Supporting Information

Room temperature synthesis of UO_{2+x} nanocrystals and thin films via hydrolysis of uranium(IV) complexes

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Table S1. Crystallographic Data for **1**	
empirical formula	$C_{36}H_{76}O_4U$
crystal habit, color	block, light blue
crystal size (mm)	0.30 × 0.41 × 0.33
crystal system	Monoclinic
space group	*P21/n*
volume (Å^3)	4060.2(1)
a (Å)	12.4277(3)
b (Å)	17.9692(5)
c (Å)	18.4162(5)
α (deg)	90
β (deg)	99.156(1)
γ (deg)	90
Z	4
formula weight (g/mol)	810.99
density (calculated) (mg/m^3)	1.327
absorption coefficient (mm^{-1})	4.028
F_{000}	1664.0
total no. reflections	78945
unique reflections	14383
final *R* indices [$I > 2\sigma(I)$]	R_1 = 0.0238, wR_2 = 0.0461
largest diff. peak and hole (e$^-$ Å^{-3})	–1.35 and 1.79
GOF	1.080

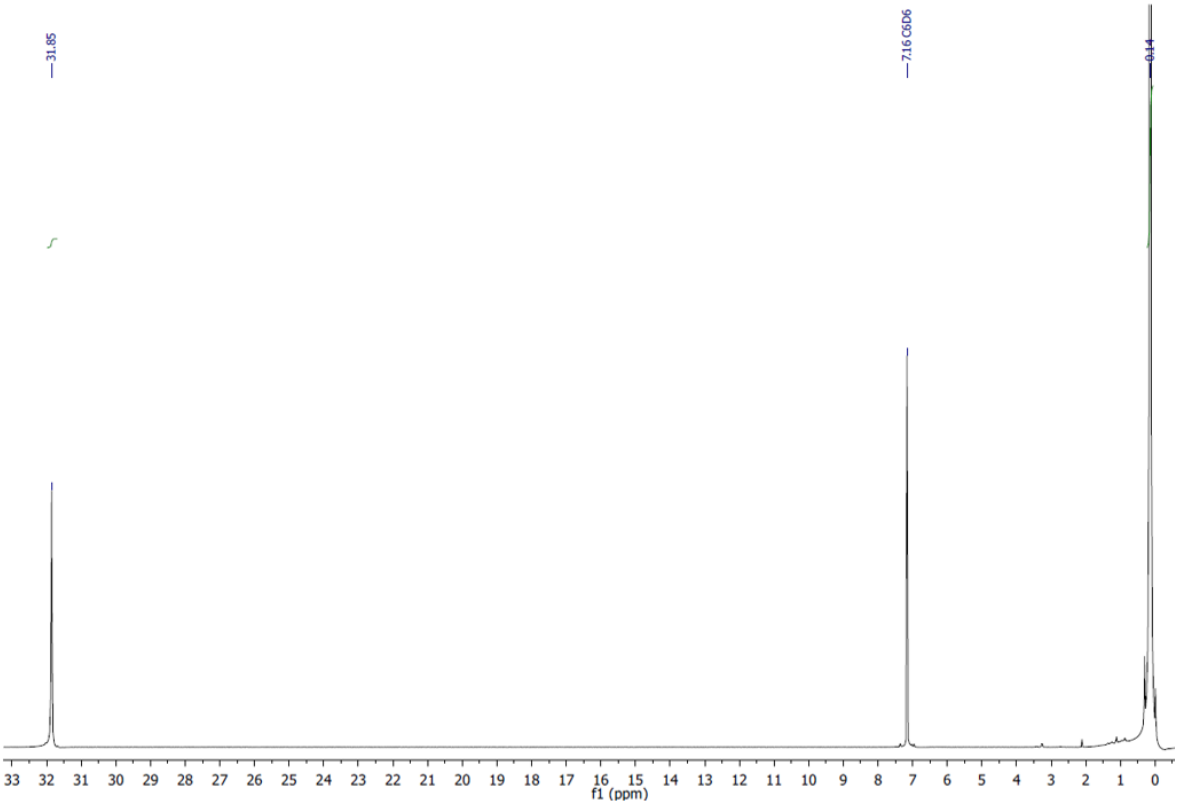

Figure S1. ^{1}H NMR spectrum of **1** in C_6D_6 (25 °C).

