Radiation Resistant Electrical Insulation Materials for Nuclear Reactors Using Novel Nanocomposite Dielectrics

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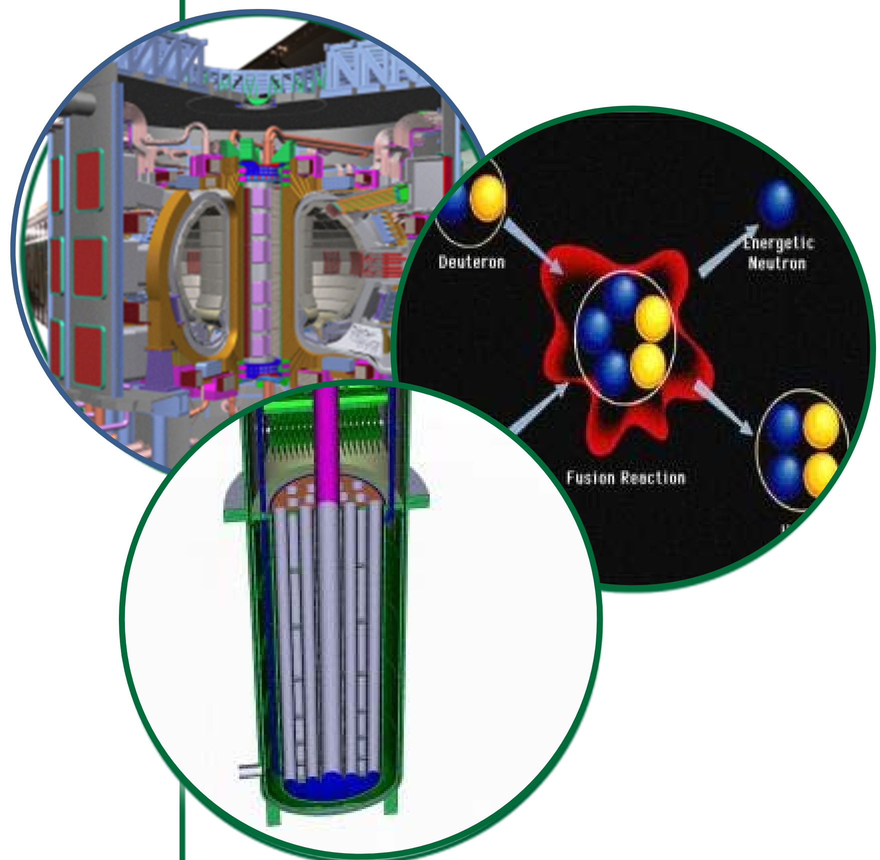

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Enabling Technology

U.S. DEPARTMENT OF ENERGY

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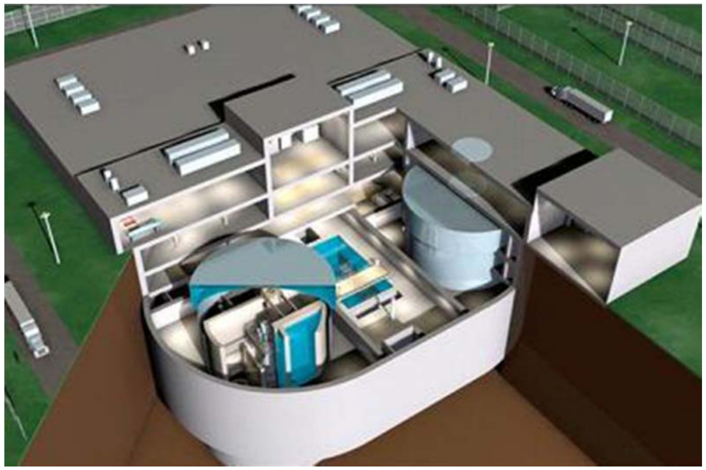
Project goal is to ***develop transformative materials using nanocomposite dielectrics for nuclear environments***

- **Due to recent renewed interest in reactor safety and many reactors approaching end of useful lifetime, emphasis on durability of power and instrumentation cabling is growing**

- **While current materials have shown suitable radiation tolerance in lab testing, combined effects of radiation, temperature, and water at normal or abnormal conditions have led to cable failures**



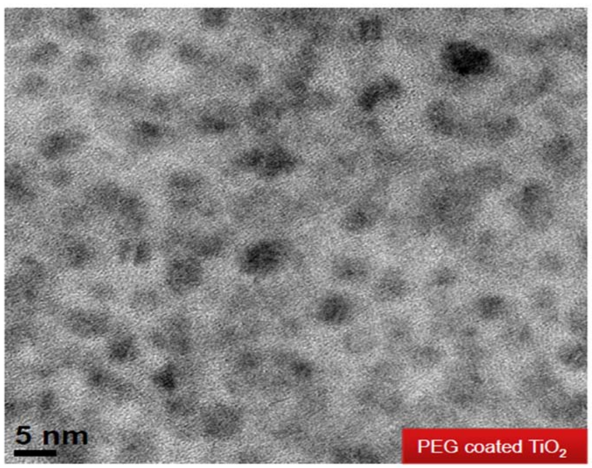
Cutaway of 2-Unit Generation mPower SMR
(courtesy of DOE NE)

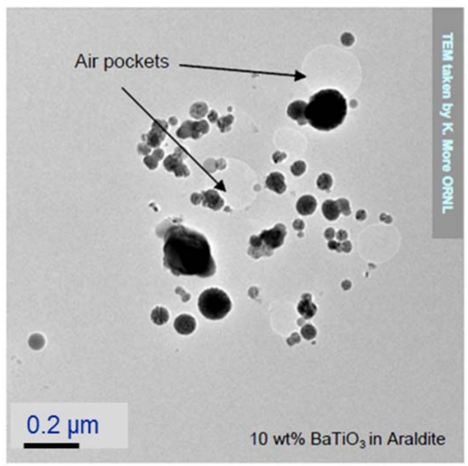
Example of degraded cables in LWR
(courtesy of Gary Toman, EPRI)

New dielectrics could help in current and future reactor deployment with respect to lifetime and performance enhancements

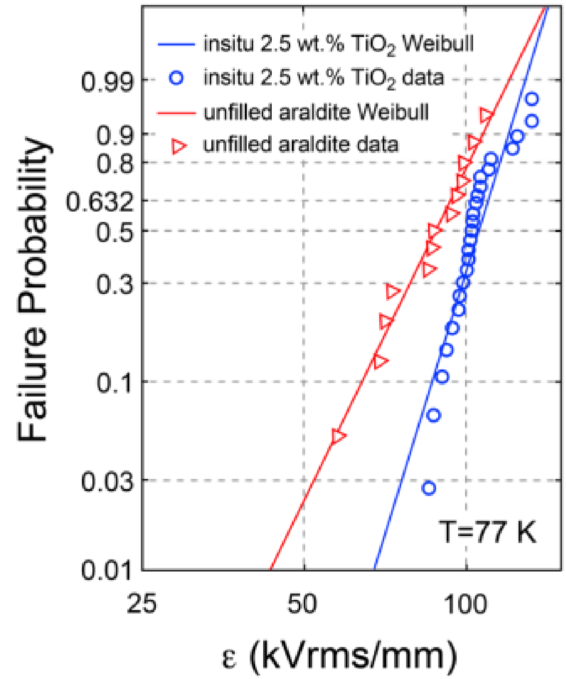
Project draws upon previous nanocomposite dielectrics experience to examine & mitigate degradation mechanisms

- As part of DOE Office of Electricity program, ORNL worked toward development of novel dielectrics for utility applications for room temperature and cryogenic environments
- Nanocomposites developed to examine aging mechanisms associated with partial discharge





