Japan), some countries of Eastern Europe (Czech Republic, Slovak Republic), as well as a number of countries that are members of the Commonwealth of Independent States (Kazakhstan). Many countries intend to create nuclear power, including: Turkey, Iran, Indonesia, Vietnam [1, 2].

Given the significant role of nuclear energy in energy supply and the low cost of electricity production at nuclear power plants, which is achieved mainly due to the fuel component, it was natural to pose the problem of creating a National Nuclear Fuel Cycle (NFC) in Ukraine, which would guarantee the provision of nuclear power plants with fresh fuel, the supply of which should be independent of political and economic relations [2].

The problem of creating a National Center for the Promotion of Culture in Ukraine is part of a complex of particularly important national tasks.

An integral part of the Comprehensive Program for the Creation of the Nuclear Power Plant is the organization of the production of zirconium rolled products - fuel element cladding tubes and fuel assembly components. Until now, fuel element cladding tubes made of zirconium alloys have not been produced in Ukraine. The technology for manufacturing fuel element cladding tubes is one of the most science-intensive in the theory and practice of pipe production.

The operation of fuel element cladding tubes in the core is carried out in the most extreme conditions: at high operating temperatures, cyclic mechanical and thermal loads, in intense radiation fluxes, in the presence of an aggressive coolant and fuel pellet environment. On this basis, a set of strict requirements is put forward for cladding tubes: high mechanical properties at different temperatures, corrosion resistance in different environments at high pressures and temperatures, the required structure, texture, surface condition, restrictions on the anisotropy of properties, orientation of hydrides, and the presence of fluorine ions on the surface [3, 4].

Purpose and tasks. It is the metallurgical problems of creating a complex of properties of fuel element cladding tubes that determine the requirements for building a technological process. In world practice, technologies for producing fuel element cladding tubes from zirconium alloys were created 50-70 years ago, in this regard, when organizing their production in Ukraine, the achievements of modern science and technology should be taken into account.

Creative generalization of world science made it possible to establish that the most complete complex of metallurgical and technological tasks of production and use of tubes in the core zone of NPPs is reduced to reducing the structural and chemical heterogeneity of metals at all stages of technological processes: metal smelting, manufacturing of billets and tubes, creation of structures of a certain type, which provide increased radiation and corrosion resistance, reduction of hydride embrittlement , improvement of the surface quality of products. The final phase of research and development is the organization of production of fuel rod cladding tubes from zirconium alloys in the conditions of Ukrainian enterprises.

Thus, the need to solve the complex scientific problem of improving the quality of pipes and organizing their production for the core of nuclear power plants (NPPs) in Ukraine determines the relevance of the problem.

Material and methods of research. The main type of reactors used at Ukrainian nuclear power plants are water-water reactors cooled by ordinary water under pressure, VVER-type shell reactors. In the active zones of the reactors under consideration, similar in type but different in design are used, rod fuel elements assembled into assemblies, the tubes of which are made of zirconium alloys Zr1Nb, and U_{O2} pellets with different enrichment in U-235 (up to 5%) are used as nuclear fuel.

Elements of tubular structures widely used in the cores of nuclear reactors, for example, fuel rods, are among the most loaded structural components of the cores of nuclear reactors.

Fuel rods are operated in very difficult conditions: in powerful radiation fields of all types of reactor radiation; at high internal temperatures of the fuel core, reaching 2000...2500°C in the center, in the cladding - 300...350°C, at fairly high coolant pressures up to 16...17 MPa, with active external corrosion action on the cladding from the coolant side and internal - from the fuel side, gaseous and volatile uranium fission products, under the action of complex stresses on the fuel rod cladding tube material , swelling fuel, thermal cycles, radiation deformation, as well as hydrodynamic effects of high-speed coolant flow. [3]. Therefore, the work carried out comprehensive testing of the metal using optical microscopy, quantitative metallography, evaluation of mechanical properties at room and elevated temperatures in the longitudinal and transverse directions, corrosion tests, evaluation of hydride orientation, ultrasonic and eddy current control. In addition, creep and low-cycle fatigue were investigated.