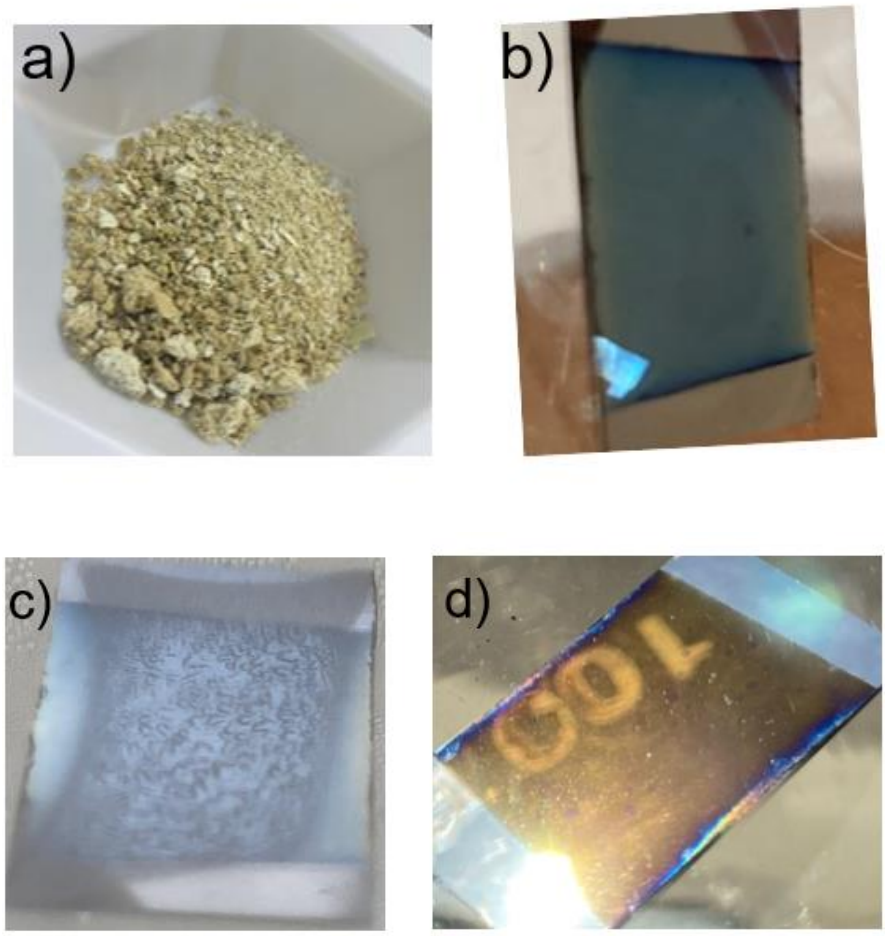

Figure S2. a) Compound **1** as a solid. b) Thin film of **1** deposited on silicon. c) Condensation of water on the film. d) Resulting UO_2-$\mathbf{1}^{\mathbf{film}}$ from the hydrolysis of **1**.

