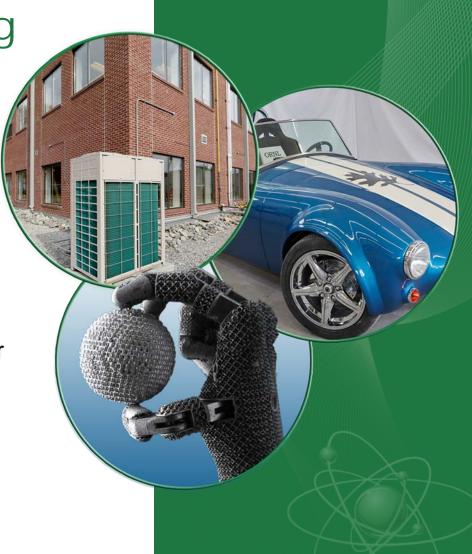
Past R2R Processing Successes at ORNL and Current/Future Opportunities

Presented by

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Presented to AMO HV R2R Workshop Alexandria, VA

12/2/15





ORNL Is Addressing R2R Processing Issues Relevant to Industry

- Annealing of materials on temperature-sensitive substrates (solid-state lithium-ion cathodes for large-scale thin-film batteries); Planar Energy Devices, 2010-2012.
- Primary drying (solvent removal) of dispersion cast coatings (replacing hazardous NMP organic solvent with water); PPG Industries, various binder suppliers, coating equipment suppliers, battery producers, 2010-2015.
- Dramatic reduction of secondary drying (removal of adsorbed moisture) time of lithium-ion electrodes in preparation for cell assembly; A123 Systems, 2012.
- UV and electron beam (EB) curing of high-solids dispersions into thick films;
 Miltec, 2011-Present; COMET Plasma Control Technologies 2015-Present.
- On-line and off-line non-destructive evaluation (NDE) for coating quality control (QC) improvement and long-term performance enhancement; NREL, battery makers, equipment suppliers, 2012-Present.
- R2R slot-die deposition of thick films for lithium-ion electrodes with graded and patterned architectures; Palo Alto Research Center (PARC), binder companies, battery producers, 2014-Present.
- Optimization of web speeds for different deposition methods and applications; ongoing.

Selected Industrial Partners















































realizing graphene's possibilities









Planar Energy Highlight – Solid-State Cathode Processing

3-4.8 V; 0.067 mA

Decay Due to Electrolyte Decomposition

125

Cycles

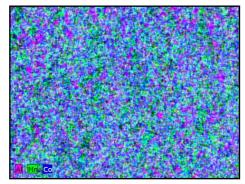
- Goal: to replace PVD and high-T furnace annealing with room-T
- furnace annealing with room-T atomized spray deposition and photonic (UV/IR) annealing.

 Current processing involves PVD for ~1 h and 650°C furnace annealing for 20 min.

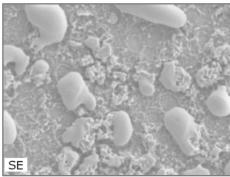
 New processing involves minutes
- of room-T deposition and <1 min of photon exposure.
- Best performance of 38% furnace annealing baseline was achieved with single voltage plateau and multiple pulses on the PulseForge 3300.
- Pulse thermal processing conditions were refined to include combinations of pulse duration ramping, 2) multiple voltage plateaus, 3) number of pulses at each voltage.
- CRADA results were sufficient for Planar Energy to make the business decision to pursue the technology for microbattery applications (i.e. sensors, smart cards, etc.).

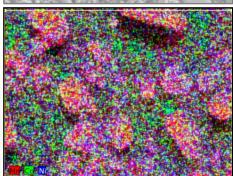


As Received



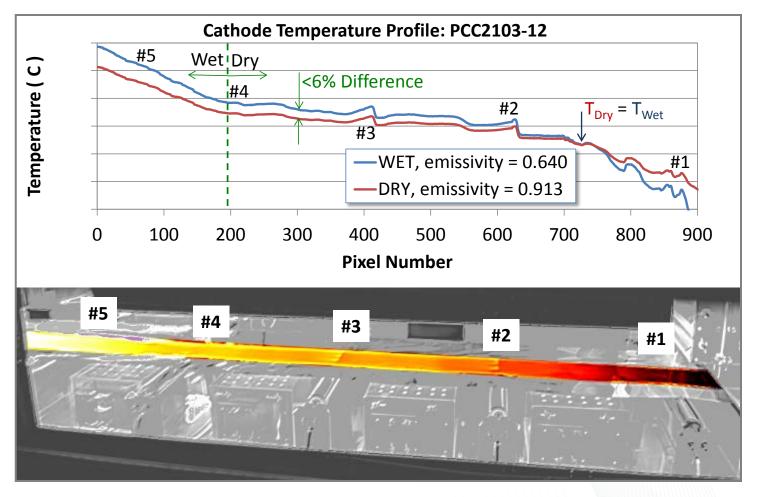
Phase Segregation Must Be Avoided After Annealing





