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Section 1. Introduction

Concepts for an inert matrix fuel, an overview

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1. Introduction

During the last five years, several national and multinational research organisations have been devoting R&D work to the transmutation of plutonium and minor actinides in thermal reactors and advanced systems by applying an Inert Matrix Fuel (IMF) concept [1–7]. This situation is a consequence of the aim of eliminating the current excesses of plutonium and other transuranium elements. These efforts are being made because of the energetic value of plutonium, proliferation risks and also because they contribute to the largest radiotoxicities of conventional spent fuels after some 100 yr [8].

Current IMF projects initially dealt with plutonium excesses from civil nuclear power production. However, after the end of the 'Cold War', excesses of plutonium from military sources had also to be considered. Today, discontinuation of the weapon production programs and increasing production of reprocessed plutonium from the electronuclear programs will yield surplus in the stocks of the order of 200 tons weapon grade plutonium and 1000 tons civilian plutonium, respectively at the end of this century [9,10].

As indicated, the global problem of plutonium excess is associated with potential hazards related to proliferation and environmental safety. The management of plutonium inventories in an effective manner has suffered a major setback due to the postponement and/or abandonment of fast breeder reactor programmes. In effect, the former goal to produce more plutonium had to change to a stock reduction strategy. However, energy production remains the most desirable disposition and to solve the problem of plutonium surplus in the short and medium term it is suggested to burn, as quickly and completely as possible, excess plutonium in existing Light and Heavy Water Reactors (L&HWRs) [9]. Since today's practice of mixed oxide (MOX) fueling in LWR (with up to 40% core loading) does not allow a rapid reduction of plutonium stock-piles [11], the replacement of uranium dioxide by an inert matrix has been recommended.

Historically, early in the sixties, IMF tests were performed searching for a matrix to hold plutonium for energy production. Irradiations of ZrO_2–UO_2 e.g. [12–14] were successfully performed in various reactors followed by the first tests with ZrO_2–PuO_2 at Hanford [15,16]. These irradiations were based on important early studies reported in the fifties [17,18].

Clearly, the current rebirth of such programmes worldwide has taken place in a new context. A synthesis of philosophy and specific recent scientific results are presented and discussed in this special issue. This represents a scientific summary of the recent activities of the organizations directly or indirectly involved in the 'Initiative for Inert Matrix Fuel'. The published contributions in this special issue represent an overview of studies that are today integrated into national or international projects initiated since the first Inert Matrix Workshops held at the Paul Scherrer Institute [19,20] and elsewhere [21] since September 1995, as well as from other specific actions.

The reported R&D work includes studies about specific properties and conditions that the inert matrix fuel must fulfil. These properties are investigated systematically according to the methodology depicted in Fig. 1. The basic strategy is to apply a once-through IMF irradiation prior to geological disposal [3]. This may, however, be extended by a specific reprocessing.

2. Fuel material selection and testing

The first requirement in the material selection is guided by the neutronic properties. The elements and/or isotopes of the inert matrix are selected according to their transparency for neutrons, thus corresponding to the original definition of the term Inert Matrix. This is

