Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.

Supporting information

Table S1 Number of molecules in unit cell

Number of carbons (n_C)	Systems	Number of molecules
0	H_3PO_4	48
1	$CH_3PO_3H_2$	48
4	$C_4H_9PO_3H_2$	32
7	$C_7H_{15}PO_3H_2$	20

Table S2 Parameters of descriptors and kernel function. See details of the symbols in main text and ref. [56]

Parameter	Value	Parameter	Value	Parameter	Value
ζ	4	σ_{atom}	0.5 Å	L_{max}	2
$\beta^{(2)}$	0.5	$R_{cut}^{(2)}$	6 Å	$N_R^{(2)}$	8
$\beta^{(3)}$	0.5	$R_{cut}^{(3)}$	4 Å	$N_R^{(3)}$	6

Table S3 Number of molecules in the unit cells for training

Number of carbons (n_C)	Systems	Number of molecules
0	H_3PO_4	32
1	$CH_3PO_3H_2$	24
4	$C_4H_9PO_3H_2$	20
7	$C_7H_{15}PO_3H_2$	12

Table S4 Elapsed time per MD step for 32 PA by the MLFF and FP method. The elapsed time was measured by using 16 cores of Intel Xeon Platinum 8358 (2.6 GHz).

	FP	MLFF
Elapse time (s/step)	92	0.024

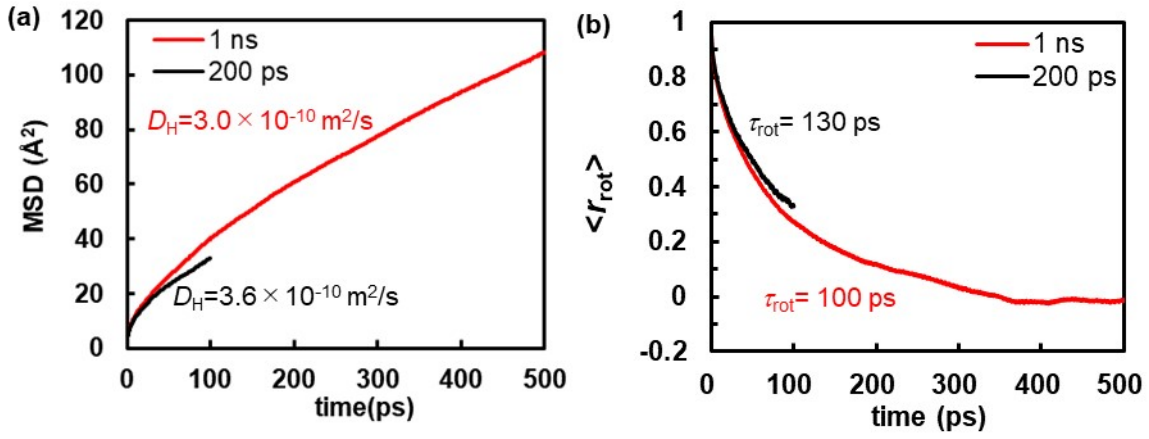

Fig. S1 Comparison of (a) MSD of hydrogen and (b) $<r_{rot}(t)>$ for n_C = 7 at 463K evaluated from 1 ns and 200 ps trajectories.

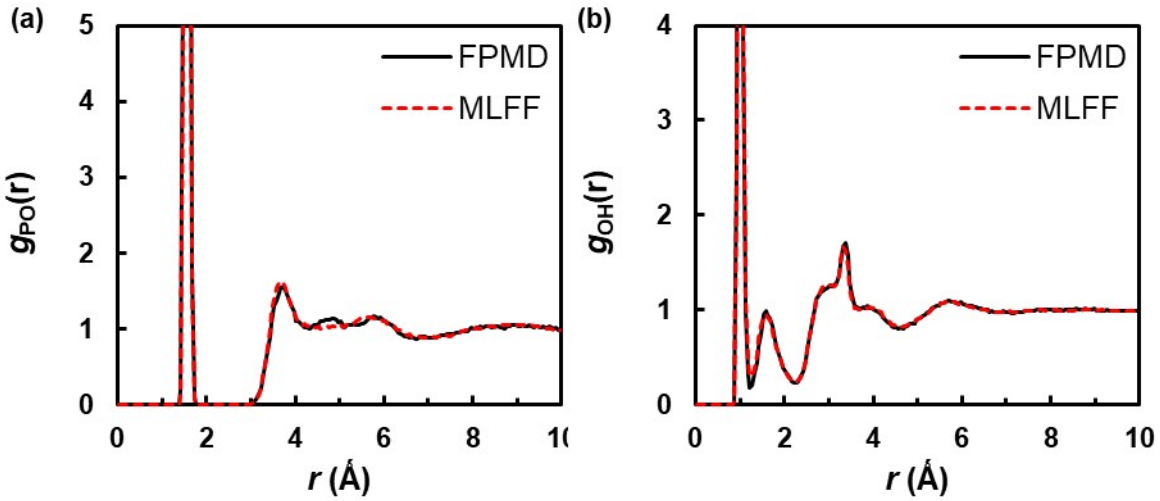

Fig. S2 Comparison of the radial distribution functions of (a) P-O (g_{PO}) and (b) O-H (g_{OH}) for liquid phosphoric acid (PA) calculated by machine-learned force field (MLFF) and First principles molecular dynamics simulation (FPMD) methods.

