Key Features of Design, Manufacturing and Implementation of Laboratory and Industrial Equipment for Mixed Uranium – Plutonium Oxide and Nitride Fuel Pellets Fabrication in Russia

A.L. Denisov[3], V. Reynaud[4], P.A. Shkurin[1], Yu.V. Chamovskikh[1], A.V. Davydov[2], A.E. Glushenkov[2], S.V. Pavlov[3], F. Renard[4], N.G. Sergeev[1], V.P. Smirnov[3].

[1]Sverdlovsk Research Institute of Chemical Engineering (SverdNIIkhimmash), Yekaterinburg, Russian Federation

[2]A.A. Bochvar High-Technology Scientific Research Institute For Inorganic Materials (VNIINM), Moscow, Russian Federation

[3]Sosny Research & Development Company, LLC (Sosny), Moscow, Russian Federation

[4]CHAMPALLE SAS (CHAMPALLE), Peronnas, France

E-mail contact of main author: adenisov@msk.sosny.ru

Abstract. The paper describes the author's experience in design, manufacturing and implementing equipment for Mixed Uranium – Plutonium Oxide and Nitride fuel pellet fabrication in Russia. The key features of mixed uranium-plutonium oxide and nitride powders are described, as well as their influence on main process (furnaces, presses) and auxiliary (gloveboxes) equipment design. Technical solutions for working with low fluidity powders, conveyance systems for boats prone to deformation, gas separation systems, furnace insulation material choice, as well as rules and regulations applicable for this kind of equipment are discussed.

Key Words: nitride fuel fabrication equipment, nitride fuel pellets, nitride sintering furnaces, nitride powder pressing

1. Introduction

Under the umbrella of the "Generation IV" International Conference developed countries and nuclear power market participants have for the past decade been designing and constructing pilot nuclear facilities of the so called "fourth generation", which are more attractive from the economic, safety, environmental impact and non-proliferation points of view than traditional nuclear power plants. Out of six concepts chosen for pilot project implementation, three are based on fast reactor technologies. Two of the three are concepts of reactors with lead and sodium coolants – which are well known in Russia from operation of nuclear submarines and BN-600 and BN-800 reactors [[1]].

At present a Russian "Federal Targeted Program" called "Next Generation Nuclear Energy Technologies for 2010-2015 and prospects for 2020" is underway. Within the framework of this program an industrial line for fabrication of mixed oxide uranium-plutonium (MOX) fuel was commissioned in September 2015. The program also provides financing to the "PRORYV" project, the goal of which is to close the nuclear fuel cycle by introducing fast reactors with lead and sodium coolants into the Russian power market. These reactors are to use mixed uranium-plutonium nitride fuel (MUPN), reprocessed and refabricated at the on-site nuclear fuel cycles. During the project implementation phase 1 of the construction of a pilot fabrication/refabrication facility (fabrication/refabrication module, hereinafter, FRM) for MUPN fuel is underway at the Siberian Chemical Combine (SCC).

2. Experience

During 2010-2017 SverdNIIKhimmash and Sosny with the support of several other enterprises including VNIINM following the order of SC “Rosatom” have been and continue to perform design and manufacturing of multiple systems for powder research and fabrication of MOX and MUPN fuel pellets. Most of them are based on chains of cells and glove boxes containing custom-designed equipment as requested by the customer’s technical specifications. The most important pieces of this equipment are presses and furnaces jointly developed with international partners such as the designer and manufacturer of powder pressing equipment - CHAMPALLE SAS. The scale of such systems varies from laboratory research to analytical support and industrial production. Table 1 provides the list of projects implemented by the author’s enterprises for the past three years.

TABLE I: PROJECTS RELATED TO MOX AND MUPN FUELS.

i.i.	Component	End-user	Type	Authors-participants	Delivered
1	MOX Fuel Production Complex	Mining and Chemical Combine	Industrial production	VNIINM, SverdNIIKhimmash, Sosny, CHAMPALLE	2015
2	Experimental system for (U,Pu,Np,Am)N Powder and Pellet fabrication	Research Institute of Atomic Reactors	R&D	VNIINM, Sosny	2015
3	Experimental Facilities Complex (КЭУ-2)	Siberian Chemical Combine	R&D	VNIINM, SverdNIIKhimmash, Sosny, CHAMPALLE	2015
4	FRM Process Support Line	Siberian Chemical Combine	Industrial production	Sosny, CHAMPALLE	2017
5	FRM Pellet and Disk Pressing Units	Siberian Chemical Combine	Industrial production	VNIINM, SverdNIIKhimmash, Sosny, CHAMPALLE	2017
6	FRM Carbothermal Synthesis unit	Siberian Chemical Combine	Industrial production	VNIINM, SverdNIIKhimmash, Sosny, CHAMPALLE	2017
7	FRM Pellet Sintering unit	Siberian Chemical Combine	Industrial production	VNIINM, SverdNIIKhimmash, Sosny, CHAMPALLE	2018

