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Silicon carbide as an inert-matrix for a thermal reactor fuel

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Abstract

This paper reports progress on work to develop methods of fabricating silicon carbide with cerium, as a substitute for plutonium, to achieve high densities at low sintering temperatures. Densities of 97–99% of TD were achieved at 1943 K for cerium oxide concentrations in the starting powders from 5 to 20 wt%. Also reported are: specific heat and thermal conductivity measurements of as-fabricated SiC; compatibility of SiC with coolant and Zircaloy-4; and accelerator simulations of in-reactor fission-fragment damage. The thermal conductivity for as-fabricated SiC with additives was 48 W m^{-1} K^{-1} at 298 K decreasing to about 18 W m^{-1} K^{-1} at 1773 K. Calculations, based on the measured thermal conductivity, show that the inert-matrix fuel could operate at 55 kW m^{-1} linear power at a centre-line temperature of only 673 K, i.e., only 100 K above coolant temperature, although it is expected that irradiation-induced degradation of thermal conductivity will lead to higher operating temperatures as burnup accumulates. The increase in central temperatures due to a possible decrease in thermal conductivity caused by fast-neutrons are calculated in the text. SiC appears to be a very promising candidate as an inert-matrix fuel for water-cooled reactors.

1. Introduction

We have been investigating silicon carbide as part of the current world effort to develop an inert-matrix material to burn minor actinides or military or civilian plutonium, while generating little or no additional actinide wastes or fissile isotopes. Silicon carbide has the following well-established properties, making it a promising candidate for investigation as an inert-matrix material:

- very high thermal conductivity (approximately as high as aluminum for very pure SiC with no porosity), giving the promise of low fuel operating temperatures and improved safety
- high melting temperature ~2973 K
- good chemical stability, e.g., strong resistance to oxidation in air and air-moisture atmospheres (fewer data are available on corrosion in high-temperature water)
- no unacceptable phase changes and
- low neutron absorption.

Issues relating to the use of SiC as a carrier for Pu, including fabrication, in-reactor behaviour and waste-disposal must be investigated. In this paper, we report on initial efforts to fabricate SiC with sintering aids and cerium as a plutonium substitute, specific heat and thermal conductivity measurements of SiC, compatibility of SiC with coolant and with Zircaloys, and accelerator-based simulations of in-reactor fission-fragment damage to SiC specimens. Reactor physics aspects have been addressed separately [1]. Long-term dissolution rates of SiC in ground water has not been studied, and remains a potential issue.

2. Fabrication

One difficulty of fabricating SiC is that it generally requires very high temperatures for pressureless sintering. In addition, it is desirable to use conventional fabrication processes and equipment, e.g., furnaces, that are compatible with UO_2 and mixed-oxide (MOX) fuel fabrication. This would mean that existing fabrication lines for commercial fuel would not need to undergo large changes. Also, existing small-scale MOX glove box fabrication facilities could easily adapt to fabricating

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inert-matrix fuel for irradiation tests. High-density fuel at reasonable sintering temperatures was the main goal. However, since the Pu has a high fissile content and tends to reside in small second-phase particles, there will be no problem in attaining sufficient fissile loading, and very high densities may not be required. Also, since the fuel will run at low temperatures, in-reactor sintering of lower density fuel may not be an issue. But higher densities are generally preferable and remained a goal throughout this work. To address these issues and test fabrication of SiC with a substitute for plutonium, three fabrication campaigns were conducted by two of the authors at Queen's University, under contract to AECL, between 1996 and 1998.

