**Supplementary Information**

**Molecular Insights of Refrigerants (R32, R125, R134a) In Amino acid based Ionic Liquids**

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**Several molecules with different colors

AI-generated content may be incorrect.**

**Figure S1.** Atomistic labeling of the molecules (a) TBP, (b) Leucine, (c) Isoleucine, (d) R32, (e) R125, (f) R134a.

**Table S1.** Partial atomic charges of the molecules.

|  |  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| **Molecule** | **Atom name** | **Charges** | **σ (nm)** | **ε (kJ/mol)** | **Molecule** | **Atom name** | **Charges** | **σ (nm)** | **ε (kJ/mol)** |
| **R134a** | CE1 | 0.79551 | 3.50E-01 | 2.76E-01 | **R32** | CC1 | 0.49544 | 3.50E-01 | 2.76E-01 |
| CE2 | -0.0298 | 3.50E-01 | 2.76E-01 | FC1 | -0.2763 | 2.90E-01 | 2.51E-01 |
| FE1 | -0.2463 | 2.90E-01 | 2.51E-01 | FC2 | -0.2761 | 2.90E-01 | 2.51E-01 |
| FE2 | -0.2462 | 2.90E-01 | 2.51E-01 | H1C1 | 0.02805 | 2.50E-01 | 1.26E-01 |
| FE3 | -0.2852 | 2.90E-01 | 2.51E-01 | H2C1 | 0.02893 | 2.50E-01 | 1.26E-01 |
| FE4 | -0.2244 | 2.90E-01 | 2.51E-01 | **R125** | CD1 | 0.65617 | 3.50E-01 | 2.76E-01 |
| H1E2 | 0.11834 | 2.50E-01 | 1.26E-01 | CD2 | 0.32562 | 3.50E-01 | 2.76E-01 |
| H2E2 | 0.11805 | 2.50E-01 | 1.26E-01 | FD1 | -0.2338 | 2.90E-01 | 2.51E-01 |
| **TBP** | P | 0.68 | 3.74E-01 | 8.37E-01 | FD2 | -0.234 | 2.90E-01 | 2.51E-01 |
| C1 | -0.31 | 3.50E-01 | 2.76E-01 | FD3 | -0.1914 | 2.90E-01 | 2.51E-01 |
| H1C1 | 0.13 | 2.50E-01 | 1.13E-01 | FD4 | -0.2166 | 2.90E-01 | 2.51E-01 |
| H2C1 | 0.13 | 2.50E-01 | 1.13E-01 | FD5 | -0.2169 | 2.90E-01 | 2.51E-01 |
| C2 | 0.01 | 3.50E-01 | 2.76E-01 | H1D2 | 0.1109 | 2.50E-01 | 1.26E-01 |
| H1C2 | 0.06 | 2.50E-01 | 1.13E-01 | **LEU** | CB1 | -0.518 | 3.50E-01 | 2.76E-01 |
| H2C2 | 0.06 | 2.50E-01 | 1.13E-01 | CB2 | 0.58385 | 3.50E-01 | 2.76E-01 |
| C3 | -0.12 | 3.50E-01 | 2.76E-01 | H1B1 | 0.11283 | 2.50E-01 | 1.26E-01 |
| H1C3 | 0.06 | 2.50E-01 | 1.13E-01 | H2B1 | 0.11618 | 2.50E-01 | 1.26E-01 |
| H2C3 | 0.06 | 2.50E-01 | 1.13E-01 | H3B1 | 0.1166 | 2.50E-01 | 1.26E-01 |
| C4 | -0.18 | 3.50E-01 | 2.76E-01 | CB3 | -0.474 | 3.50E-01 | 2.76E-01 |
| H1C4 | 0.06 | 2.50E-01 | 1.13E-01 | CB4 | 0.39954 | 3.50E-01 | 2.76E-01 |
| H2C4 | 0.06 | 2.50E-01 | 1.13E-01 | H1B4 | 0.1016 | 2.50E-01 | 1.26E-01 |
| H3C4 | 0.06 | 2.50E-01 | 1.13E-01 | H2B4 | 0.13762 | 2.50E-01 | 1.26E-01 |
| C5 | -0.31 | 3.50E-01 | 2.76E-01 | CB5 | 0.78225 | 3.55E-01 | 2.93E-01 |
| H1C5 | 0.13 | 2.50E-01 | 1.13E-01 | CB6 | -0.4485 | 3.50E-01 | 2.76E-01 |
| H2C5 | 0.13 | 2.50E-01 | 1.13E-01 | H1B3 | -0.0785 | 2.50E-01 | 1.26E-01 |
