Supporting Information

Tunable energy storage capacity of two-dimensional $Ti_3C_2T_x$ modified by a facile two-step pillaring strategy for high performance supercapacitor electrodes

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Table S1 The cationic property and the d-spacing values

	Positive charge	Ionic radius(Å)	2θ(°)	d-spacing(Å)	The number of fixed groups
Li^+	1	0.76	8.22	10.75	2
K^+	1	1.38	7.08	12.48	2
NH_4^+	1	1.43	6.96	12.69	4<X<6
Mg^{2+}	2	0.72	7.30	12.10	4
Ca^{2+}	2	1.00	7.24	12.20	4
Al^{3+}	3	0.535	7.02	12.58	6

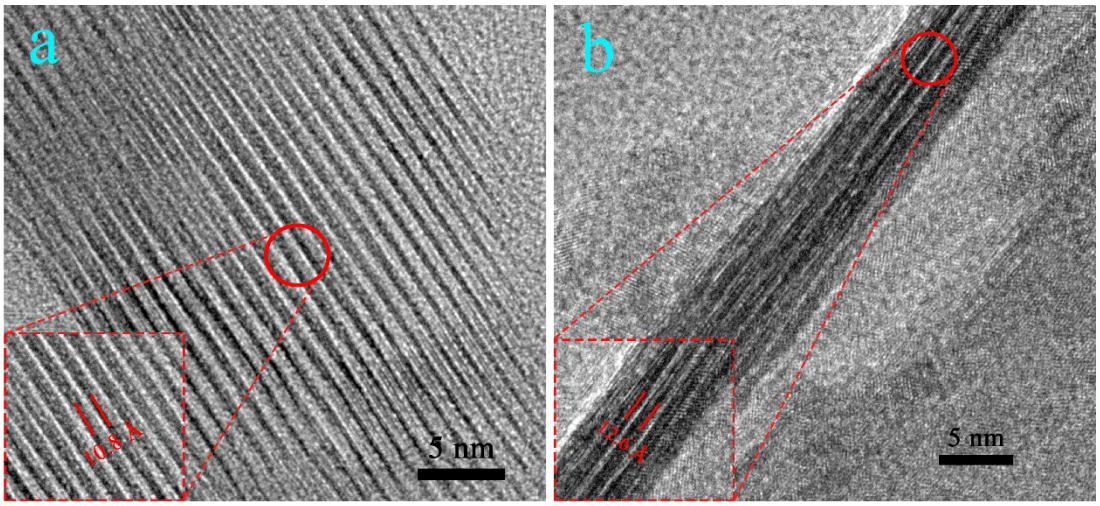

Fig. S1 HRTEM of $Ti_3C_2T_x$-Li^+; (b) $Ti_3C_2T_x$-Al^{3+}

The HRTEM of $Ti_3C_2T_x$-Li^+ and $Ti_3C_2T_x$-Al^{3+} is shown in the Fig. S1. The apparent layered structure is presented, and the test results are consistent with XRD

Table S2 Bonding ratio in Ti 2p of the similar pillared modified $Ti_3C_2T_x$

`	$Ti_3C_2T_x$-Li^+	$Ti_3C_2T_x$-K^+	$Ti_3C_2T_x$-NH_4^+	$Ti_3C_2T_x$-Mg^{2+}	$Ti_3C_2T_x$-Ca^{2+}	$Ti_3C_2T_x$-Al^{3+}
Ti-C $2p^{3/2}$	46%	37%	40%	45%	54%	45%
Ti-O $2p^{3/2}$	29%	38%	38%	27%	41%	30%
Ti-C $2p^{1/2}$	24%	18%	21%	27%	42%	22%
Ti-O $2p^{1/2}$	1%	8%	2%	2%	0	4%