In general, the following processes were used for silicon carbide fabrication:

- blend starting powders, binder (polyethylene glycol), and lubricant (oleic acid), with extra carbon to partially reduce oxide starting powders and to ensure that no free silicon was present in the final product;
- ball mill in methanol with ZrO_2 grinding media;
- dry, crush and screen through a sieve with 1.6-mm openings;
- uniaxial press (cold) at 50 MPa;
- isostatic press (cold) at 250 MPa; and
- sinter in graphite resistance furnace in static argon at 1 MPa;

The first campaign utilized Al_2O_3 (2.55 to 12.5 wt%), TiO_2 (0 and 5 wt%), Y_2O_3 (1.08 to 10.92 wt%) and SiO_2 (0.0, 1.3 and 2.6 wt%) as sintering aids (not all components at the same time) and aimed to produce silicon carbide at 93% to 95% of theoretical density (TD) at sintering temperatures below 2273 K. In addition, cerium was added as a plutonium substitute in 2 forms, CeO_2 and CeC_2, in a wide concentration range (the Ce contained in 3.4 wt% CeO_2 in SiC represents the reference weapons-grade amount of Pu for a 43-element CANDU® CANFLEX bundle design [1]) to determine what densities could be achieved and how the cerium would be incorporated in the silicon carbide matrix. Fig. 1 is a micrograph showing a typical phase structure. XRD usually showed the presence of two new phases, $AlCeO_3$ and Al_3Ce. (Phase identification was made by comparing XRD results with optical micrographs for many samples, and observing the correlation between the amounts of the various phases present in the micrographs and by XRD.) Fig. 2 shows the densities obtained when sintering with Al_2O_3 and Y_2O_3 and CeO_2 at 2133 K for 30 min. Below 5 wt% CeO_2, the densities were approximately 95% of TD. [1] At higher concentrations of CeO_2, the densities decreased, but this may also be partly due to decreasing concentrations of sintering additives.

The second campaign aimed to improve on the previous results by holding the amount of Al_2O_3 and Y_2O_3 constant at 8 wt% and 10.6 wt%, respectively, and to test with even higher CeO_2 concentrations from 0 to 30 wt%.

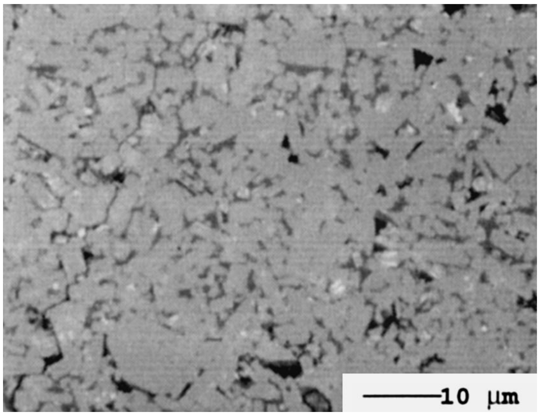

Fig. 1. Optical micrograph of a polished section of SiC fabricated with 8 wt% Al_2O_3 and 5 wt% CeO_2, sintered at 2273 K, showing $AlCeO_3$ (dark gray) and Al_3Ce (white phase). Black spots are sintering porosity; light gray spots in the background are SiC.

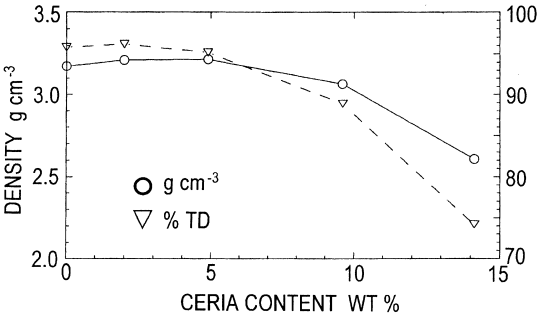

Fig. 2. Variation of density with CeO_2 content for SiC fabricated with 2.55 to 8.0 wt% Al_2O_3, and 1.1 to 3.4 wt% Y_2O_3; sintering at 2133 K for 30 min. The concentrations used of Al_2O_3 and Y_2O_3 decreased as the concentration of CeO_2 was increased.

Fig. 3 shows the surprising results. At temperatures as low as 1973 K, densities of 96 to 99% were achieved for all ceria concentrations, not including that with 0% ceria. Ceria itself has acted as an excellent sintering aid. The sample with 0% ceria sintered the poorest, except at the very highest temperatures. Even at a temperature as

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[1] For these fabrication campaigns, as well as for subsequent thermal properties measurements, approximate values and general trends were the objectives, rather than exact values, since the final product will use uranium or plutonium and, possibly, actinides instead of cerium. Therefore, no attempt was made to quantify precision or errors in quantities such as TD.

