The Way Of Nitride Fuel Producing By High Voltage Electro-Discharge Compaction

M.S. Tarasova, B.A. Tarasov, E.G. Grigoryev, D.P. Shornikov

Russia, Moscow, National Research Nuclear University "MEPhI"

yurmary@mephist.ru

The way of nitride fuel producing by high voltage electro-discharge compaction are demonstrated. General features of the parameters influence of the of high-voltage electric pulse consolidation on density of uranium nitride compacts are obtained. The main advantage of this method is the small sintering time and low processes temperature, which affect both the quality of the final product (preservation of the original phase composition) and the energy consumption during the manufacture of the product. The peculiarities of formation of macro and microstructure, as well as the phase composition of the samples are discussed.

Uranium-plutonium nitride, fuel element, high-voltage electro-discharge compaction, sintering.

1. Uranium nitride as nuclear fuel

At the Russian Federation till to 2022 it is planned to build a liquid metal fast breeder reactor with low and medium power and partially close nuclear fuel cycle. Facility for fabrication of high-density fuel will be created at the same area with the reactor. As fuel must be used: uranium mononitride UN or mixed mononitride $U_{0,85}Pu_{0,15}N$. The possibility of adding the minor actinides (Am, Cm) to fuel for further burning are considered [1, 2].

Positive aspects of using material are the high density ρ = 14,32 g/cm^3; the high thermo conductivity λ(UN) ~ 25 W/m·K and $\lambda(U_{0,8}Pu_{0,2}N)$ ~ 18 W/m·K; uranium nitride does not interact with many cladding materials (steels).

Limitations of material are: low thermal stability – uranium and plutonium nitride dissociate to nitrogen, and metal beginning from 1700 °C; the evaporation of plutonium (minor actinides) can reach 15 wt. %; complicated technology for fabrication, the possibility of oxidation; the uranium nitride has very high temperature of free sintering (over 1800 °C) [3]. Achievable relative density of UN without applying stress is less than 85% [4].

The main research problems are evaluation and study changes in the physical properties of UN and (U, Pu)N occurring under irradiation for justification performance of fuel elements; thermal stability study [5] under irradiation, as well as consideration of ways to improve it, such as nitride doping and improvement of sintering ability (increase final density products). To solute this problems we needs to study the properties and thermodynamics stability and applicate of field-assisted techniques of powders consolidation (for example high voltage electro-discharge consolidation and spark-plasma sintering).

The main purpose of this study was researching on the applicability of the methods of uranium nitride powder compaction for fabrication fuel pellets with a relative density more than 90%.

2. High voltage electro-discharge compaction HVEDC

In 1979, an “unusual” method to consolidate powders by electro-discharge compaction was proposed. The method is based on passing a high voltage current pulse (up to 30 kV) through a powder with the simultaneous application of external pressure. At a current density of (10^{8}–10^{11}) A/m^{2} and pulse duration of (10^{-3}–10^{-5}) s, the HVEDC method makes possible to obtain high density compacts of a given shape along with higher productivity of the process as compared with other compaction methods, including by reducing the number of technological operations. A great advantage of this approach is the possibility to reject a protective atmosphere or vacuum during sintering. The HVEDC method allows obtaining compacts in air without preliminary degasification of the charge material.

At National Research Nuclear University (MEPhI) it was proposed to use the HVEDC method to obtain compacts from uranium mononitride and other types of nitride fuel.

The schematic of apparatus for HVEDC (Impulse-BM, Russia) are shown in the Figure 1. Power storage unit includes a capacitor bank with a stored energy of 75 kJ, which provides a powerful energy discharge to the powder compact. The total capacitor contains 150 numbers of small capacitors with 30 µF, which allows creating up to 6 kV charge. The apparatus uses a pulsed current generated by the capacitor bank during the time of powder compaction under constant pressure. The main parameters of the process are the applied pressure on compact and the parameters of pulsed current from the battery.

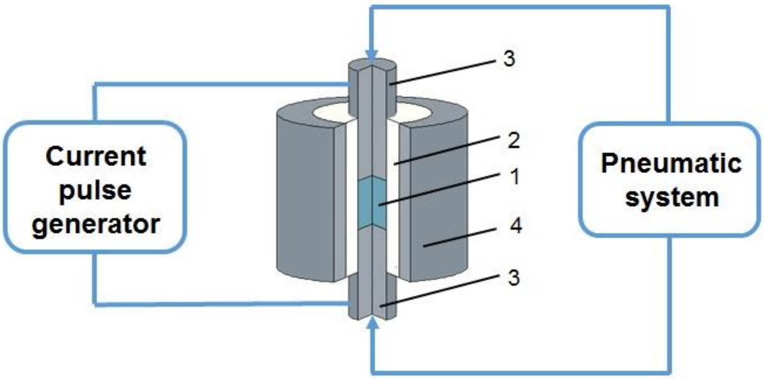


FIG.1. Schematic of the HVEDC apparatus [6]