3. Nitride Fuel Properties

MUPN fuel is a ceramic type of nuclear fuel – a mixture of depleted uranium mononitride and plutonium mononitride. At a laboratory scale uranium/plutonium mononitride can be obtained through various processes, for example by synthesis from metallic uranium/plutonium or metal melting in nitrogen, but for industrial-scale facilities the most suitable is carbothermal synthesis, which is based on the carbon reduction of uranium/plutonium oxides in nitrogen [[2]].

Having several operational advantages compared to traditional fuel types, such as higher fissile atom density and corresponding breeding ratio and orders of magnitude higher thermal conductivity as compared to oxide fuel [[3]] – MUPN fuel is significantly more complicated in production than MOX fuel due to the following reasons:

- Uranium and plutonium nitride powders, unlike oxides, self-ignite in air even at ambient temperature (so-called pyrophorosity), they are also known as efficient “getters”, adsorbing oxygen, moisture and other impurities from inert gases.
- Nitride powders are significantly less fluid and have lower pressability.
- Green MUPN pellets are more brittle than oxide pellets, thus require delicate handling.
- Nitride sintering occurs at temperatures of ~100 °C higher than conventional MOX fuel sintering and under highly specific gas-temperature conditions that prevent unwanted compounds, such as (U,Pu)2N3 (so called sesquinitride) or metallic grains from appearing.

The features listed above impose additional requirements on the equipment traditionally used in nuclear fuel production.

4. Processing steps

The main stages of the carbothermal synthesis-based MUPN pellet fabrication process implemented at the FRM are provided below:

- dosing of required portions of uranium, plutonium oxides and carbon;
- mixing;
- granulation;
- adding binder;
- disc pressing;
- bulk-loading pressed discs into a boat;
- carbothermal synthesis in batch furnaces for two days;
- nitride discs milling;
- nitride granulation;
- adding binder;
- pellet pressing;
- placing pellets into boat in an orderly fashion;
- pellet sintering in a continuous furnace for 12 hours;
- dimensional control and sorting;
- transfer to the fuel rod assembly area.

5. Glovebox ventilation systems

A qualitative comparison of the requirements for the atmosphere in the gloveboxes intended for handling uranium dioxide, MOX and MUPN fuels is given in Table 2.

TABLE II: ATMOSPHERE REQUIREMENTS FOR HANDLING UO_2, $(U,PU)O_2$, (U,PU)N

i.i.	Parameter	UO_2	$(U,Pu)O_2$	(U,Pu)N
1	Operating atmosphere	air	partially inert	mandatory inert
2	Underpressure	relaxed	strict	strict
3	Leak-tightness	relaxed	medium	strict

The glovebox atmosphere for handling uranium dioxide is conventionally air and the leak-tightness requirements are primarily based on two factors: the necessity to prevent moisture from adsorbing onto the powder that results in solidifying and worse fluidity of the powder, as well as full oxidation into U_3O_8 affecting the quality of the final product.

According to the IAEA recommendations for safety of MOX fuel fabrication facilities [3], *it may be necessary* to maintain inert atmospheres in gloveboxes to minimize the risk of a large fire.

In practice, although maintaining inert atmosphere in MOX handling gloveboxes is not mandatory, it is still maintained, mostly in the powder handling areas, i.e. starting with fuel dosing and through pellet sintering. Underpressure in MOX handling gloveboxes is controlled more stringently to limit radiation exposure (internal radiation doses) to the personnel due to release of plutonium aerosols into the operators' rooms.

As for MUPN-handling gloveboxes, the pyrophoricity of uranium and plutonium nitrides as well as the sorption of unwelcome components (oxygen, moisture) impose a strict requirement that, in addition to maintaining low pressure, the inert atmosphere shall maintain levels of oxygen and moisture contained below 50 ppm. The combination of these two requirements implies that the loss of containment will result in a fire break-out should the emergency ventilation be turned on or a release of plutonium aerosols into the operators' rooms if the emergency ventilation is turned off.

