# Supporting Information:

# Determination of infinite dilution activity coefficient of several organic solutes in N-butylpyridinium nitrate /N-octylpyridinium nitrate by blend inverse gas chromatography

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**Table S1** The specific retention volume, $V_g^0$, between probes and $[Bpy]NO_3$:$[Opy]NO_3$（1:1）at different temperatures for the hypothetical liquid at zero pressure [a]

| | Probe | | $V_g^0$/(mL·g$^{-1}$) | | | | |
|---|---|---|---|---|---|---|---|
| | | | 333.15 K | 343.15 K | 353.15 K | 363.15 K | 373.15 K |
| $[Bpy]NO_3$:$[Opy]NO_3$ | *n*-$C_6$ | 1:1 | 4.379 | 3.249 | 2.431 | 1.872 | 1.469 |
| | | 1:4 | 6.159 | 4.475 | 3.343 | 2.569 | 2.000 |
| | | 4:1 | 4.198 | 3.131 | 2.348 | 1.806 | 1.416 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | *n*-$C_7$ | 1:1 | 15.94 | 11.09 | 7.925 | 5.786 | 4.313 |
| | | 1:4 | 20.32 | 14.05 | 10.00 | 7.27 | 5.39 |
| | | 4:1 | 12.53 | 8.729 | 6.232 | 4.556 | 3.403 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | *n*-$C_8$ | 1:1 | 41.98 | 27.62 | 18.71 | 13.02 | 9.276 |
| | | 1:4 | 53.17 | 34.97 | 23.71 | 16.52 | 11.78 |
| | | 4:1 | 34.12 | 22.53 | 15.30 | 10.67 | 7.618 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | *n*-$C_9$ | 1:1 | 103.9 | 64.93 | 41.90 | 27.87 | 19.00 |
| | | 1:4 | 126.2 | 78.97 | 50.97 | 33.87 | 23.11 |
| | | 4:1 | 84.78 | 53.08 | 34.29 | 22.80 | 15.57 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | *n*-$C_{10}$ | 1:1 | 224.0 | 135.3 | 83.93 | 54.32 | 35.88 |
| | | 1:4 | 278.7 | 167.4 | 104.0 | 66.63 | 43.96 |
| | | 4:1 | 185.8 | 112.2 | 70.80 | 43.39 | 28.82 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | *n*-$C_{11}$ | 1:1 | 471.3 | 273.3 | 163.5 | 102.2 | 65.19 |
| | | 1:4 | 586.2 | 337.4 | 202.6 | 125.3 | 79.86 |
| | | 4:1 | 398.5 | 229.8 | 138.2 | 85.91 | 55.12 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | *n*-$C_{12}$ | 1:1 | 991.2 | 549.5 | 316.5 | 191.1 | 118.0 |
| | | 1:4 | 1226 | 675.9 | 390.5 | 233.3 | 144.4 |
| | | 4:1 | 859.1 | 472.2 | 271.1 | 161.8 | 99.87 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | Dichloromethane | 1:1 | 72.63 | 51.00 | 36.62 | 27.06 | 20.26 |
| | | 1:4 | 91.92 | 64.97 | 46.64 | 34.00 | 25.34 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | Acetone | 1:1 | 59.69 | 41.83 | 29.84 | 21.97 | 16.40 |
| | | 1:4 | 70.41 | 50.29 | 36.14 | 26.48 | 19.80 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | Chloroform | 1:1 | 225.2 | 150.2 | 102.3 | 71.57 | 50.99 |
| | | 1:4 | 302.6 | 200.7 | 135.9 | 94.17 | 66.56 |
| | | 4:1 | 77.02 | 53.62 | 38.56 | 28.07 | 20.96 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | Ethyl acetate | 1:1 | 63.56 | 43.19 | 29.91 | 21.21 | 15.36 |
| | | 1:4 | 77.04 | 53.21 | 37.25 | 26.38 | 19.11 |
| | | 4:1 | 64.84 | 45.18 | 32.44 | 23.61 | 17.51 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | Tetrahydrofuran | 1:1 | 65.85 | 46.50 | 33.49 | 24.95 | 18.77 |
| | | 1:4 | 79.08 | 56.74 | 41.12 | 30.23 | 22.91 |
| | | 4:1 | 224.1 | 148.5 | 101.0 | 70.10 | 49.89 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | Ether | 1:1 | 4.237 | 3.160 | 2.448 | 2.032 | 1.455 |
| | | 1:4 | 5.750 | 4.333 | 3.377 | 2.602 | 2.069 |
| | | 4:1 | 66.49 | 44.39 | 30.83 | 21.41 | 15.39 |

