

Dry Electrode Coating Technology

Hieu Duong, Joon Shin & Yudi Yudi

Maxwell Technologies, Inc.
3888 Call Fortunada, San Diego, CA 92123
hduong@maxwell.com / 1-858-503-3296

Abstract: *In this paper we report a truly solventless dry battery electrode (DBE) coating technology developed by Maxwell Technologies that can be scalable for classical and advanced battery chemistry. Unlike conventional slurry cast wet coated electrode, Maxwell's DBE offers significantly high loading and produces a thick electrode that allows for high energy density cells without compromising physical properties and electrochemical performance. Maxwell's DBE exhibits better discharge rate capability than those of wet coated electrode. Maxwell has demonstrated scalability by producing robust self-supporting dry coated electrode film in roll form with excellent long-term electrochemical cycle performance, and established large pouch cell prototypes in greater than 10Ah format.*

Keywords: Dry battery electrode; dry coated electrode; wet coated electrode; lithium ion battery

Introduction

Within this decade, there has been immense effort focused on reducing the cost of energy storage devices. The goal towards energy independence through electrification of automobiles for widespread adoption has greatly incentivized this endeavor. Strategies ranging from novel materials investigation to advanced cell manufacturing development span the cost reduction effort. Maxwell Technologies is actively engaged in this global cost reduction effort through its development and refinement of its proprietary dry electrode fabrication technology.

Classical slurry wet coating technology has drawbacks such as solvent toxicity, reactivity between electrode material and solvent and unwanted changes of physicochemical properties of coated electrodes. Maxwell's proprietary solvent-free coating technology resolves such issues [1-3]. Maxwell dry coating technology offers manufacturing cost and performance differentiation as well as novel battery chemistry enablement [4]. This paper provides the initial foundation and validation for the application of dry coated electrode technology in lithium-ion batteries.

Maxwell Technologies is a San Diego-based ultracapacitor manufacturer that uses a proprietary solvent-free electrode production process. Advanced process development without the need for solvents has enabled Maxwell's dry coating electrode production lines to operate at high throughput using a minimal manufacturing footprint. This unique electrode manufacturing process does not introduce any volatile waste products into the atmosphere or require

complex manufacturing plant arrangements. It begins with dry raw materials and maintains its liquid-free state throughout the subsequent processing steps to ultimately produce a robust high-performance ultracapacitor electrode.

For the past several years, Maxwell has been engaged in research and development efforts to expand the application space of its dry coating electrode process technology to apply to battery electrode manufacturing processes. Cell performance using prototype dry coated lithium-ion battery electrodes has been demonstrated. Dry coated electrode configuration with various architectures using a wide range of materials can be produced at thicknesses ranging from about 50 microns to about 1 millimeter. In addition to manufacturing flexibility, the cohesion and adhesion properties of electrodes derived from the dry coating process are superior in the presence of electrolyte at high temperatures compared with those produced using the wet coating technology.

This unique electrode process technology offers significant saving in manufacturing cost and helps curb CO₂ pollution in the battery electrode manufacturing process. By eliminating the use of any solvents, and the associated coating and drying complexity inherent in wet coating technology, the dry coating electrode process is environmentally friendly, and can be readily installed and commissioned with a much lower start-up capital investment. Thus, dry coating electrode manufacturing is economically attractive and socially responsible.

This paper provides recent progress in high energy dry coating electrode technology and its capacity for the enablement of advanced battery chemistries as evidenced by cell performance results witnessed from dry coated lithium-ion battery electrodes.

Experimental

Maxwell's proprietary dry coating electrode technology is comprised of three steps: (i) dry powder mixing, (ii) powder to film formation and (iii) film to current collector lamination; all executed in a solventless fashion. Maxwell's dry coating electrode process is scalable, and can accommodate current lithium ion battery chemistry and advanced battery electrode materials. In this report, the robustness of the dry coating electrode process is demonstrated using a host of commercially available anode materials such as silicon based materials and lithium titanate (LTO), as well as cathode materials such as layered Li(Ni_xMn_yCo_z)O₂ (NMC), LiNi_{0.8}Co_{0.15}Al_{0.05}O₂ (NCA),

LiFePO₄ (LFP) and sulfur. All dry powder materials were mixed using Maxwell's proprietary dry coating process to yield a final powder mixture consisting of active material, binder and conductive additive as shown in Fig. 1 (top). This powder mixture was calendered to form a continuous self-supporting dry coated electrode film that is wound in roll form (Fig. 1, bottom). A wide range of dry coated electrode configurations can be produced by the adjustment of film processing conditions to control material loading weights and active layer thickness.



Figure 1. Dry powder mixture comprised of active materials, conductive carbon and polymer binder (top) and a roll of free-standing dry coated electrode film fabricated from a pilot scale roll-to-roll equipment (bottom).



Figure 2. Dry battery electrode NMC roll (left) and graphite roll (right) double sided laminated onto current collector which is ready for cell assembly.

Once a film with target specification is achieved, it is laminated onto a current collector to yield an electrode, Fig. 2, that is ready for cell production.