So, from a regulatory point of view, the loss of containment of the MUPN handling glovebox is considered as an initiating event for a design-basis accident, and thus the gloveboxes are assigned a Safety Class of 2N according to NP-016-05, which requires, where possible, to exclude detachable joints from the glovebox design, and when impossible - to control the joint leak-tightness.

SverdNIIkhimmash, together with Sosny R&D Company, have developed a method for detachable joint monitoring based on fittings and double seals, in between which inert gas is pressurized at a pressure of 3000 Pa. If the joint leaks, the gas flow rate will immediately be detected.

Since each glovebox is usually equipped with tens of joints, the control is automated and the control systems are assembled in compact cabinets (see FIG. 1) which automatically scan each sealed joint and report the results to the control room.

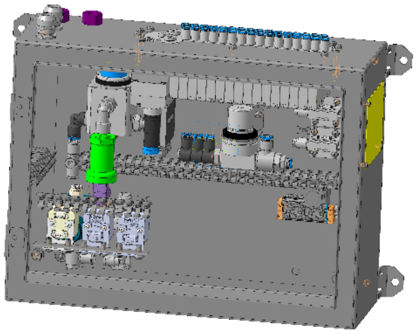

FIG.1. Glovebox joint monitoring cabinet

6. Conveyance systems

The transfer of nitride fuel powders and semi-products through and between production areas has its specifics, too. The facilities working with uranium dioxide transfer the powders in unshielded containers, while sintered pellets are openly transferred between the production areas. Due to high background radiation, both MOX and MUPN fuel fabrication facilities implement a fully automatic production process without the possibility to transfer the semi-products outside production lines. As mentioned above, in addition to the technologies used for MOX fuel production the entire production line for nitride production needs to be adapted for a nonfluid, ill-compressible and brittle-before-sintering product.

6.1.Low fluidity

In designing powder supply pipelines, all off-vertical branch pipes must have an inclination of less than 30 degrees. Additionally, the pipelines are designed and fabricated with a number of vibropercutors and a corkscrew-shaped powder agitator (Fig. 2) in the interim hopper to help propel the powder. All the equipment is adjustable and operates in automatic mode.

FIG. 2. Corkscrew-shaped powder agitator

6.2.Low compressibility

To ensure satisfactory compression of the MUPN fuel pellets, the pressing machines are equipped with a multiple compression algorithm that enables the operator to recompress the pellets with an individually set force and speed (Fig. 3). To ensure sustaniable compression results an algorithm of self-adjustment to reach the pellet density or height input by the operator is implemented.

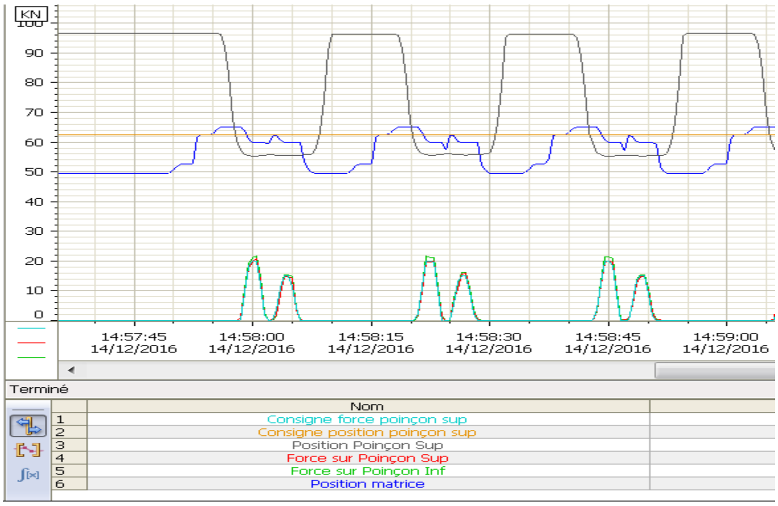

FIG. 3. Diagram of multiple compression, where green is force on the punch, yellow is the punch coordinate, blue is the matrix coordinate.

6.3.Pellet brittleness

Brittleness of green nitride pellets poses additional constraints on the procedure of their handling both individually or loaded into the boat designated for automatic transfer of the batch from the compression to the sintering area:

- Transfer of individual pellets to the loading area and loading of the pellets into boats shall be performed very carefully, the pellets cannot even tilt.
- The pellets cannot be bulk-loaded into boats.
- The pellets must be loaded into boats as close to each other as possible to prevent their tilting/movement during transfer of the boats.
- The side surfaces of the pellets must not touch to avoid non-conformities due to pellets sticking together.

