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Supplementary material

EDL structure of ionic liquid-MXene-based supercapacitor and hydrogen bond role on the interface: A molecular dynamics simulation investigation

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Table S1 Lennard-Jones components and point charge parameters for $Ti_3C_2O_2$.

Atom	L-J parameters and point charge		
	ε/kcal.mol^{-1}	σ/Å	charge/e$^-$
Ti-inner	0.6087	1.9565	0.68
Ti-outer	0.6087	1.9565	1.04
C	0.0660	3.5000	-0.74
O	0.1554	3.1656	-0.64

Table S2 Lennard-Jones components and point charge parameters for $Ti_3C_2(OH)_2$.

Atom	Lennard-Jones and point charge parameters		
	ε/kcal.mol^{-1}	σ/Å	charge/e$^-$
Ti-inner	0.6087	1.9565	0.64
Ti-outer	0.6087	1.9565	0.88
C	0.0660	3.5000	-0.76
O	0.1554	3.1656	-0.79
H	0.1554	1.5000	0.35

Table S3 The ion diffusion coefficients (10^{-6} cm^2 s^{-1}) of the ILs ([HEMIm][NTf_2] and [EMIM][NTf_2]) in the bulk

	D_{cation}	D_{anion}	D_{cation}+ D_{anion}
[EMIM][NTf_2]	2.27	1.94	4.21
[HEMIm][NTf_2]	1.77	1.41	3.18

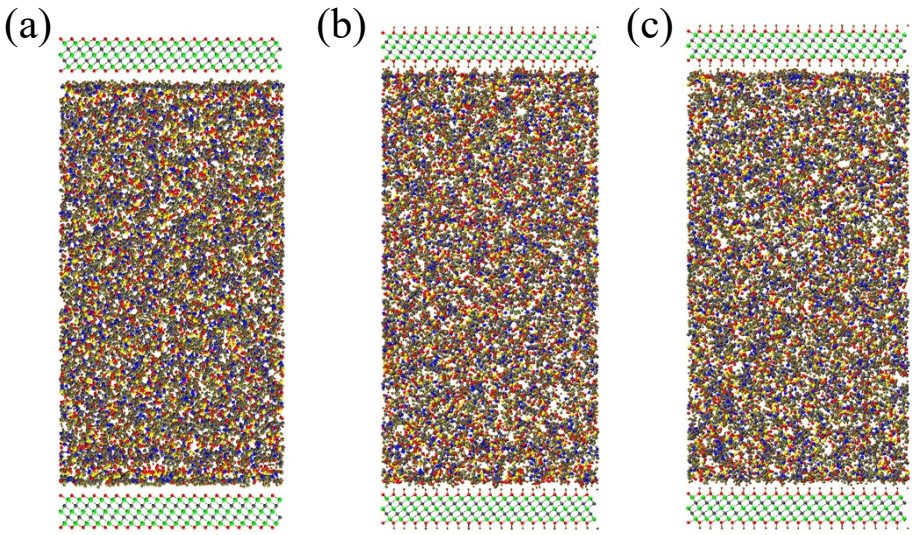


Fig. S1 A snapshot of the simulation box composed of (a) the [EMIM][NTf_2] and $Ti_3C_2O_2$, (b) the [HEMIm][NTf_2] and $Ti_3C_2(OH)_2$, (c) the [EMIM][NTf_2] and $Ti_3C_2(OH)_2$

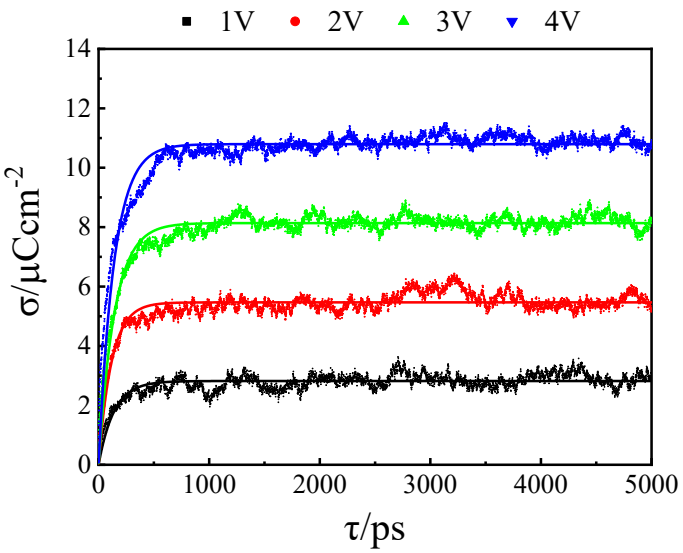


Fig. S2 Evolution of the charge density σ on electrode surface during the charging process with applying a series of potential differences to the $Ti_3C_2O_2$ electrodes for [HEMIm][NTf_2].

