UTILIZATION OF FAST REACTOR WITH URANIUM-FREE FUEL FOR MINOR ACTINIDES TRANSMUTATION

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ABSTRACT

The partitioning and transmutation of plutonium and minor actinides has been identified as being an alternative strategy for the long-term management of long-lived radioactive wastes from power reactors. New reactor developments using plutonium and minor actinides as fuel are required to cope with this problem. Design studies of actinide burner reactors have been made in Japan, France and Russia.

By now rather wide studies have been carried out on analysis on choosing different fuel compositions without uranium-238 replaced by inert diluent. But a wide range of plutonium compounds were out of consideration. The results of neutronic calculation of BN-800 reactor core with different uranium-free fuel composition for plutonium and minor actinides transmutation are presented in this report.

1. INTRODUCTION

Presently a fast reactor which uses a fuel not containing uranium-238 is considered to be one of the most efficient burner of plutonium and minor actinides (MA). The elimination of uranium, which occupied in traditional reactors from 50 to 80% of fuel pin volume, depending on reactor type (80% -for traditional reactors and 50% - for reactors-burners in increased enrichment mixed oxide (MOX) fuel) leads to appearance of essential voids, which should be filled retaining a rod-type design of fuel pins. A conception of this type reactor was developed in different directions.

IPPE was developing a conception of this type core using fuel with inert diluent. In this case a fuel material (plutonium oxide or plutonium nitride) was located in an inert matrix. ZrC, MgO, AlN, ZrN etc were considered as an inert matrix material.

The implementation of pure plutonium nitride as a fuel material was considered in the framework of CAPRA French program. In this case a ring-type fuel pin was used, in which a fuel material in the form of microgranules was located between inner and outer claddings. Empty space inside fuel pins remained non-filled or was filled by an inert diluent.

Each of these lines has its advantages and disadvantages. One should relate to the first line advantages the fact that the use of pure plutonium nitride does not require the development of any new technology for manufacture of fresh fuel and reprocessing of spent fuel. However a ring fuel pin structure is a new development and requires a more comprehensive performance substantiation.

The second line uses a traditional fuel pin design, however it requires higher expenditures for manufacture technology development and substantiation of performance for a fuel composition with inert matrix.

Thus, a list of compositions considered was limited to plutonium nitride and several compositions with inert matrix. A purpose of these studies is consideration of a more wide spectrum of materials, which can be used as a fuel material, assessment of their effect on neutronic core parameters and estimation of these cores capability to utilize MA.

2. FUEL COMPOSITION CHOOSING.

The simplest solution of fuel composition choosing problem for fast reactor with fuel not containing uranium-238 consists in implementation of known and well studied plutonium compositions[1,2,3] .

- ∗ Implementation of metal plutonium as a fuel material is not acceptable because plutonium has six allotropic modifications with low melting temperature.
- ∗ A lot of information has been accumulated recently on structure and properties of different binary plutonium alloys and intermetallic plutonium compounds, which allows the estimation of reactor systems parameters when using these compounds as a fuel material.
- ∗ Intensive studies were performed recently in creation of fuel compositions with an inert matrix. Zirconium carbide magnesium oxide, aluminium and zirconium nitrides, cerium oxide etc. were considered as an inert matrix material. The most investigated from technological point of view are PuO_2-MgO, PuN-AlN and PuN-ZrN fuel compositions.
- ∗ Among different plutonium compounds, one can separate a special group of minerals, which are compositions of plutonium with some metal oxides, a structure of which is similar to that of $CaTiO_3$ mineral. By now it has been possible to synthesise several such compositions ($BaPuO_3$, $PuVO_3$, $PuCrO_3$ etc.).

The choice of one or other material should solve the basic problems of reactor systems without uranium-238. It related primarily the reactor safety, which is mainly defined by Doppler-effect and sodium void reactivity effect (SVRE).

Below is an analysis of BN-800 reactor core neutronic parameters when using different fuel compositions both for plutonium burning and estimation of MA burning possibility.

3. PARAMETERS OF CORES WITH CERAMIC FUEL.

A power composition plutonium is considered as a fuel. The isotope composition of MA (Np, Am, Cm) corresponds to that of unloaded from VVER-1000 fuel[4].

Table I presents BN-800 reactor core neutronic parameters when using a ceramic fuel and a fuel with inert matrix from magnesium oxide, properties of which can be considered to be well studied. We will use these results as reference ones in the studies under performance.

