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Behaviour of Uranyl Phosphate Containing Solid Waste During Thermal Treatment for the Purpose of Sentencing and Immobilisation: Preliminary Results

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Thermal decomposition of the uranyl phosphate mineral phase meta-ankoleite ($KUO_2PO_4 \cdot 3H_2O$) has been considered in relation to high temperature thermal sintering for the immobilisation of a uranyl phosphate containing waste. Meta-ankoleite thermal decomposition was studied across the temperature range 25 – 1200℃ under an inert N_2 atmosphere at 1 atm. It is shown that the meta-ankoleite mineral phase undergoes a double de-hydration event at 56.90 and 125.85℃. Subsequently, synthetically produced pure meta-ankoleite remains stable until at least 1150℃ exhibiting no apparent phase changes. In contrast, when present in a mixed waste the meta-ankoleite phase is not identifiable after thermal treatment indicating incorporation within the bulk waste either as an amorphous phase and/or as uranium oxide. Visual inspection of the waste post thermal treatment showed evidence of self-sintering owing to the presence of glass former materials, namely, silica (SiO_2) and antimony(V) oxide (Sb_2O_5). Therefore, incorporation of the uranium phase into the waste as part of waste sentencing and immobilisation via high temperature sintering for the purpose of long-term disposal is deemed feasible.

Keywords: Uranium, Uranyl phosphate, Meta-ankoleite, Thermogravimetry, Radioactive waste, SOHIO process

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1. Introduction

The formation of sparingly soluble uranyl phosphates offer a route for the removal of uranium from complex aqueous waste streams [1-7]. Our recently reported method involves the removal of uranium from an industrial waste effluent via the formation of synthetic meta-ankoleite ($KUO_2PO_4·3H_2O$) [8, 9]. Final immobilisation of such a waste is of the utmost importance to ensure complete compliance with national and international concessions for radioactive waste handling and disposal.

The purpose of any immobilisation process is to stabilise the waste and prevent the release of contaminants into the environment during long-term storage and disposal. Knowledge of the chemical composition and mineral phases present in the final wasteform is required in order to forecast leaching, corrosion and general degradation phenomena of the wasteform over geological time scales. It is therefore important to characterise the mineral phases present before, during and after any immobilisation process, and understand what, if any, physicochemical changes take place. Immobilisation techniques include cementation, polymer-fixation, bituminisation, vitrification and ceramication; the latter of which exposes the wastes to high temperature processing [10-14]. Ceramication is the chosen immobilisation method for the waste in question [15-17].

Thermal immobilisation methods expose the waste to elevated temperatures, e.g. 1100℃ in the present case. Phase changes or chemical decomposition due to exposure to these elevated temperatures may result in the formation of less stable or more soluble mineral phases which would compromise the integrity of the final wasteform and leave it unfit for long term storage or disposal. Understanding the thermal behaviour of $KUO_2PO_4·3H_2O$ is necessary to ensure compatibility of the resulting uranium mineral phases with the target wasteform, and to confirm regulatory compliance.

The thermal stability of selected uranyl phosphates have been previously reported by Schaekers [18-20] ($HUO_2PO_4·4H_2O$, $NH_4UO_2PO_4·3H_2O$, $(UO_2)_3(PO_4)_2·4H_2O$, $(UO_2)_2P_2O_7$ and $U(HPO_4)_2$); Kamo et al.[21] ($UO_2(H_2PO_4)_2·xH_2O$ and $UO_2(PO_3)_2$); and by Barten et al.[22] ($(UO_2)_3(PO_4)_2$, $(UO_2)_2P_2O_7$ and $UO_2(PO_3)_2$); the latter of which reported oxygen partial pressures. Uranyl phosphates decompose at elevated temperatures depending on oxygen pressure and cation present [22]. Examples being the stability of $(UO_2)_3(PO_4)_2·4H_2O$ above 1000℃ in air, yet decomposing to a mix of U_3O_8 and $3U_2O_3P_2O_7$ in Ar and N_2 below 1000℃ [20]; or the decomposition of $NH_4UO_2PO_4·3H_2O$ starting as early as 250℃ owing to a loss of ammonia [19]. As yet there is no apparent reporting of $KUO_2PO_4·3H_2O$ thermal stability in the literature. As such, we report the stability of synthetic meta-ankoleite in relation to thermal immobilisation techniques for the formation of uranium containing wasteforms for final radioactive waste disposal.

