Supplementary information

H_2O assisted to improve the electrochemical performance of deep eutectic electrolyte formed by choline chloride and magnesium chloride hexahydrate

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Experimental Section

Preparation of electrode and physical characterizations:

To ensure that the contact angle data conforms to the real situation, the prepared electrodes were adopted in the contact angle measurement. The N, O co-doped porous carbon electrode was prepared by coating the slurries on Al foils. The slurries contain 80 wt% of active materials, 10 wt.% of acetylene black and 10 wt.% of polyvinylidene fluoride (PVDF), which are uniformly dispersed in N-methyl-2-pyrrolidone (NMP).

Theoretical Calculations

The root-mean-square displacement (MSD) and corresponding self-diffusion coefficient (D) of the four particles were calculated using the following formulas:

$$MSD = D\,t^{a} \quad (0 < a < 2)$$

$$D = \frac{1}{6t}\lim_{t\to\infty}\{[r(t) - r(0)]\}^{2}$$

Where ***t*** (ps) is the total time of the simulation process; ***a*** is a time index, commonly used to measure the particle diffusion pattern; and ***r*** (A) is the diffusion displacement.

The relevant formulas for calculating the radial distribution function (RDF) are shown in following formulas:

$$g(r) = \frac{\frac{dN}{4\pi r^{2}dr}}{\frac{N}{a \times b \times c}}$$

$$dN = \frac{g(r) \times N \times 4\pi r^{2}dr}{a \times b \times c}$$

The total number of target particles in the system is denoted as ***N***, while the length, width, and height of the theoretical calculation model are represented by ***a*** (A), ***b*** (A), and ***c*** (A) respectively. Parameter ***g(r)*** describes the ratio between the number of target particles (B) existing in the shell of an A particle (A) with radius ***dr*** and the density of B particles in the entire system. It reflects how interaction intensity between particles changes with distance and its derived meaning is the probability of an A particle finding

B particles within the entire framework. ***dN*** represents the number of B particles around A particles in this shell, and its integral area can be defined as the coordination number for each particle under the assumptions that system particles are evenly distributed and truncation radius ***r*** is reasonable.

