

Acidic dissolution behavior of U containing ZrO₂–MgO ceramics

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ABSTRACT

This study explores the possibility of dissolving zirconia–magnesia inert matrix fuel containing uranium oxide as a fissile material and plutonium homolog and erbium oxide as a burnable poison with nitric and sulfuric acid as a potential first step in a reprocessing scheme. The progress of the dissolution is followed by monitoring the amount of material in solution by inductively coupled plasma–atomic emission spectroscopy, assessing the speciation of the material by time resolved laser fluorescence spectroscopy, and determining and quantifying the crystalline phases present in the remaining residue by X-ray diffraction. This study has shown a linear incongruent dissolution of the cubic zirconia phase in concentrated nitric acid under certain chemical compositions, while the magnesium oxide phase is completely soluble. In sulfuric acid uranium, erbium, and magnesium are soluble to different extents while zirconium forms a colloidal suspension that conglomerates and settles out of solution. The feasibility of the dissolution of zirconia–magnesia inert matrix fuel with nitric and sulfuric acid for reprocessing is discussed.

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1. Introduction

There has been a recent resurgence of interest in different oxide fuel types as potential advanced burner fuel for nuclear energy systems, often highlighting the role of transuranic elements as the fissile component. Mixed oxide fuels have been proposed for the reuse of recycled plutonium and higher actinides (Chawla and Konings, 2001). These fertile fuel matrices, however, result in the production of additional plutonium and higher actinides as the initial charge is burned in the reactor. Inert fuel matrices have the potential to allow the reuse of recycled plutonium and higher actinides without the production of additional plutonium (Ledgergerber et al., 2001; Shelley et al., 2001; Carmack et al., 2006). Neutronic calculations indicate that 83% or more of the loaded plutonium can be burnt in a uranium free fuel in the thermal spectrum (Lombardi and Mazzola, 1996; Lombardi et al., 1999; Sickafus et al., 1999). Inert matrix fuel would simultaneously reduce radiotoxicity and proliferation risk in spent fuel. The fissile material in the fuel must be volumetrically diluted by an inert matrix, which is by definition neutron transparent. It must also be compatible with reactor materials such as cladding and coolant water. Additionally, a new fuel must be proliferation resistant, correspond to current safeguards and environmental safety, be economically viable, and refuel on the current time scale (Chawla and Konings, 2001; Degueldre and Paratte, 1999).

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One of the most widely studied candidate inert matrix materials is cubic zirconia. It is radiation tolerant and compatible with reactor materials (Ledgergerber et al., 2001; Shelley et al., 2001; Curran et al., 2003a,b; Degueldre and Conradson, 2001; Kamel et al., 2006; Vettrano et al., 1999). It fully incorporates the fissile material and burnable poisons. For reactor fuel applications, however, cubic zirconia is a problematic material. Due to its very low thermal conductivity, cubic zirconia-based fuel matrices can result in unacceptably high centerline temperatures. To compensate for this, a second phase, such as MgO, may be added to improve thermal diffusivity and thermal conductivity (Medvedev, 2004; Ronchi et al., 2003). Unfortunately, pure MgO cannot be used as an inert matrix fuel, because it undergoes hydrolysis and subsequent swelling in the event of a cladding failure (Medvedev, 2004; Medvedev et al., 2006). The resulting ZrO₂/MgO composite fuel matrix has the potential to combine the best fuel properties of both phases while minimizing the inherent challenges for both pure materials in a reactor environment (Medvedev, 2004).

This study is aimed at exploring the possibilities for dissolving an inert matrix fuel in acidic media in an effort to dissolve the material for reprocessing. Previous studies have used nitric acid to dissolve zirconia–magnesia inert matrix fuels without success (Medvedev et al., 2005). These studies did not use a fissile component, which does not accurately represent the material as shown in this study. This study also expands the acid media to include sulfuric acid. Samples were analyzed by inductively coupled plasma–atomic emission spectroscopy so that each element's solubility and kinetics can be established independently. Undissolved solids

left from the experiments were evaluated by X-ray diffraction. The presence of colloids and speciation was determined by time resolved laser fluorescence spectroscopy.

