

A disposal-MOX concept for plutonium disposition: Supplementary Information

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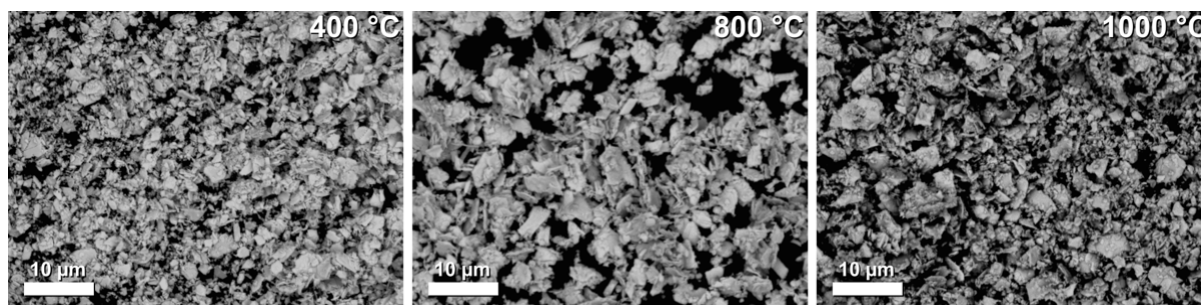
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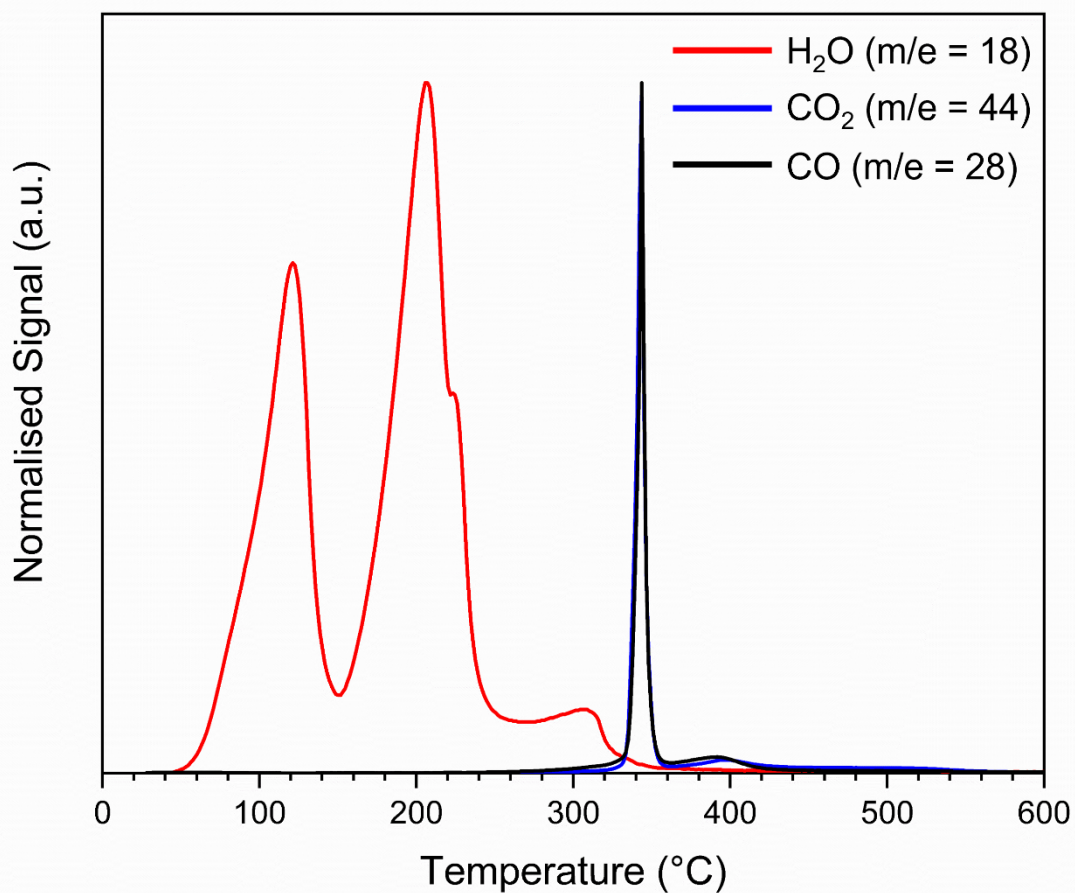
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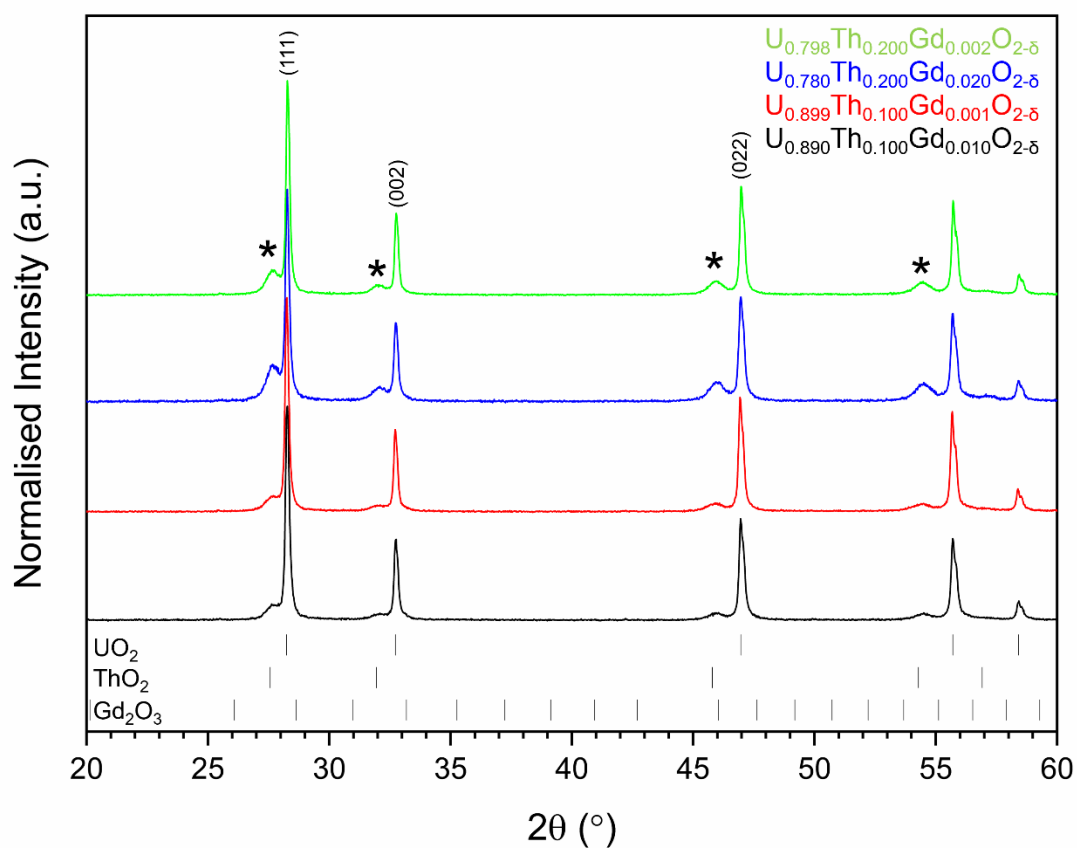
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