EAST EUROPEAN JOURNAL OF PHYSICS
East Eur. J. Phys. **3**. 110-115 (2021)
DOI:10.26565/2312-4334-2021-3-17

RESEARCH AND DEVELOPMENT OF FUEL RODS METALLURGICALLY BONDED WITH FUEL CLADDING FOR NUCLEAR INSTALLATIONS[†]

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Received June 24, 2021; revised July 29, 2021; accepted August 27, 2021

The design and scheme for manufacturing fuel rods metallurgically bonded with ribbed aluminum claddings using hot isostatic pressing and contact-reactive brazing are presented. It is shown that the developed scheme can be used both for production of dispersive fuels and high-density fuels based on uranium alloys. The results of investigations of brazed joints of aluminum cladding with a matrix composition based on aluminum and with samples of E110 alloy through copper and silumin coatings are presented. The results of research of brazed joints of an aluminum cladding with an aluminum-based matrix composition and samples of zirconium alloy E110 made through copper and silumin coating are presented. The strength of brazed joints, composition of diffusion layers formed as a result of contact-reactive brazing in a high vacuum have been determined. The modes of hot isostatic pressing that provide crimping of the ribbed cladding of fuel pellets and rods and obtaining a metallurgical bonding between their surfaces have been defined. It is shown that satisfactory bond strength is provided starting from the temperature of 610 °C. The maximum strength values obtained on the compounds Al-(Al+12% Si)-Zr and Al-Cu-Zr are 57.0 MPa and 55.3 MPa respectively. The fracture of the of aluminum samples joints, obtained with the Cu layer at a temperature of 620 °C, occurs on threaded joints at the strength value of 82 MPa. The results of research of the composition of diffusion layers formed by brazing compounds Al-(Al + 12% Si)-Zr and Al-Cu-Zr are presented. It was established that hot pressing provides the best results for manufacturing of fuel rod dummies in the studied range of modes at a temperature of 630 °C, a pressure of 380 MPa and exposure of 20 minutes.

Keywords: fuel rod, aluminum alloys, dispersive fuel, alloys of uranium, hot isostatic pressing, contact-reactive brazing.
PACS: 28.41.Bm, 81.05.Ni, 81.15.Ef, 81.40.Vw, 81.70.Bt

In connection with the implementation of nuclear non-proliferation programs, the world's initiated work is aimed at the development and production of fuel for research nuclear installations (RNI), with an enrichment of ^{235}U up to 20% [1-3]. Efforts to develop a promising fuel for most reactors are focused on solving two problems: improving existing fuel and developing new high-density fuel. In this regard, medium-alloyed uranium alloys are considered as promising fuels: U + 7% wt. Mo + 1% wt. Zr (ρU = 16.0 g/cm^3), U + 6% wt. Zr + 4% wt. Nb (ρU = 15.8 g/cm^3), U + 9% wt. Mo (ρU = 15.5 g/cm^3) and uranium compounds: U_3Si (ρU = 15.0 g/cm^3), U_3Si_2 (ρU = 11.3 g/cm^3), UO_2 (ρU = 9.2 g/cm^3) [4,5]. At the initial stage, compositions UAl_x–Al, UO_2–Al, U_3O_8–Al were used, which became widespread. Then much attention was paid to silicide fuel. The U_3Si_2–Al composition with a uranium content loading of 4.8 g/cm^3, after successful tests in Oakridge in 1987 was approved by the US Nuclear Regulatory Commission (NRC), and is widely used around the world [5, 6]. This fuel has certain disadvantages. This is a relatively low uranium content and a problem of silicide fuel processing. At the same time, denser fuels based on uranium alloys were developed. Alloys U-(8…10)% Mo by weight are considered promising [7]. Despite the fact that the study of medium-alloyed uranium alloys has produced a significant number of results, it turned out that there are some obstacles to their qualification as fuel for research reactors. The main ones can be summarized as follows [8]: under irradiation, a fuel-aluminum matrix interaction zone consisting of uranium aluminides UAl_2, UAl_3 and UAl_4 is formed; starting from some burnup, gas bubbles begin to form at the aluminum matrix - interaction zone interface due to fission products, introduced into the matrix; with increasing burnup, in the cross sections of the fuel rods located in the zone of maximum energy release, along the line of contact of the zone of interaction with the matrix, gas cavities up to 1…2 mm are formed; further increase in burnup leads to formation of gaps in the fuel core and ballooning of the cladding. Specialists in the development of fuel for research reactors have identified ways to overcome the problem of uranium alloys interaction with aluminum matrix and cladding. These ways consist in alloying of aluminum alloys with 2-8% wt. of silicon, alloying of uranium alloy U+7% wt. Mo with 1% wt. of zirconium or titanium; application of protective coatings of Si, Nb, Zr, ZrN and other materials on fuel cores. The development of fuel rods is carried out using the technology of filling the niobium layer coated fuel composition U-9% wt. Mo with a melt of silumin (Al+12% wt. Si) [6,9]. A prominent place among various designs of fuel for RNI is occupied by fuel rods in aluminum and zirconium claddings. Fuel rods in claddings with spacer ribs are considered promising [3, 8, 10]. Hexagonal, square, circular and other shape fuel assemblies are produced with such fuel rods. Moreover, in most cases they provide metallurgical bonding between the cladding and the core. This allows to significantly reduce operating temperatures of fuel compositions, increase their service life and structural rigidity [4]. Various

