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Materials science

Failure of solid-state batteries probed

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The development of a promising type of battery has been plagued by an issue that causes these devices to fail – lithium filaments grow in the electrolyte. An investigation of this failure mechanism could help to solve the problem. **See p.287**

Rechargeable lithium-ion batteries are crucial in a range of applications, including portable electronics, electric vehicles and grid-scale energy storage. Such batteries depend on the movement of lithium ions between an anode and a cathode through a liquid electrolyte. A promising strategy for the next generation of rechargeable batteries is the use of solid electrolytes and an anode made of lithium metal – such cells are known as lithium-metal solid-state batteries. However, these devices are prone to a failure mechanism in which filaments of lithium, known as dendrites, form during battery operation and pierce the electrolyte. On page 287, Ning *et al.*[1] cast light on this mechanism, revealing details that might bring practically useful lithium-metal solid-state batteries closer to reality.

Lithium-ion batteries have many potential uses because they are modular, portable and reliable. They also benefit from long lifetimes, high energy density (which prolongs use before recharging is required) and high power density (which correlates with short charging times). Nevertheless, there is still a continuous push to improve the safety, energy density and power density of these batteries.

In conventional lithium-ion batteries, the liquid electrolyte is flammable and can drive unwanted side reactions that limit the battery's lifetime. Solid-state batteries, which instead use a solid electrolyte, are being intensively researched by academic, industrial and government researchers[2], in part because of claims that such batteries are safer than their conventional counterparts[3]. Solid-state batteries that have a 'bipolar stacking' configuration and energy-dense anodes might also offer notable improvements in energy density and power density[2].

Lithium metal has many properties that make it a potentially good material for anodes in solid-state batteries. For example, it has a low density (0.534 grams per cubic centimetre), low electrode potential (−3.040 volts compared with a standard hydrogen electrode; this is beneficial for making high-voltage batteries) and high energy density (3.86 amp hours per gram). Despite this promise, and more than 40 years of research, major challenges remain that have prevented lithium metal from being adopted as an anode material in rechargeable solid-state batteries.

One vexing issue is the formation of lithium-metal dendrites. In conventional batteries containing liquid electrolytes, this problem is often ascribed to the formation of gradients in the concentration of lithium ions in the electrolyte. This can drive local charge instabilities at interfaces with electrodes, causing dendrites to grow[4]. Concentration gradients cannot form in solid electrolytes, and so this ought to solve the problem – yet solid electrolytes in batteries are still pierced by dendrites, leading to short circuiting.

Ning *et al.* now explore the underlying mechanisms of dendrite initiation and propagation in lithium-metal solid-state batteries. Maintaining contact between the solid electrolyte and lithium metal is essential for achieving reversible and uniform lithium stripping (removal of lithium from the anode during discharge) and deposition (addition of lithium to the anode during charge), both of which are necessary for successful battery operation. Unfortunately, the contact area between the anode and electrolyte can decrease when lithium metal is oxidized during discharging to produce electrons and lithium ions, a process called electrodissolution. This can leave voids in the lithium metal that accelerate battery failure. Void formation can often be counteracted by the application of pressure, but this does not solve the problem completely and can cause anode degradation through various mechanisms[5].