1 – powder; 2 – ceramic die; 3 – punches; 4 - protective cage

3. Experimental results

We use the powder of uranium nitride produced by hydriding and nitriding of pure metal in laboratory conditions. The uranium nitride powder have a medium particle size of 5-10 µm and frag particle shape (Fig. 3a). XRD data shows mainly cubic phase of uranium mononitride with small peaks of uranium dioxide phase.

The consolidation of UN was carried out at a pressure in the range of 160 to 210 MPa and at a voltage in the range of 2 to 3 kV. Fig. 2 shows the densification patterns of UN describing the powder densification dependence on the pressure and voltage. The density of the specimens increases with the increasing pressure of the pre-pressing stage. High-density material is observed to a certain value of the pressure. Further increase in pressure affects slightly on density in the studied pressure range. In particular, the behavior of the powder densification under pressure are characterized by plastic deformation of the powder particles and by brittle fracture aggregates and particles with a redistribution of the finer fractions in the spaces between the large ones.

As shown in the paper [7], the current density depends on the voltage and hence on the stored energy in the pulse is linear with the voltage. Dependence of the relative density of the specimens from the voltage on the capacitor bank is linear in the investigated range of voltages.

A consequence of the no uniform distribution of the pressure will be that the density of the center is lower and the contact area between the particles is less, which leading to greater electrical resistance, which in turn allows for more Joule heat, and hence the heating to a higher temperature.

Raising the pressure (at constant current) increases the area with the highest density, which make increase in the size of the central zone. Raising the current density (at constant pressure) make more power and heating occurs nearby the most compacted region, which also leads to an increase of the size of the central zone.

When pressing the powder in the tooling, the greatest pressure is applied on the particles close proximity to the punch. With increasing the pressure, the boundaries between the particles is changed. The friction of the particles against each other under the action of pressure causes a significant increase in conductivity of this material as compared to a less dense central part.

Maximum specimen density of UN achieved in this experiment does not exceed 96%, which is limited by the power source. Increasing the current density passing through the powder compact, obviously leads to an increase in the density of compacts. However, beyond a certain critical value, the powder will release a significant amount of voltage through the matrix. Also increasing the voltage on the punches above 4.5-5.0 kV and a pressure above 200 MPa leads to radial cracking and compacts failure.

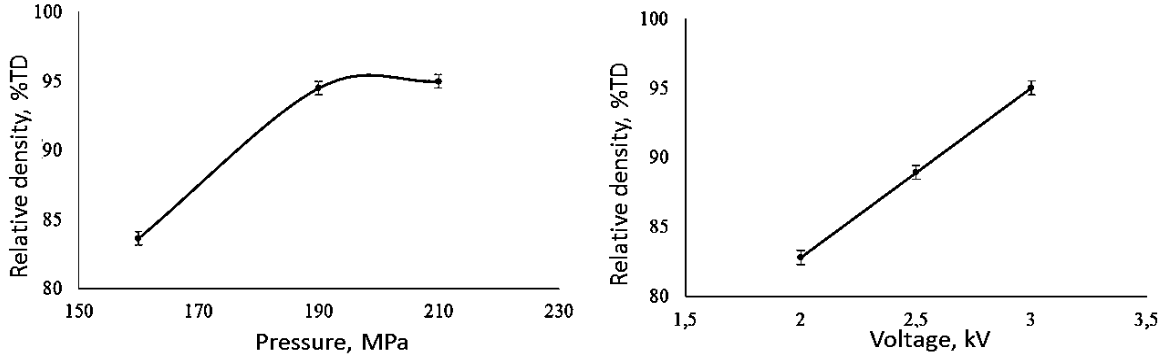
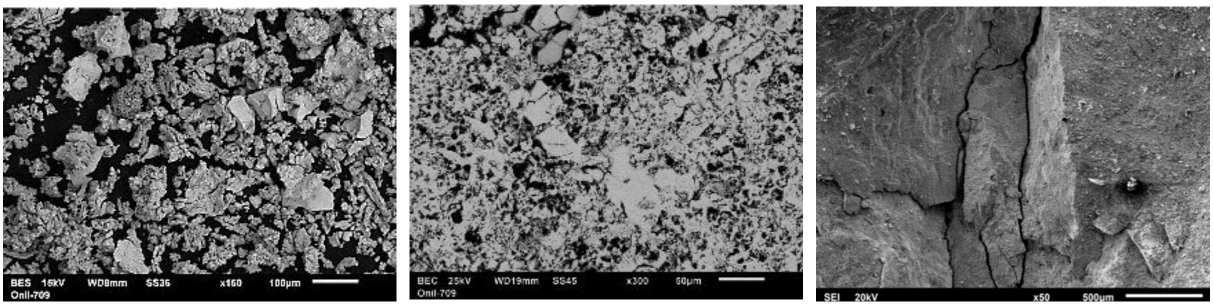