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methods are used for the metallurgical bonding of the cladding and the fuel core, such as hot isostatic pressing, drawing, extrusion, filling with braze melt, for example, with silumin. It is of interest to use the method of hot isostatic pressing (HIP) for manufacturing of rod-type fuel elements [5, 6].

This method allows separate production of claddings with a preset configuration, fuel pellets (rods), to carry out their operative control, product assembly, following which to bring into contact the connected surfaces, and to carry out their metallurgical bonding. The purpose of this work is to develop metallurgically bonded fuel rods and to study the possibility of obtaining bonds of the ribbed cladding with fuel cores by means of HIP in combination with contact-reactive brazing.

SAMPLES, FUEL ROD DUMMIES, RESEARCH METHODS

Preparation procedure, materials, design of samples

The possibility of implementation of the technological scheme for the production of metallurgically bonded fuel rods was introduced in the research. It includes the following main operations:

- production of pellets or rods;
- production of a profiled cladding and plugs of aluminum alloy;
- production of fuel pellets or cores of uranium alloy;
- applying a coating of the materials that form a liquid phase in the temperature range of 550...620 °C on the surface of the pellets;
- attaching the plug to the cladding by argon-arc welding;
- filling the inner space of the cladding with fuel pellets (or a rod-type integral core);
- fuel rod sealing;
- processing of the fuel rod dummy by the HIP method;
- fuel rod leakage control and geometry control.

The following composition of aluminum alloy 6082 (AD35) was used in the research as the cladding material (in wt.%): Al + 0.8% Mg + 1.1% Si + 0.5% Mn + + 0.27% Fe + 0.12% Cr + 0.09% Zn + + 0.1% Cu. Its chemical composition and characteristics are close to the alloys SAV-1 and 6061, that are used for production of fuel claddings for research reactors [3,4]. UO_2-Al was used as the pellet composition, and HfO_2-Al was used as the simulating composition. Powders of planetary milled alloy 6082 swarf and the industrially produced aluminum powder of the PA-4 brand were chosen as the matrix materials. Pellets of HfO_2-Al composition were produced using the powder metallurgy methods described in detail in [12-14]. Characteristics of fuel pellets are given in Table 1. Uranium alloy core dummies were rods made of alloy E110 (Zr+1% wt. Nb). The choice was made due to the fact that in the future, uranium metal alloys with a protective zirconium coating on the surface can be used.

Two methods of coating formation have been developed: vacuum arc deposition and the method using centrifugal casting in thin-walled zirconium mould [15, 16].

