



High Temperature Polymer Capacitor Dielectric Films

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APE009



Overview

Timeline

- Project start: October 2009
- Project end: September 2012
- Percent complete (70%)

Budget

- Total project funding
 - DOE: \$1950k
- Funding received in FY09, FY10 and FY11 include:
 - FY09 - \$350k
 - FY10 - \$750k
 - FY11 - \$850k

Barriers

- Barriers
 - Capacitor Cost (up to 23% of inverter)
 - Thermal control
 - Volume (up to 23% of inverter)

Partners

- Electronic Concepts, Inc.
- Sandia National Laboratories
- Penn State
- Argonne National Laboratories



Project Relevance

- **The Problem**

DC bus capacitors are currently the largest and the least reliable component of fuel cell and electric hybrid vehicle inverters. Capacitors represent up to 23% of both inverter weight and inverter cost and up to 35-40% of the inverter volume. In addition current thin polymer film capacitors have a ceiling operation temperature (105 °C). High temperature polymer dielectrics are very expensive!



Project Relevance

Objective

- Our objective is to develop and engineer novel inexpensive high temperature polymeric material systems for use as next generation dielectric materials that can be used as a replacement technology for DC bus capacitors in hybrid electric vehicles (HEV) and fuel cell vehicles.
 - Solving problems associated with transitioning from “lab-scale” to “pilot-scale” operations and to produce prototype capacitors

Addresses Targets

- Current capacitors lack the temperature, size, and price specifications required for future DC bus capacitors. Our approach simultaneously increase operational temperature ($>150\text{ }^{\circ}\text{C}$), decreases size, and lowers the price of high temperature capacitors ($\$0.015/\mu\text{F}$), while maintaining self-healing properties.

Uniqueness and Impacts

- Our approach uses inexpensive monomers/fillers to create a high temperature polymer dielectrics based on ROMP polymerizations which should meet DOE OVT requirements for high temperature capacitor dielectrics.

