Thermal Measurements and Computational Simulations of Three-Phase (CeO₂ - MgAl₂O₄ - CeMgAl₁₁O₁₉) and Four-Phase (3Y-TZP - Al₂O₃ - MgAl₂O₄ - LaPO₄) Composites as Surrogate Inert Matrix Nuclear Fuel

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Abstract

This study investigates the temperature dependent thermal conductivity of multiphase ceramic composites for simulated inert matrix nuclear fuel. Fine grained composites were made of CeO₂ - MgAl₂O₄ - CeMgAl₁₁O₁₉ or 3Y-TZP - Al₂O₃ - MgAl₂O₄ - LaPO₄. CeO₂ and 3Y-TZP are used as UO₂ surrogates due to their similar structures and low thermal conductivities. Laser flash analysis from room temperature to 1273 K (1000°C) was used to determine the temperature dependent thermal conductivity. A computational approach using Object Oriented Finite Element Analysis Version 2 (OOF2) was employed to simulate the composite thermal conductivity based on the microstructure. Observed discrepancies between experimental and simulated thermal conductivities at low temperature may be due to Kapitza resistance; however, there is less than 3% deviation between models and experiments above 673 K (400°C) for both compositions. When the surrogate phase was replaced with UO₂ in the computational model for the four-phase composite, a 12-16% increase in thermal conductivity resulted compared to single phase UO₂, in the range of 673-1273 K (400-1000°C). This computational approach may be potentially viable for the high-throughput evaluation of composite systems and the strategic

selection of inert phases without extensive sample fabrication during the initial development stages of composite nuclear fuel design.

1. Introduction

The demanding operating conditions experienced by uranium dioxide (UO_2) nuclear fuel can cause drastic microstructural changes and fracture during reactor operation [1]. Temperatures exceeding 1600 K caused by the low thermal conductivity of UO_2 (around $3 \text{ W/m}\cdot\text{K}$ at 1273 K (1000°C)) [2], create steep thermal gradients. These high temperatures induce restructuring exasperated by accumulation of fission gas bubbles and precipitation of solid fission products during burnup [3]. The development of complex microstructures not only challenges reactor performance modeling due to the conservatisms necessary in prediction of transport properties for such a complex system, but also can limit the permissible total burnup if swelling or mechanical degradation results.

To alleviate these problems, a fuel design where fissile and non-fissile phases are incorporated into a composite fuel is considered. Non-fissile phases can offer (i) increased thermal conductivity for the overall composite, (ii) crystalline phases that can accommodate fission by-products after their creation and (iii) decreased microstructural evolution by providing alternative interfaces that are more efficient sinks for point defects. Here, the fissile phase does not need to be the majority phase of the composite, although there will be a minimum fissile volume required that is governed by both the maximum feasible enrichment as well as the neutronic impact of the non-fissile phases.

Previous research has successfully demonstrated a two-phase fuel concept through the fabrication of a composite with a minority of nuclear fuel surrogate particles dispersed in an inert

host matrix [4]. More recently, MgO – Nd₂Zr₂O₇ composites for inert matrix fuel were studied to evaluate the effects that phase content has on the thermophysical properties of the composite system [5]. Other pathways to improve thermal conductivity for inert matrix nuclear fuel have also been studied using three phases, but with extremely large grains similar to those occurring naturally in rocks and minerals [6]. A key factor is that each phase must coexist without the formation of intermediate phases. Rock-like three-phase ceramic composite systems with fluorite, spinel, and corundum structures are reported to be compatible with both PuO₂ [6a] and UO₂ [6b, 7] under irradiation.

