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Materials research on inert matrices: a screening study

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Abstract

Materials research on inert matrices for U-free fuels has been extensively performed at the Institute for Transuranium Elements (ITU) for more than five years. Relevant experience, e.g. on MgO-based ceramic fuel, fabrication and irradiation of annular cercer and cermet fuel and of ThO_2-based fuel in ITU dates back to about 30 yr ago. The criteria for selecting inert matrices for Am-transmutation, their fabrication – with and without Am – and typical results on property measurements are discussed, often in comparison with UO_2, with emphasis on radiation damage formation and damage effects. The materials studied in most detail are spinel $MgAl_2O_4$, zircon $ZrSiO_4$, ceria CeO_{2-x}, yttria-stabilized zirconia $(Zr_{1-x}Y_x)O_{2-x/2}$, monazite $CePO_4$, and to a smaller degree Al_2O_3, MgO, SiC and Si_3N_4. This paper mentions and reports significant characteristics and experimental results for some of the above listed materials, as an overview of the research activities carried out at ITU. Preliminary results of first leaching experiments with Am-doped CeO_2, $MgAl_2O_4$ and $ZrSiO_4$ are also reported. Some recommendations deduced from this work are summarized.

1. Introduction

In conventional U-based nuclear fuels, higher actinides are formed by neutron capture of ^{238}U. Of these, Pu can be recycled, e.g. as MOX fuel. If Pu is separated, Am becomes the most important contributor to the radiotoxicity of the radioactive waste. Therefore, in order to reduce this radiotoxicity, the partitioning of Am from the waste and its transmutation in nuclear reactors are being studied. Promising fuel types for transmutation are the so-called inert matrices, i.e. fuels containing a support for the Am which does not produce new actinides as a UO_2-support does. Basically, different types of inert matrix fuel, IMF, are possible. The three types most frequently discussed are:

- A solid solution of an Am-compound, e.g. AmO_2 or AmN, in a suitable inert matrix (example $(Ce,Am)O_2$ or (Zr,Am)N).
- A dispersion of a ceramic Am-compound in a ceramic inert matrix not showing solubility for Am (example AmO_2 in spinel $MgAl_2O_4$). Such a configuration is often called cercer.
- A dispersion of a ceramic Am-compound in a metallic matrix (example AmO_2 in Mo). Such a configuration is called cermet, a name which dates back to the early days of nuclear energy when e.g. UO_2 in steel was investigated as candidate for reactor fuel.

Recently, the task to optimize the IMF for Am-transmutation has led to propose a ‘hybrid’ concept of incorporating Am in a stabilizing host phase which is then dispersed in an inert matrix. For example, spherical particles of $(Zr,Am,Y)O_{2-x}$ could be used as host phase in an inert spinel matrix. With such a concept, fission and α-decay radiation damage can be largely confined to the host phase whereas the inert matrix providing mechanical stability and good heat transfer is essentially subjected to neutron damage only [1].

The work at ITU was concentrated on IMF versions of the first two above mentioned types, i.e. on IMF as solid solution and IMF of the cercer type.

The following criteria were used to select inert matrix materials for more detailed studies:

- Neutron economy.
- Ease of fabrication, availability and low cost of starting materials.
- Good thermal properties: high melting point, good thermal conductivity (i.e. good thermal diffusivity

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and heat capacity), absence of phase changes and of significant dissociation at high temperatures.
- Good compatibility with cladding (Zircaloy, steel) and coolant (water, Na).
- Stability against radiation (neutrons, α-decay and fission).
- Good mechanical properties: suitable elastic constants, hardness (hot), etc., to provide mechanical stability during irradiation and benign fuel/clad interaction.

