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Oak Ridge National Laboratory

CAES — Center for Advanced Energy Studies

AML — Advanced Materials Laboratory

I. BACKGROUND

Motivation for Research

An increasing demand for nuclear energy requires reliable and efficient fuels. Release of fission gases in fuels leads to reduced thermal conductivity in the fuel-cladding gap and thus fuel reliability and efficiency(Fig.1.)[1].

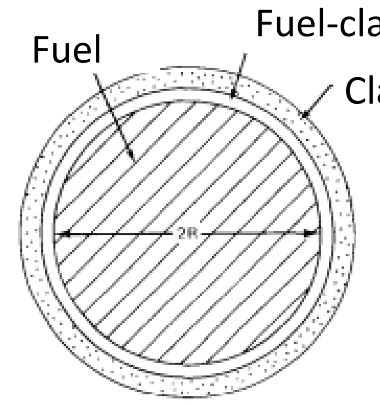

Fig.1. Cross sectional schematic of nuclear fuel-cladding system [2].

Larger grain sizes lead to improved fission product retention (Fig.2.). This study analyzes the impact of manganese dioxide (MnO_2) on the microstructure of cerium dioxide (CeO_2), a surrogate for uranium dioxide (UO_2).

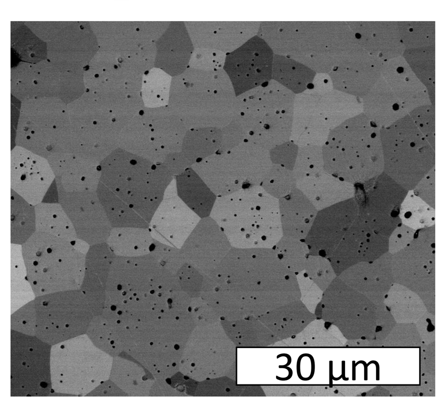

Fig.2. SEM micrograph of the microstructure of UO_2 with average grain size ~10 µm [3].

- A surrogate is used for UO_2 due to [4]:
  - Reduced radiation exposure
  - Decreased costs
  - Increased timeliness of experiments
- CeO_2 is used as a surrogate for UO_2 due to [4]:
  - Common cubic fluorite crystal structure
  - Similar melting temperature
  - Similar thermophysical properties

II. EXPERIMENTAL

Materials Synthesis

- As-received CeO_2 and MnO_2 powders were processed in a high energy planetary ball mill (HEPBM) to mix, reduce particle sizes, and to incorporate Mn^+ into the CeO_2 lattice (Fig.3. and Fig.4.).
- Milled powder was pressed using a dual action die into right cylinder pellets at 150 MPa then sintered with the profile in Fig. 5.

Fig.3. Milled 500 ppm MnO_2-doped CeO_2 powder. Pure, 1000, and 2500 ppm samples were also fabricated.

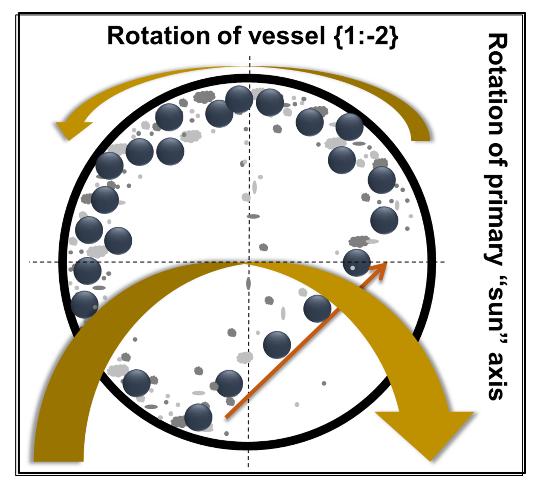

Fig.4. HEPBM motion schematic.

