Cost-Effective Fabrication of High-Temperature Ceramic Capacitors for Power Inverters*

U. (Balu) Balachandran
Argonne National Laboratory

June 7, 2017

Project ID# EDT061

***Work supported by the U.S. Department of Energy, Vehicle Technologies Program.**

Overview

Timeline

- **Project start date: January, 2014**
- **Project end date: January, 2017**
- **Percent complete: 100%**

Budget

- **Total project funding (FY14 - FY17)**
  - **DOE: $3714K. Out of this total, subcontract to Delphi/Sigma Technologies = $810K & Penn State University = $350K**
- **Funding received in FY16: $400K**
- **Funding for FY17: $0K**

Barriers addressed

- **A & C (Cost & Weight): Overall size and cost of inverters**

  Capacitors are a significant fraction of the inverter volume (≈35%), weight (≈23%), and cost (≈23%).

- **D (Performance & Lifetime): High-temperature operation**

  The performance and lifetime of polymer-based capacitors available today degrade rapidly with increasing temperature (ripple current capability decreases by 80% with temperature increase from 85°C to 105°C).

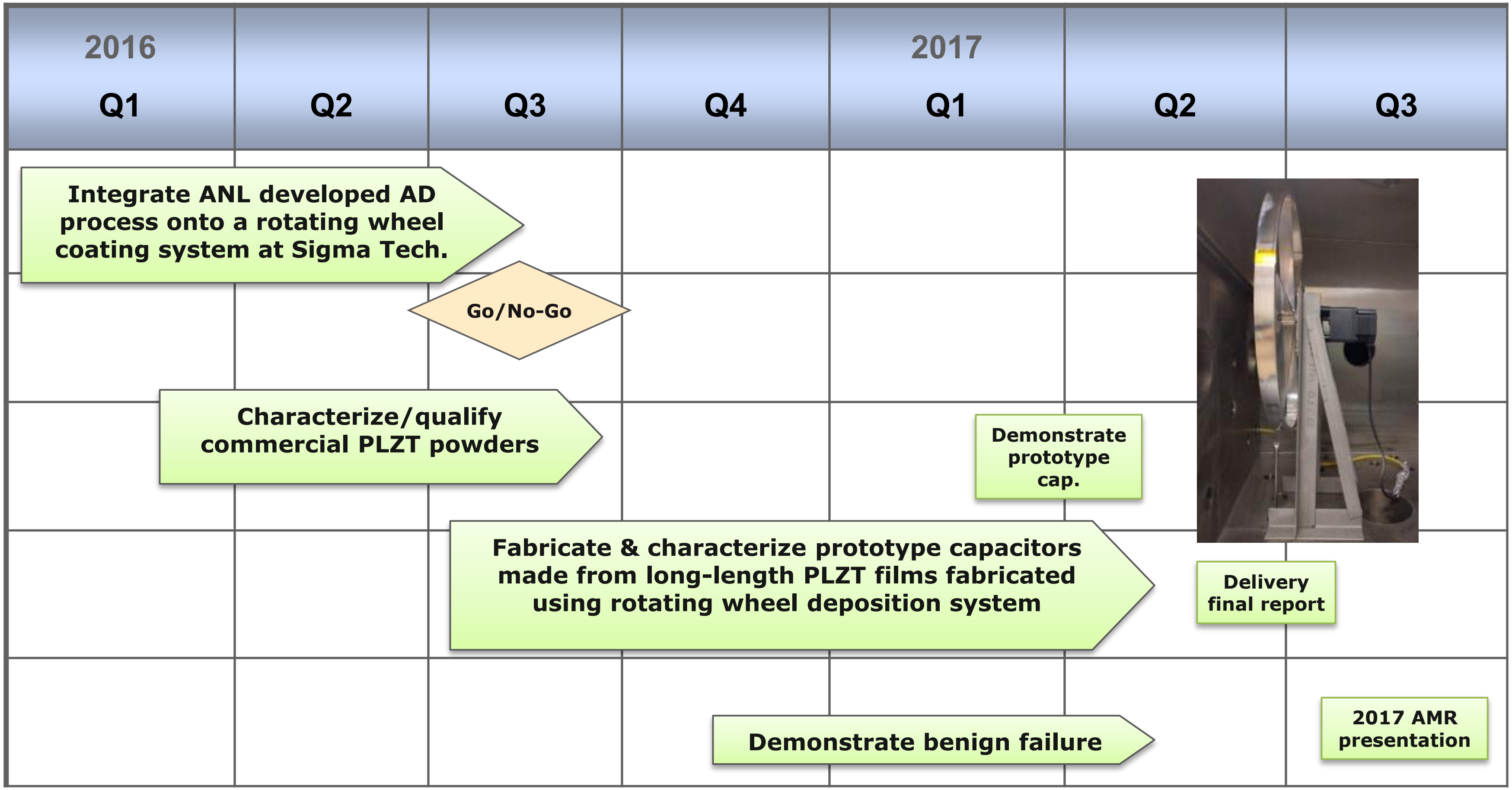
Partners

- **Delphi Electronics & Safety Systems**
- **Sigma Technologies International**
- **Penn State University**
- **Project Lead: Argonne National Laboratory**

Relevance/Objectives

- **Overall objective:** Develop an efficient, cost-effective process for fabricating Pb-La-Zr-Ti-O (PLZT)-based capacitors for advanced power inverters in EDVs. PLZT films satisfy high-temperature & volumetric requirements for advanced capacitors (140°C/650 V) (EDT Goal).
- **Relevance:** This project addresses key barriers (*capacitor volume, high-temperature operation, as well as fail-safe operation and manufacturability*). Capacitor performance improvements will reduce volumes, which will be important to achieve inverter volume targets.
- **Specific objective for March '16 – Jan. '17:** Transfer high-rate, room-temperature aerosol deposition (AD) process developed at ANL to a rotating wheel coating system at Sigma & produce prototype capacitors.
- **Impact:** Establishes an economically attractive manufacturing process to produce advanced high-temperature capacitors with high volumetric efficiency for power inverters in EDVs.