| C6 | 0.01 | 3.50E-01 | 2.76E-01 | H2B3 | 0.09678 | 2.50E-01 | 1.26E-01 |
| H1C6 | 0.06 | 2.50E-01 | 1.13E-01 | H3B3 | 0.08853 | 2.50E-01 | 1.26E-01 |
| H2C6 | 0.06 | 2.50E-01 | 1.13E-01 | H1B2 | 0.10255 | 2.50E-01 | 1.26E-01 |
| C7 | -0.12 | 3.50E-01 | 2.76E-01 | NB1 | -0.9729 | 3.30E-01 | 7.11E-01 |
| H1C7 | 0.06 | 2.50E-01 | 1.13E-01 | H1NB1 | -0.0372 | 2.50E-01 | 1.26E-01 |
| H2C7 | 0.06 | 2.50E-01 | 1.13E-01 | H2NB2 | 0.36959 | 0.00E+00 | 0.00E+00 |
| C8 | -0.18 | 3.50E-01 | 2.76E-01 | H1B5 | 0.35799 | 0.00E+00 | 0.00E+00 |
| H1C8 | 0.06 | 2.50E-01 | 1.13E-01 | OB1 | -0.69 | 3.12E-01 | 7.11E-01 |
| H2C8 | 0.06 | 2.50E-01 | 1.13E-01 | H1OB1 | 0.47018 | 0.00E+00 | 0.00E+00 |
| H3C8 | 0.06 | 2.50E-01 | 1.13E-01 | OB2 | -0.6169 | 2.96E-01 | 8.79E-01 |
| C9 | -0.31 | 3.50E-01 | 2.76E-01 | **ILE** | C1 | -0.3417 | 3.50E-01 | 2.76E-01 |
| H1C9 | 0.13 | 2.50E-01 | 1.13E-01 | C2 | 0.08507 | 3.50E-01 | 2.76E-01 |
| H2C9 | 0.13 | 2.50E-01 | 1.13E-01 | H1C1 | 0.08677 | 2.50E-01 | 1.26E-01 |
| C10 | 0.01 | 3.50E-01 | 2.76E-01 | H2C1 | 0.08681 | 2.50E-01 | 1.26E-01 |
| H1CA | 0.06 | 2.50E-01 | 1.13E-01 | H3C1 | 0.08802 | 2.50E-01 | 1.26E-01 |
| H2CA | 0.06 | 2.50E-01 | 1.13E-01 | C3 | 0.25402 | 3.50E-01 | 2.76E-01 |
| C11 | -0.12 | 3.50E-01 | 2.76E-01 | H1C2 | -0.0006 | 2.50E-01 | 1.26E-01 |
| H1CB | 0.06 | 2.50E-01 | 1.13E-01 | H2C2 | -0.0018 | 2.50E-01 | 1.26E-01 |
| H2CB | 0.06 | 2.50E-01 | 1.13E-01 | C4 | -0.0435 | 3.50E-01 | 2.76E-01 |
| C12 | -0.18 | 3.50E-01 | 2.76E-01 | C5 | 0.96599 | 3.55E-01 | 2.93E-01 |
| H1CC | 0.06 | 2.50E-01 | 1.13E-01 | C6 | -0.3527 | 3.50E-01 | 2.76E-01 |
| H2CC | 0.06 | 2.50E-01 | 1.13E-01 | H1C4 | -0.0172 | 2.50E-01 | 1.26E-01 |
| H3CC | 0.06 | 2.50E-01 | 1.13E-01 | H2C4 | 0.09295 | 2.50E-01 | 1.26E-01 |
| C13 | -0.31 | 3.50E-01 | 2.76E-01 | H3C4 | 0.08664 | 2.50E-01 | 1.26E-01 |
| H1CD | 0.13 | 2.50E-01 | 1.13E-01 | H1C5 | 0.08903 | 2.50E-01 | 1.26E-01 |
| H2CD | 0.13 | 2.50E-01 | 1.13E-01 | N1 | -0.9991 | 3.30E-01 | 7.11E-01 |
| C14 | 0.01 | 3.50E-01 | 2.76E-01 | H1N1 | 0.04692 | 2.50E-01 | 1.26E-01 |
| H1CE | 0.06 | 2.50E-01 | 1.13E-01 | H2N1 | 0.3978 | 0.00E+00 | 0.00E+00 |
| H2CE | 0.06 | 2.50E-01 | 1.13E-01 | H1C3 | 0.3758 | 0.00E+00 | 0.00E+00 |
| C15 | -0.12 | 3.50E-01 | 2.76E-01 | O1 | -0.6578 | 3.12E-01 | 7.11E-01 |
| H1CF | 0.06 | 2.50E-01 | 1.13E-01 | H1O1 | 0.42797 | 0.00E+00 | 0.00E+00 |
| H2CF | 0.06 | 2.50E-01 | 1.13E-01 | O2 | -0.6694 | 2.96E-01 | 8.79E-01 |
| C16 | -0.18 | 3.50E-01 | 2.76E-01 |
| H1CG | 0.06 | 2.50E-01 | 1.13E-01 |
| H2CG | 0.06 | 2.50E-01 | 1.13E-01 |
| H3CG | 0.06 | 2.50E-01 | 1.13E-01 |

