**TABLE CAPTIONS**

**TABLE 1.** CO2 and CH4 solubility and CO2/CH4 selectivity at about 1 bar.

**TABLE 2.** Thermodynamic properties of CO2 in [Cho][Triz]/TMS calculated by modified deactivated.

**TABLE 3.** Thermodynamic properties of CO2 and CH4 dissolution in [Cho][Triz]/TMS systems.

**TABLE 4.** Calculated ESP and Mulliken charges of the TAFILs atoms.

**TABLE 1.** CO2 and CH4 solubility and CO2/CH4 selectivity at about 1 bar.

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| Absorbents | T (°C) | mol CO2/mol absorbent | mol CH4/mol absorbent | SCO2/CH4 | Ref. |
| [Cho][Triz] | 40 | 0.5877\* | 0.0025\* | 235.1 | This work |
| [Cho][Triz]/TMS (80 wt%/20 wt%) | 40 | 0.4585\* | 0.0024\* | 191.0 |
| [Cho][Triz]/NFM (80 wt%/20 wt%) | 40 | 0.4169\* | 0.0025\* | 166.8 |
| [Cho][Triz]/TMS (60 wt%/40 wt%) | 40 | 0.3076\* | 0.0023\* | 133.7 |
| [Cho][Triz]/TMS (40 wt%/60 wt%) | 40 | 0.1885\* | 0.0023\* | 82.0 |
| TMS | 40 | 0.0132\* | 0.0021\* | 6.3 |
| NFM | 40 | 0.0149\* | 0.0017\* | 8.8 |
| [Cho][Pro]/PEG200 (1:2)b | 65 | - | - | 185.0 | 24 |
| [m-2HEA][Pr] | 40 | 0.4484 | 0.0054 | 83.5 | 23 |
| [BSmim][tos] | 25 | - | - | 47.5 | 33 |
| [BSmpy][tos] | 25 | - | - | 40.7 | 33 |
| [BHEA][Bu] | 40 | 0.2597 | 0.0083 | 32.7 | 23 |
| [NC-C1mim][Tf2N] | 40 | - | - | 18.0 | 18 |
| [E3Py][NTf2] | 40 | 0.0220 | 0.0013 | 17.2 | 17 |
| [E2Py][NTf2] | 40 | 0.0200 | 0.0012 | 17.1 |
| [E1Py][NTf2] | 40 | 0.0190 | 0.0011 | 16.9 |
| [C2mim][CH3OHPO2] | 40 | 0.0812 | 0.0048 | 16.9 | 34 |
| ChCl/ urea (1:2)a | 40 | 0.0080 | 0.0005 | 16.00 | 4 |
| [DEA][Bu] | 40 | 0.0980 | 0.0110 | 9.7 | 35 |
| [C12mim][PF6] | 30 | 0.1396 | 0.0204 | 6.8 | 15 |
| [C16mim][PF6] | 30 | 0.1626 | 0.0398 | 4.1 |
| [N2222][PF6] | 30 | 0.0898 | 0.0426 | 2.1 |
| [N4444][PF6] | 30 | 0.0936 | 0.1039 | 0.9 |  |

amolar ratio; bmass ratio; \*Since the pressure could not be accurately controlled at 1 bar during the experiment, the CO2 or CH4 solubility data at 1 bar in [Cho][Triz] binary absorbents were calculated by the modified deactivation model; The full name for absorbents was detailed in TABLE S1.

**TABLE 2.** Thermodynamic properties of CO2 in [Cho][Triz]/TMS calculated by modified deactivated model.

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| Absorbents | T (°C) | *K* | *H* | *C* |
| [Cho][Triz] | 40 | 5.15 | 72.04 | 0.69 |
| [Cho][Triz]/TMS (80 wt%/20 wt%) | 40 | 5.18 | 92.50 | 0.54 |
| [Cho][Triz]/TMS (60 wt%/40 wt%) | 40 | 5.42 | 98.31 | 0.36 |
| [Cho][Triz]/TMS (40 wt%/60 wt%) | 40 | 5.56 | 108.84 | 0.22 |
| [Cho][Triz]/TMS (20 wt%/80 wt%) | 40 | 5.77 | 112.76 | 0.12 |

**TABLE 3.** Thermodynamic properties of CO2 and CH4 dissolution in [Cho][Triz]/TMS systems.

|  |  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
|  | Absorbents | T  (°C) | *K*  (bar-1) | | *H*  (bar) | *C* | *Δrea H*  (kJ/mol) | *ΔreaS*  (J /mol/K) | *ΔreaG*  (kJ/mol/K) |
| CO2 | [Cho][Triz] | 40 | 5.15 | | 72.04 | 0.69 | -47.27 | -137.31 | -4.27 |
| 50 | 3.08 | | 84.50 | 0.61 | -136.92 | -3.02 |
| 60 | 1.73 | | 96.22 | 0.58 | -137.32 | -1.52 |
| [Cho][Triz]/TMS (80 wt%/20 wt%) | 40 | 5.18 | | 92.50 | 0.54 | -35.76 | -100.50 | -4.28 |
| 50 | 3.48 | | 93.61 | 0.48 | -100.28 | -3.35 |
| 60 | 2.27 | | 99.96 | 0.45 | -100.51 | -2.27 |
| CH4 | [Cho][Triz] | 40 | |  | 397.07 |  | -3.86 | -62.08 | 15.58 |
| 50 | |  | 408.80 |  | -61.94 | 16.16 |
| 60 | |  | 434.19 |  | -62.08 | 16.82 |
| [Cho][Triz]/TMS (80 wt%/20 wt%) | 40 | |  | 409.59 |  | -3.60 | -61.49 | 15.65 |
| 50 | |  | 432.08 |  | -61.60 | 16.30 |
| 60 | |  | 443.86 |  | -61.49 | 16.88 |

**TABLE 4.** Calculated ESP and Mulliken charges of the TAFILs atoms.

|  |  |  |  |
| --- | --- | --- | --- |
| **TAFILs** | **Atoms** | **ESP (kcal/mol)** | **Mulliken** |
| [Cho][Triz] | N-1 | -32.01 | -0.385 |
| N-2 | -34.69 | -0.278 |
| N-4 | -55.28 | -0.200 |
| [N2222][Triz] | N-1 | -35.78 | -0.351 |
| N-2 | -35.54 | -0.201 |
| N-4 | -57.39 | -0.191 |