The latter method provides not only strong adhesion of the core surface with the cladding, but also formation of a compensating central hole, moving to the central zone of impurities in the form of oxides, carbides and other compounds, additional doping with zirconium. It should be noted that the experimental metallurgically bonded fuel rods produced using centrifugal casting of uranium alloys in zirconium cladding have proved successful during the in-pile tests at the water coolant temperature of 282 °C, energy density of 21.6 kW/kg at thermal and fast neutron fluxes of 1.16×10^{17} n/m^2·s and 0.9×10^{17} n/m^2·s [16]. After a burnup of 22 MW day/kgU, they maintained integrity, metallurgical bonding of the fuel cores with the cladding and leak tightness. Testing of the technology and examination research of samples were carried out in two stages. The first stage included studying the characteristics of brazed joints of aluminum with aluminum and zirconium with aluminum made through layers of copper and silumin.

Table 1. Characteristics of dummy fuel pellets

Composition	Fuel dummies (fraction/volume ratio)	Matrix particle shape/fraction	Density of pellets (avg.)
UO_2-Al	-400 + 200 μm / 20%	Al-alloy / 112… 315 μm	0.96
Al-alloy	without dummies	grain / 112… 315 μm	0.98
ПА-4	without dummies	grain / 112… 200 μm	0.97
HfO_2-Al	-400 + 200 μm / 20%	Al-alloy / 112… 315 μm	0.96

These samples were two cylinders made of aluminum alloy 6082, with threaded ends for securing in the grips of the tensile testing machine. A recess of cylindrical shape with a diameter of 15 mm and a height of 5 mm was made in one of them to center the test samples. Some of the samples were produced of alloy 6082, and some – of alloy E110. Layers of copper or silumin with a thickness of 5…8 μm were applied on the surface of the samples by the vacuum arc deposition technique.

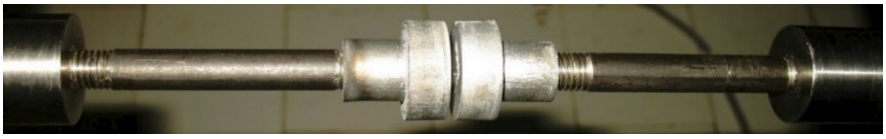
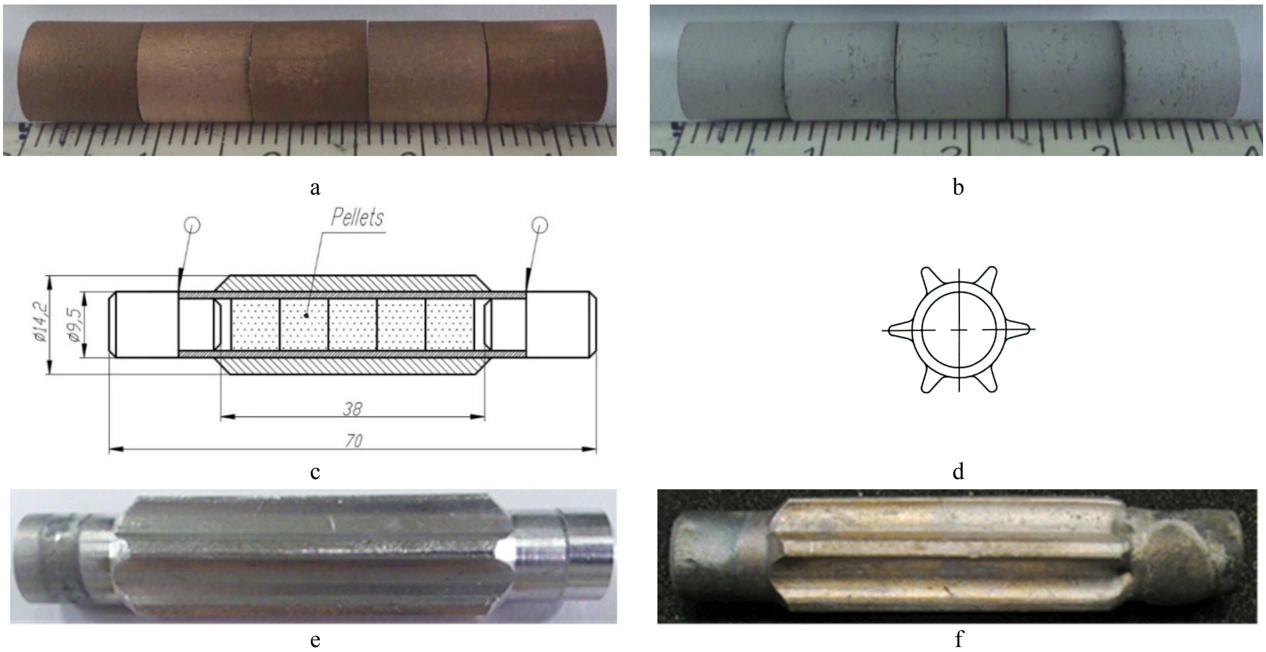


