Investigating Forensic Signatures of Studtite and its Products of Thermal Decomposition

Nathan Thompson, Prof. Neil C. Hyatt, Dr. Matthew Gilbert
Department of Materials Science & Engineering, University of Sheffield

Summary

Nuclear forensic signatures are useful in the determination of origin and processing conditions of illicitly trafficked nuclear materials. Characteristic signatures of materials may be obtained from spectroscopic analysis. In this investigation, various decomposition products of studtite ($[(UO_2)(O_2)(H_2O)_2]\cdot 2H_2O$) were examined, leading to a fractional screening experiment to test for effects of processing conditions.

Initial Analysis

1. Thermal Analysis (TGA-DSC-MS)

Analysis was carried out over a temperature range 32-1150 ˚C at a rate of 10 ˚C/min. The resulting curve showed 5 distinct phases of decomposition products over the heating process. Oxygen and water release were revealed by mass spectrometry (MS).

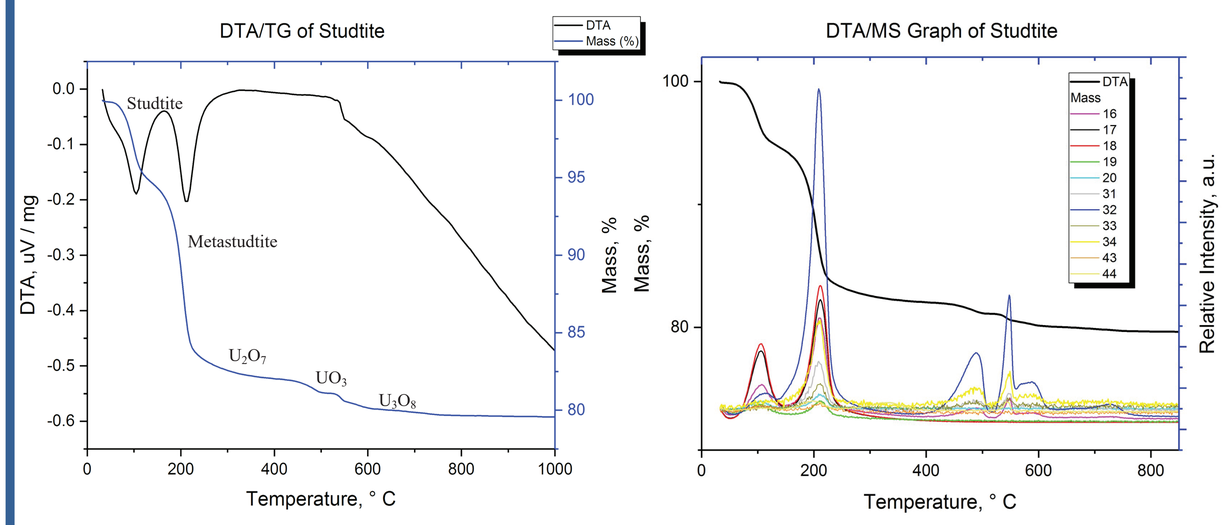

Figure 1: Heat treatments of studtite, at temperatures from room temperature to 1050˚C.

2. Heating Studtite

Studtite was heated in air to 1050 ˚C in batches at 150, 250, 520, 630 and 1050 ˚C for investigation of phase transition points found in TGA.

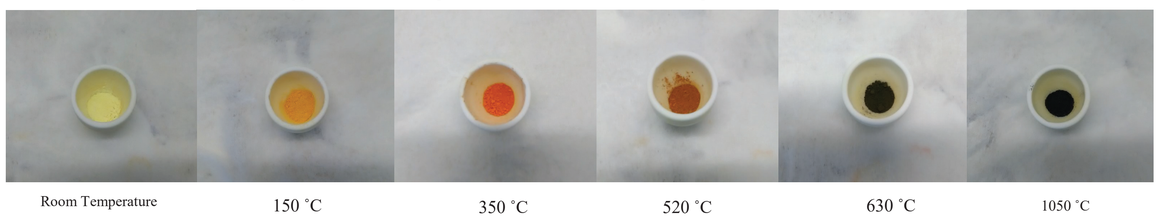

Figure 2: Heat treatments of studtite, at temperatures from room temperature to 1050˚C.

3. Scanning Electron Microscopy (SEM)

Decomposition products up to 630 ˚C were imaged by SEM. Porosity increased with gas release upon heating; morphology was retained.

