

Supplementary Material (SM)

Reactive Machine-Learned Potentials for Fluoropolymer Binders: Unified Physical and Chemical Property Validation

Huanyu Zhu, Mingjie Wen, Dongping Chen, and Qingzhao Chu*

(State Key Laboratory of Explosion Science and Safety Protection, Beijing Institute of Technology, Beijing 100081, China)

In the supplementary information, we present:

1. **Fig. S1** Comparison of MLP predictions with DFT calculations for atomic energies (a) and atomic forces (b), with the overall Mean Absolute Error (MAE) and RMSE annotated in the legends. Error distributions across the three fluoropolymers F2311, F2313, and F2314 for (c) atomic energies and (d) atomic forces.
2. **Fig. S2** Initial amorphous simulation cells for AIMD and DPMD calculations: (a) F2311, where each polymer chain contains 3 repeat units and the cell contains 5 polymer chains; (b) F2313, where each polymer chain contains 2 repeat units and the cell contains 4 polymer chains; and (c) F2314, where each polymer chain contains 2 repeat units and the cell contains 4 polymer chains.
3. **Fig. S3** Comparative probability density distributions of main-chain F-containing dihedral angles, comparing MLP predictions with AIMD reference data across varying temperatures (300 K, 400 K, and 500 K) for F2311 (a-c), F2313 (d-f), and F2314 (g-i).
4. **Table S1** Calculated BDEs for the isolated single chains of F2311, F2313, and F2314.

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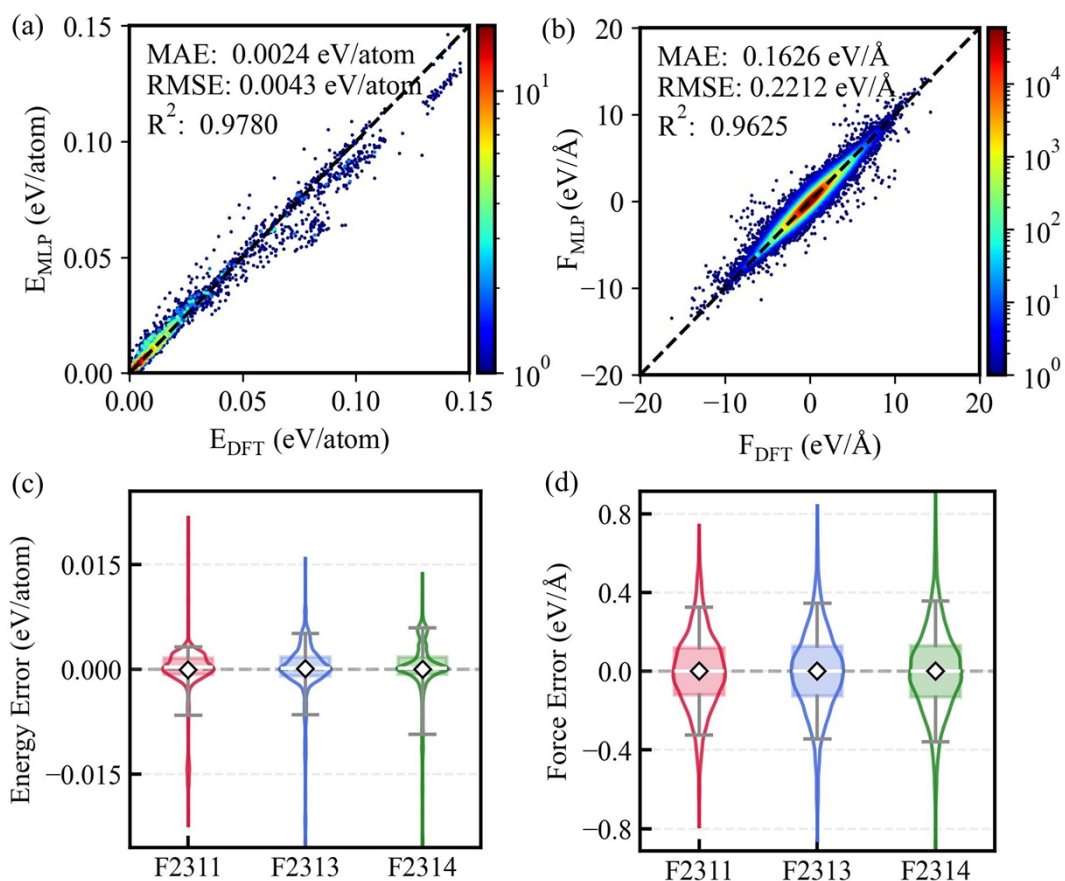


Fig. S1 Comparison of MLP predictions with DFT calculations for atomic energies (a) and atomic forces (b), with the overall Mean Absolute Error (MAE) and RMSE annotated in the legends. Error distributions across the three fluoropolymers F2311, F2313, and F2314 for (c) atomic energies and (d) atomic forces.

