Growth of PLZT Films on Metal Foils for Power Electronics Applications

Beihai Ma, M. Narayanan, S. Liu, U. Balachandran

Argonne National Laboratory

Argonne, IL 60439

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This presentation does not contain any proprietary, confidential, or otherwise restricted information

Outline

- Introduction
- Technology gap
- Argonne's advanced ceramic capacitor technology
- Summary
- Acknowledgement

Introduction
(Capacitor Basics)

Capacitor

Capacitance

$$C = \frac{Q}{V} = \varepsilon_0 \varepsilon_r \cdot \frac{A}{t}$$

ε_0: permittivity of free space

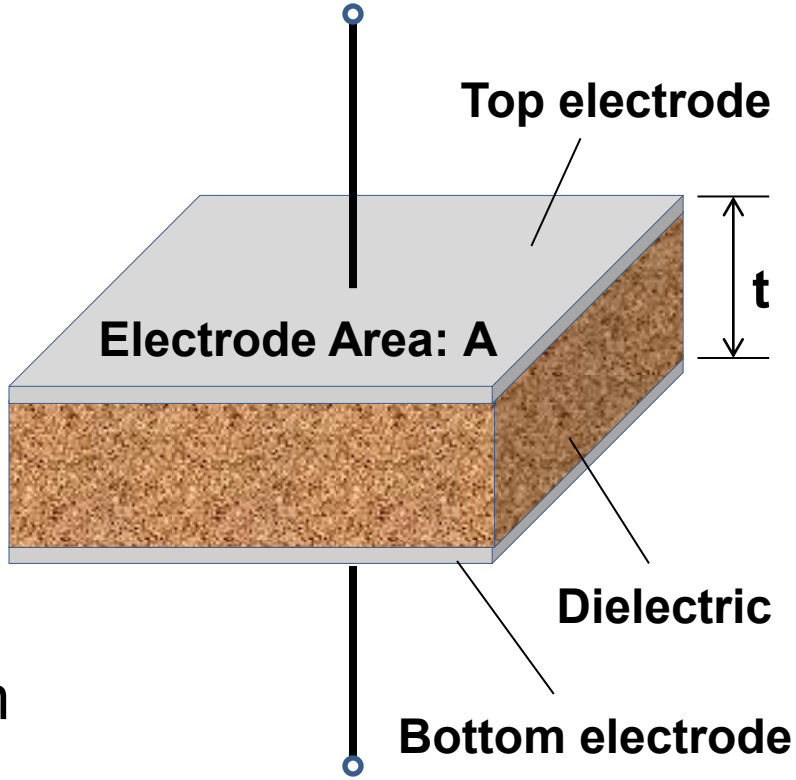
ε_r: dielectric constant

A: area (of electrode)

t: thickness of dielectric medium

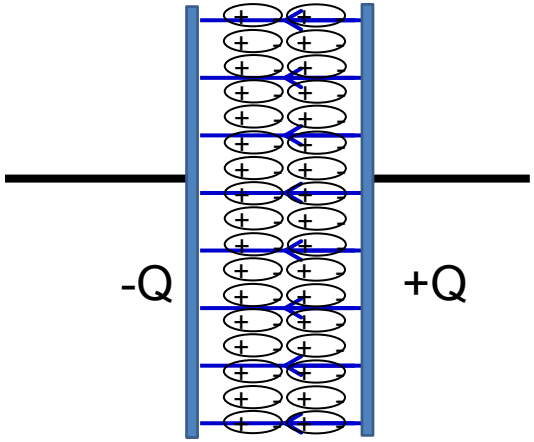
V: Applied voltage

Q: charge (on a plate)



Capacitance increases with increasing dielectric constant and decreasing thickness of dielectric medium

Energy Density



Polarization, P

Polarization

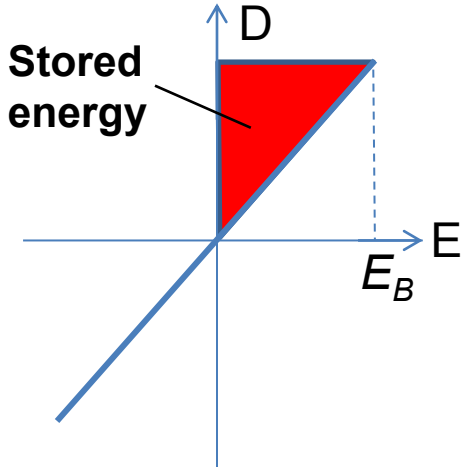
$$\vec{P} = \varepsilon_0 \chi_e \vec{E} = \varepsilon_0 (\varepsilon_r - 1)\vec{E}$$

Displacement field

$$\vec{D} = \varepsilon_0 \vec{E} + \vec{P} = \varepsilon_0 \varepsilon_r \vec{E}$$

Energy density

$$U = \int \vec{E} \cdot d\vec{D} = \varepsilon_0 \int_{E=0}^{E_B} \varepsilon_r \vec{E} \cdot d\vec{E}$$



For linear system (ε_r is independent of $\vec{E}$),

$$U = \frac{1}{2} \varepsilon_0 \varepsilon_r E_B^2$$

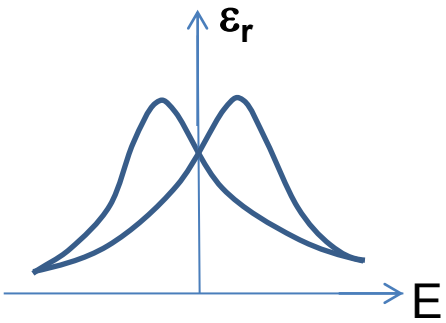
Energy density increases with increasing dielectric constant and breakdown strength

Energy Density of Nonlinear System

For example, a ferroelectric material,

ε_r is a dependent of applied field, $\vec{E}$

$$\varepsilon_r = \varepsilon_r(\vec{E})$$

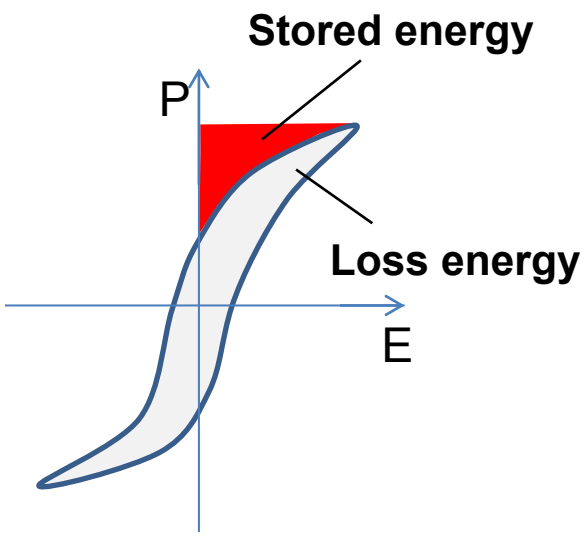


Polarization

$$\vec{P} = \varepsilon_0 \chi_e \vec{E} = \varepsilon_0(\varepsilon_r - 1)\vec{E}$$

Energy density

$$U = \varepsilon_0 \int_{E=0}^{E_B} \varepsilon_r \vec{E} \cdot d\vec{E} < \frac{1}{2}\varepsilon_0 \varepsilon_r(0) E_B^2$$

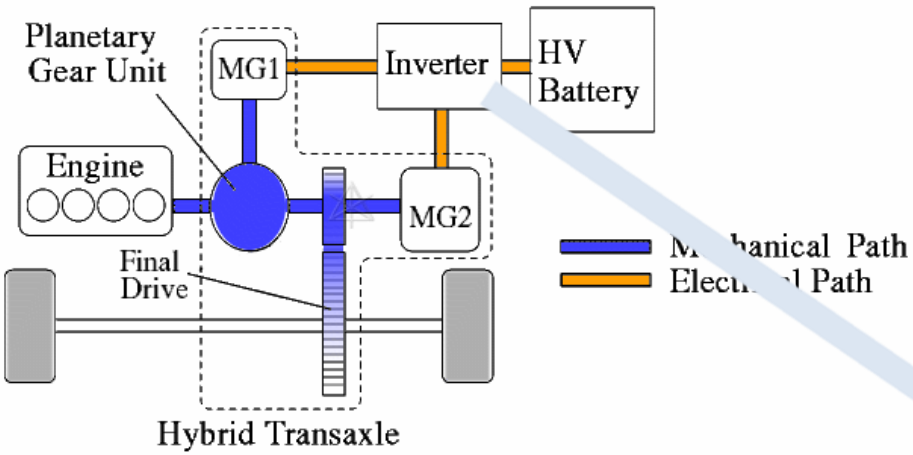


Ferroelectric PLZT $[(Pb_{0.92}La_{0.08})(Zr_{0.52}Ti_{0.48})O_3]$ exhibit high dielectric constant >1000, high breakdown strength >2 MV/cm, and high Curie temperature $T_c \approx 180°C$.

