RECENT PROGRESS IN THE INVESTIGATION OF INERT MATRIX FUEL

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ABSTRACT

The status and recent progress of the research works on the so-called Inert Matrix fuel concept for the utilization as energy resources and the incineration of excess Pu arisings as well as for an effective transmutation of minor actinides (MAs; Am, Np and Cm) and long-lived fission products (LLFPs) are reviewed in view of the materials science. Stabilized cubic phase ZrO_2, magnesium spinel ($MgAl_2O_4$) and other potential candidate materials for the Inert Matrix are compared in terms of the material properties and other particular aspects such as the behavior against irradiation, with the relevant information currently available. Strategies for the use of the Inert Matrix Fuel concept in various countries are also discussed and compared for their options in nuclear fuel cycle technology.

I. INTRODUCTION

Plutonium, minor actinides and long-lived fission products in the used nuclear fuels are unavoidable by-products of nuclear energy generation by use of the currently established nuclear fuel technology. In addition, the dismantling of nuclear weapons will give rise to an excess in separated Pu stocks. Utilizing and/or burning Pu production in the power reactors is one of the options to solve the problems caused by the surplus Pu. Van Geel et al. [1] categorized them into, namely: 1) *Pu from reprocessing spent LWR fuels* (commercial or reactor Pu, R-Pu), 2) *Pu from the dismantling of nuclear warheads* (ex-weapons or military Pu, W-Pu) and 3) *Pu contained in spent reactor fuel from electricity-producing nuclear power stations* (spent fuel Pu, SF-Pu). They discussed also the amounts of Pu arisings for above each category were forecast until 2005.

On the other hand, spent nuclear fuel annually discharged contains about 6 t Am(mostly ^{241}Am), about 4.8 t ^{237}Np and 0.2 t ^{244}Cm, 10 years after removal from the reactor. Due to their

radiotoxicity, separation (partitioning) of these minor actinides from the high level waste (HLW) containing fission products is preferred for a long term aspect of HLW disposition because of a possible deterioration of glass phases used for vitrification of the HLW. Once they are separated, they can either be stored separately or solidified in ceramic matrices (immobilization), or they can be further destroyed into non-radiotoxic and stable elements (transmutation).

Recently, an innovative fuel has been conceived and its R&D work has been initiated in order to solve the problems regarding the surplus plutonium from both warheads and worldwide reprocessing campaigns as well as for the transmutation of the MAs. The so-called 'Inert Matrix Fuel (IMF)' concept is currently being developed for its material and in-pile performance aspects and fabrication technology for the various candidate materials in connection with the remaining nuclear fuel cycle. This fuel concept is based on 'U-free' (i.e., for excluding continuous U-Pu conversion) Pu or MA compounds embedded in a material matrix (sometimes also called a *'support'* or a *'diluent'*). The stable inert matrix fuels or targets will then, after burning in a power reactor, be directly disposed of for final fuel disposal or multi-recycled, for the MA recycling in particular.

Stabilized ZrO_2-based inert matrix and $MgAl_2O_4$, in particular, have mostly been investigated up to the present time. However, there seem to be other materials such as AlN and ZrN which show interesting material properties in view of the thermal properties. In this paper, potential candidate materials for the inert matrix are compared in terms of the material properties and other behavior such as the behavior against irradiation with the relevant information currently available. Strategies for the use of the inert matrix fuel concept in various countries are discussed and compared for their options in nuclear fuel cycle technology.

II. CHARACTERISTICS OF CANDIDATE MATERIALS FOR THE INERT MATRIX

The term 'inert matrix fuel' currently employed in the area of nuclear fuel technology is used mostly to express ideally, described as above, the 'U-free Pu, MAs or LLFPs compounds embedded in inert matrices. These compounds are usually elaborated by means of solid solution formation (*homogeneous diluent*), by forming a two-phase microstructure (*heterogeneous diluent*) or even by combination of these two concepts forming solid solution inert matrix fuel dispersed in a matrix (*hybrid type*). The stable matrix will then, after burning in a power reactor via once-through mode, be directly disposed of for spent fuel disposal, or multi-recycled, for the MA recycling in particular.

