

Supplementary Informations

The initial molar fraction and molar fraction at equilibrium in solution for the binary mixture of the adsorbates at T=298 K and r=1, for the three studied systems, have been calculated (in Table 1) from the experimental isotherms. The experimental isotherms at T=298 K and r=1 are represented in Figures 3, 4 and 5 in the manuscript for the three binary systems.

Table 1:Initial molar fraction and molar fraction at equilibrium in solution for the binary mixture of the adsorbates.

Binary system	Initial molar fraction of the compound from experience			Molar fraction in mixture after adsorption from experimental isotherms		
	IL1/IL2	IL1/IBP	IL2/IBP	IL1/IL2	IL1/IBP	IL2/IBP
IL1 compound	0.225	0.248	-	0.501	0.957	-
IL2 compound	0.225	-	0.220	0.498	-	0.982
IBP compound	-	0.248	0.220	-	0.042	0.018

The initial molar fractions used for the COSMO-RS calculations are reported in Table 2. Based on the calculated partial pressure (values of partial pressure are retrieved from COSMO-RS output file), all the estimated molar fractions in mixture were also calculated. The values are reported in the Table 2.

Table 2: Initial molar fraction and molar fraction derived from calculated partial pressure for the binary mixture of the adsorbates.

Binary system	Initial molar fraction given for the COSMO-RS calculation			Molar fraction in mixture obtained from the partial pressures given by COSMO-RS calculations		
	IL1/IL2	IL1/IBP	IL2/IBP	IL1/IL2	IL1/IBP	IL2/IBP
IL1 compound	0.227	0.227	-	0.999	0.999	-
IL2 compound	0.227	-	0.227	0	-	0.045
IBP compound	-	0.227	0.227	-	0	0.954

The comparison of Table 1 and Table 2 shows obviously a disagreement between the molar fraction in mixture determined from experience and from the COSMO-RS calculation by using the calculated partial pressures in the mixture. This is because the model cannot reproduced the adsorption as it does not take into account a mobility of the adsorbates for interacting with graphene.

So, based on the chemical potentials calculated in the mixture from the COSMO-RS model, the fugacities have been deduced (Table 3) from the following equation: $\mu_i - RT \ln(P_i) = RT \ln(f_i/P_i)$, where μ_i is the chemical potential in the real mixture of the i^{th} component, P_i is the partial pressure of the i^{th} component in the ideal mixture, f_i is fugacity of the i^{th} component, T is the temperature and R the perfect gas constant.

Table 3 : Given initial molar fractions and calculated fugacities determined from the COSMO-RS model.

Binary system	Initial molar fraction given for the calculation			Fugacity of the compound calculated from the COSMO-RS model (bar)		
	IL1/IL2	IL1/IBP	IL2/IBP	IL1/IL2	IL1/IBP	IL2/IBP
IL1 compound	0.227	0.227	-	0	0	-
IL2 compound	0.227	-	0.227	0.2×10^{-3}	-	0.09×10^{-3}
IBP compound	-	0.227	0.277	-	0.03×10^{-3}	0.14×10^{-3}

Regarding the fugacity calculation, we can conclude that:

- for the IL2/IL1 system, Fugacity (IL2) >> Fugacity (IL1)
- for the IL1/IBP system, Fugacity (IBP) >> Fugacity (IL1)
- for the IL2/IBP system, Fugacity (IBP) > Fugacity (IL2).

Regarding now the adsorption isotherms at $T=298$ K and $r=1$ (and experimental molar fractions in Table 1), we noticed the results of the calculated fugacities are in good agreement with the adsorbed quantities on the activated carbon for IL2/IBP and IL1/IBP systems. Indeed for the IL1/IBP system: $Q(\text{IBP}) > Q(\text{IL1})$, and for the IL2/IBP system: $Q(\text{IBP}) > Q(\text{IL2})$.

But for the IL2/IL1 system, the fugacities values of IL1 and IL2 are not in agreement with the adsorption uptake as $Q(\text{IL2}) \sim Q(\text{IL1})$ (the adsorption uptakes of IL1 and IL2 are very close).

The conclusion is that the values of the fugacities calculated from the COSMO-RS model in the mixture, are not exactly in agreement with adsorption uptake on the carbon surface. This is because the COSMO-RS model does not allow simulating the adsorption phenomenon.