

**Table 1.** Selected Bond Valence Sum calculation for

$\text{UO}_2[\text{CH}_2(\text{PO}_3)_2] \cdot (\text{C}_2\text{H}_4\text{N}_2\text{H}_6)(\text{H}_2\text{O})$  (**UC1P2N-1**).

Atom	Bond Distances (Å)		Bond Valence Sum	Valence Sum
O1	O(1)-P(1)	1.511(7)	1.29	1.29
O2	O(2)-U(1)	2.330(6)	0.57	1.83
	O(2)-P(1)	1.518(6)	1.26	
O3	O(3)-U(1)	2.367(6)	0.53	1.76
	O(3)-P(1)	1.529(6)	1.22	
O4	O(4)-U(1)	2.393(5)	0.51	1.75
	O(4)-P(2)	1.524(6)	1.24	
O5	O(5)-U(1)	2.360(5)	0.54	1.82
	O(5)-P(2)	1.513(6)	1.28	
O6	O(6)-U(1)	2.367(5)	0.53	1.75
	O(6)-P(2)	1.531(5)	1.22	
O7	O(7)-U(1)	1.749(6)	1.79	1.79
O8	O(8)-U(1)	1.772(6)	1.71	1.71

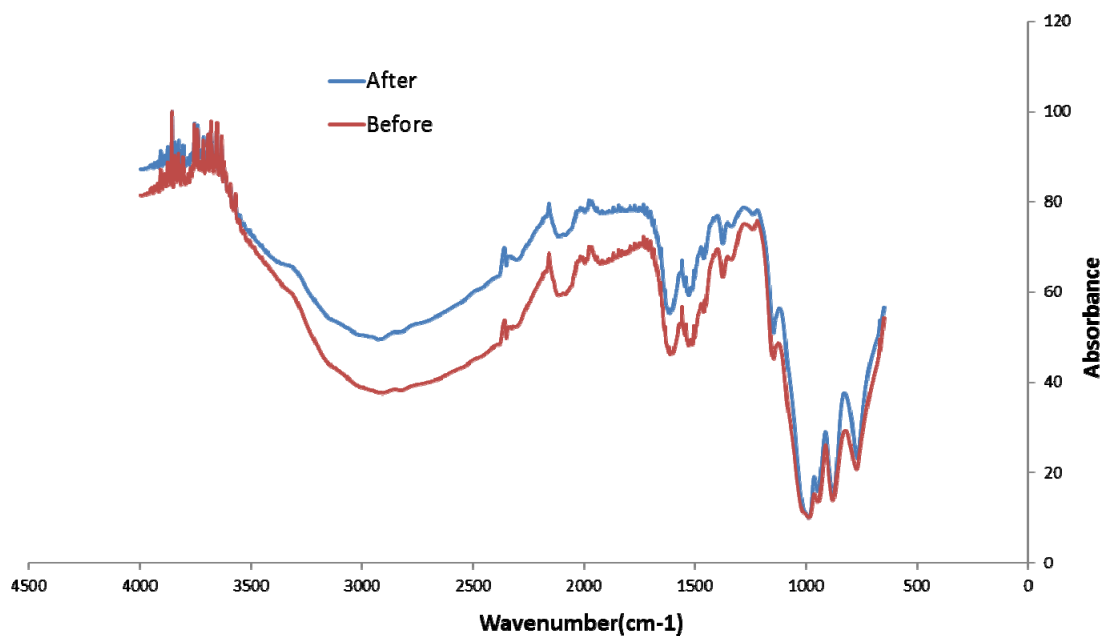


Figure S1. FT-IR spectrum of the  $\text{UO}_2[\text{CH}_2(\text{PO}_3)_2] \cdot (\text{C}_2\text{H}_4\text{N}_2\text{H}_6)(\text{H}_2\text{O})$  (UC1P2N-1) before and after the ion exchange, showing the peak of free water molecules at  $1650\text{cm}^{-1}$ .

