

5<sup>th</sup> Conference on Production Systems and Logistics

# Comparative Analysis of Lithium Metal Anode Production Methods: Evaluating Liquid-Based Manufacturing Technology for Mass Production

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## Abstract

Lithium metal anodes (LMA) have gained significant attention for their potential to revolutionize rechargeable battery technology, offering high theoretical capacity and low electrode potential. Their implementation in various applications, such as electric vehicles and portable electronics, holds the promise of significantly improving energy density and battery performance. Additionally, the successful integration of lithium metal anodes remains a crucial and yet-to-be-resolved challenge in the development of All-Solid-State Batteries (ASSBs), which aim to provide safer and more efficient energy storage systems. Overcoming the production challenges associated with lithium metal anodes is essential for realizing their full potential. This paper presents a comprehensive technology analysis and evaluation of production methods for lithium metal anodes. The analysis explores various techniques and their potential for mass production. Furthermore, this analysis evaluates the viability of each approach by considering factors such as the potential for performance improvement, cost savings, quality enhancement, and the technology readiness level. The paper outlines future directions for the development of these techniques while focusing on the liquid-based processing approach, aiming to address quality issues and enhance its scalability for large-scale production. In conclusion, this technology analysis and evaluation underscores the potential of liquid-based manufacturing technology for the mass production of high-quality lithium metal anodes and highlights the need to overcome production challenges. An approach is presented that offers a way to work through the challenge of LMA production, paving the way to next-generation battery cells.

## Keywords

Lithium Metal; Anode Production; Mass Production; Lithium Metal Anode; Solid-State Battery; Next-Generation Battery

## 1. Introduction

Lithium metal anodes have attracted considerable attention due to their potential to revolutionize rechargeable battery technology. With their high theoretical capacity and low electrode potential, LMAs promise to significantly improve energy density and battery performance. In particular, the development of solid-state batteries, which are expected to provide safer and more efficient energy storage, depends heavily on the effective incorporation of lithium metal anodes. A focus issue in the use of lithium metal is dendrite growth, which must be controlled for the safe application of battery technology. In this context, the use of a solid electrolyte is considered particularly promising. The production of lithium metal anodes is associated with considerable challenges that need to be overcome to unlock their full potential for mass production. [1,2]

## 2. Overview of production methods

Despite the potential benefits offered by lithium metal anodes, there are several challenges associated with their manufacturing and usage. For example, minor contaminants can increase lithium metal reactivity and trigger undesirable side reactions. Therefore, the processing of lithium metal anodes must occur in a dry atmosphere to prevent oxidation upon contact with air. Safety measures, including preventive fire protection, are recommended to mitigate handling risks with lithium. [3] Furthermore, lithium metal is highly adhesive, which poses challenges for roll-to-roll processes and handling tools. Special measures and supporting layers may be necessary to facilitate the processing of lithium metal anodes. [3] Additionally, uniform lithium metal deposition is crucial for future lithium metal batteries, with a required thickness range of 2–30  $\mu\text{m}$  for lithium anodes [4]. Non-uniform deposition impacts battery performance, making precise control and optimization of manufacturing processes essential in lithium metal electrode fabrication. [4] In addition to manufacturing challenges, addressing utilization and performance challenges during the usage phase is imperative. These challenges include dendrite formation, volume changes, and an unstable solid-electrolyte interphase (SEI) [5–9]. However, in this paper, the focus lies on the manufacturing process.

It is imperative to overcome these challenges to enable their mass production, improve performance, reduce costs, and enhance the overall quality of lithium metal anodes. Four production technologies are currently the focus of science to tackle these challenges.