FIG.2. Porosity of uranium nitride pellets dependence from applied pressure (a) and voltage (b)

Fig. 3 shows the typical microstructure of uranium nitride specimens obtained by high-voltage electric pulse consolidation. The SEM images show that the particle size does not exceed the initial values of particles size.



(a) (b) (c)

FIG.3. Microstructure (a) and macrostructure (b) of the consolidated sample and (c) fragment of pellet

To establish the changes in the phase composition after HVEDC the X-ray phase analysis are performed. The results are shown in Figure 4. It is shown that there is no change. The operation to air did not lead to oxidation. Uranium nitride decomposition wasn't observed.

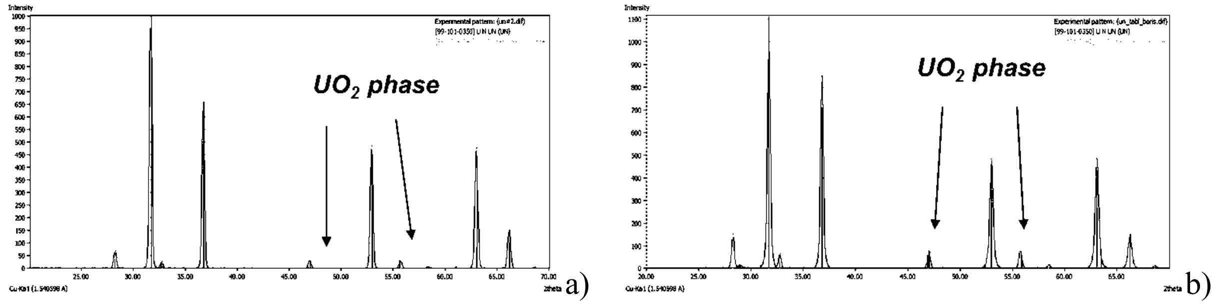


FIG.4. XRD analysis of powder (a) and sintered pellet (b)

HVEDC of model nitride nuclear fuel with plutonium imitator's (CeN, YN) is held at a voltage of 4.0 kV and a pressure of 150 MPa. According to XRD lattice parameter samples (U-10 % Ce)N , and (U-10 % Y) N is 4,905 and 4,899 Å, respectively, (evaporation of Y, Ce is not detected), the pellets achieved a relative density of 90 %.

4. Conclusions

The prospects of high voltage electro-discharge compaction to obtain tablets of powders of actinides nitrides are shown.

General features of the influence high-voltage electric pulse consolidation parameters on the density of uranium nitride compacts are obtained. It is shown that the density of the compact is linearly dependent on the voltage on the capacitor bank (pulse energy) in the investigated range of energies. Dependence of density from the applied pressure is increasing curve with the saturation. Compacts with densities of 85 - 96% are obtained. It is found that the tablets grain structure has the same size as the used powder particles. The phase state of actinide nitride (imitators) remains unchanged during the consolidation process.

[1] STREIT, M. et al. Nitrides as a nuclear fuel option. Journal of the European Ceramic Society 25 (2005) 2687–2692.
[2] KITTEL, J.H. et al. History of fast reactor fuel development. Journal of nuclear materials 204 (1993) 1-13.

[3] OSAKA, M. et al. Research and Development of Minor Actinide containing Fuel and Target in a Future Integrated Closed Cycle System. Journal of Nuclear Science and Technology 44 (2007). 309–3161.

[4] ARAI, Y. Nitride fuel. Nuclear Science and Engineering Directorate. Japan Atomic Energy Agency. Ibaraki. 2012

[5] HEDBERG M., A comparative study of nitride purity and Am fabrication losses in PuN materials by the powder and internal gelation production routes. Journal of Nuclear Materials. 482 (2016) 156-162.

[6] YURLOVA M.S., et al. Electric Pulse Consolidation: An Alternative to Spark Plasma Sintering. Journal of Materials Science 49 (2014). 952-985.

[7] GRIGORYEV E.G., et al. Wave mode high voltage consolidation of powder materials. Computational Materials Science 100 (2015). 8-14.