

Supporting Information for
Solid-Solution-Based Li-Mg Alloy for Highly Stable Lithium Metal
Anode

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Experimental section

1. Material synthesis

The main focus of the experiments was to make Li - Mg alloys. Commercial Lithium electrode (15.8 mm * 0.45 mm) and high purity Magnesium powder (74 μ m ~ 150 μ m, SCR) were prepared in a glovebox (H₂O < 0.01 ppm, O₂ < 0.01 ppm, Mikrona) with Argon-filled atmosphere ahead. Lithium electrodes was first laminated into thin pieces at 20 M Pa respectively by a hand-operate hydraulic press (MTI), in order to get much surface area as possible to contain Magnesium powder. Then using an analytical balance (Sartorius) to measure Lithium pieces and Mg powder to mix at a weight ratio of 19:1 (Mg 5 at%), 9:1 (Mg 10 at%) and 4:1 (Mg 20 at%). Hand-operating hydraulic

press were used twice at 20 °C until the mixture reached the uniform thickness of 0.45 mm.

2. Preparation of high areal mass loading LCO cathode

The aluminum foil was cut into round pieces (1.76 cm²) to prepare high areal mass loading LCO cathode. Lithium cobalt (III) oxide powder (99.8%, Macklin), PVDF and carbon black were added in a molar with the weight ratio of 482: 9: 9 (96.4% LCO) and NMP was used as a dispersant. The mixture was grinded for 1 hour and applied to the aluminum foil by a pipette gun. Then the cathodes were dried in the oven for 12 hours, weighed and their surface densities were calculated.

3. Electrochemical Studies

CR2032 coin-type cells (Canrd) were assembled in the glovebox to measure electrochemical performance and commercial Li sheet as the counter electrode. 1.0 M LiTFSI in DME: DOL = 1:1 (vol%) with 1.0 wt.% LiNO₃ additive (Canrd) was used as electrolyte in symmetric cells. All cells were assembled in a glove box full of high-purity argon with less than 0.01 ppm of O₂ and H₂O. The electrodes were separated by one layer of a polypropylene separator (Celgard 2500). The Galvanostatic charge-discharge test of symmetric cells were performed based on the ratio of 0.9 mA h cm⁻² Li stripping and plating at the current of 0.45, 0.9 and 1.8 mA cm⁻² using a Neware-MIHW-200-160CH test system.

The resembled Li and Li-Mg alloy symmetric cells were tested under reciprocating

galvanostatic discharge/charge cycles using a Neware MHWX-200 test system. Commercial LiCoO₂ cathode (LCO, areal capacity 178 mA h g⁻¹, Canrd) sheets with the active material loading mass of 10.7 mg cm⁻² were purchased and assembled full cells with Li-Mg alloy electrodes. The LCO full cells were cycled from 2.8 V to 4.2 V in this work. Moreover, the LCO cathode sheets were assembled cells with commercial Li electrodes directly. 1.0 M LiPF₆ in EC: EMC = 3:7 (vol%) with 10 wt.% FEC (Canrd) was used as electrolyte in full cells. Without specified illustration, the amount of electrolyte in this work is 100 μL.

The electrochemical impedance spectrum (EIS) was recorded using a CHI660E multi-channel cell test system in a frequency range of 0.01 Hz to 1 MHz through 10 mV AC voltage measurement. Linear scan voltammograms (LSV) and cyclic voltammograms (CV) were also tested by it.

4. Material characterization

In order to investigate the role of Li-Mg alloys in suppressing the formation of Li dendrites, X-ray diffraction (XRD) experiment were performed on a Bruker D8-Advanced diffractometer at a scan rate of 2° s⁻¹ using a Cu Kalpha radiation source ($\lambda = 1.54 \text{ \AA}$, operated at 40 kV and 40 mA). Scanning electron microscopy (SEM) images were collected through a ZEISS GeminiSEM equipped with an energy dispersive X-ray spectrometer (EDS) at an accelerate voltage of 200 kV.