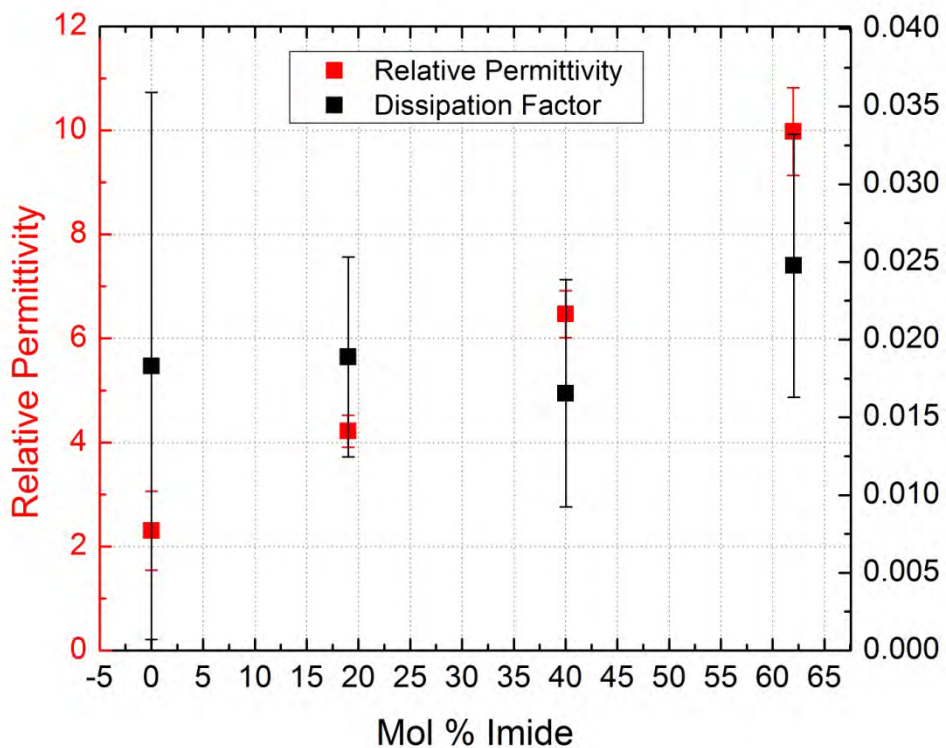


Approach

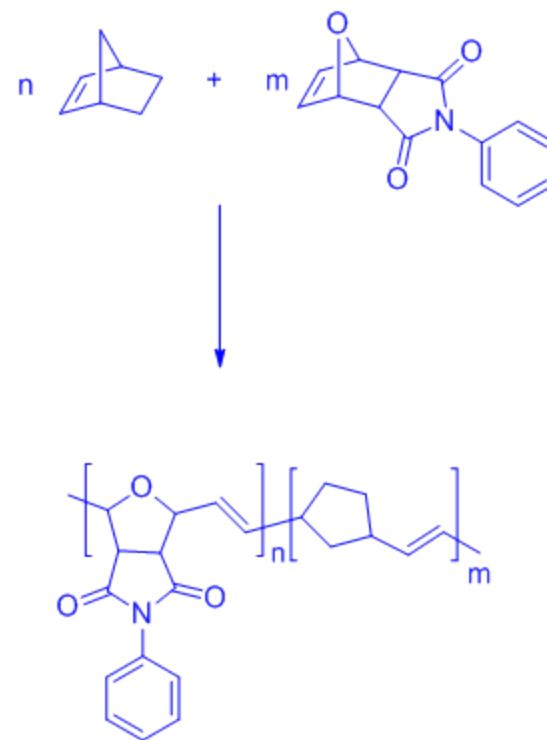
- **Developing inexpensive high temperature, high dielectric polymer capable of forming very thin films**
 - **Controlled polymerization chemistry based on the Ring Opening Metathesis Polymerization (ROMP) allows for fine control of polymer composition and molecular weight**
- **Working with ECI to produce rolls of polymer film and prototype capacitors**
 - **Solving problems realized at ECI by modifying polymer chemistry while maintaining the appropriate dielectric performance.**
 - **Hydrogenation reaction - improved performance - scaled-up**
 - **Free radical inhibitors and the Thiol-ene reaction**
- **Develop nano-composites of high temperature polymer dielectrics to improve energy density**

Technical Accomplishments

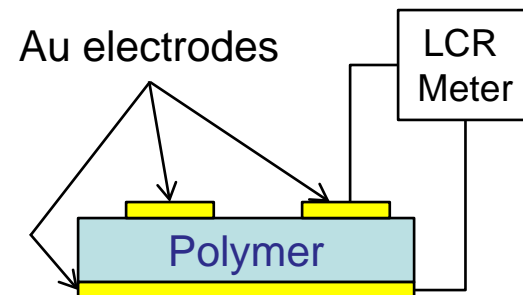
% Imide	% Norbornene	% Imide by NMR	% Norbornene by NMR
0	100	0	100
25	75	14	86
50	50	40	60
75	25	62	38



