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Sintering of Industrial Uranium Dioxide Pellets Using Microwave Radiation for Nuclear Fuel Fabrication

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Abstract: In this study, the possibility of sintering industrial pressed uranium dioxide pellets using microwave radiation for the production of nuclear fuel is shown. As a result, the conditions for sintering pellets in an experimental microwave oven (power 2.9 kW, frequency 2.45 GHz) were chosen to ensure that the characteristics of the resulting fuel pellets meet the regulatory requirements for ceramic nuclear fuel, including the following: a density of about 10.44 g/cm^3; a volume fraction of open pores of tablets of about 0.1%; an oxygen coefficient of no more than 2.002; hydrogen content of about 0.30 ppm; and the change in density after re-sintering on average no more than 1.16%.

Keywords: microwave radiation; sintering; uranium dioxide; fuel pellets

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1. Introduction

Uranium dioxide is the main nuclear fuel for operating power reactors. At present, ceramic nuclear fuel pellets are made by cold pressing UO_2 powder into pellets with their subsequent sintering into ceramic nuclear fuel in a reducing atmosphere in a multi-section electric resistance furnace (ERF) [1]. The first section of the ERF is a preheating zone, in which the tablets are heated to temperatures of 400–600 °C for 4–6 h. The second section is intended directly for sintering, in which the tablets are heated to 1750 °C and kept at this temperature for 5–6 h. The third section is the cooling zone of the obtained ceramic pellets. The duration of the whole process is from 30 to 35 h. The described technology for the manufacture of fuel pellets is currently widely used in industries both in Russia and abroad. At the same time, the technology also has a number of well-known drawbacks, which are mainly the complex design of the furnace, difficulties in commissioning and decommissioning the furnace, the duration of the process and high energy costs.

Microwave radiation is now increasingly used in thermal processes, accompanying the synthesis of organic compounds, ceramic sintering and other chemical reactions [2,3]. We have previously demonstrated the efficiency of using microwave radiation in the radiochemical technology for obtaining powders of actinide oxides [4–7]. Microwave radiation has a number of unique advantages: fast heating, heating selectivity and significantly lower power consumption.

A very limited number of literature sources report on the study of the possibility of using microwave radiation for sintering UO_2 fuel pellets. The authors have shown the promise of this sintering method by increasing the heating rate and reducing the holding time, thereby significantly reducing the sintering cycle of ceramic pellets. However, in [8], data on the temperature of fuel pellets during sintering were not demonstrated. In addition, the effect of temperature on the characteristics of sintered pellets has not been studied. Another paper [9] also showed the possibility of using microwave radiation for sintering

fuel pellets. However, the resulting tablets had numerous cracks, which indicates an incorrectly selected temperature regime of sintering. It should be noted that in the above work, the authors used hybrid heating of fuel pellets, i.e., microwave energy heated both the fuel pellets and the thermal insulation material (UO_2 powder), thereby heating the pellet material by convective heat supply. In [10], the authors demonstrated a setup for sintering fuel pellets using microwave radiation; the main difference from previous works was the presence of an IR pyrometer in the volume of the furnace resonator, which made it possible to study the temperature regimes of sintering. However, the authors failed to directly heat the tablets above 950 °C, and later they used hybrid heating using current collectors [11].

At the same time, the heating rate was low, and with its increase, tablets with cracks and traces of melting were obtained. In our work [11], the possibility of sintering compressed uranium dioxide pellets with direct high-temperature microwave heating using a laboratory unit based on a household microwave oven with a power of 2.1 kW (Samsung, Seoul, Republic of Korea) was shown. The density of the obtained pellets was 10.40 ± 0.02 g/cm^3, corresponding to the lower limit of the requirements for ceramic fuel pellets. It should be noted that a limited number (usually 1–4) of tablets were used in the mentioned works.

Based on the foregoing, the purpose of this work was to study the possibility of sintering 18 UO_2 pellets (about 100 g) with direct microwave heating of the material, to study the effect of the temperature-time regime of sintering industrial UO_2 pellets produced by the MSZ Machinery Manufacturing Plant, JSC (Elektrostal, Moscow region, Russia) using a pilot microwave oven to analyze the quality of the obtained fuel pellets and study the influence of the geometry of the arrangement of pellets in the volume of the resonator on the characteristics of sintered pellets.