For such a particularity of the materials, when used for the inert matrix, they usually require particular properties as for nuclear fuel materials; high melting points, low vapor pressure, good

thermal properties (thermal conductivity, heat capacity), good mechanical properties, good compatibility with cladding and coolant, low neutron capture cross-sections as well as availability and fabricability of the materials. Furthermore, materials should be stable, on a long term basis, against radiation and interaction with fuel materials all along the irradiation in the reactors and after irradiation should be minimized in view of the in-reactor fuel performance and disposal of spent fuel. In other words, radiation stability of the materials should be carefully examined because of the different physical and chemical effects of the different radiation sources at different stages; neutron and fission fragments during burnup, hence at elevated temperatures, and α-, β- and γ-decay events also during storage and after disposal, hence at low temperatures. An important point is also that thermodynamic entities such as the oxygen potential, solubility and phase relationship in their different chemical states must be known to reliably predict the in-reactor behavior under normal and off-normal conditions.

Based upon the above criteria, a number of inert matrix materials were pre-selected,[e.g., 2,3,4,5] mostly among the ceramic oxides and non-oxide materials. Some of the important property data for the selected materials available up to the present time, affecting the selection of materials for this inert matrix are summarized in Table 1, with regard to the material properties mostly related to their use in a nuclear reactor with fuel materials. In this table, some of non-oxide ceramics are included for their unique and potential use in fast reactors with PuN as a fuel material and consequently, examination shall be desired for their compatibility with Pu and MA nitrides, whereas most oxide ceramics can be commonly used for both thermal reactors and fast reactors. Certain nitrides, such as ZrN can also be considered as matrix materials for oxide fuel materials since oxygen can possibly be dissolved in these nitride ceramics to form a certain oxynitride phase. The formation of such phases is still unknown, and thus a study on the relevant phases has to be carried out if the material is to be applied. The materials listed in the table are of relatively better properties; their melting points are high with reasonably low vapor pressures, compatibility with their environmental species is comparatively good and their thermal conductivities are high with the exception of ZrO_2 and CeO_2.

III. RELEVANT RESEARCH PROGRAMS

Most countries that employ the recycle option of the nuclear fuel cycle are investigating, not only in their own ways but collaboratively with other nuclear relevant organizations, the 'Inert Matrix Fuel' *in their own sense*, for burning (or incineration) of (surplus) plutonium, transmutation of MAs and/or LLFPs. For the developmental works, it is desirable to perform the research programs in collaboration with other nuclear research organizations in order to

exchange, in a efficient way, the relevant information and expertise, and to commonly use their specific facilities and materials. However, there exist also, several national programs in certain countries, fitting their own fuel cycle options, in consideration of their long term nuclear fuel cycle strategies. The following are the examples of national and international collaboration programs which are being conducted in the relevant area. It is also to be noted that, for the disposition of surplus Pu (especially, from warheads), apart from the inert matrix fuel concept, there are also international joint programs with MOX and other similar options, which are discussed in detail elsewhere [6].

III.1. EFTTRA (Experimental Feasibility of Targets for TRAnsmutation) [7]

This European program is, as its name implies, an important feasibility study in the aspect of the transmutation of minor actinides and long-lived fission products, started in 1992 among European organizations, namely, CEA and EdF, France, NRG, Netherlands, FZK, Germany, and IE (Petten) and ITU (Karlsruhe), of the European Commission. Its main purpose is to perform the irradiation experiments for the transmutation of Tc and I as LLFP and Am as MA and for the observation of irradiation behavior of candidate inert matrices materials in thermal and fast reactors as well. The test reactors being employed for this feasibility study are High Flux Reactor (HFR), Petten in the Netherlands and Phénix, Marcoule in France. More detailed description is given in Table 2. The program is performed in a step-wise approach, by irradiating inert matrices alone[12,13], inert matrices with uranium dioxide[14,15,16,17], and selected inert matrices with americium[20,22]. Currently, a series of irradiation experiments, focusing on Mg-spinel and MgO, and their respective analyses are being conducted and most of their post-irradiation examination results are available in the literature (see Table 2), and the main results obtained from these experiments are discussed in the following section.

