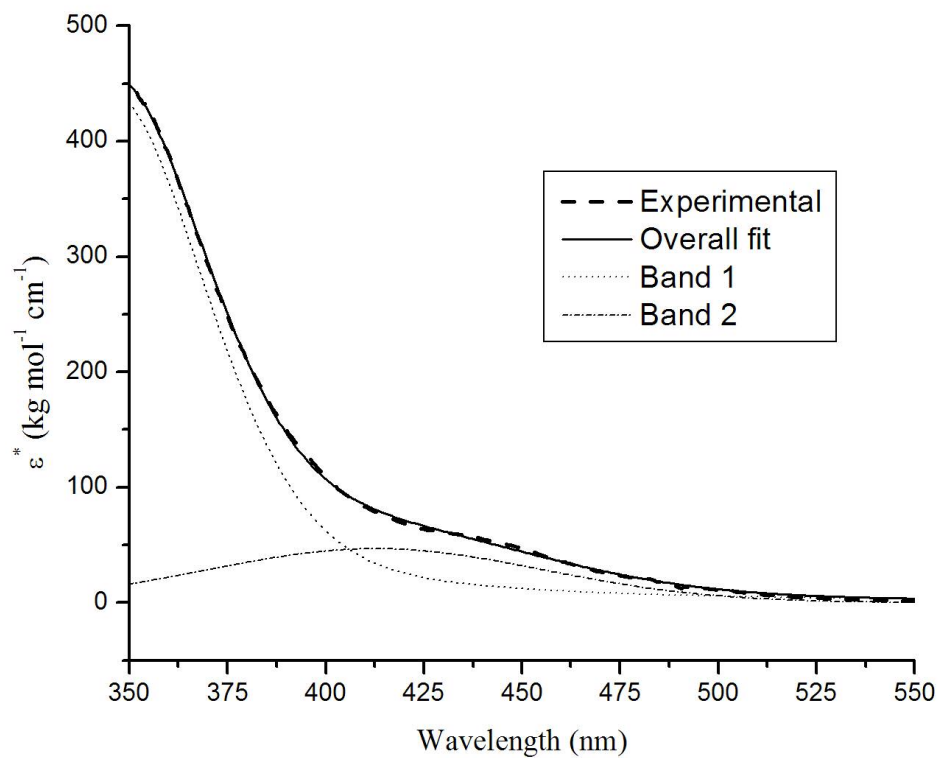
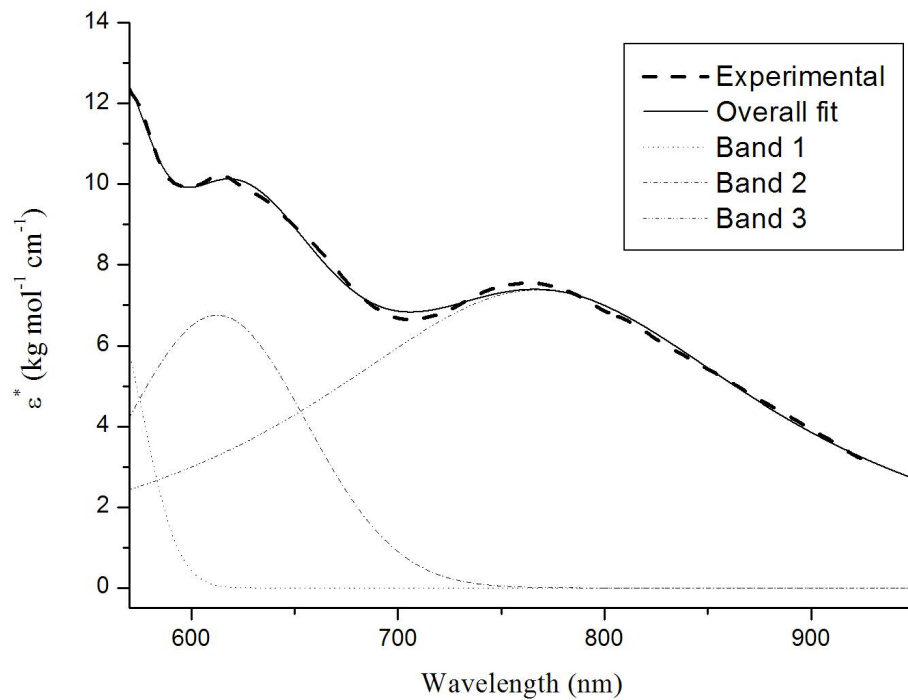