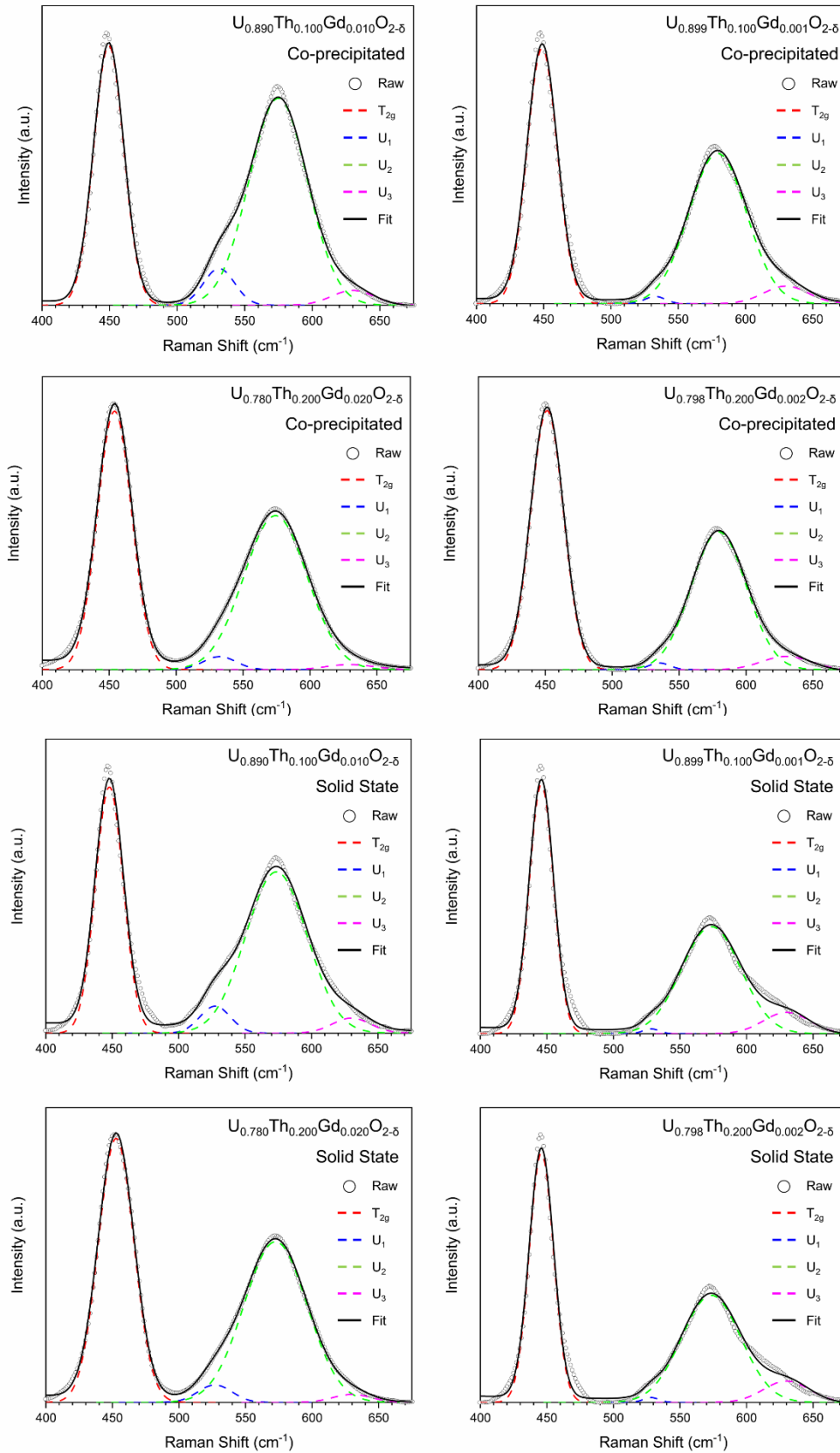
Supplementary Figure 1. SEM micrographs revealing platelet morphology of $U_{0.798}Th_{0.200}Gd_{0.002}O_{2.5}$ oxide powders obtained by calcination of co-precipitated material at 800 °C



