Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

A Neural Network Potential for IRMOF Series and Application on Thermal and Mechanical Behaviors

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Figures

Tables

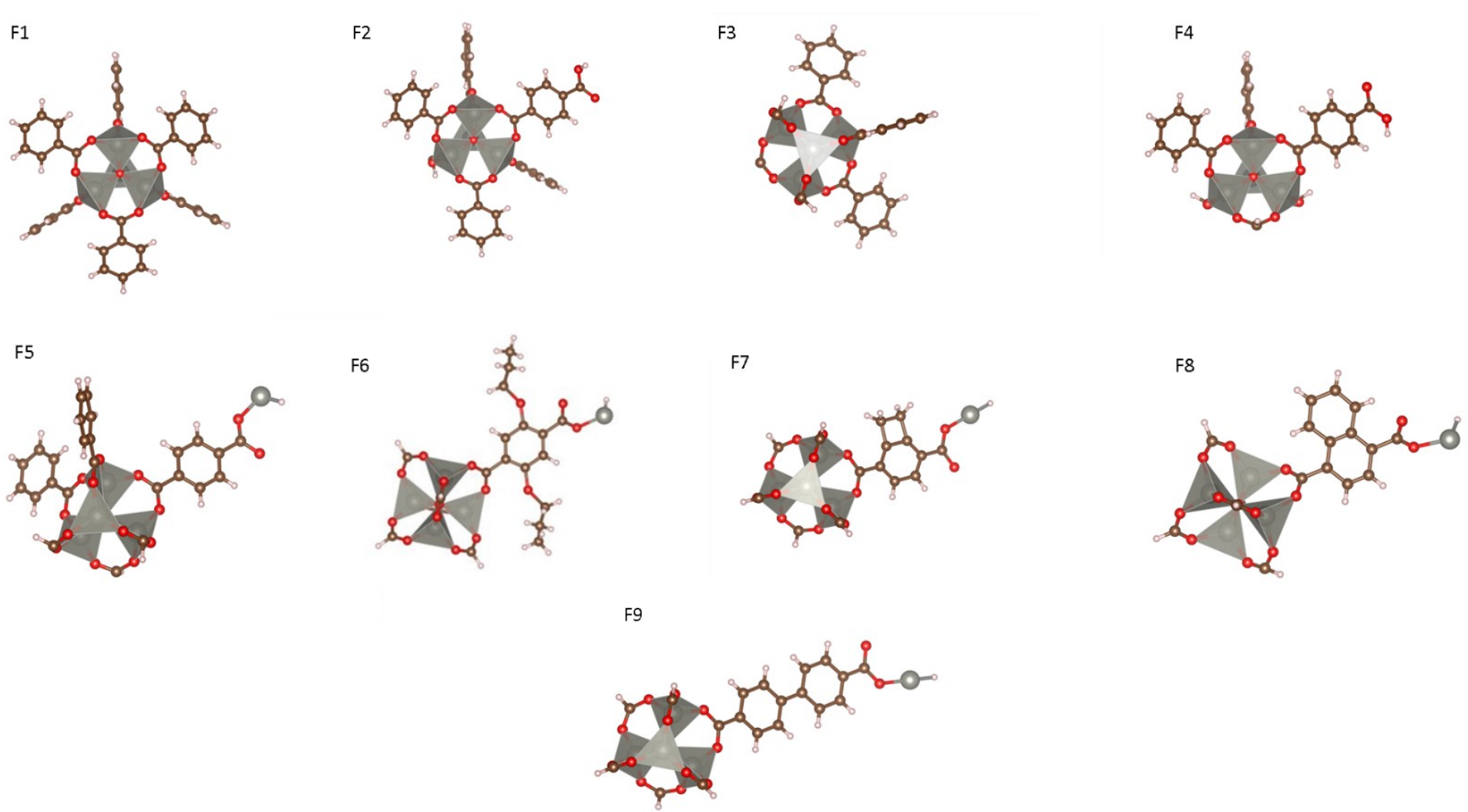

Figure S1. FigureS1: IRMOF-n (n=1,4,6,7,10) fragments. F1-F5 are generated from IRMOF-1. F6, F7, F8, F9 are from IRMOF-4, 6,7 and 10, respectively.

