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 ($[(UO_2)(O_2)(H_2O)_2]\cdot 2H_2O$)). 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.

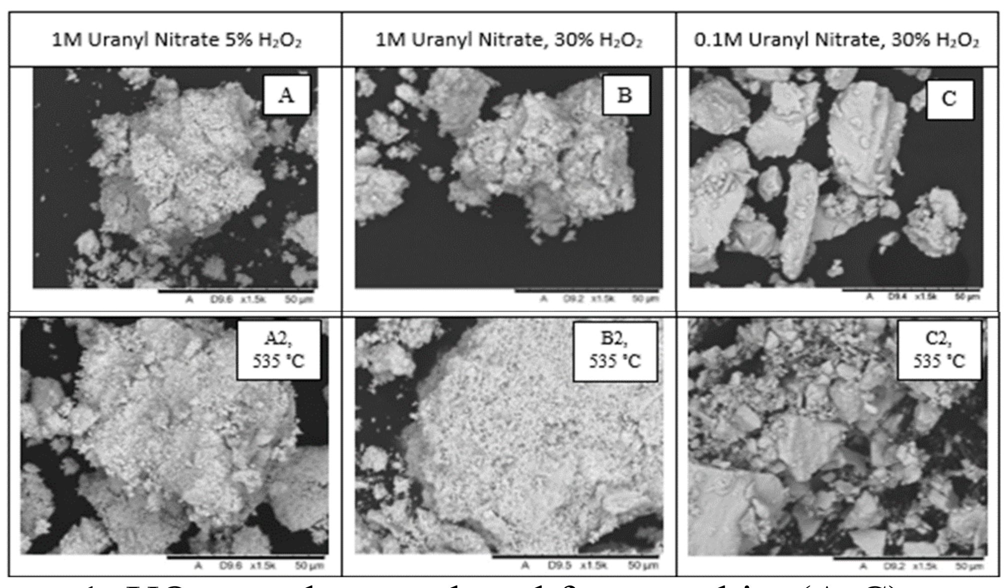
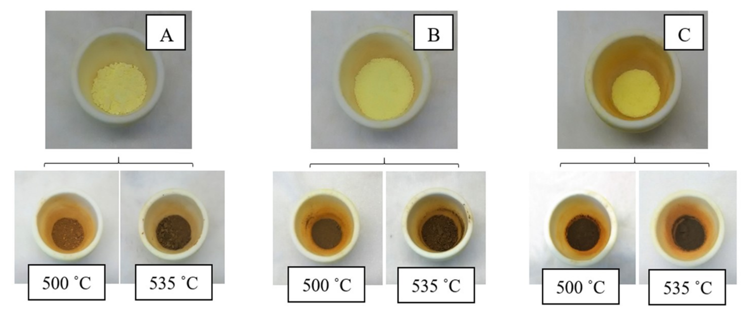


Figure 1: UO_3 powders produced from studtite (A-C) samples.

Aims

To further establish the relationships that exist between solution processing parameters (e.g. concentration, strike order, etc) and finished UO_3 powder morphology.



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_2O_2, 5 and 30 w/w%) via forward (H_2O_2 added to U nitrate) or reverse addition (U nitrate added to H_2O_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.

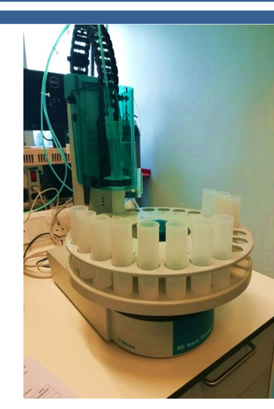


Figure 2: Metrohm Titrosampler

Factorial Matrix and Images

Design of Experiments

To test for relationships and/or interactions between effects of processing variables, a 2^{4-1} fractional factorial experiment was used, requiring a total of 8 experimental runs.

