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INVESTIGATION OF THE INFLUENCE OF THE INITIAL STATE OF POWDER, PORE-FORMING ADDITIONS, AND U_3O_8 ON THE MICROSTRUCTURE AND STRENGTH OF FUEL PELLETS

I. V. Petrov and V. V. Basov

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It is shown that the initial state of uranium dioxide powder has no effect on the density, microstructure, and strength of pellets. Pore-forming agents and U_3O_8 used in fabrication lower the pellet strength because their particles are not spherical. To increase pellet strength, it is recommended that U_3O_8 be subjected to special processing to spheroidize the particles before mixing for uranium dioxide powder.

Fuel is now used reliably in VVER reactors in a four-year run to burnup 40–50 MW·days/ton. There are plans to use fuel in a five-year cycle with some fuel assemblies retained in the sixth year as well as in a six-year cycle with some fuel assemblies retained in the seventh year, i.e., the plan is to attain average burnup 55–60 MW·days/ton. This requires increasing the quality of the fuel pellets, which is determined by, among other things, the composition, the production process parameters, and the properties of the initial uranium dioxide powder.

Numerous studies have been performed abroad of the effect of the fabrication technology on the density and microstructure of the fuel pellets (shape and size distribution of the pores, grain size, and others) [1, 2], additions of various additives and other compounds [3], sintering in an oxidizing medium [4], and the dependence of the mechanical characteristics of the pellets on their microstructure [5].

In light of this, investigations of the initial state of uranium dioxide powder, process parameters, pore-forming additions, and U_3O_8 on the microstructure (average grain size, shape and character of the pore distribution), density and mechanical strength of the fabricated fuel pellets have been undertaken at the Machine Building Works, which manufactures ceramic nuclear fuel.

Three types of powder were used for the experiments:

1) powder fabricated by the water method by means of thermal decomposition of hydrolysis-obtained uranium hexafluoride and uranium salts, precipitated from water solutions followed by hydrogen-reduction of the higher oxides of uranium to dioxide;

2) powder fabricated by the gas-flame method, in which gaseous hexafluoride is converted into oxyfluorides in an air-water flame in a special reaction apparatus followed by defluorination and reduction of the uranium oxides at 680°C in an atmosphere consisting of hydrogen and water vapor in a rotating furnace; and

3) powder obtained by dry conversion, where gaseous hexafluoride is converted by water vapor superheated to 650°C in a special fluidized bed apparatus into UO_2F_2 and then defluorinated at high temperature and reduced by a steam-hydrogen mixture to uranium dioxide.

The properties of these powders were close and satisfied the technical conditions: total specific surface area 2.5–2.52 m^2/g, bulk density with compaction by shaking 2.26–2.28 g/cm^3, and average particle size 0.48–0.5 μm. The pore-form-

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TABLE 1. Qualitative Characteristics of Fuel Pellets

Pellet batch No.	Powder production method	Additive presence	Average pellet density, g/cm^3	Average grain size, μm	Average pore size, μm	Pore shape factor	Strength, MPa
1	Water	No additives	10.61	12.4	4.81	3.7	148.3
2	Gas-flame	»	10.61	12.5	4.8	3.6	148
3	Dry conversion	»	10.61	12.5	4.81	3.6	148
4	Water	Pore-forming additive	10.3	12.3	8.42	6.3	99.3
5	Gas-flame	»	10.31	12.4	8.41	6.3	99.2
6	Dry conversion	»	10.3	12.4	8.42	6.4	99.2
7	Water	U_3O_8 pel added	10.47	12.7	4.85	3.8	148
8	Gas-flame	»	10.48	12.6	4.84	3.7	147.2
9	Dry conversion	»	10.48	12.6	4.84	3.7	147.9
10	Water	U_3O_8 st added	10.48	12.6	5.95	5.9	102.1
11	Gas-flame	»	10.47	12.7	5.98	5.8	101.9
12	Dry conversion	»	10.47	12.7	5.97	5.9	102

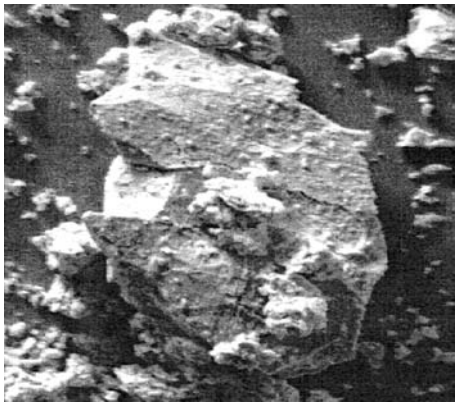

Fig. 1. Particle of untreated U_3O_8 (×2000).

ing agent azodicarbonamide and two forms of U_3O_8 – obtained using the standard technology by oxidation of wastes (U_3O_8 st) and by the method of [6] or additionally worked by pelletizing in a biconical drum (U_3O_8 pel) – were used as additives.

Twelve test batches of fuel pellets were made using a plasticizer in the form of a water solution of polyvinyl alcohol: one batch each using uranium dioxide powder obtained by the three methods indicated above without any additions and with the addition of 1 wt% azodicarbonamide and one batch each with 10 wt% U_3O_8 pel and U_3O_8 st.

To decrease the influence of random factors, the same regimes were used to prepare the compacting powders and the pellets were formed in the same press under the same pressure (1.7 tons/cm^2), sintered in the same furnace at 1750°C, and then ground on the same machine to average diameter 7.56 mm.