FY16/17 Approach and Milestones



Go/No-Go Decision: Decision on film coverage area

Phase I: (Months 1-18): Develop & demonstrate high-rate, room-temperature aerosol deposition (AD) process to fabricate PLZT films on thin polymer substrates (Argonne)

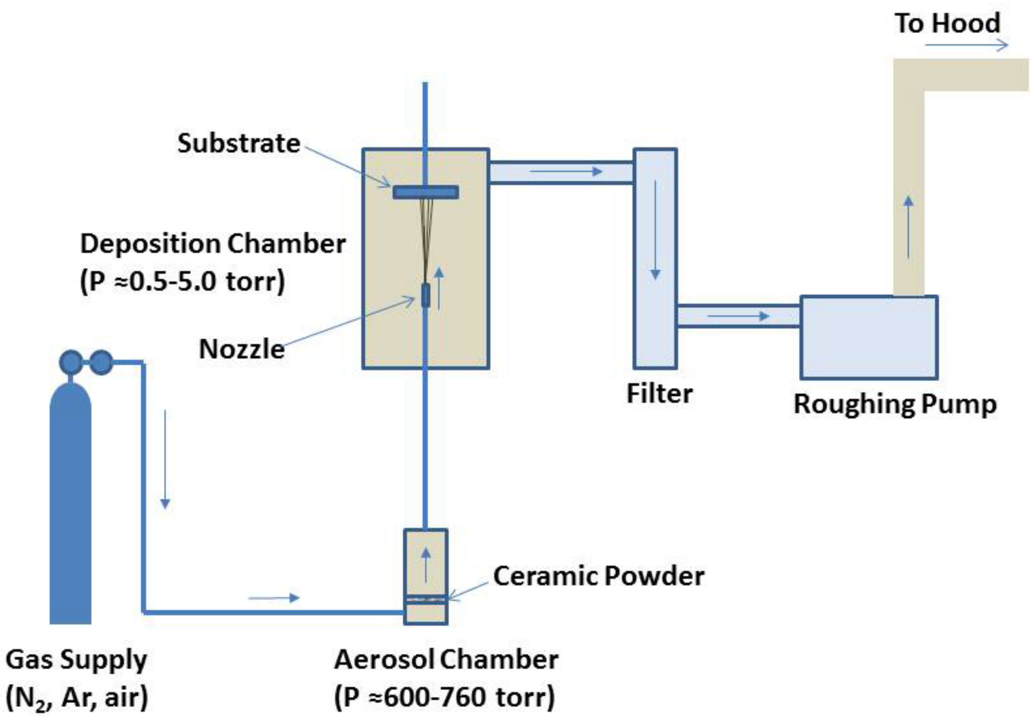
Phase II: (Months 19-36): Transfer basic AD process to a rotating wheel coating system and fabricate & characterize prototype rolled PLZT film capacitors (Sigma Tech./Delphi)

Approach/Strategy

- Higher dielectric constant (≈80 vs. ≈3 for SOA polymer-based caps.) PLZT film capacitor to achieve higher volumetric efficiency. *(Barrier C: Weight/volume reduction)*
- Higher temperature PLZT film to meet/exceed 140°C operation temperature requirement. *(Barrier D: Performance & Lifetime)*
- High-rate, room-temperature aerosol deposition (AD) process to economically manufacture the PLZT-based advanced capacitors. *(Barrier A: Cost reduction)*
- Team includes a Tier 1 automotive power electronics manufacturer (Delphi), a leading capacitor designer & capacitor OEM (Sigma), a leading university in the study of dielectrics (PSU), and a National laboratory (ANL) with strong expertise in energy technology R&D to commercialize the technology.

Our approach leads to smaller capacitor (≈0.3 L), higher operation temperature (>200°C), and lower cost, all of which significantly benefits industry and end users.

Schematic of Aerosol Deposition (AD) Process



AD Process Parameters

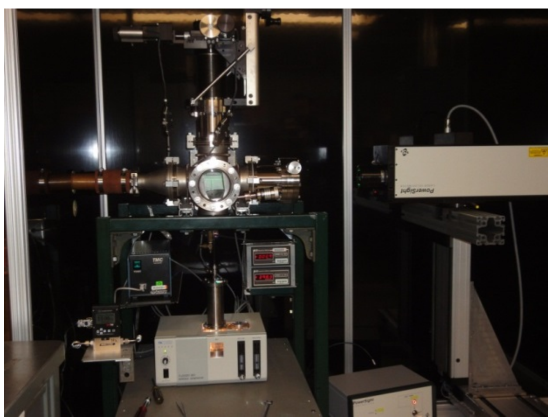
- Nozzle design
- Particle size & distribution
- Aerosol generator & deposition chamber pressure
- Particle velocity
- Distance between nozzle orifice & substrate
- Carrier gas & flow rate

The AD is a high-rate, room-temperature deposition technique that offers the possibility to deposit dense PLZT films on a variety of substrate materials (glass, metal foils, polymers, etc.).

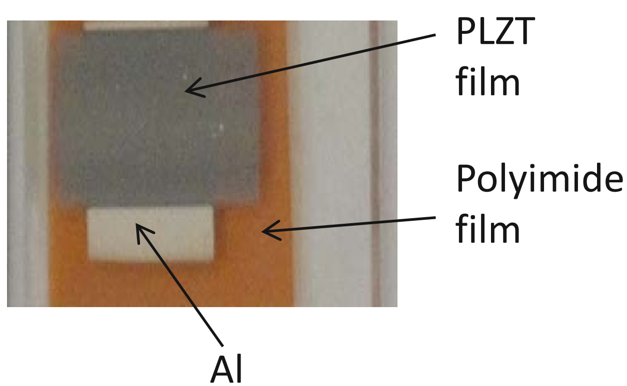
Technical Accomplishments & Progress

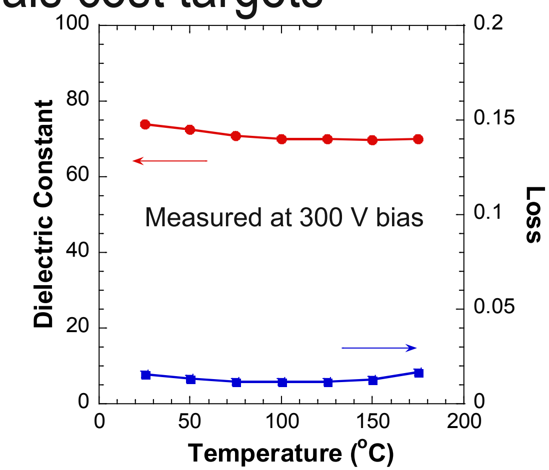
Previous accomplishments:

- Fabricated ≈8-μm-thick PLZT on aluminum metallized polyimide (PI) films by AD process in ≈20 min (*vs. 1 week by spin-coating*)
- Demonstrated that AD PLZT films on metallized PI films exhibit dielectric properties suitable for high-temperature applications
- Studied fault clearing event in AD PLZT films on PI
- Defined capacitor specification & established materials cost targets



Sample size: 7-mm x 15-mm



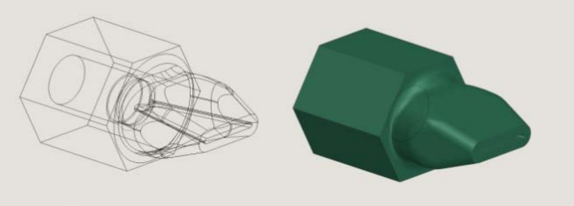


PLZT film made at ANL by high-rate AD process has high dielectric constant and low loss over the operational temperature, voltage, and frequency range.

