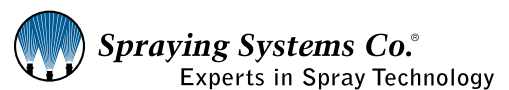


WHITE PAPER

# ELECTROSTATIC SPRAY DRYING:

A SCALABLE PLATFORM FOR ADVANCED MATERIAL  
MANUFACTURING IN ENERGY STORAGE



### AUTHORS

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### ABSTRACT

Electrostatic spray drying (ESD) integrates an applied electric field with conventional spray drying to control droplet dynamics and particle charging, offering a route to conformal, polymer-based surface modification of active battery materials at scale. Here we demonstrate ESD of silicon nanoparticles (SiNPs) using carboxymethyl cellulose (CMC) as a model coating to mitigate Si volume-change-induced degradation. We systematically vary voltage (0 vs -15 V), atomizing gas pressure (100 vs 300 kPa), and feed composition (CMC:Si = 1:1, 1:2, 1:5 w/w) and evaluate coating outcomes by SEM and EDS. Sodium in CMC serves as a tracer, enabling a Si:Na atomic ratio proxy for relative coating thickness/coverage. We find that -15 V with 300 kPa yields the most conformal, continuous coatings, even at the 1:5 CMC:Si ratio, while higher atomizing pressure produces finer droplets that favor uniform shells with thinner average coatings (higher Si:Na). These results indicate that ESD parameters can be tuned to balance coating uniformity and thickness, providing a scalable pathway to interface engineered anode powders.

**Keywords:** electrostatic spray drying; silicon anode; polymer coating; carboxymethyl cellulose; battery materials processing

## 1. INTRODUCTION

Spray drying has emerged as a versatile and scalable technique to produce engineered powders across a range of energy storage applications.<sup>1,2</sup> In battery materials synthesis, it has been widely employed to produce spherical secondary particles, control surface properties, and fabricate composite structures with tailored porosity and morphology.<sup>3</sup> Its one-step nature, compatibility with aqueous or organic solvents, and tunability of particle characteristics make spray drying especially attractive for industrial-scale processing of active materials, conductive additives, and polymer binders. Cathode materials such as lithium nickel manganese cobalt oxides (NMC), lithium iron phosphate (LFP), and lithium cobalt oxide (LCO) are also processed via spray drying to enhance particle uniformity, tap density, and surface area.<sup>4-6</sup> Similarly, for anode systems, spray drying has been used to structure composites to improve mechanical integrity and electrochemical stability.

While conventional spray drying relies solely on thermal evaporation for droplet-to-particle transformation, recent advances have integrated electrostatic fields into the process to enhance droplet dispersion, improve coating uniformity, and reduce agglomeration. This paper showcases electrostatic spray drying (ESD), an advanced technique that introduces applied voltage as an additional process input, which may influence particle morphology and surface characteristics beyond what is typically achievable with conventional spray drying. Electrostatic spray drying (ESD) is a scalable, single-step process that combines the principles of conventional spray drying with electrostatic forces to enhance particle control and coating uniformity. In ESD, a solution or suspension is atomized into fine droplets using a nozzle while an electrostatic force imparts a charge to the droplets as they are formed, which improves the drying efficiency during the process, enabling enhanced dispersion, and finer particle deposition. Unlike traditional spray drying, which relies solely on thermal evaporation, ESD provides several unique advantages for nanomaterial processing such as:

- **Improved coating uniformity:** Electrostatic forces can guide charged droplets to form conformal coatings around nanoparticles, even at low polymer loadings.
- **Processing temperatures:** ESD allows for drying at controlled temperatures, preserving the structural and chemical integrity of heat-sensitive active materials or polymer additives.
- **Tunable morphologies:** By varying operational parameters such as applied voltage, feed composition, and atomizing gas pressure, a range of morphologies can be achieved.



**Figure 1. PolarDry® 0.1**

This equipment was used for electrostatic spray drying of the materials shown and discussed in this white paper.