The abovementioned conditions required the use of a special pellet pick & place system equipped with a suction cup grapple enabling the transfer of a pellet into the boat by its butt end (Fig. 4). The pump for providing suction in the cup is located inside the glovebox and uses the internal glovebox atmosphere, however, the system can be reequipped to operate with an external nitrogen source should that be necessary.

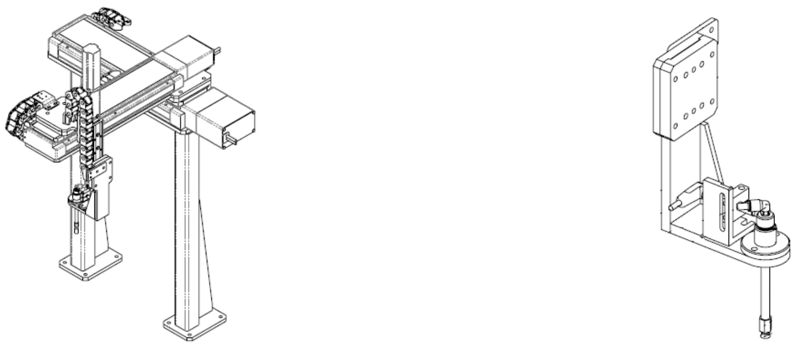

FIG. 4. Pick & place system with a suction cup (left) and the grapple (right).

7. Sintering Boats

The requirements for compact pellet arrangement in a boat impose restrictions on the boat design, for example, the bottom surface area used for pellets shall have minimally rounded corners in order to prevent any pellet from falling (Fig. 5).

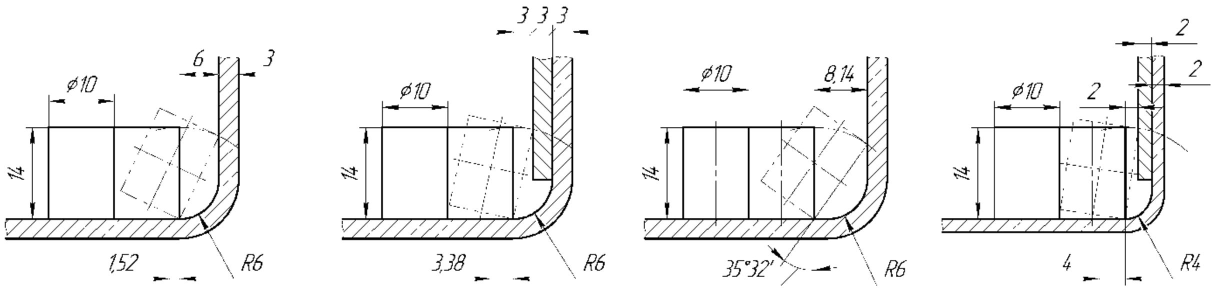

FIG. 5. Boat design affecting the risk of pellet falling

In addition, even in the case of a thick-walled boat, its long-term use in furnaces will cause the boats to become brittle and to warp, therefore the boats must be handled only by its bottom wall in case there is no tray to allow it to be pushed. Such trays must have thick walls (at least 20 mm). To comply with the abovementioned requirements, the pellet pressing unit was equipped with a lift-and-transfer system. In total 14 boat designs for the pellet sintering unit and 7 boat designs for the carbothermal synthesis process have been dismissed (Fig. 6). The final design of the boats for both sintering and carbothermal synthesis uses tungsten as the main material.

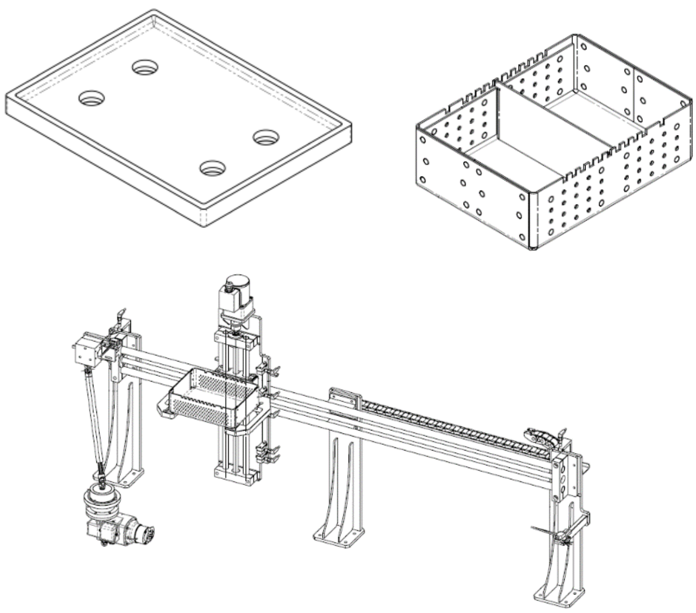

FIG. 6. Tray design option (top-left), boat design option (top-right) and pellet boat transfer system (bottom) for the FRM project