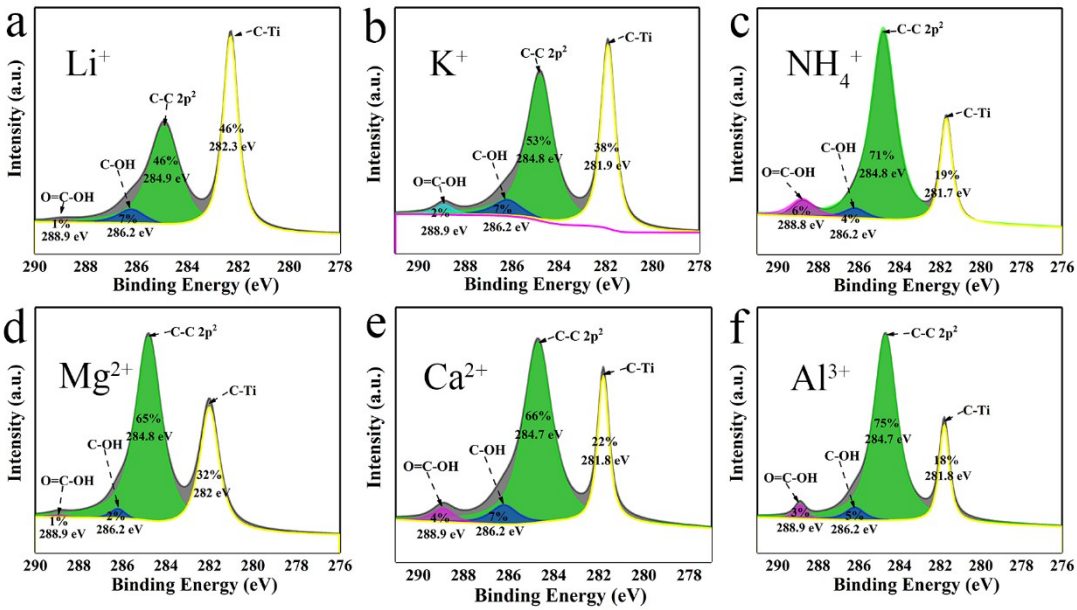

Fig. S2 C 1s spectra of (a) $Ti_3C_2T_x$-Li^+. (b) $Ti_3C_2T_x$-K^+. (c) $Ti_3C_2T_x$-NH_4^+. (d) $Ti_3C_2T_x$-Mg^{2+}. (e) $Ti_3C_2T_x$-Ca^{2+}. (f) $Ti_3C_2T_x$-Al^{3+}.

Table S3 Bonding ratio in C 1s of the similar pillared modified Ti_3C_2

	$Ti_3C_2T_x$-Li^+	$Ti_3C_2T_x$-K^+	$Ti_3C_2T_x$-NH_4^+	$Ti_3C_2T_x$-Mg^{2+}	$Ti_3C_2T_x$-Ca^{2+}	$Ti_3C_2T_x$-Al^{3+}
C-C $2p^2$	46%	53%	71%	65%	66%	75%
C-OH	7%	7%	4%	2%	7%	5%
O=C-OH	1%	2%	6%	1%	4%	3%
C-Ti	46%	38%	19%	32%	22%	18%