Research results. In Ukraine, on the basis of the State Enterprise "Scientific Research and Design and Technological Institute of Pipe Industry named after Ya.Yu. Osada" (SE "NDTI"), work was carried out on the development of technology and manufacturing of shell tubes and fuel assembly products from zirconium alloy Zr1Nb of Ukrainian production. During the work in this area, the following complex of works was carried out within the framework of the Program for the creation of the Nuclear Power Plant:

- the main technological schemes for the pilot-industrial production of casing pipes and rods have been developed in relation to the domestic production base;
- several experimental batches of fuel element cladding tubes made of Ukrainian-made Zr1Nb alloy were manufactured at various Ukrainian pipe plants under industrial production conditions;
- a comprehensive assessment of the quality of experimental batches of pipes was carried out, confirming their compliance with the main indicators with the requirements of regulatory documentation (ASTM standards);
- a package of the first versions of regulatory documentation for the production of pipes and rods (technical specifications for pilot batches of pipes and rods, technological instructions, control methods) was developed and approved;
- a set of measures and technical solutions has been developed to modernize existing and purchase new equipment, ensuring the efficiency of the technological process and product quality;
- the basic technological scheme of industrial production of zirconium rolled products in Ukraine has been determined.

As part of the work performed, a technological scheme for the manufacture of fuel element cladding tubes using new elements was proposed:

- use of cast pipe billets obtained by various smelting methods: electron beam, vacuum arc and centrifugal casting;

- high-temperature pressing in the β-region of cast pipe blanks with large degrees of deformation;

- hardening by rolling heating.

In Ukrainian pipe plants, experimental batches of cladding tubes and rods that complete fuel assemblies were rolled in industrial conditions.

New elements of the technological scheme for pipe production include, first of all, the production and use of cast pipe billets ∅150-200 mm and ∅80 mm, manufactured by various methods: electron beam remelting with electromagnetic stirring, vacuum arc remelting, and centrifugal casting.

Cast pipe blanks were comprehensively investigated: macro- and microstructure, chemical composition, mechanical properties, hardness were evaluated. Quantitative metallography and electron microscopic studies were performed [5, 6]. Fig. 1 and 2 present the macro- and microstructure of cast billets of different smelting methods.

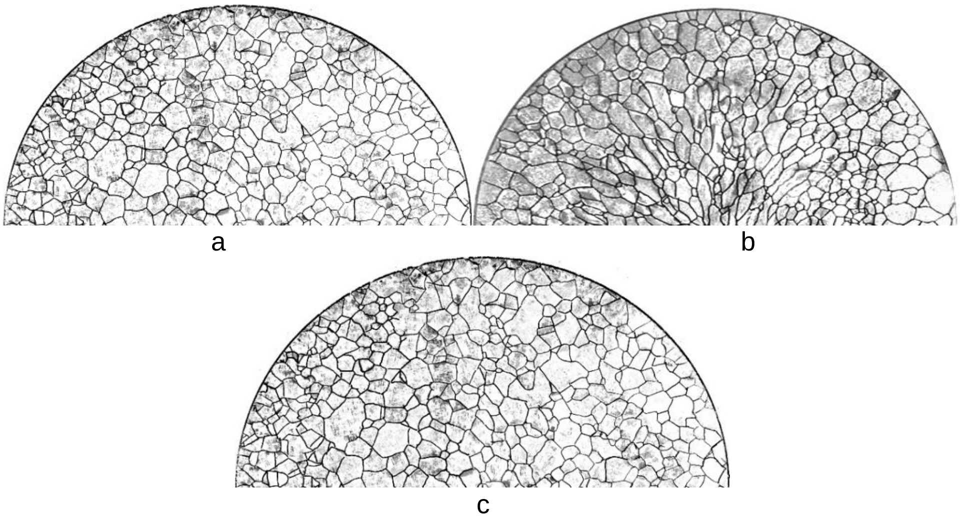


Fig. 1. Macrostructure of cast pipe billets of different smelting methods: a – electron beam remelting (EBR); b – electron beam remelting with electromagnetic stirring (GEMP);c – vacuum arc remelting (VDP)

