

Electronic Supplementary Information

Direct dissolution of UO₂ by carboxyl-functionalized ionic liquids in the presence or absence of Fe-containing ionic liquids

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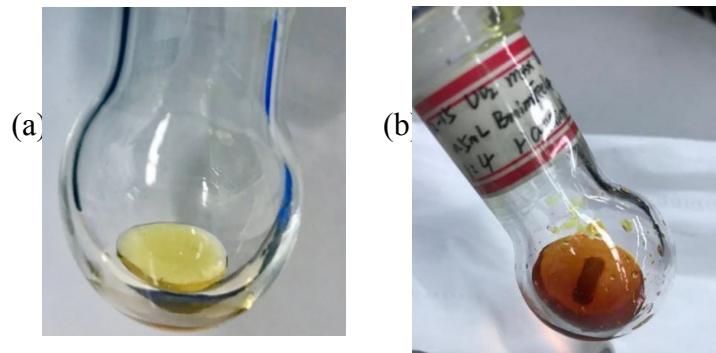


Fig. S1 Dissolution of UO_2 (a) in single $[\text{HOOCMmim}][\text{Tf}_2\text{N}]$ IL at 120°C , and (b) in the mixture of $[\text{Bmim}][\text{FeCl}_4]$ and $[\text{HOOCMmim}][\text{Tf}_2\text{N}]$ at 180°C .

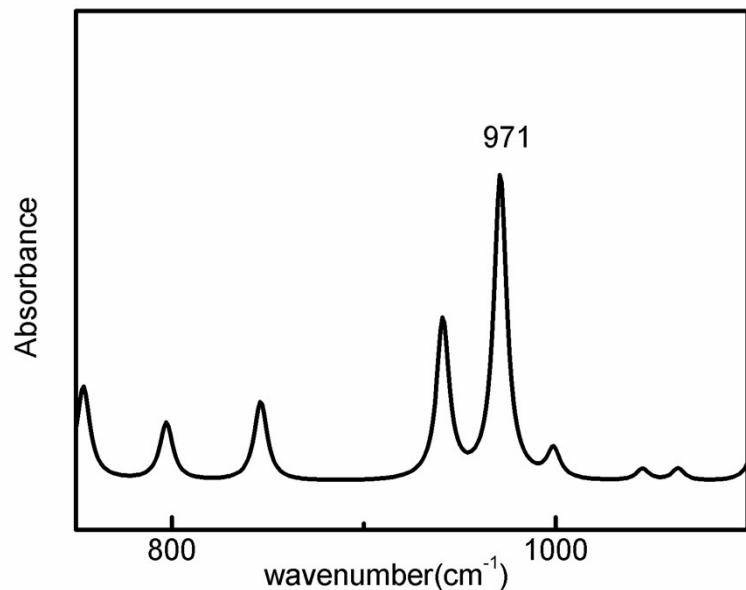


Fig. S2 The IR spectrum of $[\text{UO}_2(\text{OOCMmim})_3]^{2+}$ obtained in gas phase by the B3LYP/6-311G(d, p)/D3BJ level.

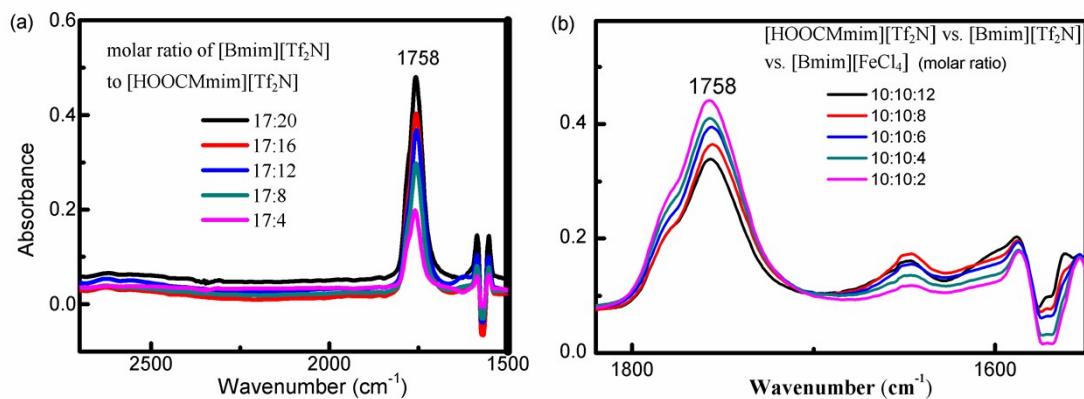


Fig. S3 IR spectra for the mixture with a various molar ratio (a) [HOOCMmim][Tf₂N] and [Bmim][Tf₂N], and (b) [Bmim][FeCl₄], [Bmim][Tf₂N] and [HOOCMmim][Tf₂N].