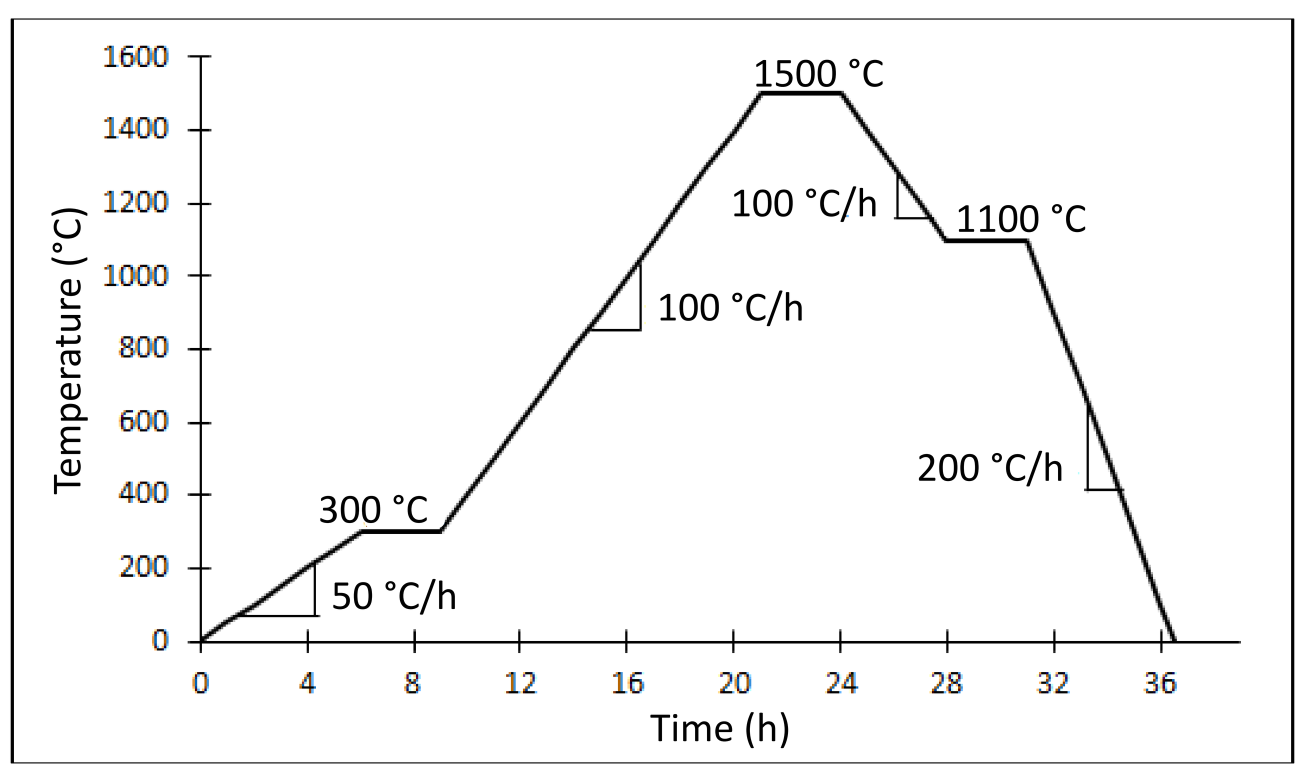

Fig.5. 1500 °C Sintering profile used for pure and doped samples. 1200 °C and 1400 °C profiles were also tested.

Characterization Techniques

- X-ray diffraction (XRD) for phase purity and dopant incorporation analysis
  - A lanthanum hexaboride (LaB_6) standard was used in the MnO_2-doped samples to identify instrumentation shifting.
- Scanning electron microscopy (SEM) for microstructural analysis
- Energy dispersive spectroscopy (EDS) for chemical analysis

