Electronic Supplementary Material (ESI) for Green Chemistry.

Supporting Information

Structure-Property Reduced Order Model for Viscosity Prediction in Single-Component CO_2-Binding Organic Liquids

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Compound Directory

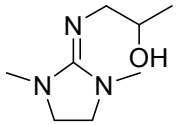
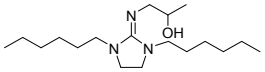
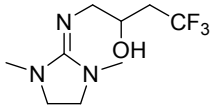
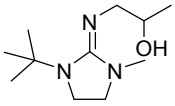
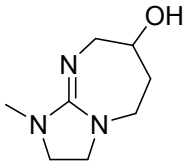
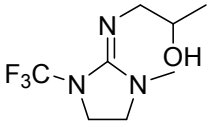
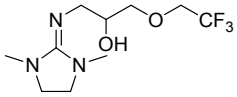
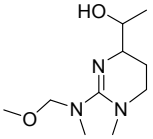
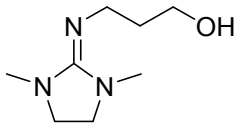
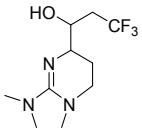
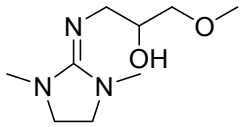
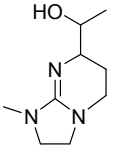
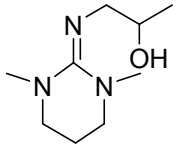
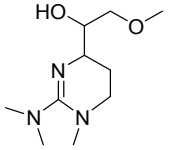
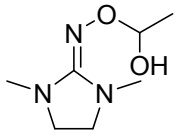
1-IPADM-2-BOL  1-((1,3-dimethylimidazolidin-2-ylidene)-amino)-propan-2-ol	**1-IPADH-2-BOL**  1-((1,3-dihexylimidazolidin-2-ylidene)amino)propan-2-ol	**1-TFMIPADM-2-BOL**  1-((1,3-dimethylimidazolidin-2-ylidene)amino)-4,4,4-trifluorobutan-2-ol	**1-IPATBM-2-BOL**  1-((1-(*tert*-butyl)-3-methylimidazolidin-2-ylidene)amino)propan-2-ol
1-EAμEM-2-BOL  1-methyl-2,3,5,6,7,8-hexahydro-1*H*-imidazo[1,2-*a*][1,3]diazepin-7-ol	**1-IPAMTFM-2-BOL**  1-((1-methyl-3-(trifluoromethyl)imidazolidin-2-ylidene)amino)propan-2-ol	**1-TFEEIPADM-2-BOL**  1-((1,3-dimethylimidazolidin-2-ylidene)amino)-3-(2,2,2-trifluoroethoxy)propan-2-ol	**1-IPAμEMMEMM-BOL**  1-(2-((methoxymethyl)(methyl)amino)-1-methyl-1,4,5,6-tetrahydropyrimidin-4-yl)ethan-1-ol
1-PADM-2-BOL  3-((1,3-dimethylimidazolidin-2-ylidene)amino)propan-1-ol	**1-TFMIPAμEMDM-BOL**  1-(2-(dimethylamino)-1-methyl-1,4,5,6-tetrahydropyrimidin-4-yl)-3,3,3-trifluoropropan-1-ol	**1-MEIPADM-2-BOL**  1-((1,3-dimethylimidazolidin-2-ylidene)amino)-3-methoxypropan-2-ol	**1-IPAμEM-2-BOL**  1-(1-methyl-1,2,3,5,6,7-hexahydroimidazo[1,2-*a*]pyrimidin-7-yl)ethan-1-ol
1-IPADM-3-BOL  1-((1,3-dimethyltetrahydropyrimidin-2(1*H*)-ylidene)amino)propan-2-ol	**1-MEIPAμEMDM-BOL**  1-(2-(dimethylamino)-1-methyl-1,4,5,6-tetrahydropyrimidin-4-yl)-2-methoxyethan-1-ol	**1-O-EADM-2-BOL**  1,3-dimethylimidazolidin-2-one *O*-(1-hydroxyethyl) oxime	

