Development of Cerium-Neodymium Oxide surrogates for Americium Oxides

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  – Am Oxides, AmO_{2-x/2} and Ce_{1-x}Nd_xO_{2-x/2} Surrogate

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Introduction: why consider surrogate for \( \text{AmO}_2-(x/2) \) using Ce and Nd?

- **Surrogate with a stable x value.**
  - Potential to study reduction behaviour of surrogate during future studies.
  - Theory dictates\(^1\) that the Nd to Ce ratio can be used to infer the Oxygen to Metal ratio.

- **Once fully investigated: CeNd Oxide solid solution production is potentially simpler than CeO\(_2-(x/2)\).**

- **Diversity in surrogates: more data to inform AmO\(_2-(x/2)\) possible behaviour.**
Am Oxides and AmO$_{2-\frac{x}{2}}$

- Relevant Oxide Forms

<table>
<thead>
<tr>
<th>Oxide</th>
<th>Symmetry</th>
</tr>
</thead>
<tbody>
<tr>
<td>AmO$_2$</td>
<td>FCC</td>
</tr>
<tr>
<td>A-Am$_2$O$_3$</td>
<td>Hexagonal</td>
</tr>
<tr>
<td>B-Am$_2$O$_3$</td>
<td>Monoclinic</td>
</tr>
<tr>
<td>C-Am$_2$O$_3$</td>
<td>Cubic</td>
</tr>
</tbody>
</table>

- Solid Solution: AmO$_{2-\frac{x}{2}}$
Ce$_{1-x}$Nd$_x$O$_{2-x/2}$ Surrogate

- Surrogates for End Phases

<table>
<thead>
<tr>
<th>Am Oxide</th>
<th>Surrogate Oxide</th>
<th>Symmetry</th>
</tr>
</thead>
<tbody>
<tr>
<td>AmO$_2$</td>
<td>CeO$_2$</td>
<td>FCC</td>
</tr>
<tr>
<td>A-Am$_2$O$_3$</td>
<td>A-Nd$_2$O$_3$</td>
<td>Hexagonal</td>
</tr>
<tr>
<td>C-Am$_2$O$_3$</td>
<td>C-Nd$_2$O$_3$</td>
<td>Cubic</td>
</tr>
</tbody>
</table>

- Ce$_{1-x}$Nd$_x$O$_{2-x/2}$ Solid Solution (SS)

<table>
<thead>
<tr>
<th>Structure</th>
<th>$x$ value</th>
</tr>
</thead>
<tbody>
<tr>
<td>F-type SS</td>
<td>$0 &lt; x \leq 0.39$</td>
</tr>
<tr>
<td>C-type SS</td>
<td>$0.39 &lt; x \leq 0.73$</td>
</tr>
<tr>
<td>C-type SS and A-type Nd$_2$O$_3$</td>
<td>$0.73 \leq x &lt; 1$</td>
</tr>
</tbody>
</table>
PART II: SURROGATE SYNTHESIS

Oxalate Precipitation

Thermal Decomposition
The Objectives

1. Can the proposed route create a solid solution?

2. Can $\text{Ce}_{1-x}\text{Nd}_x\text{O}_{2-(x/2)}$ with $0.5 < x < 0.7$ be made with repeatability?

3. How does reaction temperature and stirring rate affect oxalate particle morphology and size?

4. How do the thermal decomposition variables affect oxide properties?
Results: Oxalate Material
1. Is it an Oxalate solid solution?

- XRD and Raman Spectroscopy:
  - Rare Earth oxalate
  - No further information
1. Is the Oxide a Solid Solution?
1. Is the Oxide a Solid Solution?

- Thermogravimetric Analysis
  - Hypothesis:

4. Example of thermal decomposition of an early oxalate sample.
1. Is the Oxide a Solid Solution?

- Powder X-ray Diffraction
1. Is the Oxide a Solid Solution?

- Powder X-ray Diffraction
2. Can $\text{Ce}_{1-x}\text{Nd}_x\text{O}_{2-(x/2)}$ with $0.5 < x < 0.7$ be made with repeatability?

- Measure X-ray Fluorescence of the oxalate.

- Calculate Nd : (Ce+Nd) and compare.

- Semi-quantitative analysis
  - 3 segments of each sample.

<table>
<thead>
<tr>
<th>Oxalate Sample #</th>
<th>$x$ values</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.49; 0.59; 0.41</td>
</tr>
<tr>
<td>2</td>
<td>0.62; 0.62; 0.62</td>
</tr>
<tr>
<td>3</td>
<td>0.62; 0.51; 0.53</td>
</tr>
</tbody>
</table>
2. Can $\text{Ce}_{1-x}\text{Nd}_x\text{O}_{2-(x/2)}$ with $0.5 < x < 0.7$ be made with repeatability?

- Future:
  - collect more data to determine variability in $x$ for a given sample.
3. Oxalate Morphology and Size Variation

- Current experiments: effect of stirring rate at 25 °C.

- Future: investigate 60 °C reactions.

- SEM: shape and size estimate

- Laser Diffraction for relative size distribution
Results: Oxalate Morphology

Example: 25 °C, 250 RPM

Example: 25 °C, 400 RPM
References


Thank you for your interest, I would be happy to take any questions.