2. Experimental methods

2.1. Ceramic fabrication

A wet chemical precipitation method was chosen for ceramic fabrication because less severe sintering temperatures and times were required to produce a homogenous sample by incorporating fissile material and burnable poison into the zirconia host phase (Medvedev et al., 2006; Holliday et al., 2009). Concentrated aqueous nitrate salt solutions of zirconium ($ZrO(NO_3)_2$), magnesium ($Mg(NO_3)_2$), erbium ($Er(NO_3)_3$), and a concentrated acetate salt solution of uranium ($UO_2(C_2HO_2)_2$) were prepared. These solutions were mixed in appropriate proportions and the cations coprecipitated with an ammonia hydroxide solution saturated with ammonium oxalate. The resulting precipitate was dried, calcined, pressed, and sintered with traditional solid state synthesis methods. Details can be found in a previous publication (Holliday et al., 2009).

Uranium oxide content in the ceramics was held constant at 5% (wt/wt) as this is within the likely range of fissile material to be incorporated into an inert matrix fuel for thermal reactor applications. Neutronic calculations have shown an optimal volume ratio of burnable poison to be half that of the fissile phase (Fridman et al., 2007). Because of this, the erbium oxide content was held at 2.5% (wt/wt). The inert matrix was composed of zirconium oxide and magnesium oxide and was varied from being exclusively zirconium oxide to being completely magnesium oxide over several sample compositions. The ceramic surface area was determined by BET with a Quantachrome Nova 1000 using N_2 .

2.2. Inductively coupled plasma-atomic emission spectroscopy (ICP-AES)

Samples from dissolution studies were analyzed by inductively coupled plasma-atomic emission spectroscopy (ICP-AES) on a Thermo iCAP 6000 with iTEVA software. The instrument was run at 1150 W and the following lines were used to analyze the elements of interest: Er at 337.271 nm, Mg at 202.582 nm, U at 367.007 nm, and Zr at 343.823 nm. A seven point calibration was used, samples were measured in triplicate, and a calibration check was performed every 50 samples. Standards were purchased from Fisher Scientific and diluted using an acid matrix equivalent to that of the samples.

2.3. X-ray diffraction (XRD)

The surface of the ceramic after being exposed to the various solutions was mechanically scraped off with a spatula and spread in a thin layer over a low-background sample holder (single crystal silicon wafer) with the aid of methanol. XRD patterns were collected on a Bruker D8 Advance diffractometer, which uses a Cu anode and a Johansson-type primary monochromator (wavelength $K\alpha_1$ at 0.1540598 nm). Patterns were taken using 40 mV and 40 mA from 10 to $120^\circ 2\theta$ with a step size of $0.01^\circ 2\theta$ and 4 s per step. Phases were identified using Bruker-AXS EVA. Bruker-AXS TOPAS3 was then used to fit the diffracted intensities and to perform least-square and Rietveld analysis. Structure input parameters were taken from Inorganic Crystal Structure Database. Instrument parameter inputs were as follows: primary radius (mm) 435, secondary radius (mm) 435, receiving slit width (mm) 0.1, divergence angle ($^\circ$) 1, filament length (mm) 12, sample length (mm) 8, receiving slit length (mm) 12, secondary sollers ($^\circ$) 2.3, and Lorenz polarization factor was set to 26.6.

2.4. Time resolved laser spectroscopy

Samples were analyzed for dissolved uranyl content by time resolved laser fluorescence spectroscopy (TRLFS). The system uses a third harmonic, Nd:YAG laser (OPOTECK, Inc., Carlsbad, CA) to pump an Optical Parametric Oscillator to produce an excitation beam at 414 nm. Sample fluorescence is focused onto a PI-MAX II CCD detector (Roper Scientific/Princeton Instruments, Trenton, NJ) by means of a SP500 spectrograph (Roper Scientific) with an entrance slit width of 250 μ m. The detector and diffraction grating were calibrated against standard Hg vapor emission lines. The excitation wavelength of 414 nm was chosen as it corresponds to the largest absorption peak in the uranyl spectrum. The laser system outputs approximately 20 mJ at the specified wavelength. The fluorescence was monitored between 375 and 725 nm using a 150 l/mm diffraction grating. Samples were filtered through 0.45 μ m filters before being placed in 4.5 mL polymethyl methacrylate fluorescence cuvettes for analysis.