III. RESULTS

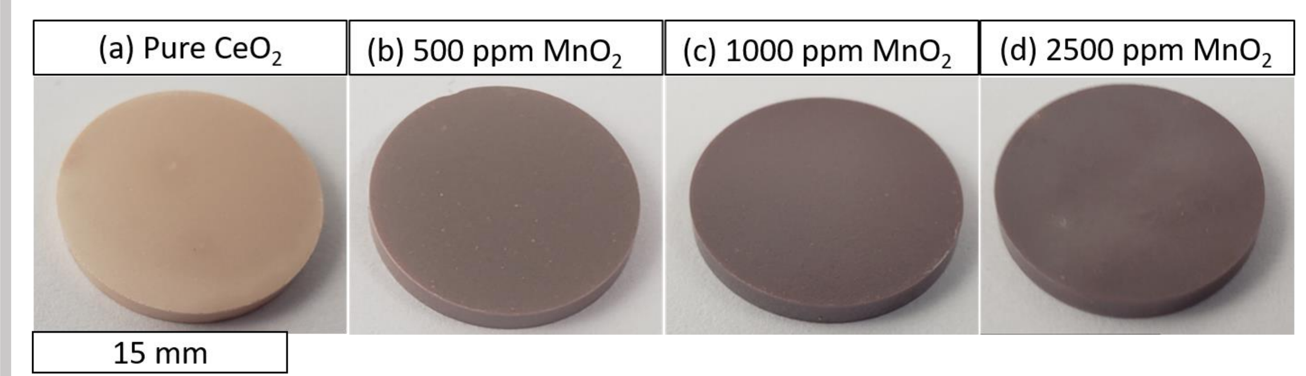

Fig.6. As-sintered specimen sintered at 1500 °C.

Table.1. Reference densities measured by multipycnometer were 7.128 and 5.13 g/cm³ for CeO_2 and MnO_2 respectively. The bulk volume method was used to determine the reference density of the doped samples. Grain size was determined using the ASTM E112−12 circular intercept method [5].

MnO_2 Concentration (ppm)	Sintering Temperature (°C)	Theoretical Density (±2%)	Average Grain Size (µm)
0	1500	94	13.3-15.9
500	1500	96	22.5
1000	1500	94	22.5-26.7
2500	1500	89	22.5-26.7
2500	1400	94	11.2-13.3
2500	1200	95	<2

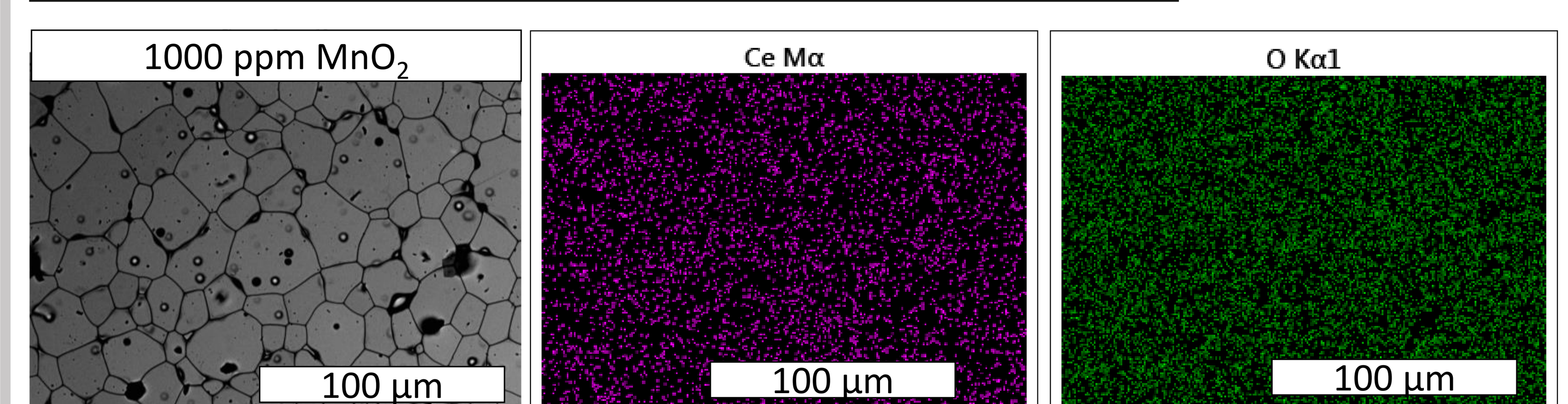

Fig 7. EDS map of 1000 ppm MnO_2-doped CeO_2.

