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STATUS OF MINOR ACTINIDE FUEL DEVELOPMENT

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STATUS OF MINOR ACTINIDE FUEL DEVELOPMENT

INTERNATIONAL ATOMIC ENERGY AGENCY
VIENNA, 2009

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FOREWORD

Following the ‘Atoms for Peace’ initiative in the 1950s, the technical feasibility and economic viability of nuclear energy were established in the 1960s, and rapid commercial deployment of nuclear energy ensued around the world. Today, there are 439 reactors worldwide with a total generating capacity of 372 GWe. In the late 1960s and early 1970s, the nuclear energy growth rate was expected to rise rapidly and the adequacy of uranium resources became a growing concern. As a result, the development of fast breeder reactors became a priority in France, India, Japan, the Russian Federation, the United Kingdom and the United States of America, as well as in other countries.

Sustainable development of nuclear energy calls for the development of innovative nuclear energy systems with highly efficient utilization of fissile and fertile resources. However, today’s commercial thermal reactors utilize less than 1% of total uranium resources. Hence, developmental efforts in recycling plutonium as well as minor actinides in current and future innovative nuclear energy systems are under way in several Member States. Additionally, plutonium recycling in LWRs has been pursued as an interim step toward an ultimate closed fuel cycle within fast breeder reactors. Furthermore, during the past two decades or so, new R&D programmes have been launched in several Member States to develop alternate reprocessing techniques and advanced partitioning processes that might aid in nuclear waste management. Current reprocessing techniques only recover uranium and plutonium. Advanced processes aim to recover minor actinides and other long lived fission products for the purpose of transmuting them in reactors.

The IAEA has been closely involved for many years in fostering collaborative R&D efforts and information exchange among Member States in development of innovative nuclear energy systems, including innovative nuclear fuel cycles approaches. The purpose of this report is to summarize the current status of minor actinide containing fuel types under development, fabrication processes for such fuels, pertinent thermal and physical properties, and irradiation test results. More importantly, these R&D programmes will be put into the context of appropriate fuel cycle options for actinide recycling and their implications on the future expansion of nuclear energy.

The contributions of the experts who helped draft and review this report, are greatly appreciated. In particular, the IAEA expresses its gratitude to M. Mayer (USA) for chairing the review group and for his significant contribution in the preparation of this report. Thanks are also extended to Y.I. Chang of the Argonne National Laboratory (USA) for a critical review of the draft and final editing.

The IAEA officer responsible for this publication was H.P. Nawada of the Division of Nuclear Fuel Cycle and Waste Technology.

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1. INTRODUCTION

Concerns about environmental preservation have increased the demand for more efficient management, and environmentally sound and sustainable development of nuclear energy [1–4]. Appropriate management of radioactive waste arising at the back end of the fuel cycle is considered to be a crucial issue of long term environmental concern in relation to the nuclear fuel cycle. There are currently 439 reactors with a total generating capacity of 372 GW(e) worldwide [5]. A further 34 units with a generating capacity of 28 GW(e) were under construction as of June 2008. A typical 1000 MW(e) light water reactor (LWR) generates about 20–30 t of spent fuel per year. The basis of environmental concern lies in whether the harmful components of this waste can be isolated from the biosphere for at least tens of thousands of years or even longer.

Most of the spent fuel mass (generally more than 98.5%) from commercial LWRs, is composed of uranium and short lived fission products, which do not pose a long term radiological hazard. Approximately 0.4 wt% of spent fuel mass is in the form of long lived fission products cesium, strontium, technetium, and iodine. Although there is some debate on the appropriate disposition path for these long lived fission products, this topic is reserved for a separate document. Of particular concern — due to long term radiotoxicity and heat load issues, as well as proliferation risk — is the approximately 1 wt% of spent fuel composed of plutonium and minor actinide isotopes. An estimate of the current and future inventory of minor actinides is shown in Fig. 1 [6]. As of the year 2006, it is estimated that about 110 tonnes of minor actinides are being contained in spent fuel storage worldwide, and an additional 40 tonnes are contained in high level waste products from reprocessing. In the absence of partitioning and transmutation, the amount will double by the year 2020.

Currently all of these materials are destined for underground storage or permanent disposal in repositories. However, the idea of developing innovative technologies for implementation in various nuclear fuel cycle schemes is receiving increasing attention in several Member States, because it could minimize radioactive waste destined for repositories. In addition, these technologies can increase the operating safety of nuclear energy systems, enhance proliferation resistance of the nuclear fuel cycle, increase the efficiency of natural resource utilization, decrease required repository space, and potentially increase the economic competitiveness of nuclear power plants.

In this context, many Member States and international organizations are working on different fuel cycle approaches that could effectively incorporate actinide recycling to reduce inventories of plutonium and minor actinides. Recognizing the potential role of actinide recycling in future nuclear fuel cycle systems, the IAEA has been coordinating studies on actinide recycling since 1980 to assist interested Member States. Recent developments in reprocessing and fuel technology, along with advances in reactor design and robotics, have led to an increased interest in actinide recycling as a key element in future nuclear energy systems.

Actinide recycling, including minor actinide processing technologies, has been the object of a large number of studies investigating its potential for waste minimization. National and international collaborative efforts over the last decade have clarified most scientific issues and associated challenges. For example, partitioning and

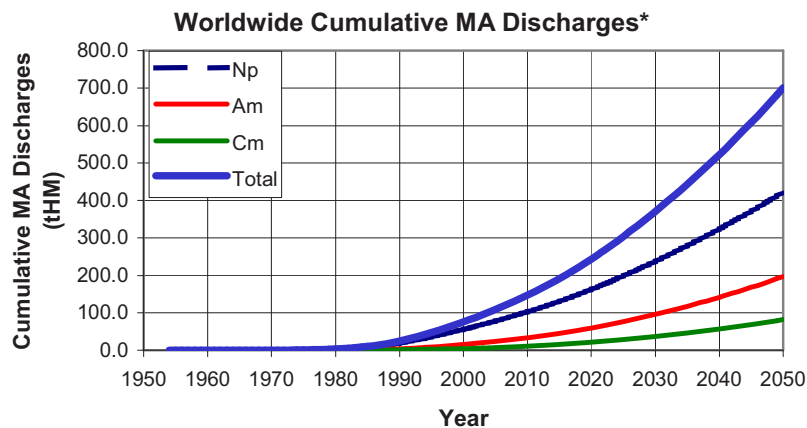


FIG. 1. Estimated inventory of minor actinides worldwide [6].

transmutation is one of the most notable research areas of EURATOM Fifth (1998–2002), EURATOM Sixth (2002–2006) and the ongoing EURATOM Seventh (2007–2011) Framework Programmes. Major national and international studies on partitioning and transmutation of minor actinides and long lived fission products generated by thermal reactors have been ongoing under different auspices, such as the SPIN Programme by Commissariat à l’Énergie Atomique (CEA) of France, the OMEGA Programme by Japan’s Atomic Energy Agency (JAEA) and Central Research Institute of Electric Power Industry (CRIEPI), the DOVITA Programme by the Research Institute of Atomic Reactors (RIAR) of the Russian Federation, and the Advanced Fuel Cycle Initiative (AFCI) Programme by the United States Department of Energy (DOE). Member States are evaluating several fuel cycle schemes to incorporate a partitioning and transmutation approach in both existing and future fuel cycles [7–10].

Despite their relatively small mass in spent fuel, transuranic elements such as plutonium, neptunium, americium, and curium, are the primary contributors to long term radiotoxicity and long term heat generation in spent fuel. Figure 2 shows that after an approximately 80 year cooling time, the heat generation rate in LWR spent fuel irradiated to 50 GWd/t is dominated by actinides, in particular ^{241}Am . The heat generation rate remains above 100 watts per tonne of heavy metal for approximately 900 years after discharge. This high, long term heat load will likely limit repository loading for scenarios planned in many countries due to potentially undesirable microstructural and chemical changes in drift wall materials. Thus, removing actinides from the waste stream could potentially increase repository space utilization by a large factor [11].

Figure 3 shows the impact of removing plutonium and minor actinides on radiotoxicity through ingestion when the long term environmental behaviour of these radionuclides is included in assessing potential dose. It can be seen that the radiotoxicity of spent fuel remains high for more than hundreds of thousands of years if left in the as-discharged form. When plutonium and minor actinides are removed and destroyed, radiotoxicity falls to a level below that of natural uranium ore within approximately 500 years. In addition, removal of plutonium would ease repository safeguards requirements. The removal of long lived radionuclides would also mitigate the effects of inadvertent intrusion scenarios.

Because of these potential waste management benefits, minor actinide partitioning and transmutation have been proposed for a variety of reactor and fuel cycle options. This document presents an overview of actinide recycling options in current and future reactor and fuel cycle systems, and the current status of minor actinide containing fuel development for use by Member States. The latest paths being pursued in the area of minor actinide fuel development — measurement of properties, processing, fabrication, and irradiation behaviour — are addressed here. Back end fuel cycle issues are also generically covered.

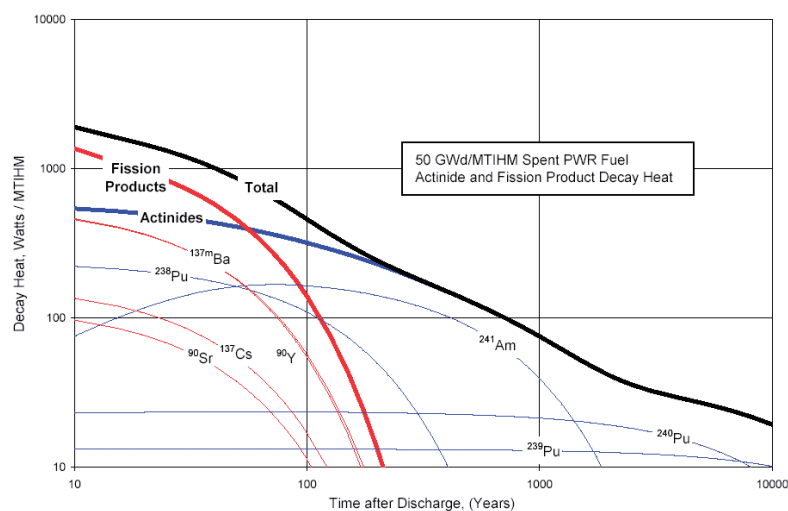


FIG. 2. Heat generation rate in LWR spent fuel irradiated to 50 GW·d/t [11].

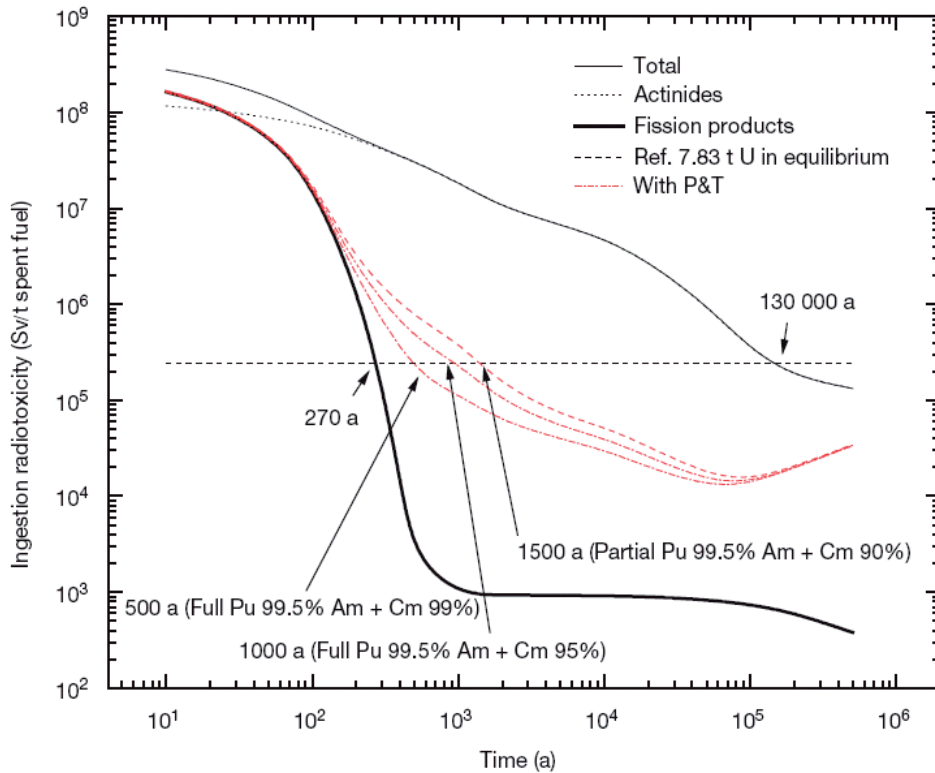


FIG. 3. The impact of removing transuranics on ingestion radiotoxicity of spent fuel [10, 12].

2. REACTOR AND FUEL CYCLE OPTIONS FOR ACTINIDE RECYCLING

In order to accrue the benefits illustrated in Fig. 3, essentially all actinides have to be extracted before disposal. In current commercial reprocessing, only uranium and plutonium are recovered, and minor actinides are discarded in the waste stream. As shown in Fig. 3, plutonium recycling alone has a rather insignificant impact on long term radiotoxicity. The first step in actinide recycling, then, is to recover minor actinides from the waste stream of current reprocessing operations. This process is commonly referred to as partitioning.

The next step is to incinerate recovered actinides, which is commonly referred to as transmutation. Transmutation occurs only when an actinide isotope fissions, resulting in short lived fission products. A simple neutron capture in an actinide isotope results in a higher mass actinide. The probability of fission depends on nuclear cross-section properties, which is a strong function of neutron spectrum. The fission probability per neutron absorption in various actinide isotopes is presented in Fig. 4 for thermal and fast spectra [13]. As an example, for ^{239}Pu , the probability of fission is 0.65 in thermal spectrum and 0.87 in fast spectrum. In general, fissile isotopes, such as ^{239}Pu and ^{241}Pu , have high fission probabilities but fertile isotopes, such as ^{240}Pu and ^{242}Pu , have very low fission probabilities in thermal spectrum. On the other hand, all actinide isotopes have reasonable fission probabilities in fast spectrum.

This drastic difference in fission probability also translates into actinides being very poor fuel in thermal spectrum but very good fuel in fast spectrum. This is one of the reasons most thermal reactors utilize a once through cycle, such as that depicted in Fig. 5. Here uranium ore is mined and fabricated into fuel. After the fuel has produced energy approximating 50 GWd per tonne of heavy metal, it is removed from the reactor and sent to permanent storage in a repository without any separation of elements.

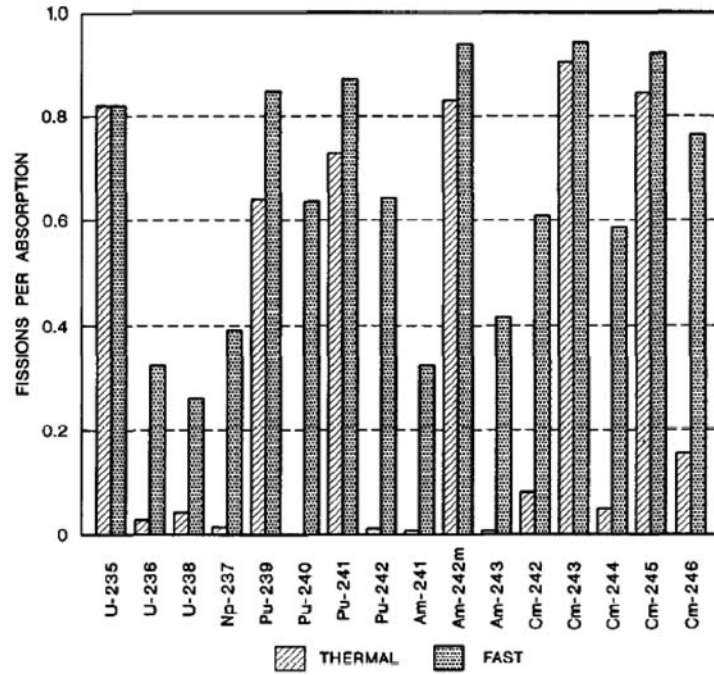


FIG. 4. Probability of fission per neutron absorbed in actinide isotopes for thermal and fast spectra [13].

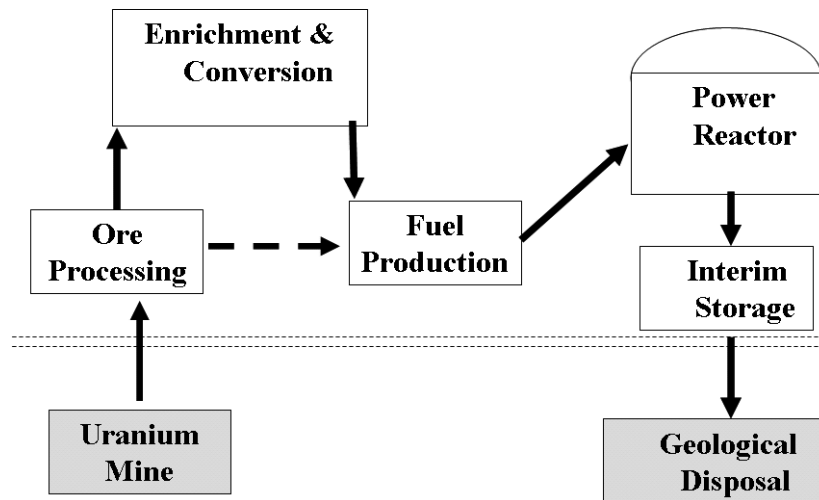


FIG. 5. Once-through fuel cycle.

The fissile contents remaining in spent fuel are envisioned to be recovered in the future if the combination of uranium resource availability and nuclear capacity growth mandates the introduction of fast reactors. In the meantime, there are large commercial reprocessing plants in operation today, such as La Hague, THORP, and Rokkasho, and some of the recovered plutonium is being recycled in thermal reactors. Thermal recycling is envisioned as an interim measure directed toward eventual full actinide recycling in fast reactors. This scenario is often referred to as double strata actinide recycling and is depicted in Fig. 6.

The first stratum is designated for current partial recycling of plutonium in thermal reactors. Second generation plutonium and other minor actinides are then transmuted in dedicated transmutation reactors; the second stratum. There are many variations of possible combinations of thermal and fast reactors (or subcritical systems), separation technologies, and material flows. There may also be additional requirements for some systems to accommodate disposition of excess weapons fissile materials as well as depleted and reprocessed uranium.

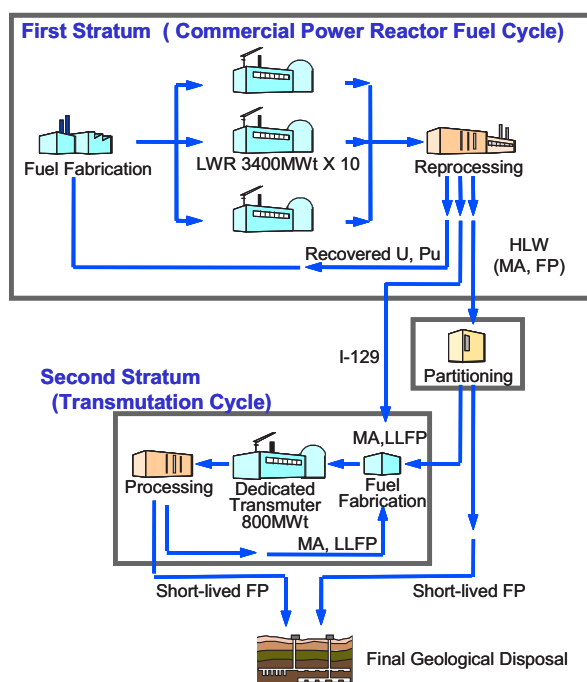


FIG. 6. Example of a double strata fuel cycle using commercial power reactors as the first stratum and a fast spectrum reactor to transmute plutonium and minor actinides [14].

Double strata actinide recycling relies on two different reactor systems to recycle different elements of actinides, in contrast to a single stratum system where a single reactor type is relied on to perform transmutation. Since the majority of installed reactors worldwide are based on water cooled thermal neutron systems, most evaluations of this approach are based on this system. Because of the large amount of fuel required for a single core loading, it is generally considered economically impractical to fabricate entire core loadings with a homogeneous distribution of minor actinides. To do so would require large, heavily shielded, remote fabrication facilities. Instead, specific elements are separated out from the bulk of used fuel and sent to a separate facility for refabrication into ‘targets’ with high minor actinide content. The targets represent a small fraction of total fuel loading, and do not impact the majority of fuel handling operations; this scheme is often referred to as ‘heterogeneous’ recycling.

It is also possible for single stratum to be based on a fast reactor system, which is often proposed to maximize homogeneous distribution of remotely fabricated minor actinides in fuel.

These strategies are all under active consideration by different Member States according to their national preferences [7–10, 15–21]. The fuels for transmutation of minor actinides in various reactor types have also been discussed over the last 10 years in several technical forums, including the ‘Information Exchange Meetings on Actinide and Fission Product Partitioning and Transmutation’⁷ (IEM P&T¹) of OECD/NEA, and ‘Advanced Reactors with Innovative Fuels’ (ARWIF²) as well as at workshops on inert matrix fuel³. Advanced fuel cycle aspects for incorporating actinide recycling have been discussed in several international working groups [7–10, 22–26].

¹ Information Exchange Meetings on Actinide and Fission Product Partitioning and Transmutation (IEM P&T) are held periodically by the Working Party on Scientific Issues of the Fuel Cycle (WPFC) of the OECD/NEA. For further details refer to the OECD/NEA’s web site <http://www.nea.fr/html/science/wpfc/index.html>

² Workshops on ‘Advanced Reactors with Innovative Fuels’ (ARWIF) are periodically conducted by the Working Party on Scientific Issues of Reactor Systems (WPRS) of the OECD/NEA. For further details on ARWIF workshops refer to <http://www.nea.fr/html/science/wprs/index.html>

³ Workshops on inert matrix fuels are normally conducted either in conjunction with ARWIF or as a special session of the annual spring meeting of the European Materials Research Society (E-MRS). Refer also to <http://www.emrs-strasbourg.com/>

The recycling of predominantly ‘aged spent fuel’ from LWRs (viz., more than 30 years old) has beneficial effects vis-à-vis recycling using fresh spent fuel [27, 28]. This scenario results in an altered transmutation pathway to produce lighter plutonium nuclides rather than heavy curium nuclides, owing to predominant transmutation of ^{241}Am grown-in by decay of ^{241}Pu (with a 14 year half-life) from aged fuels to produce lighter plutonium nuclides (^{238}Pu , ^{239}Pu , etc.) rather than heavy curium isotopes. In addition, the long aging period allows for significant decay of ^{244}Cm into ^{240}Pu [27].

The content and throughput of minor actinides in a transmutation system can be limited by reactor safety, chemical processing, or fuel performance issues. The general reactor safety problems accompanying addition of minor actinides to fuel are similar to those of plutonium based fuel, namely deterioration of neutronic characteristics of the reactor core, in particular the coolant void temperature coefficient, the Doppler coefficient, and delayed neutron fractioning. A potential fuel performance issue is associated with helium generation due to α -decays of ^{242}Cm , ^{244}Cm , and ^{238}Pu , which enhances fuel swelling and results in pin over-pressurisation during and after irradiation.

In evaluating various actinide recycling schemes, the net destruction rate of actinides is often used as a figure of merit, expressed in terms of kg/GW(e)·a, for example. Such a figure of merit leads to non-fertile target approaches or non-reactor options as an optimum strategy. Maximizing net destruction is synonymous to minimizing the conversion or breeding ratio, and hence it is counter-productive to the long term sustainability of nuclear energy. A higher conversion ratio improves uranium resource utilization. In fact, the LWR once-through cycle has a typical conversion ratio of 0.6 and about 40% of its energy is produced by fissioning of plutonium generated in situ. A higher conversion ratio also helps reduce excess reactivity requirements, which improves reactor safety by reducing the magnitude of reactivity insertion accidents.

Another misleading figure worth noting is the ratio of actinide generating reactors to actinide transmuting reactors. It creates the assumption that transmuting reactors and actinide recycling are not economic and that the goal is to reduce the number of such reactors.

The goal of actinide recycling should be to minimize the amount of actinides destined to permanent waste repositories, which has no direct relationship to maximum destruction rate. The question is how to best utilize actinides as fuel with minimum penalties in core performance, fuel performance, safety performance, and economics. Actinides cannot be simply stored or accumulated. If actinides are utilized as fuel in reactor and fuel cycle systems, not disposed of as waste, then maximum net production may become the figure of merit. This would be the case with a fast breeder reactor economy in the long term. In the near term there are many more actinides generated in current commercial reactors than can be utilized to start up fast reactors, thus initially fast reactors can operate as net burners of actinides. However, there is no reason to maximize the destruction rate as such.

2.1. ACTINIDE RECYCLING IN THERMAL REACTORS

2.1.1. LWRs

Since most commercial reactors are LWR and their spent fuels contain most of the actinides accumulated to date, actinide recycling in LWRs has been extensively evaluated [27–38]. Furthermore, there exists an extensive irradiation database on recycling of plutonium containing mixed oxide (MOX) in LWRs.

Current reprocessing processes only recover uranium and plutonium; just a limited amount of plutonium is recycled. The incentive to recycle MOX is marginal because its fabrication costs 5–6 times more than that of uranium fuel, offsetting uranium cost savings. The addition of minor actinide recycling would add additional cost penalties connected to partitioning of minor actinides from the waste stream and an even greater fabrication penalty than that for MOX fabrication. Thus, homogeneous recycling of minor actinides as part of MOX fuel is extremely difficult to justify on the basis of economics.

For this reason, separate minor actinide pins have been envisioned, sometimes referred as ‘target’ pins or ‘heterogeneous’ recycling. Actinides have large neutron absorption cross-sections and avoidance of self-shielding effects becomes important. Furthermore, pin to pin power distribution or power peaking factors also become important. Specific assembly design features and neutronic aspects have been the focus of many studies and evaluations of minor actinide recycling [29–33]. Some studies have identified that ^{241}Am can be most effectively transmuted in thermal spectrum, since neutron capture results in $^{242\text{m}}\text{Am}$ with a very large fission cross-section [29].

When actinides are recycled once in LWRs, their isotopic composition quickly evolves to higher actinides. Second generation discharge actinides have much less reactivity value than those of first generation discharge. In order to overcome this reactivity deficit, some innovative approaches have been proposed, in which reactivity deficit is made up by increased enrichment in uranium pins intermingled with continuous recycling of actinide pins in the same assembly [29]. For example, in the CORAIL concept proposed by the French CEA, the enrichment level of uranium oxide pins is increased gradually as mixed oxide pins loaded in the periphery of each PWR assembly are recycled continuously along with fresh actinides generated in the uranium oxide pins. Such self-contained actinide recycling is feasible from a neutronics point of view as long as fuel handling and fabrication challenges from increased neutron source and decay heat can be overcome and fuel irradiation performance is demonstrated.

Some safety parameter degradation may be mitigated by changes in the assembly lattice design to achieve satisfactory spectrum thermalization. This is accomplished either by an increase in moderator/fuel ratio in a high moderation PWR design [39], or by a decrease in fuel smeared density while keeping the same lattice geometry [40].

Minor actinide recycling in boiling water reactors (BWRs) has also been evaluated [41–43]. One option in BWRs is to take advantage of neutron spectrum hardening due to the increase in void fraction along the BWR axial direction. The void fraction varies from 0% to 40% along the axial direction. Since minor actinides' fission probability increases with hardening of spectrum, minor actinides can be loaded into the upper region of the core for more effective transmutation.

The common fuel form for actinide recycling in LWRs is MOX, with or without minor actinides, because of its proven irradiation performance. However, fertile free, inert matrix fuel has also been proposed only for the transmutation of minor actinides [20, 21, 44–47]. The principal motivation is to maximize actinide transmutation in LWRs without generating additional actinides in the actinide containing fuel in the process of transmuting existing inventory.

2.1.2. Heavy water reactors

Heavy water reactors, such as the Canada Deuterium Uranium (CANDU) reactor, have a superior neutron economy because of the absence of neutron absorbing hydrogen atoms. Thus, heavy water reactors can utilize natural uranium as their fuel, although discharge burnup is much less than that of LWRs. Their uranium resource utilization is better than that of LWRs and their discharge fuel contains much less fissile value, allowing heavy water reactors to operate with a once through cycle.

Although there are no plans to reprocess heavy water reactor spent fuels and to recycle actinides, there have been investigations to evaluate at least partial transmutation of actinides generated in LWRs using heavy water reactors [48–49]. This partial transmutation in heavy water reactors will improve fuel utilization and at the same time reduce the radiological toxicity inventory in storage.

The direct use of LWR spent fuel in CANDU reactors has also been proposed. In a DUPIC cycle, LWR spent fuel is mechanically refabricated into CANDU fuel for additional burnup [50–51]. The utilization of LWR spent fuel in conjunction with thorium has been evaluated for application in CANDU reactors [52].

2.1.3. Gas cooled thermal spectrum reactors

There are two general designs for thermal spectrum helium gas cooled reactors; the modular high temperature gas cooled reactor and the pebble bed reactor. Both reactor types use a basic fuel concept based on dispersion of coated particles in graphite. The modular high temperature gas cooled reactor uses massive prismatic graphite blocks containing cooling channels and is conventionally refuelled, while the pebble bed reactor uses a moving bed of spheres with a continuous on-line refuelling mechanism. The core is comprised of approximately 360 000 spherical fuel pebbles, each containing approximately 11 000 individual microspheres. The High Temperature Gas Cooled Reactor (HTGR) is also considered useful for incineration of Pu and MA, due to its unique and unsurpassed safety features, as well as to the attractive incentives offered by coated particle fuel. In the 6th Framework Programme of the European Commission (2006–2009), a project called 'PUMA' (Plutonium and Minor Actinides Management by Gas Cooled Reactors) was initiated and its objective is to provide additional key elements for the utilisation and transmutation of Pu and MA in current and future HTGR designs, thus contributing to the reduction of Pu and MA stockpiles [53].

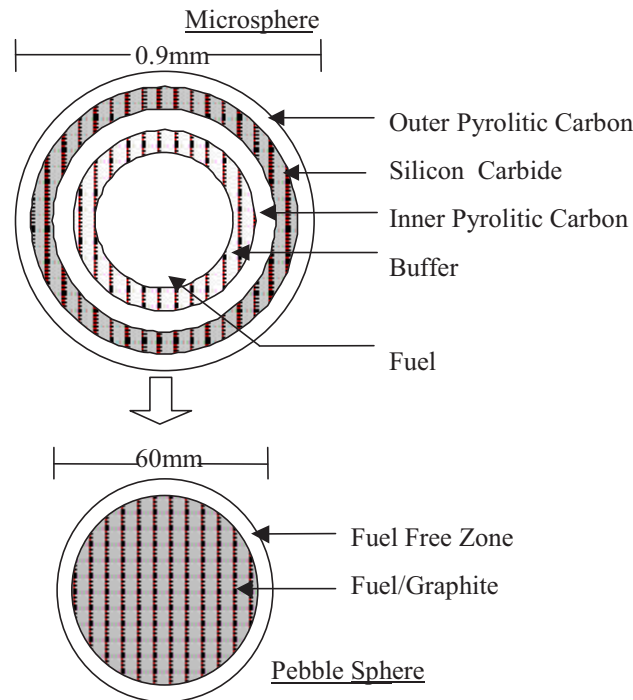


FIG. 7. Schematics of TRISO and pebble spheres.