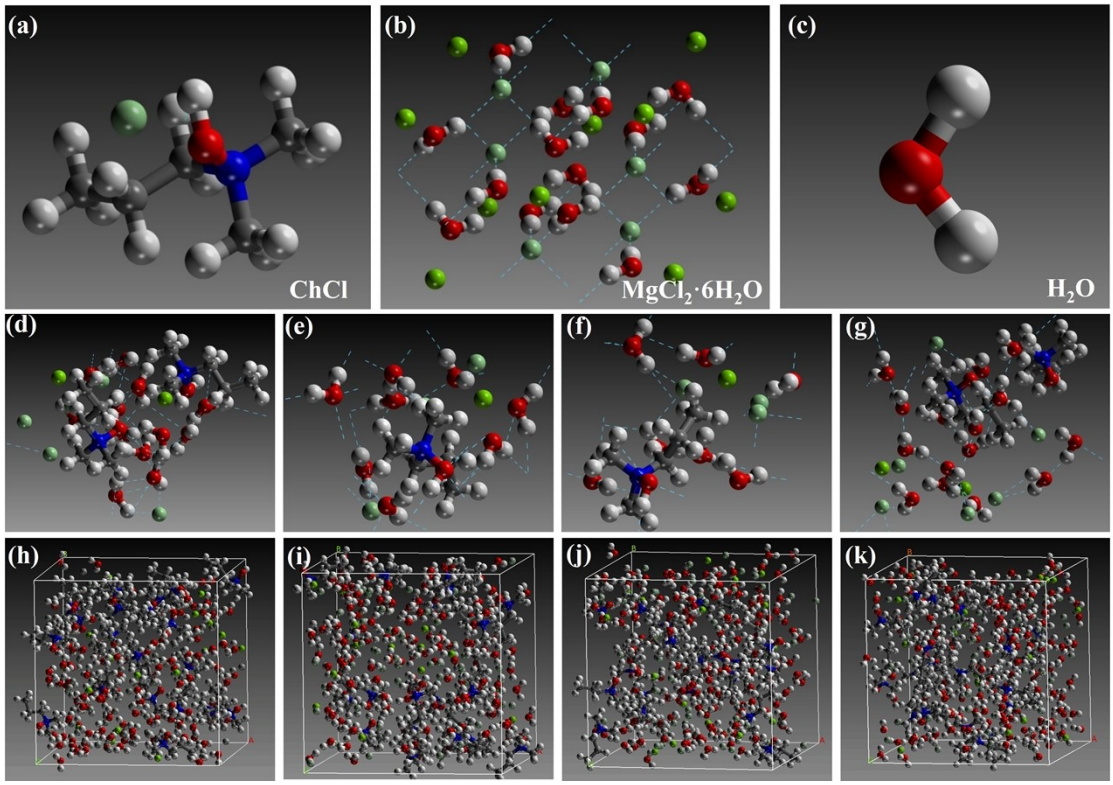

Fig. S1 The constructed modes of (a-c) choline chloride, crystalline magnesium chloride, water, and DES systems: (d, h) DES-1, (e, i) DES-1-0.5, (f, j) DES-1-1., (g, k) DES-1-1.5

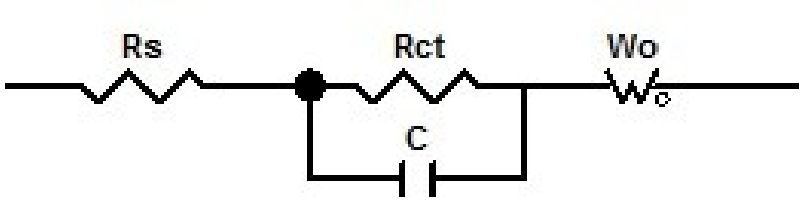

Fig. S2 The electrical equivalent circuit for the calculation of the resistances.

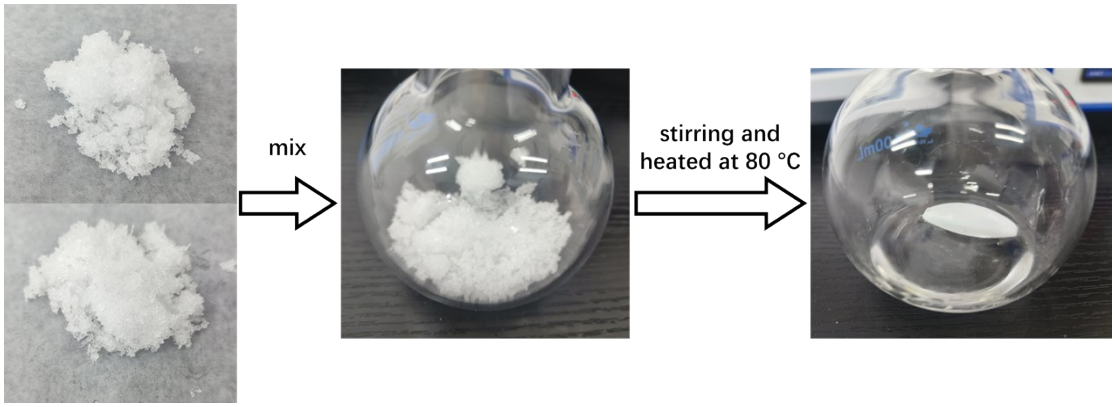

Fig. S3 Schematic illustration of the synthesis processes of DES electrolytes.

