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Zirconia inert matrix for plutonium utilisation and minor actinides disposition in reactors

Claude Degueldre *

PSI, 5232 Villigen, Switzerland

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Abstract

The radiotoxicity of the UO_2 spent fuel is dominated by plutonium and minor actinides (MA): Np, Am and Cm, after decay of the short life fission products. Zirconia ceramics containing Pu and MA in the form of an inert matrix fuel (IMF) could be used to burn these actinides in light water reactors or in high temperature reactors. Optimisation of the fuel designs dictated by properties such as thermal, mechanical, chemical and physical must be performed with attention for their behaviour under irradiation. Zirconia must be stabilised by yttria to form a solid solution such as $MA_zY_yPu_xZr_{1-y}O_{2-\zeta}$ where minor actinide oxides are also soluble. MA may act as a burnable poison reducing the reactivity at the beginning of life and yielding fertile nuclides improving the reactivity at the end of life. These zirconia cubic solid solutions are stable under heavy ion irradiation. The retention of fission products in zirconia, under similar thermodynamic conditions, is a priori stronger, compared to UO_2, the lattice parameter being larger for UO_2 than for $(Y,Zr)O_{2-x}$. $(Er,Y,Pu,Zr)O_{2-x}$ in which Pu contains 5% Am was successfully irradiated in the Proteus reactor at PSI, in the HFR facility, Petten as well as in the Halden reactor. These tests support potential irradiations of such IMF in a commercial reactor. This would allow later a commercial deployment of such a zirconia fuel for Pu and MA utilisation in a last cycle. The fuel forms namely pellet-fuel, cercer, cermet or coated particle fuel are discussed considering the once through strategy. For this strategy, low solubility of the inert matrix is required for geological disposal. As spent fuels these IMFs must be excellent materials from the solubility point of view, this parameter was studied in detail for a range of solutions corresponding to groundwater under near field conditions. Under these conditions the IMF solubility is about 10^6 times smaller than glass, which makes the zirconia material very attractive for deep geological disposal. The desired objective would be to use IMF to produce energy in reactors, opting for an economical and ecological solution.

Keywords: Inert matrix fuel; Plutonium; Minor actinide; Transmutation

1. Introduction

During the last 10 years, several research organisations have devoted work to the transmutation of plutonium and minor actinides (MA: Np, Am, Cm) in thermal reactors by applying an inert matrix fuel (IMF) concept, e.g. [1–3] based on uranium-free fuel studies, e.g. [4–6]. This situation is a consequence of the aim of eliminating the current excesses of plutonium and minor actinides. These efforts are made because of the energetic value of plutonium and of the MA's, the Pu proliferation risks and also because these nuclides contribute to the largest radiotoxicities of conventional spent fuels after some 100 years.

IMF projects initially dealt with excess plutonium produced as a result of civil nuclear electricity production. In addition, discontinuation of the nuclear weapon programs and increasing production of reprocessed plutonium from the electronuclear programs yield surplus in the stocks of the order of 200 tonnes weapon grade plutonium and 1000 tonnes civilian plutonium, respectively, at the end of the last century. In addition several tonnes of MA's are currently estimated for the world nuclear park.

The management of plutonium and MA's inventories may have suffered a major setback due to the postponement or abandonment of fast breeder reactor programmes. In effect, the former goal to produce more plutonium had to change to a plutonium reduction strategy while some countries continue to produce these actinides to use them in the next generation of reactors. However, energy production remains the most desirable disposition and to solve the problem of plutonium surplus in

* Tel.: +41 56 3104176.
E-mail address: claude.degueldre@psi.ch.

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the short and medium term it is suggested to burn [2], as quickly and completely as possible, excess plutonium in existing light and heavy water reactors (L&HWRs). Fast reactors (FR) have also been considered while their capacity is restricted for pragmatic utilisation. Since today's practice of mixed oxide (MOX) fuelling in LWR (with up-to 40% core loading) does not allow a real reduction of plutonium and MA stockpiles, the replacement of uranium dioxide by an inert matrix and an extension of the nuclear fuel cycle have been recommended.

Clearly, the current rebirth of such programmes worldwide has taken place in a new context. A new generation of IMF activities gained momentum with the first Inert Matrix Workshop held at the Paul Scherrer Institute beginning in September 1995 followed by other IMF workshops elsewhere [7–10]. The IMF philosophy and the specific scientific programs were presented and discussed in an IAEA TECDOC report [11].