## 2. CONDITIONING ACTIVE BATTERY MATERIALS

The electrochemical performance of next-generation batteries depends critically on the morphological and interfacial properties of the active materials. Conditioning these materials through surface functionalization, morphology, or composite formation can significantly enhance cycling stability, rate performance, and interface compatibility. In this work, the effectiveness of conditioning active materials is investigated, and its impact on the performance metrics, including:

<b>Active Material Property Modified by Conditioning</b>	<b>Electrochemical Impact</b>
<b>Core-shell structuring</b>	Suppressed volume expansion (e.g., in Si, Sn, Sb)
<b>Conductive additive integration</b>	Improved percolation network → higher electronic conductivity
<b>Ionic conductivity modifiers</b>	Enhanced Li <sup>+</sup> transport at electrode-electrolyte interfaces
<b>Particle dispersion and uniformity</b>	Increased packing density and rate capability
<b>Surface area</b>	Enhanced active material utilization resulting in improved capacity

PolaDry's unique ESD process can provide innovative answers to mitigating issues that are often associated with advanced materials. In this work, the effectiveness of ESD on conditioning active materials is investigated focusing on its ability to tailor surface morphology and particle structure resulting in improved key electrochemical performance metrics.

### 3. CASE STUDY

#### COATING SILICON NANOPARTICLES FOR HIGH-PERFORMANCE SILICON ANODES IN A LITHIUM-ION BATTERY

This case study demonstrates how Electrostatic Spray Drying (ESD) was used to produce **polymer-coated silicon nanoparticles** with tunable architectures designed to improve cycling stability and electrode integrity.

Silicon is a leading anode candidate for next-generation lithium-ion batteries, owing to its exceptionally high theoretical specific capacity (~3579 mAh/g). However, its practical application is limited by its large volume expansion (~300%) during lithiation, which leads to pulverization, unstable solid electrolyte interphase (SEI) formation, and rapid capacity fade. Addressing this challenge requires precise engineering of particle surfaces and interfaces.

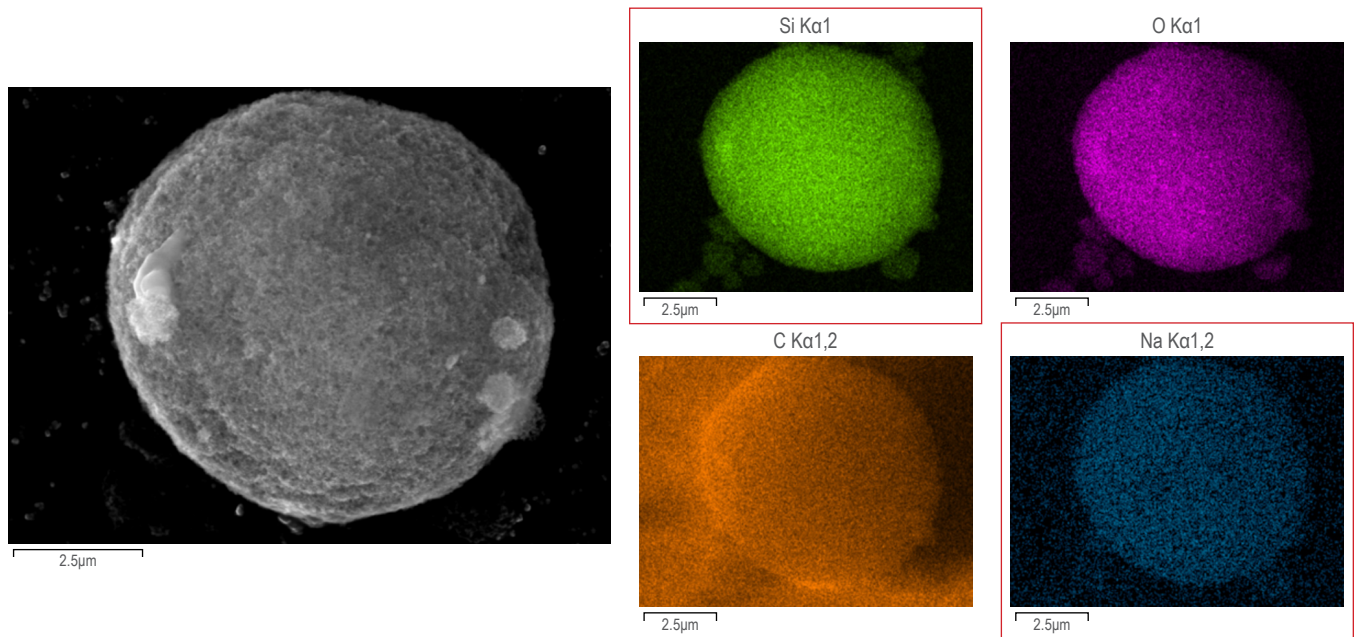
To investigate the effectiveness of electrostatic spray drying (ESD) for conditioning silicon nanoparticles, a series of formulations were prepared with varying polymer-to-particle mass ratios using carboxymethyl cellulose (CMC) as the coating agent.<sup>7,8</sup> CMC is a widely used binder in silicon anodes as it improves mechanical integrity and accommodates volume expansion during lithiation.<sup>9-11</sup> Three feedstock compositions were studied corresponding to CMC:Si weight ratios of 1:1, 1:2, and 1:5, each dispersed in deionized water. These suspensions were atomized and dried using an electrostatic spray dryer under systematically varied parameters and the feed rate, nozzle configuration, and drying temperature were optimized to ensure droplet stability and polymer integrity.