A detailed description of the application of these criteria is available elsewhere [2]. The materials selected for detailed investigations based on a literature survey were first spinel $MgAl_2O_4$, zircon $ZrSiO_4$, ceria CeO_2, and silicon carbide SiC and nitride Si_3N_4 [2]. Later, monazite $CePO_4$ [3] and stabilized zirconia $(Zr,Y)O_{2-x}$ and $(Zr,Ca)O_{2-x}$, the latter as a host matrix [4], were included. The present work will show as an example some results related to the last four criteria listed above.

2. Materials and experimental procedures

The fabrication procedures to prepare pellets of $MgAl_2O_4$, $ZrSiO_4$, CeO_2, SiC and Si_3N_4, with or without added $^{241}AmO_{2(-x)}$ or UO_2 are described in Ref. [2]. Cold pressing and sintering techniques were used. The Am was introduced into green pellets of spinel, zircon, ceria, SiC and Si_3N_4 using the soaking technique [5], i.e. by dropping known quantities of a nitric acid solution containing known concentrations of ^{241}Am on the flat surfaces of green pellets having ~50% of the theoretical density. Following the soaking, the green pellets were sintered to densities of e.g. 95% of the theoretical value in the case of spinel. The Am-concentrations in the acid solutions were selected in order to obtain sintered pellets containing 2 and 10 wt% of Am. Spinel with 11.2 wt% ^{241}Am for a reactor irradiation called EFTTRA T4 [1] [6] in the High Flux Reactor (HFR), Petten, NL, was also produced in ITU using the infiltration method, i.e. by immersing green pellets in Am-containing nitric acid solutions [7]. The details and first results of the irradiation are summarized in Ref. [6] and further post-irradiation results are included in Ref. [8]. Single crystals of stabilized cubic ZrO_2 for radiation damage studies, without and with added CeO_2, Nd_2O_3 or Er_2O_3, were produced at Los Alamos, USA [4]. UO_2-containing specimens of SiC and Si_3N_4 were produced by powder mixing (80 wt% of inert matrix and 20 wt% UO_2) followed by sintering.

The measured properties included, as also listed in the preceding section, thermal properties like specific heat (Netzsch DSC) and thermal diffusivity (laser flash method [9]), mechanical properties like hot hardness (Vickers indentations) and Young's modulus, materials compatibility properties like oxygen potential (EMF technique [10]), radiation stability properties like lattice parameter changes and swelling, etc. Particular emphasis was placed on investigating the radiation stability of the different materials against α-decay and fission damage. Lattice parameter changes as a function of the storage time, i.e. of the decay damage build-up, were measured with Am-doped specimens. Ion irradiation techniques were used to simulate (i) α-particles by irradiation with He-ions of energies up to 2 MeV; (ii) recoil atoms of the α-decay by irradiation with heavy ions in the 100 keV energy range [11]; moreover, (iii) fission product damage was simulated by irradiation with a typical fission product of fission energy (72 MeV iodine ions). To get insights into the physical mechanisms of the interaction of high energy heavy ions with the inert matrix materials, also ions of other elements (e.g. Kr, Xe, Pt, U, etc.) of even higher energy up to 2.6 GeV were used. These experiments were done in cooperation with a large number of scientists and high energy accelerators as mentioned in the list of references [12].

An important technique to assess the long term (e.g. up to million of years) immobilization and retention ability of a matrix containing actinides in a geologic repository (subjected mainly to α-decay damage) is to perform leaching tests on the so-called natural analogues, i.e. natural minerals, which contain uranium or thorium [13–15]. In the case of IMF, following reactor irradiation (hence after fission damage and fission products accumulation), reprocessing or direct disposal are envisaged. In order to study the behaviour and the stability of IMF with actinides, leaching experiments are therefore also useful. Static leaching experiments in demineralized water at room temperature and at 423 K were performed on samples of the IMF candidates spinel $MgAl_2O_4$, zircon $ZrSiO_4$ and ceria CeO_2 containing 10 wt% of ^{241}Am. The specimens were cylindrical pellets, with an average geometric surface of ~0.75 cm^2. The tests at room temperature are currently continuing and will be reported at a later time. The high temperature leaching times were 1, 25 and 240 h. The tests were performed in autoclaves, with the specimens immersed in 20 ml of water inside a teflon leaching vessel. Fresh leachant and a new leaching vessel were used for each test. The used leaching vessels were rinsed with 1N HNO_3 solution for 1 h. The leachate and acid rinse solutions were analyzed with ICP-MS.