Technical Accomplishments/Results (Cont.)

Scale-up of AD Process (*present results*)

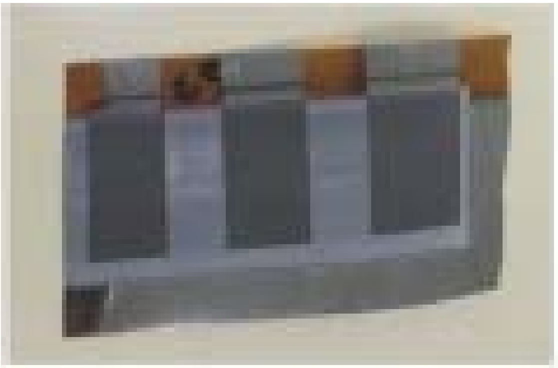
- Increased the deposition chamber size from <2 Cu.ft to ≈27 Cu.ft
- Increased the pumping capacity from ≈150 Cu.m/h (oil-based pump) to ≈650 Cu.m/h (dry pump)
- Improved the motion system from experimental X-Y table system (typical film of 7-mm x 15-mm) to rotating wheel system (≈35-mm width x 60-cm diameter)
- Changed the commercial fluidized bed aerosol generator to a proprietary design generator
- Moved from offline electrode deposition to inline sputtering process

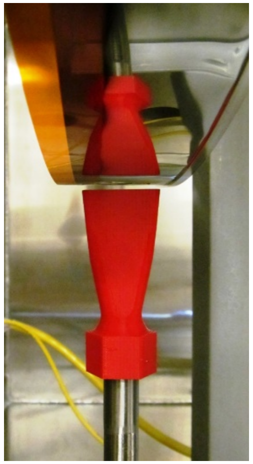


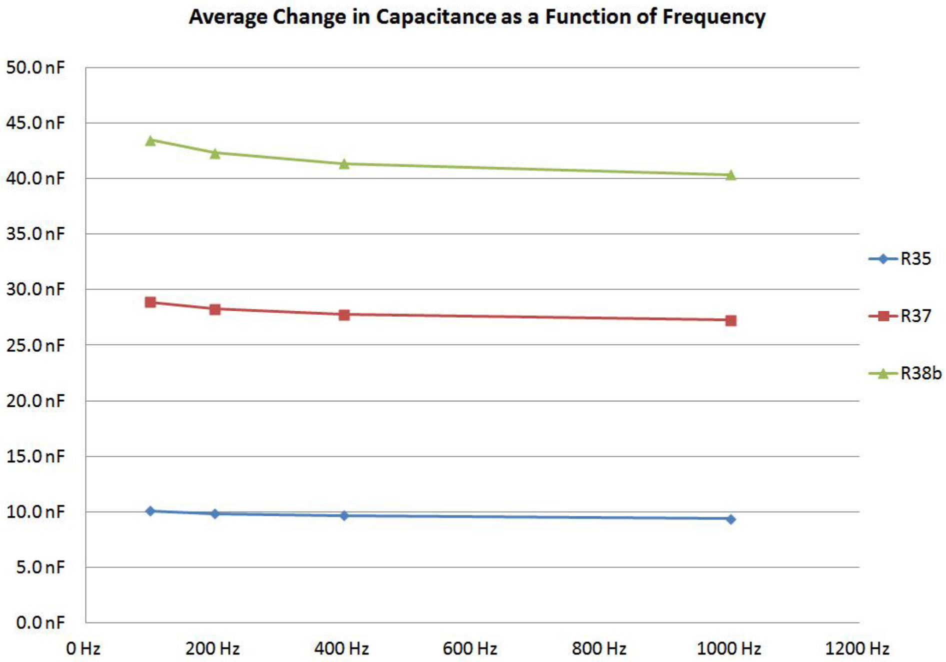
Upgraded necessary equipment & evaluated nozzle designs and feed rate capabilities to fabricate AD PLZT capacitors.

Technical Accomplishments/Results (Cont.)

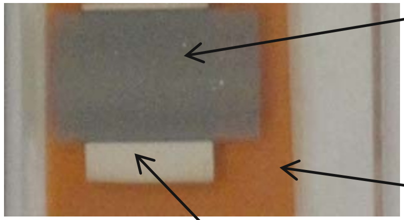
Fabrication of ~6” long PLZT tapes by AD Process (@ Sigma)







Small PLZT film made at ANL by AD process

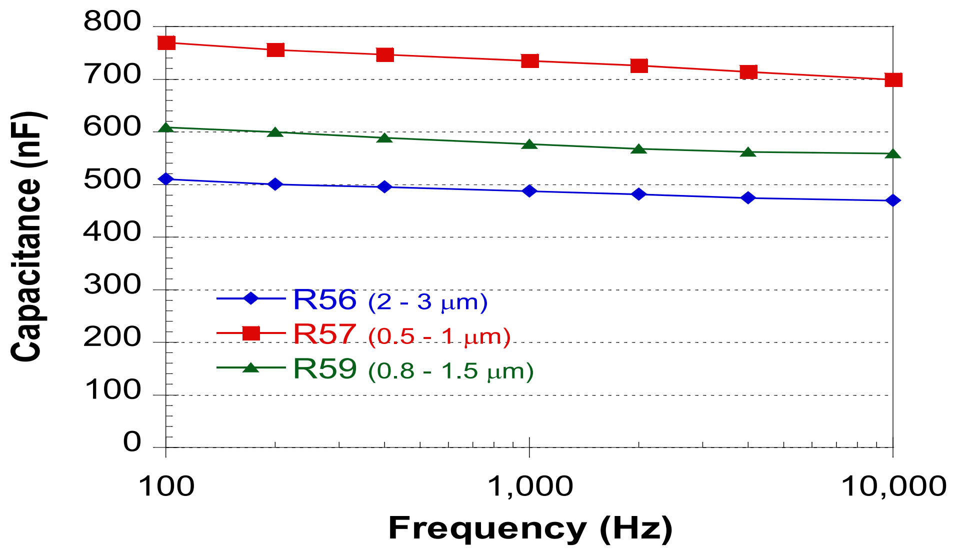


Improvements in film coating & capacitance obtained with nozzle design modification, pressure differential between chambers, and adjustment of wheel speed.