In the present work, a 4-phase composite system consisting of 3Y-TZP (3 mol.% Y₂O₃ stabilized ZrO₂), Al₂O₃, MgAl₂O₄ and LaPO₄ and a 3-phase composite system consisting of CeO₂, MgAl₂O₄ and CeMgAl₁₁O₁₉ are fabricated. The 4-phase composition is the same as used in the work of Men *et al.* [8], where no amorphization was observed after Xe and Au ion irradiation of the composite. 3Y-TZP acts as a nuclear fuel surrogate (similar to the fluorite structure and with very low thermal conductivity), non-fissile phases such as Al₂O₃ and MgAl₂O₄ improve thermal conductivity at elevated temperatures, and LaPO₄ (monazite) is used for its capability to incorporate a wide variety of the radionuclides into a single-phase structure [9]. The stability of monazite with corundum, spinel, and zirconia fluorite phases can be observed from geological deposits [10]. In the second ceramic composite, CeO₂ is used as an alternate surrogate for UO₂ (it also has the fluorite structure and similar low thermal conductivity), and spinel is included as the high thermal conductivity phase. The magnetoplumbite CeMgAl₁₁O₁₉ phase is an example of a potential deviation when phases are not stable with respect to one another at high temperatures, as it is produced by a reaction between initial starting compounds of corundum (α -Al₂O₃), spinel, and CeO₂ [11]. The relatively high

thermal conductivity values of alumina and spinel compared to the other constituent phases and UO_2 can clearly be seen in Fig. 1 [12]. Since data on the thermal conductivity $\text{CeMgAl}_{11}\text{O}_{19}$ was unavailable at the time of this study, the temperature dependent thermal conductivity of $\text{LaMgAl}_{11}\text{O}_{19}$ was used instead. Additionally, the majority of the cations in each component phase of the proposed composite systems are cations with relatively low neutron cross sections [13]. This allows for their use within the composite fuel without quantitatively impairing reactor efficiency. The goals of this paper are to i) compare the temperature dependent thermal conductivities of each composite, ii) validate these experimental measurements with those generated by microstructural finite element analysis, and iii) simulate the incorporation of UO_2 for applications in composite nuclear fuel.

Fig. 1. Temperature dependent single phase thermal conductivity values taken from literature for each component in both C-SMp and 3Y-ASM. (Al_2O_3 [12a] , MgAl_2O_4 [12b], UO_2 [2], CeO_2 [14], 3Y-TZP[12c], $\text{LaMgAl}_{11}\text{O}_{19}$ [12d] and LaPO_4 [12e])

2. Experimental Procedures

2.1. Sample Preparation and Characterization

Three-phase ceramic composites with the composition $\text{CeO}_2\text{-MgAl}_2\text{O}_4\text{-CeMgAl}_{11}\text{O}_{19}$ (abbreviated: C-SMp) were created by using starting powders of $\alpha\text{-Al}_2\text{O}_3$ (Baikowski Inter. Corp., Charlotte, NC, crystallite size of 600 nm), MgAl_2O_4 (Baikowski Inter. Corp., Charlotte, NC, crystallite size of 200 nm), and CeO_2 (Sigma-Aldrich Co., St. Louis, Missouri crystallite size of 50 nm). Excess MgAl_2O_4 and CeO_2 were included to promote the reaction given by Eqn. 1 for the in-situ formation of $\text{CeMgAl}_{11}\text{O}_{19}$ (Ce-magnetoplumbite).

This reaction with other phases occurs due to the Ce valence change from 4+ to 3+, which facilitates the formation of the magnetoplumbite structure. Magneoplumbite has been proposed for nuclear waste host applications [15], and the unusual grain morphology provided an opportunity to study an asymmetric three-phase material system.

Powders were attrition-milled for 8 hours with 5 mm zirconia balls (Tosoh, Tokyo, Japan) in isopropanol, dried for 12 hours at 393 K (120°C), crushed with an agate mortar and pestle, and sieved through an 80 μm mesh. Cylindrical samples were fabricated for microstructural characterization by cold isostatic pressing at 380 MPa for 5 minutes, followed by a sintering at 1873 K (1600°C) for 3 hours in air.

Four-phase ceramic composites with the composition $3\text{Y-TZP-Al}_2\text{O}_3\text{-MgAl}_2\text{O}_4\text{-LaPO}_4$ (abbreviated: 3Y-ASM) were created by using starting powders of 3 mol.% yttria tetragonal zirconia polycrystals (3Y-TZP, Tosoh, Japan, crystallite size of 50 nm), $\alpha\text{-Al}_2\text{O}_3$ (same as above), MgAl_2O_4 (same as above), and LaPO_4 (lab made [16], crystallite size of 200-400 nm).

Equal volume fractions of each phase were used. Powder processing steps and sintering was the same as above.

