Concerning technologies of manufacture, utilization and reprocessing of massive nuclear fuel

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Abstract. UPuO₂ oxide fuel was compared with denser UPuN, UPuZr, U fuels to look into specifics of fuel production methods, manufacture, utilization in fast reactors and reprocessing. It is shown that uranium metal offers advantages in such parameters as density, thermal conductivity and ease of manufacturing of fuel meats. At that, no additional difficulties arise with the mastered technology of irradiated fuel reprocessing. Options of non-enriched uranium metal placement into the fast reactor core with the UPuO₂ oxide fuel are provided. As regards the nuclear-physical characteristics, heterogeneous oxide-metal cores with FA, intra-FA or intra fuel elements heterogenization with the ratio of UPuO₂ to U equal to 2:1 are similar to homogeneous cores with dense UPuN or UPuZr fuels. Such cores have the breeding ratio of the core (BRC) ≥1.0 required for the reactor safety. Components of such oxide-metal heterogeneous fuel elements and FAs with uranium fuel meats of various types passed the stages of technological development, reactor testing and post irradiation examinations.

1. Introduction

In recent years, there are intense debates concerning necessity for application of so-called massive fuel in the projected fast neutron reactors (FNR). Massive fuel includes car-bide, nitride, metallic and other types of fuel which exceed the standard oxide fuel in physical density and, what is more important, in effective density (γ_eff) of heavy atoms under the fuel element shell. γ_eff values affect breeding ratio of the core (BRC). The reactor with the BRC ≥ 1.0 exhibit inherent security characteristic. The reactors with oxide fuel have the RBC < 1.0 and do not exhibit this characteristic. Use of oxide or more massive carbide, nitride or metallic fuel in the FNRs is stipulated by different density values of these fuel types as well as by different technologies of fuel and fuel assembly manufacture, specific features of certain FA operation, development of spent nuclear fuel reprocessing (SNF).

For some reasons, it was decided to use oxide fuel in the FNRs of the first generation. The technologies of oxide fuel manufacture, utilization and reprocessing were developed, investigated, mastered and successfully implemented in Russia and abroad.

During development of the projected fast neutron reactor of the new generation (Gen IV, FR-4G), it is expected to achieve the RBC ≥ 1.0 by means of massive fuel use - metallic or nitride. Selected fuel type affects significantly the majority performance parameters of the fuel assembly, reactor core, reactor and the fuel cycle in general. In this regard, there is a good reason to consider the special aspects concerning use of these types of fuel in fuel cycles.

2. Massive fuel features
The table below gives the massive fuel properties which are the most important for practical purpose, in comparison with the oxide fuel [1].

| Property of fuel | Fuel | Oxide U0.8Pu0.2O | Nitride U0.8Pu0.2N | Metallic alloy U–19Pu–10Zr | Metallic unalloyed |
|-----------------|------|------------------|-------------------|-----------------------------|-------------------|
| Density, g/cm³  | 11.0 | 14.3             | 15.7              | 18.7                        |
| t.a. density, g of t.a./cm³ | 9.7  | 13.4             | 14.1              | 18.7                        |
| Effective density in fuel assembly shell, g of t.a./cm³ | 8.0  | ≤ 11.0           | ≤ 11.0            | ≥ 13.0                      |
| Melting point, °C | 2800 | 2800             | 1130              | 1130                        |

Significant differences in physical, chemical and other properties of the considered fuel types specify the technologies of manufacture, utilization in reactors and after reactor reprocessing for these fuel types.

3. Production of fuel and cores
Nitrile fuel is produced using the standard ceramic processing like the oxide fuel. Oxides (UO₂, PuO₂) or metals (U, Pu and their alloys) may be used as raw products for manufacture of nitride fuel powder.

By now, the technology of nitride fuel powder manufacture is not selected yet permanently, that’s why it is worth to mention specific features of these two competing technologies.

When carbothermic technology is used, the essential stages of nitride fuel tablet manufacture are the following [1]:
- mixing and milling of initial UO₂, PuO₂, C powders;
- powder mixture tabletting;
- carbothermic reduction of mixture in the gaseous atmosphere (N₂ + H₂ + Ar) with cake formation;
- cake milling and addition of adhesives;
- obtained granulate compacting with production of “raw” tablet;
- tablet sintering in vacuum or in the gaseous atmosphere (Ar + H₂);
- tablet quality control (figure 1).

When “metal hydrogenation-dehydrogenation” technology is used, the essential stages of nitride fuel tablet manufacture are the following [2, 3]:
- preparation of initial metal chips;
- hydrogenation and dehydrogenation of chips with production of metallic powder;
- metallic powder nitration with production of understoichiometric nitride;
- thermal decomposition of understoichiometric nitride with production of the stoichiometric nitride powder;
- powder mixing with adhesives (figure 1).

The further process and checking operations are the same as the operation for production and control of (UPuN)-tablets using carbothermic technology (figure 1).
Figure 1. Technologies of (UPuN)-tablet production out of oxides (carbothermic technology) and metals (hydrogenation and dehydrogenation technology).

Both technologies of (UPuN)-tablets production have the following specific features:

- performance of cold process operations in the glove boxes with high purity protective atmosphere (Ar or N₂) with the volume ratio of O₂ and moisture of less than 20·10⁻⁶ (20 ppm);
- performance of hot process operations (at the high temperature - up to 1850 °C) in controlled complex gaseous mixtures;
- use of dust operations;
- use of different checkout operations at different stages;
• strict requirements to such controlled parameters as the content of corrosive and dangerous impurities (C, O₂) in tablets, to the tablet density, to the grain size, to the atmosphere used for storage of finished tablets;
• no requirements to nitrogen isotopic composition.

