Nanocrystallization of $LiCoO_2$ Cathodes for Thin Film Batteries Utilizing Pulse Thermal Processing

Oak Ridge National Laboratory (ORNL) has a unique revolutionary rapid thermal annealing capability that enables in-situ fabrication of nanoscaled materials. This technique utilizes a high density plasma arc-based technology and a methodology called Pulse Thermal Processing (PTP) that enables the manipulation of materials on the nanoscale. The unique characteristics of PTP with its high power densities (>20,000 W/cm_2), short processing time millisecond regime) and large processing area (up to 1,000 cm_2) allows for rapid thermal processing of thin film and nanoparticle material systems on flexible temperature-sensitive substrates such as polymers without thermally affecting the underlying material. Thus, with these unique processing characteristics PTP could potentially enable a multitude of thin film and nanoparticle devices such as; thin film batteries, magnetic media, photovoltaics, solid state lighting, thermoelectrics, and thin film transistors.

Nanostructured architectures can greatly increase the performance of energy-storage devices, including the capacity, charge time, cycle lifetime and overall energy efficiency of batteries. Lithium ion batteries are used primarily in consumer electronics such as cell phones, laptop computers and handheld personal digital assistants (PDA) with possible future use in electric and hybrid electric vehicles. The advantages of thin film lithium ion batteries are; all solid state construction, wide operating temperatures (-20°C to 140°C) with no degradation, can be formed into any shape and/or size, and are safe under all operating conditions (no leaking).

One of the major challenges to the improvement of thin film lithium battery technology is the efficient crystallization and sintering of the $LiCoO_2$ cathode thin-films deposited by RF magnetron sputtering. Even though the as-deposited films, which are x-ray amorphous and possibly nanocrystalline, can be used as cathodes, when crystallized to grain sizes approaching 100 nm, the cathodes can deliver a power density 10 times higher. Typically, the crystallization requires conventional furnace annealing at 550° to 700°C in an oxidizing atmosphere for several hours. Furthermore, the high temperature anneal step limits the choice of substrate materials to those stable at the high temperature oxidizing conditions. Ideally, the substrate for the thin film battery would be as thin, light, flexible, and inexpensive as possible. If the alumina substrates used in current prototype thin film batteries were replaced with Kapton® (polyimide), which can withstand temperatures to 400°C, the cathode can be properly annealed by PTP. Thereby, the gravimetric and volumetric energy and power densities of the battery could potentially be increased by nearly 10 times (from 0.1 to 1 mW/cm_2).

This nanomanufacturing concept definition study is focusing on the nanocrystallization of the $LiCoO_2$ cathode thin films on polyimide substrates and evaluate the microstructural evolution and resistance as a function of PTP processing conditions. A significant decrease in the cathode resistance as measured by liquid electrolyte testing correlates to improved capacity and charge and discharge rate of the battery. Also due to the short processing times, on the millisecond scale, the thermal budget is significantly reduced as compared to conventional annealing which occurs at 700°C for 2 hours, thus realizing a significant reduction in energy utilization during manufacturing.

Project Partners:

Oak Ridge National Laboratory
Oak Ridge, TN
Principal Investigator: Ronald Ott
E-mail: ottr@ornl.gov

ITN Energy Systems
Littleton, CO

For additional information, please contact

Gideon Varga
Technology Manager
U.S. Department of Energy
Industrial Technologies Program
Phone: (202) 586-0082
E-mail: Gideon.Varga@ee.doe.gov

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