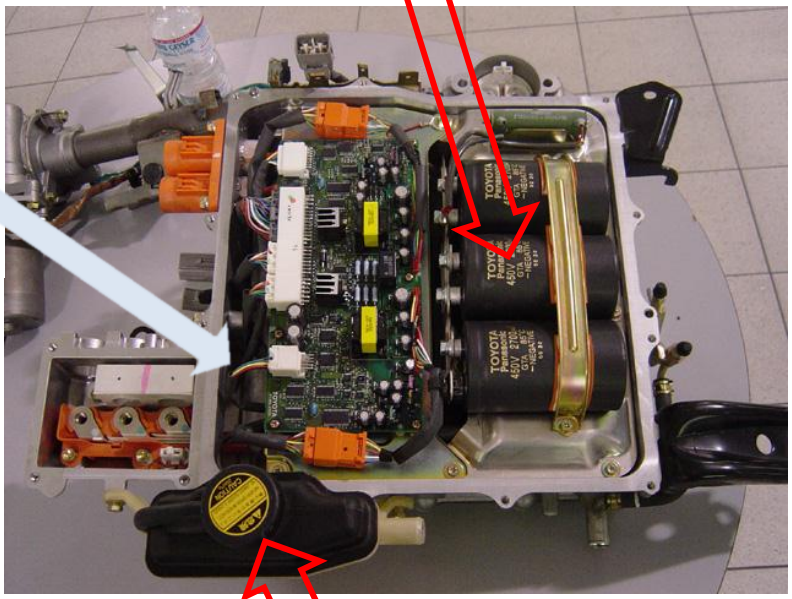
Technology Gap

Capacitor Technology in Power Electronics (Hybrid Electric Vehicles)

THS/HSD system topology



TOYOTA Prius (2004-2006 model) THS inverter unit with top cover removed. It has a bank of coke-can sized capacitors labeled "TOYOTA Panasonic 450V 2700μF GTA 85°C".



- **Overall size and cost of inverters**
  Capacitors are a significant fraction of the inverter volume (≈35%), weight (≈23%), and cost (≈23%).
- **High-temperature operation**
  The performance and lifetime of presently available capacitors degrade rapidly with increasing temperature

A separate cooling system is required for the inverter unit.

DOE Goal vs. Present Technology

Commercial film capacitor

Polypropylene film capacitor

40 μF, 600 V_{dc}
Volume = 225 cm^3
Energy Density = 0.032 J/cm^3
Capacitance Density = 0.18 $\mu F/cm^3$

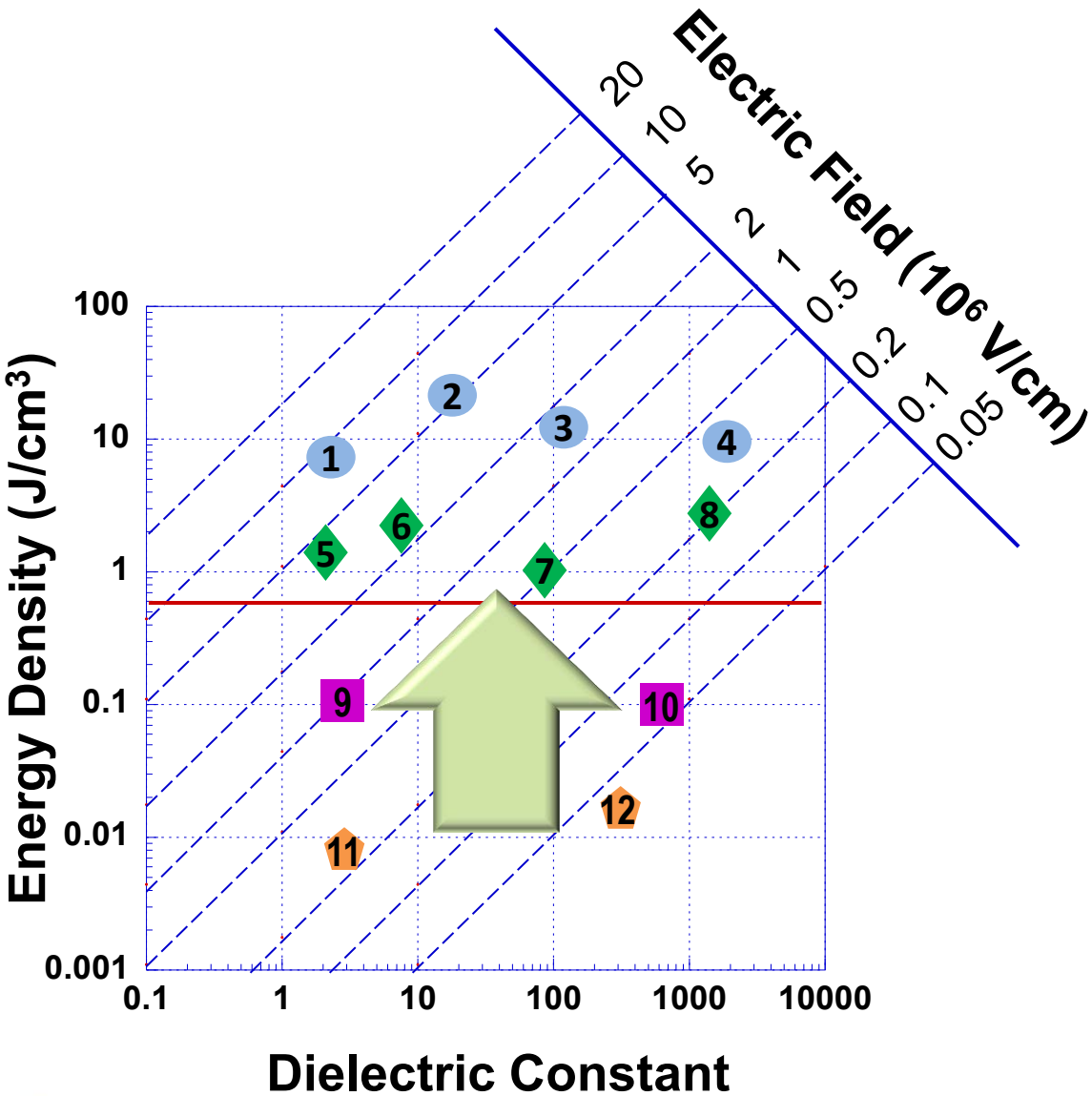
DOE goal

Capacitor volume ≤0.6 liters (600 cm^3)
Operation temperature -40 to +140°

40 μF, 600 V_{dc}
Volume = 12 cm^3
Energy Density = 0.6 J/cm^3
Cap Density = 3.3 $\mu F/cm^3$

Reduced size for improved volumetric and gravimetric efficiencies. Operation temperature, -40 to 140°C.