Relative density was measured using the Archimedes displacement method in accordance with ASTM B962-08 [17]. Distilled water was used as the immersion fluid, and the measurements were made using a beaker support positioned above the balance pan. Table 1 gives measured density and the compositional breakdown of each system.

Table 1

Volume fraction of each phase in respective composite systems, and single phase density taken from star quality powder diffraction file cards.

	Volume Fraction (%)	Density (g/cm ³)	Density Reference*
3-phase composite		4.863	
CeO ₂	30	7.215	JCPDS no. 34-0394
MgAl ₂ O ₄	25	3.579	JCPDS no. 21-1152
CeMgAl ₁₁ O ₁₉	45	4.287	**
4-phase composite		4.674	
3Y-TZP	27	6.242	JCPDS no. 60-0503
Al ₂ O ₃	25	3.984	JCPDS no. 46-1212
MgAl ₂ O ₄	25	3.579	JCPDS no. 21-1152
LaPO ₄	23	5.111	JCPDS no. 84-0600

* Joint Committee on Powder Diffraction Standards (JCPDS), ** Calculated value.

X-ray diffraction (XRD) analysis was conducted with a Smart Lab X-ray Diffractometer (Rigaku, Tokyo, Japan) using Cu-k_α radiation (wavelength 0.15406 nm) to verify phases and compositions. Scans consisted of 0.05° steps from 20° to 90° using Bragg-Brentano optical configuration. Scanning electron microscopy (SEM) was performed using a FEI XL 30 FEG (FEI, Eindhoven, The Netherlands). To prepare for SEM analysis, samples were cross-sectioned, polished to a 0.1μm finish, and thermally etched for 0.5 hours at 1273 K (1000°C). To prevent electrical charging during SEM analysis, a thin film of iridium was deposited onto the sample surface using an IBS/e Ion Beam Sputter Deposition System (South Bay Technology, San Clemente, CA). Chemical compositions were identified by energy-dispersive x-ray spectroscopy (EDS) performed with a 50mm² detector (Oxford Instruments).

2.2. Thermal Measurements

The thermal conductivity, $\kappa(t)$, for each composite is calculated using the relationship:

$$\kappa(t) = \alpha(t) \cdot c_p(t) \cdot \rho(t) \quad \text{Eqn. 2}$$

where $\alpha(t)$ is thermal diffusivity, $c_p(t)$ is specific heat capacity and $\rho(t)$ is density. Each parameter is determined separately over the temperature range of 373-1273 K (100-1000°C). Thermal diffusivity of the sintered samples was measured under ultra-high purity Ar using a laser flash apparatus (Netzsch 457 LFA, Germany). Prior to measurement, both the top and the bottom surfaces of each sample were coated with a thin layer of colloidal graphite to enhance the absorption of the laser. The specific heat capacity of each sample as a function of temperature was measured using a differential scanning calorimeter (Netzsch 404 F1 DSC, Germany). Samples were analyzed in ultra-high purity Ar at a heating rate of 20°C/min in an alumina-lined Pt crucible. Heat capacity was calculated during dynamic heating using the ratio method with sapphire as the reference. The thermal expansion of each sample was measured using a horizontal push-rod dilatometer (Netzsch 402 CD, Germany). Samples were tested under ultra-high purity Ar at a heating rate of 5°C/min using an alumina reference standard. The expansion measured was then used to determine the temperature-dependent density as required by Eqn. 2.

2.3. OOF2 Simulations

Object-oriented finite-element analysis version 2 (OOF2) [18], an open access software developed at the National Institute of Standards and Technology (NIST), is used for these simulations due to its capability to account for size, shape, and distribution of each phase within the microstructure. Two-dimensional SEM micrographs were converted to multi-color images in post-processing. Each phase, identified by backscattered phase contrast and EDS, was

represented by a single color in order to create finite-element meshes adapted to the microstructure of the material, and were assigned their appropriate single phase material property. The mesh, which is constructed from a network of nodes, segments, and elements, and which contains the physical information of the system (i.e. x-y coordinates and k values), is repeatedly refined to create a high node density at phase interfaces. This allows for a highly accurate finite element solution.