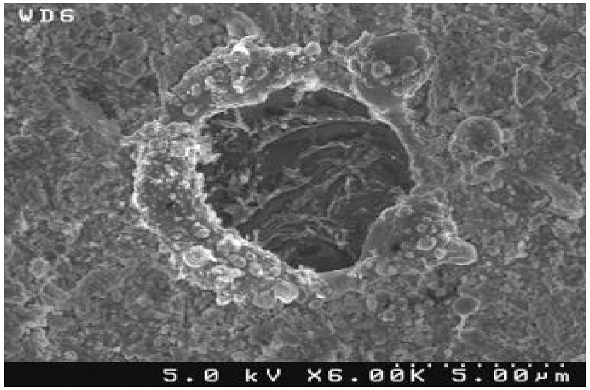
Technical Accomplishments/Results (Cont.)

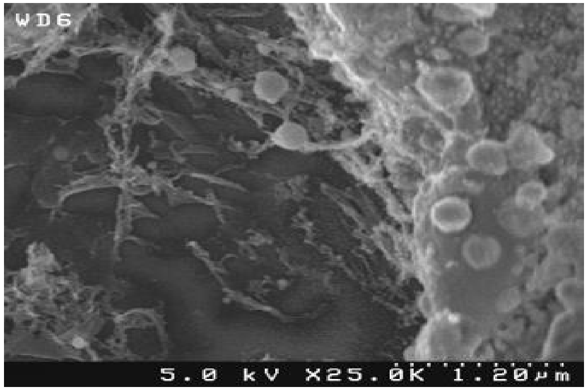
Capacitance & Self-clearing of PLZT films prior to winding

Capacitance as a Function of Frequency: Pre-winding



Self-clearing of defects in films



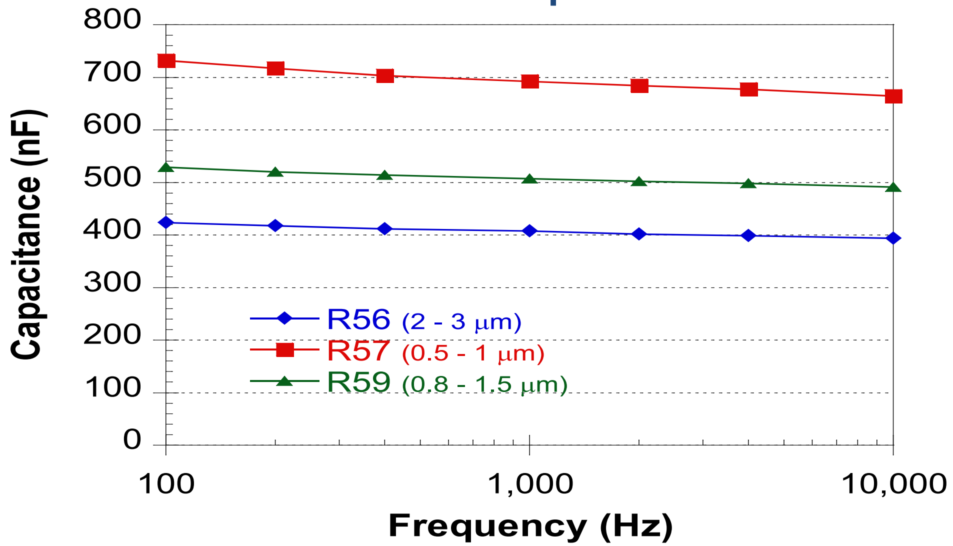


Thin aluminum electrode is removed from the defective area; the "crater" is no longer acting as a capacitor. This area is negligible compared to the entire area of the film, therefore, it does not reduce the capacitance *(similar to losing a pixel on your computer screen)*.

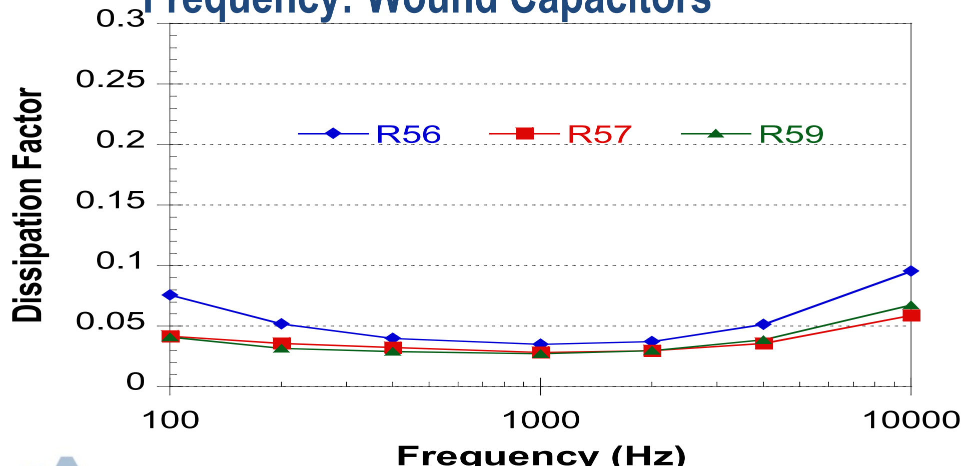
Technical Accomplishments/Results (Cont.)