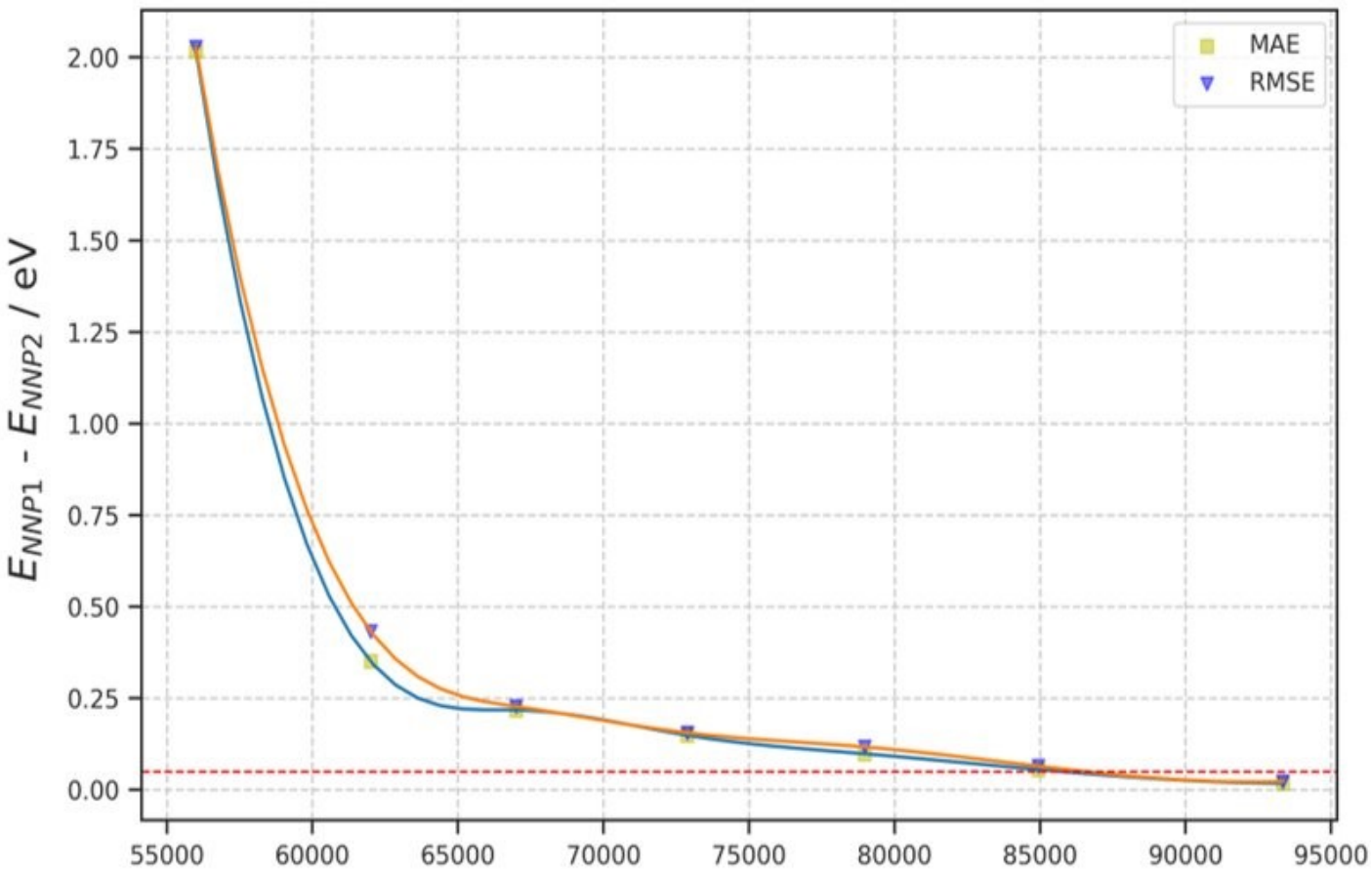

Figure S2. Uncertainty between NNP1 and NNP2 energies according to number of data.

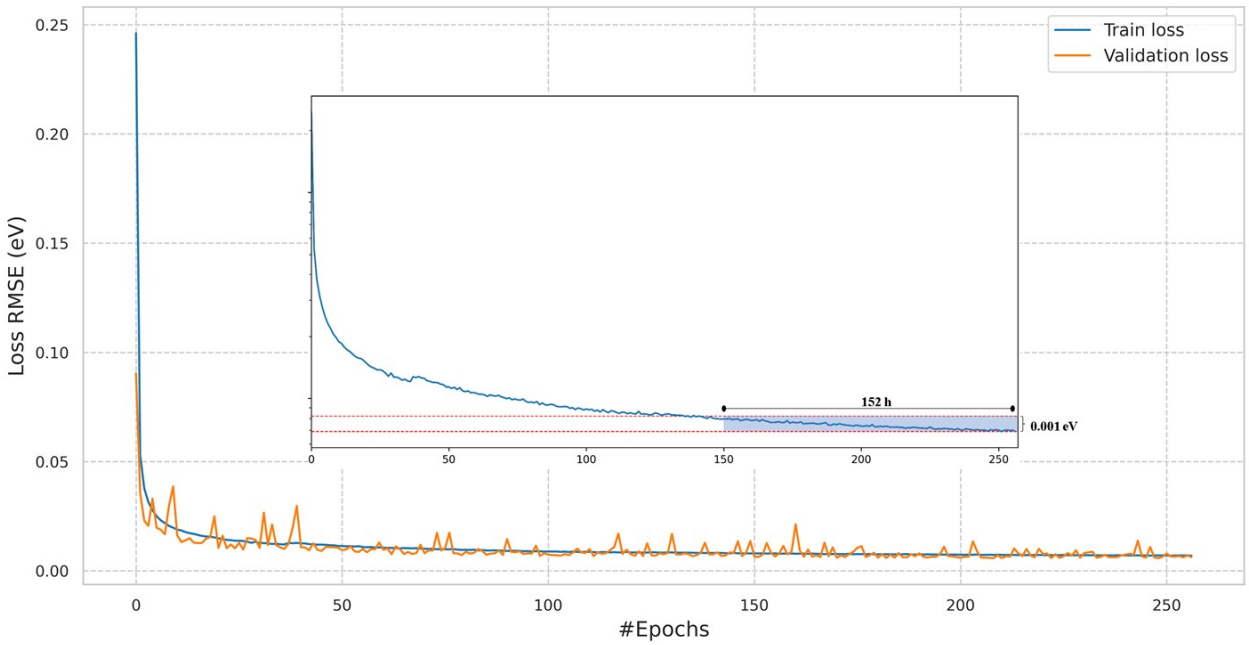

Figure S3. Training and validation loss curves. The inset is the logarithmic scale showing early stopping at 150 epoch.

