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Original scientific paper

Perspective on the mechanism of mass transport-induced (tip-growing) Li dendrite formation by comparing conventional liquid organic solvent with solid polymer-based electrolytes

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Abstract

A major challenge of Li metal electrodes is the growth of high surface area lithium during Li deposition with a variety of possible shapes and growing mechanisms. They are reactive and lead to active lithium losses, electrolyte depletion and safety concerns due to a potential risk of short-circuits and thermal runaway. This work focuses on the mechanism of tip-growing Li dendrite as a particular high surface area lithium morphology. Its formation mechanism is well-known and is triggered during concentration polarization, i.e. *during mass (Li^+) transport limitations, which has been thoroughly investigated in literature with liquid electrolytes. This work aims to give a stimulating perspective on this formation mechanism by considering solid polymer electrolytes. The in-here shown absence of the characteristic "voltage noise" immediately after complete concentration polarization, being an indicator for tip-growing dendritic growth, rules out the occurrence of the particular tip-growing morphology for solid polymer electrolytes under the specific electrochemical conditions. The generally poorer kinetics of solid polymer electrolytes compared to liquid electrolytes imply lower limiting currents,* i.e. *lower currents to realize complete concentration polarization. Hence, this longer-lasting Li-deposition times in solid polymer electrolytes are assumed to prevent tip-growing mechanism via timely enabling solid electrolyte interphase formation on fresh Li deposits, while, as stated in previous literature, in liquid electrolytes, Li dendrite tip-growth process is faster than solid electrolyte interphase formation kinetics. It can be reasonably concluded that tip-growing Li dendrites are in general practically unlikely for both, (i) the lower conducting electrolytes like solid polymer electrolytes due to enabling solid electrolyte interphase formation and (ii) good-conducting electrolytes like liquids due to an impractically high current required for concentration polarization.*

Keywords
Mass transport limitation; Li metal battery; concentration polarization; liquid electrolyte

Introduction

Li metal-based rechargeable batteries (LMBs) gain enhanced attention in battery R&D owing to higher theoretical specific and volumetric energies compared to conventional Li ion batteries (LIBs) [1-6]. However, the hazardous formation of high surface area lithium (HSAL) morphologies, *e.g.* mossy and dendritic Li, limits their application [2,4,7-9]. HSAL is reactive and leads to capacity losses, electrolyte depletion [2,9,10], as well as a potential safety risks *via, e.g.,* short circuits, which can be typically detected *via* an electrochemical voltage noise response,[11-14] also in Li ion batteries [8,15-18]. Many research efforts have been conducted towards mechanistic understanding of HSAL formation and growth, in particular in regard to the morphology of deposited Li [7,8,19-23].

A well-known HSAL-type is the tip-growing Li dendrite which grows immediate after complete concentration polarization, *i.e.* after complete limitation of (Li^+) mass transport towards Li metal electrode and can be electrochemically indicated *via* a voltage noise behavior [24-26]. In that scenario, dendrite growth relates with complete Li^+ depletion at the Li|electrolyte interface resulting in tip-growing Li dendrite propagation. It can be described as a process counteracting complete concentration polarization, leading to an over-limiting current [24-27]. This mechanism has been derived and studied for liquid organic solvent-based electrolytes, where, due to the good kinetic aspects, harsh electrochemical conditions are required, *e.g.* elevated currents and/or long inter-electrode distances to realize complete concentration polarization [26].

In this work, the aim is to give a perspective on mechanistical validation and understanding of the Li-growth mechanism with the generally poorer ion-conducting solid polymer-based electrolytes [2,28-31] under complete concentration polarization conditions, first introduced by Brissot *et al.* [32].

Discussion

Complete concentration polarization: Mass transport limitation in battery electrolytes

When applying a moderate current in Li metal batteries, extra Li^+ is provided and consumed at the anode|electrolyte and cathode|electrolyte interface, respectively [26,33,34]. As a result, a Li salt concentration gradient emerges throughout the electrolyte and reaches a steady-state after a certain time while evoking Li^+ transport *via* diffusion (Fick's laws). At kinetically harsher conditions, *e.g.* higher currents, low temperature and/or large electrolyte thickness, the applied current can force continuous growth in concentration gradient up to the maximum, *i.e.* complete Li^+ depletion at the cathode|electrolyte interface [33-36]. This 'complete concentration polarization' obeys Sand`s equation and its vertical-type polarization (overvoltage) starts at a defined Sand time [26,34]. The limiting current, defining the threshold for complete concentration polarization, is a characteristic value of DIC electrolytes and depends on Li^+ diffusion coefficient, Li^+ salt concentration and electrolyte thickness [33-35]. These relations can be simply validated in symmetric Li||Li cells as depicted in Figure 1.