This study represents a comprehensive summary of the recent activities of the organisations directly or indirectly involved in the "Initiative for Inert Matrix Fuel". The contributions and country profiles are reviewed on the basis of studies that are today integrated into national or international projects. This paper presents an overview of the philosophy and current directions of IMF in the nuclear fuel cycle.

2. Zirconia-based IMF candidates

The original aim of IMF development was the destruction of plutonium. Consequently one has to eliminate the source of these nuclide productions in the fuel that corresponds to the original concept of uranium-free fuel. The inert matrix fuel concept proceeds one step further: the elements and/or isotopes of the inert matrix are selected according to their transparency for neutrons.

Until recently, the basic strategy for IMF application has been to use a once-through irradiation prior to geological disposal. However, the IMF concept may also be extended by using additional fuel recycling steps. Multi-reprocessing is particularly compatible with IMF when the inert matrix can be partitioned from the actinides. The IMF concept has thus been extended for use in 'sustainable' fuel cycles proposed for fourth generation nuclear power plants. In order to define IMF candidates that meet all requirements for a particular fuel cycle, the properties are investigated systematically.

In order to select suitable IMF candidates, the following factors are initially considered:

- neutronics properties (reactivity effects),
- the compatibility of inert matrix components with coolant and structural materials,
- the fissile vector (first generation, second generation or weapon grade Pu, or Am) produced, and
- in advanced concepts, a suitable burnable poison and/or a fertile additive introduced in order to improve the neutronic characteristics of the fuel.

The first requirement in the material selection is guided by the neutronic properties. If the neutronic requirements of a particular reactor type cannot be met or are severely compromised by an element it is excluded. Next, the physico-chemical properties of the material are considered. These are of prime importance for understanding the fuel behaviour in reactor. The desired properties of the material(s) are: high melting point, good thermal conductivity, good compatibility with the cladding, low solubility in the coolant, good mechanical properties and high density. In all cases, the IMF candidate is compared to UO_2 as the reference. Material screening studies should be first carried out on the basis of literature reviews and knowledge gained in spent fuel and waste management studies but new experimental work are required. A list of candidate IMF materials for various applications is given in Table 1. Included are elements (metals, carbon), oxides, nitrides and carbides.

Table 1
Examples of inert matrix, additive candidates and their design as considered in this study

Inert matrix components	Inert matrix formula
Elements	C, Al, Si, Zr
Carbides	SiC, TiC, ZrC
Binary oxides	MgO, CaO, Y_2O_3, ZrO_2
Oxide solid solutions	$Ca_xZr_{1-x}O_{2-x}$, $Y_yZr_{1-y}O_{2-y/2}$
Additive type	**Additive formula**
Burnable poison	B, Gd, Er or Np or Am
Stabiliser	Y_2O_3, MgO in ZrO_2
Design	**Example**
Solide solution	$Y_yPu_xZr_{1-y}O_{2-y/2}$ [a]
Cercer	MgO–$Y_yPu_xZr_{1-y}O_{2-y/2}$ [a]
Cermet	Zr–$Y_yPu_xZr_{1-y}O_{2-y/2}$ [a]

[a] Fissile material phase.

The addition of burnable poisons is dictated by the neutronic requirements; these can be used to make the neutronic reactivity as constant as possible with values somewhat above 1.0. In addition, for safety reasons, small quantities of resonant absorber may be added to the fuel. Finally, a stabiliser may be used to fully stabilise the solid solution in an adequate thermodynamic phase. Table 1 shows some IMF additive types and formula examples.

The evaluation process has been carried out for a range of matrix candidates, and the initial IMF selection process for burning plutonium in the current fleet of water-cooled reactors has been largely completed based on these scoping studies.

3. The potential of zirconia IMF

An important criterion, for the choice of materials for IMFs, which needs to be explicitly mentioned, is that of the destination of the burnt fuel. Here: the spent IMF is sent for geological storage. Such a choice can be envisaged because the residual Pu is of very poor quality following the large reduction of the fissile isotopes (^{239}Pu and ^{241}Pu). The consequences of this choice are: the combustion of Pu must be as high as possible, in order to reduce the toxicity and the heat source in the storage, and the material which forms the IMF must be chemically very stable, such as to satisfy the criteria for long term storage (adequate immobilisation of radioactivity in geological disposal).

