Supporting Information

Superlithiophilic Graphene-Silver Enabling Ultra-stable Host for Lithium Metal Anode

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Li is the active material in the GAL composite electrode, and the relative content of Li determines the specific capacity of the GAL composite electrode. In pursuit of higher capacity, the stability of the GAL composite electrode must also be considered. In this work, the GO-Ag content in the composite electrode is about 20%, which is based on the free thermal infusion of GO-Ag and Li. In other words, when the GO-Ag content of 20%, the molten Li with free thermal infusion can be completely absorbed without a surplus. When GO-Ag content is lower, the pores formed by the limited graphene skeleton cannot accommodate too much Li, and at this time, there will be Li in a "hostless" structure, and will still undergo volume changes and form Li dendrites during repeated stripping/plating processes, which will be detrimental to the stability of GAL. The thermal infusion technology of Li is considered to be an ideal strategy for pre-storing Li because of the advantages of uniform Li deposition and no damage to the bulk structure.[1] In order to ensure the uniform dispersion of the GO-Ag composite material during the manufacture of the compound Li electrode, we mechanically stirred for 20 min after the Li was freely thermal infusion GO-Ag to obtain a viscous mixed solution and evenly coated with copper foil. Therefore, the components in the mixed GAL composite electrode can be uniformly mixed.

GO-Ag composite and polyvinylidene fluoride (PVDF), were mixed by the weight ratio of 8:2 in N-methyl-2-pyrrolidone (NMP) solution to make a paste, and coated on copper foil (0.6mg cm^{-2}). The test results are recorded in Figure S1. The coulomb efficiency of the GO-Ag composite is stable at about 98% after 10 cycles at a deposition capacity of 1mAh cm^{-2}, and a current of 1mA cm^{-2}. The high coulomb efficiency proves the excellent reversibility of Li-ion plating/stripping of the GO-Ag composite.

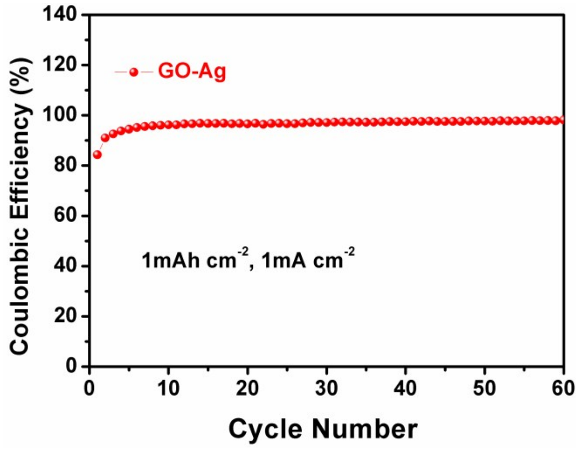

Figure S1. Coulombic efficiency at different cycles of GO-Ag composite, at a deposition capacity of 1mAh cm^{-2}, and a current of 1mA cm^{-2}.

$LiMn_2O_4$ (LMO) cathode material matching was used to evaluate the performance of GAL composite anode in full cells. The test results are shown in Figure S2. It is not difficult to find that the GAL composite anode has a higher specific capacity than the bare Li electrode. This is because the superlithiophilic silver nanoparticles of the GAL composite anode reduce the nucleation barrier of Li metal, and the continuous 3D conductive porous graphene pre-stores space for Li deposition, while balancing the electric field distribution and reducing the local current density. Thanks to these advantages, the polarization of the GAL composite anode is reduced, so LMO/GAL has a higher specific capacity.

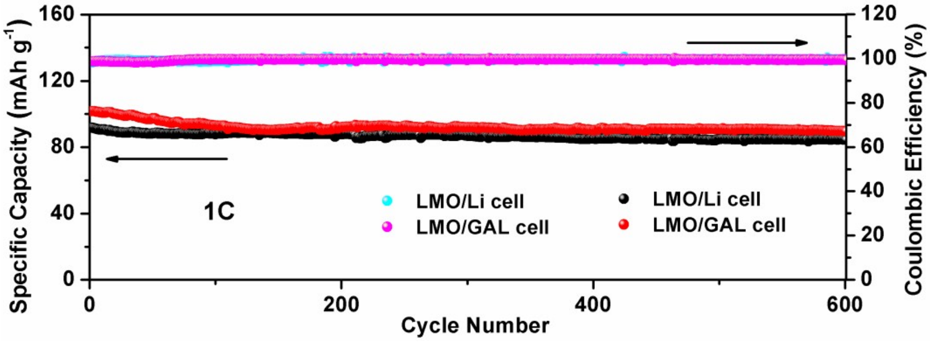

Figure S2. Discharge capacity and coulombic efficiency at different cycles of LMO/GAL and LMO/Li full cells.

Figure S3 shows the nyquist plots of LMO/GAL and LMO/Li full cells. The LMO/GAL and LMO/Li full cells exhibit a similarly shaped electrochemical impedance spectrogram (EIS), where a semicircle and a straight line were detected in the high and low-frequency regions, respectively. The semicircle of the LMO/GAL full cell is the smallest, which indicates has low electrochemical reaction resistance but excellent electron conduction and Li-ion migration rates.

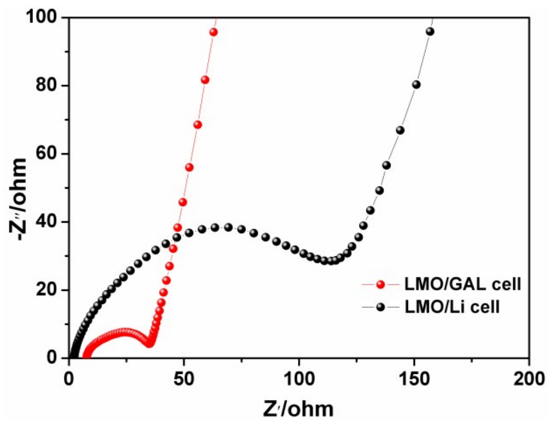

Figure S3. Nyquist plots of LMO/GAL and LMO/Li full cells.

Notes and references

S1. D. C. Lin, Y. Y. Liu, Z. Liang, H. W. Lee, J. Sun, H. T. Wang, K. Yan, J. Xie and Y. Cui, Layered reduced graphene oxide with nanoscale interlayer gaps as a stable host for lithium metal anodes, *Nat. Nanotechnol*., 2016, **11**, 626-632.