

## **Supplementary Materials**

**for**

### **The Stability of U(VI) and As(V) under the Influence of pH and Inorganic Ligands**

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## 1. The impact of pH on soluble fraction of U(VI) and As(V)

The impact of pH on soluble fractions of U(VI) and As(V), in co-existing conditions, were plotted for three experimental groups, which are: (1) the air-equilibrated group, (2) the bicarbonate group, and (3) the Ca-bicarbonate group.

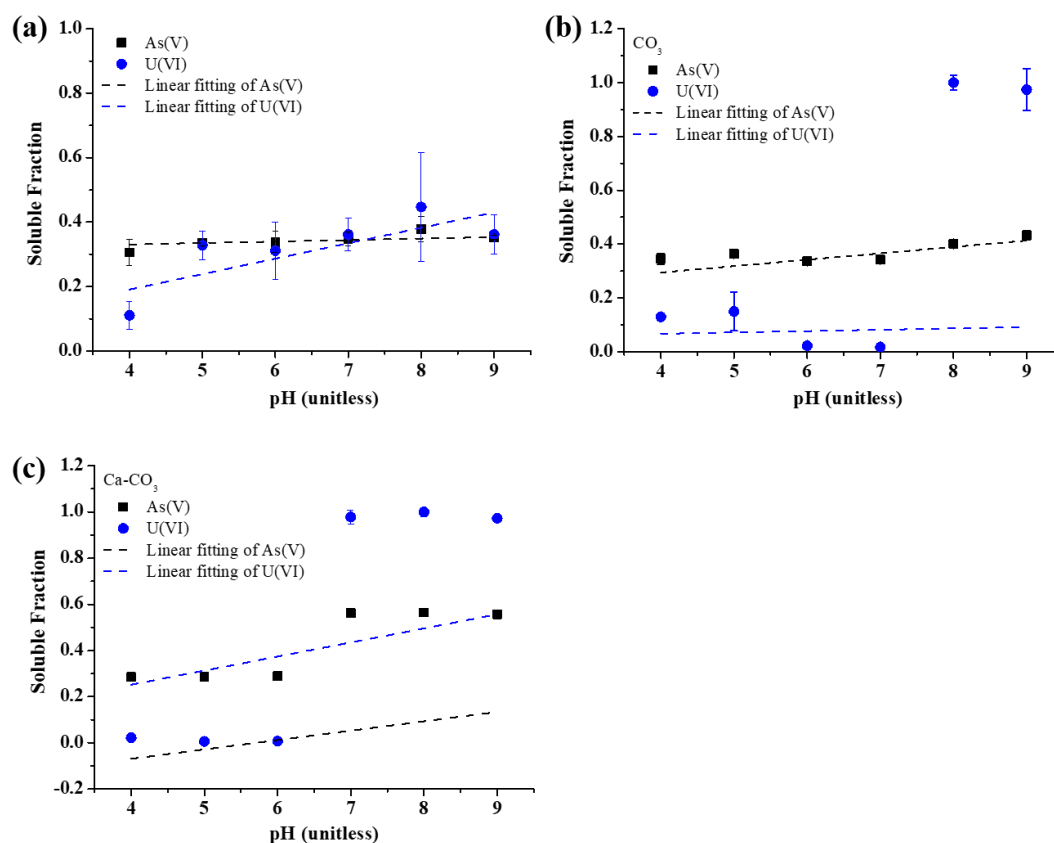


Figure S1. The change of soluble U(VI) and As(V) fractions in co-existing conditions as a function of pH. (a) the air-equilibrated group, (b) the bicarbonate group, (c) the Ca-bicarbonate group.

