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FOREWORD

Reactors around the world have produced more than 2000 tonnes of plutonium, contained in spent fuel, as separated forms through reprocessing, or as weapons-grade material. The recycling of plutonium as uranium–plutonium mixed oxide fuel derives additional energy from this resource; however, it does not speedily reduce growing plutonium inventories. The use of inert matrix fuel (IMF) in the current generation of reactors would provide a means of reducing plutonium inventories. The reduction of the accumulated plutonium by the use of IMF is a subject of great interest in several Member States. Work on IMF to date has been investigating the feasibility and reactor strategies for utilizing these fuels.

Another important application of IMF is the reduction of minor actinide content, with or without plutonium. IMF can be used both to manage plutonium inventories and to address the long term radiotoxicity of spent fuel by minor actinide reduction in today's reactors. IMF materials are also being considered for generation-IV reactors. Further development is needed before commercial utilization of IMF.

This report summarizes ongoing IMF work and strategies with emphasis on their advanced performance, and application to waste minimization in a closed fuel cycle. The status of the potential IMF candidates is reviewed. This publication encompasses several promising candidate IMF materials for both fast and thermal reactors, fabrication methods, status of irradiation tests, results of post-irradiation examination, modelling of IMF fuel performance, and system studies for identification of strategies for implementation of IMF.

This report has been prepared based on two consultants meetings (held 13–16 May 2002 and 26–28 May 2003 in Vienna with the participation of experts from Canada, France, Japan, the Russian Federation, Switzerland and the USA) and additional input from other specialists. The contributions of all who gave valuable help in drafting the report (listed at the end of this publication) are greatly appreciated. The IAEA wishes to express its gratitude to C. Degueldre (Switzerland) for chairing the group of consultants. The review of the report by H.J. Matzke (who retired recently from the Institute for Transuranium Elements, European Commission) is appreciated.

The IAEA officers responsible for this publication were F. Sokolov and H.P. Nawada of the Division of Nuclear Fuel Cycle and Waste Technology.

EDITORIAL NOTE

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1. OVERVIEW

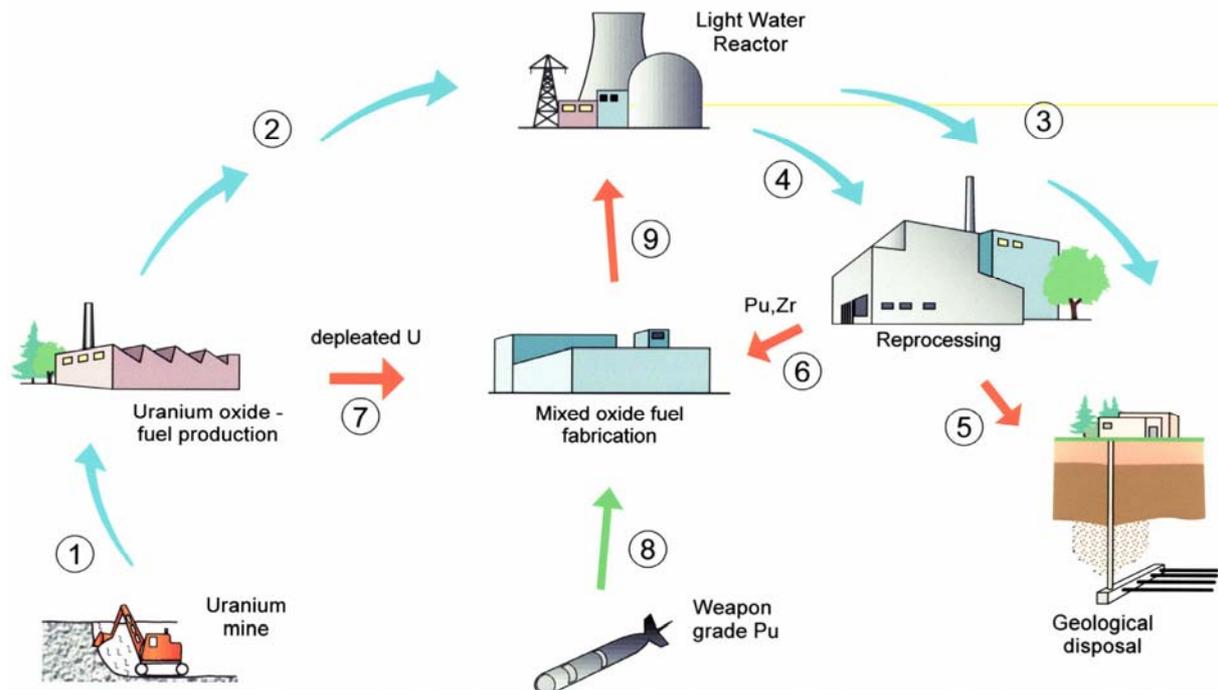
1.1. Background

During the last ten years of the former century, several national and multinational research organisations have been devoting R&D work to the incineration of plutonium and minor actinides in thermal reactors, fast reactors, and advanced systems by utilizing an Inert Matrix Fuel (IMF) concept e.g. [1–7]. This stems from the interest in the aim of reducing the current excess stockpiles of plutonium and other transuranium elements. These efforts are being made because of the energetic value of plutonium, reduction of proliferation risk consideration, and also because the transuranic elements contribute to the considerable radiotoxicities of conventional spent fuels after some few 100 years [8].

IMF is a type of nuclear reactor fuel that consists of a neutron-transparent matrix and a fissile phase that is either dissolved in the matrix or incorporated as macroscopic inclusions. The matrix plays a crucial role of diluting the fissile phase to the volumetric concentrations required by reactor control considerations, the same role ^{238}U played in conventional low enriched uranium (LEU) or MOX fuel. The key difference is that replacing fertile ^{238}U with a neutron-transparent matrix eliminates plutonium breeding as a result of neutron capture.

Current IMF projects initially dealt with excess plutonium from civil nuclear power production, however, after the end of the ‘Cold War’, excess plutonium from military sources also had to be considered. Today, reduction of weapon-materials inventories and increasing production of reprocessed plutonium from the electronuclear programmes have yielded Pu stocks on the order of 200 tons of weapons grade plutonium and 1000 of tons civilian plutonium at the end of the last century [9, 10].

As indicated, the major concern of separated plutonium 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 delays in commercial deployment of fast reactor programmes. In some Member States, the former goal to produce more plutonium has changed to a plutonium reduction strategy. However, energy production remains the most desirable option for disposition and to solve the problem of plutonium surplus in the short and medium term it is suggested by the US National Academy of sciences to burn, as quickly and completely as possible, excess plutonium in existing Water Reactors [9]. Fast neutron reactors (FRs) may also be considered for the effective incineration of minor actinides. Since today’s practice of mixed oxide (MOX) fuelling in LWRs (with up to 40% core loading) does not allow a rapid reduction of plutonium stockpiles [11], the replacement of uranium dioxide by an inert matrix and an extension of the nuclear fuel cycle (see Fig. 1) have been examined. Recently IAEA has prepared a document [12] on advanced fuels, which included also fuels to burn excessive ex-weapons plutonium in thermal power reactors. In that document more emphasis has been put on the basic properties, fabrication and irradiation behaviour of these fuels. Review of the status of development MOX fuel technology in the areas of design, fabrication, performance, in-core fuel management, transportation, spent MOX fuel management, decommissioning, waste treatment, safeguards and alternative approaches for plutonium recycling could be found in another recent IAEA publication [13]. Reactor grade MOX fuel compositions with or without additions of Am and Np could possibly be used as the primary heavy metal energy production composition in the core.



- Nuclear Fuel Cycle extension for once-through plutonium burning
- The arrows (1, 2, 3) denote the classical step of the basic nuclear fuel cycle from the uranium mine to the disposal of the non-reprocessed fuel element (e.g., Once-through Fuel Cycle).
- The arrows (4, 5) introduce the reprocessing step, Pu is produced and stocked.
- The arrows (6, 7, 9, 4) present potential extension for the MOX option
- The arrows (6, 8, 9, 3) depict the suggested fuel cycle extension. It includes spent U-fuel reprocessing plutonium fuel fabrication (U-free), Pu burning in the reactor, intermediate storage and direct geological disposal of the rock-like spent fuel.

Fig. 1. Nuclear fuel cycle extensions showing both IMF multi-recycling or “once through then out” (OTTO) options.

Clearly, the current rebirth of IMF programmes worldwide has taken place in a new context. A synthesis of philosophy and specific recent scientific programmes are presented and discussed in this TECDOC. This represents a comprehensive summary of the recent activities of the organizations 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.

IMF activities were reactivated with the first Inert Matrix Fuel Workshops held at the Paul Scherrer Institute [14, 15] and elsewhere [16–20] beginning in September of 1995, as well as from other specific actions e.g. from the OECD such as the Advanced Reactors With Innovative Fuels (ARWIF) workshops [21, 22] the Partitioning and Transmutation (P&T) workshop, held at Jeju Island [23], and European Research on Materials for Transmutation ERMT meetings [24].

The present report summarises R&D work on inert matrix fuel for plutonium and (to a lesser extent) minor actinide stock-pile reduction, and discusses the possible strategies to include inert matrix fuel approaches to the nuclear fuel cycle.

1.1.1. Definitions

The first requirement in material selection is guided by the neutronic properties. Since the aim is to burn plutonium (or minor actinides) then consequently one has to eliminate the source of production of these nuclides in the fuel. This corresponds to the original concept of uranium-free fuel.

The inert matrix fuel concept needs to go one step further; the elements and/or isotopes of the inert matrix are selected according to their transparency for neutrons. The basic strategy is to apply a once-through IMF irradiation prior to geological disposal. The fuel cycle, however, may be extended by multi-recycling. Multi-recycling could also be compatible with IMF, in particular when the inert matrix can be partitioned from actinides.

Inert Matrix properties are investigated systematically according to the methodology depicted in Fig. 2. Simple studies on neutronics properties (to characterize reactivity effects¹) are complemented by other studies on:

- compatibility of the inert matrix components with coolant and structural materials;
- the fissile vector (first generation, second generation or weapon grade Pu, or Am); 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 evaluation of Pu-incineration has been carried out for a range of elements completing the initial selection process [2].

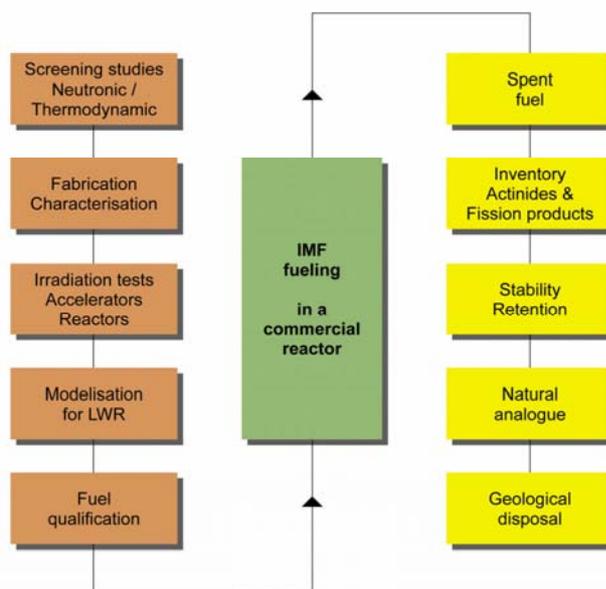


Fig. 2. Methodology used to progress towards IMF qualification for a once-through irradiation in LWR prior to geological disposal.

¹ Reactivity is the term expressing the departure of a reactor system from criticality. A reactivity addition indicates a move toward power increase (super criticality). A reactivity subtraction indicates a move toward power decrease (sub criticality).

Inert Matrix candidates

In the preliminary neutronic studies, the physico-chemical properties of the material are not considered. These are, however, of prime importance for evaluating the reactor fuel behaviour. The desired properties of the material(s) are a 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 standard UO₂ fuel as the reference [2, 25]. A critical assessment of selection of materials as diluents for burning of plutonium fuels in nuclear reactors based on their physical and chemical properties are discussed in a recent review [25]. This review appraises the out-of-pile properties of Al₂O₃, MgO, MgAl₂O₄, CeO₂, Ce₂O₃, Y₂O₃, ZrO₂, CePO₄, ZrSiO₄, B₄C, SiC, AlN, Mg₃N₂, Si₃N₄, CeN, YN and ZrN include, e.g. melting point, vapour pressure and thermal conductivity; the chemical behaviour towards steel, liquid sodium, water, air and nitric acid in the context of suitability of diluents. Material screening studies should be first carried out on the basis of literature reviews and knowledge gained in spent fuel and waste management studies. Most often new experimental work is required to supplement these databases. Candidate materials are listed in Table I. These are elements (metals, carbon [26]), oxides, nitrides, and carbides [27].

TABLE I. EXAMPLES OF INERT MATRIX CANDIDATES AND THEIR CHEMICAL FORMULAE

Inert Matrix components	Inert Matrix formula
<i>Elements</i>	C, Mg, Al, Si, Cr, V, Zr, Nb, Mo, W
<i>Inter-metallics</i>	AlSi, AlZr, ZrSi, ...
<i>Alloys</i>	Stainless steel, zirconium alloys
<i>Carbides</i>	¹¹ B ₄ C, SiC, TiC, ZrC
<i>Nitrides</i>	AlN, TiN, ZrN, CeN
<i>Binary oxides</i>	MgO, CaO, Y ₂ O ₃ , ZrO ₂ , CeO ₂
<i>Ternary oxides</i>	Mg _(1-x) Al _(2+x) O _(4-x) , Y ₃ Al ₅ O ₁₂ (YAG), ZrSiO ₄
<i>Oxide solid solutions</i>	Ca _x Zr _{1-x} O _{2-x} , Y _y Zr _{1-y} O _{2-y/2}

IMF specimens have consequently been produced on the basis of the inert matrix selection. 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.

Additives

The addition of burnable poisons is dictated by core neutronic requirements; they can be used to make the core reactivity as a constant as possible as a function of burnup. In addition, for safety reasons, small amounts of resonant additive may be used.

Finally, a stabilizer may be used to fully stabilize the solid solution in an adequate thermodynamic phase. Table II shows some IMF additive types and formula examples.

TABLE II. EXAMPLE OF ADDITIVES CONSIDERED FOR THE INERT MATRIX FUEL

Additive type	Additive formula
<i>Burnable poison</i>	B, Gd, Dy, Ho, Er, Eu
<i>Resonance additive</i>	Fe, W, Th, U
<i>Stabiliser</i>	Y ₂ O ₃ , CaO in ZrO ₂ Al ₂ O ₃ in SiC

Configurations

At the level of the fuel phase, homogenous or heterogeneous dispersions of the fissile isotopes in the matrix are considered and guide the selection of the matrix candidates. The IMF may be a single phase or multi-phase material. If the candidate is a homogeneous material, it may be a solid solution or an alloy. If the candidate materials are mixed phases, these can either be ceramics embedded in another ceramic (cercer), ceramics embedded in a metal phase (cermet), metal phase in a metal matrix (metmet). Table III gives some design examples.

TABALE III. EXAMPLE OF HETEROGENEOUS MATERIALS AS IMF's

Dispersion type	Example
cercer	MgAl ₂ O ₄ - Y _y Pu _x Zr _{1-y} O _{2-y/2} *
cermet	Zr - PuO ₂ * or Zr - Y _y Pu _x Zr _{1-y} O _{2-y/2} *
metmet	PuAl ₄ * -Al

* Fissile material phase

1.1.2. Potential IMF application

IMF in the existing fuel cycle

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 in order to increase the degree of burning/transmutation. Here the material must be leachable, if possible in the same reprocessing conditions as for UO₂ fuel. On the other hand, high burnup is not a strong criterion, since the residual fissile material will serve as fuel in another installation e.g. [28, 29].
- The spent IMF is directly sent to geological storage. Such a choice can be envisaged for a deep burn fuel because the residual Pu is of very poor quality following the large reduction of the fissile isotopes (²³⁹Pu and ²⁴¹Pu). The consequences of this choice are:
 - The material, which forms the IMF must be chemically very stable, namely 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.

IMF in existing nuclear power plants

Commercial L&HWRs are the only candidate facilities available worldwide in the near as well as mid-term to effectively transmute excess plutonium and to use its energy content. In these systems the IMF must be compatible (non-soluble, non reacting) with water, used as coolant.

As noted in Section 1.2, the objective of the studies presented in this special issue is a reduction of the current plutonium stock-pile. As a consequence, all R&D concepts are based on the following precept: the new fuel must be conceived in a such way that it should 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. 3a. The new Inert Matrix Fuel must preserve the characteristics of the reactor in terms of design, total power, length of the cycles, and safety of the plant. The loading options (Fig. 3) chosen by the different groups vary from one organisation to another [30–32].

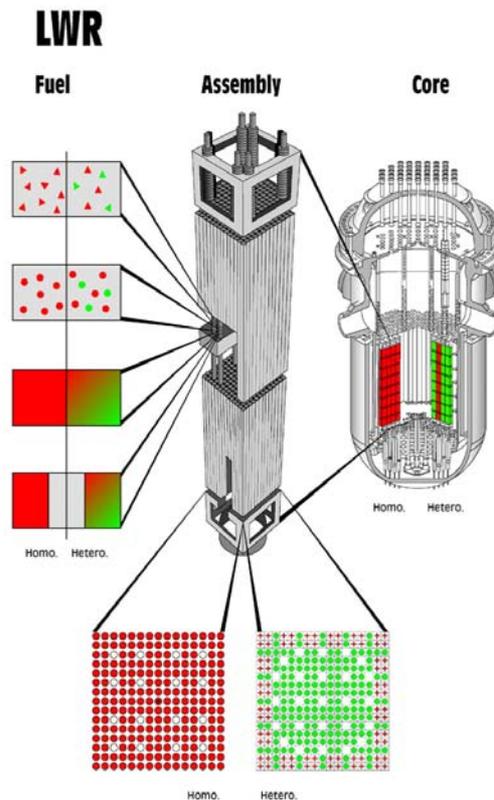


Fig. 3. The 3 main levels considered in the IMF studies for LWRs.

Some groups have studied the behaviour of 100% IMF cores (homogeneous). Other concepts foresee the partial loading of IMF assemblies in UO₂ 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 three levels for IMF utilization in light-water reactors considering homogeneous vs. heterogeneous systems at the fuel, assembly, and core levels are shown in the Fig. 3. The fuel is either a solid solution ceramic homogeneously doped with plutonium (red) or heterogeneously doped with some uranium (green), or is a composite material with particulates or micro-spheres (again plutonium-doped red, uranium-doped green) imbedded in inert matrix material. The fuel assemblies themselves may be homogeneous (all fuel rods in a given assembly contain IMF, red) or heterogeneous (red IMF rods distributed among green UO_2 fuel — e.g. the French Advanced Plutonium Assembly (APA) concept). The reactor core may also be loaded homogeneously (with red IMF assemblies), or the UO_2 core may be partially loaded with some IMF assemblies forming a heterogeneous core loading. Note: the concept of homogeneity/heterogeneity is addressed at each level; Pu phases in gray, U phases in black. 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.

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 application of IMF in PHWRs has focused on full-cores of IMF for the purpose of either actinide burning, or Pu-annihilation (with Pu from either reprocessed PWR fuel, or ex-weapons Pu).

In liquid metal cooled fast reactors (LMFR), IMFs are considered either for burning plutonium [33, 34] or for transmuting minor actinides (neptunium, americium and curium) thus reducing the radiotoxic inventory of high level wastes [35]. Here, IMF must be compatible with the liquid metal coolant used.

Transmutation of americium as an example is a complex process of neutron capture, fission and decay reactions, of which neutron capture leads to the formation of other actinides, including long-lived ones. The extent of fissioning of the produced actinides must be very high (>90%) in order to effectively reduce the amount of americium and to limit the production of curium. To achieve this, one has to optimize the neutron flux and neutron energy by choosing the highest available neutron flux and a thermalized neutron spectrum e.g. [36]. One way of achieving this is by moderating the neutron flux in a fast reactor, since flux boosting in a LWR is difficult. For such optimized conditions a once-through cycle could be sufficient. Alternatively, a multi-recycling scenario should be envisaged.

The utilization of IMF in high temperature — gas cooled reactors (HTR–GCR) may also be considered. Its advantages were recently revisited in the frame of the roadmap [37] of the Generation IV International Forum (GIF). Basically, application of heterogeneous/-homogeneous designs is feasible for the HTR as described in Fig. 4 at the kernel (fuel microsphere), pebble (assembly) or core level. The later is presented in Fig. 4 as a flow through system, while it may also be constructed as a static (bloc) system. Here, the coolant is a gas (He), and materials are not specifically affected by this inert gas.

Development of a gas cooled fast reactor (GFR) [38] is attractive due to the following characteristics: enhanced economic competitiveness, enhanced safety, the use of proliferation resistant technologies and processes, flexible and efficient use of natural resources, and a significant reduction of long life radioactive elements in produced waste.

The design goals for the core and the associated fuel cycle lead to the necessity of new fuel types fabricated with innovative materials.

Inert matrix fuels are used to satisfy the objective of large thermal margins and fission product retention, through the use of heterogeneous fuels with refractory matrices.

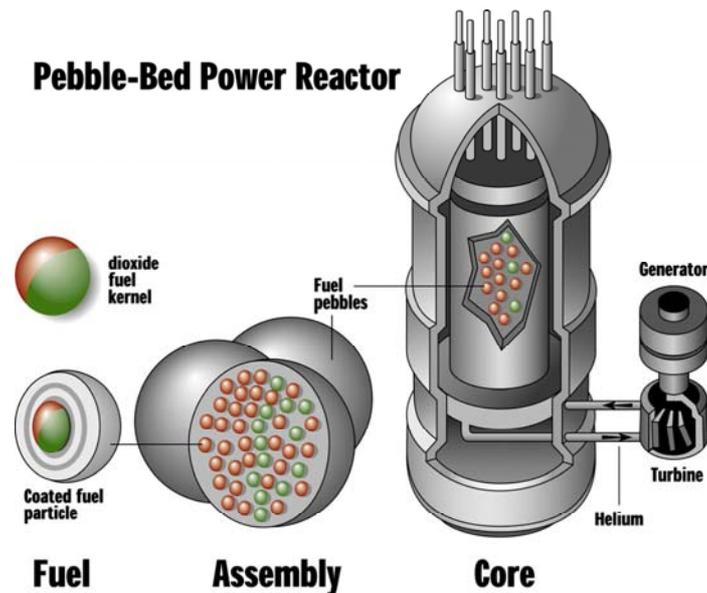


Fig. 4. The 3 main levels considered in the IMF studies for high temperature reactors.

1.2. Goals and advantages

The goals and advantages for the development IMF are described below.

1.2.1. Economics

Inert matrix fuel fabrication, to be economically attractive, should make use of fabrication technologies similar to today's standard MOX fabrication processes. Fabrication of plutonium-bearing (and MA-bearing) fuel is expensive relative to UO_2 in terms of both capital and production, cost benefit can be realized through a reduction in the number of plutonium-bearing elements required for a given burn rate. In the reactor, a fraction of the core smaller than that required with MOX is needed 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 provides some economic incentives. However a comparative cost analysis of IMF has not been carried out.

1.2.2. Ecological

The appropriate management of radioactive waste arising out of back end of the nuclear fuel cycle is considered to be one of the crucial solutions for long term environmental concern. The basis of this concern with the disposal spent nuclear fuel or high level waste (HLW) as

nuclear waste by isolation from the biosphere in a stable deep geological formation for at least tens of thousands of years or even very longer times during which this waste must be separated from the biosphere to avoid possible harmful effects. The long term hazard of the spent fuel or HLW is primarily associated with the actinides particularly the Pu and minor actinides with a long term risk linked to the likelihood of mobility of these radiotoxic elements in the geo-sphere with possibility to enter the biosphere. It is considered that the long term radiological risk is a combination of potential hazard and confining properties of the geologic strata. The measures which have to be taken for hazard reduction are very different and much more fundamental than those for risk reduction. Plutonium disposition using IMF permits a reduction of the inventory of plutonium and minor actinides in the final waste forms thus reducing the toxicity at the source itself. Since, IMF approach has the potential to reduce the repository space requirements as well as the time-period to terminate the disposal site and in addition to easing safeguards requirements for the repository. In addition, the vastly reduced inventory of radionuclides by incineration with the use of IMF will also mitigate the effects of inadvertent intrusion scenarios assuming reduced concentration and optimization of final waste form in the repositories. This reduction, of course, matches the ecological goal. However, specific environmental studies are also required to understand the long term behaviour of the spent inert matrix fuel, e.g. the role of metal for cermet (for Zr see [39]) or other ceramics [40] with regard to the conditions of a geological disposal. For example, zirconia forms a single phase with actinide dioxides or rare earth sesquioxides, and spinel fixes fission products such as alkaline earth elements. Such questions need to be addressed by specific investigations concerning the matrix solubility and corrosion rate in typical near field and far field environments and by suitable natural analogue studies.

