Supplement Data

Optimization of Copper-Ammonia-Sulfate Electrolyte for Maximizing Cu(I):Cu(II) Ratio Using pH and Copper Solubility

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Table S1. Repeatability experiments for Cu(II) Solubility (Experiment 1).

$(NH_4)_2SO_4$	NH_4OH	pH	Eh	Cu(II) Concentration
M	**M**		**mV**	**M**
0.625	4.0	7.71	273	0.483
0.625	4.0	7.82	284	0.489
0.625	4.0	7.84	276	0.486

Table S2. Repeatability experiments for Cu(I):Cu(II) ratio (Experiment 2).

Cu	$CuSO_4.5H_2O$	$(NH_4)_2SO_4$	NH_4OH	pH	Eh	Cu(I)	Cu(II)
M	M	M	M		mV	M	M
0.3541	0.3541	0.625	2.5	8.51	-104	0.563	0.187
0.3541	0.3541	0.625	2.5	8.46	-108	0.532	0.176
0.3541	0.3541	0.625	2.5	8.41	-110	0.595	0.208

Table S3. Design of experiment (DOE) for Cu(II) solubility experiments (Experiment 1).

Std. Order	Run Order	Pt. Type	Blocks	$(NH_4)_2SO_4$	NH_4OH
				M	**M**
6	1	-1	1	1.000	2.5
2	2	1	1	1.000	1.0
9	3	0	1	0.625	2.5
3	4	1	1	0.250	4.0
8	5	-1	1	0.625	4.0
4	6	1	1	1.000	4.0
5	7	-1	1	0.250	2.5
7	8	-1	1	0.625	1.0
1	9	1	1	0.250	1.0

Table S4. Design of experiment (DOE) for low pH Cu(I):Cu(II) experiments (Experiment 2A).

Std. Order	Run Order	Pt. Type	Blocks	Cu	$CuSO_4.5H_2O$	$(NH_4)_2SO_4$	NH_4OH
				M	M	M	M
24	6	2	1	0.3541	0.6295	0.625	4
14	11	2	1	0.3541	0.6295	0.25	2.5
10	13	2	1	0.6295	0.3541	0.625	1
5	16	2	1	0.3541	0.3541	0.25	1
16	18	2	1	0.3541	0.6295	1	2.5
22	19	2	1	0.3541	0.6295	0.625	1
3	21	2	1	0.0787	0.6295	0.625	2.5
6	23	2	1	0.3541	0.3541	1	1
4	25	2	1	0.6295	0.6295	0.625	2.5
9	27	2	1	0.0787	0.3541	0.625	1

Table S5. Design of experiment (DOE) for high pH Cu(I):Cu(II) experiments (Experiment 2B).

Std. Order	Run Order	Pt. Type	Blocks	Cu	$CuSO_4.5H_2O$	$(NH_4)_2SO_4$	NH_4OH
				M	M	M	M
13	2	2	1	0.3541	0.0787	0.25	2.5
18	3	2	1	0.6295	0.3541	0.25	2.5
12	4	2	1	0.6295	0.3541	0.625	4
23	5	2	1	0.3541	0.0787	0.625	4
8	8	2	1	0.3541	0.3541	1	4
21	9	2	1	0.3541	0.0787	0.625	1
20	10	2	1	0.6295	0.3541	1	2.5
7	14	2	1	0.3541	0.3541	0.25	4
25	17	0	1	0.3541	0.3541	0.625	2.5
15	20	2	1	0.3541	0.0787	1	2.5
2	22	2	1	0.6295	0.0787	0.625	2.5
26	24	0	1	0.3541	0.3541	0.625	2.5
27	26	0	1	0.3541	0.3541	0.625	2.5

Table S6. Scoping tests for constant ammonium hydroxide and varying ammonium sulfate.

Scoping Test # 01							
$CuSO_4.5H_2O$ (g/L)	20	30	40	50	60	70	80
NH_4OH (M)				2			
$(NH_4)_2SO_4$ (M)				0.25			
Complete Dissolution	YES	NO	NO	NO	NO	NO	NO
Scoping Test # 02							
$CuSO_4.5H_2O$ (g/L)	20	30	40	50	60	70	80
NH_4OH (M)				2			
$(NH_4)_2SO_4$ (M)				0.5			
Complete Dissolution	YES	NO	NO	NO	NO	NO	NO
Scoping Test # 03							
$CuSO_4.5H_2O$ (g/L)	20	30	40	50	60	70	80
NH_4OH (M)				2			
$(NH_4)_2SO_4$ (M)				0.75			
Complete Dissolution	NO	NO	NO	NO	NO	NO	NO
Scoping Test # 04							
$CuSO_4.5H_2O$ (g/L)	20	30	40	50	60	70	80

NH_4OH (M)				2			
$(NH_4)_2SO_4$ (M)				1			
Complete Dissolution	NO	NO	NO	NO	NO	NO	NO

Table S7. Scoping tests for constant ammonium sulfate and varying ammonium hydroxide.

				Scoping Test # 01			
$CuSO_4.5H_2O$ (g/L)	20	30	40	50	60	70	80
NH_4OH (M)				1			
$(NH_4)_2SO_4$ (M)				1			
Complete Dissolution	YES	NO	NO	NO	NO	NO	NO
				Scoping Test # 02			
$CuSO_4.5H_2O$ (g/L)	20	30	40	50	60	70	80
NH_4OH (M)				2			
$(NH_4)_2SO_4$ (M)				1			
Complete Dissolution	NO	NO	NO	NO	NO	NO	NO
				Scoping Test # 03			
$CuSO_4.5H_2O$ (g/L)	20	30	40	50	60	70	80
NH_4OH (M)				3			
$(NH_4)_2SO_4$ (M)				1			
Complete Dissolution	YES	YES	NO	NO	NO	NO	NO
				Scoping Test # 04			
$CuSO_4.5H_2O$ (g/L)	20	30	40	50	60	70	80
NH_4OH (M)				4			
$(NH_4)_2SO_4$ (M)				1			
Complete Dissolution	YES	YES	YES	NO	NO	NO	NO