Supplementary Information for

Formation of stabilized vaterite nanoparticles via the introduction of uranyl into groundwater

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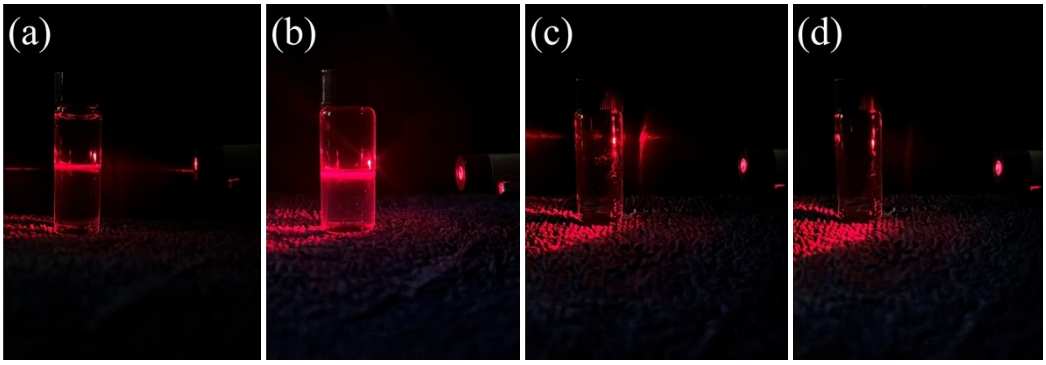
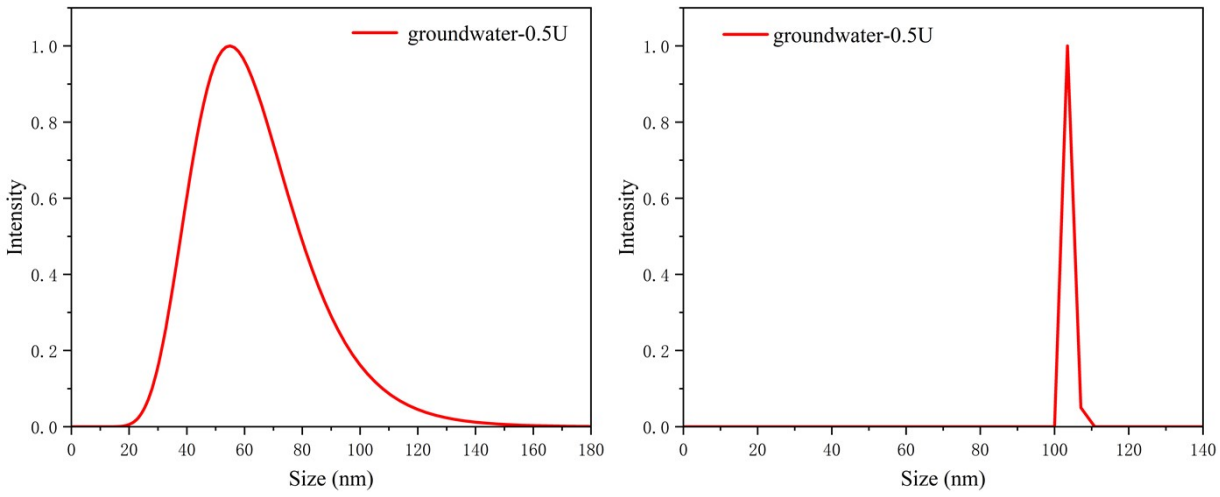


Fig. S1 Tyndall phenomena of the mixture of the Beishan groundwater and uranyl nitrate: (a) freshly prepared; (b) stored for one year at room temperature. The controlled sample of groundwater (c) and uranyl nitrate solution (d) under red laser.



(a) Log-normal (b) Multimodal

Fig. S2. The intensity-weighted log-normal (a) and multimodal (b) size distributions of hydrodynamic size.

