Supplemental Information

Ion	Bond Valence Sum	Oxidation State
K(1)	1.09	+1
K(2)	0.99	+1
K(3)	0.95	+1
U(1)	5.94	+6
U(2)	5.81	+6
U(3)	5.87	+6

Table S1: Results of bond valence sum calculations for $K_5U_5O_{17}(OH)$. All calculated BVS values fall within acceptable ranges for their respective oxidation states.

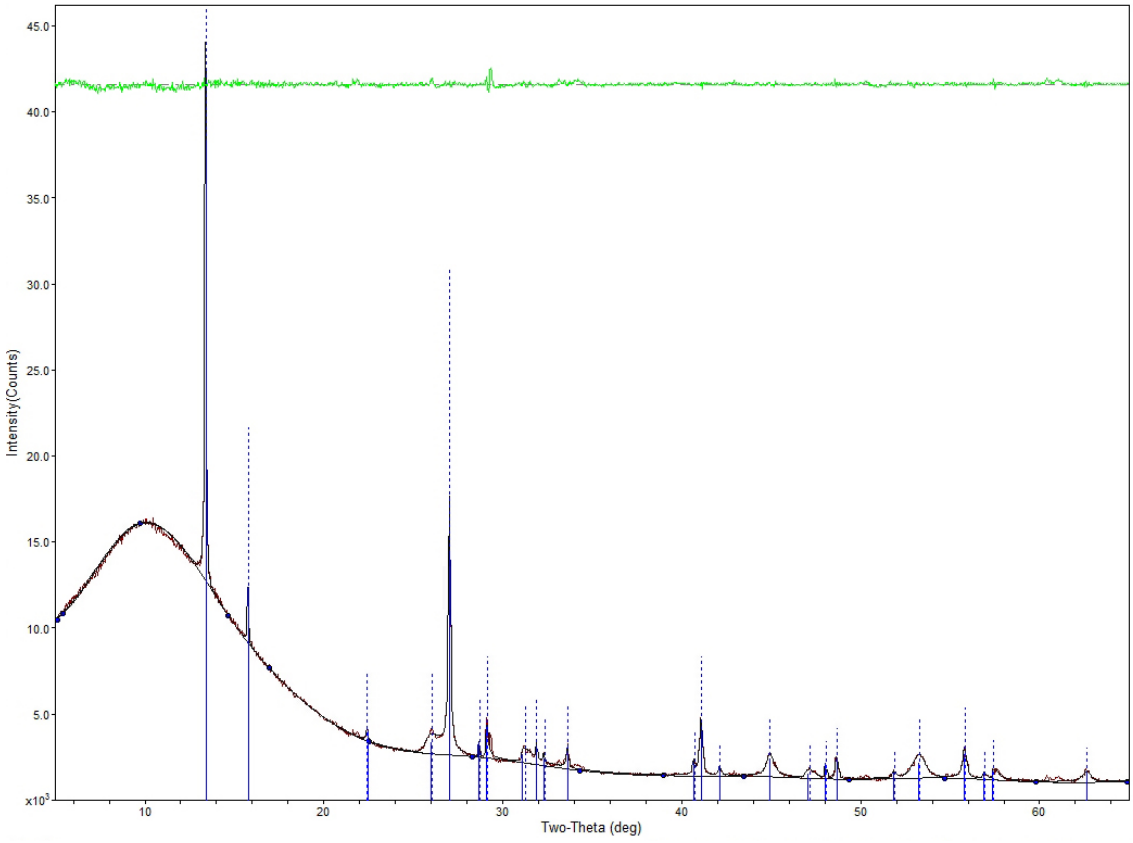

Figure S1: The powder x-ray diffraction pattern of $K_5U_5O_{17}(OH)$ compared with the calculated pattern derived from the .cif. Observed data is shown in red, and the Le Bail fit is shown in black. Peak positions are marked with blue lines, and the difference map is shown as green.