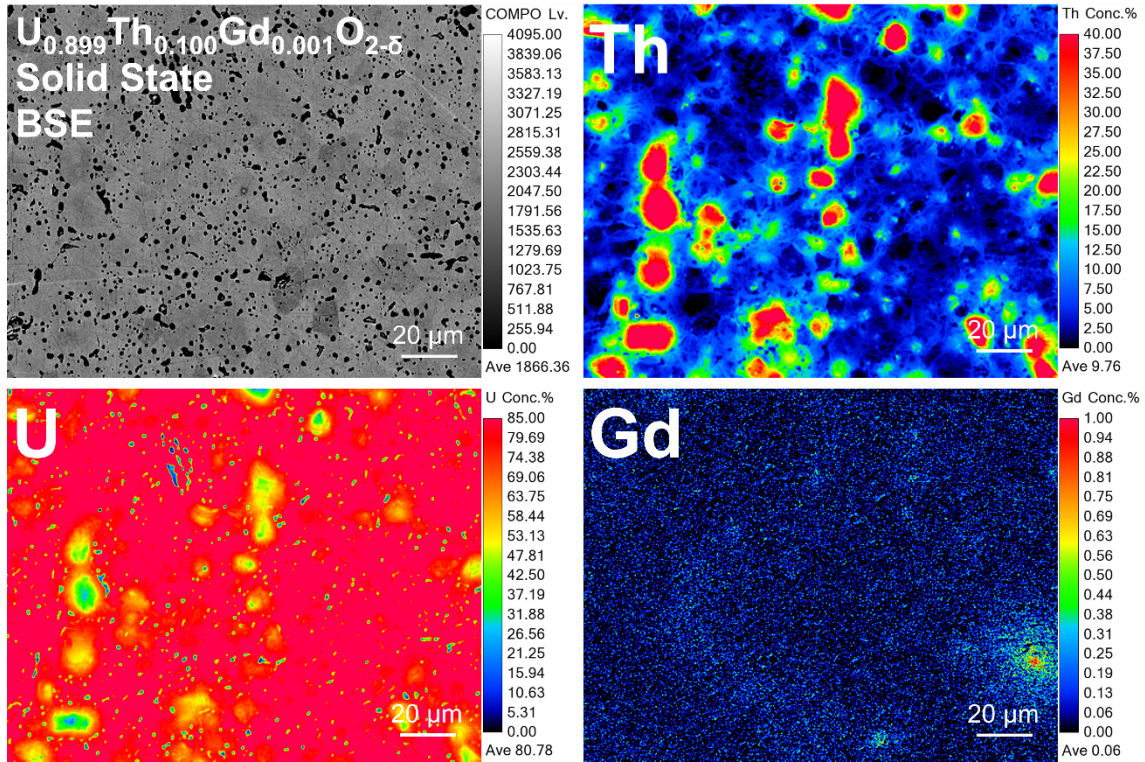
Supplementary Figure 2. TG-MS data collected during calcination of $U_{0.890}Th_{0.100}Gd_{0.010}O_{2.5}$ prepared by co-precipitation route.