1.2.3. Safety

There are two safety aspects to be discussed: one concerns the utilization of the IMF in reactors and the other one the material handling itself.

The IMF material handling prior and after irradiation must be safe compared to the classical UO₂ or MOX fuel case. Particular work addressing the high burn up objectives and the core behaviour with respect to transient and accident conditions is needed to complete IMF conceptual studies. The reactivity coefficients must be considered in the context of all applicable accident scenarios.

1.2.4. Proliferation resistance

The important factor in assessing various fuel cycle options is the relative proliferation resistance of various closed fuel cycle strategies in comparison to the once-through fuel cycle. A comparative analysis has been done recently by an international expert group for the US DOE to evaluate the non-proliferation characteristics of advanced fuel cycle [41]. The methodology created in this analysis gives a proliferation risk score for every segment of the nuclear fuel cycle and then sum the risks over time. This study has concluded that the integrated proliferation resistance measure of a fuel cycle intended to transmute minor actinides, if properly designed, has the potential to be roughly equal to that of the spent fuel standard; the Inert Matrix fuel cycle is particularly notable in this regard” [41]. Successful application of IMF will enhance non-proliferation attributes of a fuel cycle as it reduces the inventory of plutonium as well as alters plutonium isotopic composition minimizing the likelihood of diversion. The characteristics that make IMF an effective non-proliferation concept lie in its ability to utilize excess plutonium while producing no additional weapons

usable material. This will minimize the likelihood of excess defence material returning to military application by consuming material and/or reducing the isotopic quality. This also applies to civil separated plutonium.

1.3. History (before 80's)

Development of inert matrices could be traced to very early in the 1960's, with a view to develop a matrix to allow utilization of plutonium for energy production. A Westinghouse report published in 1962 [42] documents irradiations of 94 fuel elements of $ZrO_2 - UO_2$ performed in the National Research Universal reactor at Chalk River, Ontario and the National Reactor Test Station in Idaho. PIE Tests with ZrO_2-UO_2 were also completed after in-pile irradiation [43].

A description of utilization of ZrO_2-UO_2-CaO fuel in the power burst facility at the Idaho National Engineering Facility with no fuel rod failure up to a temperature to 2420 K is given in a 1966 report [41]. Between 1969 and 1974, a ternary fuel consisting of 57 ZrO_2 -38 UO_2 -5 CaO (wt%) was successfully irradiated in the Shippingport pressurized water reactor in Pennsylvania [45]. The first tests with ZrO_2-PuO_2 were performed at Hanford [46, 47]. In 1965, a study was conducted identifying ZrO_2 as a diluant for UO_2-PuO_2 [48]. These R&D projects were based on important early studies reported in the fifties and early sixties [49–51].

1.4. Current programmes

Current IMF programmes focus on specific work on fabrication, characterization, irradiation (with accelerators or in research reactors,) and modelling for predicting behaviour in commercial reactors.

Early literature is available on $MgAl_2O_4$, examination for fusion materials research programme [52], and subsequently on the IMF applications in standard reactor [53].

MgO is tested in US [54] and French programmes (MATINA in Phenix, and EFTTRA-T2 in the High Flux Reactor, Petten, Netherlands) [55] as well as in the BOR-60 and BR-10 reactors [56] showing that neutron-induced swelling is of the order of a few percent for rapid neutrons ($E > 0.1$ MeV) fluence: $2 \times 10^{26} \text{ m}^{-2}$ with plutonium to very high burn up. Currently, the US advanced fuel cycle Initiative (AFCI) LWR transmutation fuel development program plans to test inert matrix fuel in the LWR-2 irradiation series in the advanced test reactor, located at the Idaho National Laboratory. The goal of the program is to develop fuel compositions that provide for flexibility in management of the overall LWR fuel cycle and ultimately reduce the amount of material requiring repository disposal. The most direct strategy for management of the fertile burden in a nuclear fuel cycle is to replace the ^{238}U rich-depleted uranium matrix with a matrix material having a low or negligible fertile content.

Several tests and studies on cermet utilization are reported for ship-propulsion reactors [57]. The TANOX tests of a 64 Mo-36 UO_2 (vol%) cermet in the SILOE reactor, Grenoble, France, with burn up of $16 \text{ GW} \cdot \text{d} \cdot \text{t}^{-1}$, core temperature of about 793 K for a linear power density of $350 \text{ W} \cdot \text{cm}^{-1}$ reduced to about 693 K under PWR conditions [58]. U-Mo-Mg and UO_2 -Mg cermets have been in commercial operation in Russia starting with the first NPP commissioned in 1954 [59].

In the case of ceramics, fabrication of IMF powder can be produced following either a wet or a dry preparation route. 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 micro-sphere production by gelation. The products are dried and calcined [14, 15]. Hydrated salt 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 utilizing attrition milling. Pelletising is carried out prior to sintering at a given temperature and for a given time. For cermets hot extrusion is also used in some fabrication processes.

Characterization 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 microscopes are used to study pore or grain structure.

These characterization studies on IMF are also completed by irradiations works using research reactors or accelerators. Accelerators are used to study microstructural changes during irradiation due to ion bombardment. Satisfactory behaviour under ion irradiation i.e. no swelling and no amorphisation, is essential criterion for the material selection. 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 reactor. The first series of irradiations in Japanese, French, Dutch, Canadian, US and Russian reactors have already been performed. Irradiations are also carried out in the OECD Halden reactor and in the framework of joint programmes in the high flux reactor at Petten. Emphasis is given to energy production and transport (temperature measurements in the pin), mechanical behaviour, and the fission product release.

Neutronic modelling at the pellet, pin, and assembly levels is carried out prior to conceptual studies for application in commercial reactors. The components: fissile phase/burnable poison, and inert matrix components of the IMF are selected and their respective concentrations are optimized according to the neutronic characteristics.

Immense interest in the IMF programmes has induced collaborative work among several organizations. Work has been carried out or is currently in progress in Canada, at the Atomic Energy of Canada Limited; in France, at the Commissariat de l’Energie Atomique, (CEA) Saclay, Cadarache and Grenoble and at the University of Paris; in Italy at the Politecnico di Milano, the ENEA and the University of Trento; in Japan at the Japanese Atomic Energy Research Institute, at the Japan Nuclear Cycle development institute, at the University of Tohoku and at the University of Kyushu; in the Republic of Korea, at the Korean Atomic Energy Research Institute; in the Netherlands, at the Nuclear Energy Center of Petten and at the University of Delft; in Russia, at the IPPE and VNIINM; in Switzerland, at the Paul Scherrer Institute, at the Ecole Polytechnique Fédérale, Lausanne and at the University of Geneva; and in the United States, at the University of Ann Arbor, the Massachusetts Institute of Technology, Argonne National Laboratory, Los Alamos National Laboratory, and Oak Ridge National Laboratory. At the multinational level, the OECD Halden project, Norway and the European Joint Research Center at Karlsruhe, Germany have substantial projects on IMF. Institute for Transuranium Elements, JRC/EC is developing methods for the development of zirconia-based transmutation targets. Production methods based on combinations of the sol-gel and infiltration routes have been developed, both to minimize waste, and reduce potential radiation exposure to the operating personnel. The initial results on the production and property determination of (Zr,Pu) O₂ and (Zr,Pu,Am) O₂ homogeneous materials are recently published [60]. There are some studies aiming the development zirconium carbide ZrC as an

inert matrix material an alternative to SiC based inert matrix, owing to its high melting point, good mechanical property and stability against radiation is under consideration.

1.5. R&D activities for IMF qualification

Before utilization in a commercial reactor, IMF elements or assemblies require licensing, and qualification by the local nuclear regulatory commission or authorities. This demands certification of the material and design, quality control of the element production, and predicting the in-pile behaviour for core configuration optimisation.

Qualification of IMF needs to address the following items under normal reactor operation and for accident conditions:

- crystallographic evolution,
- fission product diffusion and release,
- effect of the several sources of in-pile damage,
- fuel restructuring (swelling, cracking, of redistribution of fissile isotopes),
- thermal and mechanical behaviour.
- core neutronic considerations [61].
- power burn up

This TECDOC is meant to provide an overview of the scope of current programmes on IMF. The sections, which follow develop more specific aspects of this domain concerning an important fuel cycle option, which aims at greater economy and ecological acceptability.

2. NATIONAL PROGRAMMES

The following will describe national programmes of the countries in alphabetically order.

2.1. Canada

Canadian effort on inert matrix fuels has been led by Atomic Energy of Canada Limited (AECL) with the participation of Queen's University in Ontario. This effort has focused on reactor physics analysis of the use of inert matrix fuels in a CANDU reactor, and on assessment of inert-matrix fuel candidates. The application has been either plutonium destruction or actinide burning. On-line refuelling, a simple bundle design, and high neutron economy provide the CANDU reactor with flexibility in these applications. A full core of inert matrix fuel, containing either plutonium, or a plutonium/actinide mix, could be used to fuel existing CANDU reactors, with high destruction rates.

2.1.1. Assessment of IMF candidates, SiC fuel fabrication and irradiation

AECL has done preliminary evaluations of a number of inert-matrix fuel candidates, including $ZrSiO_4$, $MgAl_2O_4$, CeO_2 , $CePO_4$, ZrO_2 doped with CaO , CeO_2 , Er_2O_3 or Y_2O_3 , and SiC. Ceria (CeO_2) has the same crystal structure as urania, and therefore was thought likely to behave well in-reactor. Zircon ($ZrSiO_4$) is a stable mineral found naturally in the earth's crust, and therefore should at least be a stable waste form, and was also being evaluated as a candidate for hosting military plutonium for direct burial in the ground. Cerium phosphate ($CePO_4$) was considered as a candidate material and studies are carried out in collaborations with ITU. Silicon carbide stood out early as a promising candidate material because of its high melting temperature and very high thermal conductivity, and because of its known resistance to attack by many corrosive agents, including oxygen, even at high temperatures.

SiC fuel fabrication

Pressureless sintering of SiC normally requires temperatures of 2 000°C or higher. AECL has worked with Queen's University to develop methods of fabrication at lower temperatures, which are more representative of UO₂ sintering. In addition, fabrication tests were performed with cerium as an additive, as a chemical substitute for plutonium.

Good progress has been made in establishing methods of fabricating SiC containing a wide range of cerium concentrations [62]. The methods are similar to those in current use for conventional UO₂ fuel production. High densities have been achieved and the required sintering temperatures have been reduced from over 2 000°C to approximately 1 700°C by judicious use of sintering additives. In fact, ceria itself has proven to be a most effective sintering additive. High-density material was fabricated without using cold isostatic pressing, a step not used in UO₂ fuel production. Cerium is usually incorporated into the SiC as an AlCeO₃ phase, often as micron-sized particles, but also as larger particles (6–8 µm) under some fabrication conditions. Above a sintering temperature of 1 830°C with Al₂O₃ and Y₂O₃ sintering aids, X ray diffraction did not reveal the evolution of any phases except SiC. This suggests that all additives are taken into solid solution, which is not fully understood.

Measurements on the specific heat capacity and thermal conductivity of SiC specimens fabricated in this programme were reported [58]. Although the thermal conductivity is lower than for pure, full-density SiC, it remains sufficiently high that the central temperature in an operating fuel at 55 kW/m would only be about 100°C higher than the coolant temperature at the beginning of life.

The thermal conductivity of such SiC-based inert-matrix fuels is high relative to UO₂ and MOX. If high conductivity is maintained during irradiation, SiC-Based IMF will provide a clear benefit from the perspective of in-reactor fuel performance and safety.

Irradiation behaviour

A critical parameter for candidate matrix materials is performance when exposed to high-energy fission fragments. AECL used its TANDEM accelerator to bombard candidate materials with a beam of 72 MeV iodine ions. This beam substitutes for fission fragments. And the bombarded areas were examined for damage, especially swelling.

The following candidate materials: ZrSiO₄, MgAl₂O₄, CeO₂, CePO₄, ZrO₂ doped with CaO, CeO₂, Er₂O₃ or Y₂O₃, as well as SiC, were tested over temperatures ranging from ambient to 1 200°C, and over a wide range of ion doses (10¹⁴–10¹⁷ cm⁻²) [63]. An ion dose of 10¹⁷ cm⁻² will corresponds approximately to a CANDU fuel burnup of 20 MW(h)/kg U. After accelerator bombardment of each sample, the surface relief of the 3 mm diameter beam spot was measured. Significant height of the spot above the original surface was taken as an indication that in-reactor swelling would occur. In general, results were not strongly dependent on dose — if a candidate material showed swelling at high dose, it also showed swelling at lower doses. The results were also reasonably independent of temperature. The materials that showed the best results (i.e. least swelling) were SiC and ZrO₂ with any dopant. These candidate materials did not show any swelling, i.e. LASER profilometry could not detect any surface relief at the implantation spots. Ceria turned out to be one of the poorest candidate materials on the basis of these tests. All other candidate materials showed some surface relief, implying that swelling could be expected when used in-reactor as a fuel.

2.1.2. Reactor physics, safety assessments and waste management

Reactor physics with plutonium/actinide mix in an inert matrix fuel

Detailed, full-core fuel management studies have been conducted [64] for an existing CANDU 6 reactor using the reactor fuelling simulation programme RFSP (Reactor Fuel Simulation Program) to assess the power distributions and reactivity effects of fuelling with an inert-matrix fuel containing either ex-weapons plutonium, or a mixture of civilian-plutonium and minor actinides.

For the burning of plutonium and minor actinides (MA) obtained from the reprocessing of spent LWR fuel, one exercise considered for simulation consists of 356 g of plutonium plus 66 g of minor actinides in the two outer fuel rings, and 20 g of gadolinium in the two inner rings of a CANFLEX bundle. The central element contains 10 g of gadolinium and the remaining 10 g of gadolinium are distributed uniformly over the seven elements in the next ring. Gadolinium is used to suppress the excessively high reactivity of the fresh fuel. Each of the two outer fuel rings contains 178 g of plutonium and 33 g of minor actinides mixed with SiC. The Pu isotopic composition corresponds approximately to spent PWR fuel having an initial enrichment of 3.5% and a burnup of ~ 35 MW·d/kg (HM). The ratio of minor actinides to plutonium is about 50% greater than that in the unadjusted spent PWR fuel. The initial material composition of the civilian (plutonium/actinide)-SiC fuel bundle is shown in Table IV.

TABLE IV. INITIAL COMPOSITIONS OF CIVILIAN PLUTONIUM-ACTINIDE IMF IN A CANFLEX FUEL BUNDLE

Isotope	Composition (g)			
	Ring 1 (1 pin)	Ring 2 (7 pins)	Ring 3 (14 pins)	Ring 4 (21 pins)
^{238}Pu	—	—	3.0	3.0
^{239}Pu	—	—	102.5	102.5
^{240}Pu	—	—	47.8	47.8
^{241}Pu	—	—	15.5	15.5
^{242}Pu	—	—	9.3	9.3
Total Pu	—	—	178.1	178.1
^{241}Am	—	—	15.37	15.37
^{243}Am	—	—	2.65	2.65
^{237}Np	—	—	14.98	14.98
Total MA	—	—	33.00	33.00
Si (natural)	139.6	977.4	1399.0	2098.5
C (natural)	59.8	418.9	599.6	2098.5
Gd (natural)	10.0	10.0	—	—

The key point is that coolant void reactivity is small and negative and the power coefficient is also negative. There would be no power pulse in a postulated “loss of coolant accident”

(LOCA), and the safety and licensing analyses would be greatly simplified. The fuel temperature coefficient is very slightly positive. However, the sign of the fuel temperature coefficient is not of great importance with this fuel, since the high thermal conductivity of SiC prevents any dramatic increase in the fuel temperature without a corresponding increase in coolant temperature. The increase in coolant temperature would reduce the coolant density and produce a negative reactivity feedback owing to the negative coolant void reactivity.

Detailed, realistic fuel management simulations were performed using RFSP for a standard CANDU 6 reactor with a full core load of civilian (plutonium/ actinide)–SiC fuel. Acceptable bundle and channel powers were obtained using a bi-directional, two-bundle-shift refuelling scheme. The refuelling rate is 9.5 bundles per full-power day, easily within the capability of the current fuel handling system.

The time average channel power distributions are very similar to those in the present natural UO₂ fuelled CANDU reactor. Bundle powers are high at the end of inlet because of the rapid depletion of the fissile plutonium. This axial power shape, which peaks towards the inlet (refuelling) end of the channel, will improve the thermal hydraulic margins (higher channel powers before dryout occurs). The maximum instantaneous channel power is less than 7.1 MW, and the maximum instantaneous bundle power is less than 990 kW. The fuel temperatures would be very low because the thermal conductivity of the SiC matrix is much greater than that of UO₂.

Table V. shows the plutonium and minor actinide content in fresh and discharged civilian (plutonium/actinide)–SiC fuel bundles. The discharge fuel burnup is 229 MW·d per bundle. In comparison, the fuel burnup of a natural-uranium CANDU fuel bundle, which contains 133 g of ²³⁵U, is about 136 MW·d per bundle.

TABLE V. PLUTONIUM AND MINOR ACTINIDE DESTRUCTION IN (PLUTONIUM/ACTINIDE)–SiC BUNDLES FOR 229 MW d

Isotope	Civilian (plutonium/actinide)–SiC fuel content		
	Fresh (g/bundle)	Discharge (g/bundle)	Destruction (%)
²³⁸ Pu	5.9	5.5	6.8
²³⁹ Pu	205.0	6.2	97.0
²⁴⁰ Pu	95.6	57.5	39.9
²⁴¹ Pu	31.0	15.8	49.0
²⁴² Pu	18.5	41.5	-
Total Pu	356.0	126.5	64.5
²³⁷ Np	29.96	14.76	49.3
²⁴¹ Am	30.73	3.69	88.0
²⁴³ Am	5.30	9.94	—
²⁴² Cm	—	8.58	—
²⁴⁴ Cm	—	3.96	—
Total MA	65.99	40.93	38.0

The civilian (plutonium/actinide)–SiC option gives a net destruction rate of 1200 kg of plutonium and 130 kg of MA per GW(e)·a. About 97% of the initial ²³⁹Pu and 88% of the

initial ^{241}Am are destroyed; the net destruction efficiency of the MA is 38%. LWRs produce about 268 kg of plutonium and about 33 kg of MA per GW(e)·a. One CANDU 6 reactor can effectively destroy much of the plutonium and minor actinides produced in three 1 GW(e)-size LWRs.

The fuel design, or fuelling strategy, has not yet been optimized to maximize the actinide destruction rate. There is great flexibility in choosing the amounts, relative fractions, and locations of plutonium and minor actinides in the bundle. The combination of on-power fuelling, high neutron economy, and simple and flexible fuel bundle design offers many possibilities for actinide burning in existing CANDU reactors.

Simple tests of fuel/cladding material compatibility at high temperature were performed by holding polished discs of SiC and Zircaloy 4 under light pressure in an argon atmosphere at temperatures of 1000, 1500 and 1700°C. The temperatures were maintained for 1 h at 1000°C and 1500°C, and for 15 min at 1700°C. After cooling and sectioning, the specimens were examined by optical and scanning electron microscopy to study the extent of interaction.

Although no significant interaction occurred at 1000°C, at 1500°C there was clear evidence of a diffusion-based reaction to form ZrC and free silicon. Diffusion of free silicon into the Zircaloy disc led to the formation of a molten zirconium–silicon rich eutectic phase. This reaction was more pronounced in the 1700°C test specimens, where the amount of molten eutectic phase was sufficient to cause partial dissolution of the molybdenum cell sidewalls.

The test results indicate that formation of a molten zirconium rich phase could conceivably occur during a hypothetical reactor accident at temperatures significantly lower than the melting point of unoxidized Zircaloy cladding (1760°C). This could allow further accelerated attack on the SiC carrier material, thereby allowing any fission products that had exsolved from the plutonium phase to be released. However, UO₂ interaction with Zircaloy begins at about 1200°C, and it dissolves into molten Zircaloy at 1760°C [65]. The rate and extent of UO₂–Zircaloy interaction depends on the amount of oxygen present. It thus appears that the SiC interaction with the cladding is, at least, no worse than that of UO₂. SiC based inert matrix fuels have shown favourable qualities which make them to stand as suitable materials for thermal reactor systems. However, extension of SiC as inert matrix form for fast reactors using steel cladding is not suitable owing to the thermodynamic incompatibility of SiC–austenitic steel system [25].

Waste management

Considerations to be borne in mind in using SiC-based fuel as a waste form for direct disposal include the following:

- The SiC matrix itself, with sintering aids used to date, will not generate long-lived activation products in-reactor. Care needs to be taken to minimize the impurity levels of nitrogen and chlorine to eliminate concerns about their activation products ^{14}C and ^{36}Cl .
- Silicon carbide itself is extremely stable and resistant to corrosion. Its performance as a waste form will depend on its capability to encapsulate and retain high burnup plutonium-containing particles. It is likely that the plutonium-rich phase will remain encapsulated, and hence be protected from leaching. This needs to be verified by post-irradiation examination and leach testing of irradiated inert-matrix fuel materials.

Plutonium destruction in an inert matrix fuel

The destruction of plutonium from dismantled warheads can also be effectively accomplished by burning in an inert matrix fuel in a CANDU reactor. The CANFLEX bundle design would be similar to that described above, with gadolinium in the central element and first ring of 7 elements, and plutonium in the outer two rings. Again, a full core of plutonium in an inert matrix can be accommodated in an existing CANDU reactor, largely due to the on-power refuelling. High neutron economy in CANDU results in high destruction rates: 98% of the initial ^{239}Pu , and 75% of the total plutonium is destroyed.

2.2. France

Studies on inert matrix fuels in France are carried out for two different programmes considering that:

- the plutonium consumption and the transmutation of wastes (mainly minor actinides) requires U-free fuels in the French standard reactors (PWR, FR, advanced PWR),
- for some types of generation IV reactors, IMF may contribute to fulfill all the requirements for these new nuclear power plants.

First, the IMF programme must be integrated in the existing fuel cycle based on the PUREX process. The introduction into reactors will be done with a special sub-assembly, composed with IMF-pins and standard ones. This special sub-assembly will have its own characteristics: adapted neutronic parameters, thermohydraulic behavior and specific irradiation conditions (2.2.1).

For new reactor types, in particular the GFR, IMF development is made in a very different context because reactor, fuel and fuel cycle are new and must be designed together at the same time. In these three fields, innovative conception is required in order to reach all the generation IV criteria that will be mentioned in paragraph 2.2.2.

Some of the IMF developmental works are carried out in CEA with the collaboration of EDF and FRAMATOME. Some works more linked to material science are studied by CEA and CNRS or other universities. Furthermore, CEA is involved in several R&D international collaborations aimed to develop technologies for transmutation or Pu burning. First of all, the Experimental Feasibility of Targets for TRAnsmutation (EFTTRA) collaboration [66] by the organizations Electricité de France (EdF) as well as CEA from France, Forschungszentrum Karlsruhe (FZK) from Germany and the Nuclear Research and Consultancy Group (NRG) as well as the Institute for Advanced Materials, from the Netherlands and the Institute for Transuranium Elements, (ITU) Karlsruhe of the Joint Research Centers (JRC-IAM and JRC-ITU) of the European Commission is working jointly on Partitioning & Transmutation issues. A valuable set of data has already been gained from the experiments performed in the HFR reactor of Petten. Various inert matrices have been tested with and without inclusions of fissile material together with experiments on long live fission products (LLFP). A collaborative work-programme of CEA and MINATOM (now called as 'ROSATOM' Russia's Federal Atomic Energy Agency), is underway in Russia. The Bora-Bora irradiation [56] in BOR60 was designed to test various fuel concepts developed in the frame of Pu management programme with the test of the composite $\text{PuO}_2\text{-MgO}$ and the solid solution (Pu,Zr)N that will bring data on the behaviour of these both fuels. The determination of fundamental properties of zirconium pyrochlore as the host phase for Am-Cm in a composite target was made with the contribution of ORNL [67]. On the mid-term and in the frame of

bilateral collaboration agreements with JNC and the Japan Atomic Energy Research Institute (JAERI) (now renamed as JAEA after merging JNC&JAERI), works are planned on oxide compounds for the homogeneous mode and also on the properties of nitrides.