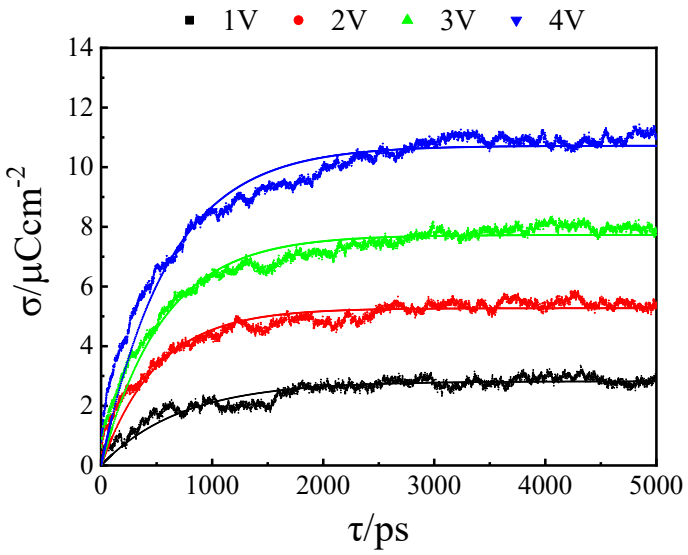


Fig. S3 Evolution of the charge density σ on electrode surface during the charging process with applying a series of potential differences to the $Ti_3C_2O_2$ electrodes for [EMIM] [NTf_2].

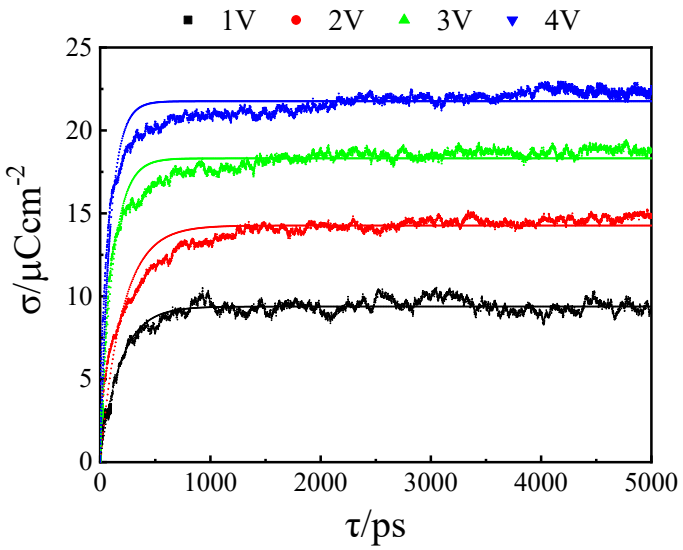


Fig. S4 Evolution of the charge density σ on electrode surface during the charging process with applying a series of potential differences to the $Ti_3C_2(OH)_2$ electrodes for [HEMIm][NTf_2].

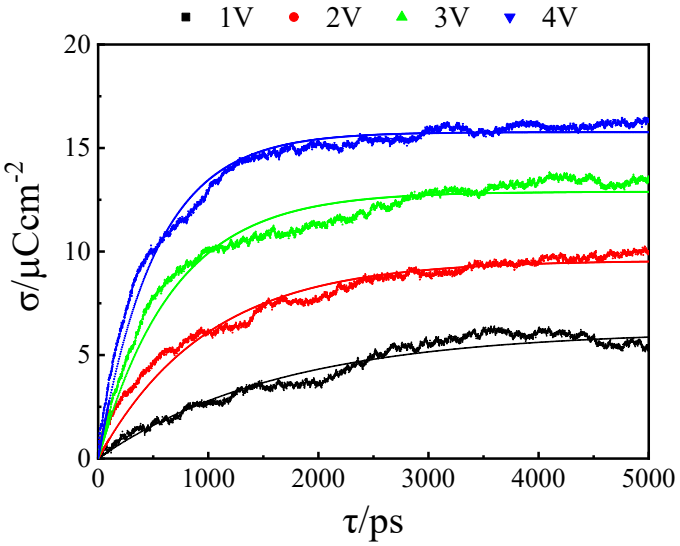


Fig. S5 Evolution of the charge density σ on electrode surface during the charging process with applying a series of potential differences to the $Ti_3C_2(OH)_2$ electrodes for [EMIM] [NTf_2].

