# TOWARDS HIGH-ENERGY-DENSITY LITHIUM BATTERIES: KEY MATERIALS AND INTERPHASE TECHNOLOGY

Luoyi Ding[1], Zheng Liang*[1]

*1. Frontiers Science Center for Transformative Molecules, School of Chemistry and Chemical Engineering, Shanghai Jiao Tong University, Shanghai, 200240, People's Republic of China*

** E-mail: liangzheng06@sjtu.edu.cn*

## ABSTRACT

Porous materials have been widely used to solve the problems related to lithium (Li) dendrites due to their unique advantages. However, the development of functional integration and self-supporting separators through elaborate molecular-level structural design has not been widely reported. Here, a polyimide aerogel (PIA) with a definite nitrogen (N) functional site was used as the functional separator of lithium metal batteries (LMBs) to promote the uniform nucleation of Li and inhibit the growth of Li dendrites.

## KEY WORDS

Several key words are shown here: polyimine aerogel Li metal battery separator engineering electron cloud density

## 1. INTRODUCTION

Lithium metal batteries (LMBs) have attracted much attention for their high performance theoretical specific capacity (3860 mAh $g^{-1}$) and lowest negative electrochemical reduction potential (-3.04 V vs SHE). Unfortunately, the formation of Li dendrites during the process can affect the performance of the LMBs due to the repeated Li plating/stripping process due to anode/electrolyte interface instability. Uncontrolled dendrite growth leads to limited coulombic efficiency (CE), unsatisfactory cycle stability, and safety hazards caused by dendrites.

Research shows that the formation of Li dendrites leads to the greatly shortened cycle life of LMBs, and the CE is significantly reduced, which limits its practical application. In recent years, porous materials have been widely used to solve the problems related to Li dendrites, especially in the field of separator engineering, because of their unique advantages such as high porosity, adjustable pore structure and easy functionalization. However, the development of functional integration and self-supporting separators through elaborate molecular-level structural design has not been widely reported.

In this article, a polyimide aerogel (PIA) with a definite nitrogen (N) functional site was used as the functional separator of LMBs to promote the uniform nucleation of Li and inhibit the growth of Li dendrites. Specifically, the imine (N1) and protonated tertiary amine (N2) sites in PIA molecular structure differ significantly in electron cloud density (ECD) distribution. The N1 site with higher ECD and the N2 site with lower ECD attract and repel $Li^+$ through electrostatic interaction, respectively. This synergistic effect of the PIA separator accelerates the diffusion of $Li^+$ at the interface over the lithium anode electrode to obtain a uniform two-dimensional Li nucleation behavior. Therefore, the N1-N2 synergistic effect of PIA separator can effectively accelerate the diffusion of $Li^+$ in the metal interface of PIA/Li, thus inhibiting the formation of local current density and promoting uniform nucleation and uniform deposition of two-dimensional lithium. In addition, the abundant multistage pore structure of PIA separator is conducive to obtaining high ionic conductivity and uniform $Li^+$ flux. In conventional carbonate electrolyte, the CE of the PIA based cell was more than twice that of the cell using PP separator, and no significant Li dendrite formation was observed. At the same time, PIA membranes are also assembled into LMBs containing $LiFePO_4$ or $LiNi_{0.8}Co_{0.1}Mn_{0.1}O_2$ at high voltage, demonstrating competitive electrochemical performance. These results indicate that the special chemical structure design and the clever combination of hierarchical pore structure can enable the separator to regulate the nucleation behavior of Li and facilitate high-performance LMBs.

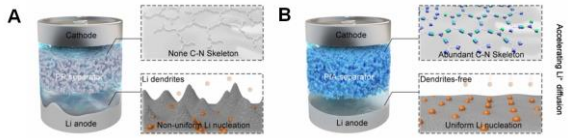


Fig.l: The interfacial $Li^+$ diffusion and Li nucleation on the Li metal in (A) PP and (B) PIA separator systems.

## REFERENCES

1 Luoyi Ding et al. *Proc. Natl. Acad. Sci. U.S.A.* 120 (2023) e2314264120.

2 Luoyi Ding et al. *Angew. Chem. Int. Ed.* (2024) e202411933.

3 Luoyi Ding et al. *Adv. Funct. Mater.* 33 (2023) 2304386.