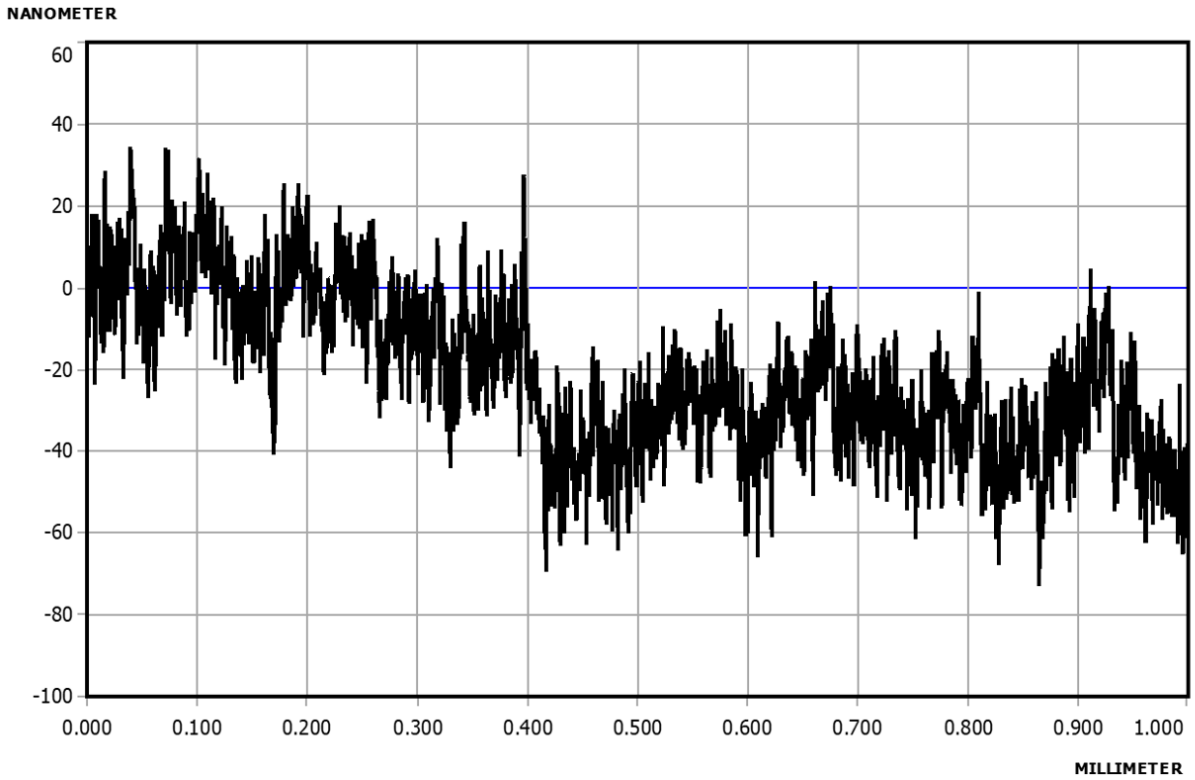

Figure S3. Step-down profile of thin film of UO_2-**1** grown on glass substrate. Step down occurs at 0.4 mm into scan.

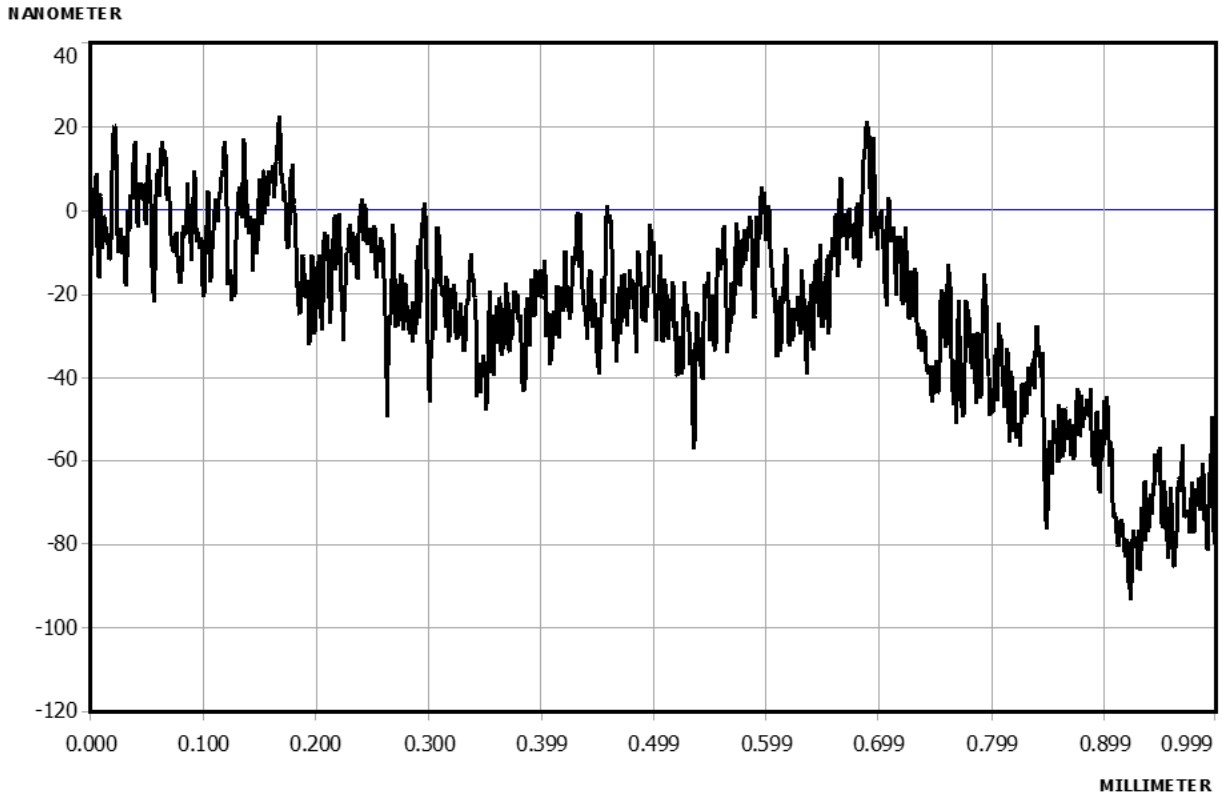

Figure S4. Step-down profile of thin film of UO_2-**1** grown on silicon substrate. Step down occurs at 0.7 mm into scan.

