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MANUFACTURING FEATURES AND CHARACTERISTICS OF URANIUM DIOXIDE PELLETS FOR SUBCRITICAL ASSEMBLY FUEL RODS[†]

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The influence of technological processes and manufacturing of uranium dioxide fuel pellets for fuel elements for experimental fuel assembly (FA-X) which was designed as an alternative fuel for the nuclear research installation (NRI) "Neutron Source Controlled by Electron Accelerator" were investigated. Unlike standard production processes of UO_2 pellets, the special feature fabrication process of this nuclear fuel type is production of uranium dioxide powder with enrichment of 4.4 %wt. of ^{235}U achieved by mixing of two batches of powders with different uranium contents: 0.4 %wt. ^{235}U and 19.7%wt. ^{235}U, as well as ensuring the required tolerance of fuel pellets without the use of machining operations. A set of design and process documentation were developed in the R&D Center at NSC KIPT. Experimental stack of fuel pellets, fuel elements and a pilot fuel assembly FA-X were fabricated and designed to be compatible and interchangeable with VVR-M2 fuel assembly adopted as a standard assembly for the first fuel loading at the "Neutron Source Driven by an Electron Accelerator" FA. As opposition to the variant of VVR-M2 fuel assembly which consisted of three fuel rods of tubular shape with dispersion composition UO_2-Al, FA-X accommodates six fuel rods of pin-type with UO_2 pellet which located in the zirconium cladding (E110) as the closest analogue of fuel rods of VVER-1000 power reactor. Inside cladding locate a 500 mm high fuel stack which is secured against displacement by a spacer. In the basic variant of FA-X the fuel pellets are made of UO_2 with ^{235}U enrichment near 4.4 %wt.
Keywords: fuel rod, pellet, fuel assembly, powder, uranium dioxide, enrichment, mixture uniformity, density, microstructure.
PACS: 28.41.Bm, 28.50.Dr, 29.25.Dz, 47.51.+a, 81.05.Je, 81.20.Ev, 83.50.Xa

SUBCRITICAL ASSEMBLY OF THE "NEUTRON SOURCE" NUCLEAR FACILITY

The subcritical assembly (SCA) of the "Accelerator Driven Neutron Source" nuclear facility serves to multiply primary neutrons from the fission of uranium-235 and includes: a core of fissile material, a moderator and reflector of neutrons, and a coolant [1].

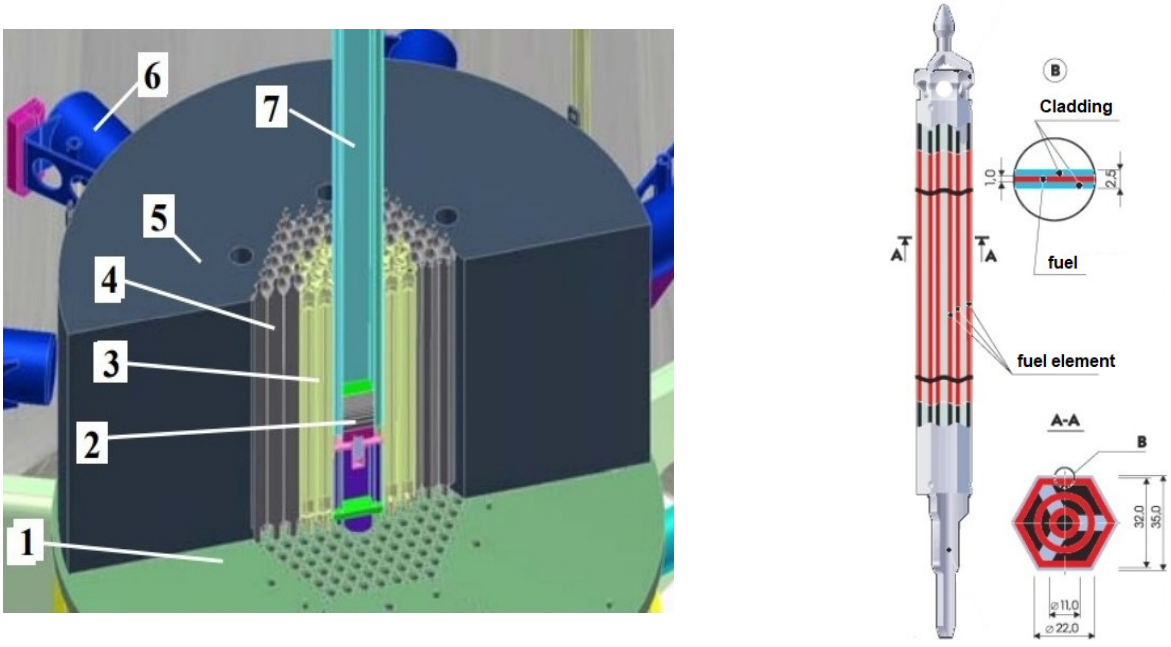

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Figure 1. 3D model of the subcritical assembly (a) [1]: 1 - baseplate; 2 - neutron-forming target; 3 - VVR-M2 type fuel assembly; 4 - beryllium reflectors; 5 - graphite reflector; 6 - neutron channel; 7 - vacuum channel of electron beam; standard fuel assemblies of VVR-M2 type (b) [2]

