Electronic Supplementary Material (ESI) for RSC Advances.

Expanding the nuclear forensic toolkit: Chemical profiling of uranium ore concentrate particles by synchrotron X-ray microanalysis

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Electronic Supplementary Information

XANES Edge Position Data

Edge positions were determined as the inflection point of the XANES spectra, defined as a maximum in the first derivative of the spectra.

Table S1 – Sample and standard XANES inflection point data

Sample	μ-XRD Phase ID	Edge Inflection Point (eV)
Rum Jungle	U_3O_8	17171.0
Olympic Dam	U_3O_8	17170.8
Key Lake	U_3O_8	17170.7
Rossing	U_3O_8	17170.8
Lucky Mc	Jachymovite	17172.0
Mindola	Copper Uranium Oxide	17171.5
Faraday	Uranium Oxide Ammonia Hydrate	17171.8
South Alligator	Uranium Oxide Ammonia Hydrate	17171.7
Mary Kathleen	U_3O_8 + Zippeite	17171.2
Ranger	U_3O_8 + Sodium Diuranate	17171.5
Everest	U_3O_8 + Uranium Oxide Ammonia Hydrate	17171.7
ESI 2	U_3O_8 + Na-Compreignacite	17171.5
U_3O_8 Standard	-	17171.0
UO_3 Standard	-	17171.7

EDX Analysis of Faraday and South Alligator UOCS

Figure S1 shows EDX analysis of particles from South Alligator and Faraday UOC samples. Particles analysed separately by synchrotron XRD were assigned as ammonium uranium oxide hydrate in both samples. However, magnesium precipitation is documented rather than ammonia, and EDX was used to examine the association of different elements with uranium.

The stoichiometry of MDU has been described as $MgU_2O_7{\cdot}3H_2O$, and of ADU as $2UO_2{\cdot}NH_3{\cdot}3H_2O$. In both of these species the distinguishing component (magnesium or nitrogen) makes up a minor component of the composition, with approximately 4.5 At % nitrogen and 6.3 At % magnesium, and therefore they will be expected to be a small component of the EDX spectrum. In addition, nitrogen K X-rays are extremely soft (0.392 keV), and are susceptible to attenuation and absorption by particle surface topography. This may explain the low yield of nitrogen X-rays in both samples.

Two populations of particles were observed in the samples, in which uranium was associated with variable but low levels of nitrogen and magnesium. In the south alligator sample uranium was associated with nitrogen and magnesium in separate populations of particles. This supports the assignment of $2UO_2{\cdot}NH_3{\cdot}3H_2O$ to both measured diffraction data, but is indicative of a heterogeneous sample, perhaps with a significant amorphous population which was not detected by XRD.