Commercial L&HWRs are the only worldwide available candidate facilities in the mid-term to effectively transmute the excess plutonium and utilise its energy content. In this application, the IMF must be compatible (non-soluble and unreactive) with the coolant: H_2O or D_2O. In practice, this condition limits the choice of IMF quite significantly. Schematic illustrations of IMF implementation into a light water reactor (PWR) and high temperature reactor (HTR) are given in Fig. 1.

The fuel in the rods/kernels themselves can be homogeneous, for instance as a solid solution of oxides of plutonium and other elements, e.g. zirconium, or a heterogeneous concept may be used to overcome the relatively low thermal conductivity of the ceramics employed. If the candidate is a homogeneous material, it may be a solid solution or an alloy. If the candidate materials are employed heterogeneously, they can either be cercer or cermet. Fig. 1 shows some design examples. The introduction of IMF rods into a UO_2 fuel assembly may be somewhat complex because of the large differences in the neutron spectra of the two cell types and their interaction with each other.

The core loading options (Fig. 1) chosen by the different groups who work on IMFs vary from one organisation to another [12–14]:

- Some groups have studied the behaviour of 100% IMF cores (homogeneous). Other concepts foresee the partial loading of IMF assemblies in UO_2 cores (heterogeneous), in an analogous way to present-day MOX fuel assemblies.
- The fuel assemblies themselves may be homogeneous, i.e. all fuel rods in a given assembly contain IMF, or heterogeneous with the IMF rods distributed among UO_2 rods.

The application of IMF in pressurised heavy water reactors (PHWR) has focused on studies of IMF full-cores for the purpose of Pu-annihilation (with Pu from either reprocessed PWR fuel, or ex-weapons Pu [15]).

4. Advantages of zirconia IMF

In order for inert matrix fuel fabrication to be economically attractive, it should make use of fabrication technology similar to today's standard fuel or MOX fabrication process. In reactor, the fraction of the core devoted to IMF is smaller than that required with MOX to balance the Pu production by the rest of the UO_2 loaded core. Consequently, for a similar fuel production cost, the lower IMF loading in core makes IMF economically competitive.

Pu disposition using IMF obviously addresses the ecological problem of radiotoxicity connected to the inventory of plutonium in the spent fuel. First, the reduction in actinide inventory helps to achieve this goal. Second, IMF can be specifically designed to provide long-term retention of actinides in spent fuel. For example, zirconia solid solution forms a single phase with actinide dioxides or rare earth sesquioxides, and spinel fixes fission products such as alkaline earth elements. Specific environmental studies are required for each IMF to understand the long-term behaviour of the spent IMF with regard to the conditions of a geological disposal. Such questions need to be addressed by specific investigation concerning the matrix solubility and corrosion rate in typical near field and far field environments and by suitable natural analogue studies.

