Supplementary Information

The Adsorption of Water-soluble Ionic Liquids on Graphene

Oxide of Different Oxygen Content

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S1: measurement of ionic liquids

The competitive sorption experiments of [bmim][Cl] and [n-bPy][Cl]: the 211 nm and 258 nm were adsorption peak of [bmim][Cl] and [n-bPy][Cl], respectively. So but a little adsorption peak of [n-bPy][Cl] was also 211 nm, so it was hard to directly measure the concentrations of [bmim][Cl] and [n-bPy][Cl] in mixture solution using UV. But a stable ratio (SR) of adsorption peaks could be found for [n-bPy][Cl] in 211 and 258 nm, so we could calculate the ABS of 211 as following:

$$ABS_{211}([n-bPy][Cl]) = \frac{ABS_{258}([n-bPy][Cl])}{SR}$$

So the ABS₂₁₁ of [bmim][Cl] :

 $ABS_{211}([bmim][Cl]) = ABS_{211}([bmim][Cl]) - ABS_{211}[n - bPy][Cl])$

And then according this formula, we could do standard curve the [bmim][Cl] and

[n-bPy][Cl], so the concentrations of [bmim][Cl] and [n-bPy][Cl] in mixture solution could be measured and calculated. Other sorption experiments were performed according to above methods.



S2: C1s and O1s XPS spectra of LGO, CGO and HGO.

Figure S1 C1s and O1s XPS spectra of LGO(a,b), CGO(c, d) and HGO(e,f);

C1s spectra were compared by deconvoluting each spectrum into three peaks that correspond to the following functional groups: carbon sp² (C=C, 284.8 eV), epoxy/hydroxyls (C-O, 287.1eV), carbonyl (C=O, 287.6eV).

S3: adsorption data fitting

The Freundlich equation (eq 1) was employed to fit the sorption data. Its general form

$$Q = k_F C_e^{n}$$
 (1)

Where Q (mol/g) was the amount of adsorbate adsorbed per unit mass of the adsorbent; C_e (mol/L) was the adsorbate concentration at equilibrium; K_F (molⁿ⁻¹ Lⁿ/g) was the sorption affinity coefficient; and n was an indicator of isotherm nonlinearity related to the heterogeneity of sorption sites.

The Langmuir equation (eq 2) was employed to fit the sorption data. Its general form was

$$q_e = kQ_{\infty}C_e/(1+kC_e) \qquad (2)$$

Where $q_e \pmod{g}$ was the amount of adsorbate adsorbed per unit mass of the adsorbent; $C_e \pmod{L}$ was the adsorbate concentration at equilibrium; $Q_{\infty} \pmod{g}$ was the maximum adsorption amount; and k (L/mol) was Langmuir sorption equilibrium constant.

S4: thermodynamic behavior of ILs on GO

Adsorption isothermals under different temperature were conducted. The adsorption of [bmim][Cl] on HGO (*fig. 2Sa*), mainly forming chemical bonds, was positive to temperature. And R. Gašparac *et al.* reported that the adsorption of imidazole and its derivatives depended on temperature¹. While the adsorption of [n-bPy][Cl] on LGO (*fig. 2Sb*), mainly forming nonchemical bonds, was negative to temperature, similar to Xu Jing's report².





Figure S2 Adsorption isotherm of [bmim][C1] on HGO (a) and [n-bPy][C1] on LGO (b) at pH=5.5 and ionic

strength I=0.1mol/L.

1. R. Gaš parac, C. R. Martin and E. Stupniš ek - Lisac, *Journal of The Electrochemical Society*, **2000**, *147*, 548-551.

2. J. Xu,L. Wang andY. Zhu, *Langmuir*, **2012**, *28*, 8418-8425.