3 mm electrodes



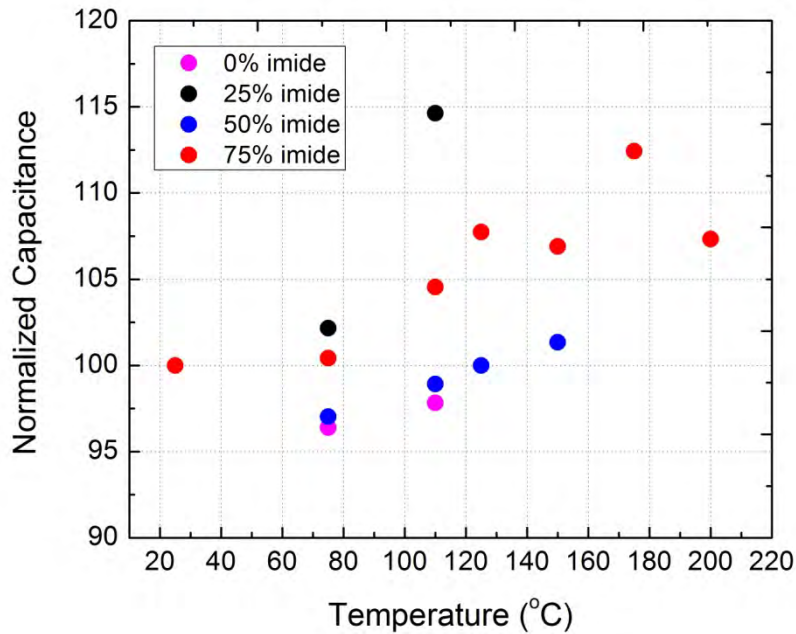
Dissipation Factor



Dielectric Materials Development

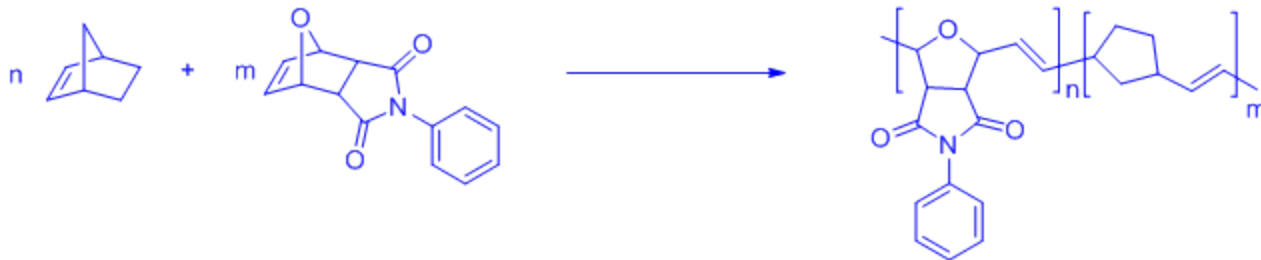
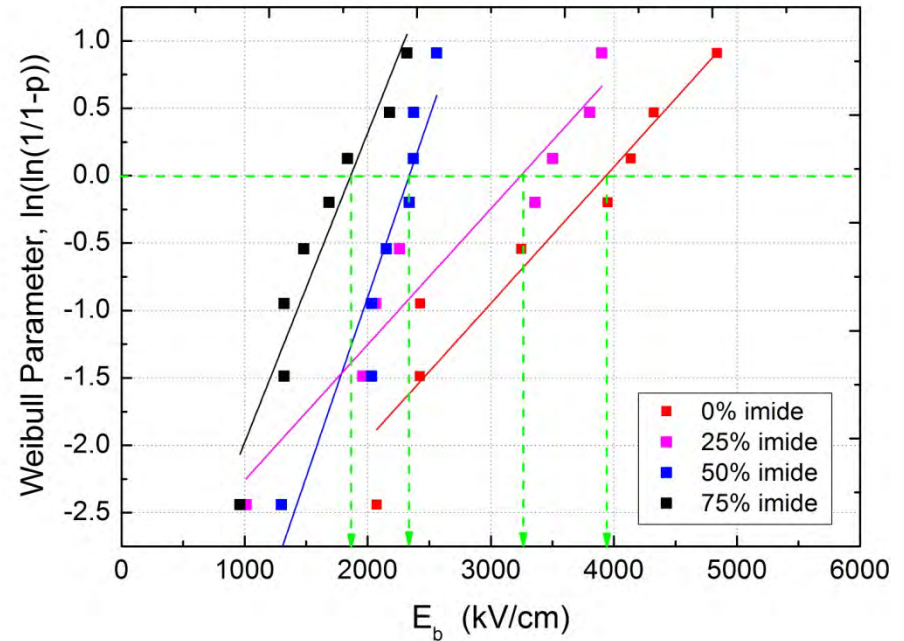
Stoichiometry optimization

- Temperature
- Dielectric properties



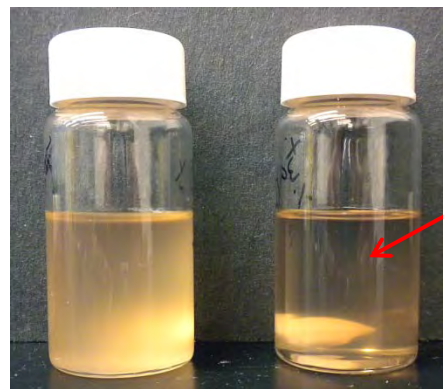
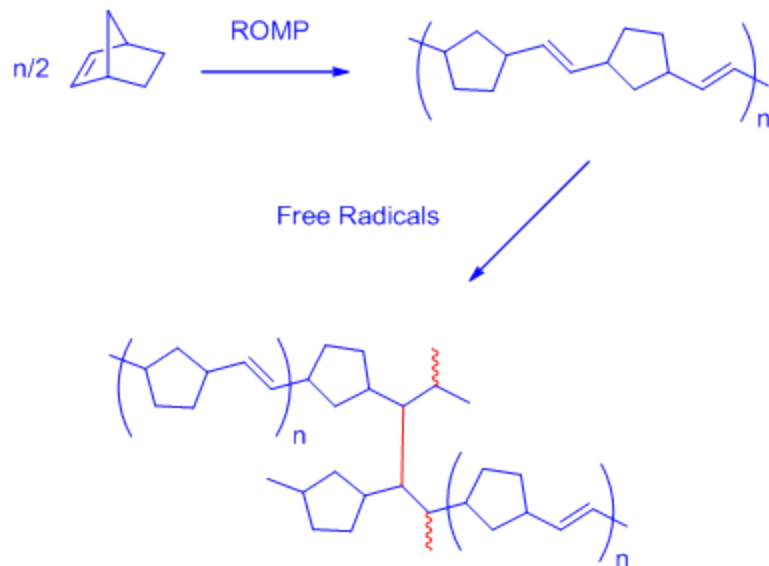
Survival probability function (Chegodayev)

$$P(i) = \frac{i - 0.3}{N + 0.4}$$



Technical Accomplishments

Spool of polymer thin film was produced at ECI using a solvent casting process.