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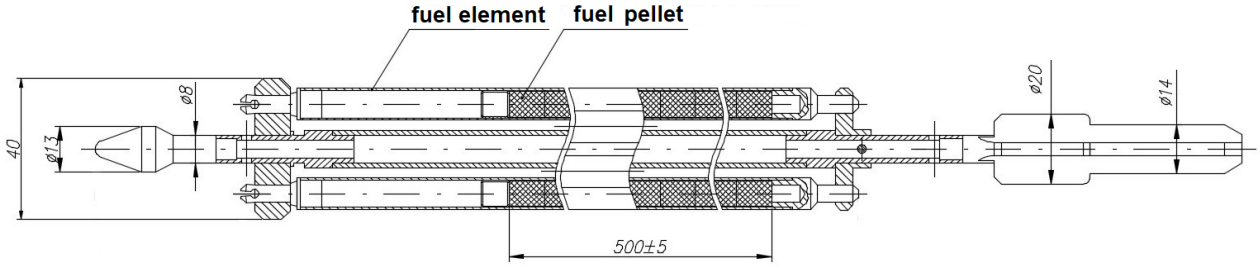
DESCRIPTION OF FA-X DESIGN

As an analog of fuel element for VVER-1000 reactor was choose a good approbated with characteristics was selected a fuel element FA-X (Fig. 2a, c) [3]: the length of the fuel element was decreased from 3800 mm to 600 mm, the height of fuel stack - from 3530 mm to 500 mm. Similar parameters of fuel elements are as follows: material and diameter of cladding - zirconium alloy E110, Ø 9.1 mm, type of the fuel composition - UO_2, enrichment of pellets with uranium-235 isotope – 4.4 %. Productions of UO_2 pellets according with technical requirements [4], must have specified characteristics (Fig. 2b): appearance, geometry, density, uranium content, oxygen coefficient, enrichment, and chemical composition.

As part of project R515 "Design and technology for fabrication of fuel pellets and fuel assemblies for subcritical assembly and testing for reliability and safety" the laboratory technology for fabrication of uranium dioxide pellet fuel with the required set of characteristics including pellet geometric dimensions, enrichment, density, uranium content was developed.

The peculiarity of the fabrication process of fuel pellets with content of ^{235}U near 4.4%wt. for FA-X is the preparation of uranium dioxide powder by mixing of two powder batches with enrichment: 0.4%wt. ^{235}U and 19.7 %wt. ^{235}U. Another peculiarity of the technology was obtaining of samples with required geometric dimensions within the tolerance range without additional mechanical operations.



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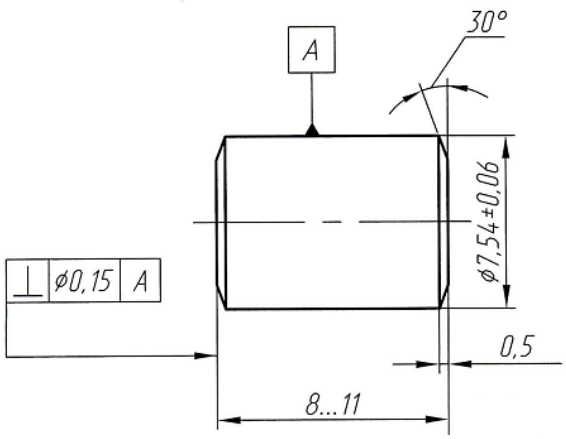
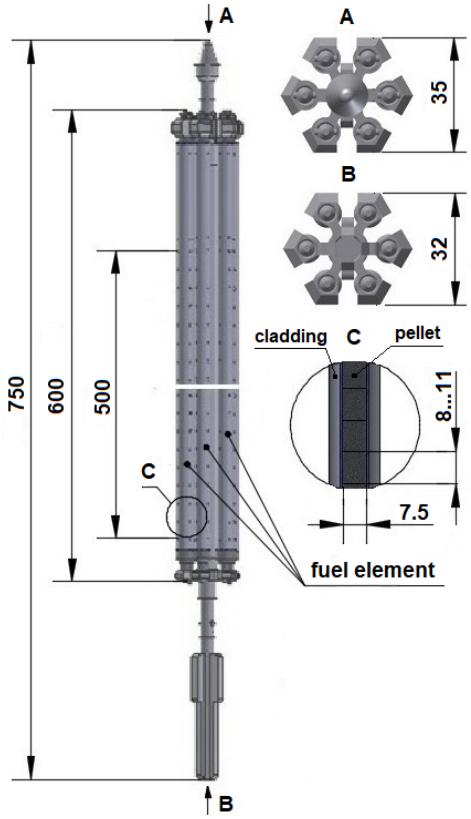