2. Experimental

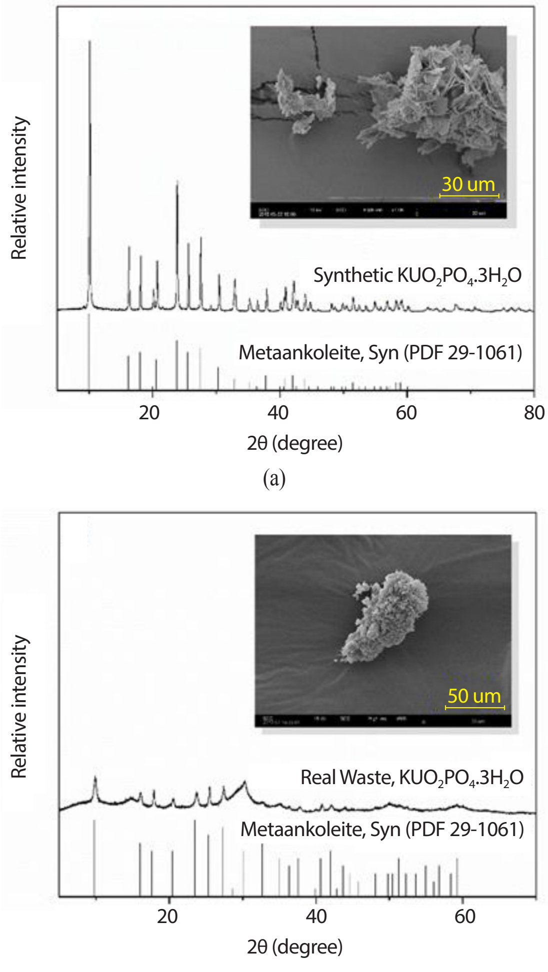
All materials were used as received with no further purification: Uranyl nitrate hexahydrate (UNH, $UO_2(NO_3)_2·6H_2O$) was obtained from Alfa Aesar; potassium phosphate (KH_2PO_4, 99.5%), potassium nitrate (KNO_3, 99%), and phosphoric acid (H_3PO_4, 85%) were obtained from SHOWA; nitric acid (HNO_3, 65%) from MERCK; and iso-propyl alcohol (IPA, 99.5%) from oriental chemical industries. Ultra-pure water (18.2 MΩ) prepared by a Milli-QPLUS (Millipore Co.) was used throughout.

The formation of synthetic meta-ankoleite was adapted for scale up from our previous publication (Eq. 1) [23]. Synthesis was as follows: To a 600 mL glass beaker 12.505 g of UNH was added followed by 250 mL KNO_3 (2 M). Stirring was provided by a magnetic stirrer bar at 400 rpm. A total of 250 mL of H_3PO_4 (1 M) was slowly added to the UNH/KNO_3 solution before being left to stir for a further 24 hours. The formed particles were then left to settle for an additional 24 hours undisturbed. The supernatant was discarded and replaced with 500 mL KNO_3 (2 M, pH 2.5–3) and left for approximately 2 weeks with periodic stirring. The

Table 1. XRF chemical compositional analysis of the real waste

Element	Concentration	
LE[1]	40.0 (±0.69)	wt%
Sb	15.5 (±0.18)	
U	14.5 (±0.17)	
Ca	13.5 (±0.16)	
K	7.7 (±0.09)	
Si	2.7 (±0.08)	
Ti	2.0 (±0.29)	
P	1.2 (±0.04)	
Fe	1.1 (±0.04)	
Cu	6595 (±168)	ppm
S	3622 (±135)	
Cl	2875 (±79)	
Bi	1559 (±110)	
Zn	1279 (±62)	
Rb	1090 (±122)	
As	1010 (±40)	

[1] Light elements. Elements lighter than Mg.



(a)

(b)

Fig. 1. X-ray diffractogram of (a) synthetic meta-ankoleite; (b) Real waste. Insert: corresponding scanning electron microscopy images.

formed fluorescent yellow precipitates were then collected by vacuum filtration, washed with ultra-pure water (3×50 mL) and once with IPA (1×50 mL) before being left to air dry. The formation of crystalline synthetic meta-ankoleite was confirmed by SEM, XRD (Fig. 1), elemental analysis and TGA: UO_2^{2+} 58.79% (58.95% theor.), PO_4^{3-} 20.42% (20.74% theor.), K^+ 6.17% (8.52% theor.) and H_2O 12.11% (11.79% theor.). Samples of the real waste, containing uranyl phosphate, were formed as part of laboratory and pilot plant trials [9, 24]. The chemical composition of the real waste is reported in Table 1.

