Design of Scalable, Next-Generation Thick Electrodes: Opportunities and Challenges

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ABSTRACT

Lithium-ion battery electrodes are on course to benefit from current research in structure re-engineering to allow for the implementation of thicker electrodes. Increasing the thickness of a battery electrode enables significant improvements in gravimetric energy density while simultaneously reducing manufacturing costs. Both metrics are critical if the transition to sustainable transports system is to be fully realized commercially. However, significant barriers

exist that prevent the use of such microstructures: performance issues, manufacturing challenges, and scalability all remain open areas of research. In this Perspective, we discuss the challenges in adapting current manufacturing processes for thick electrodes and the opportunities that pore engineering present in order to design thicker and better electrodes while simultaneously considering long-term performance and scalability.

Climate change has resulted in a global effort to reduce CO_2 emissions. As a result, there is an intensified demand for low- and zero-carbon emission transport systems. Consequently, the automotive sector is undergoing an immense transformation, such that the internal combustion engine is being replaced by lithium-ion battery (LIB) powered electric vehicles (EVs). As a result, there is ever-increasing demand for LIBs with greater energy storage and superior rate capabilities, requirements that dictate the range, performance, and, ultimately, the user uptake of EVs. In order to make EVs affordable, the battery-manufacturing process must become more economical.[1] Satisfying the growing need to improve battery performance while simultaneously reducing cost is central to emerging strategies for the design and manufacture of suitable electrodes, where thick electrodes will play central roles. Up to a certain thickness, thicker electrodes offer the possibility of a significant improvement in energy density over the present, thinner forms of lithium-ion battery electrodes.[2] These improvements are achieved *via* increasing the amount of active storage material while removing potentially redundant inactive components, which simultaneously reduces costs. However, there remain significant barriers to their implementation: Thick electrodes possess poor power density characteristics due to ion transport limitations[3] and they are difficult to manufacture at scale using present processes. In this issue of *ACS Nano*, Wu *et al*. report a novel methodology for the manufacture of thick, high-capacity electrodes,[4] highlighting the need for further discourse on the topic. In this Perspective, we consider the implications of thick electrodes on LIB performance and assess the challenges and opportunities that lie at the heart of their design, manufacture, and scalability.

At present, the average market price for LIB packs is \$137/kWh and is projected to decrease year to year until LIBs become comparable to internal combustion engines in manufacturing costs and profit margin (\$101/kWh in 2023).[5] However, in order to accelerate this process and to drive costs down even further, it is imperative to design better battery electrodes that are readily manufacturable, while realizing greater balance of power and energy density. A promising approach is the use of thicker battery electrodes. A battery electrode is a composite structure that comprises active storage materials (typically formed of graphite or silicon at the negative electrode and $LiCoO_2$ (LCO), $LiFePO_4$ (LFP) or $Li[Ni_{1-x-y}Mn_xCo_y]O_2$ (NMC) particles at the positive electrode), a nanoporous carbon black-polymeric conductive binder (CBD), and pores that permit electrolyte percolation, giving lithium ion conduction and transport. A separator permits ionic current and mass transport between electrodes, while a copper or aluminium current collector provides the outlet for electronic current flow to external loads. These structures are usually stacked, or rolled, in repeated layers forming battery cells, which are then combined to form the battery. Increasing the electrode thickness has a twofold effect: (1) If fewer inactive materials (separators and current collectors) are used in any given cell then the material costs can be reduced, and (2) the relative quantity of active storage material increases, and increased energy density then follows. For example, in the case of an NMC/graphite cell, gravimetric capacity of the cell can increase by ~17% if the thickness of a typical cathode is doubled (to 200 µm).[6] However, challenges in the design and deployment of thick electrodes remain: Greater power density must be achieved *via* pore engineering to overcome the diffusion limitations, and traditional slurry casting, drying, and calendering must be augmented to inhibit the microstructural damage that occurs during fabrication[7] while enabling the efficient implementation of the pore-engineering solutions.

Herein, we discuss the challenges and opportunities associated with the design, manufacture, and performance of thick electrodes. We describe the underlying performance-related mechanisms; examine manufacturing issues; and, finally, discuss the topic of pore engineering, detailing pathways to feasible electrode microstructural design. We address these issues under the broad theme of scalability.

