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Imitators of plutonium and americium in a mixed uranium-plutonium nitride fuel

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Abstract. Uranium nitride and mix uranium nitride (U-Pu)N is most popular nuclear fuel for Russian Fast Breeder Reactor. The works in hot cells associated with the radiation exposure of personnel and methodological difficulties. To know the main physical-chemical properties of uranium-plutonium nitride it necessary research to hot cells. In this paper, based on an assessment of physicochemical and thermodynamic properties of selected simulators Pu and Am. Analogues of Pu is are Ce and Y, and analogues Am - Dy. The technique of obtaining a model nitride fuel based on lanthanides nitrides and UN. Hydrogenation-dehydrogenation-nitration method of derived powders nitrides uranium, cerium, yttrium and dysprosium, held their mixing, pressing and sintering, the samples obtained model nitride fuel with plutonium and americium imitation. According to the results of structural studies have shown that all the samples are solid solution nitrides rare earth (REE) elements in UN.

1. Introduction

At this time discusses the use of mixed uranium-plutonium nitride (MUPN) as a fuel for fast reactors and addition to fuel minor actinides (Am, Cm, Cf). The key problem of this research is to expand knowledge of the changes of occurring in nitride fuel with the introduction of plutonium and minor actinides (MA), as well as during reactor irradiation. When the main volume of specified data is only available during reactor irradiation and subsequent studies in hot cells, which in turn is associated with a number of technological and methodological difficulties. The study of mixed uranium-plutonium fuel containing MA, also faced with the need to work in heavy protective box that greatly complicates and expensive experiments. Preliminary results (phase composition, thermal properties, etc.) can be obtained based on surrogate-materials in which radioactive elements (Pu, Am, etc.) are replaced by elements-imitators. This method is widely used for research oxide fuel [1, 2]. Therefore, the aim of this work is the rationale for the choice of imitators nitride plutonium and americium nitride.

2. Method of creation model nitride fuel

Metallic uranium initially hydrogenated at a temperature of 250-270 ° C to obtain a powder with a high active surface area that reacts with nitrogen at a faster rate than the compact metal [3]. Then uranium hydride dehydrogenation is conducted at a temperature (430-450) ° C resulting in the evolution of hydrogen gas, the destruction of the compact metal and fragmentation it into powder in its reiteration "hydrogenation - dehydrogenation" several times leads to reduction of the specific surface of the powder. Empirically it found that 3 cycles "hydrogenation - dehydrogenation" is enough to produce a powder grain size of about 20 microns. The reaction of uranium hydride and a powder of


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uranium to nitrogen is carried out in the temperature range 500-800 ° C for 2 hours. The resulting U_2N_3 decomposed in a vacuum at a temperature of about 1100 ° C. Uranium nitride has a relatively low oxygen content (100-200) ppm and the carbon (less than 100 ppm), which usually depends entirely upon the purity of the metal used. It has also been found possible hydrogenation not in hydrogen, but in argon-hydrogen mixture, in this case, due to the reduced content of hydrogen in the mixture of 9% and hence lower the partial pressure of hydrogen is necessary to raise the temperature to the hydrogenation reaction (500-600) ° C, in turn, in the dehydrogenation as necessary to reduce the temperature to 100 °C, and the process carried out at a diffusion pump for pumping within (30-40) min. At the same time, marked advantages of this method: a nitride obtained in the form of larger chips rather than powder with a characteristic gray-steel surface, the purity of the obtained product is slightly higher purity powder of size 20 microns. After receiving it mononitride uranium milled in an agate mortar, the powder particle size is about 50 microns. Nitrides of cerium and dysprosium are obtained in a similar way. Then, powders of nitrides of uranium and lanthanides is mixed in a vortex mixer was filled in a mold and compressed with a force of 200 MPa. The obtained compact was unloaded from the box into a molybdenum boat and sintered in a furnace at 1600 °C for 5 hours to a density of (80-95)% of the theoretical.

The microstructure nitride modeling performed by the scanning electron microscope JEOL 6610 LV, definition of uniformity distribution plutonium-imitators was determined by EDX microanalysis consoles.

