



High Temperature Polymer Capacitor Dielectric Films

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APE009



Overview

Timeline

- Project start: October 2008
- Project end: September 2011
- Percent complete (63%)

Budget

- Total project funding
 - DOE: \$950k
- Funding received in FY08 and FY09 include:
 - FY08 - \$100k
 - FY09 - \$150k
 - FY10 - \$750k

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

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

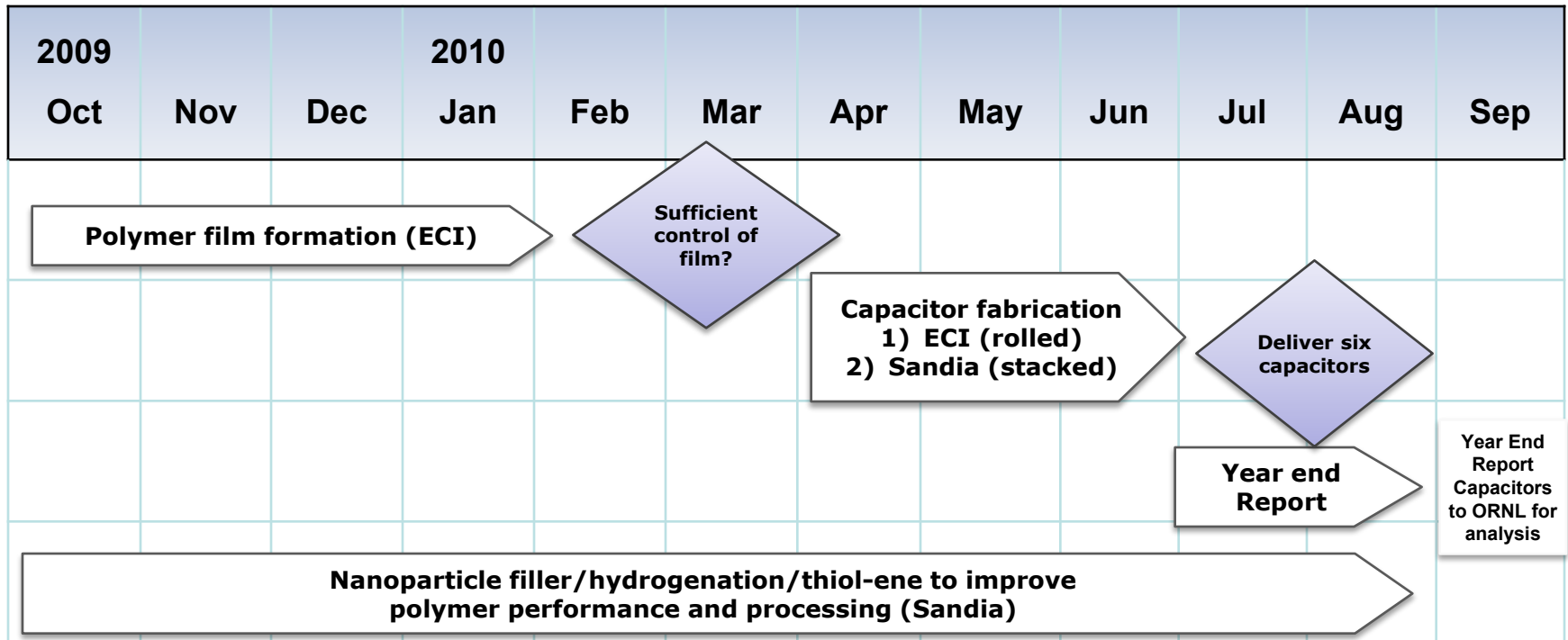
- Current capacitors lack the temperature, size, and price specifications required for future DC bus capacitors. Our approach simultaneously increase operational temperature (150 °C), decreases size, and lower the price of high temperature capacitors (\$0.015/μF), while maintaining self-healing properties.

Uniqueness and Impacts

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

Milestones

Project schedule



Go No/Go Decision Point: Is the polymer film formation process sufficient to produce 100 m of polymer film?

Challenges/Barriers: Optimizing film casting procedures to produce polymer film sufficient for rolled capacitor formation. As a fall back option, we will produce stacked capacitors at Sandia

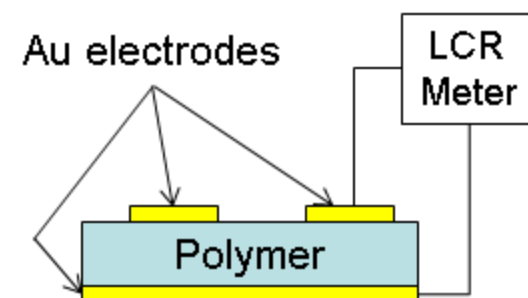
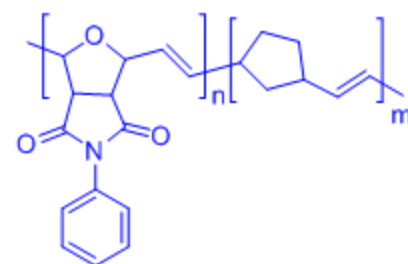
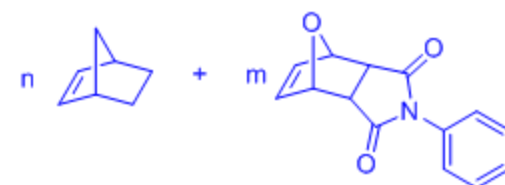
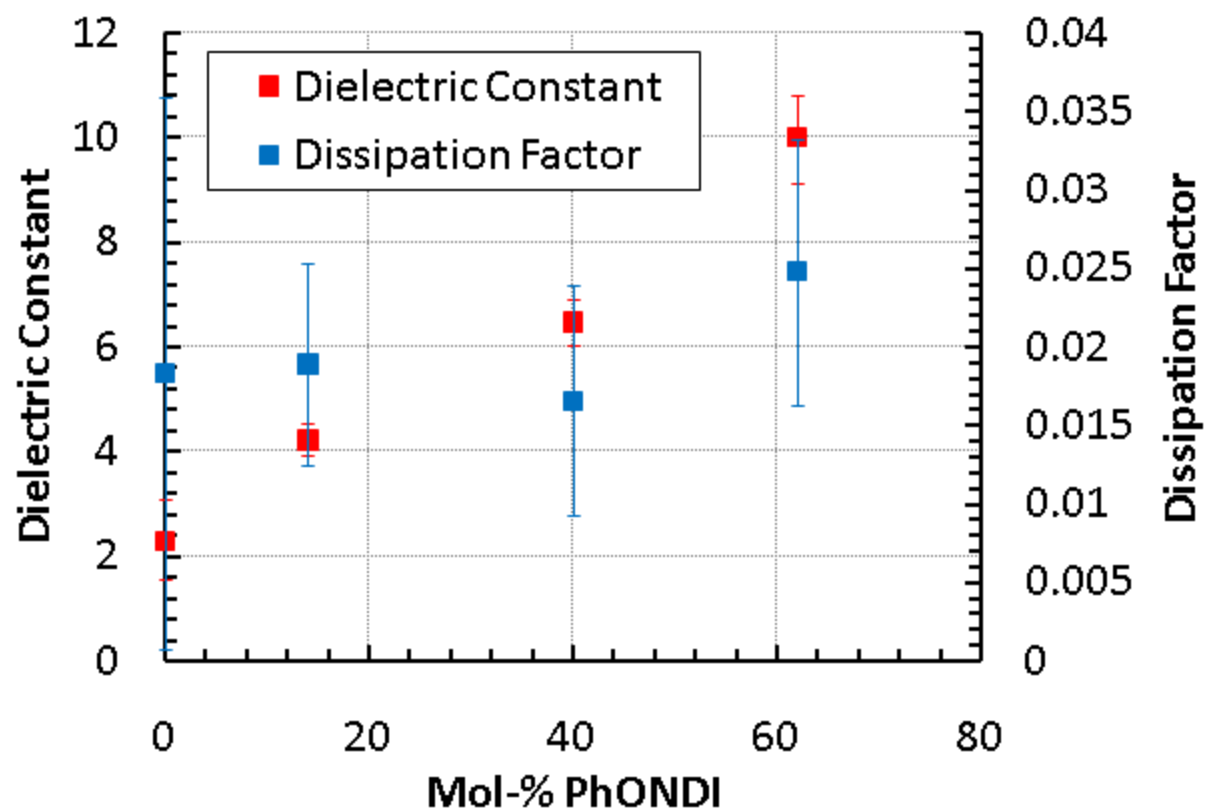