PERFORMANCE-RELATED CHALLENGES OF THICK ELECTRODES

In broad terms, the rate performance of LIBs is dictated by electrolyte diffusion limitations, such that lithium ions originating from the separator/electrode interface may or may not reach the active materials near the current collector, resulting in poor utilization and, thus, reduced storage capabilities.[8–11] The constituent materials of a conventional composite electrode are mixed such that active particles, and, in particular, the nanoporous carbon binder, provide a tortuous pathway for bulk electrolyte percolation and, thus, lithium ion mobility.[12] This highly tortuous network limits electrode behavior but, in the case of thick electrodes, the diffusion limitation is amplified such that at high charge/discharge rates the loss in capacity reaches an impractical level.[13–15] The location of the current collector and the distribution of carbon binder also affect the electrical conductivity of the electrode.[16] When all of these factors are combined, severe heterogeneities in lithium concentration are found within the particles and throughout the electrode, which originate from the steep gradients in electrolyte concentration, overpotentials, and the electrochemical reactions through the thickness of the electrode.[17] These findings are critical, as electrode microstructure greatly affects battery performance.

As an example of the influence of microstructure on battery performance, consider the modelling work of Lu *et al*.[18] The state of lithiation (SoL) or material utilization is shown in Figure 1A and B for 60 µm and 80 µm electrodes, respectively, at low and high discharge rates (1C, 3C, and 5C). At high discharge rates (5C), material utilization is poor for thicker electrodes, whereas areal capacity is improved significantly at rates typically below 1C (see Figure 1C). In Figure 1C, the authors illustrate the influence of porosity (and the associated pore tortuosity) on performance; once the electrodes are prescribed a high porosity ε_H, the areal capacity increases significantly, an effect that has greater influence for thicker electrodes. The authors' work shows a clear link between electrode thickness and porosity while highlighting the levels of heterogeneity in utilization of active material, illustrating the potential for using microstructural design techniques such as pore engineering to improve electrode capacity at high rates of discharge for thick electrodes.

