Electronic Supplementary Information for

Ordered interface regulation at Zn electrodes induced by trace gum additives for high-performance aqueous batteries

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Supplementary figures and tables

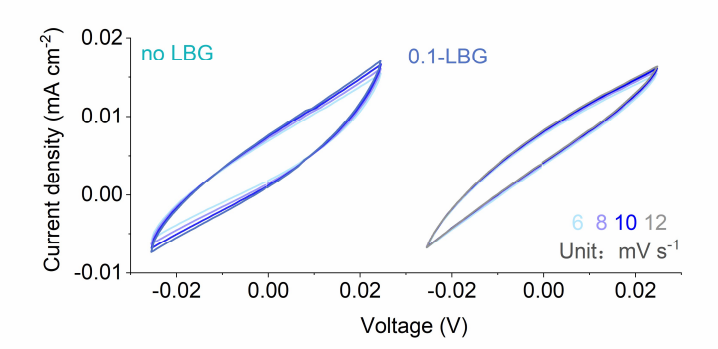

Fig. S1 CV curves of Zn symmetric cells in the non-Faradic range in 1 m $ZnSO_4$ and 0.1-LBG electrolytes.

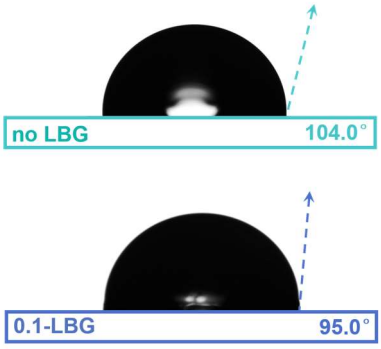

Fig. S2 Contact angles of 1 m $ZnSO_4$ and 0.1-LBG on Zn foil.

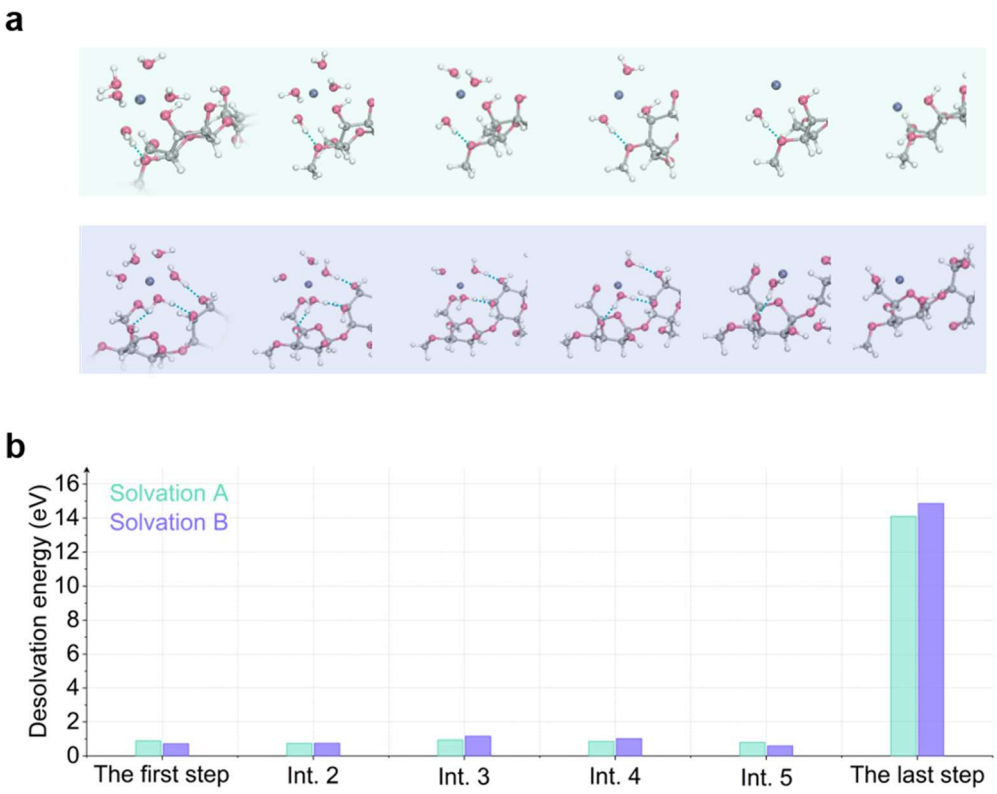

Fig. S3 a) The structures and b) energy barriers at each step along the desolvation paths of the two $Zn(H_2O)_5LBG^{2+}$ structures (A and B).