Table S1 MD calculated viscosities at 25% mol CO_2 loading and P_{int} values

Compound	**$P_{int,MD}$ (expressed as fraction of 1)**	**Viscosity**	**Method**
1-EAμEM-2-BOL	0.001	1032 *+628 -592*	Multiple temperatures
1-PADM-2-BOL	0.001	950 *+556 -505*	Multiple temperatures
1-IPATBM-2-BOL	0.02	499 *+57 -103*	Multiple temperatures
1-IPADH-2-BOL	0.06	341 *+120 -120*	Green-Kubo
1-IPAMTFM-2-BOL	0.13	328 *+415 -117*	Non-Equilibrium
1-TFMIPAμEMDM-BOL	0.15	322 *+162 -62*	Multiple temperatures
1-IPADM-3-BOL	0.21	190 *+346 -75*	Non-Equilibrium
1-IPAμEMMEMM-BOL	0.25	310 *+75 -75*	Green-Kubo
1-IPADM-2-BOL	0.34	149 *+22 -16*	Non-Equilibrium
1-TFMIPADM-2-BOL	0.34	65 *+13 -10*	Non-Equilibrium
1-TFEEIPADM-2-BOL	0.35	122 *+66 -32*	Non-Equilibrium
1-MEIPADM-2-BOL	0.52	114 *+22 -16*	Non-Equilibrium
1-MEIPAμEMDM-BOL	0.92	45 *+8 -6*	Non-Equilibrium

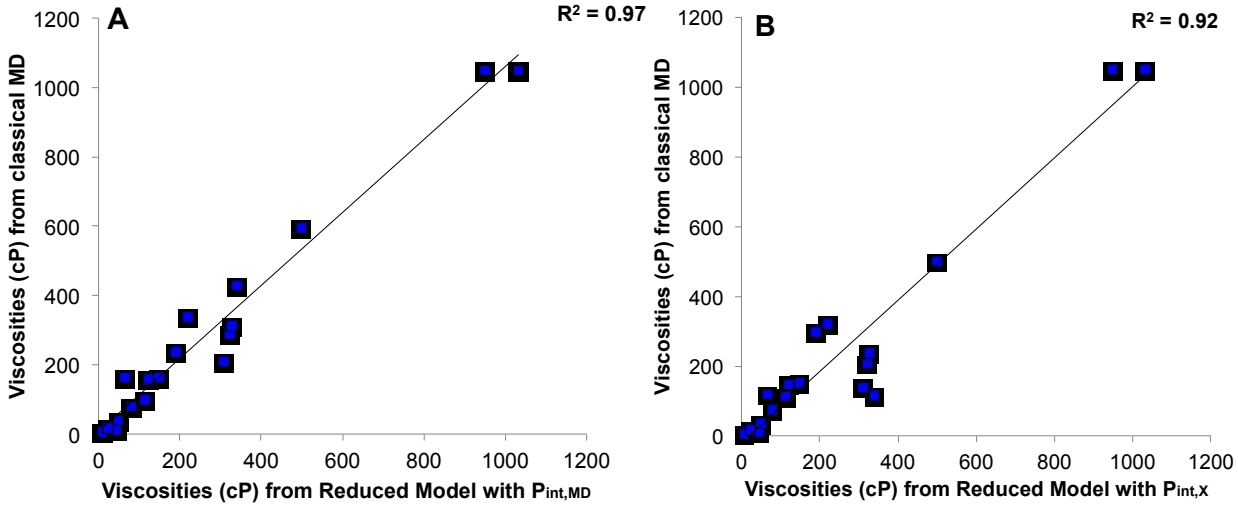
Error estimation

Viscosities calculated using the Green-Kubo approach by calculating the integral of the pressure tensor autocorrelation function. Calculation was done with the P_{XZ}, P_{XY}, and P_{YZ} tensors independently, and error estimated as the largest viscosity difference obtained between two tensors components. The non-equilibrium method was used as implemented in the g_energy program in the GROMACS package to obtain 1/viscosity values after applying an acceleration. Error calculated based on the 1/viscosity error estimate as reported in the output, that result in different + and – variations. For some compounds the Green Kubo approach was used to calculate viscosities at higher temperatures, and a exponential fit of multiple temperature points was done to estimate it at 40 °C. The lower and upper bound values for each point were also fit to an exponential to estimate the error at 40 °C. See Methods in main text for references.