Input values for the thermal conductivity as a function of temperature for each component were taken from experimental results on single-phase materials, as shown in Fig. 1. A thermal gradient was simulated in the vertical direction of the image by assigning a 10 degree difference between the top and bottom boundaries, and keeping the sides adiabatic. The heat equation was solved by the conjugate gradient method and each node of the mesh was assigned an x and y heat flux component. By setting the out-of-plane (z) heat flux components to zero, OOF2 removed the third dimension from the problem; this is analogous to plane-stress analysis used in fracture mechanics. The resulting 2D heat flux was integrated along a boundary of the image and used to determine the effective thermal conductivity:

$$k_{\text{eff}} = \frac{L_y Q}{L_x (T_{\text{bottom}} - T_{\text{top}})} \quad \text{Eqn. 3}$$

where k_{eff} is the effective thermal conductivity of the composite, Q (watts per meter of thickness in z) is the OOF2 heat flux integrated across the bottom boundary, L_y and L_x are the image dimensions, and T_{bottom} and T_{top} are the temperature values assigned to the bottom and top boundaries. The effective thermal conductivity of the composite was determined by simulating a thermal gradient across an image at various temperatures ranging from 373-1273 K (100-

1000°C). Three representative SEM micrographs were used for each composition, and the typical variability between each simulation of the same composition was less than 1%.

3. Results and Discussion

3.1. Phase and Microstructure Characterization

From the compositional breakdown and single phase densities given in Table 1, the theoretical density was calculated to be 4.988 g/cm³ and 4.752 g/cm³, for the 3-phase (C-SMp) and 4-phase (3Y-ASM) composites respectively. Using the measured densities (determined by the Archimedes method), the relative densities for each system were calculated to be 97.5% and 98.4% based upon the targeted volume fraction reported in Table 1, for C-SMp and 3Y-ASM respectively. A $\pm 0.5\%$ error in the relative densities was observed for both systems.

Fig. 2 shows the X-ray diffraction (XRD) patterns for both C-SMp and 3Y-ASM composites. Overlapping of peaks was minimal for the C-SMp system (Fig. 2a), allowing for each phase to be easily indexed. CeO₂ and MgAl₂O₄ were indexed using JCPDS no. 34-0394 and 21-1152 respectively, while CeMgAl₁₁O₁₉ was indexed using the LaMgAl₁₁O₁₉ pattern, JCPDS no. 70-6968, since there is currently no star-quality pattern for CeMgAl₁₁O₁₉ in the International Centre for Diffraction Data (ICDD) database. Numerous overlapping peaks in the 3Y-ASM system (Fig. 2b) resulted in the indexing of only the major peaks for each phase. The existence of monoclinic ZrO₂ (m-ZrO₂) can be seen in the 3Y-ASM X-ray diffraction pattern due to partial transformation from tetragonal ZrO₂ (3Y-TZP) to the monoclinic structure. Al₂O₃, LaPO₄, m-ZrO₂ and 3Y-TZP were indexed using JCPDS no. 46-1212, 84-0600, 37-1484 and 60-0503 respectively.

Fig. 2. X-ray diffraction spectrum of (a) C-SMp and (b) 3Y-ASM composites. The presence of both 3Y-TZP and monoclinic zirconia ($m\text{-ZrO}_2$) was seen in 3Y-ASM.

The surface morphology of each composite observed by SEM is shown in Fig. 3 for representative microstructures of the C-SMp and 3Y-ASM samples. In Fig. 3a, the dark phase corresponds to MgAl_2O_4 spinel, the light phase is CeO_2 , and the elongated acicular grains are $\text{CeMgAl}_{11}\text{O}_{19}$. This asymmetrical grain growth seen in $\text{CeMgAl}_{11}\text{O}_{19}$ is due to the anisotropic tetragonal crystal system with a c lattice parameter that is more than twice as large as the a lattice parameter, as well as the preferred growth occurring in the c direction. Fig. 3b shows a representative microstructure of the 3Y-ASM composite. The darkest phases are Al_2O_3 or MgAl_2O_4 , the gray phase is LaPO_4 and the lightest phase is 3Y-TZP. The similar density of Al_2O_3 and MgAl_2O_4 make phase identification through contrast alone difficult. For this reason, energy-dispersive x-ray spectroscopy (EDS) was performed on each grain in the image to determine its chemical composition (EDS analysis was also performed on the C-SMp composite to ensure correct phase identification). Although the presence of some $m\text{-ZrO}_2$ was observed in the XRD pattern for the 3Y-ASM composite, identification between the zirconia polymorphs

proved unreliable by EDS and zirconia grains were assumed to be 3Y-TZP for computational analysis (reasoning for this is discussed below).

