Supporting Information: Active-Learning Assisted General Framework for Efficient Parameterization of Force-Fields

Yati[1], Yash Kokane[2], and Anirban Mondal[1,*]

[1]Department of Chemistry, Indian Institute of Technology Gandhinagar, Gujarat, 382355, India

[1]Department of Materials Engineering, Indian Institute of Technology Gandhinagar, Gujarat, 382355, India

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E-mail: amondal@iitgn.ac.in

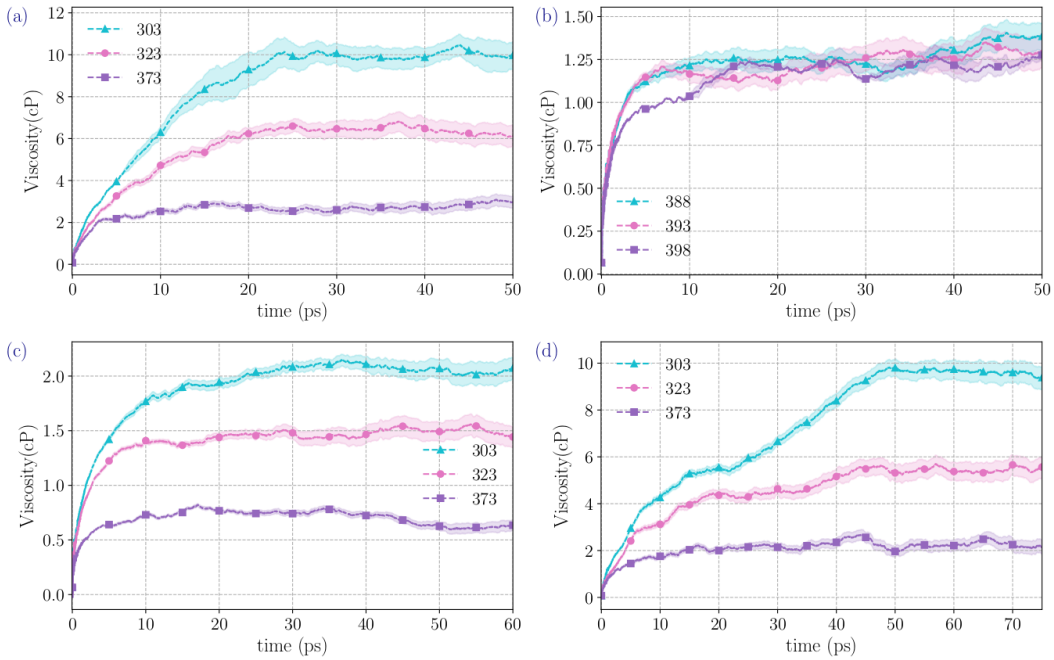

Figure S1: Running integral (see Eq. 6 of the main manuscript) representing the shear viscosities of sulfolane molecules from five independent trajectories at different temperatures: (a) SL, (b) DMS, (c) EMS, and (d) MSL. The shaded regions are the standard error of the mean.