Conc. U Nitrate (M)	Conc. H_2O_2 (w/w%)	Strike	Washing
0.1	5	Forward	W
0.1	5	Rerverse	UnW
0.1	30	Forward	UnW
0.1	30	Reverse	W
1	5	Forward	UnW
1	5	Reverse	W
1	30	Forward	W
1	30	Reverse	UnW

Scanning Electron Microscopy (SEM)

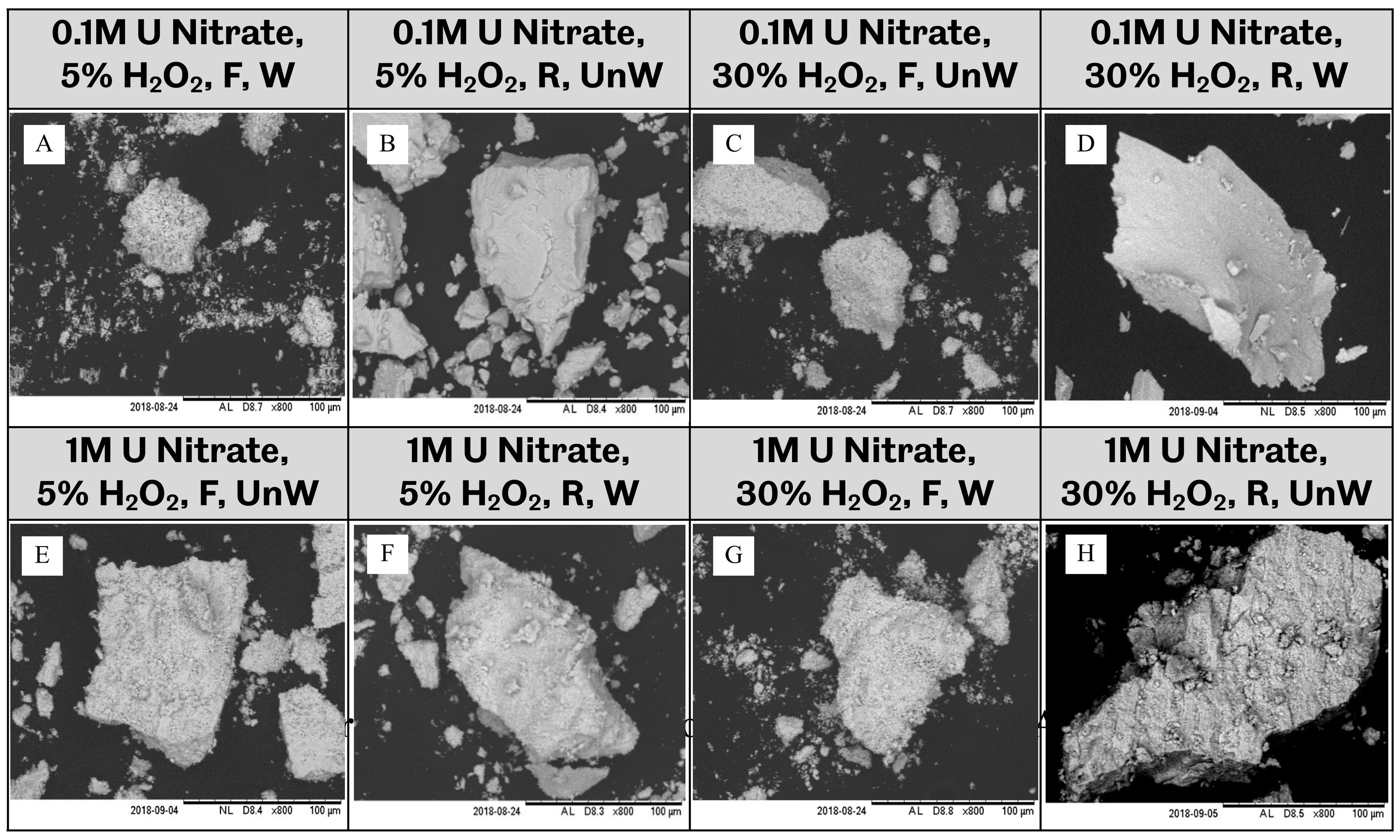


Figure 3: SEM images of ground studtite precipitates (R=Reverse, UnW=Unwashed)

