**Table S1: The values of Pearson correlation coefficients (*P*\_*x*r) between *F*P and *x*r**

|  |  |  |
| --- | --- | --- |
| **ID** | **Name** | ***P*\_*x*r** |
| *F*1 | The molar mass (*M*) of gases (CO2, CH4, N2, and H2) | 0.17 |
| *F*2 | The density (*ρ*) of gases | 0.17 |
| *F*3 | The gaseous probe size (*d*probe) of gases | 0.07 |
| *F*4 | The kinetic diameter (*d*kinetic) of gases | -0.05 |
| *F*5 | The Lennard-Jones (LJ) diameter (*d*LJ) of gases | 0.12 |
| *F*6 | The boiling point (*b.p.*) of gases | 0.17 |
| *F*7 | The melting point (*m.p.*) of gases | 0.18 |
| *F*8 | The volume derived from *d*probe of gases (*V*probe) | 0.07 |
| *F*9 | The volume derived from *d*kinetic of gases (*V*kinetic) | -0.06 |
| *F*10 | The volume derived from *d*LJ of gases (*V*LJ) | 0.12 |
| *F*11 | The specific gas constant (*R*s) of gases | -0.10 |
| *F*12 | The minimum heat of adsorption (*H*ads, min) of gases | 0.15 |
| *F*13 | The *M* of ILs (BmimTFSI, BmimPF6, BmimAc, and BmimBF4) | 0.04 |
| *F*14 | The *ρ* of ILs | 0.02 |
| *F*15 | The *m.p.* of ILs | 0.21 |
| ***F*16** | **The heat of vaporization (*H*vap) of ILs** | **0.21** |
| *F*17 | The heat capacity (*H*cap) of ILs | -0.11 |
| *F*18 | The viscosity (*η*) of ILs | 0.21 |
| *F*19 | The surface tension (*σ*) of ILs | 0.05 |

**Table S2: The values of *P*\_*x*r between *F*E and *x*r**

|  |  |  |
| --- | --- | --- |
| **ID** | **Name** | ***P*\_*x*r** |
| *F*20 | The HOMO-LUMO gap (*E*gap) of gases | -0.13 |
| *F*21 | The HOMO energy of gases | 0.16 |
| *F*22 | The LUMO energy of gases | 0.00 |
| *F*23 | The total molecular volume(*V*m) of gases | 0.12 |
| *F*24 | The polarizability (*α*) of gases | 0.13 |
| *F*25 | The quadrupole moment (*Q*) of gases | 0.18 |
| *F*26 | The electrostatic potential (*ESP*) density (*ρESP*) of gases | 0.16 |
| *F*27 | The *ESP* volume (*VESP*) of gases | 0.12 |
| *F*28 | The *ESP* surface area (*AESP*) of gases | 0.13 |
| *F*29 | The *ESP* maximum value (*ESP*max) of gases | 0.18 |
| *F*30 | The *ESP* minimum value (*ESP*min) of gases | -0.16 |
| *F*31 | The *ESP* average value (*ESP*ave) of gases | 0.17 |
| ***F*32** | **The *E*gap of ILs** | **-0.19** |
| *F*33 | The HOMO energy of ILs | 0.18 |
| *F*34 | The LUMO energy of ILs | 0.10 |
| *F*35 | The *V*m of ILs | 0.07 |
| *F*36 | The dipole moment (*μ*) of ILs | -0.06 |
| *F*37 | The *ρESP* of ILs | -0.02 |
| *F*38 | The *VESP* of ILs | 0.07 |
| *F*39 | The *AESP* of ILs | 0.09 |
| *F*40 | The *ESP*max of ILs | -0.06 |
| *F*41 | The *ESP*min of ILs | 0.04 |
| **ID** | **Name** | ***P*\_*x*r** |
| *F*42 | The *ESP*ave of ILs | -0.13 |
| *F*43 | The *E*gap of the composite system of ILs and gases | -0.19 |
| *F*44 | The HOMO energy of the composite system of ILs and gases | 0.18 |
| *F*45 | The LUMO energy of the composite system of ILs and gases | 0.11 |
| *F*46 | The *V*m of the composite system of ILs and gases | 0.09 |
| *F*47 | The *μ* of the composite system of ILs and gases | -0.04 |
| *F*48 | The *ρESP* of the composite system of ILs and gases | 0.04 |
| *F*49 | The *VESP* of the composite system of ILs and gases | 0.08 |
| *F*50 | The *AESP* of the composite system of ILs and gases | 0.08 |
| *F*51 | The *ESP*max of the composite system of ILs and gases | -0.05 |
| *F*52 | The *ESP*min of the composite system of ILs and gases | 0.18 |
| *F*53 | The *ESP*ave of the composite system of ILs and gases | -0.04 |