Particle dispersion in Araldite for in-situ (left) and ex-situ (right)

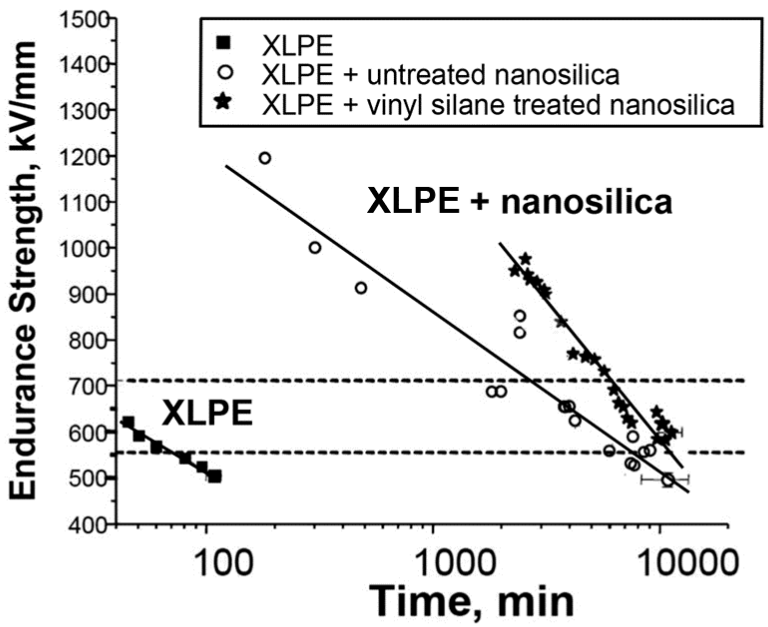


Weibull plot of breakdown strength for nanocomposite dielectric over base polymer

Current project focuses on materials development and radiation degradation mechanisms

1. **New radiation resistant nano-composites in nuclear environments**
   - » **Addition of nanoparticles that mitigate free radicals generated by radiolysis**
2. **Possible connection between degradation in electrical properties and radiation exposure**

1



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Radiation tolerant polymers and corresponding breakdown strengths for 0.125" sample thickness

Polymer	Breakdown Strength (V/mil)	Maximum useful radiation does (Gy)
PS	500	$5x10^7$
PI	560	$2x10^7$
PEEK	480	$1x10^7$
PC	380	$6x10^5$
Nylon	300-400	$2.5x10^4$

PS: polystyrene; PI: polyimide; PEEK: Polyether-etherketone; PC: polycarbonate; nylon: polyamide

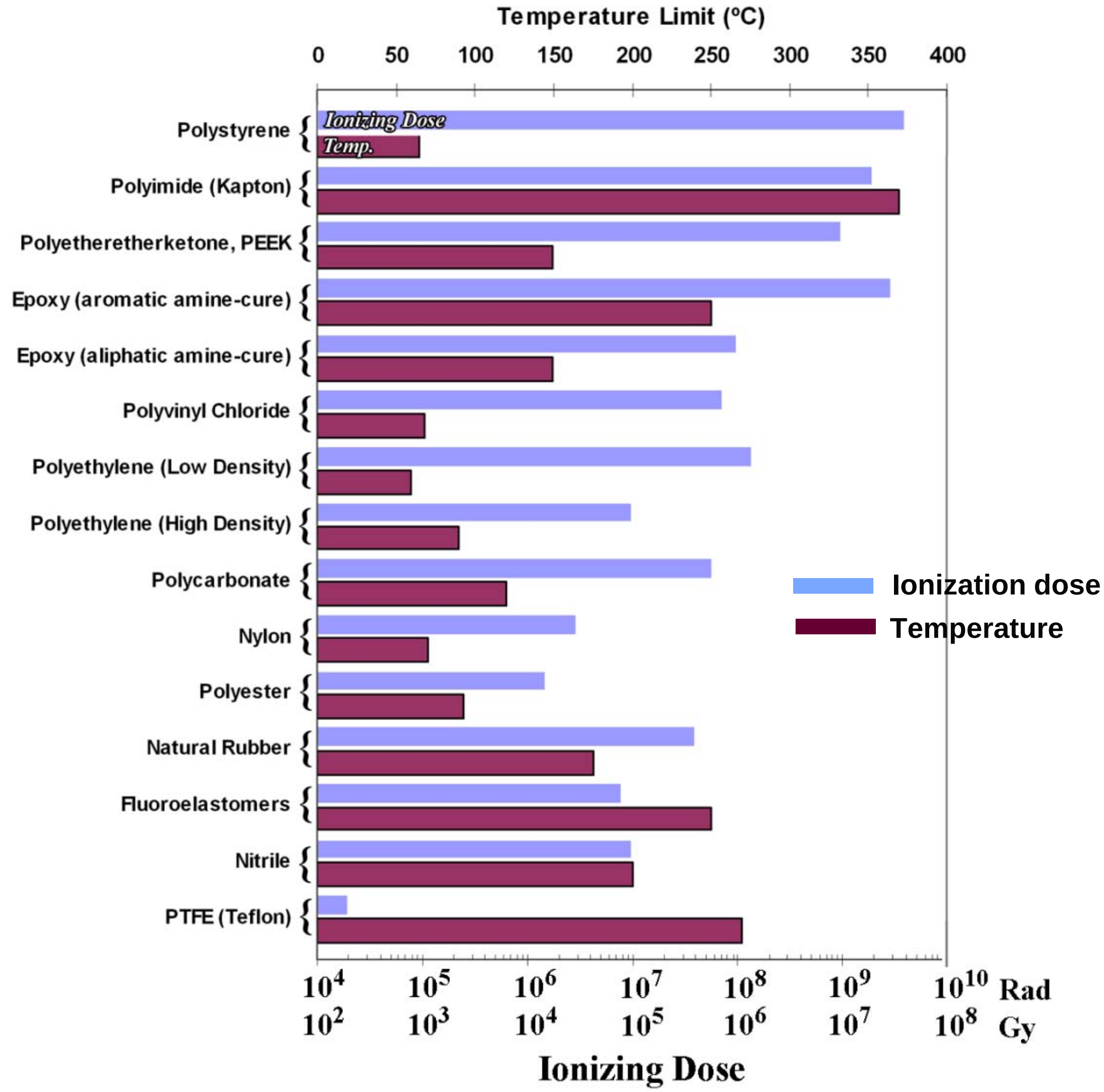
Research for the first year focuses on developing knowledge base and tools to optimize dielectric performance

- **Task Areas**
  - » **Synthesis of nanocomposite dielectrics**
    - – Add nanocomposite solutions to existing base resin materials such as cross-linked polyethylene (XLPE) and Polyvinyl alcohol (PVA)
  - » **Nanocomposite microstructural analysis**
    - – TEM, SEM, & Dielectric Relaxation Spectroscopy (DRS) are used to document changes in materials structure and properties
  - » **Irradiation of nanocomposite dielectrics**
  - » **Performance assessment of radiation response**
    - – Electrical and partial discharge measurements are made as a function of temperature
- **Milestones from September FY2012 & FY2013**

1. **Synthesis of at least two nanodielectric composite materials with XLPE or other resin base material at various particle concentrations and determine their optimum processing conditions; based on the performance assessment with respect to voltage and temperature**
2. **Successful demonstration of test assembly for irradiation of nanocomposite dielectrics at ORNL**

Work is focused on realizing improvements over current class of available insulating materials

- **Current materials utilized in instrument and power cable in nuclear reactors**
  - » PEEK
  - » EPR
  - » CSPE
  - » SiR
  - » XLPE/XLPO
- **Base dielectric materials of interest**
  - » PVA, XLPVA
  - » PE, XLPE
  - » PI

