ADVANCED MATERIALS

Supporting Information

for *Adv. Mater.*, DOI: 10.1002/adma.201804815

An Electron/Ion Dual-Conductive Alloy Framework for High-Rate and High-Capacity Solid-State Lithium-Metal Batteries

*Chunpeng Yang, Hua Xie, Weiwei Ping, Kun Fu, Boyang Liu, Jiancun Rao, Jiaqi Dai, Chengwei Wang, Glenn Pastel, and Liangbing Hu**

Supporting Information

An Electron/Ion Dual-Conductive Alloy Framework for High-Rate and High-Capacity Solid-State Lithium-Metal Batteries

*Chunpeng Yang,[1‡] Hua Xie,[1‡] Weiwei Ping,[1] Kun Fu,[1] Boyang Liu,[1] Jiancun Rao,[2] Jiaqi Dai,[1] Chengwei Wang,[1] Glenn Pastel,[1] and Liangbing Hu[1]**

Dr. C. Yang, H. Xie, W. Ping, Dr. K. Fu, B. Liu, Dr. J. Dai, Dr. C. Wang, G. Pastel, Dr. L. Hu
[1]Department of Materials Science and Engineering, University of Maryland at College Park, College Park, Maryland 20742, USA
* E-mail: binghu@umd.edu

Dr. J. Rao
[2]Advanced Imaging and Microscopy (AIM) Lab of Nano Center, University of Maryland at College Park, College Park, Maryland 20742, USA.

[‡] These authors contributed equally to this work.

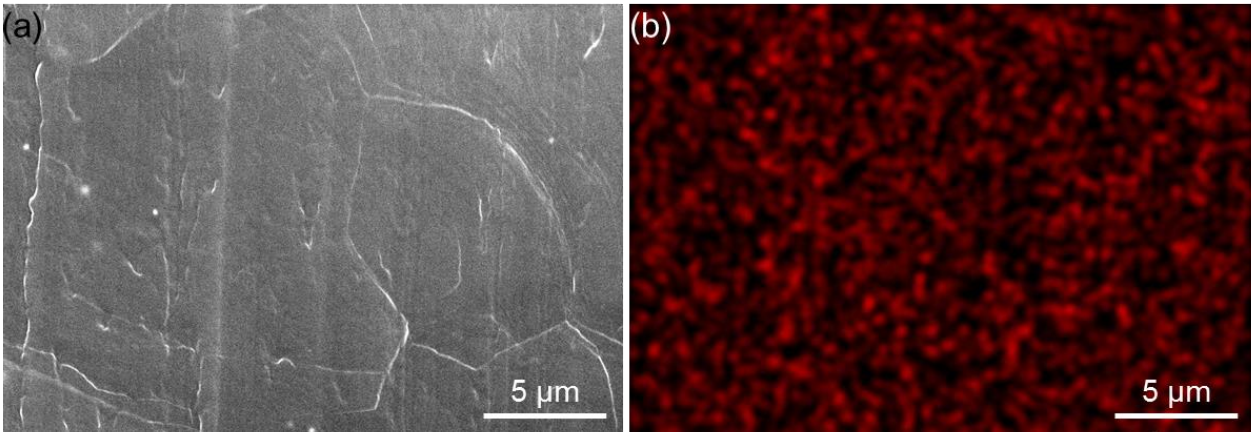

Figure S1. (a) SEM image and (b) corresponding EDX Mg elemental mapping of the Li-Mg alloy. Mg is homogeneously dissolved in the solid solution without agglomeration.

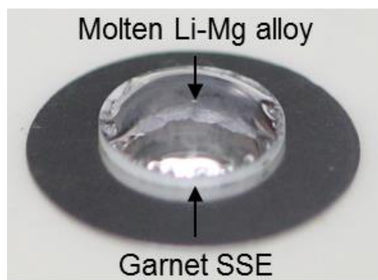

Figure S2. Digital photo of molten Li-Mg alloy on the garnet SSE. The molten Li-Mg alloy shows a small contact angle on the garnet SSE.