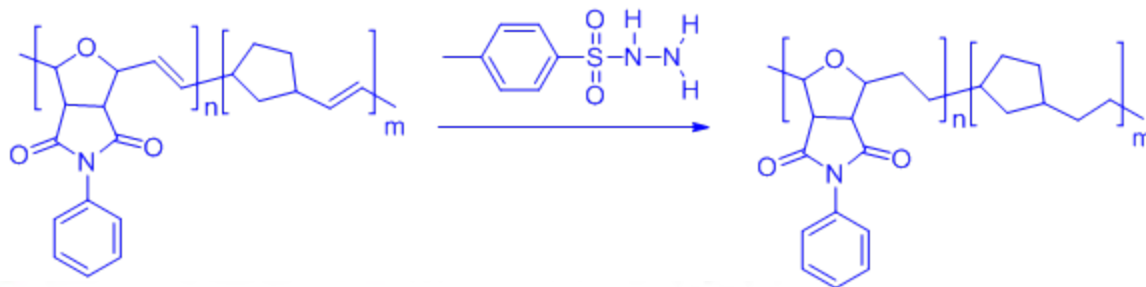


Inhibitor stops cross-linking temporarily (48 hours)

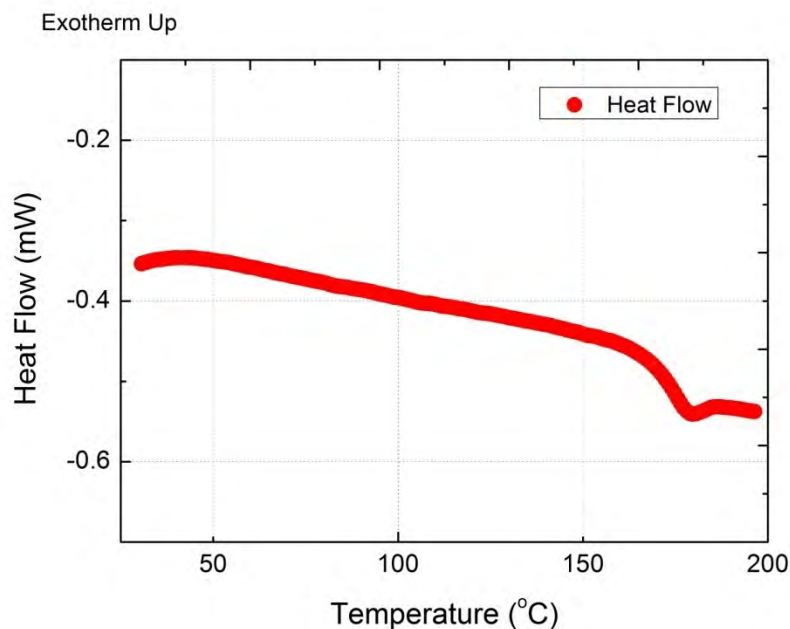
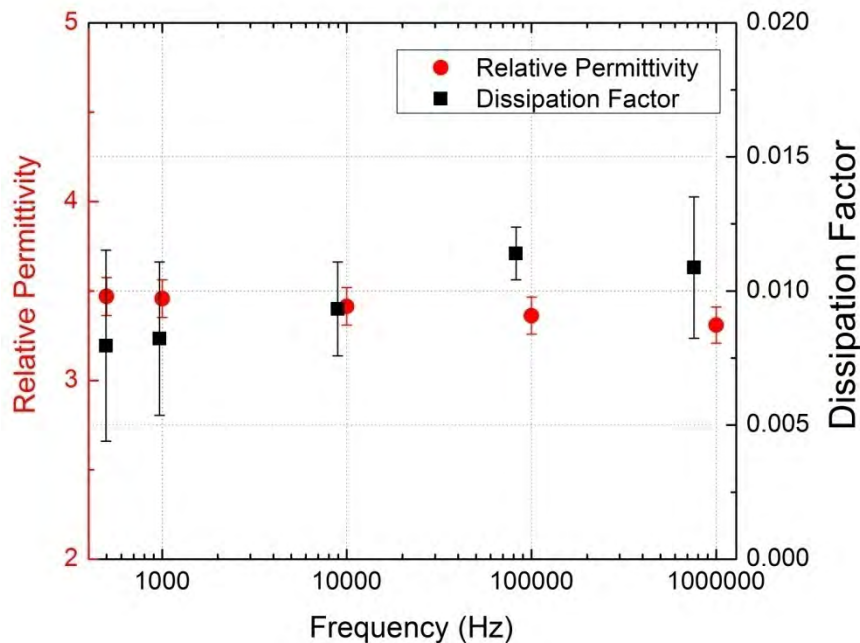
Cross-linking leads to polymer falling out of solution

Technical Accomplishments

In order to produce large lengths of polymer film we removed the double bond completely via a hydrogenation