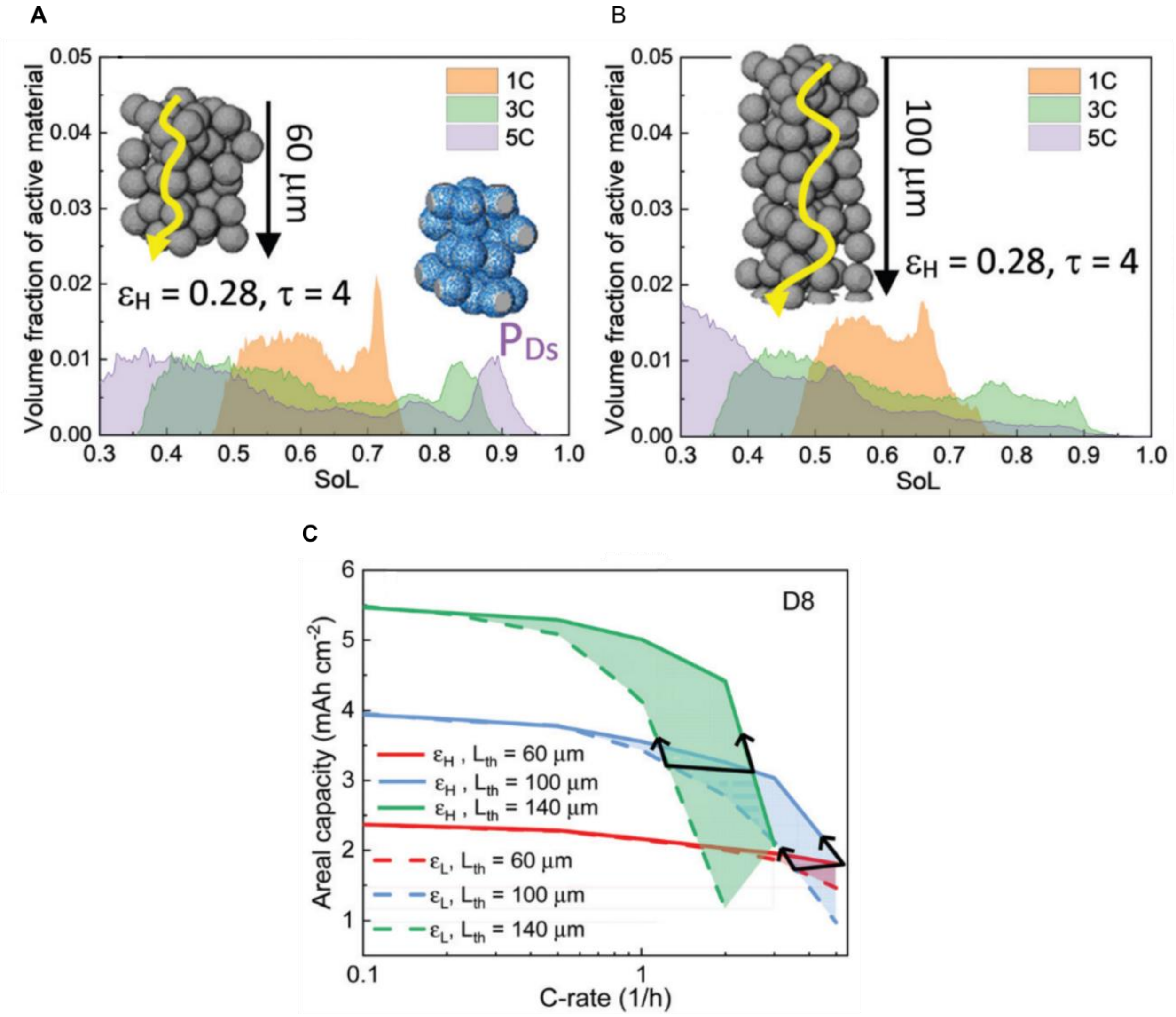

Figure 1. Histogram of state of lithiation (SoL) for discharge rates of 1C, 3C, and 5C for an electrode with **(A)** 60 µm thickness and **(B)** 100 µm thickness. The tortuous pathway for lithium ion transport is also illustrated in **(A)** and **(B). (C)** illustrates the areal discharge capacity for electrodes of varying thickness L_{th}, and a low and high porosity, $\varepsilon_L = 0.23$ and $\varepsilon_H = 0.28$ respectively. Adapted with permission from ref 18. Copyright 2021 Royal Society of Chemistry.

Long-term performance is another critical factor that has received limited attention. Zheng *et al.*, for example, cycled LFP and NMC electrodes, noting significant capacity fade, which they attribute to severe polarization and mechanical stability.[13] Both mechanical and thermal stability of thick electrodes are important considerations. There are severe electrode-level gradients in electrolyte concentration and, thus, in electrochemical reactions. Meanwhile, there tend to be particle-level gradients in lithiation due to slow diffusion in the active material. This slow diffusion can lead to through-thickness gradients in ‘damage’, which have been observed experimentally for thin/moderate electrode thicknesses (~40 μm).[19,20] In a thick electrode, high reaction rates occur near the separator, which exacerbates this issue and leads to excessive, localized particle cracking due to solid-state diffusion in the active material. Naturally, this particle cracking may lead to accelerated degradation of the electrode, as described by Zheng *et al*.[13] These heterogeneities may also manifest in another way, associated with the thermal behavior of the electrode: A thicker electrode provides greater resistance to current flow, which results in greater levels of ohmic heating. If ohmic heating is coupled with the high levels of localized electrochemical reactions (and, thus, resistance), then the thermal distributions within the electrode become an important consideration, particularly where issues of battery safety (thermal runaway) and operation at low or elevated temperatures are concerned.[21] Furthermore, elevated temperatures lead to greater thermal stresses, which may exacerbate particle damage.

Electrode and, thus, battery performance are, in general, inextricably linked to microstructural architecture. Therefore, the design of suitable electrode microstructures is a key element in the process of making thick electrodes with increased performance. One such approach is that of Wu *et al*., who used a phase inversion method to align pores hierarchically to overcome transport-related performance limitations.[4] They demonstrated improved transport kinetics, offering better power density and mechanical stability for thick electrodes (1.2 mm). In addition, they demonstrated improved cycling stability, a clear step forward in the path to thicker electrodes with improved electrochemical performances.

CHALLENGES FOR THICK ELECTRODE MANUFACTURE USING CONVENTIONAL TECHNOLOGY

Cell manufacturing is a complex process and the electrode structure, which is key to cell performance, is formed during the ‘mixing–coating–drying–calendering’ steps. After calendering, the microstructure is fixed with no further change; subsequent steps focus solely on the assembly of the electrodes into various cell configurations. If electrodes are processed incorrectly, the resulting errors in the microstructure result in lower cell performance, and there are no subsequent processing or assembly steps that can remedy or enhance cell performance. Drying and calendering processes are of critical importance in defining electrode structure and the resultant cell performance. Conventional electrode-manufacturing processes have been optimized over nearly three decades of mass manufacture; with the emerging opportunities for advanced manufacture of thick electrodes, challenges that may result from re-optimization of these critical processes need to be better understood.

Drying of the wet electrode film occurs directly after slurry coating, and its purpose is to remove the solvent used to formulate the slurry. It is the second ‘irreversible’ step during electrode manufacturing process where defects and errors will result in scrappage and will decrease yield. Solvent removal has a large impact on the final electrode structure and performance. Drying is a multistep process that involves evaporation of the solvent, sedimentation of the particles, shrinkage of the film, and upward migration of the precipitating binder and carbon black.[22] For thinner (<100 µm) coatings, the resultant buildup of mechanical stress and gradients in binder distribution may not be sufficient to overcome electrode cohesion and adhesion. However, increasing the thickness of the coating to hundreds of microns results in cracking and delamination of the electrodes and likely decreases their cycling lifetime. As with mixing, drying is a process that is mature, however, its impact on electrode electrochemical performance is not yet fully understood.