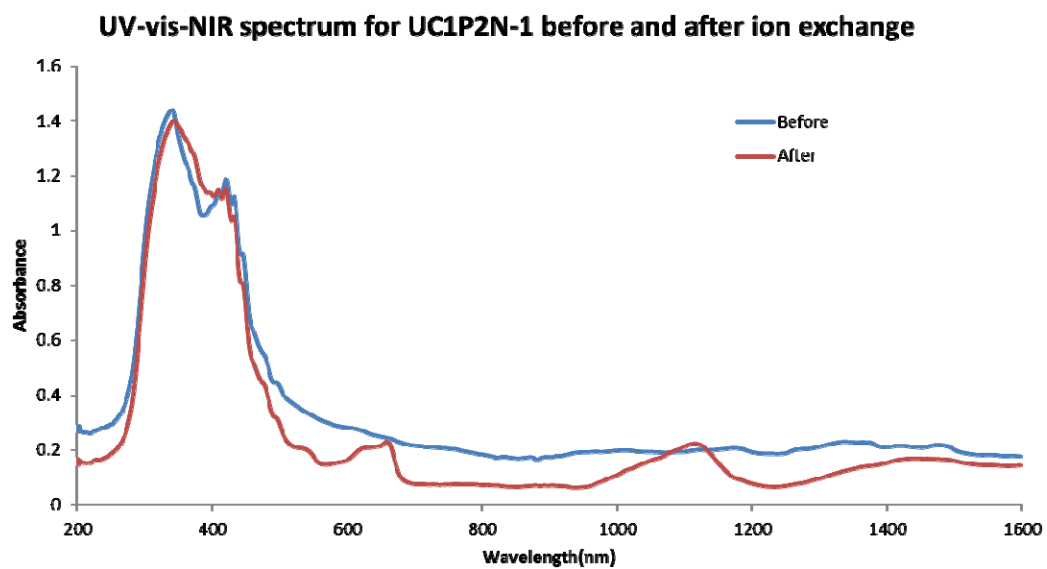


Figure S2: The UV-vis-NIR spectrum of UC1P2N-1 before and after the ion exchange with  $[\text{Co}(\text{en})_3]^{3+}$ , showing additional peaks after ion exchange.