Energy Density of Dielectrics and Capacitors



Dielectrics

1. Polypropylene
2. Fluorpolymer, PSU
3. Titania (TiO_2)
4. PZT ceramics, PSU

Pulsed power capacitor

5. Polypropylene cap
6. High-K polymer cap
7. NPO MLCC
8. X7R based MLCC

Power electronics capacitor

9. Polypropylene film cap
10. X7R Based MLCC

High temperature capacitor

11. High temperature polypropylene cap
12. High temperature 200°C MLCC

Argonne's Advanced Ceramic Capacitor Technology

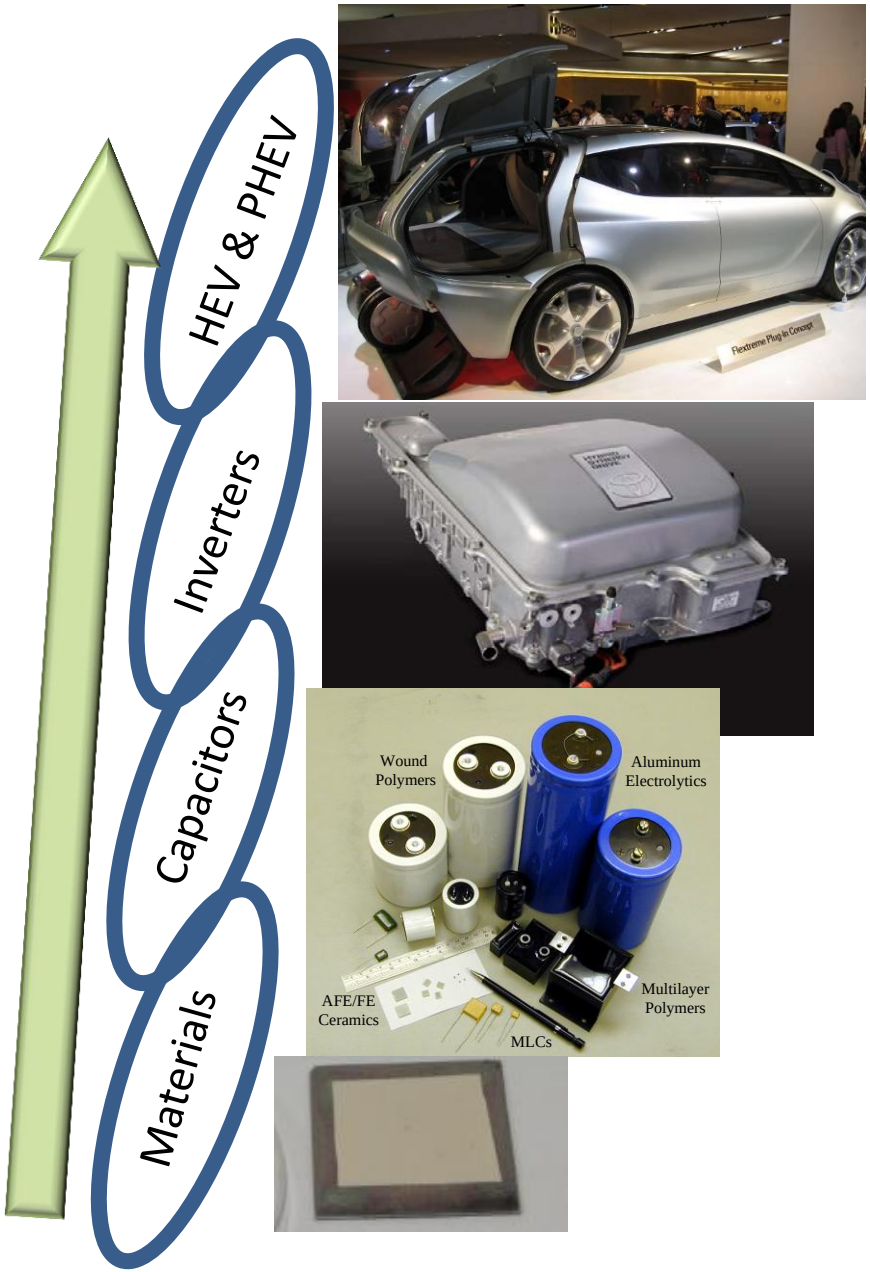
Objective

- **Our goal is to develop ceramic dielectric films that have potential to reduce size, weight, and cost, with increased capacitance density, and capable of high temperature operation, for power electronic systems.**

  DC bus capacitors for inverters

  - ***450V, 1000µF, <3mΩ ESR, 250A ripple current, 140°C, benign failure***

Materials R&D for advanced devices and to achieve better components performance, and to facilitate the implementation of electric drive vehicles.



Technology Description

- **Fabrication of high-voltage capable dielectric films on base-metal foils ("film-on-foils") that meet or exceed the needs of electric drive vehicle power inverters.**
- **Stacking high-ε_r film-on-foil dielectrics to produce capacitors with high volumetric efficiency.**
- **Embedding high-ε_r film-on-foil dielectrics within the layers of a printed wire board (PWB) to replace discreet surface-mounted capacitors is advantageous in electronics miniaturization, device reliability, and manufacturing cost reduction.**
- **We have deposited high-ε_r lead lanthanum zirconate titanate (PLZT) ceramic dielectric films on base metal foils by a chemical solution deposition technique.**
  - ***Operational temperature -50°C to +150°C, 450V DC bus capability, high ε_r (>1000), and high E_B (>2 MV/cm).***

Stacking/Embedding Architecture

Basic Element

Metal foil coated with thin film PLZT dielectric

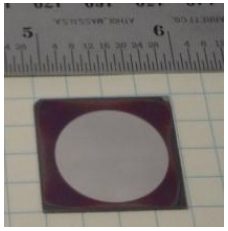
- **PLZT/Ni Film-on-Foil**
- **PLZT/Cu Film-on-Foil**

Approach

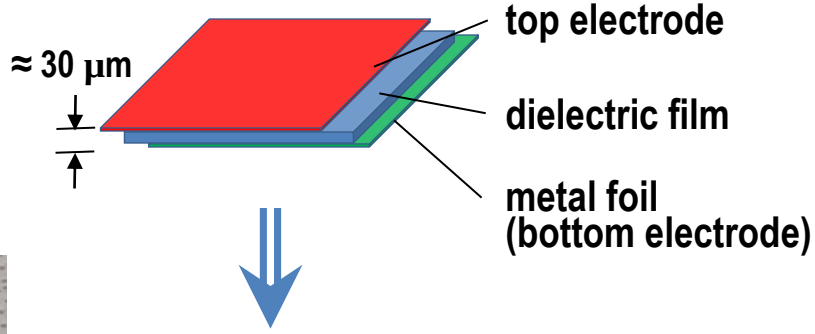
Stack or embed coated foils with interconnection for power electronics

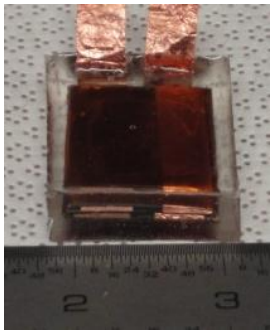
Advantages

- **Reduces component footprint**
- **Shortens interconnect lengths**
- **Reduces inductive losses & electromagnetic interference**

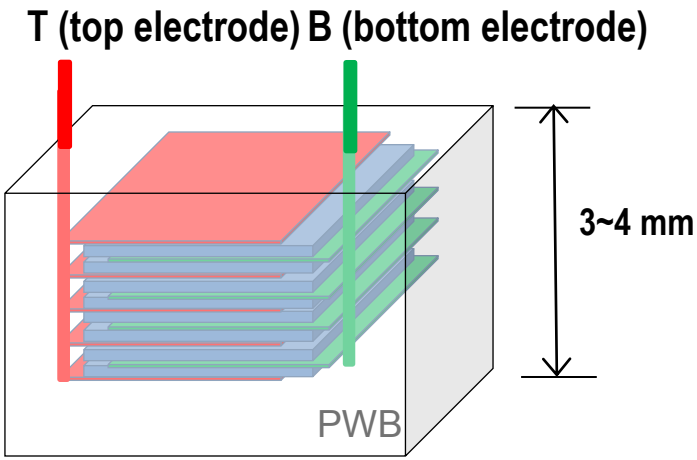


Film-on-foil





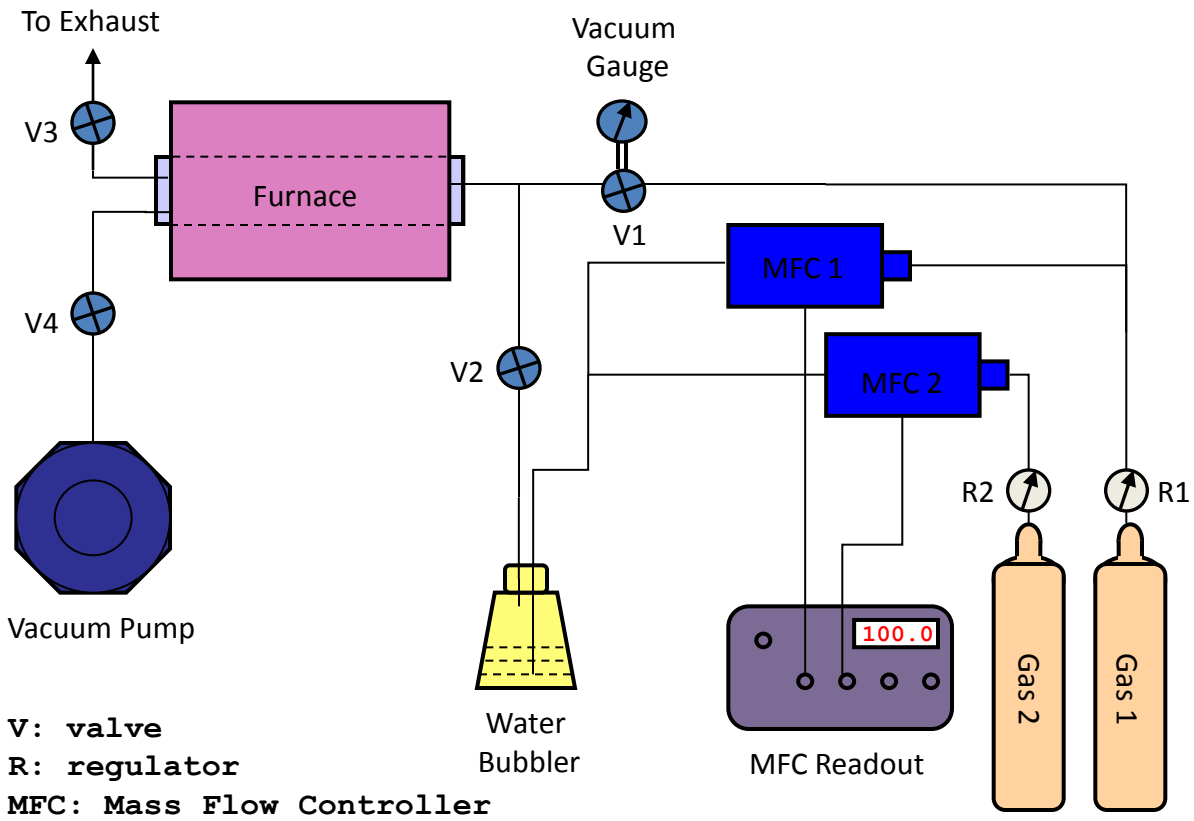
Stacked capacitor w/ interconnection



Experimental Procedure

- Prepare $LaNiO_3$ (LNO) and PLZT precursor solutions.
- Prepare base metal (nickel and copper) foils for coating.
- Grow LNO and PLZT film by chemical solution deposition,
  - LNO film coating: spin-coated at 3000 rpm/30 sec, pyrolysis ≈450°C/5 min, crystallization ≈625°C for 2-5 min. Repeat 3 times.
  - PLZT film coating: spin-coated at 3000 rpm/30 sec, pyrolysis ≈450°C/10 min, crystallization ≈650°C/2-5 min. Repeat to build desired thickness.
- No LNO buffer layer was used while fabricating PLZT films on Cu foils; all annealing and pyrolysis were done in ultra-pure nitrogen atmosphere.
- Platinum (Pt) electrodes were deposited on the PLZT films by electron-beam evaporation.
- Dielectric constant and loss measured by an Agilent E4980A Precision LCR meter, hysteresis loop measured by Radiant meters, leakage current and breakdown strength measured by Keithley 237 source meter.