Fig. 3. Scanning electron microscope image in backscatter electron mode of a) 3-phase composite where MgAl_2O_4 is the dark phase, $\text{CeMgAl}_{11}\text{O}_{19}$ is the gray phase and CeO_2 is the light phase, and b) 4-phase composite where MgAl_2O_4 and Al_2O_3 are the dark phases, LaPO_4 is the gray phase and 3Y-TZP the light phase. Energy-dispersive x-ray spectroscopy is used to differentiate each phase.

3.2. Thermal Measurements

The technical coefficient of thermal expansion (CTE) and density of C-SMp and 3Y-ASM composites, measured as a function of temperatures ranging from 373-1273 K (100-1000°C), are shown in Fig. 4. Data collected near room temperature suffered from high error due to temperature gradients that developed during initial heating of the sample and were discarded. Typical monotonic behavior for both the CTE and density as temperature increased was observed for C-SMp. In the 3Y-ASM composite, a discontinuity occurs in the temperature range of 923-973 K (650-700°C). This sudden jump in the expansion curve is interpreted as a phase transformation. Upon cooling, the tetragonal to monoclinic phase transformation occurs at 1443 K (1170°C) for pure ZrO_2 . Lower temperature transformation occurs due to yttria doping and mechanical constraints of the stiff matrix formed by the composite. Fig. 4b also shows this jump

change reflected also in the sample density, which coincides with the transformation of monoclinic ($\rho=5.817 \text{ g/cm}^3$) to tetragonal ($\rho=6.343 \text{ g/cm}^3$) zirconia. The presence of both zirconia polymorphs is seen in the XRD data of Fig. 2 from room temperature experiments, suggesting that some of the 3Y-TZP transforms to monoclinic upon cooling after sintering and transforms back to tetragonal 3Y-TZP upon re-heating. Although not shown here, a reversal of the expansion is observed upon cooling in approximately the same temperature regime. This also supports the hypothesis that Y-TZP within the sample is responsible for the observed phase transformation.

Fig. 4. Density (right axis) and coefficient of thermal expansion (left axis) measured over a temperature range of 373-1273 K (100-1000°C) for (a) C-SMp and (b) 3Y-ASM.

Fig. 5 shows the thermal diffusivity and heat capacity for each composite system. For both compositions, the thermal diffusivity decreases monotonically as temperatures increase up to 1273 K (1000°C), with values falling to 0.87 mm²/s and 0.84 mm²/s for C-SMp and 3Y-ASM, respectively. Similar to Fig. 4b, a discontinuity in the heat capacity of 3Y-ASM was observed within the temperature range of 923-973 K (650-700°C), which is attributed to the zirconia phase transformation. No discontinuity was seen for the thermal diffusivity of 3Y-ASM. The other discontinuities explained by the monoclinic to tetragonal phase transformation, are in contrast to the continuous decrease in the thermal diffusivity, likely due to thermal transport being limited by the monazite, which is the most insulating phase. As the enthalpy of transformation is an apparent specific heat observed due to the experimental technique employed, the specific heat capacity of 3Y-ASM at temperatures within the region of the phase transformation was determined by smoothing the surrounding data to remove the enthalpy peak.

Fig. 5. (a) Thermal diffusivity and (b) heat capacity measured over a temperature range of 373-1273 K (100-1000°C) for C-SMp and 3Y-ASM.

The thermal conductivity as a function of temperature for C-SMp and 3Y-ASM composite systems is shown in Fig. 6. For both systems, the thermal conductivity decreases as temperature decreases. 3Y-ASM has a higher thermal conductivity at lower temperatures than the C-SMp system (25% higher at 373 K (100°C)), but begins to converge with C-SMp as temperature increases (only a 5% greater thermal conductivity at 1273 K (1000°C), falling within the 5% error of the measurement). This effect is primarily due to the Al_2O_3 phase in 3Y-ASM

sharply decreasing in thermal conductivity. The discontinuity in density resulting from the phase transformation from 923-973 K (650-700°C) for 3Y-ASM does not affect the thermal conductivity calculated by Eqn 2. This indicates that the large decrease in thermal diffusivity is the dominant factor controlling the lower thermal conductivity with increasing temperature.