2. Materials and Methods

In the studies, we used a microwave-oven-based prototype unit manufactured by Enerzi Microwave Systems Pvt. Ltd. (model MH2918-E1-S, Belgaum, India); a photograph of the unit is shown in Figure 1a. The oven included two magnetrons of 1.45 kW each with an average operating frequency of 2450 MHz and a sealed resonator with an inner "hot" zone of 50 × 50 × 50 mm, in which the maximum microwave field strength was achieved. The oven power control range was from 0.155 to 2.9 kW, and power control was carried out in automatic or manual mode. Temperature inside the resonator in the range of 350 to 1800 °C was measured by a built-in IR pyrometer. In the experiments, we used up to 18 precompressed ("raw") cylindrical UO_2 pellets with diameters of 9.36 mm and heights of about 13.50 mm, which were placed vertically or horizontally on an alundum support plate. Then, this support plate with pellets was placed in a thermoprotective container whose carcass was made of lightweight fiber ceramic composite (LFCC, GNIIKhTEOS, Russia). The container was installed in the resonator in such a way that the viewing axis of the IR pyrometer coincided with the hole on its upper horizontal wall. In the resonator, air was evacuated through the corresponding valves, and a gas reducing mixture of argon with 7% hydrogen (TU 2114-002-05015259-97, JSC "Linde Gas Rus") was supplied to exclude oxidation of uranium.