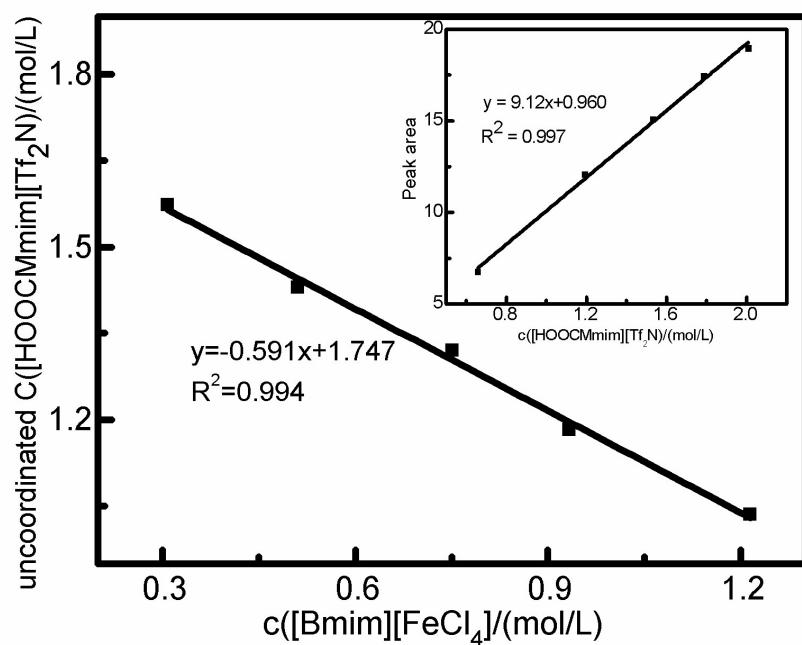


Fig. S4 The linear relationship between the uncoordinated [HOOCMmim][Tf₂N] and [Bmim][FeCl₄] (the inserted graph: the linear relationship between the peak area at 1758 cm⁻¹ and the known concentration of [HOOCMmim][Tf₂N]).

Table S1 The cyclic voltammetric behavior of [Imim][FeCl₄] in [Imim][Tf₂N].

| [Imim]FeCl ₄ | V(V/s) | E _{pc} /V | I _{pc} /μA | E _{pa} /V | I _{pa} /μA | I _{pa} /I _{pc} | E _{1/2} /V (vs Ag ⁺ /Ag) | ΔE/mV |
|--------------------------------------|--------|--------------------|---------------------|--------------------|---------------------|----------------------------------|---|-------|
| [Emim]FeCl ₄ 0.01mol/L | 0.02 | -0.579 | -12.19 | -0.509 | 12.18 | 1.00 | -0.544 | 70 |
| | 0.05 | -0.588 | -18.89 | -0.502 | 18.80 | 1.00 | -0.545 | 86 |
| | 0.075 | -0.593 | -22.13 | -0.492 | 22.18 | 1.00 | -0.543 | 101 |
| | 0.1 | -0.596 | -25.67 | -0.493 | 25.72 | 1.00 | -0.545 | 103 |
| | 0.2 | -0.614 | -33.99 | -0.476 | 33.94 | 1.00 | -0.545 | 138 |
| [Bmim]FeCl ₄ 0.01mol/L | 0.02 | -0.596 | -11.72 | -0.511 | 11.59 | 0.99 | -0.554 | 85 |
| | 0.05 | -0.605 | -14.36 | -0.499 | 14.19 | 0.99 | -0.552 | 106 |
| | 0.075 | -0.607 | -17.24 | -0.496 | 16.67 | 0.97 | -0.552 | 111 |
| | 0.1 | -0.613 | -19.18 | -0.492 | 18.58 | 0.97 | -0.553 | 121 |
| | 0.2 | -0.624 | -26.55 | -0.482 | 25.57 | 0.96 | -0.553 | 141 |

Table. S2 Dissolution amount of UO₂ in 0.5 mL of the carboxyl-functionalized ILs in atmospheric condition.

| 0.5mL [HOOCMmim][Tf ₂ N] (0.683 g) | | 0.5mL [HOOCEtmim][Tf ₂ N] (0.727 g) | |
|--|-----------------------------------|---|-----------------------------------|
| Temperature (°C) | dissolved UO ₂ (mg) | Temperature (°C) | dissolved UO ₂ (mg) |
| 60 | 1.17 | 60 | 0.73 |
| 90 | 4.82 | 90 | 7.81 |
| 140 | 16.7 | 140 | 17.5 |