

## Supporting Information

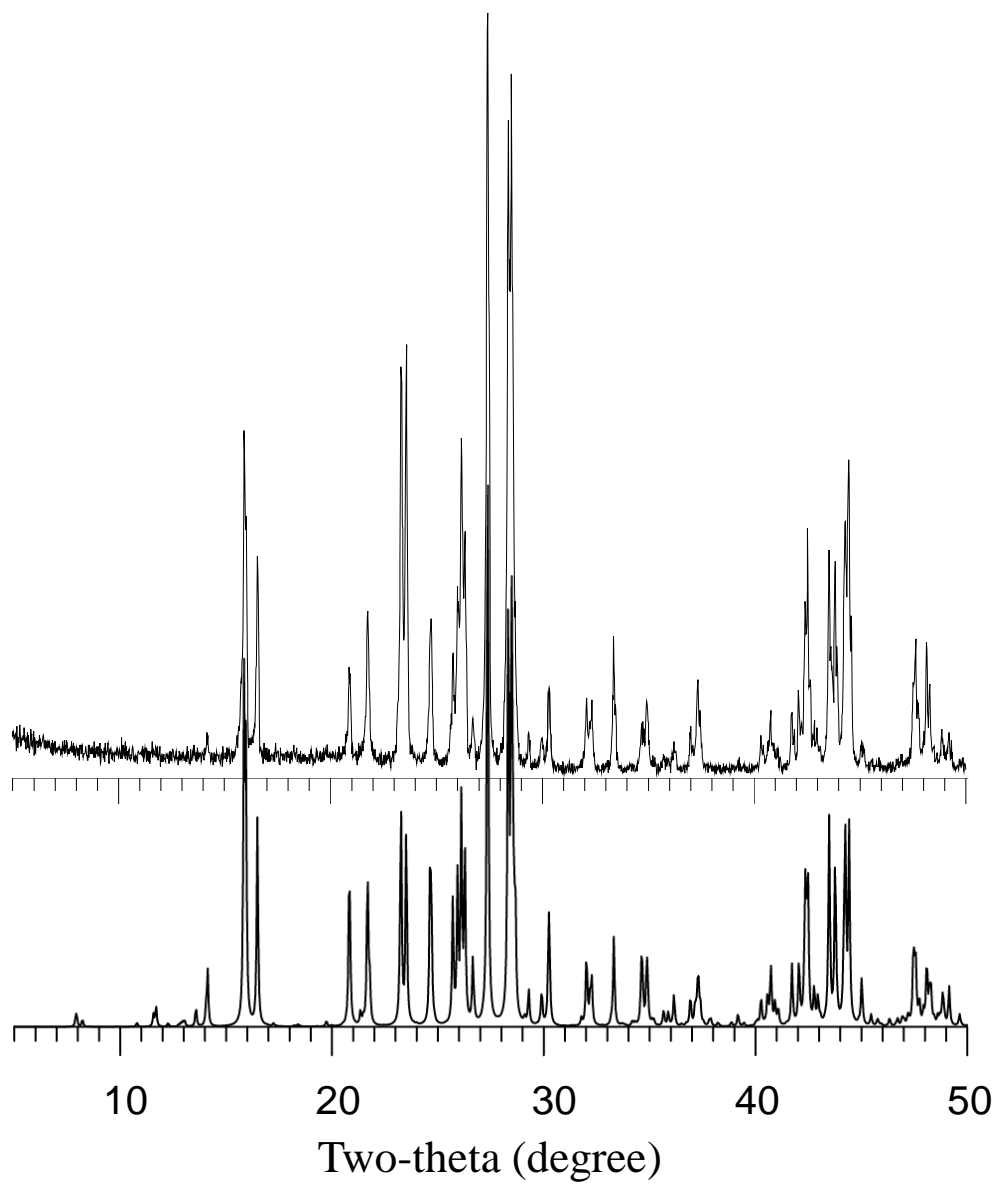
High-Temperature, High-Pressure Hydrothermal Synthesis, Crystal Structure, and Solid-State NMR Spectroscopy of  $\text{Cs}_2(\text{UO}_2)(\text{Si}_2\text{O}_6)$  and Variable-Temperature Powder X-ray Diffraction Study of the Hydrate Phase  $\text{Cs}_2(\text{UO}_2)(\text{Si}_2\text{O}_6)\cdot 0.5\text{H}_2\text{O}$