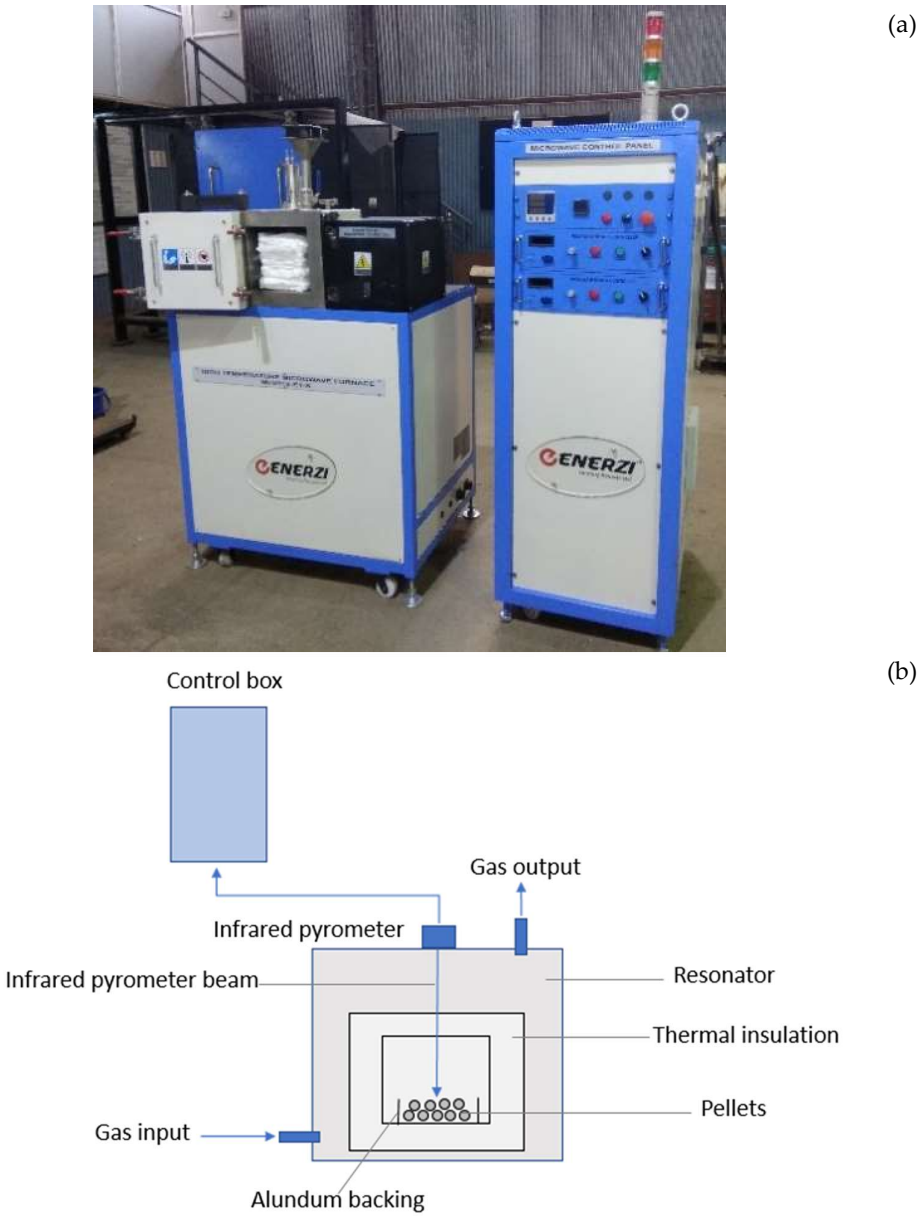


Figure 1. Experimental microwave oven for sintering fuel pellets (model MH2918-E1-S, manufacturer—Enerzi Microwave Systems Pvt. Ltd., Belgaum, India) (**a**) and schematic layout of heat insulation and pellets inside the oven resonator (**b**).

The density and porosity of sintered pellets were determined by hydrostatic weighing (Sartorius analytical scales, Germany). The microstructure of the pellets was studied with inverted metallographic microscope Olympus GX51 (Tokyo, Japan). The oxygen coefficient was determined by measuring the uranium (VI) content using the polarographic method. Determination of hydrogen content was carried out by heating the pellet to 1800 °C in hydrogen analyzer LECO RHTM-602 (LECO Instrumente, Plzen, Czech Republic). Re-sintering of the tablets was evaluated by the change in density during exposure at 1720 °C for 24 h in an argon–hydrogen environment.

3. Results and Discussion

3.1. Selection of the Microwave Sintering Mode for Fuel Pellets

The dependence of the characteristics of the pellets obtained by sintering using microwave radiation on the exposure time of the tablets at 1600 °C was investigated. In initial experiments, it was found that the pellets obtained by relatively rapid heating from

350 to 1600 °C at a rate of about 8.0 °C/min had visible cracks. For this reason, the two modes shown in Figure 2 with heating rates of no more than 4.0–5.0 °C/min were chosen as working modes.

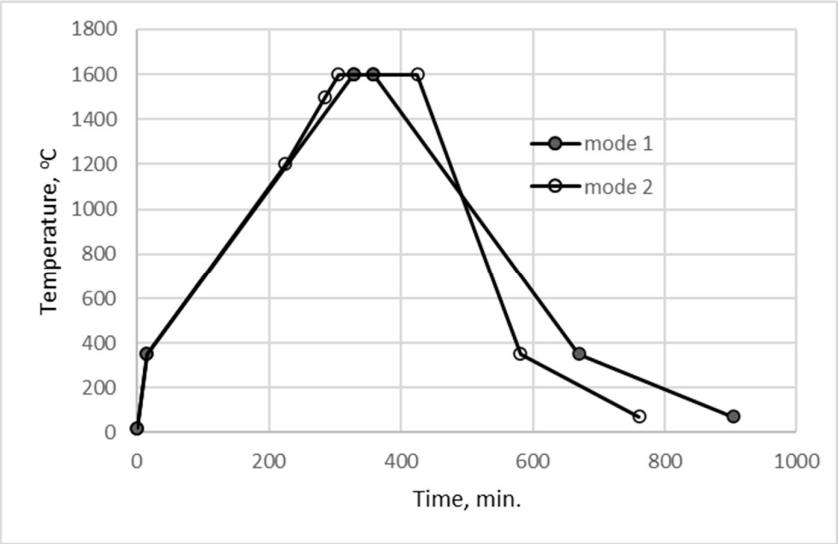


Figure 2. Sintering modes of uranium dioxide pellets on the pilot plant.