Figure 1. Sample for studying characteristics of brazed joints: Al-Cu-Zr, Al-(Al+12% Si)-Zr, Al-Cu-Al, Al-(Al+12% Si)-Al

Before coating, the surface of the samples was cleaned of oxide films by bombardment with ions of a vacuum arc at a bias voltage -800 V. The current of the vacuum arc was 120 A. The cleaning of samples was carried out in pulsed mode to avoid overheating. The temperature of the samples was monitored with a pyrometer, and it did not exceed 410 °C throughout the process. Coatings on aluminum cylindrical samples and pellets with dispersion compositions were also carried out in pulsed mode at a bias voltage -40 V. The rate of the deposition when using a silumin cathode was ~82 μm/h, and when using copper ~25 μm/h. Coating of E110 alloy products was performed in a continuous mode at a bias voltage -40 V. At the end of the process the samples temperature did not exceed 670 °C. The appearance of pellets with copper and silumin coatings is shown in Fig. 2 a, b.



a b

c d

e f

Figure 2. Fuel rod dummies before and after HIP treatment: a, b – pellets of dispersion composition HfO_2-Al with copper and silumin coatings; c, d – design of fuel rod dummy; e, f – appearance of the dummy workpiece and fuel rod dummy before and after HIP treatment

Methods

Diffusion bonding of samples (Fig. 1) was performed in a vacuum of 10^{-3} Pa in the temperature range of 600…630 °C. Exposure time ranged from 5 to 30 minutes. The heating rate was 7 °C/min, and cooling rate was 5 °C/min. In the process of testing the modes, the strength of the connection was measured, the interface was studied using metallography, SEM and EDX microanalysis with a scanning electron microscope JSM 7001F at a voltage of 10 kV. EDX microanalysis was performed with an INCA PentaFET*3 detector. Research of influence of HIP modes on characteristics of fuel rod dummies were carried out in the temperature range of 580…630 °C and the pressure range of 340…380 MPa. Exposure time was 10, 15 and 20 minutes. The microstructure of pellets and claddings, as well as the interface between them, were studied on sections produced of molded fuel rod dummies. The degree of contact of the surface of pellets with the surface of cladding was determined by the formula:

$$\varphi = \left(L_k / L_{total} \right) \cdot 100\% \qquad (1)$$

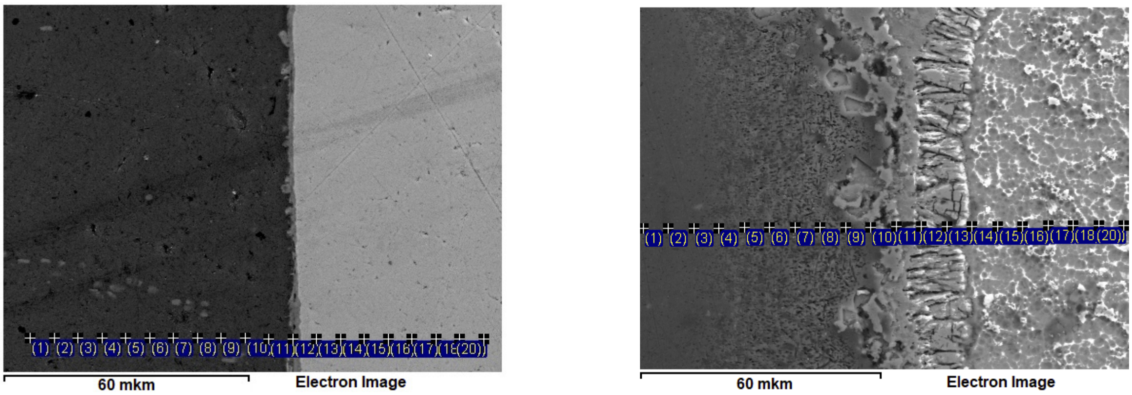
where φ is the contact factor, L_{total} is the circumference, $L_k = a \cdot R$ is the arc length of the segment (contact), R is the radius of the inner hole of the cladding; a is the angle of the segment.

EXPERIMENTAL RESULTS

Mechanical tests of brazed joints revealed that starting from a brazing temperature of 610 °C, a satisfactory bond strength is provided. The maximum strength values obtained on the bonds Al-(Al + 12% Si)-Zr and Al-Cu-Zr, respectively, were equal to 57.0 MPa and 55.3 MPa. A diffusion layer with a width of 1…6 μm of the following elemental composition (22…40)%wt. Al + (10… 18)% wt. Si + (38…64)% wt. Zr is formed at the interface of the samples with the silumin layer after brazing (Fig. 3a). When using a copper coating after the process that occurs as a result of formation of the liquid phase due to the eutectic interaction in the Al-Cu system, formation of two diffusion layers is observed (Fig. 3b). A layer 30…40 μm thick is formed on the aluminum side. Its composition is: The composition of this layer: (94… 95)% wt. Al + (3…4)% wt. Cu.

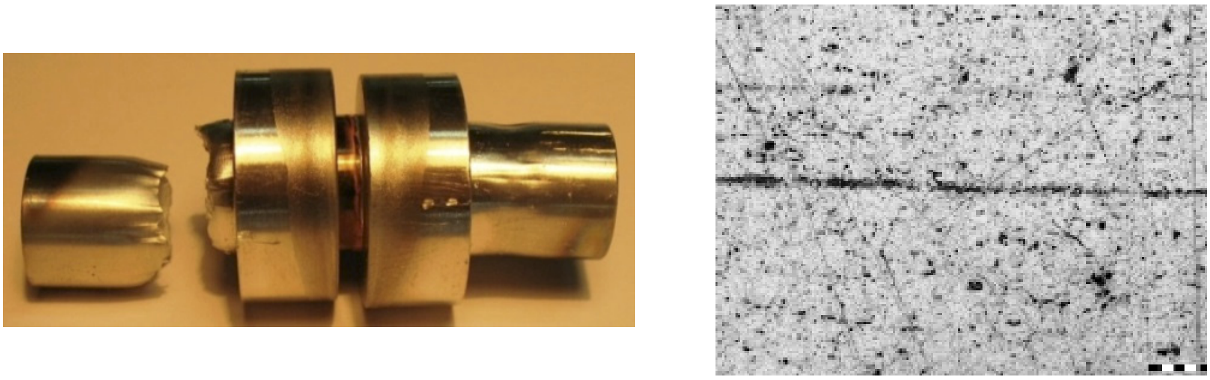
A layer 20…30 μm thick is formed on zirconium side. The concentration of elements in the layer varies in the range of (24…40)%wt. Al + (1…13)%wt. Cu + (44…65)% wt. Zr. On all tested samples, destruction occurred at the interface, i.e. along the diffusion layers. In the process of mechanical tests of aluminum samples bonded through a layer of Cu (Al-Cu-Al) at a brazing temperature of 620 °C, their destruction occurred along the thread (screw) joint (Fig. 5a).