2.2.1. Plutonium management and waste transmutation

Since plutonium is both a recyclable energy material and the main contributor to potential long term radio toxicity, the start point of all the scenarios is the management of plutonium in the current fleet of nuclear reactors and the research in transmutation is connected to the studies linked to the plutonium consumption [68], including the advanced concepts such as APA, DUPLEX programme [69, 32]. These requirements induce research to develop fuels with good thermal characteristics, able to sustain high temperatures, to reach high specific power and high burn ups.

The scientific feasibility of plutonium management and waste transmutation has been examined for the different cases [72] which constitute several scenarios with their own individual characteristics: park of reactors (one reactor type or combination of 2 types of different kinds of reactors such as thermal and fast reactors which is also termed as “double strata” scenario), ways to transmute (homogeneous or heterogeneous fuel), fuel management (multi-recycling, or once-through). The scenarios are not equivalent and must be evaluated with regard to the technical feasibility and the impact on the fuel cycle.

For the management of high level and long lived radioactive waste [73] in the frame of the French Law of December 91 [74], the potential of a partitioning and transmutation strategy to reduce the quantity and the toxicity of the waste is under evaluation (along with the other alternatives such as disposal in deep geological formations or long term interim surface or subsurface storage). The research is centered on minor actinides, which represent the majority of the long term radiotoxic elements in the waste, together with plutonium and LLFP such as technetium, iodine and caesium. The objectives of the partitioning studies are to develop chemical processes to obtain advanced partitioning of radionuclides to complete the partitioning of uranium and plutonium. Then, one way of the above scheme, consists in burning these radionuclides in reactor through dedicated targets or special fuels.

The objectives of the transmutation research programme are to evaluate the transmutation potential of long lived waste in appropriate reactors configurations (scenarios) relying on current technologies namely PWRs and FRs as well as in innovative “incinerator” reactors (with dedicated systems such as the accelerator driven systems (ADS)) and also to study the materials to be used for the new type of fuels suitable for transmutation.

These studies lead to consider the development of homogeneous recycling of Pu and MA in PWR (with multi-recycling of Pu [15]), for fast reactors (with either mono-recycling of MA and LLFP in moderated targets or multi-recycling in quasi standard fuels) and heterogeneous recycling of Pu or MA in IMF in order to prevent further formation of transuranic elements. This type of IMFs matrix are dedicated to Pu burning (sometimes with some MA) or only MA transmutation for which IMF are called targets. In both cases, IMFs can be either a solid solution of actinide elements in matrix or composite fuel or a bi-phased materials with the actinide compound and the matrix support. The R and D programme on IMF is presented below according to these topics.

IMF studies for plutonium burning

In addition to some solid-solution of plutonium and MA in homogeneous mode, which may prevent the formation of ^{239}Pu from neutron-capture of ^{238}U , two other options are under investigations using an inert matrix in which plutonium and actinides can be dispersed. These concepts are investigated considering:

- standard geometry for the rods that are of two kinds, one including UO_2 fuel and the other containing composite fuel (DUPLEX concept). The DUPLEX assembly consists of a heterogeneous arrangement of PuO_2 in an inert matrix of UO_2 rods surrounded.
- in contrast it is also considered that incineration of plutonium in thermal neutron spectra has some restrictions due to the deterioration of plutonium isotopic composition and core neutronic and safety aspects, and the limitation in the PUREX reprocessing technologies. Nevertheless some of these shortcomings are being surmounted by innovations in fuel and core design. For example CEA has proposed some innovative methods for recycling plutonium and minor actinides in PWRs based on heterogeneous distribution of uranium and plutonium pins in a fuel sub-assembly (Advanced Plutonium Assembly-APA and CORAIL MOX sub-assemblies) allowing to keep also an acceptable power shape [69, 70]. A few of these safety drawbacks may be mitigated by changes of sub-assembly lattice design in order to achieve satisfactory spectrum thermalisation. This is accomplished either by an increase of moderator/fuel ratio (up to 3.5–4) — so called high moderation PWR (HM-PWR), or by a radical decrease of the fuel smear density keeping the same lattice geometry [71].

The following inert materials have been studied:

- metal matrices for cermet: Mo [76], W [77], Zr [78],
- oxide matrices for cermet: MgAl_2O_4 [79] and Al_2O_3 [80],
- oxide matrices for solid solutions: stabilized ZrO_2 as $(\text{Zr},\text{Y},\text{Pu})\text{O}_2$, CeO_2 as $(\text{Ce},\text{Pu})\text{O}_2$.

For this programme, two composites: Mo - UO_2 and MgAl_2O_4 - UO_2 were successfully irradiated in the Siloé reactor with important results on the fuel behaviour [81].

These experimental results were completed by studies on the neutronic effects of the introduction of IMF in a PWR [82, 83] and the differences between several matrices [84]. Furthermore a prospective analysis was made on the specific behaviour of cermet fuels during severe accidents [85].

R & D programme on targets for MA transmutation: heterogeneous mode

The performances of a metallic fuel with a low percentage of minor actinides and rare-earths, are being studied in the METAPHIX experiments [86] conducted in the scope of a contract with ITU on behalf of CRIEPI in Japan. Fuel is a metallic alloy (MA,Pu,U,Zr) with a sodium bond. The aim of IMF or targets is to be efficient for transmutation in the heterogeneous mode and to allow a good level of safety in case of incident or accidents. The requirement for a transmutation strategy is fission up to 90% with a MA content of 1–2 $\text{g}\cdot\text{cm}^{-3}$. The respective composition of the different parts of these composites are optimized to take into account the different effects induced by irradiation. The main requirements are good thermal and mechanical properties for the matrix and chemical stability in the course of the evolution of the actinide phase. Possible ceramic or metal candidate materials have been selected with criteria concerning their basic properties (such as thermal and mechanical properties,

activation with neutrons, chemical compatibility with neighboring materials) and their behaviour under irradiation.

The IMF considered for this programme were both composites and solid solutions with an experimental programme based on IMF irradiations [103]. The inert matrices identified [87] for cermet or cermet fuels were MgO, MgAl₂O₄, Al₂O₃, ZrO₂, CeO₂, Y₃Al₅O₁₂, Y₂O₃, TiN, W, Nb, V and Cr.

Inert matrices

In order to design specific IMF methodologies adapted to the objectives of transmutation, the R&D programme has covered first the basic materials qualification (as mentioned above) and also the in-pile test [88]. For the material qualification under irradiation, the experiments were designed to study the different effects:

- fast neutron fluence with experiments like MATINA [57], and EFTTRA T2 [89];
- fission products damage with ion irradiations in accelerators [90],
- alpha interaction with helium implantation experiments started in the frame of the EFTTRA programme [90],
- effect of fast neutrons and fission products in irradiations like MATINA and EFTTRA T3 [91], T4 ter [55].

Fissile and fertile compounds

In the same time, several actinide compounds were studied: NpO_x, AmO_x [92] and (Am, Zr)O_x [67].

The phase diagram of americium and oxygen system is very complex. And americium oxide exhibits high 'oxygen-potential' and a high chemical reactivity towards the matrix or the coolant. The oxide form of americium has been considered in the first irradiation experiments (EFTTRA T4 [93], ECRIX [94]) and an alternative to direct oxide form of americium is now proposed with a solid solution of AmO₂ and ZrO₂: (Am,Zr)O_{2-x}. Indeed, some zirconia based solid solutions present very attractive properties considering the analogy with (U,Zr)O₂. Targets containing (Am_{0.5},Zr_{0.5})O₂ and Am₂Zr₂O₇ (pyrochlore form), were both considered and characterized at ORNL in the frame of a collaboration that may be extended to the same targets with curium dioxide.

Inert matrix fuels

Experiments are now performed to test the in pile behaviour of the different elements and of the composite itself.

The first targets were based on a concept of a micro dispersion of the actinide phase in the host matrix and many of experiments have been performed based on two types of matrices: MgO-AmO_x and MgAl₂O₄-AmO_x.

Magnesia based targets have been fabricated for the ECRIX experiments in Phénix, and samples were used for the property-measurements [95] of the composite (namely thermal characteristics, oxygen potential, melting point, heat capacity, thermal expansion and diffusivity) as well as specific tests like compatibility with sodium in collaboration between ITU and CEA. The irradiation will give indication on the behaviour of the magnesia-based concept as a candidate for americium transmutation.

Further irradiation experiments are planned to select the optimized composite and the americium targets. A research axis will concern the stabilization of the americium oxide in a cubic structure of the $(Am,Zr,Y)O_{2-x}$. The CAMIX irradiation [96] will cover the optimization of the actinide compound. A second axis of the investigation will concern the dispersion mode of the americium compound in the inert matrix considering the macro dispersed concept with $(Am,Y,Zr)O_{2-x}$ in $(Y,Zr)O_{2-x}$ and MgO. Such a process applied to different matrices is to be operational for the COCHIX irradiation [92] (optimized target in PHÉNIX) that should start in the end 2003 with the objective of a fission rate equivalent to that of ECRIX, i.e. ~30 at% of initial heavy metal [97, 98].

The first fabrication of this type of fuel was made by the impregnation process. The fuel was then irradiated in the high flux reactor in Petten in two irradiations: EFTTRA T4 and T4bis where the americium burn up reached 96% and 99.8% respectively [97] after only 358.4 and 652.6 EFPD. This gives an indication that technically high transmutation is possible. However, pellets have swollen considerably (18 and 27 vol% for T4 and T4bis), as a consequence of radiation damage and gas accumulation.

Results of the experiments indicate that the target concept is to be improved to reach the ambitious objectives for the transmutation scenario with fission rate >90%, considering high the radiation damage and gas production. Improvement of the dispersion fuels is researched with the introduction of macro masses (100–300 μm diameter) to study the radiation effects due to the fission fragments or alpha decay in a small shell [100].

The experiments THERMHET [99] and EFTTRA T3 [91], T4ter were the first tests of this concept with various microstructures and were completed with adapted modelling in order to calculate its thermo-mechanical behaviour under irradiation [100].

Conclusion

The main results come from the irradiation programme that was conducted in the frame of the European collaboration EFFTRA (where three main fields of study were identified, materials for transmutation, either matrices and actinide compounds, test of target concepts) and experiments performed in the Siloe and Phénix reactors.

Most of these materials have been tested as candidates for IMF. The examination is still underway; their potential as matrices was shown for some of them [101, 102].

The studies on these new types of fuels have yielded several results for its fabrication techniques as well as several characterization methods for the basic properties of the fuel materials and fuel-behaviour under irradiation.

With a start point based on simple chemical form of the fissile elements (AmO_2 particle) directly mixed with the matrix to obtain a micro dispersed composite, the different results obtained have lead to an optimization of the concept with a greater complexity of the various parameters, the optimization elements being:

- the choice of a spherical host phase with size ranging between 50 to 300 μm in diameter;
- a MA compound in a stabilized phase of the type $(Am,Y,Zr)O_{2-x}$;
- a matrix material leading to a composite with acceptable thermal and mechanical characteristics which can be fabricated with a not too complex process. In this domain the possible choices rely on spinel, MgO and zirconia; and

- the management of matrix damage and gas production the later being still a matter of discussion the choice going from complete gas retention to complete gas release. Some work is still necessary to evaluate the possibilities of new ways to manage great volume of gases: porosity repartition (porous matrix or “jingle” concept) or coated particles might be solutions.

A first step will be reached before 2010 with the identification of the performance potential of the tested solutions through a wide range of experiments, gathered in an experimental programme [103]. This will allow the definition of a second step for 5 to 10 years to reach the ultimate objectives fixed to the selected concepts.

Fuel for transmutation in dedicated systems

The impact of the fuel composition on the performance of innovative reactors dedicated to the minor actinides transmutation is largely investigated in Europe, Japan and the USA. Since 2000, main nuclear research institutes have developed experimental programmes to design innovative U-free fuels for accelerator driven transmutors and performing common irradiation tests to qualify their behaviour under irradiation.

Among the different fuel concepts proposed worldwide, inert matrix fuels are thought to have a great potential. Inert matrices are generally selected to enhance the thermal properties of the fuel. Improvement of fuel thermal conductivity and thermal stability (Am species are generally volatile) are some of the major developmental subjects (addressing issues such as volatility of Am species) for the use of inert matrices.

In Europe, oxide composites are preferred because of the large know-how on UOX and MOX fuel technology gained for 50 years and recent development on transmutation targets for reactors of existing technology. Cermets such as (MA,Pu)O_{2-x} or (MA,Pu,Zr)O_{2-x} diluted in MgO, or, cermets such as (MA,Pu)O_{2-x} diluted in steel, Cr, V, W or Mo are developed in the frame of the 5th European “FUTURE” framework programme [104]. Nitride fuels, such as (MA,Pu,Zr)N or (MA,Pu)N - TiN are also considered as a reference fuel concept for Japan, an alternative to metallic alloys for the USA or a backup solution for Europe. A significant effort on the nitride fuel development is therefore made in Europe through the 5th European CONFIRM programme. (Pu,Zr)N pins are being designed for an irradiation in the Studsvik reactor in 2004 [105].

From the beginning of 2003, a joint programme between CEA, ANL, LANL, ITU and JAERI was launched. It consists of irradiations in Phénix of U-free and low fertile fuels of three different types: oxide-IMF (bonded with helium), nitride-IMF (bonded with sodium) and metallic alloys (bonded with sodium). The objective of this irradiation test is to compare in similar and representative conditions (spectrum, flux and temperature) the main fuel options of dedicated fuels, as described above. Assessment and comparison of their behaviour under irradiation in a fast spectrum are required to improve fuel performance, which is so far restricted because of the poor knowledge on the basic irradiated and un-irradiated fuel properties.

A CEA/NIMATOM work programme is in progress in the Russian Federation. The BORA-BORA irradiation in BOR60 is designed to test various fuel concepts in the frame of CARRA Pu Management programme including the test of PuO₂-MgO and (Pu,Zr)N [58]. Two pins of U-free fuels: and, irradiated in the BOR60 fast reactor up to about 11.3 at% will be examined very soon.

2.2.2. IMF for reactors of Generation IV

To allow nuclear energy to be part of a sustainable energy development, future nuclear energy systems (reactors, fuel and cycle processes) must provide advances in the following directions:

- an enhanced economic competitiveness (particularly with the reduction of investment costs),
- a still enhanced safety assessment,
- the use of proliferation resistant technologies and processes
- a reduced impact on environment,
 - with flexible and efficient use of nuclear fuels and available natural resources,
 - with significant reduction of long life radioactive elements in the final waste,
- an ability to fulfill other missions than electricity generation (such as hydrogen production and water desalination).

CEA has reached the conclusion that gas cooled reactors (GCR) present attractive features likely to bring the improvements searched for [106].

For the short term, thermal neutron GCRs could combine the reference options of HTRs to recent advances on materials and turbine technology: the high coolant outlet temperature (He at 850°C) makes it possible to deliver electricity, hydrogen and process heat at a high conversion ratio and a direct cycle helium turbine promises compactness and reduction of the investment costs.

In the longer term, enhanced sustainability (efficient use of resources, waste minimization and proliferation resistance) needs to consider both neutron spectrum hardening and actinides integral recycling, motivating the development of an advanced GFR.

Innovations are especially proposed to reach the following objectives:

- to define a conceptual core design with a hard neutron spectrum and Pu balancing requirements (fast neutron fluence, fertile fuel). More specifically, innovative fuel forms are needed to preserve the most desirable properties of standard fuel particles withstanding up to 1600°C and with an excellent confinement of the fission products, while accommodating the increased heavy nuclei content and withstanding the greater fast neutron fluence;
- to cope with the decay heat removal safety function (i.e. in accidental conditions);
- to have a compact and simplified spent fuel treatment that allows quantitative recovery and recycling of all the actinides.

The use of fuel materials with high temperature margins and high capacity for fission product confinement in the fuel together with the use of engineered safety systems, mainly based on passive features, constitute a comprehensive set of safety provisions leading to a consistent implementation of the defense-in-depth principle and hence to a high degree of public acceptance (no off-site effects in case of severe accidents, and enhanced resistance to severe core damage). In order to combine fuel cycle objectives and safety objectives, a specific core power in the range of 50–100 MW/m³ has been selected for current design studies.

Fuel concepts for GFR

The key points are important constraints for the fuel element [107]: fission products retention up to 1600°C with a fast neutron fluence of $\sim 1 \times 10^{27} \text{ cm}^{-2}$ ($E > 0.1 \text{ MeV}$), good thermal conductivity and high heavy atom content including minor actinides. The temporary objective is a “reasonable” burnup of 5–8 at% (10–15% expected in a second step). Beside particle or pin fuel concepts, candidates are cermet fuels (50 to 70 % of actinide ceramic and 50 to 30% of inert ceramic) or cermet fuels (actinide ceramic and inert metal) in block or plate type geometries. For cermet fuels, the needs to have a high density of actinides leads to consider mainly carbides or nitride; for the inert matrix, ZrC and TiC are an alternative to SiC for carbide fuels and TiN is taken into account for nitride fuel as well as ZrN. Performances in reactor, safety and ability to incorporate minor actinides will be the primary selection criteria.

The process for fuel fabrication will be selected taking into account the above requirements, the fuel composition (inert matrix, coating) and geometry and also applicability for ‘an integrated cycle’. This implies that reprocessing and re-fabrication will be carried out in the same unit, using highly compatible technologies.

Fuel cycle aspects (fabrication, reprocessing)

Reprocessing of the fuel (which is roughly cooled for one year) with an aim to quantitative recover of all actinides without intra-group separation are being considered utilizing both hydro-metallurgical and pyrochemical processes.

The non-separation of the actinides, the absence of blankets, the low content of plutonium and its isotopic composition, the presence of minor actinides (Am, Cm) will provide good intrinsic proliferation resistance characteristics. The reprocessing with low losses (0.1% of heavy nuclei) reduces the waste inventories for storage, activities (short, medium and long term) and radio toxicity.

The resources utilization is also very good due to the internal breeding with no need for blanket. The fuel can come from readily available depleted uranium stocks and does not call for mining. The flexibility of the concept, which can be a breeder or a burner, allows a good adaptation in a wide variety of scenarios, in a stand-alone mode, or in synergistic interaction with other reactors (PWR, BWR or HTR-LEU fuel).

Perspectives

The prior objective is the selection of reference and back-up fuel concepts, as well as the preparation of a consistent out-of-pile and in-pile test programme. The potential advantages of hydrometallurgical and pyrochemical processes for the different types of fuels considered needs to be assessed with a view to selecting the most appropriate concepts. After process screening, mostly with surrogate material, more in-depth studies in hot laboratories will be performed, using irradiated materials provided by the fuel irradiation test programme.

2.3. Japan

As the result of a 1994 task force, the Plutonium Rock-like Fuel Integrated R&D Team (PROFIT), was established by JAERI to develop innovative fuels, plutonium rock-like oxide fuels, and their once-through burning in light water reactors (the ROX-LWR system), which enables LWRs to utilize and annihilate plutonium effectively [1]. The ROX-LWR system has the potential of burning plutonium almost completely and disposing spent fuels directly

without further processing after cooling for about 50 years. The ROX-LWR system is basically built on conventional nuclear technologies and facilities. Therefore the new system is characterized as not only safe but also strongly proliferation resistant and with a potential to reduce global environmental burden.

The R&D activities on the ROX-LWR system and some basic research on inert matrices are described below. The former activities have been conducted at JAERI in collaboration with the Universities of Nagoya, Osaka and Ibaraki, and the latter independently at Kyushu University as well as with industry (Nuclear Fuel Industries, Ltd (NFI)) and Japan Nuclear Cycle Development Institute (JNC).

2.3.1. Studies on the ROX-LWR system

ROX fuel concept and developments

ROX fuel is a type of inert matrix fuel based on mineral-like compounds such as yttria stabilized zirconia (YSZ), magnesium spinel, and corundum, which are very stable chemically and physically. Plutonium is incorporated in the YSZ phase forming a solid solution. This treatment is of importance from the non-proliferation point of view. Moreover, plutonium in spent ROX fuels is degraded in isotopic composition and will be useless for weapons use.

YSZ is one of the promising candidates for the inert matrix, because of its superior physico-chemical properties such as high melting point, high stability against irradiation damage, and durability in acid and water. In addition, it has the capability to accommodate fissile plutonium and some fission product elements stably in its lattice. However, the thermal conductivity of YSZ is quite low ($\sim 2 \text{ W}\cdot\text{m}^{-1} \text{ K}^{-1}$), which could lead to high fuel temperature and high fission gas release even under normal operating condition.

Mixing YSZ with a second inert matrix material, which has much higher thermal conductivity than that of YSZ, is a possible way to improve the bulk thermal conductivity. Spinel and corundum are possible candidates. These materials also are very stable physically and chemically, which is required for a long term storage of spent fuels. Thermal conductivity of such YSZ/spinel composites was measured and found to be slightly higher than that of UO_2 . Relevant thermo-physical properties of ROX fuel candidates were measured in collaboration with NFI [108].

Although spinel shows ample stability against neutron irradiation, it seems to be not very stable against high-energy fission fragments [109, 112]. It is thus preferable to localize fission fragment damage to a small region to avoid swelling and degradation of thermal conductivity. A new particle dispersed ROX fuel concept has been developed, where macro YSZ particles containing fissile materials are homogeneously dispersed in a spinel matrix. In this configuration the damaged volume will be limited to a thin layer ($<10 \mu\text{m}$) of the matrix near the YSZ particle surface [109].

Fuel fabrication

JAERI has developed methods to fabricate ROX fuels using 20% enriched uranium (U-ROX) in cooperation with NFI [110]. Particle dispersed U-ROX fuel pellets have been successfully fabricated for irradiation tests. About $250 \mu\text{m}$ YSZ particles containing enriched uranium were dispersed in a spinel matrix. Well-defined YSZ particles were prepared using the sol-gel technique. Mechanical blended U-ROX fuel pellets, which were prepared by mixing YSZ and spinel powders, and YSZ single-phase fuel pellets were also fabricated for irradiation testing

by the conventional powder route. Apparent densities of these pellets were more than 90 % of theoretical.

Plutonium ROX (Pu-ROX) fuels were also fabricated in collaboration with the Nuclear Research and Consultancy Group (NRG, the Netherlands) and Paul Scherrer Institute (PSI, Switzerland) for the joint irradiation experiment ‘once through then out’ (OTTO) [111]. PSI fabricated YSZ particles containing plutonium by a sol-gel method and produced macro-spheres of YSZ solid solution fuels. JAERI provided technical information on the particle-dispersed fuel pellet fabrication and supported pellet fabrication at NRG-Petten.

In-pile irradiation test

Two irradiation tests of ROX fuels were carried out in the Japan Research Reactor No. 3 (JRR-3) in JAERI at a nominal reactor power of 20 MW for 100 days. In the first test small disk-shape fuel targets containing plutonium as the fissile material were irradiated to show the feasibility of the ROX fuel concept [112]. In the second test pellet-type fuels, where 20% enriched uranium was used as fissile material, were employed to obtain data on the basic irradiation behaviour such as swelling, fission gas release, phase stability and etc. [113].

From the irradiation tests of U-ROX fuels [114], the particle dispersed fuels showed a good swelling behaviour as expected from their design. However, they showed higher fractional gas release (FGR) than those of powder mixture fuels. Micro-structure studies have revealed the formation of significant crack formation, which are due to the mismatch in thermal expansion between YSZ particles and the spinel matrix. Further improvements must be made in the area of fuel fabrication technologies, especially sintering conditions. Optimization of sintered pellet density and the gap width between YSZ particles and the spinel matrix will be the key to improve the fuel performance.

Due to very high irradiation temperatures, spinel decomposition and subsequent restructuring in the spinel matrix fuels was observed. This restructuring was considered to cause the high “fission gas release” (FGR) of spinel matrix fuels compared with the corundum matrix fuels. However, it may be possible to reduce the FGR of the spinel matrix fuels to that of the corundum ones, if the maximum fuel temperature is limited below 1700 K where neither spinel decomposition nor restructuring was observed.

ROX fuel showed a good capability to retain fission products (FPs) [115]. The main solid FPs, Nd and Ba, and metallic FP inclusions consisting of Ru and Mo were observed in the YSZ fissile phase. A fraction of Cs and Xe, which are volatile FPs, migrated to the periphery of YSZ particles and to pores in the YSZ phase. A thin reaction layer (~5µm) was found between the YSZ particle and spinel matrix. The formation of this layer could be a direct consequence of reaction between YSZ and the fission damaged spinel matrix, and gives some evidence for the limitation of the damaged area in the spinel matrix to near the surface of the YSZ particles.

The joint OTTO irradiation experiments were performed at the high flux reactor (HFR, NRG-Petten, Netherlands) to demonstrate the irradiation stability of ROX fuels [116]. The Pu-ROX fuels were irradiated successfully for 22 cycles (548 days) with Pu depletion of 50–60% under current LWR conditions. Post irradiation examinations on these Pu-ROX fuels will supply significant information of their irradiation behaviour.

