



Comparative life cycle assessment of laminated and vacuum vapor-deposited thin film solid-state batteries

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ABSTRACT

Lamination technologies used to produce lithium ion batteries are limited by the capital investment needed for multiple unit operations, solvent use for electrolyte and electrode slurries, and an inability to exert control over active material particle morphology and homogeneity. Advancements in thin-film solid-state processing using vacuum coating hold promise to overcome these challenges for batteries with superior energy density and cycle life, if cost and scalability issues can be overcome. A comparative life cycle assessment is reported for battery production using lamination and thin-film vacuum vapor deposition. Lithium vanadium oxide solid-state cells are found to have the lowest impact, per unit energy storage, in cumulative energy demand (CED), global warming potential (GWP), and six other midpoint environmental indicators. Human health and resource depletion impacts are generally higher for lithium manganese oxide and lithium cobalt oxide solid-state cells than for their laminated counterparts, whereas CED and GWP per unit energy storage are 25–65% lower for solid-state cells across all cathode chemistries. Sensitivity analysis, taking into account uncertainties related to solid-state cell properties and vacuum vapor deposition process efficiencies, indicates that CED and GWP impacts for battery electric vehicle mobility using packs with solid-state cells will be lower than those incurred using laminated cells.

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1. Introduction

Lithium ion batteries (LIBs) are displacing other battery chemistries as the preferred energy storage medium for consumer electronics and electric vehicles. The global market for rechargeable lithium ion batteries is expected to grow robustly in the next decade, with one recent forecast projecting more than a sevenfold increase in the market for LIBs in light duty consumer vehicles from \$3.2 billion in 2013 to \$24.1 billion in 2023 (Alexander and Gartner, 2014), and another analysis anticipating a nearly threefold expansion in the LIB market across all consumer sectors from \$11.7 billion in 2012 to \$33.1 billion in 2019 (Sheela, 2013). LIBs possess a higher specific energy and a longer cycle life than other battery chemistries. These features make them particularly attractive for personal mobility, where vehicle electrification is expected to fuel growth in demand for LIBs as a path toward reduction of transportation-related carbon emissions. The high energy requirements of application-oriented consumer electronic devices such as cellular

phones and PC tablets have also driven adoption of LIBs for portable power. Market penetration of LIBs however remains limited by their high cost relative to alternative battery chemistries, and by concerns about their safe use, particularly for high power density applications such as in electric vehicles where significant thermal management is required.

Conventional LIB cells are manufactured using lamination technology, wherein the electrode material is mixed with a conductivity additive and a binder to produce a slurry that is coated onto the electrode current collector. The coated foils for the cathode and the anode are dried, compressed, slit, and rolled with a separator to form a three-layer assembly that is further calendared, slit, and wound into a receiving pouch and canister filled with liquid electrolyte. Charge and discharge cycling of the sealed cell in the formation stage yields the laminated battery commonplace as the current state-of-art in LIB energy storage. More than 600 million cylindrical cells were produced using lamination in 2012 (Wu, 2013). The ubiquitous presence of LIB cells in today's consumer goods can be attributed to their proven capability to deliver energy storage at higher densities than the previous generation of electrode chemistries, while requiring relatively little maintenance, short priming, and low self-discharge. LIB cells also have the

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capacity for high-rate discharge as needed for specialized applications such as in cordless power tools.

In counterpoint to these enduring successes, LIB cells produced using wet slurry technology are limited by several key physical and chemical factors (Scrosati and Garche, 2010; Tsivadze et al., 2013). The inclusion of binder material and conductive additives in the electrodes reduces the energy and power density of a laminated cell. The cathode and anode crystalline powder morphology is difficult to control by heat treatment, and the application of the binder coating is non-uniform. The resultant inherent heterogeneity of the randomly organized electrode particle assemblies causes charge localization near non-conductive cell phases. Additionally, despite the presence of a separator and a non-aqueous electrolyte between the cathode and the anode, dendritic growth often cannot be fully arrested, shortening cycle life and increasing the risk of thermal runaway.

Laminated cell production is capital-intensive. Almost two dozen separate unit operations are required, including a kiln, mixing line, coater, roll press, laminator, slit, and dryer (Nelson et al., 2011). Many steps involve handling of sensitive electrochemical layers and evaporation of volatile organic compound solvent-based printing formulations. The winding, folding, and stacking of foil sheets introduces large stresses in electrode layers, and the inserted tab electrical leads have small contact areas and are subject to weld quality weaknesses. Coating of non-uniformities down and across web may also contribute to LIB overheating and failure.

For a laminated cell, liquid electrolyte must be supplied for the battery to function (Park et al., 2010). Many such electrolytes are flammable or toxic. Electrolyte addition requires heat drying and filling of the battery container under vacuum in a dry room to ensure that bubbles are cleared before the case is sealed, although the package will remain susceptible to gas evolution and venting. It is not uncommon for dry room costs to exceed \$1 million per month for scaled laminated LIB production facilities (Dunn et al., 2012). Prior to their use, long-term formation of laminated LIBs must be carried out with controlled cycles of charging and discharging to activate the battery anode. This extends the production time between manufacturing and testing and adds to the energy consumption for battery assembly.

