Crystal Growth Rates and Morphology Development on Plutonium(III)-oxalate by Changing Precipitation Conditions

Pacific Northwest NATIONAL LABORATORY

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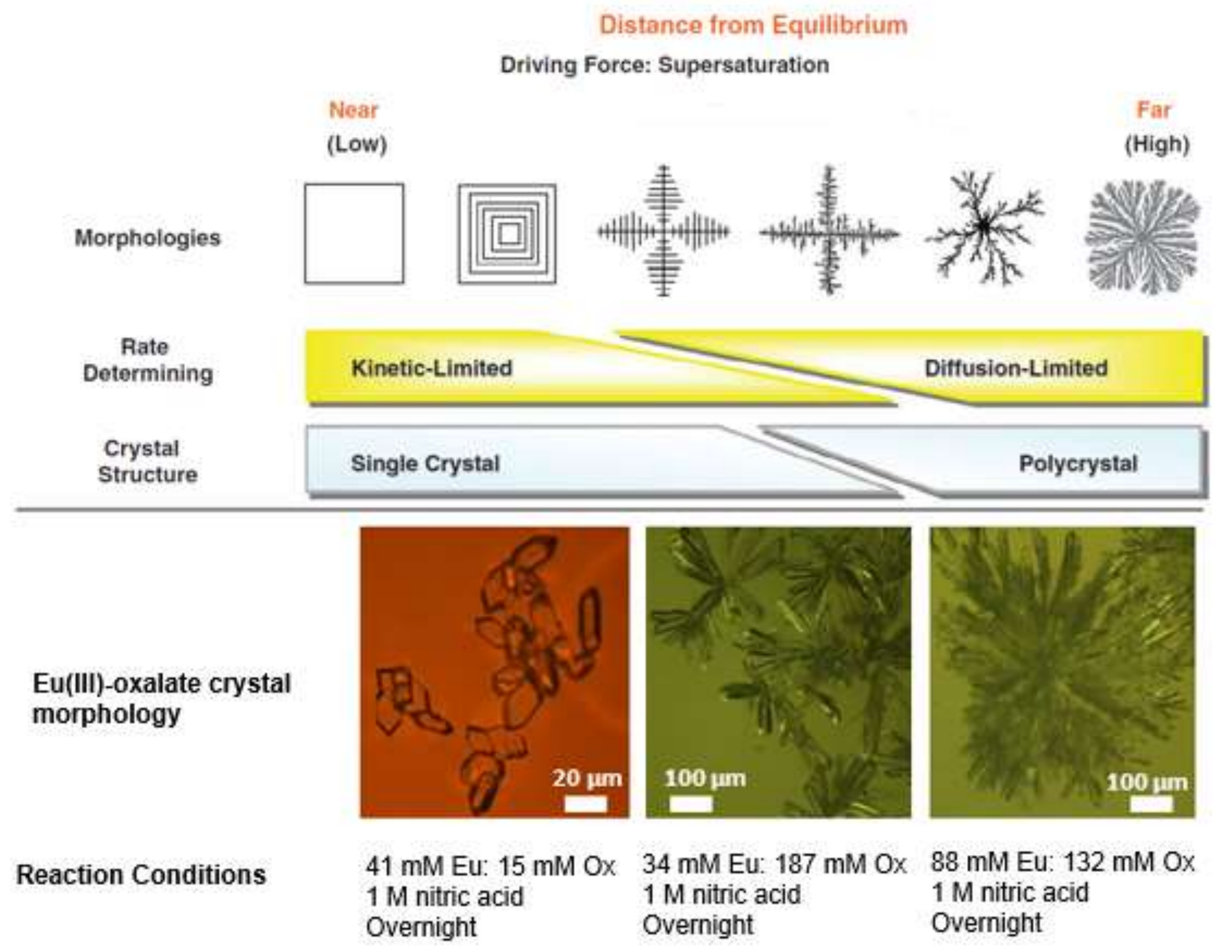
Science Problem

Research Challenge

Understanding how precipitation conditions correlate with the formation of crystal growth and final morphology on Pu(III)-oxalate crystals