In our work, we applied the Green-Kubo method1-3 to calculate the viscosities of as [TBP][LEU], and [TBP][ILE] at room temperature and pressure. These pressure fluctuations are related to viscosity through a mathematical formula known as the Green–Kubo relation.4-7 Initially, we simulate the IL at a fixed temperature. Then we track how molecules move and how pressure changes over time. We use this data to calculate how these forces are correlated at different times. And finally, we integrate these correlations to get the viscosity. This method is often used for complex liquids like ILs because it doesn’t disturb the system, and it gives viscosity values based on the liquid’s natural properties.

Since we are already studying diffusivity, combining it with viscosity data gives a more complete picture of how well these ILs might work in separation processes. By doing so, we can compare which IL has lower viscosity and may allow faster refrigerant movement, and understand if higher viscosity is limiting the diffusion of refrigerants, and support the choice of ILs for better performance in real-world separation systems.

However, there is now direct experimental data on the viscosity of these ILs [TBP][LEU], [TBP][ILE].

**Table S2.** Viscosity of amino-based ILs. The standard deviation values are given in parentheses.

|  |  |  |
| --- | --- | --- |
| **IL combination** | **Simulation (cP)** | **Experimental validation (cP)** |
| **[TBP][LEU]** | 322.445 (±113.46) | 3898h |
| **[TBP][ILE]** | 189.45 (±87.753) | 6058h |

h at 250C.



**Figure S2.** Pair distribution function plots at 0.625M of TBP with anions and refrigerants (a) PTBP with O1Anions, (b) PTBP with O2Anions, (c) PTBP with N1Anions, (d) PTBP with C1Anions, (e) PTBP with C1Refrigerants, (d) PTBP with F1Refrigerants.



**Figure S4.** Pair distribution function plots at 0.625MLEU with refrigerants (a) O1LEU-C1Refrigerants, (b) O1LEU-F1Refrigerants, (c) O2LEU-C1Refrigerants, (d) O2LEU-F1Refrigerants, (e) N1LEU-C1Refrigerants, (f) N1LEU-F1Refrigerants.



**Figure S5.** Pair distribution function plots at 0.625Mof ILE with refrigerants (a) O1ILE-C1Refrigerants, (b) O1ILE-F1Refrigerants, (c) O2ILE-C1Refrigerants, (d) O2ILE-F1Refrigerants, (e) N1ILE-C1Refrigerants, (f) N1ILE-F1Refrigerants.

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AI-generated content may be incorrect.**

**Figure S6.** Pair distribution function plots at 0.142M of C1 anions of refrigerants (a) C1LEU-C1Refrigerants, (b) C1LEU-F1Refrigerants, (c) C1ILE-C1Refrigerants, (d) C1ILE-F1Refrigerants.

A group of graphs showing different colors

AI-generated content may be incorrect.

**Figure S7.** Pair distribution function plots at 0.625M of C1 anions of refrigerants (a) C1LEU-C1Refrigerants, (b) C1LEU-F1Refrigerants, (c) C1ILE-C1Refrigerants, (d) C1ILE-F1Refrigerants.