2.5. Acid dissolution studies with heat

Sample dissolution was performed in a 250 mL round bottom flask equipped with a condenser. The flask was heated with a heating mantle. Samples were placed in the flask with 150 mL of acid and the heat was turned to maximum. The acid would typically reach its boiling point ($\sim 120^\circ$ C for nitric and 290° C for sulfuric) within 5–15 min. The first samples were taken after 1 h by pipetting a small (~ 0.5 mL) sample into a scintillation vial with a disposable pipette. The sample was allowed to cool for 15 min before a measured amount between 100 and 500 μ L was transferred by a quantitative pipette and diluting to 10 mL with deionized water. Experiments were performed in triplicate.

3. Results and discussion

3.1. Ceramic data

Uranium containing inert matrix fuel was successfully synthesized in a two phase ceramic consisting of cubic zirconia and periclase, which was confirmed by X-ray diffraction. The periclase phase remains pure MgO showing a low affinity for all other cations in the ceramic, which will allow it to retain its thermophysical properties, most importantly thermal conductivity and thermal diffusivity. Magnesium, uranium, and erbium oxides are able to substitute in the zirconia lattice at different levels to create a solid

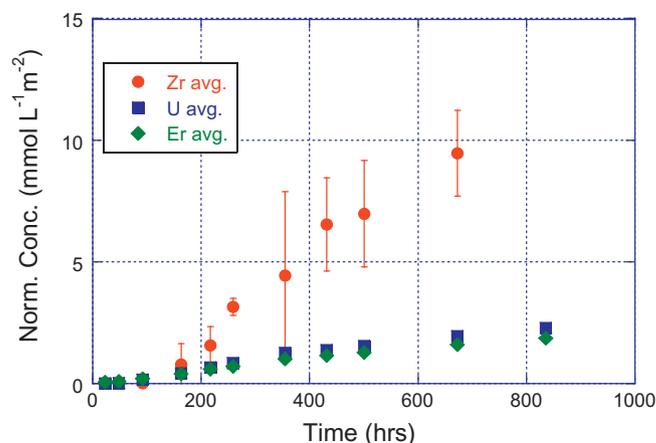


Fig. 1. Surface area normalized concentrations of $Zr_{0.070}Mg_{0.914}U_{0.009}Er_{0.006}O_{1.08}$ in concentrated nitric acid. Error bars represent standard deviation of three samples.

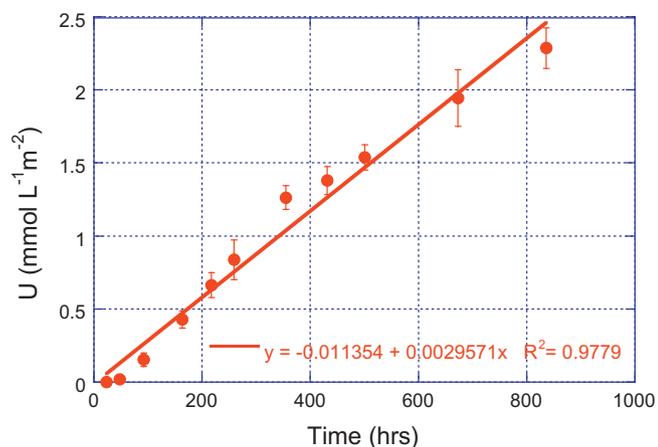


Fig. 2. Uranium concentration with rate of $Zr_{0.070}Mg_{0.914}U_{0.009}Er_{0.006}O_{1.08}$ in concentrated nitric acid. Error bars represent standard deviation of three samples.