The macrostructure of the Zr1Nb alloy melted by vacuum arc remelting (VDP), compared to electron beam remelting (EBR), is more homogeneous and fine-grained, as evidenced by the sizes of the α-zirconium and β-niobium plate packages and their periodicity (Fig. 3).

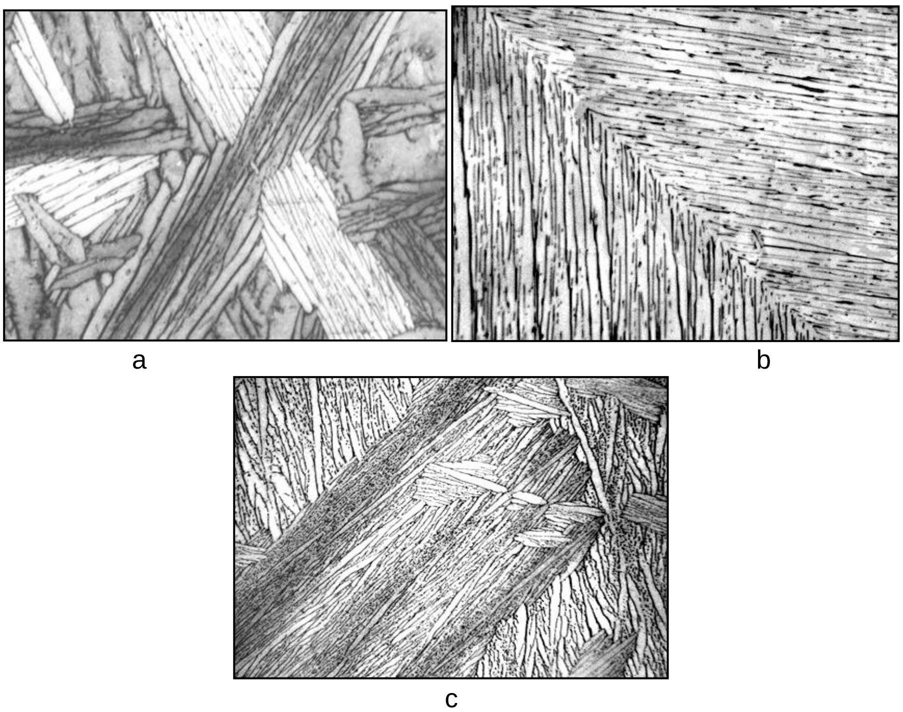
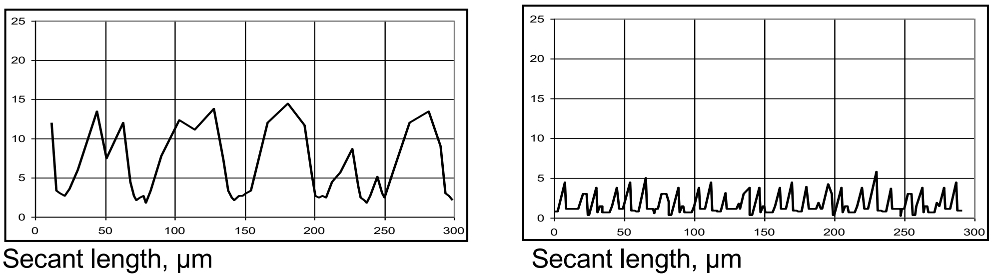


Fig. 2. Microstructure of cast pipe billets of different smelting methods: a - E P P; b - GEMP; c – VDP



a b

Fig. 3. Distribution of the width of the plates in the batch structure of castings of different smelting methods: a – EPP casting from Zr1Nb alloy; b – VDP casting

In the structure of cast blanks obtained by the VDP method, as compared to EPP, the size of the plates and the period of alternation of groups in packages are several times smaller and are 6...8 µm, as compared to 20...30 µm in EBR.