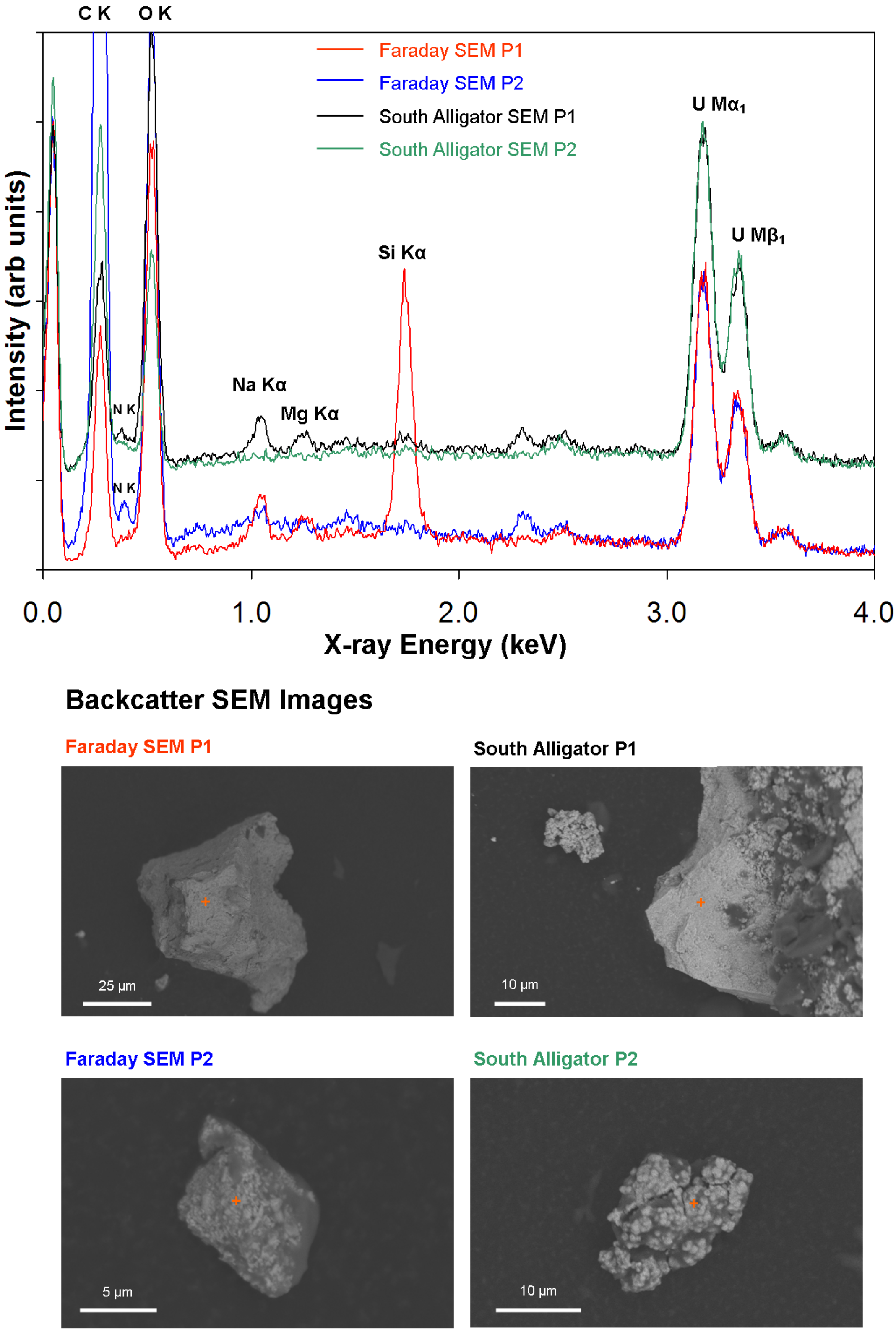

Figure S1 – Representative energy dispersive X-ray analysis of particles from Faraday and South Alligator UOC samples.

