





Research Article

On the possibility to improve mixed uraniumplutonium fuel in fast reactors

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Abstract

Accumulation of plutonium extracted from the spent nuclear fuel (SNF) of light water reactors is one of the central problems in nuclear power. To reduce out-of-the-reactor Pu inventory, leading nuclear power countries (France, Japan) use plutonium in light water power reactors in the form of MOX fuel, with half of Pu fissioning in this fuel. The rest of Pu cannot be reused easily and efficiently in light water reactors because of the high content of even isotopes. Plutonium for which there are no potential consumers is accumulated.

Unlike thermal reactors, fast reactors take plutonium of any isotopic composition. That makes it possible to improve plutonium isotopic composition and to reduce the fraction of even isotopes to the level that allows reuse of such plutonium in thermal reactors.

The idea of changing the isotopic composition of Pu in fast reactors is well-known. The originality of the research lies in applying this idea to combine the fuel cycles of fast and thermal reactors. Pu isotopic composition can be improved by combining certain operational activities in order to supply fuel to thermal and fast reactors. Scientific and technological justification of the possibility will let Russian BN technologies and French MOX fuel technologies work in synergy with thermal reactors.

Keywords

Control of plutonium isotopic composition, fast reactors, light water reactors (pressurized water reactors – PWRs), thermal reactors, improvement of plutonium isotopic composition, BN technologies

Introduction

Nowadays, global nuclear power rests on thermal light water reactors (pressurized water reactors) and uranium fuel (based on enriched uranium). Uranium is characterized by low-level radioactivity, which makes it possible to fabricate uranium fuel using manual technologies. After the fuel lifetime is spent in a power reactor, such fuel contains fission products and high-radiation plutonium consisting of five isotopes. Fission products (after their separation and required cooling decay time to reduce the radioactivity level) are immobilized and disposed of and such a technology has been developed in many countries. Plutonium is highly radioactive, with long radioactive half-life and high heat release. Standard technologies and manual operations are not possible when managing such materials; remote technologies are, therefore, required. For a number of reasons, vitrification of materials that contain plutonium is impossible as well. One of the reasons would be, for example, generation of helium during alpha-decay of plutonium, which contributes to fracture of vitreous mass. The most sensible technology solution to the problem of plutonium utilization is to use it as a fuel component both for thermal and fast reactors.

Proposal to improve the isotopic composition of plutonium

Rosatom's leadership has laid down the concept of two-component nuclear power based on the simultaneous operation of thermal and fast reactors, which is to be developed in Russia in the 21st century (Ponomarev-Stepnoy et al. 2016). More specifically, the concept suggests that, with intensive development of nuclear power, fast reactors will provide thermal reactors with fuel. No export of fast reactors or fuel cycle technologies is supposed to other countries, so there is no proliferation problem to face. In addition, plutonium extracted from the spent nuclear fuel (SNF) of PWRs will be used to fabricate mixed uranium-plutonium (MOX) fuel for sodium cooled fast reactors. Due to it, storages of PWR SNF become empty and plutonium extracted from the SNF of PWRs is concentrated in storages of SNF from fast reactors (Pu content in fast reactors is ~20 times higher than in thermal reactors). Such a strategy was also evaluated by EDF R&D in 2012 (Lemasson et al. 2012).

At the same time, absence of operating fast reactors in global nuclear power resulted in taking an interim decision to use MOX fuel for commercial light water reactors in operation. Over 40 PWR power units in Europe alone (France, Belgium, Switzerland, Germany) have a license for combined use of conventional and MOX fuel (World Nuclear Association 2017), and another 30 power units are expecting to obtain it. MOX fuel is fabricated from plutonium extracted from uranium SNF after the required cooling decay time in spent fuel storage pools. Remote technologies, which have been mastered in France (the Melox plant), for example, are used for fuel fabrication (Grouiller et al. 2017, Martsinkevich 2017, OECD/NEA 2007).

MOX fuel extracted from uranium SNF and used in light water reactors helps to reduce the total amount of plutonium to be stored by a quarter. However, reuse of plutonium from MOX SNF after irradiation in light water reactors already leads to serious changes in the isotopic composition due to an exceedingly high content of heavy (even) isotopes that do not fission in the thermal-neutron spectrum. It can also be shown that a high even plutonium isotope content causes problems of reactivity control in light water reactors (Hesketh 1995, Camarcat et al. 2019). As a result, plutonium unsuitable for use in MOX fuel of thermal reactors is produced (Shoshina et al. 2007, Grouiller et al. 2017), which hampers solution to the problem of Pu accumulation (Figure 1).