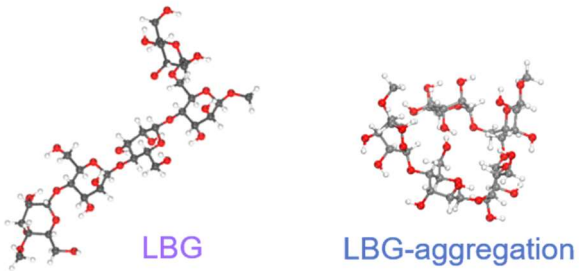

Fig. S4 The two configurations of LBG unit with a slight energy reduction of 0.11 eV after aggregation.

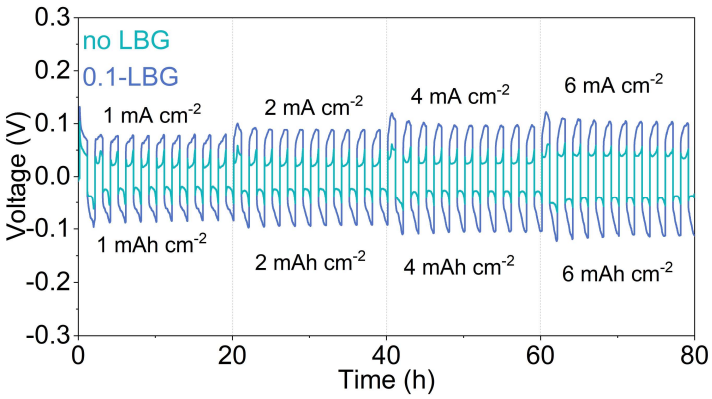

Fig. S5 The rate performance of Zn symmetric cells in the baseline $ZnSO_4$ and 0.1-LBG electrolytes.

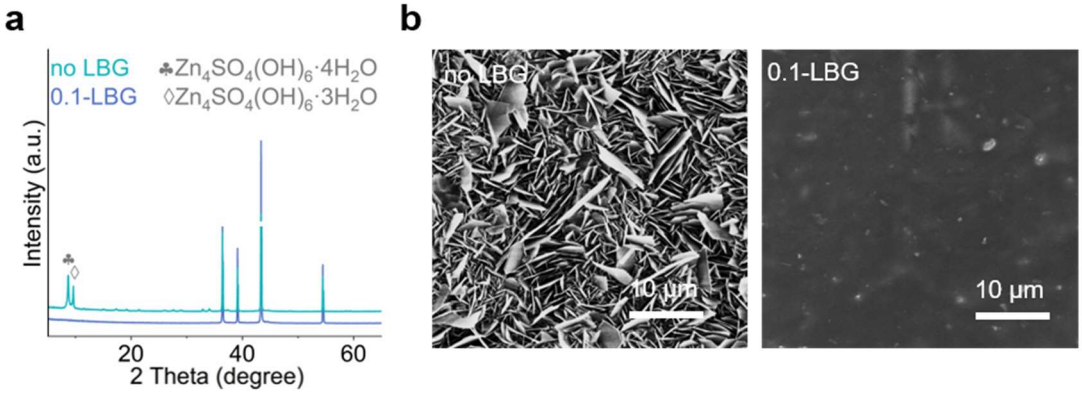

Fig. S6 a) XRD patterns and b) SEM images of Zn soaked in the baseline $ZnSO_4$ and 0.1-LBG solutions for 1 day.