In addition to scanning electron microscopy as part of this work carried out X-ray phase analysis of powders modeling nuclear fuel. The analysis was carried out in the X-ray camera RKD (X-ray Debay camera) diameter of 57.3 mm, recording the interference pattern was performed on a cylindrical film while using normal (balanced) and asymmetrical methods of charging the film. Radiation used copper Kα. Despite the presence of more modern methods of X-ray analysis, we used the powder method with the registration of the interference maxima in the film on the grounds that the method makes it possible to equip the samples with little or no risk of oxidation, as well as working with a very small sample weight.

3. Selecting simulators Pu and Am in nitride nuclear fuel

According to some literature data, such as replacement of the lanthanides may be considered. This is due to the fact that both the lanthanide and actinide series in the filling of the electrons is not the outer but the inner shell *5f*, and according to the theory Seaborg lanthanides are physico-chemical analogues actinides.

One of the key parameters that determine many properties of the compound, and is the type of lattice parameter. All the nitrides of rare earth metals, including scandium and yttrium, crystallize in the face-centered cubic lattice of the NaCl type and correspond to the composition MeN. The phases of other compounds in the rare earth metal oxide is not observed [4]. Also for lanthanide nitrides well-known pattern, is to reduce the lattice parameter with increasing atomic number of the element, the so-called effect of "lanthanide contraction."

The lattice parameters of lanthanide nitrides generally much larger lattice parameters actinide nitrides. There are four lanthanide nitrides, which have values of the lattice parameter, next to the nitrides of actinides - Ce, Dy, Gd, Y, so in the future we will stop its attention on these elements.

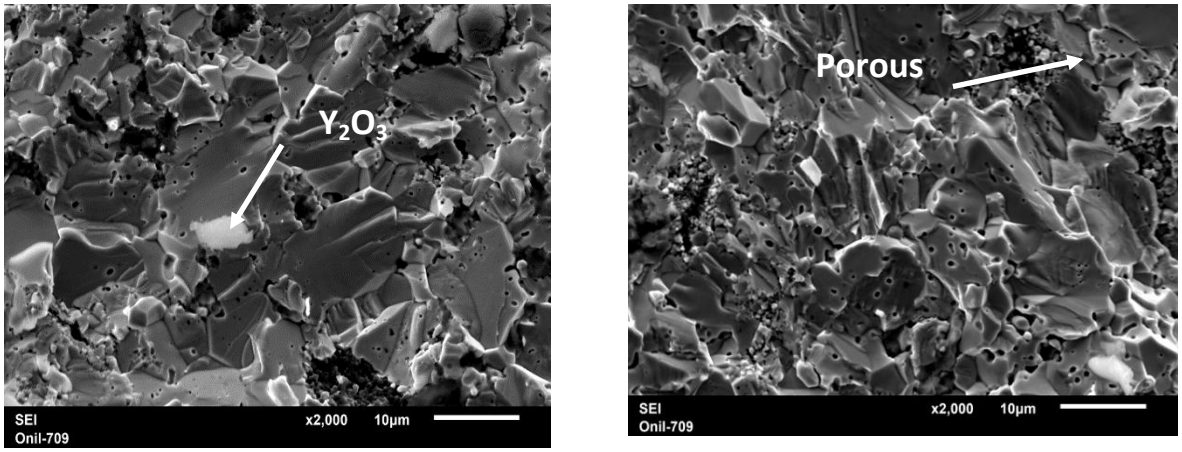
Dissolution nitrides of lanthanides nitride uranium will lead to an increase in the lattice parameter is thus the closest analogue is the material, which causes an increase in uranium nitride lattice parameter close to this nitride plutonium. Analysis of the relationship indicates that dissolution of lanthanide nitrides leads to a strong increase in the lattice constant than the dissolution of plutonium nitride. At the same time, it is clear that at low concentrations of plutonium nitride (up to 20 mol.%), The closest analogue may be considered material nitride cerium. In addition, we note that according to [4-6] dissolution of yttrium nitride uranium nitride virtually no increase in the lattice parameter of the latter, due to the proximity of the values of the lattice parameters YN and UN. In this respect yttrium nitride can be considered very close substitute nitride plutonium.