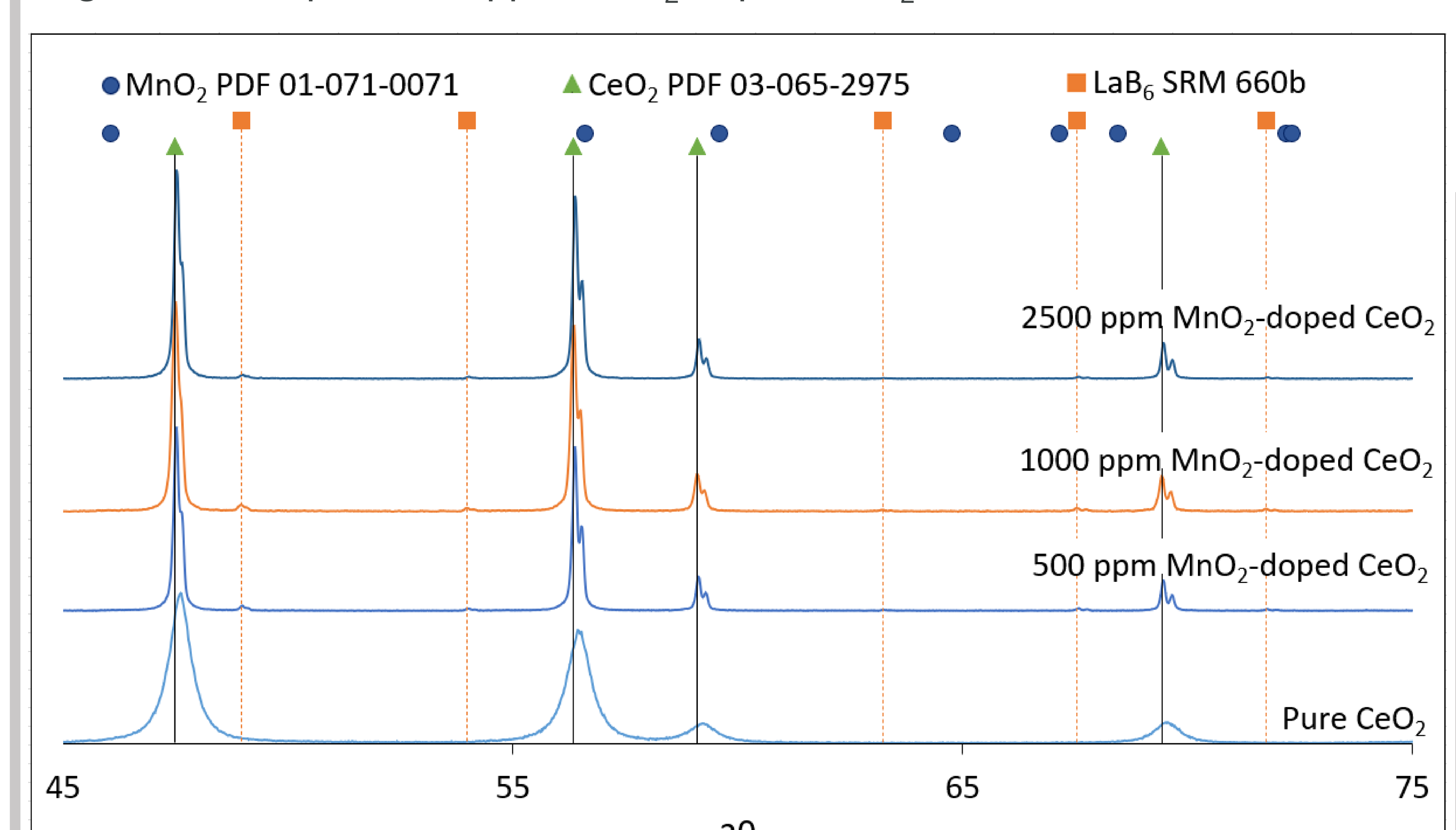

Fig.8. XRD of samples sintered at 1500 °C. The MnO_2-doped samples contain a LaB_6 standard. XRD data was adjusted 0.1° to account for instrumentation shifting identified by the LaB_6 standard. [6,7].