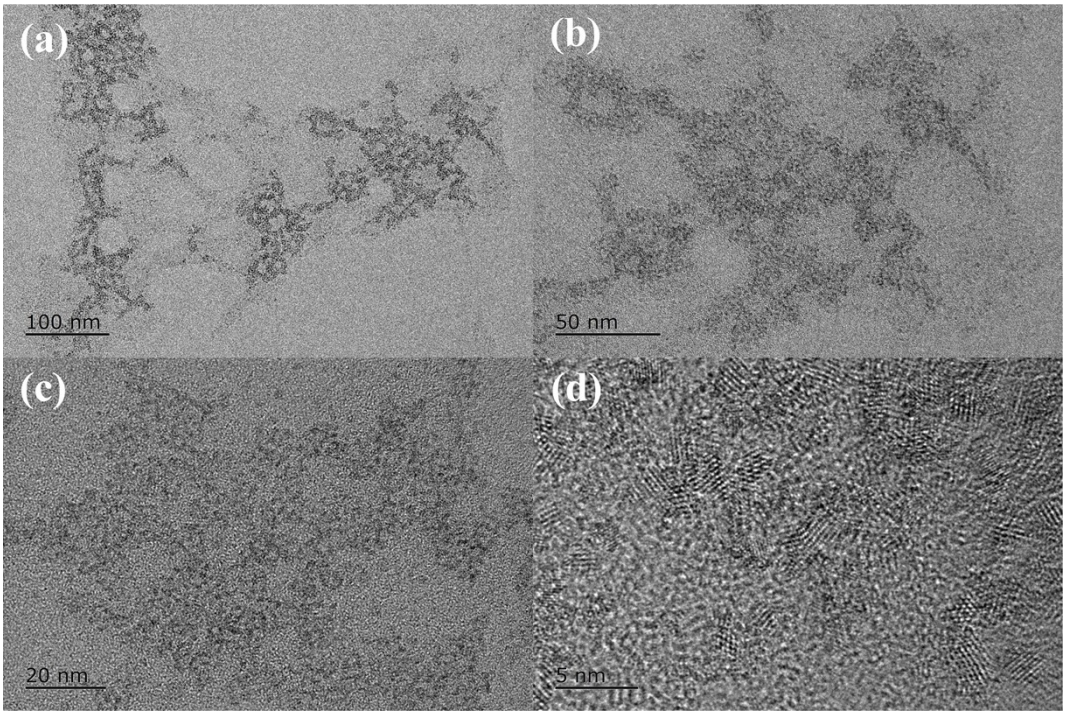


Fig. S3 TEM images obtained from the groundwater-0.5 U

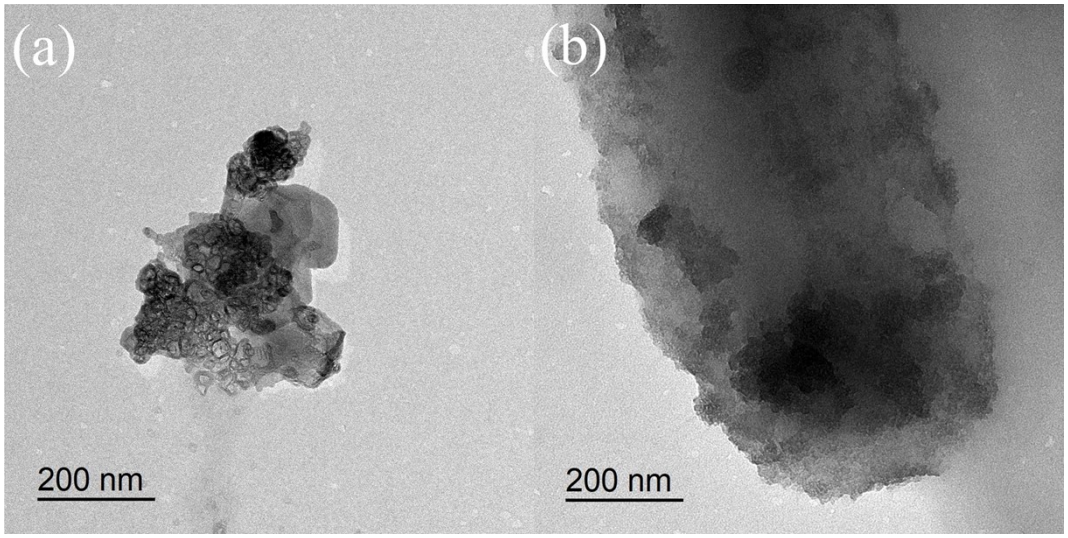


Fig. S4 TEM images obtained from the pure Beishan groundwater.