**Table S3.** RDF coordination number (Ncr) with different combinations of solvent systems.

|  |  |  |  |
| --- | --- | --- | --- |
| **System (with 0.142M)** | **Pairs** | **Peak Positions (rmax)** | **Coordination number (Ncr)** |
| **R32+LEU+TBP** | **CC1-OB1** | 0.37 | 0.068 |
| **CC1-OB2** | 0.36 | 0.057 |
| **CC1-NB1** | 0.40 | 0.120 |
| **CC1-CB1** | 0.41 | 0.133 |
| **FC1-OB1** | 0.46 | 0.176 |
| **FC1-OB2** | 0.33 | 0.024 |
| **FC1-NB1** | 0.30 | 0.022 |
| **FC1-CB1** | 0.37 | 0.059 |
| **P-CC1** | 0.54 | 0.173 |
| **P-FC1** | 0.45 | 0.099 |
| **P-OB1** | 0.46 | 0.206 |
| **P-OB2** | 0.45 | 0.217 |
| **P-NB1** | 0.43 | 0.141 |
| **P-CB1** | 0.70 | 1.774 |
| **R125+LEU+TBP** | **CD1-OB1** | 0.46 | 0.120 |
| **CD1-OB2** | 0.42 | 0.068 |
| **CD1-NB1** | 0.43 | 0.099 |
| **CD1-CB1** | 0.46 | 0.163 |
| **FD1-OB1** | 0.35 | 0.028 |
| **FD1-OB2** | 0.33 | 0.021 |
| **FD1-NB1** | 0.32 | 0.022 |
| **FD1-CB1** | 0.36 | 0.045 |
| **P-CD1** | 0.58 | 0.227 |
| **P-FD1** | 0.48 | 0.088 |
| **P-OB1** | 0.46 | 0.203 |
| **P-OB2** | 0.45 | 0.207 |
| **P-NB1** | 0.43 | 0.144 |
| **P-CB1** | 0.71 | 1.886 |
| **R134a+LEU+TBP** | **CE1-OB1** | 0.47 | 0.141 |
| **CE1-OB2** | 0.46 | 0.118 |
| **CE1-NB1** | 0.43 | 0.095 |
| **CE1-CB1** | 0.46 | 0.165 |
| **FE1-OB1** | 0.34 | 0.022 |
| **FE1-OB2** | 0.32 | 0.014 |
| **FE1-NB1** | 0.32 | 0.020 |
| **FE1-CB1** | 0.37 | 0.058 |
| **P-CE1** | 0.56 | 0.201 |
| **P-FE1** | 0.49 | 0.110 |
| **P-OB1** | 0.46 | 0.199 |
| **P-OB2** | 0.44 | 0.180 |
| **P-NB1** | 0.43 | 0.134 |
| **P-CB1** | 0.71 | 1.888 |
| **R32+ILE+TBP** | **CC1-O1** | 0.36 | 0.081 |
| **CC1-O2** | 0.34 | 0.054 |
| **CC1-N1** | 0.40 | 0.160 |
| **CC1-C1** | 0.44 | 0.392 |
| **FC1-O1** | 0.45 | 0.202 |
| **FC1-O2** | 0.44 | 0.143 |
| **FC1-N1** | 0.30 | 0.037 |
| **FC1-C1** | 0.36 | 0.142 |
| **P-CC1** | 0.54 | 0.169 |
| **P-FC1** | 0.45 | 0.092 |
| **P-O1** | 0.45 | 0.318 |
| **P-O2** | 0.41 | 0.201 |
| **P-N1** | 0.47 | 0.166 |
| **P-C1** | 0.18 | 0.593 |
| **R125+ILE+TBP** | **CD1-O1** | 0.41 | 0.078 |
| **CD1-O2** | 0.40 | 0.065 |
| **CD1-N1** | 0.43 | 0.127 |
| **CD1-C1** | 0.48 | 0.449 |
| **FD1-O1** | 0.33 | 0.026 |
| **FD1-O2** | 0.34 | 0.025 |
| **FD1-N1** | 0.32 | 0.030 |
| **FD1-C1** | 0.37 | 0.123 |
| **P-CD1** | 0.57 | 0.183 |
| **P-FD1** | 0.48 | 0.084 |
| **P-O1** | 0.45 | 0.317 |
| **P-O2** | 0.41 | 0.197 |
| **P-N1** | 0.47 | 0.164 |
| **P-C1** | 0.18 | 0.593 |
| **R134a+ILE+TBP** | **CE1-O1** | 0.42 | 0.091 |
| **CE1-O2** | 0.45 | 0.123 |
| **CE1-N1** | 0.42 | 0.120 |
| **CE1-C1** | 0.47 | 0.421 |
| **FE1-O1** | 0.33 | 0.025 |
| **FE1-O2** | 0.54 | 0.273 |
| **FE1-N1** | 0.32 | 0.031 |
| **FE1-C1** | 0.37 | 0.128 |
| **P-CE1** | 0.56 | 0.190 |
| **P-FE1** | 0.48 | 0.089 |
| **P-O1** | 0.45 | 0.314 |
| **P-O2** | 0.41 | 0.198 |