Performance of the Capacitors after Winding

Capacitance as a Function of Frequency: Wound Capacitors



Dissipation Factor (DF) as a Function of Frequency: Wound Capacitors

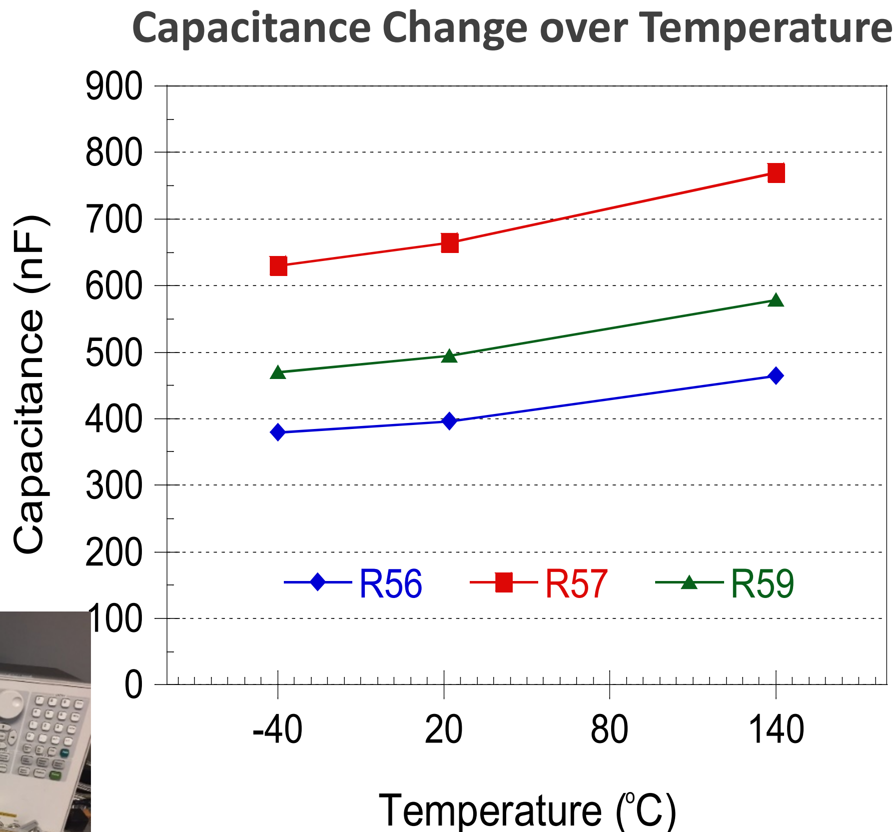


- Long-length PLZT films were wound around a ½” diameter wooden core with a separator layer of polyimide film that was trimmed to be ¼” narrower than the active capacitor.
- This left 1/8” of space on each end of the capacitor for the arc-sprayed termination metal to penetrate and make electrical contact.

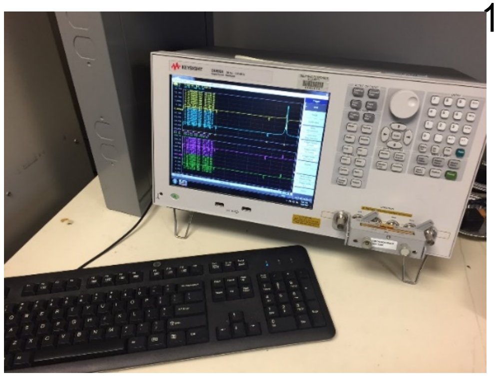
Demonstrates potential to make wound high-temperature ceramic capacitors with benign failure feature using metallization similar to wound polymer capacitors.

Technical Accomplishments/Results (Cont.)

Test/Validation of Wound Capacitors at Delphi



Venable 350C Frequency Response Analyzer



Keysight E4990A Impedance Analyzer

Delphi, a Tier 1 automotive power electronics manufacturer validated the results of the wound capacitors.

Micro Climate temperature chamber

Response to Previous Year's Reviewers' Comments

This project was not reviewed last year. Comments from the 2015 AMR are addressed here

Comment: The reviewer suggested that the investigators "more seriously study the reliability of the thin film in relation to thermal cycling to verify that cracking does not occur over time."

Response: PLZT films were thermally cycled (about 1000 cycles) by Delphi (Tier 1 inverter manufacturer) between -50°C to +150°C with no measurable degradation in dielectric properties. The performance of the PLZT thin films would have degraded drastically if cracking occurred during thermal cycling.

Comment: The reviewer would "like to have seen a real, traditional, high-volume film capacitor manufacturer on this team too just to keep it commercial."

Response: High-volume film capacitor manufacturers were not involved, because: a) Polymer film capacitor manufacturers are focusing on capacitors for inverter applications and have been developing the technology for much longer time period compared to the AD-based ceramic film capacitor, and might consider our work as competition. b) Most of the ceramic capacitor industry focuses on the larger microelectronic market, and EDVs are viewed as a small but growing market. We must educate them on the needs and size of the power inverter market. c) The AD PLZT capacitor maker(s) may not be the manufacturers of conventional multilayer ceramic capacitors (MLCCs). MLCCs are made by co-firing of dielectric tapes and electrodes and MLCCs don't have benign failure feature. The process developed in this project is more akin to making of polymer film capacitors.

Collaboration and Coordination with Other Institutions

DELPHI

Inverter design engineering (direct customer for the technology), defining overall capacitor requirements, supplying critical knowledge of automotive power electronics, testing/validating results, demonstrating DC-link capacitor with an automobile power inverter.

SigmaTechnologies

Roll-to-roll deposition systems, multilayer coating technologies, coating capacitor films to improve breakdown and self-healing properties, experience to convert lab-scale process into an industrial-scale process.

PENNSTATE

Dielectric characterization, reliability testing, electrode design & deposition, testing/validating results.

This project has a very good, complimentary team: basic science & research from Argonne; materials test & characterization from PSU; an experienced processor with knowledge to scale-up from Sigma; and a Tier 1 user of the product from Delphi.

Remaining Challenges and Barriers

- **Can PLZT with uniform film thickness & density be produced in the rotating wheel/roll-to-roll deposition process?**
  - Optimizing the nozzle slot opening & working width, carrier gas flow rate, and vacuum differential between the aerosol generator and deposition chambers will help to produce films with uniform thickness and density.
  - Sigma has in place several roll-to-roll coating systems that will be retrofitted to perform AD process on continuously moving substrates (roll-to-roll deposition).
- **Can PLZT be deposited on ≈5-μm-thick polymer films by roll-to-roll AD process?**
  - The polymer films used in the rotating wheel deposition system is ≈50-μm-thick; earlier we have deposited PLZT on ≈8-μm-thick Al foil by AD process.

We will address these challenges in our future work

Any proposed future work is subject to change based on funding levels

Proposed Future Work

- **Mechanical improvements to the rotating wheel deposition system**
  - Install an isolation valve between the two chambers
  - Investigate an alternative means of agitating the PLZT powders
- **Process improvements to obtain thickness uniformity over longer lengths (~6' long PLZT tapes)**
  - A larger aerosol generator
  - Dimensionally stable plastic substrate, such as heat-stabilized PEN, to prevent warping of thicker films
  - An abrasion-resistant nozzle material for long, continuous use
- **Transfer the AD process to a roll-to-roll deposition system**
- **Demonstrate the high-temperature DC-link capacitor in an inverter**
- **Update detailed cost & commercialization plan**

The ultimate test of this project will be when we install/test a DC-link capacitor in a traction-sized inverter.