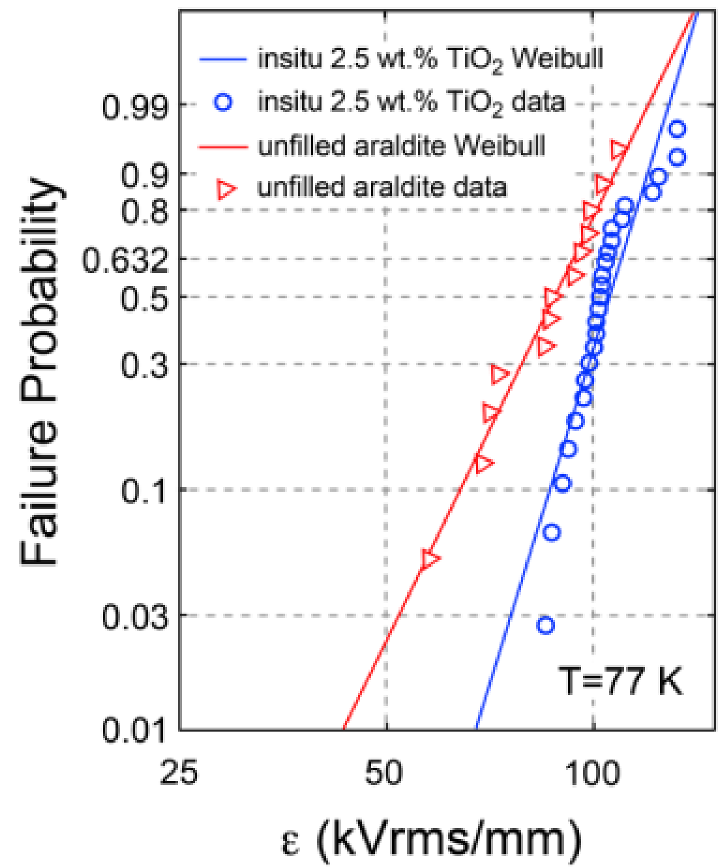


Weibull distribution discussion primer

- **Weibull distribution is utilized to help predict trends and failures with a limited number of samples**

$$F(E,\alpha,\beta) = 1 - \left\{ exp\left(-\frac{E}{\alpha}\right)^{\beta} \right\}$$

- ***α or a*** **is a measure of the parameter under test**
- ***β or b*** **is reflective of the spread of the data**



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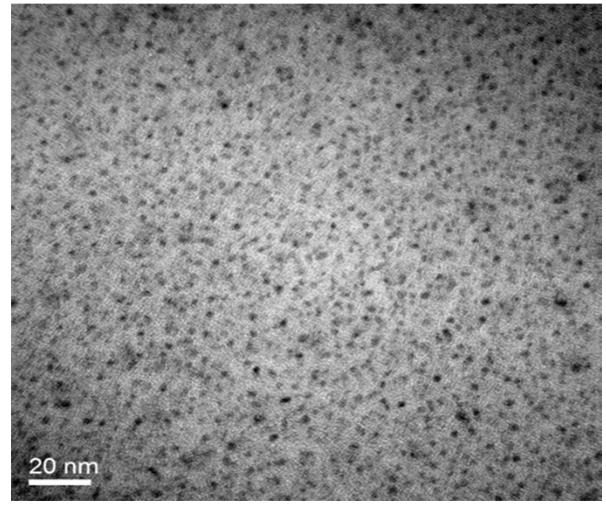
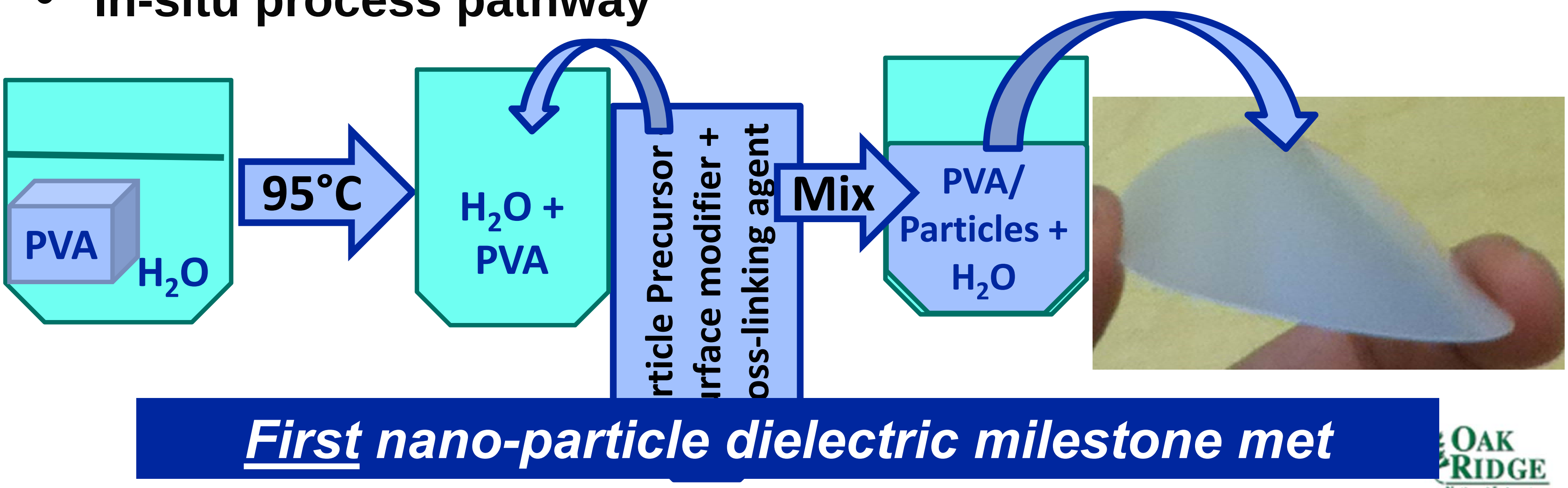
Milestone: Develop in-situ methodology to produce well dispersed nano-particles in a polymer matrix

- **Model systems: polyvinyl alcohol (PVA) and cross-linked PVA (XLPVA)**
  - » **Nano-particle variations**
    - – TiO_2 (1 to 10 wt.%)
    - – SiO_2 (1 to 10 wt.%)
    - – MgO (1 to 5 wt.%)
    - – Gd_2O_3 (1 to 5 wt.%)