In addition to these experimental activities, thermodynamic calculations were performed in order to obtain

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[1] The EFFTRA activity (Experimental Feasibility of Targets for Transmutation) is a joint action of CEA and EdF in France, ECN in the Netherlands, FZK in Germany and ITU, Karlsruhe as well as IAM, Petten of the Joint Research Centre of the European Commission.

a basis for the experimental work (THERMO-CALC [2,16]).

Finally, in close interaction with the above mentioned activities, parallel work is performed on strategic concepts for Pu-utilization and actinide transmutation including U-free fuels (e.g. pure actinide compounds) and IMF (e.g. actinide compounds with a supporting inert matrix), treating also aspects of proliferation-resistance and radiotoxicity of the different fuel cycles and strategies e.g. [17,18].

3. Results

In this section, some typical results of property measurements on the as-produced sintered matrices, on single crystalline matrices and on sintered matrices containing 10 wt% AmO_{2-x} are reported. Fig. 1 shows measurements of the specific heat of spinel, zircon and ceria pressed and sintered in ITU compared to data for UO_2 [19]. The densities for the above three materials were, respectively, 95%, 87% and 93% of the theoretical value. The thermal diffusivities of the same three materials were also measured [2]. Together with the measured heat capacity and thermal expansion data, this yielded the thermal conductivities. Knowledge of c_p is also necessary to predict fuel behaviour in reactor transients. High values imply significant amounts of stored energy. The experimental results of Fig. 1, if corrected according to the law of Neumann–Kopp to specific heat per g-atom, agree rather well with the well known relation of Dulong and Petit [20] of $c_p \sim 3R$ (where R is the gas constant). Typical values of thermal conductivity for the above three materials, in W m^{-1} K^{-1} and at 1000 K, were 5.39, 4.20 and 3.02, compared with 3.60 for typical UO_2 fuel pellets. CeO_2 shows thus a rather low conductivity. Also, it tends to become substoichiometric and reduced towards the sesquioxide Ce_2O_3. This has a still much lower thermal conductivity, lower by about a factor of two compared to CeO_2.

Fig. 2 shows the hot hardness of CeO_2 together with data for UO_2, obtained from Vickers indentations. The difference between UO_2 and ceria decreases with increasing temperature, thus indicating that a sufficient mechanical integrity of CeO_2 is maintained up to 1073 K. At low temperatures, CeO_2 is significantly softer than UO_2, which is advantageous for fuel/clad interactions in case of fuel pellet fracture and relocation of fuel pieces, as it is known to occur in UO_2 fuel. Other mechanical properties which have to be known for irradiation and which were measured included Young's modulus, shear modulus and Poisson ratio. As an example, the values for $ZrSiO_4$ were 154 GPa, 64.4 GPa and 0.19, respectively.

Fig. 3 indicates the difficulties to maintain CeO_2 stoichiometry. Sintering in strongly reducing atmosphere yields the sesquioxide form with an unfavourable microstructure of columnar grains subject to easy cracking. Therefore, 1723 K and air were used for sintering the pellets for the present work. Even when annealed in air, CeO_2 tends to loose oxygen. For instance, at 1773 K in air CeO_2 becomes slightly substoichiometric $CeO_{1.995}$. Fig. 3 shows that the oxygen potential of slightly reduced CeO_{2-x} corresponds, at

