SOME RESULTS ON DEVELOPMENT, IRRADIATION AND POST-IRRADIATION EXAMINATIONS OF FUELS FOR FAST REACTOR-ACTINIDE BURNER (MOX AND INERT MATRIX FUEL)

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Abstract

Studies performed have shown principal feasibility of the BN-600 and BN-800 cores to achieve high efficiency of Pu burning when MOX fuel with Pu content up to 45 % is used. Valuable experience on irradiation behaviour of oxide fuel with high Pu content (100%) was gained as a result of operation of two BR-10 core loadings where the maximum burnup 14 at. % was reached. Post-irradiation examination (PIE) allowed to reveal some specific features of the fuel with high plutonium content. Principal irradiation and PIE results are presented in the paper. Use of new fuel without U-238 provides the maximum burning capability as in this case the conversion ratio is reduced to zero. Technological investigations of inert matrix fuels have been continued now. Zirconium carbide, zirconium nitride, magnesium oxide and other matrix materials are under consideration. Inert matrices selection criteria are discussed in the paper. Results of technological study, of irradiation in the BOR-60 reactor and PIE results of some inert matrix fuels are summarised in this report.

1. Introduction.

Utilization of fast reactors to burn actinides is an effective way of resolving the long-lived nuclear power waste issue. In Russia extensive study of fast reactor cores which can efficiently burn plutonium and minor actinides (MA) is under way in order to demonstrate the possibility of actinide utilization in BN-600 and BN-800 reactors.

To achieve the purpose of effective actinide burning the design of the breeder reactors BN-600 and BN-800 need to be modified. Modification of BN-600 and BN-800 breeding cores with traditional oxide fuel proceeds in two stages. The first one is connected with the removal of radial and axial breeding zones from the core and their replacement by non -breeding blankets. In this case the breeding ratio 0.73., and fast reactor becomes a plutonium burner. The second direction is connected with the Pu content increase.

Studies performed have shown principal feasibility of the BN-600 and BN-800 cores to achieve high efficiency of the Pu burning, up to 360 kg/year, when MOX fuel with 45% Pu is used/1/. Technological study on development and fabrication of MOX fuel with high Pu content is under way now aiming the fuel manufacturing for the BOR-60 irradiation. Important experience on irradiation behavior of high Pu oxide fuel was gained as a result of the irradiation of two BR-10 core loadings with PuO_2 and post irradiation examination (PIE) of spent fuel pins in the hot cell. Irradiation and PIE results are summarized in this report.

Use of the inert matrix fuel without U-238 will provide the maximum burning capability as in this case the conversion ratio is reduced to zero. For the core with inert matrix fuel the fuel materials are considered on the base of zirconium carbide, magnesium oxide, aluminium nitride and some others. Calculative investigations of the BN-800 type core with different types of matrix fuel are carried out. The fundamental technological investigations are necessary in this direction on the selection and characterization of the new fuel materials.

2. MOX fuel.

2.1. Development and fabrication of MOX fuel with high Pu content.

During the 1994 the laboratory study of MOX fuel with 45% Pu fabrication was carried out in the VNIINM, Moscow. The fabrication techniques of chemical coprecipitation and mechanical mixing of oxide powders were investigated, the physical, chemical and technological properties were studied. It was shown that as for mechanical mixed so for coprecipitated powder it is possible to obtain the homogeneous fuel from the powder and the solid solution after the sintering. Adjusting some fabrication process parameters it is possible to fabricate pellets with rather wide range of density values (9.5 - 10.7 g/cm^2). The pellet solubility with high Pu content (45%) was studied also. It was found that it is necessary to increase the solution period significantly comparing with the 30% Pu MOX fuel. Presently these investigations are continued. Several fuel pins are manufactured to be irradiated in the BOR-60 reactor.

2.2. Irradiation experience.

The set of experimental subassemblies (SAs) irradiations with MOX pellet fuel were organized in BOR-60, BN-350, BN-600 reactors with an aim to study fuel performance. Besides, big experience was gained with MOX vibropacked fuel in BOR-60 (Table 1).

TABLE 1. MOX FUEL IN BR-10, BOR-60, BN-350, BN-600 REACTORS

Reactor	BR-10-PuO_2	BOR-60	BN-350	BN-600
Pellet fuel				
Pin number	3300	400	1800	1524
Max. burnup % at.	14.	24.*	10.8	10.5
Max. linear rating, kW/m	17.	50.	48.	48.
Vibropack	fuel			
Pin number	-	12800	254	762
Max. burnup % at.	-	28.*	7.2	9.8
Max. linear Rating, kW/m	-	52.	48.	48.

*) The irradiation was arranged in special dismountable assembly.
The Pu content in MOX fuel irradiated was less than 30%. All fuel pins were intact.

