

Supplementary Information

Historical development and novel concepts upon electrolytes for aqueous rechargeable batteries

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Table S1 A summary of recently representative works upon additive-added electrolytes for ARBs.

Electrolyte	Electrode	ESW	Energy density	Cyclic stability	Working temperature	Reference
SDS-added 1 M Na ₂ SO ₄ +1 M ZnSO ₄	Zn//Na ₂ MnFe(CN) ₆	2.5 V	170 Wh·kg ⁻¹ (cathode)	2000 cycles (5 C)	RT	Ref. 1
2 vol.% Et ₂ O added 3 M Zn(CF ₃ SO ₃) ₂ +0.1 M Mn(CF ₃ SO ₃) ₂	Zn//MnO ₂	n/a	n/a	4000 cycles (5 A·g ⁻¹)	RT	Ref. 2
SDBS added 1 M LiSO ₃ CF ₃ and 1 M Zn(SO ₃ CF ₃) ₂	Zn//LiFePO ₄	n/a	113 Wh·kg ⁻¹ (total electrode)	500 cycles (5 C)	RT	Ref. 3
0.5 m Me ₃ EtNOTF added 4 m Zn(OTF) ₂	Zn//VOPO ₄	2.7 V	136 Wh·kg ⁻¹ (total electrode)	6000 cycles (2 A·g ⁻¹)	RT	Ref. 4
DMSO added 2 M NaClO ₄	Na ₃ V ₂ (PO ₄) ₃ //Na ₃ V ₂ (PO ₄) ₃	3.1 V	n/a	100 cycles (1 A·g ⁻¹)	RT	Ref. 5
2 M HBF ₄ + 2 M Mn(BF ₄) ₂	Alloxazine//MnO ₂	~1.8 V	110 Wh·kg ⁻¹ (total electrode)	500 cycles (4 C)	-60 °C	Ref. 6
ZnF ₂ added 2 M ZnSO ₄ +1 M Li ₂ SO ₄	Anode-free//LiMn ₂ O ₄	n/a	n/a	100 cycles (200/400 mA·g ⁻¹)	RT	Ref. 7
5 M H ₂ SO ₄	Pb//PCHL-rGO	n/a	n/a	3000 cycles (10 A·g ⁻¹)	RT to -70 °C	Ref. 8

Table S2 A summary of recently representative works upon pH-adjusted electrolytes for ARBs.

Electrolyte	Electrode	ESW	Energy density	Cyclic stability	Working temperature	Reference
1 M ZnSO ₄ +1.5 M HOAc+1.5 M NaOAc 1 M FeSO ₄ +1.5 M FeCl ₂	Zn//Fe	n/a	n/a	200 cycles (30 mA·cm ⁻²)	RT	Ref. 9
1 M ZnSO ₄ -P2VP	Zn//I ₂	n/a	220 Wh·kg ⁻¹ (cathode)	250 cycles (0.3 A·g ⁻¹)	RT	Ref. 10
0.1 M H ₂ SO ₄ +1 M ZnSO ₄ +1 M MnSO ₄	Zn//MnO ₂	2.41 V	409 Wh·kg ⁻¹ (total electrode)	2000 cycles (30 mA·cm ⁻²)	RT	Ref. 11
6 M KOH+0.2 M ZnO+5 mM vanillin 3 M H ₂ SO ₄ +0.1 M MnSO ₄	Zn//MnO ₂	n/a	1621.7 Wh·kg ⁻¹ (cathode)	200 cycles (500 mA·g ⁻¹) ¹⁾	RT	Ref. 12
3 M NaOH+0.2 M ZnO 2 M ZnSO ₄ +0.5 MnSO ₄	Zn//MnO ₂	3.46 V	n/a	4000 cycles (10 mA·cm ⁻²) ²⁾	RT	Ref. 13
1 M LiOH+1 M Li ₂ Zn(OH) ₄ 5 M LiNO ₃	Zn//LiMn ₂ O ₄	3.0 V	208 Wh·kg ⁻¹ (total electrode)	1000 cycles (1.69 C)	RT	Ref. 14

Table S3 A summary of recently representative works upon gelled electrolytes for ARBs.