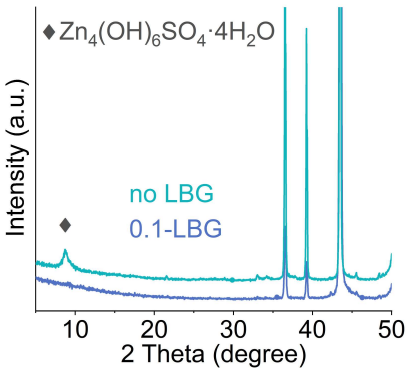

Fig. S7 XRD patterns of Zn deposition on Cu substrate for 10 mAh cm^{-2} in the baseline $ZnSO_4$ and 0.1-LBG electrolytes.

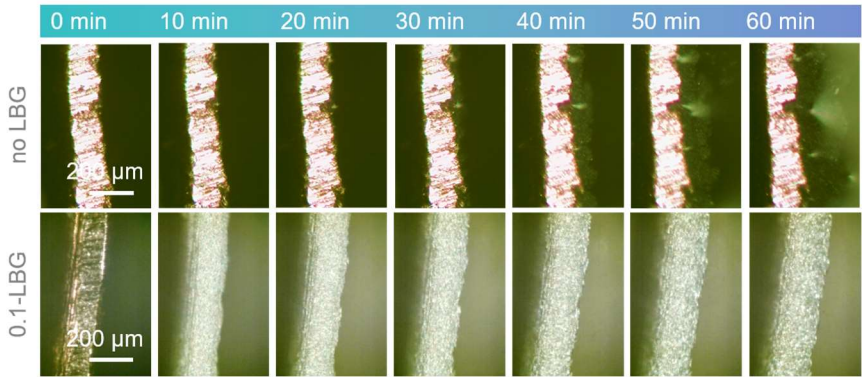

Fig. S8 Side views of in-situ optical microscopy images of Zn deposition at 10 mA cm^{-2} in the two electrolytes.

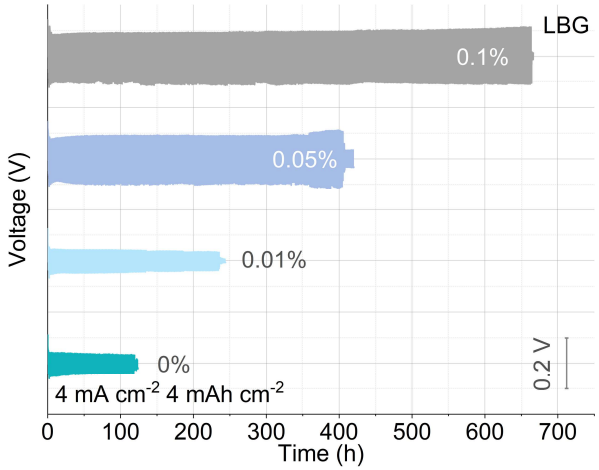

Fig. S9 Long-term cycling of Zn symmetric cells with the $ZnSO_4$ electrolytes containing different percentages of LBG.

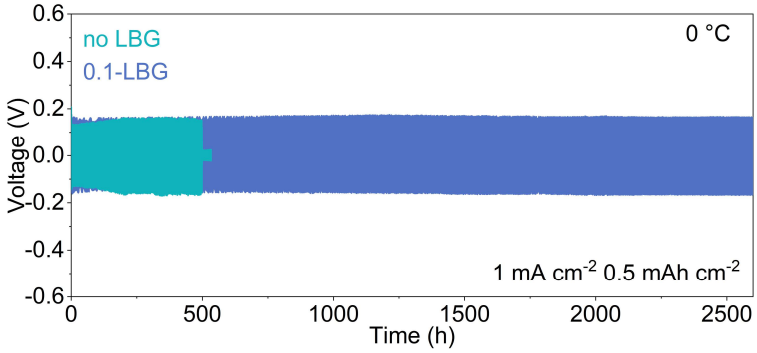

Fig. S10 Cycling performance of Zn symmetric cells at 0 °C with the $ZnSO_4$ and 0.1-LBG electrolytes.