First results on irradiation behavior of oxide fuel with high Pu content (100 %) was obtained in BR-10 where two core loadings with PuO_2 were irradiated. Principal design and operational parameters of BR-10 fuel pins are shown in Table 2.

It should be noted that in the pin design of the first core loading the fission product gas plenum was not foreseen that time because of both a small design experience and an inadequate knowledge about the fuel behavior under irradiation. As a result, the fission product gas pressure reached 30 MPa and that was the reason of fuel leakage.

Ten SAs at the peak burnup values from 2.2 to 6.7% at. were selected for PIE. Failed fuel pins were found in six SAs. Two of these 6 SAs at burnup 4.9% at. contained pin claddings with cracks. Each of the other four SAs, at burnups of 5.6, 6.1, 6.6, 6.75% at., contained two or three pins, failed at its withdrawal into two pieces with fuel bits on them.

From the beginning of the second core operation, because of manufactured microcracks in one or several fuel pin claddings, the repeated fission gas releases were detected, however, with no changes in delayed neutron activity in coolant. At burnup of 10.8% at. the failed cladding detection system (FCDS) has registered the first failed pin by delayed neutron signals. With the further core operation using the delayed neutron measurements FCDS has registered the other four signals. After appearance

of delayed neutrons in coolant the radioactivity of fission products in it increased by one order but was not too high to create obstacles to repairs of boxes in primary coolant circuit.
An out-of-reactor FCDS was used and 13 SAs with failed fuel pins were found. Pin cladding defects of the "fuel-coolant" type were found in three of them.

TABLE 2. PRINCIPAL DESIGN AND OPERATIONAL PARAMETERS OF BR-1 FUEL PINS WITH PuO_2

PARAMETER	I LOADING	II LOADING
SAs number	83	72
Fuel pins number	1577	1368
Wrapper flat-to-flat size, mm	26.	26.
Wrapper wall thickness, mm	0.5	0.5
Cladding diameter, mm	5.	5.
Clad wall thickness, mm	0.4	0.4
Core height, mm	280.	320.
Gas plenum height, mm	-	105.
Wrapper- clad material	18Cr-9Ni-Ti	18Cr-10Ni-Ti 16Cr-15Ni-3Mo
Maximum burn-up, % at.	6.6	14.1
Maximum clad temperature, C	580.	585

2.3. Results of post-irradiation examination of fuel pins.

For PIE seven SAs at maximum burnup of 1.42, 3.18, 4.62, 5.81, 7.69, 9.21, 12.06% at. with PuO_2 were selected. The following investigations were carried out:

-measurement of cladding diameter changes,
-pin cladding tightness examination by means of gas composition analysis,
-gamma-scanning of fuel pins,
-fuel and cladding microstructure investigation,
-study of mechanical properties of cladding materials,
-analysis of fission gas release rate.

As a result 8 fuel pins with gas leakage were revealed from SAs with maximum burnup of 7.69, 9.21, 12.06% at.

The following conclusions were derived from these PIEs:

-Noticeable gas release was observed at burnups of 4% at. and more.

-Solid fission products were distributed in fuel column according to the neutron fluence and had not evidently migrated along fuel column. No fuel mass transfer in the axial direction was observed either.

-Radial cracks were observed in all cross-sections of fuel column investigated. In many cross-sections the fuel had crumbled out. The central hole diameter was 1.15-1.25 and was similar for all cross-sections investigated. For the midplane three zones with the distinct types of fuel microstructure were observed: the columnar grains zone, the equiaxial grain zone and unrestructured zone. This microstructure is typical of oxide fuel irradiated in fast reactors.

-Cladding of all fuel pins at maximum burnup levels of more than 4% at. suffered internal corrosion as a result of FCCI during irradiation. FCCI varied qualitatively and quantitatively for different parts of cladding depending on irradiation conditions.
In the lower part of the fuel column no FCCI was observed with the exception of the 5 mm thick layer in which the grain boundaries were sensitive to etching. Near the core midplane the intergranular cladding penetration was 30-50 mm . In the upper part of the fuel column (at burnup of more than 8% at.) the intergranular penetration was 120 mm and followed by a partial dissolving or loss of single grains.
The perforating intergranular cracks in claddings from failed fuel pins were found near the core midplane and above. Besides the perforating cracks the numerous microcracks on the inner surface of the cladding with various penetrations were observed on these cross-sections. For these fuel pins the FCCI penetration was 90-120 mm. On the outer surface of PuO_2 pellets which were taken from the core midplane with burnup 12% at. a complex phase with Pu, Cs, Cr, Pd, Fe, Ni in its content have been found.

At present 32 fuel pins with MOX fuel from 4 SAs of BN-350 and 40 fuel pins from 5 SAs of BOR-60 have been investigated in the IPPE hot laboratory. The fuel pins were examined visually and no failures or defects were found.