III.2. CAPRA (Consommation Accrue de Plutonium dans les RApides) and SPIN (SéParation et INcinération)

These are French CEA-leading programs in collaboration with NRG, the Netherlands and ITU, JRC of EC [4]. The aim of the study in CAPRA is the plutonium incineration, whereas that of SPIN is the transmutation of MA, both in fast reactors. These programs utilize the French Phénix fast reactor to investigate the irradiation behavior of inert matrix materials and fuels and targets.

In these experiments, several candidate fuels and inert matrix materials were considered: PuO_2, PuN, PuC, Pu_2C_3, Am_2O_3, AmO_2, AmN and Am_2C_3 ceramic for pure actinide compounds, Al_2O_3, MgO, Y_2O_3, CeO_2, $MgAl_2O_4$, $Y_3Al_5O_{12}$, $CePO_4$ and $ZrSiO_4$ for inert matrices, and for compounds with Pu and Am (Actinides; Ac), $(Ac,Ce)O_2$, $(Ac,Y)_2O_3$, $(Ac,Zr)O_2$, (Ac,Y)N and

(Ac,Zr)N (solid solution), MgO-PuO_2, Al_2O_3-PuO_2 and $MgAl_2O_4$-PuO_2 (two-phase in CERCER with Pu), MgO-AmO_2, Al_2O_3-AmO_2 and $MgAl_2O_4$-AmO_2 (two-phase in CERCER with Am) and W-PuO2 and Mo-PuO_2 (two-phase in CERMET with Pu). These materials were subjected to the irradiation experiments by ion implantation and neutron as well.

III.3. OTTO (Once-Through-Then-Out) [23]

This program is an international irradiation program among JAERI, Japan, PSI, Switzerland, IE, JRC-EC and NRG, the Netherlands. The objective of the OTTO irradiation is to reach a high Pu burnup. A total of 7 pins were prepared for the irradiation test in HFR, Petten, 4 of which contain $MgAl_2O_4$ and 2 contain $(Y,Zr)O_2$, as inert matrix with 1 MOX pin for reference. The fuel materials are: $(Er,Y,Pu,Zr)O_{2-x}$, $(Y,Pu,U,Zr)O_{2-x}$ and $(Pu,U)O_{2-x}$ solid solutions and dispersion type of $(Er,Y,Pu,Zr)O_{2-x}$+$MgAl_2O_4$ and $(Y,Pu,U,Zr)O_{2-x}$+$MgAl_2O_4$ with particle sizes of <25 μm and 200-250 μm, respectively.

III.4. PSI's Advanced Nuclear Fuel Project [24,25,26]

This project is dedicated to the study of the stabilized cubic zirconia as inert matrix for the utilization of plutonium as energy resourses by burning Pu in thermal reactor in once-through and homogeneous mode. Started in July 2000, the irradiation experiment using OECD Halden material testing reactor for the comparison of the irradiation behavior between conventional U-Pu MOX and solid solution (Er,Y,Pu,Zr)O2 inert matrix fuel is currently being carried out, with a total of 6 fuel pins in one rig; 1 pin with stabilized zirconia from co-precipitation, 2 pins with stabilized zirconia from dry-milled powder mixture and 3 U-Pu MOX pins.