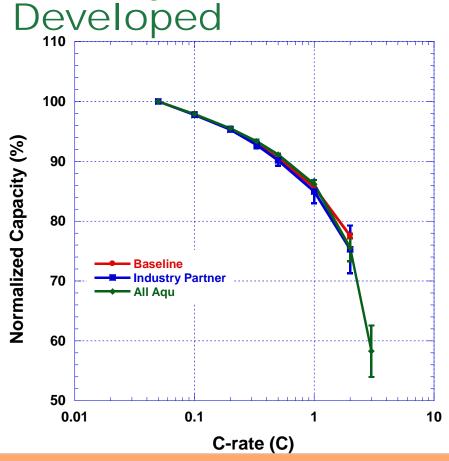
IR Imaging of Electrode Coating Line Reveals Wet-toDry Transition Points

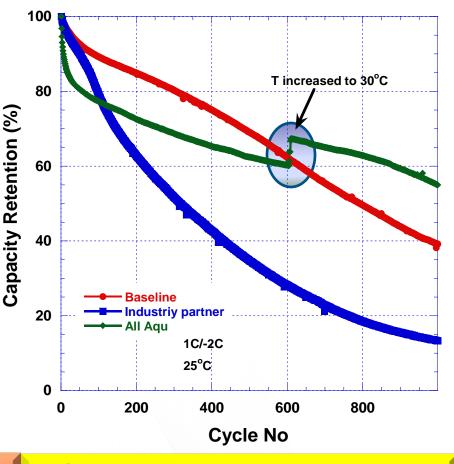




IR imaging of Dow Kokam pilot coating line showing uniform drying of cathode. The "Dry" temperature curve crosses the "Wet" temperature calibration curve at 100°C (T_{Wet} = T_{Dry} = 100°C).

Aqueous Electrode Processing - Sufficient Primary and Secondary Drying Protocols





NMC532 & CP-A12 graphite

- Baseline-all NMP based processed electrodes
- Industry partner-NMC cathode via aqueous processing
- All aqu—all aqueous processed electrodes

- Similar rate performance
- Baseline cells—best early cyclability
- Slope changes indicate different degradation mechanisms.



Approach with NREL

Problems:

- Electrode coating defects are currently identified by optical CCD cameras, which miss many of the subtle inhomogeneities.
- A *low-cost* method for *in-line* thickness and porosity is needed for optimal electrode coating QC.
- Useful feedback loops must be developed based on IR thermography input information to *prevent* coating defects and inhomogeneities.

Overall technical approach and strategy:

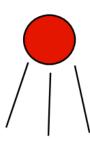
- Use white light or thermal excitation of electrode coatings to generate a IR emissivity signature from electrode coatings.
- Take measured IR emissivity and correlate it to a coating T profile for input into a mathematical model based on electrode physical properties (IR absorbance, heat capacity, thermal conductivity, bulk density, etc.).
- 3. Use model and measured heat loss down the web to generate a porosity and thickness profile.







heat source



heat loss due to convection

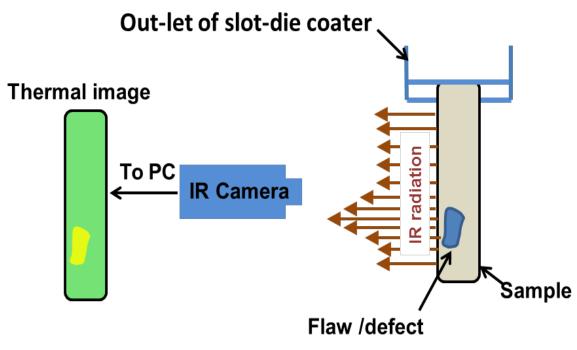


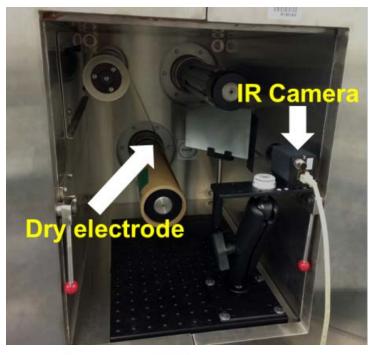
battery electrode moving at a contant velocity

NREL R2R IR
Thermography Setup



Technical Accomplishments – IR camera Installation on Slot-Die Coater



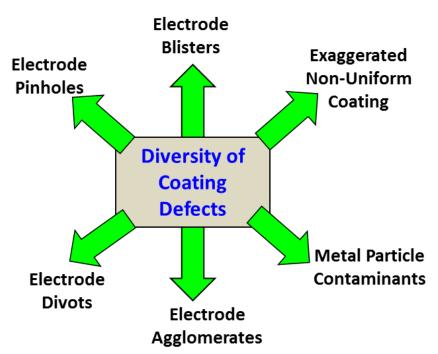


Monitor temperature profile of dried electrodes detecting any potential defects such as divots, pinholes, agglomerates, etc.