ROX core neutronic studies

Containing no fertile materials², the ROX fueled core has the inherent disadvantage of a small Doppler reactivity coefficient and a large burnup reactivity swing. The small Doppler coefficient may cause severe fuel damage under a reactivity initiated accident (RIA) condition. The large burnup reactivity swing will cause a large power peaking, which should be avoided for reasons including a loss-of-coolant accident (LOCA) safety. Improvements were performed by two approaches to overcome the above disadvantages; addition of resonant nuclides such as ²³⁸U, ²³²Th and Er to the ROX fuel [117], and composing a heterogeneous core of ROX and UO₂ [118].

Analytical results of Doppler coefficient and power peaking factor of a ROX fueled PWR core evaluated with the SRAC95 code system [119] are listed in Table VI. With addition of U and a small amount (~0.6 at%) of Er into ROX fuel, the Doppler coefficient and the peaking factor for ROX+15at%U(Er) and ROX+8at%U(Er) cores could be successfully improved to the level of a UO₂ core.

As for the partial ROX loading core (1/3ROX+2/3UO₂), satisfactory results were also obtained by adjusting the Pu and Er content as well as its distribution within ROX assemblies near the interface of ROX and UO₂ assemblies. This core configuration is very attractive and important because the Pu transmutation rate is high (99% ²³⁹Pu for W-Pu and 98% ²³⁹Pu for R-Pu) in the ROX assembly, and this is quite representative of the initial stages foreseen for ROX deployment in conventional power plants.

Accident analyses

By using the above-improved core characteristics, RIA and LOCA behaviours of ROX fuelled PWR were analyzed with the EUREKA-2 [120] and RETRAN-2 [121] codes, respectively. The results are also summarized in Table VI. Here, fuel enthalpies (*H*) are given in volumetric enthalpy, because of large difference in the density between the ROX and UO₂ fuels.

TABLE VI. IMPROVED CORE CHARACTERISTICS OF ROX FUELLED PWR CORES AT BOC, MAXIMUM FUEL ENTHALPY (*H*), FUEL TEMPERATURE (*T_F*) AND PEAK CLADDING TEMPERATURE (*T_{PC}*) IN RIA AND LOCA EVENTS

Core	Doppler reactivity (900K→1200K) $\Delta k/k$ (%)	Peaking factor	Fuel enthalpy (GJ m ⁻³)	<i>T_F</i> (K)	<i>T_{PC}</i> (K)
Weapons-grade Pu					
Original ROX	-0.098	2.7	—	>2,200	>1,470
ROX+15at%U(Er)	-0.61	2.1	4.5	1,700	1,090
1/3ROX+2/3UO ₂	-0.53	1.7	4.2	1,320	1,080
Reactor-grade Pu					
ROX+8at%U(Er)	-0.59	2.2	4.5	1,700	1,090
1/3ROX+2/3UO ₂	-0.48	1.8	4.4	1,380	1,070
UO ₂	-0.75	2.0	4.3	2,080	1,080

² Another name used for the fuel temperature coefficient of reactivity and it is defined as the change in reactivity per degree in the fuel temperature.

In the reactivity initiated accidents (RIA) analyses, fuel enthalpy and fuel temperature (T_F) of the original ROX fuel core were far larger than the UO_2 pin failure condition in RIA of 9.6 GJ m^{-3} and the ROX fuel melting temperature of 2200 K. However, all the improved ROX fueled cores address these limitations [122]. Among them, the partial ROX fuel-loading core shows even better performance than the UO_2 core due to the refinement of fuel compositions. In the LOCA analyses, the higher peak cladding temperature (T_{PC}) than the limiting value of 1473 K in the original ROX PWR is well reduced in the improved ROX fueled cores to be as low as that of the UO_2 core.

Pulse irradiation tests under RIA conditions

Along with the above calculational analyses, pulse irradiation experiments were performed on the ROX fuels in the Nuclear Safety Research Reactor (NSRR) in JAERI to investigate the fuel behaviour under RIA conditions [123, 124]. Three ROX fuels, a mechanically blended YSZ/spinel fuel (SH), a YSZ single phase fuel (Z) and a particle dispersed YSZ/spinel fuel (SD), were fabricated into short test rods with a 17×17 PWR type specification and were irradiated with a short pulse.

Peak fuel enthalpy and rod failure are summarized in Fig. 4. The horizontal dotted line of 9.6 GJ m^{-3} indicates the current rod failure threshold for the UO_2 fuel. Open marks show the intact fuel rods, while filled marks show the fuel rods failed. The rod failure threshold for the ROX fuel seems to be about 10 GJ m^{-3} and is comparable to that of the un-irradiated UO_2 fuel.

Fuel failure behaviour for the ROX fuel was, however, quite different from that of the UO_2 fuel. ROX fuels failed with fuel pellet melting and a considerable amount of the molten fuel was released to the surrounding coolant water, while UO_2 fuels remain solid at the failure enthalpy. In spite of the release, no significant mechanical energy generation due to fuel-coolant interaction (FCI) was observed in the ROX fuel test at an enthalpy range below 12 GJ m^{-3} . The absence of the FCI was likely due to the moderate release of the molten ROX fuel. These results in core neutronic studies clearly show that the ROX-LWR system is a feasible option for effective burning of excess plutonium.

Geological stability of spent ROX fuels

Direct disposal of spent ROX fuel is one of the most significant features of the ROX-LWR concept. Spent ROX fuels are planned to be disposed of in a repository located in a deep geological formation after 50 years cooling. A preliminary analysis of environmental safety was carried out for the direct disposal of ROX fuels [125]. The individual dose equivalent from actinide decay chains due to the ingesting of contaminated drinking water was evaluated based on the groundwater migration scenario. Analysis results showed that the dose equivalents from spent ROX fuels were about 2 orders of magnitude smaller than those from spent MOX fuels and the reduction of actinide nuclides in the spent ROX fuels would be beneficial to repository safety.

In order to confirm the environmental safety analysis, leaching tests have been performed for irradiated U-ROX fuels with reference to the MCC-1 method [126]. Focus was placed on the determination of not only leaching rates of long lived actinides and β -emitters but also speciation of leached elements, i.e. ions, precipitates and colloids. The latter is extremely important for evaluation of migration behaviour.

2.3.2. Basic studies

Doppler reactivity measurements at FCA

Some resonant additives have been considered to improve the small Doppler reactivity of ROX fueled core. To validate data/method for core physics calculation, Doppler effect experiments on resonant candidates such as erbium, tungsten, thorium and uranium were carried out in fast and intermediate neutron spectra using the Fast Critical Assembly (FCA) at JAERI [127, 128]. Measured data showed that the ‘Doppler reactivities’ of Er, W and ThO₂ were almost comparable to natural U in a fast neutron spectrum and were 15–25% higher in an intermediate spectrum, suggesting that these are good candidate additives for ROX fuels. Calculations using the SRAC code system with the nuclear data library JENDL-3.2 showed some discrepancies with measured Doppler effects for Er and W. Though the discrepancies were not serious, some improvements will be necessary for analytical methods and codes and possibly for the nuclear data library.

Materials research

Many efforts have been made to construct a database for basic properties of inert matrices. Under the joint research programme between JAERI and Nagoya University, cubic zirconium oxide compounds with Pu and Np oxides, and perovskite CaTiO₃ doped with Pu oxide were investigated: phase relationships and thermal expansion of YSZ solid solutions doped with Pu or Np oxide [129], thermal expansion of Pu doped lanthanum zirconate pyrochlore [130], and thermal expansion and heat capacities [131] of Pu doped CaTiO₃ have been investigated. Mössbauer spectroscopy showed that a part of the Np existed as pentavalent Np⁵⁺ in YSZ solid solutions. The finding may be extremely important for the safety assessment of spent ROX fuels after geological disposal. JAERI, in collaboration with Osaka University, investigated and revised the zirconium rich part of the ternary Zr-Pu-O phase diagram [132]. Oxygen potential measurements and phase relations were carried out for oxygen-rich pyrochlore Zr₂Pu₂O_{7+x} [133] which was a possible substitute for YSZ.

Perovskite compounds, BaUO₃, ZrUO₃ and BaMoO₃, were studied by Osaka University in collaboration with Kyoto University, JNC or NFI; Phase equilibria among these compounds [134], heat capacity [135] and thermo-chemical/physical properties such as melting point, thermal expansion, elastic moduli and Debye temperature [136] were measured.

A cermet fuel concept is a possible alternative to improve the low thermal conductivity of YSZ. With the use of metallic inert matrices, fuel centerline temperature could be kept below 1273 K. Reaction of YSZ with Zr and Si metals and Zircaloy-4 were investigated at Kyushu University in a compatibility study of cermet fuels [137]. Practically no interactions were observed below 1273 K. YSZ micro-spheres were fabricated by use of internal gelation process in collaboration with PSI [138]. A cermet sphere-pac fuel concept has been developed at JNC in collaboration with PSI as a new dust-free fuel fabrication technology. A calculation code for the thermal conductivity of cermet sphere-pac fuel pellets [139] was developed and some experimental studies on oxidation and sintering behaviour of these pellets were carried out [140] to assess the calculation model.

Radiation damage by ion irradiation

Ion irradiation coupled with in-situ observation by scanning electron microscopy (SEM) and/or transmission electron microscopy (TEM) is a powerful tool to investigate radiation damage in inert matrices. JAERI performed these studies in collaboration with Nagoya

University [141] and Ibaraki University [142] and clarified gas bubble formation behaviour, amorphization of spinel and alumina, and the annealing process of damaged matrices. This work provided useful information to understand in-pile irradiation behaviour of ROX fuels. Kyushu University found a unique radiation effect in YSZ, the anomalous formation of large defect clusters ($\sim 1 \mu\text{m}$) under electron irradiation subsequent to ion irradiation [143]. Such defect clusters were not formed solely with ion irradiation and may play an important role in property changes of YSZ under in-pile irradiation.

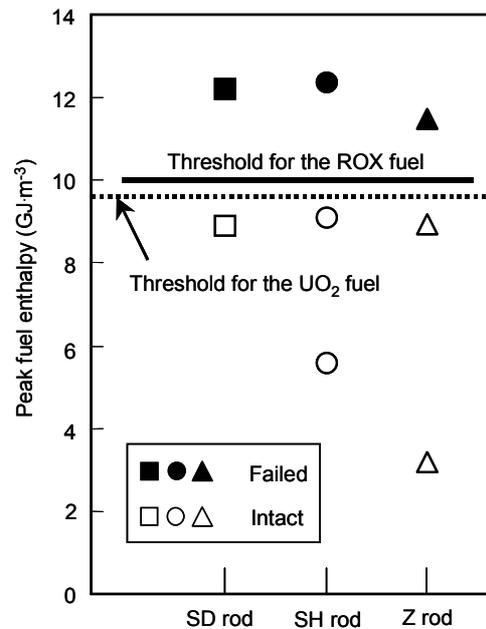


Fig. 5. ROX fuel pin failure threshold enthalpy in terms of $\text{GJ}\cdot\text{m}^{-3}$. SD: Spinel-YSZ Dispersed, SH: Spinel-YSZ Homogeneous, Z: YSZ.

2.4. Russian Federation

There are two areas of activities in the Russian Federation related to the IMF development with clear defined objectives:

- Conceptual studies on application of various IMFs for Pu and MA burning. The scope of these activities is limited to laboratory scale fabrication technology development and theoretical studies on using IMF in fast LMR and LWR. Some in-pile irradiation tests of IMF are being carried on in the frame of international cooperation.
- Cermet fuel dedicated for power reactors operating in a load-following mode. Long term operating experience with UMo-Mg and UO_2 -Mg cermet fuel has been accumulated since early 50s. This scientific and technological background is applied now in the development of cermet fuels for Russian commercial LWR (WWERs).

The IMF activities are carried out at IPPE, Obninsk. Some IMF activities are also performed at VNMIINM Moscow, and at SRIAC, Dimitrovgrad.

2.4.1. IMF fuel development and testing

Several cerium inert matrix materials were studied [144–146]: MgO, Fe-MgO, BaO, ThO₂-MgO, ThO₂, (Y,Zr)O_{2-x}, MgAl₂O₄. Short descriptions of the results from these studies are presented.

Fabrication and study of PuO₂-MgO fuel

The PuO₂-MgO fuel compound is proposed for plutonium utilization in fast-neutron sodium-cooled reactors [145]. Advantages of MgO as compared with other inert diluents are as follows: absence of intermediate phase in plutonium dioxide systems, enhanced radiation resistance, satisfactory compatibility with liquid sodium coolant, satisfactory thermal conductivity, and absence of major problems of the irradiated fuel reprocessing.

UO₂-MgO fuel pins with composition of 36% wt UO₂+64% wt MgO to simulate PuO₂-MgO fuel were manufactured in 1997. In-core testing of the fuel pins was performed in the research fast reactor BR-10 (SSC, IPPE, RF). The irradiation occurred in a BR-10 reactor cell under conditions close to the fast reactor:

- Fast neutron flux density – $8.4 \times 10^{14} \text{ cm}^{-2} \text{ s}^{-1}$;
- Fast neutron fluence – $9.76 \times 10^{21} \text{ cm}^{-2}$;
- Maximum burnup level – 1.24% (heavy atoms);
- Maximum cladding temperature – 700°C;
- Average (sodium) coolant temperature at fuel subassembly (FSA) inlet – 320°C; and
- Maximum calculated temperature at the fuel centerline – 1200°C.

Test runs demonstrated satisfactory results.

Fuel pins with 40% wt PuO_{2-x}-60 %wt MgO were fabricated in 1999. PuO_{2-x}-MgO fuel pellet fabrication process consists of chemical coprecipitation of the plutonium and magnesium compounds from solution, pelletization and pellet sintering [144–146].

In-pile test of PuO₂-MgO-fueled pins was carried out in the BOR-60 reactor (Scientific Research Institute of Atomic Reactors, RIAR, Dimitrovgrad, Russia). Fuel irradiation conditions were as follows:

- Maximum linear power rating ~13 kW m⁻¹.
- Maximum allowed fuel pin cladding temperature – 700°C.
- Average coolant temperature at FSA inlet – 330°C.
- Maximum calculated fuel centerline temperature – 1000°C.
- Irradiation time – 750 efficient days.

A part of the specimens, irradiated up to a burnup of 13% (heavy atoms), has been discharged from the reactor core and is under examination.

The study was carried out to develop a fuel composition with improved neutronics features due to neutron-absorbing additives, which enhance the efficiency of plutonium burning in the fast reactors. Absorber options (e.g. niobium, molybdenum, tungsten, iron or their oxides) were investigated.

The iron as the absorber option was proved to be most appropriate from the point of the view of manufacturing the fuel by coprecipitation.

Fabrication and study of ROX-fuel

Studies on fabrication technology and investigation of ROX-fuel aiming at plutonium utilization were carried out at the SSC and IPPE, Russian Federation. In the current work, ROX-fuel is particulates of (Pu,Zr)O₂ and (Pu,Th)O₂, which are uniformly distributed within a spinel (MgAl₂O₄) matrix [146].

Fuel fabrication processes have been developed for fuel compositions (Pu,Zr)O₂-MgAl₂O₄ and (Pu,Th)O₂+MgAl₂O₄. Granulated fuel particles were prepared by means of precipitating the components from their nitrate solutions followed by stages of precipitate calcination, compact pressing, compact milling and screening. The spinel is prepared by components coprecipitation as well. Granulated fuel in pre-sintered or non-sintered (green) state was blended with spinel powder, pressed, and sintered. Gd₂O₃-doped pellets (i.e. with an additive of the burnable poison) were fabricated as well. Characteristics of fabricated ROX-fuel samples are shown in the Table VII.

TABLE VII. ROX FUEL PELLETT CHARACTERISTICS

Fuel composition (wt%)	Density, % TD	Phase composition	Lattice parameter, (Å)
9.5% PuO ₂ +60.5% ZrO ₂ ·Y ₂ O ₃ + 30% MgAl ₂ O ₄	94.3	Solid solution, PuO ₂ ·ZrO ₂ ·Y ₂ O ₃ ; Spinel MgAl ₂ O ₄	5.151±0.001 8.072±0.002
9.5% PuO ₂ +60.5% ZrO ₂ ·Y ₂ O ₃ + 30% MgAl ₂ O ₄	99.07	—	—
19% PuO ₂ +37.8% ZrO ₂ ·Y ₂ O ₃ + 43.2% MgAl ₂ O ₄	90	—	—
18.1% PuO ₂ +36% ZrO ₂ ·Y ₂ O ₃ + 41.1% MgAl ₂ O ₄ + 4.8% Gd ₂ O ₃	92.2	Solid solution PuO ₂ ·ZrO ₂ ·Y ₂ O ₃ ; Spinel MgAl ₂ O ₄ ; Gd ₂ O ₃ -base solid solution	5.207±0.003 8.079±0.002 11.086±0.005
18.8% PuO ₂ +37.4% ZrO ₂ ·Y ₂ O ₃ + 42.8% MgAl ₂ O ₄ + 1 %Gd ₂ O ₃	95	—	—
8.8% PuO ₂ +61.2% ThO ₂ + 30% MgAl ₂ O ₄	90.9	—	—
13.6% PuO ₂ +47.9% ThO ₂ + 3.7% Gd ₂ O ₃ +34.8% MgAl ₂ O ₄	93.7	Solid solution PuO ₂ ·ThO ₂ ·Gd ₂ O ₃ PuAlO ₃ (traces) MgAl ₂ O ₄	5.550±0.003 8.080±0.002

Ingestion radiotoxicity hazard index of inert matrix spent fuels have been calculated for different cases-studies involving viz., after burning minor actinide (MA) isotopes in LWRs and compared with the hazard index of MOX and MA burning MOX (MOX+MA) spent fuels [147]. As U-free fuels, ROX: (PuO₂+ZrO₂) and TOX: (PuO₂+ThO₂) are considered, in which MA's are added as oxides. The radiotoxicity hazard index of ROX+MA spent fuel is less than that of TOX+MA and MOX+MA spent fuels due to the lower density of actinides in spent fuel. Some of cooling years the toxic yield of ROX+MA spent fuel is even less than that of MOX spent fuel, if the initial loaded MA in ROX is about 0.5 at %.

Cermet fuel development

Cermet fuel is one of the inert matrix fuel options in which the particles of UO_2 are surrounded by a metal matrix with suitable physical and thermophysical properties [148, 149].

The structure of the fuel pin is as follows: the homogeneous fuel matrix containing uranium dioxide granules surrounded by a metal matrix (silumin) has rigid cohesion with the Zr-1%Nb alloy cladding. Metallurgical bond between fuel and cladding provides practically zero-value thermal resistance at the contact boundary. The superior thermal-conductivity of the metal matrix ensures a heat conductivity of the cermet fuel composition, which is several times higher as compared to that of uranium dioxide pellets. Very high values of thermal conductivity as high as 30–40 $\text{W/m}\cdot^\circ\text{C}$ is achieved in cermet fuel when the content of uranium dioxide granules does not exceed 60%. The total uranium load for this cermet fuel is ~29% lower as compared to conventional UO_2 fuel. Assuming a 5% enrichment level, the refueling interval for WWER-1000 is about 255 effective days.

Fuel pins of the cermet fuel have a maximum centerline temperature: $T_{fuel} = 442^\circ\text{C}$ in a WWER-440; $T_{fuel} = 508^\circ\text{C}$ in a WWER-1000.

In-pile testing of cermet fuel pins was carried out in the MIR reactor (Dimitrovgrad, Russia). The testing proved that fuel pins operated up to maximum burnup $BU \approx 60 \text{ MW}\cdot\text{d/kg U}$ without loss of their integrity and demonstrated successful behaviour during reactor power changes within the operation run.

Cermet fuel unsealed pins were tested in the AM research reactor (IPPE, Obninsk). Fission product release into the reactor coolant during 2 months testing was studied and found very low.

Experimental data on the threshold value of fuel destruction enthalpy under a reactivity-initiated accident (RIA) were measured at the IGR reactor (Kazakhstan). Examination of damaged cermet fuel pins allowed to state that performance of the cermet fuel under RIA conditions was at least equal to that of the conventional fuel pins of the WWER reactors.

2.4.2. Studies on the IMF application

Application of the IMF for actinide burning in fast reactors

The use of IMFs in fast reactors for incineration of long life radioactive waste (plutonium, minor actinides MAs, and fission products) was studied [150].

Implementing the inert matrix fuel approach would allow to achieve optimum efficiency of plutonium utilization (i.e. disposition). The reactor core of the BN-800 type reactor could provide disposal up to 800 kg/yr of plutonium and from 100 to 300 kg/yr, of MA depending on core design.

Two approaches for utilizing the minor actinides are under consideration:

- Use of PuO_2 or $^{235}\text{UO}_2$ inert matrix fuel with homogeneous additives of large amounts of the minor actinides.
- Use of specially developed minor-actinide fuel subassemblies, which could be placed both in a fast reactor core as well as blanket.

ROX-fuel application for plutonium burning in thermal and fast-neutron reactors

The problem of burning the plutonium in a medium-sized light-water power reactor viz. WWER-640 type reactor was examined as a case study [151, 152]. The fuel is made of granules of plutonium dioxide dispersed within a spinel matrix (MgAl_2O_4). The option with thoria-doped matrix was analyzed as well.

Reactor characteristics were examined for three fuel loading variants as follows:

Variant-1 – complete core loading with ROX fuel.

Variant-2 – complete core loading with thoria-doped ROX fuel.

Variant-3 – combined core loading with 163 FSA when 44% of them contain ROX-fuel; the other FSAs have conventional uranium fuel (UO_2) with 4.0% enrichment.

The outcome of this study indicated that the WWER-640 does not face any principal unsolvable difficulties with the use of ROX fuel. However, an economic aspect of the issue is not considered yet.

The values of water and fuel temperature reactivity coefficients for all the three variants considered were found to be negative for entire irradiation cycle. This is a principal requirement for reactor safety. A lower value of the fuel temperature reactivity coefficient is observed for the reactor core completely loaded with ROX fuel.

Consequences of the RIA and other accident scenarios with main circulation tubing rupture have been reviewed [151]. No critical safety problems that warrant WWER-640 design modifications have been revealed.

The maximum plutonium utilization rate of 720 kg/year was achieved under the variant-1 condition (i.e. reactor core completely loaded by ROX fuel with a matrix). About 88% of the plutonium loaded was burned in the scenario.

The application of the cermet fuel concept in the WWER reactors

Cermet fuel based on particles of UO_2 in a silumin inert matrix proved to be promising for use in the WWER as this fuel has the characteristic of a low cladding stress in a cyclic reactor power change during operation. Current efforts to increase the nuclear contribution to the country electricity production make this fuel attractive even in the short term application.

Besides UO_2 cermet fuel, (U,Th) O_2 cermet fuel is suitable for WWER. Uranium enrichment does not exceed 20% in this case. Conventional UO_2 fuel could be step by step replaced by mixed U-Th cermet fuel for one of the operating WWERs.

The use of U-Th cermet fuel for WWER-1000 can be characterized by the following advantages:

- lower (three times as low) build up of plutonium and of several minor actinides;
- reduced (by 23%) poisoning of the reactor by ^{135}Xe that will contribute to the prevention of cyclic power fluctuations;
- improvement of the reactor plant safety at the expense of reducing the critical concentration of boron in the coolant in the beginning of the refueling interval from 1.07 to 0.85 $\text{g}\cdot\text{kg}^{-1}$ H_2O .

Measurement of Doppler effect of ^{239}Pu and ^{240}Pu

The Doppler effect value for plutonium isotopes is an important safety characteristic of uranium-free ROX-fuel. The experimental study of the Doppler-effect was carried out at the BFS critical assembly (IPPE, Obninsk) in the course of heat-up tests of several plutonium specimens with different composition [153].

The measurements were carried out both for a core with a fast (BFS-91-1) and a softened (BFS-91-2) neutron spectrum. Softening the spectrum occurred by means of polyethylene displacers.

PuO_2 specimens are loaded into steel-cased cylinders with diameter from 5 to 13 mm and length about 100 mm. The plutonium isotope composition is given in Table VIII.

TABLE VIII. PLUTONIUM ISOTOPE COMPOSITION (IN %) OF THE PuO_2 SPECIMENS USED IN DOPPLER EFFECT MEASUREMENT

Reference composition	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{241}Am
«intermediate»	0.23	77.86	16.76	2.26	1.02	1.87
«dirty»	0.59	67.59	21.32	4.29	2.67	3.54
«Pu-240»	0.00	9.13	89.22	0.99	0.16	0.50

PuO_2 specimens are located into an electrical heater and are heated up to 600°C. Emplacement of cold and heated plutonium-bearing specimens allows to estimating the influence of temperature on assembly reactivity change.