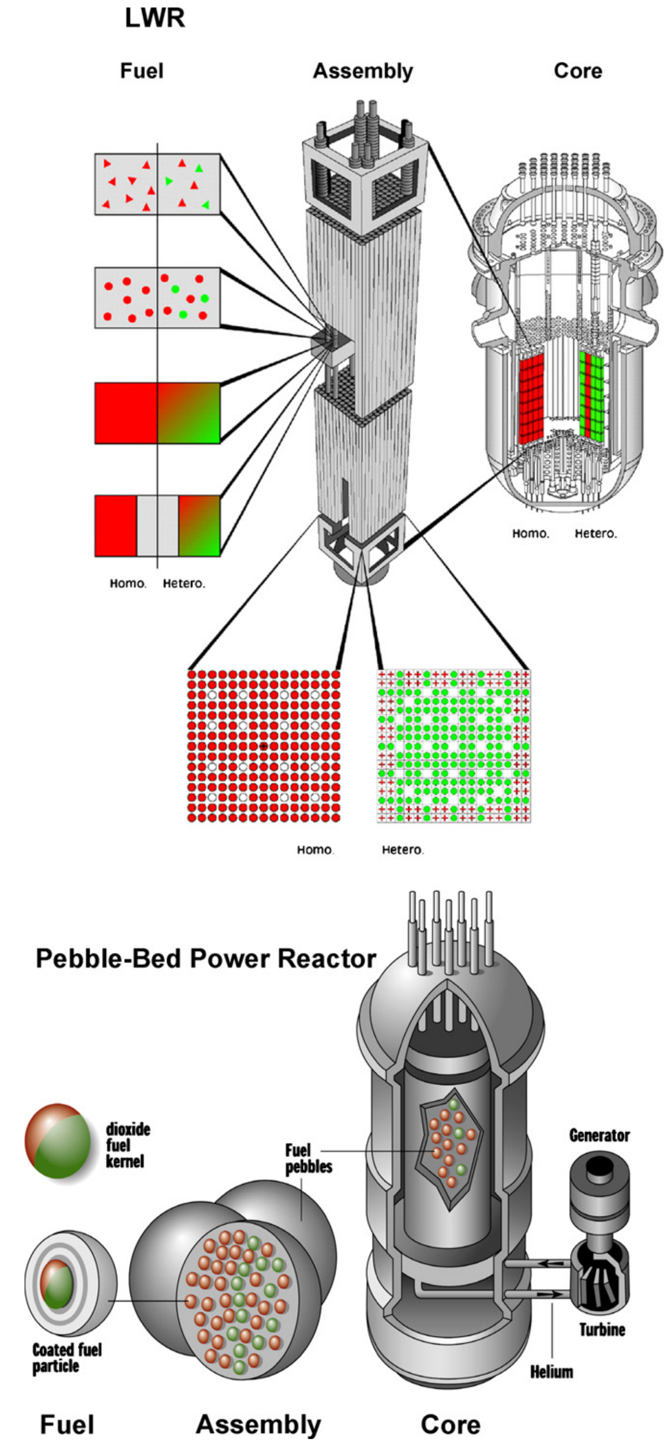

Fig. 1. The three levels for IMF utilisation in LWR and HTR considering homogeneous/heterogeneous systems at the pellet/kernel level, the assembly level and the core level, for details see [9]. Red IMF, green UO_x.

There are two safety aspects to be discussed: one concerns the utilisation of the IMF in reactor, and the other is the material handling itself. Particular work addressing the high burn-up

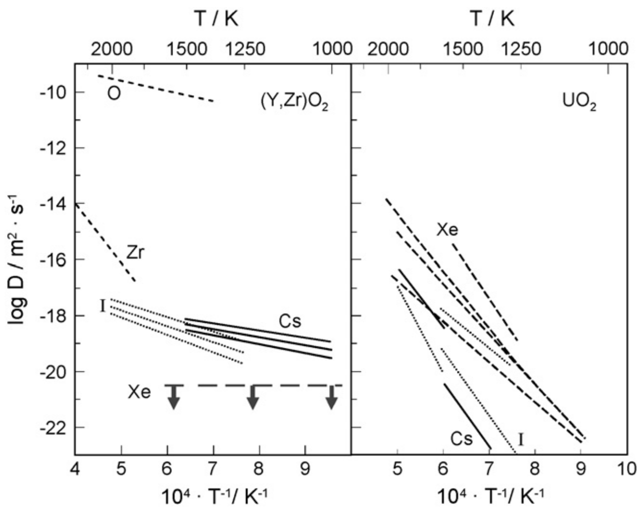

Fig. 2. Comparison of diffusion coefficient (*D*) for various elements (relevant fission products) with the intrinsic ion diffusions in zirconia. Note, for $(Y,Zr)O_2$, Cs and I follow Arrhenius law, the diffusion coefficient of Xe is below the detection limit. Data for UO_2 from: [21,22].

objectives and the core behaviour with respect to transient and accident conditions must also be noted to complete the IMF conceptual studies. The reactivity coefficients must be considered in the context of all applicable accident scenarios. However, the fuel by itself is the first retention barrier of fission products. Their diffusivity is compared Fig. 2 for Zr IMF and for UO_2 [21,22]. Clearly at the same temperature the retention in zirconia is better this is due to the smaller lattice parameter of zirconia (see Table 2) which compared to UO_2 allow less space for fission product bulk diffusion.