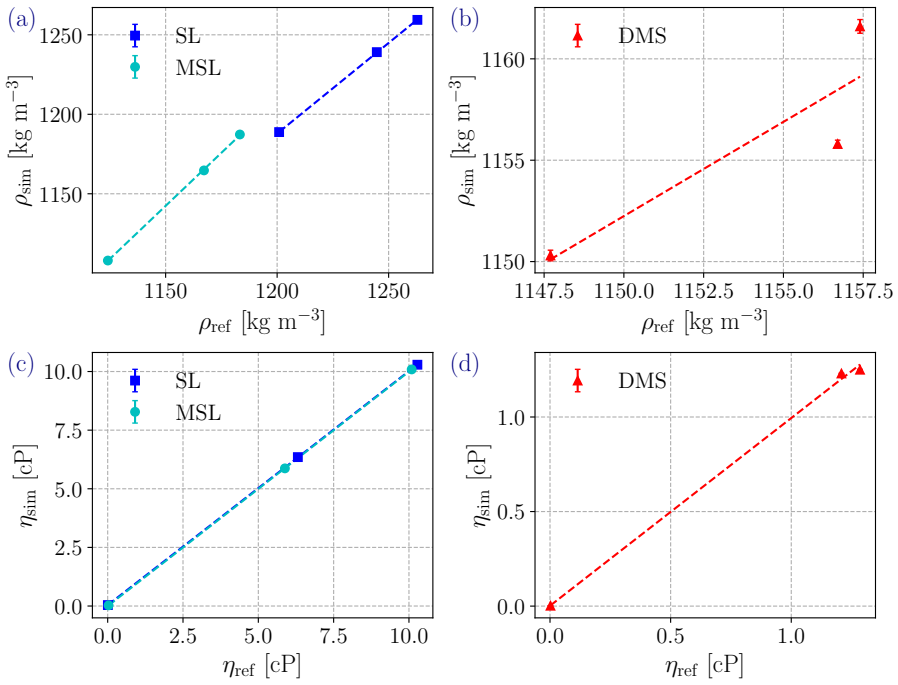

Figure S2: Density (top panel) and viscosity (bottom panel) of sulfolane molecules obtained from simulations employing the optimized force field (GA-GPR) parameters and compared against experimental measurements.

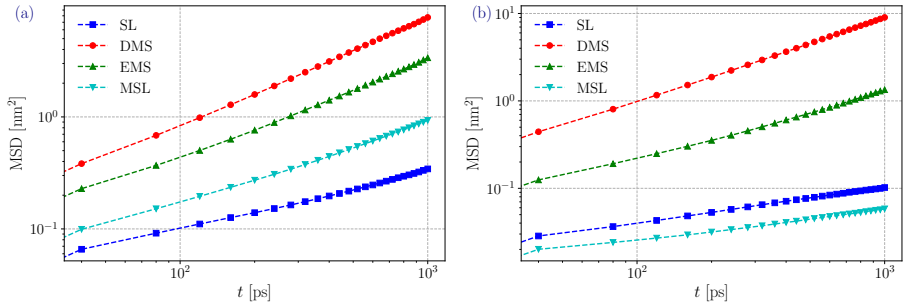

Figure S3: Mean square displacement (MSD) of center of mass as a function of time obtained from simulations employing (a) the optimized (GA-GPR) and (b) OPLS force field parameters.

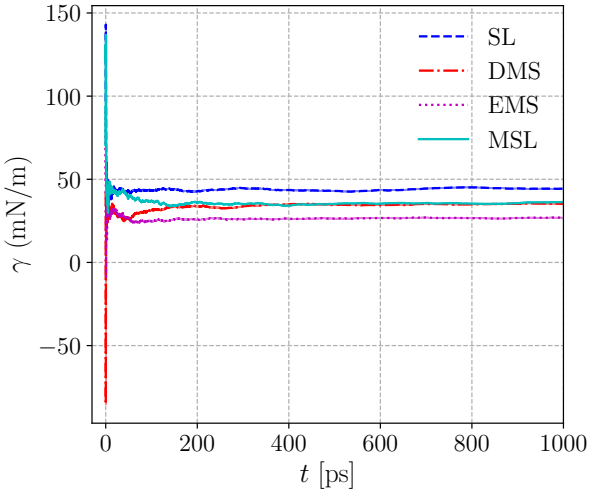

Figure S4: Running average surface tension profiles in (a) SL, (b) DMS, (c) EMS, and (d) MSL obtained from simulations employing the optimized force field (GA-GPR) parameters.