TEM image of uniformly dispersed SiO_2 nanoparticles in cross-linked PVA

- **In-situ process pathway**



***First* nano-particle dielectric milestone met**

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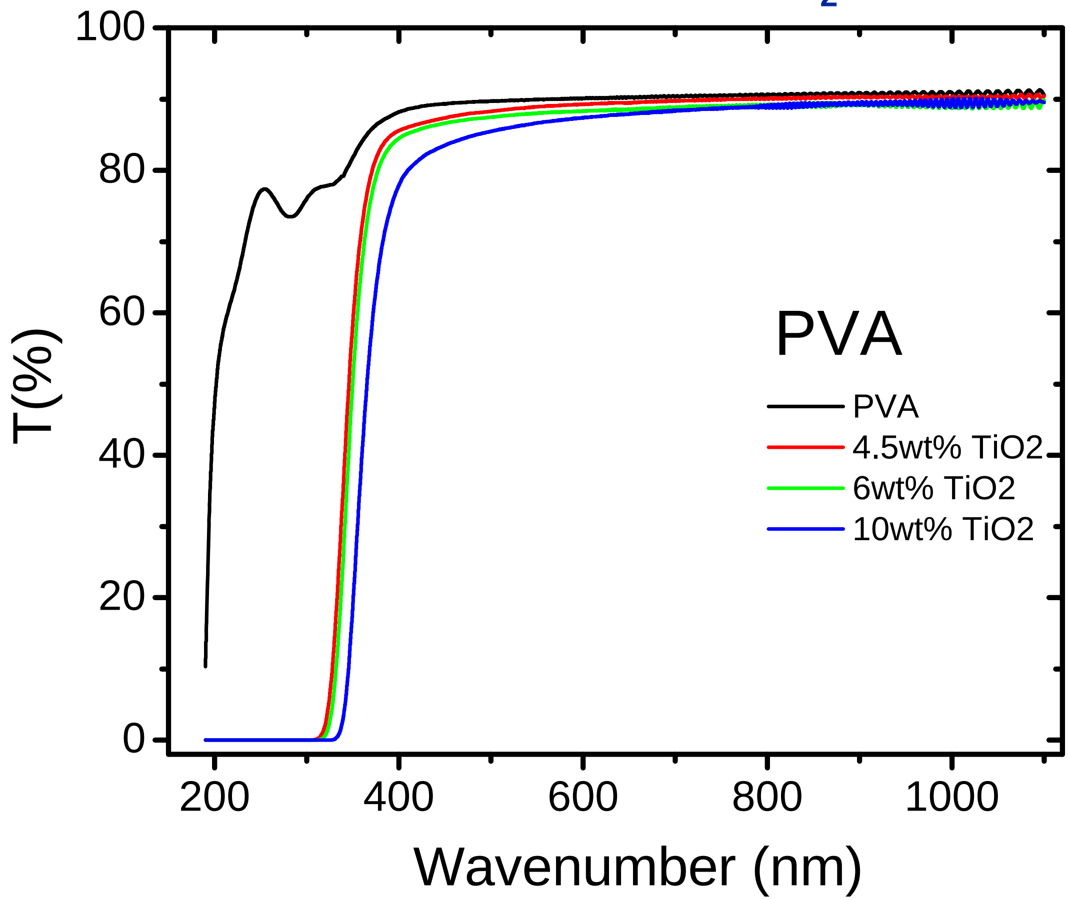
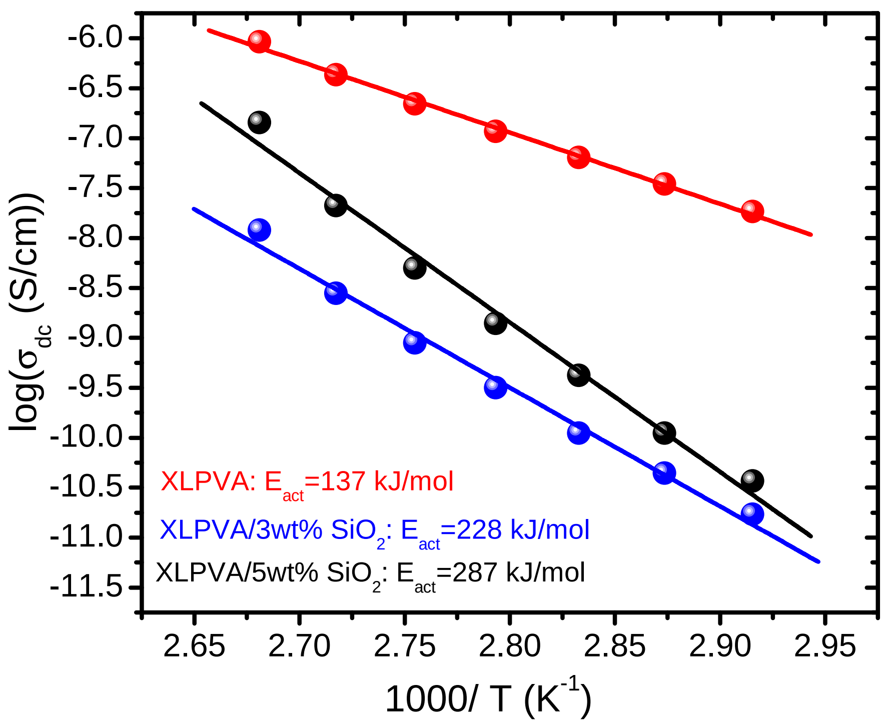
Characterization of PVA films show different trends in UV-visible absorption spectra with nanoparticle additions

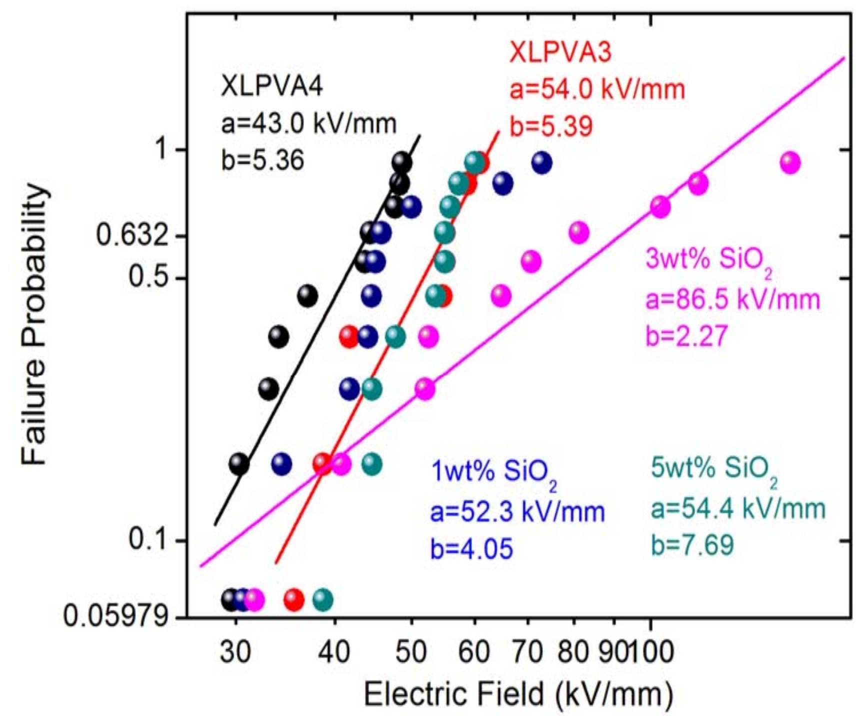
Transmittance of PVA – TiO_2 films



Systematic change in transmittance of PVA films observed with TiO_2 nanoparticle additions

XLPVA: 3 wt.% SiO_2 addition achieves optimum conditions





- ✓ **Nanocomposites exhibit lower conductivity and higher activation energies compared to pristine polymer matrix**
- ✓ **3 wt.% is the optimum nanoparticle content**