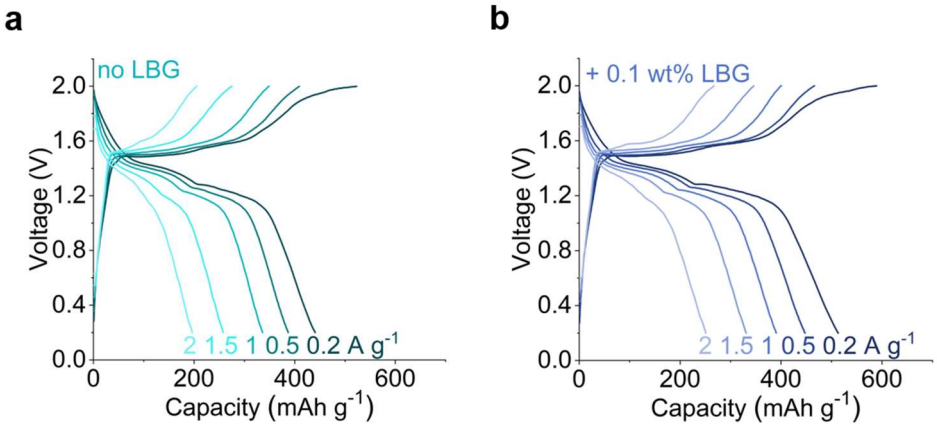

Fig. S11 Charge/discharge curves of Zn//MnO_2 cells at different current densities in a) the baseline electrolyte and b) after 0.1 wt% LBG addition.

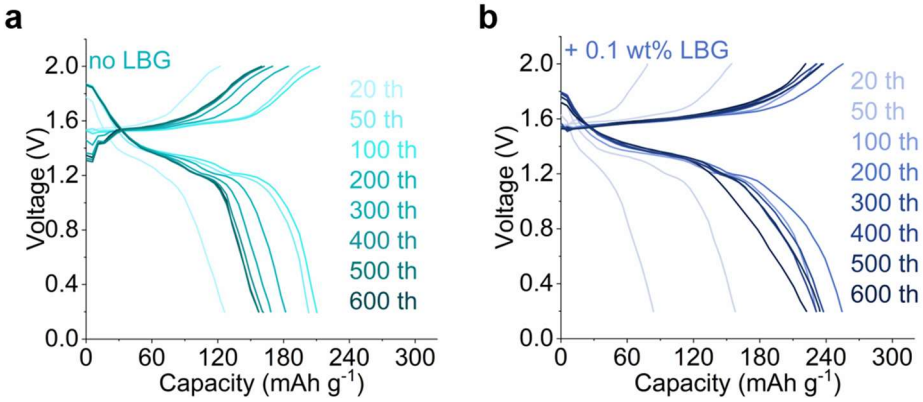

Fig. S12 Charge/discharge curves of Zn//MnO_2 cells at different cycles in a) the baseline electrolyte and b) after 0.1 wt% LBG addition.

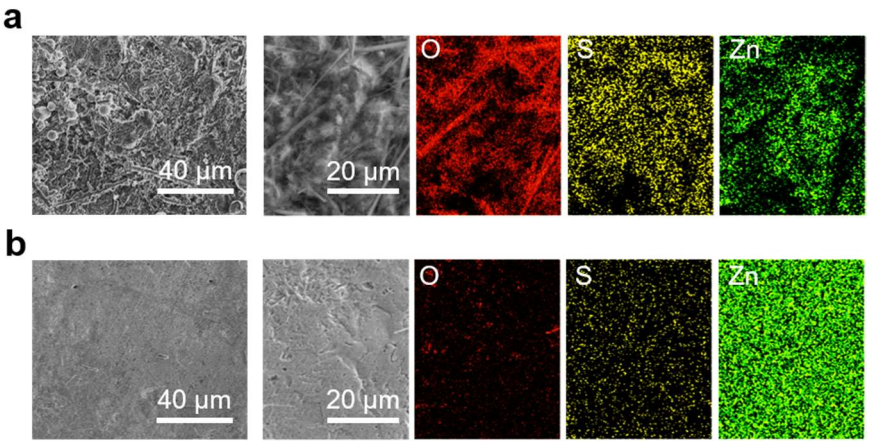

Fig. S13 SEM images and EDS mappings of Zn anode after 200 cycles at 2 A g^{-1} from full cells with a) the baseline electrolyte and b) after 0.1 wt% LBG addition.