diameter 7.48-7.60 mm
height 8-11 mm
density 10.30-10.60 g/cm^3
enrichment 4.40-4.43% wt. ^{235}U
uranium content – more 87.4%
oxygen coefficient 1.92-2.05

b



c

Figure 2. Fuel assembly (FA-X) (a — common view, c - 3D-model); UO_2 pellet and main requirements (b)

MATERIALS AND RESEARCH METHODS

Uranium dioxide powders with different ^{235}U content - 19.905±0.224 %wt. and 0.454±0.044 %wt. were used as starting materials. Fuel pellets were formed by two-sided cold pressing in a steel mold with a diameter of 10 mm. Fuel pellets were sintered in furnace with graphite heater at 1700 ºC in vacuum with preliminary annealing of binder at 600 ºC.

The control of the uranium dioxide powder mixtures and fuel pellets enrichment with isotope ^{235}U was carried out according to the methodology №07-123:2011 "Uranium and its compounds. Methods for measuring the mass fraction of uranium-235 using semiconductor gamma spectrometer CANBERRAGL-0515R and MGAU program". The enrichment value is calculated from the ratio of intensities of the obtained hardware gamma spectra of ^{235}U and ^{238}U in the energy range of 89-100 keV using the program of multigroup analysis MGAU. The technique was validated for the relative error of measurements of 4 % at the confidence probability P = 0.95.

The powders were mixed in a rotating steel drum mixer. The homogeneity (quality) of the powder mixture was estimated by methods of mathematical statistics, by the so-called key-component – the ^{235}U content.

The non-uniformity coefficient in % (Fig. 6) determined from the ratio was used as the mixture uniformity criterion:

$$V_c = \frac{100}{\bar{c}} \sqrt{\frac{1}{n-1} \sum_{i=1}^{n} (c_i - \bar{c})^2} , \tag{1}$$

where c_i mass or volume fraction of the key-component in the *i*-sample; $\bar{c}$ - the average value of the mass or volume fraction of the key-component in the *i*-samples; *n* - the total count of samples taken from the mixture.

The homogeneity *M* was determined as (%):

$$M = 100 - V_c \tag{2}$$

The quality of the mixture is considered satisfactory at V_c = 6-8% (homogeneity *M*=92-94%), good at V_c= 4-6% (*M*=94-96%) and ideal at V_c<4% (*M*>96%).

The homogeneity of the obtained mixtures, located in a cube-shaped container with a side of 40 mm, was controlled in five different "source-detector" geometries. Four measurements were carried out with the detector against the side surface of the container with a mixture of uranium dioxide powders, each measurement differed from the previous one by rotating the container by 90°. The fifth measurement was carried out with the detector against the end surface of the container.

To control the homogeneity of the mixture, the heterogeneity factor was calculated by formula (1), where the values of the content of isotope ^{235}U obtained by processing five gamma-spectra with the MGAU software were used as and values.

In addition, another approach was used to control the homogeneity of the obtained mixtures. The results of the processing of five spectra for each mixture were subjected to statistical analysis: the mean values ($\bar{E}$) of the content of isotope ^{235}U, the standard deviation of $s(E_i)$ and the standard deviation of the mean $s(\bar{E})$:

$$\bar{E} = \frac{1}{n} \sum_{i=1}^{n} E_i , \tag{4}$$

$$s(E_i) = \left[\frac{1}{n-1} \sum_{i=1}^{n} \left(E_i - \bar{E} \right)^2 \right]^{1/2} , \tag{5}$$

$$s(\bar{E}) = \frac{s^2(E_i)}{n} , \tag{6}$$

where *n* is the number of measurements; E_i is the value of uranium isotope content obtained by processing the *i*-th spectrum.

Then we calculated the confidence interval for the confidence probability P = 0.95 using the relation:

$$\Delta = \sqrt{(s(\bar{E}) \times t)^2 + \delta^2} , \tag{7}$$

where *t* - is the Student coefficient for a given confidence probability and number of measurements, equal to 2.776, δ - statistical variance of the random variable of the MGAU program.