#### 3.1 RESULTS

To evaluate the influence of electrostatic spray drying (ESD) conditions on coating performance, the coated silicon nanoparticles were characterized using scanning electron microscopy (SEM) and energy-dispersive X-ray spectroscopy (EDS). SEM imaging was used to assess particle size, surface texture, and coating morphology across varying CMC:Si ratios and process parameters. EDS mapping enabled the identification of elemental distributions, with sodium (Na) serving as a tracer for the CMC polymer. Further, to quantify the coating, Si:Na atomic ratio, derived from EDS spectra, was used as a proxy for coating thickness and coverage. These complementary techniques provided a comprehensive understanding of how applied voltage and atomizing gas pressure impacted coating formation and uniformity. Step by step analysis is provided in the sections below.

### 3.1.1 COATING CONFIRMATION VIA SEM-EDS MAPPING

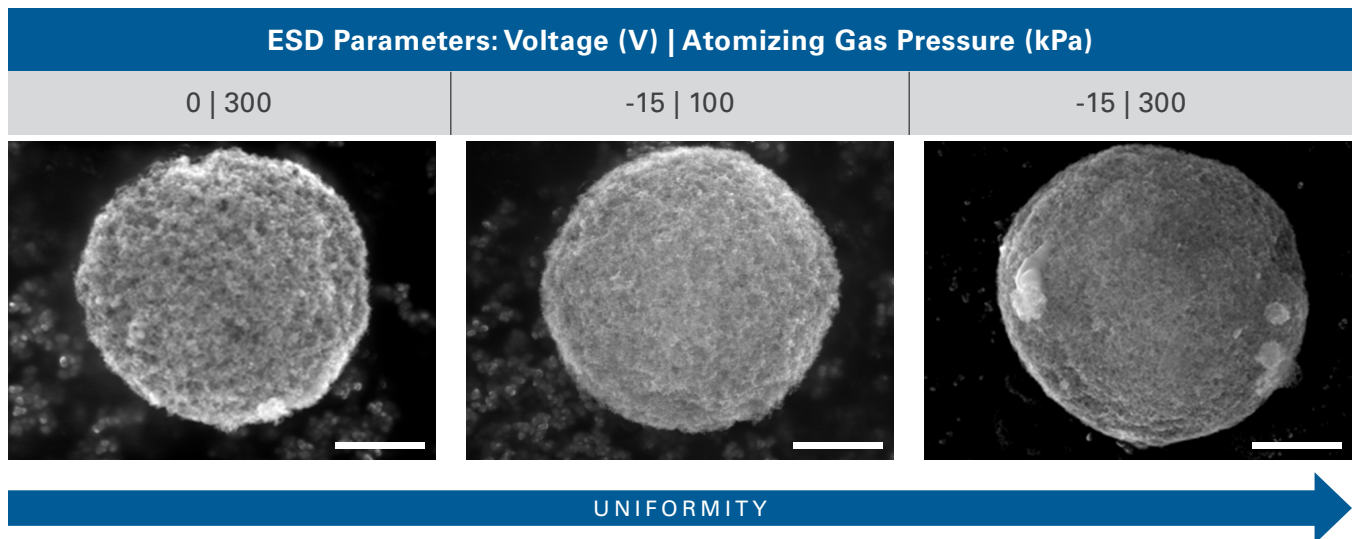
Figure 2 shows a coated silicon nanoparticle obtained after electrostatic spray drying (ESD). Elemental mapping further supports the presence of a conformal CMC shell. The silicon (Si) signal is concentrated within the core, while signals corresponding to carbon (C), oxygen (O), and sodium (Na) components of the CMC polymer are distributed across the particle surface. In particular, the Na serves as a distinctive marker of CMC, as sodium is not present anywhere except the inherent polymer's carboxymethyl functional groups.