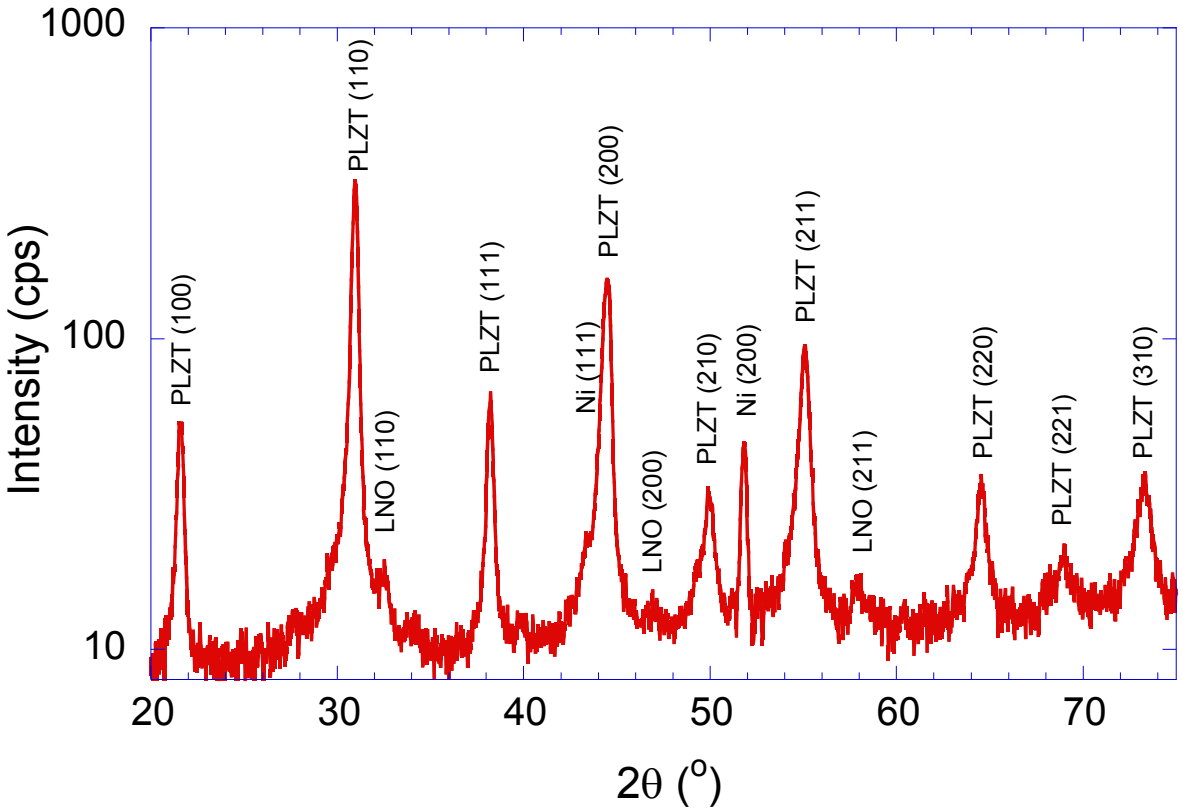
Experimental Setup
(for heat-treating in controlled atmosphere)



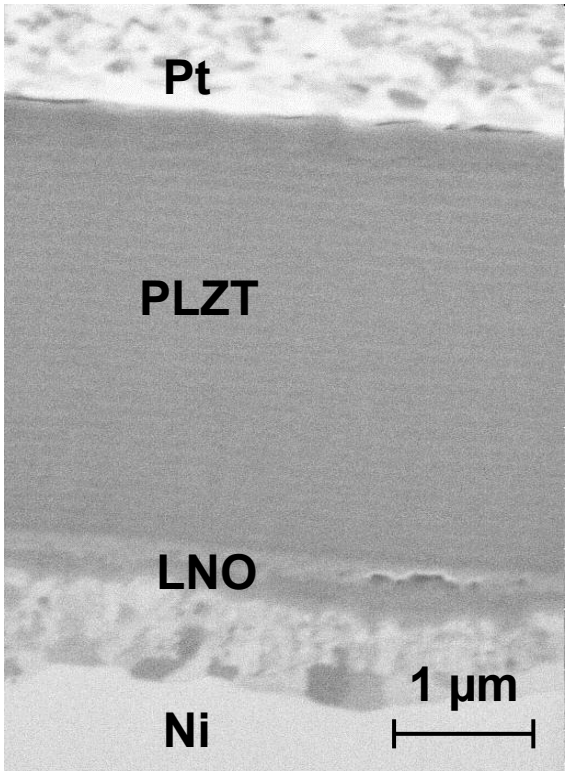
V: valve
R: regulator
MFC: Mass Flow Controller