All laminated dry coated electrodes were dried under vacuum at 120°C overnight to remove any ambient moisture before being assembled into pouch cell in an Ar-filled glove box for the small cell format or in dry-room conditions for the large cell format.

Electrochemical testing for rate capability and cycle life was carried out using an Arbin system at room temperature.

Results and discussion

Various dry coated battery electrodes were fabricated, including NMC811, NCA, LFP, LTO, sulfur/carbon and silicon composite, using Maxwell's dry coating electrode technology. Maxwell's dry coating electrode technology can be used to produce advanced high capacity NMC811 cathode and silicon-graphite composite anode that can deliver designed discharge capacity. Fig. 3 highlights discharge voltage profile for two advanced high capacity materials. The NMC811 dry coated electrode exhibited typical discharge profile with stable voltage plateau at the end of the discharge process. The Si/Graphite composite dry coated electrode produced electrochemical characteristic of Li delithiation of silicon at around 0.5V that significantly enhances energy density. Evaluations of cycling performance of high capacity NMC811 and Si/Graphite composite dry coated electrode are underway.

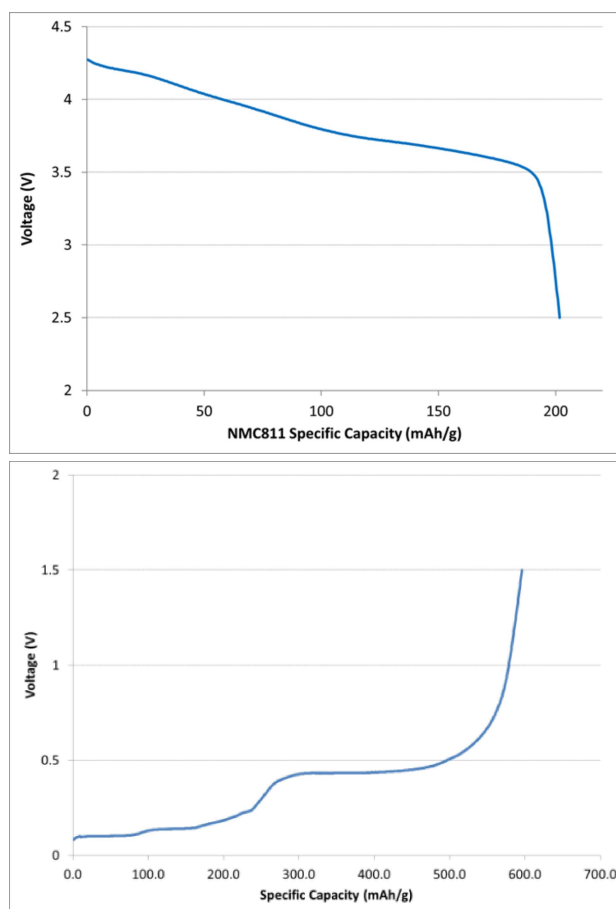


Figure 3. Discharge voltage profile of dry coated NMC811 cathode (top) and dry coated silicon-graphite composite anode (bottom) half-cell.

Maxwell's dry coating electrode technology renders a unique electrode micro-structure in which the polymer

binder network allows for high ionic conductivity and intimate electronic contact between active materials and the conductive carbon network. As a solvent-free process, the polymer binder is not dissolved; as a result, the binding mechanism is an inter-connecting network comprised of point-contacts with the active material particle surface. This dry binding structure is less obtrusive and, consequently, enables lithium ions better access to the active material particles. This feature is especially advantageous for high rate performance in high energy density electrodes. A comparative example between a dry coated versus wet coated electrode is captured in Fig. 4. Both types of electrode were prepared using NMC111 for the cathode and graphite for the anode at equaled concentration with different binder materials. A constant current at 0.1C was applied to charge the cell to 100% SOC prior to discharge at each C-rate. Under low constant current discharge, both coated electrode types yielded cell discharge capacity of 105 mAh, used to normalize as 100% capacity retention at 0.1C. This discharge rate test results indicate that the dry coated electrodes delivered higher power than wet coated electrodes in a high energy density electrode configuration.

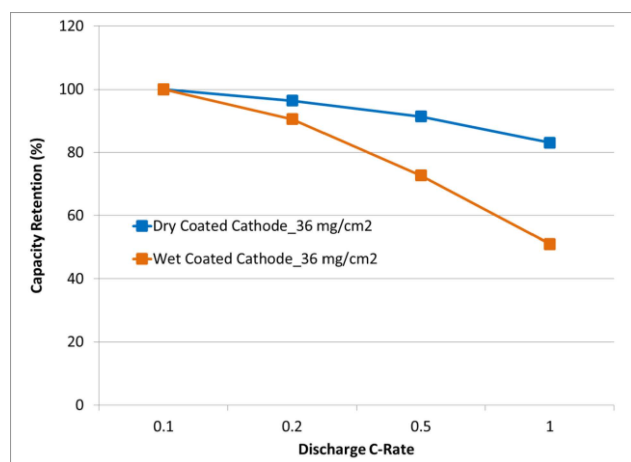


Figure 4. Discharge rate performance of dry coated NMC111/graphite full-cell in comparison to wet coated NMC111/graphite full-cell. Electrode loading is 5mAh/cm². Cut-off voltage was 4.2V and 2.8V for charge and discharge, respectively.