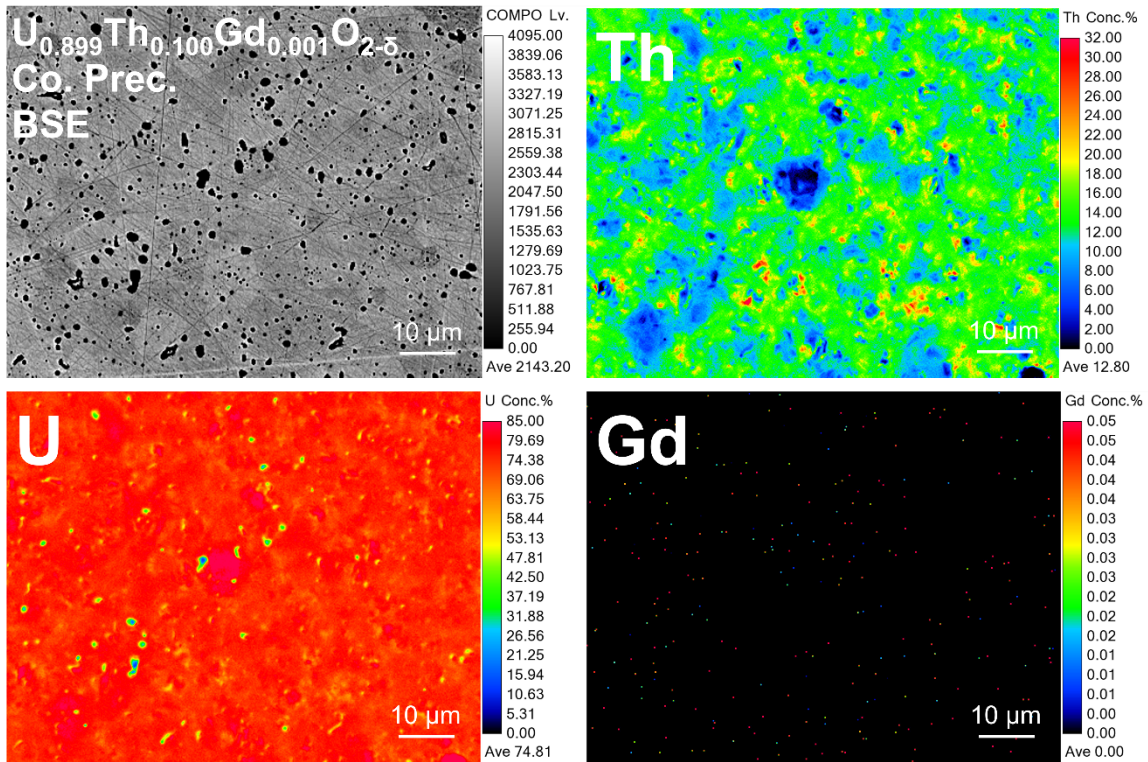
Supplementary Figure 3. XRD data for oxides obtained from decomposition of oxalates at 800 °C. Two sets of fluorite reflections were readily indexed to UO_2 (ICSD 160814) labelled with (hkl) values and ThO_2 (ICSD 253564) labelled with asterisks. The relative intensity of ThO_2 -rich peaks corresponds with measured compositions. Peak shift in the ThO_2 -rich reflections was attributed to incorporation of U^{4+} and Gd^{3+} . No peaks corresponding