Approach

- **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 in FY09 by modifying polymer chemistry while maintaining the appropriate dielectric performance.
 - Hydrogenation reaction
 - Thiol-ene reaction
- **Develop nano-composites of high temperature polymer dielectrics to improve energy density**

Technical Accomplishments

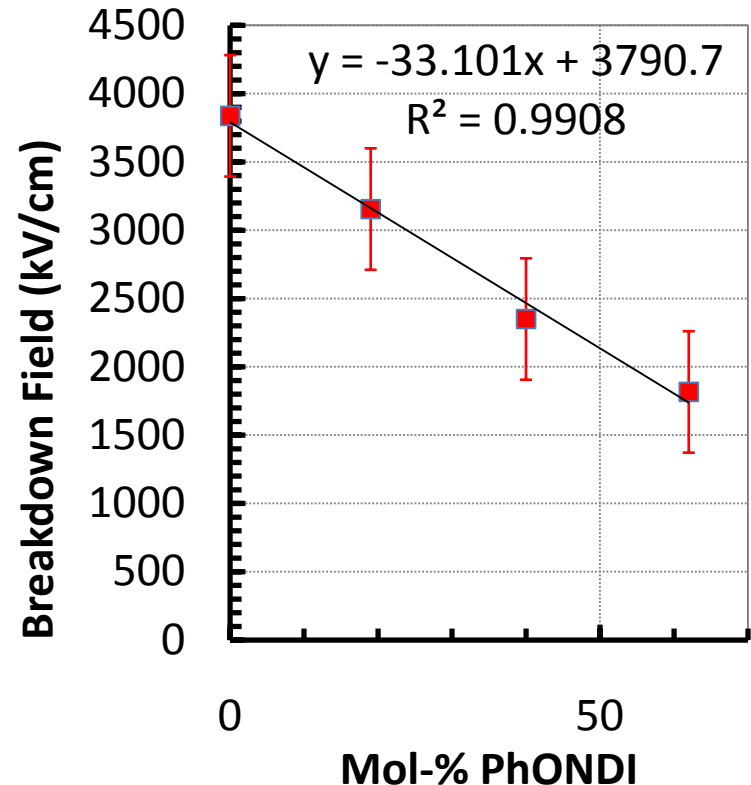
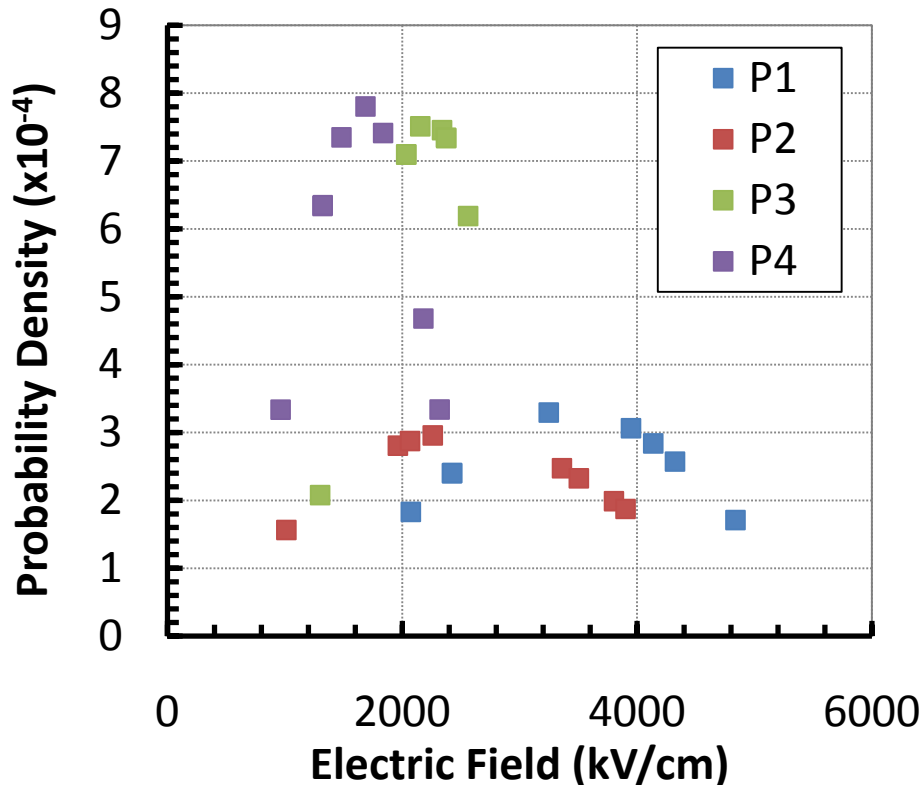
Polymer	% PhONDI	% Norbornylene	% PhONDI by NMR	% Norbornylene by NMR
P1	0	100	0	100
P2	25	75	14	86
P3	50	50	40	60
P4	75	25	62	38



Technical Accomplishments

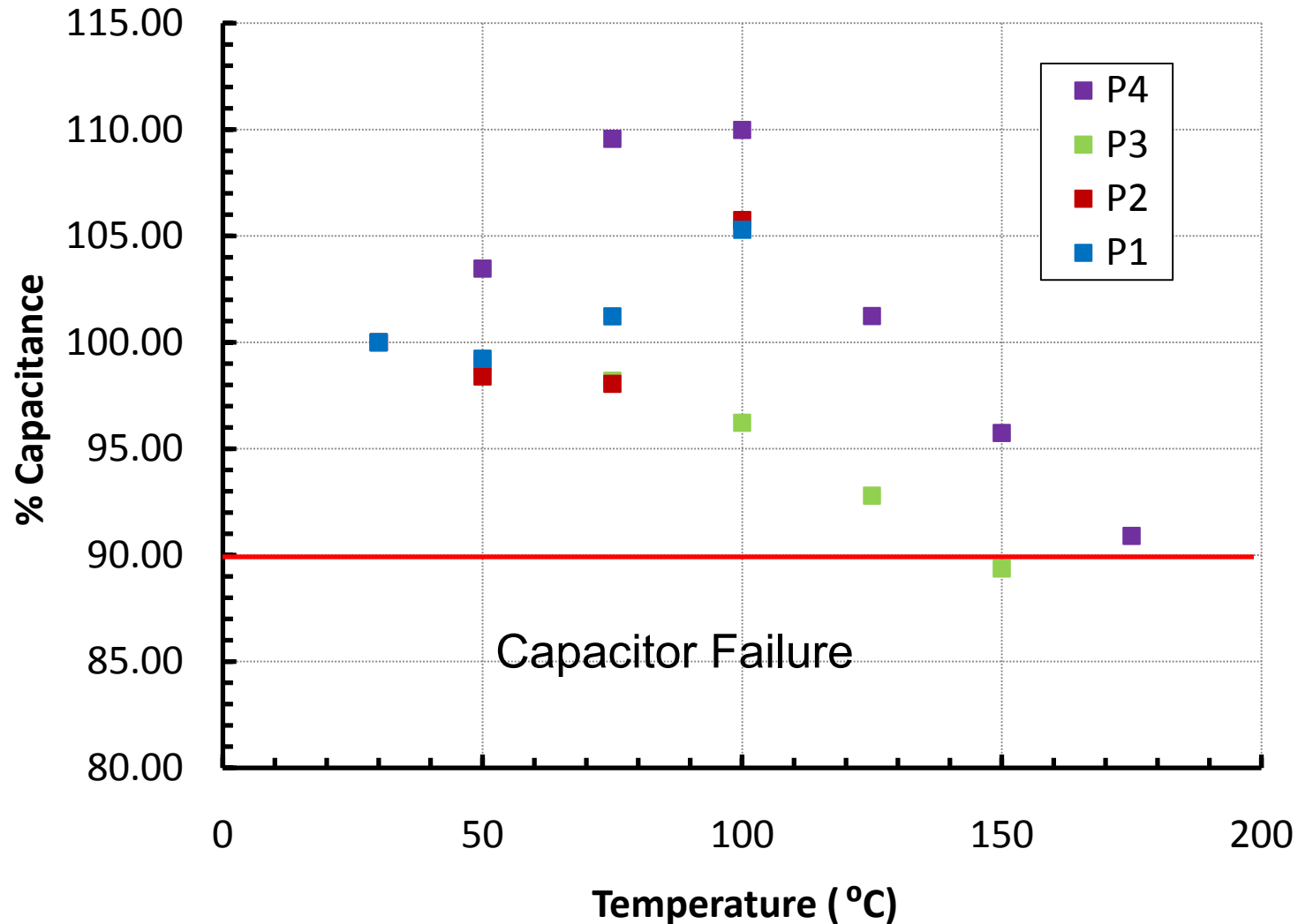
$$f(E, E_0, \beta) = \frac{\beta}{E_0} \left(\frac{E}{E_0} \right)^{\beta-1} e^{-\left(\frac{E}{E_0} \right)^\beta}$$