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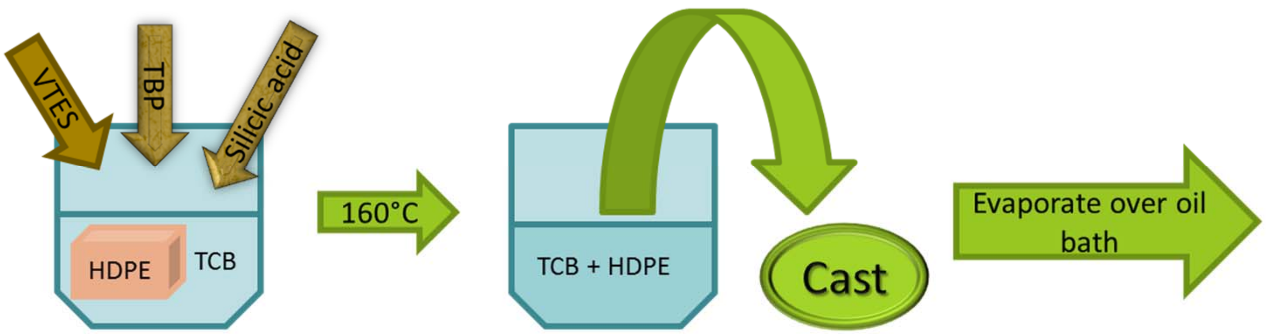
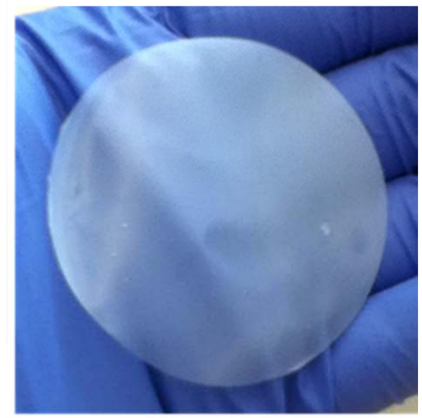
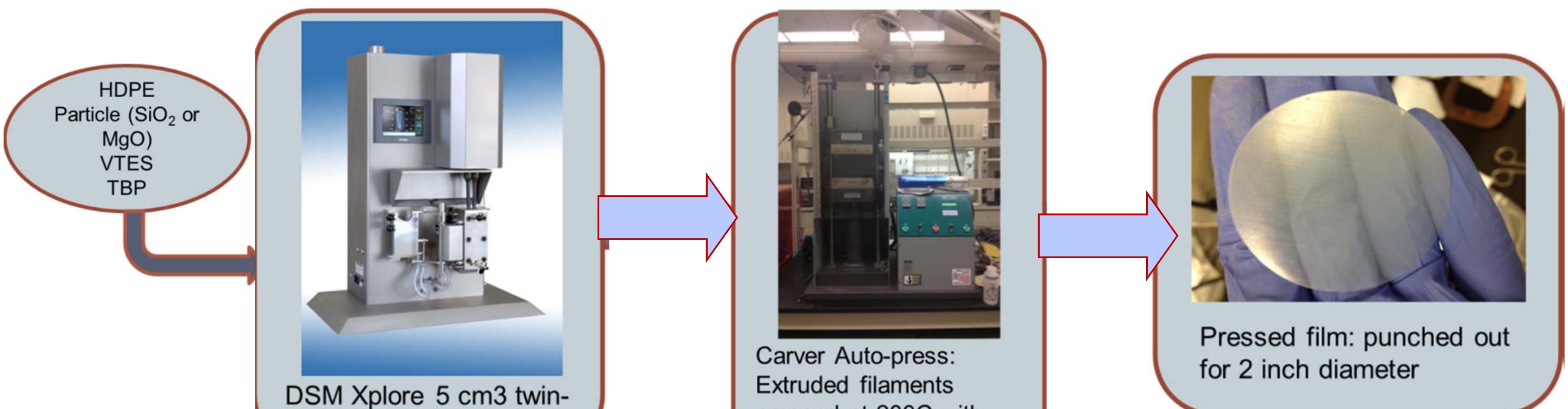
PE / XLPE: Two different process (in-situ/ex-situ) pathways were explored (both methods can be commercially scaled)