Table I. Parameters of a core with ceramic materials.

Fuel material	PuN^{nat}	PuN^{15}	PuO_2+ MgO	PuO_2
Doppler-effect, pcm	-125,0	-108,3	-310,6	-145,6
SVRE, pcm	-3812	-3744	-2223	-3932
Reactivity change rate during interval, pcm	5412	5506	5339	5641
Max linear power, kW/m	37.9	37.0	45.1	37.9
A fraction of burned plutonium from initial loading, %	25.3	25.4	29.0	25.7

For comparison, core parameters when using PuO_2 as a fuel are presented in last column. Two important points should be noted. Firstly, the introduction of MgO inert matrix into the fuel composition leads to essential Doppler-effect increase. This is connected with that MgO is a good moderator, and a contribution of Pu^{240} and Fe isotopes is higher in a thermal spectrum, as compared with fast. However, the introduction of an inert matrix leads to more than 20 % increase in the fuel pin liner power.

Table II presents the core neutronic characteristics when using the same fuel compositions, but with addition to them of some MA quantity, latter being desired by a possibility to provide a zero SVRE value ($<\beta_{eff}$) in the reactor core.

Table II. Parameters of a core with ceramic materials for minor actinide burning.

Fuel material	$(Pu+MA)N^{nat}$	$(Pu+MA)N^{15}$	(Pu+MA) O_2+MgO	$(Pu+MA)O_2$
Actinide fraction in the fuel	25	20	20	20
SVRE, pcm	+200	+60	+260	+60
Doppler-effect, pcm	-51,0	-59,5	-121,0	-62,5
Reactivity change rate during interval, pcm	5340	5855	6030	5890
Max linear power, kW/m	38.3	38.5	45.5	39.3
Quantity of transmuted MA, kg/TW*h	28.6	22.0	25.8	22.4
A fraction of transmuted MA from initial loading, %	27.0	27.0	32.5	27.0

The results presented show, that the use of nitride fuel with natural nitrogen allows the provision of a rather high MA burning efficiency, whereas the implementation of a fuel with inert matrix provides a maximum transmutation rate.

4. NEUTRONIC CORE PARAMETERS WHEN USING INTERMETALLIC PLUTONIUM COMPOUNDS.

Among different intermetallic compounds, suitable for the use as a fuel for fast reactors, one should choose those with a rather high melting temperature. The analysis of literature data shows that the most real candidates are $PuBe_{13}$ (T_m=1950 ^oC), PuBi (T_m=1300 ^oC), Pu_2Ni_{17} (T_m≈1800 ^oC), $PuRe_2$ (T_m=1500 ^oC), $PuFe_2$ (T_m=1240 ^oC).

Other intermetallic compounds of type $PuPb_3$, PuAl, $PuCo_2$, $PuMg_2$, $PuZn_2$ etc. have a melting temperature below 1000 ^oC.

Surely, the existing literature data does not allow a rather reliable substantiation of a possibility to use one or other material as a fuel. A small experience gained in the use of metal plutonium alloyed by aluminium and iron can not be practically extended to the compounds considered above, since an addition of alloying elements in these compounds did not exceed 5%.

The estimation of BN-800 reactor major neutronic characteristics, when using intermetallic compounds as a fuel, is presented in table III.

Table III. Core characteristics with fuel from intermetallic compounds.

Fuel material	$PuBe_{13}$	PuBi	Pu_2Ni_{17}	$PuRe_2$	$PuFe_2$
Doppler-effect, pcm	-324,9	-129,8	-175,2	-58,2	-107,3
SVRE, pcm	-2212	-4006	-3424	+2745	-2712
Reactivity change rate during interval, pcm	6434	6219	5733	3506	6313
Max linear power, kW/m	43.8	43.8	43.0	41.6	38.7
A fraction of burned plutonium from initial loading, %	27.7	26.0	25.9	16.2	24.3

The best neutronic parameters from the reactor safety and plutonium burning efficiency are reached with the use of $PuBe_{13}$. This compound is used presently as a neutron source, and a possibility for its implementation as a fuel should be substantiated. Other materials do not allow the provision of necessary Doppler-effect value.

As for MA transmutation the results presented in table IV show that the compositions containing effective moderators (for example, beryllium) allow to provide both a maximum transmutation efficiency and a maximum rate of MA destroying.

Table IV. Core characteristics with fuel from intermetallic compounds when burning MA.