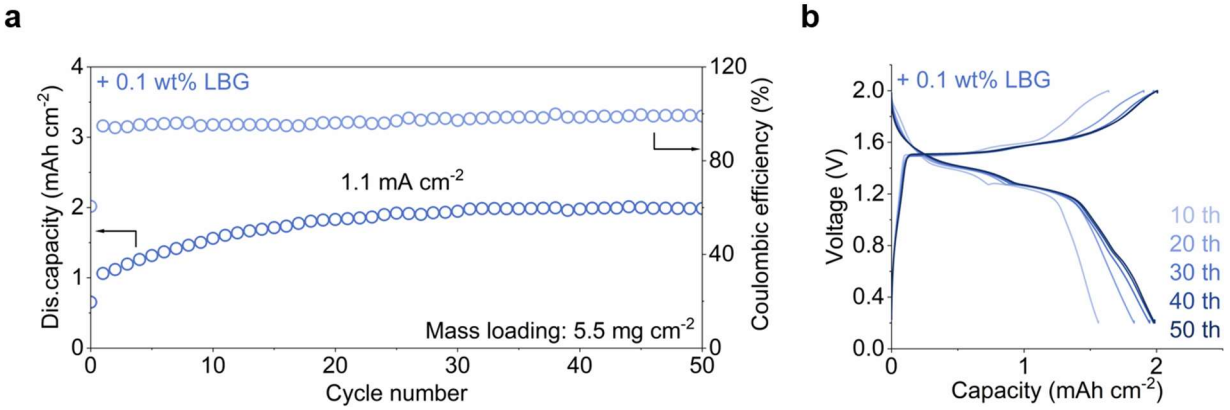

Fig. S14 a) The capacity evolution and b) charge/discharge curves at different cycles of the Zn//MnO_2 pouch cell in the LBG containing electrolyte.