The pebble bed reactor was first developed in Germany [54]. A 15 MW(e) experimental reactor, the AVR, operated from 1966 to 1988, and a full scale power station, the Thorium High Temperature Reactor (THTR-300), was also constructed. The South African electricity utility Eskom has been developing a 110 MW(e) pebble bed modular reactor. The performance of gas cooled, graphite moderated, high temperature pebble bed reactors has been evaluated for a wide range of operating conditions and fuel cycle options including the thorium cycle [55–59].

The prismatic high temperature gas cooled reactor was developed in the United States. The 40 MW(e) Peach Bottom-1 plant operated from 1967 to 1974 and the 330 MW(e) Fort St. Vrain plant operated from 1979 to 1989 [60]. The actinide burnup potential for the prismatic gas cooled reactor is similar to that of the pebble bed reactor cycle [61–64].

Both pebble bed and prismatic designs incorporate as a primary design feature coated micro fuel particles, commonly known as TRISO fuel particles, which consist of fissile material, surrounded by a coated ceramic layer of silicon carbide for structural integrity, and then by pyrolytic graphite. The detailed structure of the TRISO microsphere and overall pebble dimension are illustrated in Fig. 7. The silicon carbide layer of the microsphere is situated between protective layers of pyrolytic graphite. The buffer inside the inner pyrolytic graphite layer allows for expansion of gases generated in the fuel kernel. The innermost region of the microsphere consists of fuel, the design of which can be modified to include a minor actinide kernel.

Some development has taken place in the area of gas cooled thermal spectrum reactors for transmutation. Very high burnups of fissile material have also been obtained using TRISO coated plutonium mixed with carbon and in uranium and plutonium mixed with thorium.

TRISO coated uranium carbide and uranium oxycarbide fuels have been successfully irradiated to burnups exceeding 75% fission of initial metal atoms and highly enriched UC_2 ; they were licensed for high burnup service in the Fort St. Vrain reactor. Irradiation in the 1970s indicated that high burnup can also be achieved for TRISO coated plutonium fuel. Irradiation of TRISO coated plutonium fuels was carried out in the Peach Bottom-1 high temperature gas cooled reactor. These irradiations showed that essentially all fissionable plutonium could be fissioned, and that 65–75% of all plutonium isotopes could be burned.

The Peach Bottom-1 tests showed that for plutonium oxide fuel with a O/Pu atom ratio of 1.68, fuel behaviour was good (see Fig. 8). Coating failure by kernel migration was seen in high burnup particles with O/Pu=1.85, indicating that careful control of kernel composition is essential to achieve good performance. The TRISO coated oxide kernels containing 3Th/1Pu performed well under conditions in which essentially all fissionable plutonium

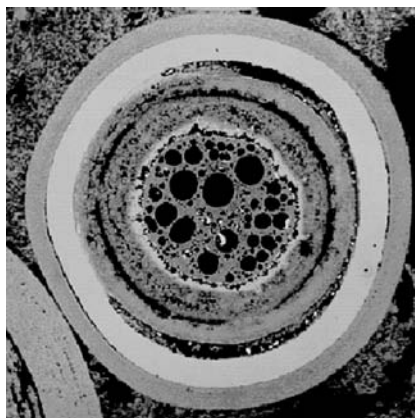


FIG. 8. TRISO coated $\text{PuO}_{1.68}$ after irradiation to $\sim 747\,000\text{ MW}\cdot\text{d/t}$ (95% ^{239}Pu fissioned) at temperatures of up to 1400°C .

was fissioned. Coating integrity of the fuel was not measured on-line in the Peach Bottom-1 experiments, and post-irradiation examinations were cursory, so the coating integrity of these samples could not be determined quantitatively. Data on the response of the coatings in high burnup plutonium fuel during accidents are not available.

2.2. FAST SPECTRUM REACTORS

Fast spectrum reactors require much higher fissile enrichment, typically in the 20–30% range, compared to 3–5% in thermal spectrum reactors. Fast spectrum reactors can operate as breeders or burners, or in a self-sufficient break even mode. Breeders incorporate external blankets, both axial and radial. When reflectors replace blankets, fast reactors become net burners of fissile material. If an appropriate amount of blanket is incorporated, then a self-sufficient mode can be maintained. In whichever mode they operate, fast reactor discharged fuel contains a large fraction of fissile inventory, and hence recycling is mandatory. As a matter of fact, resource utilization improvement is the primary rationale for fast reactors and recycling is required to achieve that goal.

Historically only uranium and plutonium have been recovered from LWR spent fuels, and only plutonium recycling has taken place or been envisioned for the fast reactor fuel cycle. However, minor actinides can be recycled along with plutonium in fast reactors. Minor actinides and even-mass isotopes of plutonium may not be attractive as fuel for thermal reactors because they have unfavourable ratios of fission to capture. These same materials, as well as odd-mass isotopes of plutonium, are fissionable in fast spectrum, where the fission to capture ratio is much more favourable, as illustrated in Fig. 4. Furthermore, the high content of recycled plutonium may require remote fabrication and hence minor actinides can be more easily incorporated into continuous recycling.

For economic and political reasons and because of proliferation concerns in some countries, fast reactors and their fuel cycle development programmes have been curtailed in many Member States since the 1990s, except in China, France, India, Japan and the Russian Federation. Fast reactor concepts for actinide transmutation have been of interest in recent international initiatives such as the International Project on Innovative Nuclear Reactors and Fuel Cycles (INPRO), Generation IV International Forum (GIF), and the Global Nuclear Energy Partnership (GNEP). Apart from these developments, the value of preserving the large technology base developed in Japan, France, Germany, the Russian Federation, the United Kingdom, and the USA, as well as information developed in other countries, has been a subject of interest for the Agency.

2.2.1. Sodium cooled fast reactors

Essentially all fast reactors which have been built and operated are sodium cooled, including: Experimental Breeder Reactor-II (62.5 MW·t/20 MW(e)), Fermi-1 (200 MW·t/61 MW(e)), and Fast Flux Test Facility (400 MW·t) in the USA; BR-5/10 (8 MW·t), BOR-60 (60 MW·t/12 MW(e)), BN-350 (1000 MW·t/150 MW(e)), and BN-600 (1470 MW·t/600 MW(e)) in the Russian Federation; Dounreay Fast Reactor (72 MW·t/15 MW(e)) and

Prototype Fast Reactor (600 MW·t/270 MW(e)) in the United Kingdom; Rapsodie (40 MW·t), Phenix (563 MW·t/250 MW(e)), and SuperPhenix (3000 MW·t/1240 MW(e)) in France; KNK-II (58 MW·t/21 MW(e)) in Germany; Joyo (140 MW·t) and Monju (714 MW·t/300 MW(e)) in Japan; and Fast Breeder Test Reactor (40 MW·t/13.5 MW(e)) in India [65].

Exceptions are sodium–potassium used in the first fast reactor, Experimental Breeder Reactor-I, and lead–bismuth used for submarine fast reactors in the Russian Federation. Of the reactors listed above, BOR-60, BN-600, Phenix, Joyo, Monju, and Fast Breeder Test Reactor are still in operation. New sodium fast reactor construction projects include the China Experimental Fast Reactor (65 MW·t/20 MW(e)) in China, the Prototype Fast Breeder Reactor (1250 MW·t/500 MW(e)) in India, and BN-800 (2100 MW·t/800 MW(e)) in the Russian Federation.

The early fast reactors — EBR-I, EBR-II, Fermi-1, and DFR — utilized metallic fuel because of its compatibility with sodium coolant, but all subsequent reactors have utilized oxide fuel, either uranium oxide or uranium–plutonium mixed oxide. Since the fast reactor fuel cycle relies on reprocessing and recycling of plutonium, the addition of minor actinide recycling should be relatively easy compared to thermal reactors. Fast reactor fuel cycles have been studied as a minor actinide transmutation scheme, either alone or in conjunction with LWRs [66–78]. Sodium cooled fast reactors can utilize either ceramic fuel types such as oxide, carbide, and nitride, or metallic fuel types.

The integral fast reactor (IFR) concept, which utilizes metallic fuel and pyroprocessing, is of particular interest because of its actinide recycling capability [79–82]. In electrorefining, which is the key processing step in pyroprocessing, all minor actinides are recovered together with plutonium. In fact, electrorefining is incapable of separating plutonium from the other minor actinides. Therefore, all actinides have to be recycled together. The injection casting fabrication technique is also a straightforward way to incorporate minor actinides in remote fabrication mode.

The French experimental irradiation programme was largely based on tests in fast neutron reactor Phénix, for which power restart was effective in late 2003 [66]. The flux conditions of Phénix allow for studies of irradiation damages of fuels and targets under representative conditions of fast and partly moderated flux, which are considered to be the most efficient for MA and long life fission product transmutation. The first irradiation experiments — MATINA 1 and 1A, and ECRIX H — complete since the restart of Phénix, are currently being examined, and preliminary post-irradiation examination results of these experiments about inert matrix behaviour under fast flux are presented in Section 6.

2.2.2. Gas cooled fast reactors

The gas cooled fast reactor is proposed as a Generation IV reactor concept combining the advantages of thermal spectrum, high temperature gas cooled reactors (such as efficient direct conversion with a gas turbine and the potential for application of high temperature processing heat) with the sustainability advantages that are possible with fast spectrum reactors, including the ability to fission all actinides and the potential for breeding.

Fast spectrum gas cooled reactors can be designed to operate at a much higher power density than thermal spectrum gas cooled systems. The system of TRISO fuel particles embedded in massive graphite blocks in thermal spectrum gas cooled reactors cannot be adapted to gas cooled fast reactor concepts due to the moderating power of graphite. The need for high fissile atom density, limitations on use of materials that are parasitic neutron absorbers in order to allow a high breeding ratio, and the requirement for fission product containment at high temperatures during loss of coolant events all combine to limit fuel types acceptable for use in gas cooled fast reactor applications.

Two categories of fuel have been proposed and experimentally explored: novel honeycomb fuel plates and dispersion fuels. Both of these fuel types are based on silicon carbide as an inert material. Initial screening studies have identified particle bed assemblies as a potential fuel type which meets set out goals, although the resolution of many technical issues remains uncertain. Pin type refractory ceramic fuel has also been considered. In gas cooled fast reactors, fuel response to core conditions following loss of coolant events is an overriding factor in the design of pin type fuels. This, coupled with core design constraints driven by plenum height restrictions, makes conventional sealed fuel pin design difficult. The issue is exacerbated by high helium gas production rates when americium is included in the fuel. Vented cladding tubes could aid in overcoming this issue.

Two primary factors are involved in selection of the fissile phase for gas cooled fast reactor fuels — core neutronics and irradiation performance. Core neutronics calculations indicate that oxide fuel is a poor performer

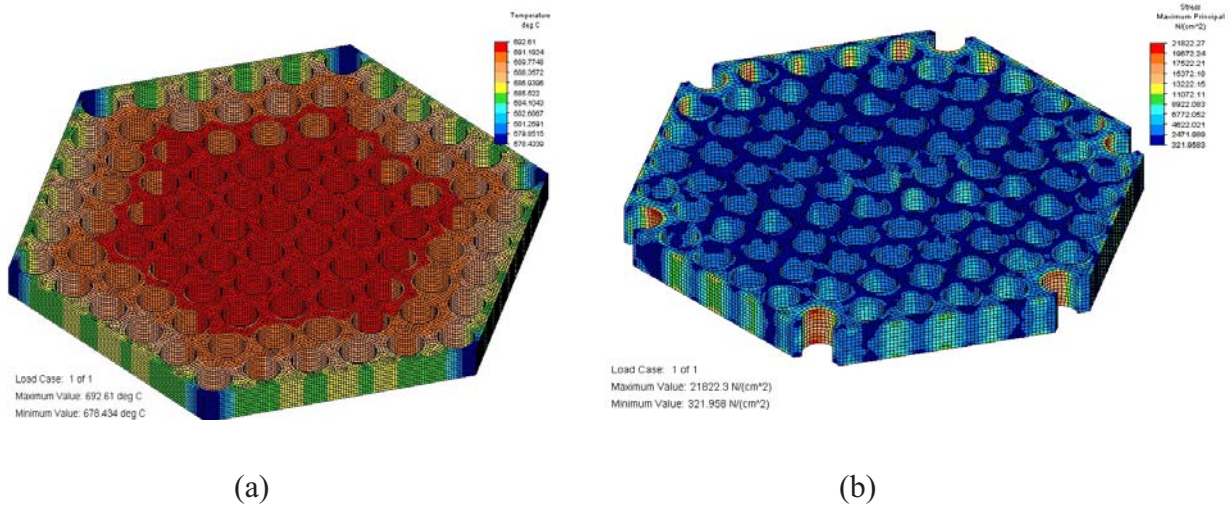


FIG. 9. (a) Temperature plot of ‘full’ hexagonal fuel element showing largest thermal gradients at the corners of the simulation model; (b) Stress plot of ‘full’ hexagonal element model with corners removed to reduce stress in these areas [83].

from the perspective of core neutronics relative to carbide and nitride fuels, due to low heavy metal density and spectral softening, because of the presence of oxygen [83–86]. Excluding oxide fuel, the primary choices of fissile phase become mixed carbide and nitride fuels. Both fuels have been demonstrated to perform well, to burnups in the order of 8–12% in sodium cooled fast reactor systems. There are no outstanding considerations related to fuel behaviour in the gas cooled fast reactor environment regarding separation of carbide and nitride fuels. At low burnups, mixed nitride fuel swelling and gas release tend to be lower than that of carbide fuel. As burnup increases, however, these differences in gas release and swelling behaviour disappear.

Since no data exists on the irradiation behaviour of proposed GFR fuels under GFR core operating conditions, ANL has made an assessment on the basis of scoping analysis of fuel concepts [83]. A combination of behaviour correlations and finite element analysis were used to make relative judgments about the impact of minor actinide loading on fuel burnup lifetime. Figure 9 displays an image of calculated temperature and stress distribution in a full hexagonal fuel element. The steepest temperature gradients, and hence the largest thermal stresses, occur at the six corners of the hexagon due to ‘overcooling’ of these areas relative to the rest of the fuel element. To eliminate these areas of high stress, a 60° wedge of material including the high stress area, was cut from the corners of the hexagon. Figure 4.8 shows that this reduced stress significantly in these locations, however the location of peak stress remains in the near surface region of the coolant channels nearest the six corners of the element [83].

In general, fuel development for gas cooled fast reactors is at an early stage, consisting of out of pile development of honeycomb and dispersion fuels in preparation for irradiation testing.

2.2.3. Lead cooled fast reactors

Lead is another liquid metal coolant option for fast reactors. The most developed lead cooled fast reactor concept is BREST, developed by the Research and Development Institute of Power Engineering (NIKIET) in the Russian Federation [87–88]. BREST was originally designed for 300 MW(e) but designs for 600 MW(e) and 1200 MW(e) have also been developed.

Because lead has a high melting temperature (372°C), coolant operating temperatures are somewhat higher than in sodium cooled fast reactors. In order to deal with higher operating temperatures, the BREST design incorporated nitride fuel. The current design does not have breeding blankets and the in-core breeding ratio is maintained at slightly above unity. The minor actinide transmutation capability of the lead cooled fast reactor is the same as for other fast reactor types.

2.3. ACCELERATOR DRIVEN SYSTEMS

Accelerator driven systems (ADSs) consist of two components: a proton accelerator and a subcritical reactor. High energy protons from the accelerator impinging on a Pb target produce spallation neutrons, which can be used for minor actinide transmutation. Since such spallation neutron sources are expensive to produce, the spallation source is surrounded by a subcritical assembly, where additional neutron multiplications occur. The subcritical assembly operates very close to criticality to maximize neutron multiplication.

Accelerator driven systems do not compete economically with critical reactors for electricity generation because a significant fraction of electricity produced is consumed to run the accelerator. Numerous studies have been performed on the application of ADSs for minor actinide transmutation [89–97], primarily in the context of a double strata scenario, in which the bulk of plutonium is recycled in commercial reactors and only small amounts of minor actinides are transmuted in dedicated second stratum systems (see Fig. 6).

In the transmutation fuel cycle (second stratum), nuclear fuel mainly consisting of minor actinides is used to enhance transmutation efficiency. A critical reactor would encounter difficulties in its safety and controllability aspects if a fuel containing only minor actinides would be used. Accelerator driven systems are perceived to have potential advantages in comparison with critical reactors: (1) various fuel compositions are acceptable, since the Doppler effect does not seriously affect system safety, and (2) small values of delayed neutron fraction may be acceptable because the margin to prompt criticality is maintained by subcriticality.

Within the EUROTRANS Integrated Project, Forschungszentrum Karlsruhe (FZK) and the Institute for Transuranium Elements (ITU) are joining together to study the behaviour of Mo-based cermet non-uranium fuel for ADS application. Contributions include core safety calculations, fuel property measurements, and irradiation experiments. Mo based cermet fuel pellets and pins loaded with Pu and Am were fabricated for irradiation programmes in Phenix (France) and HFR-Petten (the Netherlands). Thermal diffusivity and specific heat of the cermet fuels (loaded with Pu and Am) were the main properties measured [98-100].

2.4. MINOR ACTINIDE FUEL REQUIREMENTS

Some general requirements for minor actinide containing fuel are as follows:

- Fuel with high minor actinide content must be fabricated using remote processes. Many of the head end processes being considered to prepare LWR spent fuels for transmutation and the processes being considered for recycling of actinides into the transmuter would leave residual fission products in the fuel feed. Therefore, fuel fabrication would likely be conducted in a hot cell environment, demanding that fabrication processes be amenable to remote handling;
- The fuel must be compatible with fuel recycling processes. If actinides are to be recycled back into the transmuter, then some type of chemical or mechanical processing of spent fuel will be required;
- The fuel form must provide robust containment of fission products and radionuclides as a first barrier for safety during normal operations, that is, cladding failure with leakage of fission products must be a low probability event.

Another important consideration for all fuel types is volatilization of americium, leading to loss or redistribution during fabrication. The current status of fuel fabrication and fuel processing has been critically evaluated and the research and development needs of minor actinide containing fuels have been identified as below [19, 20, 101–103]:

- Development of fabrication processes and demonstration of fabricability of proposed fuel forms in a remote environment;
- Simple irradiation tests to screen samples of each fuel type for unexpected or poor performance;
- Determination of intrinsic properties or characteristics (e.g., out of pile interdiffusion behaviour of fuel and constituents and thermo-physical properties).

There are significant issues regarding development and deployment of remote fabrication and refabrication processes, which are required for the implementation of some systems for transmutation of minor actinides [104–109].

2.5. PARTITIONING PROCESSES

For actinide recycling to have any significant impact on reducing the long term radiological toxicity of high level nuclear waste streams, minor actinide recovery requires high efficiency — in the range of 99.9% [110]. In other words, the loss of minor actinides to the waste stream during reprocessing of spent fuel should be limited to about 0.1%.

A number of concepts for partitioning of minor actinides from spent fuel are based on solvent extraction processes [111–112]. PUREX is used to separate and recover uranium and plutonium. Fission products are separated from lanthanides and minor actinides using complexants, including CMPO, DIDPA, and TRPO. Separation of lanthanides from minor actinides can be achieved with CYANEX 301, TOPO, and BPT. Reprocessing of MOX spent fuel is difficult due to its high plutonium inventory and an increased level of ^{244}Cm [113]. The separation process can utilize batch, column, or counter flow methods. Variations in these separation processes exist, including targeted extraction for certain fission products and further separation of minor actinides. As part of the Advanced Fuel Cycle Initiative (AFCI) programme, the UREX+ process has also been developed and tested [114–116].

Pyroprocessing is also an available option [117–119]. Electrorefining is the key step in pyroprocessing, allowing valuable fuel constituents — uranium and actinides — to be recovered and fission products to be removed. An electrorefiner consists of an anode and a cathode submerged in molten electrolyte salt such as LiCl-KCl eutectic, which has a low melting point to allow operation at 500°C . Following disassembly of fuel assemblies, fuel pins are chopped into short lengths, loaded into perforated steel baskets, and introduced into the electrorefiner as anodes. When a direct current is passed at low voltage (typically of the order of one volt) from anode to cathode, the bulk of the current passes through electrolyte salt as uranium and actinide ions, which are reduced and collected at the cathode as metals. In other words, uranium and actinides are oxidized anodically in a molten salt electrolysis cell and deposited cathodically as solids. There are two different types of cathodes. The bulk of uranium is first collected on a solid mandrel cathode as a dendritic deposit. The remaining uranium and other actinides are collected on a second cathode, which is a ceramic crucible containing molten cadmium.

A hybrid approach is also feasible. Existing aqueous methods can recover uranium and plutonium from spent fuel and pyroprocessing can be applied to recover remaining minor actinides as a group.

Molten salt based reactor systems are a special case using on-line reprocessing. Several Member States are developing concepts for molten salt reactors, including MSTR (Molten Salt Transmutation Reactor), THORIMSNES (Thorium Molten Salt Nuclear Energy Synergetics) and AMSTER (Actinides Molten Salt TransmutER), to transmute actinides in which the fuel is in the form of molten fluorides [120–123]. The feasibility of using molten fluoride salts at high temperatures depends on the development of materials with long term resistance to corrosion.

2.6. OTHER CONSIDERATIONS

2.6.1. Thorium based fuel cycles

Some Member States are evaluating the thorium fuel cycle as an alternative disposition path for the incineration of plutonium and minor actinides in reactors and as a means for nuclear energy production with low radiotoxicity waste [124–128]. The use of uranium–plutonium mixed oxide (MOX) fuels results in production of additional second generation plutonium. A possible alternative approach is to incinerate plutonium in combination with thorium. The thorium cycle produces ^{233}U , which may offer some non-proliferation benefit relative to plutonium. First, it is contaminated with ^{232}U , which decays to highly active daughter products that provide some degree of ‘self protection’. Second, ^{233}U can easily be denatured through blending with ^{238}U . In the ^{232}Th – ^{233}U fuel cycle, a much smaller quantity of plutonium and long lived minor actinides (Np, Am, and Cm) are produced relative to the ^{238}U – ^{239}Pu fuel cycle, thereby minimizing radiotoxicity associated with spent fuel in the once through mode.

However, at the back end of the ^{232}Th – ^{233}U fuel cycle, there are other radionuclides such as ^{231}Pa , ^{229}Th and ^{230}U , which may have long term radiological impacts. R&D activities are being pursued at the laboratory scale on innovative thorium based hydride fuels for advanced plutonium and minor actinide burners with good safety characteristics [129].

A high degree of chemical stability and low solubility of thorium make irradiated thorium based fuels attractive as waste forms for direct geological disposal [130–131]. Multi-recycling and incineration of actinides in existing PWRs with an innovative concept based on thorium and fertile free fuel was analysed [132–133]. The assembly assumes a heterogeneous structure in which about 20% of UO_2 fuel pins on the assembly periphery are replaced with fertile free fuel pins containing actinides generated in the previous cycle.

2.6.2. Inert matrix fuels

Inert matrix fuel concepts have been proposed in order to minimize the number of minor actinide containing fuel pins and maximize the minor actinide transmutation rate, since no additional actinides are generated in minor actinide fuel pins. A number of inert matrices have been examined, including zirconia, zirconium nitride, and spinel. Both thermal and fast reactor systems are under investigation. The feasibility of such fuel cycles and the implications of increased neutron sources for minor actinide inert matrices have been discussed in the literature [20, 21, 46, 66, 134–136].

The CEA has devoted an extensive irradiation programme to the assessment of transmutation using minor actinide bearing inert support targets [66]. A first irradiation experiment was performed in the fast neutron reactor Phénix, in parallel to other experiments carried out in the HFR and Siloe reactors, in order to assess the behaviour of various materials under fast neutron flux intended as the inert support matrix for transmutation targets. This experiment, which included two steps — MATINA 1 (MATrices for INCineration of Actinides) and MATINA 1A — was completed in 2004 and underwent complete post-irradiation examinations (PIE); the results are discussed in Section 6. All pure inert materials showed satisfactory behaviour under fast neutrons except Al_2O_3 — which exhibits a swelling of close to 11 vol.% after irradiation. In the presence of UO_2 fissile particles, MgAl_2O_4 proved to be more stable in term of swelling as inert support than MgO and Al_2O_3 matrices, under the same irradiation conditions. A second experiment, ECRIX H (Experiment in Core for transmutation into phénIX) in Phénix involving composite pellets with an MgO matrix and AmO_{2-x} particles was completed in 2006. The initial PIE results on ECRIX H are described in Ref. 66. In light of these first experiments, a second phase dedicated to design optimization of the target was initiated and three new irradiation experiments — MATINA 2-3, CAMIX (Composites of AMericiuM In Phenix) and COCHIX (Optimized concept with respect to microstructures in phénIX) in Phénix and HELIOS in HFR — were started in 2006 and 2007.

Various programmes are running internationally on the use of plutonium in either inert matrix fuel or thorium based fuel [136–143]. Within the 6th Framework Programme EC project LWR-DEPUTY (destruction of plutonium in thermal systems), different types of Pu containing inert matrix fuels are being tested. Researchers at the Paul Scherrer Institute (PSI) launched the concept of a plutonium–erbium–zirconium oxide fuel. In this fuel, the zirconium oxide acts as the inert matrix carrier and erbium is added to reduce reactivity swing of the fuel. In Europe, three different irradiation tests were performed on zirconia based plutonium containing fuels [139]. Two irradiations (IFA-651 and IFA-652) were performed at the Halden Material Test Reactor, one focusing on yttria stabilized zirconia (YSZ) based IMF and one focussing on calcium stabilized zirconia (CSZ based IMF). The aim of these experiments is to study thermal behaviour, degradation with burnup, fission gas release and swelling under irradiation conditions similar to those in LWRs. One irradiation (OTTO: once through then out) was performed at the High Flux Reactor in Petten. Solid pellets of zirconia based Pu fuel, as well as different macro and micro dispersions of zirconia fuel spheres in a MgAl_2O_4 spinel matrix were tested. The aim of the experiments was to study swelling and integrity of the fuels and fission gas release after irradiation up to about 50–60% plutonium depletion. The result for the spinel-based fuels showed that micro-dispersed fuel underwent high swelling, which is not acceptable for commercial use. Macro dispersed, spinel based fuel showed good behaviour. Zirconia based fuels demonstrated very good behaviour under test irradiation conditions. At the beginning of irradiation densification was observed, which may be improved by optimizing the fabrication process. An Idaho National Laboratory study [141] on how to improve thermal conductivity of the zirconia based fuel was performed by adding MgO , and studying the resulting PuO_2 – ZrO_2 – MgO phase. Since neither PuO_2 nor ZrO_2 dissolves in MgO , the pure MgO phase provides a means to improve overall thermal conductivity of the fuel. Two main versions of IMF are under

development at the A.A. Bochvar Institute; heterogeneous or isolated distribution of plutonium [142]. Out of pile results on IMF loaded with uranium dioxide as plutonium simulator are presented. Fuel elements with uranium dioxide composition fabricated at the A.A. Bochvar Institute are currently undergoing MIR tests (RIAR, Dimitrovgrad). The fuel elements reached a burnup of 88 MW d kg^{-1} (equivalent to burnup of the standard uranium dioxide pelletized fuel) without loss of cladding leaktightness. The feasibility of fabricating IMF of these particular types with plutonium dioxide is considered with a view to in-pile irradiation.

Zirconium nitride is considered to be an inert matrix for burning plutonium in a closed nuclear fuel cycle [144–148]. Having fissile material diluted in a matrix softens the high fission density and consequently reduces maximum fuel temperatures that would be reached in pure PuN and PuO₂ in a fast neutron spectrum. ZrN is foreseen to be used as either a homogeneous or heterogeneous diluent for Pu, because of its chemical and thermodynamic compatibility with the actinide (and also with the liquid metal coolant). An additional advantage of nitrides is their good reprocessing behaviour (easy to dissolve in nitric acid). The characterization of thermophysical properties plays a key role in assessing performance and safety aspects related to the in-pile behaviour of nitride fuel systems. Recently significant experimental data concerning thermophysical properties of zirconium nitride were reported [144, 147]. Design and interpretation of experiments, along with characterization of both fresh and irradiated nitride fuels, have been resumed [148] at the Institute for Transuranium Elements (ITU), a joint research centre of the European Commission. Both physical and chemical phenomenological modelling and experimental activities are being included in this effort. The thermal transport properties of ZrN have been measured; the results obtained so far confirm or extend the range of data available in literature. Increasing trends with temperature have been observed, in line with predictions. These data constitute the first part of a larger programme of characterization; subsequently activities will be extended to nitride fuels and systems containing Pu and other actinides such as UN, (U_x,Pu_{1-x})N, (Zr_{0.8},Pu_{0.2})N, and irradiated nitride samples [144, 148].

3. MINOR ACTINIDE FUEL TYPES AND THEIR PERFORMANCE CHARACTERISTICS

The front end processing to incorporate minor actinides into fuels and the irradiation performance of minor actinide containing fuels are the main challenges for transmuting minor actinides. Development of minor actinide fuels (or targets) is a crucial step, since technologies for incorporating significant amounts of minor actinides into fuel are not well established. This section presents an overview of the current state of the art associated with minor actinide containing fuel types and their performance characteristics.

Several minor actinide fuel or target options are available for eventual application in nuclear transmutation systems. The choice of minor actinide fuels or targets depends on the reactor system as well as the specific application. This in turn hinges on nuclear infrastructure availability and future nuclear deployment scenarios. For example, in a multi-recycle scenario, the minor actinide burnup rate is not as important as the requirement that the fuel be easily reprocessed and refabricated into new fuel with a low minor actinide loss factor. On the other hand, a once through cycle for destruction of minor actinides 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 a long term storage facility. Thermal reactors and fast reactors have different coolants and therefore different coolant compatibility requirements. In all cases, the design of minor actinide fuel or target elements will depend on the reactor type, the primary options being conventional water cooled reactors, fast reactors, or a future dedicated system for burning minor actinides.

Minor actinide presence in fuel can necessitate the use of hot cells or hybrid laboratories, combining glovebox standards with remote operation by telemanipulators. To prevent the risk of incorporating highly toxic radioactive materials into tissue or the body while handling minor actinides (MAs), biological shielding is required. Lead (Pb) glasses provide shielding from γ emitters and a combination of water, lead and cadmium (or boron) provides shielding from neutrons emanating directly through radioactive decay (e.g. ²⁴⁴Cm) or by (α , n) reactions, which typically occur with light elements (O, N or C) in the fuel. Introduction of MAs is best undertaken in a dust free operation. For MA based fuels, processes devoid of dust production are not just desirable, they are essential.

TABLE 1. CLASSIFYING FUEL OR TARGET MATRICES ACCORDING TO THEIR CHEMICAL FORM

Matrix type	Inert matrix	Fertile matrix
Element	C, Mg, Al, Si, Cr, V, Zr, Mo, W	Th, U, Pu
Intermetallics	AlSi, AlZr, ZrSi	Th, U, Pu
Alloy	Stainless steel, zirconium alloys	Th, U, Pu
Carbide	11B ₄ C, SiC, TiC, ZrC	ThC ₂ , ThC, UC, PuC
Nitrides	AlN, TiN, ZrN, CeN,	ThN, UN, PuN
Oxide	BeO, MgO, Y ₂ O ₃ , ZrO ₂ , CeO ₂	ThO ₂ , UO ₂
Binary oxide	MgAl ₂ O ₄ , Y ₃ Al ₅ O ₁₂ , ZrSiO ₄	
Oxide solid solution	Y _y Zr _{1-y} O _{2-y/2} , Mg(1-x)Al(2+x)O(4-x)	(U,Th)O ₂ , (Pu,U)O ₂

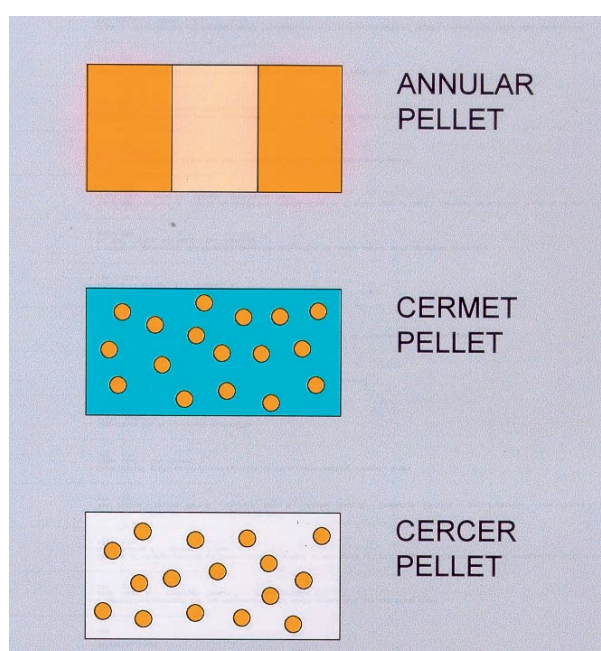


FIG. 10. Fuel and target pellet designs: annular pellets (homogeneous solid solution) and heterogeneous cermet or cercer composites.

