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New concept of designing Pu and MA containing fuel for fast reactors

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ABSTRACT

New type of metal base fuel element is suggested for fast reactors. Basic approach to fuel element development – separated operations of fabricating uranium meat fuel element and introducing into it Pu or MA dioxides powder, that results in minimizing dust forming operations in fuel element fabrication. According to new fuel element design a framework fuel element having a porous uranium alloy meat is filled with standard PuO_2 powder of <50 μm fractions prepared by pyrochemical or other methods. In this way a high uranium content fuel meat metallurgically bonded to cladding forms a heat conducting framework, pores of which contain PuO_2 powder. Framework fuel element having porous meat is fabricated by capillary impregnation method with the use of Zr eutectic matrix alloys, which provides metallurgical bond between fuel and cladding and protects it from interaction. As compared to MOX fuel the new one features high thermal conductivity, higher uranium content, hence, high conversion ratio does not interact with fuel cladding and is more environmentally clean. Its principle advantage is a simple production process that is easily realized remotely, feasibility of involving high background Pu and MA isotopes into closed nuclear fuel cycle at the minimal influence on environment.

1. Introduction

Currently as promising fuel for fast reactors two basic types of fuel are under consideration. The first one is metallic U–Pu–Zr fuel. The second one is MOX fuel $(U,Pu)O_2$ [1,2].

High-density U–Pu–Zr fuel features high thermal conductivity and a higher uranium content which provides for the high conversion ratio, negative reactivity factor and increases the passive safety of fast reactors. The metal fuel disadvantages comprise lower processability, fuel-cladding interaction, large swelling at high burn-up and the needed availability of sodium in a fuel-cladding gap.

MOX fuel is highly irradiation resistant and in terms of technology it has been more mastered. However, its low thermal conductivity restricts the heat flow in a fuel element and its insufficiently high density lowers down the major parameter of fast reactor cost effectiveness, i.e., the conversion ratio. Hence, MOX fuel is unable to provide the conversion of plutonium without a blanket. Besides, when using MOX fuel the interaction is observable between fission products and a cladding.

Therefore, the optimal solution might be to design novel combined U–PuO_2 fuel that would combine the favourable features of both fuel types as well as application of novel design and fabrication process [3].

2. Major approaches to designing combined U–PuO_2 (Metal-dioxide) fuel

The suggestion is to change over from the pelletized fuels of the container type to dispersion type fuel elements (Fig. 1) [3–5].

The dispersion fuel is known to have a high irradiation resistance and thermal conductivity as well as a metallurgical bond between cladding and fuel which not only protects a cladding against interaction with fuel and fission products but also lowers down the operating temperatures of fuel. The major drawback of dispersion fuel, viz., its low uranium content in this design is compensated for the applied high density uranium metal fuel and compatible with it zirconium alloy matrices [3,4].

In the suggested design metal fuel of U–Mo, U–Zr and U-Zr-Nb alloys forms a porous frame bonded by a zirconium matrix alloy; the pores of the frame contain PuO_2 powder manufactured by pyrochemical or other methods [3,4]. The fuel meat metallurgically bonded to a cladding promotes the high thermal conductivity to mixed U–PuO_2 fuel, while the Zr-base matrix alloy improves the compatibility between fuel components.

The process of the fuel element fabrication comprises two stages. At the first major stage of production under conventional conditions of a plant a fuel element frame is fabricated from uranium alloy particles bonded with a zirconium matrix. At the second final stage of the fuel rod fabrication under protective conditions through the fuel skeleton the standard PuO_2 powder is filled and after that a fuel element is sealed. In this way the dust

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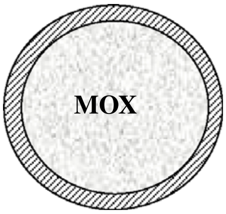
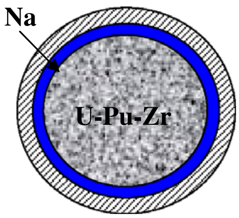
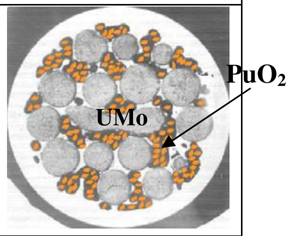
Parameters	MOX (U, Pu)O_2	**U-20Pu-10Zr** alloy	**Combined** **U-PuO_2 fuel**
Fuel element design	Container	Container	Dispersion
			
Effective density of h.a. under fuel cladding, ρ^{eff}, g/cm^3	8.3-8.5	11.0	10.8-11.0
Maximal temperature of fuel, ℃	2000-2250 (IFR, EFR)	<800 (IFR)	790 (BN-K)
Fuel (fission product) - cladding interaction	Mean	High	No interaction

Fig. 1. Design of container type fuels (MOX and U–20Pu–10Zr metal fuel) and novel combined U–PuO_2 dispersion fuels for fast reactors [3,4].

forming operation with Pu are minimized while the processability and ecology of the production are enhanced.