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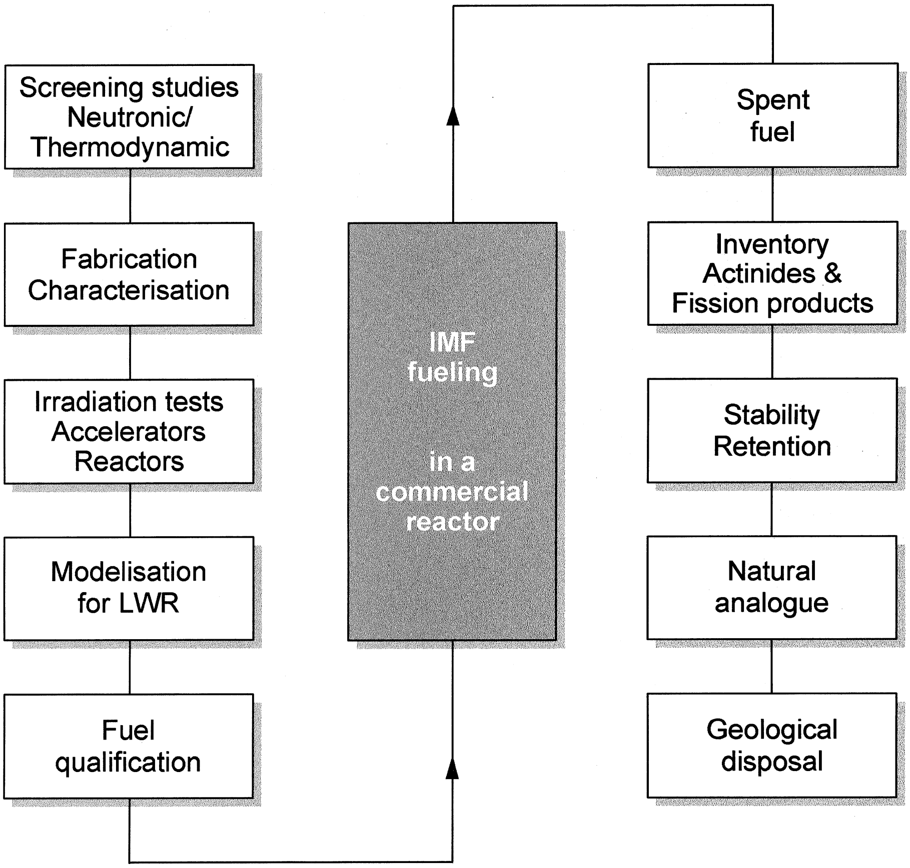

Fig. 1. Methodology used to progress towards IMF qualification for a once-through irradiation in LWR prior to geological disposal.

followed by simple neutronics cell calculations to characterise reactivity effects of:

- the inert matrix components
- the fissile vector (first generation, second generation or weapon grade Pu) and
- in some concepts, a suitable burnable poison and/or a fertile additive introduced in order to improve the neutronic characteristics of the fuel.

The evaluation was carried out for a range of elements completing the initial selection process [3,22].

In the preliminary neutronic studies, the physicochemical properties of the material are not considered. These are, however, of prime importance for understanding the fuel behaviour in reactor. The desired thermodynamic properties of the material(s) are: high melting point (e.g. ~3000 K), good thermal conductivity, good compatibility with the cladding, low solubility in hot water and high density (e.g. >95% relative density). In all cases, the IMF candidate is compared to UO_2 as the reference. Material screening studies were first carried out on the basis of literature reviews and knowledge gained in waste management studies but new experimental work was occasionally required. IMF samples have consequently been produced for the later studies. As a result of these considerations, emphasis has been given to various oxides but in some cases to metals, and even to carbides or nitrides which have been found to be acceptable in specific cases.

At the level of the fuel pellet, homogeneous or heterogeneous dispersion of the fissile isotopes in material is considered and guides the selection of the matrix candidates. The IMF may be a single phase or multi-phase material. The candidate materials are, for example: stabilised ceramics such as $Zr_{1-x}Ca_xO_{2-x}$, $Zr_{1-x}Y_xO_{2-x/2}$, CeO_2, or other ceramics such as $ZrSiO_4$, $Al_5Y_3O_{12}$ (YAG), MgO or $MgAl_2O_4$ (spinel). If the candidate materials are mixed phases, these can either be cercer (ceramics embedded in another ceramics) such as Al_2O_3–$Zr_{1-x}Y_xO_{2-x/2}$–MgO, or, cermet (ceramics embedded in metal) e.g. M–$Zr_{1-x}Y_xO_{2-x/2}$ in which case the metal M may be Mo, Cr, Zr, Al(Si). Nitrides, carbides or even carbon [23] are also investigated. The addition of burnable poisons is dictated by the neutronic requirements; they can be Er, Gd, Ho or B. Several additives are also occasionally required to stabilise the inert matrix material in presence of the plutonium compound. It must also be noted that CeO_2 is currently used as a PuO_2 surrogate for material or fabrication tests.