The main results of the above experimental study are shown in the Table IX.

TABLE IX. CHANGE OF REACTIVITY WITH TEMPERATURE INCREASE FROM 20 TO 600°C, CENTS

Specimen isotopic composition	Specimen mass (g)	BFS core name	
		BFS-91-1	BFS-91-2
«intermediate»	28.8	-0.011±0.003	-0.080±0.002
«dirty»	17.7	—	-0.092±0.002
	20.2	-0.015±0.002	—
«Pu-240»	11.4	—	-0.081±0.004
	46.4	-0.032±0.003	—
UO_2 ($x=0.4\%$)	305	-0.101±0.006	-0.353±0.005

The experiments showed that the Doppler-effect for plutonium of various isotope compositions is small enough but negative. Available computer codes and nuclear data allow to predict the Doppler-effect value with acceptable accuracy.

Conclusion

Earlier experience with IMF in Russia demonstrates advantages of this type of fuel for the improvement of performance and safety of various power reactors. Recent efforts are aimed at

meeting new requirements for nuclear power development, such as for sustainability, proliferation resistance, and for long-lived waste management. It was shown that IMF could contribute to the solution of these problems and interest in the development of the IMF exists in several research institutes. On the one hand, it is clear that further intensive and expansive R&D are required for practical implementation of IMF in today's and future reactors. In this position, the decision has not been reached how to proceed with the IMF development activities and what is the prioritization of the activities are in the view of the current country nuclear energy strategy.

2.5. Switzerland

In Switzerland the R&D work on IMF started in the 90s and involves studies at the Paul Scherrer Institut, the Ecole Polytechnique Fédérale de Lausanne and the University of Geneva with their participations in the multidisciplinary programme on zirconia based inert matrix fuel [2], which includes several irradiations in research reactors, and, at the Paul Scherrer Institut and the Polytechnicum of Zurich, with their participations in the Program CONFIRM, which concerns a zirconium nitride based inert matrix fuel [154]. In addition, since more than 10 years the Paul Scherrer Institut has played a leading role in the activities related to the promotion of the inert matrix fuel studies and PSI played a main role for the co-ordination workshops and activities in the framework of the 'Initiative for IMF'.

2.5.1. Zirconium nitride based IMFs

The CONFIRM programme foresees utilization of zirconium nitride based inert matrix fuels (Am, Pu, Zr)N for multi-recycling (with reprocessing). Its irradiation shall be performed in adequate transmutation systems with high neutron fluxes.

The main purpose of burning of americium and degraded plutonium is to reduce the radiotoxic waste inventories directed to reduce the geological repository space requirement by a factor of 100. The irradiation in an adequate transmutation system would allow reaching very high burn ups without degradation of safety margins. Nitride fuels are interesting in this context, as they are amenable to PUREX reprocessing method. The programme foresees fuel fabrication (Pu, Zr)N by the carbothermic reduction of oxides prepared by micro-spheres, using the sol-gel method, or in powder form followed by nitride preparation [155]. Irradiation is programmed with several fuel designs. Calculations establish that fuel performance for a range of porosity distributions is acceptable at $30 \text{ kW}\cdot\text{m}^{-1}$. These calculations suggest that the CONFIRM fuel is stable up to its melting point under adequate atmosphere.

From the ecological point of view the fuel material would require ^{15}N nitrides in order to avoid ^{14}C production during irradiation and its release during reprocessing. The CONFIRM irradiation is performed in the Studsvik reactor. After irradiation, a follow up project is planned including post irradiation analysis. Reprocessing of irradiated fuel samples will be performed using aqueous methods. To reach very high burn the possibility, nitride fuel fabrication in annular shape pellets (namely barrel with hole) using direct coagulation casting technique has also been attempted [156].

2.5.2. Zirconia based IMF

The programme dealing with zirconia based inert matrix fuel development involves the irradiation of a zirconia-plutonia solid solution in light water reactor in a "once through" mode (a last fuel cycle prior to geological disposal) [157].

Over the last 10 years, efforts have been carried out to derive an advanced zirconia inert matrix fuel [158, 159] from the early conceptual studies uranium-free fuel forms based on modeling [5, 61]. The studies on zirconia inert matrix fuel include:

- a suitable stabiliser to yield a better material resistance to damage under high irradiation environment and resistance with regards to the geological environment of the spent fuel disposal;
- the most favorable selected burnable poison nuclide(s) to optimise the reactivity during irradiation;
- optimum pellet design for safe utilization in light water reactor.

The evaluation of matrix materials was carried out for a range of elements in the frame of an initial selection. The first requirement in the material selection was guided by the neutronic properties. The elements and/or isotopes of the inert matrix were selected according to their transparency for neutrons. This was followed by simple neutronics cell calculations to characterize reactivity effects of:

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

In the preliminary neutronic studies, the physico-chemical properties of the material are not considered. These are, however, of prime importance for understanding the fuel behaviour during irradiation. The desired 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. As a result of screening studies, $\text{Y}_y\text{Zr}_{1-y}\text{O}_{2-y/2}$, (with $y \sim 0.1$) was selected for the above properties.

IMF pellet fabrication with optimised densities was carried out prior to characterization. The fabrication of the IMF powder can be performed following either a wet or a dry preparation route [160].

Zirconium oxide-based IMF has been prepared following the conventional pelletising route, in which an improved two-stage attrition mill was employed [9]. Different methods have been applied for the powder preparation. In addition to powder mixing, nitrate solutions have been co-precipitated to bulk gel and to micro-spheres by the internal gelation process. The calcined porous micro-spheres were crushed in the attrition mill. The powder was compared with three different mixtures of the four commercially available powders (ZrO_2 , Y_2O_3 and Er_2O_3 and PuO_2). Pellets of a standard size with relative densities higher than 90%TD and comparable grain and pore structures have been obtained. Based on these simulation tests [160], both processes are found suitable for fabricating IMF pellets, which fulfill requirements of commercial plutonium and uranium mixed oxide (MOX), where applicable. No intensive milling is required to form a solid solution in the sintering step for a $(\text{Er}, \text{Y}, \text{Pu}, \text{Zr})\text{O}_{2-x}$ pellet.

The “geometrical” density and the porosity were deduced from X ray analysis [161]. Optical and scanning electron microscopes were used to study pore or grain structure [162] and by

X ray absorption spectroscopy was used to characterize the atoms environments [163]. A good control of the sample dimensions during the fabrication could be guaranteed.

The IMF characterization was completed by irradiation studies using research institute facilities such as accelerators or research reactors. Accelerators were used to study microstructural changes during irradiation [158] and satisfactory behaviour was observed for all irradiations. Retention of cesium in yttria stabilized zirconia (YSZ) was studied experimentally and discussed on the basis of its solubility which is estimated based on chemical thermodynamic stabilities. The extent of retention of Cs and I in the YSZ matrix was investigated as a function of the temperature for cesium and iodine [165]. It is observed that in a sample implanted with both cesium and iodine, iodine becomes significantly mobile only above 1500 K approximately and cesium atoms stay immobile up to 1500 K.

Apriori studies for neutronic modelling at the pellets, pin and assembly levels were carried out for commercial reactors. The components: fissile (weapon or civilian plutonium from UOX or MOX reprocessing)/burnable poison and inert matrix components of the IMF are selected and their respective concentrations are optimised according to the neutronic characteristics [166].

PSI initiated a numerical benchmark exercise for comparing the results of different calculation methods/data sets used for the analysis of light water reactor (LWR) configurations employing Pu inert matrix fuels (IMFs). The first phase of the exercise was devoted to infinite arrays of identical IMF cells. The main feature investigated in the second phase was the influence of the neutron spectra in UO₂ and MOX cores on individual IMF cells. In the third phase: the IMF assembly was surrounded by UO₂ assemblies.

The Swiss candidate materials were first tested in research reactors. The first-of-their-kind integral measurements have been carried out at the PROTEUS facility employing a specially fabricated Pu-Er-Zr IMF rod in a heterogeneous (boiling water reactor) test lattice [167] (see Fig. 6). Analogous experiments have also been conducted with MOX and “dummy” IMF rods (without fissile material), providing the basis for a systematic comparison of experimental and calculation results. These reactor tests completed the former experiments carried out with burnable poisons (e.g. Er₂O₃) and inert matrix (ZrO₂) in the CROCUS reactor at the Swiss Federal Institute of Technology, Lausanne [168].

Stabilized zirconia ceramics were also tested in the frame of the ‘once-through-then-out’ irradiations in the high flux reactor, Petten, NL. In the OTTO plutonium incineration experiment that has been carried out by JAERI, PSI and NRG, the use of highly stable inert matrices (MgAl₂O₄ spinel and ZrO₂) [116] has been examined. These inert matrices were selected for this irradiation tests as the representative core-studies for the OTTO scenario. A total of 7 different kinds of Pu-containing targets were prepared for irradiation. The objective of the irradiation was to reach a very high Pu-burnup. The main parameters to be studied were irradiation stability, swelling, fission gas release and chemical interactions in the fuel. The fissile plutonium concentration was 0.32–0.44 g·cm⁻³. Both particle-dispersed fuel and homogeneous dispersions were fabricated in order to test the effect of the size of the fissile inclusions. The design of the experiment and the fabrication of the samples were discussed.

Recent results from the irradiation testing with a dedicated experiment in Halden, Norway, in the framework of the OECD-Halden heavy water reactor project [169] are described in the following Chapter on ‘results and outlook’. In this study emphasis is given the measurements of power distribution and centre-line temperature in the pin, the mechanical behaviour and the fission product release as well as to make also a comparative assessment with MOX fuel.

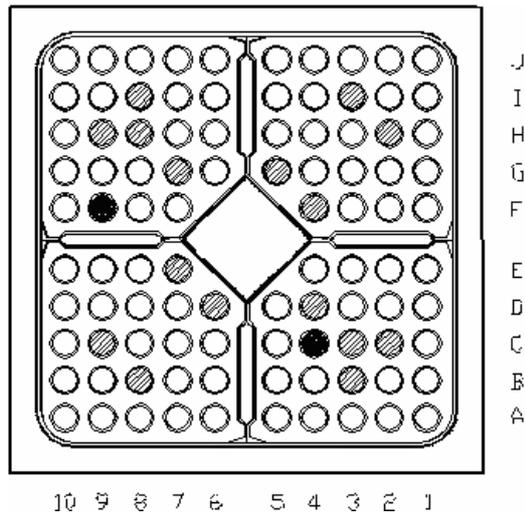


Fig. 6. Sectional view of a SVEA-96 BWR fuel assembly indicating the voided moderator conditions simulated in LWR-PROTEUS Core 3A, for details see [165] and the symbols in the figure represents O- Normal UO_2 fuel pins (^{235}U enrichments in the range 2–5%), ●- Fuel-pin positions employed for the IMF measurements and ◐ Burnable-poisoned pins.

The behaviour of IMF in LWR has also been studied analytically with emphasis on the researched goal.

The most efficient way to manage plutonium in light water reactors is to eliminate the production of plutonium generated in the UO_2 assemblies. Calculations were performed for “real-life” situations, which call for transition configurations in which mixed IMF and UO_2 assembly loadings must be considered. To determine the influence of the introduction of IMF assemblies on the characteristics of a UO_2 -fuelled core, three-dimensional full-core calculations have been performed for a present-day 1000 MW electric PWR containing up to 12 optimised IMF assemblies. The results of whole-core three-dimensional neutronics analyses indicated that the feasibility of an asymptotic equilibrium four-batch cycle fuelled solely with the proposed $\text{Er}_2\text{O}_3\text{-Y}_2\text{O}_3\text{-PuO}_2\text{-ZrO}_2$ inert matrix fuel for a present-day 1000 MW(e) PWR [166].

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.

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 types and their interaction with each other. 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 [160].

An important criterion, for the choice of materials for IMFs, which needs to be explicitly mentioned is that of the destination of the spent fuel. The spent zirconia 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). In the context of analysis of spent IMF behaviour in the geological repository, some basic R&D carried out and a short description is given below.

The solubility of zirconia in a typical radioactive waste repository environment was revisited at PSI. These solubility data were examined in the light of available thermodynamic data, with particular focus on the influence of carbonate complexes [170]. The solubility of monoclinic zirconia in a typical repository pore water was then calculated with the help of a speciation model. Although our calculations indicate that the solubility of zirconia in a bentonite-filled repository environment will probably be between 10^{-9} and 10^{-8} M, there are large unavoidable uncertainties in these predictions, due to limitations in the available thermodynamic data.

Recent experiments conducted at pH 9 and 298 K clearly indicate that addition of bicarbonate ions in aqueous solutions increases the solubility of both cubic and monoclinic zirconia by several orders of magnitude [171]. In pure water the solubility of zirconia proved to be very low (about 10^{-10} to 10^{-8} M), while in the presence of 0.05 M NaHCO_3 it increased to 10^{-6} – 10^{-5} M, indicating the formation of strong carbonate complexes. Up to a total carbonate concentration of 0.05 M, our data are consistent with the formation of a mononuclear tetracarbonate complex. PSI measurements indicated a Zr solubility in the order of 10^{-6} M for typical repository conditions, which is still 3 orders of magnitude below silica concentrations measured from the dissolution of nuclear waste glasses.

The mineral baddeleyite, ideally ZrO_2 , may be considered as a natural analogue of zirconia based inert matrix fuel [172]. A survey of the literature shows that baddeleyite is highly durable in aqueous fluids. A case study of baddeleyite from the Jacupiranga carbonatite complex of southern Brazil provides some information on the crystal chemistry, durability, and radiation damage effects of the mineral. The mineral is highly resistant to hydrothermal alteration, which affected associated uranpyrochlore crystals. Many of the baddeleyite crystals are partially enclosed within the uranpyrochlore grains, which contain UO_2 and ThO_2 . These baddeleyite crystals received maximum alpha-particle doses within $10\ \mu\text{m}$ of the uranpyrochlore grain boundary, but the intense irradiation has not affected either the structural integrity or the aqueous durability of the mineral.

2.6. United States of America

Early US interest in inert matrix fuels originated from the desire to increase the performance of nuclear power plants and for use in closed fuel cycle systems where plutonium was bred as a fuel. Zirconia matrix fuels were investigated as a potential fuel for the Shippingport reactor, magnesia and zirconia matrix fuels were also investigated for use in plutonium spike enrichment applications. Another application of inert matrix fuel was for the use of Pu-240 for reactivity control in plutonium burning reactors. Since that time, the emphasis for IMF development in the US has shifted from plutonium utilization to plutonium destruction. The IMF R&D activities in the US include ion irradiation experiments on a number of candidate materials at the University of Michigan and research at LANL focusing on ion beam irradiation of ZrO_2 and ZrN . Further research work is currently performed at the ANL, LANL and at the INEEL, as well as in Universities of Purdue, Las Vegas and at the Massachusetts Institute of Technology.

2.6.1. US Advanced fuel cycle programme

In 2003, the US's inventory of commercial spent nuclear fuel inventory totals approximately 44 000 metric tons of heavy metal (MTHM). This inventory represents approximately 400 tons of plutonium. An additional spent fuel mass of 2 000 tons is produced each year by the current commercial fleet. Current US plans involve storage of this fuel at the repository at

Yucca Mountain, Nevada. If the current inventory of spent fuel is placed in the repository along with the expected spent fuel output for a zero growth scenario for nuclear power, the fuel inventory will reach the repository statutory limit of 63 000 MTHM in 2015 [173]. The driving force behind the desire to develop inert matrix fuels has thus changed from the desire to utilize plutonium in a closed fuel cycle in the 1960's and 1970's to the rapid destruction of the current and projected plutonium and minor actinide inventories in order to reduce repository requirements today.

Recent work in inert matrix fuel in the US began on the national level with the inception of the US Department of Energy Accelerator Transmutation of Waste programme in 1998. The advanced fuel cycle (AFC) programme is the successor to that programme, and is exploring options other than accelerator driven systems for transmutation. The AFC programme is divided into two parallel sets of activities focused on intermediate and long term technology development called 'series one' and 'series two' activities respectively.

Series one activities are focused on an intermediate term solution to the currently limited capacity of the US spent fuel repository. Series one activities have focused on reducing the volume of spent fuel destined for the repository. This is accomplished by separating the uranium from the spent fuel and returning some or all of the transuranics to the current commercial fuel cycle. An additional benefit is a decrease in proliferation risk through destruction and denaturing of plutonium in the LWR infrastructure. Because series one goals are intermediate term in nature, to be deployed as quickly as possible, the strategy for plutonium destruction uses more or less conventional fuel technology. The specific choice of fuel type and isotopic composition depends on decisions related to the methodology used to achieve proliferation resistance. Currently most consideration is being given to variants of MOX, which presents a relatively straightforward path to licensing in current US commercial reactors.

Series two activities are longer term activities with the goal of significantly reducing the long term radiotoxicity and heat load of high level waste sent to a geologic repository. Series two activities emphasize the use of proliferation resistant technologies. Deployment is a longer-term objective, and technology development focuses on advanced fuels that have high potential for use in transmutation but have not been fully demonstrated. Series two activities are targeted at both fast spectrum and thermal spectrum fuels. Included in the latter will be any work on IMF destined for the current commercial fleet.

The fuel for deployment in fast spectrum transmutation system, in order to take into account proliferation resistance and account for the difficulties involved in handling large amounts of americium and curium laced with some fission products, requires fuel handling in a heavily shielded hot cell facility. In order to be successful, the fuel must be easily fabricated using remote handling technologies, must not have a negative effect on reactor safety, must be compatible with an economic recycle process, and must result in a final waste form acceptable for the repository.

Nitride and metallic fuels have been chosen as the primary fuel candidates for fast spectrum transmutation systems. Both non-fertile and low-fertile fuels are considered for use in ADS and low-conversion ratio fast reactors, respectively. The fuels for low-conversion ratio fast reactors can contain approximately 50 wt% of uranium, and are a compromise between IMF and traditional fast breeder reactor fuel. The plutonium and minor actinide burn rate are much higher while retaining some of the physics related benefits of uranium. Nitride inert matrix fuels are based on ZrN, and metal inert matrix fuels are based on zirconium alloys. The nitride

system was selected due to the simple phase equilibria relative to metallic systems, as there is expected to be a continuous range of NaCl-type (rock salt) solid solutions for all transuranic nitrides. Nitride fuels will likely require ^{15}N enrichment, however, and require more complex fabrication processes relative to metals. The TRU-Zr metallic system was selected due to the ease of remote fabrication and recycle. Metallic fuel of composition U-20Pu-10Zr was previously the reference fuel for the IFR programme in the US, where on-site molten salt electrorefining was used as the fuel recycle process. Approximately 34 900 fuel pins and 418 complete “subassemblies” were fabricated remotely in the US fuel cycle facility (FCF) hot cells in work during the 1960’s that led up to the IFR programme. All US fast spectrum fuel concepts use a sodium bond for the following reasons: (1) a large fuel to cladding gap can be used to accommodate high swelling fuels without fuel/cladding mechanical interaction, (2) the gap size is not critical to fuel behaviour and a large tolerance on fuel diameter is acceptable, eliminating the need for pellet grinding, (3) fuel centerline temperatures are lower which limits americium transport and in-pile restructuring, and will provide a larger margin to fuel melting. ANL has produced and characterized a series of non-fertile and low-fertile metal alloy test nuclear fuels for the transmutation of actinide isotopes in either fast spectrum reactors or accelerator driven systems under the advanced fuel cycle initiative (AFCI). The uranium free alloys are currently under irradiation testing in the advanced test reactor (ATR) in the USA and two of the fuel compositions will begin irradiation testing in the Phénix reactor in France under the FUTURIX program in 2006. Some of the non-fertile metal alloy fuel compositions that are being developed under study are 40Pu-60Zr; 60Pu-40Zr; 50Pu-10Np-40Zr; 48Pu-12Am-40Zr and 40Pu-10Am-10Np-40Zr.

Because of stringent fuel performance requirements posed by a fast spectrum transmutation system, namely the effects of the presence of large quantities of the minor actinides (Am, Np, Cm), high expected burnup to minimize the number of recycle passes, compatibility with an economical recycle process, and the lack of a coherent database, extensive fuel testing is necessary prior to final selection of a fuel. There no longer exists a capacity for fast spectrum fuel testing in the USA. Because of this, initial development of series two fast spectrum fuels must be completed using testing in thermal spectrum test reactors, using a filtered thermal neutron spectrum. Further development requires collaboration with parallel fuel testing programmes in other countries for access to fast spectrum test reactors.

The US AFC programme is configured in three phases. Phase I was a basic technology evaluation completed in 2002, Phase II is demonstration of proof-in-principle that the technology is viable, and Phase III is a full-scale proof-of-performance demonstration to resolve engineering and design issues. The programme is currently in Phase II. During this phase, the capability of introducing advanced transmutation systems in a cost-effective, environmentally responsible, and proliferation-resistant manner is being investigated. Research is focused on determining which technologies are most suitable to reduce the cost of spent fuel disposal and to provide more detailed projections about future repository needs based on the technologies determined to be most promising. A decision on the need for a full-scale demonstration of transmutation is expected as a result of these initial research activities. Fuel development within this phase has so far concentrated on fast spectrum fuels. Specific activities include developing fabrication processes for test fuels, fabricating fuel test specimens for reactor insertion, characterizing the microstructures of these specimens, planning for irradiation testing, and development of irradiation test rigs. Several series of irradiation tests are planned during this phase, including a joint test of fuels with the CEA and ITU in the Phénix reactor at Marcoule, France.

Consideration has also been given to oxides and cermet dispersion fuels for the fast spectrum, but these have not been experimentally investigated. Dispersion fuels are desirable for high power density applications due to high matrix thermal conductivity, however TRU density is also limited by the need to disperse the fissile phase within the matrix. The generation of large amount of helium due to ^{241}Am also decreases the probability that previous success with dispersions can be directly applied to the specific case of TRU fuel. Oxide fuels are being investigated as fast spectrum transmutation fuels and targets in Western Europe, the Russian Federation and Japan, so research is not funded in the USA.

Advanced fuels for LWRs are also considered for development in the long term series by two research programmes. A variety of MOX and IMF fuel compositions are being proposed for inclusion in the LWR-2 experiment. To address the low thermal conductivity of the ZrO₂-based IMF and the instability in water of the MgO-based IMF, the dual phase MgO-ZrO₂ ceramics are proposed as a matrix for LWR fuel for actinide transmutation and Pu burning. It is envisioned that in a dual-phase system MgO will act as efficient heat conductor while ZrO₂ will provide protection from the coolant attack [174].

New processing methods for producing SiC and BeO matrix fuel materials are under development at the Purdue University. The process is based on the 'polymer impregnation and pyrolysis' to consolidate both particulate fuels like TRISO fuels, actinide particles and waste forms, and to impregnate UO₂ fuels with pure SiC to improve their thermal conductivity. For BeO, a second approach was developed that involves just a "co-sintering" route to produce high density fuels with co-continuous BeO and UO₂ phases. The advantages of the PIP process are that it represents a non-damaging consolidation process for particulates, and forms a continuous pure SiC phase. But several impregnation cycles are necessary to obtain high density SiC. The BeO "co-sintering" process requires special powder and granule mixing techniques, but only one normal sintering cycle. The handling of BeO needs to be controlled, but the requirements are similar to those for UO₂ powders. [175]

IMF for Generation IV systems

Recent interest in GFRs has prompted an effort on fuel development in this area. New and innovative fuels are required for this reactor concept due to the challenges of maintaining passive safety response during the high temperatures possible in unprotected loss-of-coolant accidents, high heavy metal density required for fast spectrum operation, low tolerance for parasitic neutron absorbers, and the ability to use this fuel in a closed cycle. The most promising fuel for this system that meets all of these requirements is a cermet type composite using an inert refractory ceramic matrix. Development of these fuels is at the conceptual stage. In the context of the development of supercritical water reactor (SCWR) technology as a part of Generation IV development, the US and Canada are exploring the possibility of a joint INERI (International Nuclear Energy Research Initiative) project to develop IMF fuel with high thermal conductivity would be particularly beneficial to any SCWR concept and which would be applicable for plutonium and MA management.

3. INTERNATIONAL PROGRAMMES

The cooperative activities on IMF concern joint irradiations, modeling exercises and exchange of data under the initiative of the IMF workshops.

3.1. Irradiations

The irradiation experiments are performed in cooperative projects a summarized below.