After coating, the deposited material is dried in a continuous process using a coating head where the solvent is evaporatively removed from the material by heating. In the case of cathode coatings, the highly flammable and toxic solvent *N*-Methyl-2-pyrrolidone (NMP) is recovered at enormous cost to protect personnel and the environment. Transport of the current collector foil is achieved either by roller systems or by floatation air streams for single-sided and double-sided coatings, respectively. Portions of the drying tunnel are kept at different temperatures in order to produce a desired temperature profile as the coating travels though. After passing through the dryer, the foil is cooled to room temperature and, depending on the type of system, rewound (conventional) or directly coated on the second side (tandem coating). Typically, the coating will dry in 1 min, although, for thicker electrodes, drying can take more than 10 times longer.[7,23,24] As a result, long drying ovens (50–100m) are needed, and bottlenecks may occur due to the drying tunnel volume/length, which limits the coating speed rather than the coating head itself. Long ovens add significantly to the overall manufacturing line footprint and require ~40% of the energy used in the manufacturing process.[25]

As with mixing and coating, determining the required quality factors that influence the electrode performance mandate a deeper understanding of the drying mechanisms. Well-controlled temperature profiles at every position in the dryer are extremely important. Analysis of quality factors such as film adhesion, residual water content, and defect density can only be performed offline. As online measurements become available, the problem of control in the drying process becomes a challenge due to the lack of complete understanding of the drying process, and there are very few ‘levers’ to modify the process. These challenges are particularly acute for thick electrodes, because of the tradeoff between drying time and migration of functional additives such as the binder.

Controlled removal of a solvent is the single most significant problem of drying: Development of dry or semidry processing would circumvent this process entirely but it is unlikely that slurry casting will be replaced within the next five years. Solvent removal can be achieved using radiative dryers (such as infrared lamps), which heat the coating directly compared with convective air dryers. Further advances using laser drying have also been investigated, due to the smaller area that can be dried rapidly and precisely.[26]

The final step in the electrode-manufacturing process is the consolidation of the dried electrode layer, known as calendering. Calendering typically uses two large rollers to compress the dried electrode structure and finalize the electrode porosity, enhance the electronic conductivity, and improve the adhesion to the current collector foil. A common approach is to heat the metal rollers above the glass transition temperature of the binder so that the electrode will more readily plastically deform. Typically, the electrode porosity is reduced from *ca*. 50% to between 20–40%, depending on the specification of the electrode. Pore-engineered thick electrodes are often used in an uncalendered state because calendering either partially or entirely collapses the structure.[27] However, noncalendered electrodes have associated disadvantages such as low density, relatively high impedance, and surface roughness. Recently, Wu *et al*. used wet calendaring, with reasonable success, for pore-engineered thick electrodes.[28]

The quality of the calendering process is also difficult to monitor in- or online, where factors such as electrode porosity, electrode adhesion, and surface roughness are all key to the cell performance. Defects at the edge of the film with the interface of the current collector are common and also occur on intermittently coated samples where the coating will ‘smear’ under compression.

Upstream quality defects (such as variations in thickness) impact the calendering step by manifesting as variations in porosity (for a given roller gap). This phenomenon again highlights the fact that quality defects cumulatively combine to impact the process adversely. Fundamental understanding of the process combined with advances in process monitoring and control are key to overcoming the negative effects of these quality defects. Although the process may seem simplistic, there are a number of complex microstructural changes[17,29] occurring during the compression process that are poorly understood and not directly controlled in industry. Determining the exact thickness at which the calendering process is no longer effective is not yet an exact science; more fundamental research and models are needed to understand the defect categories and the microstructural evolution in thick or pore-engineered electrodes.

Advances in diameter and gap control over calenders will provide performance advances in this manufacturing step. Improvements in calendering models are closer to maturity compared with other manufacturing steps.

IMPROVING THE MICROSTRUCTURAL PERFORMANCE *VIA* PORE ENGINEERING

In the drive to increase electrode thickness to realize improved volumetric energy density at the battery cell and pack levels, the benefits of electrode pore engineering are well-established by both theory[12] and experiments.[18] If Li-ion gradients in the interconnected pore network of the electrode become significant (as is typical for the tortuous structures produced by slurry casting), energy-storage reactions of Li ions with the active material are incomplete, or advance too slowly.[30] As a result, fading of realizable capacity with increasing electrode thickness occurs, especially as the charge/discharge rate increases. In principle, pore engineering of the electrodes enables the pore fraction, dimensions, and tortuosity in each of the three orthogonal dimensions to be preselected. There has been growing interest in developing the pore-engineering processes to replace the usual tortuous, random porosity of the electrodes with a designed microstructure containing vertically oriented pore arrays through the electrode thickness. Reducing the tortuosity of the interconnected pore network through the electrode thickness facilitates faster ion transport and can partially reduce the tendency for Li-ion concentration gradients to develop as electrode thickness increases. Ideally, however, pore engineering should not increase the overall porosity of the electrode, as this can otherwise cancel energy density benefits of thick electrodes. Recent simulations by Mai *et al.* show that, even for the currently available 100 μm thick electrodes, reorganizing part of the electrode porosity into <10 μm wide, regularly spaced vertical pores can increase cell energy density by up to 30% at fast charging rates.[31]

