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Properties of UN Sintered by High Voltage Electric Discharge Consolidation

M. Yurlova*®, B. Tarasov®, D. Shornikov®, E. Grigoryev®, E. Olevsky®,

*National Research Nuclear University MEPhI, Kashirskoye shosse 31, Moscow 115409, Russia
®San Diego State University, 5500 Campanile Dr, 92182, San Diego, USA

Abstract

In the present work, the opportunity of the consolidation of uranium nitride tablets by high voltage electric discharge consolidation (HVEDC) is considered. It is shown that the consolidation by HVEDC allows the prevention of the expansion of uranium nitride powders and renders pellets with relative density of up to 97%. The thermal stability of the obtained samples has been investigated. The analysis of the microstructure of the processed samples indicates the retention of the initial powder structure.

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Keywords: high voltage; consolidation; uranium nitride; nuclear fuel

Introduction

The problem of the creation of a liquid metal fast breeder reactor with low and medium power and partially closed nuclear fuel cycle can be solved by the fabrication of high-density fuel based on uranium mononitride (UN) or on the mixed mononitride $^{235}\text{Pu}_{0.85}\text{U}_{0.15}\text{N}$. The possibility of adding minor actinides (Am, Cm) to fuel for further
burning is considered. Positive aspects of using these materials are their high density \( \rho = 14.32 \text{ g/cm}^3 \); high thermal conductivity \( \lambda(\text{UN}) = 17\text{–}30 \text{ W/m-K} \) and \( \lambda(\text{U}_{0.85}\text{Pu}_{0.15}\text{N}) = 18 \text{ W/m-K} \); and the fact that uranium nitride does not interact with many cladding materials (steels). Limitations on the usage of these materials include their low thermal stability, uranium and plutonium nitride dissociation to nitrogen and the metal at 1700 °C; the evaporation of plutonium (minor actinides) which can reach 15 wt.%; the complicated technology of their fabrication, the possibility of these materials’ oxidation; the fact that uranium nitride has very high temperature of sintering (over 1800 °C). An achievable relative density of UN is less than 85%. The main research problems are the evaluation of the changes in the physical properties of UN (U, Pu)N, occurring under irradiation. The conducted study includes the assessment of the performance of fuel elements; the analysis of their thermal stability under irradiation, as well as the consideration of ways to improve it, such as doping nitrides and improvement of the production technology of the nitride fuel (increase of the final product density). To solve these problems a the study of the applicability of field-assisted techniques of consolidation of powders (for example high voltage electro discharge consolidation and spark-plasma sintering) is carried out. Thus, the main purpose of the present study is the research the utilization of the methods of uranium nitride powder compaction (high voltage electrodischarge consolidation) for the fabrication of fuel pellets with a relative density higher than 90%.

Experimental

The traditional way to obtain the uranium nitride powders is carbothermic reduction. In this method of producing uranium nitride the initial material is uranium dioxide. The carbon and oxygen pollution of powder are occurred. For our study we used the powder of uranium nitride produced by hydriding and nitriding of pure metal. Nitride synthesis was performed in quartz tube horizontally mounted in a resistive furnace. The process was done according to next scheme:

\[
U + H_2 \xrightarrow{250^\circ C} UH_3 \xrightarrow{800^\circ C} \frac{X_2}{U} \frac{N_3}{1300^\circ C \text{ in vacuum}} \rightarrow UN
\]

For consolidation uranium nitride pellets powder of UN Among electromagnetic field assisted powder consolidation techniques it is necessary to allocate so-called High Voltage Electro Discharge Consolidation (HVEDC) technology. HVEDC is based on a single passage of a current pulse duration of less than millisecond have special place in this field [1]. A single powerful pulse of current flows through the source powder, which leads to a large electric resistance, energy transfer inside of powders, and this energy is sufficient for sintering of a variety of materials. Due to the short sintering time, substantial densifications occur with minimal changes in the microstructure, which are desirable for high mechanical strength. The high voltage electro discharge method of consolidation (HVEDC) which is based on a single passage of a current pulse duration of less than ms have special place in this field. Relative density of products can reach 95-97%. Advantages of method fuel pellet may be compacting less than 1 min. Disadvantages of method is possibility of oxidation, as the process is carried out on air.

High voltage compaction was provided by “Impulse-BM” device (“Potok” LCC, Russia) which can provide electric discharge through the specimen with a voltage up to 6 kV and pressure up to 300 MPa. Consolidation was held in an atmosphere.

Results and Discussion

The uranium nitride powder have a medium particle size 5-10 \( \mu \text{m} \) and juhfytty particle shape (fig. 2a). 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. 1 shows the densification map of ZrN 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 premolding density obtained specimens in the pressure range studied. In particular, the behavior of the powder densification under pressure are not 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 [2], the current density depends on the voltage and hence the stored energy in the pulse is linear with the voltage. Dependence of the relative density of the specimens studied compositions of the voltage on the capacitor bank is linear in the investigated range of voltages.

Maximum specimen density of UN achieved in this experiment does not exceed 96%, which is limited by the power plant. 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 failure compacts.

![Graphs showing porosity of uranium nitride pellets dependence from applied pressure and voltage.]

**Fig. 1.** Porosity of uranium nitride pellets dependence from applied pressure (a) and voltage (b).

Fig. 2 shows the typical microstructure of zirconium nitride specimens obtained by high-voltage electric pulse consolidation. The SEM images show that the particle size does not exceed the initial values of particle size powders. HVEDC of model nitride fuel with plutonium imitators nitride (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%.

![Microstructure and macrostructure images of the consolidated sample.]

**Fig. 2.** Microstructure (a) and macrostructure (b) of the consolidated sample.
Conclusions

Consolidation of uranium nitride powders by high voltage electric discharge sintering has been successfully conducted. It was shown, that there is a possibility to obtain dense, with needed porosity and structure UN pellets very quickly. Pellets after HVEDC have the good thermostability. The HVEDC is a promising method for the production of tablets toplinyh uranium nitride.

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References