Additionally, a higher rate capability has been demonstrated by dry coated electrode with areal capacity of 4 mAh/cm², as shown in Fig. 5. Capacity retention for a set of five cells incorporating dry coated electrodes at 2C discharge is above 90%. The lower charge transfer and contact resistance in the dry coated electrode offer higher energy density cell designs with improved power capability.

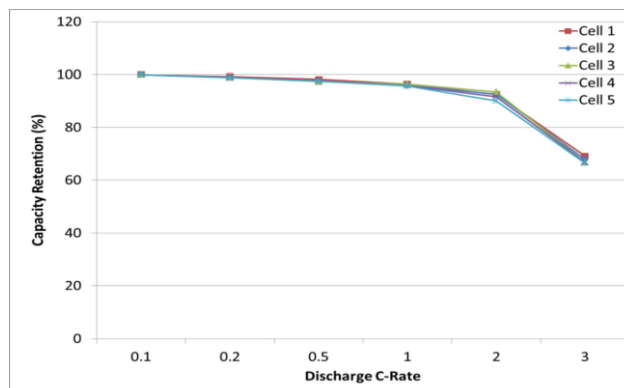


Figure 5. Rate performance of dry coated NMC111 (94 wt.% loading)/graphite (96 wt.% loading) electrode in prototype pouch cell: NMC111 electrode loading is 27 mg/cm² (4mAh/cm²). The cell was charged to 4.2V at constant current followed by constant voltage at 4.2V and discharged to 2.8V.

In addition to higher power performance demonstrated in high energy density cells using dry coating, electrodes derived from a solvent-free process can also be robustly cycled at 100% DOD using a charge/discharge rate of 0.5C/1C, respectively. It can be observed in Fig. 6 that a full-cell with a specific capacity of 4 mAh/cm² can deliver nearly 90% of its original capacity after 2000 cycles.

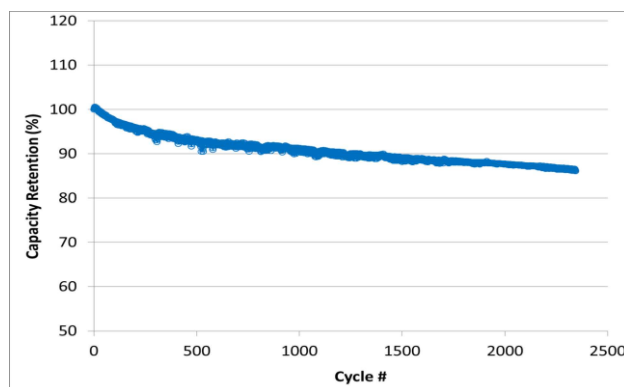


Figure 6. Cycle performance of dry coated NMC/graphite electrode in a prototype pouch cell at constant current 0.5C charge/1C discharge: Electrode loading- 4mAh/cm². Cut-off voltage was 4.2V and 2.7V for charge and discharge, respectively.

Discussion

Maxwell Technologies has successfully expanded its proprietary dry electrode process used to productize ultracapacitors to include lithium based battery electrode production. It has been demonstrated that this unique dry electrode process can be applied to both classical and advanced battery materials, and is scalable to roll-to-roll production. Maxwell's dry coated battery electrode offers extraordinary benefits, including manufacturing cost reduction, elimination of solvent toxicity, enabling the

application of liquid sensitive battery chemistries and enhancing cell performance, particularly at high loading weights when compared to conventional wet coated electrode in discharge rate studies. These results suggest that dry coated electrodes exhibit lower particle-to-particle contact resistance and charge transfer impedance, likely due to a uniform network of interconnects between binder and active material particles. Additionally, prolonged cycle life performance of dry coated electrodes in small prototype pouch cells and large format pouch cells, $\geq 10\text{Ah}$, is under evaluation.

References

1. Park, D., Canas N.A., Wagner N., Friedrich K.A., "Novel Solvent-free Direct Coating Process for Battery Electrodes and Their Electrochemical Performance," J. Power Sources, vol. 306, pp. 758-763, 2016.
2. Al-Shroofy M., Zhang Q., Xu J., Chen T., Kaur A.P., Cheng Y., "Solvent-free Dry Powder Coating Process for Low-cost Manufacturing of $\text{LiNi}_{1/3}\text{Mn}_{1/3}\text{Co}_{1/3}\text{O}_2$ Cathodes in Lithium-ion Batteries," J. Power Sources, vol. 352, pp. 187-193, 2017.
3. Ludwig B., Zheng Z., Shou W., Wang Y., Pan H., "Solvent-Free Manufacturing of Electrodes for Lithium-ion Batteries," Nature, Scientific Reports 6, no. 23150, pp 1-10
4. Duong H., Suszko A., Feigenbaum H., "Dry Electrode Process Technology," 229th ECS meeting, vol. A05, P-475, May 2016.