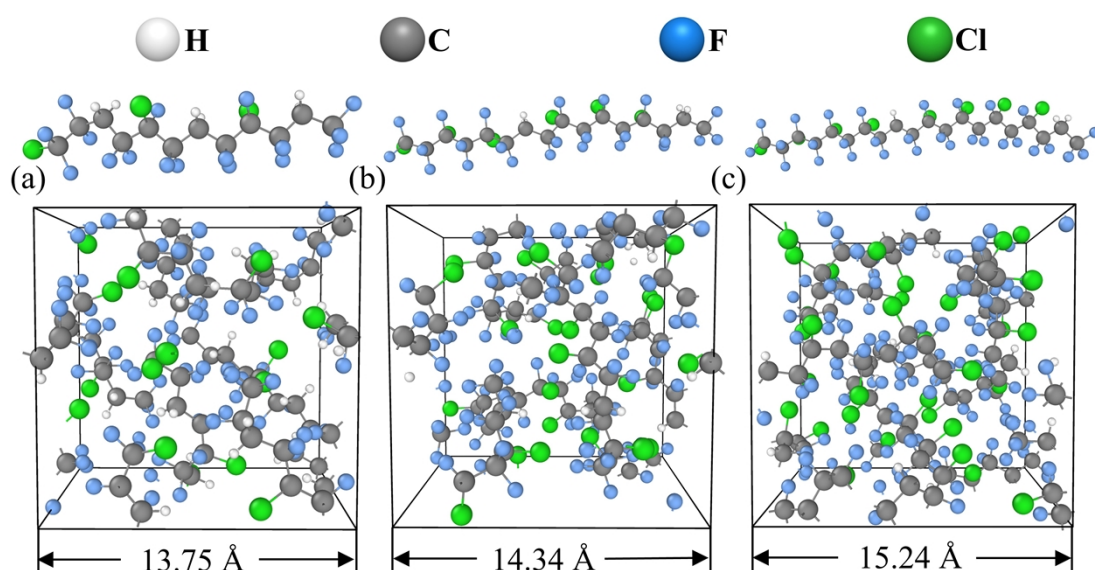


Fig. S2 Initial amorphous simulation cells for AIMD and DPMD calculations: (a) F2311, where each polymer chain contains 3 repeat units and the cell contains 5 polymer chains; (b) F2313, where each polymer chain contains 2 repeat units and the cell contains 4 polymer chains; and (c) F2314, where each polymer chain contains 2 repeat units and the cell contains 4 polymer chains.