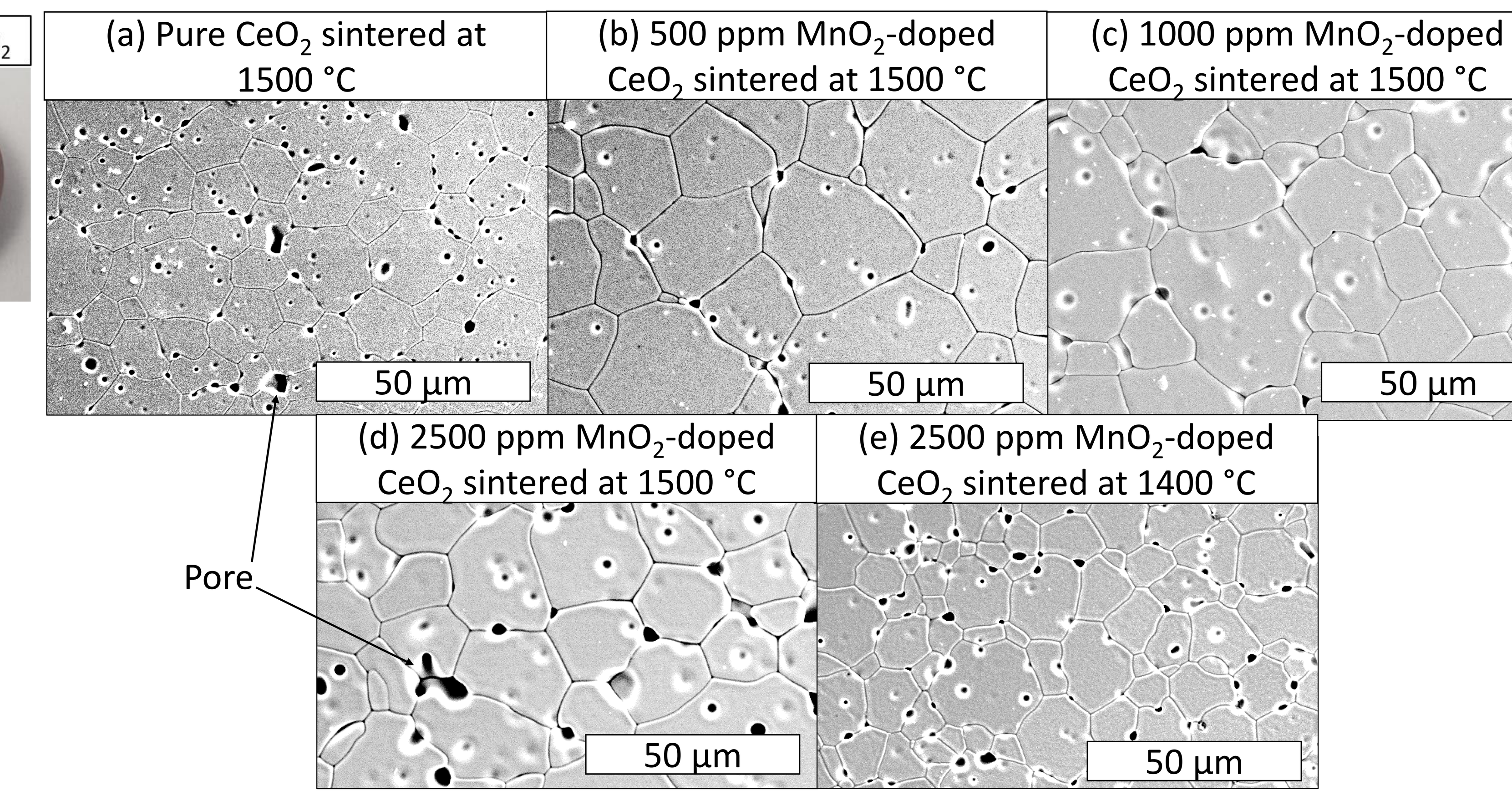

Fig.9. SEM micrographs show microstructure of pure and MnO_2-doped CeO_2 samples. The specimen were thermally etched 150°C below the sintering temperature for grain size analysis.

- The 500 and 1000 ppm samples had larger grain sizes, lower porosity and higher uniformity in their microstructures.
- EDS map of 1000 ppm MnO_2-doped CeO_2 did not detect MnO_2 precipitates.
- Surface defects were observed in the specimen doped with 2500 ppm MnO_2 in addition to having the lowest density.
- XRD analysis indicated CeO_2 phase was present, with no secondary phases. No notable peak shifting occurred, meaning Mn^+ incorporation into the CeO_2 lattice was likely substitutional instead of interstitial. A lack of MnO_2 peaks supports the attainment of a CeO_2 and MnO_2 solid solution.

IV. DISCUSSION

- Concentration of 1000 ppm MnO_2 sintered at 1500 °C increased the grain size of CeO_2, while maintaining good density (94±2 %TD).
- Due to reduced density and increased porosity, the 2500 ppm samples likely exceeded the solubility limit of MnO_2 in CeO_2
- The concentration of MnO_2 may be below the detection limit of EDS and XRD, so electron probe microanalysis will be explored as an alternative characterization method.
- Further refinement will be performed to determine the optimal MnO_2 concentration that produces enhanced grain growth. MnO_2 concentrations of 800 and 1200 ppm will be tested.

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