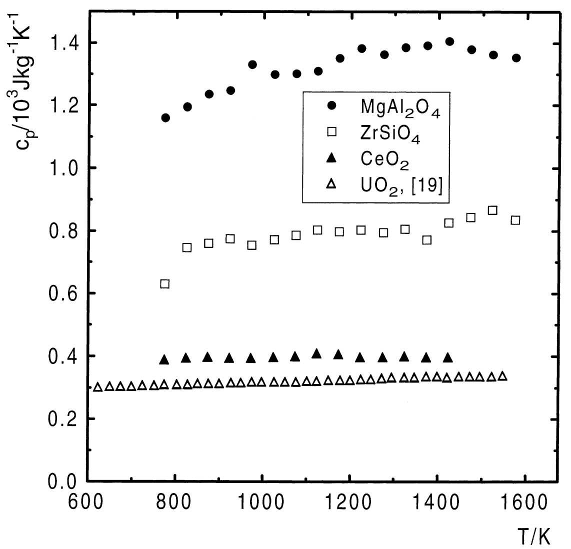

Fig. 1. Specific heat of spinel, zircon and ceria as a function of temperature, compared with results for UO_2 from Ref. [19].

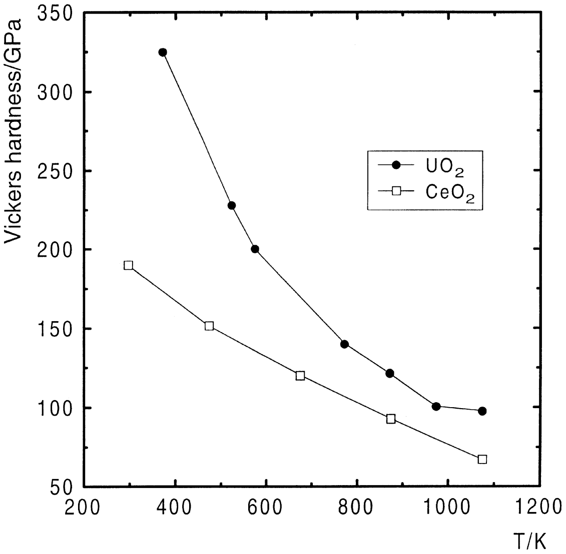

Fig. 2. Hot hardness measured with Vickers indentations of sintered CeO_2, compared to that of sintered reactor-type UO_2.

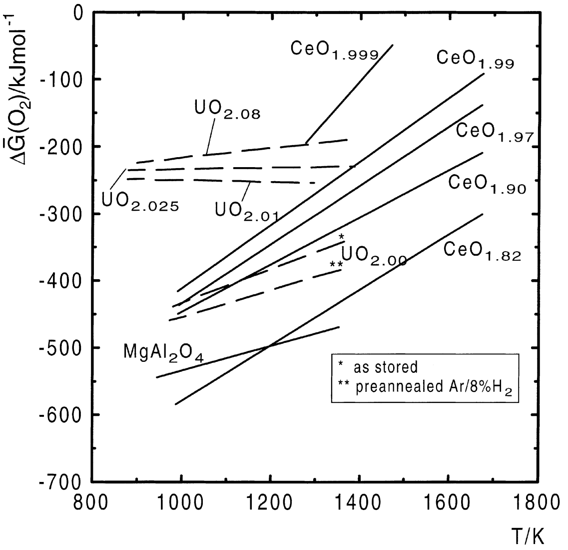

Fig. 3. Oxygen potential as a function of temperature for CeO_{2-x} and $MgAl_2O_4$ compared with data for UO_{2+x} for different deviations from stoichiometry.

typical fuel centerline temperatures, to that of hyperstoichiometric UO_{2+x} and that the oxygen potential of stoichiometric UO_2 corresponds to that of very substoichiometric CeO_{2-x} ($x \sim 0.15$). The use of stoichiometric CeO_2 would thus cause inside clad corrosion of Zircaloy. Consequently, a composition around $CeO_{1.9}$ should be produced as a compromise between clad corrosion and decreased thermal conductivity by phonon scattering at the oxygen vacancies present in substoichiometric CeO_{2-x}. Results for $MgAl_2O_4$ sintered in air [21] are also reported in Fig. 3, showing 'good' oxygen potential values, corresponding to UO_{2-x}.