| **P-N1** | 0.47 | 0.164 |
| **P-C1** | 0.18 | 0.594 |
| **System (with 0.625M)** | **Pairs** | **Peak Positions (rmax)** | **Coordination number (Ncr)** |
| **R32+LEU+TBP** | **CC1-OB1** | 0.37 | 0.045 |
| **CC1-OB2** | 0.35 | 0.028 |
| **CC1-NB1** | 0.40 | 0.074 |
| **CC1-CB1** | 0.42 | 0.111 |
| **FC1-OB1** | 0.46 | 0.116 |
| **FC1-OB2** | 0.35 | 0.025 |
| **FC1-NB1** | 0.30 | 0.012 |
| **FC1-CB1** | 0.38 | 0.058 |
| **P-CC1** | 0.54 | 1.078 |
| **P-FC1** | 0.45 | 0.626 |
| **P-OB1** | 0.47 | 0.124 |
| **P-OB2** | 0.43 | 0.091 |
| **P-NB1** | 0.43 | 0.103 |
| **P-CB1** | 0.73 | 1.442 |
| **R125+LEU+TBP** | **CD1-OB1** | 0.46 | 0.083 |
| **CD1-OB2** | 0.46 | 0.075 |
| **CD1-NB1** | 0.43 | 0.058 |
| **CD1-CB1** | 0.46 | 0.126 |
| **FD1-OB1** | 0.34 | 0.017 |
| **FD1-OB2** | 0.32 | 0.011 |
| **FD1-NB1** | 0.33 | 0.016 |
| **FD1-CB1** | 0.36 | 0.036 |
| **P-CD1** | 0.58 | 1.331 |
| **P-FD1** | 0.48 | 0.526 |
| **P-OB1** | 0.47 | 0.103 |
| **P-OB2** | 0.44 | 0.096 |
| **P-NB1** | 0.43 | 0.100 |
| **P-CB1** | 0.72 | 1.215 |
| **R134a+LEU+TBP** | **CE1-OB1** | 0.47 | 0.104 |
| **CE1-OB2** | 0.46 | 0.81 |
| **CE1-NB1** | 0.42 | 0.049 |
| **CE1-CB1** | 0.46 | 0.128 |
| **FE1-OB1** | 0.34 | 0.018 |
| **FE1-OB2** | 0.33 | 0.014 |
| **FE1-NB1** | 0.32 | 0.013 |
| **FE1-CB1** | 0.36 | 0.037 |
| **P-CE1** | 0.56 | 1.164 |
| **P-FE1** | 0.49 | 0.642 |
| **P-OB1** | 0.47 | 0.103 |
| **P-OB2** | 0.43 | 0.078 |
| **P-NB1** | 0.43 | 0.100 |
| **P-CB1** | 0.71 | 1.126 |
| **R32+ILE+TBP** | **CC1-O1** | 0.36 | 0.057 |
| **CC1-O2** | 0.34 | 0.037 |
| **CC1-N1** | 0.40 | 0.109 |
| **CC1-C1** | 0.45 | 0.314 |
| **FC1-O1** | 0.46 | 0.173 |
| **FC1-O2** | 0.44 | 0.111 |
| **FC1-N1** | 0.30 | 0.024 |
| **FC1-C1** | 0.36 | 0.100 |
| **P-CC1** | 0.54 | 1.039 |
| **P-FC1** | 0.45 | 0.578 |
| **P-O1** | 0.46 | 0.280 |
| **P-O2** | 0.41 | 0.165 |
| **P-N1** | 0.48 | 0.130 |
| **P-C1** | 0.18 | 0.594 |
| **R125+ILE+TBP** | **CD1-O1** | 0.42 | 0.080 |
| **CD1-O2** | 0.45 | 0.107 |
| **CD1-N1** | 0.43 | 0.095 |
| **CD1-C1** | 0.47 | 0.270 |
| **FD1-O1** | 0.33 | 0.024 |
| **FD1-O2** | 0.34 | 0.023 |
| **FD1-N1** | 0.32 | 0.023 |
| **FD1-C1** | 0.37 | 0.091 |
| **P-CD1** | 0.58 | 1.265 |
| **P-FD1** | 0.48 | 0.498 |
| **P-O1** | 0.45 | 0.221 |
| **P-O2** | 0.41 | 0.158 |
| **P-N1** | 0.47 | 0.092 |
| **P-C1** | 0.18 | 0.594 |
| **R134a+ILE+TBP** | **CE1-O1** | 0.46 | 0.142 |
| **CE1-O2** | 0.45 | 0.099 |
| **CE1-N1** | 0.42 | 0.086 |
| **CE1-C1** | 0.47 | 0.296 |
| **FE1-O1** | 0.33 | 0.023 |
| **FE1-O2** | 0.55 | 0.250 |
| **FE1-N1** | 0.33 | 0.029 |
| **FE1-C1** | 0.37 | 0.093 |
| **P-CE1** | 0.56 | 1.095 |
| **P-FE1** | 0.48 | 0.515 |
| **P-O1** | 0.46 | 0.263 |
| **P-O2** | 0.41 | 0.158 |
| **P-N1** | 0.48 | 0.116 |
| **P-C1** | 0.18 | 0.595 |