- **Polyethylene (PE) and cross-linked PE (XLPE) in-situ synthesis**



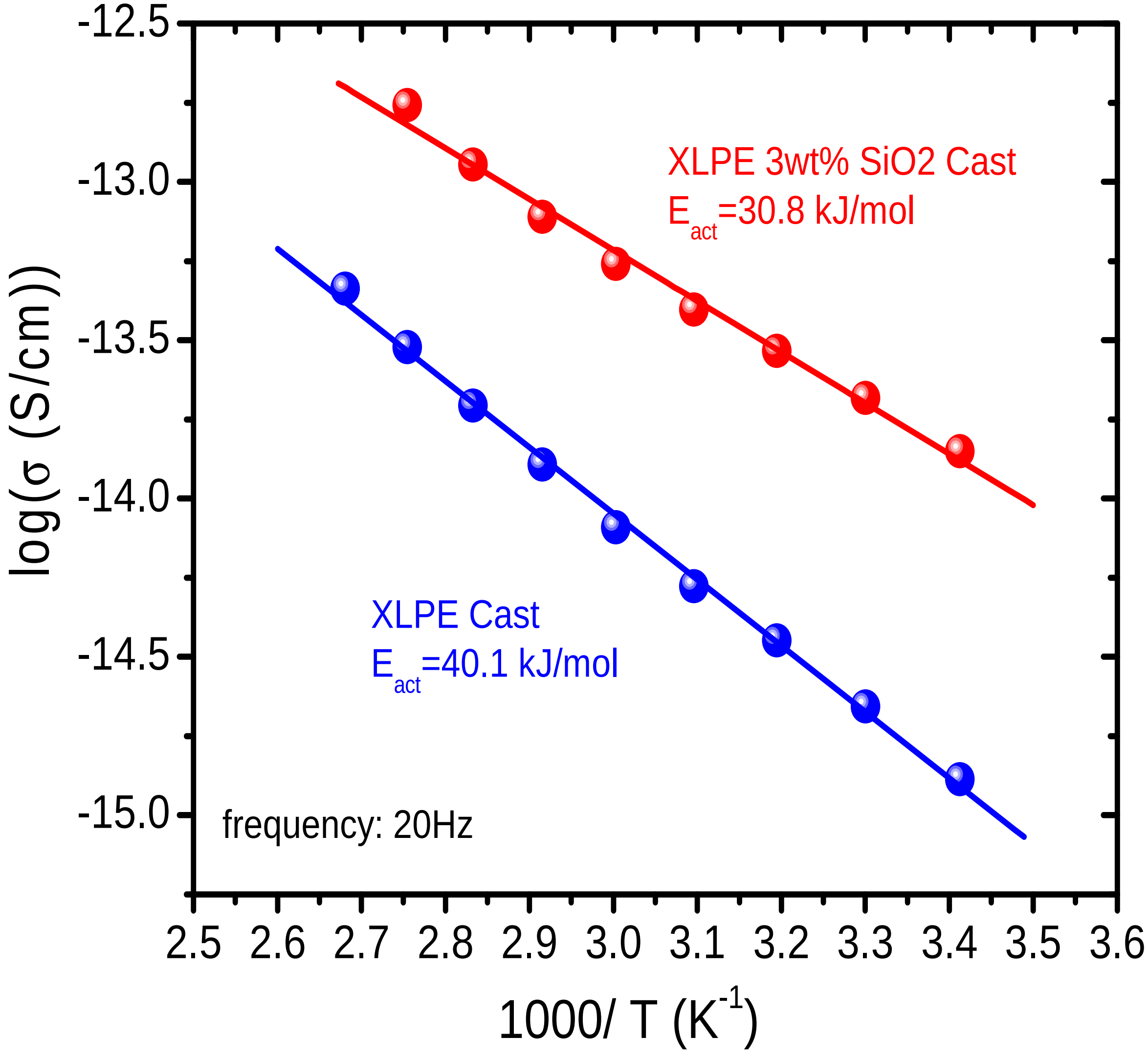


- **PE and XLPE ex-situ synthesis**



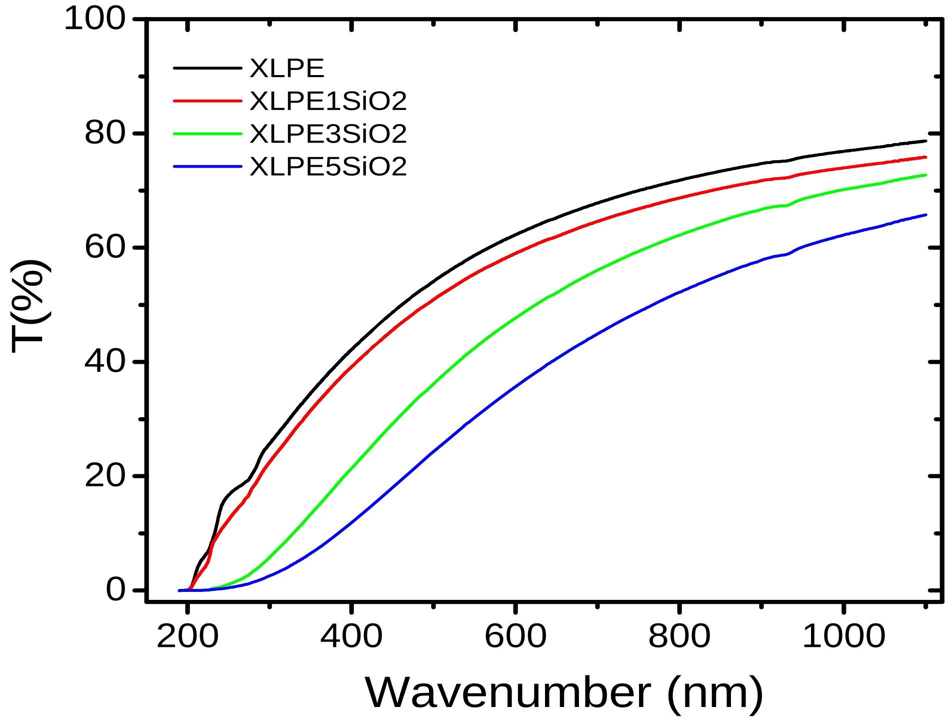
<u>Second</u> nano-particle dielectric developed to meet 1st project milestone

Characterization of XLPE films show different trends in electrical performance with nanoparticle additions

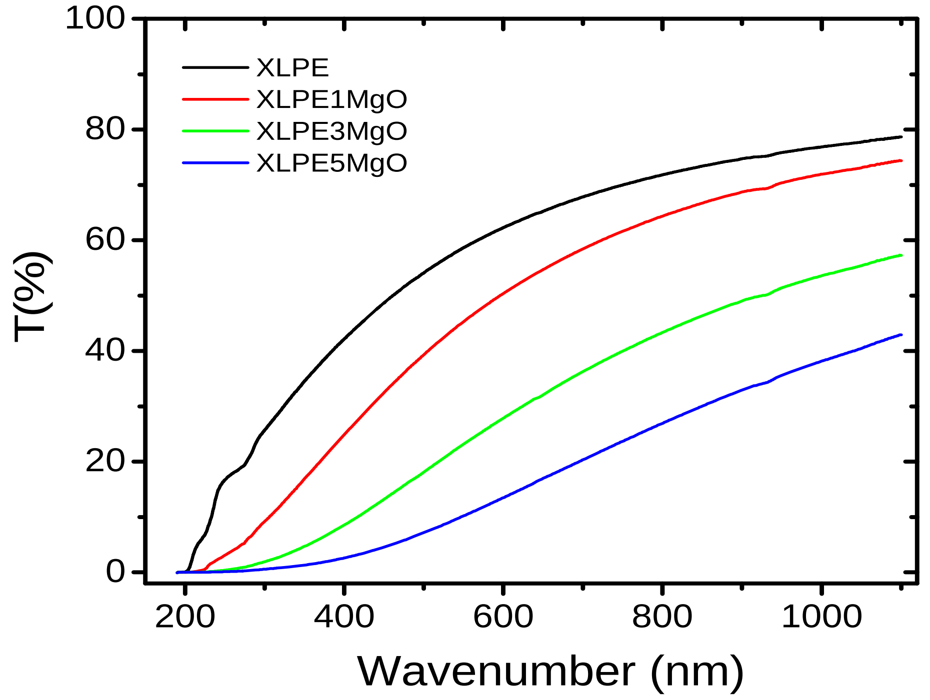


Characterization of XLPE films show different trends in UV-visible absorption spectra with nanoparticle additions

Transmittance of XLPE – SiO_2 films

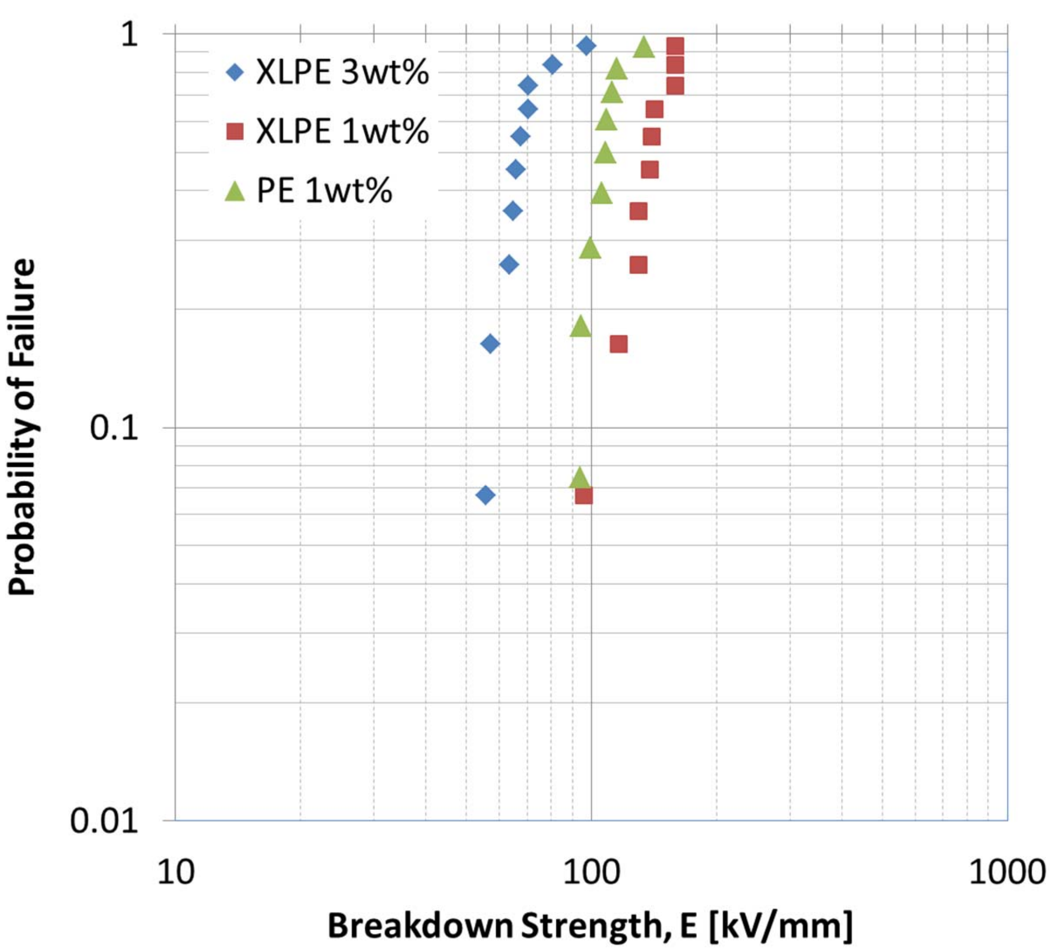


Transmittance of XLPE – MgO films



Systematic change in transmittance of XLPE films observed with nanoparticle additions