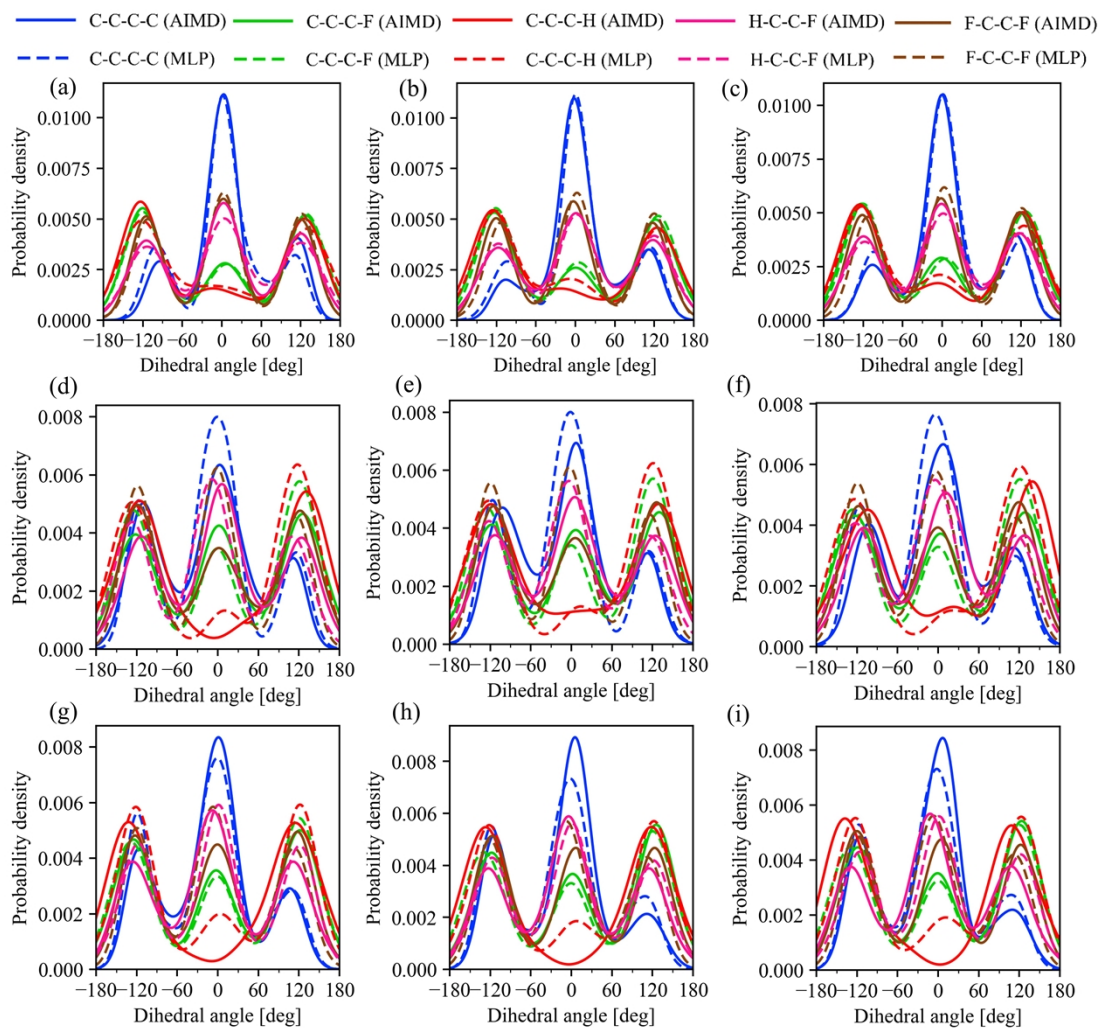


Fig. S3 Comparative probability density distributions of main-chain F-containing dihedral angles, comparing MLP predictions with AIMD reference data across varying temperatures (300 K, 400 K, and 500 K) for F2311 (a-c), F2313 (d-f), and F2314 (g-i).

Table S1. Calculated BDEs for the isolated single chains of F2311, F2313, and F2314

Bond	F2311		F2313		F2314	
	DFT(eV)	MLP(eV)	DFT(eV)	MLP(eV)	DFT(eV)	MLP(eV)
a	2.45	2.93	3.94	3.92	4.36	4.30
b	2.14	2.26	3.76	3.75	3.84	3.81
c	3.44	3.30	2.44	2.59	2.69	2.71
d	3.23	3.30	1.98	2.09	2.29	2.26
e	3.58	3.56	3.43	3.36	3.71	3.69
f	3.58	3.51	2.57	2.58	3.12	3.06
g	4.14	4.24	3.76	4.24	3.74	4.20
h	3.99	3.79	2.61	2.95	3.03	3.03
i	–	–	1.83	1.96	2.02	2.22
j	–	–	3.74	3.81	3.88	3.99
k	–	–	2.74	3.15	2.92	3.01
l	–	–	3.73	3.76	3.94	4.18
m	–	–	2.18	2.24	2.79	3.05
n	–	–	3.69	3.79	2.28	2.36
o	–	–	3.48	3.51	3.58	3.83
p	–	–	6.15	6.03	2.86	3.24
q	–	–	2.55	2.67	4.04	4.11
r	–	–	3.51	3.69	2.05	2.01
s	–	–	–	–	4.41	4.09
t	–	–	–	–	3.89	3.83
u	–	–	–	–	5.63	5.61
v	–	–	–	–	2.73	2.63
w	–	–	–	–	3.50	3.74