Removal of the double bond also enables extrusion processing



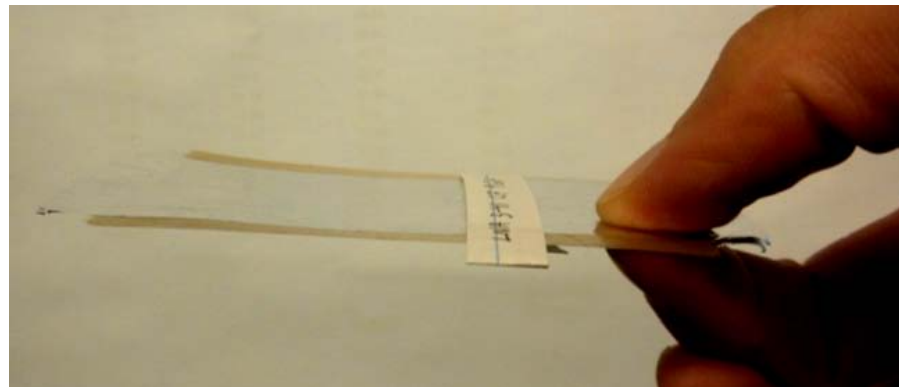
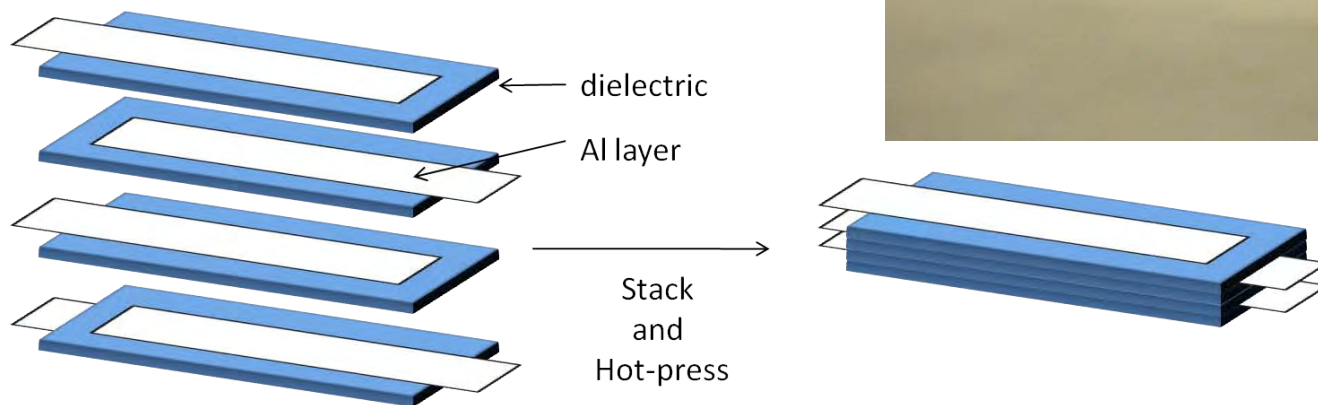
10

6.3 mm electrodes

~1 kg of hydrogenated polymer sent to ECI for film formation

Technical Accomplishments

Dielectric film processed using a drawdown machine was used as the starting material



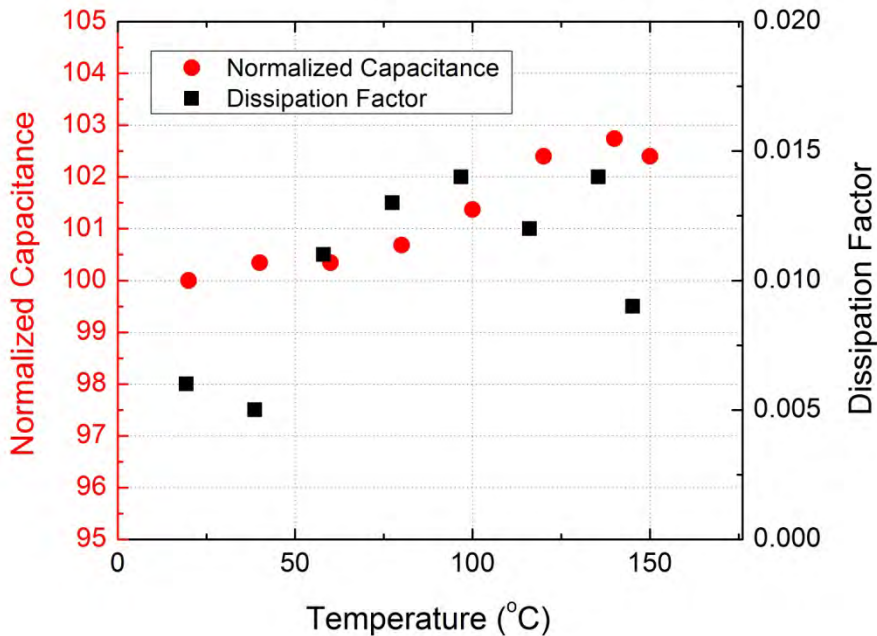
Capacitors were formed using a hot-press at a temperature above the T_g of the polymer to create a sealed capacitor. We have also begun to metalize the dielectric film with thin metal layers (50 nm) using sputter coating and have formed stacked capacitors.