Irradiation with accelerated ions (e.g. 72 MeV iodine ions) has proved to be a method to obtain qualitative results on the stability of materials against fission damage and irradiations with heavy ions of ~100 keV energy have been used to simulate the damage caused by the recoil-atoms of the α-decay [22,23]. Figs. 4 and 5 illustrate three types of investigations performed on samples irradiated with 72 MeV iodine ions. Fig. 4(a) shows a scanning electron micrograph (SEM) of sintered CeO_2 irradiated with 72 MeV iodine ions at a fluence of 10^{16} cm^{-2} at 1473 K. The surface morphology of the irradiated area provides an indication of the behaviour of CeO_2 subject to fission product impact. Polygonization, i.e. the formation of small subgrains occurs, thus largely masking the original grain boundaries. Fig. 4(b) shows a transmission electron micrograph (TEM) of CeO_2 implanted with 70 MeV iodine ions at room temperature to a fluence of 10^{11} cm^{-2}. The tracks of the ions bombarding the sample are visible as bright spots on the image, indicating the severeness of damage and the existence of thermal spikes e.g. ([24]). One of the most relevant parameters that can be observed and quantified after ion implantation is the swelling. Fig. 5 reports the results of swelling measurements on single crystals of spinel irradiated with 72 MeV iodine ions to different fluences and at different temperatures [24]. Since the irradiated region is constrained in the lateral direction by the underlying unirradiated $MgAl_2O_4$, all of the volu-

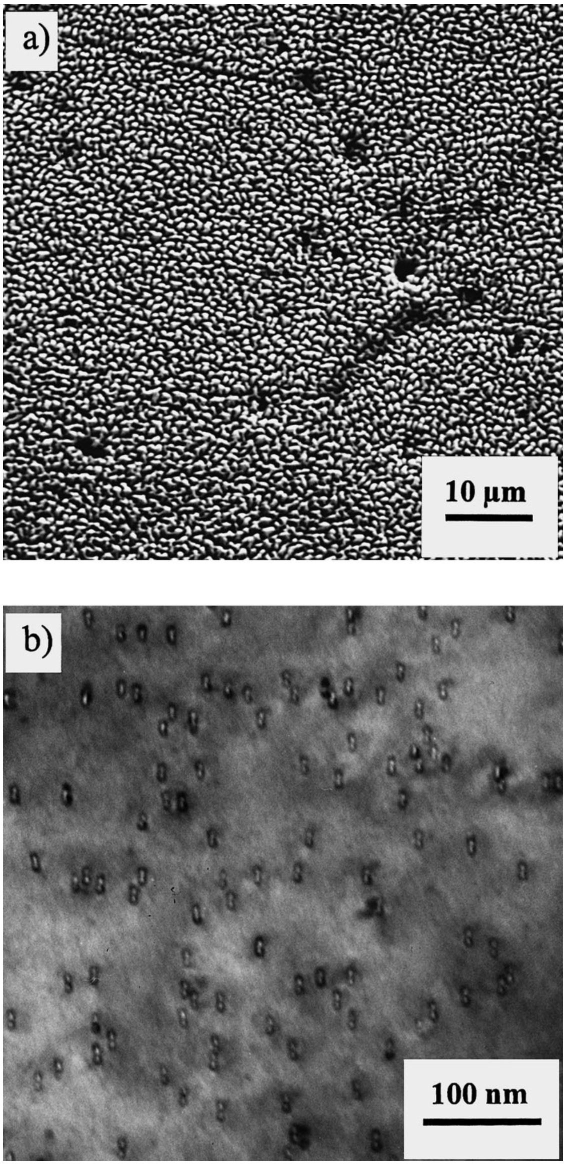

Fig. 4. (a) Scanning electron micrograph of CeO_2 following irradiation with a fission product of fission energy (72 MeV iodine, ion fluence 10^{16} cm^{-2}, 1473 K). (b) Transmission electron micrograph of CeO_2 implanted with 70 MeV iodine, ion fluence 10^{11} cm^{-2}, room temperature. The tracks of the implanting ions are visible as bright spots on the image.