### 2.1 Vapor-Based Techniques

Physical vapor deposition (PVD) techniques involve the deposition of thin films and coatings by transporting material from a condensed matter source through the gas phase onto a target surface. Unlike conventional ceramic processing methods that require high-temperature heat treatment or densification, PVD techniques offer robust and efficient alternatives [10]. The vapor deposition process has the potential to purify the lithium metal in addition to making thin lithium metal films [11]. To ensure process integrity and minimize potential sources of contamination, ultrahigh vacuum (UHV) conditions are established and maintained before and during the deposition process. PVD techniques provide a reliable and controlled approach for thin-film deposition, allowing for the fabrication of coatings with desirable properties in various applications. [1] Based on individual scientific studies, deposition rates of 1–1.5  $\mu\text{m/s}$  or correspondingly high dynamic deposition rates of 20–30  $\mu\text{m-m/min}$  are achievable for lithium. [12] PVD encompasses various methods, including electron beam evaporation, pulsed laser deposition, sputter deposition, and thermal evaporation. These techniques offer distinct advantages and are characterized by their specific operating principles and process parameters. [1,13]

#### Electron beam evaporation

Electron beam evaporation involves heating ( $T=2,000^\circ\text{C}$ ) a source material using an electron beam, which causes the material to vaporize and form a flux of atoms or molecules. [14] This vaporized material then condenses onto the substrate, forming a thin film. Electron beam evaporation offers precise control over high deposition rates and allows for the deposition of a wide range of materials. [1,14]

#### Pulsed Laser Deposition

Pulsed laser deposition utilizes a high-energy laser to ablate material from a target. The ablated material is then deposited onto the substrate to form a thin film. This technique offers excellent stoichiometric control, as well as the ability to deposit complex materials, such as oxides and nitrides, with high film quality. The process requires a deposition time of 1–4 h at pressures around  $2 \times 10^{-5}$  Pa. [1]

#### Sputter Deposition

Sputter deposition involves bombarding a target material with energetic ions, causing atoms or molecules to be ejected from the target surface. These ejected particles then deposit onto the substrate, forming a thin

film. Sputter deposition allows for precise control of film composition, thickness, and morphology, making it suitable for a wide range of materials and applications. The process promises high deposition rates and a temperature range between 100-500°C as well as pressures of  $10^{-7}$  –  $10^{-2}$  mbar. [1]

### **Thermal Evaporation**

Thermal evaporation relies on heating a source material to its vaporization temperature, allowing the atoms or molecules to escape and deposit onto the substrate. This technique is often used for materials with low melting points and offers simplicity and cost-effectiveness for thin film production. The application process takes place in a high vacuum. Thermal effects on the substrate can be excluded due to temperatures around room temperature. [1]

## **2.2 Liquid-based processing**

The liquid-based technique is a promising method for preparing lithium metal anodes for lithium metal batteries (LMBs). By utilizing the low melting point (180.5°C) of lithium, it can be easily transformed into a liquid state under anaerobic conditions and deposited onto a substrate or surface using methods like dip coating, spray coating, or doctor blading. However, the major technological challenge of this approach is the low wettability of melted lithium on various substrates due to its high surface energy. Regulating the wettability between liquid lithium and scaffold materials is crucial in this process. [1] The focus of this process is therefore specifically on the selection of the substrate, rather than the application process. Influencing parameters are accordingly the quality of the substrate, web speed, the temperature of the lithium, and, for instance in dip coating, the coating angle. [4]

## **2.3 Electrodeposition**

Electrodeposition, also known as electroplating, is a widely used technique for depositing metallic coatings onto a substrate via an electrochemical process. It involves the application of an electric current to drive the deposition of metal ions from a solution onto an electrode surface. [1] During electrodeposition, the substrate to be coated is immersed in an electrolyte solution containing metal ions of the desired coating material. An electric potential is then applied between the substrate (acting as the cathode) and a separate electrode (acting as the anode) in the electrolyte. This potential difference drives the reduction of metal ions at the cathode surface, leading to the formation and growth of a metallic layer. [1] Electrodeposition offers several advantages for coating production. It allows for precise control over coating thickness, uniformity, and adhesion strength. The process can be tailored to deposit a wide range of metals and alloys, providing versatility for various applications. Electrodeposition also enables the deposition of complex shapes and intricate geometries, making it suitable for coating objects with intricate surfaces. [15] To optimize electrodeposition, various parameters need to be carefully controlled, including the composition and concentration of the electrolyte, deposition current density, temperature, and deposition time. These parameters impact the coating quality, morphology, and physical properties. [16] Electrodeposition finds applications in diverse fields, including corrosion protection, decorative coatings, electronics, the automotive industry, and energy storage. By utilizing electrodeposition techniques, researchers and manufacturers can achieve tailored and controlled metallic coatings with desired properties, enhancing the functionality, durability, and aesthetics of various materials and components. [17,18,15]