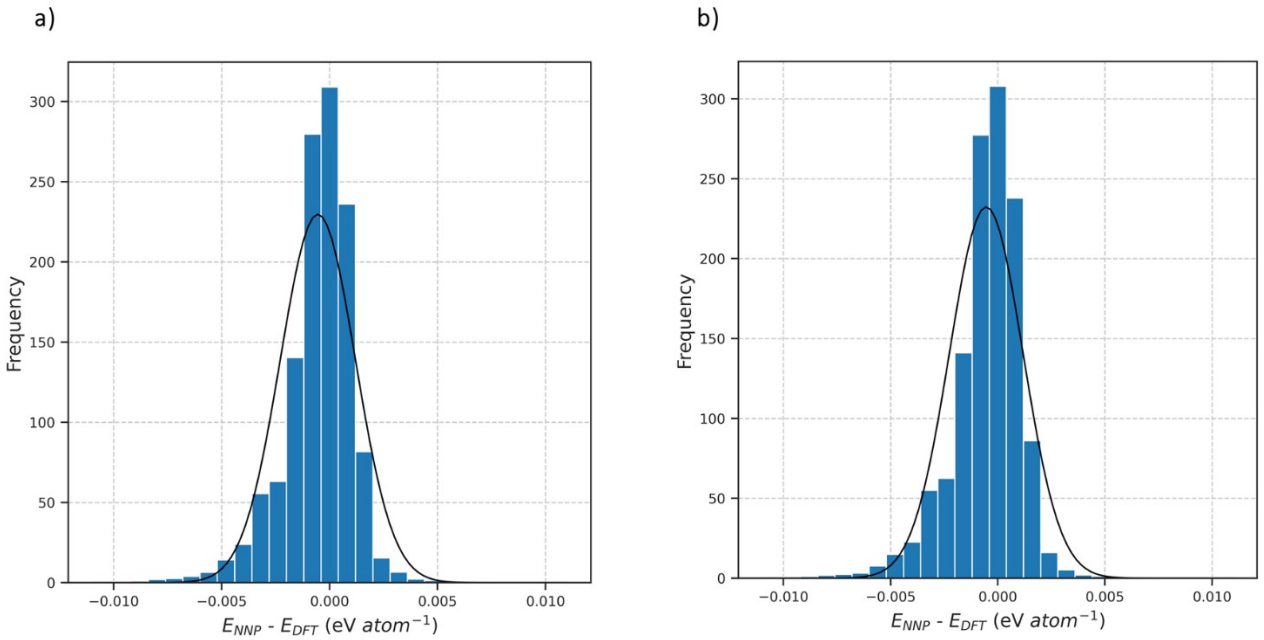

Figure S4. Histograms of energy errors show normal distribution a-training b-tests