In mode 1 (Figure 2), the heating of pressed UO_2 pellets from 350 to 1600 °C and cooling from 1600 to 350 °C were performed at the same rates, 4.0 °C/min. The duration of exposure at 1600 °C was 30 min. Cooling of tablets from 350 to ~70 °C at a rate of about 1.2 °C/min was carried out in a switched-off furnace with a supply of argon–hydrogen mixture. As a result, it turned out that some pellets had microcracks and traces of melting. Their density averaged 10.40 g/cm^3, which confirmed the data of [12]. It was noted that the diameter of the obtained pellets was 7.83 ± 0.03 mm (i.e., shrinkage by pellet diameter was on average 16.3%), which exceeded the diameter of standard industrial pellets—about 7.6–7.7 mm. The volume fraction of open pores of the pellets was 0.45 ± 0.27% and met the existing requirements for fuel pellets (not more than 1%).

When sintering in mode 2 (Figure 2), pellets were heated from 350 to 1600 °C at a rate of 5.0 °C/min, their exposure at 1600 °C was carried out for 120 min, cooling to 350 °C was conducted at a rate of 8 °C/min, and further cooling to 70 °C was achieved at a rate of ~1.6 °C/min. It was noted that some of the obtained pellets also had microcracks and traces of melting. At the same time, their density increased to 10.44 ± 0.01 g/cm^3, and the volume fraction of open pores in the pellets decreased to 0.20 ± 0.13%. Thus, mode 2 was selected as the basic mode for subsequent studies.

A series of sintering was carried out in the selected mode 2. In all cases, it was confirmed that the production of pellets met the technical requirements. The selected sintering mode of the pellets with an exposure time of 2 h at 1600 °C allowed to steadily obtain tablets with the required density and open porosity. However, as noted above, some pellets had surface defects—small cracks and melting in some places of the surface of the pellets (examples in Figure 3). The assumption was made that such defects are formed due to local overheating of the tablet surface. It is likely that local overheating occurred with insufficient thermal conductivity, with a significant increase in absorption of microwave radiation in the oxide materials with increasing temperature, as well as in the case of electrical discharges (breakdown) between neighboring pellets, between which during sintering a gap was formed due to shrinkage of the pellets. Based on the above, it was necessary to select such an arrangement of pellets in the alundum support plate, located in the resonator volume, to exclude the observed effect.

Figure 3. Fuel pellets obtained by microwave sintering in the vertical position.

3.2. Selection of the Geometry of the Placement of "Raw" Tablets in the Substrate during Sintering in a Pilot Microwave Setup

It was suggested that the surface quality of the pellets could be improved by placing the tablets in layers in the support plate while maintaining contact between them (Figure 4). In the experiment, 18 pressed tablets were placed horizontally in a boat plate in two layers so that contact between the pellets was maintained during the sintering process when the pellets shrank.

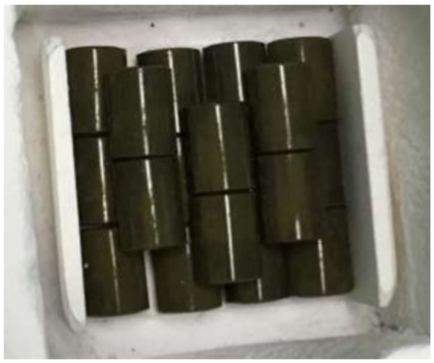


Figure 4. Location of fuel pellets in the support plate in the horizontal position.

The pellets were sintered in the selected mode 2; Figure 5 shows photographs of the sintered pellets. It was noted that the obtained pellets were not sintered to each other, and most of them, 14 out of 18 pellets, showed no traces of melting, except for 4 neighboring pellets on the edge of the plate (the red zone in Figure 5). Obviously, due to the shrinkage of the pellets, a small gap was formed between them, which could lead to an electric discharge and partial melting of their surface. The density of all obtained pellets was in the range of 10.42–10.46 g/cm^3, and their open porosity was not more than 0.1%.

Figure 5. Fuel pellets obtained by microwave sintering in the horizontal position (the red circle—arrangement of tablets with traces of melting).