## **2.4 Extrusion**

The most common lithium foils are produced by hydraulic extrusion of lithium ingots[19,20]. The lithium metal ingots are fed into the extruder, where they are compressed and forced through a slot-shaped exit cross-section. This extrusion step imparts the desired shape and dimensions to the lithium metal, transforming it into a continuous foil. After extrusion, the lithium foil undergoes mechanical rolling at controlled pressure and temperature to ensure homogeneity, surface roughness, and final film thickness. This refines surface texture and thickness uniformity, enhancing quality and performance.

The high-intensity process for lithium metal rolling is distinct from conventional techniques due to the nonporous nature of metallic lithium and the targeted thickness of 10-20  $\mu\text{m}$ . Polymeric supporting layers

help mitigate lithium's adhesive properties, allowing the production of thin, damage-free films. Commercially, this method is employed to produce limited quantities of 30-75  $\mu\text{m}$  thick lithium metal strips [21,22]. A post-production passivation coating safeguards the lithium foil, enabling processing in a dry room without the need for an inert gas atmosphere. It effectively prevents lithium reactivity, facilitating handling and integration into battery systems. [19]. Since pure lithium reacts with chromium(III) oxide, an oxide derived from the alloying element chromium in stainless steel, it is difficult to remove thin lithium from the rolls. [20] For smooth processing, use compatible rollers in extrusion and mechanical rolling. Plastic rollers, like polyacetal, are commonly used due to their compatibility with the adhesive nature of metallic lithium. [19] Due to the forming process and the resulting recrystallization, slip lines are created which would act as defects in electrochemical stripping/ plating. Depending on the technology and the manufacturer, lubricants are used that have to be removed later in the process. [23,20] Figure 1 shows schematically all previously described technologies.