Unlike thermal reactors, fast reactors take plutonium of any isotopic composition. Even isotopes (Pu-238, Pu-240, Pu-242) fission in the fast-neutron spectrum, though not so effectively as odd (Pu-239, Pu-241) isotopes do (Shoshina et al. 2007), their content tends to the so-called "equilibrium". If a fast reactor is loaded with fuel based on plutonium with a high content of even isotopes (unsuitable for a thermal reactor), the isotopic composition of such plutonium

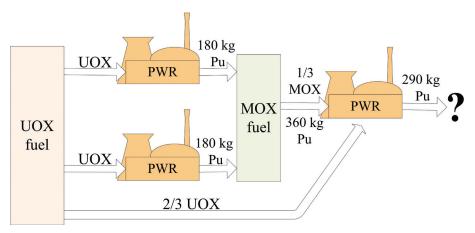


Figure 1. Utilization of Pu in thermal reactors.

will undergo a change for the better, i.e. to change in the right direction or improve, after irradiation in the fast-neutron spectrum and can become suitable for reuse in thermal reactors. If low-radiation plutonium is used, the content of even isotopes in it will increase after irradiation in the fast reactor. The idea to control the isotopic composition of plutonium by means of its irradiation in fast reactors (taking the ASTRID reactor as an example) was put forward by French specialists in 2017 (Figure 2) (Grouiller et al. 2017).

Sodium cooled fast power reactors are currently operating only in Russia. These are the BN-600 and BN-800 reactors (Matveev et al. 2012, Bagdasarov et al. 2001, Textbook 2012, Seleznev 2013). Use of MOX fuel in these reactors has been justified, and representative experimental batches of such fuel (over 30 pelletized fuel assemblies (FAs) and as many vibrocompacted FAs) have been irradiated in BN-600. The number of MOX FAs irradiated in BN-800 exceeds 100. However, this fuel was fabricated from low-radiation plutonium by using manual technologies. Remote technologies to fabricate MOX fuel for fast reactors are being developed at a plant belonging to the Mining and Chemical Complex (the town of Zheleznogorsk, Krasnoyarsk territory) in Russia.

Therefore, with the existing fast reactors (Russia) and available remote technologies for MOX fuel fabrication (France and Russia), plutonium which is so far unsuitable for reuse in thermal reactors can be improved. That does not only open up the world market of MOX fuel for reuse of plutonium in foreign thermal reactors with the help of Russian BN reactors but also ushers in a new era in the development of global nuclear power – the birth of its two-component structure based on a synergistic union of Russian BN technologies with European MOX fuel technologies.

The idea of improving mixed U-Pu fuel lies in combining operational activities in order to supply fuel to thermal and fast reactors. As already mentioned, a fast reactor aims at reducing Pu isotopic composition to equilibrium, when it remains unchanged despite Pu multiple recycle through the core. If there are more even isotopes in the isotopic composition of Pu from PWR SNF than in the equilibrium composition of a fast reactor, irradiation of Pu-containing fuel in the fast-neutron spectrum will lead to improvement. Adding Pu from fertile blankets can contribute significantly to the improvement of plutonium because such Pu is characterized by a high content of ²³⁹Pu isotope and is produced in the axial and radial blankets.

Let us illustrate the possibility to improve Pu by the example of the BN-800 reactor. For calculations, Russian «TRIGEX» diffusion code was used. The reactor criticality was assessed by using the MMKK precision code (continuous energy Monte Carlo method). The calculations suggested that only MOX fuel should be used in the BN-800 reactor (Vasiliev et al. 2016).

It is supposed to improve French plutonium extracted from the MOX SNF. Initial and final (after irradiation in BN-800) isotopic compositions of Pu are shown in Table 1:

- Pu extracted from the uranium SNF of PWR (Alyoshin et al. 2002);
- 2. Pu extracted from the MOX SNF of PWR (1/3 of the core is loaded with MOX fuel), which was irradiated in a PWR once and is no longer suitable for reuse (Camarcat et al. 2020).

Isotopic composition of Pu irradiated in BN-800 (with the fuel based on Pu extracted from the MOX SNF of PWR) is shown in column 3 (newly computed for this study).

The same isotopic composition of Pu from MOX SNF of PWR after it is irradiated in the ASTRID reactor is shown in column 4 (Grouiller et al. 2017, Venard et al. 2017).

Irradiation of Pu in BN-800 leads to decrease in the fraction of even isotopes (including Am241) from 50.7 to 47.2% and corresponding increase in the fraction of fissile (Pu-239, Pu-241) isotopes.

BN-800 requires ~2.3 tons of Pu a year, which is equal to the amount of Pu in MOX SNF of 8.5 PWR reactors (Ponomarev-Stepnoy 2016) whose core is only loaded with MOX fuel by a third. After irradiation in BN-800, the amount of improved Pu is a little less (~ by 14 kg). Extracted Pu could be used again to fabricate fuel for thermal reactors.