3. Characteristics of fuel meat

The initial components of the combined fuel are granules of depleted uranium alloys, PuO_2 powder and granules of zirconium matrix alloys. The characteristics of novel fuel and fuel composition are tabulated in Tables 1 and 2; the appearance the original components is illustrated in Fig. 2.

It is suggested to use as fuel the high density U alloys of U–Mo (from 1.5% to 9% Mo), U–Zr (from 5% to 10% Zr), U–Zr–Nb (from 5% to 10% Zr and Nb in total) systems as well as alloys of U–Mo with carbon in which carbon as an impurity forms carbide phases in the structure, enhances the irradiation resistance and compatibility between fuel and cladding. The sizes of the fuel granules range from 0.5 to 1.2 mm [4,5].

Table 1
Characteristics of combined U–PuO_2 fuel components [3,4].

	Components	Granules size (μm)	Temperature of melting (°C)
Metal fuel	U–(3–9)Mo	500–1200	1200–1300
	U–(5–10)Zr		
	U–(2–5)Zr,Nb		
	U–(3–9)Mo-(0.1–0.6)C		
Zr-matrix alloy	Zr–(1.5–2.5)Be–(4–7)Fe	100–300	800
	Zr–(6–12)Fe–(6–12)Cu		860–900
PuO_2		20–70	2200

Zirconium eutectic alloys are used as matrices [4,5]. The sizes of the matrix granules are 0.1–0.3 mm.

At the first stage frame fuel having porous meat is fabricated by the capillary impregnation method. Granules of fuel (U–Mo, U–Zr or U–Zr–Nb alloys) of depleted uranium and of a matrix are loaded into a fuel element cladding and the fuel element is heated to a temperature 50 °C higher than the melting temperature of a matrix (Fig. 3(a) and (b)) [3–5].

The matrix alloy melts down and under capillary forces moves into gaps between fuel components to form metallurgical bonds. This technology provides the formation of controllable open porosity from 30% to 40% within the resultant frame fuel element, to further accommodates oxides of plutonium and MA. A layer of a Zr matrix alloy that is available at the inner surface of a fuel cladding protects it against cesium induced corrosion which was confirmed by testing steel claddings in a Cs oxide environment at 700 °C for 1000 h. Since zirconium forms the base of the matrix alloys the alloys are compatible with high uranium content fuel both upon fabricating fuel elements and after long-term isothermal anneals of fuel compositions at 750 °C for 6000 h [3,4].

After fuel element fabrication the melting temperature of the Zr matrix alloys increases over 200–300° as its composition alters due to the ingress of components from cladding and fuel. Since the Zr matrix alloys are deep ternary and quaternary eutectics any change in the alloy composition leads to a drastic rise of its melting temperature. As a result the alloy melting temperature increases, the alloy solidifies and the further interaction stops.

This was confirmed by annealing a fuel element at 1000 °C for 30 min without any changes in the fuel dimensions and structure, as well as by in-pile irradiation of the matrix material in contact with the stainless steel cladding at 750 °C. This special design fuel

Table 2
Characteristics of combined U–PuO_2 fuel [3].

Components of fuel composition	Volume fraction (%)	U(Pu) content under fuel cladding (g/cm^3)	Temperature of melting after manufacture (°C)
Metal fuel	55–60	9–10	1300–1400
Zr-matrix alloy	7–15		1150–1250
PuO_2	10–20	0.9–1.8	2200
Pores	10–15 (30–40 without PuO_2)		