In their study, Ning *et al.* examine the

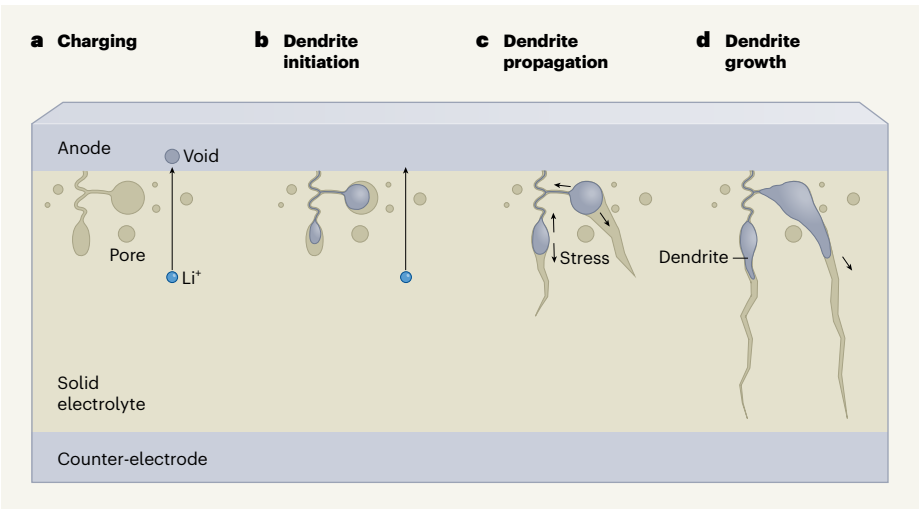

Figure 1 | Exploration of the initiation and propagation of lithium dendrites in batteries. Ning *et al.*[1] studied failure mechanisms that occur at the anode in lithium-metal solid-state batteries. Failure involves the formation of lithium filaments (dendrites) that pierce the battery's ion-conducting electrolyte. In the authors' experiments, a solid electrolyte is sandwiched between a lithium anode and a lithium counter-electrode. **a**, During charging, lithium ions (Li^+) move through the electrolyte towards the anode, where they combine with electrons to form lithium metal that is deposited on the anode. **b**, The lithium fills any voids in the anode and grows through tiny cracks in the electrolyte, filling any pores. **c**, Further charging deposits more lithium in the pores, generating stress that wedges open bigger cracks. **d**, Multiple charging and discharging cycles result in the growth of a dendrite and cracking that eventually cause catastrophic failure of the electrolyte.

physical transformations that occur in a system consisting of a lithium-metal anode in contact with a solid, lithium-ion-containing electrolyte, under conditions that simulate the operation of a battery. The authors varied the pressure applied to the lithium anode during charging, to determine the effects on crack propagation in the electrolyte. They observed that the systems studied have short lifetimes (35 cycles) at moderate pressures (around 7 megapascals) and long lifetimes (170 cycles) at low pressures (about 0.1 MPa). Lithium metal is soft, and deforms at high pressure, which should improve the contact between the anode and electrolyte and increase lifetime. But the authors find that pressure accelerates failure during charging, because it pushes lithium metal through cracks in the electrolyte that can grow during cycling.

Ning *et al*. also used a combination of imaging and modelling techniques to shed light on lithium-deposition mechanisms in micrometre-scale subsurface regions of the solid electrolyte. They observed that lithium metal grows into the solid electrolyte through micro-cracks, and remains stable in porous regions of the electrolyte during charging (Fig. 1). This growth into micro-cracks is postulated by the authors to be the initiation step for lithium-filament growth, and causes stress in the electrolyte.

Further deposition of lithium at the filled pores leads to the metal being extruded to the surface, driving local cracking of the electrolyte. This cracking can decrease lifetime, but does not result in catastrophic failure of the system. Instead, total failure occurs as a result of filament propagation through the electrolyte. This widens the cracks by wedging them open further with repeated deposition and removal of lithium in the developing cracks. Overall, Ning and colleagues' work highlights the spatio-temporal dynamics of lithium-filament formation in solid-state batteries: filament initiation and growth are highly dependent on the microstructure of the electrolyte and on the charging protocols and operating conditions (pressure and temperature).

"Filament initiation and growth depend highly on the microstructure of the electrolyte, and on the charging protocols and operating conditions."

The field of solid-state battery research has been looking for ways to operate the batteries in low-pressure environments, similar to the pressures used for conventional batteries in electric vehicles. Ning and co-workers' results suggest that low pressures can help to suppress dendrite propagation during charging, but might not be beneficial during discharging. Thus, controlling the dynamics of lithium-metal charging and discharging remains a grand challenge for solid-state battery researchers. Work is now needed to clarify how dendrite initiation and growth occur when anodes and electrolytes are coupled with a cathode that is also undergoing volume changes during battery operation.

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