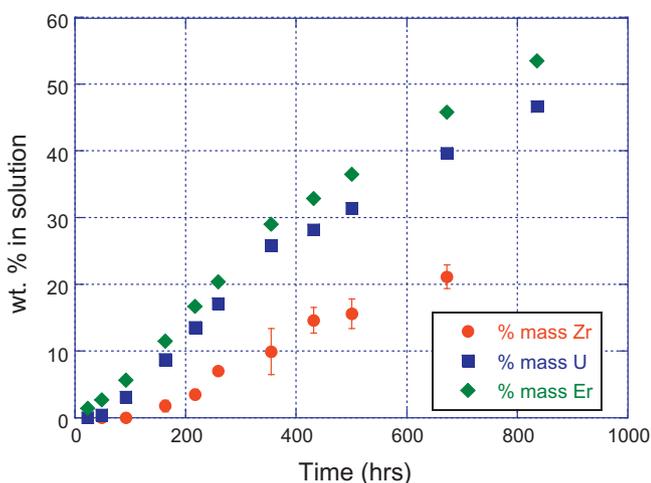


Fig. 3. Weight percent of total sample $Zr_{0.148}Mg_{0.834}U_{0.010}Er_{0.007}O_{1.16}$ in concentrated nitric acid. Error bars represent standard deviation of three samples.

solution. Magnesium concentrations within the zirconia phase remains constant at 5 wt.%, while uranium and erbium concentrations within the zirconia were as high as 20% and 10% (wt/wt) respectively in synthesized samples. Elemental concentrations were determined by electron probe microanalysis. Further detail on the material synthesis and characterization can be found in a previous publication (Holliday et al., 2010).

3.2. Nitric acid dissolution

Several sample compositions were tested for nitric acid solubility. Samples were crushed to a coarse powder and surface area was measured before being added to the flask. Concentrated nitric acid (16 M) was used for all studies. All samples showed that all free magnesium oxide was dissolved on time scales shorter than the sampling rate of the experiment (less than 1 h). Two of those sample compositions ($Zr_{0.148}Mg_{0.834}U_{0.010}Er_{0.007}O_{1.16}$ and $Zr_{0.070}Mg_{0.914}U_{0.009}Er_{0.006}O_{1.08}$) showed solubility of species other than magnesium. It was found that at these extremely high levels of magnesium oxide there are two advantages that allow for the dissolution of uranium. The first is that since the zirconia is the minor phase it has a large surface area to volume ratio after the magnesium oxide is dissolved, which will enhance the kinetics. The second advantage is that since all of the uranium is concentrated within this zirconia phase the less zirconium oxide in the sample the more concentrated the uranium within that phase. Because of this, it was observed that there is zirconium, uranium, and erbium in solution by ICP-AES. This is a substantial difference from those studies done in the absence of a fissile material or with cerium oxide as a fissile material homolog.

The sample with the highest amount of magnesium oxide, $Zr_{0.070}Mg_{0.914}U_{0.009}Er_{0.006}O_{1.08}$, shows the most material in solution. As can be seen in Fig. 1, the amount of material going into solution over time follows linear kinetics without any indication of equilibrium. This same behavior can be seen in sample $Zr_{0.148}Mg_{0.834}U_{0.010}Er_{0.007}O_{1.16}$ although less material is being dissolved for the same period of time. In both samples there seems to be a slower rate for the first 100 h than there is at longer time-scales. This initial lag in kinetics could be due to a number of factors such as an initial leaching of magnesium from the zirconia, the formation of nitrous acid, kinetics of oxidation within the lattice,