| | | | | | | | |
|---|---|---|---|---|---|---|---|
| $[Bpy]NO_3$:$[Opy]NO_3$ | Carbon tetrachloride | 1:1 | 67.82 | 46.41 | 32.62 | 23.41 | 17.09 |
| | | 1:4 | 87.62 | 60.51 | 42.18 | 30.16 | 22.40 |
| | | 4:1 | 70.18 | 49.30 | 35.59 | 25.77 | 19.08 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | Methyl acetate | 1:1 | 41.06 | 28.57 | 20.28 | 14.69 | 10.82 |
| | | 1:4 | 48.96 | 35.03 | 25.05 | 18.20 | 13.54 |
| | | 4:1 | 5.184 | 3.667 | 2.694 | 1.973 | 1.484 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | Cyclohexane | 1:1 | 15.81 | 11.29 | 8.253 | 6.159 | 4.684 |
| | | 1:4 | 20.57 | 15.41 | 11.26 | 8.368 | 6.461 |
| | | 4:1 | 67.49 | 46.05 | 32.45 | 23.06 | 16.98 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | Benzene | 1:1 | 106.7 | 73.03 | 51.04 | 36.77 | 26.91 |
| | | 1:4 | 130.5 | 90.27 | 63.40 | 45.41 | 33.17 |
| | | 4:1 | 101.6 | 69.48 | 48.71 | 34.76 | 25.50 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | Toluene | 1:1 | 221.8 | 146.5 | 99.34 | 68.93 | 48.57 |
| | | 1:4 | 273.0 | 181.22 | 122.77 | 84.87 | 59.94 |
| | | 4:1 | 199.8 | 132.2 | 89.56 | 61.76 | 43.54 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | o-Xylene | 1:1 | 611.6 | 386.3 | 251.3 | 168.0 | 114.4 |
| | | 1:4 | 756.3 | 477.7 | 309.7 | 205.8 | 140.0 |
| | | 4:1 | 541.3 | 341.0 | 221.5 | 147.6 | 100.5 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | Ethanol | 1:1 | 357.5 | 230.8 | 152.3 | 103.2 | 71.21 |
| | | 1:4 | 451.1 | 292.1 | 193.3 | 131.0 | 90.59 |
| | | 4:1 | 385.5 | 248.4 | 164.3 | 110.8 | 76.51 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | Carbinol | 1:1 | 280.2 | 185.9 | 126.0 | 87.55 | 62.13 |
| | | 1:4 | 346.7 | 231.8 | 157.8 | 109.6 | 77.50 |
| | | 4:1 | 328.4 | 217.1 | 147.2 | 101.6 | 71.87 |

[a] Standard uncertainties are $u_r(T) = 0.5$ K, and the combined relative standard uncertainty of $u_r$ $(V_g^0)=0.02$.