Many routes of pore engineering for thick electrodes have been studied, and these can largely be divided into two types: additive/subtractive manufacturing and *ex-situ*/*in-situ* templating. Additive manufacturing methods include co-extrusion of lab-synthesized rods containing a mixture of LCO and polymeric binder with an axial core of C followed by pyrolysis[32] and three-dimensional (3D) printing of acrylate-based resins followed by ultraviolet light curing and pyrolysis to form C-based anodes.[33] The advantage of additive manufacturing is an extremely ordered fabricated microstructure (*e.g.*, Figure 2A), whereas a disadvantage is large diameters of pore channels due to the resolution restrictions of the processing instrument (*e.g.*, 100 μm for 3D printing[33] compared with 15–40 μm when fabricated by other pore–engineering methods).[34] Subtractive manufacturing methods include laser patterning of the fabricated electrodes (Figure 2B,C) and offer the advantage that the electrode-casting process does not need to be changed dramatically (providing high compatibility with the existing manufacturing method). However, the disadvantages of subtractive manufacturing methods include relatively high capital costs and long financial amortization,[1] low throughput, and potential droplet formation along the laser cutting kerf due to partial electrode materials melting.[3] Furthermore, the aspect ratio between electrode thickness and pore channel diameter is low (*e.g.*, 60 µm electrode thickness with 30 µm pores on the electrode surface by laser patterning[35] compared with 500 µm–1 mm electrode thickness with similar pore sizes made by other pore-engineering methods)[36] because the laser-drilled holes start to taper with increasing electrode thickness.

The templating method involves forming a temporary scaffold to guide the movement of electrode materials during the electrode-formation process. Subsequently, the scaffold is typically removed using a solvent, heat, or combinations of approaches, leaving an electrode microstructure with structured porosity. The template can be preformed, such as with infiltration electrode materials, into a natural wood framework followed by pyrolysis to make LCO cathodes (Figure 2D).[36] Alternatively, the template can be formed *in situ* with the electrode, such as by directional ice templating (DIT, or freeze casting) of $LiFePO_4$ (LFP),[27] LCO,[37] and $LiNi_{0.8}Co_{0.15}Al_{0.05}O_2$ (NCA)[38] cathodes and graphite anodes[39] where a temperature gradient is applied to the electrode slurry to encourage vertical growth of ice columns, which are then sublimed. The magnetic templating of LCO cathodes takes place where either magnetic microrods or ferrofluid emulsion droplets are added to the electrode suspensions to encourage alignment in the applied magnetic field direction followed by removing as much Fe_3O_4 as possible.[34,40] Phase inversion of LFP cathodes accompanied by a metal mesh has also been developed, where the metal mesh is used to remove the dense skin layer at the top of the electrode slurry.[4] Although the majority of pore-engineering methods are used on LIBs with liquid electrolytes, innovative developments from these methods are starting to be applied to next-generation batteries; for example, directional freezing and polymerization (DFP) can be used to make thick electrodes containing vertically interleaving arrays of electrode and solid-state electrolyte materials for solid-state batteries.[41]