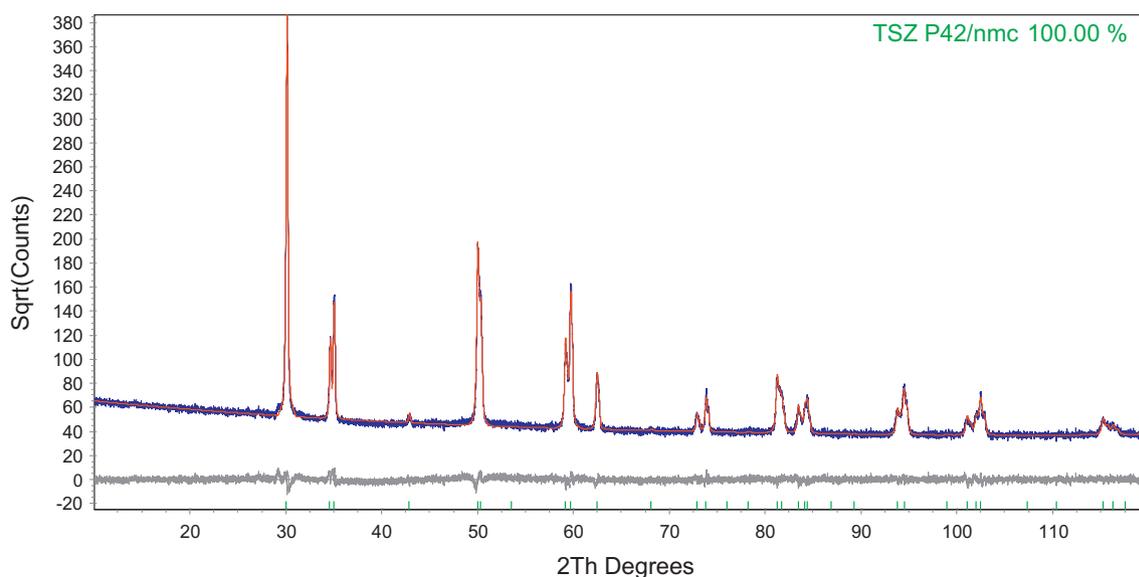


Fig. 4. X-ray diffraction pattern (blue) with fit (red) and difference curve (gray) of the residue after nitric acid dissolution of $Zr_{0.148}Mg_{0.834}U_{0.010}Er_{0.007}O_{1.16}$. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

etc. For this reason kinetics were determined from concentrations taken after 100 h. It was determined that the fastest rate of uranium dissolution, which was in sample $Zr_{0.070}Mg_{0.914}U_{0.009}Er_{0.006}O_{1.08}$, was $3.0 \mu\text{mol L}^{-1} \text{m}^{-2}$ (Fig. 2). When the zirconium oxide content was increased from 7.0 mol.% to 14.8 mol.% the rate of uranium dissolution falls to $1.2 \mu\text{mol L}^{-1} \text{m}^{-2}$. Both samples display incongruent dissolution because of the preferential leaching of uranium and erbium from the lattice. This can be seen by examining the amount of material in solution compared to the total amount of material in the sample for each component. This is shown in Fig. 3 as the wt.% of the material in solution for zirconium, uranium, and erbium. It is evident that there is a much higher percentage of uranium and erbium being dissolved than there is zirconium. The residue left at the bottom of the flask was examined by X-ray diffraction and was found to be partially stabilized tetragonal zirconium oxide (Fig. 4). This further confirms that all of the magnesium oxide is dissolved within the detection limits of this study. The partially stabilized zirconium oxide was destabilized from the leaching of ions from the crystal lattice of the parent cubic zirconia. This indicates that the chemical makeup of the zirconia has changed, which can be explained by the proposed incongruent dissolution which would strip the zirconia of the lower valent ions such as erbium and magnesium that stabilize the lattice.

This preferential leaching of uranium and erbium is only evident at extremely low concentrations of zirconium oxide. This leaching has been shown to reach as much as 50 wt.% of the total uranium and erbium within the sample over 1000 h, which proves more uranium is being dissolved than that on the original surface. This can be explained by examining the composition of the zirconia phase. At the lowest concentration of zirconium oxide the uranium and erbium content within the zirconia is 20 and 10 wt.%, respectively. Together with an additional 5 wt.% of magnesium dissolved into the zirconia, leachable material accounts for 35 wt.% of the lattice. Leaching this amount of material destabilizes the zirconia, causing it to fall apart and expose a new surface that can be leached for additional uranium, erbium, and magnesium. This mechanism not only explains the sample compositions prone to dissolution both before and after exposure to nitric acid, but also the linear kinetics of that dissolution. It is clear that zirconia alone is unable to predict the behavior of the composite and that cerium oxide is not a suitable chemical homolog for uranium oxide as it is not soluble from the zirconia matrix. Plutonium oxide will be evaluated in future studies to assess the accuracy of uranium as a plutonium homolog.