A strategy to counteract this issue relies on the use of electrolytes with high Li^+ transference numbers (immobile anions), *e.g.* single-ion conducting (SIC) electrolytes [1,37-41], that counteract the concentration polarization by enhanced migration for reasons of charge neutrality. The impact of SICs on Li deposition is discussed later.

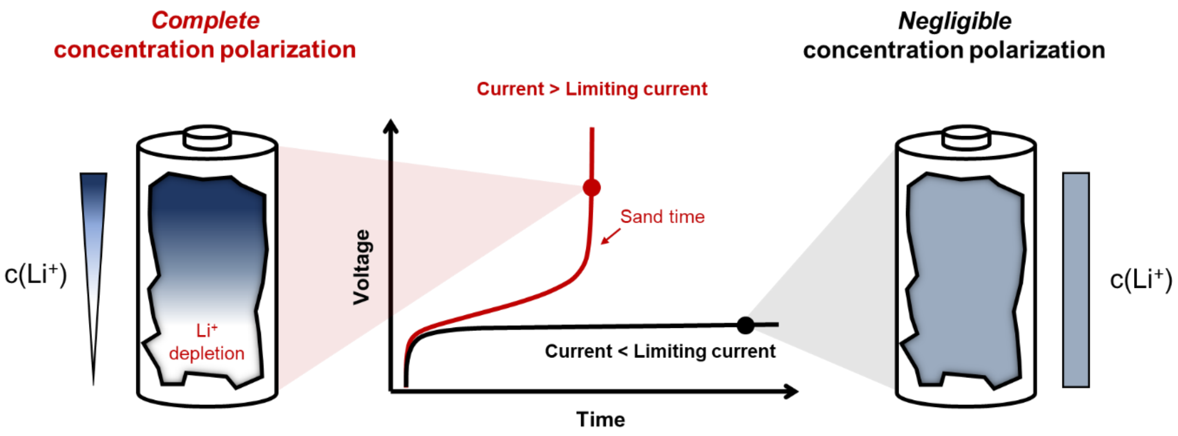

Figure 1. *Schematic illustration of the complete concentration polarization seen at a characteristic Sand time, which proceeds when applied current exceeds the limiting current (redrawn from [37])*

Complete concentration polarization: The onset of dendritic Li deposition in liquid electrolyte-based batteries

In the case of good ion conducting liquid electrolytes, *e.g.* 1 M $LiPF_6$ in a mixture of ethylene carbonate (EC) and dimethyl carbonate (DMC) [2,42-44], concentration polarization can only be observed beyond practical operating conditions, *e.g.* under impractically high currents and/or large electrolyte thickness. Bai *et al.* [26] could visually observe complete concentration polarization *via* the onset of dendritic Li deposition in symmetric Li||Li cells inside glass capillaries with an effective electrolyte thickness of about 7 mm in this model cell compared to a practical separator thickness of ~0.02 mm in real cells as depicted in Figure 2 [26]. It is demonstrated, that the onset of complete concentration polarization marks the transition between mossy and dendritic Li deposition [26]. The latter is regarded as deposition morphology occurring during over-limiting current phenomena, which circumvents Li^+ depletion and is observable by a noisy voltage response immediately after the Sand time (Figures 2a and 2 b) [26]. It has been reported, that mossy Li deposition is a root-growing phenomenon, where Li plating occurs preferably below the protective solid electrolyte interphase (SEI), while dendritic Li deposition proceeds tip-growing preferably above the SEI (Figure 2c) due to higher growing rate compared SEI formation rate, as stated in literature [24,26].