The characteristics of centrifugally cast pipe blanks according to the same parameters occupy values intermediate between EBR and VDP.

Special attention during the development of the technological scheme for the production of pipes was paid to the hot reshaping of cast pipe blanks [5]. The new solution of high-temperature pressing in the β-region with large plastic deformations was preceded by a set of studies on the influence of the deformation temperature, heating rate, degree of metal drawing (deformation), coating quality, assessment of the size of the gas-saturated layer, etc. Assessment of the properties of the metal structure by various methods made it possible to choose the optimal temperatures and degrees of metal deformation during hot pressing. Fig. 4 shows the metal structure of hot-pressed pipes at deformation temperatures in the range of 850...1100°C. The metal structure of pipes measuring ∅59×12 mm (trex pipes) is different - from the unrecrystallized one obtained at a deformation temperature of 850°C (Fig. 4 a), to bainite (Fig. 4 b) and martensitic (Fig. 4 c).

a b c

Fig. 4. Structure of hot-pressed pipes, at a deformation temperature of 850°C, not recrystallized, (a) to bainite (b) and martensitic (c)

In the manufacture of fuel element cladding tubes from zirconium alloys, the level of plasticity, which is determined by the structural state obtained at the stage of hot deformation, has a significant impact on the behavior of the metal during cold deformation. The technology developed at SE "NDTI" allows for one cycle of hot pressing to obtain trex tubes with a homogeneous, finely dispersed structure [6]. This creates the prerequisites for increasing the degree of deformation during rolling of tubes at the first cold re-section.

In the course of the research, it was established that the formation of a martensitic-type structure during hot deformation provides higher technological plasticity during the first cold reshaping.

A comprehensive assessment of the quality of fuel rod cladding tubes rolled using new technology elements showed that the tubes meet the requirements of ASTM standards in terms of their main parameters [7].

In addition to traditional tests regulated by standards, such as:

- estimation of geometric dimensions;
- evaluation of mechanical properties at room and elevated temperatures in the longitudinal and transverse directions;
- corrosion tests in steam under pressure;
- assessment of hydride orientation;
- determination of the anisotropy coefficient;
- ultrasonic control;

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- eddy current control;
- surface roughness assessment;
- the presence of fluoride ions on the surface, additional studies have been performed:
- low-cycle fatigue;
- creep;
- long-term corrosion resistance tests;
- electron microscopic studies;
- scanning electron microscopy to assess surface quality.

Analyzing the obtained results of the research on the quality of casing pipes, it should be noted the following. Research on the mechanical properties of pipes tested during stretching in the longitudinal and transverse directions at temperatures of 20°C and 350°C showed high plasticity with a high level of strength characteristics, which significantly exceed the requirements of ASTM standards and other standards (Table 1).

Table 1 - Mechanical properties of casing pipes of size 9.13× in 7.72 mm from Zr1Nb alloy

Manufacturer	Mechanical properties					
	Longitudinal direction			Transverse direction		
	Tensile strength $\sigma_в$, H/mm^2	Yield strength σ_{02}, H/mm^2	Relative lengthening δ, %	Tensile strength $\sigma_в$, H/mm^2	Yield strength σ_{02}, H/mm^2	Relative lengthening δ, %
Test temperature 20°C						
oh gti	580-590	415-425	34-36	550-600	500-533	16.1-16.7
JSC "NPTZ"	615-650	480-495	30-33	605-650	560-595	13-13.3
REQUIREMENTS TU 95-405-89 no less than	410	240	20	–	–	12
The temperature is 380°C						
oh gti	–	–	–	226-235	222-226	27-30
JSC "NPTZ"	235-265	135-153	530-560	225-240	190-200	27-36
Requirements TU 95-405-89 no less than	–	80	–	148	130	33