**Table S2** Mass fraction activity coefficients $\Omega_1^{\infty}$ of probes at various temperatures of mixture ILs for the hypothetical liquid at zero pressure [a]

| | Probe | | $\Omega_1^{\infty}$ | | | | |
|---|---|---|---|---|---|---|---|
| | | | 333.15 K | 343.15 K | 353.15 K | 363.15 K | 373.15 K |
| $[Bpy]NO_3$:$[Opy]NO_3$ | *n*-$C_6$ | 1:1 | 100.5 | 102.2 | 105.3 | 107.5 | 109.8 |
| | | 1:4 | 71.49 | 74.20 | 76.56 | 78.34 | 80.60 |
| | | 4:1 | 104.9 | 106.1 | 109.0 | 111.5 | 113.9 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | *n*-$C_7$ | 1:1 | 63.45 | 65.46 | 67.47 | 69.62 | 71.85 |
| | | 1:4 | 49.77 | 51.70 | 53.45 | 55.44 | 57.45 |
| | | 4:1 | 80.71 | 83.20 | 85.80 | 88.43 | 91.06 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | *n*-$C_8$ | 1:1 | 55.89 | 58.07 | 60.30 | 62.53 | 64.91 |
| | | 1:4 | 44.13 | 45.86 | 47.58 | 49.29 | 51.12 |
| | | 4:1 | 68.75 | 71.19 | 73.73 | 76.35 | 79.03 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | *n*-$C_9$ | 1:1 | 52.86 | 54.91 | 57.01 | 59.06 | 61.24 |
| | | 1:4 | 43.49 | 45.15 | 46.86 | 48.60 | 50.35 |
| | | 4:1 | 64.76 | 67.17 | 69.66 | 72.19 | 74.71 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | *n*-$C_{10}$ | 1:1 | 57.36 | 58.84 | 60.82 | 62.17 | 64.05 |
| | | 1:4 | 46.10 | 47.56 | 49.07 | 50.68 | 52.29 |
| | | 4:1 | 69.15 | 70.98 | 72.10 | 77.83 | 79.75 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | *n*-$C_{11}$ | 1:1 | 64.07 | 65.30 | 67.04 | 68.13 | 70.02 |
| | | 1:4 | 51.51 | 52.90 | 54.09 | 55.57 | 57.15 |
| | | 4:1 | 75.77 | 77.65 | 79.32 | 81.06 | 82.81 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | *n*-$C_{12}$ | 1:1 | 70.55 | 71.92 | 73.59 | 74.56 | 76.48 |
| | | 1:4 | 57.05 | 58.47 | 59.64 | 61.08 | 62.46 |
| | | 4:1 | 81.40 | 83.69 | 85.92 | 88.09 | 90.32 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | Dichloromethane | 1:1 | 2.354 | 2.586 | 2.835 | 3.074 | 3.346 |

| | | | | | | | |
|---|---|---|---|---|---|---|---|
| | | 1:4 | 1.860 | 2.030 | 2.226 | 2.447 | 2.675 |
| | | 4:1 | 2.220 | 2.460 | 2.692 | 2.963 | 3.233 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | Acetone | 1:1 | 7.119 | 7.647 | 8.245 | 8.782 | 9.393 |
| | | 1:4 | 6.034 | 6.361 | 6.808 | 7.286 | 7.782 |
| | | 4:1 | 6.553 | 7.081 | 7.584 | 8.171 | 8.800 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | Chloroform | 1:1 | 1.104 | 1.255 | 1.425 | 1.606 | 1.808 |
| | | 1:4 | 0.8214 | 0.9386 | 1.073 | 1.220 | 1.385 |
| | | 4:1 | 1.109 | 1.269 | 1.443 | 1.639 | 1.848 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | Ethyl acetate | 1:1 | 8.937 | 9.515 | 10.19 | 10.89 | 11.63 |
| | | 1:4 | 7.374 | 7.723 | 8.180 | 8.756 | 9.353 |
| | | 4:1 | 8.543 | 9.258 | 9.882 | 10.79 | 11.62 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | Tetrahydrofuran | 1:1 | 47.93 | 51.83 | 55.82 | 58.96 | 62.49 |
| | | 1:4 | 39.91 | 42.47 | 45.46 | 48.66 | 51.20 |
| | | 4:1 | 14.08 | 16.23 | 18.51 | 20.98 | 23.51 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | Ether | 1:1 | 40.74 | 42.93 | 44.42 | 43.66 | 50.57 |
| | | 1:4 | 30.02 | 31.31 | 32.20 | 34.09 | 35.56 |
| | | 4:1 | 2.596 | 3.056 | 3.527 | 4.144 | 4.781 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | Carbon tetrachloride | 1:1 | 4.553 | 4.959 | 5.373 | 5.814 | 6.294 |
| | | 1:4 | 3.524 | 3.804 | 4.156 | 4.513 | 4.803 |
| | | 4:1 | 4.576 | 4.999 | 5.400 | 5.900 | 6.336 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | Methyl acetate | 1:1 | 8.274 | 8.820 | 9.415 | 10.04 | 10.71 |
| | | 1:4 | 6.939 | 7.193 | 7.621 | 8.101 | 8.559 |
| | | 4:1 | 7.206 | 7.734 | 8.232 | 8.955 | 9.646 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | Cyclohexane | 1:1 | 40.92 | 42.45 | 44.02 | 45.61 | 47.24 |
| | | 1:4 | 31.46 | 31.12 | 32.27 | 33.57 | 34.25 |
| | | 4:1 | 47.94 | 49.85 | 51.79 | 53.80 | 55.86 |