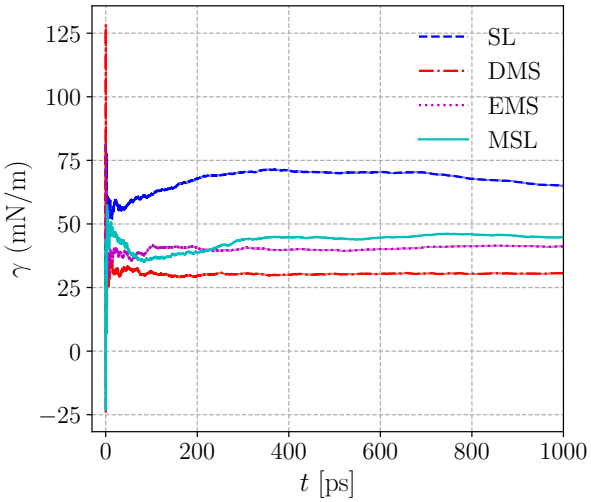

Figure S5: Running average surface tension profiles in (a) SL, (b) DMS, (c) EMS, and (d) MSL obtained from simulations employing the OPLS force field parameters.

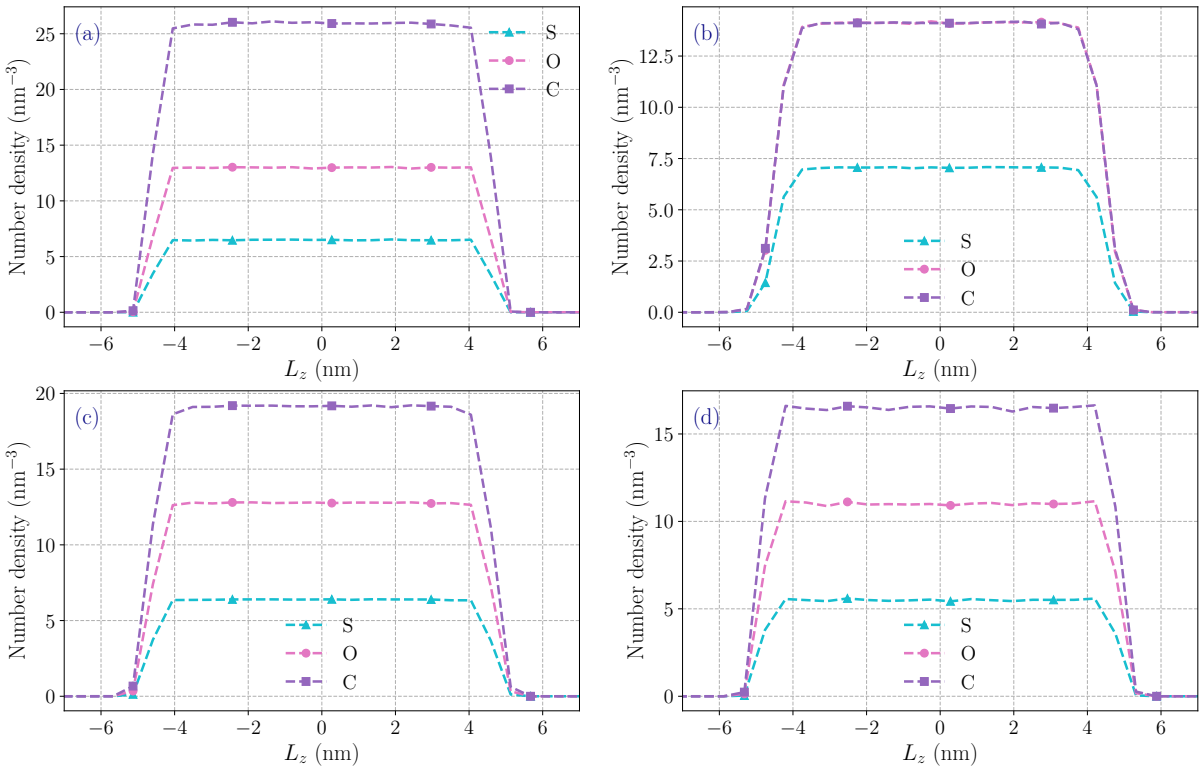

Figure S6: Density profiles of atom types in sulfolane molecules obtained from simulations employing the OPLS force field: (a) SL, (b) DMS, (c) EMS, and (d) MSL.