XRF Analysis of Mindola UOC

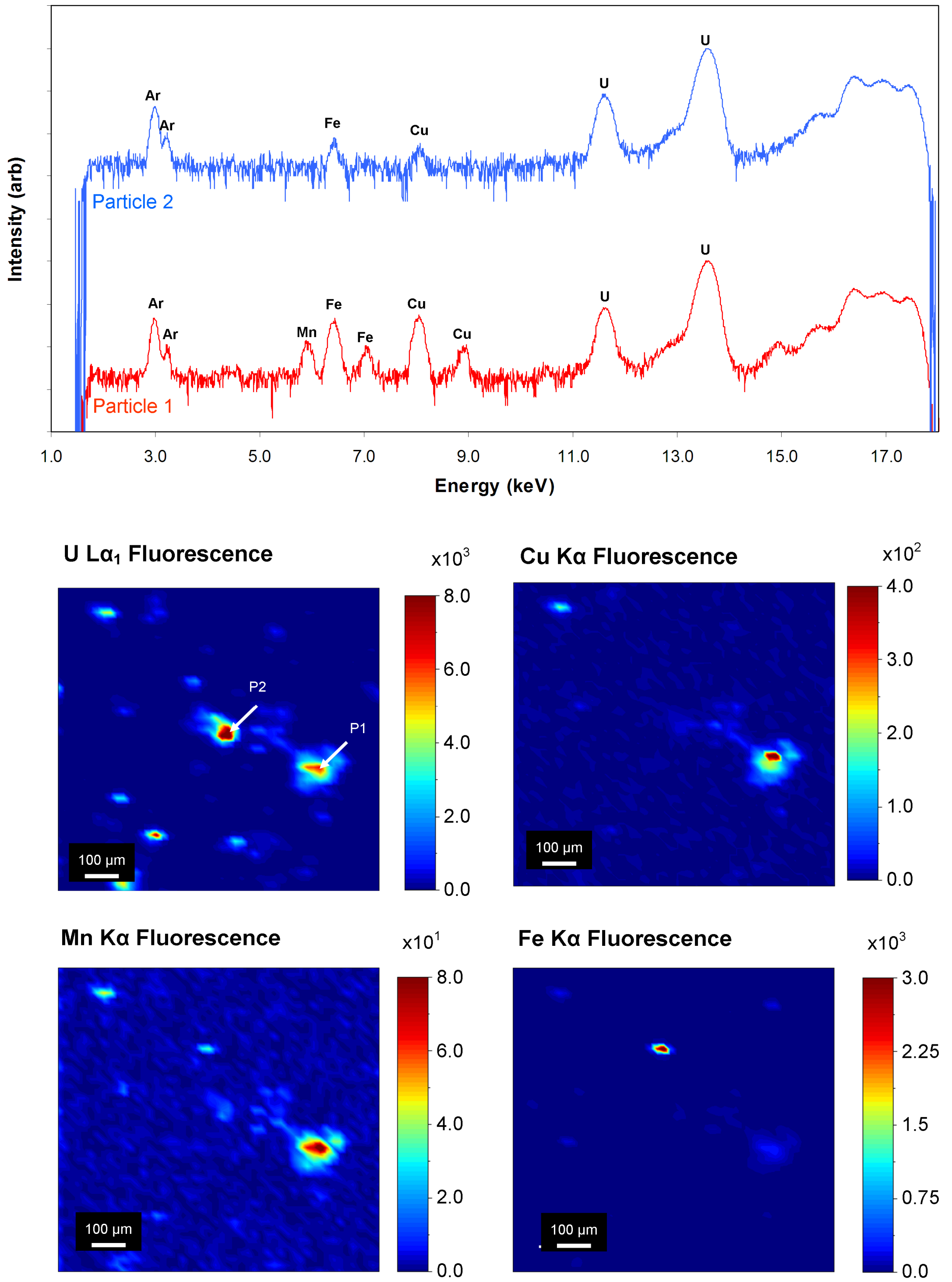

Figure S2 – Point and mapping µ-XRF analysis of Mindola UOC. A strong association of Cu and U is observed in some particles, consistent with CuU_3O_{10} observed by µ-XRD.

EDX Analysis of Everest and ESI 2 UOCs

EDX analysis of particles from Everest and ESI 2 UOCs shows a greater association of sodium and uranium in the ESI 2 sample. Low levels of sodium are associated with the Everest sample also, these may be due to impurities or X-rays generated in the sample support.

The morphologies of particles in the samples are distinctive; the ESI 2 sample displays aggregates of needle-form crystals, which are consistent with precipitation products, whereas the Everest sample has nodular appearance which is more suggestive of U_3O_8. This is consistent with µ-XRD data (Figure 3) which suggests a predominance of Na-Compreignacite in the ESI-2 sample (with secondary U_3O_8), whereas reflections for U_3O_8 are dominant in the Everest sample (with secondary Uranium Ammonia Oxide Hydrate).