$$K^+ + UO_2^{2+} + PO_4^{3-} + 3H_2O \rightarrow KUO_2PO_4\cdot 3H_2O$$
$$\text{Log } K_{sp} = 24.30 \tag{1}$$

Thermogravimetric analysis (TGA) was performed using a SDT Q600 (TA Instruments Korea) TGA-DSC instrument. Approximately 10 mg of air-dried sample was placed into a 70 μL alumina crucible. Measurements were performed under a dry N_2 atmosphere at a flow rate of 100 mL·min^{-1}. Sample mass loss was recorded from room temperature to pre-determined isothermal plateaus upto 1200℃ at a heating rate of 10℃·min^{-1}.

Mineral phase identification was performed using a Bruker D2 Phaser. Data was collected at room temperature over the range of 2θ = 5–80° with a step size of 0.01 and a counting time of 0.2 seconds per step. Collected X-ray diffraction patterns of the mineral phases were matched to International Centre for Diffraction Data database (ICDD, PDF-2 2002, Sets 1-52 plus 65 plus 70-89) using DIFFRAC. EVA V3 software.

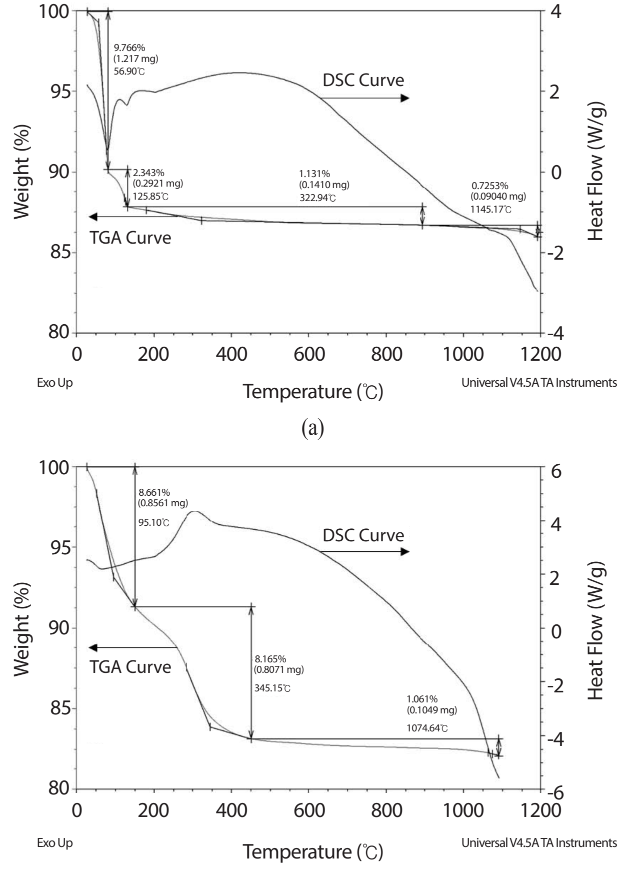


Fig. 2. Thermogravimetric data for (a) Synthetic $KUO_2PO_4 \cdot 3H_2O$, (b) Real uranyl phosphate containing waste.

3. Results & Discussion

3.1 Uranyl phosphate formation

Powder XRD phase analysis of the synthetic uranyl phosphate powders confirmed the formation of the mineral phase meta-ankoleite ($KUO_2PO_4 \cdot 3H_2O$) (Fig. 1(a)). Sharp intense peaks indicated the presence of well-defined crystal structures synonymous with uranyl phosphates [4, 23]. The crystalline nature of the sample was confirmed by scanning electron microscopy which revealed platelet-type crystals as similarly reported in our previous study (Fig. 1(a)) [23]. However, owing to the synthetic route employed, which resulted in shorter formation times, the long range order was substantially less than previously reported. Elemental analysis revealed a 1:1 U:P ratio along with TG analysis confirming the presence of three H_2O groups. However, the measured potassium concentration was less than theoretical indicating a less than perfect formation yield of 72.43% ($K_{0.72}H_{0.28}UO_2PO_4 \cdot 3H_2O$).