3.3. Sulfuric acid dissolution

A single sample composition, $Zr_{0.148}Mg_{0.834}U_{0.010}Er_{0.007}O_{1.16}$, was tested for dissolution in sulfuric acid. Experiments were performed with 5 M, 9 M, and concentrated 18 M sulfuric acid. The acid was heated to boiling and samples were immersed for over 100 h. Sulfuric acid has a stronger nucleophile, SO_4^{2-} , than that of nitric acid (NO_3^-), which may make it possible to attack the cation sites of the zirconia and dissolve the material. Sulfuric acid is not the acidic media used in reprocessing. Therefore, an additional step would be required to precipitate out the dissolved material and redissolve it in nitric acid for the purpose of reprocessing. While the use of sulfuric acid would potentially add additional complication and cost to the recycling of this fuel it would eliminate the need to use hydrofluoric acid, which is a considerable advantage to the current dissolution process. The progress of the sample dissolution was followed by examining the concentration of cations in solution by inductively coupled plasma-atomic emission spectroscopy and the nature of those species was evaluated by time resolved laser spectroscopy.

Moderate concentrations of sulfuric acid (5 M and 9 M) were unsuccessful in dissolving the zirconia phase. This was determined

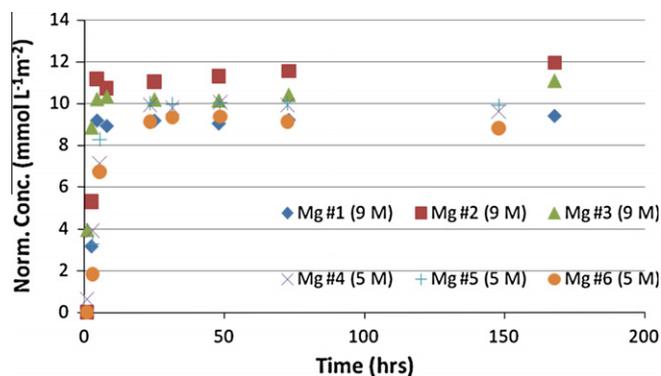


Fig. 5. Surface area normalized concentration of magnesium from six samples of $Zr_{0.148}Mg_{0.834}U_{0.010}Er_{0.007}O_{1.16}$ in 5 M and 9 M sulfuric acid.

by sampling the solution and analyzing by ICP-AES, which has a detection limit of 1–10 ppm given the current experimental conditions. Concentrated sulfuric acid was also unable to dissolve zirconium, uranium, or erbium at room temperature indicating that kinetics factors play a large role in the dissolution of this material. Magnesium oxide was easily dissolved within 10 h by all concentrations of sulfuric acid. Its kinetics were fast enough and the extent of dissolution was so complete that it seemed independent of the sulfuric acid concentration under these conditions as can be seen in Fig. 5. It is shown there that both 5 M and 9 M sulfuric acid dissolve the magnesium oxide phase to completion within 10 h at virtually the same rate. This fast, complete dissolution of magnesium oxide should leave behind a porous zirconia matrix with a high surface area to volume ratio that will improve the kinetics of the overall dissolution.

Concentrated sulfuric acid (18 M) was the only concentration that was successful in dissolving any of the zirconia phase. As can be seen in Fig. 6, concentrated sulfuric acid was able to dissolve a measurable amount of zirconium into solution and was able to solubilize an initial amount of uranium and erbium within the first 24 h of the experiment, which remained constant for the remainder of the experiment. The sample size of these experiments did not exceed 450 mg of sample in 150 mL of sulfuric acid. The amount of uranium dissolved in solution was less than 12 wt.% of