| | | | | | | | |
|---|---|---|---|---|---|---|---|
| $[Bpy]NO_3$:$[Opy]NO_3$ | Benzene | 1:1 | 6.478 | 6.961 | 7.494 | 7.990 | 8.547 |
| | | 1:4 | 5.297 | 5.631 | 6.033 | 6.470 | 6.934 |
| | | 4:1 | 6.804 | 7.317 | 7.852 | 8.452 | 9.020 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | Toluene | 1:1 | 7.400 | 7.877 | 8.388 | 8.935 | 9.572 |
| | | 1:4 | 6.000 | 6.366 | 6.788 | 7.258 | 7.757 |
| | | 4:1 | 8.200 | 8.724 | 9.305 | 9.973 | 10.68 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | o-Xylene | 1:1 | 7.882 | 8.310 | 8.759 | 9.225 | 9.770 |
| | | 1:4 | 6.374 | 6.720 | 7.108 | 7.531 | 7.984 |
| | | 4:1 | 8.906 | 9.414 | 9.938 | 10.500 | 11.122 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | Ethanol | 1:1 | 3.608 | 3.740 | 3.899 | 4.064 | 4.253 |
| | | 1:4 | 2.860 | 2.954 | 3.072 | 3.201 | 3.343 |
| | | 4:1 | 3.346 | 3.474 | 3.614 | 3.785 | 3.959 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | Carbinol | 1:1 | 3.723 | 3.926 | 4.160 | 4.398 | 4.651 |
| | | 1:4 | 3.008 | 3.149 | 3.322 | 3.513 | 3.729 |
| | | 4:1 | 3.176 | 3.362 | 3.561 | 3.790 | 4.021 |

[a] Standard uncertainties are $u_r(T) = 0.5$ K, and the combined relative standard uncertainty of $u_r$ ($\Omega_1^{\infty}$)=0.1.

**Table S3** IL-IL interaction parameter $\chi'_{23}$ at various temperatures for the hypothetical liquid at zero pressure [a]