Table S2 Compound structural parameters obtained from single molecular geometry optimizations with density functional theory, calculated X and $P_{int,X}$ values, as well as $P_{int,MD}$ values.

Compound	$P_{int,MD}$	Atomic Distances			Atomic Charges			X	$P_{int,X}$
		r_{OH}	r_{NH}	r_{NO}	q_H	q_O	q_N		
1-EAμEM-2-BOL	0.001	5.72463	When rOH>2.0 Å, $P_{int,X}$=0.001						0.001
1-PADM-2-BOL	0.001	4.45067							0.001
1-IPATBM-2-BOL	0.02	1.80186	1.03001	2.76226	0.28049	-0.82837	-0.35288	-0.11922	0.04
1-IPADH-2-BOL	0.06	1.73115	1.03772	2.71842	0.40616	-0.75798	-0.31995	-0.21385	0.50
1-IPAMTFM-2-BOL	0.13	1.68195	1.04789	2.67029	0.32287	-0.81257	-0.27905	-0.15705	0.22
1-TFMIPAμEMDM-BOL	0.15	1.75565	1.03198	2.71072	0.33625	-0.82571	-0.42826	-0.16723	0.27
1-IPADM-3-BOL	0.21	1.80903	1.02865	2.78317	0.31035	-0.78627	-0.36978	-0.14199	0.15
1-IPAμEMMEMM-BOL	0.25	1.74257	1.03372	2.69729	0.36683	-0.79730	-0.51667	-0.19846	0.43
1-IPADM-2-BOL	0.34	1.70861	1.03729	2.69645	0.36249	-0.82774	-0.39748	-0.21269	0.40
1-TFMIPADM-2-BOL	0.34	1.72621	1.03677	2.70966	0.38492	-0.81579	-0.43850	-0.19250	0.50
1-TFEEIPADM-2-BOL	0.35	1.79571	1.02929	2.75233	0.37296	-0.78458	-0.39791	-0.19371	0.40
1-MEIPADM-2-BOL	0.52	1.67597	1.03996	2.67136	0.38675	-0.80303	-0.42638	-0.24648	0.51
1-MEIPAμEMDM-BOL	0.61	1.75289	1.03251	2.70630	0.38199	-0.86820	-0.54931	-0.21620	0.51
1-IPAμEM-2-BOL	0.80	1.69120	1.03967	2.68318	0.43792	-0.83601	-0.57676	-0.27971	0.82
1-O-EADM-2-BOL	0.92	1.63505	1.04931	2.65504	0.47754	-0.84997	-0.48991	-0.31437	0.99

Figure S1 Classical MD viscosities versus reduced model viscosities using $P_{int,MD}$ (A) and $P_{int,X}$ (B).



Additionally, we calculated the average absolute percent deviation (AAPD) for the full data sets shown above:

$$AAPD = \frac{100}{N} * \sum_{i=1}^{N} \frac{\left|\eta_i^{model} - \eta_i^{MD}\right|}{\eta_i^{MD}}$$

where *N* is the number of different solvent compounds in each data set. The calculated AADP values are 29 % between the classical MD viscosities and those estimated with the reduced model with $P_{int,MD}$, and 31% between the classical MD viscosities and those estimated with the reduced model with $P_{int,X}$, showing ~70% fidelity in the prediction regardless of the method. However, we note that the AAPD analysis is not optimal because the percent deviations at the low viscosity regime are not as significant as those in the high viscosity regime.

Table S3 CO_2 capture ΔΔE values with respect to the capture energy of IPADM-2-BOL.

Compound	**ΔΔE (kJ/mol)**
1-EAμEM-2-BOL	+16.8
1-PADM-2-BOL	-31.1
1-IPATBM-2-BOL	+2.5
1-IPADH-2-BOL	-21.9
1-IPAMTFM-2-BOL	-13.0
1-TFMIPAμEMDM-BOL	+9.6
1-IPADM-3-BOL	-15.5
1-IPAμEMMEMM-BOL	+4.1
1-IPADM-2-BOL	0.00
1-TFMIPADM-2-BOL	+5.6
1-TFEEIPADM-2-BOL	-17.4
1-MEIPADM-2-BOL	-8.3
1-MEIPAμEMDM-BOL	+0.9
1-IPAμEM-2-BOL	+16.4
1-O-EADM-2-BOL	-1.2