Fuel material	$(Pu+MA)Be_{13}$	(Pu+MA)Bi	$(Pu+MA)_2Ni_{17}$	$(Pu+MA)Re_2$	$(Pu+MA)Fe_2$
Actinide fraction in the fuel	20	20	15	15	15
Doppler-effect, pcm	-206,0	-54,0	-91,0	-30,0	-61,0
SVRE, pcm	+83	+390	+180	+3400	+200
Reactivity change rate during interval, pcm	6140	6090	6130	3980	6313
Max linear power, kW/m	45.8	41.8	43.5	42.4	5890
Quantity of transmuted MA, kg/TW*h	27.6	21.5	15.6	11.7	21.6
A fraction of transmuted MA from initial loading, %	35.5	27.1	25.2	12.5	26.9

These results fully correlate with the results obtained in analysis of efficient MA transmutation in special devices containing a moderator.

Besides, the fuel composition with beryllium allow, also the assurance of the best safety characteristics (max from all compositions Doppler-effect value).

5. CORE NEUTRONIC CHARACTERISTICS WHEN USING CERAMIC PLUTONIUM COMPOSITIONS IN INERT MATRIX AS A FUEL.

5.1 THE USE OF COMPOSITIONS OF PEROVSKYTE TYPE.

When combining plutonium oxide with some metal oxides, a creation of perovskyte type compounds is possible, which have this title since their structure resembles the structure of perovskyte mineral - $CaTiO_3$. The attempts to obtain compositions $PuFeO_3$, $PuGaO_3$, $MgPuO_3$, $CdPuO_3$, $PdPuO_3$ and $BePuO_3$ failed inspire of favourable factor in some cases. In synthesis of $SrFeO_3$ and $CaPuO_3$, the structures were obtained, which could not be interpreted. Compositions $BaPuO_3$, $PuVO_3$, $PuCrO_3$, $PuAlO_3$ and $PuMnO_3$ were synthesized and interpreted. However, the last two compositions had a rather low melting temperature.

The calculation study results for BN-800 reactor core, when using the fuel compositions indicated, are presented in Table V.

The data in Table V shows that neutronic parameters of cores with different fuel compositions of perovskyte type are rather similar to each other. In our opinion, the implementation of these compositions has good prospects.

Table V. Core parameters with a fuel material of perovskyte type.

Fuel material	$BaPuO_3$	$PuCrO_3$	$PuVO_3$	$PuMnO_3$	$MgPuO_4$
Doppler-effect, pcm	-116,0	-176,2	-167,0	-159,3	-158,4
SVRE, pcm	-2512	-3622	-3801	-3754	-3214
Reactivity change rate during interval, pcm	6215	5548	5934	5819	5745
Max linear power, kW/m	38.8	39.5	39.9	38.8	40.1
A fraction of burned plutonium from initial loading, %	24.5	26.2	26.3	26.5	25.1

The analysis results of the use of cores with such type fuel for MA burning are presented in Table VI.

The analysis results show that the fuel compositions of perovskyte type considered provide practically the same MA transmutation efficiency and a rather high rate of their destroying. All this allows the conclusion on a good potential for these compositions implementation as a fuel material of cores without uranium-238. However, rather intensive studies are needed not only in neutronic, but radiation parameters as well of these fuel composition. Besides, the addition to these compositions of corresponding metal oxides (for example, BaO for $BaPuO_3$ composition) allows the transition to the traditional fuel pin design.

Table VI. Core characteristics with a fuel material of perovskyte type for MA burning.

Fuel material	Ba(Pu+MA) O_3	(Pu+MA) CrO_3	(Pu+MA) VO_3	(Pu+MA) MnO_3	Mg(Pu+MA) O_4
Actinide fraction in the fuel	25	25	25	25	25
Doppler-effect, pcm	-54,0	-87,0	-70,0	-55,0	-89,0
SVRE, pcm	+410	+450	+456	+256	+320
Reactivity change rate during interval, pcm	5530	5650	5620	5225	5310
Max linear power, kW/m	38.8	39.5	39.9	38.8	40.1
Quantity of transmuted MA, kg/TW*h	30.7	29.9	30.2	30.5	30.7
A fraction of transmuted MA from initial loading, %	29.3	28.6	29.1	29.2	29.3

5.2. THE USE OF CERMET TYPE COMPOSITIONS.

Composition of CERMET type are obtained, when using anyone metal as an inert matrix material. The works under CAPRA program have shown that a possibility exists for creation of PuO_2+Me compositions, in which Cr, V and W can serve as a metal matrix (Me). All these metals have rather good radiation parameters. Experimental studies were carried out in property studies for PuO_2+W composition, and a principal possibility was shown for manufacture of this fuel composition with a rather high density (more than 90% of theoretical).