3.1.1. Irradiation of plutonium inert matrix fuel for “once-through-then-out” option

The plutonium incineration experiment named OTTO was prepared by JAERI, Nuclear Research and Consultancy Group (NRG) of the Netherlands and Paul Scherrer Institute (PSI) of Switzerland, and has been carried out at the high flux reactor (HFR) in Petten, Netherlands [116]. The irradiation behaviour of the inert matrices MgAl_2O_4 (spinel) and $(\text{Y,Zr})\text{O}_{2-x}$ (YSZ) are examined in this experiment, because the matrices are considered to be suitable for an once-through scenario because of their insolubility in nitric acid and their geological stability for final disposal.

Sol-gel micro-sphere pelletisation was used for fabrication of the targets of controlled density, homogeneity and microstructure for this OTTO study. Seven different kinds of targets with respect to Pu-composition or microstructure were prepared. The main specifications of the seven targets are summarized in Table X. Six of the fuel compositions listed in the Table X contain α -YSZ matrix, four of which also contain α - MgAl_2O_4 matrix to form 2-phase material, and one target is MOX. Two of the four YSZ- MgAl_2O_4 2-phase materials are homogeneously blended (micro-dispersed) ones, while YSZ particles of 250 μm diameter are dispersed (macro-dispersed) in the MgAl_2O_4 matrix in the other two α -phase targets. Either Er_2O_3 or depleted UO_2 is added in the two YSZ solid solution targets, two YSZ- MgAl_2O_4 micro-dispersed targets and two MgAl_2O_4 -YSZ macro-dispersed targets. The fissile Pu concentration of the MgAl_2O_4 containing targets was slightly reduced (see Table X), because it was found that MgAl_2O_4 interacts with the YSZ containing the fissile atoms at temperatures higher than 1973 K. The diameter of the pellets is 8.00 mm. The pellets were encapsulated in Zircaloy-4 cladding tubes with an inner diameter of 8.22 mm and an outer diameter of 9.00 mm. The stack length of each target is about 67 mm. The OTTO sample holder with all the 7 target pins was placed in the HFR in-core irradiation position. The computed maximum linear heat-rate and the central temperatures of all of the seven pins are also listed in the table together with the burnup of the fuels.

The pins have been irradiated in HFR for 22 cycles i.e. 548 full power days (FPD) until the end of December 2002. Unfortunately, the neutron radiograph images have shown the failure of Pin 4 after seven cycles of irradiation. The other pins performed satisfactorily thus that the OTTO irradiation has reached efficiently a burnup of $200 \text{ GW}\cdot\text{d}/\text{m}^3$, corresponding to a Pu depletion of 50–60%. Neutron radiography showed that all fuel pins retained their initial pellet structure, with less than 2% of axial swelling. The post irradiation examination of the irradiated targets started in early 2003. The results of both non-destructive and destructive post-irradiation examination as well as the neutronics modelling after irradiation are presented in recently [116]. The total plutonium depletion at the end of irradiation was around 35% in the inert matrix fuels, whereas in the MOX reference capsule, which experienced a similar burn up, it was only 15%. The results confirmed the absence of large swelling during irradiation. The crack formation, observed in the zirconia-based targets, is comparable to that of UO_2 fuel. The fission gas release is around 10% for the zirconia-based targets, whereas for

the heterogeneous spinel-based targets, which experienced a much lower irradiation temperature, it is around 5%. The homogeneous targets show very low fission gas release.

TABLE X. TEST PINS FOR THE OTTO IRRADIATION INCLUDING THE CALCULATED VALUES FOR MAXIMUM LINEAR HEAT-RATE AT THE BEGINNING AND THE END OF IRRADIATION (BOI AND EOI), THE CENTRAL FUEL TEMPERATURE AT BOI AND BURNUP

No.	Pellet composition	Inclusion size (μm)	Pu_{fis} density (g/cm^3)	Power (W/cm)		T^{a} (K)	Burnup ($\text{GW}\cdot\text{d}/\text{m}^3$)
				BOI	EOI		
1	$(\text{Er}, \text{Y}, \text{Pu}, \text{Zr})\text{O}_{2-x}$	solid solution	0.37	280	112	2167	200.1
2	$(\text{Y}, \text{Pu}, \text{U}, \text{Zr})\text{O}_{2-x}$	solid solution	0.34	288	119	2167	208.7
3	$(\text{Er}, \text{Y}, \text{Pu}, \text{Zr})\text{O}_{2-x}$ + MgAl_2O_4	<25	0.32	220	100	1412	166.8
4	$(\text{Y}, \text{Pu}, \text{U}, \text{Zr})\text{O}_{2-x}$ + MgAl_2O_4	<25	0.31	202	106	1341	162.7
5	$(\text{Er}, \text{Y}, \text{Pu}, \text{Zr})\text{O}_{2-x}$ + MgAl_2O_4	200–250	0.31	218	99	1366	164.7
6	$(\text{Y}, \text{Pu}, \text{U}, \text{Zr})\text{O}_{2-x}$ + MgAl_2O_4	200–250	0.30	199	105	1298	161.0
7	$(\text{Pu}, \text{U})\text{O}_2$	solid solution	0.39	217	166	1640	207.5

^a central temperature

3.1.2. Halden irradiations

Two irradiations experiments are ongoing: (i) Korea Atomic Energy Research Institute (KAERI), PSI and OECD Halden tests on Pu-IMF and (ii) Politecnico di Milano (POLIMI) of Italy, ENEA, and OECD Halden test with several U-IMF.

The irradiation experiment IFA-651.1 with IMF based on yttria stabilised zirconia and with MOX fuel started as a part of the OECD Halden Reactor Project at end of June 2000 [189]. The aim of this experiment is to measure the central line temperature change with burn up, the fission gas release, the densification, the swelling and the general thermal behaviour of these fuels during irradiation. A secondary aim is the direct comparison of the fuel performance between YSZ IMF and MOX fuels. The rods were equipped with expansion thermometers (rods 3 & 5), thermocouples (rods 2, 4, 1 & 6), and/or stack elongation detectors (rods 2, 4 & 6). All rods were equipped with pressure transducers. In order to get a good record of the axial power distribution, the assembly is instrumented with three axial neutron detectors, which are placed in the axial centre of the assembly. The Halden reactor is a boiling heavy water cooled and moderated reactor with a thermal power of 18–20 MW. The moderator temperature is about 510 K and the reactor includes 35 instrumented fuel assemblies and 70 driver assemblies. For the current ongoing tests, the burn up will reach $490 \text{ kW}\cdot\text{d}\cdot\text{cm}^{-3}$, or $540 \text{ MW}\cdot\text{d}\cdot\text{kg}^{-1}$ of heavy metal (or $54 \text{ MW}\cdot\text{d}\cdot\text{kg}^{-1}$ of heavy metal for MOX) in 1000 full power days corresponding to plutonium reduction of 57%. Initial results indicate among other things considerably higher operating temperatures in the IMF rods have been observed compared with those in the MOX fuel. The higher temperatures, which were caused by the lower thermal conductivity of IMF, result also in higher FGR of the IMF rods.

The inert matrix thoria fuel experiment at Halden was presented [176] as a 6 rod irradiation involving pure calcia stabilized zirconia, this material loaded with thoria and thoria material alone. All these material are doped with HEU prior to testing Pu loaded samples. The instrumentation consists of central temperature thermometers, expansion thermometers, pressure transducer and cladding elongation detector. Irradiation is ongoing and PIE is foreseen in 2 years.

3.1.3. IMF irradiation for EFTTRA

In Europe, extensive research on americium transmutation is performed in the Experimental Feasibility of Targets for Transmutation (EFTTRA) collaboration [177], with uranium-free matrix targets for transmutation in LWRs or FRs. The EFTTRA programme is an international collaboration research project supported and performed by CEA ITU, NRG, EdF, IAM and FZK as well as JRC-IAM and JRC-ITU of the European Commission.

The EFTTRA activities are focused on the uranium-free dispersion type fuels, to limit the effect of the radiation damage by energetic fission fragments and alpha particles. Within the EFTTRA collaboration, the current emphasis is on oxides, and to a much smaller extent on nitrides and metals. Table XI shows the EFTTRA irradiations. Several selected materials are being tested by irradiation experiments in the reactor Phenix (France) and in the High Flux Reactor (HFR, Netherlands). In case of oxide fuel, americium oxide has unfavorable properties viz., low thermal conductivity, high oxygen potential and high chemical reactivity towards many matrices. A solid solution of AmO₂ in, for example, ZrO₂ is therefore proposed as an alternative. For a nitride fuel, the stability of AmN at high temperature is doubtful. Again, stabilization in a host phase could be considered, e.g. in ZrN. The most important inert matrix candidates are MgO, MgAl₂O₄, Y₃Al₅O₁₂, Y₂O₃ for oxide fuel and TiN and ZrN for nitride fuel. The americium host particle size is optimal in the diameter region between 50 and 300 μm, the lower limit being determined by the increase of the irradiation damaged fraction of the inert matrix and the upper limit being determined by the temperature gradient in the host particle.

TABLE XI. HFR IRRADIATION EXPERIMENTS PERFORMED IN THE EFTTRA ACTIVITY [95, 175]

Name	Reactor	Description	Start (year/month)	Length (EFPD)	Extent of fission (% FIMA)
T2	HFR	Neutron damage in inert matrices	96.02	386.4	—
T2bis	HFR	Neutron damage in inert matrices	96.02	101	—
T3	HFR	Neutron damage in inert matrices	97.10	199	—
		Dispersion inert matrix fuel using enriched UO ₂			18
T4	HFR	Americium in spinel	96.09	358.4	35
T4bis	HFR	Americium in spinel	97.06	652.6	~50
T4ter	HFR	central fuel temperature of spinel/UO ₂	97.06	652.6	32
T5	HFR	Hybrid fuel for Am: influence of inert matrix	Planned	350	~30

3.1.4. Inert matrix fuel for transmutation in fast reactor

Several programmes are ongoing about the use of inert matrices in fast reactors. These programmes comprise a part of the EFTTRA activities, a part of the CONFIRM activities as

well as some bilateral projects namely between (i) PSI and CEA (nitride fuel fabrication), (ii) CEA and IPPE (IMF irradiations in BR10 and BOR60) as well as (iii) local cooperative activities between JAERI and the Russian Federation. Table XII gives the main parameter for IM irradiation in Phenix in the frame of the EFTTRA programme.

TABLE XII. FAST REACTOR IRRADIATION EXPERIMENTS PERFORMED IN THE EFTTRA ACTIVITY [175]

Name	Reactor	Description	Start (year/month)
F1	Phenix	Neutron damage in inert matrices	94.12
F1A	Phenix	Neutron damage in inert matrices	98.05

3.2. Neutronic benchmarks

IMFs are neutronicly very different from standard UO_2 and MOX fuels. In a standard fuel, the presence of ^{238}U has a stabilizing effect on the neutron spectrum. In IMF, during burnup, the neutron spectrum as well as the neutron flux can vary much more than in conventional UO_2 and MOX fuels because of the larger variation of the absorption cross section due to the reduction of plutonium density. The methods and data currently used for LWR analyses have not been tested for such exotic fuel materials, and there exist no appropriate experiments for validating these tools for IMF analysis. An international exercise has accordingly been launched to compare the relative performance of different code systems and the accuracy of the basic data for the use in the analysis of LWR loaded with IMF [178, 179]. Seven institutions, CEA of France, JAERI of Japan, KAERI of Republic of Korea, NRG of the Netherlands, POLIMI of Italy, PSI of Switzerland and SKODA-Nuclear Machinery (SKODA) of Czech Republic, were participated in this activity. The codes and the nuclear data used by the participants are summarized in Table XIII.

TABLE XIII. CODES AND NUCLEAR DATABASE USED IN THE IMF PHYSICS BENCHMARKS

Institution	Code	Library
CEA	TRIPOLI-4 ^a	JEF-2.2
CEA	APOLLO-2	JEF-2.2
JAERI	SRAC-95	JENDL-3.2
KAERI	HELIOS	ENDF/B-VI
NRG	WIMS-7B	JEF-2.2
POLIMI	WIMS	WIMS-86
PSI	BOXER	JEF-1.1 ^b
SKODA	WIMS-7B	JEF-2.2

^a For beginning of life only; ^b Zr cross sections are taken from ENDF/B-IV

From the neutron physics point of view, the IMFs are characterized by large burnup reactivity swing and reduced negative reactivity coefficients. To counterbalance the above effects, it is possible to add a burnable absorber or a fertile nuclide. Alternatively, the IMF rods or assemblies can be mixed with normal UO_2 rods or assemblies. The benchmark activity therefore studied 5 different fuel materials and was divided into three phases.

The first phase of the exercise was devoted to analyze infinite arrays of identical IMF cells under conditions of a present-day PWR. Five fuel compositions covering a range of potential candidates of IMFs for Pu-burning have been studied. The materials studied are: (1) Al₂O₃-ZrO₂-MgO; (2) Al₂O₃-ThO₂-MgO; (3) ZrO₂-Er₂O₃; (4) ZrO₂-¹⁰B and (5) ThO₂. Use of reactor-grade (RG) plutonium (58 wt% ²³⁹Pu) was considered for all five IMF compositions. In addition, fuels 1, 2 and 5 were also analyzed with weapons-grade (WG) plutonium (93 wt% ²³⁹Pu).

Comparison of the results of cell burnup calculations indicate a fair agreement in k-inf, generally within 1% *dk* between participants, both at beginning of life (BOL) and end of life (EOL) after 1200 days of irradiation. At BOL, the fuel temperature coefficients also agree well among the different contributions, but as shown in Table XIV, larger differences are observed at EOL. The agreement of void coefficients is good for low voidage, but for void fractions >90%, there are large differences. The agreement in the calculated boron worth is good.

TABLE XIV. FUEL TEMPERATURE COEFFICIENTS (PCM/K) AS MEAN VALUES OVER ALL PARTICIPANTS RESULTS AND RANGE FROM THE MEAN VALUE (PCM/K)

	Inert matrix	BOL Fuel temperature coefficient (pcm/K)		EOL Fuel temperature coefficient (pcm/K)	
		Mean	Range from mean	Mean	Range from mean
WG-1	Al ₂ O ₃ -ZrO ₂ -MgO	-1.22	-0.16 to +0.06	-0.66	-0.24 to +0.31
RG-1	Al ₂ O ₃ -ZrO ₂ -MgO	-1.34	-0.24 to +0.24	-0.59	-0.37 to +0.55
WG-2	Al ₂ O ₃ -ThO ₂ -MgO	-3.19	-0.12 to +0.09	-2.95	-0.13 to +0.13
RG-2	Al ₂ O ₃ -ThO ₂ -MgO	-3.29	-0.13 to +0.31	-3.13	-0.19 to +0.35
RG-3	ZrO ₂ -Er ₂ O ₃	-1.66	-0.17 to +0.21	-1.29	-0.20 to +0.25
RG-4	ZrO ₂ - ¹⁰ B	-0.73	-0.27 to +0.24	-1.06	-0.31 to +0.36
WG-5	ThO ₂	-3.71	-0.10 to +0.11	-3.52	-0.21 to +0.14
RG-5	ThO ₂	-3.71	-0.20 to +0.35	-3.63	-0.24 to +0.41

The basic configuration treated in the second phase of the benchmark consists of a sub-assembly with 5 × 5 square fuel rod positions. The central position is occupied by an IMF rod of the same five materials as Phase 1, the four corners are empty and filled with water, while the other positions are occupied either by UO₂ or by MOX rods. The main objective for this phase of the exercise is to check the modelling of the strong interaction between the dominating neutron spectrum of the surrounding UO₂ or MOX and the different materials of the IMF cells.

The first comparisons have been made for BOL against results of the Monte Carlo code TRIPOLI-4 using a cross section library based on JEF-2.2 data. Most of the infinite multiplication factors for the IMF cells as calculated by the deterministic codes are greater than the reference Monte Carlo values by up to about 4% (Table XIV). All the infinite multiplication factor values for the UO₂ and MOX regions are smaller than the reference TRIPOLI-4 results by about 1%. Table XV also shows that the power densities in the IMF cells, as predicted by the deterministic codes, are always higher than the reference values by 2 to 11%.

TABLE XV. RANGE FROM TRIPOLI-4/JEF-2.2 RESULTS OF EACH PARTICIPANT'S RESULTS FOR MULTIPLICATION FACTORS ($k\text{-inf}(IMF)$) AND POWER DENSITIES ($pd_{(ave.)}$ IS AVERAGE IMF POWER DENSITY RELATIVE TO COMPLETE SUBASSEMBLY) IN THE IMF ROD IN PHASE 2 SUBASSEMBLIES AT BOL

	Inert matrix	TRIPOLI-4/JEF-2	Range from TRIPOLI-4
$k\text{-inf}(IMF)$			
UO ₂ -RG1 ^a	Al ₂ O ₃ -ZrO ₂ -MgO	1.576 ±0.37%	+0.33 to -0.40 %
UO ₂ -WG1	Al ₂ O ₃ -ZrO ₂ -MgO	1.708 ±0.38%	+0.79 to -0.05 %
UO ₂ -RG2	Al ₂ O ₃ -ThO ₂ -MgO	1.450 ±0.37%	+0.97 to -0.37 %
UO ₂ -RG3	ZrO ₂ -Er ₂ O ₃	1.304 ±0.37%	+1.83 to -0.22 %
UO ₂ -RG4	ZrO ₂ - ¹⁰ B	1.214 ±0.36%	+0.54 to -1.29 %
UO ₂ -RG5	ThO ₂	1.394 ±0.36%	+1.27 to -0.43 %
UO ₂ -WG5	ThO ₂	1.447 ±0.37%	+1.68 to +0.44 %
MOX-RG3	ZrO ₂ -Er ₂ O ₃	1.121 ±0.50%	+3.87 to -0.37 %
MOX-RG5	ThO ₂	1.184 ±0.49%	+3.52 to -1.15 %
$pd_{(ave.)}$			
UO ₂ ^b	—	1.605 ±0.26%	+10.6 to +2.3 %
MOX	—	0.814 ±0.36%	+11.4 to +1.8 %

^a This means that the value of $k\text{-inf}$ predicted by the Monte-Carlo code TRIPOLI is 1.576, with a statistical uncertainty of ±0.37%; the deterministic calculations varied about this value by +0.33% to -0.40%, e.g. The largest value was 1.576+0.33%, and the smallest value was 1.576-0.4%.

^b In the UO₂ lattice with a central IMF element, the relative power density of the IMF rod to the cell-average was predicted by the TRIPOLI code to be 1.605 (with statistical variation of 0.26%); the deterministic calculation were higher than this by a range between 10.6% and 2.3%.

At EOL, with respect to the average over all participants' results, the infinite multiplication values for the complete sub-assembly show differences of about 1%. The spread in the infinite multiplication factor for the IMF cell itself at EOL is the largest in the material 1 case to be 5–6%. Rather large differences have been noted in the prediction of kinetic parameters. Discrepancies are up to 10% for the delayed neutron fraction, and 30% for the neutron generation time.

Phase 3 of the activity is concerned with the more realistic situation of an IMF assembly surrounded by UO₂ assemblies. This is quite representative of the initial stages foreseen for IMF introduction in PWR power plants, namely with only a few IMF assemblies loaded into a UO₂ fueled core. Only UO₂, and not MOX, environment has been considered in Phase 3. Four types of IMF assemblies have been considered: fuel material 1 with RG- and WG-Pu, material 3 with RG-Pu and material 5 with RG-Pu. Moreover, a MOX fuel assembly in UO₂ assemblies has been included in the list, in order to compare the relative difficulties of treating IMF and MOX assemblies in a UO₂ core.

For the multiplication factor of the complete configurations, the agreement is fairly good at BOL, as well as at EOL. The deviations of individual results from the average are within 1%. Discrepancies in the infinite multiplication factors of IMF assemblies shown in Table XVI are much larger, especially at high burnup. The most discrepant results become larger than the average by up to 6% at EOL. Contrary to the spreads in infinite multiplication factor (neutron balance) differences for the IMF, the results for the maximum power density (always at IMF/UO₂ interface) are found to agree well within about 2%.

TABLE XVI. INFINITE MULTIPLICATION FACTOR OF IMF AND MOX ASSEMBLIES AS MEAN VALUES OVER ALL PARTICIPANTS RESULTS AND RANGE FROM THE MEAN VALUE

IM composition		<i>k-inf</i> (IMF) at BOL		<i>k-inf</i> (IMF) at EOL	
		Mean	Range from mean	Mean	Range from mean
WG-1	Al ₂ O ₃ -ZrO ₂ -MgO	1.6426	-0.44 to +0.32 %	0.7872	-2.38 to +5.37 %
RG-1	Al ₂ O ₃ -ZrO ₂ -MgO	1.4853	-0.78 to +0.35 %	0.7130	-4.91 to +6.25 %
RG-3	ZrO ₂ -Er ₂ O ₃	1.1612	-1.63 to +2.62 %	0.9635	-0.81 to +1.08 %
RG-5	ThO ₂	1.2397	-1.10 to +0.57 %	0.9642	-0.98 to +0.78 %
MOX	.	1.2685	-0.30 to +0.74 %	0.9883	-0.56 to +0.68 %

This study is an interesting and useful exercise in a new field of reactor physics. Generally the agreement between the results of the participants is not bad. However there some important discrepancies were found: In Phase 1 of the benchmark, large differences were found in the fuel temperature coefficient values at EOL. In Phase 2, discrepancies were observed for power peaking effects and kinetic parameters. The differences in the power peaking effect were not so large in Phase 3, in the more realistic configurations, while the discrepancies in neutron balance for the IMF assembly at EOL were found to be larger than in Phase 1. The fuel temperature coefficient, the power peaking effect and kinetic parameters are related to reactor safety. Through further detailed analyses based both on calculations and experiments, useful information would be obtained on IMF reactor physics and safety.

3.3. IMF workshops

In 1994, in the IAEA Technical Committee Meeting on Unconventional Options for Plutonium Disposition held at Obninsk, Russian Federation [180], JAERI, POLIMI and PSI presented their research works on the use of inert matrices for plutonium incineration fuel in LWRs. After the discussion between these three institutions, there was a proposal of having an international workshop on the use of IMF, because it became clear that there was a common interest in IMF in many countries. The first IMF workshop was thus held at PSI, Switzerland on 20 September 1995 with 12 participants from 4 institutions of 3 countries, Italy, Japan and Switzerland.

After the first IMF workshop, 9 workshops were held in France, Italy, Japan, Netherlands, Switzerland, and the United Kingdom. The numbers of the participants and the papers presented in the workshop are increased year by year. The 4th workshop, also at PSI, on 19–20 October 1998, was held in conjunction with the OECD/NEA ARWIF workshop and the workshop was successful with 64 participants from 11 countries and 1 international organization. The 6th and 10th IMF workshops were held as a special session of the European

Materials Research Society Spring Meeting at Strasbourg. In the last 8 workshops that were held before 2003, a total of more than 400 participants have taken part from 17 countries, Australia, Belgium, Canada, Czech Republic, France, Germany, India, Israel, Italy, Japan, Republic of Korea, Netherlands, Russian Federation, Sweden, Switzerland, the UK and the USA, and 3 international organizations, EC, IAEA and OECD Federation.

The IMF workshops cover not only plutonium burning but also minor actinides transmutation, materials both as a fuel and as a target in reactors (thermal and fast) and accelerator driven systems. The subject-areas of the workshop ranges from the materials studies (such as basic thermal properties), fuel fabrications, compatibilities, fuel behaviour under ion irradiation and in-pile irradiation, benchmark calculations on data and method, physics experiments, core and assembly design and safety analyses. Until around the 4th IMF workshop, the discussion on the physics benchmark calculations were cited extensively, whereas the results of irradiation tests are becoming available in the recent workshops.

Since the 4th workshop, the proceedings were published in a journal every 2 years. The proceedings of the 4th IMF workshop were published as a special issue of Journal of Nuclear Materials: Volume 274. The proceedings of the 6th workshop were published in Progress in Nuclear Energy: Volume 38, and the proceedings of the 8th workshop were published in Journal of Nuclear Materials: Volume 319. A total of 111 papers have been published in these 3 journals.

4. RESULTS AND OUTLOOK

Several IMF options are available for eventual application in L&HWR, FR, and HTGR systems. The choice of IMF depends heavily on the reactor system as well as on the specific application. This in turn, hinges on the present nuclear infrastructure and future nuclear scenarios. For example, in a multi-recycle scenario, the plutonium burnup potential is not as important as the requirement that the fuel be easily reprocessed and refabricated into new fuel. On the other hand, a once through cycle for destruction of plutonium requires that the fuel be capable of high burnup and have low solubility in groundwater under repository conditions so that it may be safely placed in long term storage. Fast reactor, gas cooled reactor, and thermal reactor systems have different coolants and therefore different coolant compatibility requirements. In all cases the IMF and the design of the whole fuel element will depend on the reactor type: conventional L&HWR, future FRs, or a dedicated system for burning MAs such as ADS. During fuel development, fuel behaviour calculations are first conducted to establish concepts that appear to be promising and to optimise fuel design parameters based on available data.