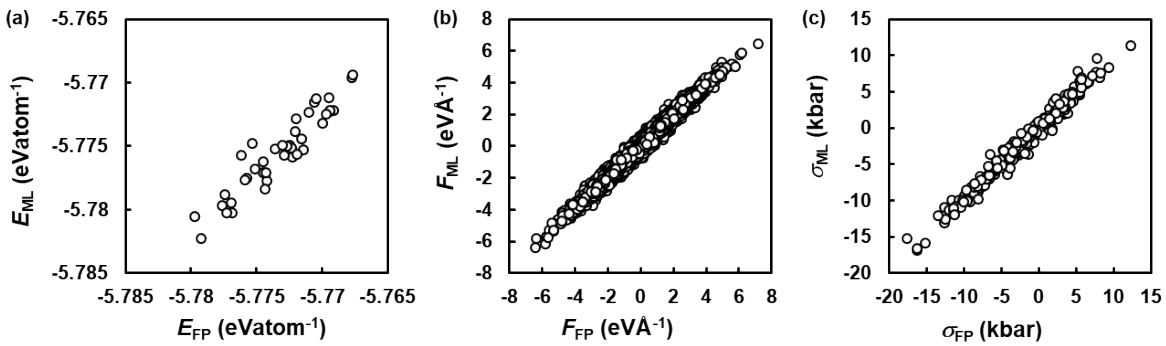

Fig. S3 (a) Energies, (b) forces, and (c) stress tensor components of liquid PA predicted by the MLFF and FPMD methods.