Meets temperature and energy density goals (energy densities range from 1.46 - 1.86 J/cm³ but we are not done yet...what about high temperature operation?



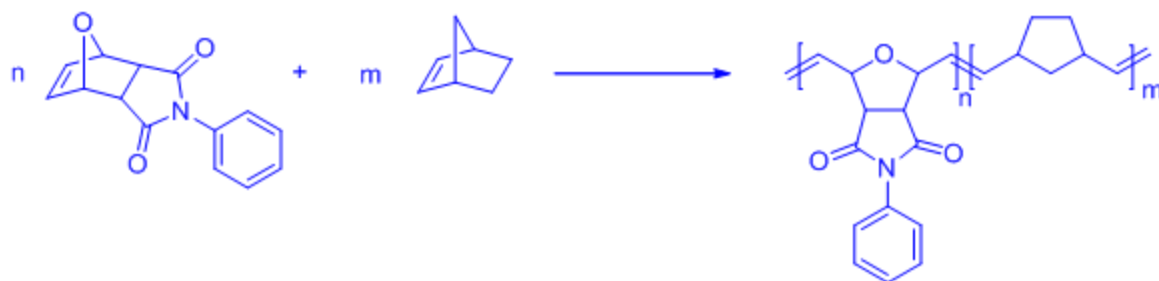
P1 = 0% PhONDI $E_0 = 3837$ kV/cm $\beta = 3.30$, **P2** = 14% PhONDI $E_0 = 3154$ kV/cm $\beta = 2.28$,
P3 = 40% PhONDI $E_0 = 2349$ kV/cm $\beta = 4.73$, **P4** = 62% PhONDI $E_0 = 1816$ kV/cm $\beta = 3.70$

Technical Accomplishments



P1 = 0% PhONDI, P2 = 14% PhONDI, P3 = 40% PhONDI, P4 = 62% PhONDI

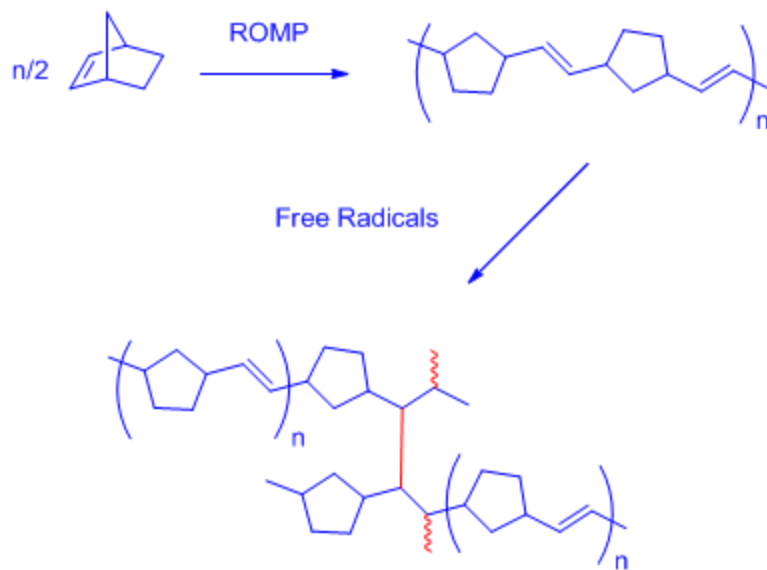
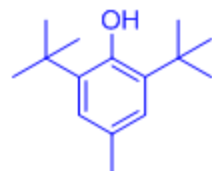
Technical Accomplishments



- Working with ECI to fabricate rolls of polymer thin film using a solvent casting process. A spool of 100 m length is required to fabricate rolled capacitors.
- Still working to solve scale-up problems
 - Solvent change led to improved film formation, but solution stability is still a problem...

Technical Accomplishments

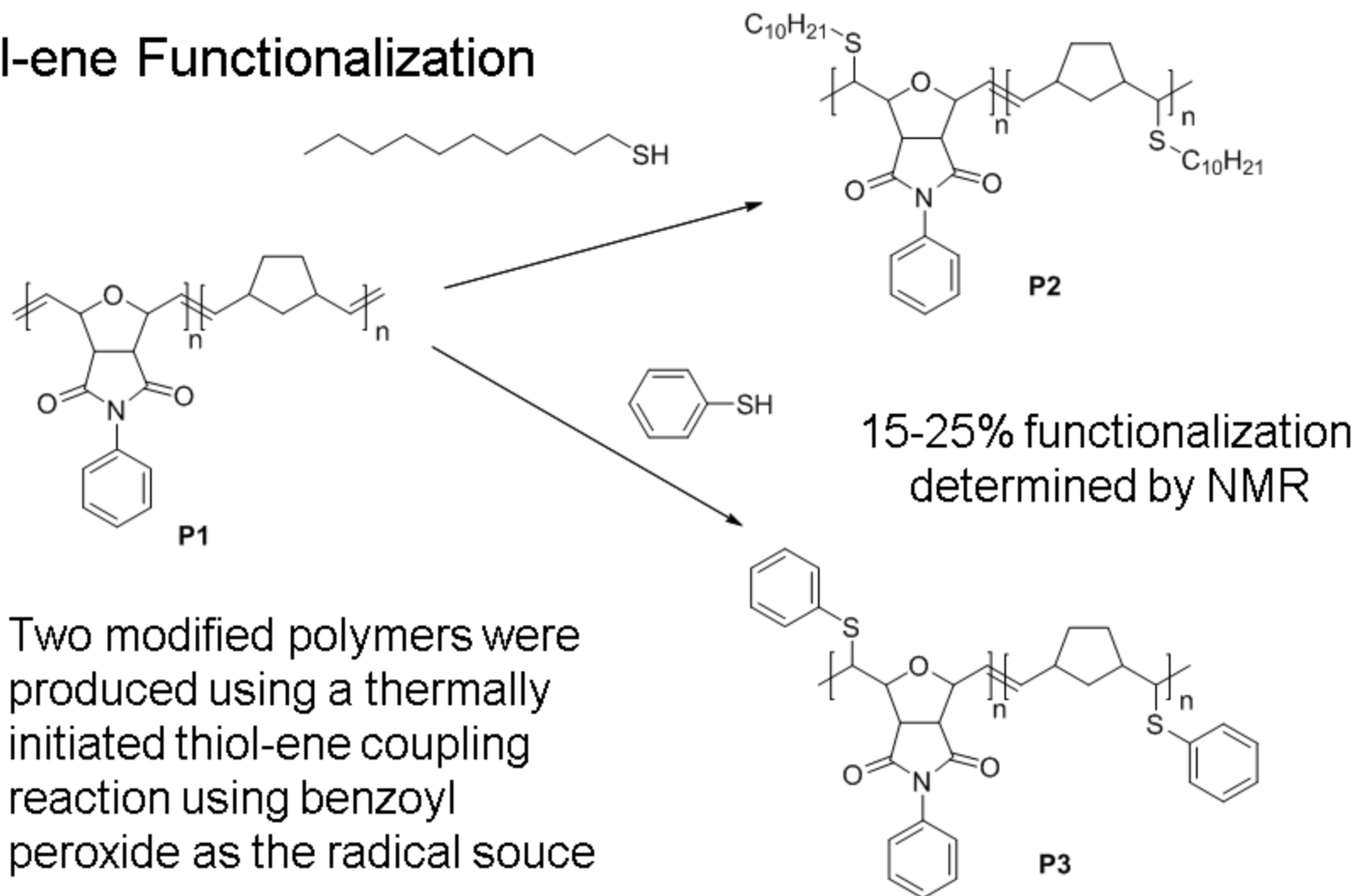
- Identified the double bonds as a source of gelation that shortens polymer solution lifetimes.
- Exploring two chemical methods to remove double bonds without sacrificing dielectric properties.
 - Hydrogenation
 - Thiol-ene reaction
- Exploring classical double bond containing solution stabilization methodology using free radical inhibitors like BHT