Table S1. The fitting results of soluble U(VI) and As(V) fractions as function of pH

Condition	Species	Slope	Adjusted R <sup>2</sup> value
Air-equilibrated	U(VI)	0.047	0.76
	As(V)	0.004	0.83
Bicarbonate	U(VI)	0.004	0.83
	As(V)	0.023	0.03
Ca-bicarbonate	U(VI)	0.008	0.316
	As(V)	0.040	0.940

## 2. The speciation of soluble U(VI) and As(V)

The soluble speciation of U(VI) and As(V) were calculated using PHREEQC using LLNL database file. Detailed information regarding the calculation were given in the materials and methods section. The soluble species considered for As included:  $\text{AsO}_4^{3-}$ ,  $\text{HAsO}_4^{2-}$ ,  $\text{H}_2\text{AsO}_4^{3-}$  and  $\text{H}_3\text{AsO}_4$ . On the other hand, for U(VI), the species that accounted for more than 0.1% of total U(VI) at each pH values were considered. It should be noted that, the soluble species of U(VI) were calculated under the presence of  $\text{Ca}^{2+}$  and bicarbonate, which is a scenario that included all the possible soluble U(VI) species in our experimental settings.

The speciation of soluble U(VI) and As(V) were tabulated in tables S2 and S3.