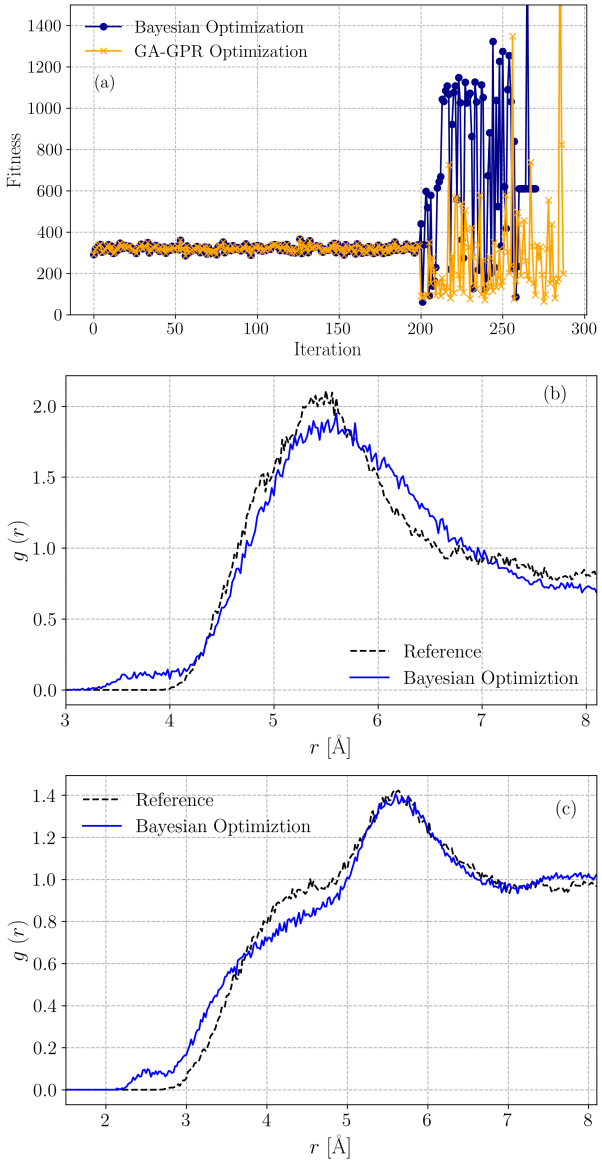

Figure S7: (a) Evolution of the fitness function during Bayesian optimization and GA-GPR training: illustrates the progressive improvement in the fitness function as new individuals are added using active learning. Radial distribution function obtained using best parameters (lowest fitness) from Bayesian optimization, compared against AIMD reference: (b) SS and (c) OO.

Table S1: Atom names and their corresponding charges, σ, and ϵ parameters for SL molecule.

Atoms	Charges (e)	σ (nm)	ϵ (kJ/mol)
S1	1.187726	0.34443	1.13170
O1	-0.633322	0.32411	0.41901
O2	-0.633059	0.32411	0.41901
CS1	-0.318247	0.35781	0.25916
HS1	0.140548	0.23111	0.13015
HS2	0.149467	0.23111	0.13015
CC1	-0.124087	0.34880	0.2595
HC1	0.099307	0.23822	0.13955
HC2	0.092293	0.23822	0.13955
CC2	-0.124899	0.34880	0.25952
HC3	0.092745	0.23822	0.13955
HC4	0.099093	0.23822	0.13955
CS2	-0.317703	0.35781	0.25916
HS3	0.149774	0.23111	0.13015
HS4	0.140364	0.23111	0.13015

Table S2: Atom names and their corresponding charges, σ, and ϵ parameters for EMS molecule.

Atoms	Charges (e)	σ (nm)	ϵ (kJ/mol)
S1	1.555625	0.32019	1.04355
O1	-0.691636	0.36028	0.58288
O2	-0.691633	0.36028	0.58288
CS1	-0.555699	0.17855	0.40939
HS1	0.146207	0.21544	0.25787
HS2	0.146206	0.21544	0.25787
HS3	0.154753	0.21544	0.25787
CB1	-0.314231	0.32883	0.23501
HB1	0.101949	0.20275	0.13916
HB2	0.101949	0.20275	0.13916
CC1	-0.303628	0.19429	0.20673
HC1	0.109445	0.19234	0.12642
HC2	0.120345	0.19234	0.12642
HC3	0.120348	0.19234	0.12642

Table S3: Atom names and their corresponding charges, σ, and ϵ parameters for MSL molecule.