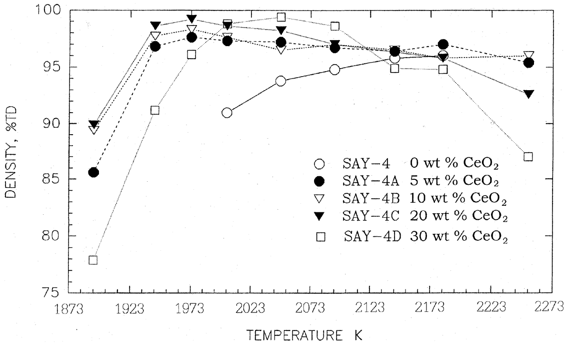

Fig. 3. Variation of density with sintering temperature of SiC fabricated with (before sintering values) 8 wt% Al_2O_3, 10.6 wt% Y_2O_3 and 0–30.0 wt% CeO_2.

low as 1943 K, densities of 97 to 99% were achieved for all ceria concentrations used, except the highest – 30 wt%. X-ray diffraction showed that Al_2O_3, Y_2O_3, and CeO_2 reacted during sintering to produce two clearly identifiable compounds, $3Y_2O_3{\cdot}5Al_2O_3$ garnet (YAG) and cerium aluminate ($CeAlO_3$). The amount and form of these two phases was dependent on the composition and the sintering temperature. For example, between 1893 K and 2143 K the garnet phase XRD peaks were not present for the sample with 30 wt% ceria and were significantly reduced in intensity for the sample with 20 wt% ceria. Surprisingly, above 2143 K XRD did not show the presence of either of the two phases, garnet or cerium aluminate. The reason for this phenomenon is not understood. Optical microscopy (e.g., Fig. 4) showed that the phase identified as garnet plus cerium aluminate (dark gray) was finely distributed between micrometre-sized β-SiC grains. For 20 and 30 wt% concentrations of ceria, the garnet/cerium-aluminate phase was larger in size (6 to 8 μm).

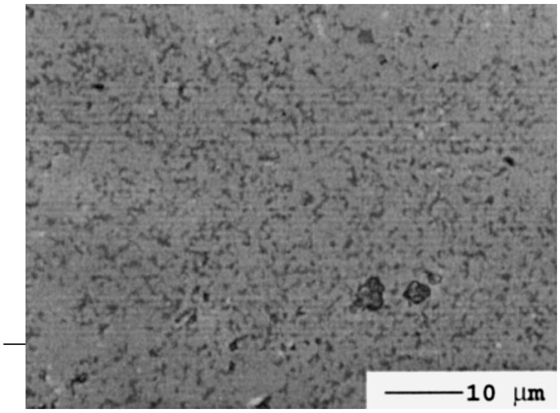

Fig. 4. Optical micrograph of a polished section of the sample (SAY-4B) with 10 wt% CeO_2. Phase identification is the same as for Fig. 2.

The main objective of the third campaign was to find an acceptable fabrication route that did not require the isostatic pressing stage that had previously been used in all fabrication tests. Isostatic pressing is not used in commercial UO_2 fabrication methods for power reactors, and the objective was to make the SiC fabrication route more acceptable to fuel fabricators. In addition, somewhat lower amounts of sintering aids, Al_2O_3 and Y_2O_3, were used. The starting materials were 7 wt% Y_2O_3, 7–8 wt% Al_2O_3, 5–10 wt% CeO_2, 1 wt% oleic acid (lubricant) and 3–11 wt% polyethylene glycol (binder). Sintering temperatures ranged from 1983 K to 2163 K. Cold uniaxial pressing at pressures of 50, 70, 100, 150 and 200 MPa were used. In addition, for comparison, tests were made using an isostatic pressure of 300 MPa.