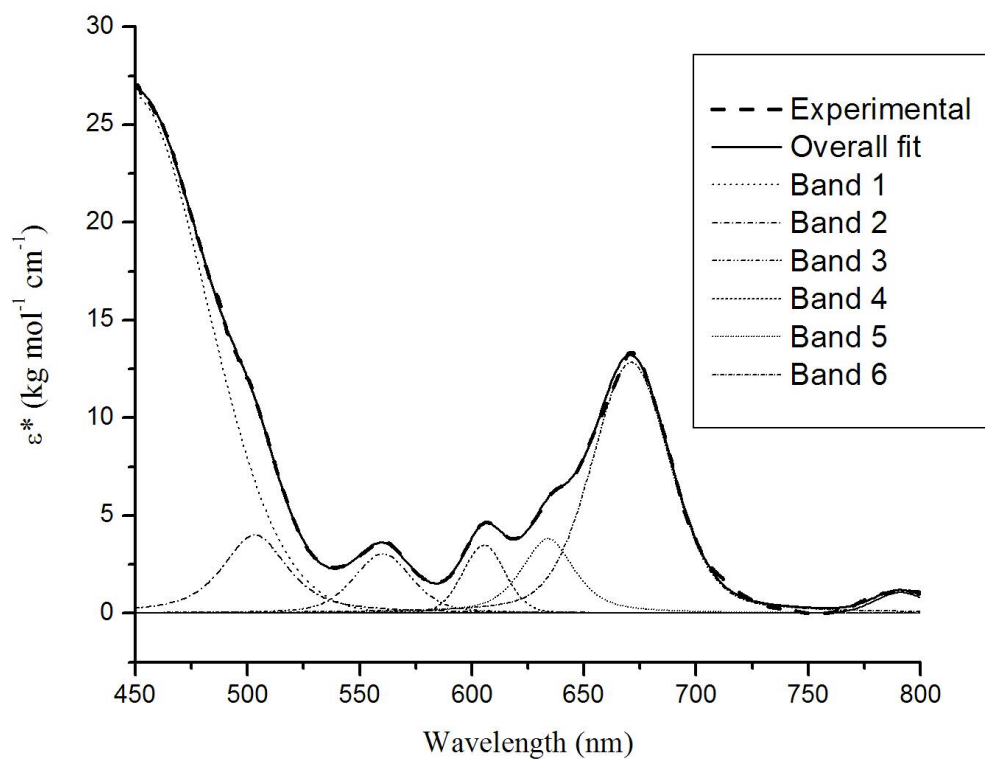
## Supporting Information



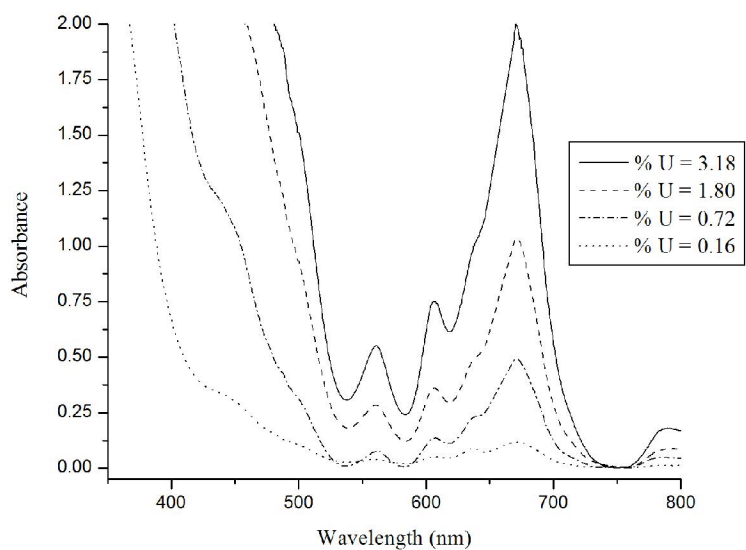
**SI 1.** Electronic absorption spectrum of  $\text{UO}_3$  exposed to HCl in LiCl-KCl at  $450^\circ\text{C}$  – *i.e.*  $\{\text{UO}_2\}^{2+}$  - with fitted peaks.