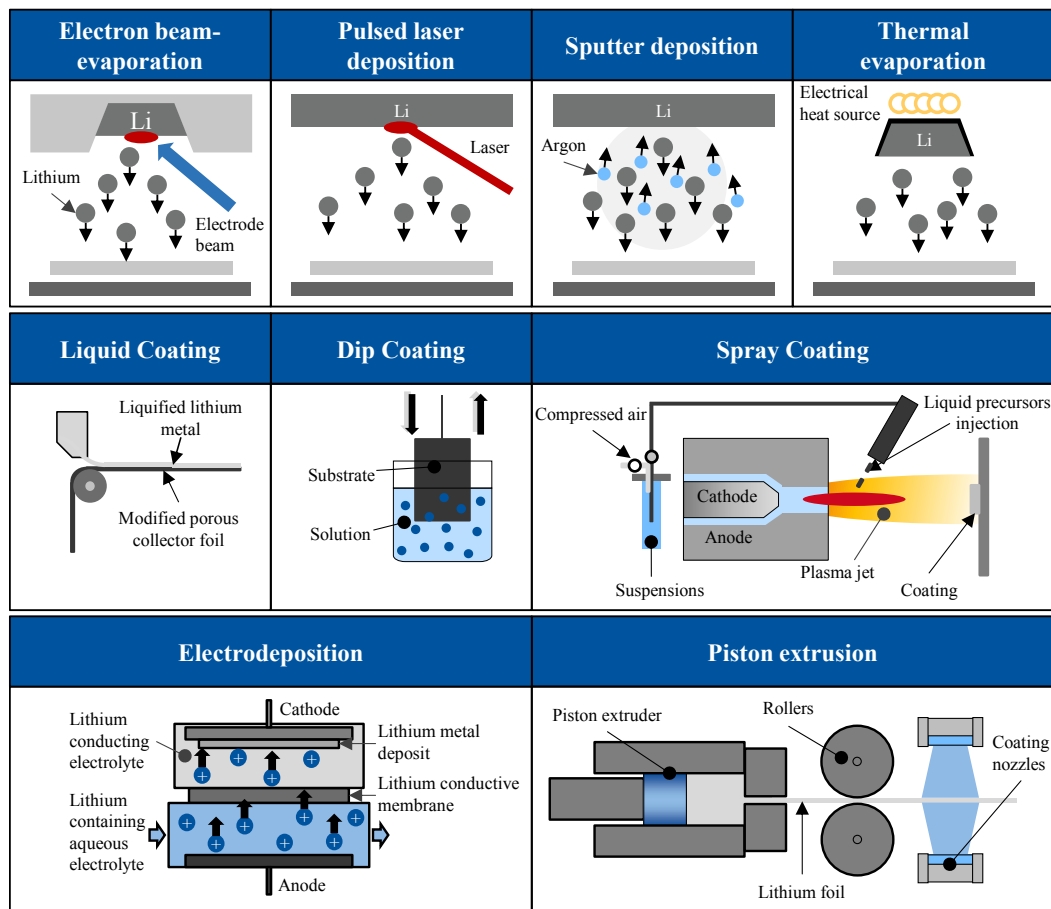


Figure 1: Schematic illustration of described coating processes

### 3. Methodology

Specific evaluation criteria have been established to evaluate selected production processes for the mass production of lithium metal anodes. In the style of the evaluation method of DEGEN & KRÄTZIG, evaluation parameters are selected based on which the selected processes are evaluated [24]. Due to the significant difference in the development stages and the limited research in this field of production technology, no conclusive validation can be made:

Technological maturity:

- *Technology readiness level:* The current stage of development and readiness for practical implementation of the production method [25].

Technology parameter:

- *Performance improvement potential*: The ability of the method to enhance the performance characteristics of lithium metal anodes, such as capacity, cycling stability, and rate capability.
- *Cost-effectiveness*: The economic feasibility of the production method, considering factors such as equipment costs, material costs, and process efficiency.
- *Quality enhancement*: The impact of the method on the quality and consistency of the produced lithium metal anodes, including factors such as uniformity, porosity, and surface morphology.

The benchmarking results employ a systematic evaluation approach, including the Technology Readiness Level (TRL) assessment on a scale of 1 to 9 [26,25]. This allows for a standardized assessment of the technological maturity of each method. This standardizes the assessment of technological maturity for each method. Additionally, performance improvement potential, cost-effectiveness, and quality enhancement are assessed using separate scales, carefully chosen for a comprehensive comparison. For instance, performance potential can be rated as lower than reference (-; 1 point), same as reference (0; 2 points), or higher than reference (+; 3 points). Cost-effectiveness considers equipment, material costs, and process efficiency, while quality enhancement evaluates factors like uniformity, porosity, and surface morphology. Using these evaluation scales enables a thorough and standardized comparison, clarifying the methods' relative strengths and weaknesses in technological readiness, performance improvement, cost-effectiveness, and quality enhancement.

The evaluation aims to identify potential production processes that could provide an alternative to the extrusion process in the future. In the following sections, the critical components and sub-processes are analyzed to provide recommendations for scalable production and increasing the TRL. To achieve further development of the manufacturing process, the current status and influencing parameters of the respective process must be identified. These are examined in more detail and defined based on parallels with existing production to define a methodological framework for equipment-side process and product optimization. [27]

#### 4. Results and discussion

The evaluation of the technology is presented in Tables 1-4 and is in each case qualitatively compared to the benchmark process of mechanical rolling following the extrusion process.

