



Modeling and analysis of solvent removal during Li-ion battery electrode drying



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HIGHLIGHTS

- Drying of lithium-ion battery electrode is studied using continuum modeling approach.
- Studied the effect of temperature and air velocity on electrode drying.
- Water dries ~4.5 times faster and requires ~10 times less energy as compared to NMP.
- At least 50% of the overall energy demand can be reduced using a multi-zone drier.

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ABSTRACT

In this work, we study the design aspects and process dynamics of solvent removal from Lithium-ion battery electrode coatings. For this, we use a continuum level mathematical model to describe the physical phenomenon of cathode drying involving coupled simultaneous heat and mass transfer with phase change. Our results indicate that around 90% of solvent is removed in less than half of the overall drying time. We study the effect of varying temperature and air velocity on the drying process. We show that the overall drying energy can be reduced by at least 50% by using a multi-zone drying process. Also, the peak solvent flux can be reduced by at least 40%. We further present the effect of using an aqueous solvent instead of *N*-Methyl-2-pyrrolidone (NMP) in electrode drying. Our results indicate that Water dries nearly 4.5 times faster as compared to NMP and requires nearly 10 times less overall drying energy per kg of solvent.

1. Introduction

Lithium-ion batteries are becoming increasingly popular for their use in a variety of consumer applications and especially, in hybrid and electric vehicles. Consequently, the development of low cost and high energy density lithium-ion batteries is receiving ever greater interest from both academia and industry. The research effort in developing efficient and affordable batteries can be broadly classified into three main areas of interest. They are increasing energy density, ensuring operational safety, and reducing cost. These objectives are mainly addressed in the studies developing new active materials and benchmarking process control practices. In this regard, optimization of battery manufacturing operations and the in-depth understanding of underlying physical/chemical processes have a great potential to offer crucial insights in addressing some of the prevalent problems in battery research, especially in improving quality and cost.

One of the major components of the overall lithium-ion battery cost

is the processing cost of an electrode [1]. The physical properties of an electrode coating, such as coating structure and the distribution of material components within the coating layer (including active materials, additives, and polymer binders), greatly influence its electrochemical performance [2–6]. The production of an electrode involves a series of steps including mixing of materials, coating of material-solvent slurry on a current collector, drying of electrode coatings, and then calendaring of dried coatings. A number of publications [4,5] study the impact of slurry mixing on the quality of electrodes. Also, some studies address the effect of calendaring [3,7] and coating methods [8,9] on the electrochemical performance of electrodes. However, studies focusing on the analysis of drying process and its impact on the cost [10] and quality of an electrode are limited.

The process of drying has a significant impact on: (i) the distribution of materials within the coating due to the interplay of various forces (such as capillary action, diffusion, and convection), and (ii) the structure of the porous coatings. In addition, drying as a unit operation

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is inherently slow, expensive, and energy intensive [10]. Thus, the study of drying process in electrode coatings is of great importance (i) to understand the interplay of mass transport and heat transfer processes, particularly if they can be experimentally validated and correlated to electrode quality, and (ii) to extract insights on improving the processing speed and energy demand. Consequently, the study of electrode drying and its effect on electrode quality (and cost) is receiving an increasing attention from the research community [1,10,11].

Drying of coatings in general has been studied for several decades in a variety of industries including paper, paint, textile, food, agriculture, wood, ceramics, pharmaceutical products, and wastewater sludge. A variety of mathematical models have been proposed to explain the process and phases of drying [2,12–14]. Over the last few decades, the concept of simultaneous heat and mass transfer processes has been widely adopted to explain drying in various coatings or materials. Defraeye [13] presents a comprehensive account of various approaches and models proposed so far to study drying in a variety of systems. The drying of porous coatings and its impact on the coating properties has been extensively studied, particularly, in the paper industry [12,13,15,16]. In contrast, similar research efforts in electrode coatings with application to lithium-ion batteries are limited. Although, both of these applications have similarity, there are significant differences that require them to be studied separately. However, findings from one can very well be adopted as a basis to develop models for the other [17]. Gutoff [18,19] proposed a series of simple models considering simultaneous heat and mass transfer for drying of coatings on continuous coated webs. He studied the rate of drying and temperature profile in the coating through an iterative algorithm implemented on an excel worksheet. Kim and Rhee [20] presented the effect of using near infrared (NIR) drying method instead of the conventional oven drying on the electrochemical properties of cathodes. The study of binder particle re-distribution within the coating layer during drying of electrode has attracted a significant attention in the literature [17,21–24]. This is because the distribution profile of binder particles within coating layer has a significant impact on the quality of coating [25]. Li et al. [25] studied the distribution of binder, Polyvinylidene fluoride (PVDF), in LiCoO_2 based coatings in aqueous and organic [N-methyl-2-pyrrolidone (NMP)] mediums during drying to highlight its impact on the electrochemical performance of the resulting electrode. Lim et al. [26] studied the development of stress during the drying of anodes coated with the graphite and sodium carboxymethyl cellulose (CMC) or styrene butadiene rubber (SBR) slurry. Westphal et al. [27] showed the effect of process parameters such as temperature and nozzle speed on the elasticity, electrical volume resistivity and adhesion strength between electrode coating and current collector. Similarly, Baunach et al. [2] studied the impact of drying rate on the coating adhesion of a graphite based anode. They highlighted that the quality of electrode can be controlled by solving an optimization problem with a tradeoff between drying rate and cost. Jaiser et al. [14] studied the phenomenon of film solidification in lithium-ion battery anodes. They investigated the impact of slurry conditions and process parameters during drying on the adhesion of coated film on the current collector. Jaiser et al. [28] proposed a strategy to reduce time for drying time and maintain the quality of the coating through a 3-stage drying process. They vary the rate of drying in the three stages by varying temperature and convective flow of moisture carrier. They defined characteristic points for changing the drying rates based on the amount of moisture present in the coating. Li et al. [29] investigated the impact of residual moisture after drying on the electrochemical performance of electrodes. They compared the performance of a $\text{LiNi}_{0.5}\text{Mn}_{0.3}\text{Co}_{0.2}\text{O}_2$ (NMC532) cathode in both aqueous and NMP solvent after drying. They showed that the residual moisture in both the cases are comparable and in general lower residual moisture favored coulombic efficiency in the first cycle. Schunemann et al. [30] identified that apart from the material costs, drying contributes the maximum cost to the overall cost of a Li-ion based cell and showed the potential savings in cost by increasing the solids content (or reducing solvent) in the slurry making process. They further highlighted drying is also a very high energy consuming step in the battery