Thus, it was found that during microwave sintering of pellets it is necessary to place them so that the contact between the tablets is kept. This is possible by stacking the pellets in layers or, as well as in bulk, in a boat plate. In addition, it is advisable to use a substrate with the bottom angled so that during sintering the tablets move lower due to gravity, touching each other, and thus the contact between them remains.

3.3. Microstructure, Chemical Composition and Re-Sintering of Ceramic Fuel Pellets Produced Using Microwave Radiation

When analyzing the microstructure of the obtained ceramic pellets, it was found that the pellets sintered in the vertical position had areas of large and small grains with average sizes of 30.9 (Figure 6a) and 5.7 µm (Figure 6b), respectively (light and dark areas, respectively, in Figure 6c). Differences in grain size may be due to differences in the temperatures reached in the individual fragments of the pellet. Temperature distribution on the surface and in the volume of pellets during high-temperature microwave heating is quite complicated. It is necessary to consider the uneven distribution of heat release during microwave heating in the presence of the insulator and support plate, heat loss due to thermal radiation from the surface of the pellets, and the thermal conductivity of both tablets and the support plate. As a result, different areas of the tablets reach a given temperature at different times. The same effects can lead to overheating of individual pellets or their parts. It is likely that in the lower part of the tablet an increase in temperature relative to the set sintering temperature occurred (1600 °C). Another reason could be the diffusion of aluminum from the alundum plate, the incorporation of which in the fuel also leads to an increase in grain size. The elucidation of the reason for the difference in grain size in the pellets requires further investigation. It was also noted that the investigated pellet had microcracks (Figure 6d), the total size of the cracks being 545 µm. The diameter of the maximum projection (Feret diameter) of an individual pore in the thin section field was 375 µm, and the quantitative proportion of pores with effective diameters from 1 to 10 µm in the plane of the thin section was about 98%.

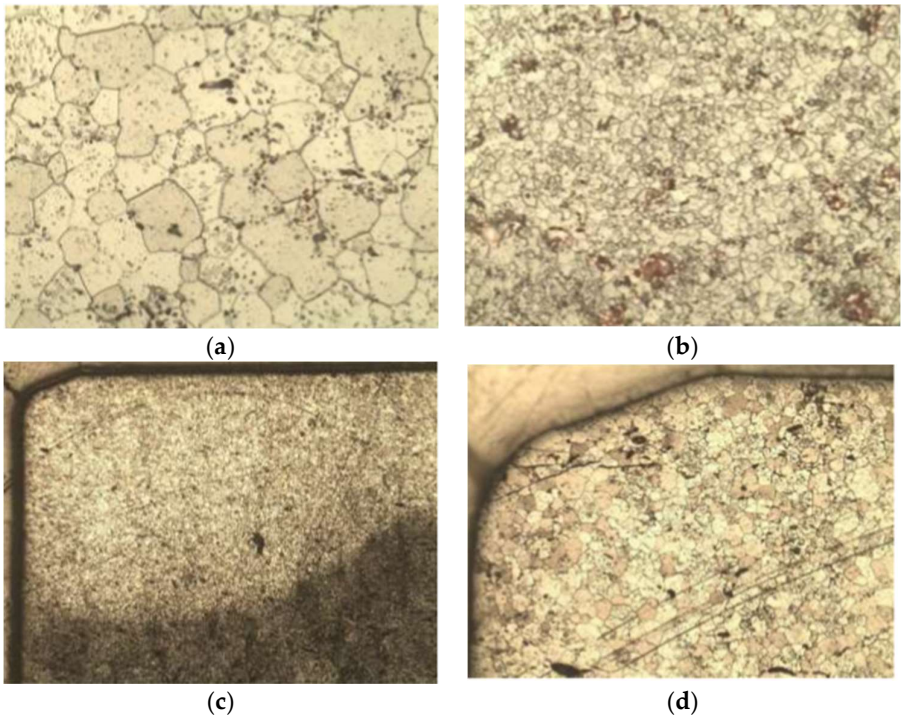
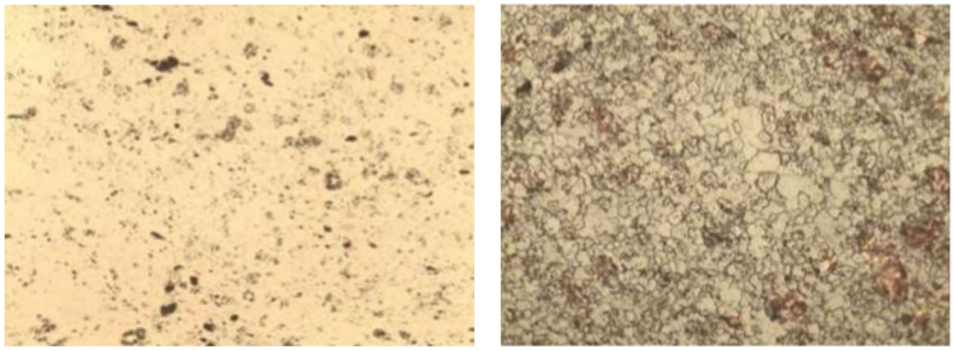