The IMF material handling prior and after irradiation must be safe compared to the classical UO_2 or MOX fuel case. As there do not appear to be any significant differences, this point applies more to the ecological impacts of spent fuel storage.

The non-proliferation aspects concern the safety through the IMF extension of the nuclear fuel cycle with regards to fissile material diversion. The characteristics that make the IMF an effective non-proliferation concept lie principally in its ability to utilise excess plutonium while producing no additional weapons usable material. IMF provides an efficient method for rapid destruction and degrading of the isotopic composition of weapons plutonium, which will minimise the likelihood of excess defence material returning to military application. It is likewise an efficient method to consume and reduce the quality with regard to isotopic composition of excess civil separated plutonium. IMF designed for a once-through cycle can also be made difficult to dissolve. Plutonium separation from fresh or spent fuel material is thereby made more difficult. Low leaching potential in aqueous solutions, brine, acidic, or basic heated or super-heated, molecular or ionic liquids such as molten salts is important in this regard.

The underlying incentives for all points developed above are common in that IMF could provide the option of balancing fissile material production and consumption in the current fuel cycle. This would negate the need and expense of long-term monitored storage of this material, contributing to the sustainability of the nuclear enterprise.

Table 2
Comparing lattice parameter of cubic stabilise zirconia inert matrix, with actinide dioxides (IMF 20–30 pm lower a values enhancing FP retention)

AnO_2 or ZrO_2-IMF	Lattice parameter (pm)	References
UO_2	547.0	[16]
PuO_2	539.5	[17]
AmO_2	538.8	[18]
ZrO_2	512.0	[19]
$(Er,Y)_yPu,Zr_{1-y}O_{2-y/2}$	517.9–520.8	[20]

5. Current programs

Current IMF programs focus on specific work in the areas of fabrication, characterisation, irradiation with accelerators or in research reactors, and development of models for predicting behaviour in commercial reactors. IMF fabrication with optimised densities is carried out prior to characterisation. The fabrication of ceramic IMF powder can be performed following either a wet or a dry preparation route. The wet route starts with nitrate solutions of all components and coprecipitates the oxi-hydroxides from concentrated or highly concentrated solutions. The latter is required in case of microsphere production by gelation. The products are dried and calcined. Hydrated salts thermolysis has also been occasionally applied. The dry route involves mixing and milling of powders. Milling is performed in batch in a discontinuous way using ball milling or by a continuous process utilising attrition milling. Pelletising is carried out prior to sintering at a given temperature and for a given time. For cermet fuels, hot extrusion is also occasionally used.

Characterisation of the pellet or of the material is carried out at both macroscopic and microscopic levels. The "geometrical" density is first measured and the porosity is deduced from the theoretical density, which itself may be derived from X-ray diffraction analysis. At the microscopic level, optical microscopy and scanning electron microscopy are used to study pore or grain structure. The IMF characterisation is completed by irradiation studies using research institute facilities such as accelerators and research reactors. Accelerators have been used to study microstructural changes during irradiation. Stabilised zirconia was found to be very resistant to irradiation, e.g. [23]. To complete the basic knowledge gained using accelerator irradiation, in-pile tests irradiation are done in research reactors. The first series of IMF irradiations in Japanese, French, Dutch, Canadian, American and Russian reactors have already been performed and irradiations are also planned in the OECD Halden reactor and in the framework of joint programs in the high flux reactor at Petten. Emphasis is given to energy production and transport in the fuel (temperature measurements in the pin), the mechanical behaviour and the fission product release. Irradiations are also planned in the ATR reactor at the Idaho National Laboratory, USA.

Neutronic modellings studies at the fuel, assembly and core levels have been carried out for conceptual studies to ensure the feasibility of IMF application in commercial reactors. The

