



## Review

## Recent progresses in the suppression method based on the growth mechanism of lithium dendrite

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## ABSTRACT

Lithium secondary batteries (LSBs) with high energy densities need to be further developed for future applications in portable electronic devices, electric vehicles, hybrid electric vehicles and smart grids. Lithium metal is the most promising electrode for next-generation rechargeable batteries. However, the formation of lithium dendrite on the anode surface leads to serious safety concerns and low coulombic efficiency. Recently, researchers have made great efforts and significant progresses to solve these problems. Here we review the growth mechanism and suppression method of lithium dendrite for LSBs' anode protection. We also establish the relationship between the growth mechanism and suppression method. The research direction for building better LSBs is given by comparing the advantages and disadvantages of these methods based on the growth mechanism.

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## 1. Introduction

In the early 1960s, the concept of lithium second battery (LSB) was initialized [1,2]. LSBs have been widely used in portable electronic devices (PEDs) in the past decades [3,4]. Recently, it is still the object of intense research with the aim at further improving electrochemical performance for the requirements of electric vehicles (EVs), hybrid electric vehicles (HEVs) and smart grids (SGs) [5–10]. However, these performance requirements with high energy and high power may accelerate the growth of lithium dendrite on the anode surface; it leads to safety concern and low coulombic efficiency [11–13]. Therefore, the investigation of lithium dendrite has become the focus on energy storage of LSBs [14,15].

In the early 1990s, rechargeable lithium ion battery (LIB) with graphite anode was announced to significantly overcome the dendrite problem because graphite anode involves intercalation/deintercalation of lithium ion in graphite lattice during cycles [16]. Regrettably, the energy density of graphite anode is approximate  $380 \text{ Wh kg}^{-1}$  in LIB with a  $\text{LiCoO}_2$  cathode [17], which could not meet the requirement for the development of next generation energy storage equipment [18,19]. Aimed at an energy density beyond the practical  $500 \text{ Wh kg}^{-1}$ , lithium sulfur battery and lithium air battery with lithium metal anode are intensively investigated [20–24]. In addition, lithium dendrite also forms on the surface of the recycled graphite anode [25,26]. Therefore, metallic lithium is becoming the most promising anode candidate again. Finding the effective way to suppress the growth of dendrite becomes more and more important for the better application of lithium metal anode [27–30].

Over the past 20 years, various attempts were made to solve the problems of safety concerns and low coulombic efficiency [31,32]. In 1997, Takehara [33] reviewed the suppression methods

of lithium dendrite formations during the charging process. In 1999, Song et al. [34] reported the gel-type polymer electrolytes for LIBs. In addition, they pointed out that these electrolytes could suppress the growths of lithium dendrites. In 2012, Wen et al. [35] summarized the mechanisms of lithium dendrites in the review about LIBs safety issues. In 2014, Li et al. [36] reviewed the deposition criteria and models, and stated that the morphology control might be the key to suppress the initiation and propagation of lithium dendrites in LSBs.

Particularly in 2017, Guo et al. [37] reviewed the behavior of lithium ions upon deposition/dissolution and the failure mechanisms of lithium-metal anodes, as well as the protection method of lithium anode including, tailoring the anode structures, optimizing the electrolytes, building artificial anode electrolyte interfaces, and functionalizing the protective interlayers. Yang et al. [38] outlined the approaches to protect lithium metal anodes from liquid batteries to solid-state batteries and discussed to facilitate the practical application of lithium metal batteries. Zhang et al. [39] reviewed some typical micro/nanostructured lithium metal anodes and presented the suppression of lithium dendrite growth. It illustrated the principles and current situations of micro/nanostructured lithium metal anodes. Cheng et al. [40] presented an overview of the lithium metal anode and its dendritic lithium growth and endeavor to realize the practical applications of lithium metal batteries.

Although lithium dendrite growth received attention, it is quite difficult to completely eliminate dendrites. It is necessary to establish a link between the growth mechanism and the suppression method of dendrite for searching a better way to suppress its growth. Here we reviewed the growth mechanism and suppression method of lithium dendrite. The growth mechanism includes growth model and experimental research. Based on the growth mechanism, the suppression method is divided into two categories: interface modification and manufacturing three dimensions (3D) pore structure. In addition, the link between the growth mechanism and the suppression method is established.

## 2. Growth mechanism

Lithium dendrite is a kind of dendritic crystal, which forms in the condition of deviation from balance. As shown in Fig. 1, this was the typical dendritic morphology, which was reported by Tatsuma et al. [41]. Various researches prove that the current density and the working temperature have great influences on the growth of lithium dendrite [42–44]. In general, both the high current density and the low working temperature can increase the mass transport resistance on the lithium ion and reduce surface reaction resistance offered by a thinner solid electrolyte interphase (SEI) layer, therefore, which causes and accelerates the growth of lithium dendrite [45,46].

### 2.1. Model of dendrite growth

In the theoretical research, a series of models are proposed to describe the thermodynamics and dynamics behaviors of dendrite formation. Yamaki et al. [47] proposed the classical theoretical model in 1998, it was the deposition/dissolution model (Fig. 2). This model was described below: (a) Lithium metal was deposited under the SEI film. (b) Supplied with an external power, lithium ions in the electrolyte transported to lithium metal surface through the protective SEI film. The deposition sites on the protective film exhibited higher lithium ion conductivity, thus crystal defects and grain boundaries in the SEI initiated the continuous deposition of lithium. (c) The mechanical stress within the lithium anode induced an asymmetrical deposition of lithium, resulting in the formation of lithium dendrites.

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