

## Supporting information

### Effects of Cu(I) contents on voltammetric behavior and electrodeposition mechanism of bimetallic composite ionic liquids

Ping Ouyang<sup>a</sup>, Rui Zhang<sup>a</sup>, Jian Zhou<sup>a</sup>, Haiyan Liu<sup>a</sup>, Zhichang Liu<sup>a</sup>,  
Chunming Xu<sup>a</sup>, Shaojuan Zeng<sup>b</sup>, Qian Su<sup>b</sup>, Xiangping Zhang<sup>b</sup>, Xianghai  
Meng<sup>a</sup>, \*

<sup>a</sup> State Key Laboratory of Heavy Oil Processing, China University of Petroleum, Beijing, 102249, China

<sup>b</sup> State Key Laboratory of Multiphase Complex System, Beijing Key Laboratory of Ionic Liquids Clean Process, Institute of  
Process Engineering, Chinese Academy of Sciences, Beijing, 100190, China

[Corresponding author: mengxh@cup.edu.cn](mailto:mengxh@cup.edu.cn)

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Fig. S1 The color change of wet potassium iodide starch paper during the electrodeposition of (a) Et<sub>3</sub>NHCl-1.54AlCl<sub>3</sub>-0.010CuCl IL and (b) Et<sub>3</sub>NHCl-1.54AlCl<sub>3</sub>-0.23CuCl IL.

**Table S1** Actual molar coefficient of bimetallic composite ILs.

Theoretical molar ratio	Contents of Al (wt%)	Contents of Cu (wt%)	Molar coefficient		
			Et <sub>3</sub> NHCl	AlCl <sub>3</sub>	CuCl
Et <sub>3</sub> NHCl-1.8AlCl <sub>3</sub>	11.96	0	1	1.49	0
Et <sub>3</sub> NHCl-1.8AlCl <sub>3</sub> -0.005CuCl	12.01	0.10	1	1.51	0.005
Et <sub>3</sub> NHCl-1.8AlCl <sub>3</sub> -0.01CuCl	11.68	0.19	1	1.42	0.010
Et <sub>3</sub> NHCl-1.8AlCl <sub>3</sub> -0.02CuCl	11.67	0.36	1	1.43	0.020
Et <sub>3</sub> NHCl-1.8AlCl <sub>3</sub> -0.05CuCl	12.35	0.60	1	1.66	0.034
Et <sub>3</sub> NHCl-1.8AlCl <sub>3</sub> -0.1CuCl	12.25	1.28	1	1.67	0.074
Et <sub>3</sub> NHCl-1.8AlCl <sub>3</sub> -0.2CuCl	11.70	2.23	1	1.54	0.11
Et <sub>3</sub> NHCl-1.8AlCl <sub>3</sub> -0.5CuCl	11.46	3.95	1	1.57	0.23
Average	-	-	-	1.54	-

**Table S2** Variation of copper and aluminium contents with potential in electrodeposits from Et<sub>3</sub>NHCl-1.54AlCl<sub>3</sub>-0.010CuCl on silver electrode.

Electrodeposition potential (V)	Copper (wt%)	Aluminium (wt%)	Other elements (wt%)
-1.5	89.48	2.89	7.63
-1.8	70.97	1.17	27.86
-2.6	10.22	40.75	49.03

**Table S3** Variation of copper and aluminium contents with potential in electrodeposits from Et<sub>3</sub>NHCl-1.54AlCl<sub>3</sub>-0.23CuCl on silver electrode.

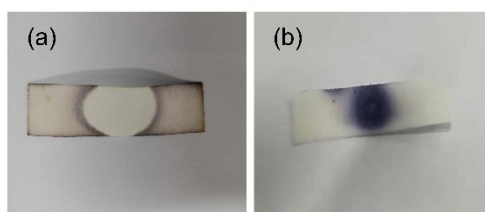
Electrodeposition potential (V)	Copper (wt%)	Aluminium (wt%)	Other elements (wt%)
-1.2	94.25	0.48	5.27
-1.8	98.58	0.35	1.07
-2.6	96.40	0.30	3.30

**Table S4** Contents of copper and aluminium in electrodeposits from Et<sub>3</sub>NHCl-1.54AlCl<sub>3</sub>-xCuCl ILs on silver electrode at the potentials of -1.8 V vs. Pt. (Corresponding to Fig.6).

Actual molar ratio	Copper (wt%)	Aluminium (wt%)	Other elements (C, O, Cl, Ag) (wt%)
Et <sub>3</sub> NHCl-1.54AlCl <sub>3</sub> -0.005CuCl	54.99	4.53	40.48
Et <sub>3</sub> NHCl-1.54AlCl <sub>3</sub> -0.010CuCl	70.97	1.17	27.86
Et <sub>3</sub> NHCl-1.54AlCl <sub>3</sub> -0.020CuCl	69.27	7.70	23.03
Et <sub>3</sub> NHCl-1.54AlCl <sub>3</sub> -0.034CuCl	87.89	4.75	7.36
Et <sub>3</sub> NHCl-1.54AlCl <sub>3</sub> -0.074CuCl	90.62	1.59	7.79
Et <sub>3</sub> NHCl-1.54AlCl <sub>3</sub> -0.11CuCl	98.58	0.35	1.07
Et <sub>3</sub> NHCl-1.54AlCl <sub>3</sub> -0.23CuCl	97.02	0.15	2.83

**Table S5** Contents of copper and aluminium in electrodeposits from Et<sub>3</sub>NHCl-1.54AlCl<sub>3</sub>-xCuCl ILs on silver electrode at the potentials of -2.6 V vs. Pt. (Corresponding to Fig.6).

Actual molar ratio	Copper (wt%)	Aluminium (wt%)	Other elements (C, O, Cl, Ag) (wt%)
$\text{Et}_3\text{NHCl}-1.54\text{AlCl}_3-0.005\text{CuCl}$	1.37	34.09	64.54
$\text{Et}_3\text{NHCl}-1.54\text{AlCl}_3-0.010\text{CuCl}$	10.22	40.75	49.03
$\text{Et}_3\text{NHCl}-1.54\text{AlCl}_3-0.020\text{CuCl}$	18.82	34.28	46.90
$\text{Et}_3\text{NHCl}-1.54\text{AlCl}_3-0.034\text{CuCl}$	19.87	33.43	46.70
$\text{Et}_3\text{NHCl}-1.54\text{AlCl}_3-0.074\text{CuCl}$	41.21	9.67	49.12
$\text{Et}_3\text{NHCl}-1.54\text{AlCl}_3-0.11\text{CuCl}$	96.40	0.30	3.30
$\text{Et}_3\text{NHCl}-1.54\text{AlCl}_3-0.23\text{CuCl}$	94.21	0.31	5.48



**Fig. S1** The color change of wet potassium iodide starch paper during the electrodeposition of (a)  $\text{Et}_3\text{NHCl}-1.54\text{AlCl}_3-0.010\text{CuCl}$  IL and (b)  $\text{Et}_3\text{NHCl}-1.54\text{AlCl}_3-0.23\text{CuCl}$  IL.