Control of trace impurities in fuel pellets was performed using the methodology №47:2018 from 28.06.2018 "Methodology for measuring the mass fraction of elements in uranium oxides, metallic uranium and its alloys by inductively coupled plasma mass spectrometry". Measurements are performed using an ELEMENT 2 type mass spectrometer. The technique covers measurement of mass concentration of such elements as uranium, chromium, iron, calcium, molybdenum, tungsten, silicon and vanadium.

The density of fuel pellets was determined by hydrostatic weighing in distilled water. The oxygen coefficient was determined by the calculation method based on weighing powders or UO_2 pellets before and after their calcination at 1000 ºC in a muffle furnace.

INFLUENCE OF TECHNOLOGICAL PARAMETERS FUEL PELLETS PRODUCTION ON THEIR CHARACTERISTICS

Many years of worldwide experience in nuclear fuel fabrication have developed the basic technological methods for producing uranium dioxide powders and fuel pellets using them, as well as methods for their control. Pressing and sintering of pellets are typical technological operations. A decisive role is also played by the preparation of powder materials before pressing, the type and amount of binder, and the method of mixing. Apart from the characteristics of powders themselves, the quality and characteristics of obtained fuel pellets depend on the design of used equipment and peculiarities of the technological process at all its stages.

Uranium dioxide powder with a nominal enrichment of 0.4 wt.% was used to develop the modes of pressing and sintering fuel pellets ^{235}U.

As a result of studies of the influence of the type of organic plasticizer (binder) in the form of polyethylene glycol and calcium stearate on the characteristics of pressed and sintered pellets, the optimal pressure, which provides the integrity and a necessary density of formed pellets was determined. Fuel pellets were formed at a pressure of 2.1-4.1 t/cm^2 (Fig. 3). Pressure exceeding 4 t/cm^3 negatively affects the quality of the pellets due to the appearance of cracks and delamination when pressing them out.

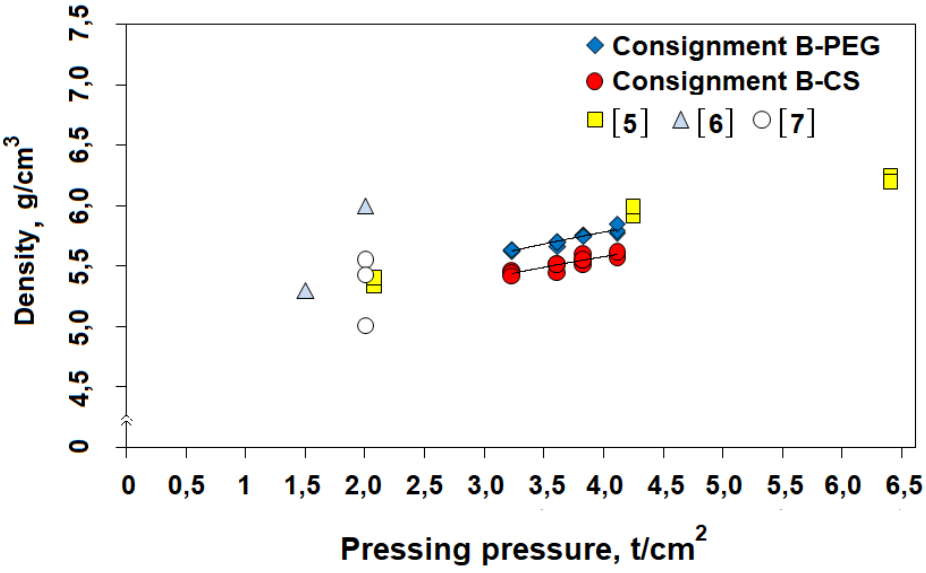


Figure 3. Dependence of the compressed (raw density) pellets on the pressing pressure and type of binder (in [5-7] literature data are given for comparison)

The optimal pressing pressure was 3 t/cm^2, at which two experimental batches of fuel pellets were subsequently pressed: V-CS (15 pcs.) and V-PEG (46 pcs.). The obtained data on the density of compressed tablets are in good agreement with other experimental data [5–8]. Sintered pellets were characterized by uniform shrinkage in height and diameter at the level of 20.2±0.6%, had no chips or cracks. Their appearance is satisfactory, which corresponds to the technical requirements of TR No. 12-1-081 [4] (Fig. 4).