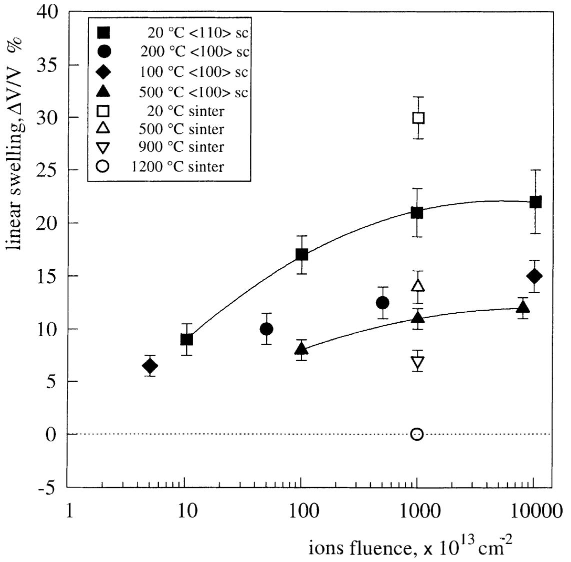

Fig. 5. Swelling of spinel single crystals as a function of dose following irradiation with a fission product of fission energy (72 MeV iodine ions). The small pictures in the diagram represent (a) typical surface profilometry of an implanted specimen; (b) optical microscope image of the same region.

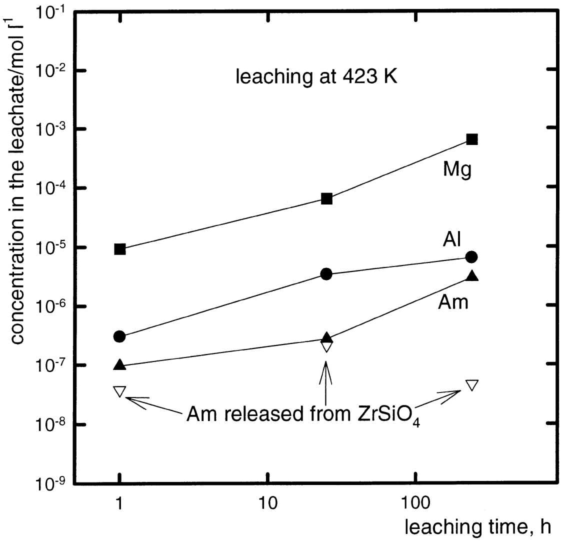

Fig. 6. Concentrations of Mg, Al and ^{241}Am measured in solution after leaching of spinel with 10 wt% AmO_{2-x} in demineralized water at 423 K as a function of time. The concentrations of Am found in the leachates for $ZrSiO_4$ are also plotted on the figure. Zr and Si were not detected, and neither Am nor Ce were detected in any solution for CeO_2 leaching.

metric swelling occurs in the direction normal to the surface (linear swelling). Also shown on the diagram (labeled respectively, a and b) are a surface profile of the implanted region measured by profilometry and an optical micrograph of the same region. Swelling causes a 'pop-out' of the irradiated area. The swelling data presented in Fig. 5 are based on the assumption that the measured elongation (pop-out) is due to swelling of the total implanted volume defined by the range of the fission product iodine. Recent cross-sectional microscopy [25] shows that the irradiated spinel is turned amorphous along only a part of the range. If swelling is due to the amorphized part only, the swelling values of Fig. 5 have to be corrected upwards accordingly (see also Ref. [22]).