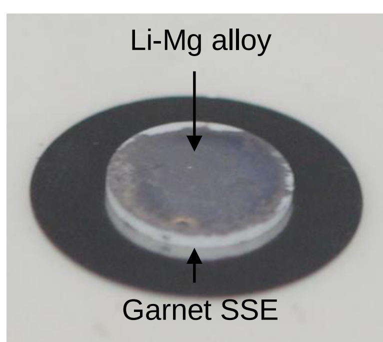

Figure S3. Digital photo of the Li-Mg alloy on the garnet SSE after melting and solidifying.

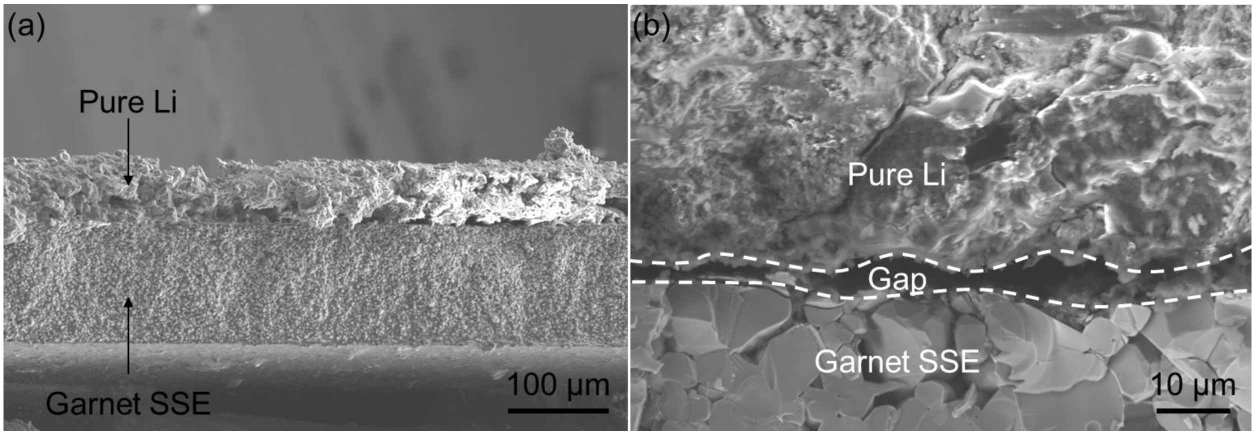

Figure S4. Cross-sectional SEM images of pure Li melted on the garnet SSE at different magnifications. Significant gaps are observed at the Li-garnet interface indicate poor interface contact of pure Li melted on the garnet SSE.

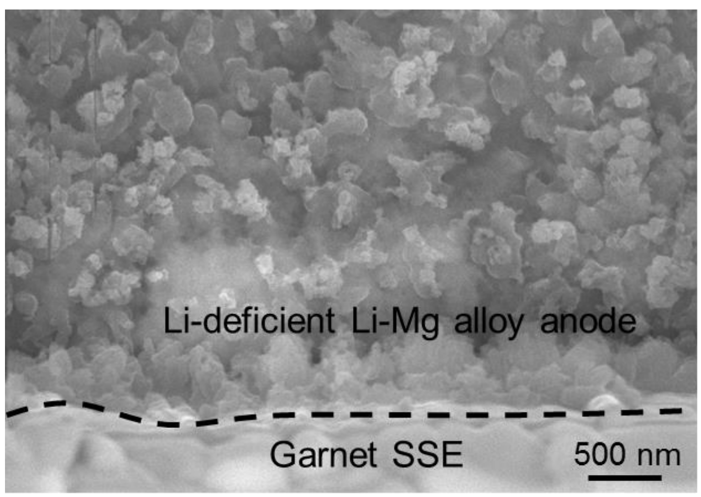

Figure S5. Cross-sectional SEM image of the Li-Mg alloy on the garnet SSE after stripping 2 mA h cm^{-2} of Li. The Li-deficient Li-Mg alloy becomes a porous framework after dealloying, but maintains good interface contact with the garnet.