Crosslinking leads
to polymer falling
out of solution

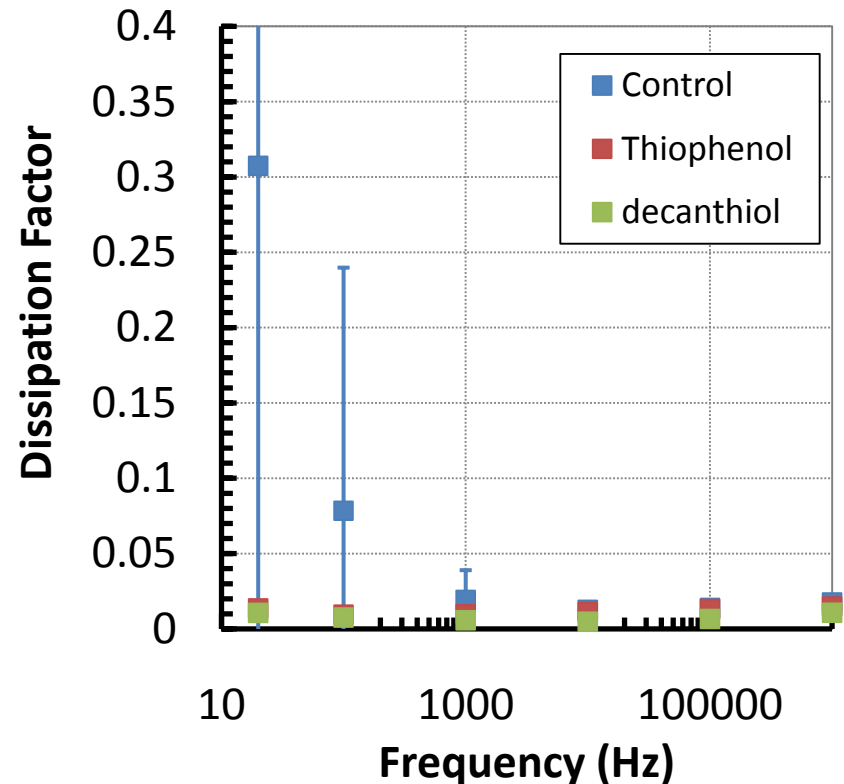
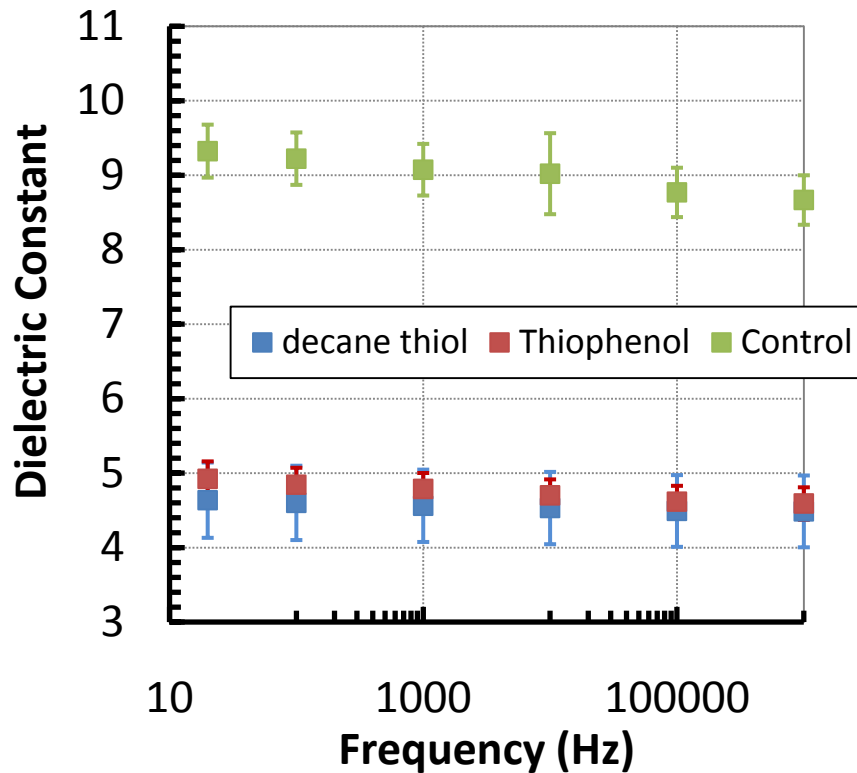
Technical Accomplishments

Thiol-ene Functionalization



- Two modified polymers were produced using a thermally initiated thiol-ene coupling reaction using benzoyl peroxide as the radical source