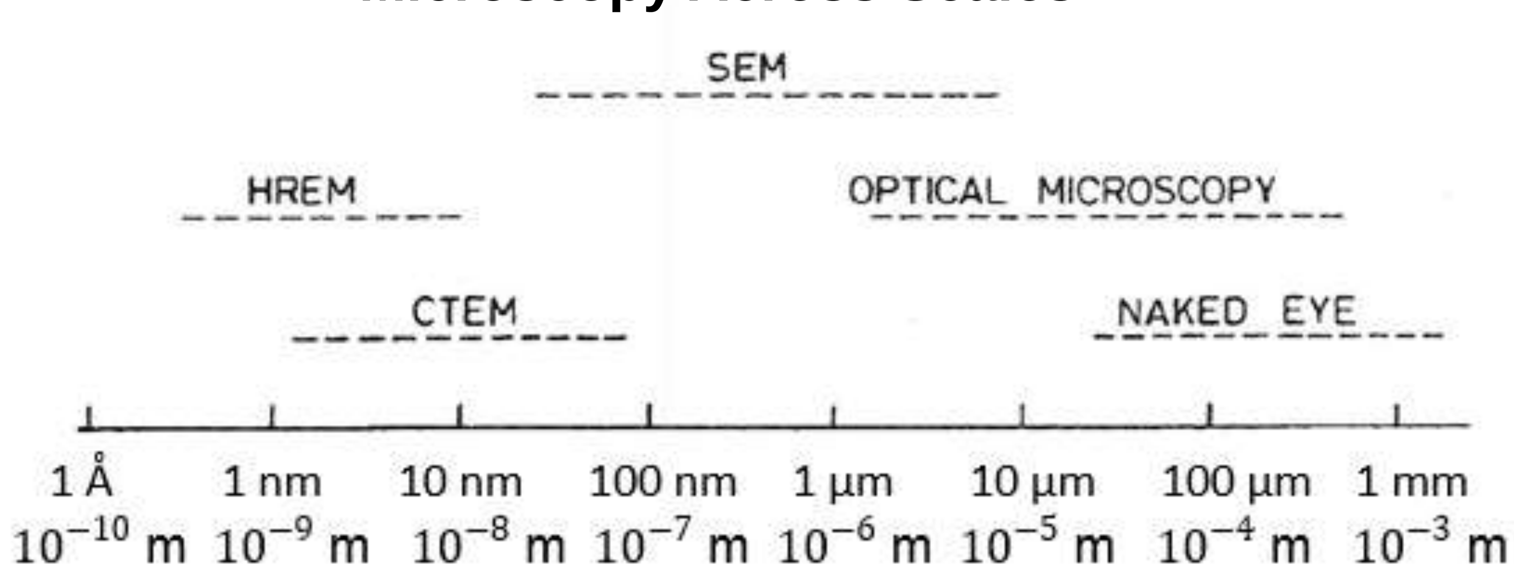


Schematic model for morphological variation obtained from literature[1] and experimental europium(III)-oxalate crystals were added.

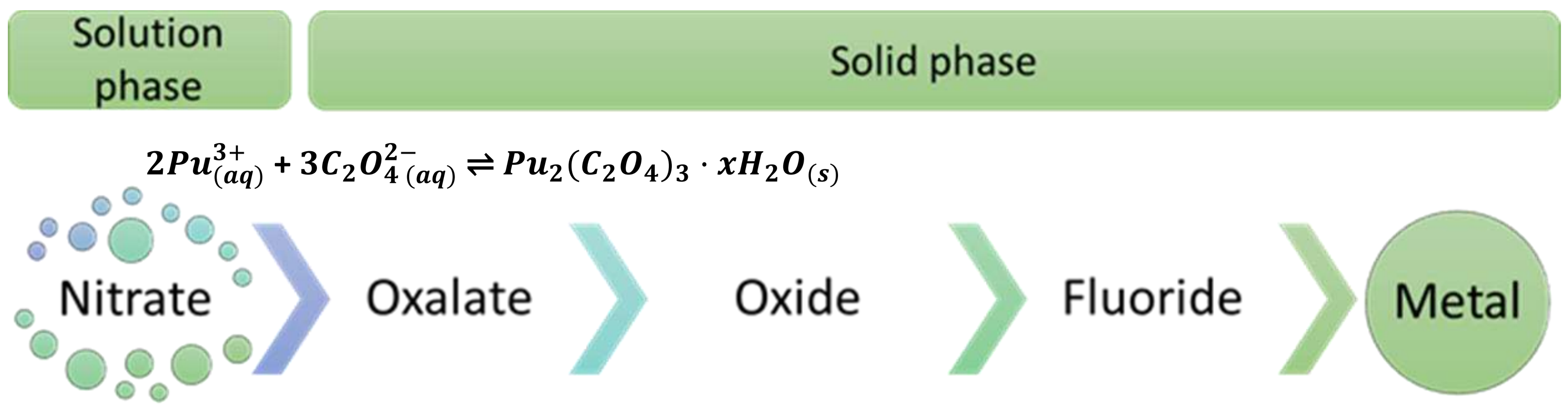
Technical Challenge

Crystal growth begins as ions (~10^{-10} m) and ends as ~10 µm (10^{-5} m) particles

Microscopy Across Scales



Background



Precipitation of plutonium from an acidic solution by the addition of oxalic acid has become a major step in the large-scale separation and purification processes of plutonium. A kinetic model for crystal growth by diffusion-limited mechanisms was rigorously developed by Lifshitz, Slyozov, and Wagner, also known as the LSW model. The LSW model predicts that the average particle radius should evolve as a function of time according to the following equation:

$$R^3 - R_0^3 = kt$$

where R is the average particle radius, R_0 is the average initial particle radius, t is the time, and k is the rate constant.