Table 1: Rating of production methods for lithium metal in terms of their TRL

Technology	TRL	Explanation
Extrusion	9	Technology is widely implemented and utilized on a large scale in various industries. [1]
Vapor-Based techniques	7	Development phase for scalable component production for lithium anodes. [12,28]
Electrode-position	4	Successful experiments conducted in research laboratories [29,30,15]
Liquid-based processing	5	Development phase for scalable component production for lithium anodes. [4]

Table 2: Rating of production methods for lithium metal in terms of their process performance potential

Technology	Rating	Explanation
Extrusion	<i>Ref.</i>	The piston extrusion method is widely used for mass-producing lithium anodes in current polymer solid-state batteries. It offers high efficiency, scalability, and significant benefits for fabrication. [31,32]

Vapor-Based techniques	+	Vapor-based PVD methods, such as sputtering and evaporation, hold potential for lithium metal foil production. They deposit purer and more reactive lithium compared to rolling and electrodeposition. These established processes allow broad coating, including roll-to-roll applications, on diverse substrates like alloys. [1,20]
Electrodeposition	-	Lithium electrodeposition has mixed process performance. It has strengths in its established use across industries and the ability to structure deposited films. However, its commercial availability is limited, and it is less commonly applied compared to other metals. [1,30,15]
Liquid-based processing	+	Lithium melt deposition shows promising process performance, creating thin lithium metal anodes with high utilization and precise coating control. Scalable roll-to-roll processing allows efficient high-volume production with variable dimensions. [1,4]

Table 3: Rating of production methods for lithium metal in terms of their economic efficiency

Technology	Rating	Explanation
Extrusion	<i>Ref.</i>	Piston extrusion for lithium metal foil is economically efficient, offering potential cost reduction benefits. Its ability to create thin, continuous foils with high material utilization and scalability contributes to its favorability. Further multistep calendaring leads to a total cost of about 250\$/kg. [31,33,34]
Vapor-Based techniques	-	Vapor-based techniques for lithium metal foil production may face economic challenges due to higher costs related to equipment, vacuum sealing, and processing thin foils. The vapor also deposits on the walls of the vacuum chamber, resulting in a waste of material. [1,31,12,4,20]
Electrodeposition	0	Electrodeposition for lithium metal foil production holds promise for lower costs due to lower process temperatures and raw material expenses. However, roll-to-roll processes may result in slower production rates and significant electrolyte recycling costs, impacting overall profitability. [1,31]
Liquid-based processing	+	Liquid-based processing for lithium metal foil production shows promising economic efficiency. High-quality and thickness-controlled layers on the copper current collector, achieved through ultrathin lithium metal films, lead to overall thickness reduction and higher energy densities on the cell level. Less subsequent operations after extrusion lead to cost savings. [1,33,4]

Table 4: Rating of production methods for lithium metal in terms of their quality potential

Technology	Rating	Explanation
Extrusion	<i>Ref.</i>	Piston extrusion can produce thin, continuous foils with attributes like homogeneity, uniformity, and precise thickness control. However, the process may also exhibit strong texture behavior due to shear stress. [11,34]
Vapor-Based techniques	+	Vapor-based processes such as thermal evaporation in PVD enable precise, defect-free coatings for efficient lithium metal foil production at lower temperatures. They can remove impurities from the lithium, although controlling the deposition rate remains a challenge. [11,4,34]
Electrodeposition	+	Electrodeposition techniques promise high-quality lithium metal foil with pure and homogeneous layers, allowing precise control over morphology and thickness. Despite challenges in achieving uniform morphologies and high

coulombic efficiencies, conformal coatings improve battery performance and adhesion. [1,35,15]

Liquid-based processing	0	Liquid-based processing offers promising quality potential for producing thin lithium metal foil. The low melting point of lithium allows an easy transition to the liquid state, enabling standard wet coating methods. However, previous attempts at melt deposition led to unsuitable film thicknesses and weak adhesion. Hence, the development of efficient and scalable coating methods in the desired thickness range is essential. [1,4]
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Figure 2 shows the cumulative evaluation of manufacturing processes based on their TRL. The analysis demonstrates the potential of the liquid application of lithium, offering opportunities for process optimization and further development to raise the TRL evaluation. To raise the TRL from level 5 to at least level 6 and optimize manufacturing technology, a conceptual design of a scalable approach is essential [36]. Achieving these results without changing the process necessitates a close examination of the sub-processes used.