Fig. 6. Thermal conductivity of C-SMp and 3Y-ASM calculated by the laser flash method over a temperature range of 373-1273 K (100-1000°C).

3.3. OOF2 Simulations

Single phase temperature-dependent thermal conductivity values, taken from Fig. 1, were inputted into OOF2 software along with a solid color representation of a SEM micrograph in order to approximate the effective thermal conductivity for each composite system. Fig. 7a shows the color representation of the original SEM image of C-SMp from Fig. 3a. An adaptive mesh was fitted to the grain boundary interface of each phase (Fig. 7b), where a finer mesh and higher node density can be seen along the phase interface. After the simulation, a heat flux map was generated (Fig. 7c), showing regions of high and low heat flux. Aside from the porosity, $\text{CeMgAl}_{11}\text{O}_{19}$ appears to be the phase with the lowest heat flux, which is expected due to its low thermal conductivity. The temperature-dependent thermal conductivity of air, which ranges

from 0.023 W/m·K at 300K to 0.083 W/m·K at 1300K [19], was used for the thermal conductivity of porosity. Considering that porosity makes up <2% of the composite, it can be assumed to have a negligible contribution to the overall heat conduction.

Fig. 7. OOF2 images of the C-SMp composite shown in Fig. 3a (a) color representation of the microstructure, (b) finite element meshing, and (c) heat flux map generated by the simulation. Al_2O_3 is represented by red, $\text{CeMgAl}_{11}\text{O}_{19}$ blue, CeO_2 green and porosity black.

Fig. 8. OOF2 images of 3Y-ASM composite shown in Fig. 3b (a) color representation of the microstructure, (b) finite element meshing, and (c) heat flux map generated by the simulation. MgAl_2O_4 is represented by red, LaPO_4 blue, Al_2O_3 green, 3Y-TZP white and porosity black.

Fig. 8 depicts a similar set of images as Fig. 7 for the 3Y-ASM composite system. Blocking of heat flux pathways is observed in Fig. 7c due to the irregular acicular elongated particle morphology of the $\text{CeMgAl}_{11}\text{O}_{19}$ phase. This magnetoplumbite is the lowest thermal conductivity phase in this system, and its particle morphology prevents the higher thermal conductivity alumina from forming an interconnected network. Although each phase is above the

percolation limit, and interconnected pathways are not present in the 2D representation of the microstructure, that does not rule out the existence of percolation pathways in 3D.

In contrast, Fig. 8c clearly shows heat flow pathways formed between the higher thermal conductivity phases, i.e. Al_2O_3 and MgAl_2O_4 . These pathways are a result of all phases being above the percolation limit, an even distribution of each phase, and a uniform grain morphology seen in the microstructure. Fig. 9 shows how Al_2O_3 and MgAl_2O_4 grains, along with small regions of 3Y-TZP, create a pathway for heat flow from the bottom of the image to the top. This contiguity of highly conducting phases allows the composite to maintain an overall high thermal conductivity despite containing 25 vol % LaPO_4 , which is well known for its extremely low thermal conductivity.

Fig. 9. Heat flux pathway formed by network of highly conducting Al_2O_3 (A) and MgAl_2O_4 (S) grains, along with small regions of 3Y-TZP (Z) gains.

3.4. Surrogate Comparison

The simulated thermal conductivity of C-SMp and 3Y-ASM via OOF2 was compared to the experimentally determined thermal conductivity (from Fig. 6) and is shown in Fig. 10. A discrepancy between the experimental and simulated thermal conductivities for C-SMp is

observed at the low temperature range 373-673 K (100-400°C), while above 673 K (400°C) the two data sets are convergent. Similarly, a low temperature discrepancy is observed in 3Y-ASM. For the C-SMp composite, a difference of 9.1% at 373 K (100°C) and 1.1% at 1273 K (1000°C) was measured between the laser flash method (LFM) and OOF2. In comparison, the 3Y-ASM composite showed a difference of 17.6% at 373 K (100°C) and 0.3% at 1273 K (1000°C). One aspect that may contribute to the greater deviation of the 3Y-TZP compared to C-SMp at low temperatures is the approximation that was made in the simulation that the 3Y-TZP is completely tetragonal zirconia. Above 673 K (400°C), the percent difference between LFM and OOF2 is <3% for both composite systems, which falls with the standard 5% error of the measurement.