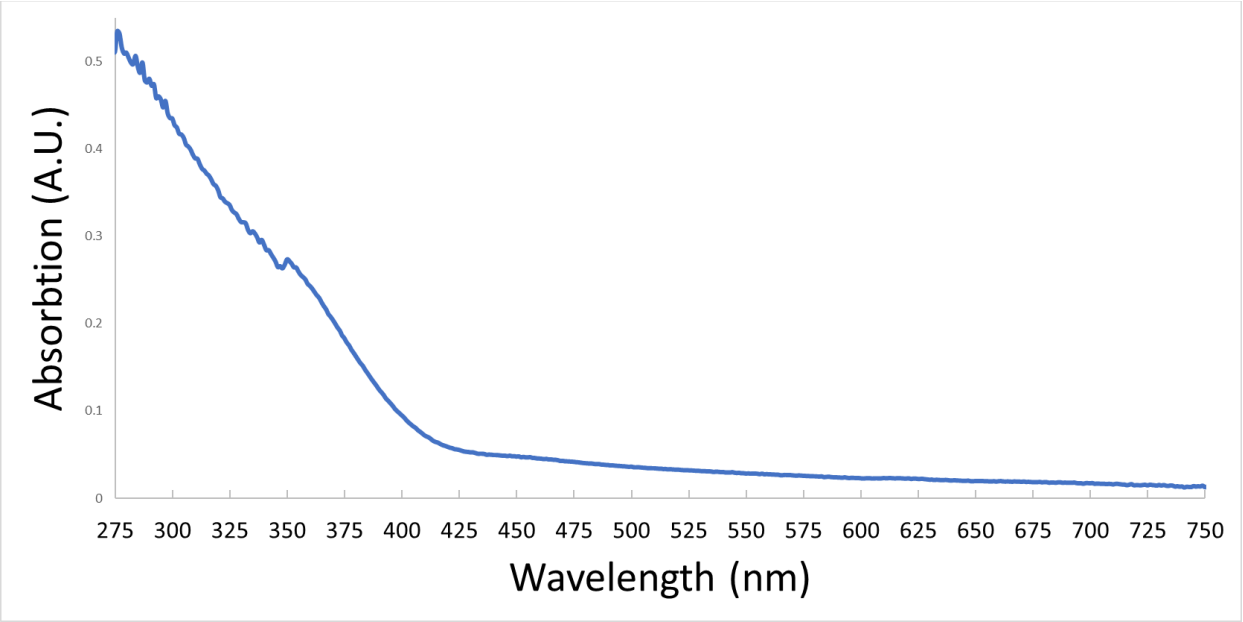

Figure S5. Room temperature electronic absorption spectra of UO_2-**1**$^{\mathbf{film}}$ formed on glass substrate**.**