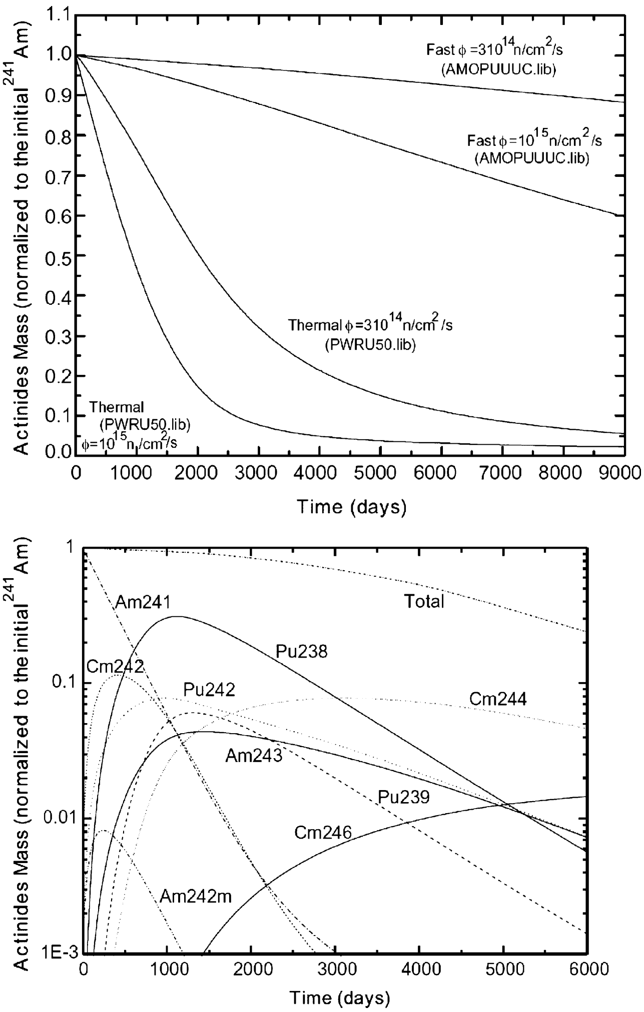

Fig. 3. (a) Total actinide mass as function of time for different neutron spectra and fluxes and (b) isotopic evolution in a thermal spectrum with a constant neutron flux of 3×10^{14} cm^{-2} s^{-1}. Actinide mass normalised to the initial isotope mass.

components: (i) fissile (weapon or civilian plutonium from UOX or MOX reprocessing), (ii) fertile in certain cases, (iii) burnable poison and (iv) inert matrix components of the IMF are selected and their respective concentrations are optimised according to the neutronic characteristics. Plutonium consumption is optimised in IMF because of the absence of ^{238}U. The MA (e.g. ^{241}Am) consumption is also significant in thermal flux as calculated in detail for thermal and fast neutron flux (Fig. 3).

Waste management issues are relevant from a sustainability point of view. The limited evidence from the literature suggests that baddeleyite may be very resistant to the effects of alpha-decay damage from ^{232}Th and ^{238}U atoms incorporated in dilute solid solution, e.g. [24]. This evidence is consistent with previous ion irradiation studies and is also supported to a certain extent by the case study presented above. However, the TEM results described in the case study document the potential modification of the microstructure of baddeleyite under intense alpha-particle irradiation by an adjacent actinide-rich phase. In this example, the bombardment by alpha-particles apparently

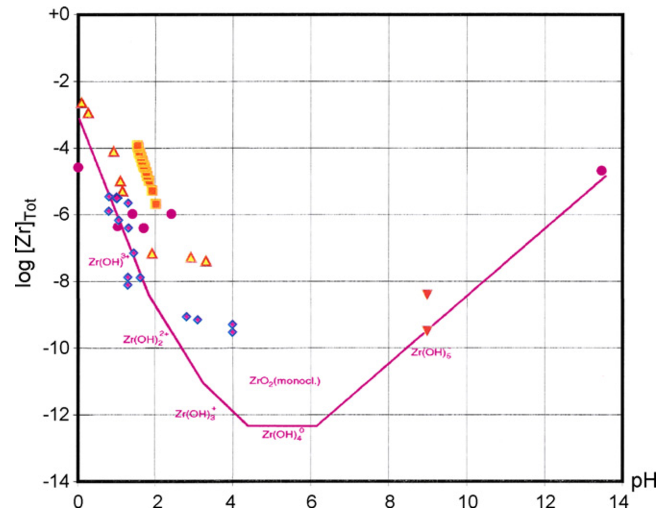

Fig. 4. Solubility of monoclinic zirconia and oxy-hydroxide precipitate as a function of pH. Note the Zr solubility ranges from 10^{-9} to 10^{-8} M. Data from: (■) Kovalenko and Bagdasarov [29]; (●) Adair et al. [30]; (▼) Pouchon et al. [31]; (▲) Ekberg et al. [32]; (◆) Michel [28].

caused the development of a domain structure and fine scale modulations, but otherwise did not significantly degrade the crystalline structure of the material.