III.5. ROX Program in JAERI [27,28,29,30]

The Pu rock-like (ROX) fuel concept was studied in the Japanese former PROFIT (Plutonium Rock-like Fuel In-reactor Technology) project, since as early as the beginning of 1990's, which is essentially a study on burning dismantled warhead plutonium in LWR (ROX-LWR system) after the START. The composition of ROX fuel is based on the multi-phase mixtures of mineral-like (or rock-like) compounds such as yttria-stabilized zirconia (YSZ), corundum and Mg-spinel including *'quasi-inert'* thoria. In this concept, on which the program is currently being carried out, the homogeneous type with Pu solid solution with YSZ, Mg-spinel, corundum, or with mixture with two different oxides, and heterogeneous type with Pu-YSZ solid solution dispersed in Mg-spinel or corundum ('hybrid' type) are examined and tested for in-pile irradiation behavior.

IV. IRRADIATION EXPERIMENTS FOR SELECTED CANDIDATE MATERIALS

Apart from the material properties investigated on the topics described above, studies on the in-reactor behavior have been initiated using various techniques such as fission products implantation [31,32], irradiation with Xe and I ions [5,33] using accelerator and/or ion implanter and neutron irradiation [13,21,33] and, some significant results came to be published and show indications that;

- Spinel ($MgAl_2O_4$) and CeO_2 are relatively stable against radiation (though spinel swells at the fission energy level and is polygonized.[2])
- The volume change is smaller than 1% for Spinel($MgAl_2O_4$) and excessive (about 4.2%) for α-Al_2O_3 at a neutron fluence of 1.7×10^{26} m^{-2}.[5]
- For ZrO_2, the monoclinic phase in PSZ (Partially Stabilized Zirconia) was observed to disappear by neutron irradiation while the cubic phase remained intact.[31])
- Zircon ($ZrSiO_4$) has high dissociation at high temperature and Monazite has poor radiation stability and thermal conductivity.

These results are likely to lead to a preliminary selection of spinel ($MgAl_2O_4$) and stabilized zirconia as inert matrix materials for further studies. In the irradiation experiments in EFTTRA, ROX and OTTO, Mg- spinel inert matrix material was always included and recent observations reveal that Mg-spinel alone has good neutron irradiation stability and high dimensional stability. Mg-spinel inert matrix fuel with UO_2 or Am showed high swelling rate due to the crack formation or He accumulation, respectively. In this respect, the hybrid concept would be more favorable, if the problem of gap between the solid solution inert matrix fuel phase and surrounding inert matrix phase can be overcome.

V. STRATEGIC VIEW OF THE FUEL CYCLE

The innovative nuclear fuel concept, once it is conceived and decided to be implemented, calls for modifications to the existing fuel cycle activities. These modifications may come from any variations in fuel material utilization and supply, the international framework related to nuclear activities and strategies in the nuclear community..

As mentioned before, the Inert matrix Fuel concept is being pursued due mainly to the surplus R-Pu accumulated and radioactive waste disposition (HLW). For the surplus W-Pu arising from dismantling warheads, U.S. decided to follow the once-through (U-Pu)O_2 MOX option for which related R & D activities have been initiated and are on-going [e.g., 34], keeping pace with the European community in the waste disposition aspect of the inert matrix materials.

Japan has its unique fuel cycle project, integrating not only the organizations related to the fuel cycle, but also all the options for transmutation of actinides with various reactors including an actinide burner including the conventional recycling option, called the OMEGA (Options for Making Extra Gains from Actinide and fission products) Project. This project is also naturally integrated with existing Japanese fuel cycle scheme, the recycle option and, for each option, the fuel types to be developed are different with respect to their characteristics. In the case of European countries, the introduction of this innovative fuel concept does not seem to alter, in general, their nuclear fuel cycle strategies and, rather, they tend to expand their recycle options in more efficient ways, hence, to activate more nuclear fuel cycle programs.