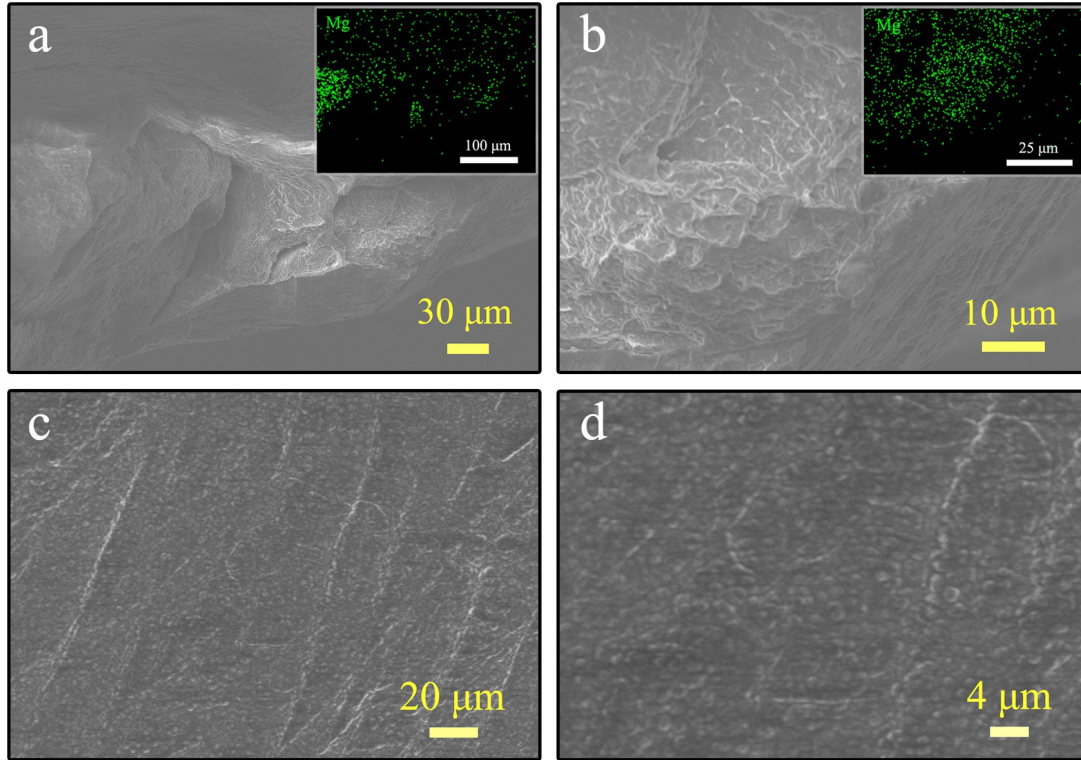


Figure S1. (a), (b) Cross section SEM images with EDS patterns of uncycled Li-Mg alloy (10 at% Mg). (c), (d) Top view SEM images of Commercial Lithium electrode.

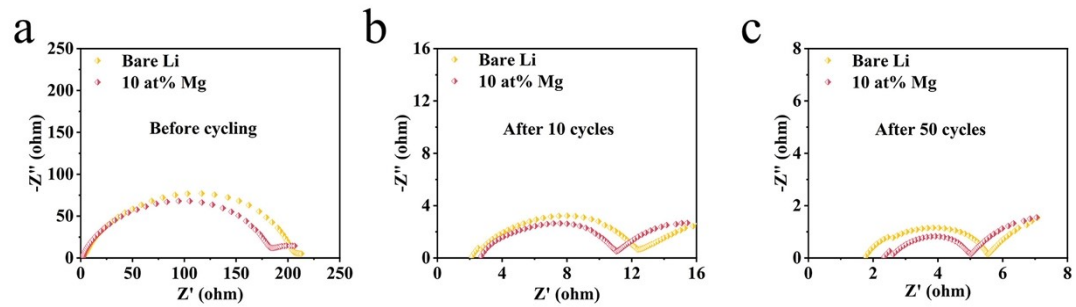


Figure S2. (a), (b), (c) EIS of Li//Li and Li-Mg alloy//Li-Mg alloy symmetric cells cycled different times.

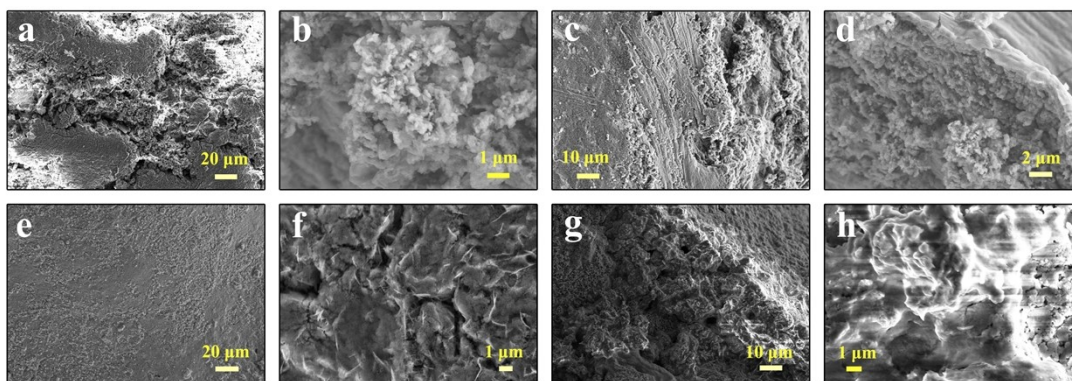


Figure S3. (a)(b)(c)(d) SEM micrographs of Li symmetric cells after reciprocating galvanostatic charge-discharge test for 100 cycles. (e)(f)(g)(h) SEM micrographs 10 at % Mg Li-Mg alloy symmetric cells after reciprocating galvanostatic charge-discharge test for 100 cycles at 0.9 mA h cm^{-2} .

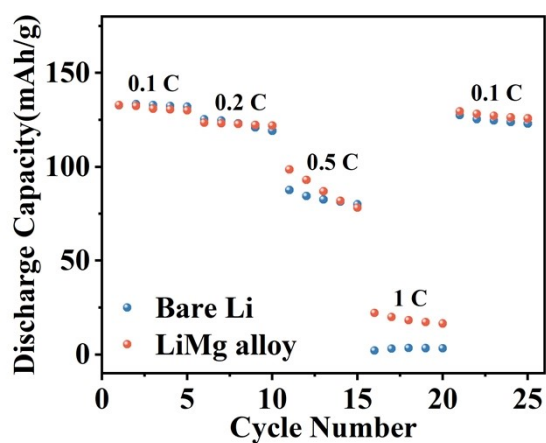


Figure S4. Rate performance of Li-Mg alloy//LCO ($4.30 \text{ mA h cm}^{-2}$) and bare Li//LCO full cells at various rates from 0.1 C to 1 C.