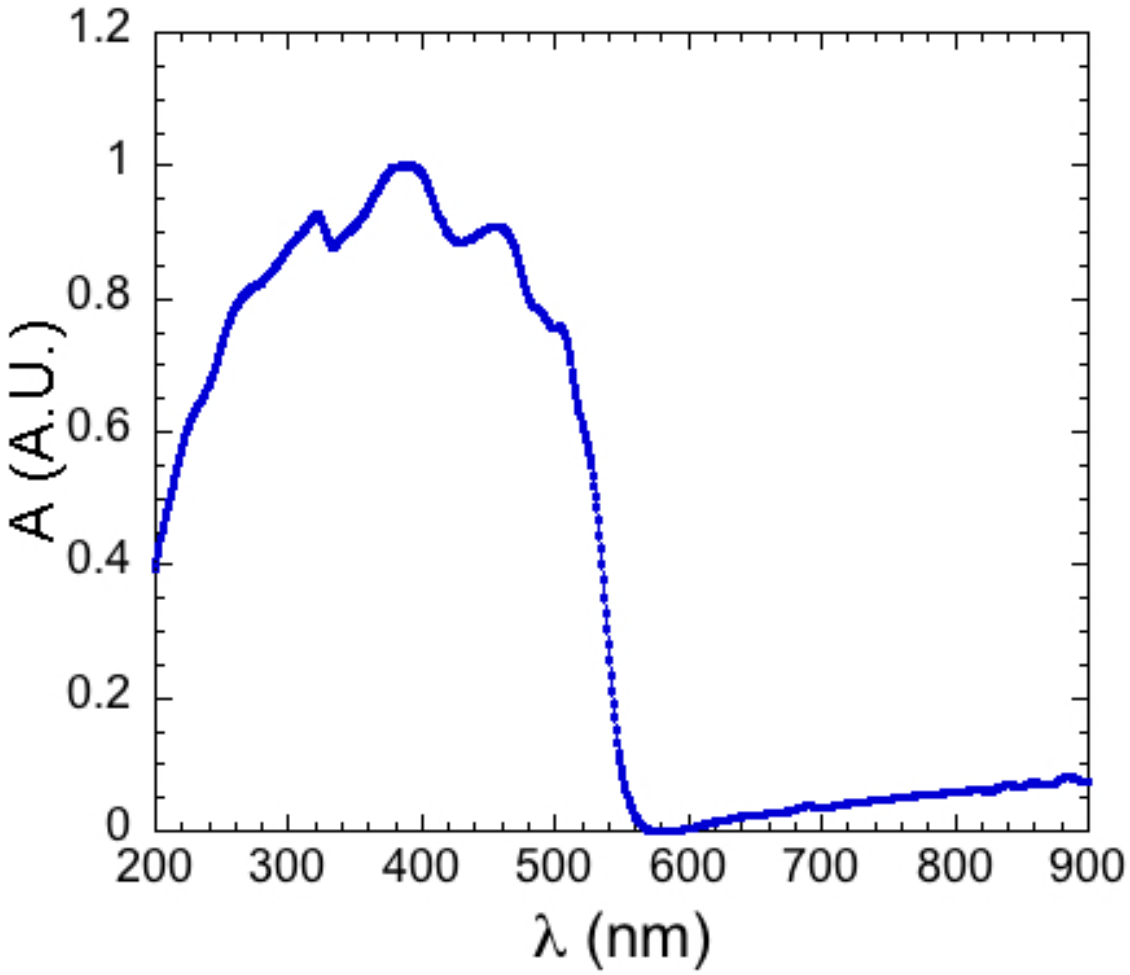

Figure S2: The UV-visible absorption spectrum of $K_5U_5O_{17}(OH)$. The spectrum shows a sharp absorption edge at 550 nm and the fine structure corresponding to the U 4*f* – 4*f* electronic transitions can be seen above the absorption band.

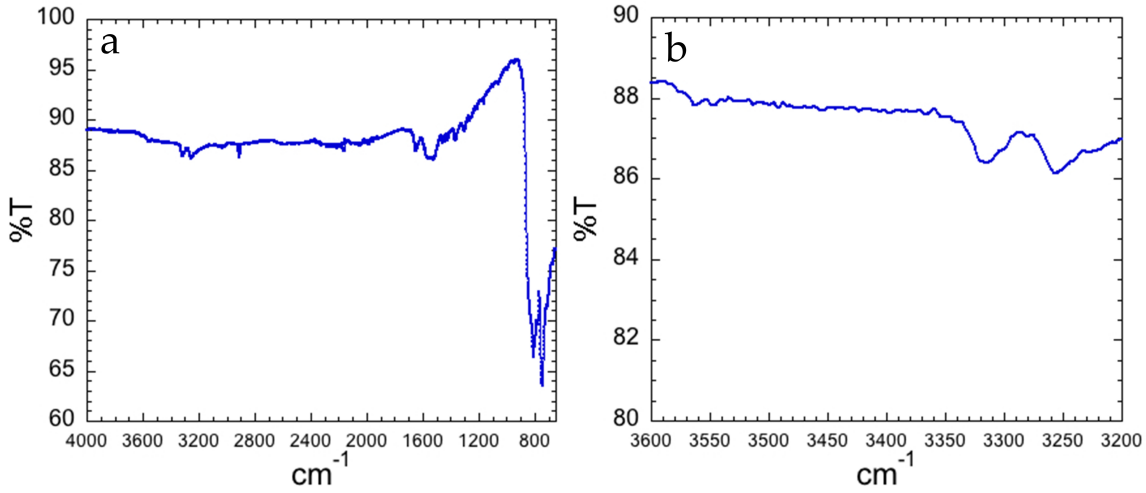

Figure S3: The IR spectrum of $K_5U_5O_{17}(OH)$. **a)** The full range spectrum from 4000 cm^{-1} to 650 cm^{-1}. **b)** A zoomed-in plot of the IR spectrum from 3600 cm^{-1} to 3200 cm^{-1}. Plot b highlights the weak O-H stretching bands. The weakness of the bands can be attributed to the low concentration of hydroxyl ions in the structure.