PLZT on LNO Buffered Ni Substrates

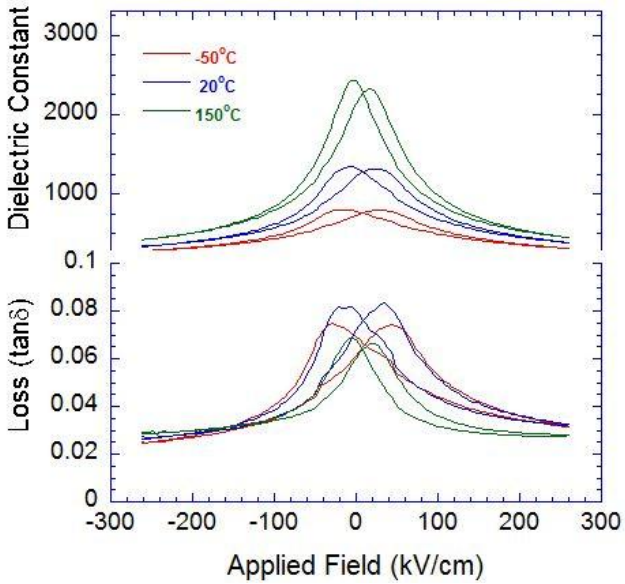
X-ray Diffraction Pattern

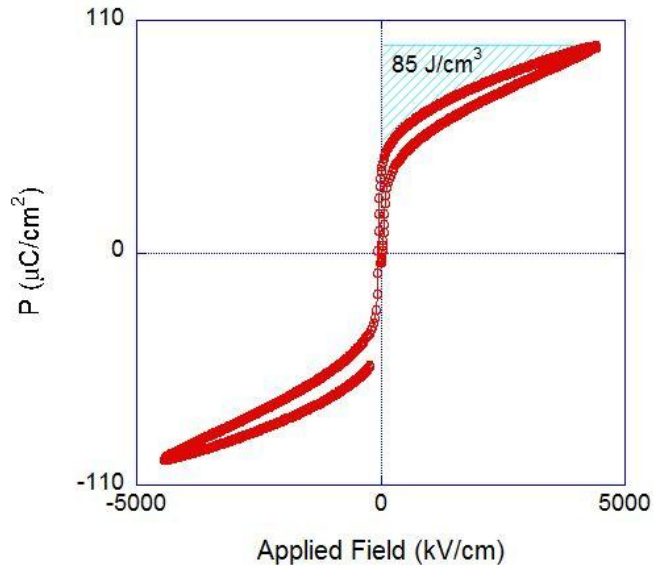


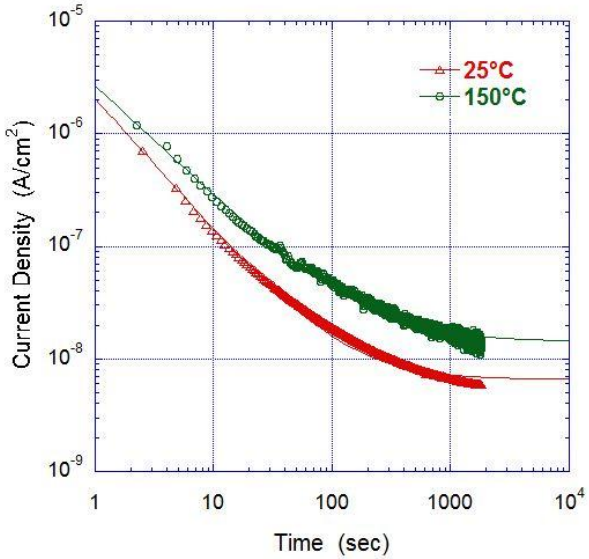
Cross-sectional SEM



Dielectric Properties of PLZT/LNO/Ni

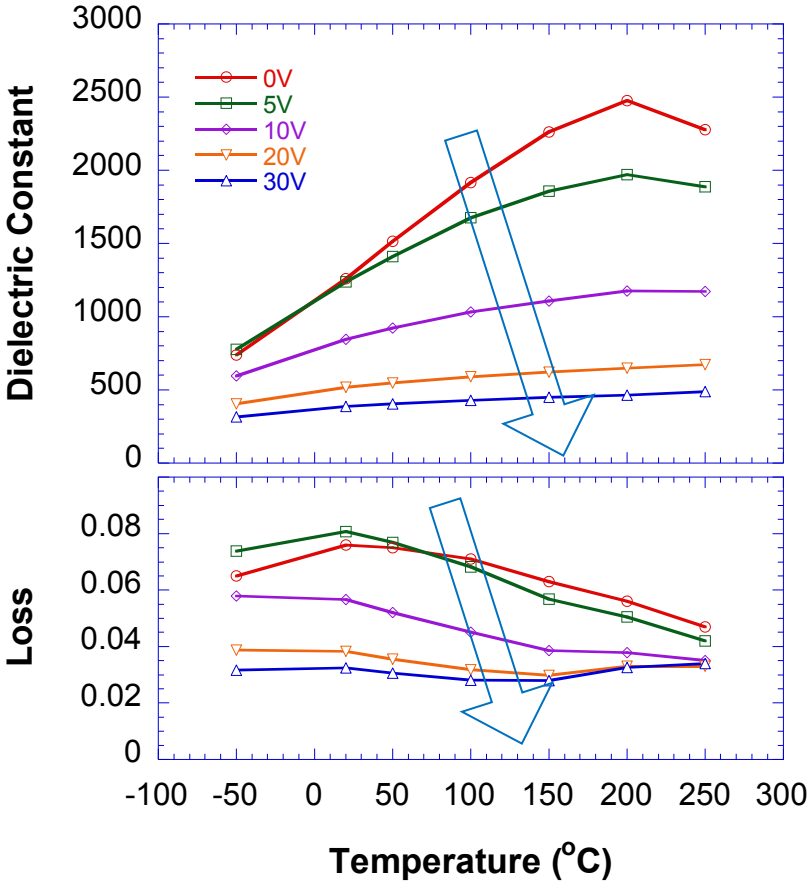


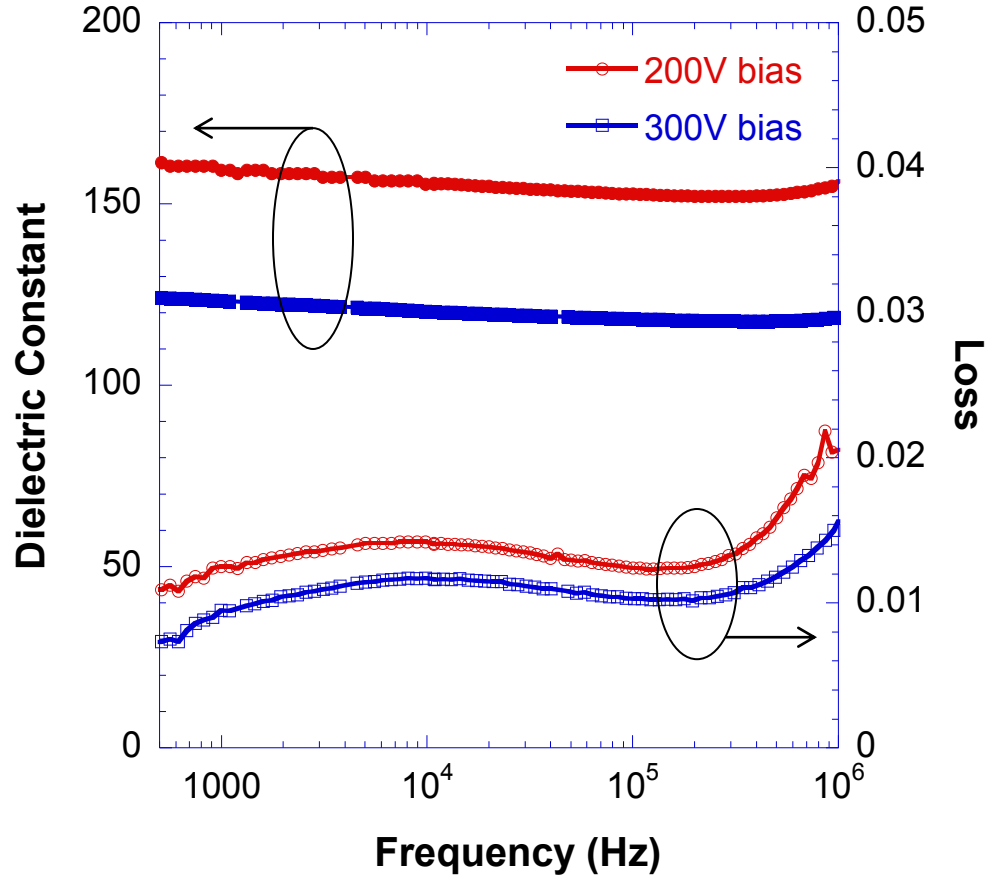




- **Fabricated PLZT on Ni foils with ε_r ≈700 & DF ≈0.07 @ -50°C; ε_r ≈1300 & DF ≈0.08 @ room temperature; and ε_r ≈2200 & DF ≈0.06 @ 150°C.**
- **Leakage current <7x10^{-8} A/cm^2 at RT and <2x10^{-7} A/cm^2 at 150°C.**
- **Measured energy density ≈ 85 J/cm^3 @1400 V and ≈15 J/cm^3 @ 600 V**

Dielectric Properties under Bias Field





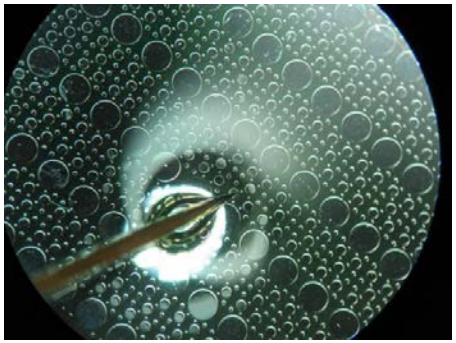
Both ε_r and Loss ↘ with ↗ bias field

ε_r ≈120 and Loss ≈1% at 300 V bias

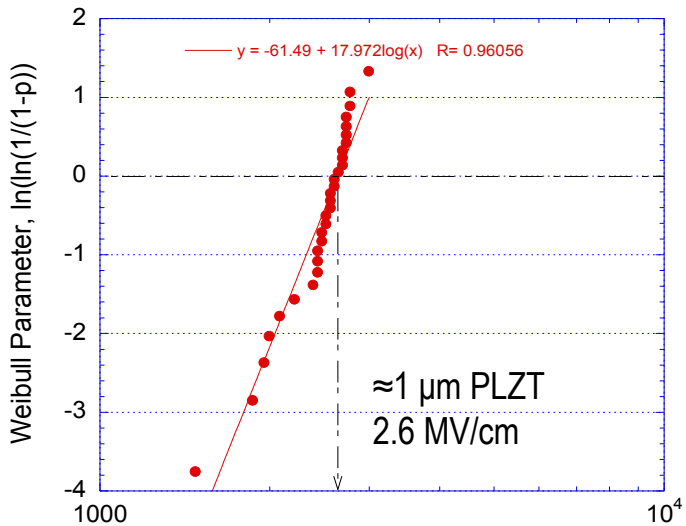
Breakdown Strength of PLZT/LNO/Ni

Breakdown strength is dependent of

- **stress level (V)**
- **stress duration (t)**
- **temperature (T)**



Measured current: I(V, t, T)

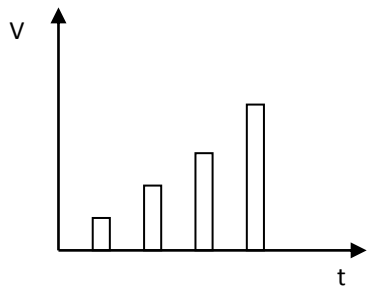


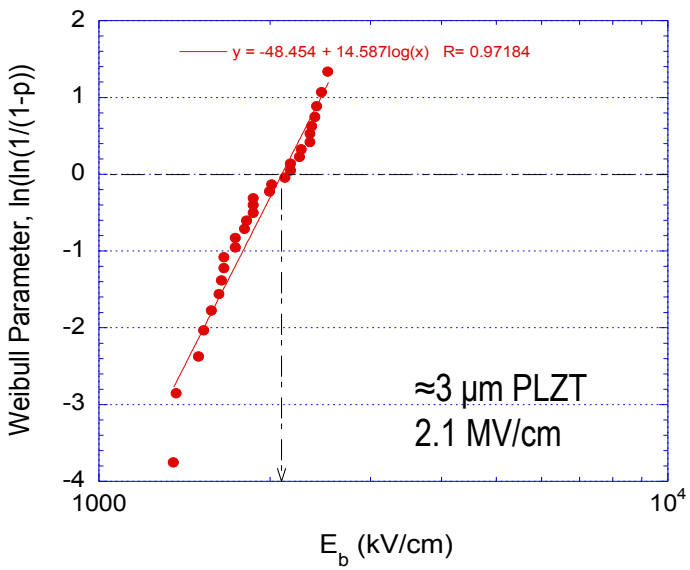
Test condition:

voltage: 100~1000V

step time: 0.5 sec

zero time: 1 sec

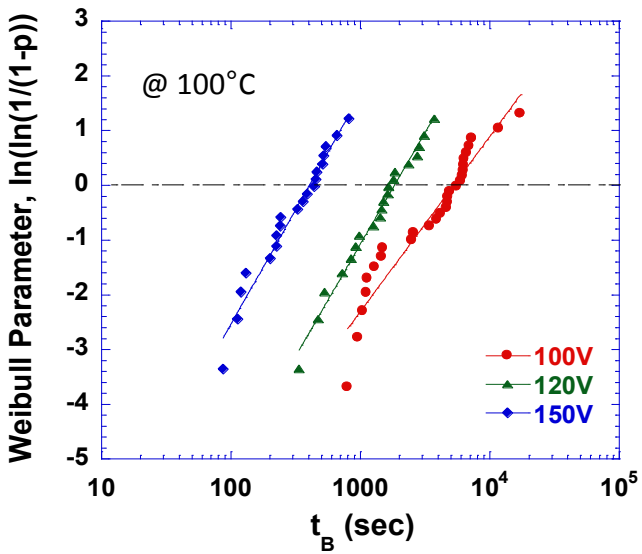


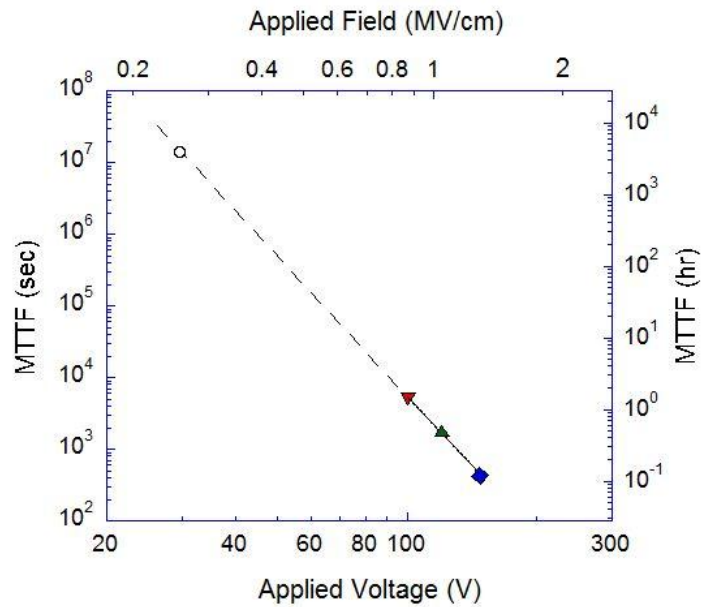


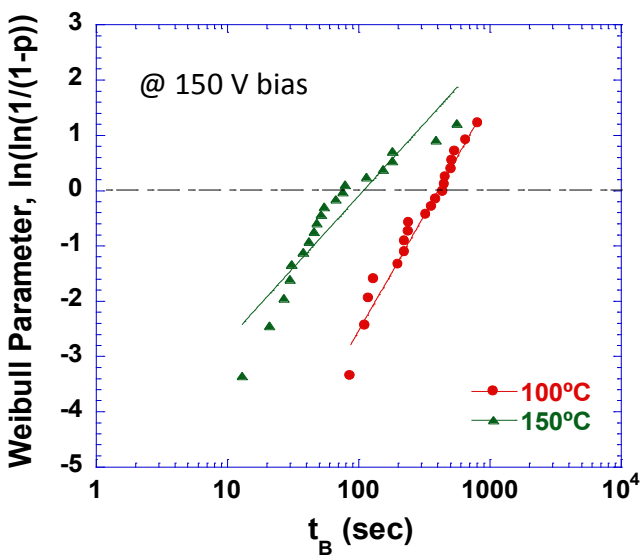
Two-parameter Weibull analysis

$$F(t) = 1 - \exp\left[-\left(\frac{t}{\eta}\right)^{\beta}\right], \qquad p = \frac{r - 0.3}{N + 0.4}$$

Life Prediction HALT Analysis





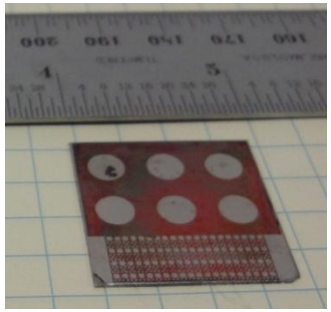


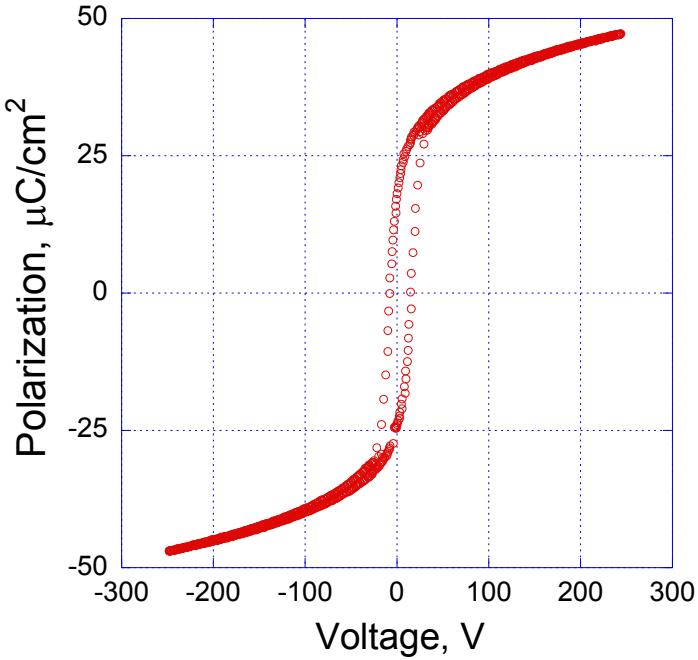
$$t = C \cdot V^N \exp\left(\frac{E_a}{k_B \cdot T}\right)$$

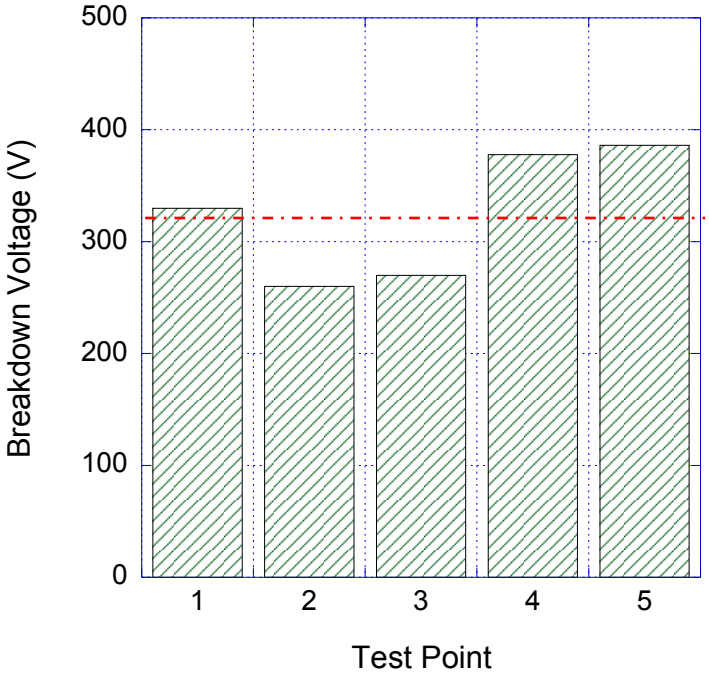
- **Lifetime estimated by measuring time to failure vs. applied voltage as function of temperature.**
- **Mean time-to-failure (MTF), voltage acceleration factor (N), and activation energy (E_a) are obtained from reliability measurement.**

Properties of 5-mm-dia. Caps

Hysteresis loop of a ≈3μm-thick capacitor with 5mm-dia Pt top electrode measured up to 250V