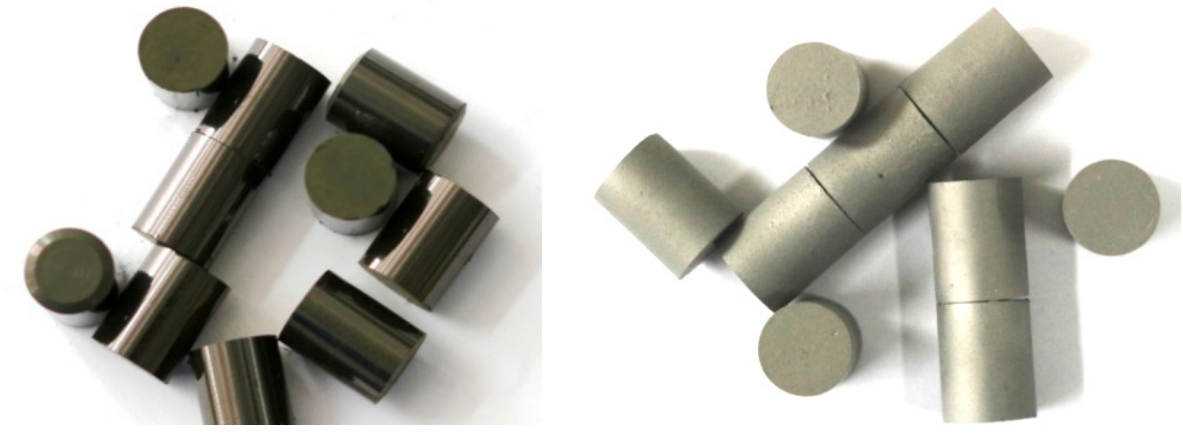


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Figure 4. Appearance of raw (a) and sintered (b) pellets

The results of measuring the geometric dimensions and density of fuel pellets are shown in Fig. 5.

Fuel pellets pressed on a dry binder - calcium stearate (B-CS batch), having an initial density of ~ 5.5 g/cm^3, after sintering have a density of 10.3-10.4 g/cm^3. Tablets pressed on a liquid binder - polyethyleneglycol (B-PEG batch), having an initial density of ~ 5.9 g/cm^3, after sintering have a density of 10.3-10.6 g/cm^3 (Fig. 5 b).

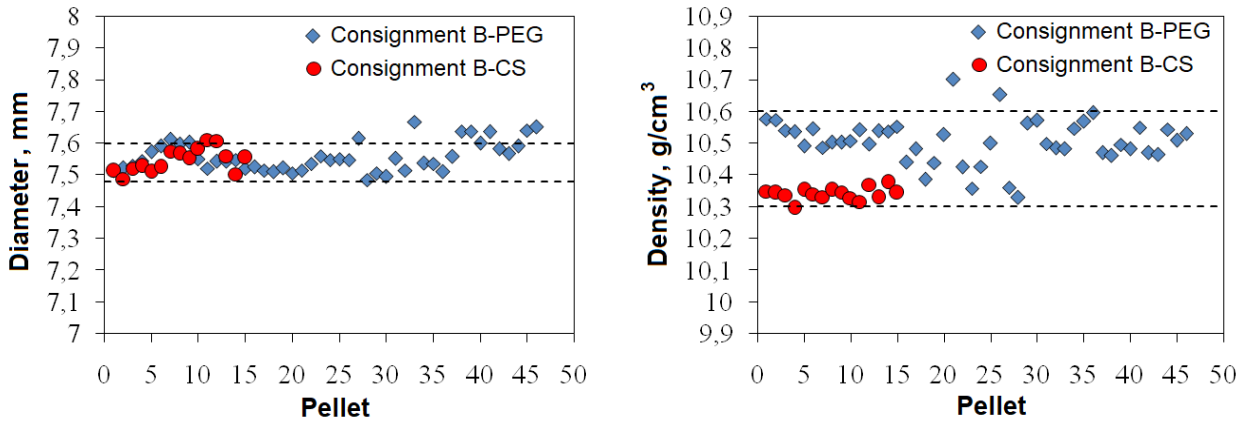

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Figure 5. Diameter (a) and density (b) of sintered pellets UO_2

It follows from the data obtained that a significant part of them meets the technical requirements for diameter (Ø7.54±0.06 mm) and density (10.45±0.15 g/cm^3). It follows from the conducted studies that in order to obtain the maximum density of fuel pellets, a liquid plasticizer, polyethyleneglycol, should be used as a binder. A study of the effect of isothermal holding time of 2 and 4 hours at 1700 ºC in vacuum did not reveal a significant increase in the density of sintered pellets. The average value of the density of tablets sintered at 1700 ºC for 4 hours for the B-PEG batch was 10.5 g/cm^3, and for the B-CS batch – 10.4 g/cm^3. The study of the microstructure of the pellets showed a uniform distribution of pores in their volume, the grain size was 2-7 μm, and the microhardness was 6-7 GPa.