The precipitates obtained during the treatment of the real waste resembled a fine powder that was distinctively amorphous in appearance. Powder XRD phase analysis of the real waste confirmed the formation of the crystalline uranyl phosphate mineral phase meta-ankoleite (Fig. 1(b)); although the lower relative intensity and broader peaks indicates a more amorphous character and/or a decrease in grain size compared to the meta-ankoleite formed under idealistic synthetic conditions. The co-contaminant antimony was found to be present in the +5 oxidation state as antimony(V) oxide (Sb_2O_5), but again peak broadening indicates amorphous character and small grain sizes. The other co-contaminants (Table 1), in particular silicon, were not identifiable in the XRD as crystalline phases either due to being present as amorphous phases (i.e. Silica, SiO_2) or below the detection limit. The amorphous appearance of the precipitates was confirmed by SEM analysis (Fig. 1(b)). This is likely explained due to the rapid and turbulent formation conditions in which the precipitates are formed and also the presence of un-dissolve amorphous silica particles from previous process steps; as seen previously [9].

3.2 Thermal treatment

Thermogravimetic analysis of synthetic uranyl phosphate revealed two distinct de-hydration events both occurring below 150℃ (Fig. 2(a)). The first occurs between 28–81℃ (onset: 56.90℃) and with a mass loss of 9.766% (1.217 mg) equivalent to loosing 2.5 H_2O molecules (theoretical weight loss: 9.825%) (Eq. 2). The second de-hydration event occurs between 81–131℃ (onset: 125.85℃) with a mass loss of 2.343% (0.2921 mg) which is assigned to losing the remaining 0.5 H_2O (theoretical weight loss: 2.179%) (Eq. 3). Sharp well-defined endothermic heat flow peaks accompany both de-hydration events. The de-hydration of meta-ankoleite is seen to follow the same steps as both acid uranyl phosphate (AUP, $HUO_2PO_4 \cdot 4H_2O$) and uranyl ammonium phosphate (UAP, $NH_4UO_4PO_4 \cdot 3H_2O$) as reported by Schaekers [19].

$$\text{Step I} \quad KUO_2PO_4 \cdot 3H_2O \rightarrow KUO_2PO_4 \cdot 0.5H_2O + 2.5H_2O \quad (2)$$

$$\text{Step II} \quad KUO_2PO_4 \cdot 0.5H_2O \rightarrow KUO_2PO_4 + 0.5H_2O \quad (3)$$

A third mass loss of 1.131% (0.141 mg) was observed with an onset of 322.94℃ (Fig. 2(a)). No further decomposition route for dehydrated meta-ankoleite (KUO_2PO_4) would logically result in only a 1% mass loss. In contrast, Schaekers previously reported a further two decomposition steps for AUP beyond the initial double-dehydration event (Eqs. 4, 5). The latter decomposition steps result in the release of water and oxygen molecules as noted by Eq. 6 and 7, respectively. The decomposition of HUO_2PO_4 (Eq. 6) is reported to occur over the temperature range of 192–610℃, while decomposition of the resulting $(UO_2)_2P_2O_7$ compound into $U_2O_2P_2O_7$ and oxygen (Eq. 7) is reported over the temperature range of 800–931℃ [19]. Both decomposition events occur within the temperature range of the third mass loss seen in the TG result of $KUO_2PO_4 \cdot 3H_2O$ (Fig. 2(a)).

$$\text{Step I} \quad HUO_2PO_4 \cdot 4H_2O \rightarrow HUO_2PO_4 \cdot H_2O + 3H_2O \quad (4)$$

$$\text{Step II} \quad HUO_2PO_4 \cdot H_2O \rightarrow HUO_2PO_4 + H_2O \quad (5)$$

$$\text{Step III} \quad 2HUO_2PO_4 \rightarrow (UO_2)_2P_2O_7 + H_2O \quad (6)$$

$$\text{Step IV} \quad 2(UO_2)_2P_2O_7 \rightarrow 2U_2O_3P_2O_7 + O_2 \quad (7)$$

Considering Eqs. 6 and 7, the decomposition of a single HUO_2PO_4 would lead to the formation of ½H_2O and ¼O_2 molecules (Eq. 8). This would represent a theoretical mass loss of 4.64%. Based on the elemental analysis results, the meta-ankoleite yield was approximately 72% with the remaining yield likely comprising of AUP, based on experimental conditions used, resulting in a mixed solid with an approximate empirical formula of $K_{0.72}H_{0.28}UO_2PO_4 \cdot 3H_2O$. Assuming a K:H ratio of 0.72:0.28 and the subsequent decomposition of AUP as described by Schaekers, then a theoretical mass loss of 1.299% (4.6% × 0.28) over the temperature range of 192–931℃ can be predicted. It is therefore concluded that the third mass loss of 1.131% (0.141 mg) observed in Fig. 2(a) is in fact associated with the decomposition of AUP present due to incomplete formation of meta-ankoleite and not as a result of meta-ankoleite itself thermally decomposing.