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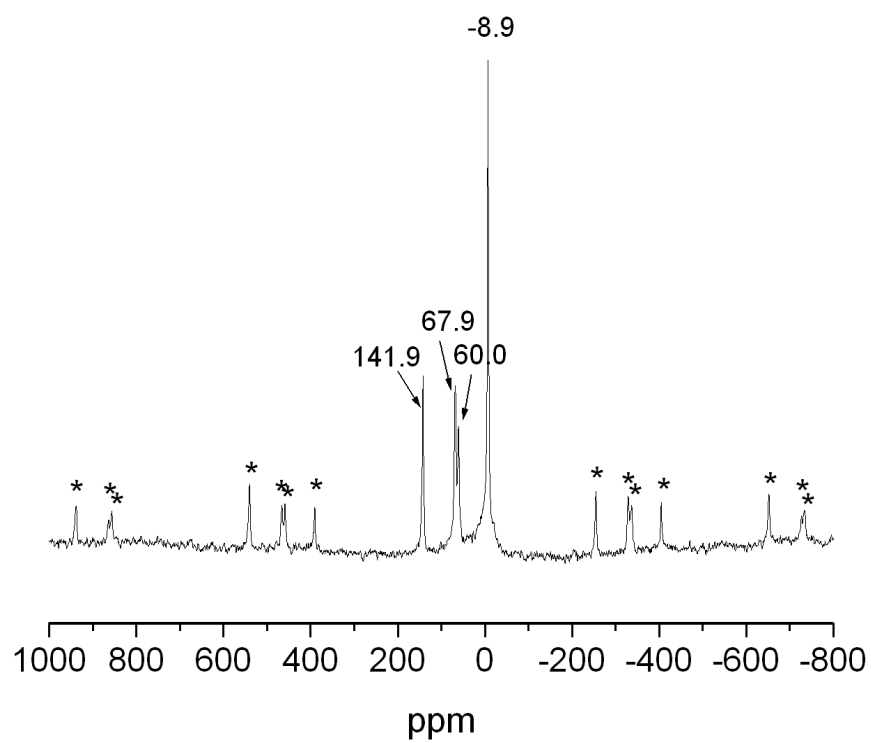
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**Figure S1.** Experimental X-ray powder pattern (top) and simulated powder pattern (bottom) based on the results from single-crystal X-ray diffraction for  $\text{Cs}_2(\text{UO}_2)(\text{Si}_2\text{O}_6)$ .

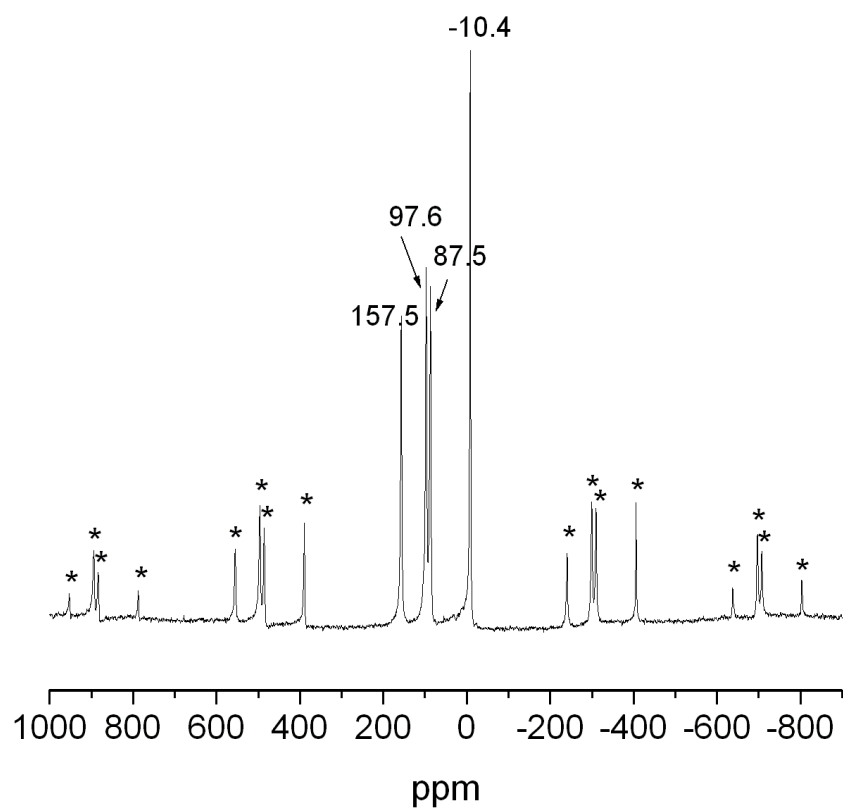
**Figure S2.**  $^{133}\text{Cs}$  MAS NMR spectra of  $\text{Cs}_2(\text{UO}_2)(\text{Si}_2\text{O}_6)$  (a) and  $\text{Cs}_2(\text{UO}_2)(\text{Si}_2\text{O}_6)\cdot 0.5\text{H}_2\text{O}$  (b) acquired on a NMR spectrometer of 11.7 T at a spinning speed of 26 kHz. The isotropic peaks are labeled. Asterisks denote spinning sidebands.



**Figure S1**



**Figure S2(a)**



**Figure S2(b)**