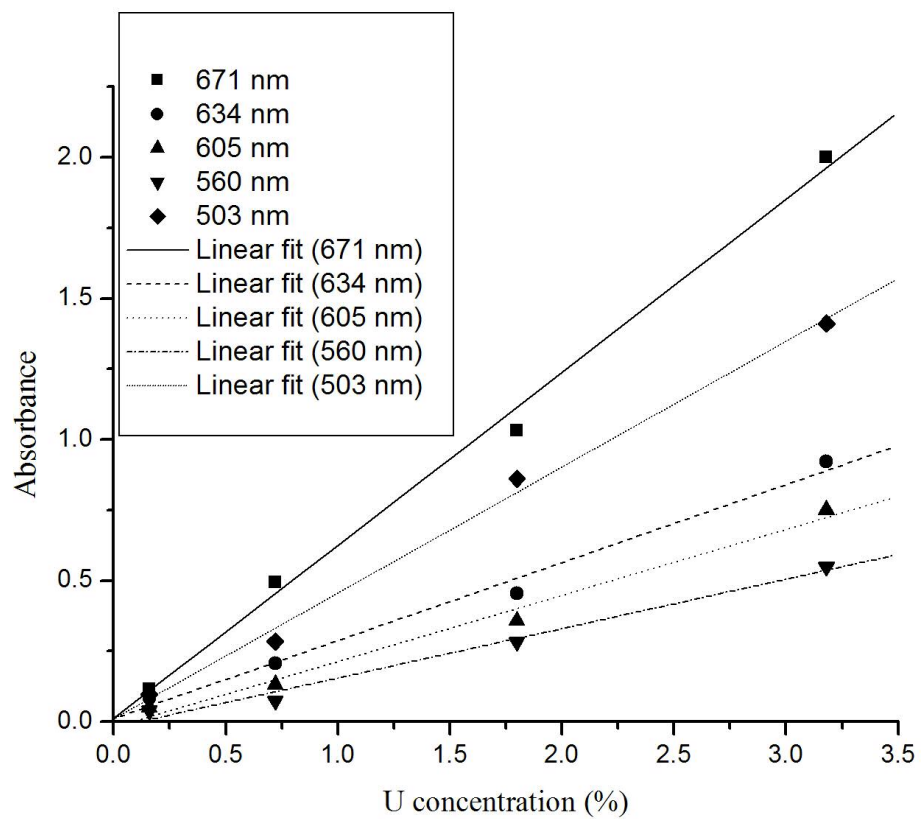
**SI 2.** Electronic absorption spectrum of  $\text{UO}_2$  exposed to HCl in LiCl at 750 °C - *i.e.*  $\{\text{UO}_2\}^+$  - with fitted peaks.