VI. CONCLUDING REMARKS

In this paper, potential candidate materials for the Inert Matrix are reviewed on the current issues being discussed and compared in terms of the material properties and the behavior against irradiation with the relevant information currently published and available. Up to the present, for the incineration of plutonium and transmutation of MA, there are two potential candidate materials for oxide fuel materials; stabilized zirconia and magnesium aluminum spinel, which are focused on in the experiments of the dedicated research programs being conducted in the various research organizations. For nitride fuels, which are supposed to be burnt more favorably in a fast reactor, AlN and ZrN can be considered. Strategies for the use of the Inert Matrix Fuel concept in various countries discussed above seem to keep their directions further until the final decision will be made in the future, always keeping their options in the nuclear fuel cycle technology in favorable considerations.

ACKNOWLEDGEMENTS

This project has been carried out under the Nuclear R&D Program of Korean Ministry of Science & Technology.

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Table 1. PROPERTIES OF SELECTED CANDIDATE MATERIALS

Materials / Properties	Al_2O_3	$MgAl_2O_4$	CeO_2	ZrO_2	SiC	ZrN	AlN
Melting point(□)	2054	2105	~2400	2710	2830 (peritectic)	2960	Incongruent
Vapor pressure (bar)	$\sim 10^{-6}$ (total, at 1950□)	-	6×10^{-5} (total, at 2000□)	$\sim 10^{-8}$ (total, at 2000□)	1.4×10^{-4} (total, at 2000□)	$\sim 10^{-7}$ (N_2 partial, at 2008□)	10^{-1} (2417□-decomposition)
Thermal conductivity (W/Km)	1000□: 8.2 1500□: 5.8	1000□: 7.7 1500□: 8	1000□: 1.2 1500□: 0.9	1000□: 2.2 1500□: 1.5	1000□ 47(α), 40(β) 1500□ 31(α), 30(β)	1000□: 23 1500□: 26	1000□: 36 1500□: 27
Compatibility with · Austenitic steel - Coolant · Na (liq) · Water	No reaction <900□ No reaction Not dissolvable	- No reaction -	No reaction <650□ Incompatible ($NaCeO_2$ formation) Not dissolvable	No reaction <1200□ No reaction -	Compatible <700□ No reaction <500□ Compatible	Possible formation of Zr_4M_2N - Not dissolvable	Compatible <1000□ compatible slow hydrolysis
Other remarks			· complete solid solution formation with PuO_2		· Stable modification Hex. α-SiC · Si part. P at 2000□, 1×10^{-4} · compatible with air <1500□		· N_2 part. P at 2000□, 10^{-3}

Table 2. EFTTRA IRRADIATION EXPERIMENTS

Experiment	Purpose	Materials	Remarks	References.
T1	Transmutation of fission products, Tc and I	- Metallic Tc - NaI, PbI_2, CeI_3	- no change - NaI to be selected	8, 9,10
T2 T2-bis	- Tc, inert matrices - neutron damage in inert matrices	Al_2O_3, $MgAl_2O_4$, $Y_3Al_5O_{12}$ Al_2O_3, $MgAl_2O_4$, $Y_3Al_5O_{12}$, CeO_2	- $MgAl_2O_4$, $Y_3Al_5O_{12}$. good irradiation stability . high dimensional stability	11 12,13
T3	- neutron damage in inert matrices - behavior of particle-dispersed fuel	CeO_2, Y_2O_3, Al_2O_3, MgO, $MgAl_2O_4$, $Y_3Al_5O_{12}$, $BaZrO_3$	- 4.2% vol. Change in Al_2O_3 - <1% in $MgAl_2O_4$ and $Y_3Al_5O_{12}$	14,15,16,17
T4 T4-bis T4-ter	- behavior of AmO_2-dispersed spinel " Behavior of UO_2-dispersed spinel	$MgAl_2O_4$ " "	- swelling up to 18% due to He accumulation - 94% transmutation - 28% fission	18,19,20 21,22 21,22