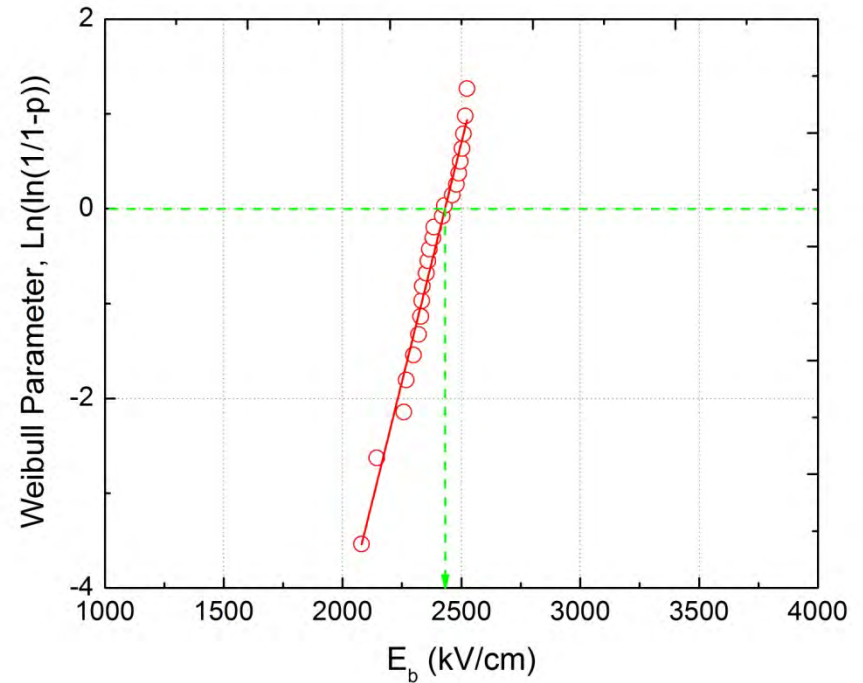
Technical Accomplishments

Stacked Capacitor Performance

Calculated Energy Density at 1 kHz: 0.88 J/cm^3



Three-layer stacked capacitor
with capacitance of 2.9 nF

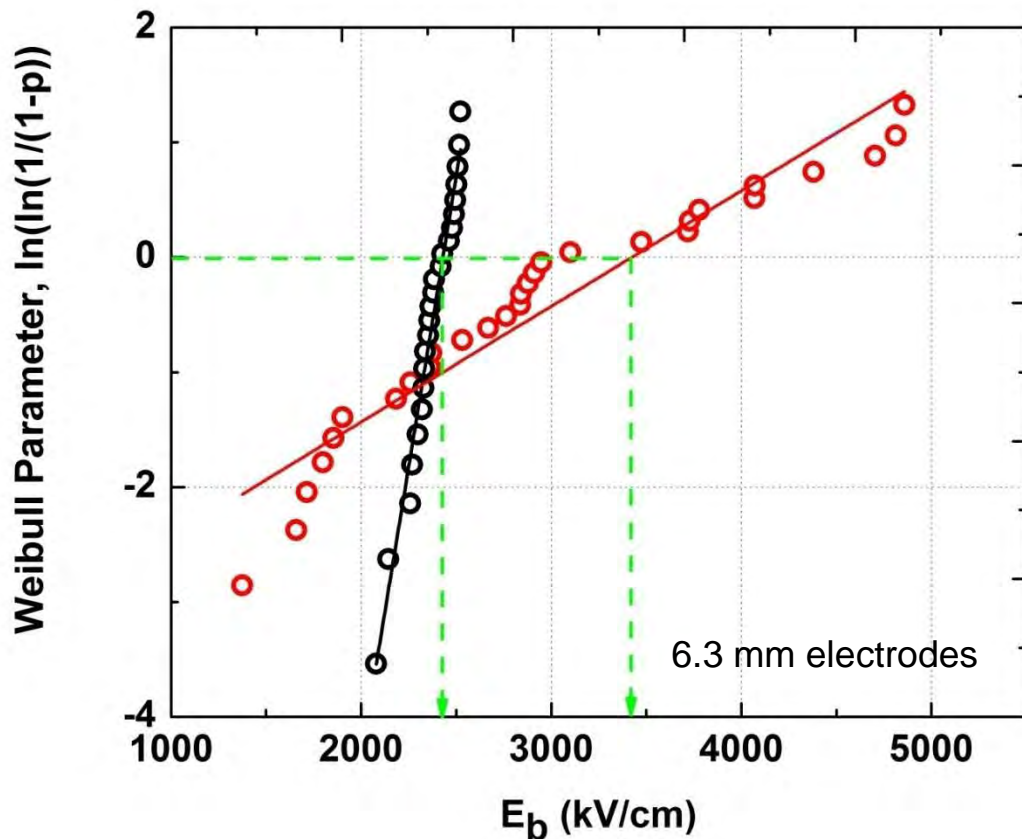


Survival probability function
(Chegodayev)

$$P(i) = \frac{i-0.3}{N+0.4}$$