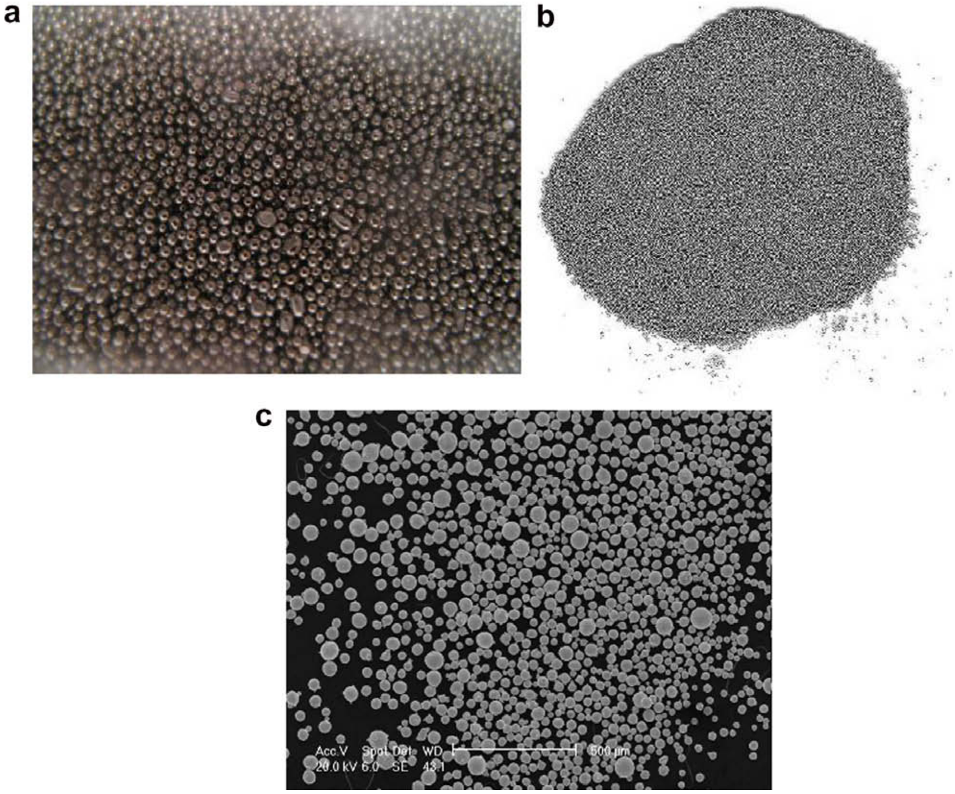


Fig. 2. Appearance of initial components of combined U–PuO_2 fuel, (a) U–Mo alloy granules, (b) Zr–Fe–Cu alloy matrix granules, (c) PuO_2 powder manufactured by pyrochemical method [3,5,6].

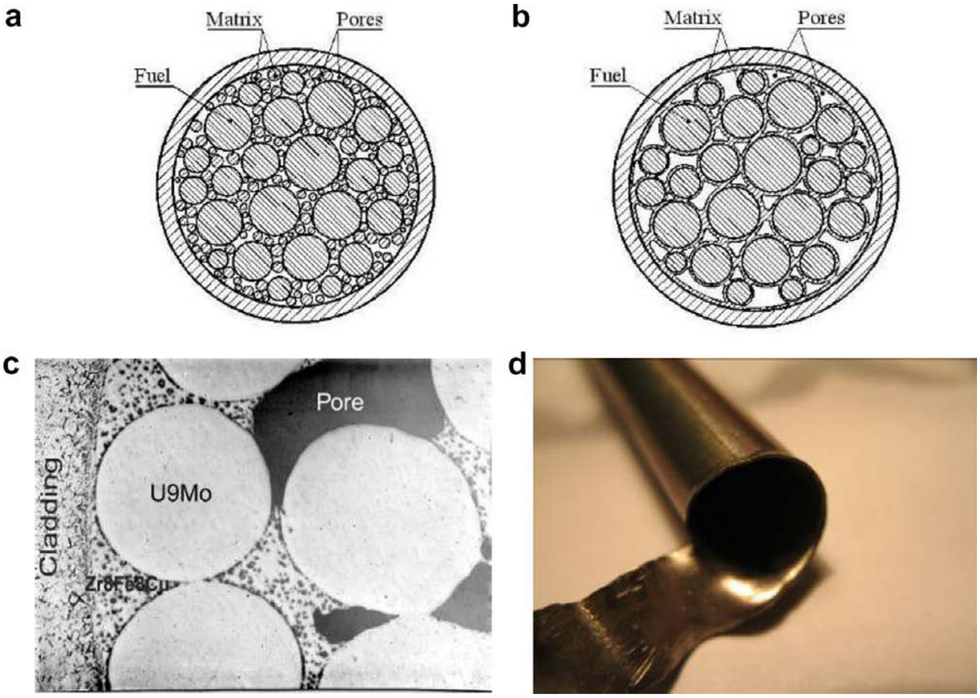


Fig. 3. Schematic presentation of fuel element cross-section, (a) as vibropacked, (b) as capillary impregnated, (c) structure of fuel meat, (d) internal zirconium alloy coat at inner surface of steel cladding [3,5].

element for Pu incineration reached the burn-up of 1.5 g fiss-fragm/cm^3 under the fuel element cladding with the temperature of steam up to 600 °C [3,4].