Any proposed future work is subject to change based on funding levels

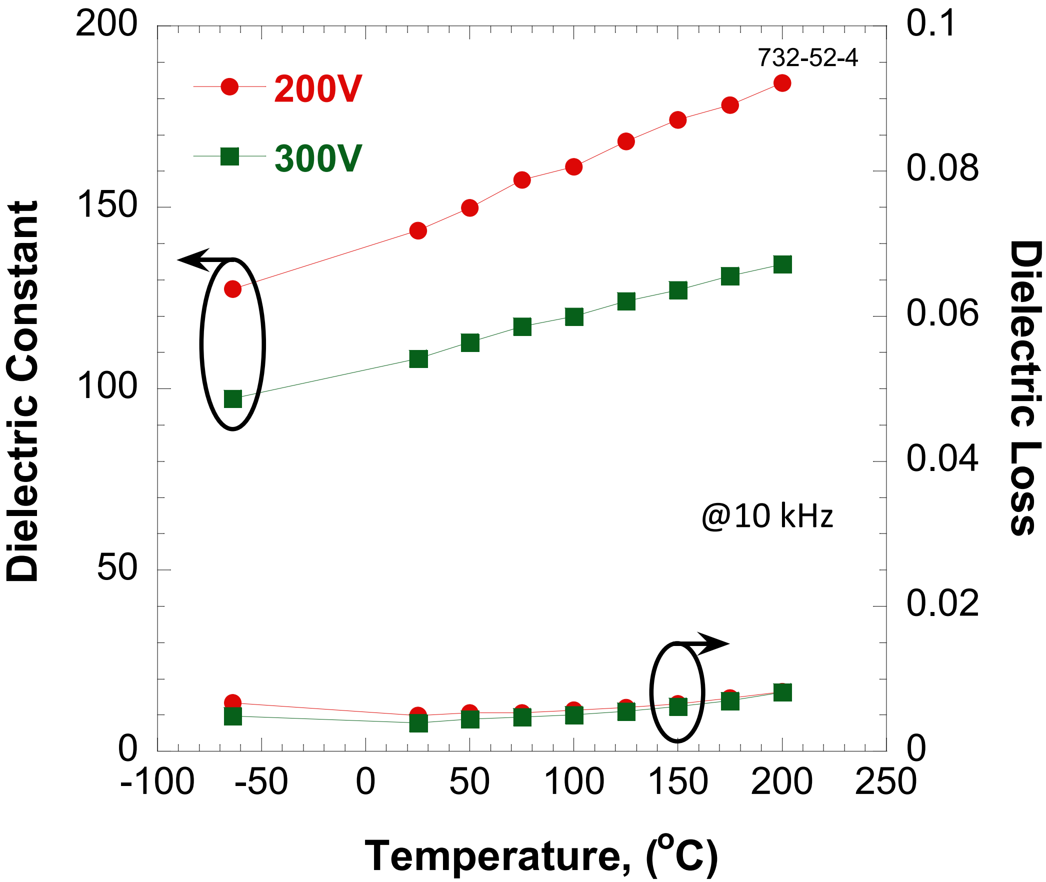
Summary

- The project addresses key barriers *(size, high-temperature capability, as well as manufacturability)* to achieve advanced DC-link capacitors.
- Aerosol deposition (AD), a high-rate, room-temperature film deposition process, is developed to reduce capacitor cost.
- Demonstrated that AD PLZT films on metallized PI films exhibit dielectric properties suitable for high-temperature applications.
- The team includes a Tier 1 automotive power electronics manufacturer (Delphi), a leading capacitor designer & capacitor OEM (Sigma), a leading university in the study of dielectrics (Penn State), along with ANL, a National laboratory with strong expertise in energy technology R&D, to commercialize the technology.
- Demonstrated wound ceramic capacitors with benign failure features.
- Patents: Nine issued; Publications/Presentations: >60 made.

Tier 1 user of the developed capacitor (Delphi) validated the results.

Technical Back-up Slides

Temperature Dependent Dielectric Properties of PLZT Films at 200V & 300V (made by spin-coating)



Measured k ≈110 & loss ≈ 0.004 (i.e., 0.4%) @ 10 kHz & 300 V bias on a 3-µm-thick PLZT on Ni-foil

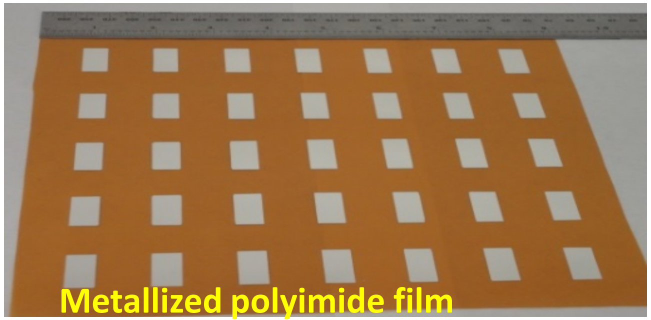
ESR = DF/2πfc (DF = loss factor; f = frequency; c = capacitance).

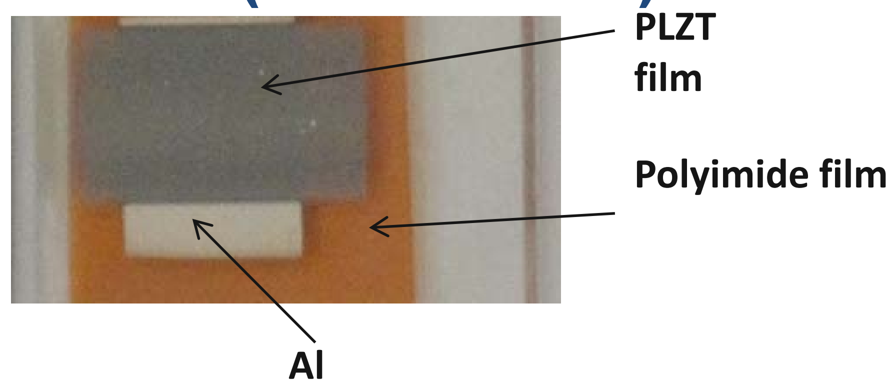
Calculated ESR for 1000 µF cap. based on measured material properties

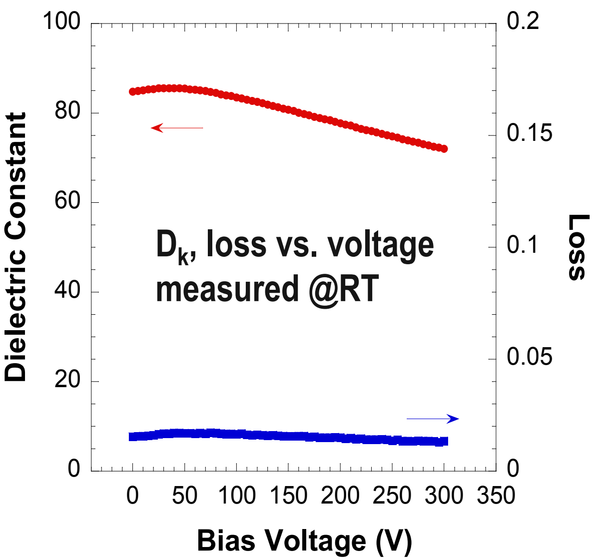
Bias Voltage (V)	ESR @ -64°C (mΩ)	ESR @ RT (mΩ)	ESR @ 150°C (mΩ)
200	0.11	0.08	0.10
300	0.08	0.06	0.10