Fig. 6 shows the preliminary results of leaching pellets in demineralized water at 423 K of spinel containing 10 wt% of americium as a function of leaching time. The concentrations of Am, Mg and Al measured in the leachate are plotted as a function of leaching time. The amounts released for all three elements were relatively high and increased with leaching time. The concentrations of Am measured in the leachate from the tests with $ZrSiO_4$ are also plotted. The pH values did not show significant changes during the tests and were comprised between 5.5 and 5.9. After each leaching time, the concentration of dissolved Mg was the highest, followed by that for Al and that for Am. This behaviour, which is in contrast with what was observed when leaching spinel not containing Am (in this case neither Mg nor Al were detected in solution), might indicate a selective solubility of the MgO sublattice of spinel, possibly due to a phase separation of MgO and Al_2O_3 caused by radiation damage, or might be due to poor properties of the batch of material used in these tests. For indicative comparison, the solubility values in water of MgO at room temperature and at 353 K are, respectively, $\sim 1.5 \times 10^{-4}$ mol l^{-1} and $\sim 2.1 \times 10^{-3}$ mol l^{-1} [26]. In the case of aluminium oxide, a solubility value in water of $\sim 1.9 \times 10^{-5}$ mol l^{-1} is reported for corundum (α-Al_2O_3) at room temperature [26]. A lower value (about 10^{-8} mol l^{-1}) is reported for Gibbsite (γ-$Al(OH)_3$) at 323 K in the pH range measured for these tests [27]. The highest concentration values for Am measured in the leachates of spinel at 423 K ($\sim 3 \times 10^{-6}$ mol l^{-1}) are in substantial agreement with preliminary results of calculations of the solubility of Am at room temperature performed using the PHREEQCI code. Further studies will be performed in order to better characterize the effects due to the presence of Am (radiation damage, radiolysis, chemical interactions) on the leaching behaviour of this and other inert matrices. For instance, separate effect studies are planned to investigate effects of water radiolysis due to the γ- and α-radiation of ^{241}Am and of α-decay damage in the solid matrices. Also, model calculations as they are presently performed in the institute on leaching of spent UO_2 fuel [28] will be made once more experimental data are available.

Neither Am nor Ce were detected in any of the CeO_2 leachate or rinse solutions. This material appeared to be the most resistant to leaching under the conditions used. In the case of zircon, only Am was detected in the solutions analyzed, in amounts similar to those found for spinel (except after the longest leaching time; see Fig. 6) and without a clear trend as a function of leaching time. Neither Zr nor Si were detected in the leachate solutions, while significant amounts of materials were found in the acid rinse solutions. However, it was often noticed that fragments of $ZrSiO_4$ broke off the edges of the samples during the tests and during handling. The presence of small fines left in the leaching vessels after removing the specimen is the probable cause for the concentrations measured in the rinse solutions.

4. Conclusions and summary

The investigations on inert matrices for Am-transmutation performed at ITU have yielded expertise for fabrication, data on properties and information on radiation stability. Some of the candidates could be rejected, e.g. Al_2O_3 because of excessive swelling, zircon because of its dissociation upon annealing to high temperatures and its poor stability against radiation and monazite because of the poor radiation stability and the poor thermal conductivity [3]. For other candidates, like spinel and (Zr, Y)O_2, an innovative optimized concept ('hybrid' concept [1]) could be developed to exploit the good mechanical and thermal properties of $MgAl_2O_4$ and the good resistance against fission damage of ZrO_2 while avoiding the poor stability of spinel under fission product impact. More property measurements, tailor-made irradiation experiments and coordinated activities, at ITU and in the other cooperating laboratories, are needed to assess the inert matrix candidates already proposed and to prove the validity of the new concepts [1] based upon the available data.

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