| ILs | Probes | | $\chi'_{23}$ | | | | |
|---|---|---|---|---|---|---|---|
| | | | 333.15 K | 343.15 K | 353.15 K | 363.15 K | 373.15 K |
| $[Bpy]NO_3$:$[Opy]NO_3$ | *n*-$C_6$ | 1:1 | 1.935 | 2.074 | 2.09 | 2.221 | 2.272 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | | 1:4 | 4.005 | 4.005 | 4.012 | 4.13 | 4.128 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | | 4:1 | 1.856 | 2.028 | 2.06 | 2.182 | 2.229 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | *n*-$C_7$ | 1:1 | 2.196 | 2.201 | 2.196 | 2.182 | 2.162 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | | 1:4 | 3.614 | 3.574 | 3.548 | 3.5 | 3.453 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | | 4:1 | 0.7987 | 0.8091 | 0.8004 | 0.7949 | 0.7892 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | *n*-$C_8$ | 1:1 | 2.128 | 2.07 | 2.023 | 1.985 | 1.932 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | | 1:4 | 3.502 | 3.443 | 3.402 | 3.37 | 3.323 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | | 4:1 | 0.9524 | 0.9178 | 0.8881 | 0.8596 | 0.8245 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | *n*-$C_9$ | 1:1 | 1.959 | 1.908 | 1.858 | 1.823 | 1.786 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | | 1:4 | 3.06 | 3.013 | 2.965 | 2.923 | 2.891 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | | 4:1 | 0.8115 | 0.7702 | 0.7282 | 0.6909 | 0.6663 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | *n*-$C_{10}$ | 1:1 | 1.6 | 1.642 | 1.635 | 1.713 | 1.736 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | | 1:4 | 2.856 | 2.861 | 2.866 | 2.875 | 2.889 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | | 4:1 | 0.5592 | 0.5973 | 0.7062 | 0.4219 | 0.4815 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | *n*-$C_{11}$ | 1:1 | 1.498 | 1.509 | 1.518 | 1.549 | 1.513 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | | 1:4 | 2.753 | 2.714 | 2.75 | 2.708 | 2.667 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | | 4:1 | 0.5847 | 0.5604 | 0.6025 | 0.5957 | 0.6004 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | *n*-$C_{12}$ | 1:1 | 1.238 | 1.315 | 1.363 | 1.464 | 1.51 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | | 1:4 | 2.453 | 2.495 | 2.564 | 2.594 | 2.659 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | | 4:1 | 0.4883 | 0.5087 | 0.5352 | 0.5579 | 0.6054 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | Dichloromethane | 1:1 | -1.619 | -1.639 | -1.659 | -1.608 | -1.583 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | | 1:4 | -0.2501 | -0.2263 | -0.2469 | -0.2882 | -0.2906 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | | 4:1 | -1.029 | -1.105 | -1.114 | -1.163 | -1.154 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | Acetone | 1:1 | -5.279 | -5.474 | -5.721 | -5.965 | -6.246 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | | 1:4 | -4.376 | -4.446 | -4.644 | -4.92 | -5.191 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | | 4:1 | -4.527 | -4.761 | -4.965 | -5.285 | -5.611 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | Chloroform | 1: | -0.9546 | -1.047 | -1.087 | -1.072 | -1.052 |