Fuel matrix can be inert, fertile or fissile. Examples of fuel chemical forms are presented in Table 1. Fuel types can be divided into two major categories based on fuel microstructure. These categories are: (1) heterogeneous fuel, in which minor actinides are distributed in inert matrices separated from other fissile elements, and (2) homogeneous fuel, in which minor actinides are uniformly distributed together with the other fissile elements.

Figure 10 shows examples of both homogeneous and heterogeneous fuel types, many of which are discussed in the following sections. Work to date has established the feasibility of minor actinide fuel/target materials, and the core loadings and reactor strategies for utilizing these fuels. Further development is required before commercial deployment of minor actinide fuels or targets. Additional in-pile irradiations are required for both normal operating and accident conditions. Further safety analysis and tests are required. Some development is needed for analysis tools and computer programmes. Finally, proof of performance requires fuel irradiations starting with small test pins or segments, proceeding to full size pins in fully prototypic irradiation environments, and finally to qualification through irradiation of multiple prototypic full size elements.

An exhaustive overview of CEA's experience in selection criteria, synthesis, fabrication, design, physical properties evaluation, irradiation testing and irradiation damage of MA containing fuels and transmutation targets

(and dissolution) describes some of the following developments in detail [19, 135]. Along similar lines, recent developments which have taken place at the Institute for Transuranium Elements (ITU) [149] and in the United States of America by the Department of Energy (DOE) [67] have been utilized in the following sections.

3.1. HOMOGENEOUS FUELS: OXIDE, NITRIDE, CARBIDE, METAL–ALLOY, AND MOLTEN SALT

Solid solution fuels utilize a homogeneous distribution of fissile and minor actinide elements throughout the fertile fuel phase. Ideally, these element distributions are uniform over unit cell distances within a crystal structure, although this may be difficult or unnecessary to achieve in practice. In the case of solid solution fuels, fission damage is more or less uniformly distributed throughout the fuel. The response of fuel matrix to fission fragment damage, fission gas production and diffusion, and solid fission product formation affects fuel performance behaviour. Two different approaches are used in the design of solid solution fuel. A metallic fuel design allows for a large amount of free swelling and high gas release. This design requires an initial thermal bond between metal fuel and cladding, 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.

3.1.1. Oxide solid solutions

Solid solution ceramics are attractive for transmutation applications because they represent a relatively small perturbation compared to established and extensively tested uranium dioxide and MOX fuels currently used in a great majority of reactors worldwide. As such, solid solution fuels for application to minor actinide burning in current generation power reactors has been the subject of substantial theoretical and experimental development [104, 108, 149–151].

MOX fuel has been extensively developed and is in common commercial usage for electricity generation; MOX technology has been reviewed by the Agency [152]. Minor actinides can be added to MOX type fuel for transmutation in either light water or fast reactors. The use of uranium based fuel for transmutation was demonstrated in the SUPERFACT experiment (Minor Actinide Irradiation in Phénix) [153, 154]. Fuel compositions representing both small deviations from MOX fuel and high minor actinide content uranium oxide based transmutation targets were fabricated, characterized, and irradiated. This experiment is described in a greater detail in Section 6.

Oxide ceramic solid solution fuels can also incorporate minor actinides and fissile materials with 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 the addition of yttria (Y_2O_3), calcia (CaO) or magnesia (MgO) [46]. Plutonium dioxide (PuO_2), or minor actinide dioxides, also have fluorite crystalline structure, and two major components form a continuous solid solution. Burnable poisons such as rare earth elements erbia (Er_2O_3) or gadolinia (Gd_2O_3) may be incorporated into the solid solution if required by reactor system neutronics, and can also act as cubic stabilizing additions. Fission products may also be found in the solid solution and may contribute to stabilizing zirconia, as shown by X ray absorption spectroscopy.

The possibility of developing fuels in the ZrO_2 – PuO_2 system was realized in the early 1960s [155]. A large body of information specific to the in-reactor use of zirconia–plutonia based fuels has been compiled within the last decade. This database includes fabrication development, core physics, phase equilibria studies, microstructural characterization, thermal property measurement, corrosion testing, and ion and neutron irradiation [44, 46, 156–159].

In general, fabrication may be carried out either by a co-milling process similar to current industrial MOX fabrication or by co-precipitation from an aqueous solution [46]. The latter option 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.

New methods of fabrication are being developed that could minimize radioactive waste generation, for example infiltration processes for the fabrication of minor actinide fuels, targets and conditioning matrices. Transmutation targets can be fabricated using a sol gel method for the production of porous inactive beads, which are then infiltrated by transuranic solutions [160]. The new concept utilises powder metallurgy routes (compaction, crushing and sieving) to produce porous, nearly dust free granules, which are infiltrated with actinide nitrate. The method has been developed using yttria stabilised zirconia and alumina, and has been demonstrated for the production of Al_2O_3 – AmO_2 targets for neutron capture investigations.

Co-conversion processes play an important role by closing actinide separation–purification operations and at the same time producing mixed actinide solid compounds for the fabrication of fresh fuel. Handling of actinides mixtures, from the initial solution up to the solid product, requires innovative synthesis methods and structures, particularly for minor actinides such as americium and curium. Considering the different designs of future nuclear fuels, various uranium–actinide co-conversion routes are currently being investigated at the CEA-ATALANTE facility [161, 162]. Research and development of minor actinide containing fuels and targets, i.e., (Pu,Am)O₂–MgO, (Pu,Np)O₂–MgO, and (U,Pu,Np)O₂ for use in a future integrated closed cycle system that includes fast reactor and accelerator driven subcritical systems is described [104].

The effect of oxygen potential on the sintering behaviour of MOX fuel containing Am, which has high oxygen potential, was studied using ceramography and metrology [163]. The phase relation of the fabricated Am-MOX was obtained by ceramography, X ray diffraction and differential thermal analysis. Current studies involve characterization of precursor powder morphology effects in the areas of flowability and porosity in order to predict manufacturability. Preproduction pellets were characterized in order to determine impurity levels and locations prior to final production of pellets to be inserted into the reactor [151]. Fuel compositions evaluated in order correlate to irradiation performance of oxide fuels with high concentrations of Pu, Np and Am as actinide transmutation fuels. Some experiments with Np and Am have been performed in France and Japan, but these would be some of the first in the United States of America [151, 153]. The three compositions to be studied are: one MOX composition, (U_{0.80}Pu_{0.20})O_{1.98}, and two MA-MOX compositions with oxygen to metal ratios (O/M) of (U_{0.75}Pu_{0.20}Am_{0.03}Np_{0.02})O_{1.98} and (U_{0.75}Pu_{0.20}Am_{0.03}Np_{0.02})O_{1.95}.

The Japan Atomic Energy Agency (JAEA) has developed homogeneous MOX containing minor actinides (MA-MOX) as a fuel for advanced fast reactors [150]. Physical property measurements (phase state and phase separation, lattice parameters, oxygen potentials, melting temperatures and thermal conductivities) of Am and Np bearing MOX are reported. The composition was in the following range: U_{1-z-y'-y''}(Pu_zAm_{y'}Np_{y''})O_{2.00-x} where Pu : Z = 0–97.9% ; Am : y' = 0.5–7.2% ; Np : y'' = 0–12% ; x : x = 0–0.282 ; and O/M=2.00–1.718.

Irradiation damage studies have been carried out using both neutron and ion irradiation techniques. Ion irradiation is used to simulate fission fragment damage. Work in this area prior to 2008 has been summarized [164]. Breakaway swelling of fuels is often triggered by transition from an ordered crystalline structure to an amorphous phase, with 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 amorphization of stabilized zirconia, nor was it observed after irradiation with 72 MeV I⁺ ions to a dose of 5 × 10¹⁹ cm⁻² at 300–1770 K. Laser profilometry of specimen surfaces after irradiation showed no evidence of ‘popout’ features, indicating that swelling was <2%. Amorphization has not been observed in any case for fully stabilized ZrO₂, indicating that this material can be irradiated to a dose exceeding 100 dpa without occurrence of this phenomenon [46].

The behaviour of inert matrix ceramics like MgO, MgAl₂O₄ and cubic ZrO₂ oxides under swift heavy ion irradiation is also reported on [165]. These oxides have been irradiated with swift heavy ions at CIRIL/GANIL (Centre Interdisciplinaire de Recherches avec les Ions Lourds/Grand Accelérateur National Lourds) to simulate fragment fission effects. Irradiation with different heavy ions (from S to Pb) with energy between 91 and 820 MeV, have been realized at room temperature or 500°C. The fluencies were between 5 × 10¹⁰ and 5 × 10¹⁵ ions/cm². Alumina Al₂O₃ has been also studied as a reference for ceramic materials. The polished faces of sintered polycrystalline disks or single crystal slices have been characterized before and after irradiation using X ray diffraction and optical spectroscopy. The apparent swelling evaluated from surface profile measurements after irradiation is very important for spinel and zirconia, compared to that of magnesia or alumina. Amorphisation seems to be the origin of this swelling, and the electronic stopping power of the ions is the most influential parameter for irradiation damage. Point defects characterized by optical spectroscopy show a significant amount of damage in the oxygen sub-lattice of irradiated oxides. F⁺ centres are present in all irradiated oxides. However, new absorption bands are observed and cation clusters cannot be excluded in magnesia and spinel after irradiation.

There have been cursory studies conducted on other minor actinide fuels or targets as candidate materials for solid solution fuels, most notably ceria and yttria. CeO₂ adopts the fluorite phase and is isostructural with PuO₂ and UO₂. Y₂O₃ is nearly so, exhibiting a bixbyite structure that resembles the fluorite structure with one fourth of the anions missing. Specimens (Pu,Ce)O₂ and (Pu,Ce)O_{2-x} were irradiated as a split pellet stack in the TRABANT-1 irradiation unit at HFR-Petten [166]. Here, Ce may be considered a surrogate for Np or Am. In these fuels, 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/t, the TRABANT-1 experiment was removed from the reactor due to failures in two pins. Post-irradiation examination showed that the fuel had melted, presumably due to low thermal conductivity of the substoichiometric composition [167–168].

The project FUTURE (Fuels for Transmutation of Transuranium Elements) was initiated under the FP5 programme to compare the performance of different oxides viz. (Pu,Am)O₂, (Th, Pu, Am)O₂ and (Pu,Am,Zr)O₂ as fuel for ADS [19]. The first phase of this programme involves developing methods of synthesis and measuring thermal and chemical properties of these compounds at high temperatures; then characteristics of the fuels are evaluated. The different types of fuels studied are fissile compounds cited above in the form of solid solution matrix composites ZrO₂ or MgO, or matrix composites tungsten, chromium or vanadium. The incorporation of curium into these compounds is taken into account in assessing the performance of these fuels. In terms of matrices, neutron studies promote magnesia, zirconia, chromium and molybdenum, which can maintain a matrix volume fraction exceeding 50%, the lower limit value necessary for the manufacture of composites. The very poor thermal conductivity of zirconia resulted in reduced margins. First tests have shown the manufacturing risk of interaction between chromium and the fissile phase; studies are now focused on magnesium and molybdenum. Concerning fissile phases, the goal is to produce compounds of composition close to (Pu_{0.5}Am_{0.5})O₂. In this context, CEA Atalante developed a co-precipitation process and has made a few batches of composition (Pu_{0.8}Am_{0.2})O₂, (Pu_{0.5}Am_{0.5})O₂ and (Pu_{0.2}Am_{0.8})O₂. Finally, the irradiation Metaphix (described in 6.2.4) on the homogeneous transmutation mode, aims to study the behaviour of metallic fuels containing a small quantity of minor actinides.

Ceria based fuel was also found to disintegrate on immersion in molten sodium at 550°C for 6 h, making it unappealing for use in sodium cooled reactors [166]. TANDEM accelerator tests conducted at AECL showed high swelling of ceria when irradiated with 72 MeV iodine ions [169]. Heavy ion irradiation experiments of Y₂O₃ reveal unusual behaviour, with cubic to monoclinic transformation occurring under irradiation with molybdenum and lead ions [170]. The irradiation behaviour of UO₂ in yttria and ceria macrodispersion, however, indicates that the response of the material to the combined effects of neutrons and local fission fragment damage 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.

3.1.2. Nitride solid solutions

Nitride fuels are attractive primarily due to their high thermal conductivity relative to actinide oxides, their high actinide density, and their simple phase equilibria [144–148, 170–172]. Typically ZrN, TiN, YN, or AlN are proposed as inert matrix phase for nitride fuels. More emphasis has been placed on specimen fabrication of these minor actinide fuels or targets 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 JAEA through carbothermic reduction of oxides [170]. Solid solution (Pu,Zr)N pellets and two phased TiN + PuN pellets have been fabricated and are undergoing irradiation testing [171]. Fuel fabrication activities focussed on (Pu,Zr)N are also being conducted in Switzerland as part of the CONFIRM programme [172] and in the United States of America as part of the AFCI programme [67]. Both of these programmes use fabrication techniques that rely on carbothermic reduction of oxide powders. The fuels tested in the CONFIRM irradiation project, as well as in Japanese programmes, are helium bonded, while the fuels in the United States of America test programme are sodium bonded. The initial results of these tests will provide the first scoping data on inert matrix nitride fuels, from which fuel design can be improved for potential application in fast spectrum systems.

Transuranic nitrides of NpN, PuN, AmN and CmN have the same NaCl type crystal structure with similar lattice parameters. Actinide mononitrides would form a solid solution, which could accommodate a wide range of actinide compositions. This characteristic may be important for minor actinide bearing fuel since the isotope vector of actinides strongly depends on commercial spent fuel history. The formation of solid solution has been experimentally confirmed in the Np–Pu–Am–Cm nitride system.

The melting point of nitride fuel is higher than that of metal fuel and comparable to that of oxide fuel. Thermal conductivity is higher than that of oxide fuel and comparable to metal fuel.

Actinide nitrides, in particular AmN and CmN, have a hygroscopic nature and also react with oxygen in the air. It is necessary to handle them in an inert atmosphere. A potential issue confronting the use of this fuel is the need to enrich nitrogen in ¹⁵N isotopes in order to avoid the production of large amounts of hazardous ¹⁴C in reactor through the ¹⁴N(n,p)¹⁴C reaction. Highly enriched ¹⁵N may have to be used for nitride fuel in order to prevent this

from occurring. Applying pyrochemical process in the treatment of spent fuel, could allow ^{15}N to be recovered and recycled.

Knowledge of ZrN radiation stability is limited. The radiation stability of ZrN using a 2.6 MeV proton beam at 800°C has been recently reported upon [173]. Irradiated microstructure and hardening were investigated and compared with annealed samples. A high density of nano sized defects was observed in samples irradiated to doses of 0.35 and 0.75 dpa. Some defects were identified as vacancy type pyramidal dislocation loops using lattice resolution imaging and Fourier filter image processing. A very slight lattice expansion was noted for the sample which received a dose of 0.75 dpa. Hardening effects were found for samples irradiated to both 0.35 and 0.75 dpa using Knoop indentation.

3.1.3. Carbides

Carbide fuels are also attractive, due to high thermal conductivity relative to actinide oxides, high actinide density, and expected simple phase equilibria. There are no known tests for carbide fuels designed for transmutation of minor actinides.

Although much smaller than that for either metal or oxide fuels, the irradiation performance database for (U, Pu)C mixed carbide fuels is considerable [174–177]. Over 470 mixed carbide fuel pins were irradiated in EBR-II (Experiment Breeder Reactor) and over 200 mixed carbide pins were irradiated in FFTF (Fast Flux Test Facility). Mixed carbide fuel with 70% PuC content is being used as driver fuel in the Fast Breeder Test Reactor (FBTR) in Kalpakkam, India [178–180]. This fuel crossed a peak burnup of 150 000 MWday/tonne in 2005, which is the maximum limit per design [73, 180]. The FBTR discharged spent nuclear fuel of very high burnup was successfully reprocessed [181].

Carbide fuel failures typically result from mechanical fuel cladding interactions. Fuel swelling is greater than that in oxide pellets, which leads to early fuel/cladding gap closure. Since carbide fuel generally operates at a low temperature relative to its melting point, fuel creep is not effective at relieving cladding stress. For this reason, mixed carbide fuel pin design must incorporate a large fuel/cladding gap and make use of a low density fuel in order to delay the onset of mechanical fuel cladding interactions. While cladding carburization has been a historical concern for mixed carbide fuels, and was observed for Type 316SS clad pins irradiated in EBR-II, no fuel failures have been attributed to this phenomenon.

3.1.4. Metal alloys

Metal alloy transmutation fuels are of interest primarily for closed fuel cycle fast spectrum systems when coupled to an electrorefining based pyroprocessing, due to the potential for a compact integral reactor/recycle system [182]. Recent fabrication and irradiation test results, discussed more extensively in Section 6, have established the basic viability of metal fuel for transmutation of minor actinides [183].

The high swelling behaviour of metallic fuel is compensated for by fuel design, which allows for a certain amount of free swelling to occur. Swelling is accommodated by fabricating the fuel with a sodium filled fuel to cladding gap. The gap is sized so the fuel occupies approximately 75% of the cross-sectional area inside the cladding at the beginning of life. The sodium filled gap minimizes 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% in the early stage of fuel burnup, and causes the fuel to contact the cladding wall. At this point however, the fuel forms a network of interconnected porosity for fission gas release to the plenum, and exerts almost no stress on the cladding.

High fission gas release leads to pin pressurization, which drives cladding creep. This is controlled by appropriately sizing the gas plenum region of the fuel pin. At 18–20 at.% 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 [184]. Since metal fuel is engineered to allow for swelling and promotion of gas release, the issuing of helium generation due to americium and curium is a matter of: (1) sizing the gas plenum to prevent excessive gas pressure driven cladding creep, and (2) ensuring that gap size is adequate to accommodate solid fission product swelling. The recycling technology for metal fuel has been established by engineering scale pyroprocessing. Remote fabrication was established as part of the EBR-II development programme in the 1960s with the remote fabrication of more than 39 000 fuel pins in a hot cell [185].

Metal fuels may be used as either inert matrix fuels or fertile fuels. Recent work on inert matrix fuel has focused on fabrication, characterization, and irradiation testing of metal alloy fuels in the Pu–Zr alloy system [186]. The Pu–Zr phase equilibrium diagram is well characterized [187]. The addition of zirconium as an alloying agent limits fuel cladding chemical interaction and raises the fuel melting temperature. Compositions of interest are generally within the face centered cubic δ -phase field, which has an zirconium content upper boundary of approximately 40 wt.%. The lower boundary for plutonium content is set by margin to melting requirements. Early work established the acceptable low burnup behaviour of Pu–Zr alloys [188].

An important issue related to the use of U–Pu–Am–Np–Cm–Zr alloys as transmutation fuels is the incomplete knowledge of phase equilibria in the multi-component alloy system. The potential for immiscibility and formation of an inhomogeneous microstructure is not a fuel performance issue, as shown by the excellent performance of multi-phase U–Pu–Zr fuel [189]. Rather, the formation of low melting phases in the complex alloy system is an issue that must be experimentally determined. Recent experimental irradiations and out of pile studies conducted as part of the Advanced Fuel Cycle Initiative (AFCI) programme indicate that this is not an issue. Further details of AFCI and METAPHIX (Metal fuel irradiation in Phénix reactor) irradiation tests are discussed in Section 6.

3.1.5. Molten salts

Molten salt reactor concepts for minor actinide burning require reconsideration of earlier molten salt concepts, including optimization of neutron spectra in the core and selection of salt composition and approaches to its treatment.

Recent studies have demonstrated that a broad range of molten salt reactors with PuF₃ and minor actinides as the startup fuel is conceptually feasible. The basic reactor flowsheet is essentially the same as that for the Oak Ridge National Laboratory's (ORNL) earlier molten salt reactor design [190–192]. The only differences are in core/blanket configuration, details of fuel salt composition and the fission product cleanup system. The use of uranium free fuels as well as ThF₄ and UF₄ in a homogeneous solution is conceptually feasible. Desirable characteristics of these innovative fuels are: (1) stability and negligible pressurization at high temperatures; (2) good hydrodynamic and heat transfer properties; (3) stability in a high radiation field; (4) low neutron absorption cross-sections; and (5) low solubility of gaseous fission products to allow high burnup [193].

Molten salt reactor design concepts include both thermal and fast neutron spectrum systems. At one end is a high power density, well thermalized graphite moderated reactor, in which fuel consists of a molten mixture of ⁷Li, Be/F and Na, Be/F, containing appropriate quantities of plutonium and minor actinides as trifluorides [194–196]. At the other end is a fast spectrum reactor without a graphite moderator, in which the solvent system consists of NaF–⁷LiF (and/or other possible constituents, like CaF₂, ZrF₄, or PbF₂).

Trivalent plutonium and minor actinides are the only stable species in various molten fluoride salts. Tetravalent plutonium could transiently exist if salt redox potential is high enough, but for practical purposes (stability of potential container material), the salt redox potential should correspond to the stability area of Pu(III) [197]. Trifluoride species, PuF₃, AmF₃, CmF₃, NpF₃ and rare earths have a limited solubility in fluoride salt mixtures and tend to co-precipitate when their solubility is exceeded.

PuF₃ solubility is high in pure LiF or NaF and decreases with the addition of BeF₂ and ThF₄. The solubility decrease is greater with the addition of BeF₂. The solubility of PuF₃ in LiF–BeF₂ and NaF–BeF₂ solvents is temperature and composition dependent, and PuF₃ solubility seems to be minimal in neutral melts. PuF₃ solubility peaks at about 0.5 mol.% at the liquidus temperature of 600°C and increases to about 2.0 mole % at 800°C [198]. These values are adequate for the use of plutonium as a fuel and would allow accumulation of rare earth fission products for as long as 10 years without processing, if these were desirable. Substitution of a small quantity of minor actinides scarcely changes the phase behaviour of the solvent system [199–200].

The single fluid fast spectrum molten salt reactor concept requires a concentration of PuF₃ much higher than that for the 2LiF–BeF₂ system with graphite moderator. The LiF–PuF₃ system has eutectic at 743°C and 20 mol.% PuF₃. The NaF–PuF₃ system has eutectic at 24 mol.% PuF₃, and melting at 727°C. Inspection of the phase diagram for the LiF–NaF–PuF₃ system reveals that a considerable range of compositions with about 10 mol.% PuF₃ will be completely molten at 600°C. It is possible that some ternary or quaternary additions (e.g. CaF₂) would lower the liquidus temperature. As expected from the general similarity between PuF₃ and minor actinide trifluorides, the substitution of a relatively small quantity of AmF₃, CmF₃, or NpF₃ for PuF₃ has a minimal effect on phase behaviour and the phase behaviour of a fuel will be determined by the behaviour of the

LiF–NaF–PuF system. However, significant quantities of minor actinides in the mixture complicate phase behaviour of the fuel system.

3.2. HETEROGENEOUS FUELS

Heterogeneous fuel designs (e.g. composite fuels such as cermet or cermet) may be desirable for safety reasons and to obtain high transmutation rates. Heterogeneous fuels consist of discrete particles which may or may not be embedded in a solid matrix. The matrix, if present, may be fissile, fertile, or inert. Special cases of vibro-pac and sphere-pac fuels can be represented as a dispersion of discrete particles with no matrix, and are discussed in this section; they should be distinguished from pellet type fuels. Examples of some possible chemical forms of heterogeneous fuels are given in Table 1. Some general information on heterogeneous fuels is provided in this section.

3.2.1. Vibro-pac and sphere-pac fuels

Heterogeneous fuels consist of a distribution of discrete fuel particles. The particles may or may not be embedded in a metal or ceramic matrix.

Vibro-pac fuels with minor actinide additions have been developed at the Research Institute of Atomic Reactors (RIAR) in Dimitrovgrad, the Russian Federation, using a pyroelectrochemical process in chloride melts (oxide electro-winning) [201-202].

Three oxide fuel batches of fuel composition (U - 2%Pu - 3%Am)O₂, (U - 8%Pu - 3%Am)O₂ and (U - 51%Pu - 2%Am)O₂ were produced. Three fuel pins were fabricated from these batches for irradiation in the BOR-60 reactor in the spring of 2005. Detailed compositions of these fuels are summarized in Table 2.

In addition, uranium oxide and uranium–plutonium oxide vibro-pack fuels with addition of Np have been produced. The pyroelectrochemical codeposition technique for uranium and plutonium dioxides was adapted by taking into account the introduction of Np into UO₂ and (U,Pu)O₂ matrices. Experiments based on the codeposition technique resulted in fuel batches containing neptunium. Detailed compositions of neptunium containing vibro-pac fuel batches are summarized in Table 3.

Sphere-pac fuel may offer several advantages. It allows for almost dustless production with no pressing procedure, which is especially advantageous for remote controlled production. Only two size fractions are necessary to reach high filling densities, and either parallel filling or infiltration filling can be applied.

Sixteen fuel segments were fabricated as part of the FUJI project (Fuel irradiation for JNC and PSI) which is a collaboration project between the Japan Nuclear Cycle Development Institute (JNC), the Nuclear Research & Consultancy Group (NRG) and PSI [203]. The experiment included three types of fuel: nine sphere-pac fuel segments, two vibro-pac fuel segments and five pellet fuel segments. Two sphere-pac segments contained neptunium and had the following composition: (5%Np,20%Pu,75%U)O₂.

TABLE 2. MAIN CHARACTERISTICS OF FUEL BATCHES CONTAINING AMERICIUM

Batch No.	Batch mass (g)	Mass portion (%)			Oxygen factor, O/M	Pycnometric density (g/cm ³)
		U	Pu	Am		
1	38.4	34.8	50.8	2.1	1.989	9.71
2	42.6	77.6	7.6	3.0	1.991	9.80
3	94.6	81.9	2.6	3.2	1.984	9.80

TABLE 3. VIBRO-PAC FUEL BATCHES CONTAINING NEPTUNIUM

Batch No.	Batch mass (g)	Mass percentage (%)		
		U	Pu	Np
Co-deposited homogeneous fuel (U, Np)O ₂				
1	92.6	93.4	0	6.6
2	103.5	94.7	0	5.3
Heterogeneous fuel as a mechanical mixture of (U, Np)O ₂ +PuO ₂				
3	200	77	20	3
4	200	74	20	6

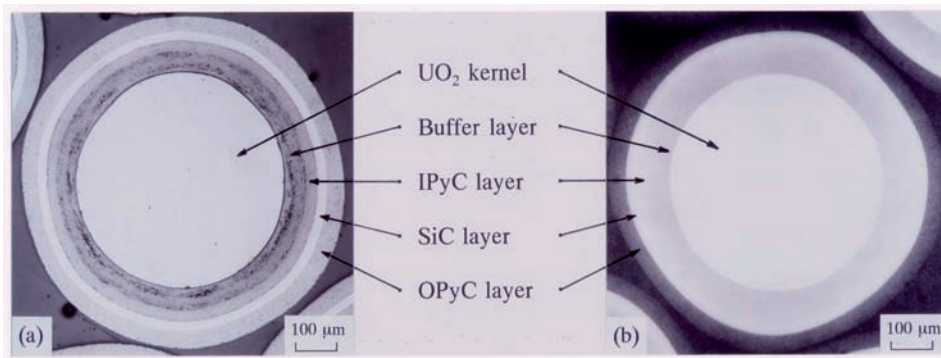


FIG. 11. Ceramograph and X ray microradiograph of TRISO coated fuel particle.

3.2.2. Coated particles

Coated particle fuels are a subset of ceramic composite (cercer) fuel which have been uniquely applied to gas cooled thermal spectrum reactors [56]. Coated particle fuel technology has been demonstrated on a large scale in several prototype and production gas cooled reactors, and a good theoretical basis exists for the development of fuels designed for plutonium burning. Application to plutonium transmutation has been demonstrated experimentally on a limited scale. This technology is of interest primarily in future nuclear scenarios employing high temperature gas cooled reactors, since no commercial scale, high temperature, gas cooled reactors currently exist that make use of this fuel type.

The concept of spent fuel transuranic (TRU) destruction in a TRISO-fuelled (TRI-structural ISO-tropic) gas cooled reactor is known as ‘deep burn’ [204, 205]. The term ‘deep burn’ reflects large fractional burnup of up to 60–70% fissions per initial metal atoms (FIMA), which may be achievable with a single pass, multi-cycle irradiation in very high temperature reactors (VHTRs). The deep burn capability of this fuel makes once through thermal cycles attractive for plutonium and minor actinide transmutation. Coated particle fuel consist 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. The structure of a typical TRISO particle is shown in Fig. 11.

Oxides, carbides, and oxycarbides have been used as kernels. Properly executed, TRISO type fuels are robust and capable of high burnup. Fuel performance is highly dependent on the fabrication technique and redox thermochemistry within the coated particle as a function of burnup [56]. Burnups beyond 900 GW·d/t have been reported for a few particles of PuO_{2-x} TRISO type fuel [206]. Innovative fabrication methods of oxide kernels containing transuranic elements have been developed [207]. Uranium based particles have been shown to withstand

extended exposure to temperatures of 1600°C, with a failure rate in the order of 10^{-4} . Recent work has focused on improving fuel particle fabrication techniques and on developing improved coating materials, particularly ZrC coatings 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 and silver, into the reactor coolant [208–209].

Coated fuel particles are consolidated with graphite powder to form cylindrical fuel compacts or spherical fuel elements. To improve the irradiation performance of coated fuel particles at high temperatures and high burnups, the use of a zirconium carbide layer in place of silicon carbide has been studied.

Coated fuel particles for application in fast spectrum gas cooled reactors have not been well developed or tested, although some exploratory investigations have been made using titanium nitride coatings and coatings derived from non-halide precursors [210]. The use of TiN-coated particle fuel in a subcritical helium cooled core for very high transmutation of minor actinides is being considered [211], as the hard neutron spectrum manifests in further limiting buildup of Cm and Cf to a high burning rate of minor actinides with low plutonium loading.

3.2.3. Dispersion fuels

Dispersion fuels as considered here consist of a fuel phase dispersed in an inert, non-fuel matrix. In an ideal case, the matrix remains largely unaffected by neutron, fission fragment, and α -particle damages from fission events that take place in fuel particles. Only those fissions taking place close enough to the particle surface to allow fission fragments and α particles to escape into the matrix contribute to matrix damage. In the case of an ideal dispersion microstructure consisting of a uniform distribution of spherical particles of one size, the fraction of undamaged matrix is a function of fuel particle radius, fuel particle volume fraction, and fission fragment range in the matrix [212]. The matrix must also not chemically react with fuel particles during irradiation. Through an appropriate choice of metal or alloy for the matrix phase, a metal matrix dispersion fuel can be operated at extremely high temperatures. The selection of tungsten as matrix material in the nuclear rocket programme, for example, allowed the fuel to operate with a maximum centerline temperature of almost 2500°C [213].