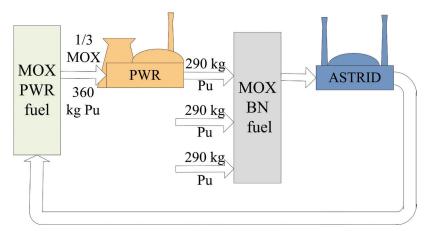


Figure 2. Recycle of plutonium in the fuel cycle of thermal reactors.

	PWR			BN-800	ASTRID	
Reactor	1	2	1	3	4	
	UOX SNF	MOX SNF		BN-800 SNF	ASTRID SNF	
Isotope	(Alyoshin et al. 2002)	(Camarcat et al. 2020)	STRID	(newly computed for this study)	(Grouiller et al. 2017,	
			S		Venard et al. 2017)	
Pu238, %	2.5	4.02]7	2.95	2.7	
Pu239, %	57.6	37.88	800	44.28	45.8	
Pu240, %	21.7	33.48	-8	31.77	33.6	
Pu241, %	11.6	11.42	M	8.47	6.0	
Pu242, %	5.7	12.04	E.	11.26	11.9	
Am241, %	0.9	1.16	tion	1.26	_	
Fraction of even	30.8	50.70	iat	47.24	48.2	
isotopes (+Am241)			radiat			
Pu loading, kg/year	0 (100%UOX)	360 (33%MOX)	Ē	2275	-	
				(7.8 PWR)		
Pu discharge, kg/year	180	290	1	2261	_	

Table 1. Illustration of the change in the isotopic composition of Pu being irradiated in BN-800.

Table 2. Pu isotopic composition in BN-800 zones.

Isotope	Initial Pu (MOX SNF of PWR)	Core	Bottom axial FB	Lateral FB	Total	
	(Camarcat et al. 2020)					
Pu238, %	4.02	3.17	0.02	0.06	2.95	
Pu239, %	37.88	40.63	95.45	91.88	44.28	
Pu240, %	33.48	33.70	4.32	7.53	31.77	
Pu241, %	11.42	9.07	0.21	0.49	8.47	
Pu242, %	12.04	12.08	0.00	0.01	11.26	
Am241, %	1.16	1.32	0.00	0.01	1.26	
Fraction of even isotopes	50.70	50.27	4.34	7.59	47.24	
(+Am241), %						

Let us consider the mechanism for improving plutonium. Not only fissile but also even Pu isotopes fission in fast reactors. Besides, an important feature of fast reactors is a smaller portion of Pu isotope radiation captures. As a result, the final composition of Pu (including Pu-239 formed from U-238) is improved (Table 2). Almost pure Pu-239 from fertile blankets, which mixes with Pu from the core when reprocessed, also contributes to the result.

Apart from reactor physics, there may also be technological limits in the fuel cycle plants when each of the 5 isotopes of plutonium reaches the value of the left hand side column of Table 2. In particular, the Pu-238 proportion is challenging for fuel fabrication due to thermal loading and alpha-irradiation that could degrade lubricant and pore formers. The global improvement of the BN-800 scheme in the right hand side column should be judged not only with respect to the even isotopes but also with respect to each component of the isotopic vector.

Conclusion

Accumulation of plutonium extracted from the SNF of light water reactors is one of the central problems in nuclear power. To reduce out-of-the-reactor Pu inventory, leading nuclear power countries (France, Japan) use plutonium in light water power reactors in the form of MOX fuel, with a significant part of Pu fissioning in this fuel. The rest of Pu cannot be reused in light water reactors because of the high content of even isotopes.

Plutonium of any isotopic composition can be used in fast reactors. That makes it possible to improve plutonium in principle, i.e. to improve its isotopic composition, and to reduce the fraction of even isotopes to the level that could allow reuse of such plutonium in thermal reactors. Further improvements should be made and other options will be studied, such as decreasing the content of the plutonium in the fuel loaded in the fast reactors, or turning to fast reactors with higher breeding ratios. The possibility also depends on the isotopic composition of initial Pu intended for improvement and on the requirements to the isotopic composition of plutonium for MOX fuel of thermal reactors.

Scientific and technological justification of the possibility will let Russian BN technologies and French MOX fuel technologies work in synergy with thermal reactors. That does not only open up the world market of MOX fuel for Russian nuclear technologies with their BN reactors that can improve plutonium in foreign thermal reactors but also ushers in a new era in the development of global nuclear power as a two-component structure (Baryshnikov 2016, 2017).

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