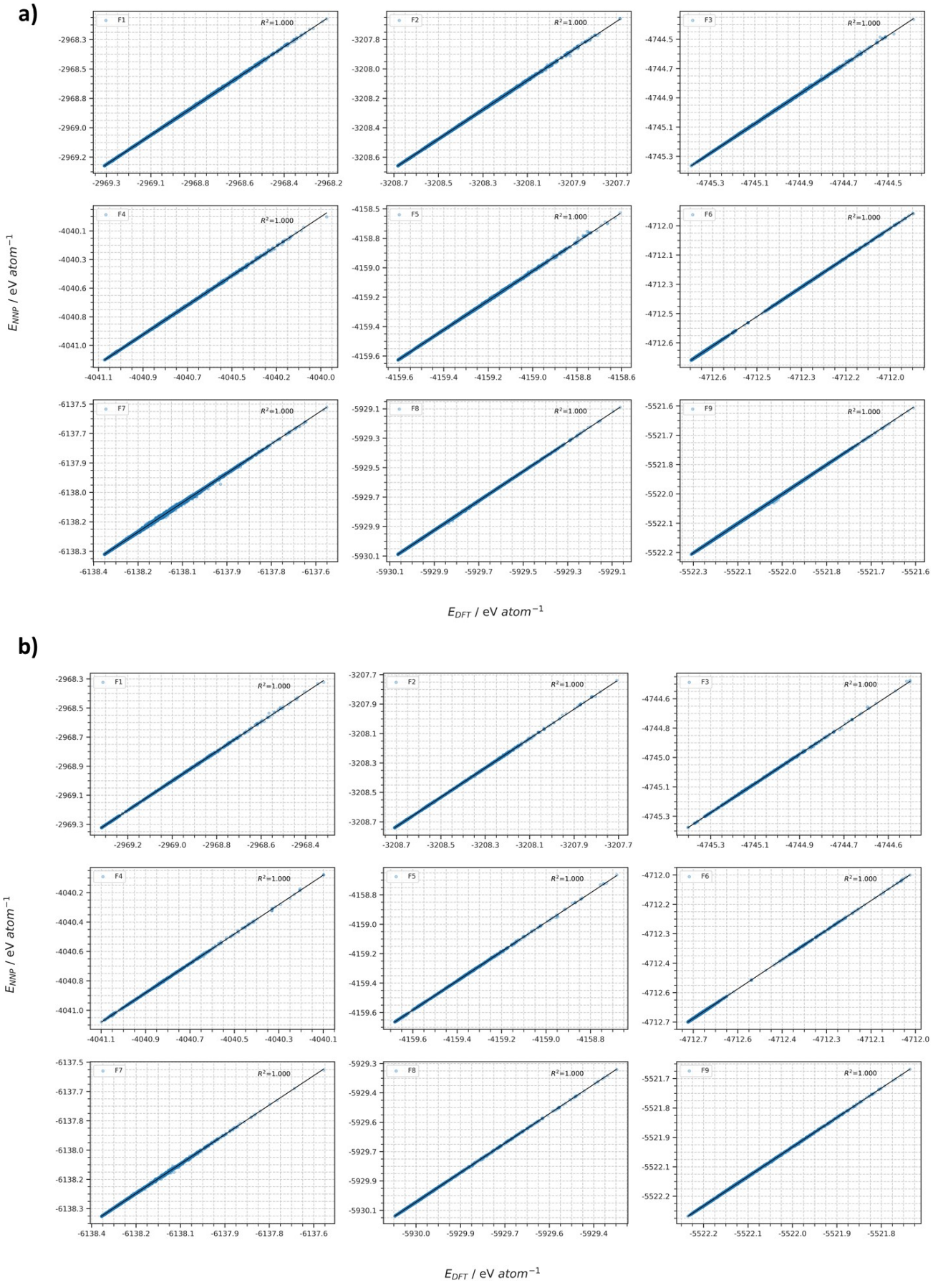

Figure S5. DFT vs NNP energies of 9 fragments for a-training b-test

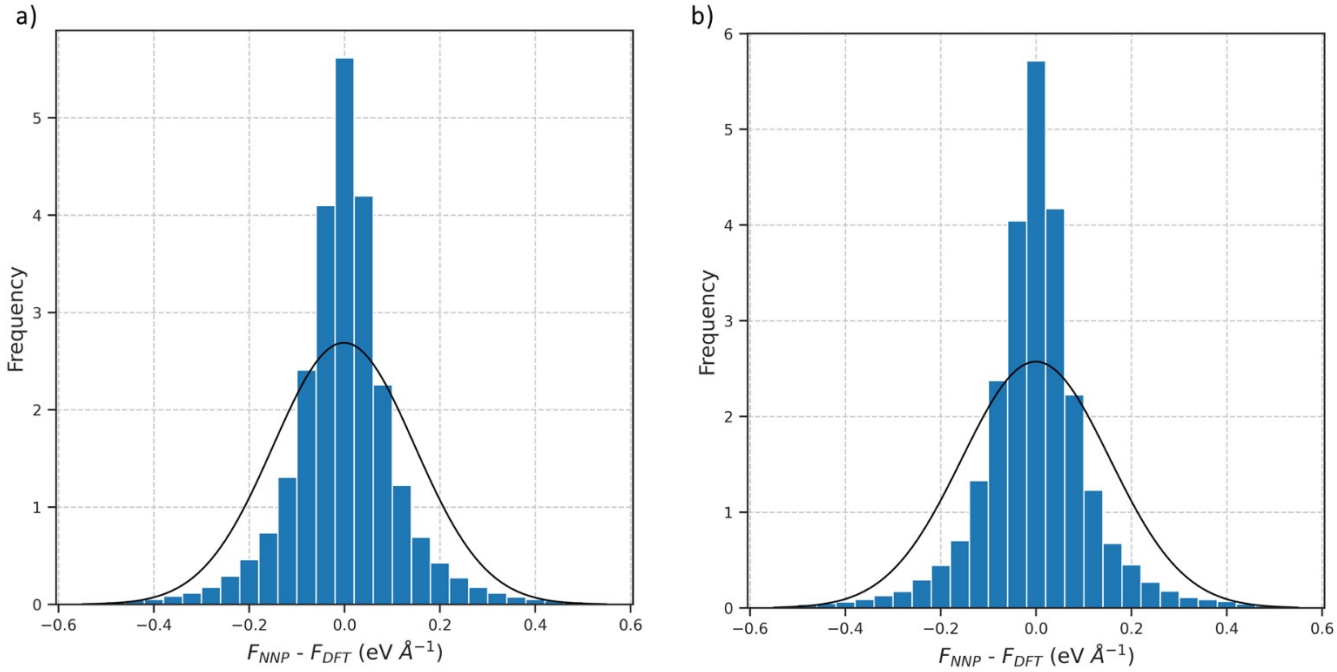

Figure S6. Histograms of force errors show normal distribution a-training b-tests