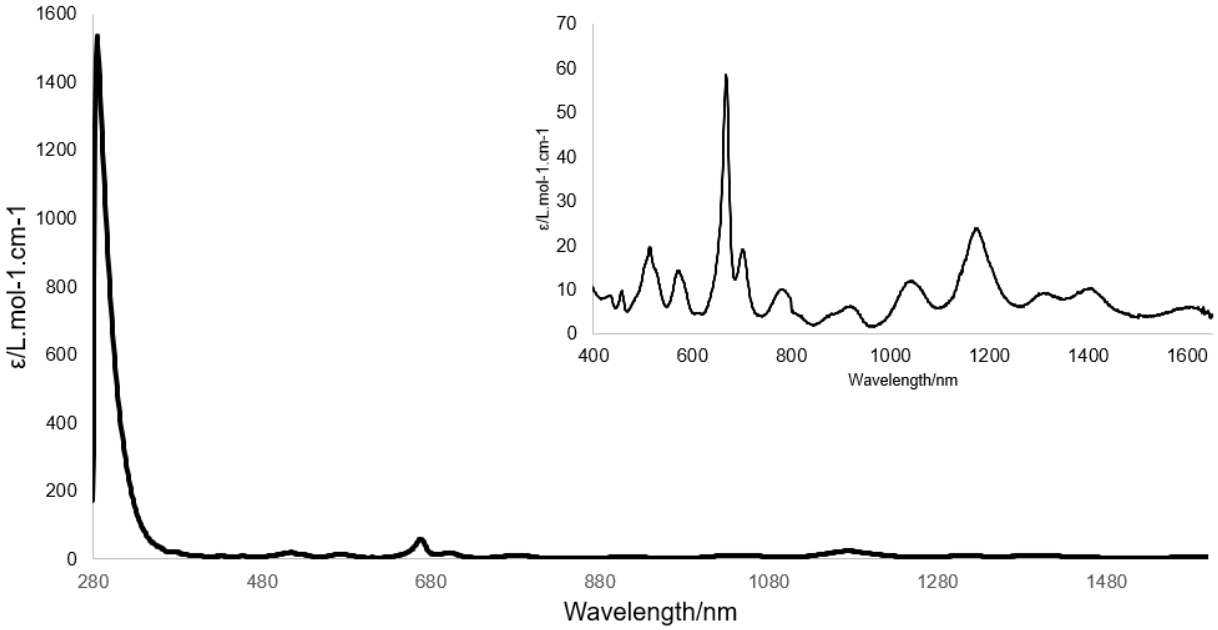

Figure S6. Room temperature electronic absorption spectra of **1**. Inset shows featured 400-1600nm in higher magnification. 1.60 mM solution in toluene.

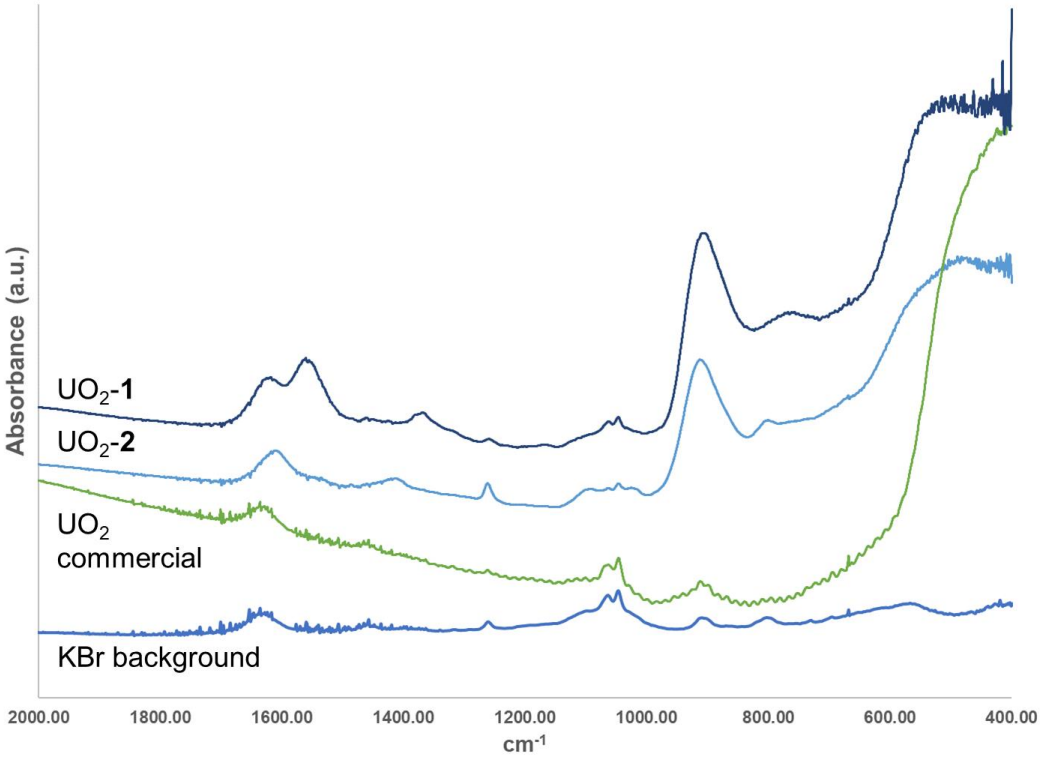

Figure S7. FT-IR spectra of UO_2-**1**, UO_2-**2**, commercial source UO_2 (KBr pellets) and the KBr background.