**Figure 2.** SEM-EDS elemental mapping of a silicon nanoparticle coated with carboxymethyl cellulose (CMC) via electrostatic spray drying. The SEM image (left) shows a spherical morphology with a smooth surface indicative of uniform coating. Elemental maps (right) confirm the coating: Si is concentrated in the core, while Na, a dominant characteristic of CMC, is uniformly distributed over the particle.

### 3.1.2 COATING MORPHOLOGY ANALYSIS UNDER DIFFERENT ESD CONDITIONS

To assess the influence of electrostatic spray drying (ESD) parameters on coating morphology, silicon nanoparticles were processed under distinct conditions, two voltages (0 V and -15 V) with two atomizing gas pressures (100 kPa, low and 300 kPa, high), across three CMC:Si feed ratios (1:1, 1:2, and 1:5). Representative SEM images for each condition for 1:5 ratio are presented in Figure 3.



**Figure 3.** Representative SEM images of coated nanoparticles at distinct electrostatic spray drying conditions (voltage | atomizing gas pressure) for 1:5 ratio. Coating uniformity improves significantly with application of voltage and higher atomizing pressure, with -15 V | 300 kPa condition producing the most conformal and smooth shells.

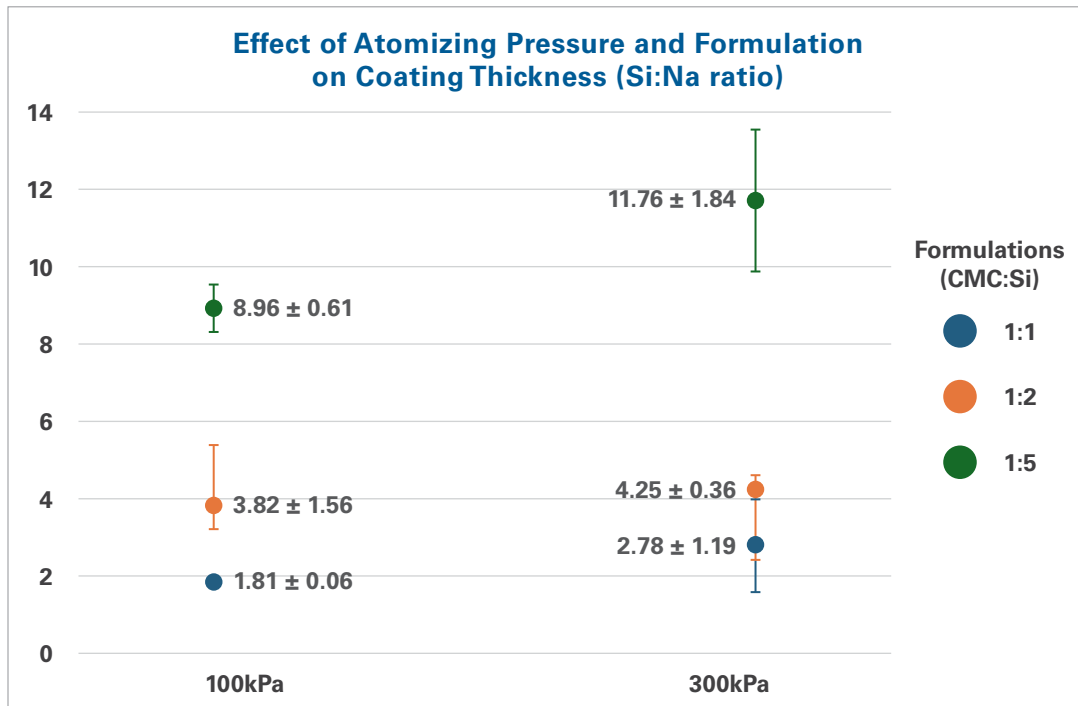
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Particles processed without an applied voltage (0 V) and at high atomizing pressure (300 kPa) showed irregular coatings across all ratios. These samples exhibited rough surfaces, indicating insufficient deposition and poor coating adhesion, suggesting that high atomizing force alone is insufficient for forming conformal coatings without electrostatic assistance. Samples processed under -15 V with low atomizing pressure (100 kPa) showed improved coating coverage but still showed rough surfaces which is likely due to larger droplet formation and slower evaporation kinetics at low pressure, which again led to non-uniform shell morphologies. The most conformal and continuous coatings were observed under -15 V with 300 kPa pressure. These conditions formed finer droplets with directed deposition, resulting in uniform, smooth shells with minimal agglomeration or surface defects. Even at the lowest polymer loading (1:5), this condition yielded uniform surface coverage, indicating enhanced coating efficiency driven by the synergy of electrostatic forces and high atomization energy.

These observations highlight the critical role of both electrostatic field and atomizing pressure in determining final coating morphology, with high-voltage and high-pressure conditions offering the most effective route for producing uniform, application-ready coated particles.