Particle Morphology Discussion

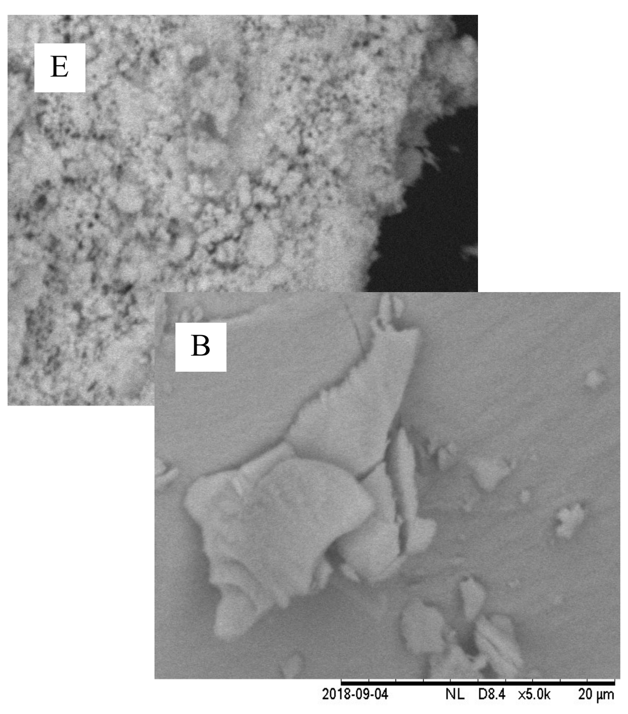


Figure 4: Samples E and B at 20 µm scale

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_2O_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

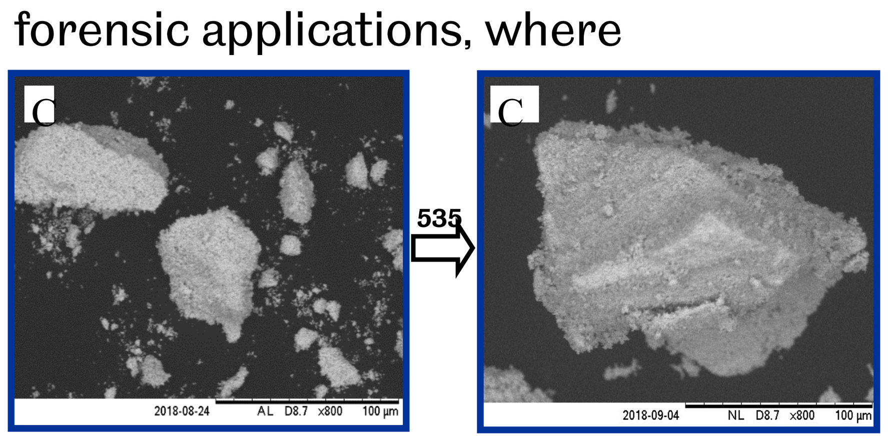


Figure 6: 0.1M 30% R, W at RT (C) and 535˚C (C1)

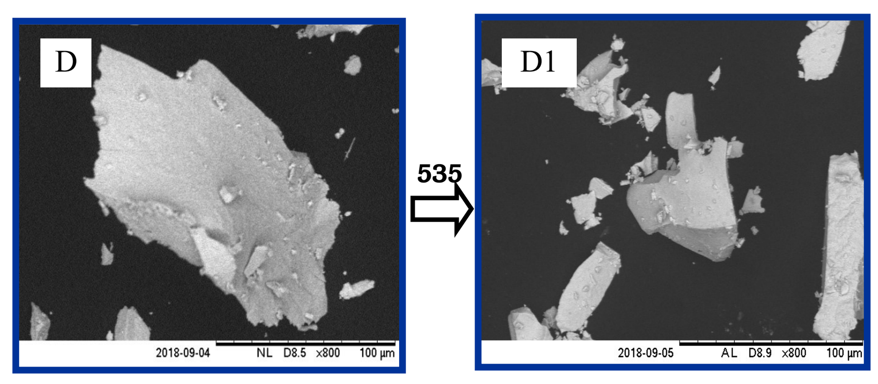


Figure 5: 0.1M 30% F UnW at RT (D) and 535˚C (D1)

a sample of UO_3 (or other thermal product of studtite) may be traced to its starting material using the UO_3 morphology, compared to a set of known standards.

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^{4-1} 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.

Acknowledgements

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