Sintered densities of 95% to 97% of TD for the samples with 5 wt% CeO_2 were achieved at all pressures (and also for the samples given an isostatic pressing). However, the pressure used affected the formation of cracks in the green compacts, which generally became visible to the naked eye after sintering. No cracks formed at 50 MPa (or for the isostatically pressed samples) at all concentrations of binder. At 70 MPa pressing pressure, only samples made with 5% binder formed with no cracks (four samples made at each composition and pressing pressure). Results were generally the same for the 10 wt% ceria samples. Thus, by judicious selection of the amount of binder and powder pressing pressure, crack-free samples could be made.

3. Specific heat and thermal conductivity

The use of SiC as an inert-matrix carrier for fuel carries the promise of high thermal conductivity and low fuel operating temperatures. Although high thermal conductivities are reported for synthetic single crystals [2], measurements were needed of samples fabricated by us – polycrystalline, slightly porous SiC with additives – to determine if that promise would be fulfilled. Therefore, measurements of specific heat and thermal diffusivity were made on two of the samples fabricated at Queen's University:

- sample 31-A with 8.0 wt% Al_2O_3, 5.0 wt% CeO_2 and 2 wt% C (plus lubricant and binder) concentrations in the powders prior to sintering and sintered at about 2273 K to about 94% TD,
- and sample SAY-6AI with 6.9 wt% Al_2O_3, 5.0 wt% CeO_2, 7.2 wt% Y_2O_3 and 5 wt% binder starting concentrations, pressed uniaxially at 70 MPa (no isostatic pressing), sintered at a temperature between 1800 to 2163 K to about 97% of TD.

Specific heat measurements were made from ambient room temperature (RT) to 1273 K using both a Perkin–

Elmer Model DSC-2 Differential Scanning Calorimeter with sapphire as the reference material for measurements from RT to 973 K and a Netzsch Model 404 DSC to 1273 K. This work was done by Thermophysical Properties Research Laboratory, Inc., 2595 Yeager Rd., West Lafayette, IN 47906, USA on contract to AECL. Plotted results are a smooth curve through the combined results. Measurements were made during both heating and cooling of the sample. Results for sample SAY-6AI are shown in Fig. 5, along with literature results for pure SiC for comparison. Only at high temperatures do the additives make any significant difference from pure SiC. The values for the apparent specific heat of sample 31-A were similar to that for SAY-6AI, showing, however, more separation between the heating and cooling curves above 500 K and a small spike at about 873 K on the heating curve. These differences probably indicate a small degree of phase-volume adjustment during this temperature cycle. (This may be a simple technique to ensure in future that a product being developed for irradiation with uranium or plutonium is stable against phase-volume adjustments.) For subsequent calculations of thermal conductivity of each sample, the specific heat values obtained during the heating and cooling portions of the cycle were smoothed and averaged.

Thermal diffusivity measurements (α) were made using the laser flash diffusivity method (see, e.g. [5]). These values were then combined with measurements of the geometric density (ρ) of the samples and specific heat (c_p) to arrive at thermal conductivity (κ), according to the formula

$$\kappa = \rho \cdot \alpha \cdot c_p$$

Results are given in Fig. 6 along with literature values for UO_2 and for pure SiC for several fabrication routes for comparison. Thorne [7], Kennedy [8] and Price [9] have all reported that the thermal conductivity of SiC irradiated in a fast-neutron flux decreases significantly. Price's measurements were for pyrolytic cubic SiC irradiated at 773 K to 1373 K to doses of 2.7 to 7.7×10^{25} m^{-2} ($E > 0.18$ MeV). It is not clear at present whether neutron-irradiation in combination with fission-fragment damage in SiC would also cause such a reduction in thermal conductivity. Complete lack of swelling in the tests described below (see the section 'Accelerator Simulations of In-Reactor Fission-Fragment Damage') suggest that localized heating due to fission-fragment spikes may anneal the damage. This would not, of course, apply to portions of the lattice that do not experience fission-fragment spikes.

