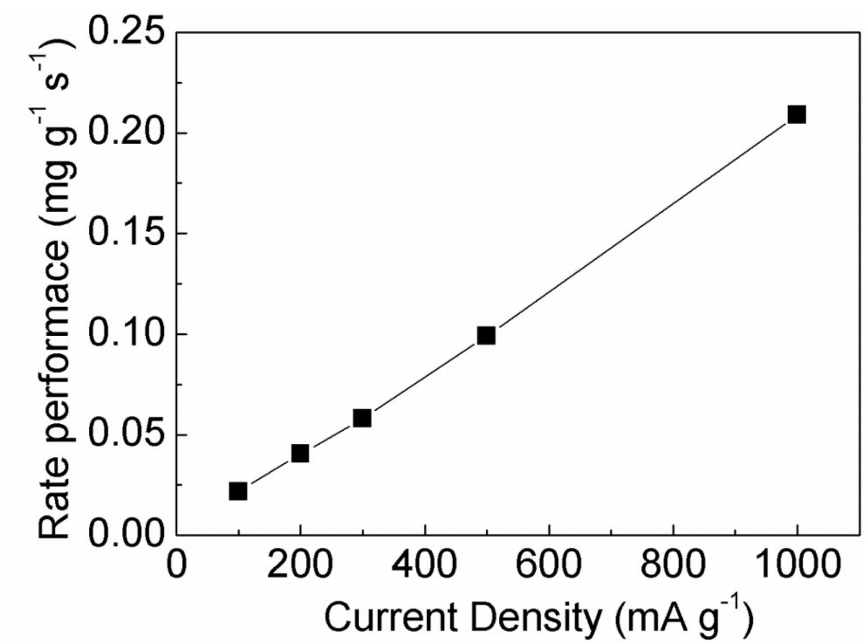
Supplementary Information

A dual-ion electrochemistry deionization system based on AgCl-$Na_{0.44}MnO_2$ Electrodes

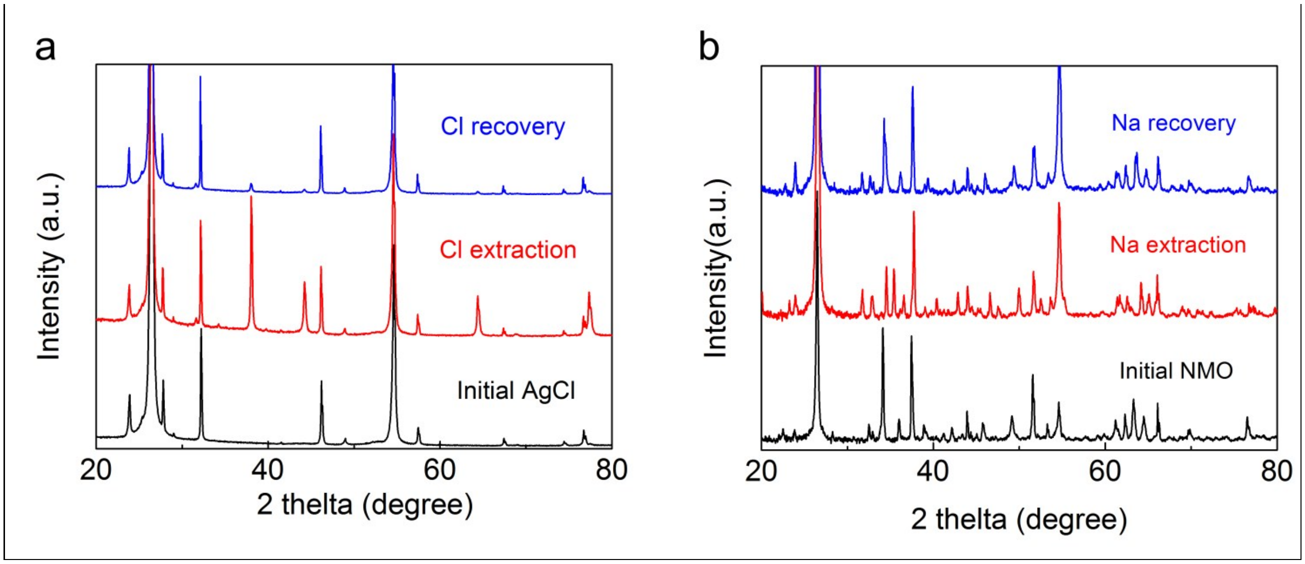
*Fuming Chen, Yinxi Huang, Lu Guo, Meng Ding, Hui Ying Yang**