Scientific Approach

Experimental Overview

- Measure the initial rate of crystal growth by only increasing nitric acid concentration
- Measure the initial rate of crystal growth by only increasing oxalic acid concentration
- Measure specific surface area and density on the final morphology of Pu(III)-oxalate crystals

Instrumentation for Experiments

- Optical microscopy
- Scanning electron microscopy (SEM)
- Transmission electron microscopy (TEM)
- Brunauer-Emmett-Teller (BET)
- Gas pycnometry

Preliminary Results

Microscopy Imaging → Image Analysis → Crystal Growth Rates

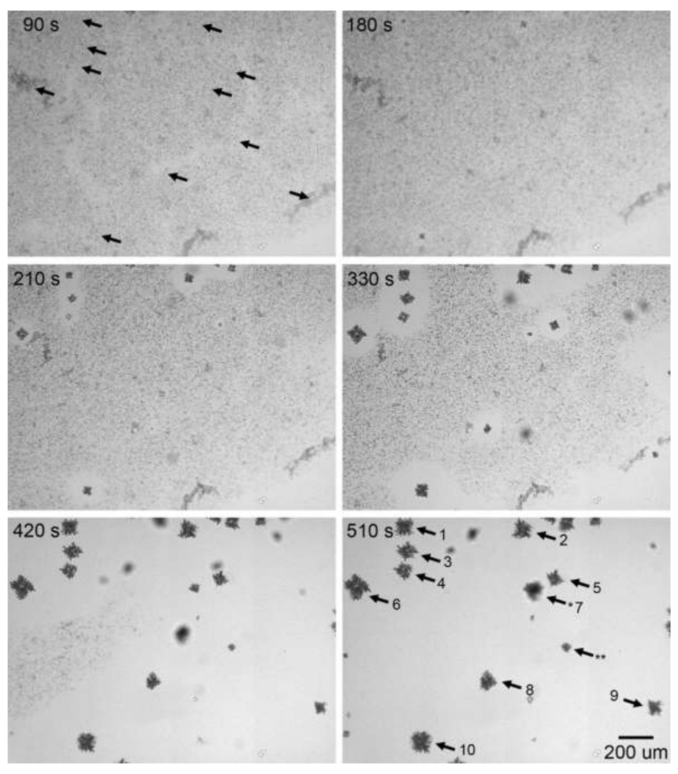


Figure 1. Optical microscopy images of europium(III)-oxalate crystals growing in solution over 9 minutes.

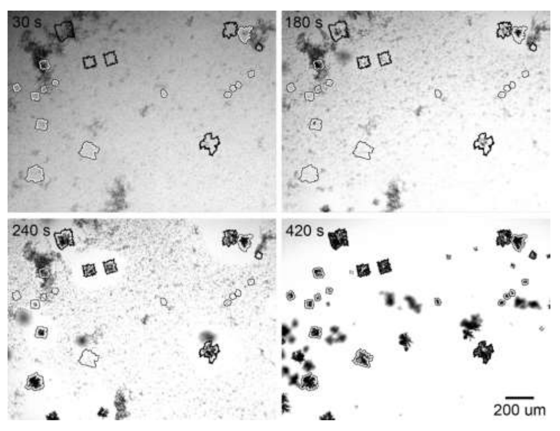


Figure 2. The outer boundary's outline of the particles using Image J software for particle analysis.

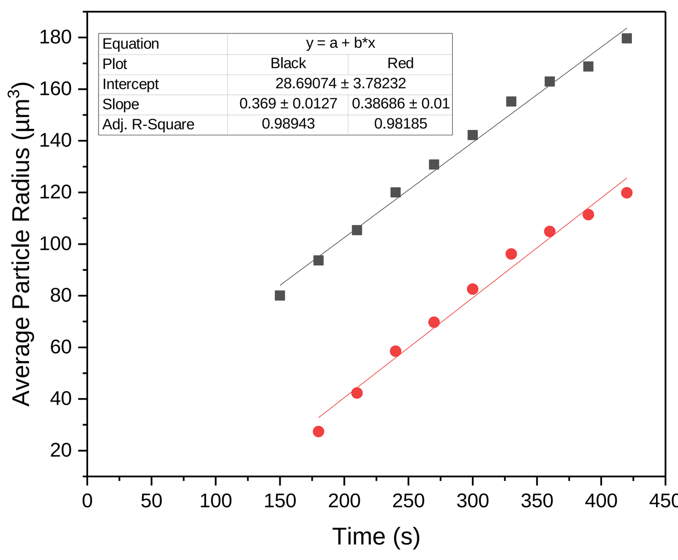


Figure 3. A plot that depicts the average particle radius as a function of time for europium(III)-oxalate. The crystal growth rate is approximately 0.4 µm³/s.

Future Work

- Use the developed microscopy technique on plutonium by varying nitric and oxalic acid to measure crystal growth rates
- Measure specific surface area and density on plutonium(III)-oxalate crystals
- Provide fundamental understanding on correlating morphology to precipitation conditions

References

1. Imai, H., Oaki, Y. Bioinspired Hierarchical Crystals. MRS Bulletin, 2010, Vol.35, pp 138-144

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