The higher level of mechanical properties is due to the significantly increased oxygen content in the Zr1Nb alloy (0.12...0.16%). Studies conducted at the National Scientific Center “Kharkiv Institute of Physics and Technology” at the electron microscopy level have established that this has a positive effect on the behavior of the metal when irradiated.

The values of the increase on samples of Ukrainian-made pipes of different melts and manufacturing methods are close to each other and are 14-16 m^2 /dm^3, which does not exceed the requirements of TU and ASTM standards.

The study of corrosion of Ukrainian-made Zr1Nb alloy pipes under three different autoclaving regimes showed that the corrosion rates are similar for the materials under study. An additional test regime for 72, 500, 1000 hours did not reveal any signs of nodular corrosion, fracture sites, or surface whitening (Fig. 6).

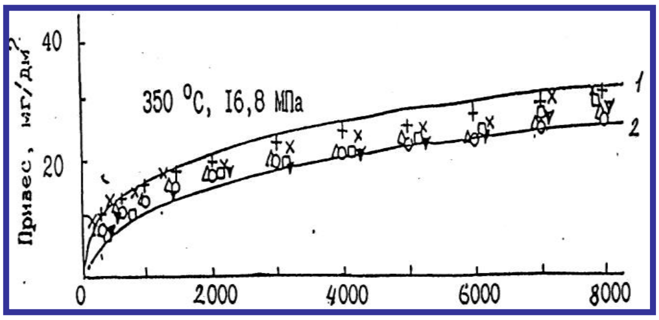


Fig. 6. Corrosion kinetics of samples of experimental batches of fuel element cladding tubes: 1 - alloy E-110; 2 - alloy CTC (calcium-thermal zirconium)

Studies of long-term strength and creep at loads from 157 to 227 MPa and test temperatures of 380°C showed higher plasticity of the Zr1Nb alloy in creep resistance tests. The time to failure - the moment of the appearance of the first microcracks at P=157 MPa for pipes made of Zr1Nb alloy was 1206 hours, the average creep rate ε=3-10-4%h, and for the E-110 alloy - 1170 hours at ε = 5-10-3%h (Fig. 7).

The microstructure of the finished pipes, evaluated using optical and electron microscopy, regardless of the technological options for production, is equiaxed. recrystallized grains of the α-phase with dispersed inclusions of the β-Nb phase (Fig. 8), grain size 3-10 μm. The recrystallization processes were complete, which was confirmed by electron microscopic studies.

Most grains (90%) are oriented with the basal plane (0001) almost parallel to the pipe surface, as shown by the texture study data.

The average orientation coefficient of hydrides in rolled pipes is 0.2-0.35 (Fig. 9).

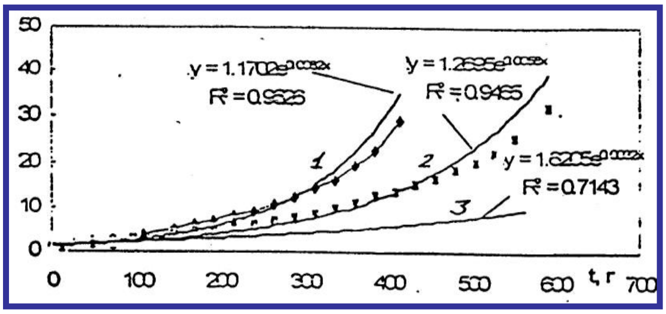
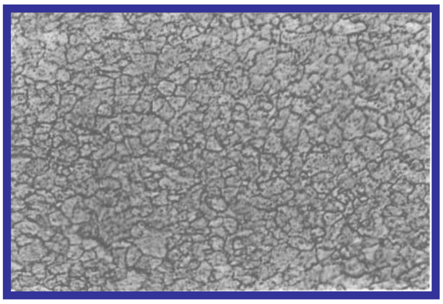
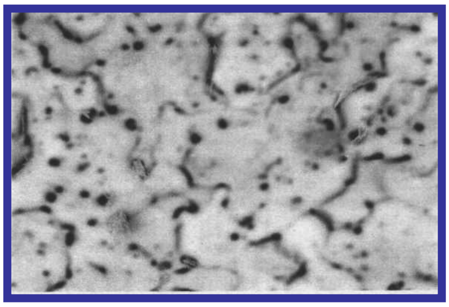


