The Effect of Laser Ablation and Oxygen Atmospheric Conditions on the Formation of Cerium Oxide Particulates

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Abstract

We generate cerium oxide particulates using nanosecond pulsed or continuous wave (CW) lasers in different concentrations of gaseous oxygen.

Motivation

Early models of fallout chemistry relied on empirical observations from historical fallout, but improved models require a fundamental understanding of fireball chemistry.

Results

<table>
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<tr>
<th>Pulsed ablation</th>
<th>CW heating</th>
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<tr>
<td><strong>Low oxygen</strong> (1%)</td>
<td>&quot;defective&quot; CeO₂, CeO₂ &amp; Ce₂O₃</td>
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<tr>
<td><strong>High oxygen</strong> (&gt;20%)</td>
<td>CeO₂</td>
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Table 1: Summary of particulates observed

SEM of particulates found outside the crater (a) and inside the crater (b) were found to be defective CeO₂ and Ce₂O₃, respectively.

The defective particulates are unstable. When heated with <10 mA·W from the CW Helie Raman laser, the defective particulates changed in real-time to form the fluorite mode (465 cm⁻¹).

In the CW heating experiments in 1% O₂, some heated particulates in the vicinity of the laser impingement were found to have the A-type CeO₂ structure.

Conclusions

Assigning Raman active modes more accurately to the cerium CeO₂ and Ce₂O₃ phases, the CW heating experiments were repeated in O₂, and isotopic shifts were measured around the point of the CW laser impingement.

The shift is proportional to the change in reduced mass ν [cm⁻¹] = ν [cm⁻¹] with the 409 cm⁻¹ mode predicted at ~388 cm⁻¹ for the CeO₂ mode with the Ce₂O₃ mode shifted as expected.

Methods

Scanning electron microscopy (SEM) and Raman spectroscopy were used to investigate the composition and structure of the particulates formed.

Variations in bond length or the formation of oxygen vacancy defects directly impact the oscillation frequency and are detectable in Raman spectra.

This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344, is supported by LLNL Strategic Initiative LDRD project 20-SI-008.