The choice of matrix material is complex, and depends on factors including melting temperature, thermal expansion, response to neutron irradiation and fission fragment implantation in the matrix, chemical inertness with respect to the fuel, clad and coolant, degree of activation during irradiation and types of activation products produced, neutron absorption cross section, ability to be fabricated, resistance to corrosion, degradation of strength and thermal conductivity, during irradiation.

One of the most important parameters in fuel design is the fuel's margin to melting during normal and off-normal reactor operating conditions. To give a general picture of the situation in this regard, the thermal conductivity (κ) for some proposed matrix materials is plotted versus the melting/decomposition temperature (T_m) as shown in Fig. 7. The thermal conductivity used in Fig. 7 is based on available literature data. In some cases, there is a high degree of uncertainty or scatter in the data, or significant changes in conductivity as a function

of temperature. The appropriate materials that are most desirable in terms of the combined parameter κ/T_m fall in the upper right hand corner of the plot.

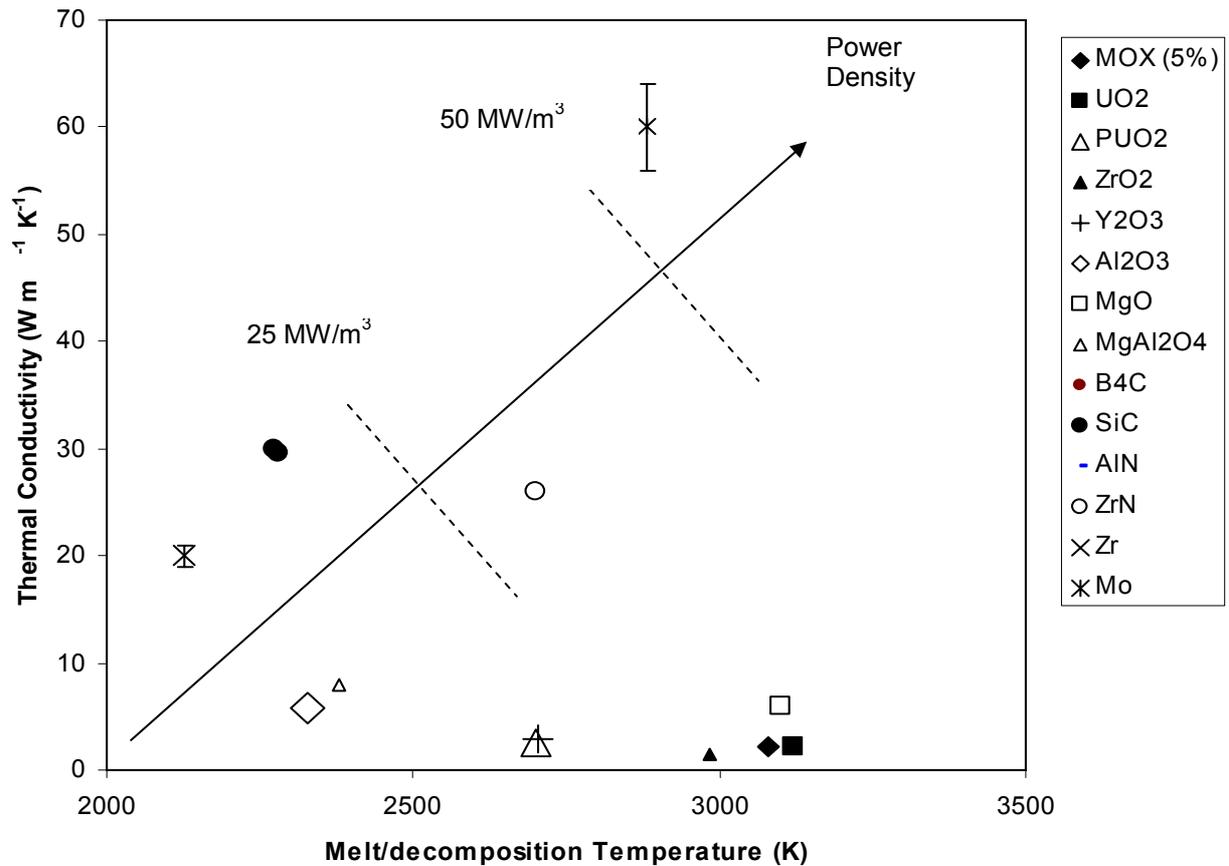


Fig. 7. Thermal conductivity plotted versus melting temperature for some materials considered as inert matrices for dispersed fuel. MOX and UO_2 are plotted for comparison.

Inert-matrix fuels can be roughly divided into two major categories based on fuel microstructure. These are homogeneous and heterogeneous fuel, both discussed in the following sections.

4.1. Homogeneous fuels

Solid solution fuels utilize a homogeneous distribution of fissile atoms throughout the fuel matrix. In the case of solid solution fuels, fission damage is more-or-less uniformly distributed throughout the fuel. The response of the inert matrix to fission fragment damage, namely in terms of (i) fission gas production and diffusion, and (ii) formation of separate solid phases of fission products become the features most important to fuel behaviour. Two different tactics are used in the design of solid solution fuel. Metallic fuel design allows for a large amount of free swelling and high gas release due to the metal bonding and is applicable to fast reactors. Ceramic solid solution fuels rely on materials that exhibit low swelling and gas release and use a tightly controlled fuel-to-cladding gap to control fuel temperature.

4.1.1. Oxide solid solution fuels

Solid solution ceramics are attractive because they are extensively tested as power reactor fuel (i.e. uranium oxide clad in zirconium alloy and MOX) and they exhibit a relatively small perturbation over large range of power-levels. As such, solid solution fuel for application to

plutonium burning in current generation power reactors has been the subject of substantial recent development activity.

Ceramic solid solution fuels incorporate fissile and inert components into a homogeneous mixture consisting of a single crystalline phase. A commonly considered inert matrix is zirconia (ZrO_2) stabilized in the cubic fluorite phase by additions of yttria (Y_2O_3) or calcia (CaO) [181, 182]. Plutonium dioxide (PuO_2) or minor actinide dioxides (MAO_2) also have the fluorite crystalline structure and form a continuous solid solution. Burnable poisons such as the rare earth elements erbia (Er_2O_3) or gadolinia (Gd_2O_3) can be incorporated into the solid solution and can also act as cubic stabilizing additions. Some of the fission products may also be found to stabilize zirconia as shown by X ray absorption spectroscopy e.g. [163].

The possibility for the development of fuels in the ZrO_2 - PuO_2 system was realized in the early 1960s, see for example [51]. Irradiation testing of ZrO_2 - UO_2 fuels was conducted at this time [42]. A large body of information specific to in-reactor use of zirconia-plutonia based IMF has been compiled within the last decade. This data includes fabrication development, core physics [166], phase equilibria studies, microstructural characterization, thermal property measurement, corrosion testing [170], and ion [183] and neutron irradiation. Thoria doped zirconia (as well as zirconia-spinel (MgAl_2O_4) based ceramic – ceramic composites) are also considered and fabricated by conventional powder metallurgical route comprising of milling, compaction and sintering.

In general fabrication may be carried out either by co-milling processes similar to current industrial MOX fabrication routes or by coprecipitation from an aqueous solution [184]. The latter route leads to a more homogeneous distribution of elemental constituents within the fuel. Densification of powder compacts is carried out by sintering in a more-or-less conventional manner.

Irradiation damage studies have been carried out using both neutron and ion irradiation techniques. Ion irradiation is used to simulate fission fragment damage. A summary of the work in this area prior to 1999 is given in [183]. Breakaway swelling of fuels is often triggered by transition from an ordered crystalline structure to an amorphous phase (no long range order); this behaviour is undesirable in an inert matrix. Several studies using Xe^+ and Xe^{++} irradiation of cubic YSZ at 60–400 keV or 1.5 MeV and temperatures as low as 20 K did not result in amorphisation of stabilized zirconia, nor was it observed after irradiation with 72 MeV I^+ ions to a dose of $5 \times 10^{19} \text{ m}^{-2}$ at 300–1770 K. Laser profilometry of the specimen surfaces after irradiation showed no evidence of ‘popout’ features, indicating that swelling was <2%. Amorphisation has not been observed in any case for fully stabilized ZrO_2 indicating that this material can be irradiated to a dose exceeding 100 dpa without the occurrence of this phenomenon [183].

Implantation of xenon in yttria stabilized ZrO_2 has also been used to study mobility of this fission gas. Results obtained by measuring the Xe depth profile as a function of annealing temperature using Rutherford Backscattering indicate no measurable mobility of Xe to 1773 K [185]. Low gas release is a desirable attribute of commercial reactor fuel when coupled with low swelling behaviour. Additional work has focused on detailed analysis of the microstructure and crystalline structure [162, 186], much of the work being completed on materials containing ceria as a surrogate for plutonia. Zirconia has low solubility in water and is known to be resistant to leaching in water [171] and also difficult to dissolve in nitric acid [2]. These latter features make the fuel desirable for direct disposal in a geologic repository, but undesirable for a fuel cycle that relies on reprocessing.

The properties, fabrication, and microstructure of zirconia-based ceramic solid solution fuels are well understood due to recent efforts in out-of-pile development and testing. Due to their similarity to existing L&HWR fuels, these fuels could be one of the potential options for deployment as lead test assemblies in the existing fleet of commercial water reactors within near future. The primary uncertainty involved with using zirconia as an IMF is its low thermal conductivity. Questions about the effect of the low thermal conductivity on irradiation behaviour can only be answered by in-reactor testing, which is currently underway. Thermal conductivity of $(\text{Er}_{0.07}\text{Y}_{0.10}\text{Ce}_{0.15}\text{Zr}_{0.68})\text{O}_{1.915}$ and $(\text{Er}_{0.07}\text{Y}_{0.10}\text{Ce}_{0.15}\text{Zr}_{0.63})\text{O}_{1.89}$ measured in the range of 300–1000 K was found to be 1.90 ± 0.15 W/m·K with almost no temperature dependence within that temperature range [187], which agrees approximately with published data for calcia stabilized zirconia [188]. Comprehensive analysis of the thermal conductivity data of zirconia based IMF has been conducted [158]. The low thermal conductivity results in high fuel centre-line temperatures relative to MOX fuel; for example, calculated pellet centre-line temperatures at a linear heating rate of 24 kW/m are 1250 K for MOX fuel and 1700 K for zirconia IMF at the BOL, a difference of 450°C [184]. The temperature independent nature of the thermal conductivity, however, is beneficial in limiting the positive fuel temperature feedback caused by (typically) decreasing conductivity as a function of temperature during thermal excursions. Thermal conductivity is foreseen to have little variation with the burnup [158]. The values of the in-pile thermal conductivity will be influenced by the fission-related processes, therefore only instrumented in-reactor testing will be used to determine the thermal response of zirconia-based IMF as a function of power, power history, and burnup. The OTTO test in HFR-Petten was successfully completed in December of 2002 [111], and a joint test in Halden is in progress [169, 189]. The irradiations are performed at Halden reactor with a target burnup of $\sim 400\text{--}450$ kW·d/cm³, which is equivalent to $\sim 40\text{--}45$ MW·d/kg. Initial irradiation results indicate that higher operating temperatures in all the different IMF rods have been observed compared with those in the MOX or thoria fuel [191]. The higher temperatures observed were caused by the lower thermal conductivity of IMF, resulting in higher fission gas release of the IMF rods than the MOX or thoria rods. If fuel thermal profiles during irradiation prove unacceptable, annular pellets [151], such as used in WWERs [192], fuel with cruciform shape [193], fuel columns with high-conductivity disks between pellets (so-called zebra configuration), or pellets with a non-fuelled centre region, might be used to reduce peak fuel temperature.

Deployment will thus depend on successful completion of the irradiation testing and post-irradiation examination of rod let specimens currently under irradiation in research reactors. Fuel transient behaviour must also be established through the completion of rapid power ramp testing in a test reactor and transient testing of irradiated fuel in a transient test reactor. A discussion of fuel safety behaviour is included in Section 4.4.

There have also been cursory studies conducted on other IMF's as candidate materials for solid solution fuels, most notably ceria and yttria. CeO₂ exists in the fluorite phase and is isostructural with PuO₂ and UO₂. Y₂O₃ is nearly so, exhibiting the bixbyite structure that resembles the fluorite structure with one fourth of the anions missing. (Pu,Ce)O₂ and (Pu,Ce)O_{2-x} specimens were irradiated as a split pellet stack in the TRABANT 1 irradiation in HFR-Petten. [194]. In these fuels, the plutonium content was high, with a cerium to total metal ratio of 0.56–0.59 for both fuel types. After irradiation to ~ 4.6 MW·d/MT, the fuel specimens were removed from the reactor due to failure of two pins in the TRABANT 1 experiment. On post-irradiation examination, it was found that the fuel had melted, presumably due to the low thermal conductivity of the sub-stoichiometric composition. [195, 196] The ceria-based fuel was also found to disintegrate on immersion in molten sodium at

550°C for 6 hours, making it unappealing for use in sodium-cooled reactors [194]. TANDEM accelerator tests conducted at AECL also showed high swelling of ceria when irradiated with 72 MeV iodine ions [197]. Heavy ion irradiation experiments that have been conducted on Y_2O_3 , reveal unusual behaviour, with a cubic to monoclinic transformation occurring under irradiation with molybdenum and lead ions [198]. The irradiation behaviour of macrodispersion of UO_2 in yttria and ceria (discussed in section 4.2.1), however, indicates that the response of the material to the combined effects of neutrons and local fission fragment damage of both ceria and yttria appears to be acceptable under the irradiation conditions tested. More comprehensive irradiation testing is required to understand the behaviour of these latter fuels in-pile.

4.1.2. Nitride solid solution fuels

Nitride fuels are attractive primarily due to high thermal conductivity relative to actinide oxides, high actinide density, and simple phase equilibria – only a rock salt (face-centered cubic) phase is known to form in the Np, Pu, Am, and Cm systems. The major issue confronting the use of this fuel is the need to enrich nitrogen in the ^{15}N isotope in order to avoid production of large amounts of ^{14}C in reactor through the $^{14}N(n,p)^{14}C$ reaction. Typically ZrN, TiN, YN, or AlN are proposed as the inert matrix phase for nitride fuels. Recent emphasis has been on fabrication of these IMFs of specimens for irradiation testing. NpN, (Np,Pu)N, (Np,U)N, AmN, (Am,Y)N, (Am,Zr)N, and (Cm,Pu)N have been recently prepared by JAERI by the carbothermic reduction of oxides [190]. Pellets of (Pu,Zr)N and two phased TiN + PuN have been fabricated and these pellets are undergoing irradiation testing [191]. Activities on (Pu,Zr)N fuel fabrication are also conducted in Switzerland as part of the CONFIRM programme [150] and in the US as part of the AFCI programme. Both of these programmes use fabrication routes that rely on the carbothermic reduction of oxide powders.

The fuels tested in the CONFIRM and Japanese programmes are He bonded; the US programme will test sodium bonded fuels. The initial results of these tests will provide the first scoping data on inert matrix nitride fuels, from which the fuel design can be improved for potential application in fast spectrum systems.

4.1.3. Metal alloy fuels

Metal alloy fuels are unique to application in liquid metal cooled fast spectrum systems. The high swelling behaviour of metallic fuels is taken into consideration by a fuel design, which allows for a certain amount of free swelling to occur. Fabricating the fuel with sodium filled fuel-to-cladding gap accommodates swelling; the gap is sized so that the fuel occupies approximately 75% of the internal cross-sectional area of the cladding at beginning of life. The sodium-filled gap minimizes the temperature rise across the gap and limits fuel centerline temperatures. Typically, fission gas-driven swelling of the alloy results in a volume increase of approximately 30% within the first 2% of (HM) burnup, and causes the fuel to contact the cladding wall. At this point however, the fuel has formed a porous mass with a high creep rate, and exerts almost no stress on the cladding. High fission gas release leads pin pressurization, which drives cladding creep. This is controlled by appropriately sizing the gas plenum region of the fuel pin. At 18–20 at.% HM burnup, solid fission product swelling of the fuel can lead to fuel/cladding mechanical interaction. Metal fuels have been experimentally tested to burnups of approximately 200 GW·d/t, see for example [201].

Recent work in this area has focused on fabrication and characterization of metal alloy fuels in the Pu-Zr alloy system [202]. The Pu-Zr phase equilibrium diagram is well characterized [203]. The addition of zirconium as an alloying element limits fuel cladding chemical interaction and raises the melting temperature. Compositions of interest are generally within the face-centered cubic δ -phase field, which has an upper bound of zirconium content at approximately 40 wt%. The lower bound on plutonium content is set by margin to melting requirements. Earlier work established the acceptable low-burnup behaviour of Pu-Zr alloys [204].

Metallic fuels are of interest primarily for closed fuel cycle fast spectrum systems when coupled to an electrometallurgical fuel recycle process due to the potential for a compact integral reactor/recycle plant [205]. Although metal fuel technology is well established, inert matrix metallic fuel concepts have not been proven through a programme of extensive irradiation testing.

4.2. Heterogeneous fuels

Heterogeneous fuels consist of a distribution of discrete fuel particles embedded in a metal or ceramic matrix. In the ideal case, the matrix remains largely unaffected by neutron, fission fragment, and α -particle damage from the fission events that take place in the fuel particles. Only those fissions taking place close enough to the surface of the particle to allow fission fragments and α -particles to get recoiled into the matrix contribute to matrix damage. Figure 8 is a schematic of the irradiation damage processes occurring in heterogeneous dispersion.

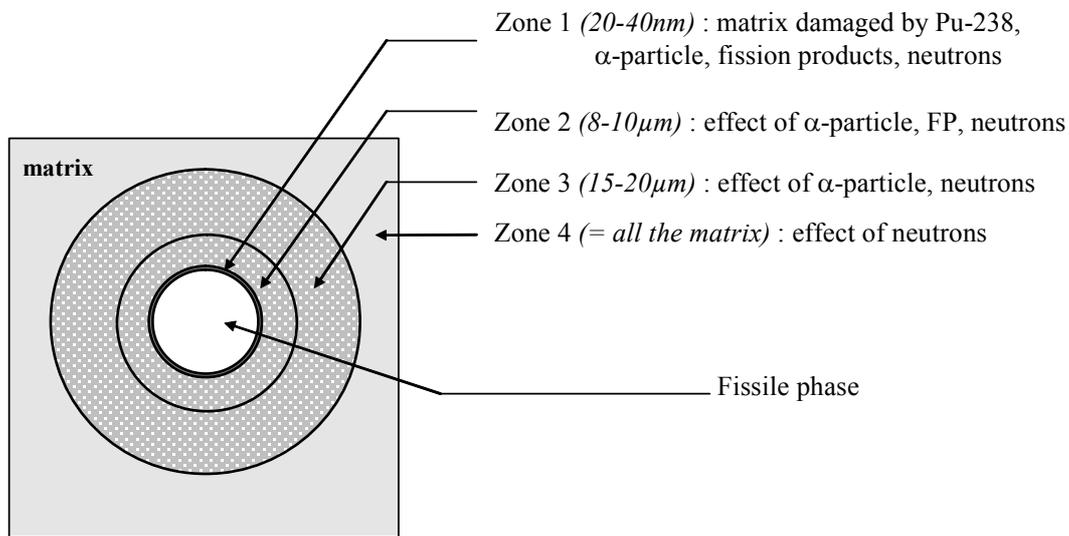


Fig. 8. Schematic presentation of the radiation damage in a dispersed IMF.

In the case of an ideal dispersion microstructure consisting of a uniform distribution of spherical particles of one size, the fraction of undamaged matrix [206] is given by equation (1):

$$V_m = 1 - \frac{V_f}{1 - V_f} \left\{ \left(1 + \frac{2\lambda_m}{D} \right)^3 - 1 \right\} \quad (1)$$

where V_m is the volume fraction of undamaged matrix, r is the fuel particle radius, V_f is the fuel particle volume fraction, D is spherical particle diameter, and λ_m is the fission fragment range in the matrix. The dependence of V_m on volume loading of fuel and the ratio of fission fragment range to particle diameter (λ_m/D) could be described qualitatively for small values of (λ_m/D) using the equation 1. The matrix must also not chemically react with the fuel particles during irradiation.

Fuel centerline temperature for a specific power density and coolant conditions is governed by the thermal conductivity of the dispersed fuel core. Thermal conductivity can be estimated using a variety of correlations, many based on variants of the Maxwell equation. For the case of dispersions with low volume fraction of fuel, the Maxwell-Eucken equation [207, 208],

$$K_f = K_m \frac{2K_m + K_p - 2V_p(K_m - K_p)}{2K_m + K_p + V_p(K_m - K_p)} \quad (2)$$

generally provides an adequate description of a two phase system if there is good interface between the matrix and particles. Here K_f is fuel bulk conductivity, K_m and K_p are matrix and particle thermal conductivity, and V_p is again the volume fraction of particulate loading in the case of fresh fuel.

4.2.1. *Cercer dispersion fuels*

Cercer fuels incorporate a macroscopic or microscopic dispersion of the fissile phase in a ceramic matrix. Typically, cercer fuels are made into pellet form and encased in standard cladding with a pellet to cladding gap. The primary advantages of cercer fuel are the ability to partially capitalize on the existing understanding of clad pellet-type fuel behaviour coupled with the potential for an additional barrier to fission gas release, a lower fuel centerline temperature due to localization of fission fragment damage, which also introduces the potential for higher burnup.

Cercer fuel performance depends heavily on the microstructural characteristics of the fuel. Two primary strategies have been pursued for optimizing fuel behaviour through manipulation of microstructural variables. Macrodispersions attempt to maintain the thermal conductivity and structural integrity of the matrix during irradiation through the use of relatively large and widely distributed fissile phase particles. Microdispersions incorporate a fine dispersion of the fissile phase into a neutronically inert matrix. This is often done as a convenience for fabrication. For example, a porous ceramic body can be infiltrated with a solution of fissile material [93]. The pellet is then dried and calcined to convert the solution to a solid phase. The pellet may then be sintered to achieve the desired density. In the case of a microdispersion, fission related damage occurs over a larger fraction of the matrix volume relative to a macrodispersion.

A wide variety of choices for the ceramic matrix phase are available including oxide, carbide, and nitride ceramics as binary, ternary, and higher compounds. An example of this concept would be a dispersion of PuO_2 particles of size on the order of $100 \mu\text{m}$ in MgO . Burnable poisons for reactivity control in thermal spectrum systems can be included either in solution with the matrix phase, as a separate particulate phase, or in solution with the fuel particle.

Recent work has focused on determining the viability of cercer concepts for plutonium burning through fabrication studies [209, 210], out-of-pile characterization, and irradiation

testing of materials that employ uranium as a fissile surrogate for plutonium [24, 211]. More details are presented in the Russian country profile in the Section 3.4. Fabrication and neutronic studies of SiC microdispersions using surrogates have also been conducted [64, 62]. More details are presented in the Canadian country profile (Section 3.1). These materials are of interest due to the known good dimensional stability of SiC under irradiation and high thermal conductivity relative to oxides.

The results of a few recent experimental irradiation tests are available. The MATINA experiment [55] was designed to investigate the behaviour of candidate inert matrix materials (MgO, MgAl₂O₄, Y₃Al₅O₁₂, Al₂O₃, Nb, Cr, V, W and TiN) and dispersions of uranium oxide (UO₂) in magnesia (MgO), alumina (Al₂O₃), and magnesium aluminate spinel (MgAl₂O₄) under fast neutron irradiation in the Phénix reactor. The experiment was stopped at low exposure due to an extended reactor shutdown. In general, all inert materials except Al₂O₃ performed acceptably at a fast neutron fluence ($E > 0.1$ MeV) of $1.95 \times 10^{26} \text{ m}^{-2}$. Al₂O₃ showed volume swelling on the order of 4%. The uranium oxide cercer dispersions were irradiated to approximately 1.3% burnup at a maximum linear power of 4.3 kW/m. A low temperature thermal spectrum experiment (THERMHET) [99, 212] using similar fuels showed markedly different behaviour for micro- and macrodispersions of UO₂ in MgO. In this experiment, the microdispersion showed much greater swelling and restructuring than the macrodispersed specimens. The macrodispersions showed some matrix cracking. Comparison of the differences in behaviour of similar spinel-based fuels in the MATINA and THERMHET experiments showed that the behaviour of certain ceramic matrix materials, spinel in particular, can vary widely with irradiation temperature. Also included in the THERMHET test was the so-called ‘jingle’ variant of the macrodispersed concept, which incorporates free space between the fuel particle and matrix. This concept shows promise in reducing matrix fracture due to fuel/matrix mechanical interaction, although some matrix cracking was still observed.