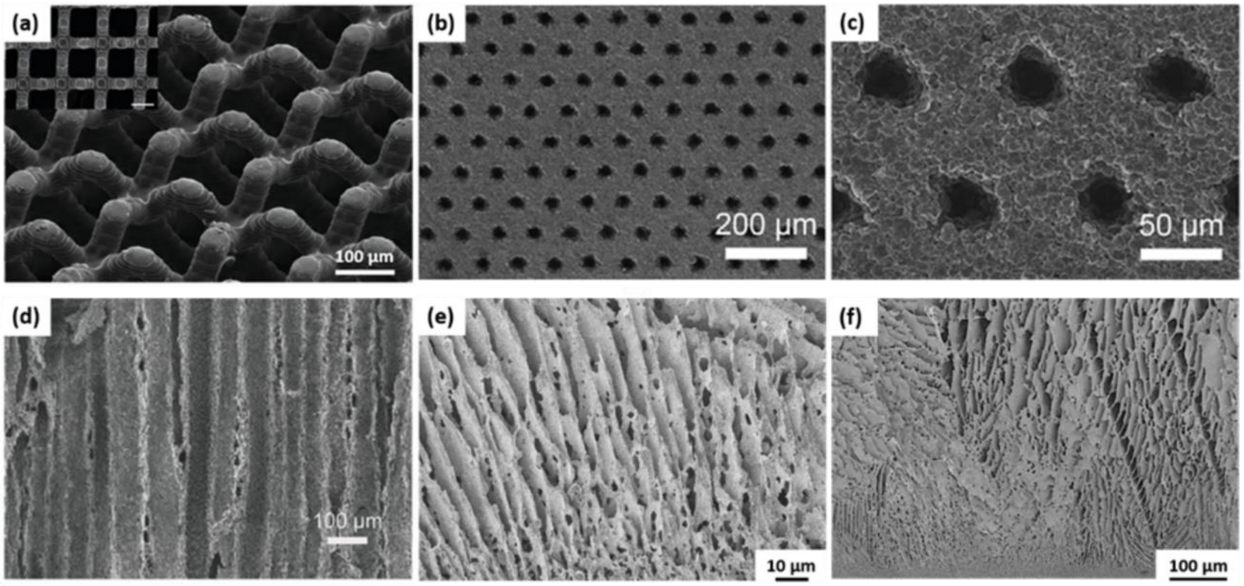

Figure 2. Top view scanning electron microscopy (SEM) images. **(A)** C-based anode made by three-dimensional printing. Adapted with permission from ref 33. Copyright 2021, John Wiley & Sons **(B, C)** C-based anode made by laser patterning. Adapted with permission from ref 35. Copyright 2020 Elsevier. **(D)** Cross-sectional SEM images of a $LiCoO_2$ cathode made by wood templating. Adapted with permission from ref 36. Copyright 2018, John Wiley & Sons. **(E, F)** $LiFePO_4$ cathode made by directional ice templating. Adapted with permission from ref 27. Copyright 2019 Royal Society of Chemistry.

Some *in-situ* templating methods have been investigated in detail to understand the effects of the processes on the resulting microstructure of thick electrodes. For example, Delattre *et al.* investigated different cooling rates of 5, 7.5, and 10 °C min^{-1} for the DIT method; they found that increases in cooling rate led to significant changes in electrode morphology and an increase of pore tortuosity from 1.74 ± 0.09 to 2.09 ± 0.1.[38] In addition, Li *et al.* investigated different binders for the magnetic alignment method, including polyvinyl alcohol (PVA), a commercial M-binder, and polyacrylic acid (PAA). They observed varying interfacial stability between the different binders and both the magnetic nanoparticles (containing oil droplets) and the water solvent, resulting in different pore morphologies.[40] The advantage of *in-situ* templating methods is that by tuning the key processing factors, finer pore channel diameters can be fabricated (*e.g.*, <7 μm for DIT[3,37,40] and magnetic alignment[34] as shown in Figure 2E, compared with 30–40 μm by *ex-situ* templating methods).[36] However, methods that utilize the mobility of liquids as a means to influence the movement of electrode materials during electrode formation, such as ferrofluid emulsion droplets, usually have restrictions on the electrode thickness (*e.g.*, 200–310 μm)[34] because controlling the process becomes more difficult with increasing thickness. In this sense, the 1.2 mm ultra-thick electrodes made by phase inversion reported by Wu *et al.*[4] have successfully pushed the boundaries of the electrode thickness that can be achieved with an aligned microstructure formed by a solvent-based alignment mechanism.

The two key challenges to advancing pore-engineered electrodes and their benefits on practical scales are low volumetric capacity and lack of scalability. As well as containing the aligned pore structures, the pore-engineered electrodes usually contain relatively high porosity (40–71 vol%[4,40] compared with 20–40 vol% for conventional slurry-cast electrodes). Because volumetric energy density is an important performance metric for batteries, different approaches have been developed to reduce the porosity of the pore-engineered electrodes, including densifying the electrode material columns through sintering at 700–1000 °C[4,32,36] and fabricating density gradient electrodes that have small pore channel diameters (~500 nm) in the region nearest the current collector and large pore channel diameters (1–20 μm) in the region nearest the separator to maintain a relatively low overall porosity (36 vol%, Figure 2F).[27] The second challenge, a lack of scalability (speed and throughput), will be key for battery gigafactories to reduce costs through economies of scale. Pore-engineering methods have primarily been demonstrated at the laboratory scale to date, and they involve several additional process steps. However, there are also promising early signs (*e.g.*, with the maturity of phase inversion in the polymer membrane industry[4]), that the simple, fast, and up-scalable character of pore-engineering methods means that they have the potential to be adapted to current battery production lines.