The calculation results for core neutronic characteristics when using metal matrices are presented in Table VII.

Table VII. Core characteristics when using metals as inert matrix.

Matrix material	Cr	V	W	C
Doppler-effect, pcm	-187,5	-196,7	-236,5	-396,3
SVRE, pcm	-3612	-3644	+2499	-3456
Reactivity change rate during interval, pcm	5012	5015	3534	5766
Max linear power, kW/m	43.8	44.0	45.8	44.2
A fraction of burned plutonium from initial loading, %	26.2	26.0	15.4	30.0

It is seen from the Table VII results that W can not be called an inert material, since it effects essentially on the neutron spectrum, which leads to a large increase in sodium void reactivity effect. The results for the use of graphite as a matrix material are given in fourth column of Table VII. Neutron spectrum softening, caused by introduction of an effective moderator (as was noted many times) led to increase in Doppler-effect almost twice, as compared with other metal matrices. This matrix ensures also a maximum plutonium burning efficiency.

Neutronic characteristics of the cores with these matrices for MA burning are presented in Table VIII.

Table VIII. Core parameters when using a metal as inert matrices for minor actinide burning.

Fuel material	Cr	V	W	C
Actinide fraction in the fuel	15	15	15	20
Doppler-effect, pcm	-97,0	-101,1	-197,0	-145,0
SVRE, pcm	+210	+250	+2830	+30
Reactivity change rate during interval, pcm	7280	7290	4410	5960
Max linear power, kW/m	43.5	44.9	45.8	44.9
Quantity of transmuted MA, kg/TW*h	16.2	15.9	12.8	26.7
A fraction of transmuted MA from initial loading, %	27.0	27.0	14.2	33.6

From the standpoint of MA burning efficiency, as it was expected, the best results were provided by a graphite matrix. One should pay attention to the fact that the introduction of MA into a fuel composition with W matrix has a less effect on core neutronic parameters as compared with the use of fuel composition with other type of matrices.

5.3. IMPLEMENTATION OF CERCER TYPE COMPOUNDS.

Among different ceramic materials, the following compositions are considered for the use as possible fuel compositions: MgO, Al_2O_3, CeO_2, $MgAl_2O_4$, SiC, ZrC, ZrN, TiN etc.

By now, PuO_2+MgO composition properties have been well studied, the technology having been developed by IPPE. Irradiation of this composition specimens in BOR-60 is planned. The experimental studies of different ceramic materials irradiated in reactors EBR II, Phenix, HFR etc. have demonstrated good radiation characteristics of MgO, MgAl2O4, TiN, ZrN.

In our studies we will limit ourselves to a comparison of BN-800 reactor parameters when using a fuel with MgO, $MgAl_2O_4$, TiN and ZrN inert matrices. We will consider additionally inert matrices from spinels, similar to magnesium spinel (MgO+Al_2O_3): $NiFe_2O_4$ and $MgFe_2O_4$ with a high iron and nickel content to enhance the Doppler-effect. The calculation study results are presented in Table IX

Table IX. Core neutronic parameters when using ceramic inert matrices.

Fuel material	PuO_2			PuN	
Matrix material	$MgAl_2O_4$	$MgFe_2O_4$	$NiFe_2O_4$	TiN	ZrN
Doppler-effect, pcm	-314,2	-350,4	-373,9	-287,6	-349,9
SVRE, pcm	-2439	-2667	-2334	-2412	-2022
Reactivity change rate during interval, pcm	6834	6009	6434	5765	5756
Max linear power, kW/m	42.5	45.7	44.7	42.5	43.0
A fraction of burned plutonium from initial loading, %	29.0	28.0	27.2	24.7	24.9

The results presented show, that the use of different ceramic matrices gives practically the same results in assessment of reactor neutronic parameters. We note a rather high Doppler-effect value, which is provided when using this type of matrices. However, the composition with nitride fuel provide a somewhat less plutonium burning capability.

The analysis results of a possibility for MA burning are presented in Table X.

Table X. Core neutronic parameters when using ceramic inert matrices.