| | | | | | | | |
|---|---|---|---|---|---|---|---|
| | | 1 | | | | | |
| [Bpy]$NO_3$:[Opy]$NO_3$ | | 1:4 | 0.8128 | 0.685 | 0.6022 | 0.5556 | 0.5226 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | | 4:1 | -0.7839 | -0.9208 | -0.9702 | -1.009 | -0.9965 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | Ethyl acetate | 1:1 | -3.781 | -4.03 | -4.359 | -4.751 | -5.206 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | | 1:4 | -2.699 | -2.84 | -3.096 | -3.497 | -3.951 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | | 4:1 | -3.28 | -3.647 | -3.955 | -4.487 | -4.995 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | Tetrahydrofuran | 1:1 | -0.5499 | -0.5366 | -0.5189 | -0.3553 | -0.3116 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | | 1:4 | 0.4712 | 0.5905 | 0.6496 | 0.7234 | 0.8167 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | | 4:1 | 0.07531 | 0.05288 | 0.08679 | 0.05916 | -0.00385 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | Ether | 1:1 | -0.5203 | -0.917 | -1.049 | -0.3673 | -1.707 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | | 1:4 | 1.314 | 0.9852 | 0.8935 | 1.079 | 0.437 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | | 4:1 | 1.022 | 0.273067 | -0.2102 | -0.3631 | -1.37662 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | Carbon tetrachloride | 1:1 | -0.0961 | -0.2054 | -0.2494 | -0.3206 | -0.364 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | | 1:4 | 1.41 | 1.361 | 1.262 | 1.167 | 1.237 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | | 4:1 | 0.07279 | -0.05663 | -0.08247 | -0.2176 | -0.2075 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | Methyl acetate | 1:1 | -5.321 | -5.611 | -5.953 | -6.393 | -6.864 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | | 1:4 | -3.951 | -4.056 | -4.348 | -4.765 | -5.173 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | | 4:1 | -4.202 | -4.538 | -4.86 | -5.431 | -5.966 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | Cyclohexane | 1:1 | 2.273 | 2.157 | 2.055 | 2.007 | 1.947 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | | 1:4 | 4.222 | 4.42 | 4.319 | 4.244 | 4.285 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | | 4:1 | 1.422 | 1.289 | 1.176 | 1.111 | 1.035 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | Benzene | 1:1 | -0.2437 | -0.3529 | -0.4057 | -0.4013 | -0.403 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | | 1:4 | 1.294 | 1.256 | 1.236 | 1.202 | 1.188 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | | 4:1 | -0.3707 | -0.483 | -0.5153 | -0.5744 | -0.5577 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | Toluene | 1:1 | 0.3364 | 0.3646 | 0.3588 | 0.3331 | 0.302 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | | 1:4 | 1.93 | 1.981 | 1.967 | 1.916 | 1.9 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | | 4:1 | -0.1443 | -0.1126 | -0.1287 | -0.1958 | -0.2228 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | o-Xylene | 1:1 | -2.238 | -2.284 | -2.334 | -2.348 | -2.42 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | | 1:4 | -0.6268 | -0.6713 | -0.7455 | -0.7971 | -0.8754 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | | 4:1 | -2.848 | -2.912 | -2.972 | -3.006 | -3.078 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | Ethanol | 1:1 | -1.806 | -1.852 | -1.9 | -1.934 | -1.978 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | | 1:4 | -0.0597 | -0.08382 | -0.1141 | -0.1465 | -0.1778 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | | 4:1 | -1.103 | -1.16 | -1.194 | -1.26 | -1.301 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | Carbinol | 1: | -2.845 | -2.802 | -2.813 | -2.823 | -2.817 |

| | 1 | | | | | |
|---|---|---|---|---|---|---|
| $[Bpy]NO_3$:$[Opy]NO_3$ | 1:4 | -1.228 | -1.135 | -1.119 | -1.128 | -1.147 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | 4:1 | -1.589 | -1.57 | -1.58 | -1.633 | -1.649 |

[a] Standard uncertainties are $u_r(T) = 0.5$K, and the combined relative standard uncertainty of $u_r$ ($\chi'_{23}$)=0.2.

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**Table S4** Activity coefficients at infinite dilution $\gamma_{12}^{\infty}$ of probes at various temperatures of mixture ILs for the hypothetical liquid at zero pressure [a]