**A group of graphs showing different colors

AI-generated content may be incorrect.**

**Figure S8.** Centre of mass of Pair distribution function plots at 0.142M of (a) TBP-TBP, (b) TBP-LEU, (c) TBP-ILE, (d) LEU-LEU, (e) ILE-ILE.

In the label of graph, R32LT: system [R32] [LEU][TBP], R32IT: system [R32] [ILE][TBP], R125LT: system [R125] [LEU][TBP], R125IT: system [R125] [ILE][TBP], R134aLT: system [R134a] [LEU][TBP], R134aIT: system [R134a] [ILE][TBP].

A group of graphs showing different colors

AI-generated content may be incorrect.

**Figure S9.** Centre of mass of Pair distribution function plots at 0.625M of (a) TBP-R32, (b) TBP-R125, (c) TBP-R134a, (d) R32-R32, (e) R125-R125, (f) R134a-R134a, (g) TBP-LEU, (h) TBP-ILE.

A group of graphs showing different colored lines

AI-generated content may be incorrect.

**Figure S10.** Centre of mass of Pair distribution function plots at 0.625M of (a) LEU-REFRIG, (b) ILE-REFRIG, (c) TBP-TBP, (d) LEU-LEU, (e) ILE-ILE.

**Table S4.** RDF COM coordination number (Ncr) with different combinations of solvent systems.

|  |  |  |  |
| --- | --- | --- | --- |
| **System (with 0.142M)** | **Pairs** | **Peak Positions (rmax)** | **Coordination number (Ncr)** |
| **R32+LEU+TBP** | **TBP-R32** | 0.51 | 0.156 |
| **TBP-LEU** | 0.61 | 0.755 |
| **TBP-TBP** | 0.96 | 2.921 |
| **LEU-LEU** | 0.64 | 1.562 |
| **LEU-R32** | 0.52 | 0.242 |
| **R32-R32** | 0.45 | 0.155 |
| **R125+LEU+TBP** | **TBP-R125** | 0.59 | 0.246 |
| **TBP-LEU** | 0.61 | 0.758 |
| **TBP-TBP** | 0.97 | 3.075 |
| **LEU-R125** | 0.55 | 0.216 |
| **LEU-LEU** | 0.64 | 1.529 |
| **R125-R125** | 0.52 | 0.176 |
| **R134a+LEU+TBP** | **TBP-R134a** | 0.56 | 0.185 |
| **TBP-LEU** | 0.61 | 0.747 |
| **TBP-TBP** | 0.96 | 2.852 |
| **LEU-LEU** | 0.64 | 1.599 |
| **LEU-R134a** | 0.57 | 0.295 |
| **R134a-R134a** | 0.52 | 0.202 |
| **R32+ILE+TBP** | **TBP-R32** | 0.52 | 0.174 |
| **TBP-ILE** | 0.61 | 0.637 |
| **TBP-TBP** | 0.94 | 2.834 |
| **ILE-R32** | 0.56 | 0.516 |
| **ILE-ILE** | 0.55 | 0.600 |
| **R32-R32** | 0.45 | 0.148 |