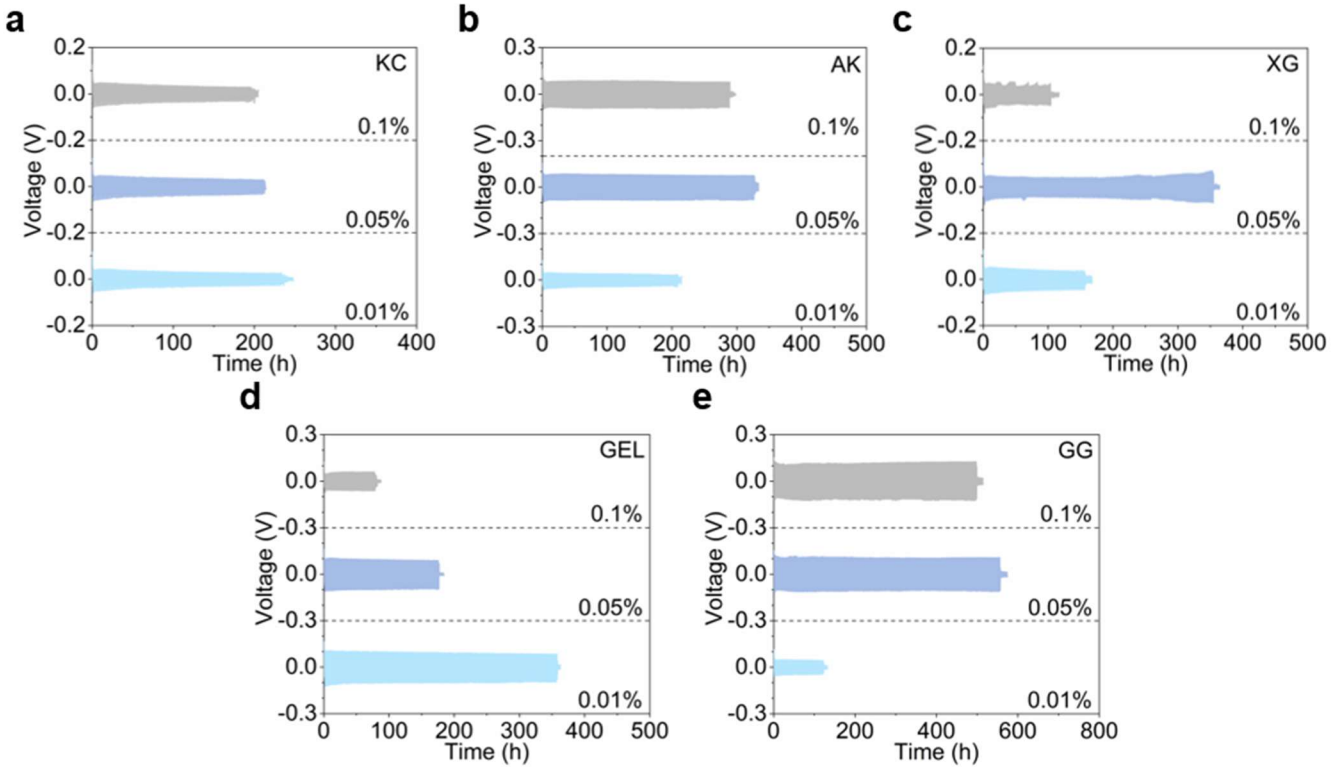

Fig. S15 The cycling performance of Zn symmetric cells with different gum additives at 4 mA cm^{-2} and 4 mAh cm^{-2}.

Table S1 The desolvation energy barrier at each step of $Zn(H_2O)_6^{2+}$ and the three $Zn(H_2O)_5LBG^{2+}$ solvation structures

Desolvation step	$Zn(H_2O)_6^{2+}$	Solvation A	Solvation B	Solvation C
I	1.0 eV	0.9 eV	0.7 eV	0.9 eV
II	1.3 eV	0.7 eV	0.7 eV	0.4 eV
III	2.1 eV	0.9 eV	1.2 eV	1.0 eV
IV	2.7 eV	0.9 eV	1.0 eV	1.2 eV
V	4.0 eV	0.8 eV	0.6 eV	1.1 eV
VI	4.6 eV	14.1 eV	14.9 eV	14.6 eV

Table S2 The H-bond lengths among solvated LBG and water during the desolvation paths of the three $Zn(H_2O)_5LBG^{2+}$ solvation structures, which are all shorter than 1.92 Å for typical H_2O-H_2O.

Desolvation step	Solvation A	Solvation B	Solvation C
$Zn(H_2O)_5LBG^{2+}$	1.64 Å	1.86/1.66/1.60 Å	1.61/1.66/1.63 Å
$Zn(H_2O)_4LBG^{2+}$	1.66 Å	1.75/1.56/1.62 Å	1.67/1.58 Å
$Zn(H_2O)_3LBG^{2+}$	1.62 Å	1.71/1.64/1.52 Å	1.63/1.62 Å
$Zn(H_2O)_2LBG^{2+}$	1.64 Å	1.73/1.92/1.64 Å	1.66/1.60 Å
$Zn(H_2O)LBG^{2+}$	1.64 Å	1.67 Å	1.60 Å

Table S3 Comparison of additive costs and concentrations in the electrolytes with previous studies.

Additive category	Additive	Additive concentration (g L^{-1})	Additive price (USD $L_{electrolyte}^{-1}$)
Organic powder	Na_4EDTA	29.1	0.73
	Arginine	17.4	0.94
	PEO	1.2	1.86
	TMAC	75.8	9.1
	Dopamine	9.5	11.5
	Succinonitrile	985	177.3
	TEHC	206.5	262.2
	Maltose	420	283.9
Organic solvent	Methanol	396	3.96
	NMP	50	8.5
	DMF	45	10.8
	TEP	535	925.6
	DMSO	203	5.48
Salt	KPF_6	8.3	0.28
	LiCl	9.2	0.52
	$Zn(H_2PO_4)_2$	84.8	1.02
	$Ce_2(SO_4)_3$	7.4	1.41
	$NaClO_4$	28.4	6.8
	EMImCl	2528.3	480.4
Gum (This work)	LBG	1	0.064
	GG	0.5	0.087
	AK	0.5	0.043
	XG	0.5	0.021
	KC	0.1	0.009
	GEL	0.1	0.006