Solid-state battery processing, using thin film deposition with a conductive solid electrolyte, has the potential to reduce or eliminate many of the problems associated with laminated LIB assembly (Quartarone and Mustarelli, 2011), while simultaneously enhancing the physical and chemical properties of the produced cells. High-speed vacuum coating manufacturing has been successfully applied in the roll-to-roll production of capacitors, optical filters, coated foils, solar thermal collectors, and solar photovoltaic panels, with scaled capabilities from multiple global suppliers ranging from tens to hundreds of thousands of square meters of coated product per worker shift (Espinosa et al., 2012). A variety of techniques have been investigated for deposition of thin-film batteries, including radio-frequency sputtering (Kanekori et al., 1983), magnetron sputtering (Xu et al., 2010), pulsed laser deposition (Zhao et al., 2002), plasma-assisted directed vapor deposition (Kim and Wadley, 2009), electron beam evaporation (Liu et al., 2004), ion beam directed assembly (Vereda et al., 2000), or a combination of methods thereof (Bates et al., 2000, 1994; Dudney and Jang, 2003).

In the roll-to-roll production of double-sided, roller-mounted polymer photovoltaic cells (Sondergaard et al., 2012), rotary screen printing has been used to fabricate high-conductivity electrodes (Manceau et al., 2011) and active layers (Zhang et al., 2009) for solar cells, while slot-die coating has been applied in the one-dimensional patterning of cells at speeds of several meters per minute (Krebs et al., 2009). Post-processing of screen-printed and

slot-die coated cells typically involves hot-air drying or curing with ultraviolet light, followed by lamination with a pressure-sensitive or hot melt adhesive. The thin-film vacuum vapor deposition procedure used in the production of the solid-state lithium batteries modeled in this study follows a broadly similar process to that described here for roll-to-roll production of polymer photovoltaic cells.

For the physical vapor deposition of the solid-state battery cells, assembly occurs in deposition zones within a single, compact sealed vacuum chamber (Albano et al., 2011). All materials are useable in their as-delivered form, as there is no required heat treatment to form complex crystal structures. Also, no dry room or clean room is needed, which reduces energy consumption and the infrastructure footprint. Charge concentration can be mitigated by depositing homogeneous single-phase materials tuned for single functionality in each layer. The solid-state formulation replaces the liquid electrolyte and separator with a solid electrolyte that is more likely to arrest dendrite growth on account of its higher stiffness (Takada, 2013; Tarascon and Armand, 2001). Moreover, no physical separation of anode and cathode or burr free cutting is required, as the moving substrate in roll-to-roll processing produces full-size stacked and terminated batteries in a single non-contact unit operation. Folding and handling stresses are thereby minimized, and *in situ* full area contact termination provides lower resistance and better safety. Lastly, for solid-state batteries no formation or storage-aging work in process is needed to activate the batteries.

Solid-state fabrication of thin film lithium batteries presents a set of challenges distinct from those encountered in laminated LIB cell production. In thin film batteries, mass transport limitations restrict the rate capability and utilization of the electrode active materials, reducing the achievable specific energy and power (Patil et al., 2008). Control of the interface between the solid electrode and the solid electrolyte remains problematic. Scaling of roll-to-roll production for thin film batteries beyond the prototype stage also remains to be demonstrated. The prospective performance advantages of solid-state electrochemical cells have spurred research and entrepreneurial interest to overcome these challenges, in pursuit of the fabrication of lithium batteries using vacuum deposition methods for consumer electronics and BEV battery packs. Recently, an energy density of 1143 Wh/L was reported for a prototype solid-state vacuum vapor-deposited lithium cell, more than double the energy density of current laminated lithium ion batteries (Fletcher, 2014).

It is timely to consider, from a process-based, attributional perspective, the environmental impacts of manufacturing lithium batteries using solid-state rather than lamination technology. To this end, life cycle inventories (LCIs) for a representative lithium-based electrochemical cell were constructed in this study for laminated and thin film solid-state battery production methods. A comparative life cycle assessment was then conducted over a range of impact categories, first considering manufacturing-phase impacts per unit electrochemical energy storage, and then broadening the scope of the comparison to the entire life cycle environmental impacts for prospective BEVs powered by battery packs containing either laminated or solid-state cells.

In the remainder of this article, the functional unit and system definition, battery characteristics, and battery inventory are described. Thereafter, the principal results of the LCA are presented, and the implications of the research findings are discussed in the context of prior work on this subject.

2. Research scope and method

A streamlined life cycle approach, following the operational guide based upon ISO standards (Guinee, 2002), is employed in this

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