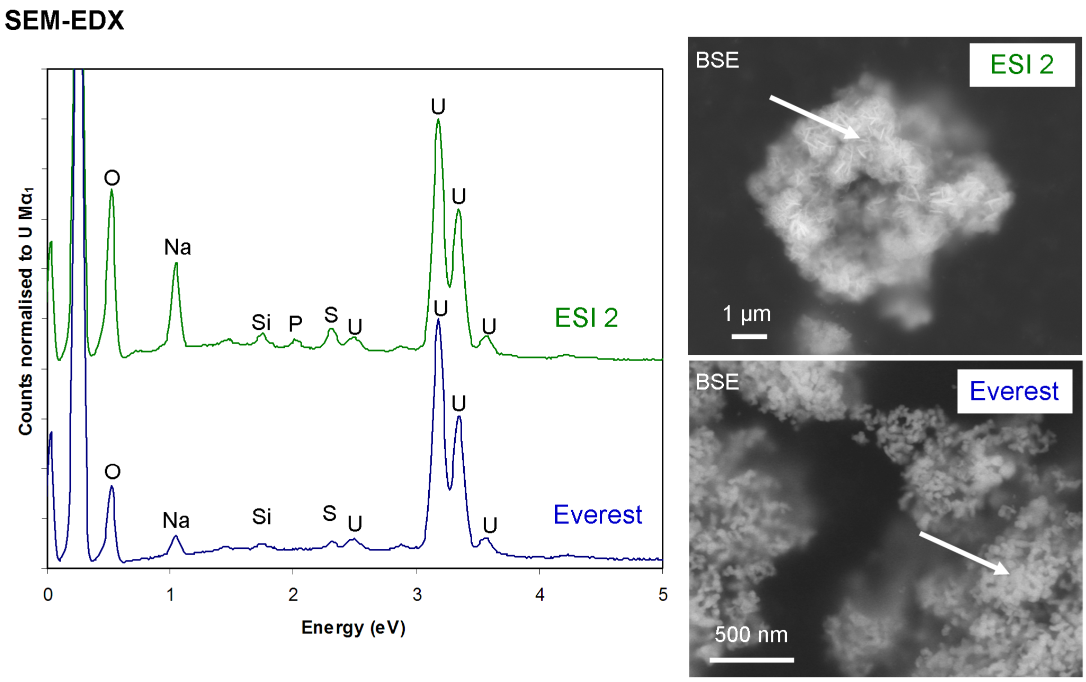

Figure S3 – Representative energy dispersive X-ray analysis of particles from Everest and ESI 2 UOC samples

Polycrystallinity of Olmypic Dam and Rum Jungle UOC Particles

Figure S4 shows crystallite size effects in the Olympic Dam and Rum Jungle U_3O_8 UOCs. The spotty nature of the Olympic Dam diffraction patterns is due to a lower number of randomly oriented crystallites in the beam interaction volume. The grain size appears to be variable also between the two example particles, with particle 1 displaying more complete diffraction rings. In contrast, the Rum Jungle sample likely comprises of a greater number of smaller crystallites, resulting in a diffraction pattern which approximates a powder.

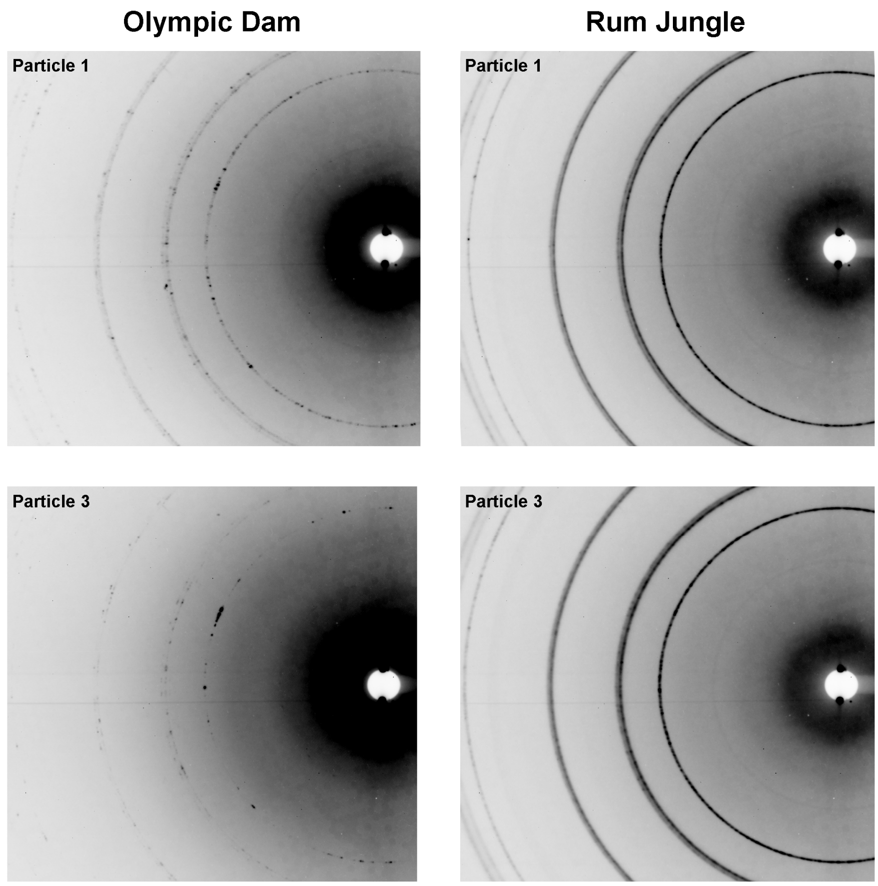

Figure S4 – Two dimensional diffraction patterns from particles in U_3O_8 containing UOC samples.