3.2.3.1. *Metmet fuel*

Recent developments have taken place regarding fissile metal dispersions (U–Mo alloys) in an aluminium matrix for use as research reactor fuel [214–216]. Prior to this, there had been extensive testing of dispersions of so-called Pu–Al alloy fuels, namely the dispersion of PuAl₄ precipitates in aluminium. Hundreds of fuel specimens of this fuel type have been irradiated, some to burnups as high as 90% fissioning of plutonium atoms. More than 1500 zircaloy clad fuel pins irradiated in the Plutonium Recycle Test Reactor at Hanford were included in the testing to demonstrate use of this fuel in a PWR environment [217].

Although promising results have been obtained, the application of this technology to modern commercial reactors is uncertain due to the low melting temperature of aluminium and corrosion of aluminium matrix by coolant. Extensive safety testing of this type of fuel would be required. Corrosion behaviour can be improved by the use of silumin (AlSi).

3.2.3.2. *Zr–hydride fuel*

The majority of research efforts for zirconium hydride involve uranium fuels and TRIGA (Training, Research, Isotopes, General Atomics) reactors [218–221]. The fuel used consists of actinide zirconium hydride matrix. TRIGA fuel is considered safer than other nuclear fuels because even at a high temperature, the hydrogen's cross section in the fuel is shifted to higher energies, allowing more neutrons to be lost, and less to be thermalized. Most cores that use this fuel are 'high leakage' cores, in which excess leaked neutrons can be utilized for research. Neutron physics considerations, materials properties, irradiation performance, fission product release, pulse heating, and limiting design basis for zirconium hydride fuels have been evaluated. Fuel performance is independent of uranium content up to 45 wt% U, indicating high loading is possible for these fuel types. Reactors with mixed U–Zr hydride fuels have a relatively large negative prompt temperature coefficient of reactivity. Other studies indicate that the driver fuels can exploit thermal properties of ZrH. Experiments on ZrH fuel behavior above 900°C have been performed in TRIGA reactors. After significant repetitive pulsing there was some evidence that at hot spot regions the fuel surface reached about 1200°C. These tests demonstrated that ZrH fuel can operate at

required high temperature levels safely with a reasonable core life. Studies on the feasibility of transmutation for the most toxic isotopes in conventional spent fuel (viz. ^{79}Se , ^{90}Sr , ^{93}Zr , ^{99}Tc , ^{129}I , ^{135}Cs , ^{137}Cs , ^{147}Sm , ^{237}Np , ^{241}Am , ^{243}Am , ^{244}Cm , ^{240}Pu , and ^{242}Pu) with use of TRIGA fuels in ADS are being researched for future innovative nuclear energy systems.

3.2.3.3. *Cermet fuel*

A primary advantage of cermet fuels are their robust behavior. A large amount of work was performed on cermet fuels in the late 1950s and early 1960s as an alternative to oxide pellet fuel in LWR and fast reactors. 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. Aluminium matrix cermet fuels 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 behavior in case of cladding breach. The high burnup capability of carefully fabricated cermet fuels was demonstrated in the early 1960s, validating general principles of cermet dispersion fuel behaviours [222]. Although cermet fuels have demonstrated high burnup capability, they were set aside as a candidate for commercial reactor fuel in the 1960s, possibly due to its more difficult fabrication and lower heavy metal density relative to pellet type oxide fuels.

For LWR plutonium transmutation applications using plutonium oxide as the dispersed fuel phase, particle loading will be in the range of 6–12 vol.% for a system utilizing a burnable poison to control reactivity [223]. Burnable poisons (such as erbia and gadolinia) for reactivity control in thermal spectrum systems can be included as a separate particulate phase, or in a solution with fuel particle. The reactivity swing can also be compensated to some extent by the use of a neutron absorbing matrix such as tungsten or molybdenum [46]. The formulation of a definite criterion to predict failure in these fuels remains elusive due to a complex stress state and unknown material properties during irradiation, although early works included some attempts to quantify failure modes [224].

Recent activities in this area include conceptual studies of the application of cermets to Pu transmutation in LWRs [225], the irradiation testing of aluminium matrix dispersions of UO_2 as fuel for LWRs [226], and the testing of molybdenum based cermets under thermal spectrum conditions [224]. The irradiation of a 36 wt.% UO_2/Mo cermet in the TANOX facility at the SILOE research reactor in France showed that the fuel performed well to a burnup of 55.4 GWd/t, and also maintained integrity during post-irradiation heating to 1580°C . The corrosion behavior of aluminium 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 aluminium matrix fuels, the cermets for Pu and minor actinide transmutation are only in the conceptual phase at this time. Questions remain in the area of core neutronic analysis and in-reactor mechanical behavior of long rods. Safety testing, including transient testing, must be conducted prior to deployment in commercial reactors. If fuel recycling is to be considered, then the compatibility of a large amount of matrix metal with conventional recycling processes must be assessed.

3.2.3.4. *Cercer fuel*

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 fuels are their ability to partially capitalize on the existing understanding of clad pellet type fuel behavior, coupled with the potential for an additional barrier to fission gas release, and 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 microstructural characteristics of the fuel. Two primary strategies have been pursued for optimizing fuel behavior through manipulation of microstructural variables. Macrodispersions attempt to maintain 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 [227]. 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 micro-dispersion, fission related damage occurs over a larger fraction of the matrix volume relative to macro-dispersion.

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 the order of 100 μm size into MgO . Burnable poisons for reactivity control in thermal spectrum systems can be included either in a matrix phase solution, as a separate particulate phase, or in a solution with fuel particles.

Studies have focused on determining the viability of cercher concepts for plutonium transmutation through fabrication studies [46], out of pile characterization, and irradiation testing of materials that employ uranium as a fissile surrogate for plutonium. Fabrication and neutronic studies of SiC micro-dispersions using surrogates have also been conducted [228]. These materials are of interest due to the known good dimensional stability of SiC under irradiation and high thermal conductivity relative to oxides.

The combination of low neutron absorption cross section, high thermal stability for a wide range of ceramic compounds, and similarity to well known pellet type uranium and mixed oxide fuels makes the cercher concept attractive. The response of many ceramic materials to neutrons and fission fragments, however, is a complex function of cercher micro-structure, irradiation temperature and dose history. Ceramics are also generally brittle and care must be taken so that thermal and mechanical stresses in the matrix do not exceed the elastic limit of the material. Otherwise, matrix cracking may occur, negating the fission product barrier, which is a useful attribute 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 minor actinide burning. Cercers can thus be regarded as an attractive fuel concept in the early stages of development.

The potential to incinerate minor actinides in a subcritical ADS using cercher type IMF is the subject of study in several countries. The performance of MYRRHA experimental ADS, as to the transmutation of Am and Cm in IMF samples consisting of 40 vol.% ($\text{Cm}_{0.1}\text{Am}_{0.5}\text{Pu}_{0.4}\text{O}_{1.88}$) fuel and 60 vol.% MgO matrix in three various spectrum regions, were analysed at the Belgian nuclear research centre SCK-CEN [229]. The total amount of MA is shown to decrease in all three considered cases. For Am, the decrease is largest in the reflector but at the cost of a net Cm production. The reduction in Pu content is also the highest in the reflector position. In the other positions, the incinerated amount of Pu is much smaller.

4. PERTINENT THERMAL AND PHYSICAL PROPERTIES FOR FUEL DESIGN, FABRICATION PROCESSES AND FUEL PERFORMANCE

4.1. PROPERTIES CRITICAL TO FUEL DEVELOPMENT

The physiochemical properties of materials involved in the fuel design and fabrication of minor actinide fuels are needed to understand and improve performance of fuel elements exposed to high temperatures, corrosive coolants and a radioactive environment. The objectives for development of a physiochemical properties database are to assemble a database on fuel properties and performance sufficient to support a safety/licensing case, and to develop a fabrication and quality assurance process that will enable effective and economic fuel fabrication [230–232].

The required physiochemical properties include a range of thermodynamic, transport, and mechanical properties, as summarized in Table 4. These properties are needed to understand fuel performance and are also required for fuel behaviour modelling. Relevant experimental data for all minor actinide elements are not available for all properties, and may include datasets for a mixture of actinides. The database is also sparse for metal, oxide, nitride and other potential fuel forms of the minor actinides. A database is available for a few Np compounds, while data for Am and Cm are sparse. The collection of property data for minor actinides is an ongoing effort.

TABLE 4. DATABASE OF PROPERTIES NEEDED FOR MINOR ACTINIDE FUELS

Thermodynamic	Transport	Mechanical
Enthalpy and heat capacity	Thermal conductivity	Hardness
Melting temperature	Thermal diffusivity	Creep
Enthalpy of fusion	Emissivity and optical constants	Thermal shock
Vapour pressure		Swelling
Vaporization		
Thermal expansion		
Density		
Surface tension		
Surface energy		

4.2. STATUS OF PROPERTY MEASUREMENT AND MODELLING

Property measurements of the minor actinides are complicated by the requirement of performing experiments with suitable quantities of high activity materials. These experiments necessitate the use of a glove box with appropriate shielding containing suitable equipment with the capacity to properly maintain the chemical composition of examined material. Suitable measurement methods include X ray diffraction, laser flash diffusivity, and calorimetry. The levels of oxygen and water must be strictly controlled to prevent variations in chemical composition during measurements. Methods that account for decay heat, production of daughter isotopes, and material damage due to radiolysis must be employed to obtain useful data. Facilities to perform these experiments are available at a handful of laboratories and in some cases further expansions are planned.

Because of the difficulty in making empirical determinations, computer modelling is often utilized in fuel design. There are a number of models available for uranium, plutonium, and thorium fuels for fission gas release, corrosion, and other thermal related characteristics. There are also ongoing efforts to develop scale linked models that can predict properties from fundamental atomic and electronic behaviours. However, for minor actinide containing fuels, there is a lack of model development [233–237].

Global fuel performance models depend heavily on fuel thermal and physical properties to predict performance. For example, thermal conductivity influences restructuring and transport processes during fuel irradiation, as these depend upon temperature gradients inside the fuel pellet. Phase stability and mutual solubility behaviour might also restrict fuel performance limits of minor actinide bearing fuels.

When property values have not been measured, they must be calculated taking into account the microstructure of minor actinide materials. For example, a model for thermal conductivity of actinide oxides, including mixed oxides, has been based on the Debye–Einstein theory of thermal energy of ionic dielectrics and correlations between thermoelastic properties of solids. Thermodynamic calculations, such as CALPHAD, try to combine experimental data with theoretical data to predict phase diagrams of minor actinide based systems which can be employed to explore the phase behaviour of minor actinide fuel compositions and assess its performance limits. The need for development of a property database for minor actinides must proceed in conjunction with the development of property models to ensure that data is available for benchmarking and validation.

The IAEA has conducted a comprehensive review of thermodynamic data of all actinide elements and compounds and published a series of 13 reports⁴ between 1976 and 1992. This publications have become a standard for chemical thermodynamic data of actinides and their compounds. The series includes an assessment of thermodynamic actinide data covering actinide elements, binary alloys, aqueous ions, carbides, halides, hydrides, gaseous ions, and miscellaneous compounds. Furthermore, chemical properties of all actinide elements, namely uranium, plutonium and minor actinides, as well as their compounds and alloys, are compiled in a five volume book published in 2006 [231].

⁴ INTERNATIONAL ATOMIC ENERGY AGENCY, The Chemical Thermodynamics of Actinide Elements and Compounds, Parts 1–13, IAEA, Vienna (1976–1992).

An IAEA activity currently under way is targeted at assembling a database (minor actinide database MADB⁵) from existing published literature on minor actinides, especially thermo-physical and thermochemical properties pertinent and applicable to fuels as well as advanced partitioning methods. The present web based database only offers bibliographic information together with technical data whenever available. The property database intends to cover thermodynamic, electrochemical, phase diagram and physical properties for respective distinct unary and binary as well as multi-component oxide/nitride/halide/metal minor actinide systems in solid and molten phases.

Development of a predictive tool for assessment of thermodynamic properties and phase diagrams of advanced fuels, based on the careful critical analysis of experimental data and a complete modelling of solid and liquid solutions is ongoing in several Member States. Such a thermodynamic tool could be useful for the development of advanced nuclear fuels in the following areas: (1) sintering conditions for manufacturing; (2) chemical interactions between fuel components; (3) compatibility between fissile material and coating; (4) gas partial pressures, with impact on thermo-mechanical behaviour; and (5) localization of fission products. In this context, ITU has built a material property database (f-MPD⁶) which is a compilation of physical, thermophysical and thermochemical properties of compounds relevant to lanthanide and actinide (the f elements) research in general, and specifically for advanced fuel cycle studies, i.e. fuel fabrication, fuel behaviour and pyrochemical processing. In the f-MPD project, the properties of lanthanide and actinide elements and their oxides, nitrides and halides are being collected.

4.3. PROPERTIES OF ACTINIDE ELEMENTS

The following sections provide a summary of property data available for minor actinide elements and selected compounds.

4.3.1. Elemental (metallic) actinide

Actinide elements produce a plethora of interesting physical behaviours owing to the 5f electronic states. Moore and van der Laan recently published a review which compiles and analyzes progress in the understanding of the electronic and magnetic structure of 5f states in actinide metals [238]. The results of the theoretical studies are interwoven and discussed in relation to published experimental data. Table 5 presents elemental data for actinides. The values in Table 5 are nominal values at room temperature. The atomic mass of minor actinides varies according to isotopic composition, which is linked to both material source and decay time since production. Density data are for pure metal at room temperature. Plutonium, in particular, has a complicated phase equilibrium and forms seven solid phases with different densities. Equilibrium phases of minor actinides (and uranium) and approximate phase transition temperatures are provided in Table 6. Room temperature lattice parameter data are presented in Table 7.

TABLE 5. ELEMENTAL DATA FOR ACTINIDE ELEMENTS [239]

Element	Atomic number	Atomic mass	Nominal density (g/cm ³ at 298K)
U	92	238.0289	18.95
Np	93	(237.0482)	20.25
Pu	94	(244.0642)	19.84
Am	95	(243.0614)	13.67
Cm	96	(247.0703)	13.51

⁵ To be published as a web based database shortly; for further details refer <http://www-nfcis.iaea.org/>

⁶ The f-MPD database can be consulted at www.f-elements.net.

TABLE 6. EQUILIBRIUM PHASES AND APPROXIMATE PHASE TRANSITION TEMPERATURES OF PURE ACTINIDE ELEMENTS

Element	Phase	Crystal structure	Transition	Temperature (K)
U	α	orthorhombic	$\alpha - \beta$	942
	β	tetragonal	$\beta - \gamma$	1049
	γ	b.c.c	$\gamma - L$	1408
Pu	α	monoclinic	$\alpha - \beta$	397.6
	β	b.c.m.	$\beta - \gamma$	487.9
	γ	f.c.orthorombic	$\gamma - \delta$	593.1
	δ	f.c.c.	$\delta - \delta'$	736
	δ'	b.c.t.	$\delta' - \epsilon$	755.7
	ϵ	b.c.c.	$\epsilon - L$	913
Am	α	d.h.c.p.	$\alpha - \beta$	923
	β	f.c.c.	$\beta - \gamma$	1350
	γ	b.c.c.	$\gamma - L$	1449
Np	α	orthorombic	$\alpha - \beta$	553
	β	tetragonal	$\beta - \gamma$	849
	γ	b.c.c.	$\gamma - L$	912

TABLE 7. LATTICE PARAMETERS OF ACTINIDE ELEMENTS

Element	bcc lattice parameter a_M (nm)	fcc lattice parameter a_M (nm)
U	0.3524	—
Np	0.3518	—
Pu	0.3636	0.4637
Am	—	0.489
Cm	—	0.5065

Available thermal expansion data for actinide elements of interest are presented in Fig. 12 [240–241]. The coefficient of thermal expansion of α americium metal (dhcp structure) is given as $\alpha_a = .75 \pm 0.2 \times 10^{-6} \text{ K}^{-1}$ and $\alpha_c = .62 \pm 0.2 \times 10^{-6} \text{ K}^{-1}$ [231]. Measured data for Pu-40Zr is also presented [242]. Heat capacity data for minor actinide elements and zirconium is presented in Fig. 12 [67, 243–244]. The data in Fig. 13 include values for Pu-4Am-2Np-60Zr alloy (wt.%) based on the Neumann–Kopp model [245]. Thermal conductivity data for actinide elements is presented in Fig. 14 [239, 241, 246–249]. Thermal conductivity data for americium and curium are estimated.

4.3.2. Actinide nitride

Lattice parameter data for actinide nitrides are presented in Table 8 [232, 250–253].

Thermal expansion data of NpN and AmN determined from the temperature dependence of lattice parameters measured by high temperature X ray diffraction are shown in Fig. 15 [251–252]. The measurements were carried out in flowing N_2 in the temperature range 300–1369 K for NpN and 296–1488 K for AmN. Reported values for thermal expansion of UN and PuN are also plotted.

Specific heat capacities of UN, PuN, NpN, and AmN measured using differential scanning calorimetry have been reported [243, 254–255]. Data for UN and PuN are also available in literature [246, 256–257].

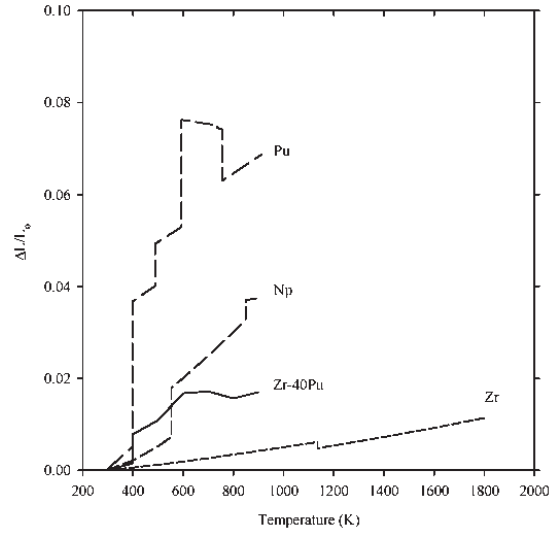


FIG. 12. Thermal expansion data for actinide elements.

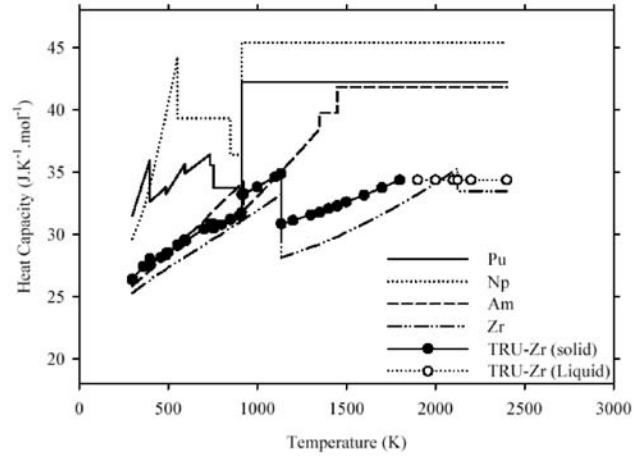


FIG. 13. Heat capacity of minor actinide elements and calculated values for a TRU-Zr model alloy.

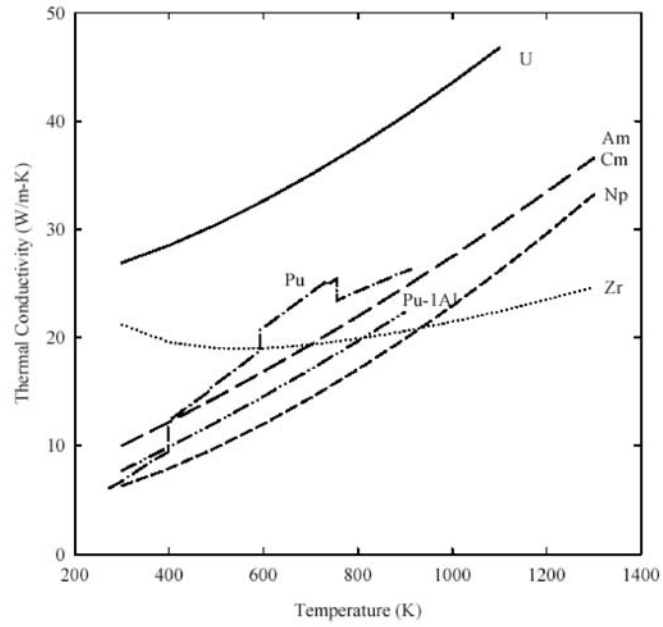


FIG. 14. Thermal conductivity for actinide elements, stabilized plutonium alloy, and zirconium.

TABLE 8. LATTICE PARAMETERS

Nitride	fcc lattice parameter a_{MN} (nm)
UN	0.4888
NpN	0.4897
PuN	0.4905
AmN	0.4991
CmN	0.5027
ZrN	0.4577

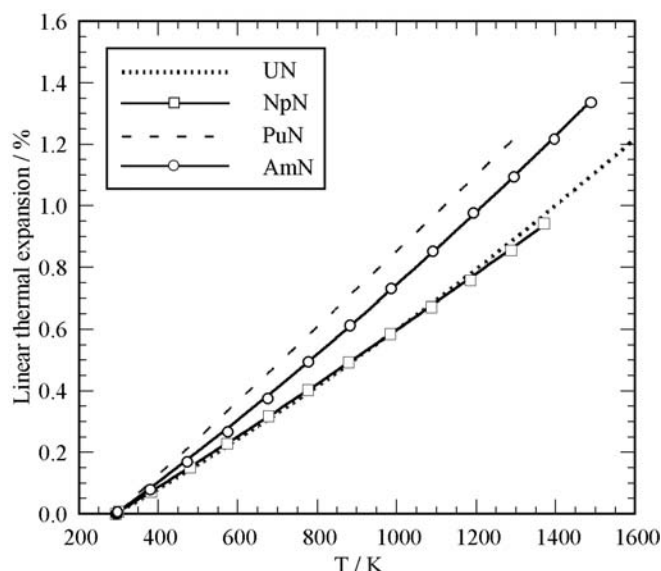


FIG. 15. Thermal expansion data of NpN and AmN, together with those of reported values for UN and PuN for comparison.

Thermal diffusivity of AmN was measured using a laser flash method from 298 to 1473 K for a specimen with a density of approximately 77% of the theoretical density [258]. From this measurement, and using the specific heat capacity of PuN, thermal conductivity of AmN was obtained [232]. Thermal diffusivities of UN, NpN, PuN, and solid solutions of (U, Pu)N, (U, Np)N and (Np, Pu)N have been measured using the laser flash method from 740 to 1630 K, and thermal conductivities have been reported as a function of temperature [232, 259–261]. Figure 16 shows the thermal conductivity of AmN, together with that for PuN, NpN, and UN.

Systematics of the evaporation behaviour of actinide mononitrides, including NpN and AmN, as well as mixed nitrides with plutonium, are also reported [262].

4.3.3. Actinide oxide

Thermal expansion of stoichiometric actinide dioxides (ThO_2 , UO_2 , NpO_2 and PuO_2) was investigated using a high temperature X ray diffraction method [263–267]. Selected values of these data are given in Table 9, which lists lattice parameters of actinide oxides at room temperature.

The thermal expansion of AmO_2 and $\text{AmO}_{1.5}$ was measured using high temperature X ray diffractometry [263, 266]. The measurements were performed in flowing air for AmO_2 and flowing N_2 for $\text{AmO}_{1.5}$. Figure 17 shows the thermal expansion of AmO_2 , together with reported values for UO_2 [239, 258, 266], NpO_2 [263, 266], and PuO_2 [263, 264, 266]. The thermal expansion of AmO_2 is smaller than that of others, though the difference is not large. Figure 18 shows the thermal expansion of $\text{AmO}_{1.5}$, which has a hexagonal close packed structure. Volume expansion of $\text{AmO}_{1.5}$ is about 1.5 times that of AmO_2 .

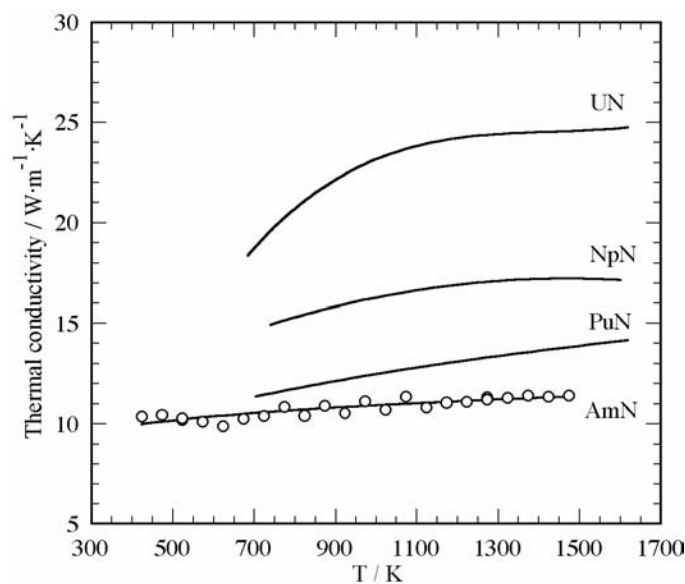


FIG. 16. Thermal conductivity of AmN, together with that of reported values for UN, NpN, and PuN for comparison.

TABLE 9. LATTICE PARAMETERS OF ACTINIDE DIOXIDES

Oxide	fcc lattice parameter a_{MO_2} (nm)
ThO ₂	0.5597
UO ₂	0.54704
NpO ₂	0.54334
PuO ₂	0.53960
AmO ₂	0.53730
CmO ₂	0.53590
ZrO ₂ (c)	0.5120

The thermal conductivity of minor actinide oxide was measured [268–272], the phase behaviour of plutonium oxide with the addition of 9% americium was studied [273], and oxygen potentials of homogeneous minor actinide oxides were measured at temperatures of 1473–1623 K using a gas equilibrium method with (Ar, H₂, H₂O) gas mixture [274, 275]. Vapourization behaviour of irradiated oxide fuel as well as (Cm,Pu)₂O₃ is also reported [276].

4.3.4. Actinide carbide

Little thermophysical property data is available for minor actinide carbides. Table 10 presents lattice parameter data of uranium, neptunium, and plutonium monocarbides [175, 180, 277–279].

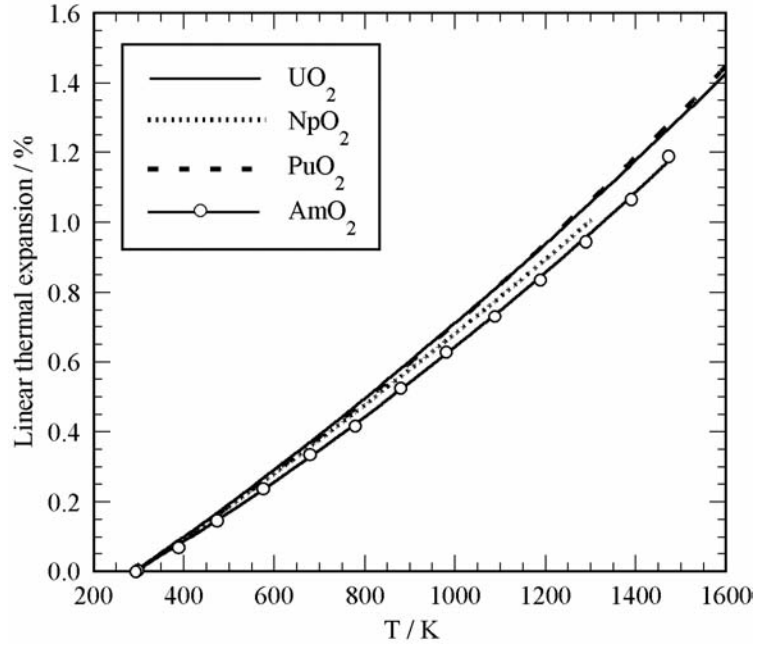


FIG. 17. Thermal expansion of AmO_2 , together with that of reported values for UO_2 , NpO_2 , and PuO_2 for comparison [266].

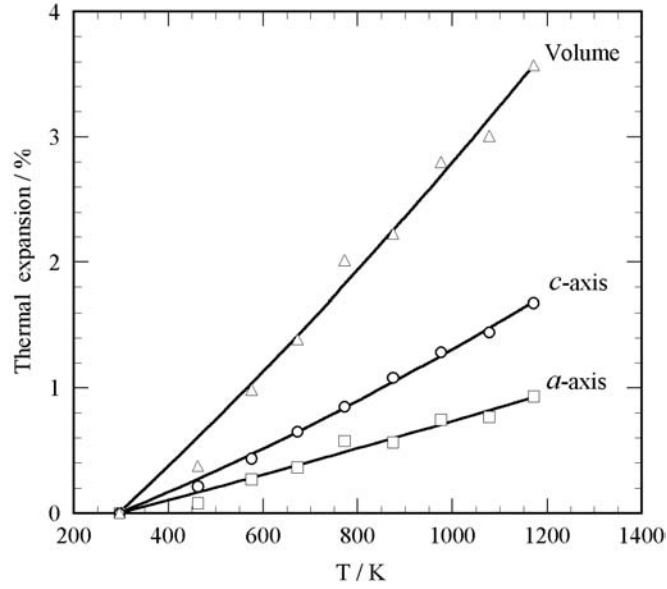


FIG. 18. Thermal expansion of $\text{AmO}_{1.5}$ with hexagonal, close packed structure [266].

TABLE 10. LATTICE PARAMETERS OF ACTINIDE MONOCARBIDES

Nitride	fcc lattice parameter a_{MC} (nm)
UC	0.4961
NpC	0.5005
PuC	0.4973
AmC	—
CmC	—

5. FABRICATION PROCESSES FOR FUELS CONTAINING MINOR ACTINIDES

High alpha activity, high decay heat, high neutron emission rate, and high γ activity relative to uranium oxide fuel all require special consideration during the fabrication of minor actinide fuels. Table 11 lists the decay heat and neutron production rate of primary minor actinide isotopes present in transuranic fuel.

Because the α activity of minor actinide fuel is many orders of magnitude higher than that of uranium, the fabrication of fuel with significant minor actinide content requires utilization of α tight containment boxes. The standard industrial practice for MOX fuel fabrication requires fabrication in a α tight glovebox lined with local shielding. The addition of 5 wt% ^{241}Am to MOX fuel increases α activity by roughly one order of magnitude. The further addition of 1.5 wt% ^{244}Cm results in α activity more than two orders of magnitude greater than that of MOX. More important to the fabrication of fuel are the implications of penetrating radiation (γ and neutrons). It is not likely that material with any significant curium content can be fabricated outside of a heavily shielded hot cell facility because of high associated radiation fields. Depending on minor actinide content, a fully remote facility may be required, with a provision disallowing human entry. Some proposals for transmutation of actinides advocate incorporation of some level of fission products in the fuel to further increase radiation field and hence proliferation resistance. In this case, a fully remote hot cell facility is required.

An additional consideration for pellet type ceramic fuel is the effect of radiation and decay heat on the binders used during pellet pressing. Traditional organic or metal-organic binders tend to decompose quickly upon incorporation into a system with high minor actinide loading.