Estimations revealed that the strength of the connection in this case exceeds 82 MPa. A thin diffusion layer 1…3 μm thick is formed at the interface of brazed samples (Fig. 5b). The results of research of the influence of HIP modes on the characteristics of joints in the fuel rod dummies are presented in Table 2 and in Fig. 6. The obtained results reveal that in mode 1 (Table 2), there is only 45…50% contact of the cladding and the core (Fig. 6a). Increasing the temperature up to 610 °C and the pressure up to 360 MPa leads to an increase in the contact surface to ~ 65…75% (Fig. 6b). The pellet-to-cladding gap varies from 18 μm to 211 μm. In mode 3 (T = 630 °C, P = 380 MPa, τ = 20 min), the coefficient of contact of the core with the cladding is ~ 90…95% (Fig. 6c).



a b

Figure 3. Brazed joint interface: a – Al-(Al+12% Si)-Zr; b – Al-Cu-Zr

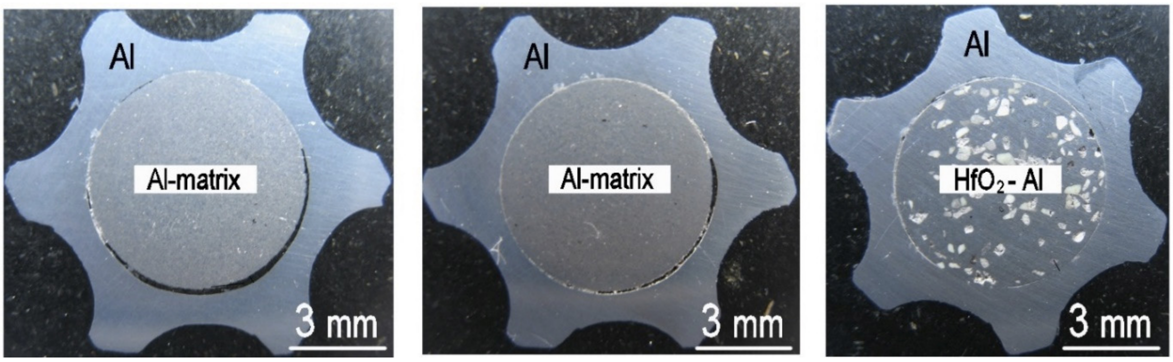


a b

Figure 4. Results of mechanical tests and research of Al-Cu-Al connection:
a – the destruction after mechanical tests; b – interface of the section

Table 2. Influence of HIP modes on the characteristics of fuel rod dummies

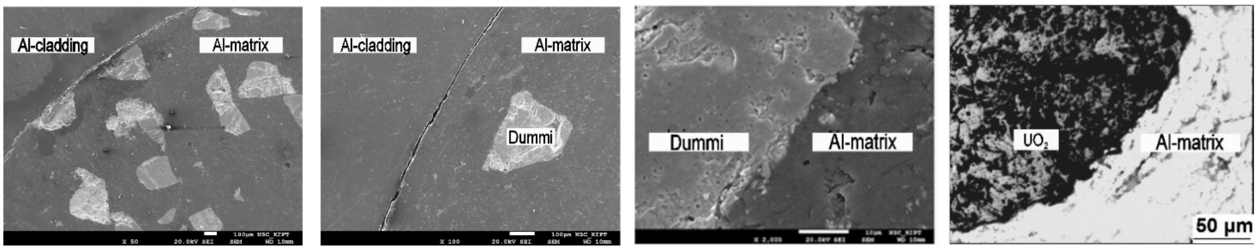
Test modes	Temperature, °C,	Pressure, MPa	Exposure, min	Contact factor (φ),%
#1	590	340	10	40…50
#2	610	360	15	65…75
#3	630	380	20	≥95



a b c

Figure 6. Appearance of cross sections of fuel rod dummies after HIP treatment in different modes:
a – mode #1; b – mode #2; c – mode #3