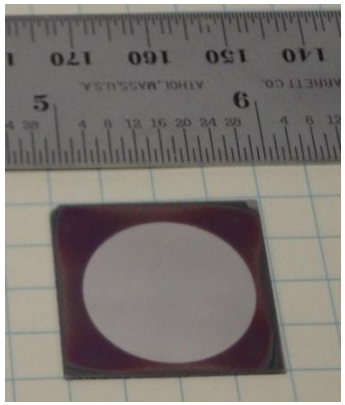


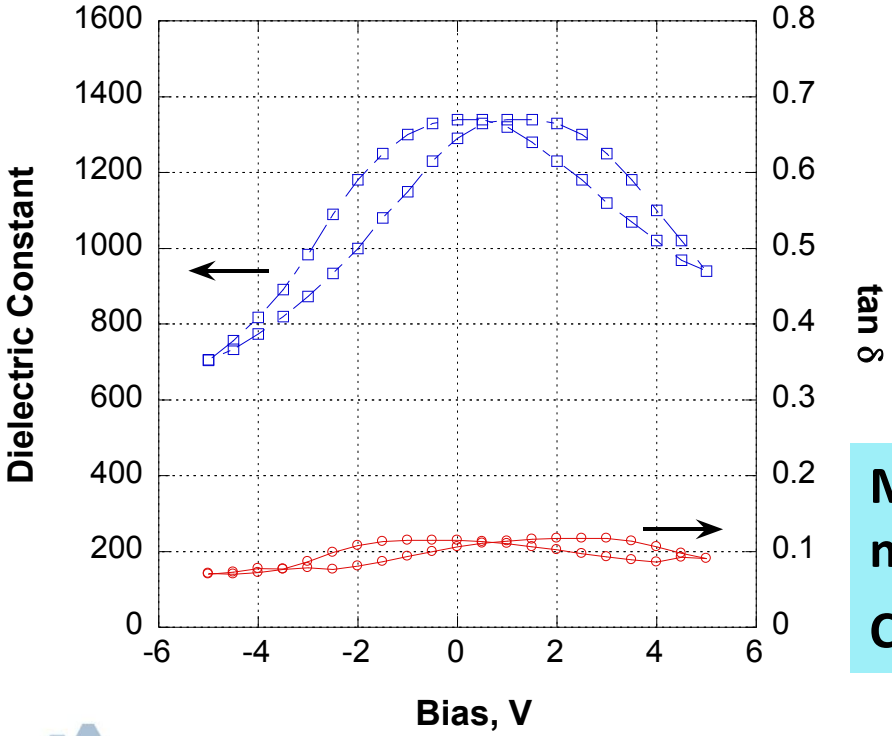


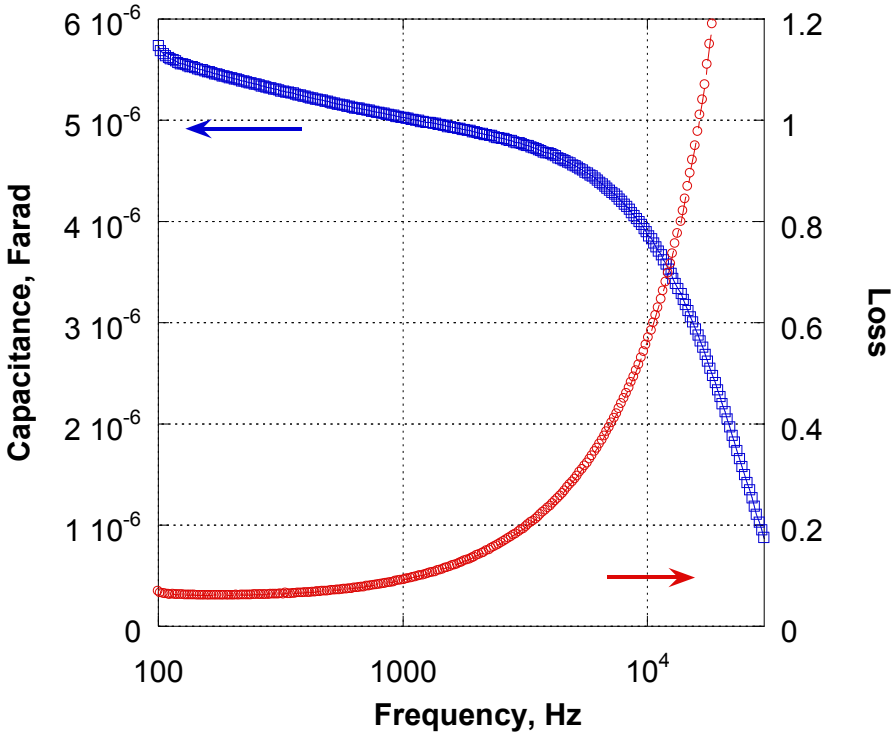
Breakdown voltages for the 5mm caps are: 330V, 260V, 270V, 378V, 386V (Average ≈325V)

Properties of Large Area Caps

20 mm dia.
Film-on-foil



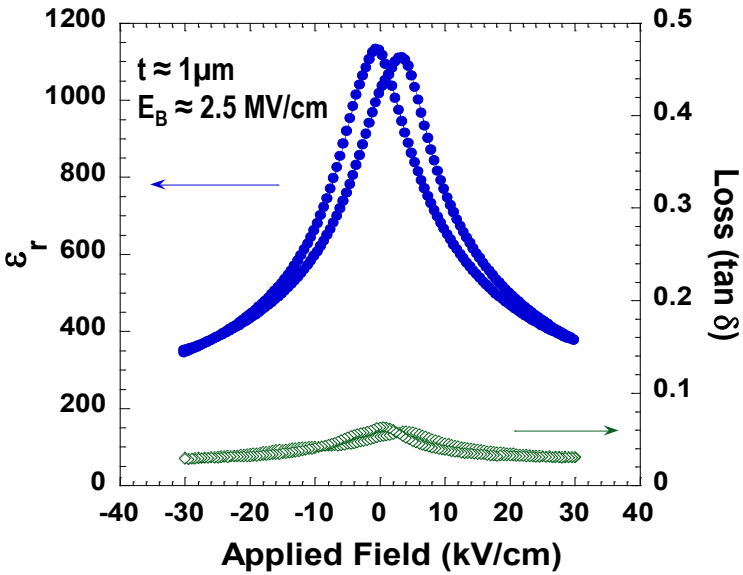


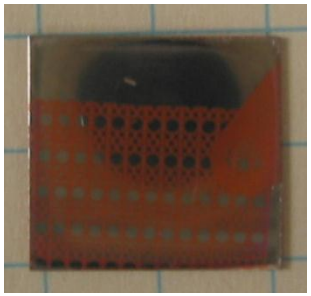


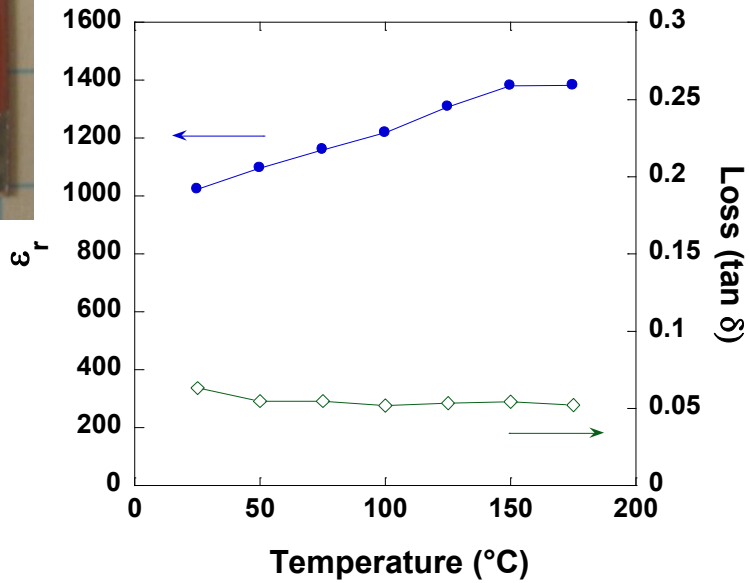
Measured on a 0.7 µm-thick PLZT film on nickel foil with 20-mm-dia Pt top electrode

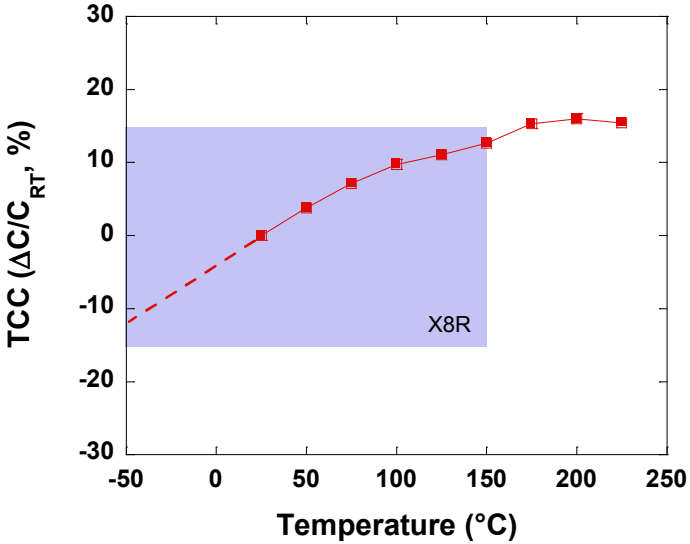
Capacitance ≈5 µF and Loss ≈0.1 at 1 kHz