8. Sintering Furnace Type

The concept of a system consisting of several horizontal batch furnaces was found to be unacceptable for a production-scale nitride fuel sintering process. Firstly, the thermal cycling without forced cooling significantly affects the furnace capacity and service life. Secondly, each batch sintering furnace will have its manufacturing features and errors related to the process gas flow rate, pressure and temperature. The study of available technologies has shown that a continuous pusher sintering furnace is preferred.

9. Furnace Gas Media

In order to prevent the forming of any less-dense nitride states in the fuel composition, the atmosphere of the carbothermal synthesis furnace and the atmosphere inside the sintering furnace shall strictly comply with the requirements not only for moisture and oxygen but also with ones related to the nitrogen content.

In contrary to the MOX fuel sintering process, where in order to prevent generation of U_3O_8 the sintering atmosphere is changed just once (air-nitrogen and back), the MNUP fuel sintering process requires that:

- In the carbothermal synthesis cycle all preheating and holding stages shall be passed in nitrogen media followed by a nitrogen-hydrogen mixture (in order to remove excess of carbon), after which prior to cooling the gas mixture has to be replaced by argon in order to prevent the generation of sesquinitrides;
- In the sintering cycle all pellet heating and cooling operations shall be performed in argon containing less than 0.1 % of nitrogen. The sintering shall be performed in the atmosphere of at least 50% volumetric of nitrogen in order to prevent the nitrogen loss and generation of metal inclusions in the pellets.

The above requirement for the sintering cycle was the most difficult to meet because it was necessary to provide efficient inert gas separation in a single channel of the continuous furnace and out of reparability reasons no channel-separating barriers closer than 50 mm to the boat wall were allowed.

ANSYS FLUENT code was used to perform a series of CFD calculations which allowed to find an optimal flow rate of the gases, and to determine the furnace design (Fig. 7) that provided efficient gas separation within the required temperature range (Fig. 8).