| **R125+ILE+TBP** | **TBP-R125** | 0.59 | 0.236 |
| **TBP-ILE** | 0.61 | 0.625 |
| **TBP-TBP** | 0.94 | 2.747 |
| **ILE-R125** | 0.59 | 0.499 |
| **ILE-ILE** | 0.55 | 0.611 |
| **R125-R125** | 0.53 | 0.196 |
| **R134a+ILE+TBP** | **TBP-R134a** | 0.57 | 0.211 |
| **TBP-ILE** | 0.61 | 0.629 |
| **TBP-TBP** | 0.95 | 3.000 |
| **ILE-R134a** | 0.59 | 0.517 |
| **ILE-ILE** | 0.55 | 0.604 |
| **R134a-R134a** | 0.52 | 0.187 |
| **System (with 0.625M)** | **Pairs** | **Peak Positions (rmax)** | **Coordination number (Ncr)** |
| **R32+LEU+TBP** | **TBP-R32** | 0.52 | 1.183 |
| **TBP-LEU** | 0.61 | 0.444 |
| **TBP-TBP** | 0.99 | 2.159 |
| **LEU-R32** | 0.52 | 0.167 |
| **LEU-LEU** | 0.64 | 2.011 |
| **R32-R32** | 0.45 | 1.090 |
| **R125+LEU+TBP** | **TBP-R125** | 0.60 | 1.712 |
| **TBP-LEU** | 0.61 | 0.399 |
| **TBP-TBP** | 1.030 | 2.174 |
| **LEU-R125** | 0.55 | 0.162 |
| **LEU-LEU** | 0.64 | 2.173 |
| **R125-R125** | 0.52 | 1.189 |
| **R134a+LEU+TBP** | **TBP-R134a** | 0.58 | 1.542 |
| **TBP-LEU** | 0.61 | 0.396 |
| **TBP-TBP** | 1.010 | 2.071 |
| **LEU-R134a** | 0.56 | 0.197 |
| **LEU-LEU** | 0.64 | 2.127 |
| **R134a-R134a** | 0.52 | 1.387 |
| **R32+ILE+TBP** | **TBP-R32** | 0.52 | 1.102 |
| **TBP-ILE** | 0.61 | 0.421 |
| **TBP-TBP** | 0.97 | 2.064 |
| **ILE-R32** | 0.57 | 0.426 |
| **ILE-ILE** | 0.55 | 0.623 |
| **R32-R32** | 0.45 | 1.066 |
| **R125+ILE+TBP** | **TBP-R125** | 0.60 | 1.629 |
| **TBP-ILE** | 0.62 | 0.462 |
| **TBP-TBP** | 1.000 | 1.915 |
| **ILE-R125** | 0.59 | 0.401 |
| **ILE-ILE** | 0.55 | 0.627 |
| **R125-R125** | 0.52 | 1.136 |
| **R134a+ILE+TBP** | **TBP-R134a** | 0.58 | 1.452 |
| **TBP-ILE** | 0.62 | 0.469 |
| **TBP-TBP** | 0.99 | 1.955 |
| **ILE-R134a** | 0.59 | 0.403 |
| **ILE-ILE** | 0.55 | 0.639 |
| **R134a-R134a** | 0.52 | 1.336 |

**A group of graphs showing different colored lines

AI-generated content may be incorrect.**

**Figure S11.** Mean square displacement at 0.142M with (a) R32 with Anions, (b) R125 with Anions, (c) R134a with Anions, (d)TBP with R32, (e) TBP with R125, and (f) TBP with R134a.

