Electrostrictive Graft-Elastomers (G-Elastomers)
-Materials, Devices and Applications-
A Review

Ji Su
Advanced Materials and Processing Branch
NASA Langley Research Center, Hampton, VA 23681, USA
(ji.su-1@nasa.gov)

Chiba University
Chiba, Japan
August 26, 2019

Electroactive Polymers (EAP)

ELECTRONIC EAP

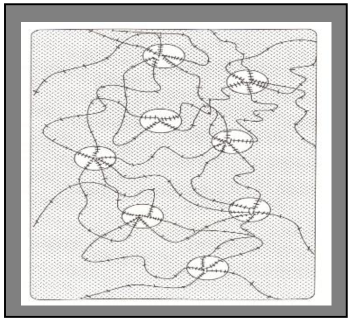
- Electroactive Semi-crystalline Polymers
  - Ferroelectric and piezoelectric polymers
    - Poly (vinylidene-flouride) (PVDF) and its copolymers
    - Odd-numbered nylons
- Electroactive Elastomers
  - Polyurethane (PU)
  - Dielectric Elastomers (DE)
  - Electrostrictive Graft Elastomers (G-elastomers)
  - Electro-Viscoelastic Elastomers (EVE)
  - Liquid Crystal Elastomers (LCE)
- Electroactive Papers
- Carbon Nanotubes (CNT)

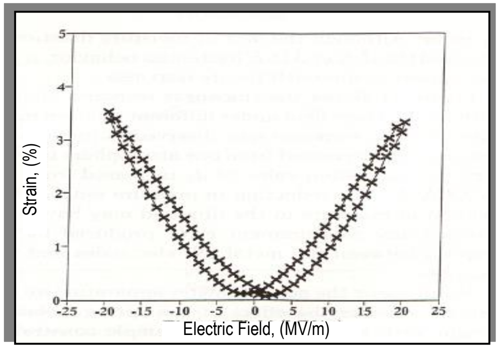
IONIC EAP

- Conductive Polymers (CP)
- Electro Rheological Fluids (ERF)
- Ionic Polymer Gels (IPG)
- Ionic Polymer Metallic Composite (IPMC)

Electromechanically Functional Elastomers

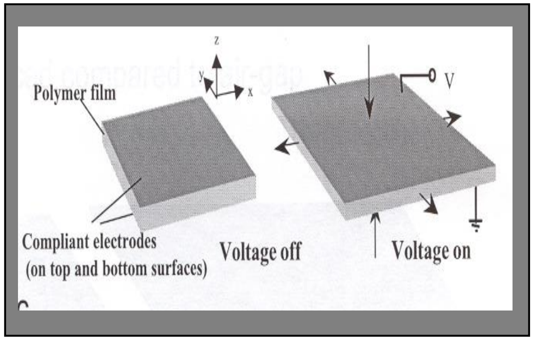
Polyurethane elastomers

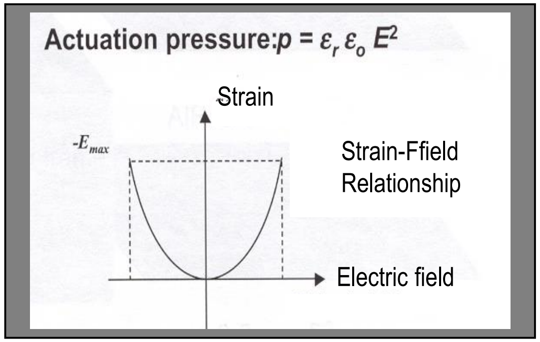




Physically cross-linked (hydrogen bonds) elastomers that offer field-induced strain by a combined mechanism.

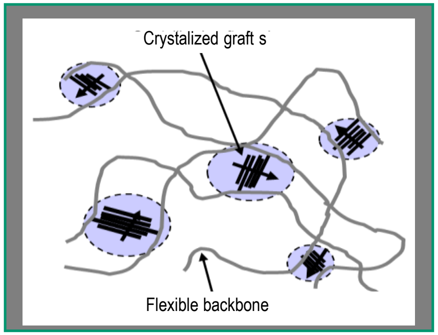
Electrostatic/dielectric elastomers

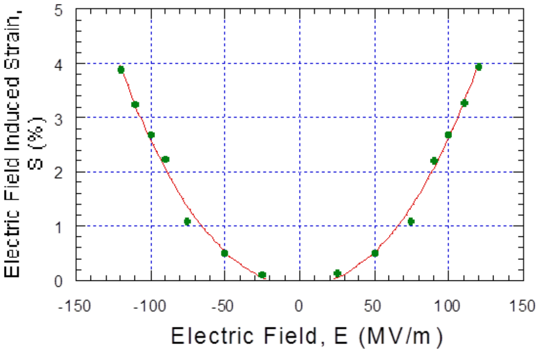




Chemically cross-linked soft elastomers that can offer a large ffield-induced strain due to the Maxwell Stress effect.