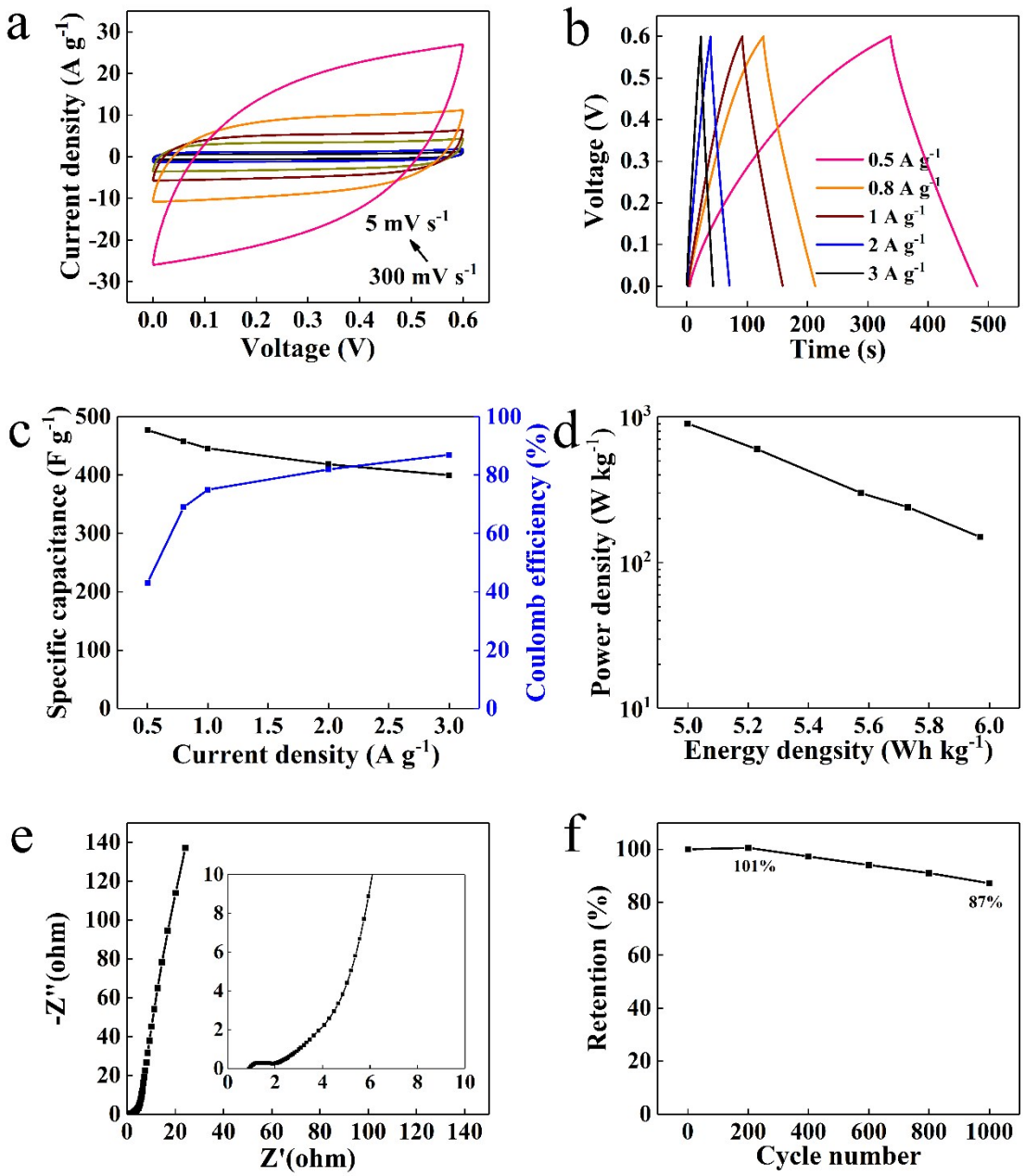

Fig. S3 Electrochemical performance of the $Ti_3C_2T_x$-Al^{3+}|| $Ti_3C_2T_x$-Al^{3+} SC device in 1 M KOH electrolyte: (a) Cyclic voltammetric curves at various scan rates (5-300 mV s^{-1}), (b) Constant current charge-discharge curve at various current densities (0.5-3 A g^{-1}), (c) Specific capacitance and Coulomb efficiency *vs.* current densities (0.5-3 A g^{-1}), (d) Ragone plots, (e) The EIS curves, (f) Capacitance retention, of the symmetrical supercapacitor

Table S4 Comparison of electrochemical performance of $Ti_3C_2T_x$ electrode

Electrode material	Specific capacitance	Scan rate (mV s^{-1})	Electrolyte	Ref.
Ti_3C_2	100F g^{-1}	2	H_2SO_4	1
Ti_3C_2	117 F g^{-1}	2	KOH	2
Ti_3C_2	70 F g^{-1}	20	EMITSI	3
$Ti_3C_2T_x$	63 F g^{-1}	5	KOH	4
d-$Ti_3C_2T_x$ paper intercalated	122 F g^{-1}	5	KOH	4
d-$Ti_3C_2T_x$ film on NF	140 F g^{-1}	5	KOH	5
TiO_2-Ti_3C_2	143 F g^{-1}	5	KOH	6
Delaminated-Ti_3C_2	150 F g^{-1}	5	H_2SO_4	7
CNT-Ti_3C_2	85 F g^{-1}	2	EMITSI	8
Ti_3C_2-216h etching	118 F g^{-1}	5	KOH	9
Li-intercalated $Ti_3C_2T_x$	134 F g^{-1}	20	KOH	10
$Ti_3C_2T_x$-Al^{3+}	154 F g^{-1}	5	KOH	This work

Table S5 Bader charge distributions of different cations pillaring $Ti_3C_2T_x$

Cations	Al^{3+}	NH_4^+	Mg^{2+}	K^+	Ca^{2+}	Li^+
Bader charge of the nearest Ti	2.60	2.58	2.41	2.23	2.20	2.19
Number of outer electrons of Ti	4	4	4	4	4	4
Charge transfer	1.40	1.42	1.59	1.77	1.80	1.81

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