Analysis [7,8] showed that in the range of operating temperatures (the sintering temperatures) (1700-2000 K) the partial pressure of nitrogen and the metal nitride plutonium fairly close to the pressure of yttrium nitride, lanthanum nitride curves and cerium are somewhat lower, but when the temperature rises more than 2500 K, the curves are the same.

Thus, on the basis of this analysis it is expedient to choose as the simulator nitride plutonium when tested at low temperatures and high temperatures - nitrides of yttrium and cerium, respectively. The simulator can select americium nitride dysprosium, all the above materials are available and can be synthesized in the laboratory.

4. Results

Uranium nitride is a powder with a particle size of from 0.1 to 10 microns. After compacting and sintering powder of uranium mononitride obtained tablets. The tablet has few pores, cracks are absent. As a result of electron microprobe and X-ray phase analysis of the oxide phase is not detected. Figure 1 shows the microstructure of tablets Mononitride uranium with addition of 5 wt. % Yttrium. Figure 1 (a) visible white phase separation, according to microanalysis phase defined as yttrium oxide (Y_2O_3). Its occurrence is associated with short-term stay of the sintered sample mints in the air. Subsequently mints treatment technology has been changed and the oxide phase of the samples is not detected. Figure 1 (b) clearly visible large pores in which the particles were yttrium nitride, which during sintering dissolved uranium nitride.

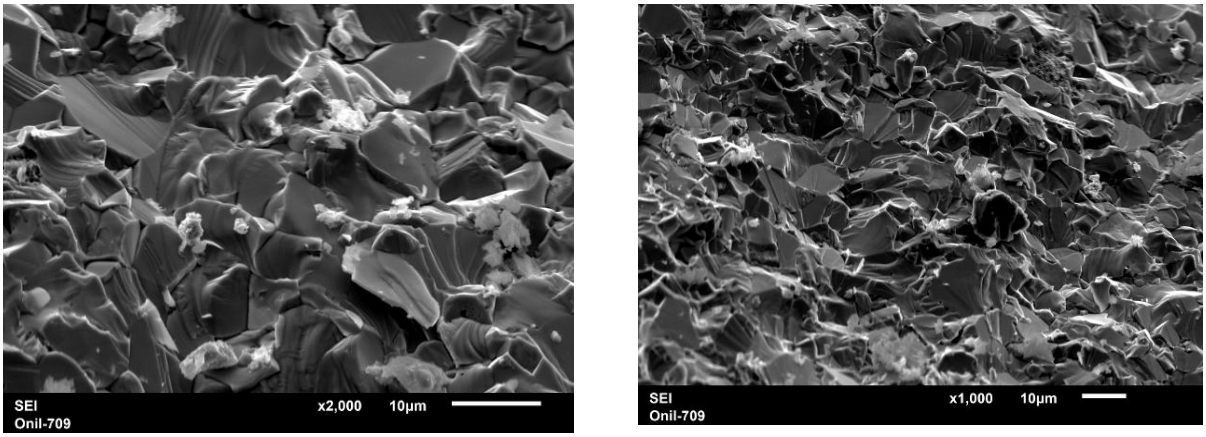


(a) (b)

Figure 1. (a) The microstructure of tablets UN-5 wt. % YN, contaminated yttrium oxide; (b) the microstructure of the sintered tablet UN-5 wt. % YN, without yttrium oxide.

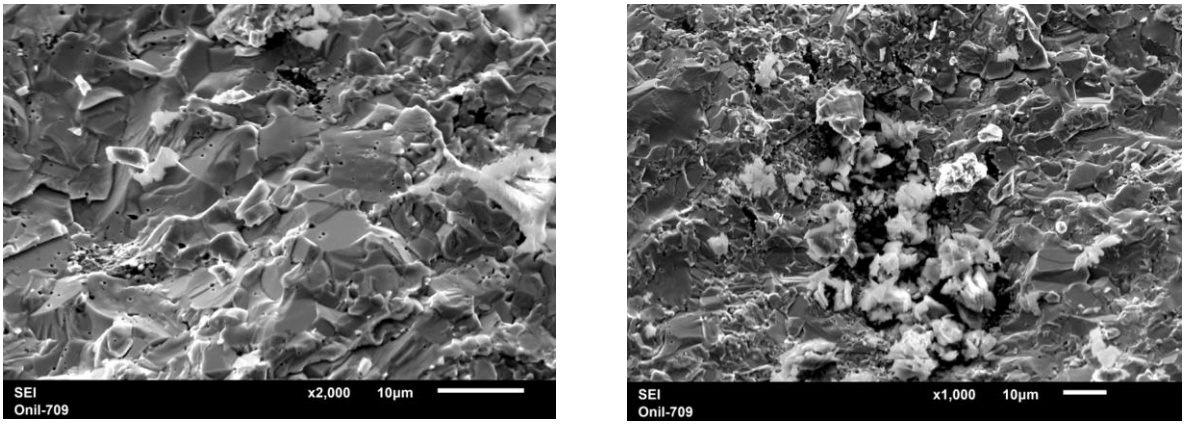
A similar situation was observed for the mixture of uranium nitride - nitride, cerium, in one of the samples were in the air, traces of the oxide phase. This fact is clearly demonstrated in Figure 2 (a). Figure 2 (b) shows the microstructure of a sample in which no traces of the oxide phase.