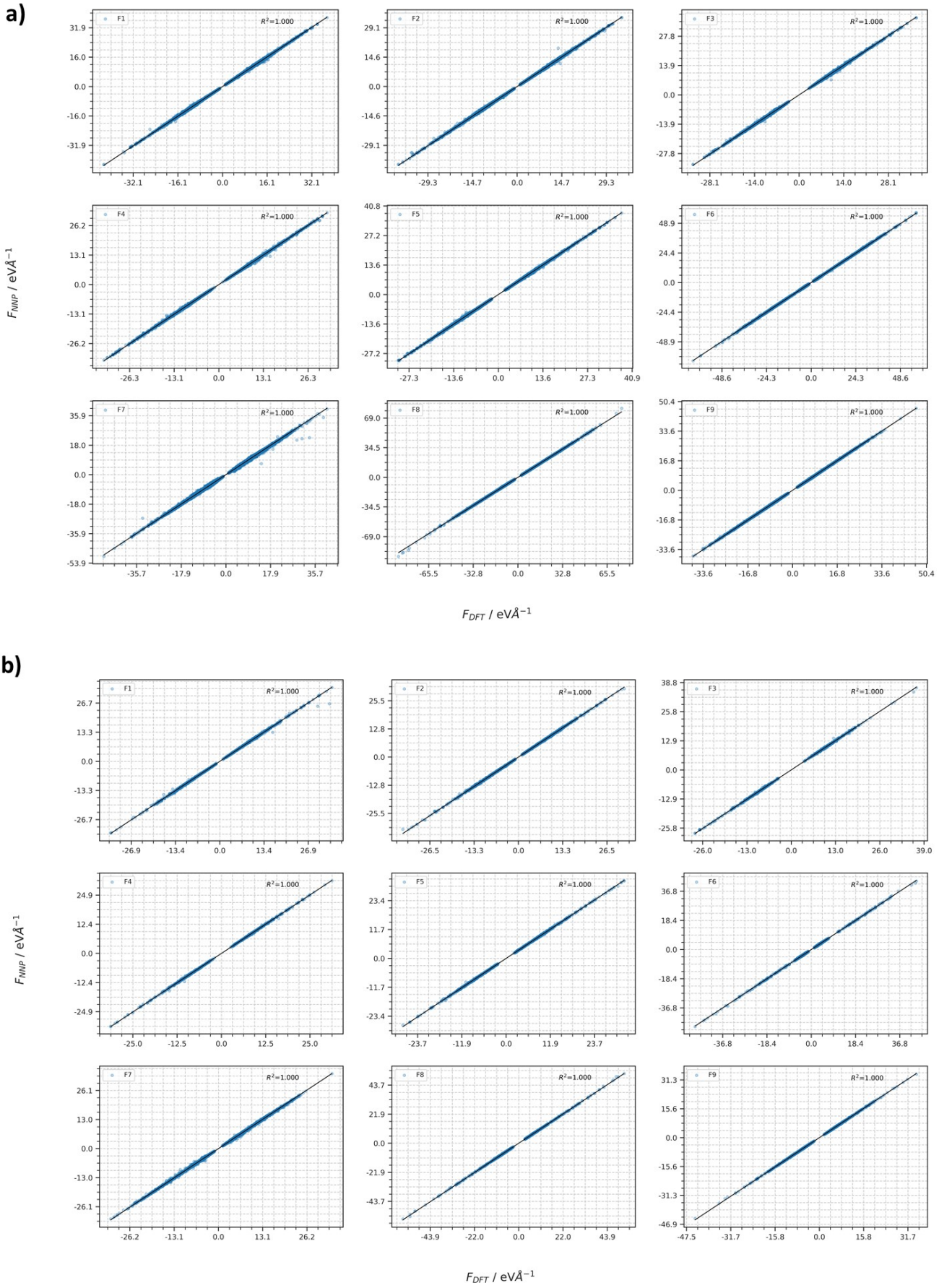

Figure S7. DFT vs NNP forces of 9 fragments for a-training b-test

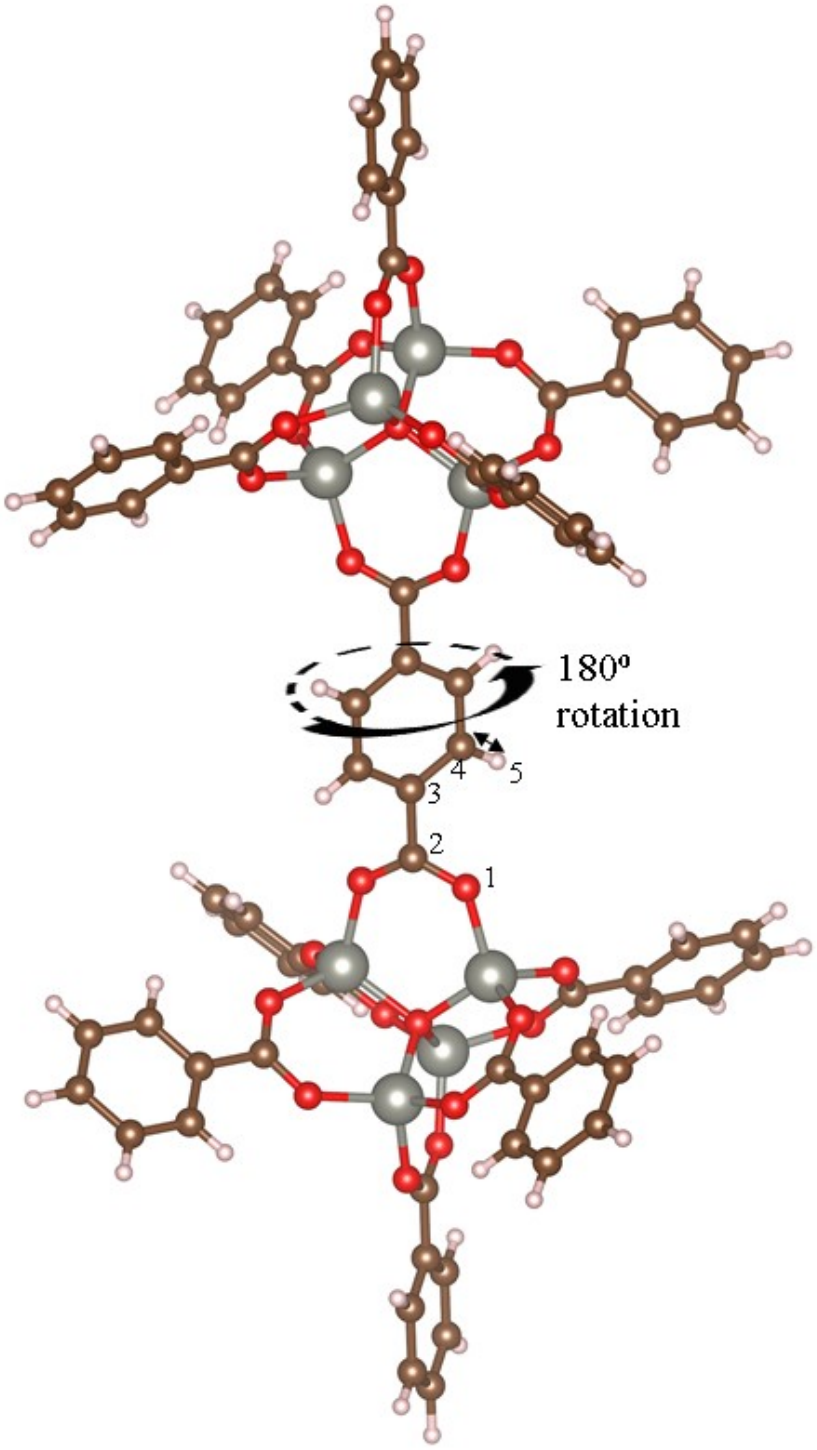

Figure S8. IRMOF-1 truncated structure for phenylene ring torsion by scan of dihedral angle, (Φ1234) from 0 to 180º and aromatic C-H distance of ($r45$) from 1.02Å to 1.16Å

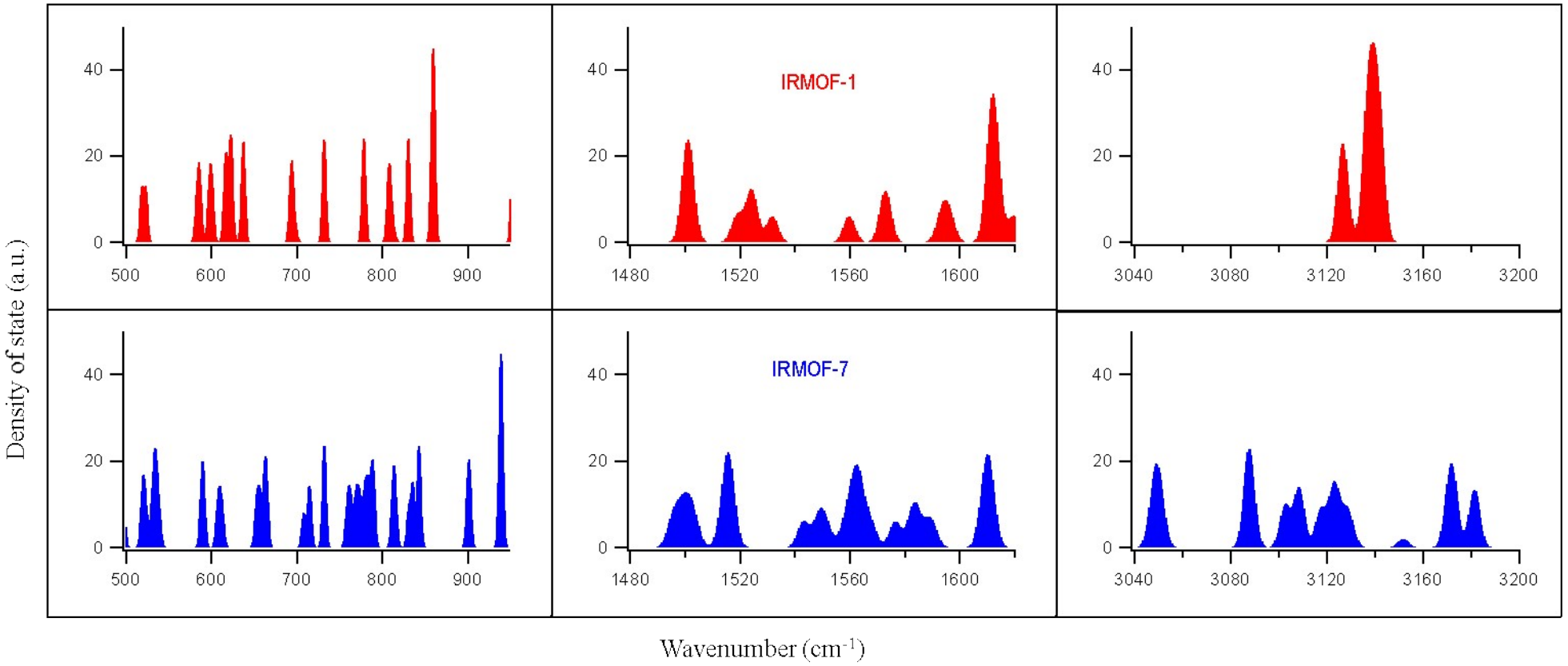

Figure S9. IRMOF-1 and IRMOF-7 vibrations zoomed in different regions

Non-linear thermal expansion coefficient

The linear equation between temperature and volume in equation **(1)** no longer holds for these unusual MOFs with simultaneous PTE and NTE. The thermal expansion coefficient must be a function of temperature; and thus, at a distinct temperature, these MOFs should have neither positive nor negative thermal expansions (PTE to NTE switching temperature). In order to cover these MOFs, we can modify the equation **(1)** by defining thermal expansion coefficient as a function of temperature:

$\alpha_0 = f(T) = m + nT + ...$

For the first order approximation, temperature dependent thermal expansion coefficient is in the form of

$\alpha_0 = m + nT$

Substituting this equation in the integration will yield:

$\langle \ln (V) \rangle_P \approx \frac{nT^2}{2} + mT + C$

where m and n are the coefficients and can be determined from the nonlinear (quadratic) fit of the ln(V)-T plot.