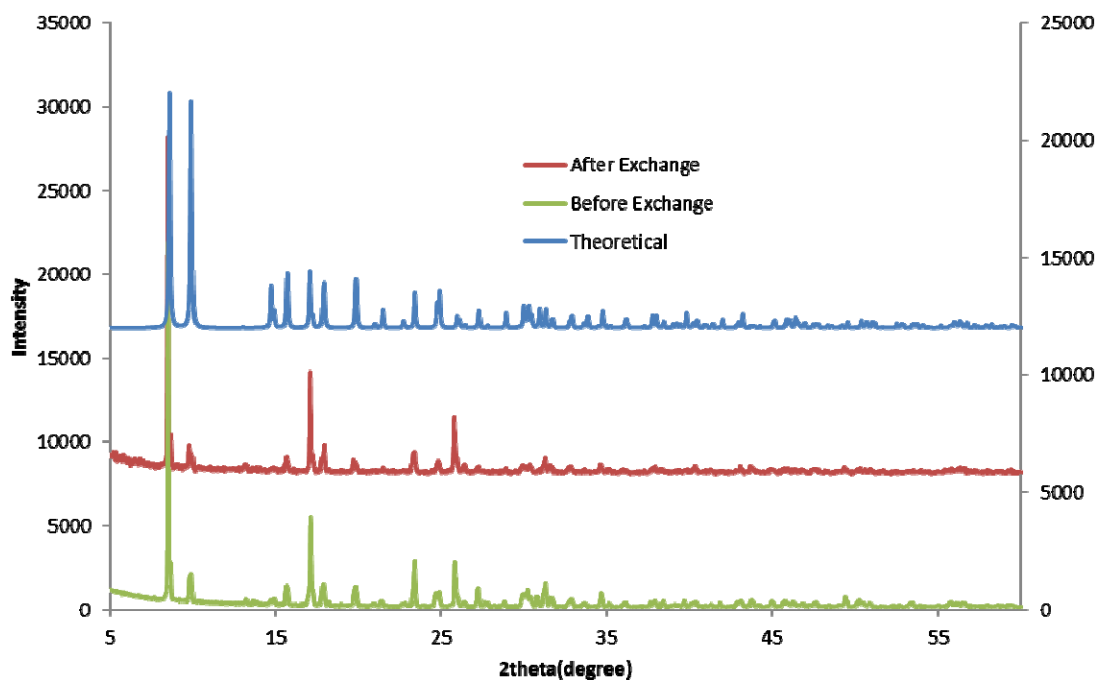
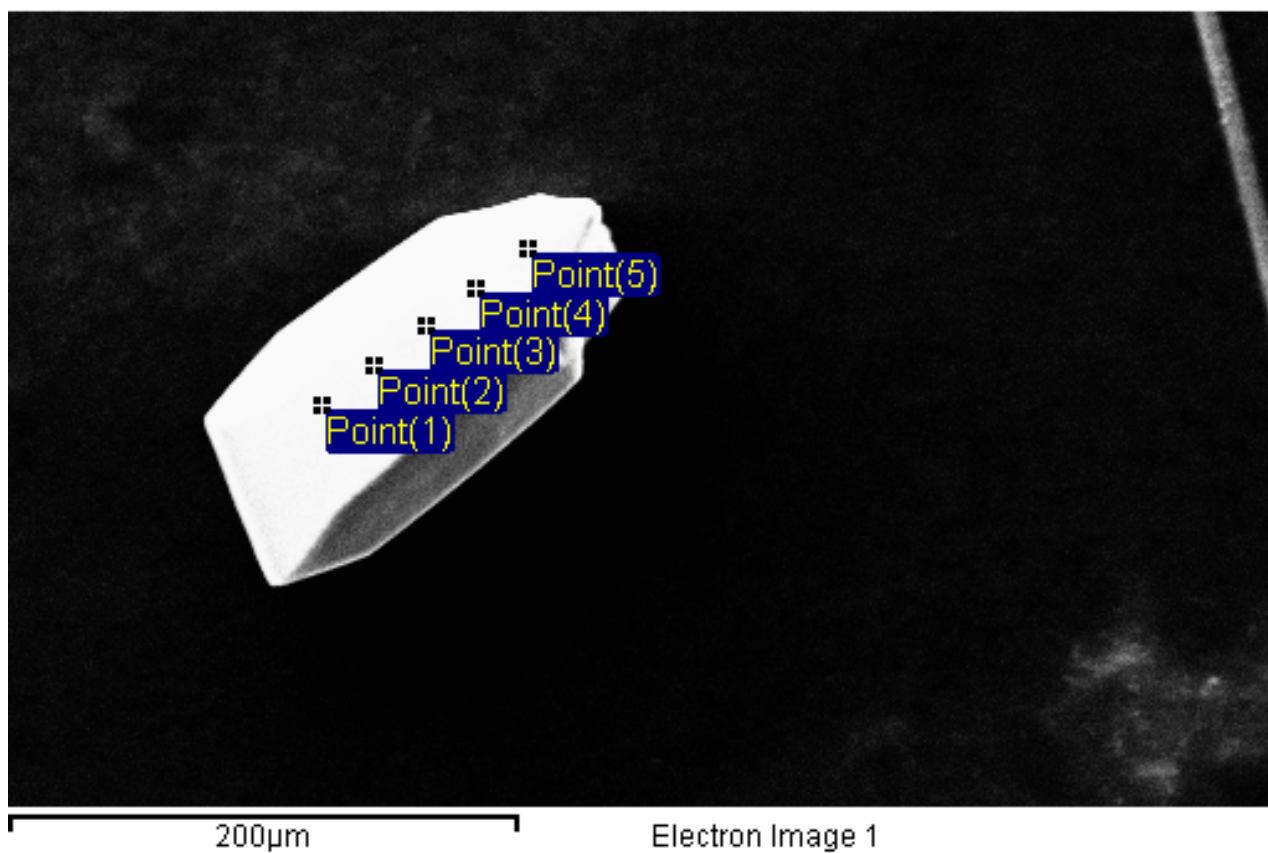


Figure S3. The powder X-ray diffraction pattern showing that the  $\text{UO}_2[\text{CH}_2(\text{PO}_3)_2] \cdot (\text{C}_2\text{H}_4\text{N}_2\text{H}_6)(\text{H}_2\text{O})$  (**UC1P2N-1**) is a pure sample and stable during the ion exchange process.

Table S2. EDS results showing the existence of Co in the crystal.

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Processing option : All elements analysed (Normalised)

Spectrum	In stats.	P	Co	U
Point(1)	Yes	67.41	2.65	29.94
Point(2)	Yes	66.48	2.92	30.60
Point(3)	Yes	66.83	2.24	30.93
Point(4)	Yes	46.50	6.81	46.69
Point(5)	Yes	66.35	1.52	32.13
Mean		62.71	3.23	34.06
Std. deviation		9.07	2.07	7.11
Max.		67.41	6.81	46.69
Min.		46.50	1.52	29.94

All results in atomic%