At the second stage of the fuel element fabrication the powder of PuO_2 is filled into the frame (Fig. 4) [3]. The PuO_2 powder is manufactured by the already mastered methods, viz., the

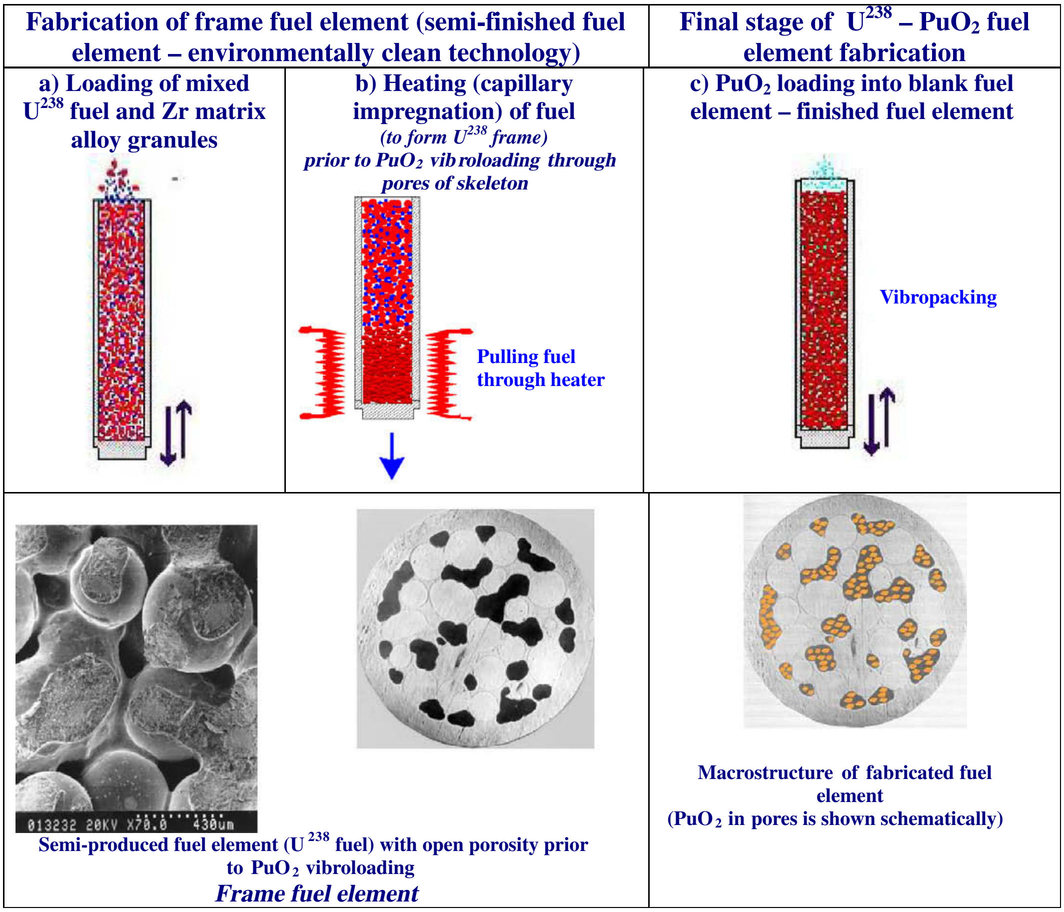


Fig. 4. U–PuO_2 dispersion combined fuel in place of MOX and fabrication stages [3].

pyrochemical one [6] or using the GRANAT type process etc. The sizes of the powder particles are 0.02–0.07 mm. Then the second plug is welded up, and the finished fuel elements are subjected to the instrumental control.

In this method of fabrication the dust forming technological operations minimized and all benefits of dispersion type fuel – (high uranium density, low fuel temperature, workability in transients, high burn-up) – will remain. It is one of the options of closing fuel cycle.