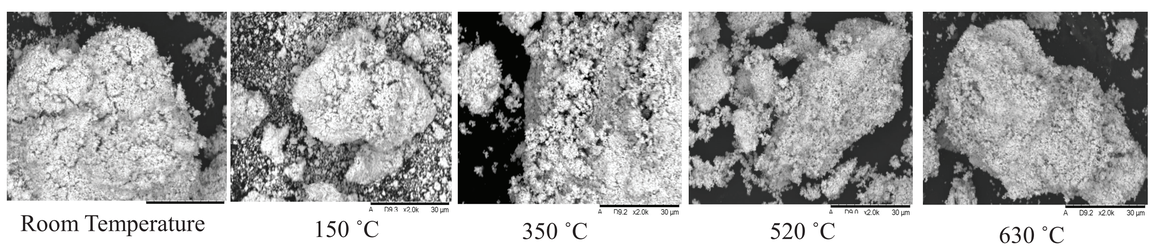

Figure 3: SEM images of studtite decomposition products, over heating period to 630 ˚C.

Screening Experiment

Design of Experiments

To test for relationships or interactions between effects of processing variables, a fractional factorial experiment was designed.

	500 ˚C, 5% H_2O_2	500 ˚C, 30% H_2O_2	535 ˚C, 5% H_2O_2	535 ˚C, 30% H_2O_2
0.1 M Uranyl Nitrate		+/- (C1)		+/- (C2)
1 M Uranyl Nitrate	+/- (A1)	+/- (B1)	+/- (A2)	+/- (B2)

Table 1: Different conditions used for producing UO_3 from studtite.

1. Colours of UO_3

All powders were brown upon heating studtite. Powders were darker in colour at the higher (535 ˚C) calcination temperature.

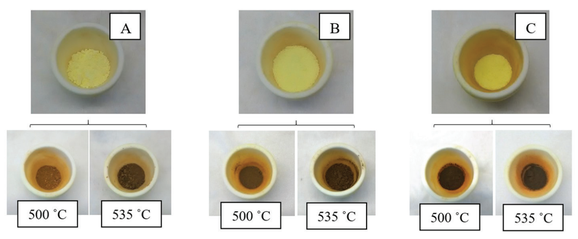

Figure 4: UO_3 powders produced from studtite (A-C) samples.

2. X-Ray Diffraction (XRD) Analysis

Certain process conditions produced either hexagonal or orthorhombic crystal structures of UO_3.

	500 ˚C, 5% H_2O_2	500 ˚C, 30% H_2O_2	535 ˚C, 5% H_2O_2	535 ˚C, 30% H_2O_2
0.1 M Uranyl Nitrate	-	Hexagonal *P3m1*	-	Hexagonal *P3m1*
1 M Uranyl Nitrate	Orth. *C2mm*	Orth. *C2mm*	Orth. *C2mm*	Orth. *C2mm*

Table 2: Crystal structures from various production conditions

3. Scanning Electron Microscopy (SEM)

The shape of the powder was retained during heating. From a lower concentration, the UO_3 product appeared more faceted and ordered.

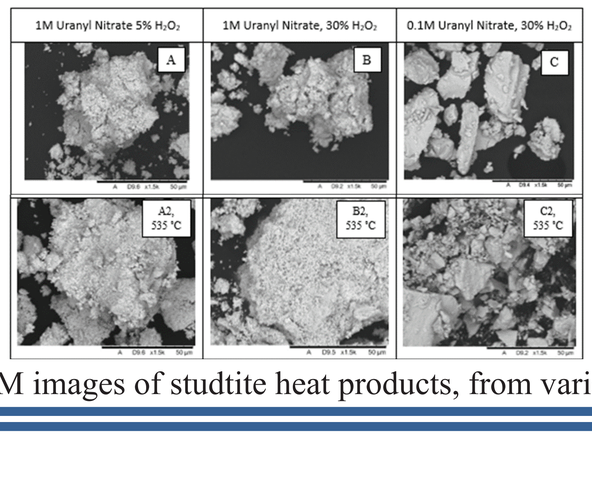

Figure 5: SEM images of studtite heat products, from varied conditions.

Conclusion

A process involving initial analysis (e.g. TGA) on individual decomposition products has been useful in understanding the chemical changes in heating studtite, and accompanying forensic signatures. A screening experiment has demonstrated that it is possible to obtain different crystal structures of UO_3 from different processing conditions. The concentration of precursor has an effect on the final UO_3 morphology.

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@ISL_Sheffield
Department of Materials Science & Engineering, University of Sheffield
Robert Hadfield Building, Mappin Street, Sheffield, S1 3JD

nbathompson1@sheffield.ac.uk
https://uk.linkedin.com/in/nbathompson