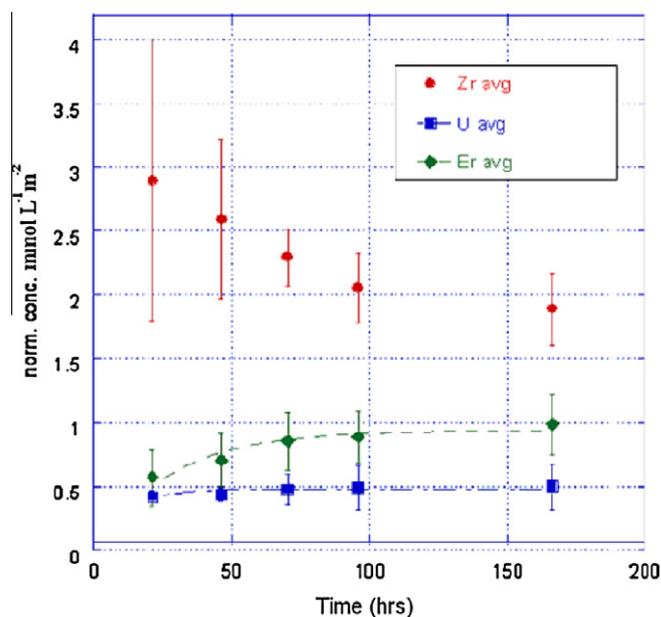


Fig. 6. Surface area normalized concentrations of $Zr_{0.148}Mg_{0.834}U_{0.010}Er_{0.007}O_{1.16}$ in concentrated sulfuric acid. Error bars represent standard deviation of three samples.

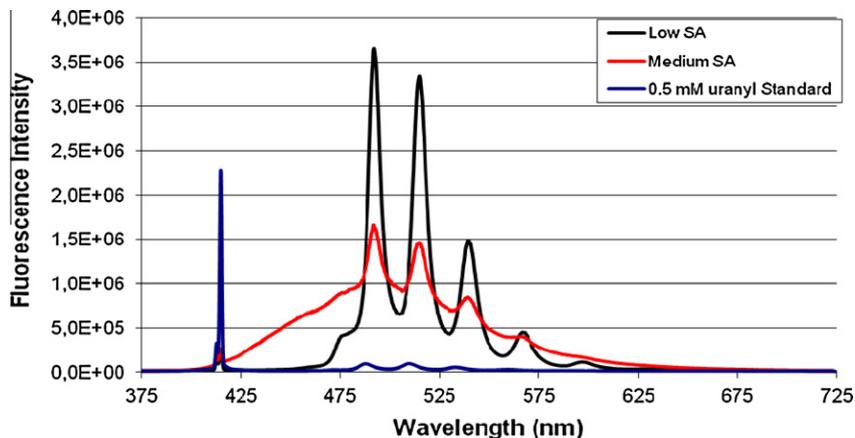


Fig. 7. Time resolved laser fluorescence of two samples of $Zr_{0.148}Mg_{0.834}U_{0.010}Er_{0.007}O_{1.16}$ labeled by relative surface area to volume ratios and a uranyl standard 200 ns after fluorescence. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

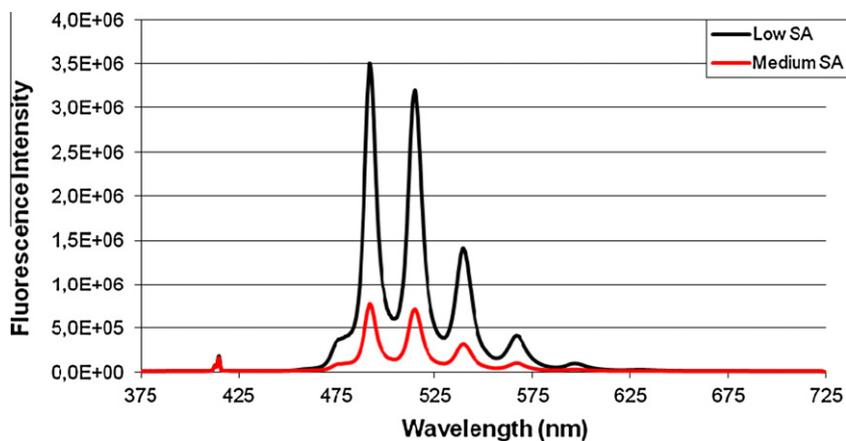


Fig. 8. Time resolved laser fluorescence of two samples of $Zr_{0.148}Mg_{0.834}U_{0.010}Er_{0.007}O_{1.16}$ labeled by relative surface area to volume ratios 1000 ns after fluorescence.

the total uranium in the sample. The erbium was significantly more soluble with 46 wt.% of the total erbium dissolved in concentrated sulfuric acid.