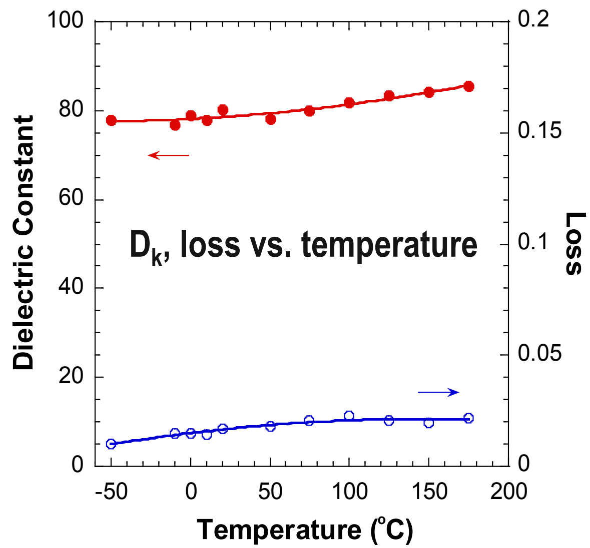
PLZT film has high-temperature capability; high dielectric constant (high volumetric efficiency); and very low ESR

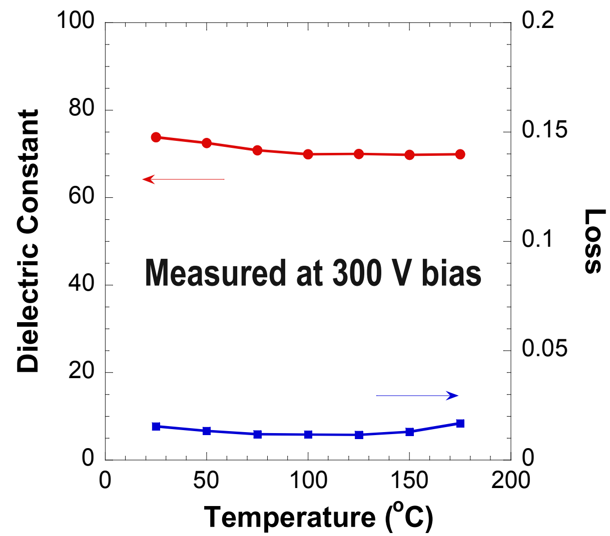
Properties of ≈8-µm-thick PLZT on Al-metallized polyimide films by AD process (made at ANL)