### 3.1.3 QUANTITATIVE ANALYSIS OF COATING COVERAGE

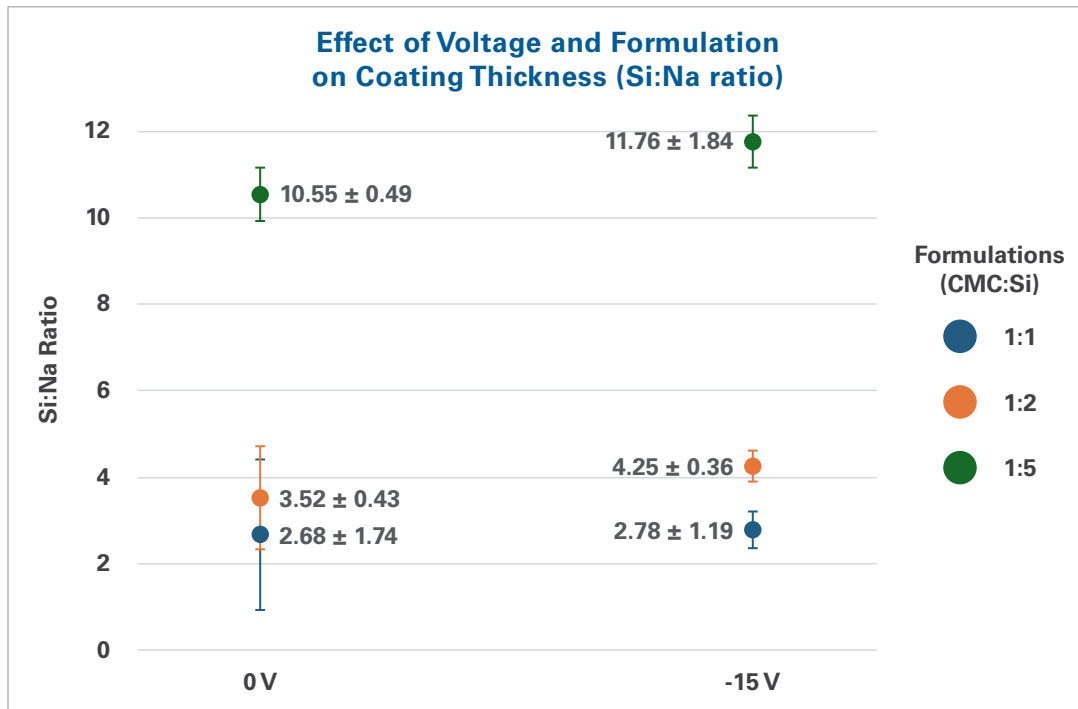
To quantitatively evaluate the extent of CMC coating across electrostatic spray drying (ESD) conditions, energy-dispersive X-ray spectroscopy (EDS) was performed on coated silicon nanoparticles. Weight percentages of key elements (Si, O, Na, and C) were extracted from the spectra, and the weight percentages of Si and Na were used to evaluate coating thickness, where sodium marked CMC presence and the Si:Na ratio quantified coverage. Since most carbon weight percentage comprises the carbon tape used to prepare the sample, it is not used for any analysis. Additionally, EDS elemental mapping confirmed the presence of sodium on the particle surfaces in all samples, indicating that coating was achieved even in cases with high Si:Na ratios. Therefore, the Si:Na ratio serves as a reliable indicator of relative coating thickness.



**Figure 4.** Effect of atomizing gas pressure at constant voltage (-15V) on Si:Na atomic ratios across different CMC:Si feed ratios.

Figure 4 shows the Si:Na atomic ratios, derived from EDS spectra, as a function of CMC:Si feed ratio under two electrostatic spray drying (ESD) conditions differing in atomizing gas pressure. Across all formulations (1:1, 1:2, 1:5), the Si:Na ratio increases with decreasing CMC content, indicating progressively thinner polymer coatings at lower feed concentrations. Notably, the low-pressure condition consistently resulted in lower Si:Na ratios, than the high-pressure condition. This change can be attributed to enhanced atomization at higher pressure, which produces finer droplets that promote more homogeneous dispersion and coating coverage, even when polymer availability is limited.

However, due to the reduced droplet size and faster solvent evaporation, the resulting coating is thinner, as reflected in the higher Si:Na ratios. In contrast, lower atomizing pressure leads to larger droplets and slower drying, allowing for thicker deposition but often resulting in uneven surface coverage.



**Figure 5.** Effect of applied voltage at constant pressure (300 kPa) on Si:Na atomic ratios across different CMC:Si feed ratios.

Figure 5 illustrates the effect of applied voltage on the Si:Na ratio at a constant atomizing gas pressure of 300 kPa. Across all CMC:Si feed ratios, a moderate increase in Si:Na is observed upon applying -15 V relative to 0 V. This trend suggests that the electrostatic field may influence the distribution and deposition of CMC, resulting in thinner but potentially more conformal coatings. The increased Si:Na ratios under applied voltage indicate a reduction in relative coating thickness, consistent with EDS mapping observations. At lower CMC loadings (1:5), where coating uniformity becomes more sensitive to process parameters, the distinction between voltage conditions remains statistically significant, further supporting the influence of electrostatic forces on coating efficiency.

Together with pressure-dependent results from Figure 4, these findings demonstrate that both atomizing gas pressure and applied voltage systematically modulate the ESD coating process. ESD enables fine-tuning of coating thickness and surface composition through independent adjustment of formulation and process inputs, with reproducible results across conditions.