Table S1. Thermal expansion coefficient calculations based on quadratic and linear equations. ST shows PTE to NTE switching temperature

							α_0=m+nT		α_0=constant
IRMOF-n	m x 10^6	n x 10^8	Quadratic equation	Linear equation	ST=-m/n		200K x 10^6	300K x 10^6	T independent α_0
1	-60.42	4.52	2.2576E-08T^2 - 6.0422E-05T + 9.7841E+00	-4.6876E-05T + 9.7822E+00		Only NTE	-51.392	-46.876	-46.900
4	662.76	-251.42	-1.2571E-06T^2 + 6.6276E-04T + 9.6715E+00	-9.1506E-05T + 9.7784E+00	263.60671	PTE to NTE	159.920	-91.500	-91.506
6	-25.74	-2.94	-1.4719E-08T^2 - 2.5735E-05T + 9.7972E+00	-3.4566E-05T + 9.7984E+00		Only NTE	-31.623	-34.566	-34.566
7	1459.31	-512.78	-2.5639E-06T^2 + 1.4593E-03T + 9.4736E+00	-7.9032E-05T + 9.6915E+00	284.58793	PTE to NTE	433.750	-79.030	-79.032
10	-20.12	-10.04	-5.0190E-08T^2 - 2.0119E-05T + 1.0644E+01	-5.0233E-05T + 1.0648E+01		Only NTE	-40.195	-50.233	-50.233

Table S2 Equilibrium lattice parameters produced by NNP at different temperatures.

	Lattice constant (a=b=c) in Angstrom					ln(<Volume>)				
T (K)	IRMOF-1	IRMOF-4	IRMOF-6	IRMOF-7	IRMOF-10	IRMOF-1	IRMOF-4	IRMOF-6	IRMOF-7	IRMOF-10
200	25.98776171	25.81962824	26.14978993	25.05992992	34.67394602	9.77286941	9.753397	9.791518	9.663732	10.63796
250	25.96690189	25.8814584	26.13413159	25.16866906	34.64504411	9.770458158	9.760572	9.789719	9.676775	10.63546
300	25.94791248	25.82383728	26.12004725	25.19608271	34.61687294	9.76826027	9.753879	9.788099	9.680009	10.63301
350	25.92291174	25.80800564	26.10659322	25.13368317	34.59628239	9.76536445	9.752037	9.786552	9.672566	10.63123
400	25.90853144	25.66023341	26.08842062	24.91328927	34.55357941	9.76369724	9.734788	9.78446	9.646079	10.62752