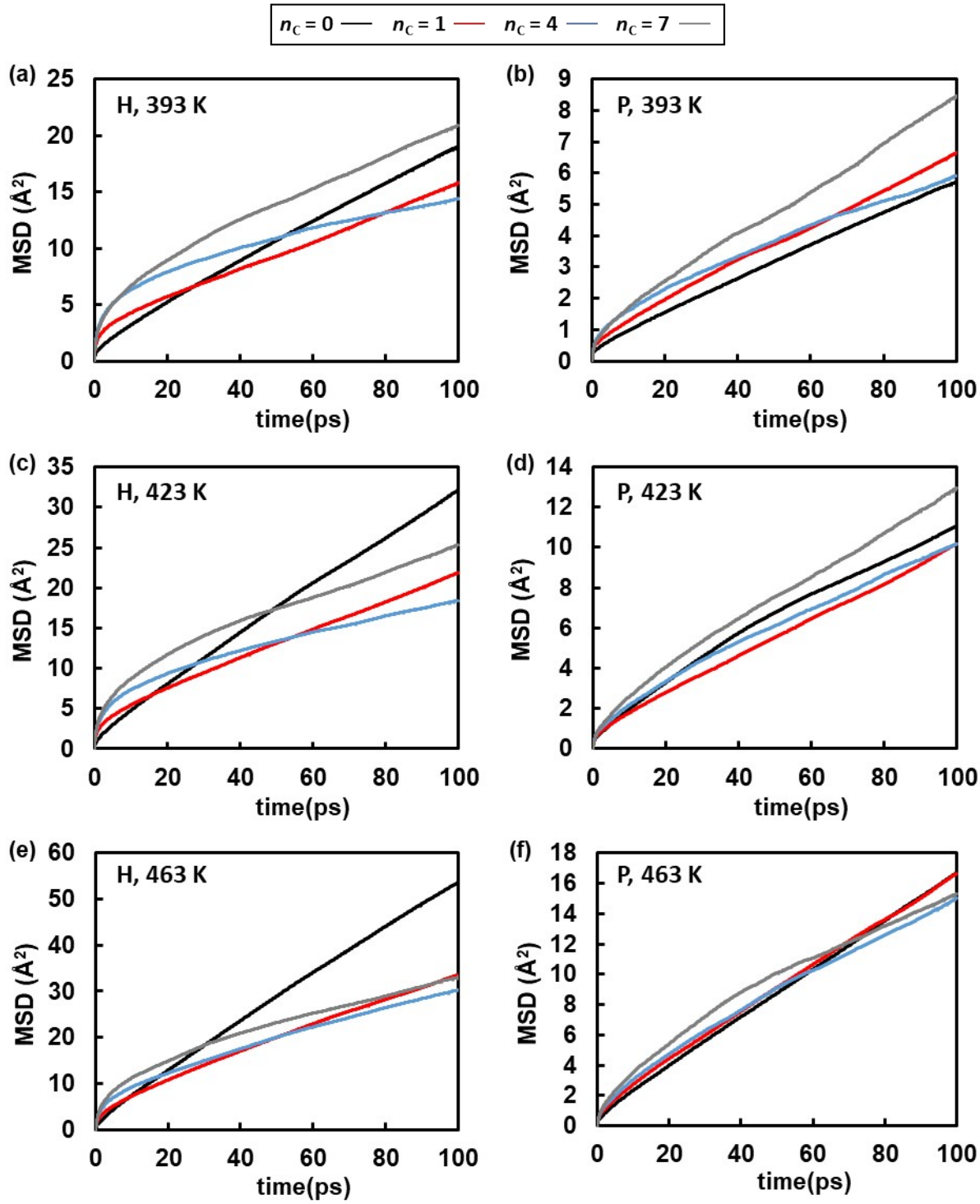

Fig. S4 Mean square displacements (MSDs) of hydrogen and phosphorus atoms at 393, 423, and 463 K.

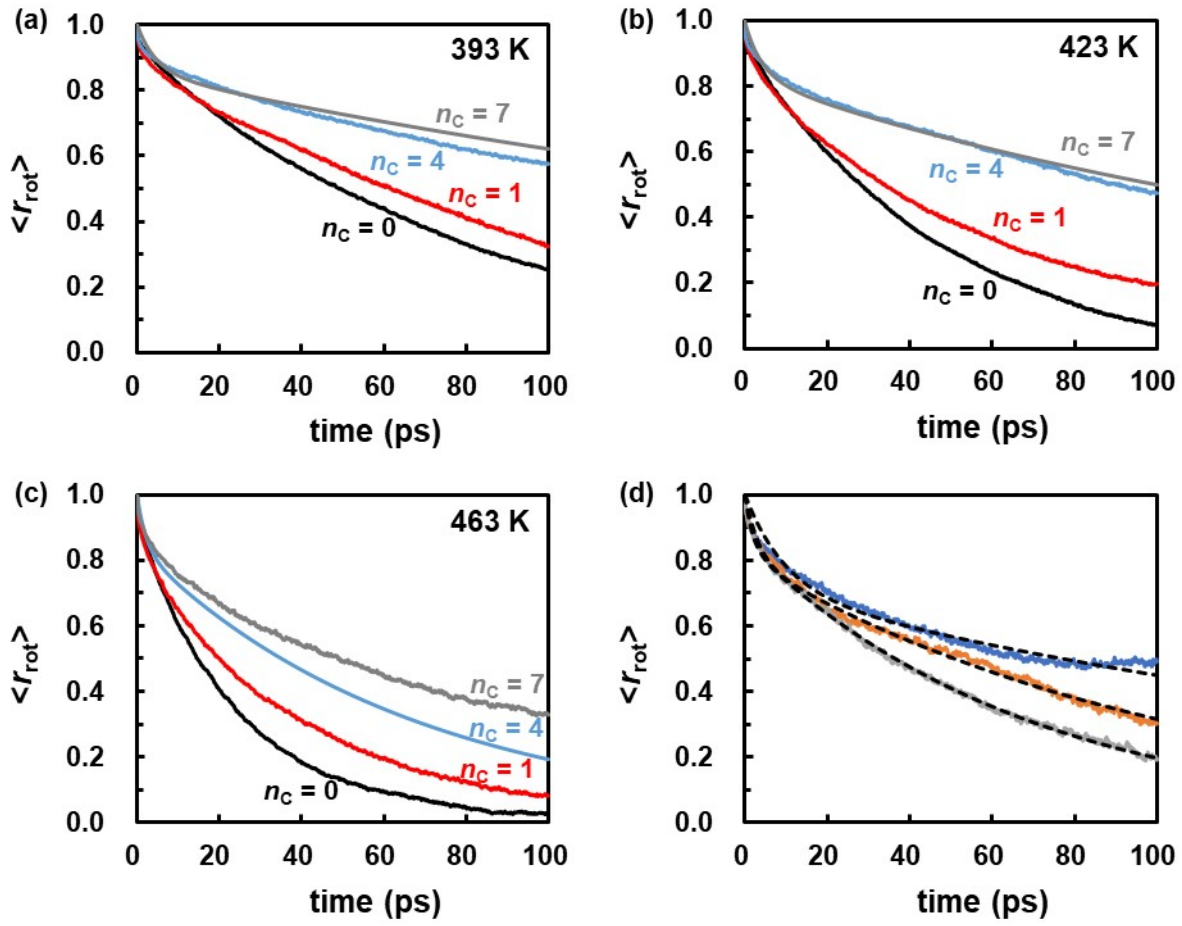

Fig. S5 Autocorrelation functions of P-O vectors ($\langle r_{rot}(t) \rangle$) defined using Eq. (5) at (a) 393, (b) 423, and (c) 463 K. Each data point is average over three MD trajectories that started from different initial structures. (d) The raw data before averaging (solid line) and fitted lines determined using Eq. (6) (dashed line) for n = 7 at 463 K.