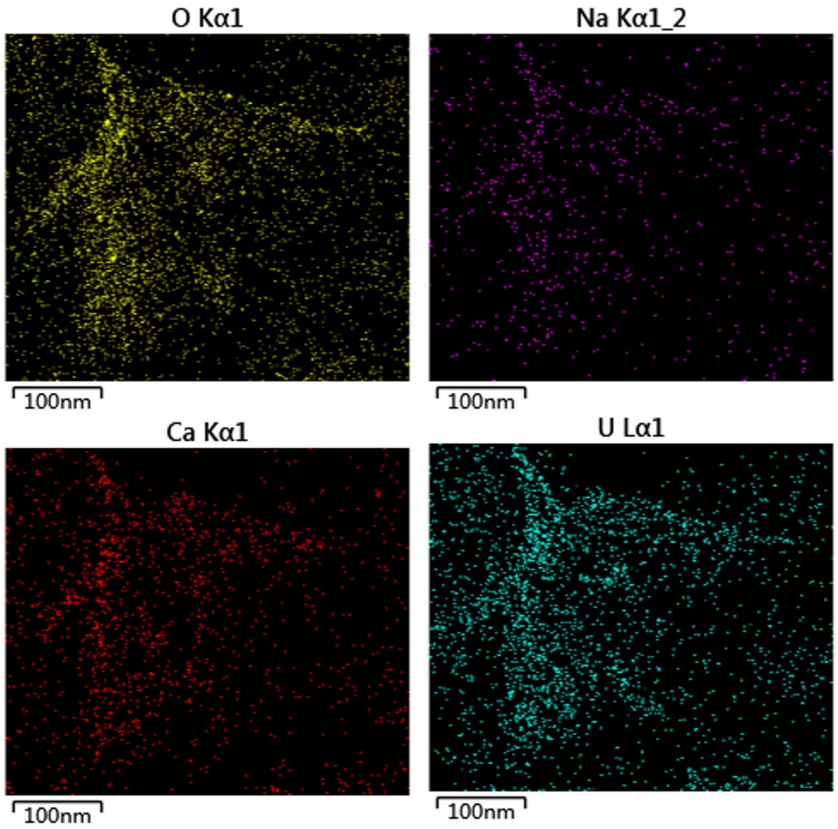
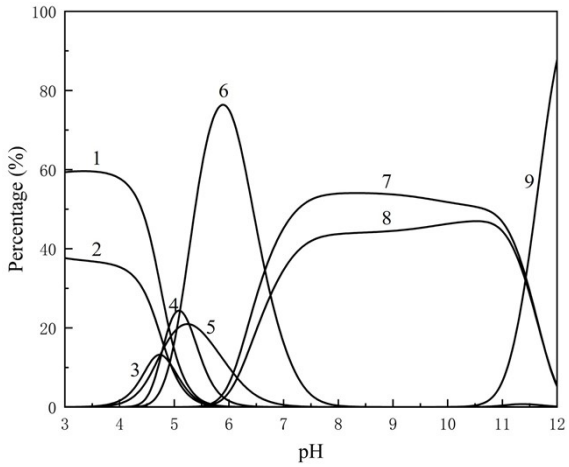


Fig. S5 TEM element mapping images of O, Na, Ca, and U obtained from groundwager-3U.



1——$UO_2SO_4(aq)$；2——UO_2^{2+}；3——$(UO_2)_2(OH)_2^{2+}$；4——$(UO_2)_3(OH)_5^{+}$；5——$UO_2CO_3(aq)$；
6——$(UO_2)_2CO_3(OH)_3^{-}$；7——$Ca_2UO_2(CO_3)_3(aq)$；8——$CaUO_2(CO_3)_3^{2-}$；9——$UO_2(OH)_3^{-}$

Fig. S6 Influence of pH on the distribution of uranium species (>5%) in groundwater-0.5 U ([U] = 0.5 mmol/L, T = 25 °C).

Table S1 Thermodynamic data of uranium at 25 ℃.