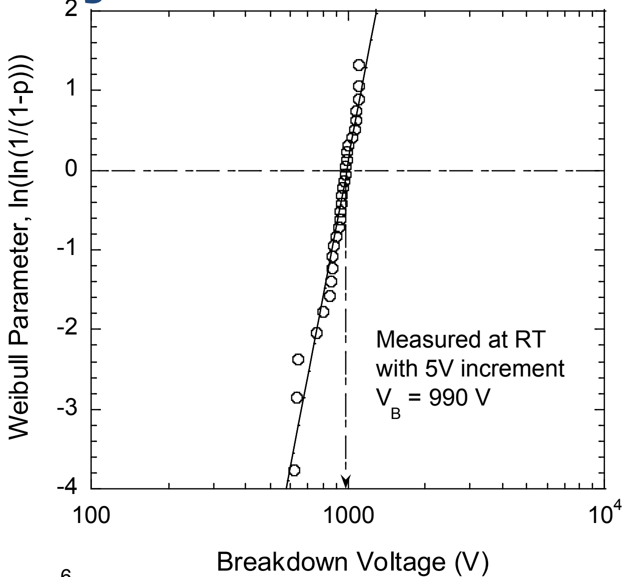


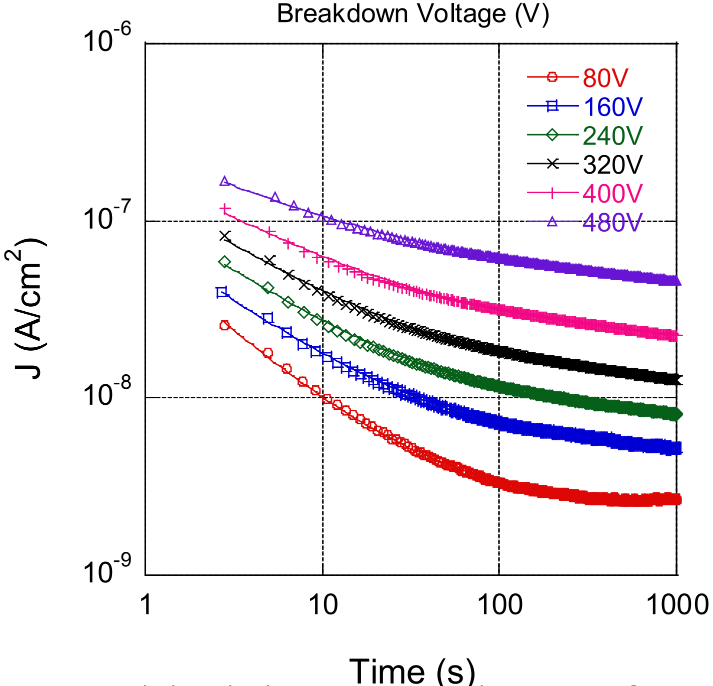


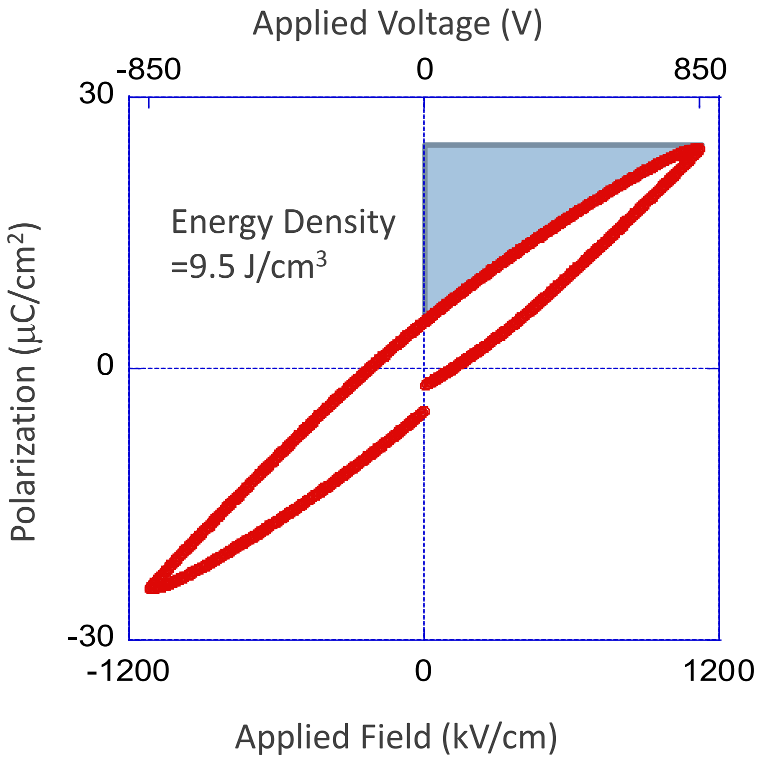


Deposited ≈8-µm-thick PLZT film *in ≈20 min by AD (vs. ≈5 days by spin coating)*.
PLZT has high dielectric constant (≈70 - 80) and low loss (<2%) at 300V bias.

Properties of ≈8-μm-thick PLZT on Al-metallized polyimide films by AD



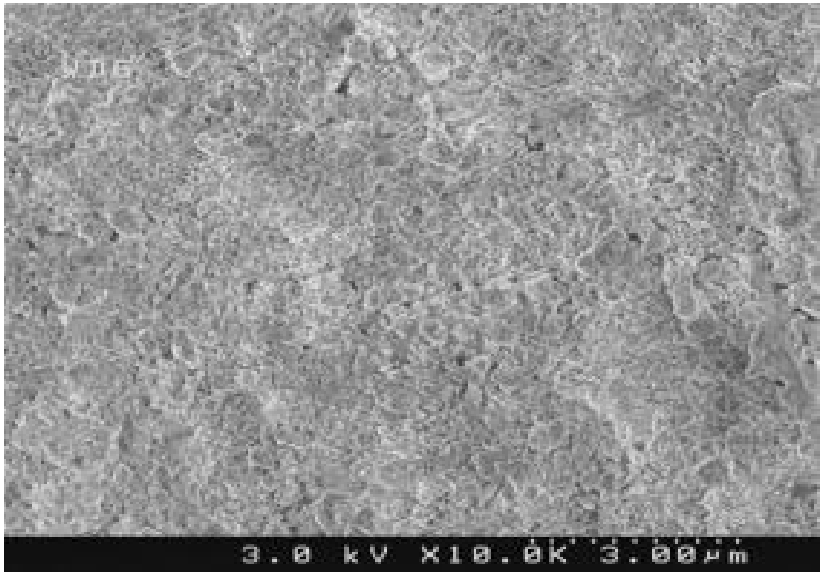




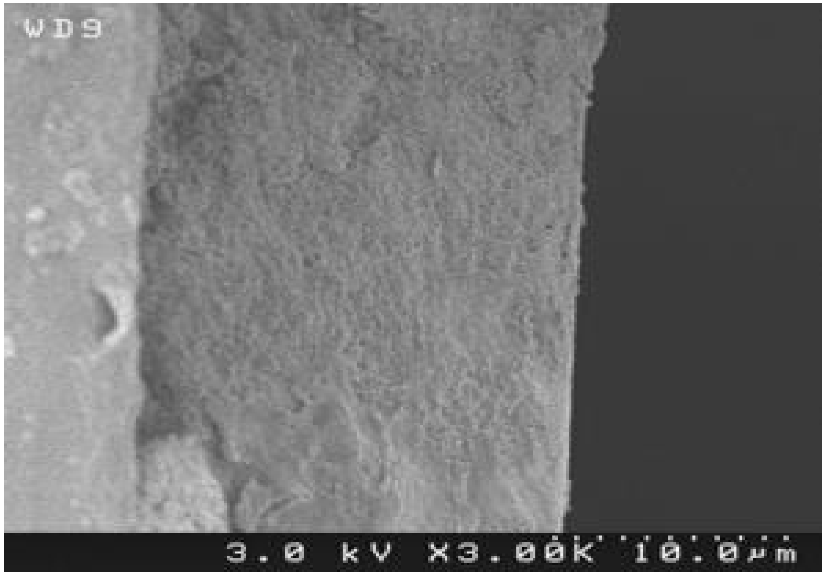
PLZT film made by high-rate AD process has breakdown voltage ≈130 V/μm, energy density ≈10 J/cm^3, and low leakage current (<10^{-7} A/cm^2) at ≈500 V.

SEM Pictures of the PLZT Film Fabricated in the Rotating Wheel Deposition AD System

SEM of the surface of the film (R-53)



SEM of the X-section of the film (R-53)



Dense thick PLZT films are fabricated on metallized polyimide sheets by rotating wheel AD system.

Specifications for DC-link capacitor & technology that enables PLZT-based capacitor to meet specifications

Feature	Specification	Proposed technology to meet specifications
Volume	≤0.6 L	High dielectric constant; ≤0.3 L
Voltage	nominal: 325 V; peak: 600 V	High breakdown strength (> 200 V/µm)
Capacitance	700 µF ± 10%	High dielectric constant (D_k ≈100@ 450V)
Temperature	-40°C to 140°C	High temperature dielectric material with ferroelectric Curie temperature, Tc ≈200°C
Ripple Current	165 Arms continuous, 295 A peak	Low ESR (≤1 mΩ); PLZT performs better as it becomes hotter
Frequency	≥50 kHz	Ferroelectric based capacitors are used for high frequency applications
Dissipation Factor (DF)	≤0.4% at 1 kHz	DF of PLZT is ≈0.4% at 300 V; DF decreases with increase in bias voltage
ESL	≤5 nH	ESL ≤1 nH have been reported for ferroelectric-based multilayer capacitors
Failure Mode	Benign (open)	Benign failure in single-layer PLZT has been demonstrated. Proposed electrode architecture to obtain self-clearing in multilayer and wound capacitor
Life @ operating conditions	>13,000 h	Highly accelerated life testing (HALT) is proposed to predict capacitor life-time.
Cost	≤$30	Inexpensive PLZT films and base-metal electrodes will be continuously deposited as is conventionally carried out in the polymer film industry.