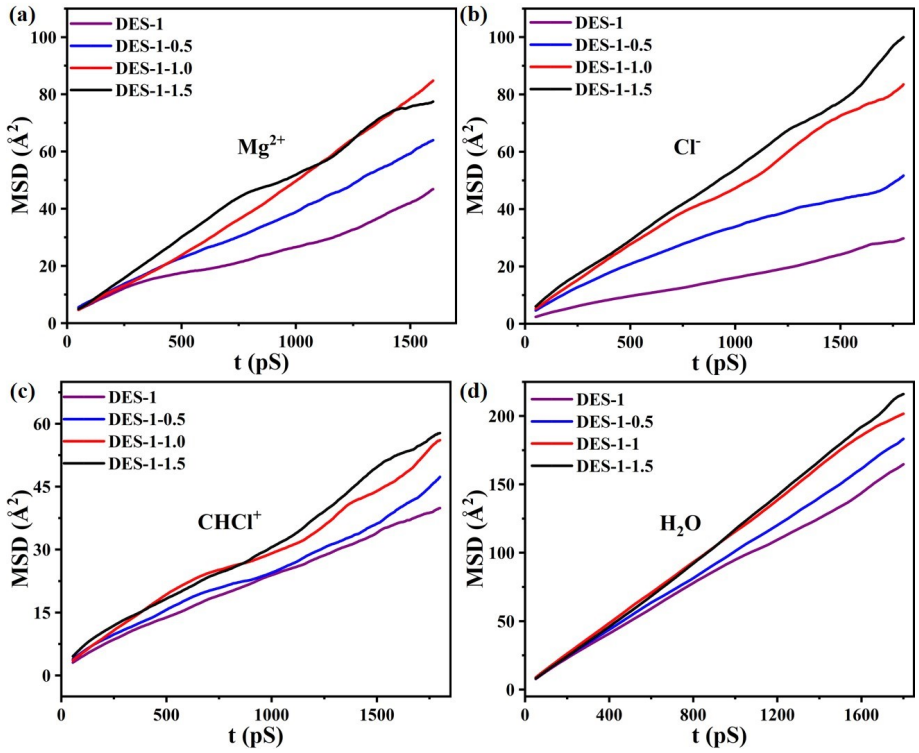

Fig. S4 The Mean square displacement (MSD) of different atoms (a) Mg; (b) Cl; (c) N; (d) H_2O in various systems

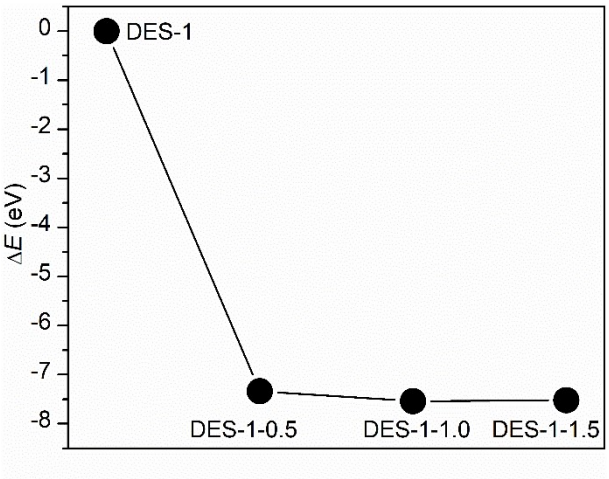

Fig. S5. The average adsorption energy of the DES-1, DES-1-0.5, DES-1-1.0, DES-1-1.5.