Thus, the studies carried out made it possible to establish the optimal modes for obtaining fuel pellets, as well as to develop and manufacture molds in order to ensure the required tolerance for their diameter. The results of these studies made it possible to proceed to the manufacture of pilot batches of fuel pellets from a mixture of uranium powders with a given enrichment in ^{235}U according to the developed scheme. The results of measuring the ^{235}U content in the initial UO_2 powders were 19.905 ± 0.224 wt.%. and 0.454±0.044 wt.%.

Based on the obtained data, the mass of the initial powders was calculated to obtain a powder mixture with an enrichment of 4.4% wt. by ^{235}U. Figure 6 shows the results of monitoring the homogeneity of the mixtures obtained at different mixing times of the powders.

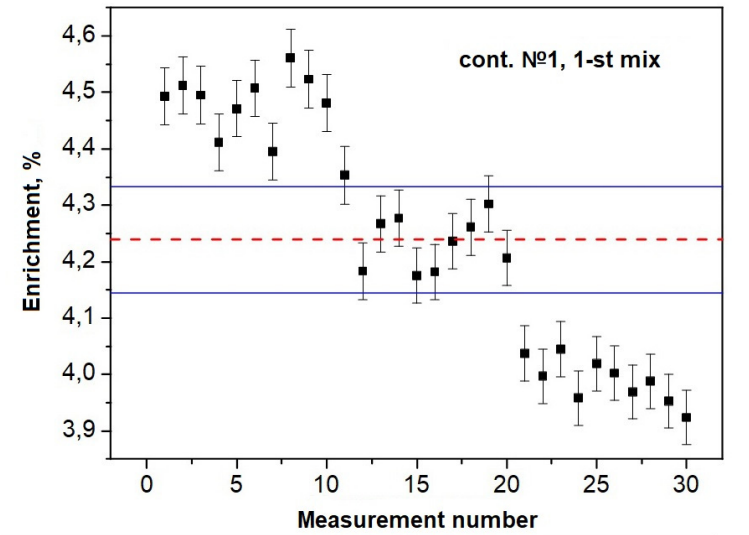

Figure 6. Dependence of the value of the coefficient of heterogeneity and the degree of mixing of UO_2 powders on time

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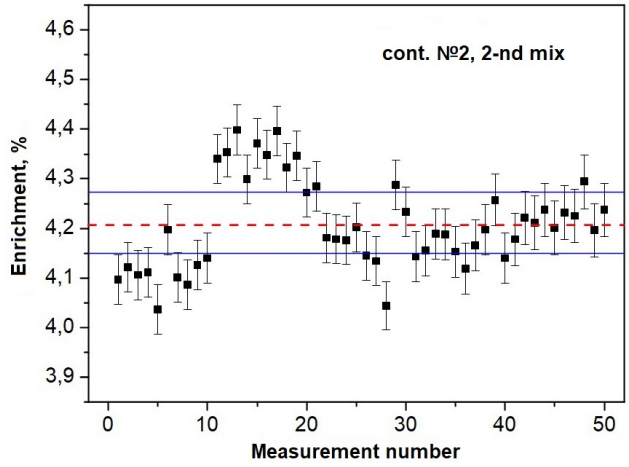
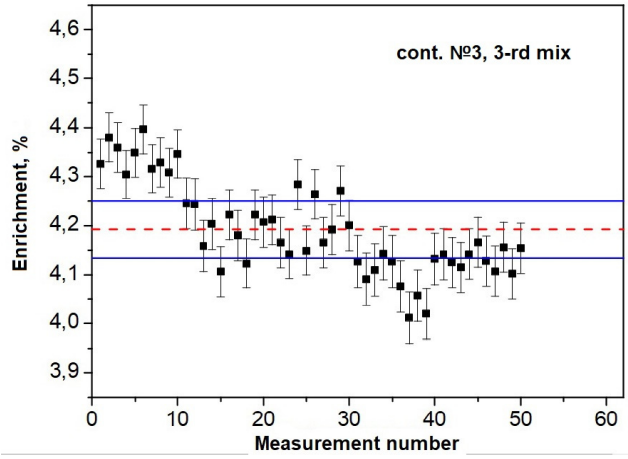


Mixing 3 hour

Heterogeneity coefficient
V_c = 2.14 %
Mixing degree
M=97.86%



Mixing 8 hour

Heterogeneity coefficient
V_c = 2.24 %
Mixing degree
M=97.76%

Figure 6. Dependence of the value of the coefficient of heterogeneity and the degree of mixing of UO_2 powders on time

Thus, in this approach, the criterion for the homogeneity of the obtained mixtures of powders is the maintenance of the condition $\varepsilon \leq 4\ \%$ which will indicate that the possible inhomogeneity of the distribution of the ^{235}U isotope in the mixture is lower than the relative measurement error of the technique used.