**Table S3: The values of *P*\_*x*r between *F*I and *x*r**

|  |  |  |
| --- | --- | --- |
| **ID** | **Name** | ***P*\_*x*r** |
| *F*54 | The pair of ILs (*N*ILs) of NC-ILMs | -0.18 |
| *F*55 | The pressure (*P*) of NC-ILMs | 0.15 |
| *F*56 | The volume (*V*) of NC-ILMs | -0.16 |
| *F*57 | The interaction energy of ILs to ILs (*E*l/l), averaged by *N*ILs | 0.02 |
| *F*58 | The interaction energy of gases to gases (*E*g/g), averaged by the number of gas molecules (*N*gas) | 0.11 |
| *F*59 | The interaction energy of ILs to gases (*E*l/g), averaged by *N*gas | -0.06 |
| *F*60 | The interaction energy of anions to gases (*E*a/g), averaged by *N*gas | -0.06 |
| *F*61 | The interaction energy of cations to gases (*E*c/g), averaged by *N*gas | -0.05 |
| ***F*62** | **The interaction energy of cations to anions (*E*c/a), averaged by *N*ILs** | **0.20** |

**Table S4: The values of *P*\_*x*r between *F*AC and *x*r**

|  |  |  |
| --- | --- | --- |
| **ID** | **Name** | ***P*\_*x*r** |
| *F*63 | The fractional angstrom cavity (*FAC*), with the *r*P = 0 nm (*FAC*0) | 0.00 |
| *F*64 | The *FAC* detected with the positronium as the probe, where the *r*P is 0.11 nm (*FAC*0.11) | 0.50 |
| *F*65 | The *FAC* detected with the oxygen molecule as the probe, where the *r*P is 0.173 nm (*FAC*0.173) | 0.69 |
| *F*66 | The number of *CAC* (*NCAC*) | 0.04 |
| *F*67 | The total volume of *CAC*(*VCAC*, total) | 0.61 |
| *F*68 | **The average volume of *CAC* (*VCAC*, ave)** | **0.84** |
| *F*69 | The standard deviation of volume of *CAC* (*VCAC*, dev) | 0.79 |
| *F*70 | The maximum volume of *CAC* (*VCAC*, max) | 0.69 |
| *F*71 | The *FAC*0 in the near-boundary layers (*FAC*B, 0) | 0.13 |
| *F*72 | The *FAC*B with *r*P = 0.11 nm (*FAC*B, 0.11) | 0.60 |
| *F*73 | The *FAC*B with *r*P = 0.173 nm (*FAC*B, 0.173) | 0.71 |
| *F*74 | The proportion of *FAC*B, 0 to total *FAC*0 (*FAC*B/*FAC*0) | 0.08 |
| *F*75 | The proportion of *FAC*B, 0.11 to total *FAC*0.11 (*FAC*B/*FAC*0.11) | 0.20 |
| *F*76 | The proportion of *FAC*B, 0.173 to total *FAC*0.173 (*FAC*B/*FAC*0.173) | 0.18 |
| *F*77 | The *FAC*0 in the center of BLGOs (*FAC*C, 0) | -0.07 |
| *F*78 | The *FAC*C with *r*P = 0.11 nm (*FAV*C, 0.11) | 0.16 |
| *F*79 | The *FAC*C with *r*P = 0.173 nm (*FAC*C, 0.173) | 0.40 |
| *F*80 | The proportion of *FAC*C, 0 to total *FAC*0 (*FAC*C/*FAC*0) | -0.08 |
| *F*81 | The proportion of *FAC*C, 0.11 to total *FAC*0.11 (*FAC*C/*FAC*0.11) | -0.20 |
| *F*82 | The proportion of *FAC*C, 0.173 to total *FAC*0.173 (*FAC*C/*FAC*0.173) | -0.18 |