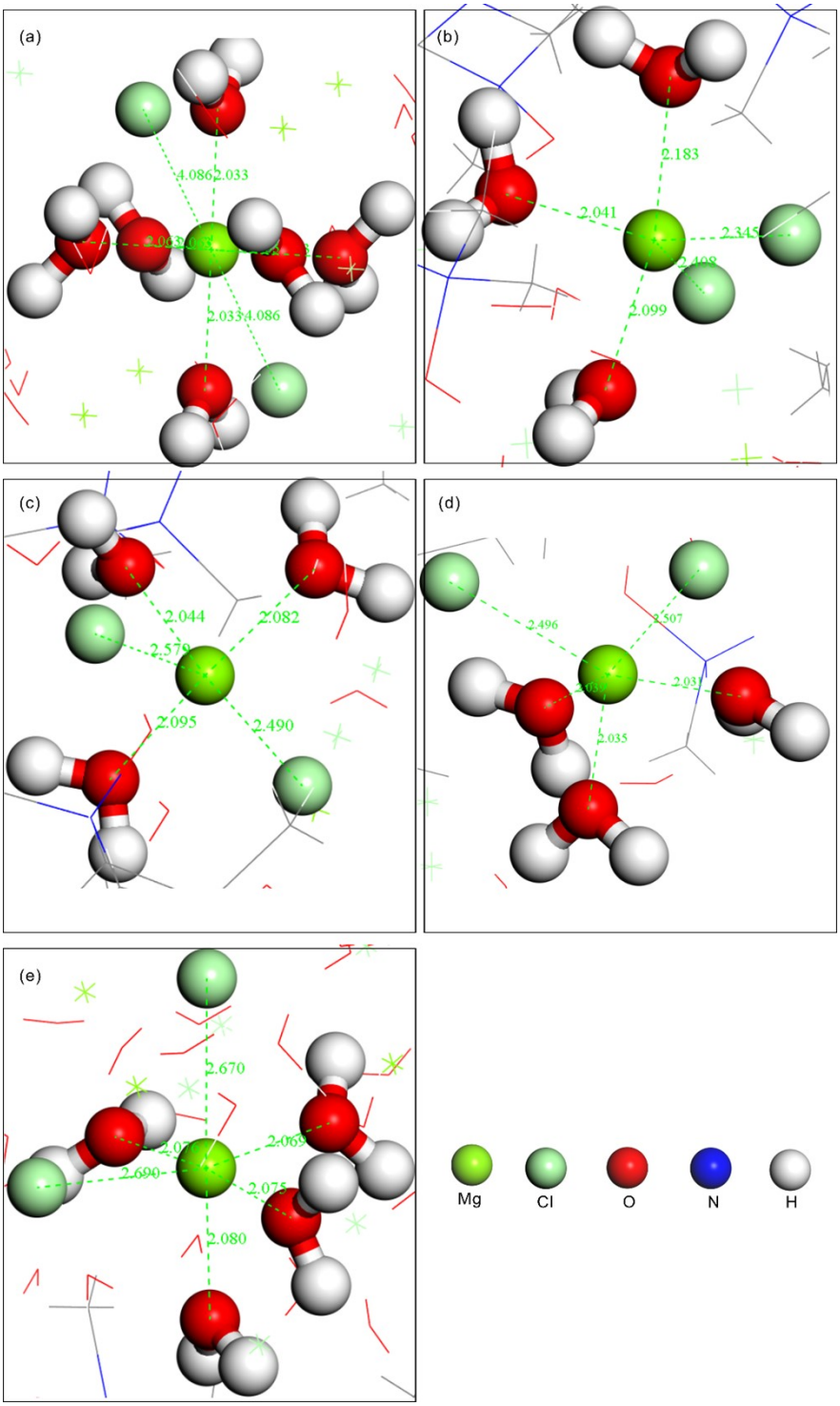

Fig. S6. The coordination structure of Mg in different systems and the corresponding bond lengths. （a）The bulk of magnesium chloride hexahydrate.（b）DES-1, (c) DES-1-0.5, (d) DES-1-1.0, (e) DES-1-1.5.

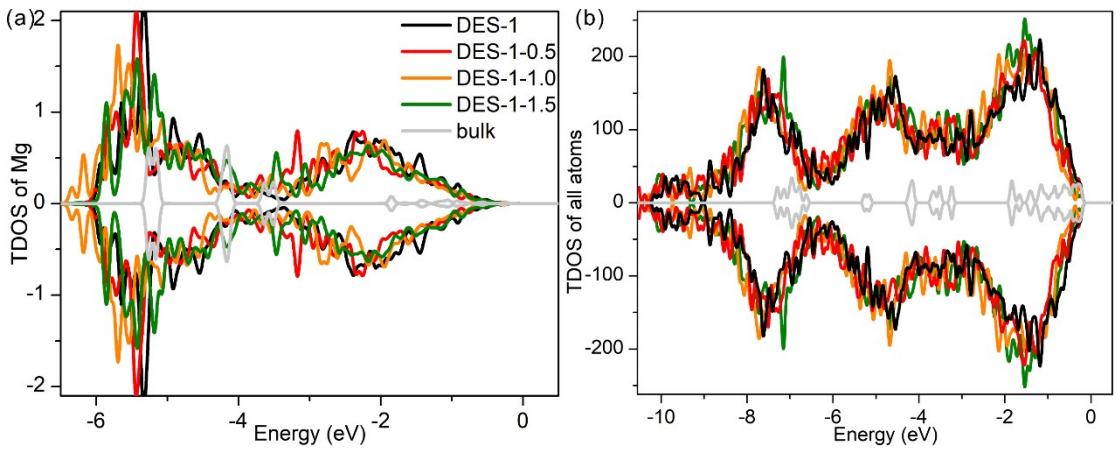

Fig. S7. The total density of states (TDOS). (a) TDOS of all Mg atoms, and (b) TDOS of all atoms in DES-1, DES-1-0.5, DES-1-1.0 and DES-1-1.5.

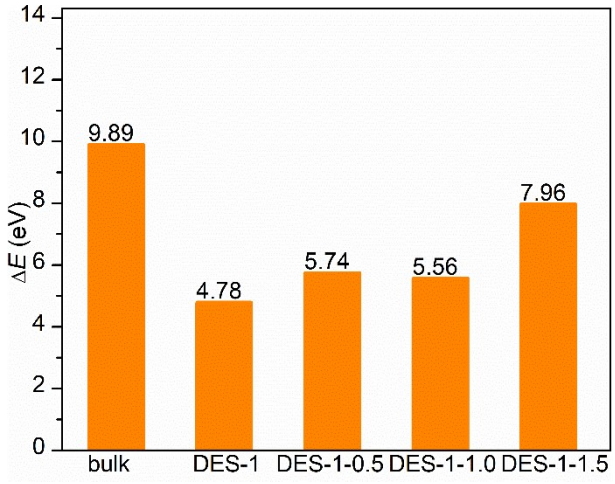

Fig. S8. The energy required to remove the $MgCl_2$ unit in DES-1, DES-1-0.5, DES-1-1.0 and DES-1-1.5.