MgO, MgAl₂O₄, Y₃Al₅O₁₂, Y₂O₃, and CeO₂ were irradiated in the EFTTRA-T3 experiment, with and without low volume loadings of UO₂ (2.5 vol.%) or (Y,U)O_x (19.6 vol.%) fissile particles [213]. All compositions were fabricated as macro-dispersions; in addition, a micro-dispersion of UO₂ in MgAl₂O₄ was irradiated. BaZrO₃ was irradiated without fissile inclusions. The specimens were irradiated at low power ratings (max. 6.3 kW/m) to burnups on the order of 17.2–19.8% FIMA. In general, it was found that the hybrid fuel concept ((Y,U)O_x dispersions) was beneficial in reducing fission gas release. The micro-dispersion of UO₂ in MgAl₂O₄ performed well, has been found to be unsuitable as a fuel in other experiments. Y₃Al₅O₁₂ was found to exhibit high swelling and extensive matrix cracking leading to high fission gas release (47%); BaZrO₃ pellets also broke apart on irradiation. MgO, CeO₂, and Y₂O₃ performed well, exhibiting low swelling and fission gas release in the range of 3–8%. The CeO₂ and Y₂O₃ fuel specimens showed less matrix cracking, indicating that the matrices may have the ability to deform plastically during irradiation to accommodate particle swelling.

Similar experiments comparing macro- and microdispersions of particles of (Zr,U)O₂ solid solution in MgAl₂O₄ and Al₂O₃ have been carried out as a part of the Japanese programme for development of ROX (rock-like) fuels. Irradiations in JRR-3 (Japan Research Reactor 3) [114] to approximately 10 GW·d/t indicate that swelling was greater for the microdispersions, and that fission gas release was greater for the macrodispersions due to matrix cracking. The MgAl₂O₄ matrix fuels swelled more and had higher fission gas release than the Al₂O₃ matrix fuels tested. MgAl₂O₄ matrix fuels also exhibited redistribution of MgO from regions in the pellet where the temperatures were calculated to exceed 1 700K.

The combination of low neutron absorption cross section, high thermal stability for the choice of a wide range of ceramic compounds, and similarity to well known pellet-type uranium and mixed oxide fuel make the cercher concept attractive. The response of many ceramic materials to neutrons or decay fission fragments, however, is a complex function of cercher microstructure, irradiation temperature and dose history. Ceramics are also generally brittle and care must be taken that thermal and mechanical stresses in the matrix do not exceed the elastic limit of the material, or matrix cracking may occur, negating a fission product barrier and one of the useful attributes of this fuel. Although some transient testing has been performed, further safety testing of specific fuels will be required prior to large-scale deployment in commercial reactors. These issues must be further understood before cercers can be demonstrated to be a viable fuel for plutonium burning. Cercers can thus be regarded as an attractive fuel concept in the early stages of development.

4.2.2. Coated particle dispersion fuels

Coated particle fuel technology is a subset of cercher fuel that has been uniquely applied to gas cooled thermal spectrum reactors. The deep burn capability of this fuel makes it attractive for once through cycles for plutonium destruction. Coated particle fuel consists of a fissile kernel contained within a multi-layered shell. Fission products are retained locally within the shell. Typically, as in a TRISO particle, the shell is composed of four layers. These consist of a low-density pyrocarbon ‘buffer’ layer that accommodates fission gas and fuel swelling, an inner pyrocarbon layer, a pyrolytic layer of silicon carbide, and an outer pyrocarbon layer. Oxides, carbides, and oxy-carbides have been used as kernels. Properly executed, TRISO-type fuels are quite robust and capable of very high burnup. Fuel performance is highly dependent on fabrication technique and the redox thermochemistry within the coated particle as a function of burnup, however [214]. Very high burnups of beyond 900 GW·d/t have been reported for a few particles of PuO_{2-x} TRISO-type fuel [215]. Uranium-based particles have been shown to withstand extended exposure to temperatures of 1600°C with failure rates on the order of a few in 10⁴. Recent work has focused on improving fuel particle fabrication techniques and on developing improved coating materials, particularly ZrC coatings [216,217] in place of SiC to extend the temperature range of this fuel and in an attempt to slow or prevent migration of fission products, particularly palladium [218] and silver, [219] into the reactor coolant. Novel Pu based kernels have been fabricated at ITU using an innovative fabrication route based on actinide infiltration of porous yttria stabilised zirconia kernels [220]. This process is very attractive for Pu or minor actinide applications. No radioactive liquid waste is produced. In the early 1970’s several successful tests were made on plutonium based fuel in the experimental HTR Peach Bottom in US and Dragon in UK [221] test reactors. Fuel burnups of 750 GW·d/t were demonstrated.

Coated particle fuel technology for gas-cooled reactor has been demonstrated on a large scale in several prototype and production gas reactors, and a good theoretical basis exists for the development of fuels designed for plutonium burning. The concept of plutonium or minor actinide incineration is often known as the “deep burn” concept [222]. The deep burner modular helium reactor (DB-MHR), was proposed by General Atomics (GA) to fit sustainability objectives e.g. incineration of fissile plutonium and reduction of waste toxicity after one hundred year cooling-time. When used to destroy transuranics waste, the DB-MHR three-ring active core contains two different kinds of fuels: The driver fuel (DF), consisting of the plutonium and neptunium discharged from LWRs. The transmutation fuel (TF), consisting of the minor actinides also discharged from LWRs, plus the transuranics left in the DF after a three-cycle irradiation. Fuels are packaged in TRISO micro particles that are assembled in ceramic (e.g. IMF) compacts, which are retained in graphite fuel elements. The DB-MHR has

a net thermal power outlet of 600 MW. The plutonium content for each re-loading ranges between 400 and 500 kg. The average operating temperature is 900°C. A negative temperature coefficient is granted by the graphite and a Doppler contribution from actinides. The prompt neutron lifetime is about 1 ms. After a complete irradiation, the residual mass of transuranics and the radiotoxicity of wastes depend on the transmutation ratio (TRR). TRR was originally evaluated at more than 80%. In the GA's double-strata approach, the remainder of the fuel was recycled in an ADS, to achieve an almost complete incineration of the actinide charge. Analyses, conducted jointly by FRAMATOME-ANP and GA, estimated TRR more precisely at $65\% \pm 5\%$. Recent works demonstrated that the TRR can be increased up to 70% for a critical and controlled operation. Present results allow adopting a TRR value of $70\% \pm 5\%$ as a design basis for an industrial DB-MHR. For the disposal of military Pu, a gas turbine modular helium reactor (GT-MHR) is now being designed and developed in Russia [223, 224]. The programme to develop coated particle fuel for disposal of excess Russian weapons plutonium is being carried out by Russian nuclear labs and industrial organizations with support from US specialists at General Atomics and Oak Ridge National Laboratory. Sol-gel process for manufacture of hypo-stoichiometric plutonium oxide ($O/Pu = 1.67-1.75$) particles $\sim 200 \mu\text{m}$ in diameter was investigated at VNIINM with use of experience of previous investigations on preparation of uranium dioxide micro-spheres [224]. PuO_{2-x} micro-spheres with the good characteristics were prepared in laboratory experiments: diameter 200 ± 25 micron, density $10.2-10.4 \text{ g/cm}^3$ [224].

4.2.3. Cermet dispersion fuels

The primary advantage of cermet dispersion fuels are robust behaviour. A large amount of work was performed on cermet fuels in the late 1950's and early 1960's as an alternative to oxide pellet fuel in fast and light water reactors. In application, the cladding is often metallurgically bonded to the matrix; the absence of a fuel to cladding gap dramatically improves fuel to coolant heat transfer and lowers fuel centerline temperature. A barrier is often required to prevent reaction of the matrix and the fuel particle. Aluminum matrix cermet fuels (list types including silicides, oxides) designed in this way remain the fuel of choice for research and test reactors, which operate at high power density (up to 500 W/cm^3 in the fuel) to high burnup and require benign behaviour in case of cladding breach. The high burnup capability of carefully fabricated cermet dispersions was demonstrated in the early 1960's, validating the general principals of cermet dispersion fuel behaviour, see for example [225]. Thurber summarized data on dispersions of UO_2 particles in stainless steel matrices available at about 1964 as a function of fuel surface temperature and fission density [226, 227]. It can be seen that fuel burnup lifetime is inversely proportional to fuel temperature. Although cermet fuels have demonstrated high-burnup capability, they were set aside as a candidate for commercial reactor fuel (as were many other fuels) in the mid-1960's possibly due to the more difficult fabrication and lower heavy metal density relative to pellet-type oxide fuels.

Plutonium oxide as the dispersed 'fuel' phase could be used with particle loading in the range of 6–12 vol.% [227] in a system utilizing a burnable poison to control the reactivity for plutonium burning applications. Burnable poisons (such as erbia and gadolinia) for reactivity control in thermal spectrum systems can be included as a separate particulate phase, or in solution with the fuel particle. Reactivity swing can also be somewhat compensated for by the use of a neutron absorbing matrix such as tungsten [228] or molybdenum [229]. The formulation of definite criterion to predict failure in these fuels remains elusive, due to the complex stress state and unknown material properties during irradiation, although early work included some attempts at quantifying failure [230]. Recent activity in this area includes

conceptual studies of the application of cermets to Pu-burning in LWRs [227], irradiation testing of aluminum matrix dispersions of UO_2 as fuel for LWRs [148], and the testing of molybdenum-based cermets under thermal spectrum conditions [229]. The irradiation of a 36 wt% UO_2/Mo cermet in the TANOX facility at SILOE showed that the fuel performed well to a burnup of 55.4 GW·d/t, and also maintained integrity during post-irradiation heating to 1 580°C. Corrosion behaviour of aluminum clad fuels in an LWR environment can be improved by the use of silumin (AlSi).

Although a significant database for fuel performance has been accumulated for steel and aluminum matrix fuels, cermets for Pu and MA burning are only in the concept phase at this time for use in commercial reactors. Questions remain in the area of core neutronic analysis and in-reactor mechanical behaviour of long rods. Safety testing including transient testing must be conducted prior to deployment in commercial reactors. If fuel recycle is to be considered, then the compatibility of the large amount of matrix metal with conventional recycle processes must be assessed.

4.2.4. Metmet dispersion fuels

There has been recent development of dispersions of fissile metal (U-Mo alloys) in an aluminum matrix for use as research reactor fuel [231–233]. Prior to this, there had been extensive testing of dispersions of so-called Pu-Al alloy fuels, actually a dispersion of PuAl_4 precipitates in aluminum due to the low solubility of Pu in aluminum. Hundreds of fuel specimens of this fuel type have been irradiated, some to burnups as high as 90% (plutonium atoms fissioned). In testing more than 1 500 zircaloy clad fuel pins were irradiated in the plutonium recycle test reactor (PRTR) at Hanford as a demonstration of the use of this fuel in a pressurized water environment. A summary of Pu-Al fuel irradiation data is given by Freshley [234].

Although promising results have been obtained, the application of this technology to modern commercial reactors is uncertain due to the low melting temperature of aluminum, and corrosion of the aluminum matrix by coolant would require extensive safety testing of this type of fuel. Corrosion behaviour can be improved by the use of silumin (AlSi). There are some considerations given to the advantages of using aluminum alloy dispersion fuel as IMF. These fuels have desirable attributes such as low fuel centre-line temperatures, viability for high burn ups, serviceability in transients and environmentally friendly process of fuel rod fabrication. At A.A. Bochvar Institute two main versions of IMF are under development, viz., having heterogeneous and isolated distribution of plutonium. Fuel elements with dioxide uranium composition fabricated at this institute are currently under MIR irradiation tests (RIAR, Dimitrovgrad). The fuel elements have reached burnup 88 MW·d/kg U (equivalent to the burn up of the standard uranium dioxide pelletized fuel) without loss of tightness by cladding. The feasibility is considered of fabricating IMF of the particular type with plutonium dioxide to be in-pile irradiated. Various versions low melting point zirconium brazing alloys are being investigated to improve IMF. Upon heating they melt down and due to capillary properties they penetrate fuel particle joints and ensure metallurgical contact between both cladding and fuel component and between fuel components themselves. These brazing alloys have melting temperatures of 690 to 860°C and are produced in the form of both granules and amorphous (metglass) strips. Zirconium brazing alloys used in IMF increases the thermal conductivity and allows a high-quality bond to fuel cladding, which will result in higher serviceability of fuels under transient conditions.

4.3. Fuel safety performance

4.3.1. Solid solution type fuel

Important safety studies have been made on the yttria stabilized zirconia solid solution (sol-sol) fuel. This type of fuel, as well as YSZ based ceramics-ceramics (cercer) type fuel such as YSZ-MgAl₂O₄ (spinel) fuel, has the significant safety problem of fuel temperature coefficient (FTC). The small FTC of the fuels causes very severe transient behaviours especially during reactivity initiated accident (RIA).

In order to confirm the RIA behaviour of YSZ based fuels, pulse irradiation experiments were performed at nuclear safety research reactor (NSRR) of JAERI, Japan by using UO₂-YSZ fuel test pins. The pulse irradiation of test fuel pin was carried out several times with different peak thermal energy increases in the test fuel pellets, and it was found that the YSZ sol-sol type IMF pin failure threshold in RIA condition is close to that of UO₂ pin, in terms of volumetric fuel enthalpy, at about 10 GJ/m³ [123]. Under RIA condition, therefore, the enthalpy increase of the YSZ sol-sol fuel should be comparable to that of conventional UO₂ fuel. For this purpose, from the analytical studies on Japanese rock-like oxide (ROX) fuel behaviour in RIA (Section 2.3), it seems sufficient to improve the FTC value of YSZ based fuel to about 2/3–3/4 of that of UO₂.

The small FTC of YSZ based fuels can be improved, for example, by adding poison or fertile material to the fuel. The international physics benchmark calculations (Section 3.2) generated information that is quite useful in understanding the effect of poison or fertile additive. In this benchmark calculation for the fuel material YSZ-Al₂O₃-MgO that did not include any poison nor fertile isotope, has shown the FTC is very small, especially at EOL. For the fuel material YSZ-Er₂O₃ with Er poison, although there are discrepancies between the results of benchmark participants, an improvement in the FTC is clearly observed. The same statement can be made for with fertile Th (fuel material ThO₂-Al₂O₃-MgO). It is also shown in the benchmark results that Er₂O₃ additive has larger FTC effect than ThO₂.

There are still uncertainties in the FTC values calculated in the IMF physics benchmark calculations. Because of the importance of the precise estimation of FTC of Pu inert matrix fuels, experimental data of FTC are very valuable. From this viewpoint, Doppler reactivity effect measurement of PuO₂ samples were carried out at the fast critical assembly BFS of IPPE, Obninsk Russian Federation (see Section 2.4). In this measurement, PuO₂ samples with three different Pu isotope compositions were prepared, and placed in the experimental channel at the center of the core BFS-91-1, where the neutron spectrum of the test zone is similar to that of IMF LWR.

4.3.2. Cercher type fuel

Most cercher fuels have the same basic FTC problem as YSZ based cercher type fuel i.e. low fuel temperature coefficient. Improvement of FTC is essential. In addition to FTC, the melting temperature of cercher type fuels tends to be lower than that of the component phases. The lower melting temperature may lead to a RIA behaviour different than that of sol-sol type fuels. In the NSRR experiment, pulse irradiation testing was also carried out for two-phase fuels of micro- and macro-dispersed UO₂-YSZ in spinel. The pulse irradiation test results have shown that the YSZ based cercher type fuel pin failure threshold energy is almost the same (10 GJ/m³) as that of UO₂ and YSZ sol-sol type fuels, even though the pin failure mechanism is completely different from UO₂. It is an important point that the same RIA safety limit can be applied for all UO₂, YSZ sol-sol and YSZ-spinel cercher fuels.

4.3.3. *Cermet type fuel*

The thermal conductivity of the ceramics-metal (cermet) fuel is high, and fuel temperature does not increase significantly both in the operating and accident conditions. [235, 236] The FTC problem is thus not so serious in the cermet type fuel case, yet it is important to study the accident behaviours of the cermet fuels. An RIA test on $\text{UO}_2 + \text{silumin (SiAl)}$ cermet fuel was performed by Russian researchers at the IGR impulse reactor in Kazakhstan [149]. The fuel enthalpy increase during a pulse was of 200 to 230 cal/g fuel. No fuel damage was observed and it can be concluded that the failure threshold of the cermet fuel pin in RIA condition is higher than this value.

From the safety point of view, the lower melting temperature of metals than ceramics is an important factor. Although the temperature of cermet fuel is expected to have a large margin to melting even under accident conditions, the behaviour of this composite fuel under accident condition is not yet well known. A preliminary analysis was therefore carried out in France on re-criticality of molten metal with ceramics fuel particles at the lower part of the vessel, in order to eliminate the serious risks even in the most severe calculation model [237].

5. CONCLUSION

The 400 reactors around the world produce about 100 tonnes of plutonium annually, in spent fuel and separated through reprocessing. There is worldwide interest in the investigation of IMF for scenarios involving stabilization or burn down of plutonium in the fleet of existing commercial power reactors. IMF offers the potential advantage for more efficient destruction of plutonium relative to MOX fuel. Greater efficiency in plutonium reduction results in greater flexibility in managing plutonium inventories and in developing strategies for disposition of MA. This approach is viable in existing cores that already utilize MOX fuel. The choice of matrix material may be manipulated either to facilitate fuel recycling or to make plutonium recovery extremely difficult.

Another important application of IMF is the destruction of minor actinides, with or without plutonium. So IMF can be used both to manage plutonium inventories and to address the long term radiotoxicity of the spent fuel by minor actinide destruction. In addition to plutonium/actinide management, some of the inert matrix fuels are having high thermal conductivity may have operational and safety benefits; lower fuel temperatures could be used to increase operating and safety margins, up rate reactor power, or a combination of both. Some of these IMF materials are also being considered for Gen IV reactors, because of their advanced performance, economics, safety features, sustainability, and application to waste minimization in a closed fuel cycle. Combinations of varied fuel compositions will help achieve a reliable, proliferation resistant LWR fuel cycle. Each proposed fuel composition could serve a specific purpose. For example, an inert matrix fuel composition with additions of Am and Np could be used for their destruction. In practice a limited number of inert matrix fuel pins could be distributed throughout the reactor core to maintain a homogeneous power profile.

IMF fabrication with optimized densities is carried out prior to characterization. In the case of ceramics, fabrication of IMF powder can be produced following either a wet or a dry preparation route. Several promising candidate materials have been identified for both fast and thermal reactors: MgO , ZrO_2 , SiC , Zr alloy, SiAl (silumin), ZrN ; some of these have

undergone test irradiations and PIE. Modelling of IMF fuel performance and safety analysis and tests has advanced. 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.

Neutronic modelling at the pellet, pin, and assembly levels is carried out prior to conceptual studies for application in commercial reactors. The components: fissile phase/burnable poison, and inert matrix components of the IMF are selected and their respective concentrations are optimized according to the neutronic characteristics. 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 tests are required. Some development is needed for analysis tools and computer programs. Irradiations in commercial reactors should be undertaken in a staged approach as soon as possible: segments in pins, full pins, and then assemblies.

Materials research, development and evaluation are presently a top priority in fortifying a case for burning plutonium and minor actinides using IMF. The research in the field is expected to continue until a creation that meets general criteria outlined above is developed and licensed for use in current commercial thermal reactor by a nuclear regulatory body. IMF can play an important role in the future of nuclear power.

IMF technology is believed to have a great potential to improve the efficiency of in-reactor disposal of plutonium, and provide opportunities for disposal of neptunium, americium and curium. Estimates have shown that if plutonium is used as a fissile phase, at least 90% of it will be destroyed [238].

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ABBREVIATIONS

ADS	Accelerator Driven System
AFC	Advanced Fuel Cycle
APA	Advanced Plutonium Assembly
APOLLO	Neutronic Code Name
ARWIF	Advanced Reactors with Innovative Fuels
BFS	Critical Assembly System, at IPPE
BN-800	Fast Reactor, 800 MW(e) Capacity (under construction in the Russian Federation)
BOL	Beginning of Live
BOR-60	Experimental Fast Reactor (in operation, thermal power 60 MW, Dimitrovgrad, RIAR, Russian Federation)
BOXER	Neutronic Code Name
BR-10	Research Reactor at IPPE
BWR	Boiling Water Reactor
CADRA	Consommation Accrue des Déchets dans les Réacteurs Rapides (English translation: Increased consumption of Waste in the Fast Reactors)
CAMIX	Composites of AMericiuM In phéniX (English translation: Composites of AMericiuM In phoenix)
CANDU	CANadian-Deuterium Uranium
CANFLEX	CANdu FLEXible programme
CAPRA	Consommation Accrue du Plutonium dans les réacteurs RAPides (English translation: Increased consumption of Plutonium in the Fast Reactors)
Cercer	Ceramic-ceramic Composite
Cermet	Ceramic-metal Composite
COCHIX	Concept Optimisé vis à vis des miCrosstructures en pHénIX (English translation: Optimized concept with respect to miCrosstructures in pHénIX)

CONFIRM	Collaboration on Nitride Fuel Irradiation and Modelling
DB-MHR	Deep-Burner - Modular Helium Reactor
DUPLEX	Heterogeneous Assembly
ECRIX	Expérience en Cœur pour tRansmutation en phénIX (English translation: Experiment in Core for transmutation into phénIX)
EdF	Electricité De France (English translation: Electricity of France)
EFPD	Energy Full Power Day
EFTTRA	Experimental Feasibility of Targets for Transmutation
ENEA	Ente per le Nueve tecnologie, l'Energia e l'Ambiente
EOL	End of Live
EPR	European Pressurized Reactor
ERMT	European Research Material for Transmutation
FCA	Fast Critical Assembly
FGR	Fission Gas Release
FIMA	Fission per Initial Metal Atom
FP	Fission Product
FPD	Full Power Days
FR	Fast Reactor
FTC	Fuel Temperature Coefficient
GA	General Atomics
GCR	Gas Cooled Reactor
GT-MHR	Gas Turbine-Modular Helium Reactor
HELIOS	Neutronic Code Name
HEU	Highly Enriched Uranium
HFR	High Flux Reactor, Petten, Netherlands
HFR	High Flux Reactor
HLLW	High Level Liquid Waste

HM	Heavy Metal
HTR	High Temperature Reactor
HWR	Heavy Water Reactor
IMF	Inert Matrix Fuel
ITU	Institute for TransUranium elements
JEF	Joint European File, data library
JRC	Joint Research Centre
JRR	Japanese Research Reactor
LEU	Low Enriched Uranium
LMFR	Liquid Metal Fast Reactor
LLFP	Long Live Fission Products
LOCA	Loss of Coolant Accident
LWR	Light Water Reactor
MA	Minor Actinide
MATINA	MATrices for INcineration of Actinides
Metmet	METal-METal composite
MIR	Materials Irradiation Reactor of pool/channels type, thermal power 100 MW (in operation, RIAR, Dimitrovgrad, Russian Federation)
MOX	Mixed OXide (Pu,U)O ₂
MTHM	Metric Ton Heavy Metal
NSRR	Nuclear Safety Research Reactor
OTTO	Once-Through-Then-Out
P&T	Partitioning and Transmutation
Pcm	Per Cent Mil
PIE	Post Irradiation Examination
POLIMI	POLItecnico di Milano, Italy, (English Translation: Polytechnic of Milan)
PROFIT	Plutonium Rock like Fuel Integrated R&D Team

PUREX	Plutonium Uranium Extraction
PWR	Pressurised Water Reactor
RETRAN	Neutronic Code Name
RFSP	Reactor Fuel Simulation Program for PWR
RG-Pu	Reactor Grade Plutonium
RIA	Reactivity Induced Accident
ROX	Rock-like Oxide Fuel
SILOE	Research Reactor, CEA-Grenoble, France
TANDEM	Accelerator Name
TANOX	Irradiation System in SILOE
TD	Theoretical Density
THERMHET	THERmique des HETérogènes, irradiation name
TRIPOLI	Neutronic Code Name
TRISO	TRiple ISOtropic coated particle
TRR	TRansmutation Ratio
TRU	TRansUranium element
WG-Pu	Weapon Grade Plutonium
WIMS	Neutronic Code Name
WWER	Water cooled Water moderated Energy Reactor
YSZ	Yttria Stabilised Zirconia

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