Fuel material	$(Pu+MA)O_2$			(Pu+MA)N	
Matrix material	$MgAl_2O_4$	$MgFe_2O_4$	$NiFe_2O_4$	TiN	ZrN
Actinide fraction in the fuel	20	20	20	20	15
Doppler-effect, pcm	-121,0	-140,8	-120,0	-62,0	-150,0
SVRE, pcm	+260	+423	+445	+380	+160
Reactivity change rate during interval, pcm	6030	6110	5910	4800	6280
Max linear power, kW/m	42.9	45.9	45.3	43.0	43.3
Quantity of transmuted MA, kg/TW*h	25.8	25.1	24.5	31.7	16.7
A fraction of transmuted MA from initial loading, %	32.5	30.8	29.8	27.7	26.5

5.4. IMPLEMENTATION OF CERAMIC MATRICES WITH RESONANCE ABSORBERS.

One of lines of studies in a possibility for Doppler-effect increase in cores without uranium-238, when using ceramic matrices, was a possibility study to introduce into the fuel composition of some resonance absorbers, which could effect on the neutron spectrum and to provide in absence of uranium a rather high Doppler-effect value.

We will consider iron, nickel, chromium and niobium as such absorbers. The calculation results are presented in Table XI

Table XI. An effect of resonance absorbers on BN-800 reactor core neutronic parameters for plutonium burning.

Matrix material	MgO	MgO+ Fe	MgO+ Nb	MgO+ Ni	MgO+ Cr
Doppler-effect, pcm	-310,6	-352,5	-351,5	-351,0	-195,7
SVRE, pcm	-2216	-2242	+3366	-2514	-1908
Reactivity change rate during interval, pcm	6915	6111	3845	5866	6001
Max linear power, kW/m	45.1	45.7	45.6	44.9	45.7
A fraction of burned plutonium from initial loading, %	29.0	26.1	16.8	25.0	26.3

It is seen from the table data that the introduction of such resonance absorbers as iron, nickel and niobium leads to Doppler-effect increase and in this case a fuel effect component exceeds 80% of the integral effect. However, the introduction of large quantity of niobium (~60% of fuel pin volume) leads to essential increase in the void effect. It is noteworthy to pay attention to the fact that the introduction of resonance absorbers decreases somewhat a relative plutonium burning efficiency.

The calculation results for a core with MA are presented in Table XII.

Table XII. An effect of resonance absorbers on neutronic parameters of BN-800 reactor for MA burning.

Matrix material	MgO	MgO+ Fe	MgO+ Nb	MgO+ Ni	MgO+ Cr
Actinide fraction in the fuel	20	15	15	15	15
Doppler-effect, pcm	-121,0	-195,0	-276,0	-116,0	-107,0
SVRE, pcm	+260	+172	+3760	+171	+102
Reactivity change rate during interval, pcm	6030	6620	1220	6120	6650
Max linear power, kW/m	45.8	46.3	45.8	45.8	46.1
Quantity of transmuted MA, kg/TW*h	25.8	16.6	13.4	16.5	16.5
A fraction of transmuted MA from initial loading, %	32.5	25.4	16.6	26.2	27.9

The introduction of resonance absorbers into a fuel composition with inert matrix from magnesium oxide leads to some SVRE increase, which limits a MA fraction introduced into fuel. Besides, the introduction of resonance absorbers leads to spectrum hardening, which leads in its turn to decrease in relative MA transmutation efficiency.

CONCLUSIONS

A brief analysis performed of a possibility for implementation of different fuel compositions in reactor cores with fuel free of uranium-238, designed for efficient burning of plutonium and minor actinides, allows the following preliminary conclusions.

From the physical standpoint, a rather wide choice exists of fuel compositions without uranium-238, both pure plutonium – plutonium nitride, compositions of perovskyte type, intermetallic compounds etc. and diluted by inert matrices. The implementation of pure fuel compositions, due to necessity to meet the heat engineering requirement, leads to a need for development of new ring type fuel pin. Materials with inert matrices can be used in fuel pins of traditional design.

Noteworthy also that among different fuel compositions the most interesting are plutonium nitride (with enriched (by N^{15}) or natural nitrogen), compositions of perovskyte type, compositions with plutonium oxide or plutonium nitride, located in inert matrices with addition of resonance absorbers to solve the problem of fuel Doppler reactivity coefficient increase. Among resonance absorbers, iron and nickel attract the greatest interest.

From the standpoint of MA burning the most efficient are compositions containing a large amount of moderators.

Besides, it is of interest to develop operation regimes for such type reactor, which could provide a more deep fuel burn up.

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