Atoms	Charges (e)	σ (nm)	ϵ (kJ/mol)
S1	1.493091	0.33140	1.71473
O1	-0.679080	0.35990	0.35096
O2	-0.665534	0.35990	0.35096
CS1	-0.400675	0.32714	0.23181
HS1	0.126726	0.16952	0.17702
HS2	0.120162	0.16952	0.17702
CA1	0.081547	0.36170	0.21388
HA1	0.056889	0.23590	0.15386
CB1	-0.349532	0.36159	0.20522
HB1	0.103750	0.23136	0.15570
HB2	0.116082	0.231336	0.15570
HB3	0.103603	0.23136	0.15570
CC1	-0.156752	0.37414	0.25074
HC1	0.088810	0.23279	0.17560
HC2	0.082456	0.23279	0.17560
CS2	-0.361935	0.32714	0.23181
HS3	0.118429	0.16952	0.17702
HS4	0.121963	0.16952	0.17702

Table S4: Atom names and their corresponding charges, σ, and ϵ parameters for DMS molecule.

Atoms	Charges (e)	σ (nm)	ϵ (kJ/mol)
S1	1.596340	0.33885	1.15816
O1	-0.684529	0.34916	0.52055
O2	-0.684529	0.34916	0.52055
CS1	-0.560950	0.35509	0.35276
HS1	0.149103	0.18692	0.14873
HS2	0.149103	0.18692	0.14873
HS3	0.149103	0.18692	0.14873
CS2	-0.560950	0.35509	0.35276
HS4	0.149103	0.18692	0.14873
HS5	0.149103	0.18692	0.14873
HS6	0.149103	0.18692	0.14873

Table S5: Comparison of viscosity (cP) obtained from classical molecular dynamics simulations employing OPLS (η^{OPLS}) and GA-GPR ($\eta^{\mathrm{GA-GPR}}$) force fields, with experimental ($\eta^{\mathrm{Exp.}}$) viscosities at different temperatures.

Sulfones	323 K			373 K		
	$\eta^{\mathrm{Exp.}}$	η^{OPLS}	$\eta^{\mathrm{GA-GPR}}$	$\eta^{\mathrm{Exp.}}$	η^{OPLS}	$\eta^{\mathrm{GA-GPR}}$
SL	6.312[1,2]	26.25 ± 0.10	6.35±0.04	2.57[1]	8.9 ± 0.06	2.77±0.03
EMS	-	4.2 ± 0.002	1.56±0.002	-	1.43 ± 0.002	0.72±0.001
MSL	5.88,[2] 5.882[1]	20.46 ± 0.18	5.87±0.03	2.289[1]	20.00 ± 0.11	2.35±0.02
Sulfones	388 K			398 K		
	$\eta^{\mathrm{Exp.}}$	η^{OPLS}	$\eta^{\mathrm{GA-GPR}}$	$\eta^{\mathrm{Exp.}}$	η^{OPLS}	$\eta^{\mathrm{GA-GPR}}$
DMS	1.285[1]	0.94 ± 0.001	1.25±0.002	1.14[1]	0.95 ± 0.01	1.10±0.00

S1 References

[1] Casteel, J. F.; Sears, P. G. Dielectric constants, viscosities, and related physical properties of 10 liquid sulfoxides and sulfones at several temperatures. *J. Chem. Eng. Data* **1974**, *19*, 196–200.

[2] Vaughn, J. W.; Hawkins, C. F. Physical Properties of Tetrahydrothiophene-1, 1-Dioxide and 3-Methyltetrahydrothiophene-1, 1-Dioxide. *J. Chem. Eng. Data* **1964**, *9*, 140–142.