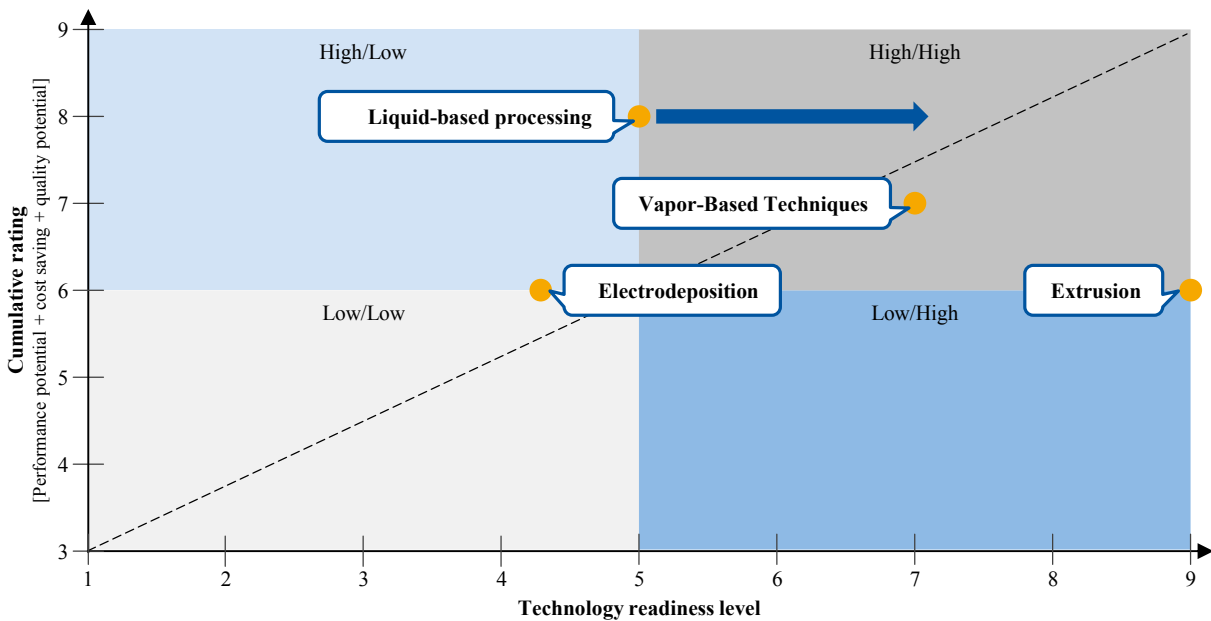


Figure 2: Attractiveness of lithium metal production methods in terms of TRL and overall cumulative rating.

The evaluation carried out shows that the application of liquid lithium has potential in the context of lithium metal anode production. SCHÖNHERR'S approach shows that there is a general feasibility, but there are still some challenges in industrializing this approach. For this purpose, it is necessary to take a closer look at the process with the individual technologies used and to identify limit values that must be considered during industrialization.

The current development status of liquid lithium deposition can be divided into individual sub-processes. In an upstream process, the surface is modified to produce a lithiophilic surface. Subsequently, liquid lithium is applied in a glovebox filled with argon. Within this inert mini-environment, the lithium metal anode is wound up and the substrate is implicitly tempered by dipping it into the heated lithium coating. The overall process can thus be divided into four sub-functions in which the control of the atmosphere is not initially considered due to the inconsistency over the entire process.