## 4. CONCLUSION AND FUTURE WORK

This study demonstrates a stepping stone to a series of studies highlighting the benefits of ESD via PolarDry® technology to create uniformly coated active materials for silicon. PolarDry® is an effective and scalable method for coating silicon nanoparticles with polymer layers for battery applications. By tuning voltage, atomizing gas pressure, and CMC:Si feed ratio, the process yielded conformal coatings with controllable thickness, confirmed through SEM and EDS analysis. Compared to conventional spray drying or multi-step coating techniques, PolarDry® offers key advantages by providing enhanced coating uniformity and compatibility with sensitive materials. Importantly, the technology is scalable available in benchtop (Model 0.1), R&D (Model 001/004), pilot (Model 004/032), and full-scale production systems (Model 032/050), allowing direct translation of lab-scale results to manufacturing environments positioning PolarDry® as a versatile solution for advanced battery material development. These attributes make it particularly suitable for advanced battery materials where effective interfacial engineering is critical. While silicon was used as a model system, the approach is broadly applicable to other anode and cathode materials.

The results support PolarDry's potential as a high-precision, scalable platform for commercial powder coating in energy storage. Future studies will include different battery materials with electrochemical characterization testing using coin cells. While this work demonstrates proof of concept at the single-particle level, additional research will be needed to assess batch-level particle size distribution and post-processing behavior, which are critical for real-world applicability. Following this, material testing in full electrochemical cells will be required to correlate coating quality with performance improvements.

## 5. ACKNOWLEDGMENTS

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## REFERENCES

1. Vertruyen, B. *et al.* Spray-Drying of Electrode Materials for Lithium- and Sodium-Ion Batteries. *Materials* **11**, 1076 (2018).
2. Nandiyanto, A. B. D. & Okuyama, K. Progress in developing spray-drying methods for the production of controlled morphology particles: From the nanometer to submicrometer size ranges. *Advanced Powder Technology* **22**, 1–19 (2011).
3. Stunda-Zujeva, A., Irbe, Z. & Berzina-Cimdina, L. Controlling the morphology of ceramic and composite powders obtained via spray drying – A review. *Ceram Int* **43**, 11543–11551 (2017).
4. Li, Y., Wan, C., Wu, Y., Jiang, C. & Zhu, Y. Synthesis and characterization of ultrafine LiCoO<sub>2</sub> powders by a spray-drying method. *J Power Sources* **85**, 294–298 (2000).
5. Rigamonti, M. G. *et al.* Influence of atomization conditions on spray drying lithium iron phosphate nanoparticle suspensions. *Can J Chem Eng* **97**, 2251–2258 (2019).
6. Ihalainen, M. *et al.* Synthesis of solid NMC622 particles by spray drying, post-annealing and lithiation. *Advanced Powder Technology* **34**, 104187 (2023).
7. Nguyen, C. C., Yoon, T., Seo, D. M., Guduru, P. & Lucht, B. L. Systematic Investigation of Binders for Silicon Anodes: Interactions of Binder with Silicon Particles and Electrolytes and Effects of Binders on Solid Electrolyte Interphase Formation. *ACS Appl Mater Interfaces* **8**, 12211–12220 (2016).
8. Ding, N. *et al.* Improvement of cyclability of Si as anode for Li-ion batteries. *J Power Sources* **192**, 644–651 (2009).
9. WANG, X. *et al.* Influence of Degree of Substitution of Carboxymethyl Cellulose on High Performance Silicon Anode in Lithium-Ion Batteries. *Electrochemistry* **87**, 94–99 (2019).
10. Ndour, M. *et al.* The formulation of a CMC binder/silicon composite anode for Li-ion batteries: from molecular effects of ball milling on polymer chains to consequences on electrochemical performances. *Mater Adv* **3**, 8522–8533 (2022).
11. LESTRIEZ, B., BAHRI, S., SANDU, I., ROUE, L. & GUYOMARD, D. On the binding mechanism of CMC in Si negative electrodes for Li-ion batteries. *Electrochem Commun* **9**, 2801–2806 (2007).