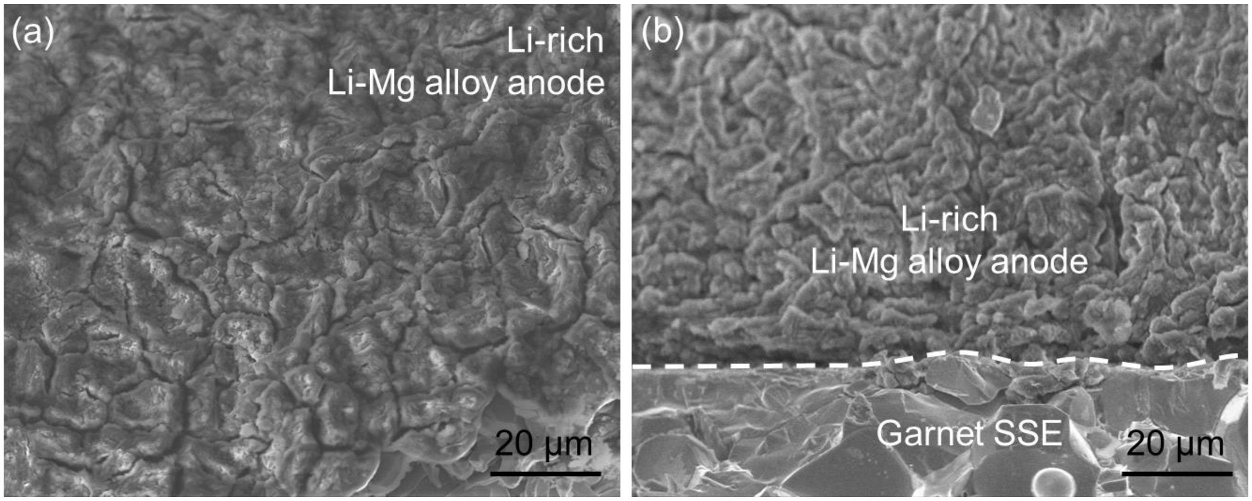

Figure S6. (a) Top view and (b) cross-sectional SEM images of the Li-Mg alloy on the garnet SSE after stripping and plating 2 mA h cm^{-2} of Li. The Li-Mg alloy after a Li stripping/plating cycle became a relatively dense Li-rich alloy, without Li dendrite formation or Li accumulation on the surface or at the alloy-garnet interface.