**Table S5: The values of *P*\_*x*r between *F*CS and *x*r**

|  |  |  |
| --- | --- | --- |
| **ID** | **Name** | ***P*\_*x*r** |
| *F*83 | The near-boundary layer thickness (*d*B) of NC-ILMs | -0.40 |
| *F*84 | The proportion of cations (Bmim+) in the near-boundary layers to the total confined space (*P*com-B) | -0.11 |
| *F*85 | The *P*com-B of anions (TFSI-, PF6-, Ac-, BF4-) in NC-ILMs | -0.19 |
| *F*86 | The *P*com-B of gases (CO2, CH4, N2, H2) in NC-ILMs | 0.24 |
| *F*87 | The *P*com-B of all components in NC-ILMs | -0.15 |
| *F*88 | The maximum value obtained from *ρ*number profile of cations in the near-boundary layer (*ρ*cation-B, max) | 0.05 |
| *F*89 | The maximum value obtained from *ρ*number profile of anions in the near-boundary layer (*ρ*anion-B, max) | -0.38 |
| *F*90 | The maximum value obtained from *ρ*number profile of gases in the near-boundary layer (*ρ*gas-B, max) | 0.22 |
| *F*91 | The maximum value obtained from *ρ*number profile of cations in the center of BLGOs (*ρ*cation-C, max) | 0.10 |
| *F*92 | The maximum value obtained from *ρ*number profile of anions in the center of BLGOs (*ρ*anion-C, max) | 0.30 |
| *F*93 | The maximum value obtained from *ρ*number profile of gases in the center of BLGOs (*ρ*gas-C, max) | -0.16 |
| *F*94 | The value at *Z* = 0 obtained from *ρ*number profile of cations (*ρ*cation, *Z* = 0) | -0.12 |
| *F*95 | The value at *Z* = 0 obtained from *ρ*number profile of anions (*ρ*anion, *Z* = 0) | 0.11 |
| *F*96 | The value at *Z* = 0 obtained from *ρ*number profile of gases (*ρ*gas, *Z* = 0) | -0.20 |
| *F*97 | The value at *Z* = 0 obtained from *ρ*mass profile (*ρ*mass, *Z* = 0) | -0.03 |
| *F*98 | The maximum value obtained from *ρ*mass profile (*ρ*mass, max) | 0.05 |
| ***F*99** | **The value at *Z* = 0 obtained from *ρ*charge profile (*ρ*charge, *Z* = 0)** | **-0.40** |
| *F*100 | The positive peak value obtained from *ρ*charge profile (*ρ*charge, peak+) | 0.03 |
| *F*101 | The negative peak value obtained from *ρ*charge profile (*ρ*charge, peak-) | 0.18 |

**Table S6: The values of *P*\_*x*r between *F*CI and *x*r**

|  |  |  |
| --- | --- | --- |
| **ID** | **Name** | ***P*\_*x*r** |
| *F*102 | The interaction energy of BLGO to BLGO (*E*s/s) | -0.20 |
| ***F*103** | **The interaction energy of BLGO to gases (*E*s/g), averaged by *N*gas** | **-0.38** |
| *F*104 | The interaction energy of BLGO and ILs to gases (*E*o/g), averaged by *N*gas | -0.24 |
| *F*105 | The interaction energy of BLGO to ILs (*E*s/l), averaged by *N*ILs | -0.29 |
| *F*106 | The interaction energy of BLGO to cations (*E*s/c), averaged by *N*ILs | -0.27 |
| *F*107 | The interaction energy of BLGO to anions (*E*s/a), averaged by *N*ILs | -0.25 |
| *F*108 | The self-diffusion coefficient of ILs (*D*ILs) | -0.04 |
| *F*109 | The self-diffusion coefficient of gases (*D*gases) | -0.07 |
| *F*110 | The diffusion duration derived from *D*gases (*D*t) | -0.07 |