Fig. 7 Creep curves of fuel element cladding tube samples: 1 - alloy KTC 110 (P=221 MPa); 2 - alloy KTC 110 (P=219 MPa); 3 - alloy E 110 (P=119 MPa).



a



b

Fig. 8. Microstructure (a), ×200 and separation of the second phase (b), ×1000 (b) in pipes with dimensions of 9.13×7.72 mm made of ZrINb alloy

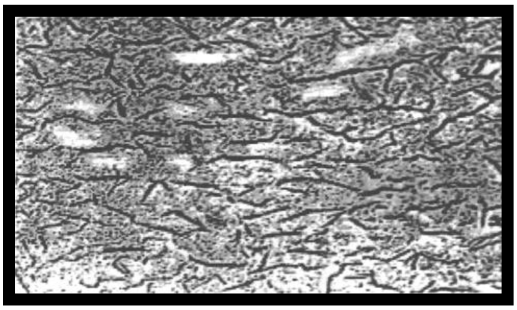


Fig. 9. Orientation of hydrides in the metal of pipes measuring 9.13×7.72 mm in size made of ZrINb alloy

Experimental batches of tubes were used to manufacture fuel rod models, which were tested in conditions close to reactor conditions. All models withstood the tests.

The main results of the work carried out to develop the technology of zirconium rolling are briefly presented above. The entire complex of works on the smelting of the billet, chemical treatment, calculations of tool calibrations, acoustic emission studies on the selection of the limiting degrees of deformation, studies of crack resistance, etc. are not reflected.

Unfortunately, in recent years, work in this area has been discontinued in Ukraine.

The plant, which was supposed to house the production of zirconium rolled products and other types of pipes for the needs of nuclear energy, - the Experimental Plant of the Pipe Institute (OZ GTI, and then DZPT) - was completely destroyed, and the Pipe Institute (SE "NDTI" named after Ya.E. Osada) is in critical condition.

Ukraine still has scientific and production potential that needs to be used in the near future. For example, the production of zirconium rolled products can be located in Nikopol at the OSKAR plant (the former shop for the production of pipes for nuclear power TVC-4 PTZ).

Conclusions

The conducted research allowed us to develop a basic technological scheme for the production of zirconium rolled products: fuel element cladding tubes, zirconium alloy rods. The first version of regulatory documentation was created.

With a state approach, in the context of the approved decision to build a nuclear fuel production plant and the availability of a zirconium raw material base in Ukraine, the creation of the production of components for the manufacture of fuel assemblies, primarily fuel element cladding tubes, should not be postponed for decades. It is necessary to use the achievements of the Ukrainian scientific school in the field of obtaining zirconium alloys and rolling from them.

The primary tasks for organizing the production of zirconium rolled products are:

1. Obtaining zirconium sponge and ingots from it.
2. Creation of production facilities for the production of ingots and trex pipes.
3. Reconstruction and acquisition of equipment for rolling and quality control of fuel rod cladding tubes at the OSKAR enterprise (Nikopol, former TVC4).
4. Resume research and development work to support the organization of production and rolling of zirconium and its alloys in Ukraine.

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