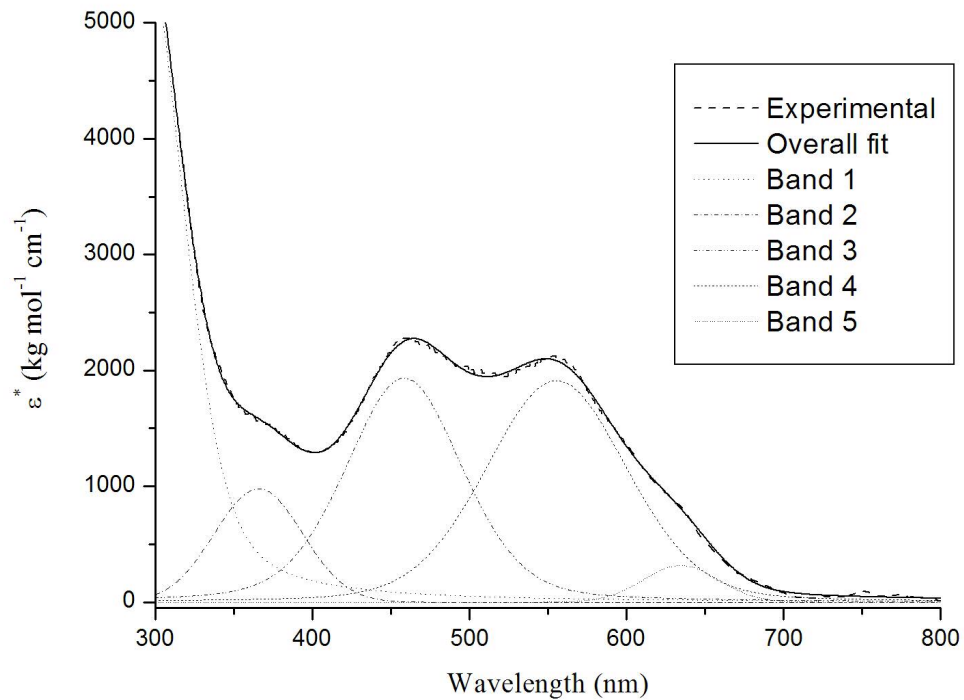
**SI 3** Electronic absorption spectrum of  $\text{UO}_2$  exposed to HCl in LiCl-KCl at 450 °C - *i.e.* U(IV) - with fitted peaks.



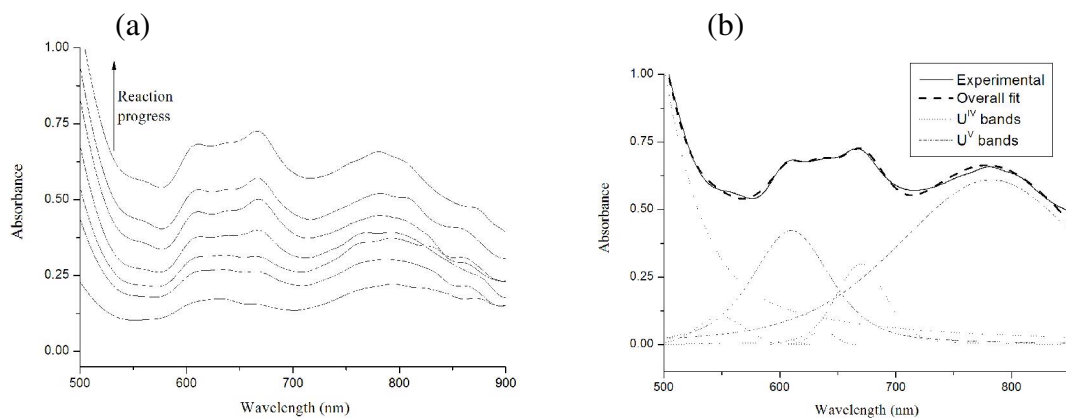
**SI 4.** Electronic absorption spectra of  $\text{UO}_2$  exposed to  $\text{HCl}$  in  $\text{LiCl-KCl}$  at  $450^\circ\text{C}$  - *i.e.*  $\text{U(IV)}$  - at various uranium concentrations.



**SI 5.** Plot of intensities of absorption maxima from the electronic absorption spectra of the reaction of  $\text{UO}_2$  with  $\text{HCl}$  in  $\text{LiCl-KCl}$  at  $450^\circ\text{C}$  with respect to uranium concentration



**SI 6** Electronic absorption spectrum from the reaction of U metal with  $\text{UCl}_4$  in LiCl-KCl eutectic at 450 °C – *i.e.* U(III) - with fitted peaks.



**SI 7.** Electronic absorption spectra monitoring the reaction progress of  $UO_2$  with HCl in LiCl-KCl eutectic at 750 °C (a), and the final spectrum with fitted peaks (b).