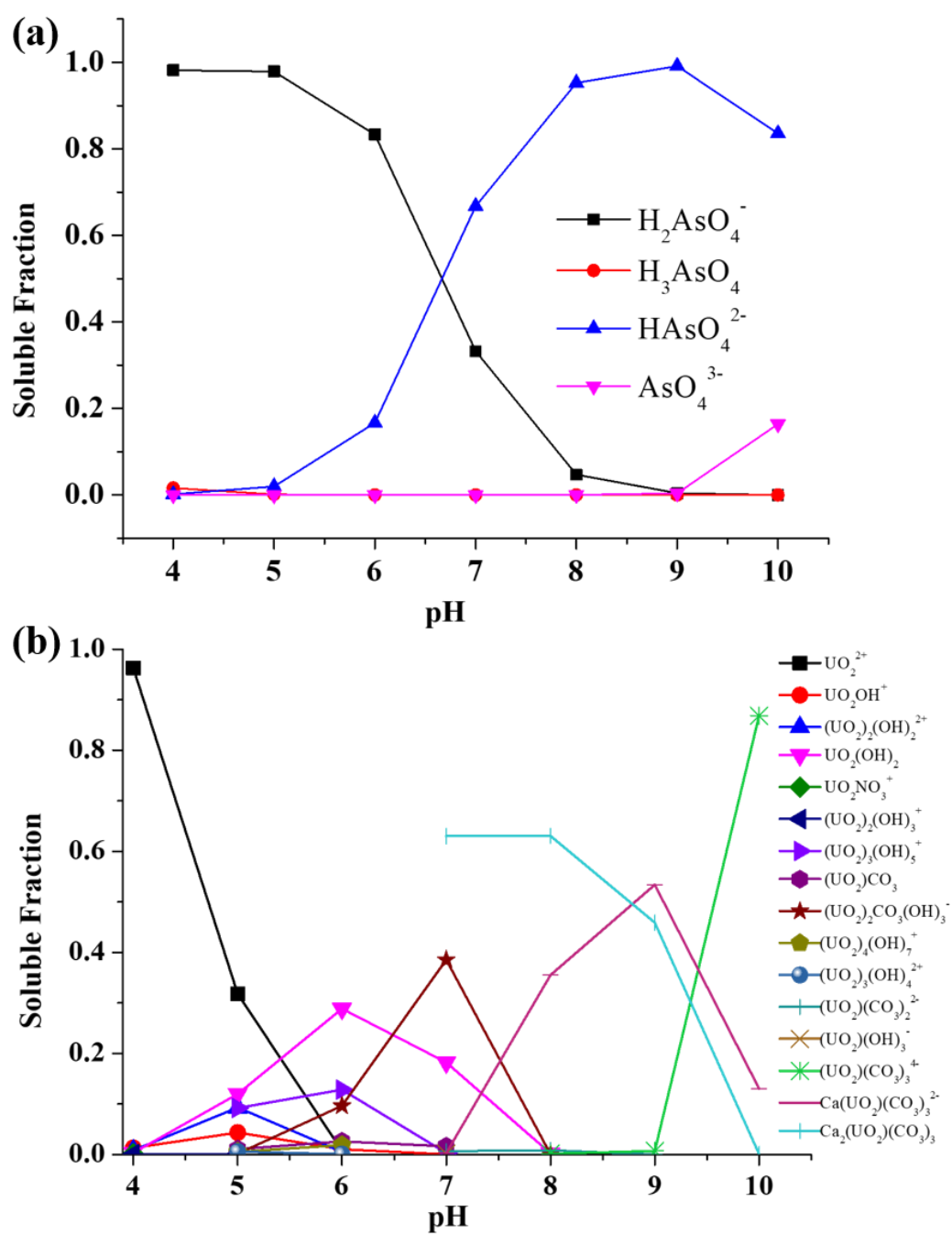


Figure S2. The soluble species of As (a) and U (b) in the pH range studied

Table S2. The soluble fraction of As(V) in the studied pH range

<b>pH</b>	<b>H<sub>2</sub>AsO<sub>4</sub><sup>-</sup></b>	<b>H<sub>3</sub>AsO<sub>4</sub></b>	<b>HAsO<sub>4</sub><sup>2-</sup></b>	<b>AsO<sub>4</sub><sup>3-</sup></b>
4	0.9818	0.01623	0.00197	6.96E-11
5	0.9788	0.00162	0.01962	6.89E-09
6	0.833	1.38E-04	0.1669	5.86E-07
7	0.3322	5.49E-06	0.6677	2.36E-05
8	0.04703	7.76E-08	0.9526	3.41E-04
9	0.00447	7.17E-10	0.9914	0.00411
10	1.48E-04	1.81E-12	0.8357	0.1642

Table S3. The soluble fraction of U(VI) in the studied pH range

pH	Species							
	$\text{UO}_2^{2+}$	$\text{UO}_2\text{OH}^+$	$(\text{UO}_2)_2(\text{OH})_2^{2+}$	$\text{UO}_2(\text{OH})_2$	$\text{UO}_2\text{NO}_3^+$	$(\text{UO}_2)_2\text{OH}^{3+}$	$(\text{UO}_2)_3(\text{OH})_5^+$	$\text{UO}_2\text{CO}_3$
4	0.9624	0.01314	0.0085	0.00362	0.00142	9.60E-04	/	/
5	0.3178	0.04352	0.09302	0.1199	/	0.00105	0.09212	0.01085
6	0.00765	0.01047	0.00539	0.2886	/	/	0.12848	0.02612
7	/	6.62E-04	/	0.18234	/	/	0.00324	0.0165
8	/	/	/	0.0023	/	/	/	/
9	/	/	/	/	/	/	/	/
10	/	/	/	/	/	/	/	/
pH	Species							
	$(\text{UO}_2)_2\text{CO}_3(\text{OH})_3^-$	$(\text{UO}_2)_4(\text{OH})_7^+$	$(\text{UO}_2)_3(\text{OH})_4^{2+}$	$\text{UO}_2(\text{CO}_3)_2^{2-}$	$\text{UO}_2(\text{OH})_3^-$	$\text{UO}_2(\text{CO}_3)_3^{4-}$	$\text{Ca}(\text{UO}_2)(\text{CO}_3)_3^{2-}$	$\text{Ca}_2(\text{UO}_2)(\text{CO}_3)_3$
4	/	/	/	/	/	/	/	/
5	0.00167	0.00558	0.00572	/	/	/	/	/
6	0.09646	0.01874	7.97E-04	/	/	/	/	/
7	0.3854	/	/	0.0066	0.00241	/	0.0031	0.63
8	6.15E-04	/	/	0.00841	/	0.00185	0.3552	0.6304
9	/	/	/	2.32E-04	/	0.00727	0.5338	0.4588
10	/	/	/	/	/	0.8682	0.13066	0.00121

Note: For simplicity, only the species whose soluble fraction were larger than  $1 \times 10^{-4}$  were tabulated; “/” means that the fraction of the corresponding specie is lower than  $1 \times 10^{-4}$ .