

Supporting information

A High Areal Capacity Hybrid Magnesium–Lithium-Ion Battery with 99.9% Coulombic Efficiency for Large-scale Energy Storage

Hyun Deog Yoo,[†] Yanliang Liang,[†] Yifei Li,[†] and Yan Yao^{†,‡,*}

[†] *Department of Electrical and Computer Engineering and Materials Science and Engineering Program, University of Houston, Houston, Texas 77204, United States.*

[‡] *Texas Center for Superconductivity, University of Houston, Houston, Texas 77204, United States.*

*Corresponding Author: yyao4@uh.edu

Table S1. Comparison of this work with previous reported literatures in MLIBs

Electrolyte				Cathode			Ref.
Mg salt (Li salt / solvent)	Mg anode CE ^a	$c(\text{Li}^+)$ [M]	Voltage window [V _{Mg}]	Li-ion cathode / Average voltage	Specific capacity / Cycle achieved (C-rate, retention %)	Areal capacity / Rate achieved (retention %)	
DCC ^b (LiCl / THF)	100%	0.5	2.0	Mo ₆ S ₈ 1.3 V	113 mAh g ⁻¹ -	-	23
APC (LiCl / THF)	100%	0.5	2.9	LiFe _{0.2} Mn _{0.8} PO ₄ 2.6 V	-	-	28
APC (LiCl / THF)	100%	0.25	2.9	V ₂ O ₅ 2.35 V	120 mAh g ⁻¹ -	-	13
APC (LiBF ₄ / THF)	60%	0.2	2.5	(Li)FePO ₄ 2.5 V	120 mAh g ⁻¹ -	-	24
Mg(BH ₄) ₂ (LiBH ₄ / DG) ^c	100%	1.0 – 2.0	1.6	Mo ₆ S ₈ 1.3 V	100 mAh g ⁻¹ 300 cyc. (0.1C, 90%)	-	26
APC (LiCl / THF)	100%	0.5	2.9	Activated carbon 2.5–0.5 V	40 mAh g ⁻¹ 4500 cyc. (6C, 79%)	0.6 mAh cm ⁻² 25C (50%)	32
APC (LiCl / THF)	100%	1.0	2.9	Mo ₆ S ₈ 1.3 V	126 mAh g ⁻¹ 3000 cyc. (10C, 95%)	0.3 mAh cm ⁻² 30C (88%)	25, 27
APOC ^d (LiPF ₆ / THF)	98%	0.5	5.0	LiMn ₂ O ₄ 1.8 V	100 mAh g ⁻¹ -	-	29
APC (LiCl / THF)	100%	0.5	2.9	TiS ₂ 1.4 V	170 mAh g ⁻¹ 400 cyc. (0.3C, 99%)	0.2 mAh cm ⁻² 2C (40%)	31
Mg(BH ₄) ₂ (LiBH ₄ / TG) ^e	70%	1.0 – 2.0	2.0	TiO ₂ 0.9 V	150 mAh g ⁻¹ 90 cyc. (0.2C, 93%)	0.1 mAh cm ⁻² 2C (57%)	30
APC (LiCl / THF)	100%	1.0	2.9	TiS ₂ 1.4 V	220 mAh g ⁻¹ 2000 cyc. (1C, 95%)	~ 2.0 mAh cm ⁻² 10C (57%)	This work

^a CE: Coulombic efficiency. ^b DCC: dichloro-complex. ^c DG: diglyme. ^d APOC: magnesium tetraphenol-aluminate. ^e TG: tetraglyme.

Table S2. Electrochemical properties measured in a three-electrode cell in Figure 2a. Pt wire was used as the working electrode and 0.25 M APC solution with 0.0, 0.5, and 1.0 M of LiCl as the electrolyte

c (LiCl) [M]	Coulombic efficiency of Mg deposition/dissolution [%]	Anodic potential limit [V]	Mg deposition overpotential [mV]
0.0	100.0	2.6	-155
0.5	99.6	2.6	-130
1.0	99.3	2.8	-206