There is no industrial scale experience in fabricating high minor actinide content fuels specifically designed for transmutation. Fuel fabrication processes are currently being carried out on a laboratory scale, generally in support of irradiation tests. In this environment, the objective is a study of process parameters. Radiation dose is often controlled through limiting material quantities and local shielding. Details of fabrication process equipment required for industrial production such as remote operation, equipment maintenance, and fuel cooling are not addressed, and further development of engineered fabrication systems as well as chemical and physical process optimizations is still required.

The following sections describe the current state of laboratory scale fabrication processes developed for minor actinide oxide, cermet, cermet, metal, and nitride fuels.

TABLE 11. CHARACTERISTICS OF TRANSURANIC NUCLIDES

Isotope	Half-life (a)	Spontaneous neutrons (n/g-s)	Decay heat (W/g)
Np-237	2.14×10^6	0.90	0.00002
Pu-238	87.7	3.5×10^4	0.56
Pu-239	2.41×10^4	96.0	0.002
Pu-240	6.54×10^3	1.3×10^3	0.007
Pu-241	14.4	1.23	0.004
Pu-242	3.76×10^5	2.0×10^3	0.0001
Am-241	432.2	7.0×10^3	0.11
Am-243	7.38×10^3	5.4×10^2	0.007
Cm-242	0.446	2.9×10^7	120.0
Cm-243	28.5	1.3×10^5	1.7
Cm-244	18.11	1.2×10^7	2.8

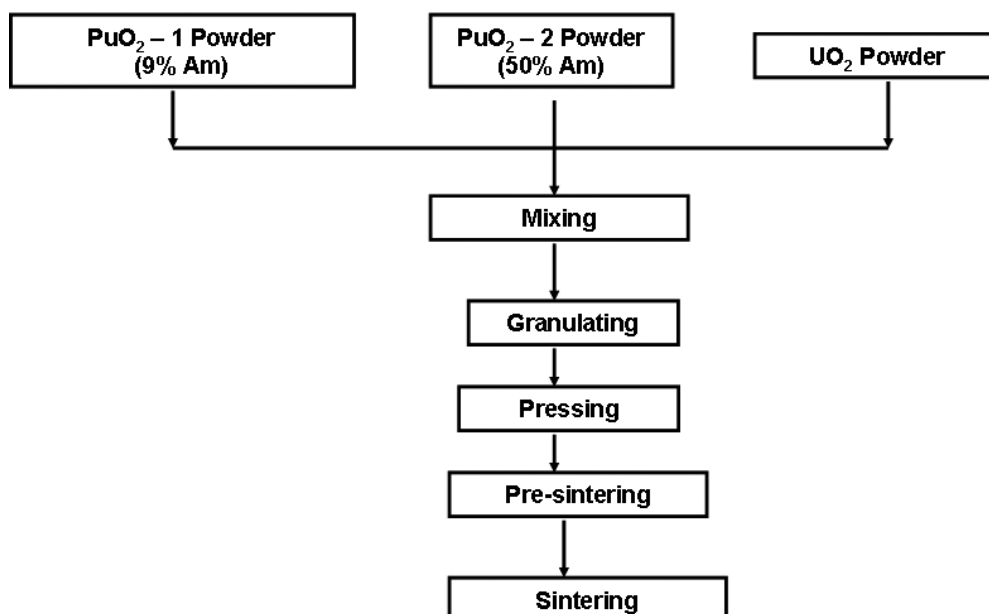


FIG. 19. Flow diagram of Am-MOX fuel pellet fabrication.

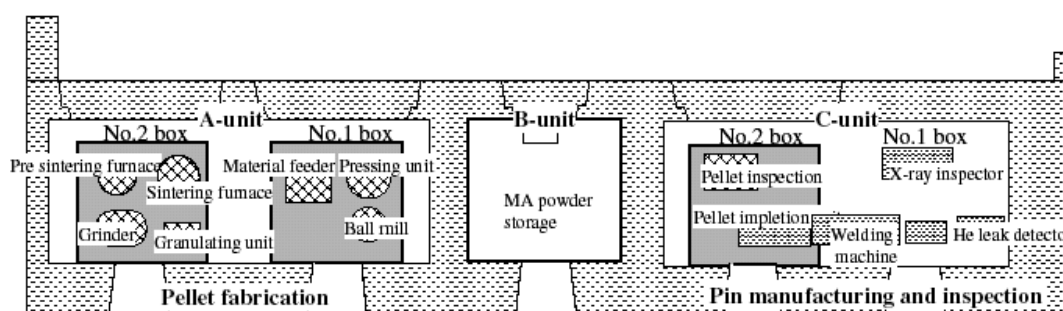


FIG. 20. In-cell layout of equipment used in Am-MOX fuel pellet fabrication and pin manufacturing.

5.1. FABRICATION PROCESSES FOR OXIDE FUEL

5.1.1. Powder metallurgy

An in-cell remote fabrication technique has been developed for MOX fuel pellets containing 3% and 5% americium at the Japan Nuclear Cycle Development Institute (JNC). Fuel pellets were fabricated using a conventional powder processing route shown in Fig. 19. A series of fuel pin fabrication apparatus were installed in the hot cell to protect workers from gamma ray exposure.

Figure 20 shows the layout of fabrication apparatus in the hot cell. All pellet fabrication equipment systems were installed within two stainless steel boxes inside the A-unit cell. In a second cell, the B-unit, raw powder and pellets were stored. A third cell, the C-unit was used for quality inspection and fuel pin manufacturing.

The fabrication apparatus were systematically placed in the hot cell to ensure smooth flow of the fabrication process. Materials used in fabrication were transported between stations with a conveyor system attached beneath stainless steel boxes. Fabrication processes were remotely controlled from a panel in an operating room outside the hot cell as much as possible, and the frequency of manual operations using remote manipulators was minimized.

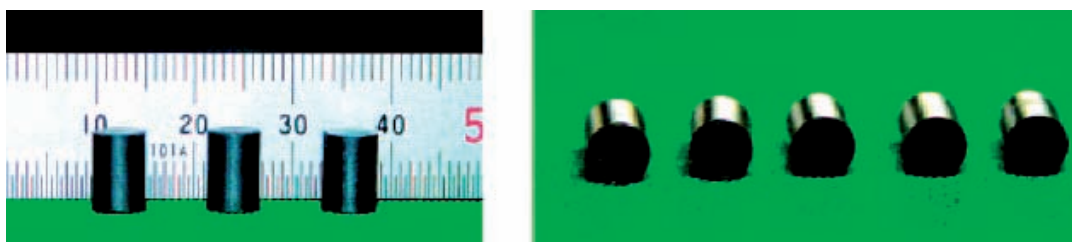


FIG. 21. Pictures of 5 wt.% Am-MOX pellets remotely fabricated using powder processing.

To batch the fuel, UO_2 and Am-Pu O_2 powders were weighed within an error of 1 mg using a vibrator and an electronic balance to adjust the Am O_2 content to 3 or 5 weight percent. Powders were then blended with an organic binder in a ball mill. The stainless steel ball mill pot (200 mL capacity) used to blend and pulverize the powder simultaneously had an inside surface lined with silicon rubber and used tungsten balls 10 mm in diameter. The ball mill was designed to swing along a longitudinal axis while rolling in order to enhance mixing of the powders.

Initially Am-MOX fuel pellets containing 3 wt.% Am with lower density (88.2% of theoretical density) were obtained, which is below the specified value. The suspected reason for this low density value is that UO_2 and Am-Pu O_2 powders had differing morphologies and were not uniformly homogenized. Examination of ball milling time on pellet density revealed that a ball milling time of more than 10 hours was needed.

Taking into account experience gained during fabrication of 3wt.% Am-MOX fuel pellets, 5wt.% Am-MOX fuel pellets were fabricated using the same method. The density of these fuel pellets was above 93% of theoretical values. Ceramography showed homogeneous fuel pellets without agglomeration of plutonium. Figure 21 shows pictures of 5% Am-MOX fuel pellets.

5.1.2. Sol-gel process

The sol-gel process has been used for the synthesis of thorium, uranium, and plutonium fuels, including the incorporation of burnable poisons as well as minor actinide bearing fuels [19, 22, 149, 280–288].

The fundamental property of the sol-gel process is that it is possible to generate ceramic material at low temperatures. The sol gel process involves transition of a system from a liquid ‘sol’ (mostly colloidal) into a solid ‘gel’ phase. The starting materials used in the preparation of ‘sol’ are usually inorganic metal salts or metal organic compounds such as metal alkoxides. The precursor is subjected to a series of hydrolysis and polymerization reactions to form a colloidal suspension, or a ‘sol’. During the sol-gel process, the particles condense in a new phase in which a solid macromolecule is immersed in a liquid solvent phase. Through drying the solvent with low temperature treatments, it is possible to obtain porous solid matrices. Applied to the manufacturing of nuclear fuel microspheres, actinide compounds can be dissolved in superheated water with nitric acid and agitated until a stable sol or solution forms. Chemical reactions and physical changes, such as reduction, precipitation, hydrolysis, peptization, addition of surface active agents, and heating, convert the material to a solid gel.

Actinide oxide microspheres produced by the sol-gel process have been used as the kernel in experimental TRISO fuels as well as the starting material for actinide carbides and nitrides. The sol-gel process has also been used for formation of zirconia as an inert matrix for minor actinides. The ceramics resulting from the sol-gel process have densities greater than 97% of theoretical densities at relatively low sintering temperatures. Sintering temperature is shown to impact final density, with lower densities occurring at lower temperatures.

Spheres of MOX and Np-MOX were produced by an internal gelation process at the Paul Scherrer Institute, in connection with the FUJI irradiation experiment. The starting uranyl nitrate, plutonium nitrate, and neptunium nitrate solutions were prepared by dissolving actinide oxide powders in nitric acid. The targeted metal proportions were 20Pu:80U and 20Pu:5Np:75U. The mixed metal solutions were then blended with a cooled hexamethylenetetramine (HMTA) + urea solution to result in a feed solution at about 273 K.

Figure 22 shows a schematic of the gelation unit setup. In general, feed solution flows into a feed line ending in a capillary which generates droplets. Depending on the desired sphere size, capillary diameter is adjusted and a vibration is applied. The droplets fall into silicone oil at a temperature of 376 to 379 K. Within the oil, droplets

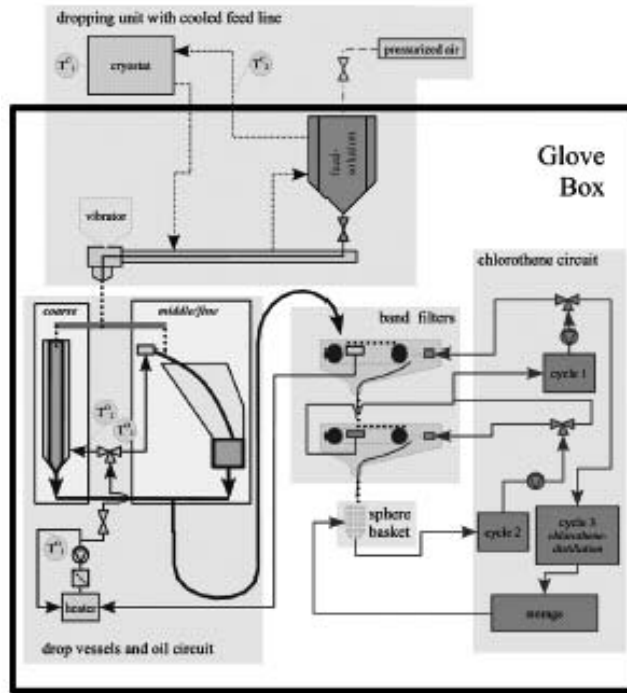


FIG. 22. Schematic of a gelation unit within a glove box.

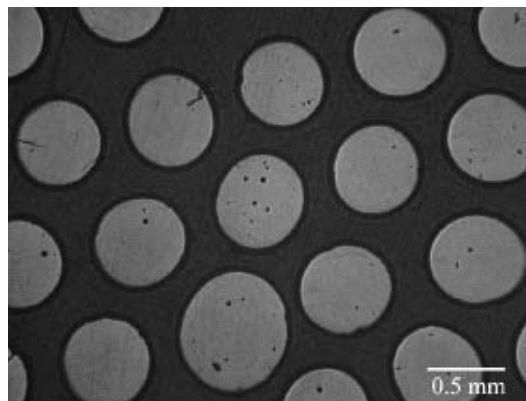


FIG. 23. Ceramograph of coarse Np-MOX sphere particles fabricated using internal gelation.

transform into spheres and solidify. The fabrication of coarse spheres follows a slightly different scheme than the fabrication of mid size and fine spheres. Coarse spheres directly fall into a hot silicone oil bath. Middle and fine spheres are injected into an oil jet, which then flows into an oil bath. An oil line transfers the spheres to a system of band filters.

With the first filter, the oil is removed by suction and transferred back to the oil circuit, and the spheres are rinsed by 1,1,1-trichloroethane. The spheres and 1,1,1-trichloroethane are transferred to a second band filter where potentially oil contaminated 1,1,1-trichloroethane is removed by suction and the spheres are rinsed with fresh 1,1,1-trichloroethane. The spheres are then collected in a basket. The gelation process is accomplished with a final washing step, in which residual reaction products and excesses of nitrate are removed using a diluted ammonia solution.

The spheres are dried in hot air and then calcined in Ar-7%H₂ at 873 K to convert hydroxides into oxides. Finally the spheres are sintered in a reducing atmosphere of Ar-7%H₂ at 1673 K for 4 hours. Humidity is used to adjust the oxygen to metal ratio to the required value of 1.97. Fig. 23 shows a ceramographic cross-section of a batch of Np-MOX particles. The sol gel process is utilized to group conversion of actinides and further fuel fabrication [150].

5.1.3. Sphere-pac process

Sphere-pac fuel consists of a blend of spheres of two or three different size fractions contained in a fuel rod. By controlling the relative fraction of the spheres, the smear density of the sphere-pac fuel column can be adjusted to values required for LWR fuel pellets ($\approx 91\text{--}95\%$) and/or to values typical of the fast reactor oxide fuel column ($\sim 85\%$) by controlling relative size fractions. Sphere-pac technology was developed between 1960–1990 for thermal and fast spectrum reactors of nearly all types of fuel cycle combinations (such as U–Th, U–Pu fuel and MA containing fuels, oxide and carbide fuels). Development of sphere-pac technology was motivated by the need for remote fabrication. For transmutation fuels, sphere-pac technology has the potential to be a lower cost alternative while also offering flexibility in tailoring fuel elements to match the requirements of any particular reactor core at any given time in the cycle. In fact, the blend of spheres can be adjusted to offer a different composition for each fuel pin or group of pins in a given fuel element. Moreover, it can provide a vertical composition gradient in a single fuel pin. For minor actinide bearing fuels, the sphere-pac form is likely to accept large helium releases from ^{241}Am transmutation with less difficulty than pellet forms and is especially well suited to remote fabrication of dustless fuel forms that require a minimum number of mechanical operations. The sphere-pac (and vi-pac) fuel forms are promising routes for the fabrication of MA bearing fuels.

5.1.4. Oxide electro-winning and vibro-pac process

Since the 1960s, the Research Institute of Atomic Reactors (RIAR) in the Russian Federation has been conducting research and development of an advanced fuel cycle based on the following elements:

- Pyroelectrochemical reprocessing of irradiated oxide fuel leading to production of reprocessed fuel in granulated form;
- Use of reprocessed oxide granules as vibro-pac fuel;
- Remotely operated recycling stages as part of a remotely controlled production line.

The pyrochemical technology being used is based on the production of pure and mixed actinide oxides from molten chloride systems. The behaviour of actinides such as Th, U, Np and Pu in molten alkali chlorides has been studied in detail [289–296]. The production of mixed actinide oxides is based on the following key properties:

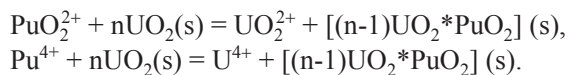
- In molten chlorides, uranium, plutonium and neptunium form complex oxygen containing ions of MeO_2^{n+} which are recovered as dioxides during electrolysis;
- Increasing oxygen content in molten salt causes decomposition of Th, Np and Pu chlorides into insoluble dioxides. They can undergo an exchange reaction with UO_2 and form solid solutions;
- Uranium dioxide is a conductor at temperatures above 400°C . During electrolysis of the melt containing UO_2^{2+} ions, crystals form and grow. Since the electric conductivity of UO_2 is less than that of molten salt, electrolysis results in stable plane crystallization and formation of compact cathode deposits. The electric conductivity of thorium, neptunium and plutonium oxides is negligible.

The mixed uranium and plutonium oxide production process has been studied and developed. The process is based on cathode collection of UO_2^{2+} , PuO_2^+ and PuO_2^{2+} ions in a molten $\text{NaCl}\text{--}2\text{CsCl}$ bath. During electrolysis, the UO_2 deposition rate is controlled by current density. The PuO_2 deposition rate is limited by diffusion of PuO_2^+ and PuO_2^{2+} to the cathode surface. The content of plutonyl ions in the melt is regulated by varying the oxygen potential of the melt using a $\text{Cl}_2\text{--O}_2$ gas mixture according to a specially developed algorithm.

Mixed oxides deposit on the cathode as a dense, two phase conglomerate of separate UO_2 and PuO_2 crystals. The deposit structure, such as dimension and form of separate crystals, and the strength of their coalescence to each other, depends on process parameters. Usually dimensions of PuO_2 crystals vary from 1–2 to 10–20 μm . Crystals of UO_2 are larger in size and contain 3%–7% Pu in solid solution. The formation mechanism of two phase deposits is based on substantial differences in their deposition potential (about 0.7 V).

Deposit density usually ranges from 80% to 87% of theoretical density, and depends on electrolysis conditions and the U to Pu ratio. Formation of pores during crystal growth results in a density of less than the theoretical density of the phases. As Pu content increases, the deposit density decreases. The correlation between deposit density and some characteristics of granulated product, such as vibropacking density and distribution of components by particle size, has been observed. An increased deposit density improves fuel performance.

Along with the direct electrochemical reduction of plutonyl ions, PuO_2 can be deposited on UO_2 substrate under the exchange reactions:



It is possible that these reactions provide for the formation of homogeneous solid solutions of $(\text{U,Pu})\text{O}_2$ containing up to 8% Pu.

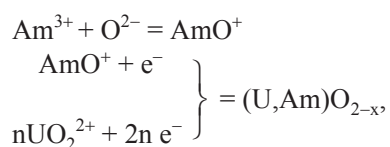
The electrochemical production of mixed deposits $(\text{U,Th})\text{O}_2$, $(\text{U,Np})\text{O}_2$, and $(\text{U,Pu,Np})\text{O}_2$ is based on the same principles: co-precipitation and/or exchange reactions. The electrical conductivity of Np and Th oxides is low, and passivation of the cathode is observed. Passivation increases as the content of these elements rises until the process terminates.

Neptunium is close to plutonium in its physical–chemical behaviour in molten chlorides. Research related to development of the method and processes for introducing Np into fast reactor MOX fuel has been carried out since the early 1990s. Based on the deposition potentials of NpO_2 in the main process melts (NaCl-KCl and NaCl-2CsCl), a codeposition model for U and Pu dioxides was adapted to incorporate Np into UO_2 and $(\text{U,Pu})\text{O}_2$ matrices. Experiments based on the model enabled the following fuel batches containing Np to be fabricated:

- $(\text{U},5\%\text{Np})\text{O}_2$,
- $(\text{U},3\%\text{Np})\text{O}_2 + 20\% \text{PuO}_2$,
- $(\text{U},6\%\text{Np})\text{O}_2 + 20\% \text{PuO}_2$.

Fuel pins with $(\text{U},5\%\text{Np})\text{O}_2$ have been irradiated in the BOR-60 reactor. Fuel pins in a mechanical mixture of $(\text{U,Np})\text{O}_2 + \text{PuO}_2$ are being prepared for irradiation.

The mechanism of incorporating Am into the growing cathode deposit of UO_2 may be described by the following reactions:



These reactions occur at high oxygen partial pressure and with a cathode potential of between -1.3 and -2.0 V.

The introduction of Am into UO_2 and the $(\text{U,Pu})\text{O}_2$ matrix using electrocodeposition has been experimentally investigated. Three compositions have been obtained from laboratory experiments:

- $(\text{U,Pu,Am})\text{O}_2$ containing 49% Pu and 2% Am;
- $(\text{U,Pu,Am})\text{O}_2$ containing 8% Pu and 5% Am;
- $(\text{U,Pu,Am})\text{O}_2$ containing 2% Pu and 3.5% Am.

These batches are being used to prepare experimental fuel pins for BOR-60.

All pyrochemical reprocessing spent nuclear fuel operations for the production of vibro-pac pins are carried out remotely in hot cell facilities [297]. Granulated fuel is ready for vibropacking into pins with a smear density of more than 8.8 g/cm^3 .

5.2. FABRICATION PROCESSES FOR CERCER FUEL

Cercer fuels have been fabricated using two principal techniques: (1) powder processing and (2) infiltration of a porous ceramic body with an actinide precursor. Several variations of each process are possible.

5.2.1. Fabrication of cercer fuel through powder processing

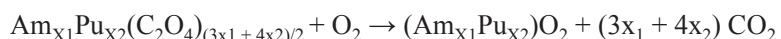
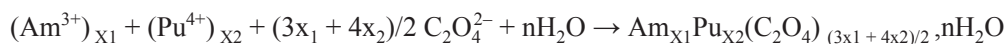
Fabrication processes for cercer fuel pellets through dry or wet mixing of ceramic matrix and actinide powders followed by pellet pressing and sintering have been demonstrated on a laboratory scale. Production of the actinide phase has generally involved precipitation of single actinide elements or co-precipitation of mixed phases.

5.2.1.1. Precipitation of the actinide phase

Precipitation of oxalates has been used to fabricate both AmO_2 and $(\text{Pu}_x, \text{Am}_{1-x})\text{O}_2$ at the CEA's ATALANTE laboratory [19]. This wet process route is a good alternative to minimize airborne contamination, although an aqueous waste stream is produced.

To produce americium oxide, americium was dissolved and purified through chromatographic extraction of Am(VI). Once reduced, Am(III) was precipitated as americium oxalate, filtered, calcined at 1073 K and sieved to produce pure americium dioxide granules of approximately 80 μm .

To produce a mixed americium–plutonium oxide solid solution, the oxalic co-precipitation of Am and Pu followed by calcination of the precipitate in an oxidizing atmosphere was undertaken. Sintering of this mixed oxide in a reducing atmosphere leads to $(\text{Pu}_{1-x}\text{Am}_x)\text{O}_{2-y}$ as shown below:



A single phase mixed oxide structure was confirmed by X ray analysis. Particle size distribution covers a narrow range, with over 99% of grains being smaller than 7 μm and the specific area value offers good performance for pressing. Photographs of mixed oxalate precipitate and calcined oxide power are shown in Fig. 24.

5.2.1.2. Fabrication of pellets

Americium oxide produced through the oxalate route was fabricated as microdispersion into an MgO matrix using the process flow shown in Fig. 25 for the ECRIX irradiation experiment, conducted at the CEA ATALANTE laboratory.

Magnesia powder was mixed with americium oxide through a classical powder metallurgy route. Magnesia powder was calcined at 1073 K to eliminate adsorbed water. MgO powder was compacted into disks at 150 MPa, which were then crushed and sieved to obtain granules with a diameter of 150 μm . The MgO granules were then

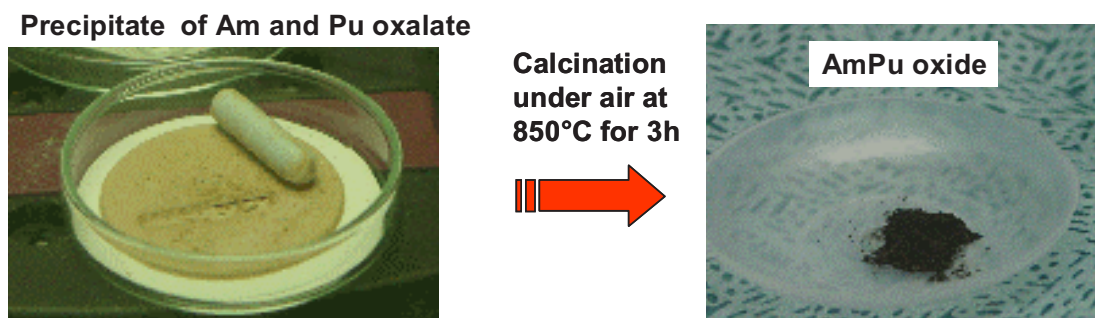


FIG. 24. $(\text{Pu}, \text{Am})\text{O}_2$ powder from Am and Pu oxalate.

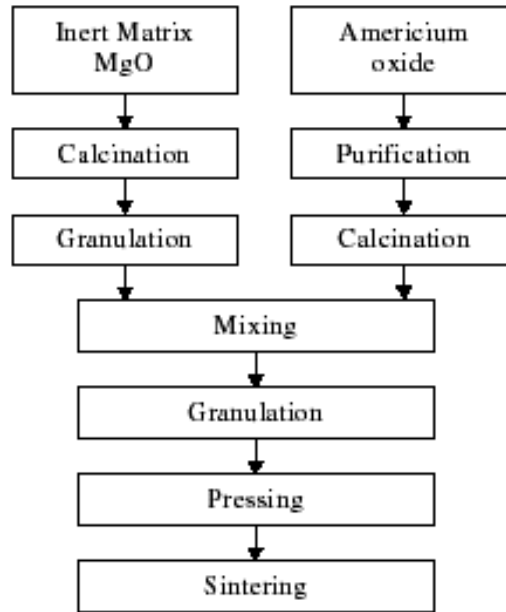


FIG. 25. ECRIX fabrication flowsheet.

mixed with americium oxide using an ellipsoidal movement in a mechanical mixer. In order to improve the distribution homogeneity of both components, the mixed powder was compacted into pellets at 150 MPa, and then pulverized through a sieve (in a specially designed granulator with low powder retention) to obtain granules with a diameter of about 150 μm . These granules were remixed and re-pressed in a single acting press at 250 MPa to fabricate green cerer pellets. The pellets were sintered in two steps, first at 1273 K for 4 hours in an argon/hydrogen atmosphere to fix the oxygen to metal ratio of the americium oxide, then at a temperature of 1773 K in an argon atmosphere for 4 hours.

The fabrication process of magnesia matrix uranium/plutonium oxide pellets for irradiation in the FUTURIX-FTA (FUels for Transmutation of transURanium elements in phenIX - Fortes Teneurs en Actinides (high actinide content) experiment is shown in a flowchart (Fig. 26). The fabrication process has been widely investigated with surrogates and active precursors not described here [298].

In summary, mixed Pu/Am oxide and magnesia were blended in a ball type mixer mill, and then formed by uniaxial pressing. The sintering profile was determined based on the sintering behaviour of MgO matrix. The holding temperature was set at 1600°C and the atmosphere was changed from argon/hydrogen to a neutral atmosphere at 1000°C to avoid reducing americium oxide to metal form.

Photographs of sintered $(\text{Pu}_{0.5}\text{Am}_{0.5})\text{O}_2 + \text{MgO}$ pellets are shown in Fig. 21. Two pellet compositions were fabricated: $(\text{Pu}_{0.5}\text{Am}_{0.5})\text{O}_2 + \text{MgO}$ and $(\text{Pu}_{0.2}\text{Am}_{0.8})\text{O}_2 + \text{MgO}$. Figure 27 also shows that the microstructure of pellets produced through this route is homogeneous.

5.2.2. Infiltration of porous ceramic matrix

An alternative fabrication method for minor actinide bearing cerer fuels and targets is through infiltration of porous ceramic bodies with a liquid solution of minor actinides. This process has been developed principally at the Institute for Tranuranium Elements (ITU) [108]. As an example, the fabrication process for $(\text{Am}_{0.06}\text{Zr}_{0.78}\text{Y}_{0.16})\text{O}_{1.89}$ target material for the CAMIX-1 experiment is shown schematically in Fig. 28.

Yttria-stabilized zirconia (YSZ) spheres were produced using a sol-gel process. Feed solutions with a determined Y/(Zr+Y) ratio of 0.17 were prepared from Zr oxychloride and Y chloride in distilled water. Following the addition of a surface active agent and an organic thickener, the solution was dispersed into droplets using a rotating cup atomizer. The droplets were collected in an ammonia bath, where spontaneous gelation occurs. After aging, the resulting spheres are washed with distilled water, dried using an azeotropic (C_2Cl_4) distillation procedure, and calcined at 1123 K. These spheres have a polydisperse size distribution in the 40 to 150 μm range, a specific

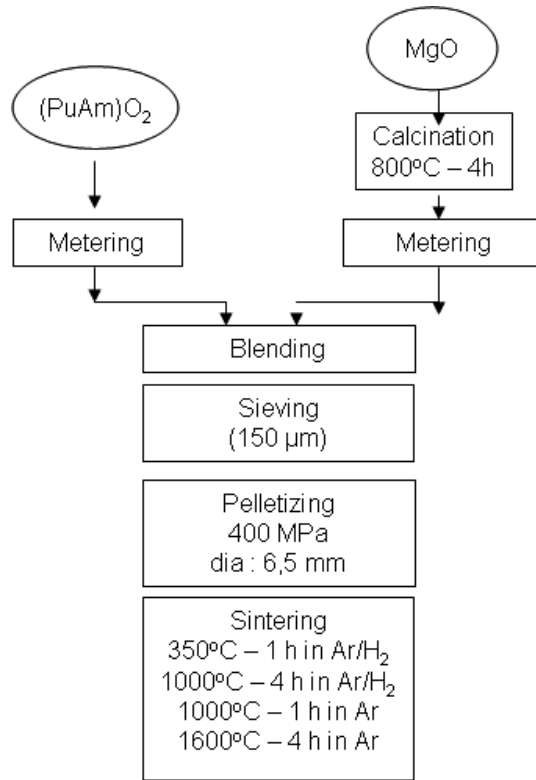


FIG. 26. $(Pu_{0.5}Am_{0.5})O_2 + MgO$ and $(Pu_{0.2}Am_{0.8})O_2 + MgO$ composite fabrication process.

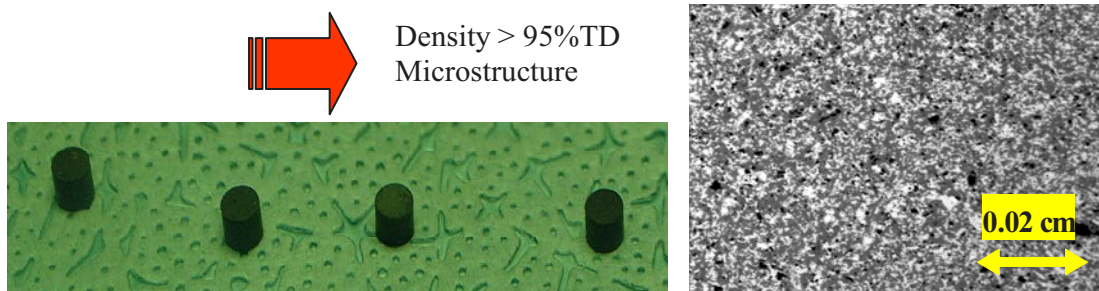


FIG. 27. Photograph of sintered $(Am_{0.3}Pu_{0.3})O_2 + MgO$ pellets at left. The micrograph at right shows the homogeneous distribution of phases in the microdispersion.

surface area of $67.2 \text{ m}^2/\text{g}$, and a porosity of approximately 80% of theoretical density. X ray diffractometry of sintered beads indicated a cubic crystal structure with a lattice parameter of $0.5140 \pm 0.0003 \text{ nm}$, which is in agreement with the value for $(Y_{0.15}Zr_{0.85})O_{1.93}$ ($0.5139 \pm 0.0001 \text{ nm}$).