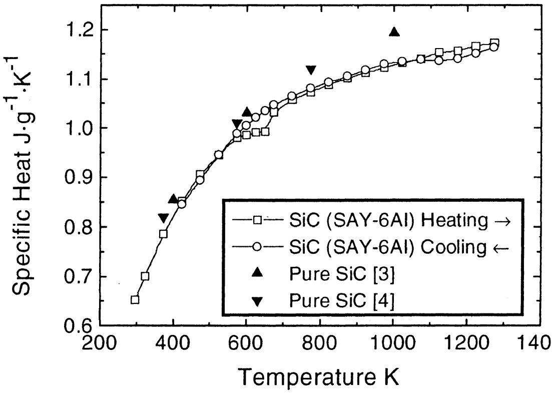

Fig. 5. Apparent specific heat capacities of SiC sample SAY-6AI with 8 wt% alumina and 5 wt% ceria additives, obtained during heating to 1273 K and cooling back to 298 K. Literature values for pure SiC are also shown for comparison.

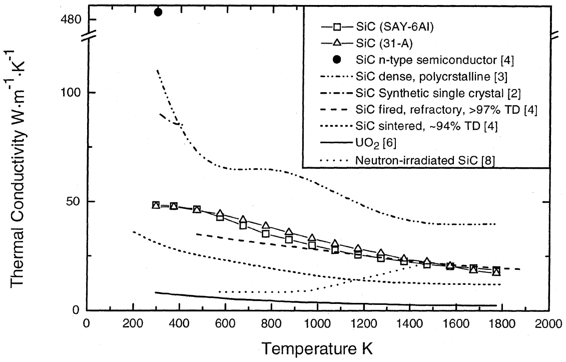

Fig. 6. Thermal conductivity of two samples of as-fabricated SiC with additives. The Figure also shows literature values of thermal conductivity of pure SiC and UO_2 for comparison. The values for pure SiC vary greatly (e.g., 31 to 490 W $m^{-1}K^{-1}$ at 300 K) depending on the fabrication route [2–4,6,8].

These values of thermal conductivity can be used to obtain an approximate value of the centre temperature of SiC-based matrix materials using the formula

$$P = 4 \cdot \pi \int_{T_S}^{T_C} \kappa \cdot dT,$$

where P is the linear power of the fuel element and T_S and T_C are the surface and centre temperatures of the fuel. The validity of this formula depends on uniform heat production in the fuel rod, and so loses accuracy when the fissile loading is such that self-shielding is significant. With this limitation, the formula provides linear power as a function of centre temperature. For example, if the surface temperature is 573 K and the linear element power is 55 kW m^{-1} (approximate peak power in a CANDU reactor), the centre temperature would be only 673 K based on our measured values of thermal conductivity shown in Fig. 6. This graphically demonstrates the advantage of using a high thermal conductivity material-low operating fuel temperatures. Using the thermal conductivity for neutron-irradiated SiC shown in Fig. 6, the centre temperature would be about 1100 K, which is still low for such a high linear-heating rate and for a refractory ceramic with a melting temperature of ~2973 K. For UO_2, the same conditions would produce a calculated central temperature of about 1773 K.

4. Compatibility of SiC with coolant and Zircaloy

Compatibility measurements of SiC in deoxygenated H_2O at 593 K, pH 10.3 for 90 d [10] and at 573 K, pH 3 for 32 d [11] have been previously reported. At 593 K, pH 10.3, an 880 mg sample (cubic shape-cut from a pellet) showed a linear (with time) weight loss of about 2% (16 mg) after 90 d (after an initial apparent weight increase of 0.5%). This weight change converts to a loss of about 20 μm from the surfaces. For the pH 3 tests, eleven samples of SiC, 75 to 400 mg, fabricated for the inert-matrix program were also tested at 573 K. Some samples broke into two or more pieces, and a grayish appearance formed on the surfaces, but the weight changes were only 1 to 1.5 mg. Thus the corrosion rate for the pH 3 tests were lower than for the pH 10.3 tests.