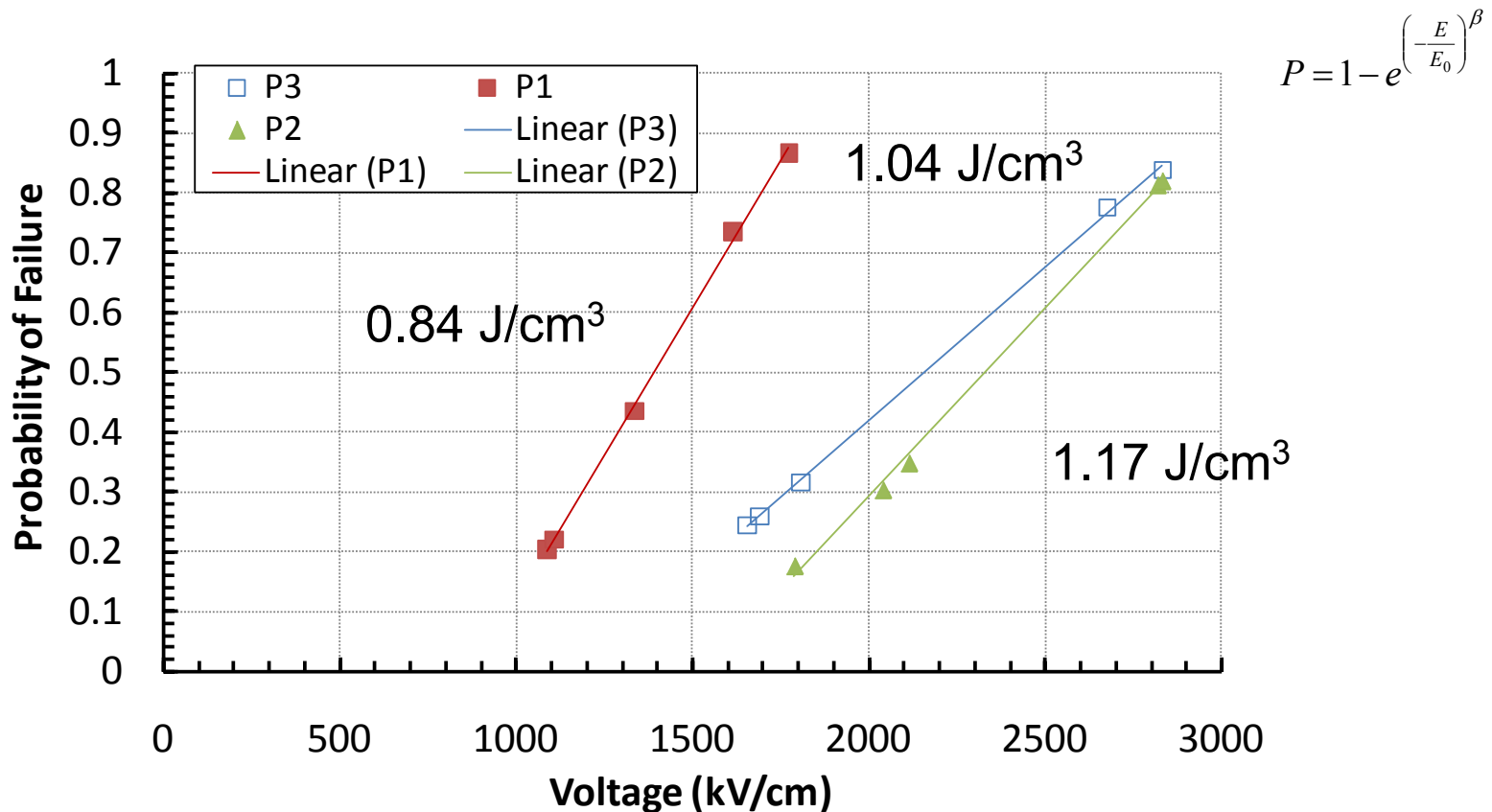
Technical Accomplishments



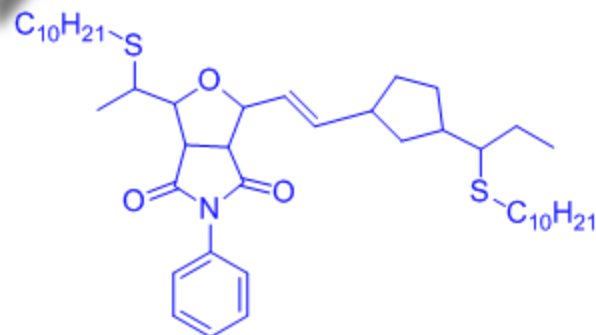
The inclusion of the thioether decreases dielectric constant, however, the dissipation factors are improved at low frequencies due to the removal of a fraction of double bonds in the polymer backbone.

Technical Accomplishments

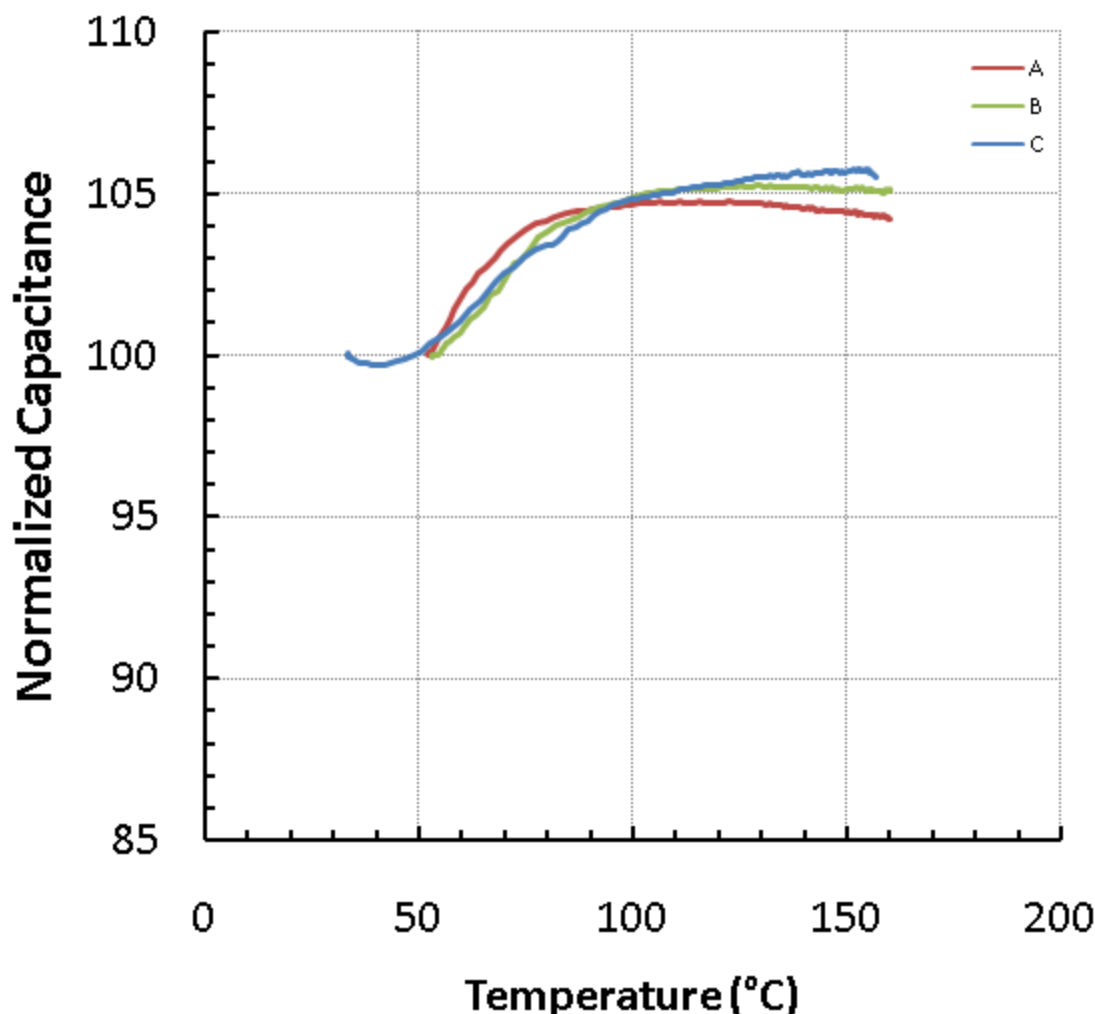
- The inclusion of the thioether improves breakdown strength by reducing the fraction of double bonds in the polymer backbone.