| IL | Probes | | $\gamma_{12}^{\infty}$ | | | | |
|---|---|---|---|---|---|---|---|
| | | | 333.15 K | 343.15 K | 353.15 K | 363.15 K | 373.15 K |
| [Bpy]$NO_3$:[Opy]$NO_3$ | *n*-$C_6$ | 1: 1 | 27.83 | 26.68 | 25.97 | 25.09 | 24.27 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | | 1:4 | 18.41 | 18.03 | 17.57 | 17.01 | 16.59 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | | 4:1 | 31.35 | 29.9 | 29.04 | 28.09 | 27.19 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | *n*-$C_7$ | 1: 1 | 20.38 | 19.84 | 19.32 | 18.86 | 18.45 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | | 1:4 | 14.88 | 14.58 | 14.24 | 13.98 | 13.73 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | | 4:1 | 28 | 27.23 | 26.53 | 25.88 | 25.25 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | *n*-$C_8$ | 1: 1 | 20.43 | 20.03 | 19.65 | 19.29 | 18.97 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | | 1:4 | 15.01 | 14.72 | 14.43 | 14.15 | 13.91 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | | 4:1 | 27.15 | 26.52 | 25.95 | 25.43 | 24.95 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | *n*-$C_9$ | 1: 1 | 21.66 | 21.23 | 20.83 | 20.43 | 20.08 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | | 1:4 | 16.59 | 16.25 | 15.94 | 15.64 | 15.36 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | | 4:1 | 28.66 | 28.05 | 27.49 | 26.97 | 26.45 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | *n*-$C_{10}$ | 1: 1 | 26.03 | 25.2 | 24.62 | 23.82 | 23.27 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | | 1:4 | 19.47 | 18.96 | 18.49 | 18.07 | 17.67 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | | 4:1 | 33.89 | 32.83 | 31.52 | 32.21 | 31.28 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | *n*-$C_{11}$ | 1: 1 | 31.88 | 30.67 | 29.77 | 28.64 | 27.9 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | | 1:4 | 23.85 | 23.12 | 22.35 | 21.74 | 21.2 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | | 4:1 | 40.72 | 39.39 | 38.04 | 36.8 | 35.64 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | *n*-$C_{12}$ | 1: 1 | 38.19 | 36.75 | 35.55 | 34.11 | 33.17 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | | 1:4 | 28.74 | 27.81 | 26.82 | 26 | 25.21 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | | 4:1 | 47.59 | 46.19 | 44.83 | 43.52 | 42.31 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | Dichloromethane | 1: 1 | 0.6592 | 0.6853 | 0.7118 | 0.7325 | 0.7579 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | | 1:4 | 0.4848 | 0.5006 | 0.52 | 0.5427 | 0.5638 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | | 4:1 | 0.6715 | 0.7039 | 0.73 | 0.7627 | 0.791 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | Acetone | 1: 1 | 1.353 | 1.374 | 1.403 | 1.418 | 1.441 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | | 1:4 | 1.067 | 1.064 | 1.078 | 1.095 | 1.111 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | | 4:1 | 1.345 | 1.374 | 1.394 | 1.425 | 1.458 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | Chloroform | 1: 1 | 0.4317 | 0.4641 | 0.4992 | 0.5341 | 0.5714 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | | 1:4 | 0.299 | 0.3231 | 0.3498 | 0.3777 | 0.4074 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | | 4:1 | 0.4684 | 0.5069 | 0.5461 | 0.5889 | 0.6309 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | Ethyl acetate | 1: 1 | 2.839 | 2.582 | 2.619 | 2.657 | 2.697 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | | 1:4 | 1.969 | 1.95 | 1.957 | 1.988 | 2.018 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | | 4:1 | 2.648 | 2.713 | 2.743 | 2.842 | 2.908 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | Tetrahydrofuran | 1: 1 | 11.43 | 11.67 | 11.88 | 11.89 | 11.95 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | | 1:4 | 10.18 | 10.23 | 10.35 | 10.49 | 10.47 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | | 4:1 | 11.52 | 11.83 | 12.01 | 12.37 | 12.64 |
| [Bpy]$NO_3$:[Opy]$NO_3$ | Ether | 1: 1 | 10.22 | 10.19 | 10.58 | 9.328 | 10.47 |