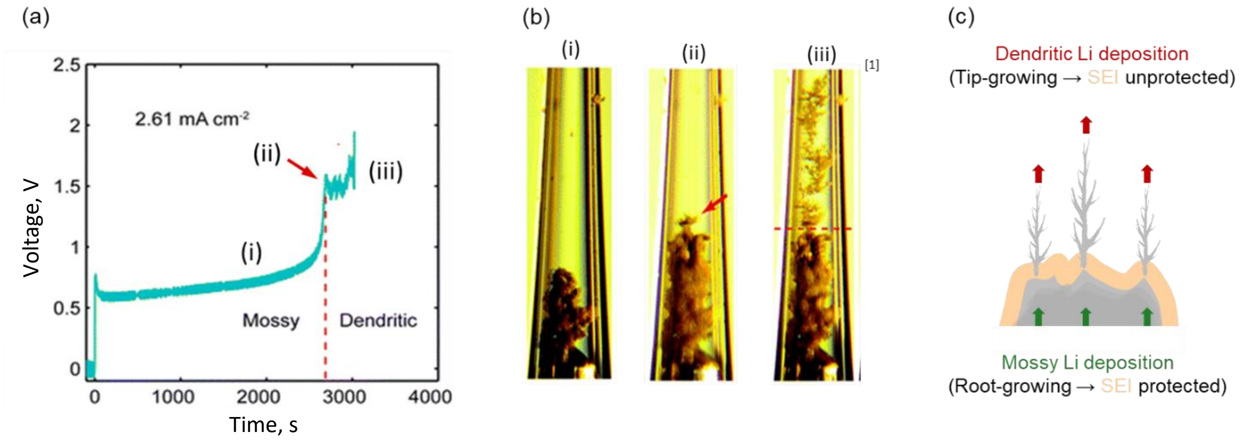

Figure 2. *(a) Voltage as a function of time for an over-limiting current in glass capillary-based Li||Li cell with liquid electrolyte (1M $LiPF_6$ in EC/DMC), derived from Bai et al.[26] (b) Visual observation of the onset of complete concentration polarization (ii) and immediate transition from mossy (root-growing) to dendritic (tip-growing) Li deposition (iii), derived from Bai et al.[26] (c) Schematic illustration of tip-growing (dendritic) and root-growing (mossy) Li deposition, which among others differs in the presence of SEI on the high surface are lithium (HSAL), depending on HSAL growth kinetics relative to SEI formation kinetics.[24,26] (redrawn from [26])*

No impact of complete concentration polarization on dendritic Li deposition in solid polymer electrolytes (SPEs)

Because of considerably poorer kinetic transport properties (*e.g.* lower diffusion coefficients), thus lower limiting currents, observing the concentration polarization in SPEs is practically easier than in liquid electrolytes and can be investigated in conventional cell configuration with conventional *inter*-electrode distances, *e.g.* coin cells [34,45]. Interestingly, contrary to the observations derived by Bai *et al.* for liquid organic electrolytes, the SPE, *i.e.* solid poly(ethylene) oxide (PEO)/Lithium bis(trifluoromethanesulfonyl)imide (LiTFSI), within Li||Li cells proceeds without the characteristic voltage noise immediately after concentration polarization, as shown Figure 3a [32-37]. It can be concluded that a direct transition from mossy Li towards tip-growing dendritic Li is absent with this type of electrolyte. A reason might be an interplay/competition between the rate of Li deposition and rate of SEI growth, which has been suggested by Bai *et al*. [26] for liquid electrolytes (Figure 3b) [24,26]. A requirement for dendritic (tip-growing) Li deposition is a higher rate of deposition (according to the applied current rate) compared to the rate of SEI growth, as otherwise the SEI would passivate the metallic Li deposits and lead to probably mossy (root-growing) Li morphologies, as stated in literature [24]. Hence, it is speculated that the tip-growing dendritic Li deposition might be less favored with SPEs as a consequence of rather lower limiting current, *i.e.* longer lasting deposition times.