The calcined $(Y,Zr)O_2$ spheres were infiltrated with cerium and plutonium nitrate solutions. For cerium, an infiltration solution with various metal concentrations (from 125 up to 200 g/L) was used, while for plutonium solutions the metal concentration was $216 \pm 1.2 \text{ g Pu/L}$, which is the maximum obtainable without the risk of Pu polymerization and precipitation.

Several infiltration/calcination cycles were carried out. Beads were thermally treated at 1073 K after each infiltration step to convert infiltrated metal nitrates into corresponding oxides. Metal content was determined by gravimetric analysis of the spheres before infiltration and after calcination. The resulting infiltrated beads were free flowing, and, due to their size and integrity, their physical manipulation did not produce dust in the following fabrication steps. At this stage in the fabrication process, little interdiffusion occurred, and X ray diffractometry measurements show that the materials are comprised of PuO_2 or CeO_2 phases dispersed in YSZ.

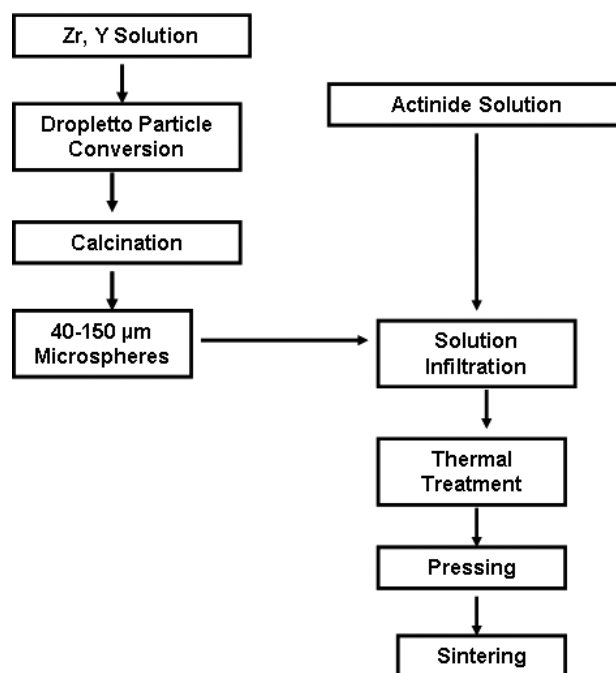


FIG. 28. Process flowsheet for fabrication of $(Am_{0.06}Zr_{0.78}Y_{0.16})O_{1.89}$ target material for the CAMIX-1 experiment.

5.2.3. Infiltration/powder processing

Minor actinide particles produced through infiltration can be formed into a cermet type composite by dispersion of minor actinide bearing microspheres in a suitable matrix. As an example, $(Am_{0.20}Zr_{0.66}Y_{0.14})O_{1.83}$ microspheres were dispersed in an MgO matrix for the CAMIX-2/COCHIX experiment [299–300].

The fabrication process for CAMIX-2 and COCHIX-3 pellets, shown schematically in Fig. 28, partially overlaps with that of the CAMIX-1 experiment described above. YSZ spheres were produced as described above. The $(Y,Zr)O_2$ spheres were then sieved and specific size fractions of 40–63 and 100–125 μm were selected. The sphere size fractions were next infiltrated in two consecutive steps with either a cerium nitrate solution (300 g Ce/L) or a plutonium nitrate solution (216 ± 1.2 g Pu/L). After each infiltration step, they were thermally treated at 1073 K in air for two hours to convert the infiltrated nitrate phase to a corresponding oxide. The $(Me,Y,Zr)O_{2-x}$ ($Me = Ce, Pu$) beads were mixed with a commercial MgO powder. Prior to mixing, the MgO was calcined at 1073 K and compacted into discs, which were then crushed and sieved to obtain granules.

Possible difficulties in powder processing lie in the potential for segregation of materials with differing densities and in accommodation of different granule and matrix sintering rates to prevent matrix cracking. Systematic investigations were performed to determine the optimum size of MgO granules and minimize these effects. These studies have shown that 50–71 μm MgO granules are required to produce cermet pellets without cracks and with a random distribution of isolated spheres. Pressing tests on MgO granules without inclusion of YSZ spheres led to densities of greater than 95% of the theoretical value after sintering. For composite fabrication, the granules were mixed with infiltrated YSZ spheres, compacted into pellets following the addition of zinc stearate as lubricant, and sintered at 1923 K for 8 hours in argon. Fig. 29 shows the resulting microstructures of fabricated pellets.

5.3. FABRICATION PROCESSES FOR CERMET FUEL

Cermet fuel and target materials are similar in concept to cermets, with substitution of a metal matrix phase for ceramic matrix. Metals in general have higher thermal conductivity than ceramic materials, and in some cases remain ductile during irradiation, which may prevent matrix cracking during irradiation [301].

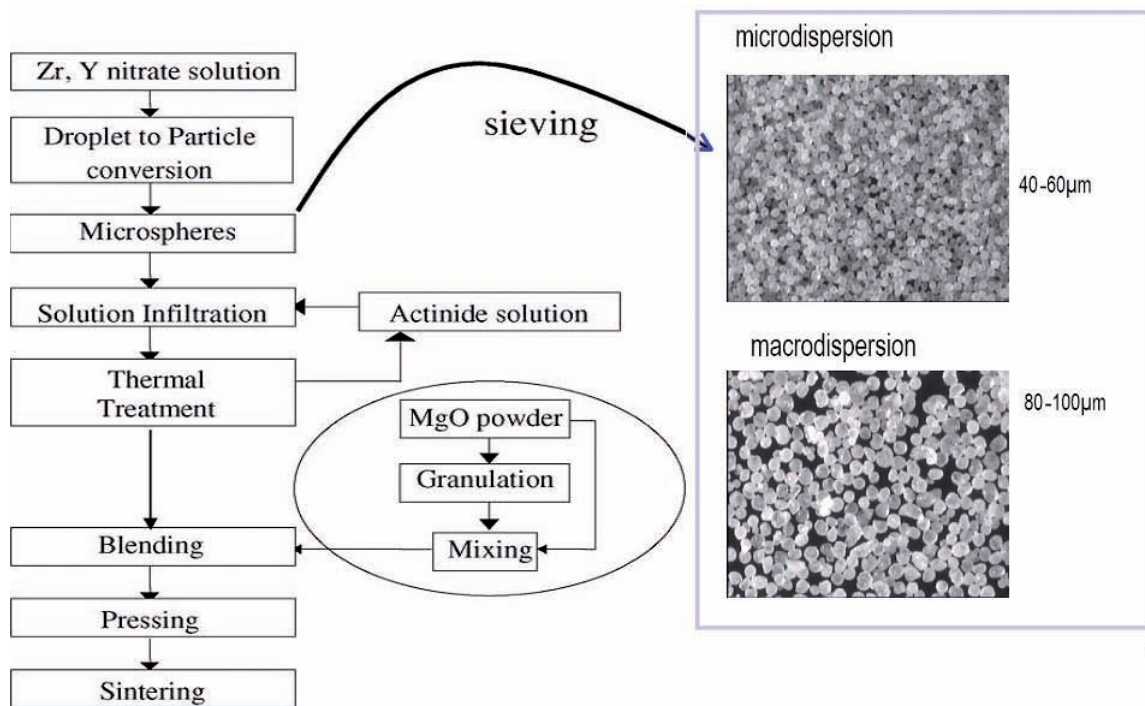


FIG. 29. CAMIX-2 and COCHIX-3 fabrication flowsheet showing the fabrication process for MgO matrix targets using the infiltration of microspheres followed by powder processing.

5.3.1. Infiltration/powder metallurgy

Molybdenum matrix cermet pellets with a dispersion of $(\text{Pu}, \text{Am})\text{O}_{2-x}$ particles of 3.05 g/cm^3 at actinide loading have been fabricated at ITU for inclusion in the FUTURIX-FTA experiment.

Preparation of these pellets is based on the fabrication of particles containing actinide phase through a combination of external gelation and infiltration methods [302–303]. Porous spheres of PuO_2 were produced by the external gelation method. After calcination at 1073 K in air, the spheres were infiltrated with an americium nitrate solution at 300 g Am/L. To reach a high americium concentration, the infiltration step was repeated following an intermediate thermal treatment to evaporate excess water and convert the infiltrated americium nitrate phase to oxide.

The final americium content — determined by gravimetric analysis of the beads before and after infiltration — was 21.7% by weight, which leads to a composition of $\text{Am}_{0.244}\text{Pu}_{0.756}\text{O}_{2-x}$. The infiltrated $(\text{Pu}, \text{Am})\text{O}_2$ actinide microspheres were mixed with molybdenum metal powder. Following addition of zinc stearate as lubricant, the powder mixtures were compacted into pellets using biaxial pressing. Sintering was performed at 1873 K for 6 hours in an Ar/H_2 atmosphere. The final density of pellets was in the range of 90% to 95% of the theoretical density. Characterization of the fuel shows a homogeneous phase distribution. Photographs of fuel pellets are shown in Fig. 30. Fabrication of the fuel requires use of an Ar/H_2 mixture in the sintering step. Thus, actinide oxide is reduced to an O/Am ratio of about 1.6. The oxygen to metal ratio has not yet been determined for Pu, but an O/Pu ratio in the range of 1.8 to 1.9 can be expected. The chemical activity of oxygen freed through the fission process under these substoichiometric conditions is not expected to be high enough to cause significant oxidation of the Mo.

5.4. FABRICATION PROCESSES FOR METAL FUEL

5.4.1. Injection casting

The metal injection casting process utilizes differential pressure to drive molten fuel alloys into molds, where the metal is rapidly solidified. A typical injection casting system consists of an induction heated graphite crucible

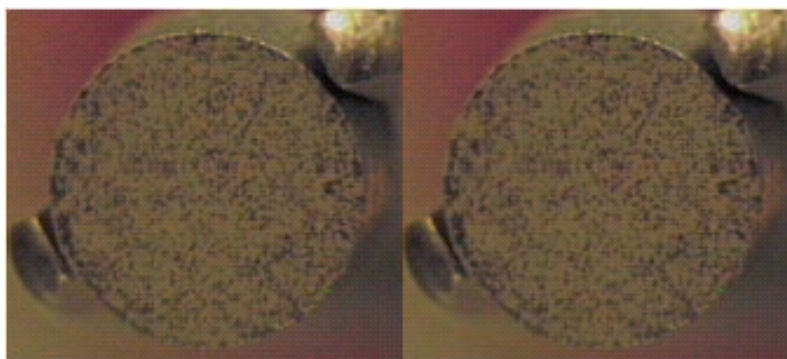


FIG. 30. Photographs of $(Am_{0.244}Pu_{0.756})O_{2-x}$ — Mo pellets loaded with 30 vol% of the ceramic phase.

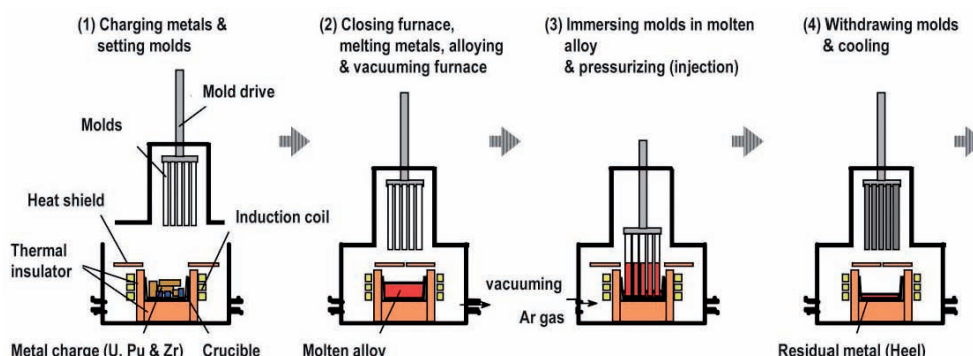


FIG. 31. Injection casting system for metal fuel [304].

and a moveable mold pallet containing 100 or more quartz molds. The crucible, induction coil, and mold pallet are enclosed in a vacuum/pressure vessel. A system for furnace evacuation and gas pressurization is used to control the injection casting process. A typical system schematic and process flow is shown in Fig. 31.

Fuel charges to be melted are loaded into a graphite crucible, and brought up to a chosen temperature under an inert gas atmosphere for a sufficient time to homogenize the melt. After homogenization, the melt temperature is adjusted to the casting temperature and the furnace chamber is evacuated, thereby evacuating the casting molds. The mold pallet is then rapidly lowered so that the ends of evacuated molds are held below the surface of the melt, and the system is pressurized, forcing molten fuel into the molds. The mold pallet is withdrawn and allowed to cool. After cooling, the molds are removed from the fuel, which is cut to size. End crops are recycled into the next melt. The charge is generally only limited by criticality considerations, and a large amount of fuel rods can be fabricated quickly.

EBR-II experimental fuel pins for the X501 minor actinide transmutation experiment [20] (see Section 6.2.3) were fabricated in this way. A considerable amount of americium was lost in the injection casting process due to volatilization of calcium and magnesium impurities present in the americium feed stock [305]. Such impurities are not expected in normal feedstocks, and substantial improvements with minimal americium volatilization are predicted with properly prepared feed stocks and optimized casting parameters.

5.4.2. Arc casting

An arc-casting process was developed at JRC-ITU in cooperation with CRIEPI. U–Pu–Zr and U–Pu–MA–Zr alloys were prepared by arc melting in an argon atmosphere. To homogenize the alloys, a U–Pu or U–Pu–MA alloy was first prepared by arc melting and then molten Zr was added. On the other hand, it is difficult to prepare homogenized alloys containing highly dispersed rare earth metals only through melting because the miscibility between rare earth metals and the U–(Pu–MA)–Zr matrix is low even in the liquid phase. In these cases, powders of U–Pu–Zr and rare earths were first prepared and then blended mechanically before melting. According to the

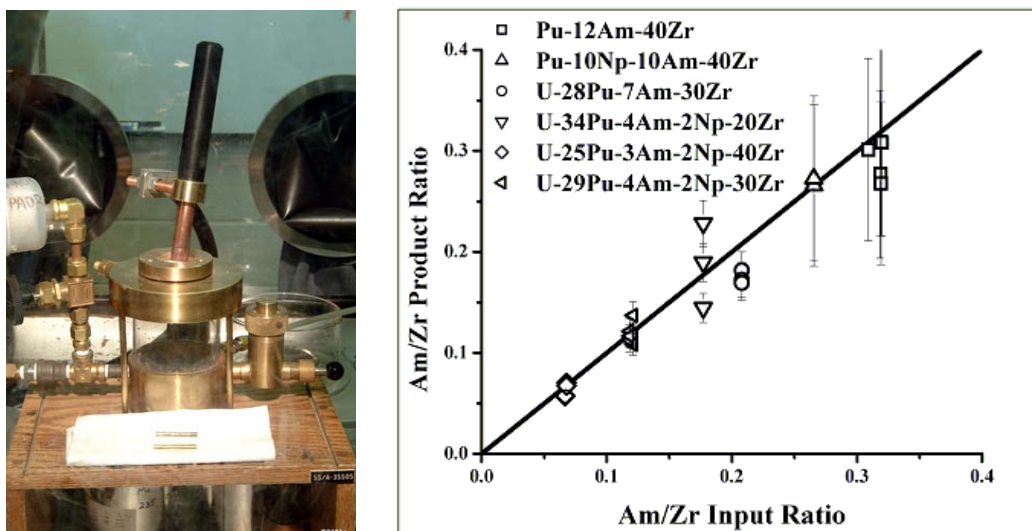


FIG. 32. Laboratory scale arc casting furnace and Am retention results.

metallography of an alloy obtained through arc melting, in U–Pu–Zr powder containing 7 wt% rare earths, the rare earth phase dispersal in actinide matrix was sufficiently homogeneous. Four types of alloy were cast into metal fuel rods in yttria molds which prevented reaction with molten alloys, including rare earths. Some of the fuel rods were irradiated in the METAPHIX experiment in Phenix.

The arc casting process was also developed to fabricate metal alloy fuel slugs at Idaho National Laboratory for the AFC-1 series of irradiation tests in the Advanced Test Reactor (ATR). The apparatus is shown in Fig. 32. Over 75 cm of 4 mm diameter test fuel pins were fabricated for ATR irradiation using a gravity drop method. Buttons of fuel alloy were first prepared using a standard arc melting hearth to produce a homogeneous product. The homogeneously mixed alloy button was then transferred to a casting hearth designed to allow the alloy button to be liquefied and poured through an opening in the hearth base into an yttria-coated quartz tube mold. Americium loss was tracked by chemical analysis before and after melting. The significance of this development is that rapid melting techniques have been shown to reduce the loss of volatile americium during the fabrication of Am containing alloys. Americium losses are typically within the uncertainty in chemical analysis, as shown in Fig. 32.

The drop casting technique proved to be inconsistent when attempted with 5 mm diameter fuel slugs (a 56% increase in cross sectional area) for insertion in the FUTURIX-FTA experiment. Greater fabrication consistency was achieved through adaptation of the injection casting process, in which differential pressure was used to draw metal alloy into the quartz mold. FUTURIX-FTA fuel pin slugs in the order of 2–3 cm in length and having densities of $\geq 95\%$ of the theoretical amount were produced using this technique. Although successful for fabrication of laboratory scale test fuels, the arc casting method, regardless of variation, is not intended for scale-up.

5.5. FABRICATION PROCESSES FOR NITRIDE FUEL

5.5.1. Carbothermic reduction/powder metallurgy

Nitrides in the Np–Pu–Am–Cm system have been fabricated at JAEA using carbothermic reduction [306–309]. With this method, actinide oxides are mixed with carbon, compacted, and heated in flowing N_2 gas to effect the following reaction:



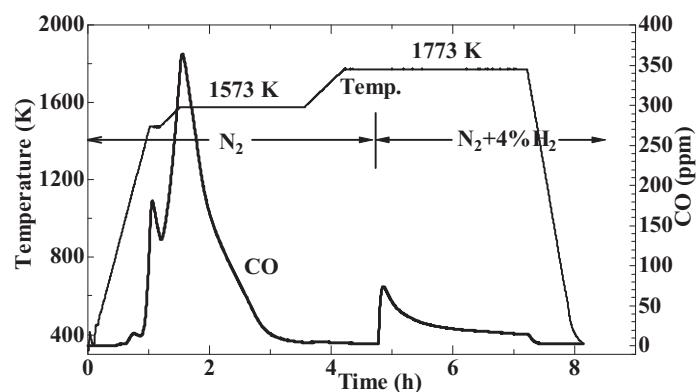


FIG. 33. Typical CO release curve during carbothermic reduction of oxides with carbon under a nitrogen atmosphere.

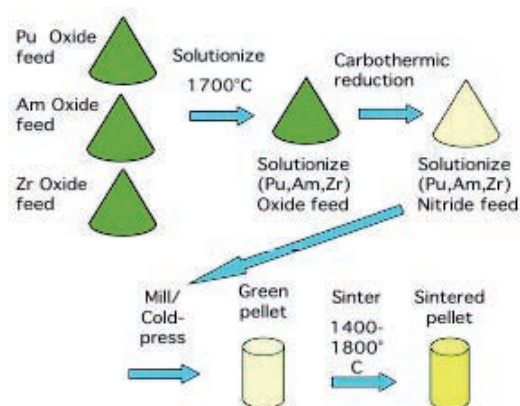


FIG. 34. Actinide nitride fuel processing schematic showing ‘solutionization’ of oxide feed materials prior to carbothermic reduction.

The initial carbon/metal ratio is usually chosen to be in 20–30% excess of the final stoichiometric ratio in order to obtain a nitride product with low oxygen content. The carbothermic reduction process is performed at 1573–1773 K in flowing N_2 gas. During heating, CO gas is released. Monitoring of the CO gas production rate (with an infrared spectrometer, for example) provides a useful tool for process control, as shown in Fig. 33. After confirming that the release of CO is complete, the flowing gas mixture is changed from N_2 to N_2-H_2 , and the process temperature is increased to approximately 1773 K to eliminate residual free carbon contained in the product.

Nitride fuel has also been fabricated using standard cold press and sintering methods beginning with feedstock powder in oxide form at LANL (see Fig. 34). The oxide powders are blended, milled, and then thermally treated to form a solid solution in oxide phase in an attempt to bind americium in a multi-constituent phase and minimize americium boil off. After carbothermic reduction and nitridation, nitride powder is milled, pressed, and sintered.

During fabrication into pellets, the sintering temperature selected was a tradeoff between sintered density and americium loss. The resulting process achieved a target density of 80% of the theoretical amount for non-fertile fuel composition, $(Pu_{0.50},Am_{0.50})N-36ZrN$. X ray diffraction data show at least two distinct phases in the sintered product, and difficulty in attaining a higher density for non-fertile fuel is most likely due to the lattice parameter mismatch between ZrN (inert phase) and the actinide nitride solid solution phase.

A low fertile composition $(U_{0.50}Pu_{0.25}Am_{0.15}Np_{0.10})N$ has proven to be easier to fabricate with a density of 90% of the theoretical amount readily achievable. X ray diffraction results for low fertile material show a single solid solution phase. Recent development work optimizing the carbothermic reduction process has led to a three fold reduction in residual carbon and oxygen levels below 5000 ppm for both low fertile and non-fertile compositions.

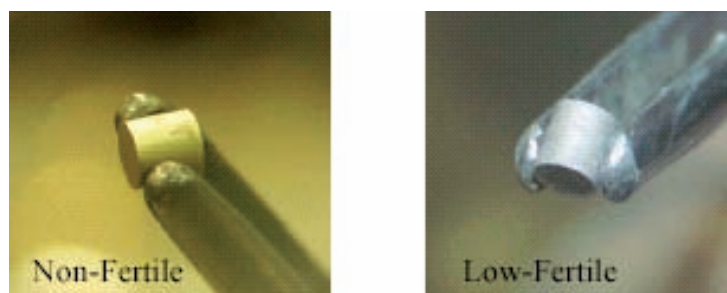


FIG. 35. AFCl non-fertile and low fertile nitride pellets.

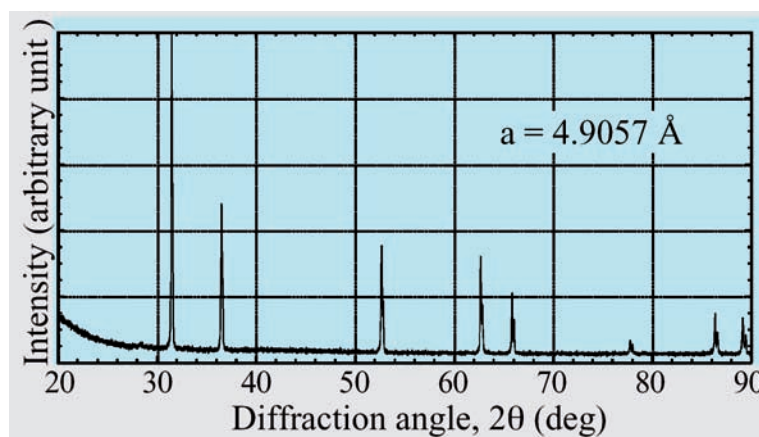


FIG. 36. X ray diffraction pattern of PuN recovered by a nitridation–distillation combined process.

Actinide nitride fuel pellets fabricated in this way have been used in the ATR to acquire irradiation data and also to establish process parameters for FUTURIX-FTA experiment designated fuel. Both non-fertile and uranium based fuels have been fabricated and irradiated, and the representative fuel pellets are shown in Fig. 35.

An alternative to mixing carbon and metal powders prior to carbothermic reduction is to incorporate carbon into the oxide feedstock using the sol-gel method. In this process, an actinide nitrate solution is mixed with an aqueous solution containing urea, hexamethylenetetramine, and dispersed carbon-black. The mixed solution is dropped into hot silicon oil to form ADU particles with internally dispersed carbon. The particles, after being washed and dried, are calcined at 600–700 K, and then heated in flowing N_2-H_2 to obtain nitride particles through carbothermic reduction.

5.5.2. Direct nitridation of metal

When pyrochemical processes are applied to nitride fuel, actinides are recovered at the liquid cadmium metal cathode [310–311]. When cadmium alloy containing plutonium and uranium are heated at 973 K under a nitrogen gas stream, the actinides are almost completely nitridated and cadmium is distilled simultaneously. Using this combined nitridation–distillation process, actinide nitrides with high purity have been successfully recovered. An X ray diffraction pattern of nitride material obtained using this process is shown in Fig. 36.

6. OVERVIEW OF IRRADIATION TESTS ON MINOR ACTINIDE FUELS

6.1. IRRADIATION BEHAVIOUR OF MINOR ACTINIDE FUELS

The irradiation behaviour of minor actinide fuels may vary from that of conventional fuels in several ways. Most prominent is the increased fuel pin helium gas inventory due to capture and decay sequences associated with ^{241}Am and a significant amount of ^{242}Cm , which decays by α emission (half-life of 169 days) to ^{238}Pu . The additional helium gas inventory can lead to higher fuel swelling rates and is an additional source term for fuel pin over-pressurization.

An additional interesting phenomenon relates to the evolution of isotopic mixtures in fuel with high ^{241}Am and ^{237}Np loadings, and the effect this evolution has on pin power. As plutonium isotopes are bred in fuel from neutron capture during irradiation, particularly in the thermal spectrum, fuel rod power increases as a function of irradiation time.

Other issues to be studied include fission product behaviour, and optimization of the oxygen to metal ratio in oxide fuels. Due to the shift in isotopic composition of the starting fuel, the isotopic and chemical distribution of fission products also shifts relative to that of standard MOX fuel. The distribution of fission products has not been quantified.

6.2. DESCRIPTION OF RELEVANT IRRADIATION TESTS

6.2.1. SUPERFACT-1

The SUPERFACT-1 experiment was developed to study irradiation behaviour of uranium oxide or MOX based fuels containing neptunium and americium in fast spectrum [22, 153, 154, 302, 303]. Minor actinide concentration in the fuel ranged from 2 wt.% to 45 wt.% of total heavy metal. The experiment was also designed to prove the feasibility of fabricating such fuels using standard processes. The experiment was performed jointly by the CEA and ITU, and irradiation took place in the French Phénix fast reactor between October 1986 and January 1988.

The SUPERFACT (Minor Actinide Irradiation in Phénix) experiment was irradiated in a standard 19 pin experimental Phénix cluster, in which 8 pins were fuels containing minor actinides. The pins had standard Phénix type external geometry using 15-15 titanium cold-worked (Ti CW) stainless steel cladding. Pellet diameter was 5.42 mm and height was 7 mm.

Two modes of transmutation were demonstrated; the homogeneous mode (no separation of Pu from minor actinides) used MOX based fuel with 2 wt.% NpO_2 or AmO_2 , while the heterogeneous recycling mode (separation of Pu from minor actinides) consisted of uranium based fuels containing up to 45 wt.% minor actinides. Table 12 shows initial fuel compositions. Three additional pins were MOX fuel containing titanium oxygen getters.

Experimental fuels were irradiated in the most outboard position (fourth row) of the Phénix reactor driver core. Maximum burnup of the actinide pins was calculated to be 6.8 at.% for low actinide concentration pins and around 4.5 at.% for high actinide concentration pins. Burnup determination by chemical analyses of ^{148}Nd showed strong agreement with data from calculated burnup. Standard oxide pins reached a burnup of 8.5 at.% within 383 effective full power days.

For standard pins, the linear power rate continuously decreased from 430 W/cm at the beginning of life to 380 W/cm at the end of life. The irradiation conditions for actinide pins were clearly different. Contrary to the behaviour of standard MOX or UO_2 fuel, the linear heat generation rate of pins with high actinide concentration increased continuously during irradiation. This behaviour is due to the formation of ^{238}Pu and ^{239}Pu from the ^{237}Np n- γ reaction and from alpha decay of ^{242}Cm in americium containing fuels.

Post-irradiation examinations of the fuel pins were completed, including profilometry measurements, gamma scanning, fission/helium gas measurements, metallography, burnup analysis, and microprobe analysis. Table 13 shows measured and calculated transmutation rates for neptunium and americium based on these analyses.

TABLE 12. COMPOSITION OF FUELS AND IRRADIATION CONDITIONS IN PHÉNIX

Pin Number and Type	Composition	Burnup (at%)		Linear Power (W/cm)
		CEA Calculation	ITU Measurement	CEA Calculation BOL and EOL
1, 2, 9, 10, 11, 12, 17 and 18, standard	(U _{0.718} Pu _{0.282})O _{1.983}	8.5		430/380
7 and 13, 2%Np	(U _{0.741} Pu _{0.244} Np _{0.0150})O _{1.973}	6.8	6.4	380/325
4 and 16, 2%Am	(U _{0.745} Pu _{0.237} Am _{0.0184})O _{1.957}	6.8	6.4	380/325
5 and 15, 45%Np	(U _{0.552} Np _{0.4482})O _{1.996}	4.6	4.5	206/283
6 and 14, 20%Np-20%Am	(U _{0.596} Np _{0.2118} Am _{0.1918})O _{1.926}	4.3	4.5	174/273

TABLE 13. COMPARISON OF CALCULATED AND MEASURED TRANSMUTATION RATES

Fuel Type	Np depletion (%)		Am depletion (%)	
	Calculation	Measurement	Calculation	Measurement
2%Np	24.3–24.7	30.2	—	—
2%Am	—	—	24.9–25.3	28.1
45%Np	26.3–27.6	26.3	—	—
20%Np–20%Am (pin 14)	29.0	34.4	26.6	26.0

Profilometry measurements showed normal clad deformation with the exception of the pins containing a high level of americium (pins 6 and 14), which presented slightly higher values of swelling indicative of the onset of fuel clad mechanical interaction. An axial expansion of the fuel column greater than 2% was also noted by neutron radiography for pins containing high levels of minor actinides (pins 5, 6, 14, and 15). Gamma scanning showed that fission products such as ¹³⁴Cs and ¹³⁷Cs were axially displaced in the fuel column. Both the degree of this movement and the axial locations of accumulation were similar to standard oxide pins irradiated under the same conditions.

Release of xenon and krypton fission gases was 60–70% of fission gas yield for all actinide pins. Fission gas release data are shown in Table 14. This fission gas release rate is similar to that expected from standard oxide pins under the same irradiation conditions. The pins containing americium had a notably elevated volume of helium gas. The helium production rate for pin 6 (20% Am — 20% Np) was 50 cm³ of He per gram of ²⁴¹Am transmuted, typical for irradiation in fast spectrum reactors. The volume of helium measured in the plenum of the 2% Am pin was roughly four times greater than that measured in standard MOX fuels. This additional gas inventory has little impact on pressure driven cladding creep for fuels with low americium content, but may determine fuel lifetime at higher americium loading. More than 80% of the gas inventory in the 20Am–20Np pin was helium. Retained helium was measured using sublimation in this pin. Results are consistent with the hypothesis that 100% of helium produced upon irradiation is released from fuel pellet to plenum, while only about 58% of that produced by decay of ²⁴²Cm during cooling after irradiation is released, resulting in a high occluded helium gas inventory. This may be of consequence, for example, in fuel that has been irradiated, cooled over a significant time period, and then subjected to a startup transient.

TABLE 14. GAS RELEASE DATA FROM SUPERFACT-1

Fuel Type	Calculated Xe+Kr inventory (cm ³)	Measured Xe+Kr in plenum (cm ³)	Xe+Kr release (%)	He production irradiation/ cooling	Measured He in plenum (cm ³)	Percent He in plenum gas ^a
Pin 7 2%Np	226.68	150.7	66.5%	—	14.2	8.6%
Pin 4 2%Am	225.0	154.1	68.5%	—	39.7	20.5%
Pin 5 45%Np	82.2	50.4	61.3%	—	8.1	13.8%
Pin 6 20%Np-20%Am	74.5	48	64.4%	Calculated: 147/134 Measured:285	225	82.4%
Pin 11 Std. MOX	250.3	169.3	67.7%		10.0	5.6%

^a Does not include helium fill gas.