The zirconium exhibits different behavior in that its concentration reaches a maximum at 24 h with a significant standard deviation between samples and then drops to a constant concentration of around 0.2 mmol/L. This is due to the low solubility of zirconium in sulfuric acid. The sulfuric acid is successful in breaking up the matrix and releasing the cations within the zirconia. Based on the observed concentration trends, it appears that the uranium and erbium are released from the zirconia matrix and dissolved in solution, to an extent. The zirconium appears to form colloids, which are initially suspended in solution. As the colloids conglomerate, they settle to the bottom of the vessel, leaving only an equilibrium concentration of dissolved zirconium in solution. This also accounts for the large range of concentrations at the maximum zirconium concentration as this is based on suspended colloids which is highly influenced by particle size distribution, which varies from sample to sample of crushed ceramic. At this time, it is unclear if the uranium and erbium are leached from the zirconium phase, leaving the zirconia as an intrinsic colloid, or if the zirconia is congruently dissolved and the zirconium is forming colloidal species.

Colloidal suspension presented more of a problem with samples that were more finely crushed than those that had a slightly lower surface area to volume ratio. This was confirmed by time resolved laser spectroscopy. At 200 ns after the laser pulse (Fig. 7) the sample with a small surface area to volume ratio shows a typical

fluorescence pattern due to the presence of uranyl, though enhanced because of the high concentration of sulfate. The sample with a slightly higher surface area to volume ratio has a significant background due to scattering in addition to typical uranyl fluorescence spectra. This background was proven to be due to colloidal scattering by taking a spectra at 1000 ns after the laser pulse at which time the scattering process has concluded. At this time, the background seen in the previous spectra is not present and only the contribution due to uranyl fluorescence is present as seen in Fig. 8. This colloidal suspension explains the anomalous behavior of zirconium in sulfuric acid.

4. Conclusions

The potential for dissolving ZrO_2/MgO -based inert matrix fuels was examined using both a nitric and sulfuric acid dissolution process. These studies differ from previous work because of the incorporation of uranium and erbium oxides. This changed the dissolution behavior of the ceramic and future studies will need to be performed with plutonium oxide to accurately represent the behavior of the potential inert matrix fuel. This is best illustrated in the dissolution behavior of the material in concentrated nitric acid. It was shown that at low levels of zirconium oxide, the high concentration of uranium within the zirconia phase and high surface area to volume of that phase enables the leaching of uranium, erbium, and magnesium from the zirconia lattice. It is un-

clear from this study whether surface area or uranium content is the dominant factor in the dissolution. It is suggested that at significantly high levels of these substituting ions it is possible to destabilize the lattice due to leaching and consequently dissolve the zirconia phase through a linear incongruent dissolution process. This process does however require a great deal of time and only happens under specific chemical compositions. Because of this, sulfuric acid was also explored as a means of dissolving the fuel. It was found that only 12 wt.% of the uranium in the sample was dissolved under current experimental conditions. This would mean that large ratios of sulfuric acid to fuel would be needed to dissolve the material completely, making it a highly inefficient process. In addition there is significant formation of colloids that conglomerate and settle to the bottom of the vessel. This could pose engineering problems in the processing of large amounts of material. Both sulfuric and nitric acid pose problems to the dissolution of zirconia–magnesia inert matrix fuels. Future studies will use plutonium as the fissile component of the fuel and compare chemical behavior to that of the uranium and cerium homolog studies. In addition future studies will also incorporate the use of hydrofluoric acid to further enhance the dissolution process with nitric acid.

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