to cubic C-type Gd_2O_3 (ICSD 94892), or monoclinic Gd_2O_3 were observed.



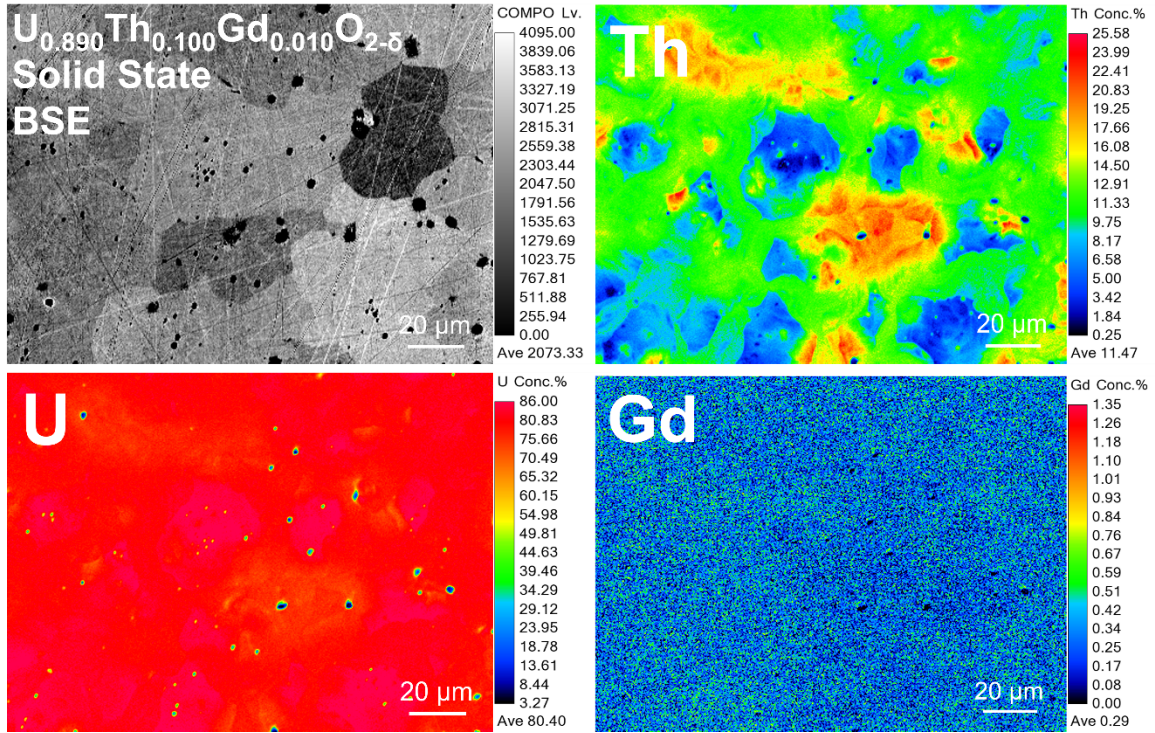
Supplementary Figure 4. Complete set of deconvoluted Raman spectra for disposal MOX materials fabricated by co-precipitation and solid state route



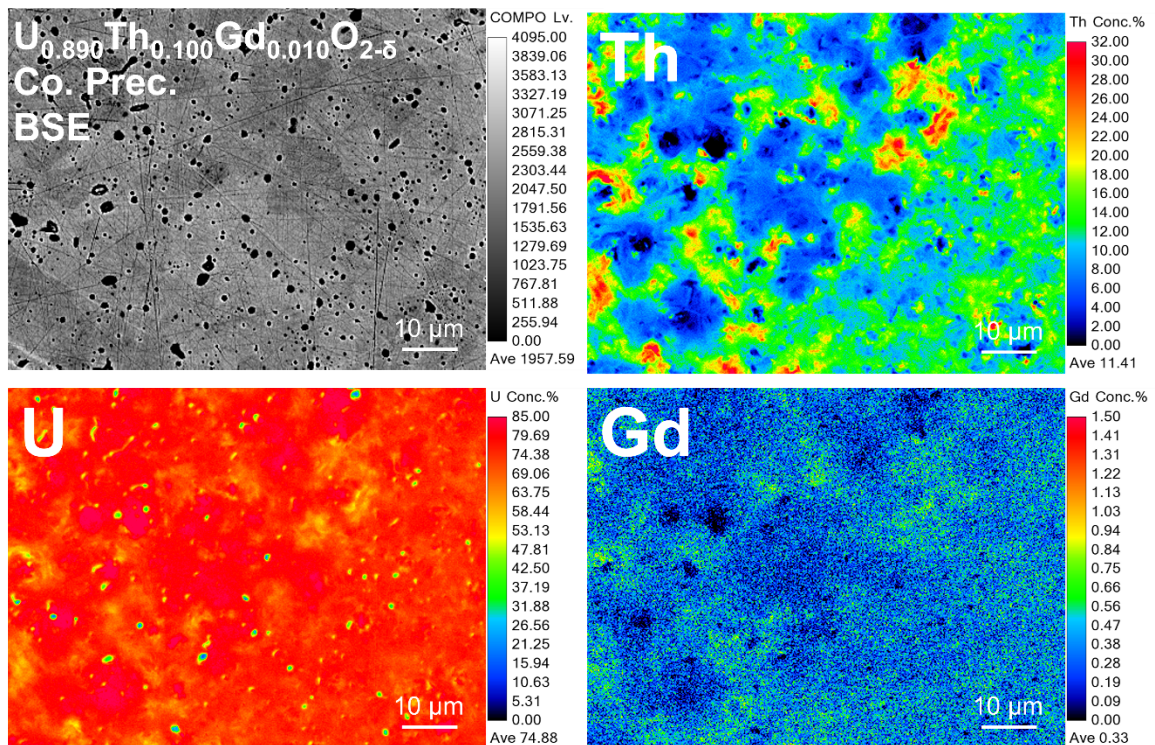
Supplementary Figure 5. EPMA maps for U, Th and Gd present in disposal MOX with nominal formulation $U_{0.899}Th_{0.100}Gd_{0.001}O_{2-\delta}$ fabricated by the solid-state route.