Technical Accomplishments

Further improved the performance by removing side product from the hydrogenation



Survival probability function
(Chegodayev)

$$P(i) = \frac{i - 0.3}{N + 0.4}$$

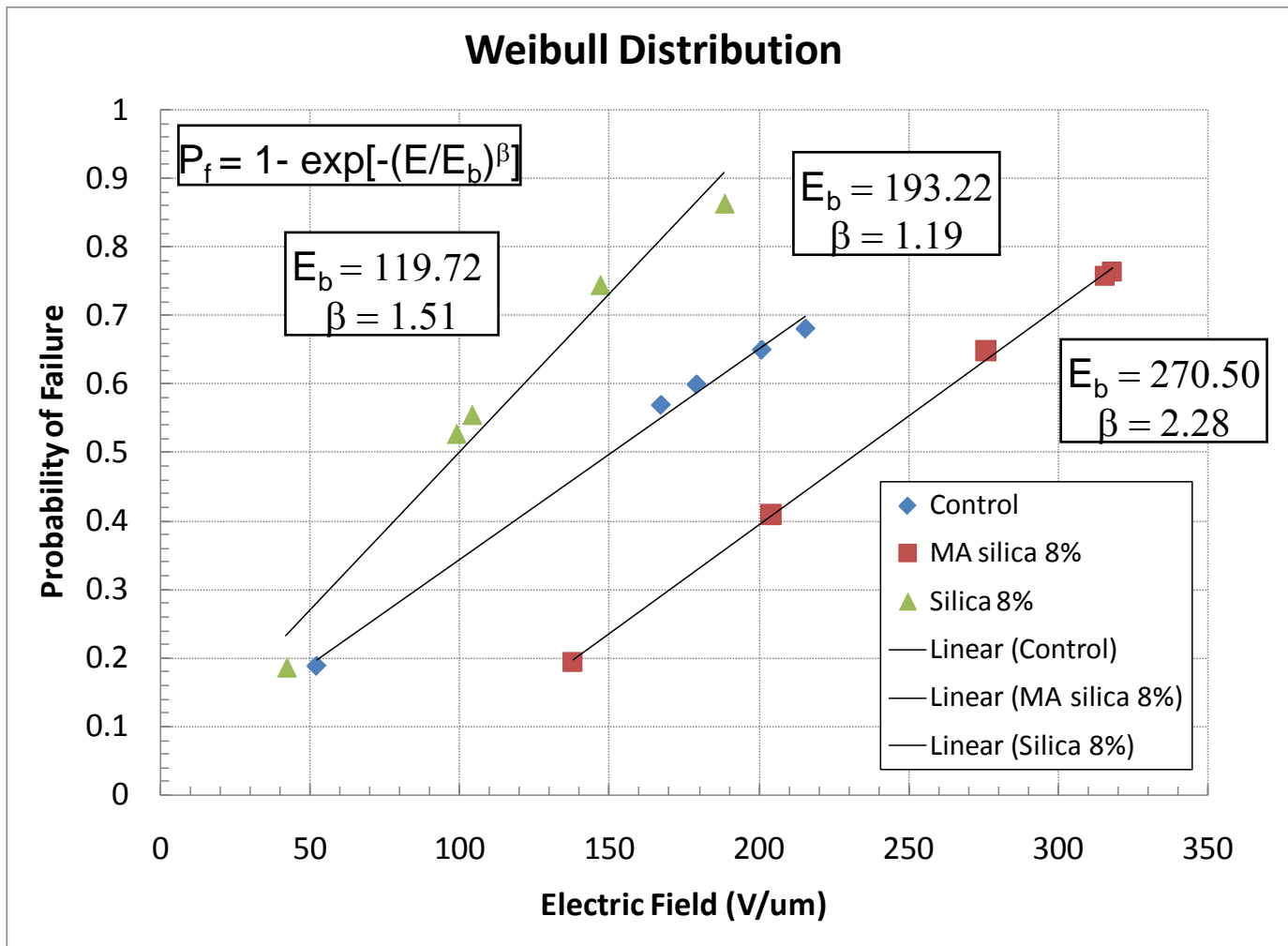
Aryl sulfinate anion



Fabricated small
rolled capacitors "in-house"