Other experimenters have measured even lower corrosion rates for de-oxygenated water. Hirayama et al. [12] measured corrosion of 1.5-g samples of sintered SiC (mainly α-SiC) containing boron and carbon as sintering additives. They used water solutions of pH 4, 6 and 10 at 563 K. For all pH values, deoxygenated and oxygen-saturated solutions were tested, and in each case, the oxygenated solutions corroded the SiC faster than the deoxygenated solutions did, specifically by factors of 10, 15 and 100 for pH values of 4, 6 and 10, respectively. The fastest corrosion rate, 3 μm over 72 h, was that for oxygen-saturated water at pH 10. For de-oxygenated water, the rate, about 0.03 μm over 72 h, was considerably slower than for our tests, but different additives may have an effect, and the level of de-oxygenation may have been greater. In any case, corrosion of SiC would not be a cause for concern in case of cladding breach.

Initial tests of SiC interaction with Zircaloy-4 were performed by pressing polished disc specimens of SiC and Zircaloy-4 under a light pressure and an argon atmosphere in a molybdenum cell at 1273 K, 1773 K and 1973 K. Temperatures were maintained for 1 h at 1273 K and 1773 K and for 15 min at 1973 K. Two types of SiC specimens, fabricated at Queen's University, were used; both contained alumina as a sintering aid; and one contained titania, and the other ceria. Extra carbon had been added to all specimens during sintering, to ensure that no free Si was present in the final products. After cooling and sectioning, the specimens were examined by optical and scanning electron microscopy, to study the extent of interaction.

Although no significant interaction occurred at 1273 K, at 1773 K there was clear evidence of a diffusion-based reaction to form ZrC and free Si. Diffusion of free Si into the Zircaloy disc led to the formation of a molten Zr–Si-rich eutectic phase. This reaction was more pronounced in the 1973 K test specimens, where the amount of molten eutectic phase was sufficient to cause partial dissolution of the Mo-cell sidewalls. Further details are provided in Ref. [8].

The test results indicate that formation of a molten Zr-rich phase could conceivably occur during a hypothetical reactor accident at temperatures lower than the melting point of unoxidized Zircaloy cladding (2033 K). This would allow further attack on the SiC carrier material, thereby allowing any fission products that had exsolved from the plutonium phase to be released. On the other hand, UO_2 interaction with Zircaloy begins at about 1473 K [13] and UO_2 dissolves into molten Zircaloy at 2033 K. The rate and extent of UO_2–Zircaloy interaction depends on the amount of oxygen present; however, it seems that the SiC interaction with sheath is, at least, no worse than the UO_2 interaction. Tests using SiC pellets clad with Zircaloy-4 sheathing, rather than pressed discs in a Mo cell, would provide a better comparison.

5. Accelerator simulations of in-reactor fission-fragment damage

The Tandem Accelerator at the Chalk River Laboratories was used to simulate the severe damage inert-matrix materials will experience from in-reactor fission fragments by generating a beam of 72 MeV iodine (typical fission fragment at a typical fission-fragment energy). This beam was used to bombard the surfaces of candidate inert-matrix materials over a wide ion fluence-range (10^{14} to 10^{17} cm^{-2}) and a wide temperature range (RT to 1473 K). (A ion fluence of 10^{17} cm^{-2} is approximately equivalent to a fuel burnup of 12 MW d kg^{-1} heavy element.) Samples tested were $ZrSiO_4$ (zircon), $MgAl_2O_4$ (spinel), CeO_2 (ceria), $CePO_4$, ZrO_2-doped with Ca, Ce, Er or Y, as well as SiC. After accelerator bombardment of each sample, the surface relief of the 3-mm-diameter beam spot was measured. Significant height of the spot above the original surface was taken as an indication that in-reactor swelling would occur. In general, results were not strongly dependent on dose or temperature.