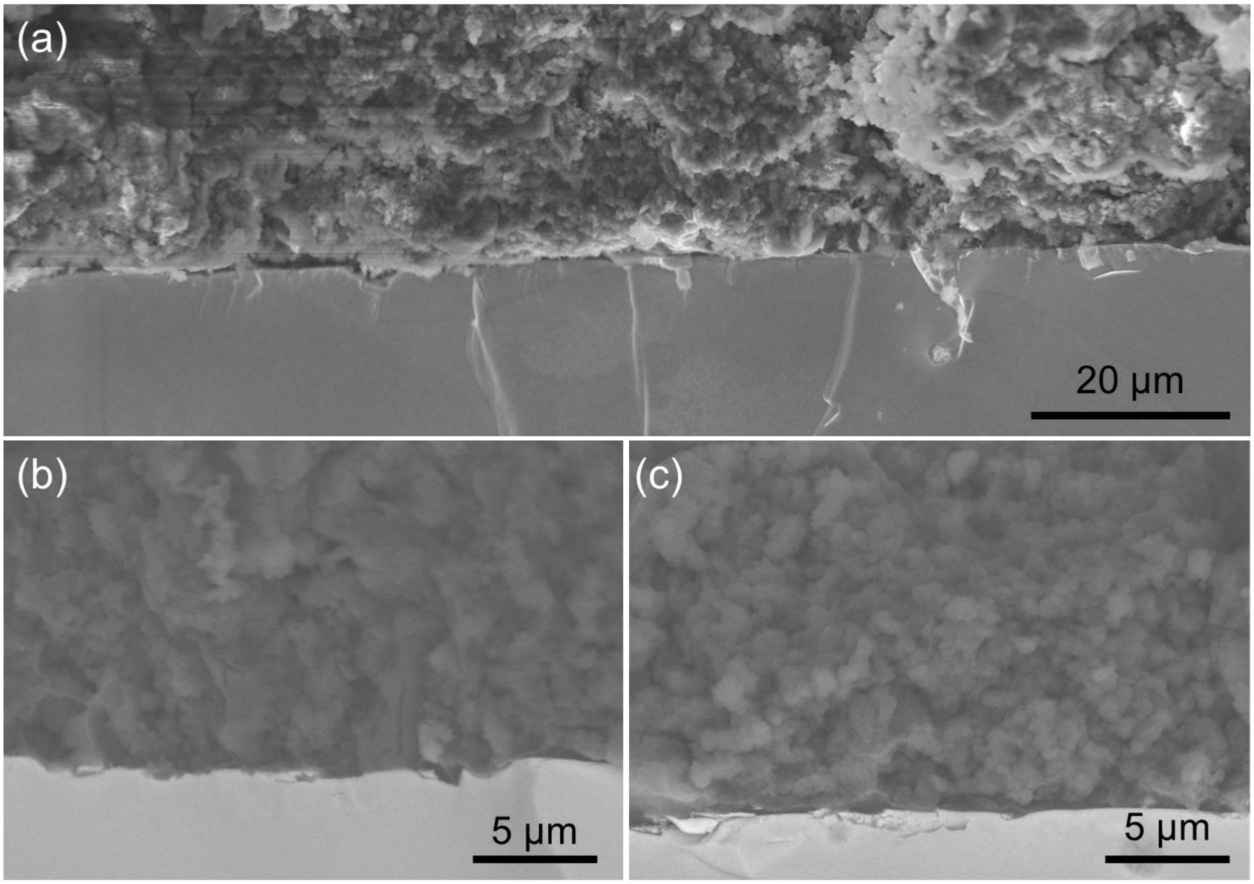

Figure S7. Cross-sectional SEM images of the Li-Mg alloy on the garnet SSE after 500 stripping/plating cycles: (a) low-magnification secondary electron image, (b) and (c) high-magnification backscattered electron images. Li-Mg alloy after the cycling remains relatively intact with good interface contact with the garnet SSE, without Li dendrite penetration.

Table S1. Cycling performance and conditions in this work and representative publications in Figure 5.

Point	Reference	Electrolyte	Testing temperature	Current density (mA cm^{-2})	Cumulative capacity plated (mA h cm^{-2})
a	*Nat. Mater*. **2017**, *16*, 572-579	Garnet	r.t.	0.1 0.2	0.35 9
b	*ACS Appl. Mater. Interfaces* **2016**, *8*, 10617	Garnet	r.t. 80 °C	0.08 0.25	6 18.75
c	*Nano Lett*. **2017**, *17*, 565	Garnet	r.t.	0.1	2.4
d	*Chem. Mater*. **2017**, *29*, 7961-7968	Garnet	r.t.	0.2	20.8
e	*Angew. Chem. Int. Ed*. **2017**, *56*, 14942	Garnet	r.t.	0.1	1.9
f	*Adv. Energy Mater*. **2018**, *8*, 1701963	Garnet	r.t.	0.05	2.5
g	*J. Am. Chem. Soc*. **2018**, *140*, 6448	Garnet	65 °C	0.1	22.5
h	*Mater. Today* **2018**, *21*, 594	Garnet–PEO	60 °C	0.05 0.1 0.2	5.75 13.75 30.75
i	*Proc. Natl. Acad. Sci*. **2016**, *113*, 13313	$LiZr_2(PO_4)_3$	80 °C	0.05 0.1 0.2 0.1	3 7 12 14.5
j	*J. Am. Chem. Soc*. **2015**, *137*, 1384	$Li_7P_2S_8I$	r.t.	0.2	160
k	*Adv. Energy Mater*. **2018**, *8*, 1703644	Li_2S-P_2S_5	25 °C 60 °C 100 °C	0.3 0.6 1.5	30 60 150
This work		Garnet	r.t.	1 2	250 750