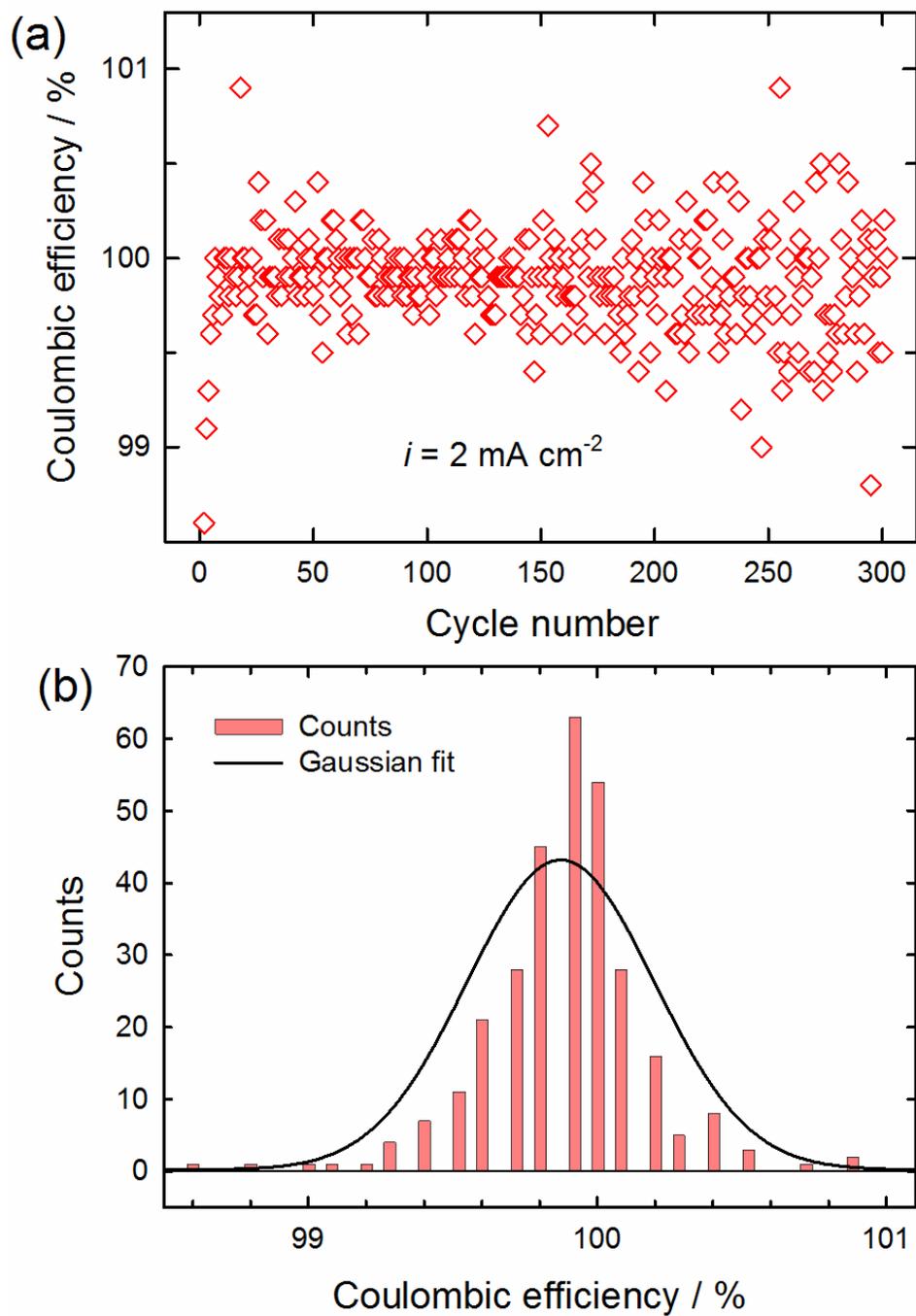


Figure S1. (a) Enlarged view of coulombic efficiency of the hybrid MLIB with capacity of 1.9 mAh cm⁻² at rate of 2 mA cm⁻². (b) Gaussian fitting of the distribution of Coulombic efficiency (black line) shows the average coulombic efficiency of $99.9 \pm 0.3\%$.



Figure S2. The image of a glass fiber separator in the Li cell after exposure to air for several minutes. The previous metallic dark brown part became whitish, indicating the formation of lithium hydroxide (LiOH).

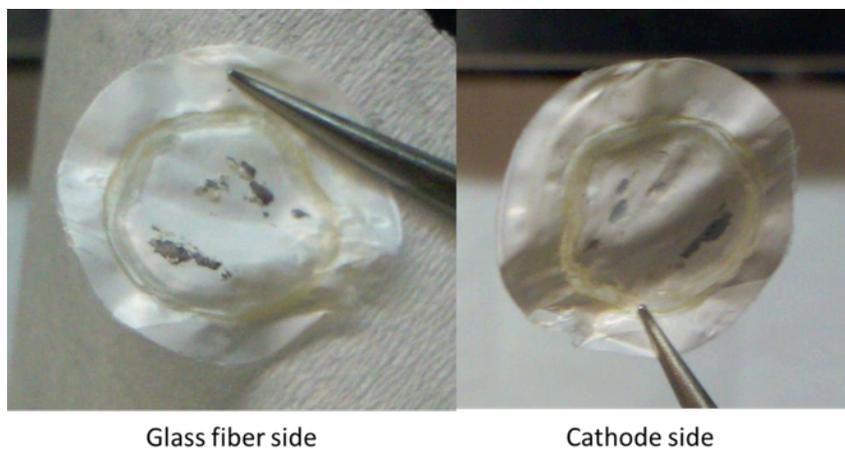


Figure S3. Images of the PP/PE/PP separator in the Li ion cell after 100 cycles at 2 mA/cm².