Chemical reactions	log K
$UO_2^{2+} + H_2O = UO_2OH^+ + H^+$	-5.25
$UO_2^{2+} + 3H_2O = UO_2(OH)_3^- + 3H^+$	-20.25
$UO_2^{2+} + 2OH^- = UO_2(OH)_2$	-10.31
$2UO_2^{2+} + H_2O = (UO_2)_2OH^{3+} + H^+$	-2.70
$2UO_2^{2+} + 2H_2O = (UO_2)_2(OH)_2^{2+} + 2H^+$	-5.62
$3UO_2^{2+} + 4H_2O = (UO_2)_3(OH)_4^{2+} + 4H^+$	-11.90
$3UO_2^{2+} + 5H_2O = (UO_2)_3(OH)_5^+ + 5H^+$	-15.55
$3UO_2^{2+} + 7H_2O = (UO_2)_3(OH)_7^- + 7H^+$	-32.20
$4UO_2^{2+} + 7H_2O = (UO_2)_4(OH)_7^+ + 7H^+$	-21.90
$UO_2^{2+} + SO_4^{2-} = UO_2SO_4$	3.15
$UO_2^{2+} + 2SO_4^{2-} = UO_2(SO_4)_2^{2-}$	4.14
$UO_2^{2+} + Cl^- = UO_2Cl^+$	0.17
$UO_2^{2+} + Cl^- = UO_2Cl_2$	-1.10
$UO_2^{2+} + CO_3^{2-} = UO_2CO_3$	9.94
$UO_2^{2+} + 2CO_3^{2-} = UO_2(CO_3)_2^{2-}$	16.61
$UO_2^{2+} + 3CO_3^{2-} = UO_2(CO_3)_3^{4-}$	21.84
$2UO_2^{2+} + 3H_2O + CO_3^{2-} = (UO_2)_2CO_3(OH)_3^- + 3H^+$	-0.86
$Ca^{2+} + UO_2(CO_3)_3^{4-} = CaUO_2(CO_3)_3^{2-}$	5.33*
$2Ca^{2+} + UO_2(CO_3)_3^{4-} = Ca_2UO_2(CO_3)_3$	8.70*
$Mg^{2+} + UO_2(CO_3)_3^{4-} = MgUO_2(CO_3)_3^{2-}$	2.61*

*Jo Y, Kirishima A, Kimuro S, et al. Formation of $CaUO_2(CO_3)_3^{2-}$ and $Ca_2UO_2(CO_3)_3$(aq) complexes at variable temperatures (10 – 70 ℃). Dalton Transactions, 2019, 48(20): 6942–6950.

* Jo Y, Kim H K, Yun J I. Complexation of $UO_2(CO_3)_3^{4-}$with Mg^{2+} at varying temperatures and its effect on U(VI) speciation in groundwater and seawater. Dalton Transactions, 2019，48(39): 14769–14776.

Table S2 *d*-spacings derived from the SAED and HRTEM image of groundwater-3U.

JCPDS 87-1714 Clarkeite [$Na(UO_2)O(OH)$]		SAED	HRTEM
hkl	*d* (Å)	*d* (Å)	*d* (Å)
003	5.89		5.86
012	3.19	3.20	
104	2.71	2.71	
113	1.87	1.92	
116	1.64	1.61	

Table S3 The possible precipitated minerals with SI > 0 from the Beishan groundwater simulated by PHREEQC ([U] = 0.5 mmol/L, T = 25 ℃, pH = 7.0).

Phase	Chemical formula	Log K	Log IAP	SI
Andersonite	$Na_2CaUO_2(CO_3)_3 \cdot 6H_2O$	-37.50	-34.47	3.03
Bayelite	$Mg_2UO_2(CO_3)_3 \cdot 18H_2O$	-36.60	-34.93	1.67
Becquerelite	$Ca(UO_2)_6O_4(OH)_6 \cdot 8H_2O$	40.5	40.88	0.38
Clarkeite	$Na(UO_2)O(OH)$	9.02	10.23	1.21
Calcite	$CaCO_3$	1.85	1.97	0.13
Liebigite	$Ca_2UO_2(CO_3)_3 \cdot 10H_2O$	-37.90	-33.92	3.98
Magnesite	MgCO3	-8.91	-8.80	0.11