| | | | | | | | |
|---|---|---|---|---|---|---|---|
| $[Bpy]NO_3$:$[Opy]NO_3$ | | 1:4 | 9.400 | 7.952 | 7.751 | 7.792 | 7.724 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | | 4:1 | 8.980 | 9.442 | 9.012 | 10.33 | 10.83 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | Carbon tetrachloride | 1: 1 | 2.285 | 2.353 | 2.415 | 2.48 | 2.551 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | | 1:4 | 1.646 | 1.68 | 1.738 | 1.791 | 1.811 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | | 4:1 | 2.48 | 2.562 | 2.622 | 2.718 | 2.773 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | Methyl acetate | 1: 1 | 1.98 | 1.989 | 2.005 | 2.023 | 2.044 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | | 1:4 | 1.545 | 1.51 | 1.511 | 1.519 | 1.52 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | | 4:1 | 1.862 | 1.884 | 1.894 | 1.949 | 1.988 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | Cyclohexane | 1: 1 | 11.22 | 11.01 | 10.81 | 10.63 | 10.47 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | | 1:4 | 8.028 | 7.511 | 7.378 | 7.283 | 7.061 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | | 4:1 | 14.2 | 13.96 | 13.74 | 13.55 | 13.36 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | Benzene | 1: 1 | 1.968 | 1.999 | 2.038 | 2.061 | 2.095 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | | 1:4 | 1.249 | 1.257 | 1.268 | 1.293 | 1.313 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | | 4:1 | 1.571 | 1.600 | 1.633 | 1.685 | 1.744 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | Toluene | 1: 1 | 2.702 | 2.718 | 2.74 | 2.767 | 2.815 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | | 1:4 | 1.681 | 1.702 | 1.735 | 1.775 | 1.833 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | | 4:1 | 2.64 | 2.654 | 2.679 | 2.723 | 2.769 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | o-Xylene | 1: 1 | 3.215 | 3.201 | 3.192 | 3.187 | 3.204 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | | 1:4 | 2.013 | 2.010 | 2.009 | 2.012 | 2.021 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | | 4:1 | 3.650 | 2.546 | 2.552 | 2.570 | 2.598 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | Ethanol | 1: 1 | 0.6584 | 0.6433 | 0.6333 | 0.6243 | 0.6189 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | | 1:4 | 0.3988 | 0.3909 | 0.3862 | 0.383 | 0.3833 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | | 4:1 | 0.5383 | 0.5269 | 0.5176 | 0.5126 | 0.5079 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | Carbinol | 1: 1 | 0.4755 | 0.4738 | 0.4752 | 0.4765 | 0.4787 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | | 1:4 | 0.2942 | 0.2933 | 0.295 | 0.2979 | 0.3043 |
| $[Bpy]NO_3$:$[Opy]NO_3$ | | 4:1 | 0.3577 | 0.3578 | 0.3587 | 0.362 | 0.3649 |

[a] Standard uncertainties are $u_r(T) = 0.5$ K, and the combined relative standard uncertainty of $u_r$ ($\gamma_{12}^{\infty}$)=0.2.

**Table S5** Solubility parameter $\delta_2$ of mixture ILs at various temperatures for the hypothetical liquid at zero pressure

| ILs | $\delta_2$ /(J·cm$^{-3}$)$^{0.5}$ | | | | | linear equation | $R^2$ |
|---|---|---|---|---|---|---|---|
| | 333.15 K | 343.15 K | 353.15 K | 363.15 K | 373.15 K | | |
| 1:1 | 18.59 | 18.41 | 18.15 | 17.93 | 17.59 | Y=26.89-0.0248 9X | 0.985 2 |
| 1:4 | 18.26 | 18.03 | 17.83 | 17.53 | 17.23 | Y=26.82-0.0256 X | 0.990 0 |
| 4:1 | 19.85 | 19.48 | 19.09 | 18.86 | 18.47 | Y=31.07-0.0338 X | 0.992 4 |
| [Bpy]NO$_3$ | 21.28 | 20.75 | 20.16 | 19.64 | 19.09 | Y=39.57-0.0549 0X | 0.999 6 |
| [Opy]NO$_3$ | 18.13 | 17.88 | 17.71 | 17.43 | 17.16 | Y=26.12-0.0239 6X | 0.999 9 |