Results of the SUPERFACT experiment indicate that the irradiation behaviour of MOX fuel with elevated minor actinide content designed for homogeneous transmutation was satisfactory and not greatly different from the behaviour of standard mixed oxide fuels at up to 6.4 at.% burnup. Likewise, although some changes in behaviour were noted for fuels with high neptunium content, behaviour appeared to be satisfactory up to 4.5 at.% burnup. Fuel cladding chemical interaction was observed in minor actinide bearing pins, although no correlation between either type of actinide or actinide concentrations could be made. The measured depths of interaction layers were within the conventional range for standard MOX fuels. In fuels with high americium content, the generation of large quantities of helium may have increased swelling rate and caused mechanical interaction between fuel and cladding for pins 6 and 14, resulting in a higher than normal increase in fuel pin diameter during irradiation. This factor must be taken into account in fuel pin design.

6.2.2. EFTTRA-T4

The EFTTRA-T4 experiment was a pilot study for transmutation of americium in an inert matrix target (cercer type) [22, 227, 312–313]. A target containing americium dispersed in MgAl₂O₄ was irradiated in the High Flux Reactor (HFR) at Petten over 358 effective full power days from August 1996 to January 1998. The project was performed by members of the Experimental Feasibility of Targets for Transmutation (EFTTRA) group. EFTTRA is a network of research organizations in France (EDF and CEA), Germany (JRC-ITU and FZK) and the Netherlands (JRC-IAM and NRG); it was formed in 1992 to study transmutation of both americium and long lived fission products (⁹⁹Tc and ¹²⁹I).

The inert matrix pellets for EFTTRA-T4 were prepared by infiltration of porous MgAl₂O₄ pellets with an americium nitrate solution. The sintered pellets had a diameter of 5.39 ± 0.03 mm with a density of $96.5 \pm 0.5\%$ of the theoretical value. It was found that during the sintering process, a reaction between americium oxide and spinel occurred to form a species with a perovskite structure. The actual composition was probably MgAl₂O₄ + AmAlO₃. Am content in the pellets was found to vary between 9.7 and 11.9 wt.%, with the mean value being 11.1 ± 0.7 wt.%.

The target pin contained 10 pellets loaded under an inert atmosphere (He with O₂ < 0.5% and H₂O < 10 ppm) into a 15/15 Ti stainless steel cladding tube (inner diameter 5.65 mm, outer diameter 6.55 mm). The pellet column was 70 mm long and was terminated at each end by a spinel pellet (MgAl₂O₄) with a length of 4.9 mm, a Hf pellet with a length of 6.0 mm, and a stainless steel pellet with a length of 17.0 mm. The cladding was welded using the tungsten inert gas (TIG) method.

The EFTTRA-T4 sample was irradiated in a channel of a standard in-core TRIO-131 rig in HFR. The irradiation lasted for a period of 14 cycles, equivalent to 358 effective full power days. The first eleven cycles were performed in core position C5 and the last three cycles in C7. Irradiation was interrupted after the first nine cycles

(231 effective full power days) for a period of one cycle in order to inspect the target capsule using neutron radiography.

The results of burnup calculations are shown in Table 15. They indicate that the extent of transmutation of ^{241}Am is about 96%, with ^{241}Am being transformed mainly to ^{238}Pu and ^{242}Cm . The extent of fission, the actual measure for destruction of actinides, is 28%. Also shown in Table 15 is actinide composition one year after the end of irradiation. This corresponds to the time of target capsule puncturing. It can be seen that at this time the amount of ^{242}Cm has been reduced by about 80% to ^{238}Pu as a result of alpha decay.

Non-destructive examinations, such as gamma tomography, profilometry, X ray radiography and gas puncturing were carried out on the irradiated capsule, and ceramography was carried out on the irradiated material. The most notable observations were large swelling of the matrix (~18%) and the formation of considerable porosity (~15%). Gamma tomography revealed that the diameter of pellets was about 5.75 mm after irradiation; about 6% larger than the initial value. A radial gamma scan recorded with higher accuracy has confirmed this value. The 5.75 mm pellet diameter was larger than the inner diameter of the cladding tube (5.65 mm) before irradiation. Expansion of the cladding must therefore have occurred and was confirmed by profilometry analysis.

The high swelling rate could be attributed to accumulation of helium (produced by alpha decay of ^{242}Cm) in gas bubbles. It is possible that the matrix was amorphous at the end of irradiation and may have recrystallized during the cooling period prior to examination. Large bubbles formed in the matrix mainly from helium generated by the ^{241}Am capture and decay sequence, but also from fission gases produced during irradiation. Fission products were distributed throughout the matrix and five metal precipitates were found in the matrix, often in association with gas bubbles.

Since the MgAl_2O_4 matrix shows a pronounced plastic deformation and associated high swelling, it should no longer be considered a candidate micro-dispersed inert matrix for actinide transmutation. Its unpredictable behaviour is linked to its poor chemical stability when used in conjunction with americium and its transmutation products, and its predilection to amorphise when irradiated in this configuration.

The latter conclusion has to be taken into account in the design of fuels and targets for americium transmutation. More recent concepts for heterogeneous targets mainly deal with minimizing the effects of radiation damage to the matrix. To achieve this, a dispersion of $(\text{Zr,Y,Am})\text{O}_{2-x}$ host particles between 50 and 300 μm in size in an inert matrix has been designed to restrict fission fragmenting and alpha particle damage almost completely to the host particle (see Section 6.2.7). In this case, helium gas (and fission gases) will accumulate inside the host particle and at the interface of the host particle and matrix.

As in the case of MOX produced using the MIMAS process, swelling might still occur at high burnup, when destruction of grains causes gas release to the host phase. This potential issue could be avoided by inclusion of tailored porosity to accommodate swelling or by providing channels for gas release to fuel pin plenum through decreasing the smeared density of fuel to allow swelling, or through use of non-pellet type designs such as sphere-pac fuels or targets.

TABLE 15. COMPOSITION OF MAIN ACTINIDE ISOTOPES IN THE EFTTRA-T4 TARGET AS OBTAINED BY POST TEST MCNP/FISPACT CALCULATIONS

Actinides	Initial (%)	Final (% initial mass of Am)	
		End-of-irradiation	EOI + 1 year
^{241}Am	100	3.7 ± 0.2	3.8 ± 0.2
^{243}Am	—	5.1 ± 0.2	5.1 ± 0.2
^{238}Pu	—	24.1 ± 0.1	36.3 ± 0.1
^{239}Pu	—	6.5 ± 0.1	6.5 ± 0.1
^{242}Pu	—	7.4 ± 0.5	7.5 ± 0.5
^{242}Cm	—	15.6 ± 0.1	3.1 ± 0.1
Total actinides	100	72.1 ± 0.2	72.1 ± 0.2

6.2.3. X501

The X501 experiment was conducted in the EBR-II reactor as part of the Integral Fast Reactor (IFR) programme to demonstrate minor actinide burning through use of a homogeneous recycling scheme [20, 314, 315]. The X501 assembly contains two metallic fuel pins loaded with relatively small quantities of americium and neptunium. Fuel slugs were fabricated into EBR-II Mk-IV fuel pin configuration. Chemical analysis and physical attributes of these pins are given in Table 16. Two minor actinide bearing pins were inserted into a standard EBR-II assembly while the remaining fuel pin locations were filled with U-10Zr driver fuel.

The X501 assembly was inserted into the EBR-II in February 1993 and withdrawn just prior to its shutdown in September 1994. Total irradiation time was 339 effective full power days. The burnup calculated by REBUS/RCT/ORIGEN was 7.6% with transmutation of 9.1% of ²⁴¹Am. The peak linear heat generation rate was estimated to be 45 kW/m (13.7 kW/ft), while peak fuel centerline and cladding inner surface temperatures were approximately 700°C and 540°C, respectively.

Of primary interest in this experiment was the effect of minor actinides on fuel cladding chemical interaction, the redistribution behaviour of americium, the quantity of helium gas release from the fuel and any effects of helium on fuel performance.

A partial post-irradiation examination was completed on X501, including gamma scanning, optical microscopy, microprobe analysis, and metallography. Gamma scans showed normal metallic fuel fission product behaviour; ¹³⁷Cs alloying with bond sodium and migrating to the region near the top of fuel slug. A microscopic examination of the inside cladding surface was made to determine whether the inclusion of minor actinides to U–Pu–Zr fuel has an effect on fuel cladding chemical interaction. Optical microscopy showed no evidence of reaction layer formation on the inner cladding wall or the outer surface of the fuel slug. A gap was visible between fuel and the cladding wall at all locations. These preliminary results indicated that under typical metal fuel operating conditions, fuel cladding chemical interaction behaviour was not affected by small amounts of americium or neptunium.

The irradiated fuel showed a microstructure typical of U-20Pu-10Zr, in which constituent radial redistribution resulted in the formation of three microstructural zones within the fuel. Microprobe analysis revealed that the outer zone, near the cladding, was enriched in zirconium, the intermediate zone was zirconium depleted, and the central zone was enriched in zirconium and depleted in uranium. Plutonium content remained relatively uniform. Americium appeared in the form of features with high elemental concentrations, generally depleted in U, Pu, and Zr. The morphology of americium rich phases could not be determined from micrographs.

TABLE 16. ATTRIBUTES OF X501 MINOR ACTINIDE BEARING FUEL PINS

Composition (average, wt%)	U-20.2Pu-9.1Zr-1.2Am-1.3Np
Major impurities (average, wt%)	Si: 0.26, Al: 0.089, Ca: 0.067, Cr: 0.0025, Mg: 0.009, Fe: 0.001, Mn: 0.001
U ²³⁵ enrichment (nominal)	60%
Fuel mass	77.5 g
Fuel length	34.3 cm (13.5 in.)
Element length (nominal)	74.9 cm (29.5 in.)
Cladding type	HT-9 steel
Cladding OD	5.84 mm (0.230 in.)
Cladding wall	0.457 mm (0.018 in.)
Slug diameter	4.27 mm (0.168 in.)
Plenum volume	7.1 cm ³
Plenum gas	75He–Ar
Smeared density	75%
Fuel slug density (% theoretical)	99.5%

Comparison of pre-irradiation and post-irradiation wavelength dispersive spectroscopy line scans suggested that americium rich features were formed during irradiation. Americium was present only in uranium depleted central and outer zones, indicating that americium migration had occurred, along with the migration of uranium and zirconium. Local radial redistribution of americium to the cladding inner wall did not occur; however these results do not rule out the possibility of condensation of americium in the plenum region above the fuel column.

Plenum gas sampling results were combined with ORIGEN calculations to estimate fuel fission gas and helium release rates. The results are shown in Table 17. A radioactive xenon tag gas was used in this experiment, which displaced some helium. The ratio of tag gas to helium displaced was estimated using several methods. Based on these estimates, it appears that approximately 90% of the helium gas produced was released to the plenum. The fission gas release was 79%, typical of U–Pu–Zr fuel at this burnup.

6.2.4. METAPHIX

The METAPHIX (Metal fuel irradiation in Phénix reactor) is an experiment devoted to homogeneous transmutation of minor actinides. The fast reactor U–Pu–Zr based metal fuel pins, containing 2 wt.% minor actinides with 2 wt.% rare earth elements, 5 wt.% minor actinides without rare earth elements, and 5 wt.% minor actinides with 5 wt.% rare earth elements were irradiated in the French fast reactor Phenix. The METAPHIX experiments are performed by the Central Research Institute of Electric Power Industry (CRIEPI) in cooperation with JRC-ITU and CEA [19, 316].

Nine metal fuel pins with four compositions were fabricated for the irradiation test: three pins of U-19Pu-10Zr, three pins of U-19Pu-10Zr-2MA-2RE, and three pins of U-19Pu-10Zr-5MA/U-19Pu-10Zr-5MA-5RE. The length of minor actinide containing specimens was limited to 10 cm due to the limited quantity of minor actinides available for this study. Minor actinide containing slug segments were sandwiched between two U-19Pu-10Zr slugs of 10 cm and 28.5 cm in length to make up an axial power distribution. CW15-15Ti cladding was provided by CEA/Cadarache for irradiation in the Phenix reactor. The fuel was sodium bonded and seal welded in a helium atmosphere. Fabricated metal fuel pins specifications are shown in Table 18.

TABLE 17. X501 GAS PRODUCTION AND RELEASE

BOL ²⁴¹ Am content	0.972 g
EOL ²⁴¹ Am content	0.884 g
²⁴¹ Am transmutation	0.088 g (9.1%)
Measured He release	3.1 cm ³
Calculated He inventory, 1 yr. decay	3.4 cm ³
He release	90%
Fission gas release	79%

TABLE 18. METAL FUEL PIN SPECIFICATIONS

Pin length (mm)	1,793
Outer cladding diameter (mm)	6.55
Cladding thickness (mm)	0.45
Fuel length (mm)	485
Fuel diameter (mm)	4.9
Initial fuel–cladding gap (mm)	0.375
Fuel smear density (%TD)	75.2
Sodium level above fuel (mm)	~10
Plenum length (mm)	464

Three experimental metal fuel pins of different compositions were arranged in each of three irradiation capsules, each capsule consisting of three metal fuel experimental pins and 16 oxide fuel driver pins. The three capsules correspond to three different discharge burnup targets: 2.4 at.% (120 EFPD, METAPHIX-1), 7 at.% (360 EFPD, METAPHIX-2), and 11 at.% (600 EFPD, METAPHIX-3).

The experimental capsules for METAPHIX-1 and METAPHIX-2 were loaded in the inner core and those of METAPHIX-3 were loaded in the outer core of the Phenix reactor in October 2003. Table 19 shows the calculated irradiation parameters.

The first capsule, METAPHIX-1, was discharged in August 2004. Irradiation was carried out up to a maximum burnup of 2.4–2.6 at.% and a maximum fast fluence of 3.8×10^{22} n/cm².

After cooling, the assembly containing the METAPHIX-1 experiment was dismantled. Non-destructive post-irradiation examinations in the irradiated element cell (IEC) of Phenix were conducted to study visual appearance and to examine the pins for local defects using photographs and videos. The length and diameter of irradiated capsules and pins were measured to detect deformation or swelling with $\sim 1/1000$ mm accuracy. No excessive deformation or critical damage was observed. The integrity of metal fuel pins containing minor actinides and rare earth elements was to be further examined.

Based on examination results, the remaining two METAPHIX capsules were irradiated without interruption towards the middle and high burnup goals of 7 at.% and 11 at.%. The fuel pins will be subject to detailed destructive examinations to characterize microstructure of the irradiated metal fuel and redistribution of minor actinides and fission product constituents. Further, minor actinide transmutation rates will be determined based on precise burnup measurements.

6.2.5. ECRIX-B and -H

The goal of the ECRIX-B and -H experiments was to test the feasibility of transmuting americium in heterogeneous targets in the context of monorecycling in fast reactors [317–318]. Two identical experimental pins were placed in the Phenix reactor under different irradiation conditions. Inside each pin was a 20 cm high column

TABLE 19. CALCULATED IRRADIATION PARAMETERS FOR METAPHIX-1, -2, AND -3

	No.1 U-19Pu-10Zr	No.2 U-19Pu-10Zr- 2MA-2REE	No.3 (bottom) U-19Pu-10Zr- 5MA	No.3 (top) U-19Pu-10Zr- 5MA-5REE
METAPHIX-1				
120 EFPD				
Linear power (W/cm)	330	308	325	313
Cladding temperature (°C)	572	572	572	572
Burnup (at%)	2.4 (max.)	2.5 (max.)	2.4 (max.)	2.6 (max.)
Cladding damage (dpa)	18 (max.)	18 (max.)	18 (max.)	18 (max.)
Fluence >0.1MeV (10^{22} n/cm ²)	3.8 (max.)	3.8 (max.)	3.8 (max.)	3.8 (max.)
METAPHIX-2				
360 EFPD				
Linear power (W/cm)	295	276	294	282
Cladding temperature (°C)	556	556	556	556
Burnup (at%)	6.9 (max.)	7.1 (max.)	7.0 (max.)	7.5 (max.)
Cladding damage (dpa)	54 (max.)	54 (max.)	54 (max.)	54 (max.)
Fluence >0.1MeV (10^{22} n/cm ²)	11 (max.)	11 (max.)	11 (max.)	11 (max.)
METAPHIX-3				
600 EFPD				
Linear power (W/cm)	268	251	269	256
Cladding temperature (°C)	543	543	543	543
Burnup (at%)	10.9 (max.)	11.2 (max.)	11.2 (max.)	11.9 (max.)
Cladding damage (dpa)	90 (max.)	90 (max.)	90 (max.)	90 (max.)
Fluence >0.1MeV (10^{22} n/cm ²)	19 (max.)	19 (max.)	19 (max.)	19 (max.)

made of cermet type composite pellets, in which particles of americium oxide were micro-dispersed in an inert matrix of MgO. The size of americium containing particles was between 1 and 50 μm . A classical powder metallurgy process was used successfully for pellet fabrication.

The irradiation undertaken for these experiments was completed in the Phenix reactor in mid 2007. Post-irradiation examination results indicated that magnesia based inert matrix targets experience moderate swelling under irradiation, even for significant values of helium production and burnup (in comparison with values corresponding to recycling studies in ADS and/or FR). In particular, measured experimental swelling is inconsistent with transmutation fuel pin and assembly pre-design [319].

6.2.6. CAMIX-COCHIX

The CAMIX-COCHIX experiment follows the ECRIX experiment and will test the behaviour of optimized transmutation targets [153, 317–318]. Evolutions in the experiment involve the mode of dispersion of particles into inert matrix and the stabilization of americium containing crystalline structure. This experiment will be irradiated in a moderated spectrum in the Phenix under conditions similar to those of the ECRIX-H experiment.

CAMIX-1 is a homogeneous americium–zirconium–yttrium ($\text{Am}_{0.06}\text{Zr}_{0.78}\text{Y}_{0.16}\text{O}_{1.89}$) mixed oxide target. CAMIX-2 is a cermet type composite in which particles of an americium–zirconium–yttrium oxide compound ($\text{Am}_{0.20}\text{Zr}_{0.66}\text{Y}_{0.14}\text{O}_{1.83}$) are microdispersed in a MgO matrix. The size of the americium containing particles is between 30 and 50 μm . COCHIX-3 is a cermet type composite in which particles of americium–zirconium–yttrium oxide compound ($\text{Am}_{0.20}\text{Zr}_{0.66}\text{Y}_{0.14}\text{O}_{1.83}$) are macro-dispersed in MgO. Each americium particle is between 90 and 130 μm in size.

6.2.7. FUTURIX-FTA

The FUTURIX-FTA (FUEls for Transmutation of transURanium elements in phenIX — Fortes Teneurs en Actinides [high actinide content]) is an international irradiation programme pursued jointly by the United States of America Department of Energy (DOE), the Institute for Transuranium Elements (ITU), the Japan Atomic Energy Research Institute (JAERI), and the CEA. The programme is designed to provide comparative data on the technical feasibility of burning minor actinides in fast spectrum reactors, mainly from the standpoint of fuel behaviour [320–321].

The irradiation test plan includes eight compounds summarized in Table 20. FUTURIX irradiation specimens, with the exception of nitride fuels, were inserted in the Phenix reactor during the first half of 2007 and scheduled for 240 effective full power days of irradiation. Calculated linear power, burnup and transmutation rates are also shown in Table 20.

TABLE 20. FUTURIX-FTA EXPERIMENTAL FUELS AND CALCULATED IRRADIATION PARAMETERS

Pin ID	Fuel Type	Composition	<i>Am/TRU</i> <i>content</i> (g/cm ³)	Max linear power (W/cm)	Burnup (% fima)	²⁴¹ Am Transmutation rate (%)
DOE1	Metal (low fertile)	35U-29Pu-4Am-2Np-30Zr (wt %)	0.5 / 4.0	272	7.0	20
DOE2	Metal (U-free)	48Pu-12Am-40Zr (wt %)	1.2 / 5.8	320	11.4	18
DOE3	Nitride (low fertile)	(U _{0.5} Pu _{0.25} Am _{0.15} Np _{0.10})N	1.7 / 5.7	327	5.5	20
DOE4	Nitride (U-free)	(Pu _{0.21} Am _{0.21} Zr _{0.58})N	2.7 / 5.3	287	10.2	20
ITU5	Cermet (U-free)	(Pu _{0.8} Am _{0.2})O ₂ + 86 vol% Mo	0.3 / 1.3	98	13.4	20
ITU6	Cermet (U-free)	(Pu _{0.23} Am _{0.25} Zr _{0.52})O ₂ + 60 vol% Mo	1.0 / 1.8	96	9.6	20
CEA7	Cermet (U-free)	(Pu _{0.5} Am _{0.5})O ₂ + 80 vol% MgO	1.0 / 2.0	104	9.2	20
CEA8	Cermet (U-free)	(Pu _{0.2} Am _{0.8})O ₂ + 75 vol% MgO	2.0 / 2.5	87	6.1	20

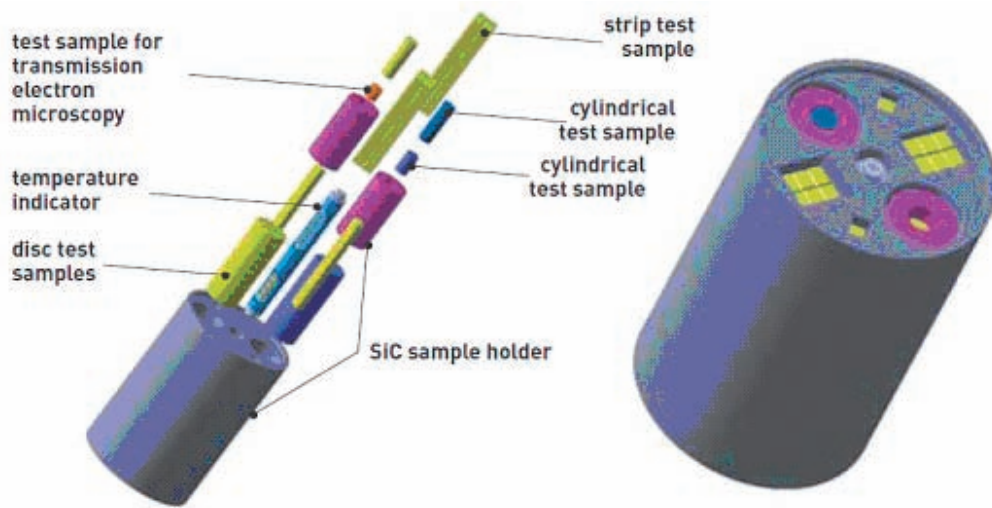


FIG. 37. Samples, and sample holder for the FUTURIX-MI experiment. (from Ref. 323).

FUTURIX concepts

In order to collect information on the behaviour of future GFR (gas cooled fast reactor) fuel under fast neutron irradiation, an experimental irradiation programme, called ‘Futurix-concepts’ [322, 323] has been launched at CEA. The concept under consideration is a composite material made up of a fissile fuel embedded in an inert matrix. Fissile fuel pellets are made of UPuN or UPuC, while matrices are SiC for carbide fuel and TiN for nitride fuel. The UPuC pellets are manufactured using metallurgical powder processes for the fabrication of four pins. Initial work is more focused on the development of a new fuel design (see Fig. 37). Development of innovative processes (for the shape of the fissile fuel and welding of the inert matrix) and characterization of different pellets are under way. Fabrication of the sample to be irradiated was undertaken in 2006 and irradiation began in May 2007 in the PHENIX reactor.

6.2.8. FUJI

The FUJI experiment (Fuel irradiation for JNC and PSI) is a collaborative project of JNC, NRG and PSI [203, 324, 325]. The performance of various MOX fuel configurations, including those containing Np, are compared through fuel fabrication and irradiation in order to obtain information on their suitability as fast reactor fuels. For this purpose, conventional pellets, vibro-pac, and sphere-pac fuels have been investigated. All of these fuels were fabricated at PSI and delivered to the High Flux Reactor (HFR) at Petten, the Netherlands.

Sixteen segments of three types of fuel were fabricated: nine sphere-pac fuel segments, two vibro-pack fuel segments, and five pellet fuel segments. Two sphere-pac segments had a composition of (5%Np,20%Pu,75%U)O₂.

Sphere-pac fuel segments were fabricated through infiltration filling using two MOX microsphere sizes (776 µm and 67 µm), with the exception of segment R21L. Segment R21L, intended to test low smear density sphere-pac fuel, was fabricated through simultaneous filling of MOX microspheres of two sizes (776 µm and 195 µm). Microspheres were fabricated using an internal gelation process described in Section 4. Table 21 summarizes characteristics of the FUJI fuel pins.

TABLE 21. CHARACTERISTICS OF FUJI FUEL PINS

	Sphere-pac fuel	Vibro-pac fuel	Pellet fuel
Fuel component	(Np _{0.05} Pu _{0.2} U _{0.75})O ₂ , (Pu _{0.2} U _{0.8})O ₂	(Pu _{0.2} U _{0.8})O ₂	(Pu _{0.2} U _{0.8})O ₂
Smear density	79.1~81.5%TD (R21L: 70.6%TD)	75.8~75.9%TD	88.8~89.8%TD
Fuel effective length	250 mm	250 mm	50 mm
Cladding dimension	7.50 mm OD, 6.70 mm ID		

Pin segments were connected two by two to form one pin, which was placed in a double walled stainless steel container at NRG. In each test, two fuel pins consisting of four segments were simultaneously irradiated in the pool side facility at HFR in the KAKADU irradiation rig. Irradiation test conditions resulted in higher linear power (53 kW/m) and lower cladding temperature (673 K) relative to conditions in fast reactors such as Joyo (41 kW/m and 783 K).

Testing focused on thermal behaviour of the fuels. Three restructuring tests and a power to melt test were performed. Restructuring tests IST (initial sintering test), RT1 (1st restructuring test), and RT2 (2nd restructuring test) were performed to obtain restructuring data for sphere-pac fuels. Irradiation patterns were: (1) after initial startup ramp in 36 hours to 53 kW/m, (2) after initial startup ramp and subsequent holding time at the linear heat rate of 53 kW/m for 48 h, and (3) after initial startup ramp and subsequent holding time at the same linear heat rate for 96 hours.

The power to melt test was performed to evaluate the melting linear heat rate of sphere-pac fuels in a fast reactor as well as to apply data for validation of sphere-pac fuel design codes. In the power to melt test, two sphere-pac fuel segments (P12U, P12L) and two pellet fuel segments (P11H, P11L) were irradiated. P12L had the composition (5%Np,20%Pu,75%U)O₂, while all other segments contained (20%Pu,80%U)O₂.

The results of these tests indicate that fast reactor particle fuels undergo a rapid structural change during the early phase of irradiation. The fuel may undergo sintering and pore migration to form a central void within a matter of hours, depending on fuel temperature. The central void formation and formation of a sintered zone adjacent to the central void result in a decrease in fuel temperature, providing a negative fuel temperature feedback mechanism. It was concluded that particle shape made little difference to fuel behaviour.

6.2.9. AFC-1 test series

The AFC-1 irradiation test series is designed to evaluate the feasibility of actinide bearing fuel forms in sodium cooled fast reactors for the transmutation of actinides from spent nuclear fuel. AFC-1B, AFC-1F, and AFC-1Æ irradiation test capsules provided irradiation performance data at intermediate burnups of 4 to 8 at.% on non-fertile and fertile actinide transmutation fuel forms containing plutonium, neptunium and americium isotopes [67, 326]. AFC-1D, AFC-1G, and AFC-1H capsules will extend fuel performance. AFC fuel test hardware was designed to simulate fast reactor test conditions in Idaho National Laboratory's advanced test reactor. The fuel rods had the same diameter as EBR-II fuel, but were reduced in length. All fuels, both nitride and metal, were sodium bonded inside stainless steel Type 421 (HT-9) cladding with an inert plenum gas. Six fuel rods were contained in each stainless steel capsule. The experiment was shrouded in a cadmium thermal neutron filter. Details of capsule and rodlet radial dimensions of metallic and nitride fuel specimens can be found in Refs [327–330].

The AFC-1B test capsule contained four non-fertile metallic fuel compositions, Pu-12Am-40Zr, Pu-10Am-10Np-40Zr, Pu-60Zr, and Pu-40Zr. The AFC-1F capsule contains four uranium bearing metallic fuel compositions, U-28Pu-4Am-2Np-30Zr, U-27Pu-3Am-2Np-40Zr, U-34Pu-4Am-2Np-20Zr, and U-29Pu-7Am. The AFC-1Æ capsule contained two non-fertile nitride fuel compositions, (Pu_{0.5}, Am_{0.5})N-36ZrN and (Pu_{0.5}, Am_{0.25}, Np_{0.25})N-36ZrN and one fertile nitride fuel composition (U_{0.5}, Pu_{0.25}, Am_{0.15}, Np_{0.10})N.

Gas release and microstructural data from 11 metallic fuel specimens indicate that these fuels follow the same behavioural trends as U-xPu-10Zr fuel when correlated with fission density [328]. There is a negligible amount of fission gas release until a fission density of $\sim 6.0 \times 10^{20}$ f/cm³. At this threshold burnup, there is a rapid increase in fission gas release rate, similar to U-xPu-10Zr fuel. In the fission density range of the samples examined, there was no difference noted in fission gas release behaviour between non-fertile (non-uranium bearing) and low fertile (uranium bearing) compositions. Based on gas release at this intermediate burnup, it is predicted that fission gas release for actinide containing fuels at higher burnup will show a slightly increased plateau. This will be evaluated during post-irradiation examinations of sibling experiments AFC-1D and AFC-1H, to be irradiated to a peak burnup of 40 at.% of heavy metal.

Comparisons of microstructures of fuels with high minor actinide contents to those for U-Pu-Zr fuel indicate similar microscopic and swelling characteristics at equivalent fission densities. Metal alloy fuels typically exhibit a well defined sequence of swelling and gas release behaviour. There is an incubation period characterized by a very low swelling rate up to a threshold fission density of $\sim 0.35 \times 10^{21}$ f/cm³ (1.0 at.% burnup). During the transition period between 0.35 and 1×10^{21} f/cm³, rapid fission gas driven swelling occurs until a network of open porosity develops. At an areal swelling of approximately 25%, the fuel contacts the cladding and the majority of the fission

gas inventory has been released to the plenum. Eventually, a stable plateau in swelling rate and fission gas release rate is reached. Swelling in this phase is primarily due to accumulation of solid fission products. Metallographic examination of low fertile metallic alloy composition at two burnups, 0.42 and $0.60 \times 10^{21} \text{ f/cm}^3$, indicate that the transition in swelling behaviour of minor actinide bearing metallic fuel occurs in a fission density range of $0.6\text{--}1.2 \times 10^{21} \text{ f/cm}^3$.

- (1) Fission gas release and helium burnup dependence of the five minor actinide nitride rodlets shows a threshold in fission gas release, with no release below $0.45 \times 10^{21} \text{ f/cm}^3$ [329]. Above this threshold, fission gas release ranges between 5 and 20%. Nitride composition with uranium has the highest burnup, but shows a moderate gas release of 10%. This may indicate lower fission gas release from low fertile nitride compared to non-fertile compositions. Metallographic examinations of $(\text{Pu}_{0.5}\text{Am}_{0.5})\text{N-36ZrN}$ and $(\text{U}_{0.5}\text{Pu}_{0.25}\text{Am}_{0.15}\text{Np}_{0.10})\text{N}$ samples exhibited slight areal swelling and significant cracking, fragmentation, and radial relocation of fuel. One specimen exhibited a fragment shard that would likely have caused premature failure in an He bonded fuel rod. The sodium bond ameliorates this fragment induced failure mechanism by lowering the thermal gradient and eliminating local hot spots from small fragments.