PLZT on Copper Foil



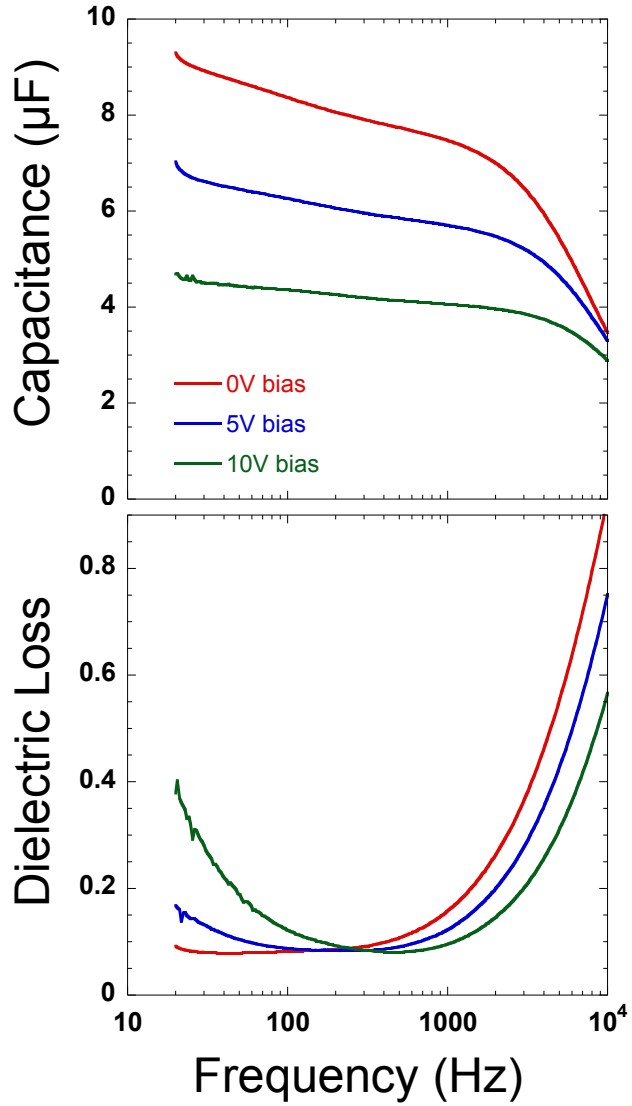




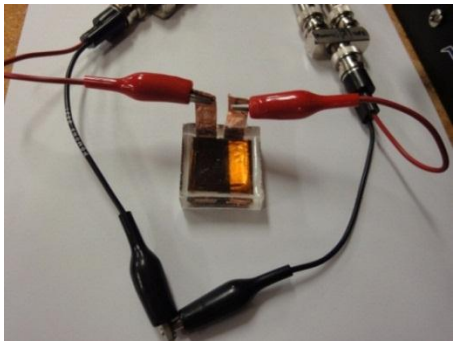


- Dielectric constant increased from ≈1100 to ≈1400 and loss factor decreased from 0.07 to 0.05 when temperature was increased from 25°C to 150°C.
- mean breakdown strength >2.0 MV/cm.
- Temperature variation is within the X8R specification.

Prototype Multilayer Capacitor with Leads



25 mm x 25 mm x 3 mm;
Capacitance ≈9 μF
Cap. density ≈4.8 μF/cm^3



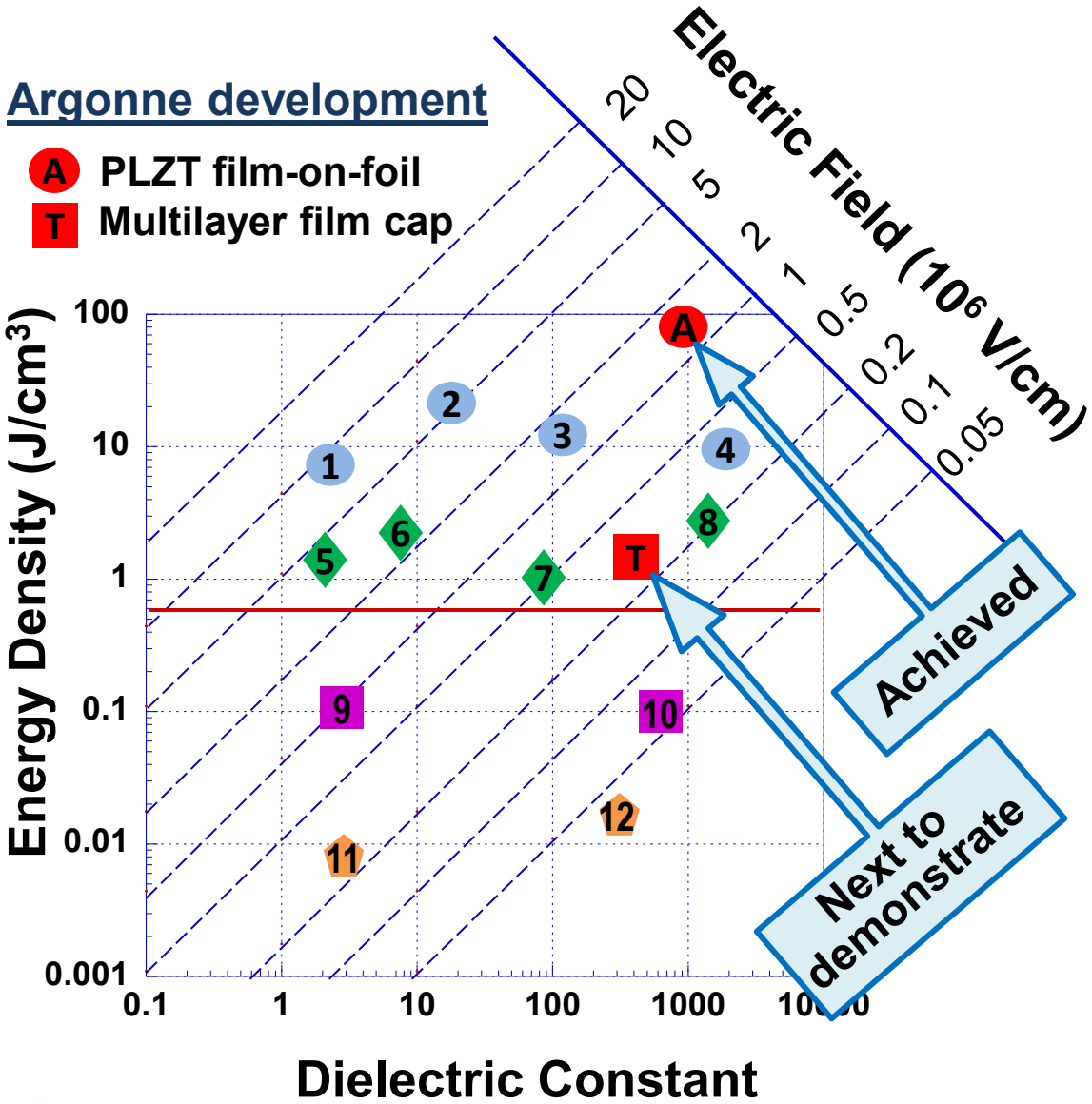
This capacitor was made with:

- Four film-on-foils that were coated with 20-mm-dia top electrodes
- Ni foil thickness ≈400 μm
- PLZT film thickness ≈1 μm
- Two Cu ribbons (thickness ≈80 μm)

PLZT film capacitors have potential to reduce size, weight, and cost, increase capacitance density, and enable high temperature operation, for use in power electronic systems in electric drive vehicles.

Achievement (Summary)

Energy Density of Dielectrics and Capacitors



Dielectrics

1. Polypropylene
2. Fluorpolymer, PSU
3. Titania
4. PZT ceramics, PSU

Pulsed power capacitor

5. Polypropylene cap
6. High-K polymer cap
7. NPO MLCC
8. X7R based MLCC

Power electronics capacitor

9. Polypropylene film cap
10. X7R Based MLCC

High temperature capacitor

11. High-T polypropylene cap
12. High-T 200°C MLCC

Summary

- Fabricated PLZT on Ni foils with $\varepsilon_r \approx 700$ & DF ≈0.07 @ -50°C; $\varepsilon_r \approx 1300$ & DF ≈0.08 @ room temperature; and $\varepsilon_r \approx 2200$ & DF ≈0.06 @ 150°C.
- Measured breakdown strength E_B >2.6 MV/cm, and leakage current density $I_{leakage}$ <10^{-8} A/cm^2.
- Measured energy density ≈85 J/cm^3 in a ≈3 μm-thick PLZT film-on-foil.
- Measured high dielectric constant ≈120 and loss ≈0.01 under bias field ≈1 MV/cm (300V bias).
- Fabricated PLZT on Cu foils with ε_r increased from ≈1100 to ≈1400 and loss factor decreased from 0.07 to 0.05 when temperature was increased from 25°C to 150°C, Breakdown strength >2 MV/cm, and $I_{leakage}$ <10^{-8} A/cm^2.
- Film-on-foil dielectrics were thermally cycled (≈1000 cycles) between -50°C and +150°C with no measurable degradation in capacitance.
- Fabricated a prototype multilayer capacitor of ≈9 μF capacitance with leads attached.

R&D Magazine
2011
R&D 100 Winners

Press release: *R&D* Editors Announce 2011 R&D 100 Awards

The 2011 R&D 100 Award Winners are listed below in alphabetical order by the name of the primary developer company.

Primary Developer	2011 R&D 100 Winning Technology (Category)	Co-developers/Contributors
A123 Systems Inc., USA	A123 Systems Nanophosphate Engine Start Battery (Energy Devices)	
Agilent Technologies, USA	Infiniium 90000 X-Series Oscilloscope (Electronic Instrumentation)	
Argonne National Laboratory, USA	Advanced Ceramic Film Capacitors for Power Electronics in Electric Drive Vehicles (Electrical Devices)	
Argonne National Laboratory, USA	Enhanced Renewable Methane Production System (Energy Devices)	Eurisko Scientific LLC
Argonne National Laboratory, USA	Integrated RF MEMS Switch/CMOS Device (Communication Technologies)	MEMtronics Corporation Innovative Micro Technologies Peregrine Semiconductor Advanced Diamond Technologies
Argonne National Laboratory, USA	Photoacoustic Spectroscopy (PAS) System for Remote Detection of Explosives and Chemicals (Safety and Security)	
Battelle Memorial Institute, USA	Resource Effective Bioidentification System (REBS) (Environmental Technologies)	
Black Box Network Services, USA	VirtuaCore Desktop Virtualization Computer Sharing (Information Technologies)	Fiddlehead Inc.

http://www.rdmag.com/Awards/RD-100-Awards/2011/06/R-D-100-2011-Winners-Overview/

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Collaborators:

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Thank you!