Studying the Effects of Solution Processing Conditions on Morphology of Studtite and UO$_3$

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Summary
Nuclear forensic signatures are useful in the determination of origin and processing conditions of illicitly trafficked nuclear materials. In this investigation, UO$_3$ was examined (as a product of heating studtite starting material ([(UO$_2$)(O$_2$)(H$_2$O)$_2$]·2H$_2$O)). Based on previous screening experiments, relationships and interactions between processing variables were studied for their effects on the characteristics of studtite and UO$_3$ powder.

Background and Aims

Screening Experiments

Initial screening experiments helped to establish the processing conditions that may affect the physical and chemical properties of UO$_3$ product. A potential relationship between solution concentration and powder morphology (shape and form) was found, leading to further investigation by a fractional factorial matrix.

Material Preparation

Studtite and UO$_3$ powder synthesis

Synthetic studtite was prepared by mixing aqueous uranyl nitrate (0.1 and 1 M) and hydrogen peroxide (H$_2$O$_2$ 5 and 30 w/w%) via forward (H$_2$O$_2$ added to U nitrate) or reverse addition (U nitrate added to H$_2$O$_2$). All mixing was performed robotically by a Metrohm Titrosampler, filtered under vacuum and dried for 48 hours. UO$_3$ was obtained directly by heating studtite powder to 535 °C in nitrogen (N$_2$) at 10 °C/min.

Factorial Matrix and Images

Design of Experiments

To test for relationships and/or interactions between effects of processing variables, a 2$^{30}$ fractional factorial experiment was used, requiring a total of 8 experimental runs.

<table>
<thead>
<tr>
<th>Concentration (M)</th>
<th>Concentration (w/w%)</th>
<th>Strike</th>
<th>Washing</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.1</td>
<td>5</td>
<td>Forward</td>
<td>W</td>
</tr>
<tr>
<td>0.1</td>
<td>30</td>
<td>Reverse</td>
<td>UnW</td>
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<tr>
<td>1</td>
<td>5</td>
<td>Forward</td>
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<tr>
<td>1</td>
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<tr>
<td>1</td>
<td>30</td>
<td>Reverse</td>
<td>UnW</td>
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</tbody>
</table>

Particle Morphology Discussion

Particles generally appeared faceted and angular when produced by reverse-strike addition, particularly at low U nitrate concentrations. Reverse-strike precipitation will occur with slower growth rate than forward, due to immediate excess dilution in H$_2$O$_2$. Washing appears to make little difference to morphology. Forward-strike samples appeared rounded and clumped when either i) nitrate-peroxide concentration levels were equal and ii) washing had occurred after precipitation. Further work will be considered to discern the main effect contributing to these observations.

UO$_3$ Morphology

Heating studtite to 535 °C in N$_2$ at a rate of 10 °C/min appeared to have no effect on the particle morphology. This result may prove relevant to nuclear forensic applications, where

Conclusion

Initial screening experiments demonstrated that it is possible to obtain different morphologies of studtite, and therefore UO$_3$, from different solution processing conditions. By running a 2$^{30}$ fractional factorial matrix, it was found that the morphology of studtite and its heat products are affected by a combination of concentration, concentration ratio and strike order of reagent during the studtite precipitation stages.

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