Figure 6. Photos of microstructure of pellet sintered in vertical position (1600 °C, 120 min), at different magnifications: (**a**) grain, lower part of the pellet, 500×; (**b**) grain, upper part of the pellet, 500×; (**c**) thin section after etching, 25×; (**d**) delamination, 100×.

Figure 7 shows that in tablets sintered horizontally, the grain size distribution is homogeneous, which confirms the uniformity of the temperature achieved during sintering. The average grain size in the tablets was about 8–9 µm. Obviously, it is possible to increase the grain size by increasing the holding time at 1600 °C or by increasing the temperature. The maximum diameter of a single pore in the thin section field of pellets

sintered horizontally was 139 µm, and the quantitative proportion of pores with effective diameters from 1 to 10 µm in the thin section plane was about 96%.



(**a**) (**b**)

Figure 7. Photo microstructure of pellet obtained by sintering in horizontal position (1600 °C, 120 min), at different magnifications: (**a**) grain of pellet, 500×; (**b**) porosity of pellet, 10×.

The data on the oxygen and hydrogen content in the pellets obtained in the vertical and horizontal position in the microwave resonator of the oven are shown in Table 1. As can be seen from the above data, the oxygen ratio was not more than 2.0024, and the mass fraction of hydrogen was about 0.30 ppm.

Table 1. Oxygen coefficient and hydrogen content in pellets obtained in vertical and horizontal position during microwave sintering.

Parameters	Position of Pellets During Microwave Sintering		*Parameter Requirements*
	Vertical	Horizontal	
Oxygen coefficient	2.0007	2.0024	*2.00–2.01*
Hydrogen concentration, ppm	0.29	0.35	*Not more than 0.60*

The results of the pellet re-sintering study are shown in Table 2. From Table 2, it can be seen that the probability of re-sintered pellets increased by an average of no more than 1.1%, which corresponds to a maximum limit—no more than 1.3%. At the same time, it was noted that the density of some finished pellets was higher than this limit, which confirms the conclusion that the temperature or exposure time in the selected mode should be increased.

Table 2. Re-sintering of ceramic pellets.

Density of Sintered Pellets, g/cm^3	Density of Pellets after Re-Sintering, g/cm^3	Change in Density, %
10.42	10.49	0.7
10.52	10.57	1.1

4. Conclusions

The possibility of sintering the main fuel pellets with direct microwave heating without the use of current collectors was established. Tablets were obtained during the work. In the work, important optimal temperature and time parameters for the sintering cycle of UO_2 fuel pellets were found at which fuel pellets without cracks and melting were obtained. It was found that the geometry of placing pellets in the resonator volume has an effect on the microstructure of sintered pellets, and this fact allows us to significantly advance research in this area.

It should be noted that the proposed technology can be especially effective in the manufacture of regenerable mixed fuel (REMIX fuel, 1.5%wt ^{239}Pu) for fast neutron reactors.

It is known that this stage is subject to strict requirements for physical personnel, so the REMIX fuel factory can be organized remotely in protective chambers and with remote maintenance. The proposed method of microwave preparation of fuel pellets requires pilot testing.

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