### Surface treatment

The lithiophilic layer is produced in SCHÖNHERR'S process using a muffle furnace. For the production of copper(I) oxide ( $\text{Cu}_2\text{O}$ ), temperatures of at least  $200^\circ\text{C}$  are necessary, as a result of which a chemical reaction with oxygen occurs. [37] When molten lithium interacts with the  $\text{Cu}_2\text{O}$  layer, it is consumed, resulting in

Li<sub>2</sub>O and Cu species that are no longer present as a compact film and are bound in the boundary region. The reaction is as follows:  $\text{Cu}_2\text{O} + 2 \text{Li} \leftrightarrow 2 \text{Cu} + \text{Li}_2\text{O}$  [38,4]. The Cu<sub>2</sub>O layer's formation depends on temperature and duration. Excessive energy input at elevated temperatures can lead to porosity and the formation of copper(II) oxide (CuO), observable from 320°C [38]. Delamination can occur at a layer thickness of 1500 nm, and CuO is less ideal for the interlayer formation. [39]. This suggests the reaction:  $\text{CuO} + 2 \text{Li} \leftrightarrow \text{Cu} + \text{Li}_2\text{O}$  [40].

To determine parameters for liquid application production, assume web speeds of 80 m/min based on the conventional and commercially used rolling process. [41–43,34] For an inline production of Cu<sub>2</sub>O substrate and to increase TRL, a layer with a thickness of several hundred nanometers, not exceeding 1500 nm, must be produced as quickly as possible. A temperature of approx. 320°C should not be exceeded in order to maintain the quality or to achieve no loss of quality compared to the current process. The substrate's quality is directly influencing the coating process. [44,4]

### Coating process

The liquid application coating process uses dip coating with lithium bath, bath heater, and drive as functional carriers in a glovebox filled with argon, primarily to avoid reactions with atmospheric humidity. Under normal conditions, lithium reacts slowly with air. It can be assumed that a dry air atmosphere has less effect on the lithium metal anode in an accelerated manufacturing process. [36] The process's physical conditions depend on web speed and temperature-dependent rheological properties of lithium. [27,45,36,44] At the melting point of about 180.5°C, lithium has a viscosity of  $\eta = 0.645$  mPas, a density of  $\rho = 516$  kg/m<sup>3</sup> and a surface tension of  $\sigma = 0.396$  N/m<sup>2</sup>. [46] The coating thickness (h) as a function of web speed (U) is determined by the following equation [47]:

$$h = 0.944 * \frac{\mu U^{\frac{1}{6}}}{\sigma} * \frac{\mu U^{\frac{1}{2}}}{\rho g}$$

The coating's inclination was not considered due to uneven thickness distribution in case of coating on both sides. The equation represents web speed, and with a targeted thickness of around 20 μm, the maximum web speed is  $U = 4.3$  m/min. At a production speed of 80 m/min, this results in a coating thickness of 140 μm. Dip-coating is not suitable for scaled consideration; therefore, alternative coating technologies like slot dies must be taken into account. Slot dies have easily scalable process parameters, with slurry viscosity ranging from 1 mPas to multiple kPas and surface tension from less than 25 mPas to more than 500 mPas, both matching liquid lithium requirements. [48] According to KRAYTSBERG et al. a coating with a slot die is considered stable if the ratio of the distance of the slot die (h) in relation to the wet film thickness (t) meets the following requirement:

$$\frac{h}{t} \leq 1.49 * \frac{\mu U^{-\frac{2}{3}}}{\sigma}$$

At a web speed of 80 m/min and lithium at the melting point, the wet film thickness requires a minimum spacing of about  $h = 1.7$  mm. [48,49] In summary, increasing quality can be achieved through controlled substrate treatment and varying the application system, leading to a relatively simple potential increase in TRL. [50]

## 5. Outlook and conclusion

This paper discusses current manufacturing processes and approaches for lithium metal anode production and evaluates and classifies them on the TRL scale. A cumulative rating, was determined, considering process performance, economics and quality potential, with the liquid-based processing application receiving the highest rating but having a relatively low TRL. To optimize equipment, a product structure analysis is conducted. Relevant process parameters are presented as framework conditions for product development and process improvement. An alternative coating technology is proposed after a critical



analysis of the coating process. Detailed system design and development based on liquid lithium application offer a promising alternative and can enhance quality through controlled parameters. This paper serves as a basis for further development and accelerated progress in lithium metal anode production, which is crucial for future generations of battery cells.

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