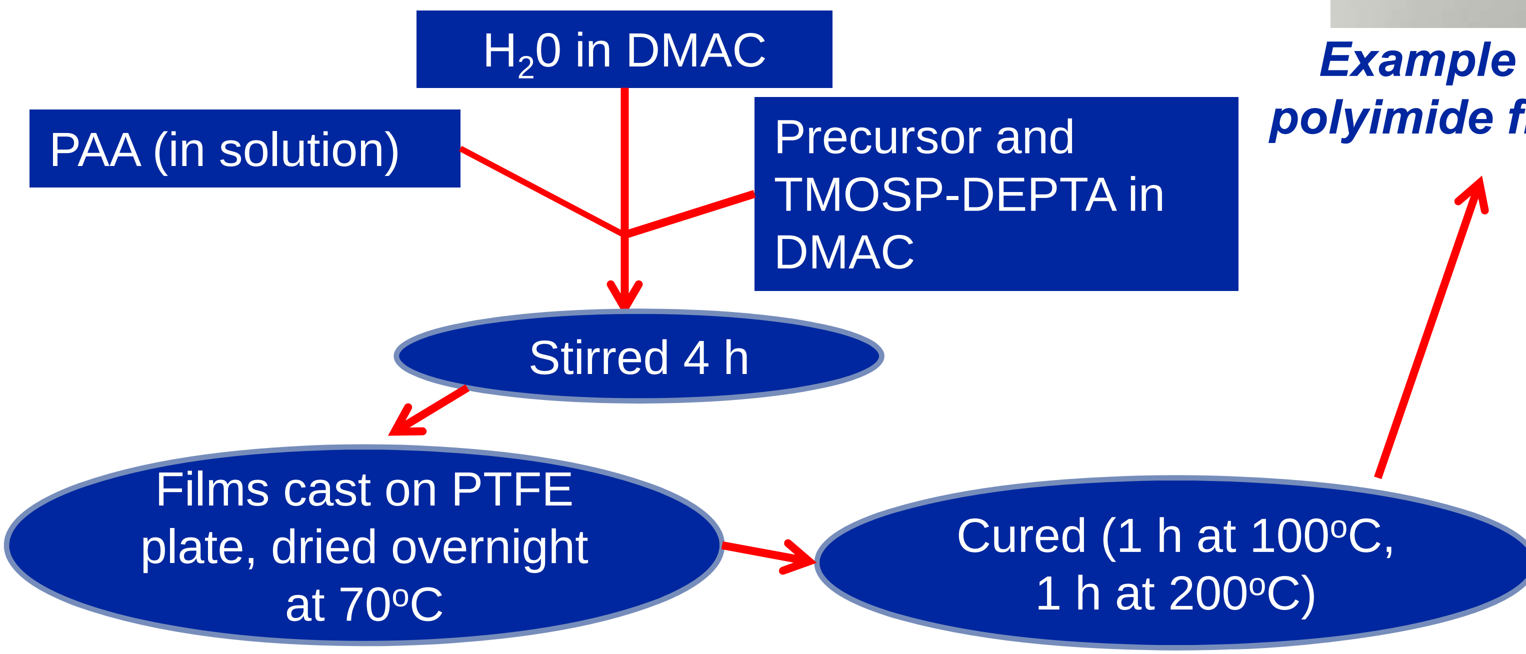
Characterization of (PE/XLPE) films show different trends in dielectric performance with nanoparticle additions



Sample	α (kV/mm)	β (-)
XLPE 3wt.% SiO_2	74.6	6.25
XLPE 1wt.% SiO_2	146.0	6.85
PE 1wt.% SiO_2	113.7	9.25
PE 3wt.% SiO_2	115.3	14.02
PE 5wt.% SiO_2	116.8	8.23
PE (pristine)	87.6	12.75

Polyimide: Third polymer nano-particle dielectric system aims to enable operation at higher temperatures

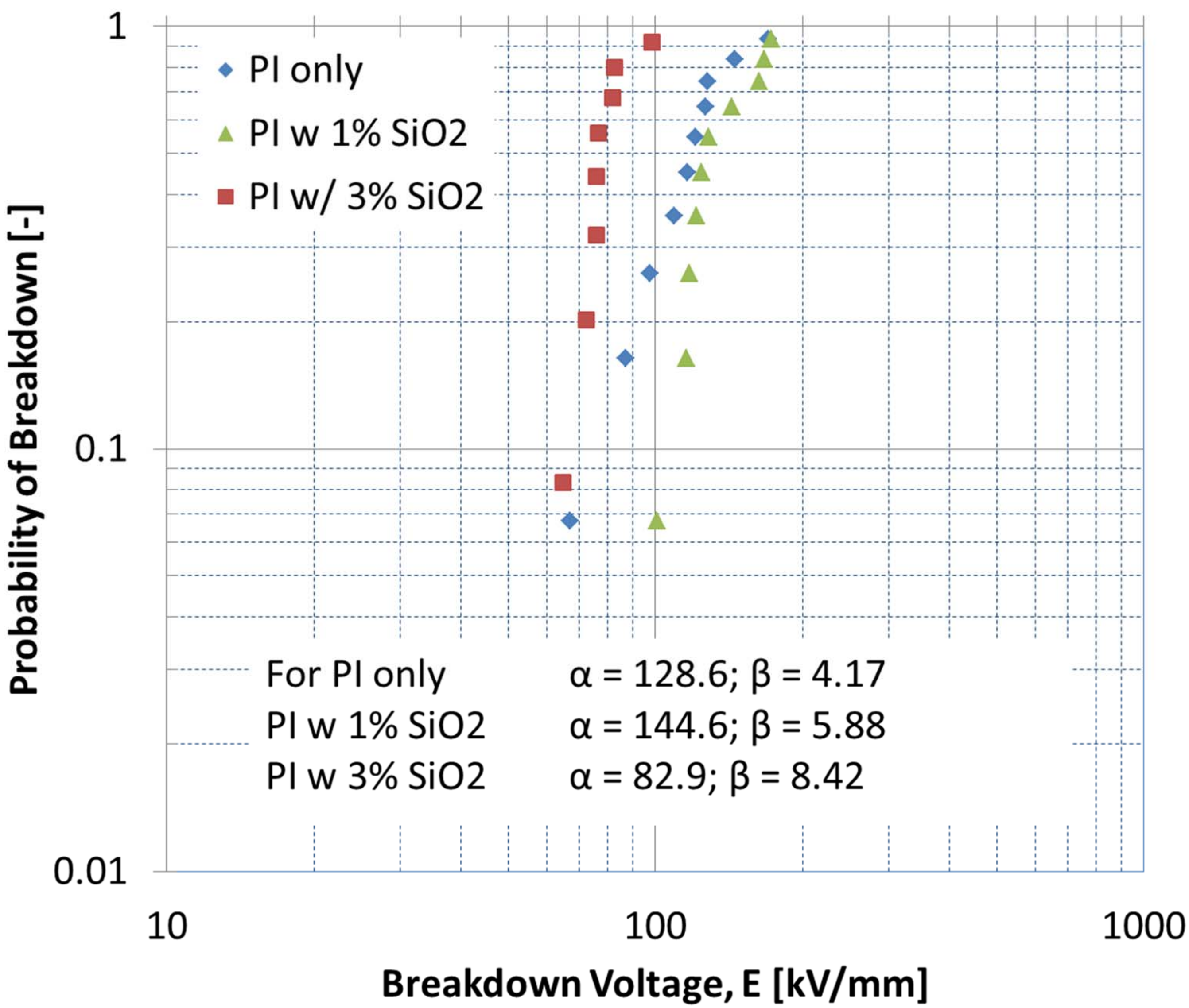
- **Polyimide (PI) often found in magnet applications for high-energy physics and fusion applications**
- **In-situ synthesis**