Pillar of Engineering Product Development, Singapore University of Technology and Design, Singapore 487372

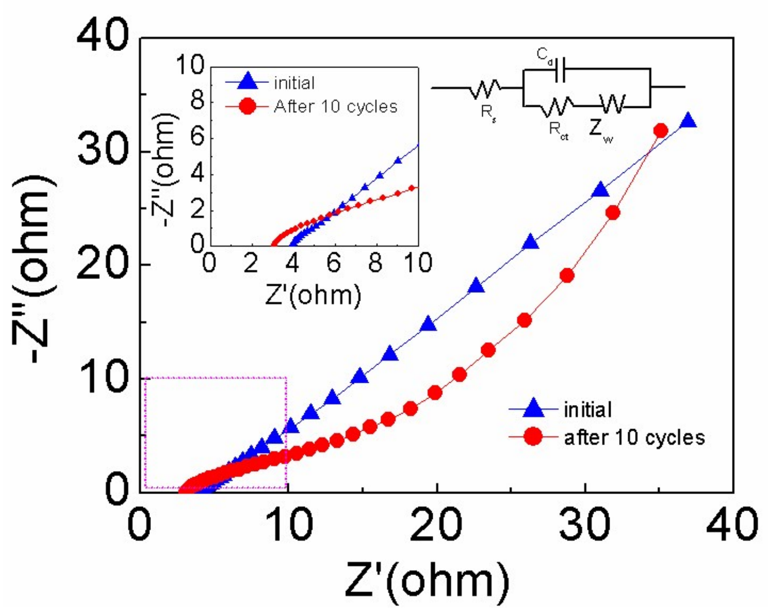
Supplementary Figures:



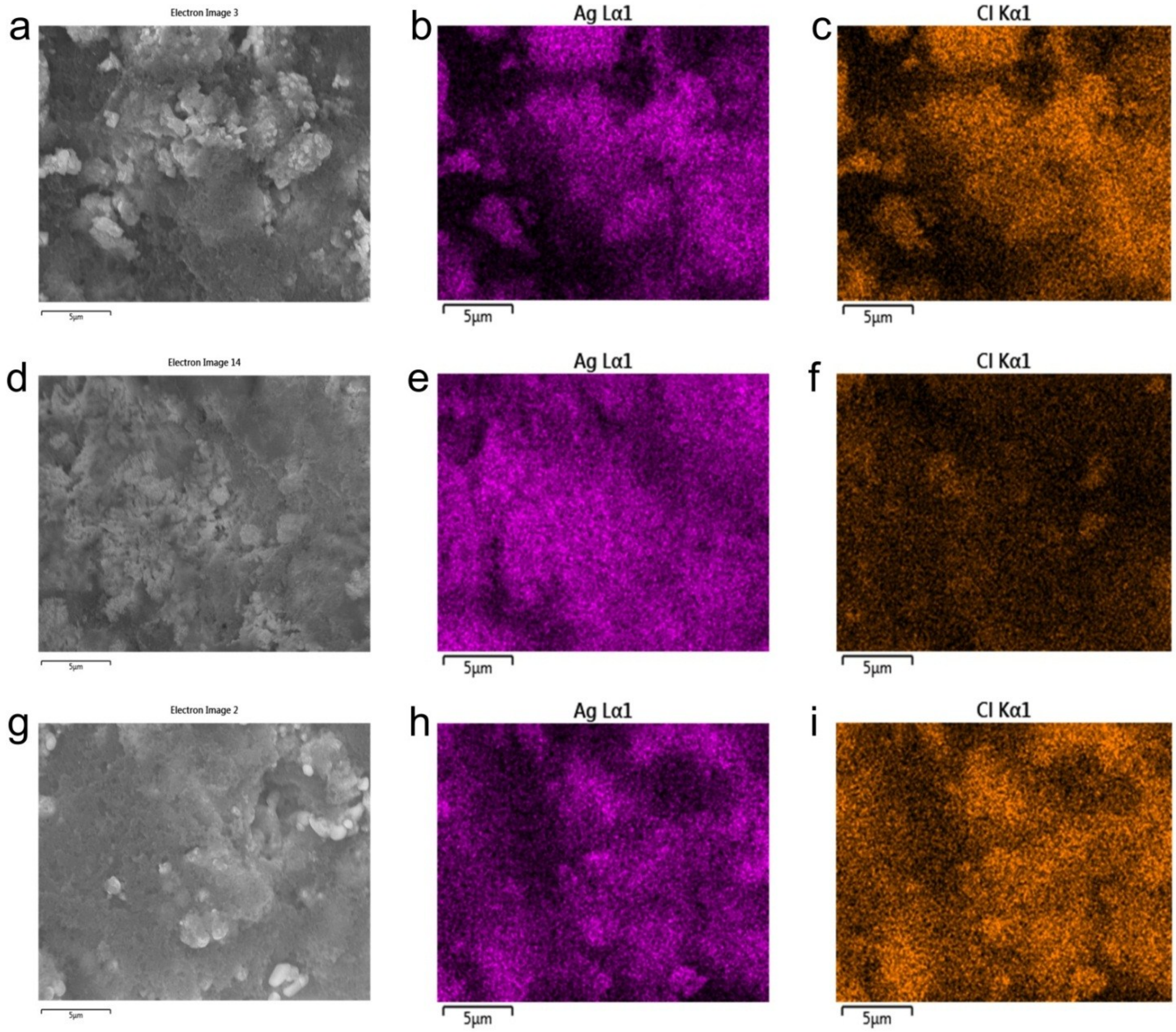
Supplementary Figure 1. The deionization rate performance at different current density