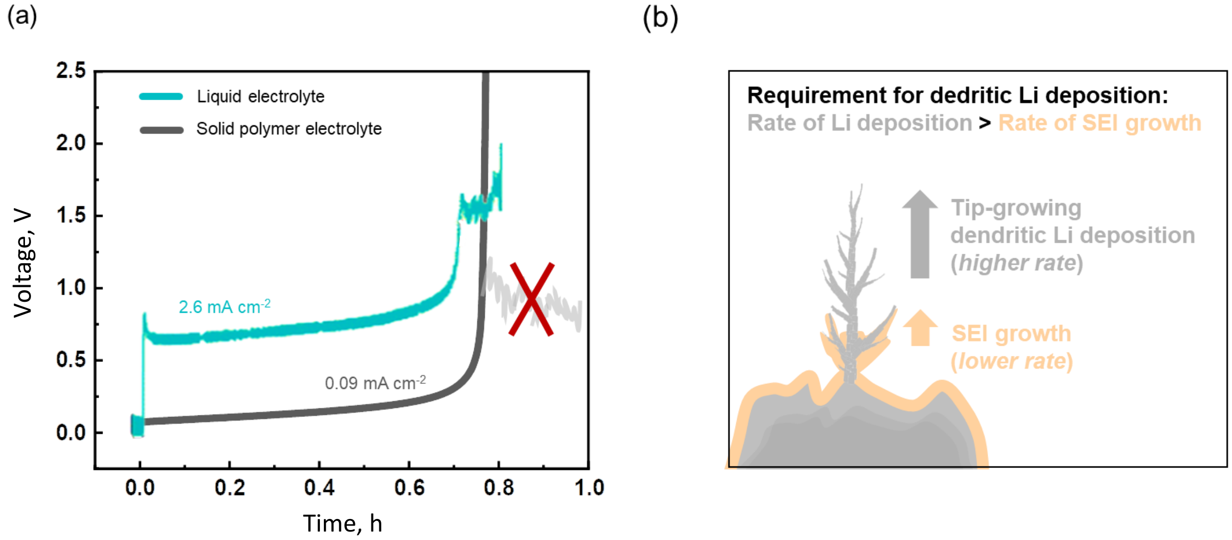

Figure 3. *(a) Voltage as a function of time for over-limiting currents in liquid electrolyte (Bai et al. [26])* vs. *SPE-based Li||Li cells. Complete concentration polarization initiates an onset of dendritic (tip-growing) Li deposition in liquid electrolytes, seen as voltage noise, while such voltage response is missing with SPE. Missing voltage noise points to absence of dendritic Li in SPE [26,34], (b) which is assumed to relate with lower rates (lower limiting currents), which realize sufficient time for SEI growth on the Li deposits, consequently leading to different HSAL shapes (redrawn from [26])*

Interestingly, Brissot *et al.* [32] detect a change in Li plating morphology at high currents, suggesting dendritic Li deposition, though, it is not observed in the first mass transport limiting polarization, *i.e.* first cycle (as seen in liquids), but only after subsequent cycling. This is probably caused by local defects in the SEI. A theoretical possible difference might be also related with electroconvection [24,27,46]. where overpotentials required for tip-based dendritic growth might be significantly higher compared to liquid-based systems due to structural rigidity of SPEs. However, practically they are not observable in voltage ranges up to 10 V, which is significantly higher than voltage ranges in conventional batteries (<5 V); thus relativizing its practical relevance.

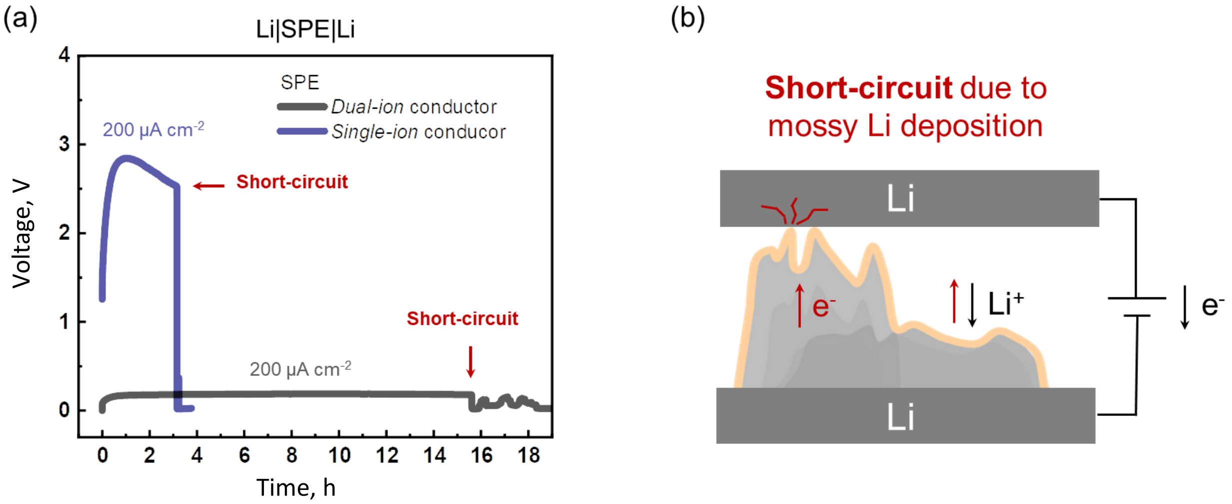

Figure 4. *(a) Voltage as a function of time at moderate areal current (200 μA g^{-1}) of DIC- and SIC-SPE-based symmetric Li||Li cells, both leading to short-circuits.[37] (b) As schematically illustrated, short circuits are likely caused by inhomogeneous mossy Li depositions and growth throughout the SPE after several hours*

Still, short-circuits based on inhomogeneous Li deposition are still relevant in SPEs and happen also below the limiting currents, *i.e.* below onset of complete mass transport limitations/concentration polarizations [11-14]. Though, tip-growing dendritic Li is practically unlikely, other HSAL morphologies like mossy Li deposition still play a major role and also can lead to short circuits *via* penetration as they obviously cannot be mechanically suppressed by the flexible SPE membranes as depicted in Figure 4 for SIC and DIC electrolytes in symmetric Li||Li cells.