Additional measurements of the enrichment of sintered fuel pellets located in the container amounted to 4.382±0.078% wt. by ^{235}U.

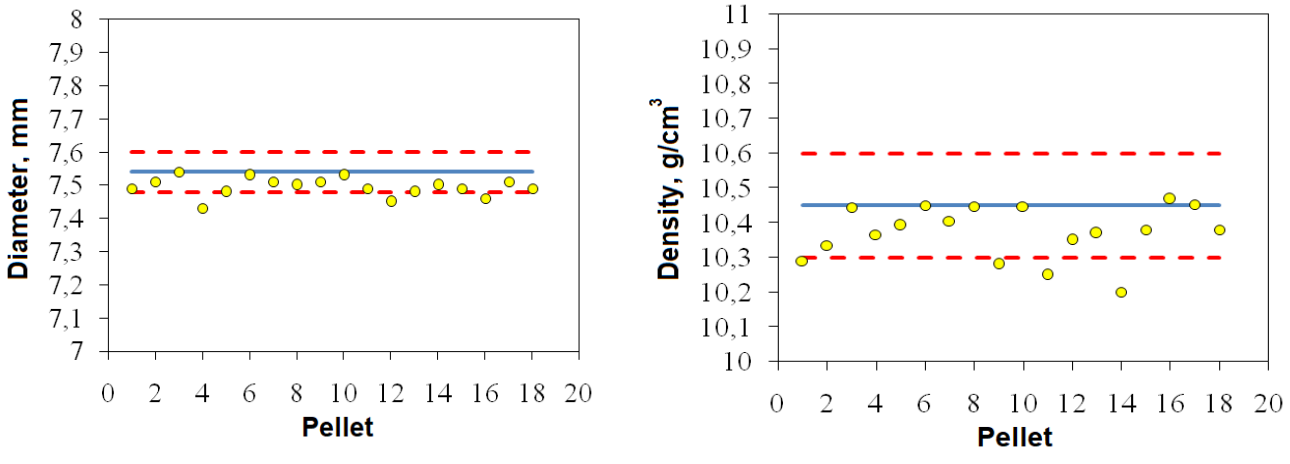


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Figure 7. Diameter (a) and density (b) of sintered fuel pellets UO_2

The pellets after sintering had a diameter of Ø7.48-7.60 mm (Fig. 7a) and a height of 8.6-10.7 mm. The density of sintered pellets was in the range of 10.4-10.6 g/cm^3 (Fig. 7b). The appearance of the pellets is satisfactory, there were no chips on the chamfers, and their microstructure is shown in Fig. 8.

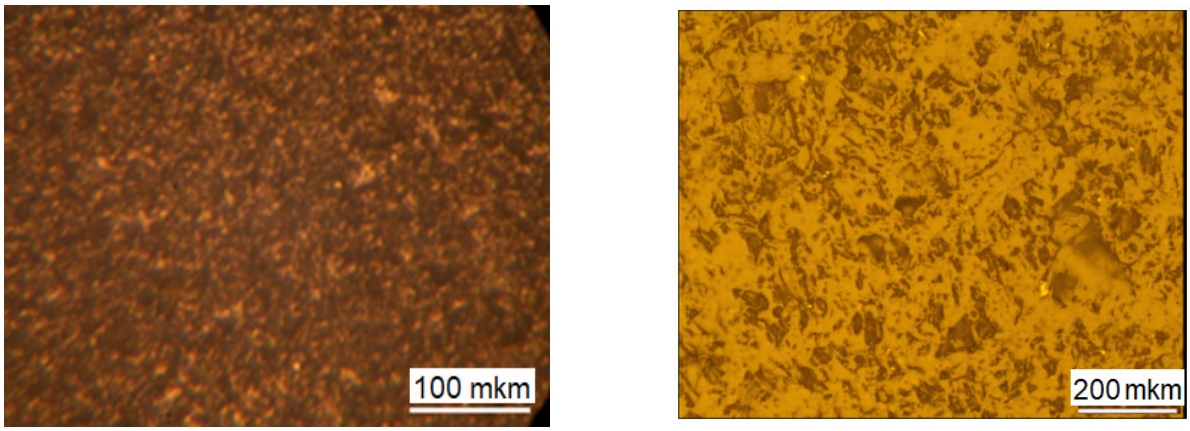

Figure 8. Microstructure of fuel pellets UO_2

The measured enrichment of fuel pellets was 4.395±0.058% weight ^{235}U (Table 1). The additionally measured average fuel enrichment of the experimental FA-X was 4.4±0.18 wt%. ^{235}U.