TOWARD IMPROVED THICK ELECTRODE PRODUCTION AND PERFORMANCE USING PHASE INVERSION

The phase inversion technique used by Wu *et al.*[4] utilized the difference between the high solubility of typical polymeric electrode binders in organic solvents (such as NMP), as opposed to the insolubility of such binders in water. When the slurry of energy-storing powder (here, LFP), conductive additive, and polymeric binder dissolved in NMP is put in contact with water, the diffusion of water destabilizes the binder solution, causing liquid–liquid demixing and formation of binder-rich and binder-lean phases within the slurry.[42] The supersaturation of the binder in the binder-rich phase leads to its rapid precipitation and solidification, resulting in macroscopically more uniform distribution of the binder in an electrode. Consequently, such electrodes are more mechanically stable compared to electrodes fabricated *via* conventional (and slower) drying, where binder migrates and aggregates at the top region of the electrode, giving poor adhesion to the current collector.[43,44]

In addition to being mechanically stable, the 1.2-mm thick electrodes created by Wu *et al.* exhibited 100 μm-wide, vertical pores that protruded through the entire composite layer, as shown in Figure 3.[4] The formation of such pores can be related to the convective flow of water quickly penetrating the slurry during phase inversion.[42] Similar structures could also be induced thermally by using slurries that contain two solvents with different boiling points and different affinities to dispersed solids, which, upon consecutive evaporation during drying, would result in the formation of vertical pores in solidified electrodes.[45,46] Although vertical pores in phase-inverted electrodes are typically capped with a solid thin layer, Wu *et al.* found a way to remove this layer using a metal mesh peel.[4] Intriguingly, the authors noticed that the distribution of vertical pores in the fabricated electrodes partially followed the distribution of the metal mesh apertures, a finding that may be promising for future optimization of pore distribution in thick electrodes.

Thanks to the presence of low-tortuosity vertical pores, nearly 100% of the typical LFP capacity (160 mAh/g) could be reached in a 1.2 mm-thick electrode (with 87% active mass loading) during cycling at 0.05 C, and almost 50% of that capacity was delivered at 0.5 C.[4] Such rate performance, albeit inferior to the typical < 100 µm thick LFP electrodes, is undoubtedly significant for an ultra-thick electrode. At low C-rates, the demonstrated electrodes could offer up to 32% higher gravimetric energy density of a Li-metal cell compared to a cell made of conventional, thinner electrodes. The somewhat thinner (~500 µm) electrodes also retained 99% of their initial capacity after 120 cycles at 0.5 C, although much faster deterioration was noticed for the 1.2 mm-thick electrodes. Although the volumetric energy density and long-term stability of demonstrated electrodes need to be improved for their practical application, the work of Wu *et al*. shows how a technologically simple method of microstructure engineering can enable the fundamentally challenging fabrication and utilization of ultrathick electrodes.

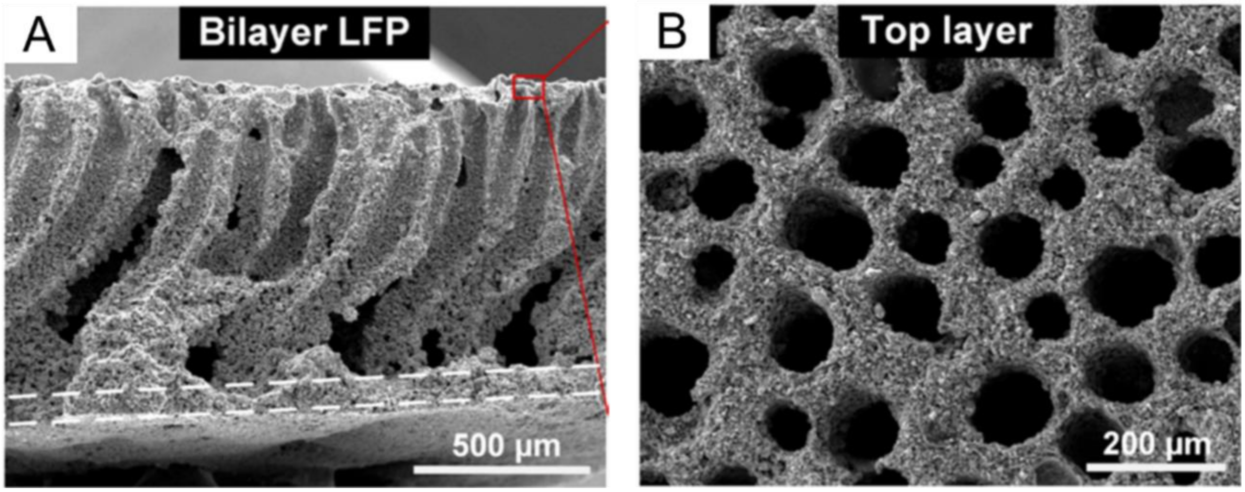

Figure 3. Scanning electron microscopy images of 1.2 mm thick $LiFePO_4$ electrodes with vertical channels obtained *via* phase inversion, projected in **(A)** cross-section and **(B)** top view. Adapted from ref 4. Copyright 2021 American Chemical Society.