IMF pellet fabrication with optimised densities is carried out prior to characterisation. The fabrication of the IMF powder can be performed following either a wet or a dry preparation route [24]. The wet route starts with nitrate solutions of all components and coprecipitates the oxy-hydroxides from concentrated or highly concentrated solutions. The latter is required in case of microsphere production by gelation. The products are dried and calcinated. Hydrated salts thermolysis has also been occasionally applied [25]. 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, hot intrusion 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 and scanning electron microscopies are used to study pore or grain structure.

The IMF characterisation is completed by irradiation studies using research institute facilities such as accelerators or research reactors. Accelerators are used to study microstructural changes during irradiation [24]. Satisfactory behaviour under ion irradiation i.e. no swelling and no amorphisation, is essential for the material selected. Implantation is also performed to study specific thermodynamic behaviour (e.g during annealing or in contact with water). To complete the basic knowledge gained using accelerator irradiation, in-pile tests irradiation are done in research reactors. First series of 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 the energy production and transport (temperature measurements in the pin), the mechanical behaviour and the fission product release.

Neutronic modelling at the pellets, pin and assembly levels is carried out prior to conceptual studies for commercial reactors. The components: fissile (weapon or civilian plutonium from UOX or MOX reprocessing)/fertile (^{238}U and/or ^{232}Th) in certain cases [25]/burnable poison and inert matrix components of the IMF are selected and their respective concentrations are optimised according to the neutronic characteristics.

3. Reactor studies

Commercial L&HWRs are the only worldwide available candidate facilities in the mid-term to effectively transmute the excess plutonium and realise its energy content. The objective of the studies presented in this special issue is a reduction of the current plutonium stock-piles. As a consequence, all concepts developed in this direction are based on the following precept:

> “The new fuel must be conceived such as to be suitable for loading into present-day power reactors, without any geometrical modifications of the core”.

In practice, this condition limits the choice of IMF quite significantly. A schematic illustration of IMF implementation into, for example, a Pressurized Water Reactor (PWR), is given in Fig. 2. The new Inert Matrix Fuel must preserve the characteristics of the reactor in terms of:

- design
- total power
- length of the cycles
- safety of the plant

The loading options (Fig. 2) chosen by the different groups who work on IMFs vary from one organisation to another [22,26–28]:

- 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 [27], or heterogeneous with the IMF rods distributed among UO_2 rods [28].
- Finally, the fuel in the rods 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. In the later case, the grains of the solid solution may be dispersed into a metallic matrix (cermet) or into other ceramics with a higher thermal conductivity (cercer). Another approach consists in producing annular pellets, which may be clad also on the inside to allow cooling from both inner and outer surfaces. Such a concept must, however, be carefully optimised to avoid modifications affecting the thermalhydraulic behaviour of the IMF assemblies.

The heterogeneity of IMF at the rod level normally has little influence on its neutronic behaviour: the grains of the phases containing the fuel are sufficiently small to be considered as homogeneous from the viewpoint of the neutronic modelling.

The introduction of IMF rods into a UO_2 fuel assembly is, on the contrary, more complex because of the large differences in the neutron spectra of the two cell