The accumulated PIE results are the followings.

-Solid fission products were distributed in fuel column according to the neutron fluence (similar to the UO_2 fuel) and had not evidently migrated along fuel column.

-Gas release increased noticeably with burnup increase after 3% at. Gas release from MOX was almost the same as from uranium dioxide and reached the value of 95% at burnups more than 10% at. Besides the burnup gas release depended on fuel fabrication method , fuel composition, O/M ratio. With the equal burnup values gas release from the coprecipitated fuel was less. Gas release from the fuel with O/M=2. was less than from the fuel with O/M<2. The fuel with lower density retained more fission products , that probably could be connected with the dependence between open and closed porosities. With linear rating increase (37.- 50.W/cm) gas release increased by 15 - 20%.

-MOX fuel had typical microstructure consisting of three zones whose size was basically dependent on linear rating value in cross-sections investigated. The cladding - fuel gap contained fission products. In the outermost part of mechanical mixing fuel pellets precipitates of UO_2 and PuO_2 were found in the columnar grain zone." Sol-gel" fuel was more homogeneous after irradiation , though the phases consisting of fission products were observed in fuel microstructure.
-In the mechanical mixing fuel with O/M =2. the Pu enrichment of columnar grain zone near the central hole was observed (15 - 20% more than the initial structure). In a more homogeneous fuel such type of Pu enrichment was not more than 2 - 7%. It depended on O/M ratio of solid solution. The less O/M ratio was, the less Pu enrichment of central pellet part was. No Pu redistribution along fuel column height was found.

-FCCI depends on oxygen chemical potential as a function of O/M ratio in fuel. Depending on the initial O/M ratio the radial redistribution of oxygen took place during irradiation. With the O/M initial ratio increase and burnup increase the oxygen potential increase was observed especially in the outermost part of pellets that led to FCCI increase. No oxygen redistribution along the fuel column height was observed. Radial oxygen redistribution was seen primarily in midplanes and above (cross-sections with higher level of linear rating).

-FCCI depth increased with burnup, temperature, linear rating, initial O/M ratio increase. Intergranular and matrix type of cladding penetration with microcracks on an inner cladding surface was observed. The chemical compatibility of MOX pellet fuel with cladding materials used was just the same as that of oxide pellet fuel for equal irradiation parameters. No influence of fabrication method and plutonium composition was found.

3. INERT MATRIX FUEL.

3.1. Criteria on inert matrix selection.

The following criteria should be taken into account when inert matrix material is selected:
-workability,
-compatibility with fuel in the total range of operational temperatures,
-high melting point,
-compatibility with clad and coolant,
-irradiation stability (no big size increase, structural integrity and so on),
-thermal conductivity,
-mechanical properties (ductility, strength, high linear thermal coefficient),
-low neutron cross section,
-low activation,
-solubility when reprocessing.

3.2. Development and fabrication.

On the base of above mentioned criteria the list of inert matrix materials was selected to carry out further technological investigations : MgO, MgO+Me (IPPE, Obninsk), ZrC, ZrN, AlN (VNIINM, Moscow).

-PuO_2+MgO

On the first stage the work was carried out with Th and U as simulators instead of Pu. The coprecipitatipon process was used to obtain UO_2-MgO and ThO_2-MgO. The technological route is shown on Fig.1.

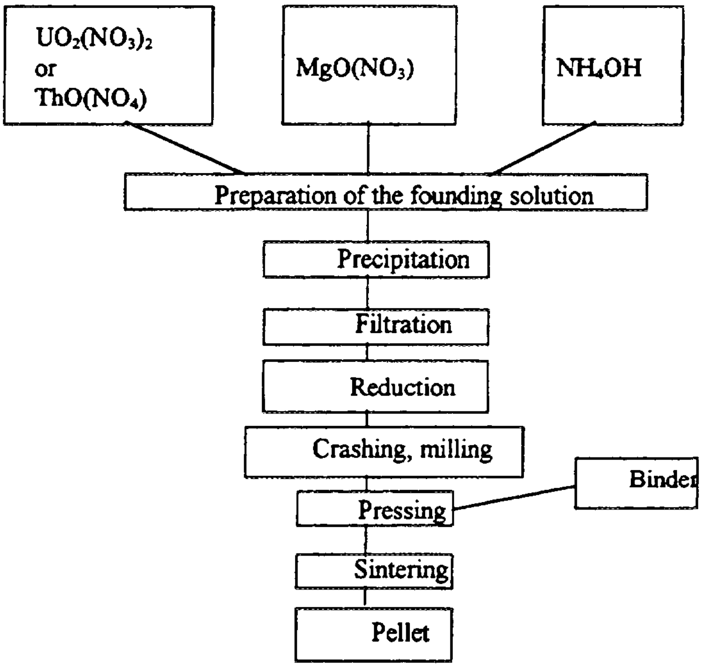

Fig.1. Cercer fuel fabrication route.