Polycrystallinity and Reproducibility of South Alligator and Faraday UOC Particles

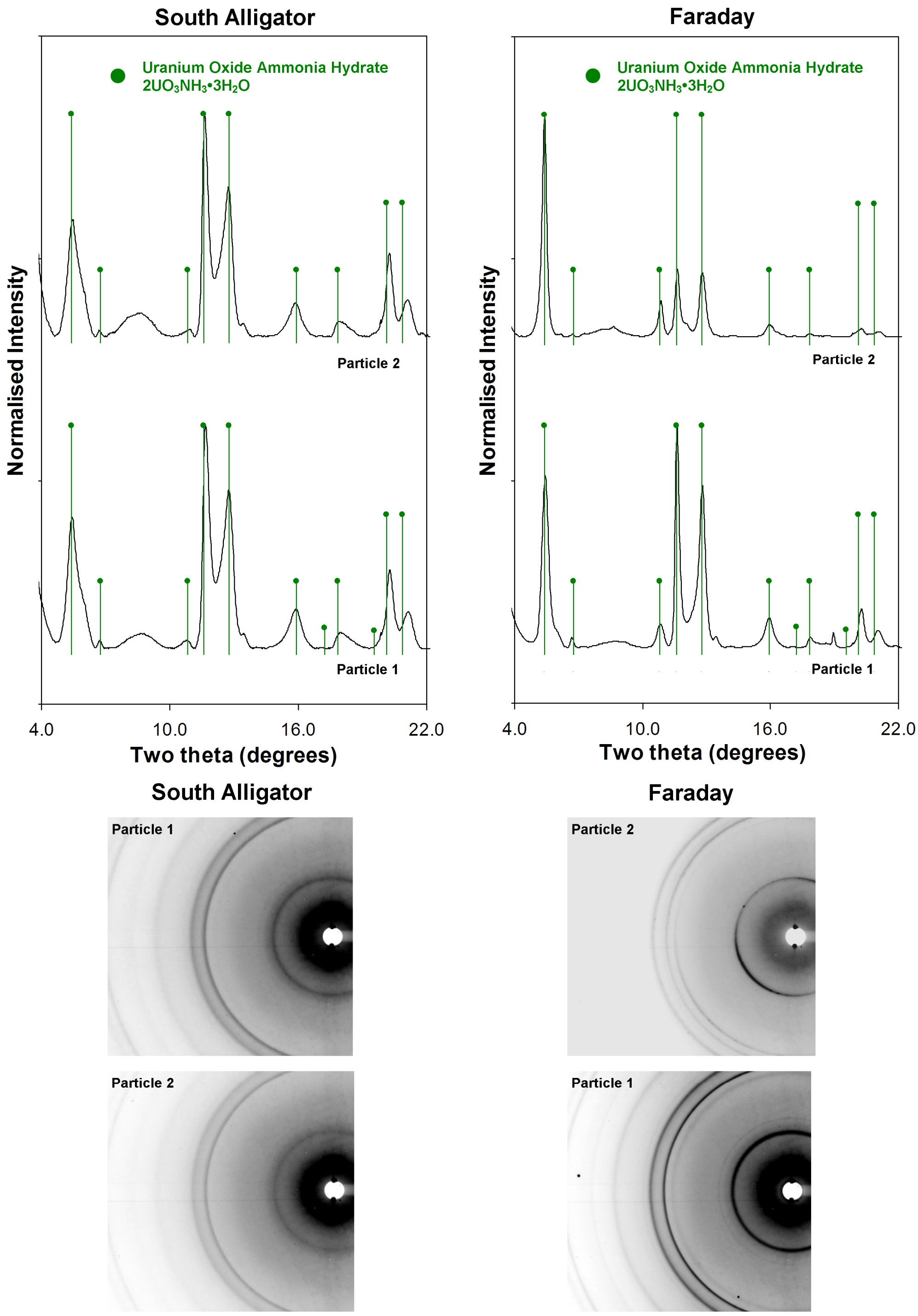

Figure S5 - µ-XRD data for two UOCs identified as containing only ammonium-uranium oxide hydrate. The broader peak width is consistent for both South Alligator particles, whereas the Faraday samples have narrower reflections. The difference in intensity between the two Faraday samples is due to crystallite orientation effects, as observed for the Particle 1 innermost ring which has a large non-uniform section.