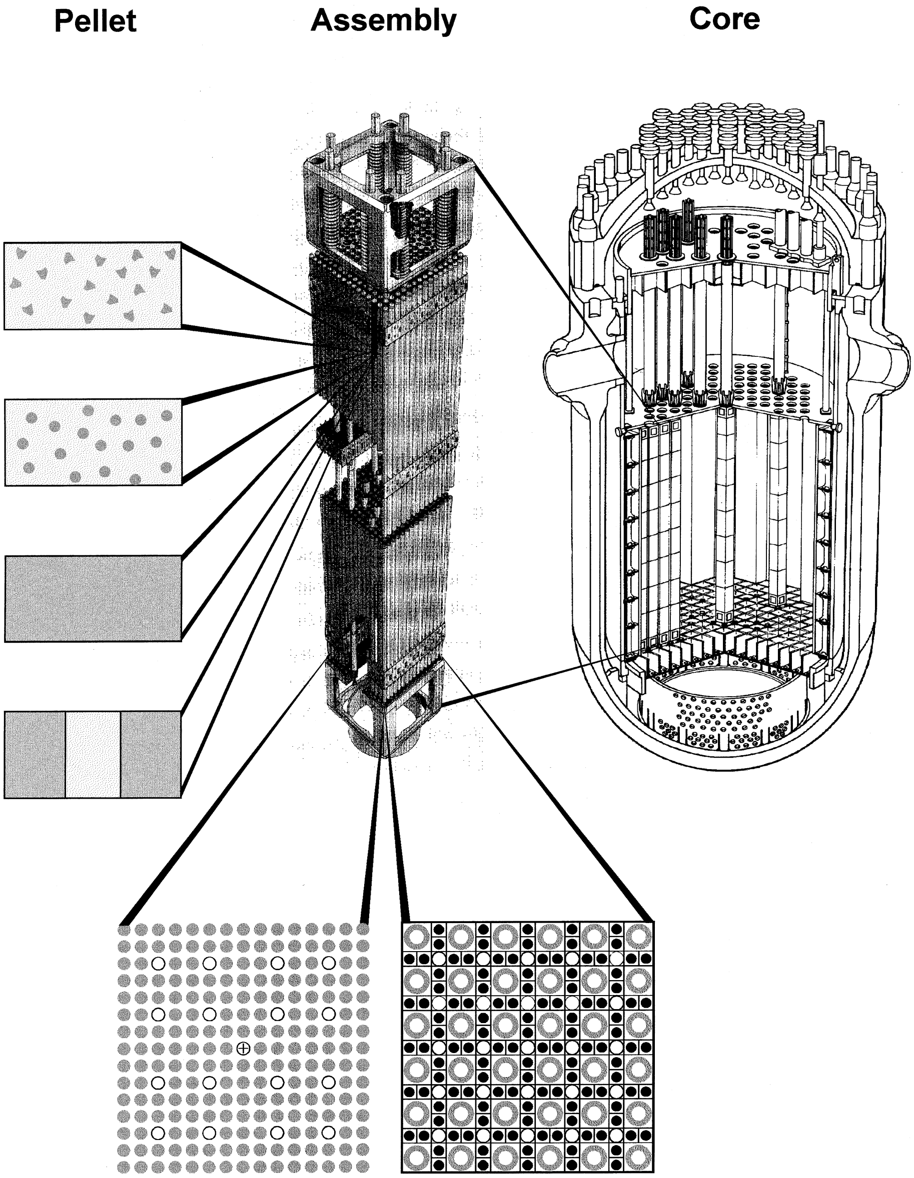

Fig. 2. The three main levels considered in the IMF studies. Pellet (annular, full) and material design (composite, with microspheres), pin and assembly (IMF, APA [28]), core IMF loading. Note: the concept of homogeneity/heterogeneity is addressed at each level; Pu phases in gray, U phases in black.

types and their interaction with each other. On this level, the calculation methods need more validation. It is for this reason that an international modeling benchmark exercise has been under way in order to compare the results of different calculation schemes.

In the case of a heterogeneous core, the homogeneous IMF assemblies are distributed within an environment of UO_2 fuel assemblies. Here also, strong variations in the neutron spectrum occur at the interfaces between fuel assemblies of different types. Large power ‘peakings’ take place in the IMF rods at beginning of life (BOL), which makes it necessary to reduce the Pu density in the IMF rods on the outer rows of the fuel assembly, especially on the corners, or to add some elements which act as additional burnable poisons, e.g. erbium or gadolinium [27].

A homogeneous core, i.e. one which is loaded to 100% with IMF assemblies, is easier to conceive and to calculate. In spite of that, three significant problems are to be solved.