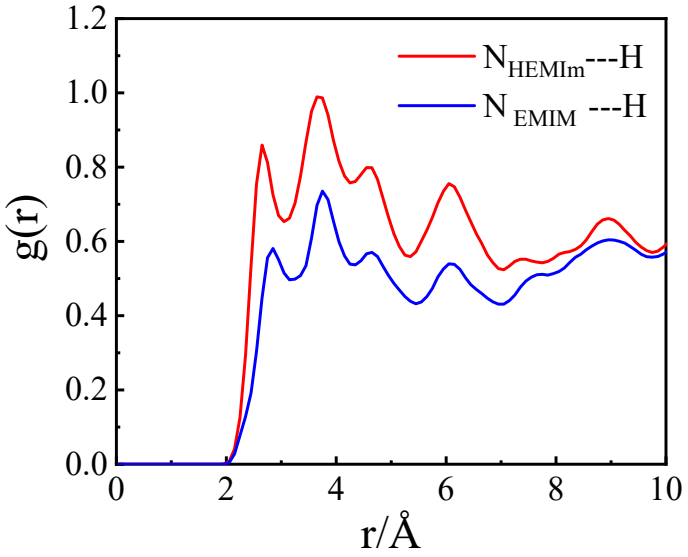


Fig. S6 Radial distribution function between N atoms in cations and H atoms in the $Ti_3C_2(OH)_2$ electrodes at PZC.

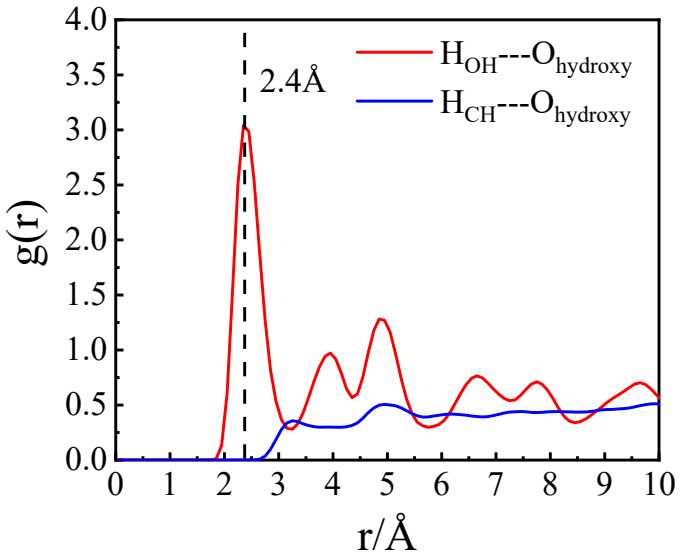


Fig. S7 Radial distribution function between H atoms of cations and O atoms of the $Ti_3C_2(OH)_2$ electrodes at PZC.

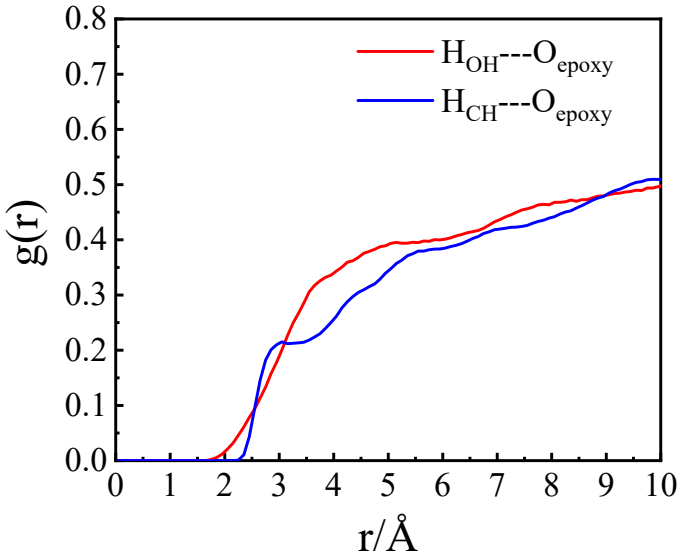


Fig. S8 Radial distribution function between H atoms of cations and O atoms of the $Ti_3C_2O_2$ electrodes at PZC.