Electrolyte	Electrode	ESW	Energy density	Cyclic stability	Working temperature	Reference
25 m LiTFSI-PVA	LiVPO ₄ F// LiVPO ₄ F	3.2 V	141 Wh·kg ⁻¹ (total electrode)	4000 cycles (20 °C)	RT	Ref. 15
2 m LiTFSI-PEG	Li ₄ Ti ₅ O ₁₂ //LiMn ₂ O ₄	3.2 V	110 Wh·kg ⁻¹ (total electrode)	300 cycles (1 °C)	RT	Ref. 16
1 M NaCl+1 M ZnCl ₂ -SA	Zn//CuHCF-CNT	2.72 V	440 Wh·kg ⁻¹ (cathode)	450 cycles (1 A·g ⁻¹)	RT	Ref. 17
1 M ZnSO ₄ -gelatin 1 M CuSO ₄ -gelatin	Zn//S	n/a	2372 Wh·kg ⁻¹ (cathode)	100 cycles (500 mA·g ⁻¹) 1)	RT	Ref. 18
7 M LiCl-SBMA+HEA	AC//AC	2.2 V	n/a	10000 cycles (5 mA·cm ⁻²)	-30 °C, RT, 60 °C	Ref. 19
2 M ZnSO ₄ +0.2 M MnSO ₄ -PVA-B-G	Zn//MnO ₂	~2.0 V	25.8 mWh·cm ⁻³	2000 cycles (1 A·g ⁻¹)	-30 °C	Ref. 20

Table S4 A summary of recently representative works upon interface-tuning electrolytes for ARBs.

Electrolyte	Electrode	ESW	Energy density	Cyclic stability	Working temperature	Reference
20 mM $\text{Zn}(\text{NO}_3)_2$ +3 M $\text{Zn}(\text{OTf})_2$	Zn// MnO_2	n/a	168 $\text{Wh}\cdot\text{kg}^{-1}$ (total electrode)	700 cycles (10 C)	RT	Ref. 21
21 m LiTFSI+7 m LiOTf-PVA	LiTFSI- HFE+PEO+DMC/graphite// LiMn_2O_4 or LiVPO_4F	~3.1 V	approaching state- of-art LiBs	50 cycles (0.3 C)	RT	Ref. 22
9.5 M LiNO_3 + LiOH (pH=10)	AC// LiNO_2	n/a	n/a	50 cycles (1 C)	RT	Ref. 23
15 m NaClO_4	TiS_2 - Al_2O_3 //MFCN- TiO_2	3.5 V	100 $\text{Wh}\cdot\text{kg}^{-1}$ (total electrode)	200 cycles (1 C)	RT	Ref. 24
2 M ZnSO_4	Zn-graphite// MnO_2	n/a	164 $\text{Wh}\cdot\text{kg}^{-1}$ (total electrode)	1000 cycles (8 $\text{mA}\cdot\text{cm}^{-2}$)	RT	Ref. 25
2 M $\text{Al}(\text{OTf})_3$	Zn/Al// Al_xMnO_2	n/a	n/a	80 cycles (100 $\text{mA}\cdot\text{g}^{-1}$) 1)	RT	Ref. 26

Table S5 A summary of recently representative works upon beyond concentrated electrolytes for ARBs.

Electrolyte	Electrode	ESW	Energy density	Cyclic stability	Working temperature	Reference
33 m LiNO ₃ -PVA	VO ₂ //LiNi _{0.5} Mn _{1.5} O ₄ or LiMn ₂ O ₄	3.2 V	176 Wh·kg ⁻¹ (total electrode)	700 cycles (3 C)	RT to 80 °C	Ref. 27
1 m Zn(ClO ₄) ₂ +10 m LiClO ₄ -PVA	Zn//LiMn ₂ O ₄	3.3 V	183 Wh·kg ⁻¹ (cathode)	300 cycles (1 C)	RT to 80 °C	Ref. 28
1 m Zn(OAc) ₂ +40 m KOAc-PAA	Zn//MnO ₂	3.45 V	386 Wh·kg ⁻¹ (cathode)	2000 cycles (5 C)	RT to 80 °C	Ref. 29
1 M Zn(ClO ₄) ₂	Zn//AC	n/a	n/a	70000 cycles (1 A·g ⁻¹)	-30 °C & -60 °C	Ref. 30
45 m ZnBr _{0.5} Cl _{1.5} +1 m Zn(OAc) ₂	Zn/GFF//PGA	n/a	908.5 Wh·kg ⁻¹ (cathode)	500 cycles (1 A·g ⁻¹)	RT	Ref. 31
42 m LiTFSI+21 m Me ₃ EtN·TFSI	Li ₄ Ti ₅ O ₁₂ //LiMn ₂ O ₄	3.25 V	145 Wh·kg ⁻¹ (total electrode)	150 cycles (0.2 C)	RT	Ref. 32

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