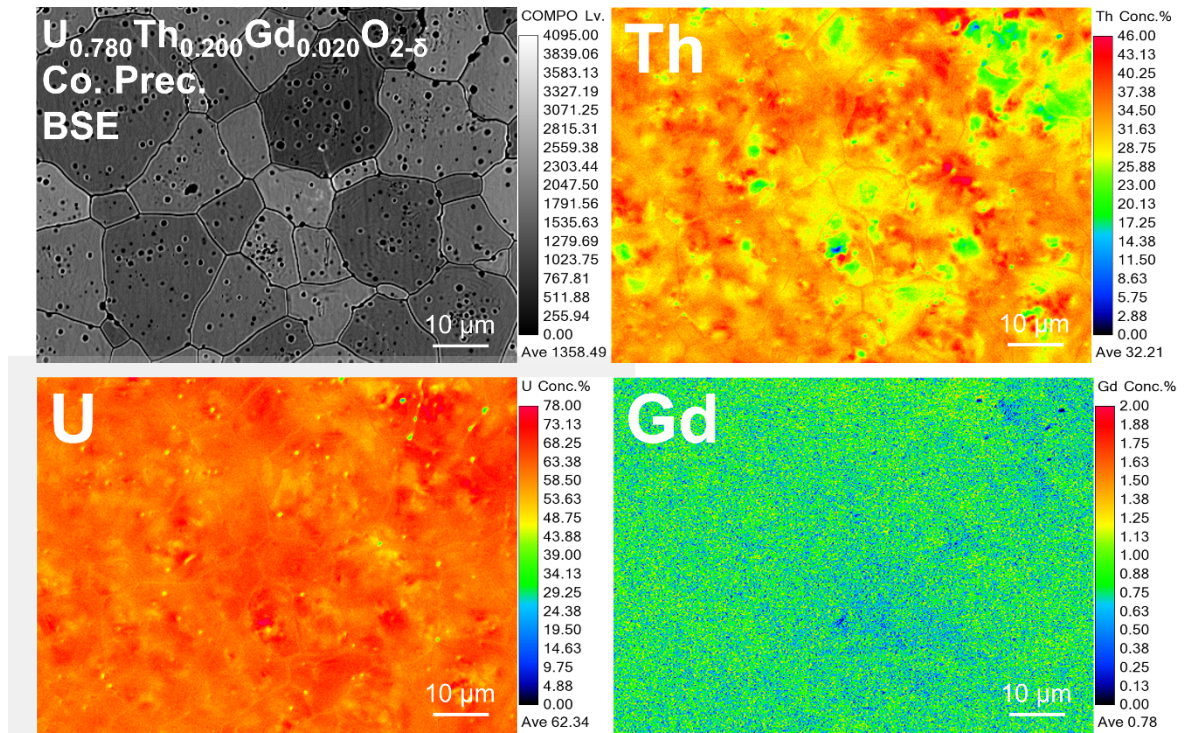
Supplementary Figure 6. EPMA maps for U, Th and Gd present in disposal MOX with nominal formulation $U_{0.899}Th_{0.100}Gd_{0.001}O_{2-\delta}$ fabricated by the co-precipitation route.



Supplementary Figure 7. EPMA maps for U, Th and Gd present in disposal MOX with nominal formulation $U_{0.890}Th_{0.100}Gd_{0.010}O_{2-\delta}$ fabricated by the solid-state route.