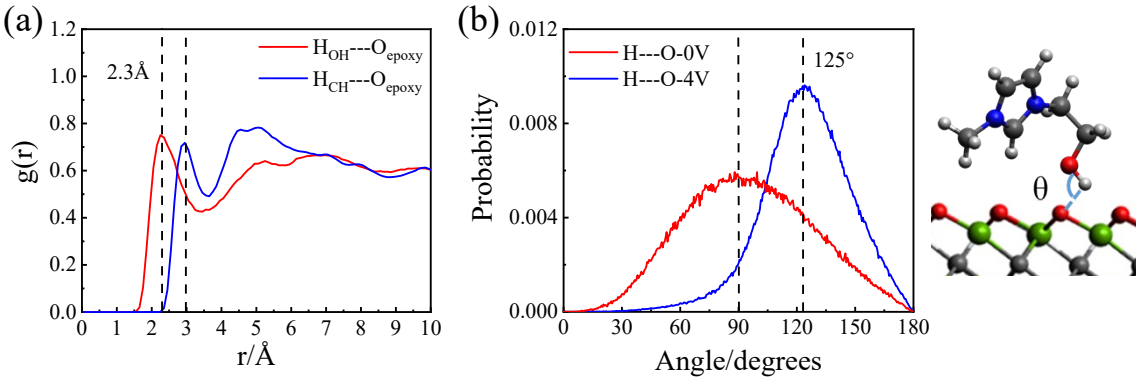
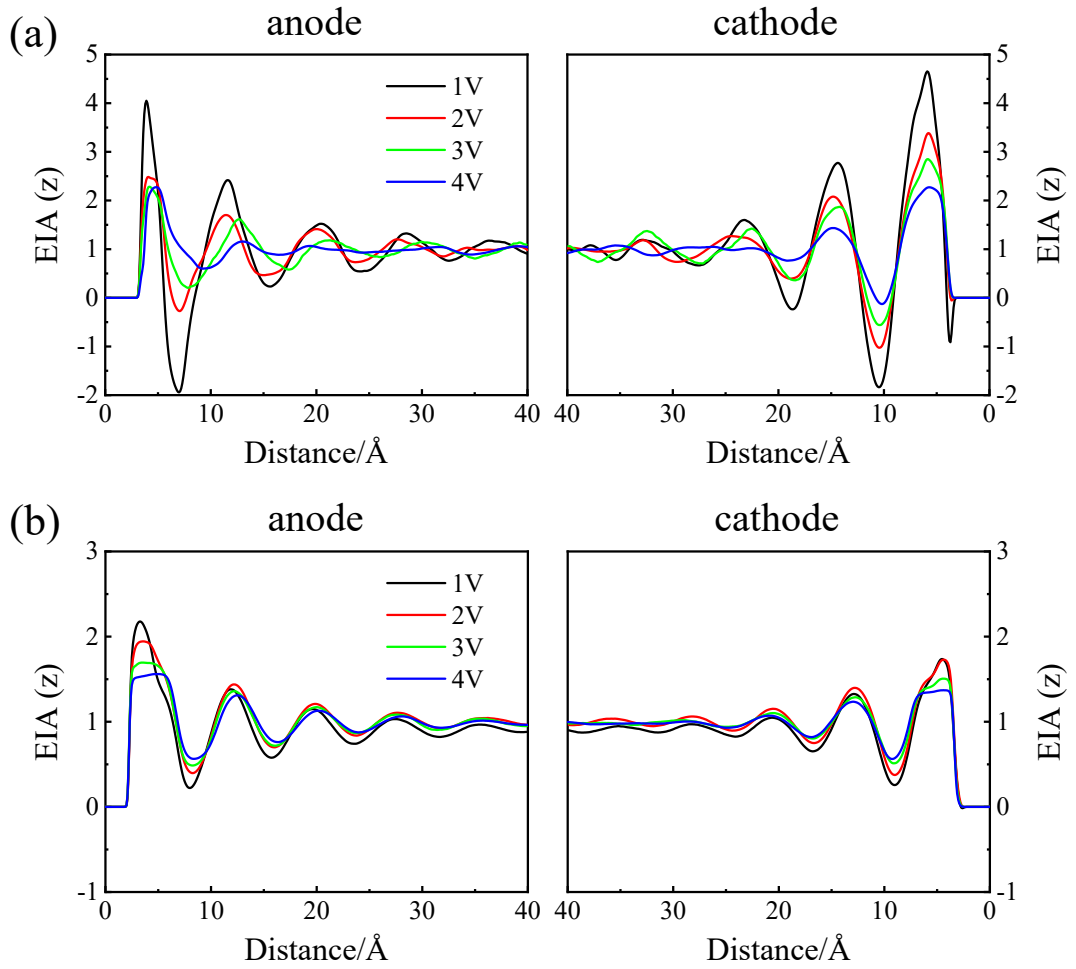


Fig. S9 (a)Radial distribution function between H atoms of cations and O atoms of $Ti_3C_2O_2$ electrodes at $\Delta\Psi = 4V$. The oxygen atoms in termination of the $Ti_3C_2O_2$ electrodes are denoted by O_{epoxy}. (b) distribution of the O-H…O angle between H_{OH} and O_{epoxy} within the distance less than 3 Å at $\Delta\Psi = 0$ and $4V$.