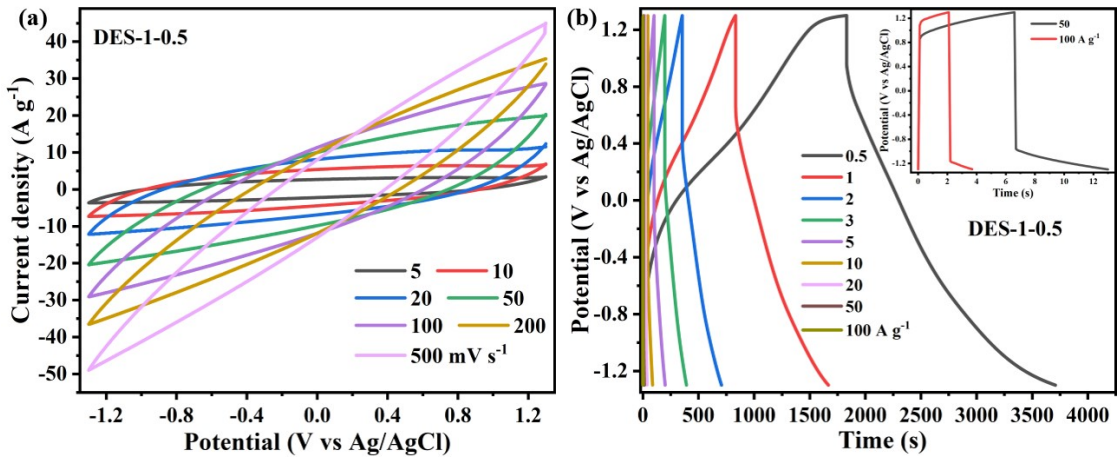

Fig. S9 (a) CV curves at different scan rates and (b) GCD curves at different current density of the three-electrode system using DES-1-0.5 electrolyte.

Table S1. The calculation of the total number, average lengths and total energy of H-bonds in DES-1, DES-1-0.5, DES-1-1.0 and DES-1-1.5.

Sample	**N_{Hbond}**	**Average (Å)**	**E_{Hbond} (eV)**
DES-1	220	2.09	-156.77
DES-1-0.5	237	2.07	-159.58
DES-1-1.0	252	2.03	-165.16
DES-1-1.5	268	2.04	-166.89

Table S2. Relative concentrations of H-bonds in DES-1, DES-1-0.5, DES-1-1.0 and DES-1-1.5

Raman shift (cm^{-1})	DES-1	DES-1-0.5	DES-1-1.0	DES-1-1.5
3215	21.50%	22.14%	20.96%	19.57%
3386	62.60%	60.89%	59.49%	55.55%
3484	14.20%	14.64%	17.10%	21.87%
3578	1.26%	1.18%	1.26%	1.59%
3644	0.44%	1.16%	1.20%	1.43%

Sample	DES-1	DES-1-0.5	DES-1-1.0	DES-1-1.5
Viscosity (Pa s)	1.802	0.464	0.074	0.037
Conductivity (mS/cm)	2.75	6.45	14.59	18.36

Table S3. Viscosity and conductivity of DES-1, DES-1-0.5, DES-1-1.0 and DES-1-1.5

Table. S4 Comparisons of electrochemical performances of the recently reported symmetric SCs using other state-of-the-art electrolytes.

Sample	Electrolytes	Operating Voltage window (V)	Energy density (Wh/kg)	Power density (kW/kg)	Ref.
Three-dimensional graphene like material	Reline DES	2.2	19.5	1	20
Active carbon	$NaNO_3$ /Formamide DES	2.6	12.55	17.37	21
Active carbon	37 mol/kg KFSI in H_2O	2.3	20.5	2.3	46
YP-50F	17 M $NaClO_4$ in H_2O	2.3	23.7	1.15	47
N/O/S ternary-doped porous carbon	7 M LiTFSI in H_2O	2.3	34.3	0.575	48
Hierarchically porous carbon	$EMIMBF_4$ in FEC/EA (1:1 in volume)	2.5	35.7	21.1	49
Activated carbon	SBS-g-PEGBEM	2.4	28.1	3.08	50
N, O co-doped hierarchically porous carbon	**DES-1-1.0**	**2.6**	**87.7**	**1.69**	**This work**