For this strategy, low solubility of the inert matrix is required for geological disposal. As spent fuels these IMFs must be excellent materials from the solubility point of view. This parameter was studied in detail for a range of solutions corresponding to groundwater under near field conditions. Under these conditions the zirconia IMF solubility is 10^6 times smaller than glass [25–28]. Even in carbonate solution the zirconia solubility is very low (see Fig. 2) which makes the zirconia material very attractive for deep geological disposal (Fig. 4).

Internationalisation of the IMF programs has induced collaborative work among several organisations. Work has been

Table 3
Organisations involved in the efforts of the inert matrix fuel initiative

Country	Organisation
Canada	Atomic Energy of Canada Limited
France	Commissariat de l'Energie Atomique, Saclay, Cadarache and Grenoble; University of Paris
Italy	Politecnico di Milano; ENEA, Bologna; University of Trento
Japan	Japanese Atomic Energy Research Institute, Japan Nuclear Cycle Development Institute; University of Tohoku; University of Kyushu
Korea	Korean Atomic Energy Research Institute
The Netherlands	Nuclear Energy Centre, Petten; University of Delft
Russia	IPPE, Obninsk; A. Bochvar Institute, Moscow
Switzerland	Paul Scherrer Institute; Ecole Polytechnique Fédérale de Lausanne; University of Geneva
USA	University of Ann Arbor; Massachusetts Institute of Technology, Idaho National Laboratory; Argonne National Laboratory; Los Alamos National Laboratory; Oak Ridge National Laboratory
Multilateral CEC	European Joint Research Centre, Karlsruhe
Multilateral NEA	OECD Halden project
Multilateral UN	IAEA, Vienna

carried out or is currently in progress in various organisations around the world (see Table 3).

6. Conclusions

The 441 reactors around the world produce about 100 tonnes of plutonium and about 10 tonnes of minor actinide annually, in spent fuel, some of which is separated through reprocessing. While the recycling of plutonium as MOX fuel derives additional energy from this resource, it does little to address the issue of growing plutonium inventories. If a societal objective is to reduce the amount of plutonium, then IMF provides an attractive option for plutonium destruction. More generally, the utilisation of plutonium in IMF provides flexibility in balancing the quantity of plutonium production and consumption by enabling either the net burning of plutonium or a balanced production and consumption. This approach is viable in existing cores that already utilise MOX fuel. Due to the added dimensions in managing the fuel cycle that IMF allows, it can play an important role in the future of nuclear energy.

Another important application of IMF is the destruction of minor actinides, with or without plutonium. IMF can also be used both to manage to address the long-term radiotoxicity of the spent fuel by minor actinide destruction. IMF s are also being considered for Gen IV reactors, because of their advanced performance, economics, safety features, sustainability, and application to waste minimisation in a closed fuel cycle.

Several promising candidate materials have been identified for both fast and thermal reactors: ZrO_2 solid solutions or composite with MgO, SiC or Zr alloys; some of these fuel candidates have undergone test irradiations and PIE. Modelling calculations of IMF fuel performance and safety analysis as well as tests have progressed. Fabrication methods have also been developed or adapted from existing technologies. System studies have identified strategies for both implementation of IMF fuel in existing reactors in the shorter term, as well as in new reactors in the longer term.

The work to date has established the feasibility of these IMF materials, and core loadings and reactor strategies for utilizing these fuels. Further development is required before commercial deployment of IMF, which will require additional resources. Additional in-pile irradiations are required, both for normal operating and accident conditions. Further safety analysis and safety testing is required. Some development is needed in the area of analysis tools and fuel performance codes. Irradiations in commercial reactors should be undertaken in a staged approach as soon as possible, beginning with segments in pins, full pins, then finally, lead test assemblies.

The desired objective would be to use IMF to produce energy in reactors, opting for an economical and ecological solution. The Swiss IMF results are reported in the proceedings of the IMF workshops published in several issues of J. Nucl. Mater. (1999, 2003, 2006) [7,9,10] and Prog. Nucl. Energy (2001) [8].

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