The worst situation is with pills containing nitrate, dysprosium, whose microstructure is shown in Figure 3. Clearly visible white discharge oxide phase oxidation occurs even during a short stay in the air while carrying samples of argon box under a microscope. Due to the lack of homogenization of a large number of dysprosium nitride is insoluble in uranium nitride. Subsequently, dysprosium oxides to nitride oxide.



(a) (b)

Figure 2. (a) The microstructure of tablets UN-10 wt. % CeN; (b) the microstructure of the sintered tablet UN-20 wt. % CeN.



(a) (b)

Figure 3. (a) and (b) the microstructure of tablets UN-10 wt. % DyN.

Table 1. Results of measurement and calculation of the lattice parameter.

Composition	Calculated *a* (pm)	Referential *a* (pm)
UN – 5 % YN	488.4	488.5
UN – 10 % YN	489.9	488.5
UN – 10 % CeN	490.5	490.8
UN – 20 % CeN	491.7	491.9

According to the analysis we can conclude a good agreement between the results obtained in this study with the literature data. Also, comparison of the data in table 1 suggests that in the synthesis of nitride powder and sintering was noticeable evaporation simulators plutonium nitride (yttrium and cerium).

5. Conclusion

The results of the work can be done preliminary conclusions:

1. On the basis of an assessment of physicochemical and thermodynamic properties of selected lanthanides, actinides are the closest analogues. Analogues of plutonium are cerium and yttrium, americium and analogues - dysprosium. The introduction of these elements in mononitride uranium produces fuel Snoop model for the study of a number of its properties;

2. A method of obtaining a model Snoop-based fuels and lanthanide nitrides Mononitride uranium.

3. The method of hydrogenation-dehydrogenation, nitration obtained nitride powders of uranium, cerium, yttrium and dysprosium, conducted their mixing, pressing and sintering, obtained samples of the model nitride fuel with plutonium and americium imitation of a given composition; According to the results of structural studies have shown that all the samples are solid solution of nitride of REE in uranium nitride. It is found that the sintered pellets Snoop model fuel are heavily oxidized in air, as opposed to tablets Mononitride uranium.

Acknowledgements

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References

[1] Bibilashvili J K, Baranov V G and Godin Y G 2002 *Problems of Atomic Science and Technology. Series: Materials and New Materials* **59** 55
[2] Laverov N. P. et al. 2006 *Geology of Ore Deposits* **48 5** 335
[3] Brauer G 1985 *Guidelines for inorganic synthesis* 4 (Moscow Mir)
[4] Samsonov G V 1964 *Refractory compounds of rare-earth metals with nonmetals* (Moscow Metallurgy)
[5] Yasufumi S, Yasuo A, Takashi I and Toshihiko O 1990 *J. Nuc. Sci. and Techn.* **28** 689
[6] Tennery V J, Bomar E S 1971 *J. Amer. Ceram. Soc*. **54** 247.
[7] Benedict U 1979 *Thermodynamics of nuclear materials* **1** 453
[8] Wicks C E and Block F E 1961 *Thermodynamic properties of 65 elements: their oxides, halides, carbides, and nitrides* (Bureau of Mines, Albany, OR (USA). Albany Metallurgy Research Center) **BM-BULL-605**