Table 1. Results of measurements a fuel pellets enrichment

№	Measured enrichment value, *X*	Instrumental measurement error, *d*	Statistical parameter	Value
1	4.392	0.049	Number of measurements, *n*	5.000
2	4.391	0.048	Mean, *Xcp*	4.395
3	4.415	0.048	Standard deviation, *S*	0.026
4	4.355	0.048	Standard deviation of the mean values, *Scp*	0.012
5	4.420	0.049	Student's coefficient for P=95%, *t*	2.776
6	4.392	0.049	Average instrumental error, *dcp*	0.048
			Confidence interval, *D*	0.058

At the measuring of impurities concentration in fuel pellets, it was found that the concentrations of such impurities as iron (Fe), chromium (Cr), molybdenum (Mo), tungsten (W) and others are quite low and do not exceed the limit values established by the ASTM standard [9], as well as the technical requirements of TR No. 12-1-081 [4] (Table 2). The limits of the relative error in measuring the mass contents of the analyzed elements do not exceed ± 20% at P = 0.95.

Table 2. Elemental composition of fuel pellets

Element	Element content, %wt.	
	Requirements № 12-1-081	Measurement
U	≥87.4	87.7
Cr	≤0.01	0.0086
Fe	≤0.03	0.0239
Na	≤0.01	0.0033
Mn	≤0.01	0.0065
Mo	≤0.01	0.0039
W	≤0.01	0.0047
Ca	≤0.015	0.0075
Si	≤0.01	0.0084
V	≤0.01	0.0006

CONCLUSION

Some characteristics of the subcritical assembly of the nuclear facility "Source of Neutrons" are given, as well as a description of the standard fuel assembly of the WWR-M2 type, used during the first fuel loading of the core, and the experimental FA-X, developed for subsequent loads.

A description of the technological scheme for the manufacture of fuel pellets with an enrichment of 4.4% by weight is presented on the ^{235}U isotope by mixing two powders with different uranium content: 0.4% wt. ^{235}U and 19.7% wt. ^{235}U.

The influence of technological regimes and parameters for the manufacture of fuel pellets from the obtained mixtures on their characteristics, such as enrichment, geometric dimensions, density, established in the design and technical documentation, has been studied.

On the basis of the conducted studies, the correctness of the choice of the technological scheme for the manufacture of fuel pellets and fuel rods of FA-X was shown.

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ОСОБЛИВОСТІ ВИГОТОВЛЕННЯ І ХАРАКТЕРИСТИКИ ПАЛИВНИХ ТАБЛЕТОК ІЗ ДІОКСИДУ УРАНУ ДЛЯ СТРИЖНЕВИХ ТВЕЛІВ ПІДКРИТИЧНОЇ ЗБІРКИ

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Досліджено вплив технологічних режимів виготовлення паливних таблеток із діоксиду урану для твелів розроблюваної дослідної тепловидільної збірки ТВЗ-Х, призначеної в якості альтернативного палива для дослідницької ядерної установки (ДЯУ) «Джерело нейтронів, яке керується прискорювачем електронів». На відміну від стандартних технологічних процесів виготовлення таблеток UO_2, особливістю виготовлення даного типу ядерного палива, є одержання порошку діоксиду урану зі збагаченням 4.4% ваг. по ^{235}U шляхом змішування двох партій порошків з різним вмістом урану: 0.4% ваг. ^{235}U і 19.7% ваг. ^{235}U, а також забезпечення необхідного допуску паливних таблеток без застосування операцій механічної обробки. У НТК ЯПЦ ННЦ ХФТІ розроблено комплект конструкторської та технологічної документації, виготовлені експериментальні зразки паливних таблеток, твелів та дослідної тепловидільної збірки ТВЗ-Х, яка спроектована сумісною та взаємозамінною з ТВЗ ВВР-М2, яка прийнята в якості штатного при першому паливному завантаженні в ДЯУ «Джерело нейтронів, яке керується прискорювачем електронів». На відміну від варіанту ТВЗ типу ВВР-М2, що складається з трьох твелів трубчастої форми з дисперсійною паливною композицією UO_2-Al, в ТВЗ-Х розміщені шість твелів стрижневого типу з таблетковим паливом UO_2 в оболонці з цирконієвого сплаву Е110, як найближчого аналога твела енергетичного реактора ВВЕР-1000. Всередині оболонки розташований стовп паливних таблеток висотою 500 мм, який зафіксований від переміщення розпірною втулкою. В основному варіанті ТВЗ-Х паливні таблетки виготовлені з UO_2 зі збагаченням по ^{235}U 4.4% ваг.

Ключові слова: тепловидільний елемент, тепловидільна збірка, порошок, діоксид урану, збагачення, однорідність суміші, щільність, мікроструктура.