Example of a typical polyimide film (2 inches)

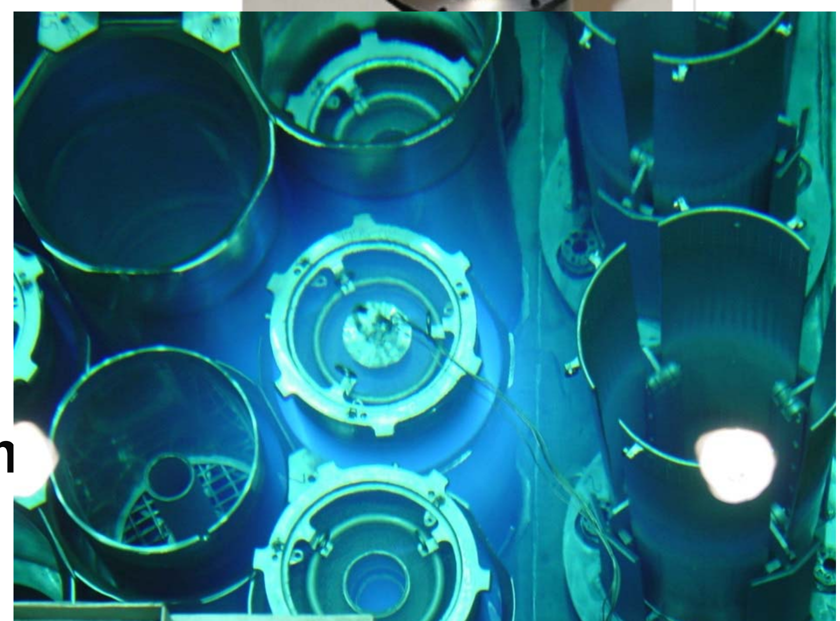
Polyimide nano-particle composite dielectrics developed

Characterization of polyimide films show different trends in performance with nanoparticle additions



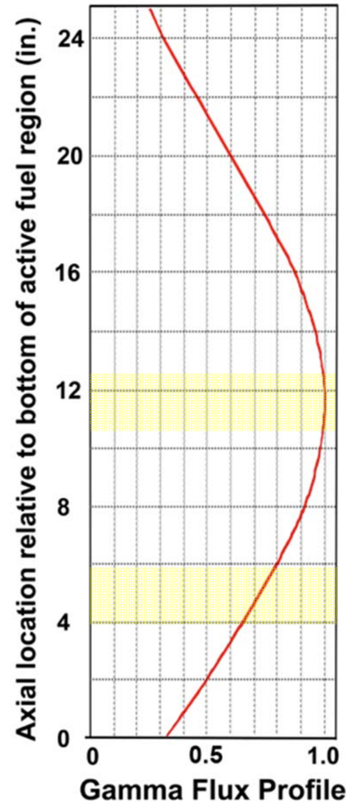
Electrical and microstructural investigations form foundation to evaluate tolerance to extreme environments

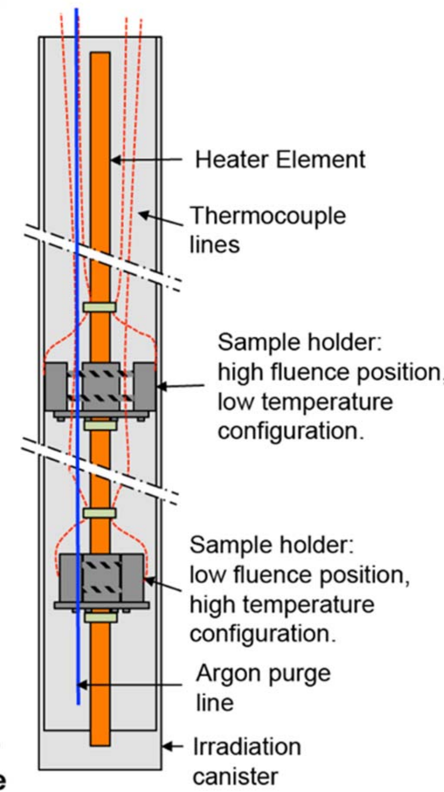
- **Neutron and gamma irradiation: yield different degradation mechanisms in polymers. Most cabling will be used in areas dominated by gamma radiation conditions**
- **Experiments performed at the Gamma Irradiation Facility (GIF) using spent HFIR fuel cores**
  - » **Limit study to one primary radiation type**
  - » **Control over variables: temperature, dose rate, environment**
  - » **Design flexibility and cost effective**
  - » **Significant gamma radiation database for data comparisons**
- **New dielectric composites will be exposed to radiations ranging from 35 to 100 Mrad at temperatures up to 150ºC**

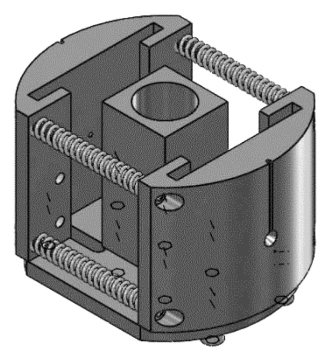


Gamma Irradiation Test Fixture

- **Variable position sample holders**
- **Up to three possible gamma dose positions per run**
- **Dosimeter packets positioned next to samples**
- **Each holder contains two equal loading positions for sheet samples**
- **Unheated condition: springs push holder to canister wall for maximum cooling**
- **First irradiation run (unheated): Max T_{irr} = 40°C, T_{pool} = 37°C**
- **Heated condition: springs removed and sample holders moved inward to heater rod - max temperature ~150°C**







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First irradiation run completed on June 27th – ahead of schedule. Post Irradiation Evaluation underway

- Materials: PVA/XLPVA & PE/XLPE with TiO_2, SiO_2, MgO, Gd_2O_3
- 42 Samples Irradiated
- Irradiation Specs: Fuel element from cycle 442 (discharged June 2012), peak gamma flux = 0.768 MR/h (7.68 kGy/h)

Run	1	2	3*	4
Position 1	3.2 MRad (32 kGy)	10 MRad (100 kGy)	20 MRad (300 kGy)	10 MRad (120 kGy)
Position 2	10 MRad (100 kGy)	20 MRad (200 kGy)	40 MRad (500 kGy)	20 MRad (200 kGy)
Temperature	40°C	40°C	95°C	150°C
Completion Date	June 27th, 2013	Sept 4th, 2013	TBD	TBD

* Exact conditions and the number of runs are based on results of previous experience.

- Run 2 scheduled (September) – XLPE & PI with SiO_2, MgO, Al_2O_3
- Conditions of run 3 & 4 will be determined based on the findings of first two runs

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Conclusions and future work

- ✓ **Three distinct nanocomposite dielectric material systems have been developed**
  - » **PVA/XLPVA with TiO_2, SiO_2, MgO, Gd_2O_3**
  - » **PE/XLPE with SiO_2, MgO, Al_2O_3**
  - » **PI with SiO_2, MgO, Al_2O_3**
  - » **Invention disclosures have been filed for all three material systems**
- ✓ **Initial irradiation of samples completed and comparison of sample performance due to irradiation underway**
  - » **In addition to discussion with industrial partners on scale-up, the outcome of this will help down selection of material systems for second year of project**
- ✓ **All milestones for first year completed on time and ahead of schedule**

Multidisciplinary effort benefited greatly from contributions from diverse expertise of project partners

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  - Chemical Sciences (CSD)
  - Material Science and Technology (MSTD)
  - Measurement Science and Systems Engineering (MSSED)

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