○ unpurified ○ Re-precipitated polymer

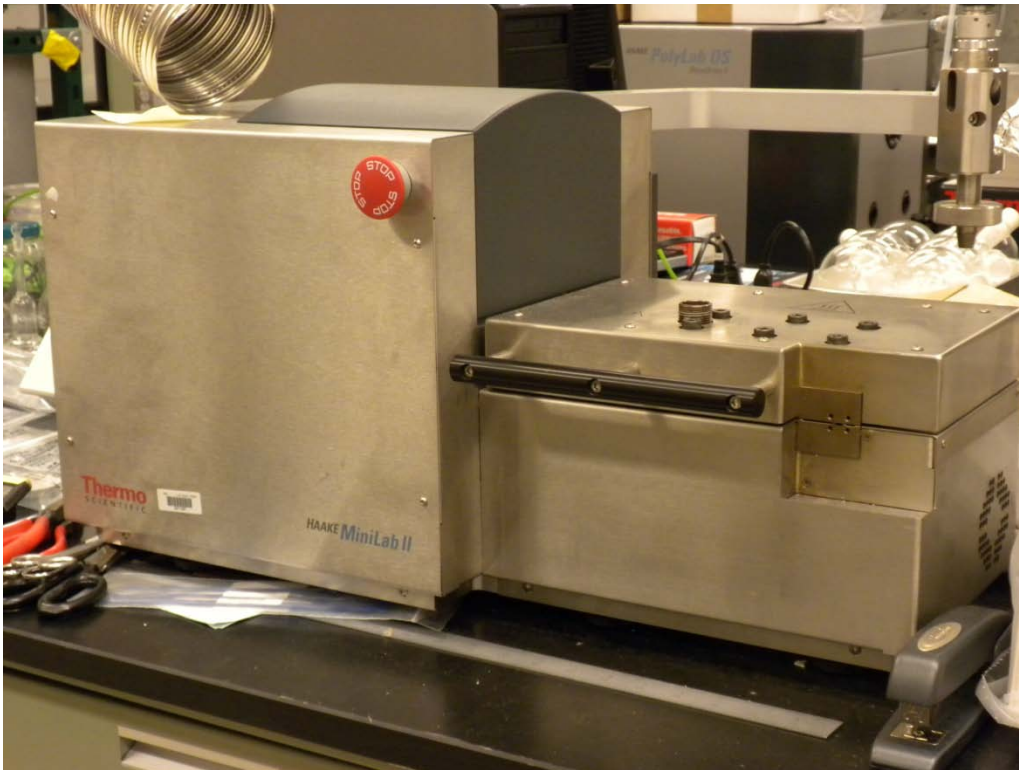
Technical Accomplishments



- To further improve energy density we are evaluating nanoparticle fillers to improve breakdown strength. This work was conducted with non-hydrogenated polymer. Developed process to disperse nanoparticles in many polymers

Technical Accomplishments

Extrusion experiments have been initiated. Easy to extrude polymer – harder to get thin films



Two extruders set-up and ready to use

Thermo Scientific
Haake MiniLab II – small scale extrusion evaluation

Thermo Scientific
Haake PolyLab OS – large “lab scale” film production”



Collaborations and Coordination with Other Institutions

- Working to cast polymer films
 - Joe Bond

- Coordination
 - Penn State
 - Mike Lanagan
 - Argonne National Laboratories
 - Uthamalingam (Balu) Balachandran





Future Work

- **Continue transition polymer film technology to industry - Producing films and prototype capacitors at ECI**
 - **A specific goal is to produce 100 m of capacitor film**
 - Fabrication of capacitors at ECI with film produced from hydrogenated polymer
- **Producing six stacked capacitors “in-house”**
- **If larger scale experiments using nanoparticle loaded material shows improvement in breakdown strength, begin production of prototype capacitors and evaluate**
 - **A specific goal will be the production of a prototype “stacked capacitor”**



Summary

- We have characterized the high temperature film electrical to provide the stoichiometry that meets high temperature performance metrics while allowing for film processing
- Working with ECI to produce prototype capacitors and solving problems as they occur related to transitioning from a laboratory to a pilot scale operation
 - Exploring several options including thiol-ene, hydrogenation, and radical inhibitors to improve polymer solution stability. Hydrogenation looks most promising.
 - Sent hydrogenated material to ECI
- Working to produce “in-house” fabricated stacked capacitors -
Delivered very small capacitor to ORNL
- Developing nanocomposite chemistry to increase energy density of high temperature dielectrics