OUTLOOK

Although a wide range of solutions exists to overcome the performance-related issues associated with thick electrodes, a major advance is required to realize their efficient manufacture on the scale of the modern gigafactory. Some promising manufacturing alternatives have recently been reported,[47] however, the traditional slurry cast manufacturing approach remains dominant, and replacing this process is perhaps unrealistic in the short term. Hence, solutions are required that minimally alter, or align with, these practices. For example, casts might be altered to incorporate pore channels, or spray coating[16] could be implemented in tandem without disrupting the casting process.

If manufacturing processes are to be adapted to accommodate thicker electrodes, then there are gaps in fundamental understanding that need to be addressed. For example, the drying process in slurry casting is not suitable for the effective removal of solvent from slurry cast thick electrodes because severe microstructure cracking and delamination from the current collector occurs due to shrinkage.[7] These mechanisms must be inhibited completely for the scalable use of thick electrodes. Although Wu *et al*. demonstrated rapid replacement of solvent and structure stabilization,[4] it remains to be seen whether this method can be implemented on a large-scale basis. Furthermore, the calendaring process remains another open issue: If pore engineering produces a scalable solution to improving ion transport in the electrode, then how does rolling a well-designed microstructure influence the original intended performance?

It is also worth noting that adapting manufacturing processes for thick electrodes can offer nonobvious benefits to the cost structure of electrode fabrication. For example, introducing phase inversion to battery fabrication may alter the management of solvent waste, which contributes significantly to battery-manufacturing costs. Contrary to condensing the solvent from its vapors during typical electrode drying, the water-NMP waste generated during phase inversion can be gathered and shipped for processing in separate, dedicated facilities, using alternative methods (*e.g.*, membrane filtration) at much larger scales than in typical battery manufacturing lines. As a result, the complexity, potential safety hazards, and overall cost of battery fabrication may be reduced.

Performance-based understanding of thick- and pore-engineered electrodes is another key area of research that must be addressed. For example, LIBs degrade and experience capacity fade *via* a wide range of electrochemical, thermal, and mechanical phenomena;[48] degradation is an ongoing subject of research for thin/moderate electrodes, and it has yet to be comprehensively understood. If thick electrodes are introduced into cells, then further degradation-based understanding is required, primarily due to the inherent severe heterogeneities that are introduced into the microstructure. For example, we do not fully understand the long-term-capacity loss of thick electrodes in general, which is linked to mechanical and thermal degradation mechanisms, among others. Relatedly, there are gaps in our understanding of battery safety and long-term stability, such as how these electrodes perform under low- and high-temperature operating conditions. If a major step forward is to be taken in the use of new electrode architectures, then comprehensive analyses must be carried out on their long-term performance. For example, advanced characterization techniques such as X-ray diffraction and X-ray computed tomography (CT) can be used to gain deeper understanding of the underlying microstructures. These techniques can then be combined with multiphysics simulations (*i.e.* image-based models, typically based on X-ray CT images), which can facilitate rational design of pore-engineered electrodes using realistic microstructures as the basis for simulation.[49]

Significant opportunities exist for a stepwise change in LIB technology *via* the simple act of increasing the electrode thickness. However, the coupling of fundamental research and large-scale engineering efforts is required to overcome the manufacturing- and performance-related issues that currently impede the use of such batteries. Scalability lies at the heart of this problem, and is yet to be satisfactorily addressed; solving the problem of scalability will require synergy between pore engineering and advanced manufacturing techniques. Wu *et al.*, with their phase-inversion technique, have begun to find such a path forward.[4] However, the question remains as to whether pore engineering and its associated manufacturing techniques can meet the challenges related to the utilization of thicker, next-generation chemistries such as sodium-ion, silicon-based anodes, and solid-state batteries, among others. More generally, combining insights from particle engineering; advanced electrode manufacturing, characterization, and modelling; metrology; and optimization of processing parameters is required. This holistic philosophy is a cornerstone of the Faraday Institution's Nextrode project, which is tackling the challenges of next-generation electrode design and manufacture. A paradigm shift in electrode manufacture, targeting simultaneous improvements to performance and cost, requires a coordinated and multidisciplinary approach; achieving this within time scales commensurate with Net Zero ambitions is truly a grand challenge.

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