manufacturing process. Ahmed et al. [10] developed a process model to estimate the cost and energy demand of drying the cathode and recovering the solvent (NMP). They showed that the energy demand for drying is over 10 kWh per kg of NMP vaporized for the given plant capacity, which is about 45 times in excess to the actual heat required for vaporization of equivalent solvent. Also, the authors estimated that the cost of drying and recovery of solvent amount to about 3.4% of the overall battery cost.

The aforementioned review of existing literature highlights an increasing interest in studying the drying of electrodes for lithium-ion battery application, especially, in the last couple of years. However, most of these publications [14,17,23,24,28] focus on studying the change in binder concentration profile within the coating layer during drying and then associating this phenomenon to the quality of resulting electrode. Other publications [1,10,11] have focused on the energy demand and economic aspects of drying. Clearly, few works focus on studying the process of electrode drying by understanding the mass transport and heat transfer processes within the coating layer. Also, the effect of different solvents (aqueous and organic) on drying process has not been studied. This forms the primary focus of our work.

In this paper, we develop a mathematical model to study and understand the physical processes such as mass transport and heat transfer during drying of Li-ion battery electrode coating. We present a set of cases to study solvent drying in electrode coatings during the manufacturing process of battery electrodes. We further present an analysis on the effect of various (process and system) parameters on the drying of electrodes. We then discuss potential causes that can lead to quality defects and present an analysis of strategies to mitigate such issues. Finally, we study the effect of using aqueous versus organic solvents on drying.

2. Drying process model

The drying of porous electrode coatings is inherently very complex and involves simultaneous heat and mass transfers in all three phases (solid, liquid, and gas) represented by transport equations based on conservation laws for mass and energy. We follow the volume averaging technique proposed by Whitaker [31,32] and popularized by his co-workers [33] and others [12,18,33–35] to develop a macro-scale model for drying. Here, the porous material is assumed to be in a continuum and the dependent variable (such as liquid phase density or gas phase density) is averaged over a small representative volume consisting of all three phases [34,35]. Now, every point in the coating layer is assumed to be the center of such a representative volume, which can change composition (for liquid and gas phases) based on space and time. The volume averaging technique and the derivation of the transport equations for drying have been presented by a number of articles in the literature [31,32].

An essential factor in modeling is to understand the process conditions where the formulated equations and assumptions are physically valid. In other words, it is essential to define the scope of a mathematical model with respect to process/physical conditions of the system (the drying material). In general, the drying literature has classified drying operation into a number of sub-categories or regimes based on solvent concentration, drying rate, and moisture content. The process of drying porous coatings and solids, typically, consists of two phases. In the first phase, the solid particles are suspended in a pool of solvent and drying is mainly a surface phenomenon. Here, the rate of solvent drying is dependent only on the rate of evaporation at the surface of solvent-solid reservoir. As long as the process conditions remain constant, the rate of drying in this phase remains constant and so, is also referred to as the constant rate period. This first drying phase concludes as the solid particles come closer to form a porous structure. This porous structure is completely saturated with the liquid phase solvent. As drying proceeds the porous structure is no longer saturated with liquid phase solvent alone as the pore volume is now filled with both vapor phase and liquid phase, as shown in Fig. 1. At this time, the rate of drying is governed by the complex interplay of multiple physical phenomena within the porous electrode structure involving solvent

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