Supplementary Figure 8. EPMA maps for U, Th and Gd present in disposal MOX with nominal formulation $U_{0.890}Th_{0.100}Gd_{0.010}O_{2-\delta}$ fabricated by the co-precipitation route.



Supplementary Figure 9. EPMA maps for U, Th and Gd present in disposal MOX with nominal formulation $U_{0.780}Th_{0.200}Gd_{0.020}O_{2-\delta}$ fabricated by the co-precipitation route.

Supplementary Table 1. Tabulated values for deconvoluted Raman spectra

| Synthesis | Composition | T _{2g} Pos. (cm ⁻¹) | T _{2g} FWHM (cm ⁻¹) | U ₁ Pos. (cm ⁻¹) | U ₁ FWHM (cm ⁻¹) | U ₂ Pos. (cm ⁻¹) | U ₂ FWHM (cm ⁻¹) | U ₃ Pos. (cm ⁻¹) | U ₃ FWHM (cm ⁻¹) | χ ² |
|-------------|---|--|--|---|---|---|---|---|---|----------------|
| Co-prec. | U _{0.899} Th _{0.100} Gd _{0.001} O _{2-δ} | 448.82 ± 0.09 | 26.39 ± 0.23 | 532.84 ± 2.74 | 17.53 ± 7.28 | 579.29 ± 0.48 | 50.39 ± 1.42 | 630 ± 0.00 | 38.73 ± 8.25 | 0.09 |
| Co-prec. | U _{0.890} Th _{0.100} Gd _{0.010} O _{2-δ} | 449.34 ± 0.08 | 26.24 ± 0.23 | 531.01 ± 1.07 | 27.07 ± 2.41 | 574.80 ± 0.27 | 50.70 ± 1.32 | 630 ± 0.00 | 36.95 ± 8.81 | 0.09 |
| Co-prec. | U _{0.798} Th _{0.200} Gd _{0.002} O _{2-δ} | 451.30 ± 0.05 | 28.93 ± 0.14 | 535.30 ± 1.82 | 18.49 ± 4.91 | 579.64 ± 0.39 | 48.52 ± 1.04 | 630 ± 0.00 | 43.04 ± 8.35 | 0.03 |
| Co-prec. | U _{0.798} Th _{0.200} Gd _{0.002} O _{2-δ} | 453.95 ± 0.03 | 30.31 ± 0.09 | 532.44 ± 1.49 | 29.24 ± 3.71 | 574.24 ± 0.22 | 55.81 ± 1.36 | 630 ± 0.00 | 41.56 ± 13.96 | 0.01 |
| Solid State | U _{0.899} Th _{0.100} Gd _{0.001} O _{2-δ} | 445.83 ± 0.09 | 21.46 ± 0.23 | 528.24 ± 3.74 | 13.98 ± 10.13 | 573.45 ± 0.59 | 52.31 ± 1.83 | 630 ± 0.00 | 40.81 ± 7.06 | 0.12 |
| Solid State | U _{0.890} Th _{0.100} Gd _{0.010} O _{2-δ} | 447.83 ± 0.09 | 24.91 ± 0.25 | 527.43 ± 1.39 | 26.98 ± 3.31 | 573.55 ± 0.36 | 54.38 ± 1.68 | 630 ± 0.00 | 34.10 ± 7.63 | 0.11 |
| Solid State | U _{0.798} Th _{0.200} Gd _{0.002} O _{2-δ} | 448.71 ± 0.08 | 27.06 ± 0.23 | 531.44 ± 2.09 | 19.33 ± 5.64 | 575.94 ± 0.32 | 51.38 ± 1.26 | 630 ± 0.00 | 38.09 ± 7.80 | 0.08 |
| Solid State | U _{0.798} Th _{0.200} Gd _{0.002} O _{2-δ} | 452.74 ± 0.04 | 21.45 ± 0.13 | 526.98 ± 1.27 | 30.09 ± 2.84 | 572.48 ± 0.22 | 55.89 ± 0.95 | 630 ± 0.00 | 34.43 ± 6.89 | 0.02 |