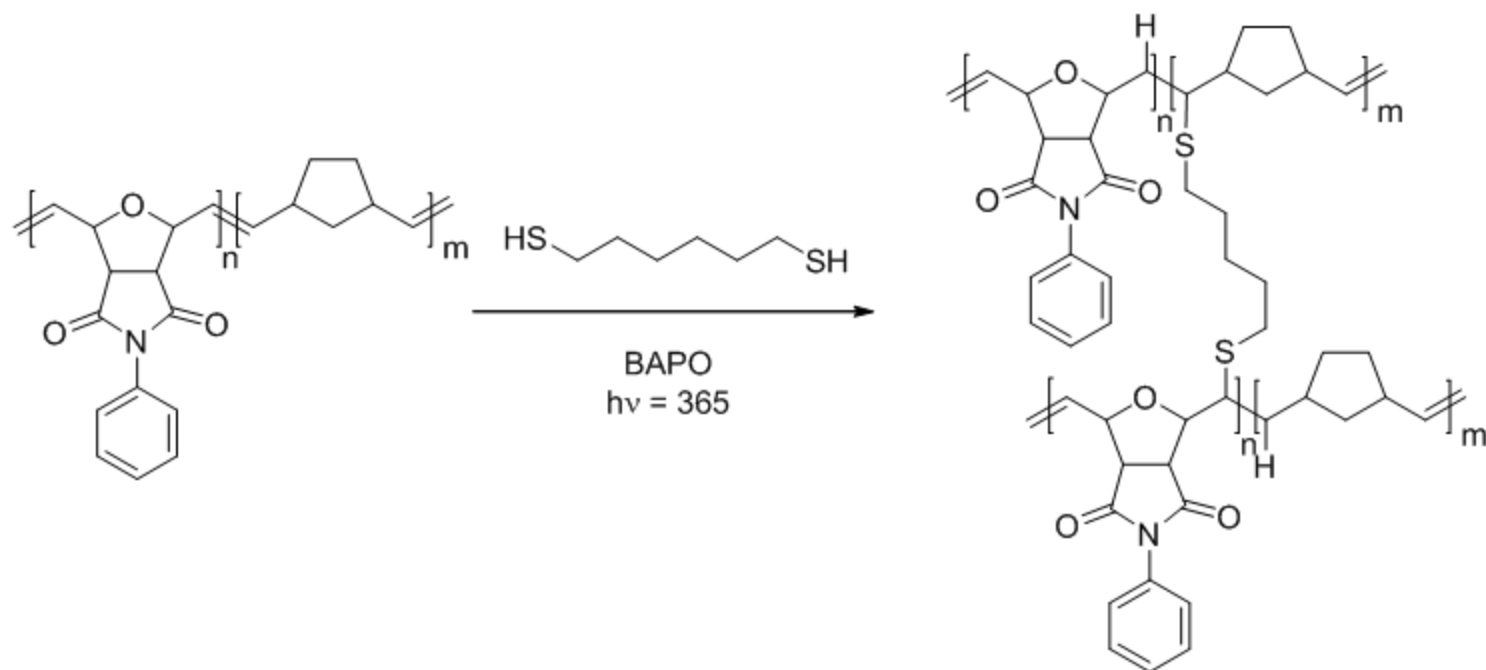
Technical Accomplishments



- Polymer films were cast (much more soluble than precursor polymer)
- Dozens of 50 pF capacitors were fabricated and evaluated.
- Dielectric breakdown strength increased 27% as ~15% of the double bond content was removed.



Technical Accomplishments



BAPO

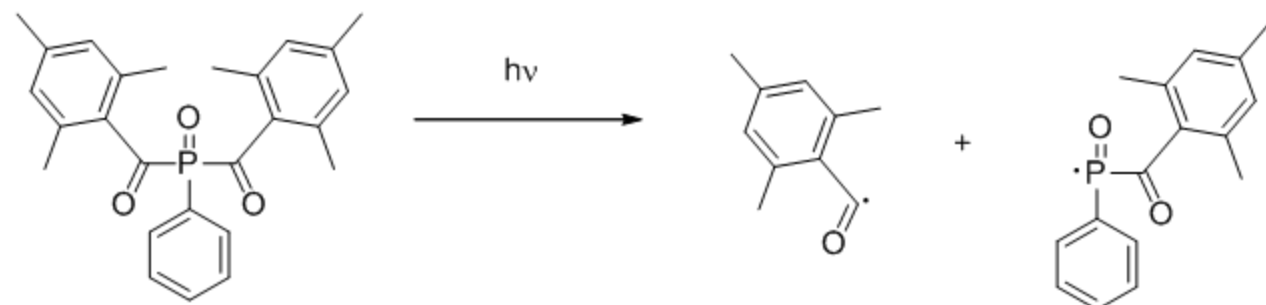
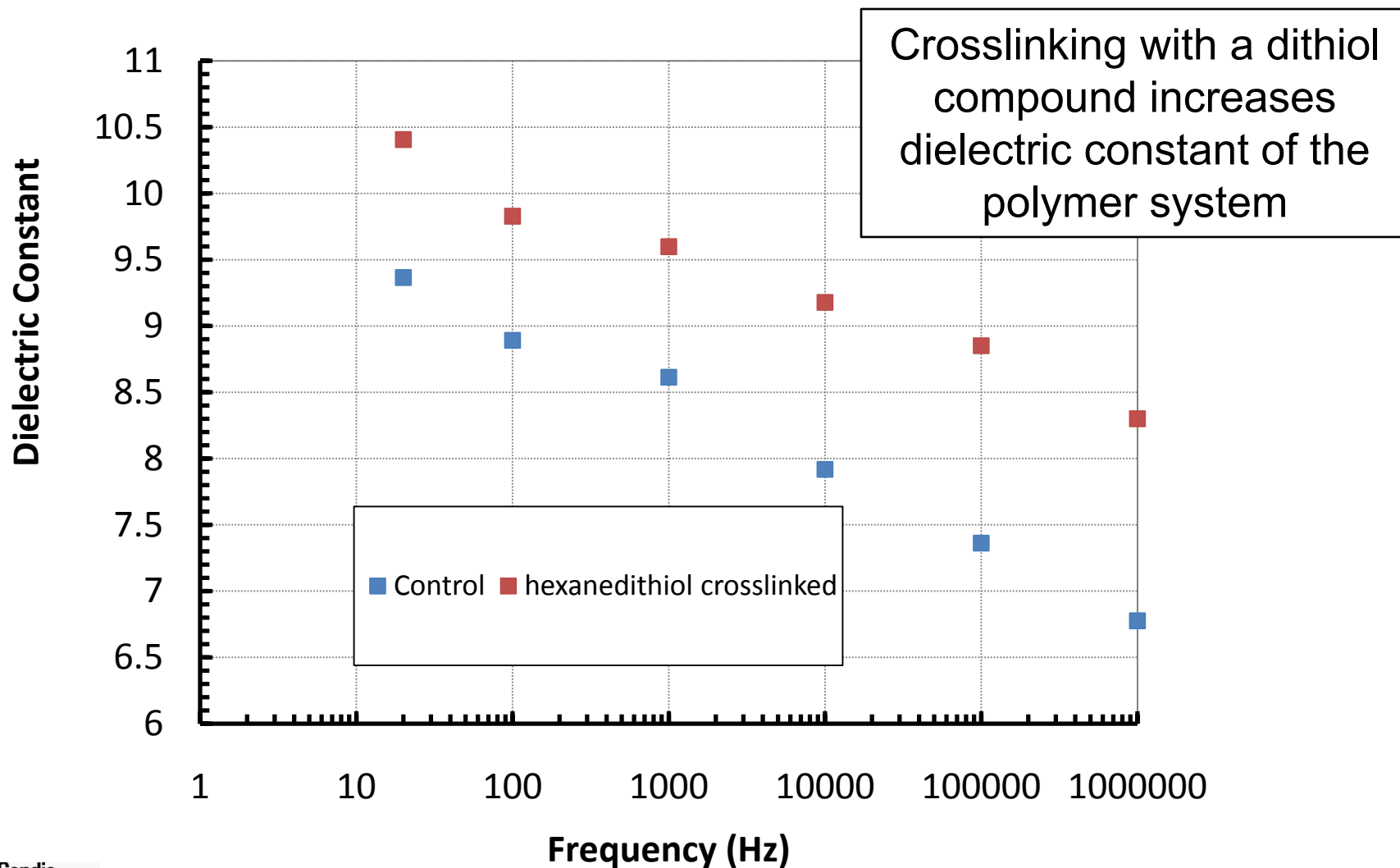
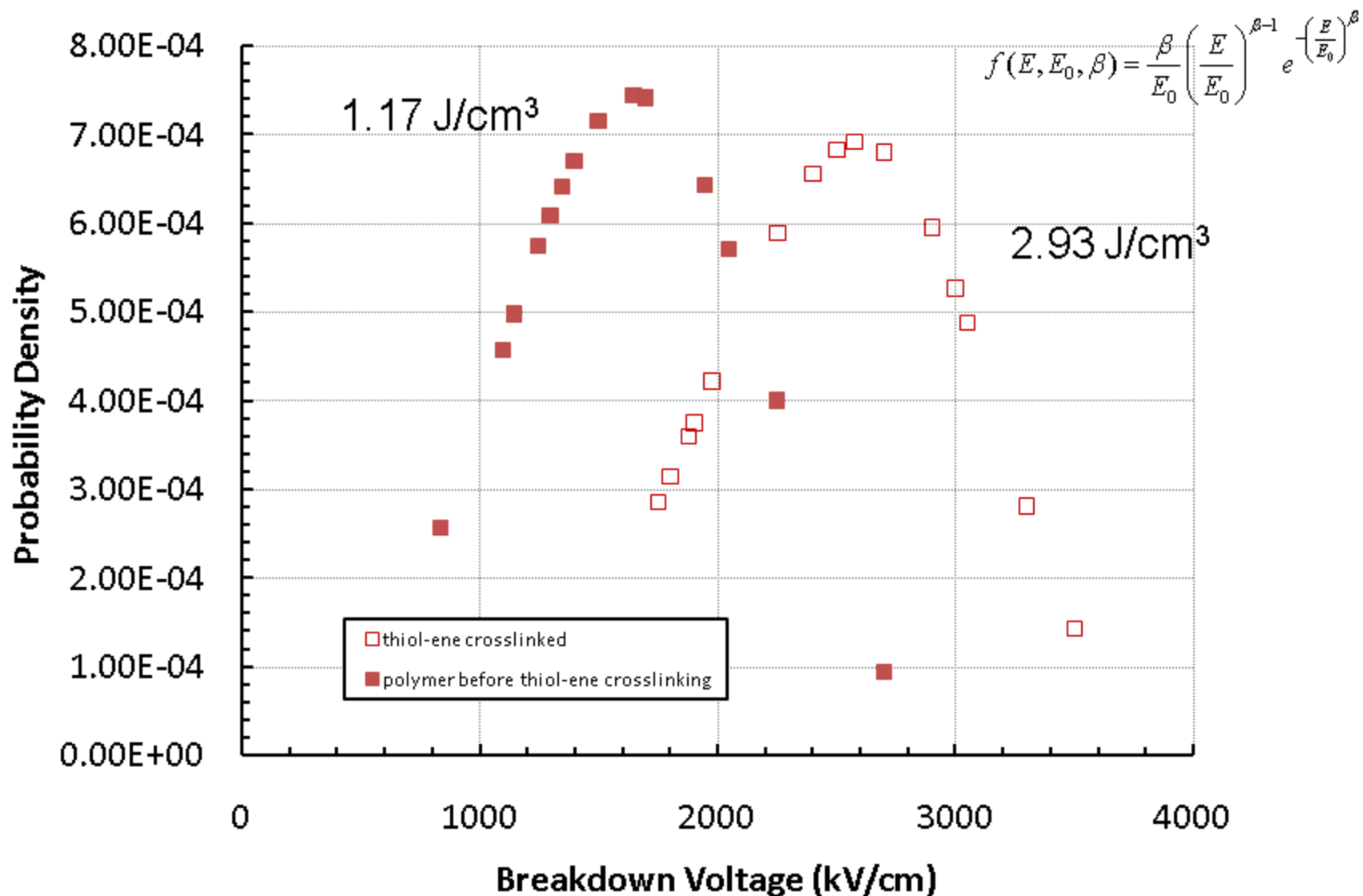