3.1. Alloy metallic fuel

The alloying addition (10 %Zr) to the metallic fuel is selected and justified by the US professionals [1]. Intense and continuous advertising of its specific properties encouraged selection of this fuel composition (as massive) for the projected FNRs designed in the USA and other countries.

Cylindrical cores made out of these fuel can be manufactured by the injection molding method (fusion absorption) using disposable quartz tubes (casting molds). The melting and cast temperature is about 1600 °C. The cores released from the casting moulds do not undergo machining except end cuts.

The technology of core injection molding should be improved. In particular, in order to reduce the hard waste volume by decrease of the temperature which is dangerous for alloy melting in the blacklead crucible and ensure fusion suction into the quartz forms, the alloys with reduced zirconium content (down to 3–6 %Zr) and replacement of quartz forms by the thin-wall zirconic tubes with metallic cohesion with the core are under study now.

Also, the centrifugal atomization method is being developed now. This method enables production of spheroidized pellets of U–10Zr alloy with the size of ab. 100 mkm. This pelleted fuel can be used for manufacture of vibropacked fuel columns [1].

3.2. Unalloyed metallic fuel

It bears mentioning that the special properties of unalloyed metallic fuel include not only maximum density and thermal conductivity but also high processability. The cores with different shape may be produced using different (non high temperature) methods (figure 2). This reduces essentially the overloaded requirements to impurities content in the fuel, to gaseous atmosphere composition at core manufacture and storage, to scope and number of check operations. Many process operation are performed in the third class facilities.

![Figure 2. Shapes of metallic cores in studied elements.](image)

Foremost, these simplifications refer to the cores made out of unenriched unalloyed (technical grade) metallic uranium recommended [4] for use in the lateral blankets, the side breeder blanket, the inner breeder blankets of heterogeneous oxide metallic reactor cores of different types (figure 3).
Figure 3. Variants of oxide metallic heterogeneous cores with uranium: the NOX-fuel part is ~ 67 %

The core metallic component in the fast neutron reactors with oxide metallic heterogeneous core consists of the elements or FA with unenriched metallic uranium. Plutonium is re-produced and then the accumulated plutonium is partially burn out in these elements or in the fuel assembly.

The oxide component in this core consists of fuel assemblies and fuel elements containing the MOX-fuel with increased uranium content. This fuel, fuel assemblies and fuel elements with MOX-fuel are produced in the first class facilities. At that, the scope of these works is reduced by ab. 30%.

4. Fuel element design and manufacturing process

According to some known radiative and thermal properties of the nitride and metallic alloy fuel, it is appropriate or necessary to manufacture the fuel elements containing this fuel with liquid metal filling with high thermal conductivity (Pb or Na for UPuN, Na for UPuZr). Such filling and its quality control require development and mastering in the industry, where the fuel elements of thermal and fast neutron power reactors are traditionally willed with helium.

The fuel elements with liquid metal filling should be equipped with top gas receiver which increases the total length of the fuel element with corresponding burden. All domestic FNRs are designed for use of the helium filled fuel elements with bottom gas receiver. BREST-300-OD fast neutron reactor was initially designed for use of cold (slightly swelled) nitride fuel (fuel elements with lead filling). The results of post irradiation examination of experimental fuel elements show unacceptability of this filling. Alternative domestic filling mediums are being developed including helium but the gas receiver location remains the same.

According to the known radiative and thermal properties of unalloyed metallic uranium, the elements with uranium core can be produced with helium filling and bottom gas receiver.

This allows to use traditional production technologies and the standard design of the fast reactor elements and fuel assemblies designed for use of oxide fuel.

The elements and/or fuel assemblies with unenriched metallic uranium in the fast neutron reactors with oxide metallic heterogeneous core may be interchangeable with the standard fuel elements and/or fuel assemblies with the MOX-fuel. Also, the reactor cores (Fig. 3) with the ratio between the oxide fuel component and the metallic reproducing component equal to 2:1 is similar to the homogenous reactor zones with massive fuel (nitride or alloy metallic) by its nuclear and physical properties.
At the present time, the industrial power SNF is reprocessed in all countries using aqueous processes (PUREX-process and its modifications). These processes are considered to be the primary in the foreseeable future for the operating and constructed power reactor designed to use oxide fuel. In our countries, these aqueous processes have been used by decades up to the present moment including, without limitation, in SNF reprocessing for already shut-down uranium-graphite production reactors where the fuel element cores are made out of metallic technical grade uranium. There is no room for doubt applicability of these aqueous processes for reprocessing of the FNR elements filled with helium which contain unalloyed metallic uranium cores.

As for the sodium filled fuel elements containing nitride or alloy metallic fuel cores, it is necessary to apply non-aqueous charts for their reprocessing, for example, pyroelectrochemical charts using molten salts.

Due to their characteristics, these technologies are considered to be the most promising and suitable for reprocessing of different SNF, but they have not industrial application by now. Nevertheless, only this technology is deemed to be acceptable at designing of BREST-300-OD station rector of the closed fuel cycle.

5. Conclusion
Application of massive fuel in the fast neutron reactor core enables to increase the part of reproducing $^{238}\text{U}$ to achieve the $\text{RBC} \geq 1.0$.

This goal may be achieved in the designed reactors BREST-OD-300 and BN-1200 when using fuel elements with uranium and plutonium nitride fuel only in the presence of the positive results of the scheduled technological developments, reactor testings and post irradiation investigations.

For BN-1200 reactor, this goal may be achieved by means of arrangement of the heterogeneous oxide and metallic reactor core. The elements of this core (fuel elements, reproducing elements, FAs) passes the stages of technological developments, reactor testings and post irradiation investigations.

References
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