These data indicate the presence of metallurgical bonding between the cladding and the surface of the pellet. The boundary of the connection of the cladding with the aluminum matrix and the core dummy with the matrix are shown in Fig. 7. Vickers microhardness values of the aluminum cladding, aluminum matrix and fuel dummies are: 500…560 MPa, 608…620 MPa and 686…735 MPa respectively.



a b c d

Figure 7. SEM microstructure cross-section of fuel rod dummy with dispersion composition pellets of UO_2 and dispersion composition UO_2–Al: a – ×50; b – ×100; c – ×2000; d – dispersion composition UO_2–Al

CONCLUSIONS

The directions for the development of fuel compositions and designs of fuel rods for research nuclear installations have been determined. The possibility of implementation realization of the technological scheme for production of rod-type metallurgically bonded fuel elements with ribbed cladding by hot isostatic pressing in a combination with contact-reactive brazing is shown. The modes of applying copper and silumin coatings on aluminum and zirconium samples by vacuum arc deposition technique and obtaining joints by contact-reactive brazing and hot isostatic pressing have been worked out. It is shown that satisfactory bond strength is provided starting from the temperature of 610 °C. The maximum strength values obtained on the compounds Al-(Al+ 12% Si)-Zr and Al-Cu-Zr are 57.0 MPa and 55.3 MPa respectively. The fracture of the of aluminum samples joints, obtained with the Cu layer at a temperature of 620 °C, occurs on threaded joints at the strength value of 82 MPa. The results of research of the composition of diffusion layers formed by brazing compounds Al-(Al + 12% Si)-Zr and Al-Cu-Zr are presented. It was established that hot pressing provides the best results for manufacturing of fuel rod dummies in the studied range of modes at a temperature of 630 °C, a pressure of 380 MPa and exposure of 20 minutes.

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РОЗРОБКА ТА ДОСЛІДЖЕННЯ ТВЕЛІВ З МЕТАЛУРГІЙНИМ З'ЄДНАННЯМ СЕРДЕЧНИКА І ОБОЛОНКИ ДЛЯ ЯДЕРНИХ УСТАНОВОК

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Приведені конструкція та схема виготовлення стрижневих твелів з ребристою алюмінієвою оболонкою зчепленого варіанту з використанням методів гарячого ізостатичного пресування та контактно-реактивної пайки. Показано, що розроблена схема може бути використана як для виготовлення дисперсійного, так і високощільного палива на основі сплавів урану. Представлені результати досліджень паяних з'єднань алюмінієвої оболонки з матричною композицією на основі алюмінію та із зразками зі сплаву Е110 через мідне та силумінове покриття. Визначені міцність паяних з'єднань, склад дифузійних шарів, що утворюються в результаті контактно-реактивної пайки у високому вакуумі. Визначені режими гарячого ізостатичного пресування, що забезпечують обжимання ребристою оболонкою паливних таблеток і стрижнів та одержання металургійного з'єднання між їхніми поверхнями. Показано, що починаючи з температури 610 °C забезпечується задовільна міцність зчеплення паяних з'єднань, а максимальні значення міцності, отримані на з'єднаннях Al-(Al+12%Si)-Zr і Al-Cu-Zr, відповідно, становлять 57,0 МПа і 55,3 МПа. Приведені результати досліджень складу дифузійних шарів, що утворюються в результаті пайки з'єднань: Al-(Al+12%Si)-Zr і Al-Cu-Zr. Встановлено, що при використанні гарячого пресування найкращі результати для виготовлення макетів твелів в досліджуваному діапазоні режимів забезпечуються за температури 630 °C, тиску 380 МПа і тривалості процесу 20 хвилин. Відпрацьовані режими нанесення мідного і силумінового покриттів на алюмінієві та цирконієві зразки вакуумно-дуговим методом (КІБ) та одержання з'єднань контактно-реактивною пайкою в комбінації з гарячим ізостатичним пресуванням.

Ключові слова: стрижневі твели, сплави алюмінію, дисперсійне паливо, сплави урану, гаряче ізостатичне пресування, контактно-реактивна пайка.