Results, with figures showing surface relief of many of the samples, were reported in Ref. [10]. Usually samples showed either no surface relief or relief of 1–2 μm (ion-bombardment depth was 10 to 20 μm, depending on material). A benchmark test on Al_2O_3, which is well-known to vitrify and swell in-reactor when used as a fuel-containing matrix, showed surface relief of about 1.6 μm. Similarly, tests on UO_2 did not show swelling (no observable surface relief). Of all the mate-

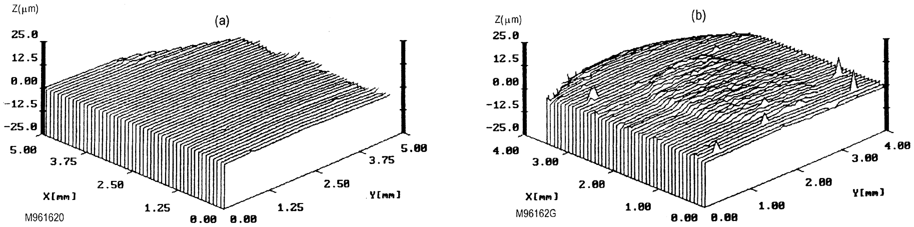

Fig. 7. Surface relief of two samples bombarded in a 72 MeV iodine beam. (a) SiC^{-3} separate bombardments (I fluence 1×10^{15} cm^{-2} at 1473 K, 1.3×10^{14} and 1×10^{15} at ambient temperature); (b) $ZrSiO_4$ (I fluence 1×10^{15} cm^{-2} at 1473 K). The SiC sample shows no evidence of the implantations, but the $ZrSiO_4$ does.

rials tested, the least amount of swelling was observed for UO_2, SiC and stabilized ZrO_2 with any of the dopants. These materials did not show *any* swelling; i.e., laser profilometry could not detect any surface relief at the implantation spots. Fig. 7(a) shows the lack of any surface relief for a typical test on SiC; Fig. 7(b), for comparison, is a typical for samples that did show surface relief. All other materials showed some surface relief, implying that swelling could be expected when used in-reactor as a fuel.

6. Conclusions

Progress has been made in establishing methods of fabricating SiC containing a wide range of cerium, which was used as a plutonium substitute. The methods are similar to those in current use for UO_2 fuel production. High densities were achieved and the required sintering temperatures have been reduced from over 2273 K to approximately 1973 K by judicious use of sintering additives. In fact, ceria itself has proven to be a most effective sintering additive. High density material was fabricated without using cold isostatic pressing, a step not used in UO_2 fuel production. When isostatic pressing was not used, care was required in the selection of uniaxial pressing pressures and binder concentrations to eliminate cracks in the sintered product. Cerium is usually incorporated into the SiC as an $AlCeO_3$ phase, often as micron-sized particles, but also as larger particles (6–8 μm) in the samples fabricated with 20–30 wt% ceria. Above 2103 K sintering temperature, with Al_2O_3 and Y_2O_3 sintering aids, XRD did not show the existence of any phase except SiC. This suggests a total solid solution of all additives; the reason for this is not understood.

Measurements of specific heat capacity and thermal conductivity of SiC specimens fabricated in this program were made; although the thermal conductivity is lower than pure, fully dense SiC, the conductivity is still so high that the central temperature in an operating fuel at 55 kW m^{-1} would only be about 100 K higher than the coolant temperature. Literature values of neutron-irradiated SiC suggest that the thermal conductivity would decrease to the extent that the central temperature would be about 500 K over coolant temperature at a linear heat rating of 55 kW m^{-1}, still low for a refractory ceramic operating at such a high power (c.f., about 1500 K over coolant temperature for UO_2).

In conclusion, silicon carbide containing cerium as a plutonium substitute and a few weight per cent sintering additives has been fabricated using equipment that is compatible with conventional UO_2 or MOX fabrication technology. The thermal conductivity of such SiC-based inert matrix fuels is very high – a clear benefit from the perspective of in-reactor fuel performance and safety considerations. Accelerator simulations of in-reactor damage indicate that such a fuel would be dimensionally stable. Conventional pressurized water coolants and Zircaloy claddings are also compatible with SiC.

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