The insufficient suppression in HSAL penetration/short-circuits of SPE- compared to liquid electrolyte-based battery cells is likely due to rather poor mechanical stability [12,14] combined with lack in homogeneity (*e.g.* salt distribution), which consequently enhance the risk of inhomogeneous plating [11], thus the growth throughout the SPE-based membrane [13].

Oxidative SPE decomposition is unlikely, as the Li$^+$ depletion occurs at the negative electrode; also, no signs of decomposition (*e.g*. potential plateau) are observable, as would be the case during electrochemical decomposition [14,47,48].

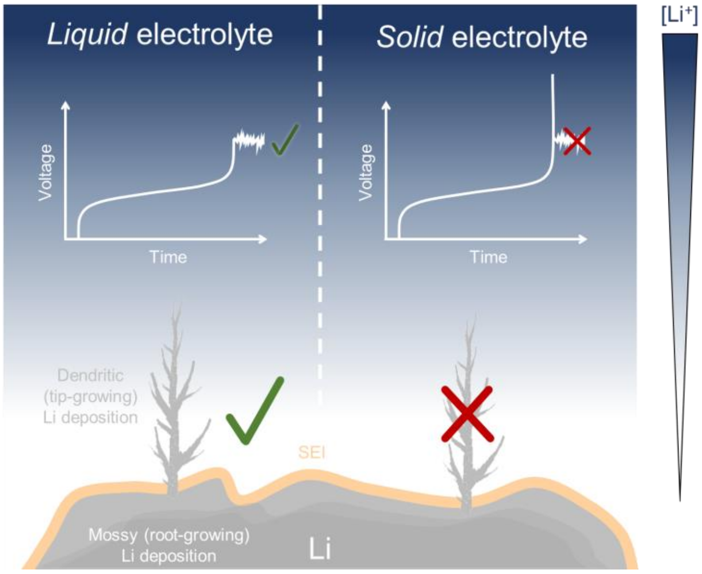

Figure 5. *Schematic comparison of Li interface in contact with liquid- and solid electrolyte at mass-transport limiting (i.e., Li$^+$ depleted) conditions. The tip-growing Li dendrite in liquid electroyltes, seen by characteristic voltage noise, is absent in solid electrolytes*

Conclusion and future perspective

Lithium metal electrodes can potentially raise the energy density and specific energy compared to Li ion batteries, but suffer from growth of reactive HSAL during charge/discharge cycling. Drawbacks like electrolyte depletion, capacity losses, safety issues and short-circuits require intense R&D efforts.

In literature, a complete Li^+ (mass) transport limitation, that is, complete concentration polarization is linked to the onset of a particular HSAL shape, *i.e.* dendritic (tip-growing) Li deposition and has been thoroughly investigated in Li||Li cells with liquid organic solvent-based electrolytes. This HSAL shape is indicated by the characteristic over-limiting current and seen *via* voltage noise [24,26].

Interestingly, with SPEs this particular voltage noise is absent during the complete concentration polarization, pointing to absence of dendritic tip-growing Li growth under the applied specific electrochemical conditions [33-35,37], as summarized in Figure 5. Given the poorer ion transport properties of SPEs compared to liquid electrolytes [28,31,49], the considerably lower limiting current/rate in SPEs [33,34] is suggested to provide sufficient time for formation of SEI on freshly deposited Li, thus being a crucial difference to the literature known situation in liquid electrolytes, where the rate of Li tip-growing is faster than the rate of SEI formation [24,26]. Still, dendritic Li deposition has been observed in SPE-based systems as discussed by Brissot *et al.* [32], but only after subsequent cycling, probably due to defects in the SEI [32,50], in other words, not observed in the initial charge as is seen for liquid-based electrolytes.

It can be concluded, that tip-growing dendritic morphology is generally unlikely in Li metal batteries with both, (i) good-conducting electrolytes (*e.g.* liquid) as impractically high currents and/or *inter*-electrode distances are necessary to achieve complete concentration polarization [26], or (ii) poor-conducting electrolytes (*e.g.* SPEs) as the rate of tip-growing mechanism is slower than the SEI-formation rate, as also stated in literature [24].

Still, short-circuits, most likely due to mossy Li penetration, are still an issue and observed for dual ion conducting (DIC)- and single ion conducting (SIC)-SPE-based Li battery cells [37]. These insights suggest that developing electrolytes with higher cation transference number is not expedient for suppression of the particular tip-growing Li dendrite. Other factors like mechanical stability of SPE and/or homogeneity aspects of Li deposition, have likely a stronger effect [11].

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