We quoted the calculation method of effective ion accumulation factor (EIA) to evaluate the degree of screening efficiency, following Feng

$$EIA(z) = \frac{\int_0^z \left[\rho_{N,\alpha}(s) - \rho_{N,\alpha}{}'(s)\right] ds}{|\sigma|/e}$$

where z is the distance from the electrode surface, σ is the absolute value of the charge in the electrode, e is the electron charge, and $\rho_{counter-ion}$ and ρ_{co-ion} are the number density of the counterion and coion, respectively.



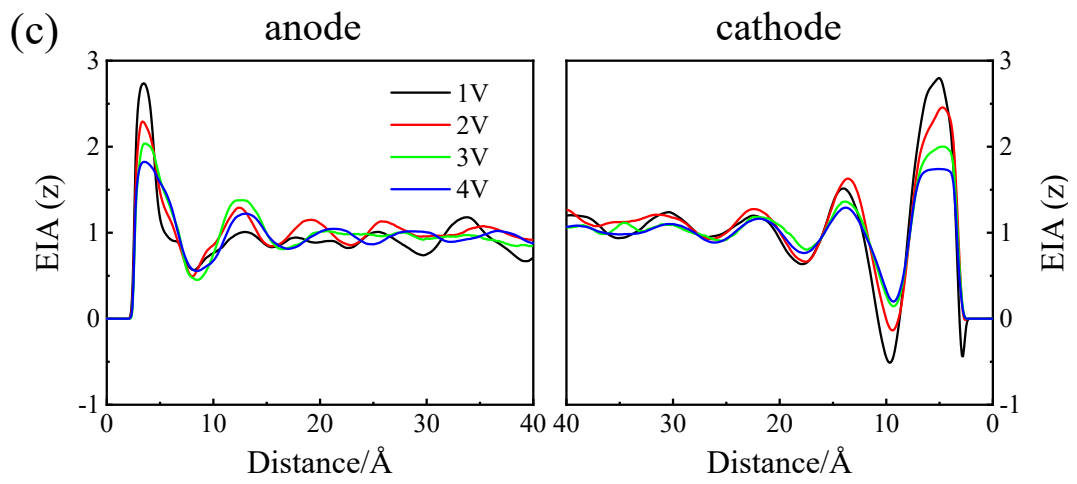


Fig. S10 EIA factor as a function of distance from the MXene electrode with different applied potentials. EIA profile for [EMIM][TFSI] at $Ti_3C_2O_2$ electrode and $Ti_3C_2(OH)_2$ electrode (a, c) and for [HEMIm][NTf_2] at $Ti_3C_2(OH)_2$ electrode (b).

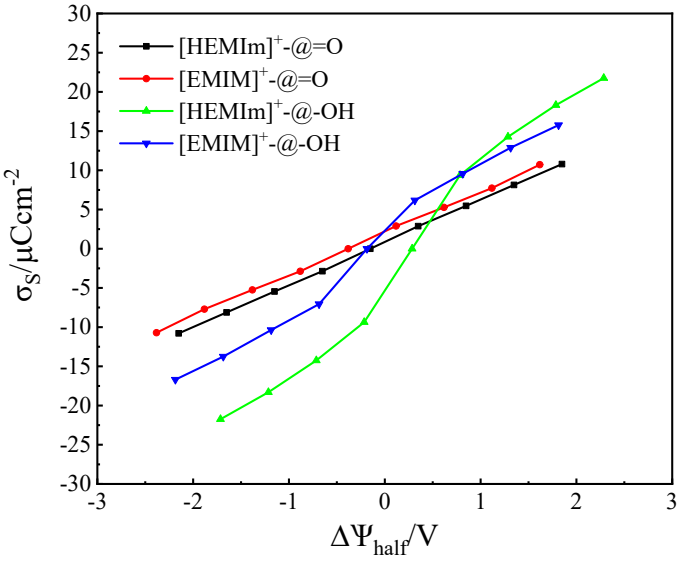


Fig. S11 Surface charge σ_s as a function of the half potential drop across the interface ($\Delta\Psi_{half} = \Psi^{\pm} - \Psi^{bulk} -$ PZC) obtained in two ILs at both electrodes.