The reactivity curve of the fuel as a function of the burnup must be adjusted. IMF cells show a much steeper reactivity variation than UO_2 cells, because, contrary to the case where the reduction of ^{235}U is partially compensated by the production of ^{239}Pu, no new fissile nuclides are created during burnup when fertile material is absent. The excess reactivity to be invested at BOL is therefore very large and it cannot be any longer compensated by the boron dissolved in the moderator. Some burnable poisons must be added to the fuel. The choice of the poison and of its concentration is difficult: taking into account the limits imposed by the chemistry/metallurgy, an element must be found whose neutron absorption correctly decreases during burnup and for which the residual absorption at end of life is as small as possible, the condition for the maximum reduction of the Pu. Erbium seems to impose itself as the best compromise, in spite of a fairly large residual absorption.

The fuel temperature coefficient must be sufficiently negative. In the UO_2 cells, the fuel temperature coefficient is mostly driven by ^{238}U, which is absent in the IMF cells. The presence of a burnable poison with a large absorption cross section in the thermal energy range e.g. gadolinium, or boron, reduces the coefficient even further. The problem is more severe in the case of weapons-grade Pu than for reactor-grade Pu, where the large concentration of ^{240}Pu ameliorates the situation. The fuel temperature coefficient for IMF requires particular attention as the fraction of delayed neutrons (β_{eff}) is about three times smaller than in the UO_2 case. Thanks to its resonances, Er (if present) contributes significantly to improvement of the coefficient.

The moderator void coefficient must be negative in all cases. In the calculation of infinite arrays of certain types of IMF cells, the moderator void coefficient for large voidages can be found to be positive at the beginning of irradiation. After a few weeks at full power, even such coefficients become strongly negative. There is thus no practical problem in a reactor core where fuel assemblies with a range of different burnups are present at a given time.

Specific work addressing the high burnup objectives and the core behaviour with respect to transient and accident conditions must also be noted to complete the IMF conceptual studies.

4. Spent fuel disposition

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. Two schools of thought can be found here.

- The used IMF is reprocessed for a later irradiation. Here the material must be leachable, if possible in the same reprocessing conditions as for UO_2 fuel. On the other hand, a maximal combustion is not a strong criterion, since the residues will serve as fuel in another installation [29].
- The used IMF is directly 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 material which forms the IMF must be chemically very stable, such as to satisfy the criteria for long term storage (adequate immobilization of radioactivity in geological disposal).
- The combustion of Pu and of the minor actinides must be as high as possible, in order to reduce the toxicity and the heat source in the storage. This can have some consequences for the choice of IMF loading strategies to be implemented.

Specific environmental studies are required to understand the long term behaviour of the spent fuel or the inert matrix e.g. metal for cermet (for Zr see [30]) or other ceramics [31] with regard to the conditions of a geological disposal. For example zirconia solid solution forms a single phase with actinide dioxides or rare earth sesquioxides, and spinel fixes fission products such as earth alkaline elements. 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.

5. IMF R&D programs

Internationalisation of the IMF programs has induced collaborative work among several organisations. Currently, work is carried out at the Paul Scherrer Institute, at the Ecole Polytechnique Fédérale, Lausanne and at the University of Geneva, Switzerland; at the Japanese Atomic Energy Research Institute and at the University Kyushu, Japan; at the Korean Atomic Energy Research Institute, Korea; at the Politechnico di Milano, the ENEA and the University of Trento, Italy; at the Nuclear Energy Center of Petten, the Netherlands; at the University of Ann Arbor, the Massachusetts Institute of Technology and the Los Alamos National Laboratory, USA; at the Commissariat de l'Energie Atomique, Saclay, Cadarache and Grenoble, France and inter alia at IPPE and VNIINM, Russia. At the multinational level, the OECD Halden project, Norway and the European Research Center at Karlsruhe, Germany devote substantial activities for the IMF projects.

This brief presentation has meant to provide an overview of the scope of current programmes on IMF. The articles which follow develop more specific aspects of this vast investigation domain concerning an important fuel cycle option which aims at greater economy and ecological acceptability.

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