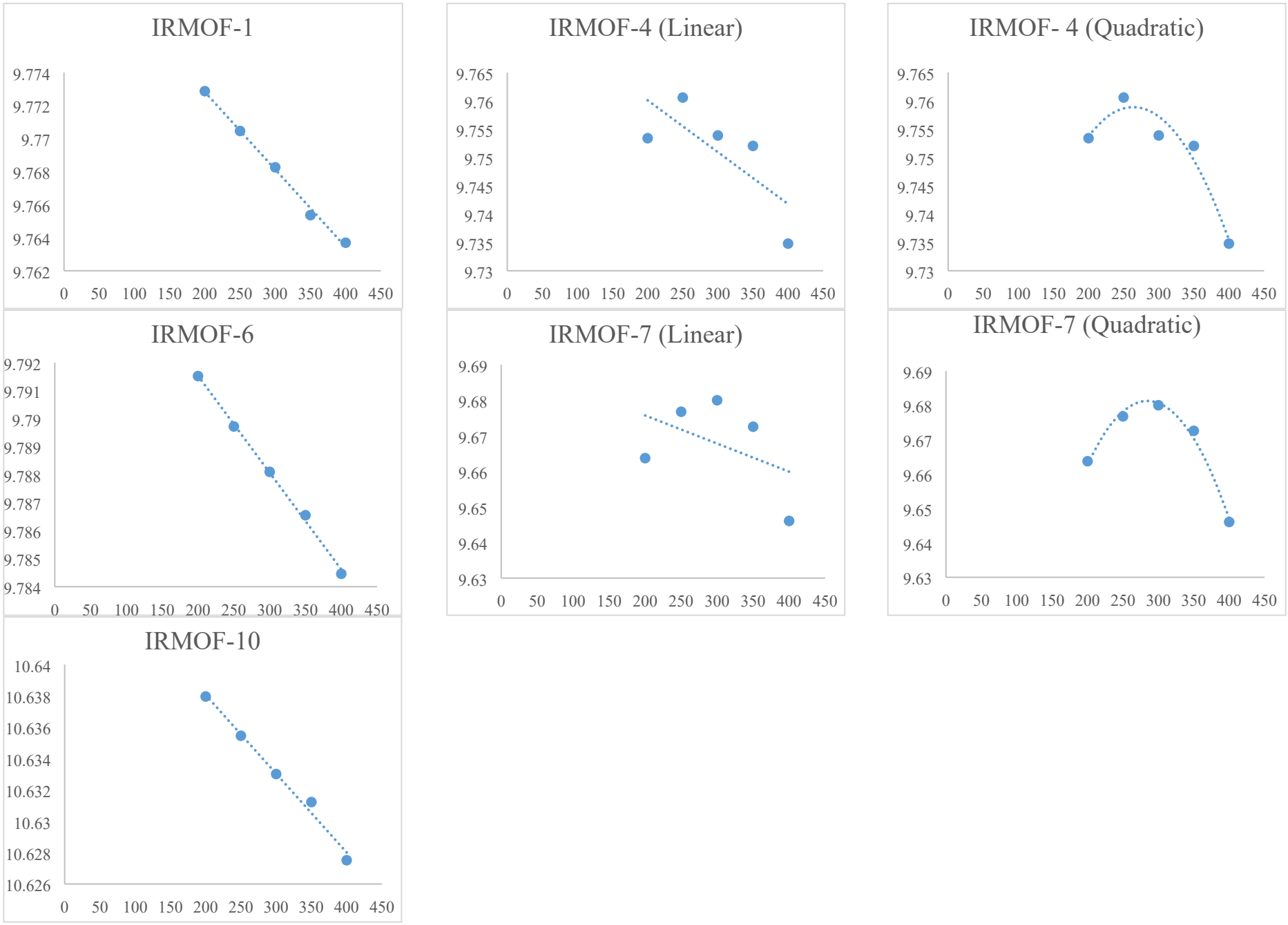

Figure S10. ln(<V>) vs. T (in Kelvin) plots for thermal expansion coefficients

Table S3 Thermal pressure coefficients calculated from MD simulations using NNP

IRMOF-n	Thermal Pressure Coefficent, γ ($MPaK^{-1}$)		
	NNP (100K)	NNP (300K)	Literature (300K) [[1]]
1	-0.6768	-0.4205	-0.408
4	2.0070	-0.7558	-
6	-0.4614	-0.4079	-
7	2.8541	-0.1818	-
10	-0.2794	-0.2627	-0.233

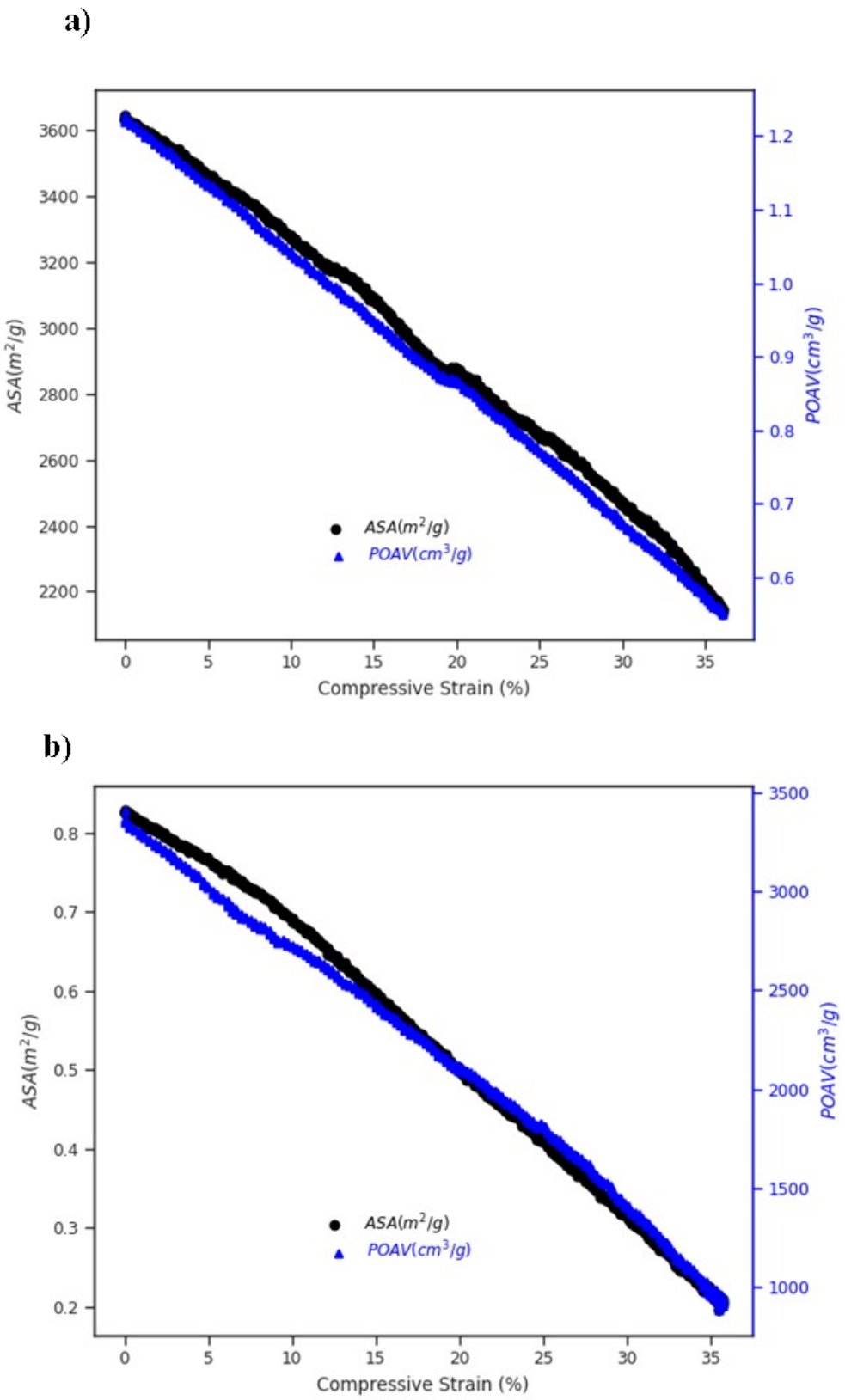

Figure S11. ASA and POAV change of a) IRMOF-1 and b) IRMOF-7 upon stress applied along [001] direction