At 500–800 °C the thermal conductivity of the porous fuel meat makes up 20–35 W/m deg and increases with the temperature. Depending on the alloy used and its volume fraction the content of uranium per a unit fraction under the fuel cladding makes up 9–10 g/cm^3 that together with PuO_2 in the fuel element gives the effective density of heavy atoms up to 11 g/cm^3.

4. Comparative characteristics and advantages of novel fuel

Comparative characteristics of fuel are shown in Table 3, taken from INL and DOE review on promising fuel for fast reactors – MOX and metallic U–Pu–Zr fuel. We have introduced into the table the version of new alternative combined U–PuO_2 fuel.

MOX fuel has a high irradiation resistance but low thermal conductivity and a relatively low uranium content which decreases the main parameter of fast reactor – conversion ratio. The U–Pu–Zr metal fuel having high uranium content interacts with a fuel cladding and is more intricate in terms of the fabrication technology.

The suggested U–PuO_2 combined fuel retains the advantages inherent in metallic and ceramic types of fuel. Since the contribution into the total burn-up is made not only by the metallic fuel frame at the final irradiation stage as plutonium is generated, but also by plutonium dioxide at the initial irradiation stage, the total swelling of the metallic fuel frame is reduced due to a less build-up of fission fragments in it.

Novel fuel having a high conversion factor and thermal conductivity like metallic fuel does not interact with a fuel cladding since Zr matrix coats the cladding and protects against fuel-cladding interaction. Fabrication technology is simple and environment-friendly.

Table 3
Comparative characteristics of various fuel versions for fast reactors [1,3].

Parameters	MOX $(U,Pu)O_2$	U–20Pu–10Zr	UMo–PuO_2
Content of fuel in fuel element (% of theoretical)	80–85%	75%	75% (55% + 20%)
Fuel – cladding gap	0.1 mm He	0.7 mm Na	Metallurgical bond
U+Pu content under fuel cladding	8.3 g/cm^3	11.0 g/cm^3	11.0 g/cm^3
Thermal conductivity of fuel (W/m deg)	2–4	15–20	20–30
Adaptability of fuel element fabrication	Mean	Mean	High
Environmentally clean production	Low	Low	High

4.1. Major advantages of novel combined U–PuO_2 fuel

High contents of U and Pu compared to MOX fuel and, hence, high conversion ratio which makes it feasible to close nuclear fuel cycle.

Lower damage of fuel by fission products and, hence, lower swelling compared to that of U–Pu–Zr fuel.

Novel fuel is dispersion type fuel, therefore the existence of metallurgical bond between fuel and cladding not only decrease fuel temperature, but also protects fuel cladding from interaction with fuel and fission products.

High fabrication adaptability and ecology of production – actually the main part of fuel element fabrication is carried on under conventional conditions and only the final operation of fuel element fabrication needs remote implementation. In this case PuO_2 is used as a powder not as pellets. All this minimizes process operations with Pu and makes the fuel element fabrication environmentally clean.

5. Conclusion

For fast reactors a novel type of promising combined U–PuO_2 fuel is proposed that is based on dispersion fuel elements. Novel fuel features higher characteristics in comparison to metallic or MOX fuel its fabrication technology is readily accomplished and is environmentally clean.

References

[1] Dauglas Crawford et al., J. Nucl. Mater. 371 (2007) 202.
[2] IAEA-TECDOC-1374, 2003.
[3] A.M. Savchenko, I.I. Konovalov, A.V. Vatulin, O.I. Uferov, E.M. Glagovsky, New generation fuel element for different purpose power reactors, In: Proceedings of All-Russia Conference MAYAT-2007, Zvenigorod, 19–22 October 2007.
[4] A. Savchenko, I. Konovalov, A. Vatulin, A. Morozov, V. Orlov, O. Uferov, et al., J. Nucl. Mater. 362 (2007) 356.
[5] A.M. Savchenko, A.V. Vatulin, A.V. Morozov, I.V. Dobrikova, S.A. Ershov, S.V. Maranchak, Z.N. Petrova, Y.V. Konovalov, J. Nucl. Mater. 352 (2006) 334.
[6] A. Bichkov, O. Skiba V. Kisly, M. Kormilitsin, Experience in closing nuclear fuel cycle developments, In: Proceedings of All-Russia Conference MAYAT-2007, Zvenigorod, 19–22 October 2007.