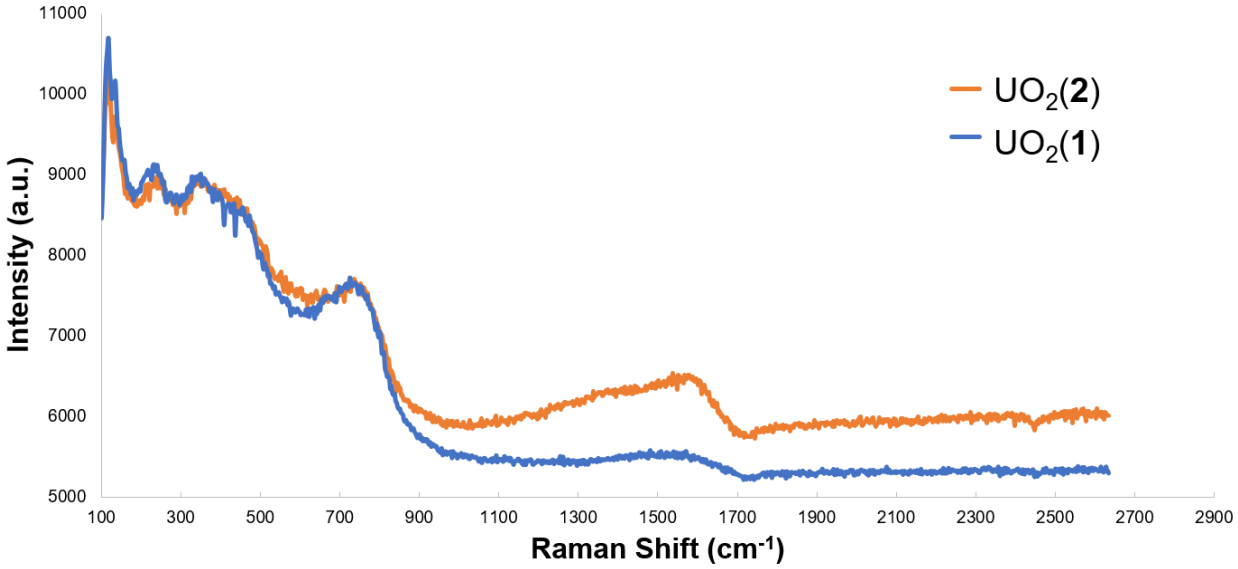

Figure S8. Raman spectra of UO_2-**1** and UO_2-**2** powder samples.

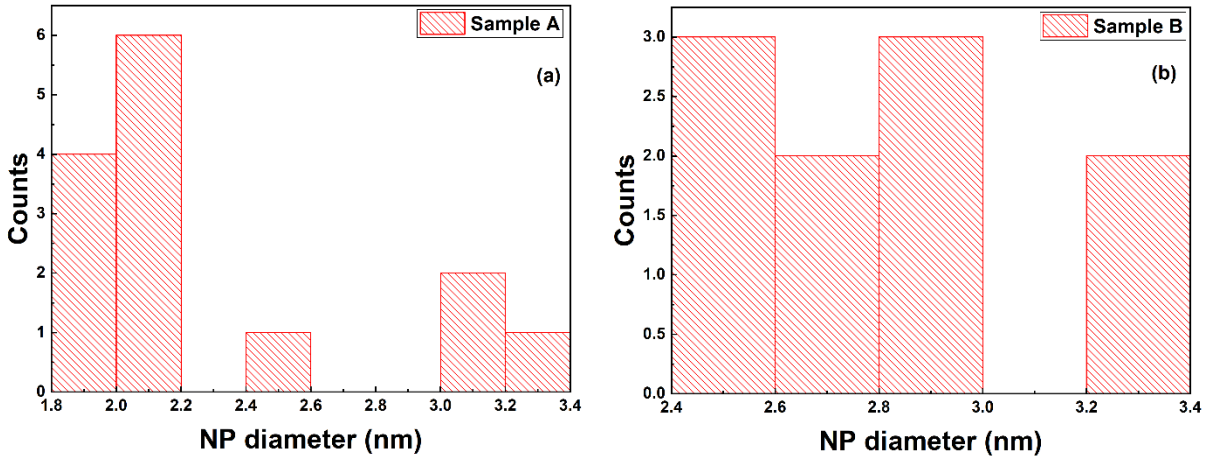

Figure S9. ImageJ generated histogram of NP size distribution in UO_2-**1** (A) and UO_2-**2** (B). It can be observed from the histrograms that larger NPs are formed in case of sample UO_2-**2**, which is due to the coalescence of smaller NPs. However, discrete NPs are formed in case of UO_2-**1**, and the size of most of the NPs is below 2.2 nm.