However, the disagreement between the experimental and computational results at low temperatures for both systems could be partially due to the thermal resistivity of the grain boundary interface, known as the Kapitza resistance [20], not being taken into consideration in the OOF2 simulations. This would affect the models in two ways. First, the Kapitza resistance is higher at low temperatures, which could contribute to the low temperature discrepancy between the computational modeling and experiments. Second, the Kapitza resistance is typically higher for interfaces between dissimilar phases than for grain boundaries between similar phases [21]. Information on the Kapitza resistance of single phase oxide materials such as Al_2O_3 [22] and two phase systems such as YSZ/ Al_2O_3 [23] has been previously published, but to the authors' knowledge, no data on three or four phase systems is currently available in the literature. Work is currently underway to incorporate grain boundary and interface effects into the OOF2 simulations with known and predicted Kapitza resistance data.

Fig. 10. Thermal conductivities simulated by OOF2 and measured through the laser flash method (LFM) for C-SMp and 3Y-ASM. A good agreement between methods is observed at temperatures above 673 K (400°C).

Hypothetical composites with UO_2 instead of the surrogates, labeled U-SMp and U-ASM for the 3-phase and 4-phase compositions, respectively, were also simulated using OOF2. UO_2 single phase thermal conductivity values replaced CeO_2 in the C-SMp composite, and replaced 3Y-TZP in the 3Y-ASM composite. The thermal conductivity over the temperature range 373-1273 K (100-1000°C) for the two hypothetical composites was determined by OOF2 using the same procedures described above. Fig. 11 shows the results of the OOF2 simulations and includes the thermal conductivity of single phase UO_2 for comparison. U-SMp had an even lower thermal conductivity than single phase UO_2 from 373-973 K (100-700°C), and less than a 6% increase in thermal conductivity compared to pure UO_2 above 973 K (700°C). The non-uniform morphology and extremely low thermal conductivity of the large volume of $\text{CeMgAl}_{11}\text{O}_{19}$ is clearly detrimental to overall thermal conductivity of the composite system. In contrast, the composite $\text{UO}_2\text{-Al}_2\text{O}_3\text{-MgAl}_2\text{O}_4\text{-LaPO}_4$ had a 12-16% increase in thermal conductivity from 673-1273 K (400-1000°C) compared to single phase UO_2 . Even in this case, the incorporation of extremely low thermal conductivity LaPO_4 limits the magnitude of the

increase in conductivity, and therefore compositions with alternate phases to monazite is currently being evaluated.

Fig. 11. OOF2 simulated composites where UO₂ is substituted for CeO₂ in C-SMp (U-SMp) and 3Y-TZP in 3Y-ASM (U-ASM). A single phase thermal conductivity of UO₂ is shown for comparison.

4. Conclusions

In this paper, inert matrix 3-phase and 4-phase ceramic composites to be used as surrogate nuclear fuels were prepared and characterized using CeO₂ or 3Y-TZP as surrogate phases for UO₂. Thermal properties such as temperature dependent density, thermal expansion coefficient, specific heat capacity, thermal diffusivity and thermal conductivity were reported for each system. Computational simulations using OOF2 were used to calculate the effective thermal conductivity and showed good agreement with experimental methods within the error of the experimental technique at temperatures above 673 K (400°C) for both composite systems. The ability to predict the thermal behavior of UO₂ incorporated nuclear fuel composites, without the need for synthesis of a diverse set of sample compositions, makes this approach valuable to screen non-fissile material candidates and to construct various microstructural designs. It may be

possible to determine the optimal microstructure and phase ratio required for a composite nuclear fuel design prior to fabrication, enabling a more efficient and effective design process using this approach.

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