Photo-crosslinking
(with 10 % thiol)
removes double
bonds using a
process that is easily
transferred to
industry

Technical Accomplishments

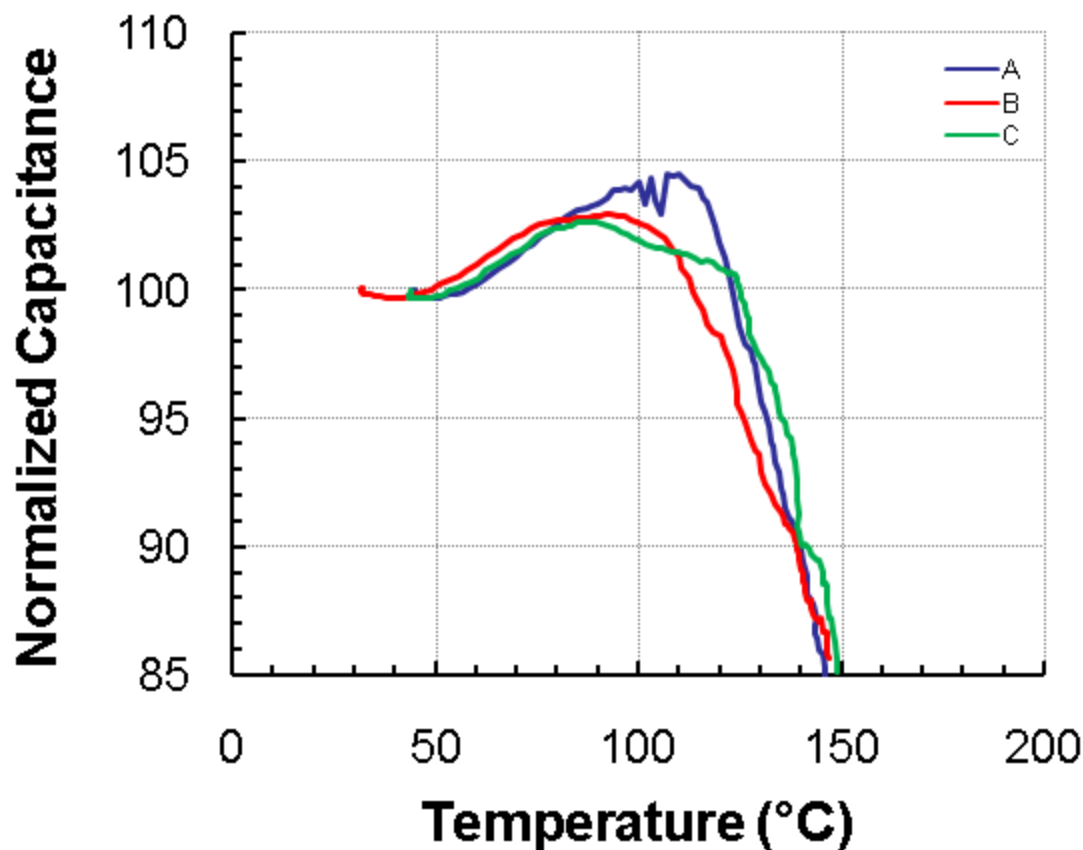
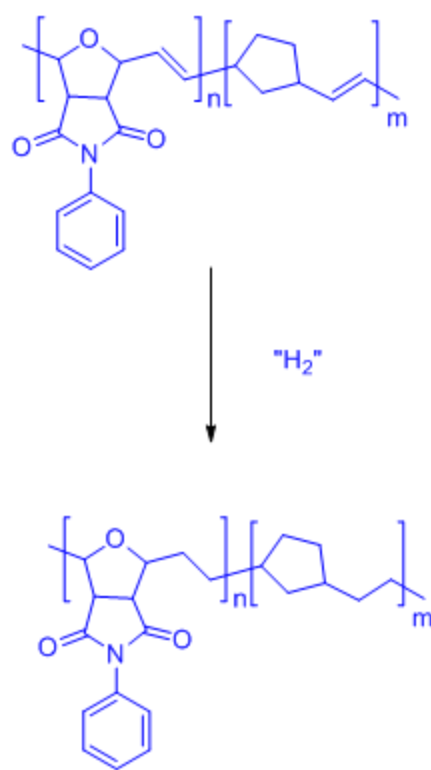