A group of graphs showing different colored lines

AI-generated content may be incorrect.

**Figure S12.** Mean square displacement at 0.625M with (a)TBP with R32, (b) TBP with R125, (c) TBP with R134a, (d) R32 with Anions, (e) R125 with Anions, and (f) R134a with Anions.

**Table S5.** Diffusion behavior of Refrigerants (R32, R125, and R134a) with different combinations of cation and anions in the system. The standard deviation values are given in parentheses.

|  |  |  |  |  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| **Self-diffusivity, D\*105; cm2/sec** | | | | | | | | | | | | |
| **Mixed system** | **R32** | | **R125** | | **R134a** | | **TBP** | | **LEU** | | **ILE** | |
| **System 0.142M** | | | | | | | | | | | | |
| **R32** | 0.883 | (0.043) | - | | - | | 0.036 | (0.006) | 0.017 | (0.000) | - | |
| **R125** | - | | 0.372 | (0.024) | - | | 0.026 | (0.001) | 0.014 | (0.000) | - | |
| **R134a** | - | | - | | 0.332 | (0.022) | 0.024 | (0.002) | 0.012 | (0.002) | - | |
| **R32** | 1.144 | (0.121) | - | | - | | 0.123 | (0.007) | - | | 0.141 | (0.009) |
| **R125** | - | | 0.534 | (0.045) | - | | 0.097 | (0.003) | - | | 0.111 | (0.002) |
| **R134a** | - | | - | | 0.571 | (0.027) | 0.108 | (0.003) | - | | 0.131 | (0.001) |
| **System 0.625M** | | | | | | | | | | | | |
| **R32** | 2.793 | (0.046) | - | | - | | 0.314 | (0.012) | 0.078 | (0.003) | - | |
| **R125** | - | | 0.819 | (0.029) | - | | 0.134 | (0.005) | 0.040 | (0.005) | - | |
| **R134a** | - | | - | | 0.922 | (0.014) | 0.181 | (0.005) | 0.044 | (0.003) | - | |
| **R32** | 2.783 | (0.023) | - | | - | | 0.437 | (0.043) | - | | 0.481 | (0.037) |
| **R125** | - | | 0.952 | (0.012) | - | | 0.255 | (0.018) | - | | 0.276 | (0.053) |
| **R134a** | - | | - | | 1.055 | (0.056) | 0.229 | (0.017) | - | | 0.277 | (0.020) |

For the validation of Self-diffusivity results, the available data is provided below:

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| **Molecules** | **Simulation Self-diffusivity, D\*105; cm2/sec** | | **Literature reported ‘D’ values of different refrigerants, cation, and anions ;(105 cm2/sec)** | |
| **DCation** | **DAnion** | **DCation** | **DAnion** |
| **TBP + LEU** | 0.0192 (0.0036) | 0.0162 (0.0021) | 0.1101 (0.0093)9d | 0.1144 (0.0091)9d |
| **TBP + ILE** | 0.0876 (0.0037) | 0.1010 (0.0086) | - | - |

d at 442 K.

**Table S6.** Free-energy solvation in the mixture and ionic liquid. The standard deviation values are given in parentheses.

|  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- |
| **System** | **Ele, (KJ/mol)** | | **vdW, (KJ/mol)** | | **ΔGsolvation, (KJ/mol)** | |
| **R32LT** | -8.38 | (0.09) | 1.98 | (0.76) | -6.4 | (0.85) |
| **R125LT** | -8.92 | (0.2) | 0.28 | (0.55) | -8.64 | (0.75) |
| **R134aLT** | -9.78 | (0.09) | -2.08 | (0.57) | -11.86 | (0.66) |
| **R32IT** | -9.99 | (0.38) | 1.6 | (0.34) | -8.39 | (0.72) |
| **R125IT** | -8.92 | (0.53) | -3.64 | (0.65) | -12.56 | (1.18) |
| **R134aIT** | -9.51 | (0.39) | -1.99 | (0.81) | -11.5 | (1.2) |

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AI-generated content may be incorrect.

**Figure S13.** The dU/dλ as a function of (a) λele for Refrigerants, (b) λvdw for Refrigerants in the presence of LEU+TBP system, (c) λele for Refrigerants, (d) λvdw for Refrigerants in the presence of ILE+TBP system.

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