6.2.10. DOVITA

Since 1992, the Research Institute of Atomic Reactors (RIAR) in the Russian Federation has worked with the DOVITA programme concept (Dry reprocessing, Oxide fuel, Vibro-pack, Integral, Transmutation of Actinides), which is aimed at demonstrating new fuel cycle technologies for the transmutation of Np, Am and Cm [289–297, 331]. RIAR uses the BOR-60 reactor as the experimental base, because its fast neutron spectrum is beneficial for minor actinide burning.

Batches of $\text{UO}_2\text{-20PuO}_2\text{-(3-6)NpO}_2$ fuel have been produced and physicochemical and physicochemical properties of their components have been investigated. Fuel pins containing $(\text{U-5Np})\text{O}_2$ granulated fuel fabricated using vibro-pack technology do not differ qualitatively from regular BOR-60 vibro-pack fuel pins. The fuel pins were irradiated in the BOR-60 reactor to a peak burnup of between 12.5 and 19.5% heavy atoms. The fuel pin 20-H was irradiated up to the maximum damage dose for ChS-68 alloy cladding (85 dpa) allowed for BOR-60 fuel pins and reached a high burnup value. Destructive examinations for the 20-H fuel pin have been completed. The ^{237}Np mass reduction (^{237}Np transmutation) was 16.9% and 19.0% in the fuel pin central plane.

Irradiation conditions are graphically represented in Fig. 38. After irradiation, the fuel pins were investigated using non-destructive methods including visual inspection, eddy current defectoscopy, profilometry, and gamma scanning. Visual inspection and gamma scanning results are shown in Fig. 39. An example of ceramography is shown in Fig. 40, revealing good compatibility between ChS-68 alloy cladding and $(\text{U,Np})\text{O}_2$ fuel, as well as structural stability of the fuel. No essential differences in transmutation efficiency between fuel pins with UNpO_2 fuel and the fuel pins containing UO_2 or $(\text{U,Pu})\text{O}_2$ fuel were found. Distribution of main fission products in the irradiated fuel was typical of hypo-stoichiometric oxide fuel.

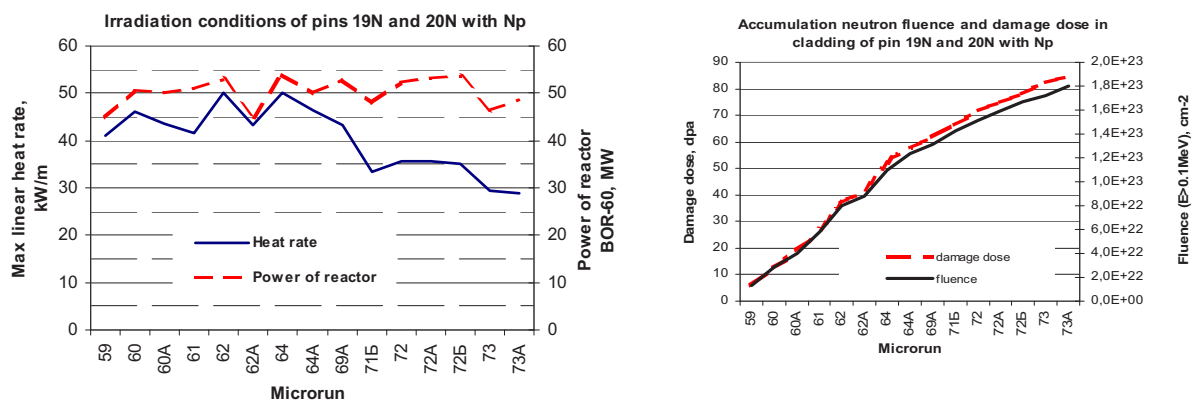
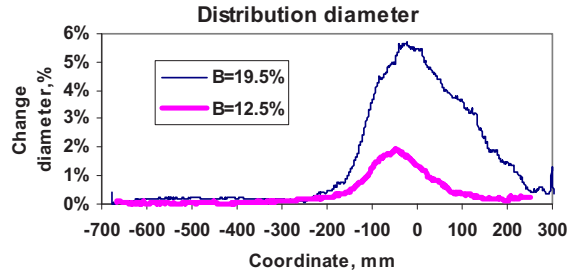
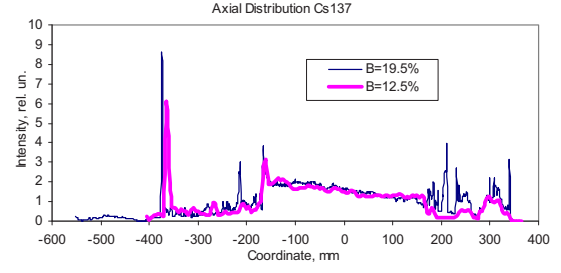


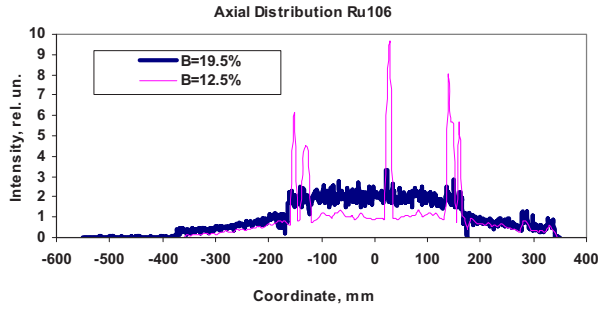
FIG. 38. Irradiation conditions of pins with $(\text{U, Np})\text{O}_2$ fuel in the BOR-60 reactor.



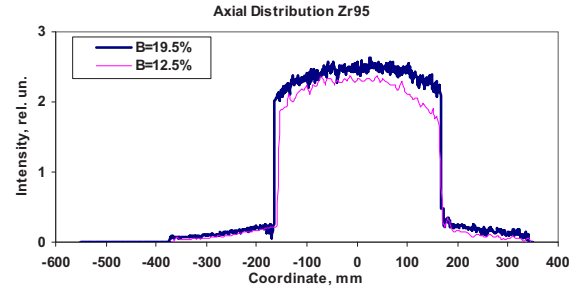
(a)



(b)



(c)



(d)

FIG. 39. Distribution diameter (a) and fission products, Cs(b), Ru (c), and Zr (d) along the fuel pins at 12.5 and 19.5% burnup.

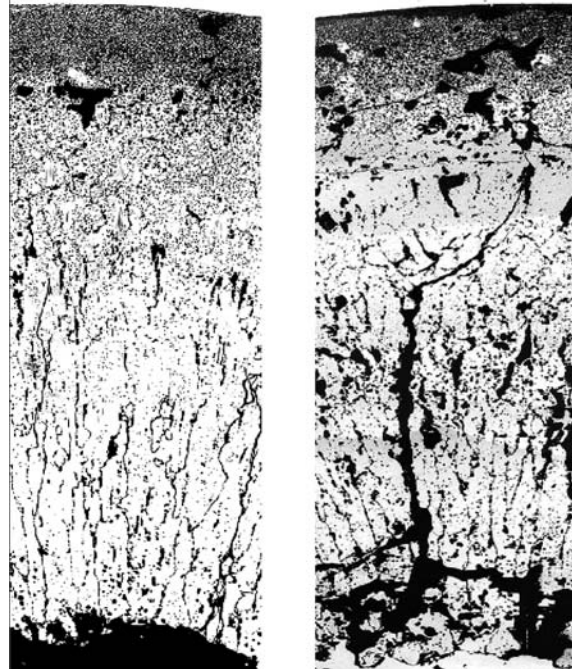


FIG. 40. Microstructure of UNpO_2 irradiated fuel and fuel-cladding boundary at 12.5% HM burnup.

The DOVITA programme has been slightly modified by RIAR to include ‘on-site reprocessing’ and ‘integration steps into fast reactor fuel cycle’, and is called now DOVITA-2 (DOVITA-2 = Dry technologies, On-site reprocessing, Various type of fuel with MA, Integration of MA recycling into FR closed fuel cycle, TA - Transmutation of Actinides (2006-8)). Thermodynamic measurements of Cm in molten chlorides have been reported [332] under the recent DOVITA-2 programme.

6.2.11. HELIOS

Previous irradiation experiments carried out on ^{241}Am bearing spinel targets have shown significant volume swelling resulting from helium production. As a consequence, helium release and trapping is considered a key parameter for fuel and target design. The HELIOS irradiation experiment (an irradiation test in HFR to study in-pile behaviour as a function of U-free Am target temperature) aims to increase knowledge between microstructure and in-pile fuel behaviour for minor actinides transmutations in 'once through' heterogeneous mode. For this programme, five pins containing either Am based cermet or cermet (solid solution) were prepared. The CEA was responsible for fabricating the pin containing $\text{Am}_2\text{Zr}_2\text{O}_7$ pyrochlore in MgO matrix, which was chosen for its satisfying thermal properties and irradiation behaviour. Research, development and fabrication steps carried out in the Cadarache and Atalante facilities to achieve this compound preparation are described in Refs [333–335]. In the first results on obtained pyrochlore, the materials exhibited the expected properties.

6.2.12. Am-1

The ‘Am-1’ programme is being conducted in order to investigate the irradiation behaviour of americium containing MOX fuel in the experimental fast reactor Joyo [336–341]. Two irradiation experiments were conducted in the Joyo MK-III 3rd operational cycle to research early thermal behaviour of MA–MOX fuel. Six prepared fuel pins included MOX fuel containing 3% or 5% americium (Am–MOX), MOX fuel containing 2% americium and 2% neptunium (Np/Am–MOX), and reference MOX fuel as illustrated in Fig. 41. The Am-1 programme consists of

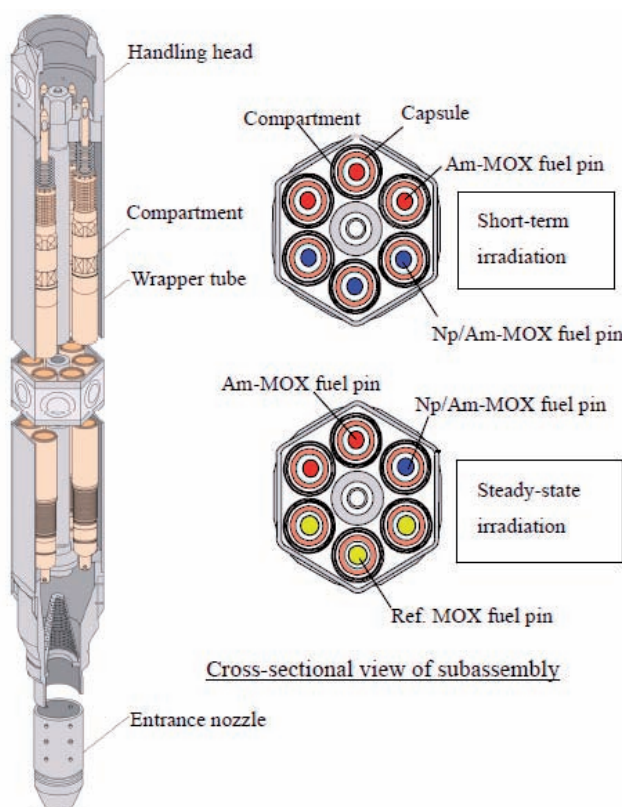


FIG. 41. Schematic view of capsule type irradiation test subassembly [338].

two short term irradiation tests of 10 min and 24 hours and a steady state irradiation test. The short term irradiation tests were completed and post irradiation examinations (PIE) are in progress. PIE results for Am containing MOX fuel irradiated for 10 minutes have been recently presented [337]. MOX fuel pellets containing 3% or 5% Am were fabricated in a shielded airtight hot cell using a remote handling technique. The oxygen to metal ratio (O/M) of these fuel pellets was 1.98. They were irradiated at a peak linear heating rate of about 43 kW m^{-1} . Focus was placed on migration behaviour of Am during irradiation. Ceramography results showed that structural changes such as lenticular pores and a central void occurred early, within the brief 10 min of irradiation. The results of electron probe microanalysis revealed that the concentration of Am increased in the vicinity of the central void. Post irradiation examination of these pins to confirm fuel melting and local concentrations under irradiation of NpO_{2-x} or AmO_{2-x} in the $(\text{U,Pu})\text{O}_{2-x}$ fuel are underway. Test results are expected to reduce uncertainties in the design margin for the thermal design of MA-MOX fuel.

6.2.13. AMBOINE

RIAR, in collaboration with CEA, France initiated a programme called AMBOINE-1 (design and production of americium target fuel pin for in-pile tests in the BOR-60 reactor) in December 2001 [332]. The main objective of this study is americium recycling and its separation from rare earth elements using pyrochemical process. The development and fabrication of $(\text{UAm})\text{O}_2$ fuel pins and $\text{UAmO}_2 + \text{MgO}$ as axial blankets as well as testing in the BOR-60 was completed in 2006. Investigations on americium/rare earth element separation through selective oxide precipitation in molten salts has been carried out. The possibility of AMBOINE-2 continuation by including a comparative study of different fabrication techniques (vi-pack, sol-gel, GRANAT⁷) and further irradiation in the BOR-60 reactor is under consideration.

6.2.14. MATINE (ISTC Project#2680)

RIAR has also been studying transmutation of nitrides with MA covering modelling and measurements of out of pile properties since 2004 under International Science and Technology Center (ISTC) Project#2680, also called 'MATINE' in collaboration with IPPE, and the Bochvar Institute of the Russian Federation, CEA, France and KTH, Sweden [99, 332]. The objectives of the project are: (a) performance modelling of $(\text{Pu,Am,Cm,Zr})\text{N}$ (with $\text{ZrN} = 55\text{--}65\%$, $\text{Pu/Am/Cm} = 40/50/10$); (b) irradiation of fuel under fast spectrum to high burnup in order to compare relative performance of helium; and (c) develop sodium and lead-bismuth bonded pins. Two different fuel forms (pellet and vibro-packed) are made under this project. An experimental study of $(\text{Pu,Zr})\text{N}$ properties is in progress. This programme might be continued under a new project (ISTC Project #3608, MATINE-2) encompassing in particular the fabrication of $(\text{Pu,Am,Cm,Zr})\text{N}$ microtargets containing about 1 g of ^{244}Cm using the vi-pack technique; further irradiation in the BOR-60 reactor is also under consideration.

6.2.15. CONFIRM

The project 'CONFIRM' (Collaboration On Nitride Fuel IRradiation and Modelling) under Framework Programme 5 (FP5) was meant to develop methods of manufacturing fertile-free fuel viz., without uranium nitrides and to model and test their performance under irradiation in the thermal reactor at R2 Studsvik in Sweden [19, 92, 172]. The CONFIRM project is a collaboration on nitride fuel irradiation and modelling including the following partners: KTH (coordinator) and Studsvik of Sweden, PSI of Switzerland, ITU, EU, SercoAssurance and BNFL of the United Kingdom, and CEA of France. $(\text{Pu,Zr})\text{N}$ pellets with good mechanical properties were fabricated at PSI. Measurements of thermophysical properties for sample pellets were performed at CEA in Cadarache. Measured thermal conductivity was well in line with prior theoretical assessment. At ITU, fabrication of $(\text{Ce, Zr})\text{N}$ and $(\text{U, Zr})\text{N}$ pellets was undertaken. This work will be followed by the production of $(\text{Pu,Zr})\text{N}$ and $(\text{Am,Zr})\text{N}$ pellets for material characterisation. These fabrication experiments revealed that the thermodynamic conditions required to

⁷ A fuel fabrication technology being used by the Russian Federation involving a co-precipitation technique with ammonium hydroxide and polyacrylamide is called the GRANAT process. This process produces particle sizes which can be labelled granulated, and results in the lowest dusting level.

form and the content of nitride solid solutions are difficult to control and often lead to compounds with high levels of impurities, including oxygen. Samples were characterized at CEA/Cadarache (LEFCA). Due to the Studsvik R2 reactor shutdown, CONFIRM irradiation of (Pu,Zr)N fuels was moved to HFR in Petten. The CONFIRM fuel pins fabricated at PSI, were subjected to an irradiation campaign which ended in July 2008.

6.2.16. GACID

Under the framework of the GEN-IV sodium fast reactor programme, an international collaboration programme called GACID (Global Actinide Cycle International Demonstration) is being conducted (2007-2016) with participation of CEA of France, DOE of the United States of America, and JAEA of Japan. The objective of the programme is to demonstrate the transmutation of minor actinides in a 20% Pu MOX fuel. Transmutation of (U, Pu, Am, Np, Cm)O₂-x will be demonstrated in the Monju fast reactor located in Tsuruga, Japan. The fuel pins will be manufactured at the ATLANTE hot cells in Marcoule, France using MA feedstocks supplied by the United States of America. Data obtained from the GACID irradiation project will provide a feasibility assessment of MOX fuel matrix for transmutation [17, 342].

7. CONCLUSIONS

In recent years, transmutation of minor actinides has received much attention. This is motivated by the fact that long term radiological toxicity of spent fuel is dominated by minor actinides, in particular neptunium, americium, and curium. Although it is desirable to eliminate long lived minor actinides from a permanent repository, the partitioning and transmutation of minor actinides should not be viewed by Member States as an end goal for all nuclear programmes. Rather, it should be considered in the context of overall nuclear deployment scenarios, which may dictate different fuel cycle options depending on each Member State's nuclear policy, cost-benefit considerations, and public acceptance.

In fact, the once through fuel cycle with direct disposal of spent fuel is a reference nuclear deployment scenario in many Member States. Technically sound engineered barrier concepts make direct disposal a viable option. Therefore, the question is not whether minor actinides have to be partitioned and transmuted or not, but rather how to best manage minor actinides given a particular envisioned future nuclear deployment scenario. In the same context, the minor actinide transmutation rate per se has no particular significance depending on the future deployment scenario, and hence should not be adopted as a figure of merit without understanding its implications.

In a once through fuel cycle, the development of a durable engineered barrier concept would be given priority. In a closed fuel cycle with thermal reactors only, nuclear properties dictate that minor actinides cannot be fully transmuted. Thus, cost-benefit considerations will be a strong factor in deciding the minor actinide management option. If no fast reactors are envisioned, then other dedicated transmutation systems, such as an accelerator driven system, could be considered for minor actinide transmutation. If fast reactors are envisioned, then it would be best to dedicate minor actinide transmutation to fast reactors, although some partial plutonium recycling can be and is being carried out in thermal reactors in the near term. Even in this case, depending on 'cost-benefit considerations' and 'nuclear energy deployment scenarios' one should not eliminate the possibility of introducing 'dedicated transmutation machines' like ADS in 'double-strata strategies'. This is because minor actinides can be more effectively transmuted in fast spectrum strategies and because they become only a small fraction of fast reactor fuel, which typically requires higher fissile contents. On the other hand, thermal reactors have advantages for minor actinide transmutation because the fuel cycle infrastructure already exists for plutonium recycling, and hence some partial minor actinide recycling can be implemented with incremental effort.

At the present time, we cannot predict future nuclear energy deployment scenarios with any reasonable certainty. Therefore, we cannot decide on an optimum approach to manage minor actinides, which is applicable to all future scenarios. There is no urgency today to settle on a particular approach. In the meantime, the necessary R&D and demonstrations to enable minor actinide recycling can be carried out to broaden technical options for managing minor actinides, and in fact, important R&D activities are being carried out in several Member States.

This report attempted to review and summarize all available information as well as ongoing R&D results related to minor actinide transmutation. Reactor and fuel cycle options for actinide recycling were summarized in Section 2, various minor actinide bearing fuel types and their performance characteristics in Section 3, pertinent thermal and physical properties in Section 4, their associated fabrication processes in Section 5, and an overview of irradiation tests on minor actinide fuels in Section 6.

Because minor actinide bearing fuel is a new field, a tremendous amount of excellent research has been accomplished to date. It appears that the most emphasis has been placed on heterogeneous target fuel concepts with a high content of minor actinides only (no plutonium). This trend may have been driven by the desire to set the performance envelope, namely to understand and prove performance characteristics in extreme cases. However, this approach may overlook some practical approaches amenable to commercial scale deployment in practice.

Many fuel types, encompassing different matrices and chemical forms, have been considered for minor actinide bearing fuels. Several unconventional fabrication techniques have been explored as well. The in-reactor irradiation tests conducted to date are all very promising. As post-irradiation examination results become available, they will provide valuable information to guide additional future irradiation tests. Obviously, more R&D and in-reactor irradiation tests are required to qualify minor actinide bearing fuels.

The incorporation of MA has some impact on the physico-chemical properties of fuel material. Some results are available for the incorporation of MA in MOX fuel (for example lower melting temperature, influence of stoichiometry on thermal conductivity, redistribution of Am). But additional data are needed to guarantee the safe operation of reactor and fuel cycle facilities (fuel fabrication and reprocessing).

The SUPERFACT irradiation in Phenix (1986-1988) represents the main body of existing knowledge on in-pile behaviour of MOX fuel loaded with MA. This project demonstrated the feasibility of Am or NP incorporation of up to 2% in MOX fuels. The main constraint in introducing minor actinides into the core (homogeneous recycling mode) is linked to their impact via core reactivity and kinetic factors. For heterogeneous recycling and in particular in the case of blankets loaded with MAs, high MA content raises the question of managing the large quantity of He produced. Hence, a specific transmutation fuel microstructure must be developed, which requires envisioning several innovative steps in irradiation systems.

Most fast reactor irradiation tests were done in the French Phenix reactor, which is scheduled to be shut down in 2010. Since there are only a handful of fast reactors still in operation which can provide prototypic irradiation test environments in fast spectrum, an international collaboration to expand test capabilities and optimize the limited availability of irradiation test facilities would also be desirable.

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ABBREVIATIONS

ADS	Accelerator driven system
AECL	Atomic Energy of Canada Limited
AFC	Advanced fuel cycle
AFCI	Advanced fuel cycle initiative
AMBOINE-1	Design and production of americium target fuel pin for in-pile tests in BOR-60 reactor
AMSTER	Actinides molten salt transmuter
ANL	Argonne National Laboratory (USA)
ASTRID	Advanced sodium test reactor for industrial demonstration
ATR	Advanced test reactor
BN-800	Fast reactor, 800 MW(e) capacity (under construction in the Russian Federation)
BOL	Beginning of life
BOR-60	Experimental fast reactor (in operation, thermal power 60 MW, Dimitrovgrad, RIAR (Russian Federation))
CALPHAD	Calculations of phase diagram
CAMIX	Composites of americium in Phénix
CEA	Commissariat à l'énergie atomique
Cercer	Ceramic-ceramic composite
Cermet	Ceramic-metal composite
CIRIL	Centre Interdisciplinaire de Recherches avec les Ions Lourds
COCHIX	Conception optimisée vis-à-vis des microstructures dans Phénix
COGEMA	Compagnie générale des matières nucléaires Vélizy
CONFIRM	Collaboration on nitride fuel irradiation and modelling
CORAIL	Combustible recyclage a ilot
CRIEPI	Central Research Institute of the Electricity Power Industry, Japan
DB-MHR	Deep-burner-modular helium reactor
DOVITA	Dry reprocessing, oxide fuel, vibro-pac, integral, transmutation of actinides
DOVITA-2	Dry technologies, on-site reprocessing, various type of fuel with MA, integration of MA recycling into FR closed fuel cycle, TA-transmutation of actinides
EBR	Experimental Breeder Reactor
ECRIX	Expérience en Cœur pour transmutation en Phénix
EdF	Electricité de France
EFPD	Effective full power day
EFTTRA	Experimental feasibility of targets for transmutation
EOL	End of life
EPR	European Pressurized Reactor
ERMT	European research material for transmutation
FBTR	Fast Breeder Test Reactor, Kalpakkam (India)
FGR	Fission gas release
FFTf	Fast flux test facility
FIMA	Fission per initial metal atom
FP	Fission product
FPD	Full power day
FR	Fast reactor
FUJI	Fuel irradiation for JNC and PSI
FUTURE	Fuels for transmutation of transuranium elements

FUTURIX/FTA FZK	Fuels for transmutation of transuranium elements in Phénix / Fortes Teneurs en Actinides Forschung Zentrum Karlsruhe (Germany)
GANIL	Grand Accélérateur National d'Ions Lourds, Caen (France)
GCR	Gas cooled reactor
GFR	Gas cooled fast reactor
GNEP	Global Nuclear Energy Partnership
GT-MHR	Gas turbine–modular helium reactor
HELIOS	HFR irradiation test to study the in-pile behaviour of U-free Am targets as a function of temperature
KÜFA	Kühlfinger apparatus
HEU	High enriched uranium
HFR	High flux reactor, Petten (Netherlands)
HLLW	High level liquid waste
HM	Heavy metal
HMTA	Hexamethylenetetramine
HTGR	High temperature gas cooled reactor
IAM	Institute of Advanced Materials, Petten (Netherlands)
IFR	Integral fast reactor
IMF	Inert matrix fuel
INEEL	Idaho National Engineering and Environmental Laboratory (USA)
INPRO	International Project on Innovative Nuclear Reactors and Fuel Cycles (IAEA)
IPPE	Institute for Physics and Power Engineering, Obninsk (Russian Federation)
ISTC	International Science and Technology Center (Russian Federation)
ITU	Institute for TransUranium Elements (Germany)
LWR-DEPUTY	Destruction of plutonium in thermal systems — European Commission project
KAERI	Korea Atomic Energy Research Institute
LANL	Los Alamos National Laboratory (USA)
LMFR	Liquid metal fast reactor
LLFP	Long live fission products
LOCA	Loss of coolant accident
MA	Minor actinide
MATINA	Matrices for incineration of actinides
MATINE	Minor actinide transmutation in nitrides
MCNP	Monte Carlo N-particle transport code system
METAPHIX	Metal fuel irradiation in Phénix reactor
Metmet	Metal-metal composite
MIMAS	Micronized master blend
MIMOX	Minor actinides in mixed transuranium oxide
MINOX	Minor actinides in mixed oxide
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
MSTR	Molten salt transmutation reactor
MTHM	Metric tonnes heavy metal
MW·d/kg U	Megawatt days per kg of uranium
MYRRHA	Multipurpose neutron source for R&D applications at SCK-CEN (Belgium)

NFI	Nuclear Fuel Industry Ltd (Japan)
NRG	Nuclear Research Groupe (Netherlands)
OMEGA	Program for options for making extra gains from actinides and fission products
ORIGEN	Oak Ridge National Library-Isotope Generation and Depletion Code
ORNL	Oak Ridge National Laboratory (USA)
OTTO	Once through then out
P&T	Partitioning and transmutation
PIE	Post irradiation examination
PSI	Paul Scherrer Institute (Switzerland)
PUREX	Plutonium uranium extraction
PUMA	Plutonium and minor actinide management by gas cooled reactors
RIAR	Research Institute of Atomic Reactor, Dimitrovgrad (Russian Federation)
SCK-CEN	Studiecentrum voor Kernenergie — Centre d'Étude de l'Énergie Nucléaire, Mol (Belgium)
SILOE	Research reactor, CEA-Grenoble (France)
SPIN	Séparation-incinération
SUPERFACT	Minor actinide irradiation in Phénix
TANDEM	Accelerator name
TANOX	Irradiation system in SILOE
THTR-300	Thorium high temperature reactor 300 MW(e)
THERMHET	Thermique des hétérogènes, irradiation name
THORIMS-NES	Thorium molten-salt nuclear energy synergetics
TRISO	Triple isotropic coated particle
TRIGA	Training, research, isotopes, general atomics reactor
TRU	Transuranium element
VHTRs	Very high temperature reactor
WWER	Water cooled water moderated energy reactor
YSZ	Yttria stabilized zirconia

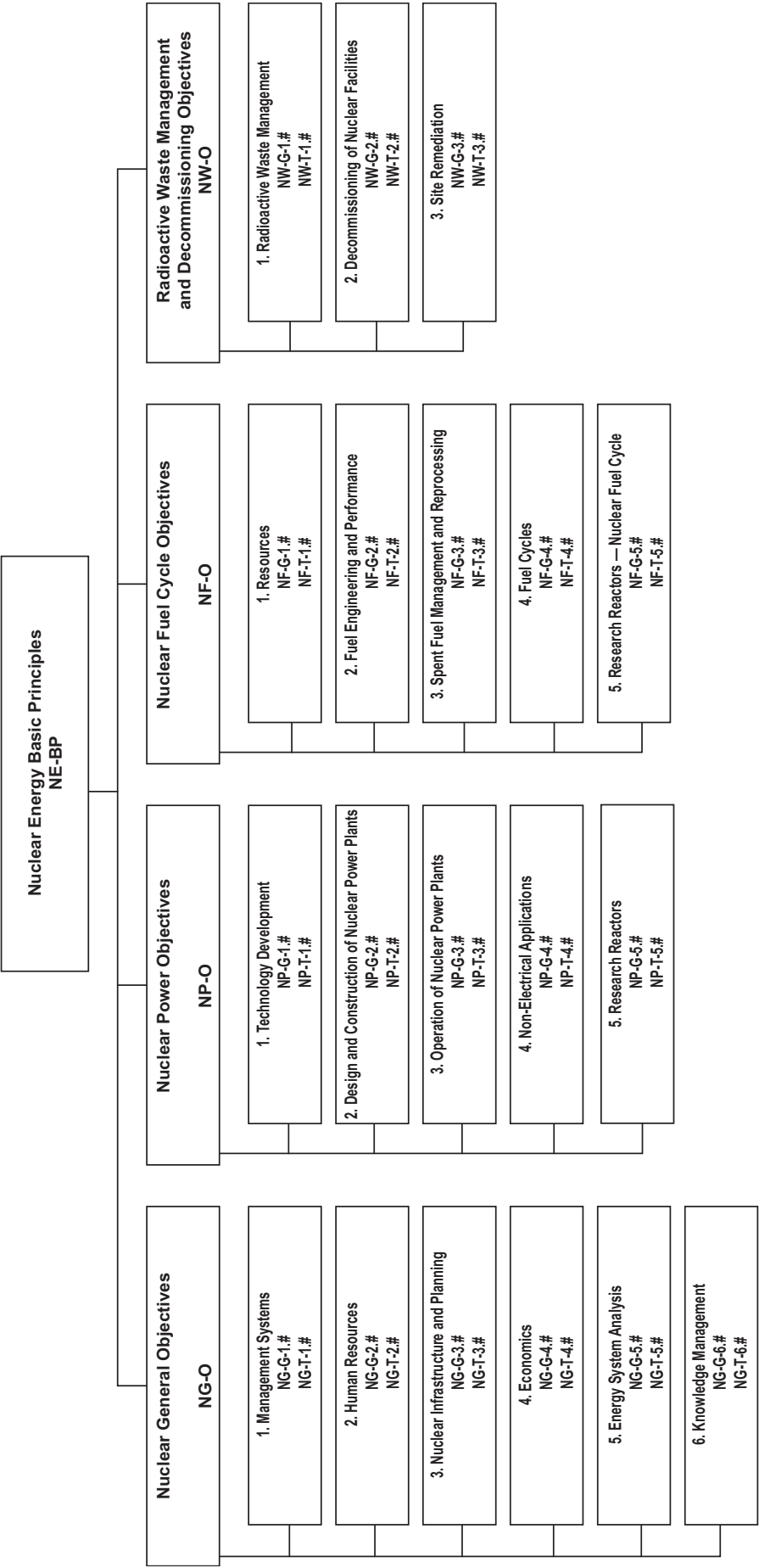
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