The ratio O/U [7], which satisfied the requirements given in the technical documentation and remained in the interval 2.002–2.007, and the density [8] were determined for the pellets in all batches. The room-temperature mechanical strength of

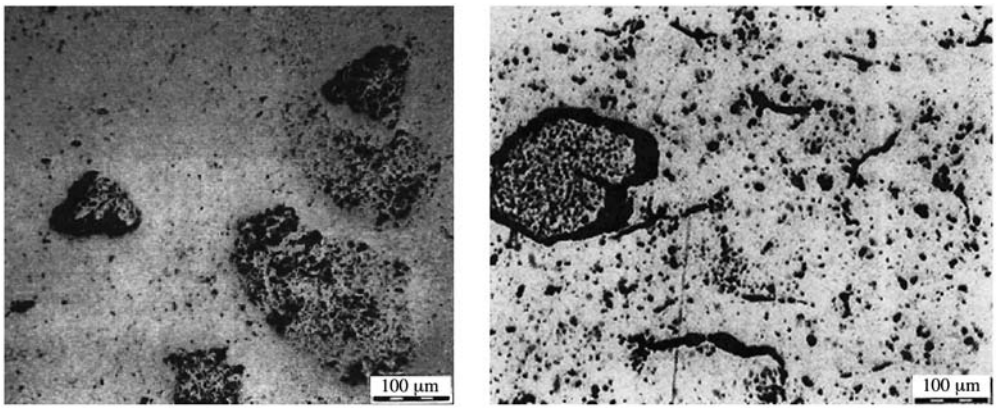

Fig. 2. Characteristic porous sections in the microstructure of pellets obtained from uranium dioxide powder with U_3O_8 addition.

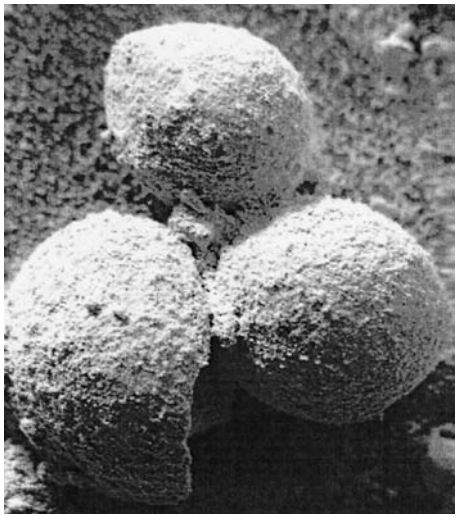

Fig. 3. U_3O_8 particle after additional working (×700).

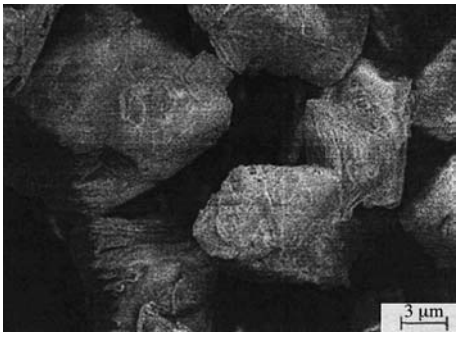

Fig. 4. Particles of the pore-forming agent.

the pellets was determined by crushing in a hydraulic press, following the procedure RA 0335 of the Siemens Company (Germany). Ten pellets were placed inside a shell on a bearing plate. The top pellet protruded 2–3 mm above the top edge of the shell. Then a punch was used to apply a load until an audible crack appeared and the applied force at this moment was recorded. The strength of the pellets in each batch was evaluated according to the average values for 10 repeated measurements [5].

It should be noted that inserting into the technical documentation requirements for the room-temperature mechanical strength of the pellets is probably insufficient because in so doing the brittleness of the pellets, which has a negative effect on fuel behavior in a reactor, is not regulated.

After each crushing test, the microstructure of crushed pellets and two uncrushed pellets were analyzed, the density, porosity, and conventional average grain size were determined, and the shape coefficient of the largest pores, which characterizes the deviation from sphericity, and the average pore size were calculated [8, 9].

According to the results obtained (see Table 1), the uranium dioxide powder, which satisfied the technical conditions, did not affect the density, the characteristics of the microstructure, or the strength of the sintered fuel pellets (the data for batches Nos. 1–3). At the same time, when additions of U_3O_8 st were made, characteristic porous sections shaped just like the particles introduced (Fig. 1) appeared in the microstructure of the pellets (batches Nos. 11–12). The crack embryos observed in the process explain the observed decrease of the pellet strength (Fig. 2). The more the shape of the U_3O_8 particles differs from a sphere (Fig. 3), the lower the mechanical strength of the pellets (batches Nos. 10–12). This also concerns pellets containing azodicarbonamide additions (Fig. 4) (batches Nos. 4–6). The negative effect of the U_3O_8 st additions was surmounted (Nos. 7–9) by changing the technology used to obtain U_3O_8 [6] or by introducing an additional pelleting operation in order to spheroidize the particles of U_3O_8 st obtained by the standard technology using oxidation of production wastes.

In summary, the present investigations allow the following conclusions to be drawn:

1) the initial state of the uranium dioxide powder has no effect on the microstructure, density, or mechanical strength of the fuel pellets which are fabricated under identical technological regimes;

2) the introduction of a pore-forming agent and U_3O_8 st results in the appearance, in the microstructure of the pellets, of porous sections with crack embryos and decreases the mechanical strength in crushing tests at room temperature; and

3) spheroidization of U_3O_8 particles by the new technology or by introduction of an operation to pelletize U_3O_8 st in addition to the standard technology gives the required microstructural parameters and strength, equal to the strength of uranium dioxide pellets with no U_3O_8 st additions.

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