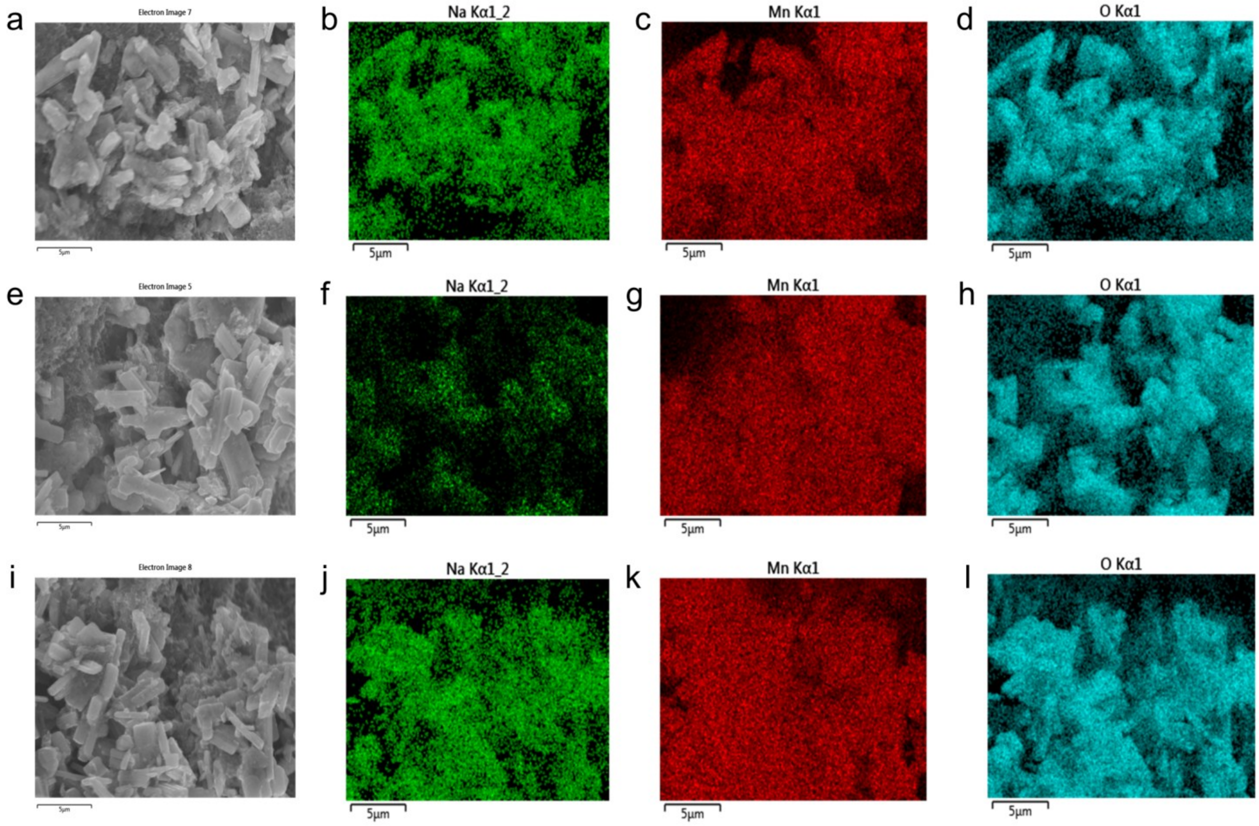
Supplementary Figure 2. (a) XRD patterns of the initial AgCl (black), the state of chloride extraction (red), and the state of chloride recovery (blue) with wide diffraction angle. (b) XRD patterns of as-prepared NMO electrode (black), the state of sodium extraction (red), and the state of sodium recovery (blue).The reflection peaks appearing at 2 theta = 26.2^{o}, 54.3^{o} are assigned to the graphite substrate.



Supplementary Figure 3. Nyquist impedance spectra of the initial state and after 10[th] cycling in the frequency regions 1M Hz-0.01Hz , Inset shows the zoom-in area of the high frequent part and the circuitry model for The Nyquist diagram. Rs: the resistance between the current collector and electrolyte; Rct: charge transfer resistance; C_t: double layer capacitor; Z_w: the Warburg impedance related to the ion diffusion.



Supplementary Figure 4 the SEM images of as-prepared AgCl (a), the corresponding EDX mapping of silver (b), chloride (c); the SEM images of chloride extraction state of AgCl (d), the corresponding EDX mapping of silver (e), chloride (f); the SEM images of chloride recovery state of AgCl (g), the corresponding EDX mapping of silver (h), chloride (i).



Supplementary Figure 5. the SEM images of as-prepared NMO (a) and the corresponding EDX mapping of sodium (b), manganese(c), and oxide (d); the SEM images of sodium extraction state of NMO (e) and the corresponding EDX mapping of sodium (f), manganese(g), and oxide (h); the SEM images of sodium recovery state of NMO (i) and the corresponding EDX mapping of sodium (j), manganese(k), and oxide (l)

Supplementary Tables:

Supplementary Table 1. The comparison of the two groups of electrode system (Ag-NMO and AgCl-NMO)