Technical Accomplishments

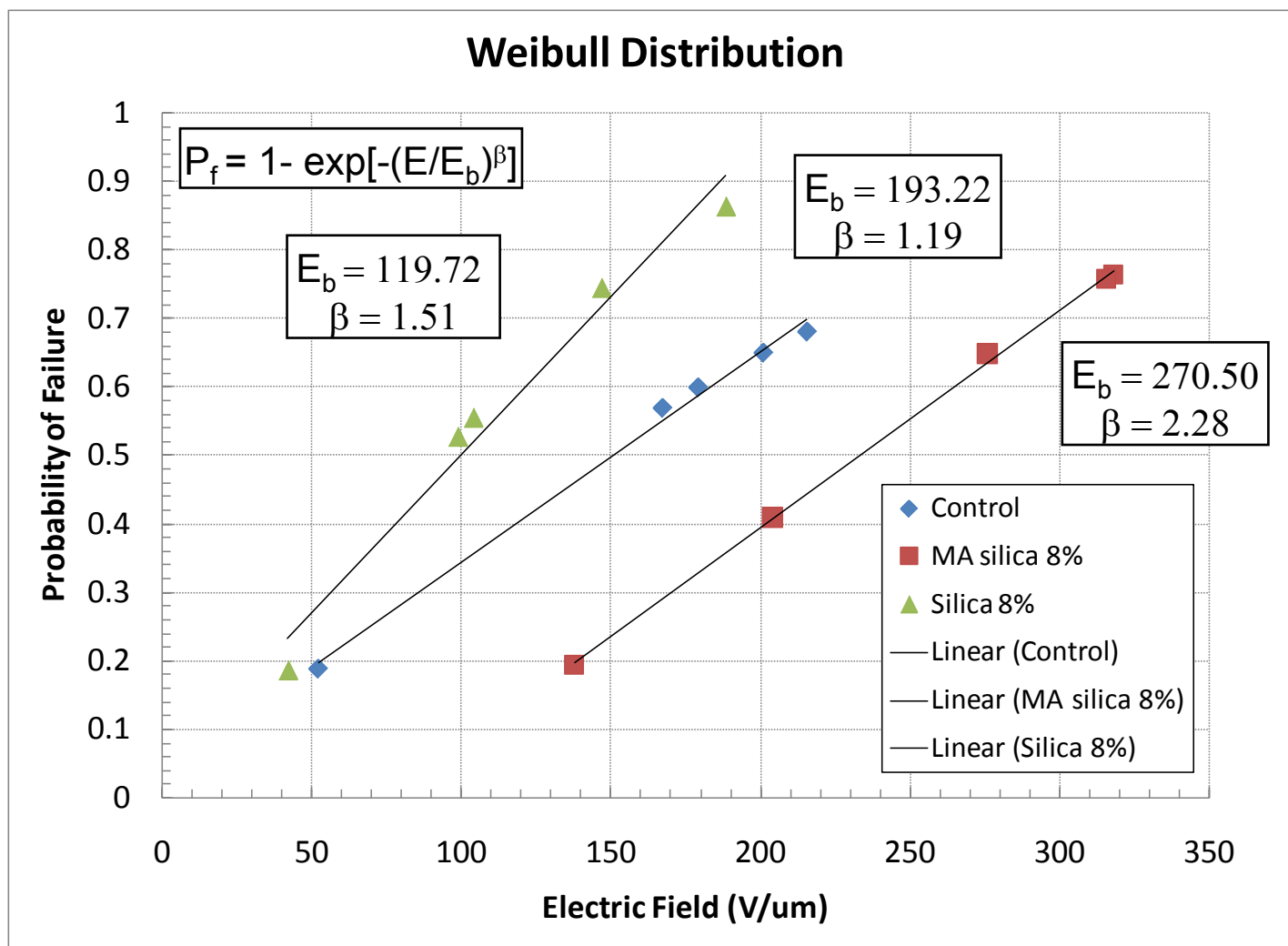


Technical Accomplishments

- Removal of the double bonds will eliminate free radical induced crosslinking and enable extrusion of the polymer.

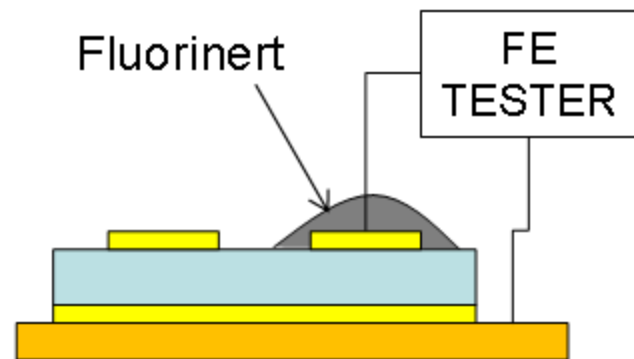
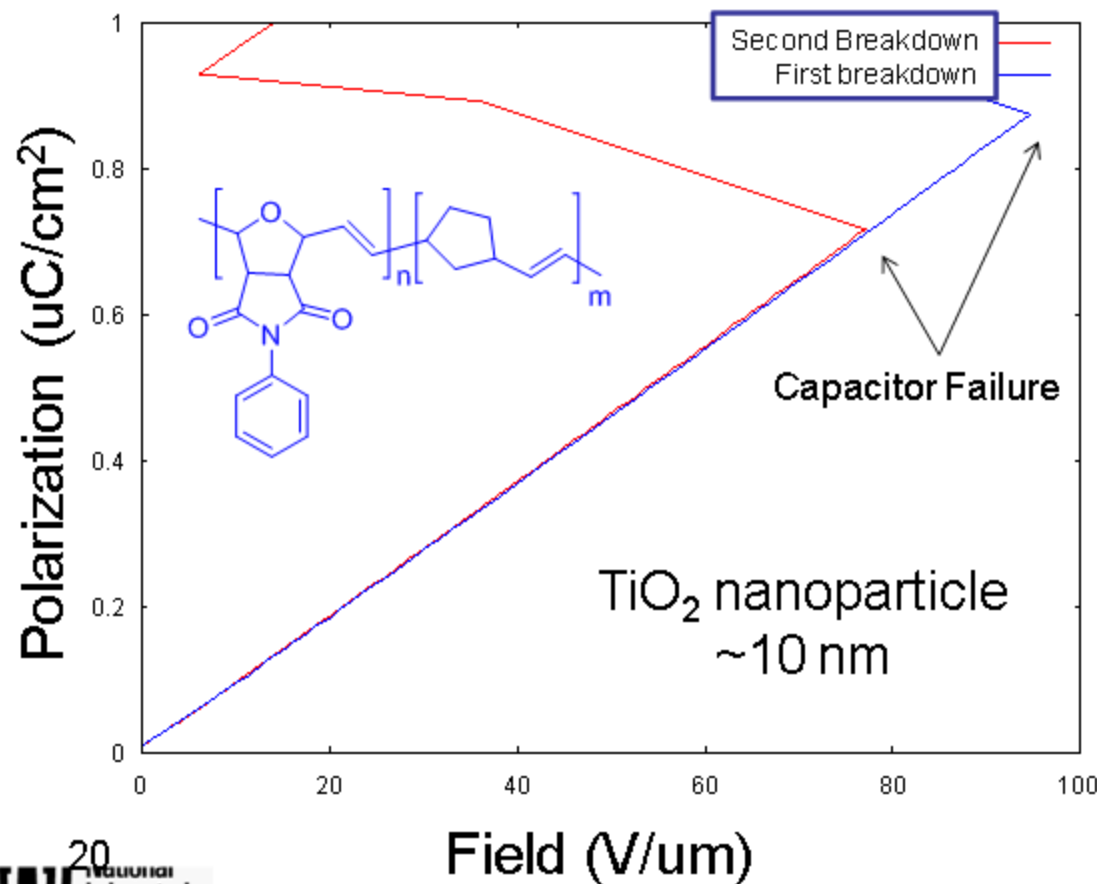
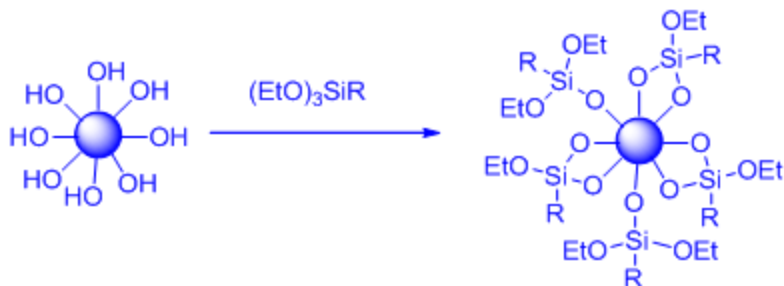


Nanoparticle Additives



- To further improve energy density we are evaluating nanoparticle fillers to improve breakdown strength.

Accomplishments to Date



- Nanoparticle filled dielectric based capacitors were evaluated using a ferroelectric tester. These capacitors demonstrate graceful failure mechanism repeatedly.



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**
 - Scale-up synthesis of thiol-ene polymer formulation to transition to ECI for casting
- Producing six stacked capacitors “in-house”
- If nanoparticle loading 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.
- Working to produce “in-house” fabricated stacked capacitors
- Developing nanocomposite chemistry to increase energy density of high temperature dielectrics