$$HUO_2PO_4 \rightarrow \tfrac{1}{2}U_2O_3P_2O_7 + \tfrac{1}{2}H_2O + \tfrac{1}{4}O_2 \quad (8)$$

A fourth and final mass loss of 0.7253% (0.0904 mg) is observed in the TG of synthetic meta-ankoleite with an onset temperature of 1145.17℃. Under inert atmostpheres it has also been shown that $U_2O_3P_2O_7$ further decomposes with the loss of oxygen at temperatures above 1000℃ (Eq. 9) [19]. Combining Eqs. 8 and 9, the decomposition of a single HUO_2PO_4 would eventually give rise to an additional ¼O_2 lost through the decomposition of $U_2O_3P_2O_7$ representing a theoretical mass loss of 2.19%. Again, assuming a K:H ratio of 0.72:0.28, then a theoretical mass loss of 0.612% at temperatures over 1000℃ can be predicted. This is slightly lower than the 0.725% mass loss recorded.

$$2U_2O_3P_2O_7 \rightarrow 2(UO)_2P_2O_7 + O_2 \quad (9)$$

Beyond the first two dehydration events no further mass loss associated with meta-ankoleite was observed until 1150℃ indicating that the remaining KUO_2PO_4 structure does not decompose before or during the targeted temperature range of 1000–1100℃ used for ceramication of the residue wastes [15-17]. Over 500℃ a large endothermic peak was observed likely due to melting or phase change phenomena.

A similar thermal profile was obtained for the real

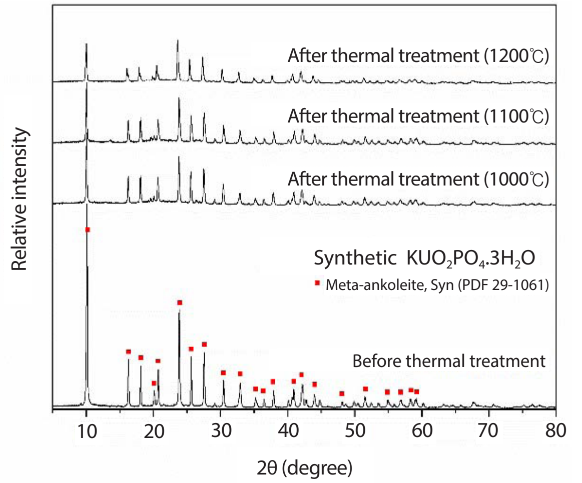


Fig. 3. X-ray diffractogram of synthetic meta-ankoleite both before and after thermal treatment at 1000℃, 1100℃, and 1200℃.

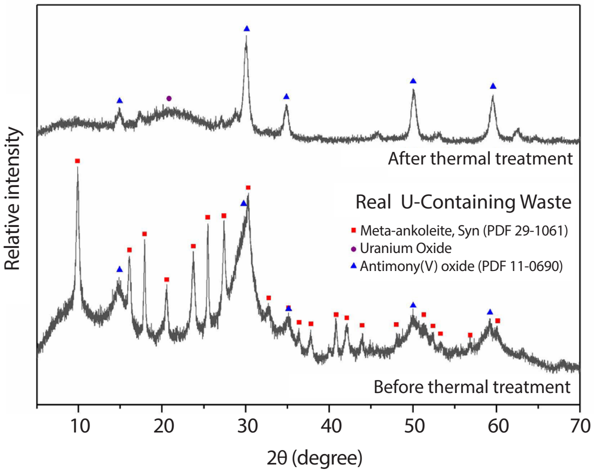


Fig. 4. X-ray diffractogram of the real meta-ankoleite containing waste both before and after thermal treatment at 1100℃.