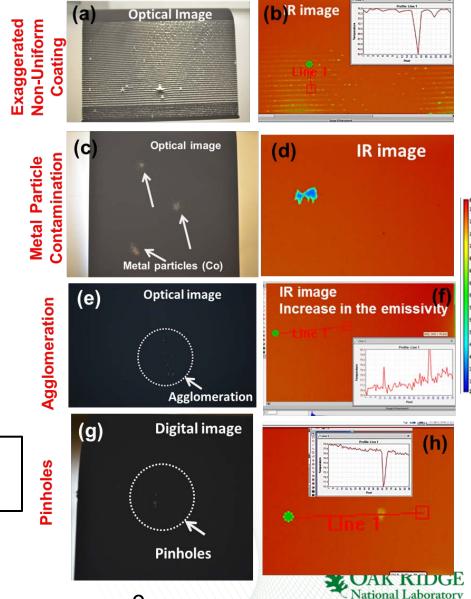


Systematic Study of Electrode Coating Defects Reveals Differing Performance

Effects

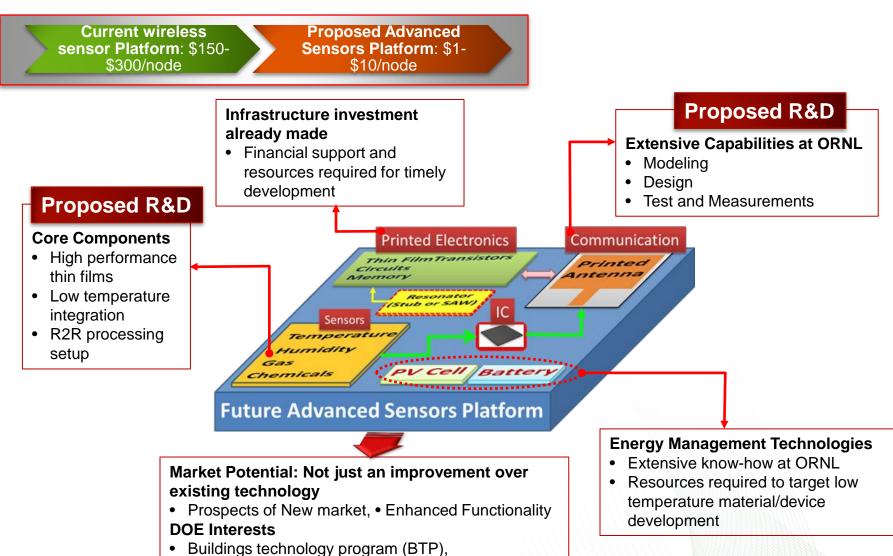


6 types of defects are being studied to determine relative importance.

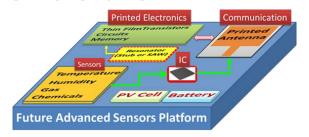


Low-Cost, Multi-Sensor Wireless Platform for SMART Buildings

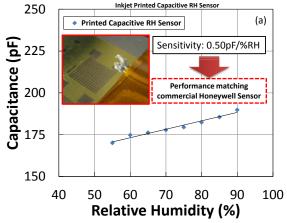
Advanced manufacturing office (AMO)

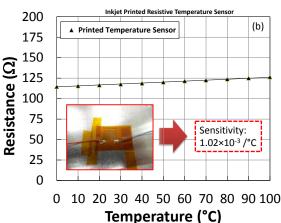


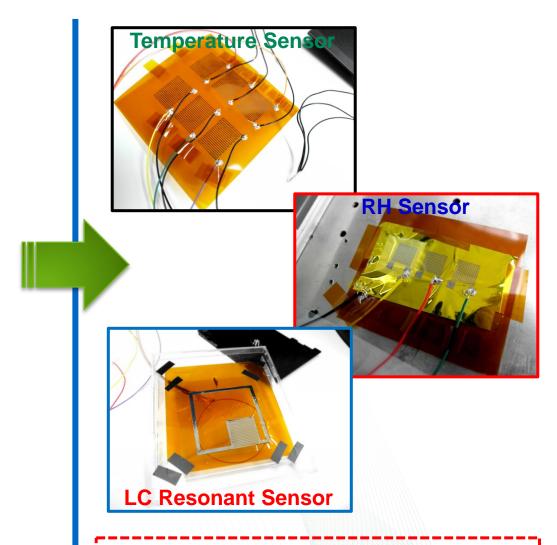
Multi-Sensor Integration and Performance Evaluation



Discrete T and RH Sensors







Multi-sensor Printing and Calibration for system level integration und

Future Challenges and Opportunities

- Raw material variation effects on coating deposition quality and associated device performance.
- Can similar equipment and approaches be used thick and mid-range coatings (~1-1000 μm)?
- Addressing difficulties of developing R2R thin-film (nm-scale) deposition processes.
- Identification of locations in R2R processes where in-line QC is needed.
- Successful technology transfer of developed in-line NDE and QC methods to industry.
- Understanding effects of defects on device performance; what constitutes a defect worth detecting?
- At ORNL, translate battery R2R processing successes to other applications such as supercapacitors, energy conversion (fuel cells and membrane electrolyzers), flexible displays, and flexible sensors.
- New R2R methods needed for 2-D material production.



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