The most optimum parameters of fabrication process were chosen. Pellets fabricated have the structure with homogeneously distributed oxides UO_2 (ThO_2) and MgO. Pellet density, thermal conductivity were measured. Several pellets were reprocessed . The optimum solution parameters have been found (without special dopes). Presently the work extends aiming the fabrication of 2 -3 fuel pins with PuO_2 - MgO for BOR-60 irradiation.

-PuN+ZrN, PuC+ZrC.

One of the principal criterion of fuel with inert matrix its reprocessing ability. From this point of view solid solutions of plutonium carbides, plutonium nitrides and inert matrix ZrC, ZrN are seems to be the best candidates.

Two synthesis processes of UC-ZrC were developed /2/:
-from initial metals,
-from the initial oxides.

The problem was to define experimentally the optimum carbon quantity to exclude the sesqui-phase. Using these techniques the following fuel was fabricated:
-56% UC +44% ZrC, 55% PuC+45% ZrC for the core region,
-15% UC +85% ZrC for the blanket region
The solid solution obtained consisted of two phases. The nonuniformity of Pu distribution was less than 5%.

Technological research on synthesis of solid solution of UN+ZrN from initial oxides and fabrication of fuel columns with different density , shape and size was performed also. As it was mentioned above the synthesis method from oxides of PuC+ZrC solid solution was developed. It's properties are close to the properties of PuN+ZrN solid solution. This fact and existing experience of UN+ZrN fabrication proved the feasibility of carbothermal synthesis of PuN +ZrN from the initial oxides. The feasibility of fabrication of UN+ZrN, PuN+ZrN solid solution from the initial metals was demonstrated also.

3.3. Irradiation.

The subassembly with 19 fuel pins was irradiated in the BOR-60 , 7 fuel pins contained 55% PuC+45% ZrC fuel , 12 fuel pins contained 56% UC+44% ZrC. Irradiation parameters are shown in Table 3.

TABLE 3. IRRADIATION PARAMETERS OF INERT MATRIX FUEL IN BOR-60

Parameter	Value
Max.burn-up,% at.	8.
Max.fluence,cm^{-2},E>0.1Mev	$4.43*10^{22}$
Max. linear ratiNG, kW/m	40.2
Max clad temperature, C	$635{\pm}25$

All fuel pins were intact.

The principal PIE results of inert matrix fuel are the following:
-no gas release from the fuel,
-fuel swelling is equal to 1% per 1% of fuel burnup,
-the fine-grain structure and round-form voids uniformly distributed through the fuel (as for the unirradiated fuel),

-instead of initial two phases only one phase was observed , which seems to be rather favourable factor for fuel performance,
-homogeneous distribution of Pu,
-local carburization of cladding only in the upper part of pins.

4. CONCLUSION.

1. To achieve the purpose of effective actinide burning, the design of the breeder reactors BN-600 and BN-800 have to be modified. The modification being considered include :
-elimination of blanket region,
-increase in the Pu content in MOX fuel,
-use of Pu fuel with an inert matrix.

2. Laboratory study on fabrication and reprocessing of MOX fuel with 45% Pu was carried out . Several fuel pins are manufactured for the BOR-60 reactor. Important experience on irradiation behavior of oxide fuel with high Pu content (100%) was gained as a result of the irradiation of two BR-10 core loadings. The results of PIE on PuO_2 fuel rods performed which were irradiated in the BR-10 reactor up to the burn-up 12 % at. and on traditional MOX fuel rods which were irradiated in BN-350 and BN-600 reactors allowed to make the following conclusion on irradiation behavior of high Pu oxide fuel:
- There is no difference between PuO_2 or UO_2 or MOX fuels for swelling, gas release, fission products behavior, microstructural changes. These properties changes depend on fuel burn-up and temperature.
-Fuel-cladding interaction increases with the Pu content increase. The lowering of the initial O/M ratio in the PuO_2 fuel and the utilization of improved cladding steel could probably decrease cladding corrosion damage to the level of UO_2 fuel pins.

3. Use of the fuel without U-238 provides the maximum burning capability. Inert matrix materials under consideration are magnesium oxide, zirconium carbide, zirconium nitride and some others. The laboratory study on oxide fuel with MgO inert matrix was carried out. The fuel was fabricated, some physical. chemical and technological properties were studied. The first experience on irradiation behavior of PuC+ZrC, UC+ZrC fuels was obtained in the BOR-60 reactor. The experiment demonstrated the real possibility to use the inert matrix fuel as a fast reactor fuel.

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