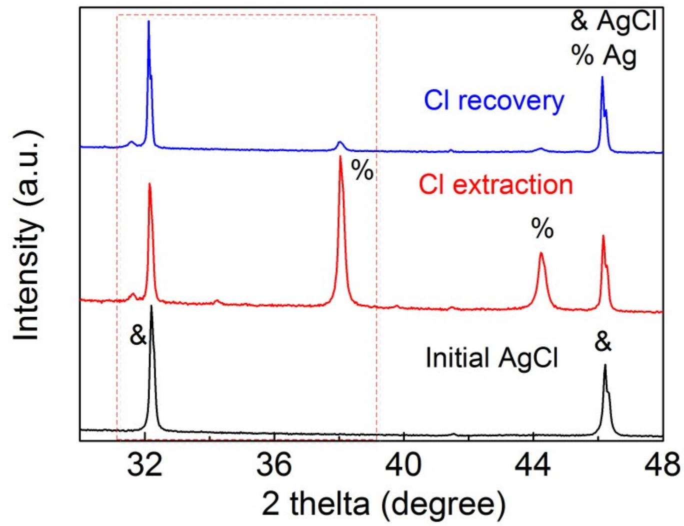
System	Initial state	Role of NMO	Remark
Ag-NMO	charge state	Na acceptor	Ref. 1, energy storage device with the function of salt removal at static 300 micro litre of feed condition, one cycle demonstrated, no capacity reported. The overall reaction: $Ag + Na_{0.44}MnO_2 + NaCl = AgCl + Na_{0.44+x}MnO_2$
AgCl-NMO	discharge state	Na donor	Current research, deionization devices with stable and reversible salt removal capacity with 50 ml flow electrolyte. 100 cycles demonstrated, capcity reported. The overall reaction: $xAgCl + Na_{0.44} MnO_2 = xAg + Na_{0.44-x}MnO_2 + xNaCl$

Supplementary Table 2. The deionization types and features

Deionization technology	**Features**	**Max absorption capacity mg/g**	**References**
Conventional CDI	Carbon materials, ion absorption of electrical double layer, ion physical absorption by carbon electrodes	7-17	[2-12].
Hybrid CDI	Sodium ions battery materials for chemical capture at one electrode side, chloride ions are physical adsorbed by carbon materials.	31.2	[13]
		30.2	[14]
Battery deionization	Dual-ion electrochemical technology, sodium ions battery materials for chemical capture of sodium at one electrode side, chloride ions battery materials for electrochemical intercalation of chloride	57.4	This work

Supplementary Table 3. the peak intensity ratio (silver at 38.0°/AgCl at 32.1°) of initial state, the state of chloride extraction, and the state of chloride recovery of AgCl electrode.

From Figure 5(a)



	AgCl electrode
i.e. circled in red	Peak intensity ratio (% at 38.0°/& at 32.1° marked in red area)
Initial state	0.00
Chloride extraction	1.25
Chloride recovery	0.06

Supplementary Table 4. Fitting results of the EIS spectra with the configurative circuitry model in Fig. S3

Sample	R_s (Ω)	R_{ct} (Ω)
Initial	3.773	11.78
After 10 cycle	2.802	13.57

Supplementary Table 5. the element composition of Ag and Cl in the as-prepared AgCl, chloride extraction state and chloride recovery, and the calculated the atomic ratio of Cl:Ag

atomic %	*Ag*	*Cl*	*Cl:Ag*
Initial AgCl	52.3	47.7	**0.912**
Chloride extraction	84.3	15.7	**0.186**
Chloride recovery	53.8	46.2	**0.859**

Supplementary Table 6. the element composition of Na, Mn, and O in the as-prepared NMO, sodium extraction state and sodium recovery state, and the calculated the atomic ratio of Na:Mn

atomic %	*Na*	*Mn*	*O*	*Na:Mn*
Initial NMO	17.7	34.6	47.7	**0.51**
Sodium extraction	9	40.7	50.3	**0.22**
Sodium recovery	14.5	36	49.5	**0.40**

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