



Conductive porous carbon film as a lithium metal storage medium

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ABSTRACT

The Li metal anode boasts attractive electrochemical characteristics for use in rechargeable Li batteries, such as a high theoretical capacity and a low redox potential. However, poor cycle efficiency and safety problems relating to dendritic Li growth during cycling should be addressed. Here we propose a strategy to increase the coulombic efficiency of the Li metal electrode. Conductive porous carbon films (CPCFs) were prepared by distributing amorphous carbon nanoparticles within a polymer binder. This porous structure is able to provide enough conductive surfaces for Li deposition and dissolution, which reduce the effective current density. Moreover, the pores in these films enable the electrolyte to easily penetrate into the empty space, and Li can be densely deposited between the carbon particles. As a result, dendritic Li growth can be effectively prevented. Electrochemical tests demonstrate that the coulombic efficiency of the porous electrode can be greatly improved compared to that of the pure Cu electrode. By allowing for the development of robust Li metal electrodes, this approach provides key insight into the design of high-capacity anodes for Li metal batteries, such as Li-air and Li-S systems.

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1. Introduction

With the rapid development of portable electronic devices and electric vehicles, an increasing demand for rechargeable batteries with high energy densities has intensified concerns regarding the use of Li metal as an alternative electrode material [1]. Li metal exhibits beneficial electrochemical properties such as a low redox potential of -3.04 V (vs. standard hydrogen electrode) and a huge theoretical capacity of 3860 mA h g^{-1} ; it therefore is used as the anode in high-energy Li rechargeable batteries, such as the Li-air and Li-S varieties [2]. However, it is still difficult to commercialize Li-metal-based rechargeable batteries because of their poor cycle performance and their associated safety problems. In order to solve these problems, the dendritic growth of Li metal – which leads to the degradation of the coulombic efficiency as well as safety issues resulting from internal short circuit – should be addressed. Therefore, various studies have been performed on electrolyte [3–7], additive [8–16], optimal plating conditions [17,18] and theoretical models [19–21]. However, the problems mentioned above have not been satisfactorily solved, and issues relating to the

efficiency, cycling stability, Li ionic conductivity, and the chemical reactivity still remain [7,22].

When Li-metal-based batteries are cycled, Li deposition and dissolution repeatedly occur, with the morphology of the Li metal electrodes depending on the applied current density [22,23]. When high current density is applied, some current is able to converge on a local area, which results in irregular surface morphologies, such as dendrites and moss [24,25]. The Li dendrites can penetrate separators and lead to short circuits in the cell, which constitutes a safety concern. In addition, the repeated deposition and dissolution of Li metal with high surface area and specific volume induces large strains and cracks in previously formed solid electrolyte interphase (SEI) films [23]. At this stage, the freshly exposed Li reacts with the electrolyte and the SEI can be formed again. One consequence of this consumption of fresh Li is a lowered coulombic efficiency.

To address the issues mentioned above, ideal Li deposition substrates that are able to alleviate the fluctuation of Li ion flux on the electrode/electrolyte interface should be considered. If the current is uniformly distributed across the surface of the substrate, morphological regularity can be achieved. Furthermore, free space in the substrate is required to accommodate the volumetric Li growth. Some recent studies have shown that Li metal storage electrodes can maintain their capacities by means of storing Li in the empty space of the substrate. The substrate materials and structures employed for this purpose are TiO_2 nanotubes [26],

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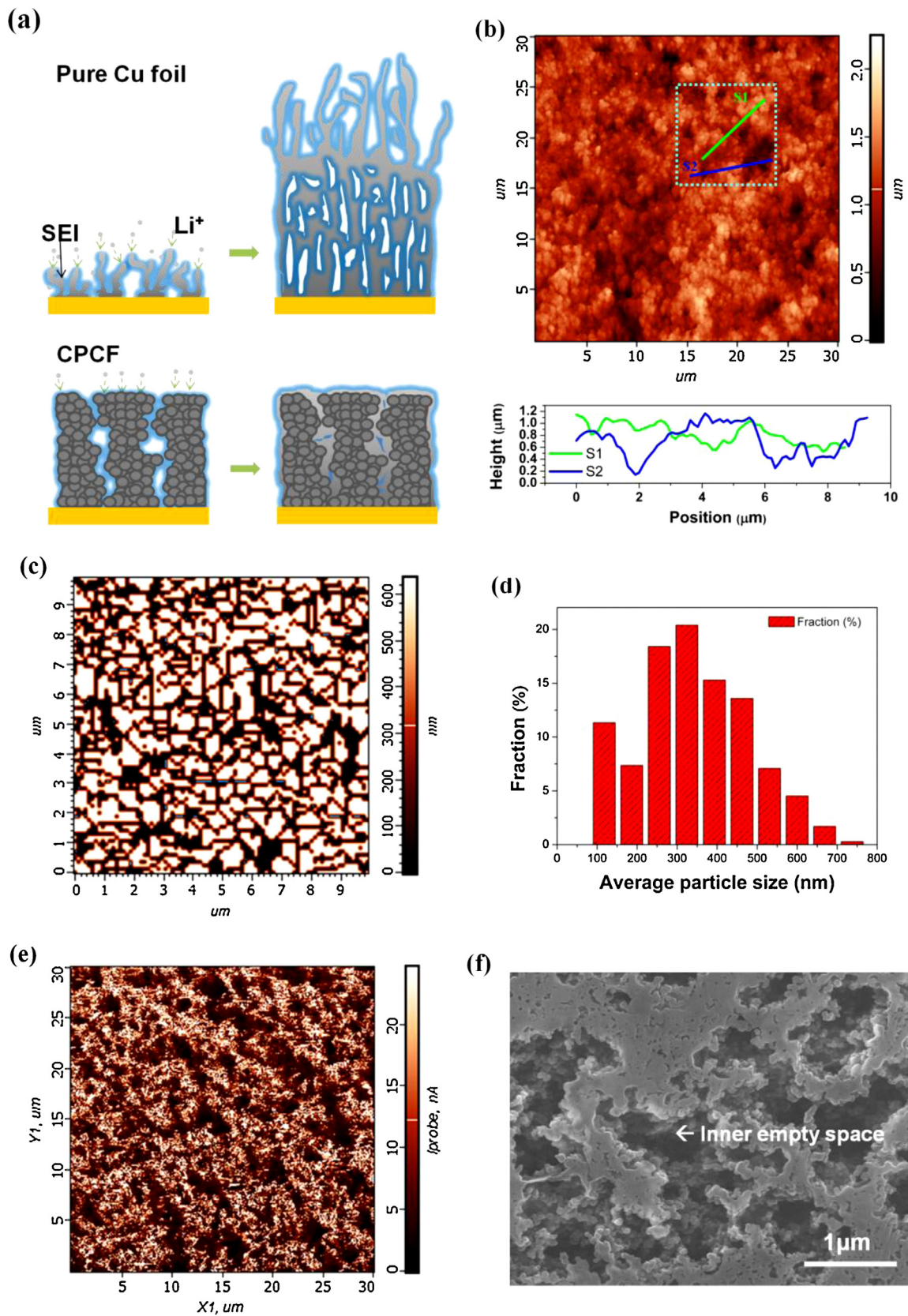


Fig. 1. (a) Schematic of metallic Li storage concept in CPCF electrode, (b) AFM image of the film surface with line profiles, (c, d) analysis for size distribution of carbon particles, (e) electrical conductivity mapping image by scanning spreading resistance microscopy based on AFM image, and (f) cross sectional FE-SEM image of CPCF.

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