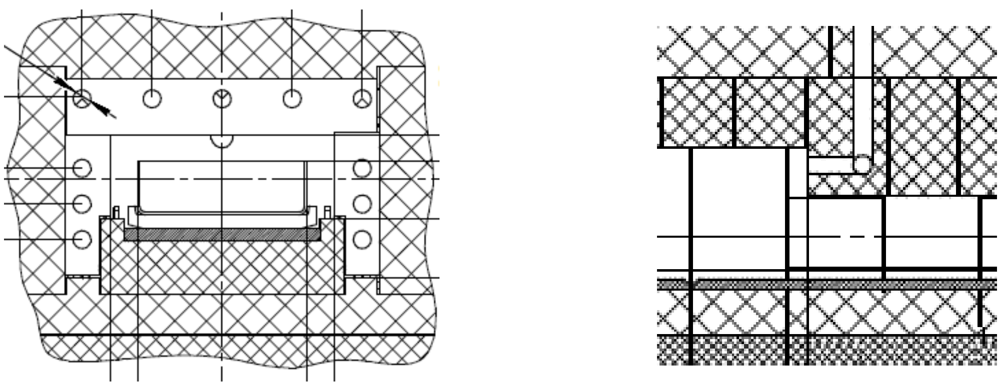

FIG. 7. Continuous furnace gas barrier

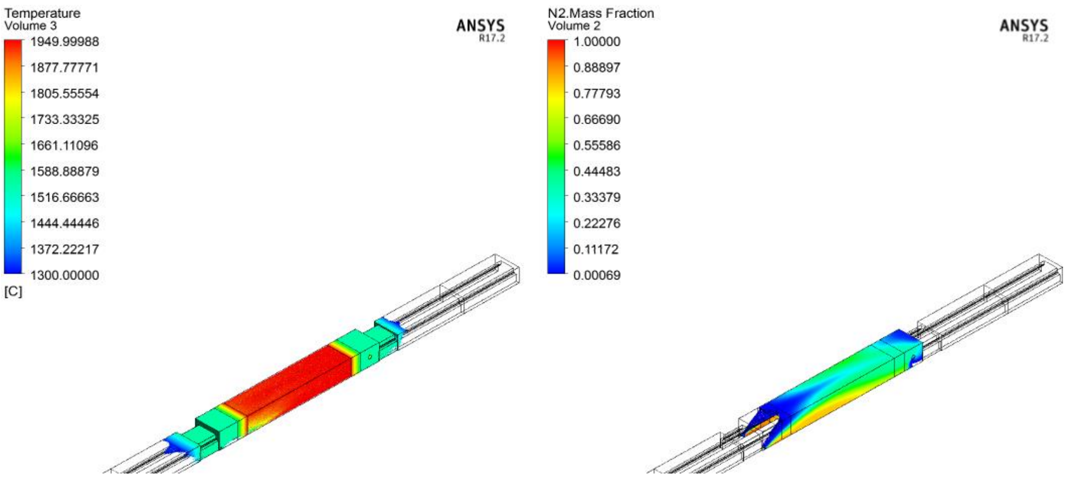

FIG. 8. CFD calculation results of temperature distribution (left) and gas distribution (right) obtained for one of the gas barrier options

10. Material Choice

The choice of process gas resistant materials was a separate task to be performed during the nitride fuel fabrication furnace equipment design. The minimal temperatures supporting the carbothermal synthesis reaction and the pellet sintering are 1700 °C and 1950 °C, respectively. The gas media is a mixture of nitrogen and hydrogen, containing carbon, carbon monoxide, hydrocyanic acid and hydrocarbons. The aluminum oxide, which is conventionally used in MOX fuel furnaces, had evaporated under the pellet sintering conditions.

As a result of endurance tests performed by VNIINM in 2016 (Fig. 9), various grades of high-purity and doped aluminum oxide, zirconium oxide stabilized by calcium and magnesium, graphite, silicon carbide and other composite carbon-containing materials have been rejected.

The best result for its use in the high-temperature zone of the sintering furnace was obtained for yttria stabilized zirconium oxide. As none of the graphite or ceramic materials were strong enough to be used in carbothermal synthesis, it was decided to manufacture the furnace crucible out of tungsten.

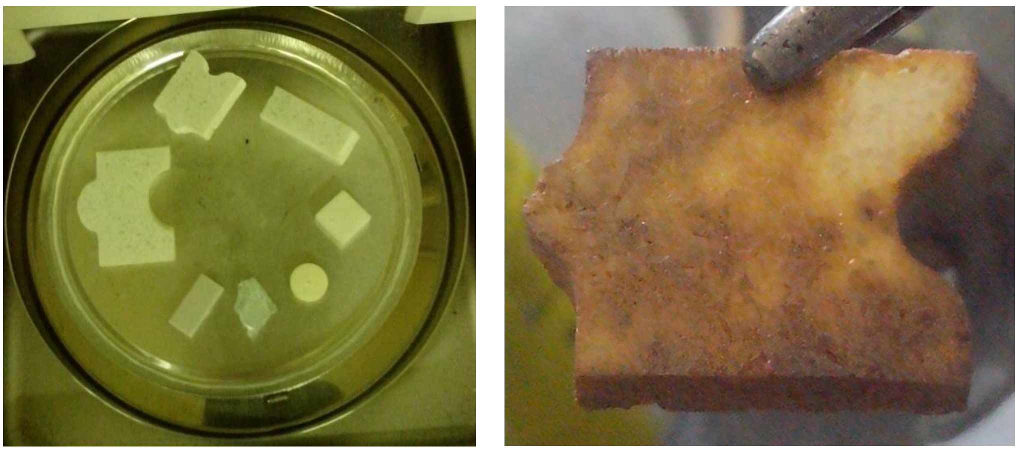

FIG. 9. Photos of samples before (on the left) and after (on the right) life tests

11. Conclusions

Both production and laboratory applications of the nitride fuel fabrication process impose more stringent requirements on auxiliary and main process equipment. Nevertheless, the current state of science and technology allow to reach the objectives set for nitride fuel fabrication equipment.

The experience gained from this work can be used in newbuild fuel fabrication plants that follow the on-site nuclear fuel cycle concept and in other projects aimed at research or production of highly-toxic and atmosphere-sensitive powders both in the international nuclear industry and in neighbouring fields of application.

12. References

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