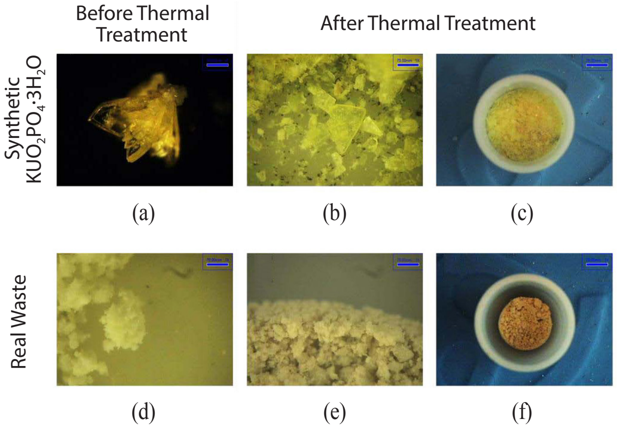


Fig. 5. Photographs of synthetic meta-ankoleite and the real waste before and after thermal treatment.

waste (Fig. 2(b)). Double de-hydration events occurred over a wider temperature range 28–150℃. The events were accompanied by two endothermic heat flows. Both of these peaks were broader than those recorded for the synthetic uranyl phosphate indicating non-uniformed dehydration events were taking place. This is likely due to the amorphous nature of the sample leading to a non-homogeneous crystal structure. An as-yet un-assigned exothermic peak was identified at 345.15℃ associated with a mass loss of 8.165% (0.8071 mg), likely due to the decomposition of low molecular weight compounds and further dehydration events. Again, no further mass loss was observed until 1150℃ with the corresponding endothermic heat flow likely due to melting or phase change phenomena.

3.3 Post thermal treatment analysis

Powder XRD diffraction patterns of synthetic meta-ankoleite after thermal treatment at 1000, 1100 and 1200℃ are shown in Fig. 3. For comparison, the XRD pattern before thermal treatment is also shown. Analysis of the XRD patterns indicates that the meta-ankoleite phase remains stable on account of no changes to the recorded peaks. This suggests, along with the TG analysis results, that after a two-step de-hydration event synthetic meta-ankoleite does not further decompose or undergo further phase changes. Visual inspection of the sample before and after thermal treatment were comparable in appearance (Fig. 5(a), (b) & (c)), thus further supporting the idea that wide spread degradation of the crystal structure does not occur.

Recorded XRD patterns of the real waste both before and after thermal treatment are shown in Fig. 4. Before thermal treatment uranium (14.5wt%) was present as meta-ankoleite while antimony (15.5wt%) was present as antimony(V) oxide (Sb_2O_5) [25]. No further crystalline phases were identifiable. The relative intensity and broadness of the peaks indicates a lack of long range crystallinity, particularly for those identifiable with antimony(V) oxide, which suggests the crystal phases possess a degree of amorphous character and also smaller grain sizes.

After thermal treatment at 1100℃ a complete loss of peaks associated with the meta-ankoleite mineral phase was observed; with the emergence of a peak at 20.8° assigned to uranium oxide [17]. In contrast, antimony(V) oxide remains with associated peaks being clearly observed (Fig. 4).

The real waste as received was a semi-amorphous powder (Fig. 5(d)). Following thermal treatment visual inspection indicates that a degree of self-sintering had taken place (Fig. 5(e) & (f)). Silica, antimony oxides and phosphates are all known glass former materials. It has been previously shown that they form a glass-ceramic composite upon thermal treatment [17].

Based on these preliminary results, it would appear that the uranium phosphate mineral phase meta-ankoleite is incorporated within the bulk waste either as an amorphous phase and/or as uranium oxide. Incorporation of the uranium phase into the waste is suitable for real thermal processes and therefore the use of meta-ankoleite formation for the sequestering of uranium is unlikely to pose any significant challenges later in the process as part of waste sentencing and immobilisation for the purpose of long term disposal [17].

4. Conclusions

Thermodynamic properties of the uranyl phosphate mineral phase meta-ankoleite ($KUO_2PO_4.3H_2O$) and a uranyl phosphate containing waste have been studied and compared. It has been shown that the meta-ankoleite mineral phase undergoes a double de-hydration event at 56.90 and 125.85℃. This having been observed in both the synthetic and real waste samples. Synthetically produced meta-ankoleite remains stable until at least 1150℃. In contrast, when present in the real waste the meta-ankoleite phase is not identifiable after thermal treatment indicating incorporation within the bulk waste either as an amorphous phase and/or as uranium oxide.

Co-contaminants in the real waste, namely, antimony have been shown to remain unchanged under the applied thermal conditions. The co-contaminants are glass former materials and have been observed to have the added benefit of being self-sintering.

Based on the preliminary results presented in this study, thermal treatment of the uranyl phosphate mineral phase meta-ankoleite, present within the waste, for the purpose of immobilisation should pose no detrimental effects with regards to the final wasteform.

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