Electrostrictive graft elastomers





Physically cross-linked (formed crystal domains) elastomers that offer field-induced strain by primarily electrostriction.

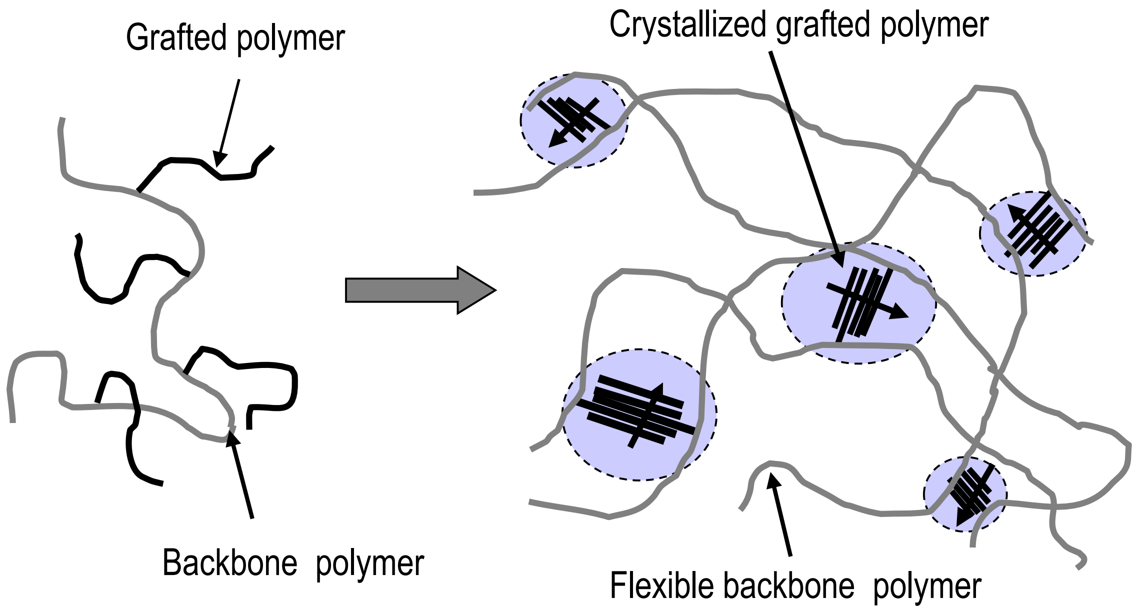
Ref.: Z. Ma, etc. p. 2721, Vol.32, Journal of Polymer Science: Part B, Polymer Physics (1992)
Ref.: "Electroactive Polymer (EAP) Actuators as Artificial Muscles: Reality, Potential and Challenges" by J. Bar-Cohen (SPIE, 2003).

Electrostrictive Graft Elastomers (G-elastomers)

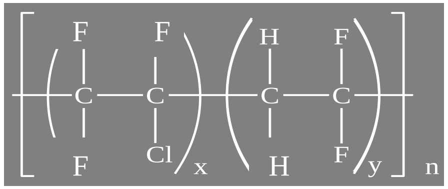
A Two Constituent System

1) Flexible backbone polymer that functions as a main contributor for elastomeric three-dimensional molecular network.

2) Grafted polymer that can crystallize (a) to form physical cross-linking sites for elastomeric three-dimensional network and (b) to produce polar domains that are electric-field responsive.

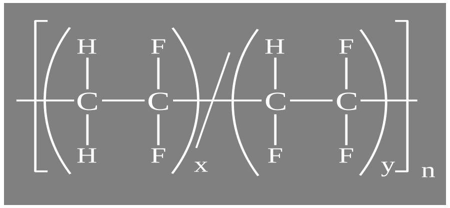


Structures and Functions of the Constituent Polymers



Backbone Polymer
chlorotriflouroethylene-vinylidene flouride copolymer
Nature: **non-crystallizable and flexible**

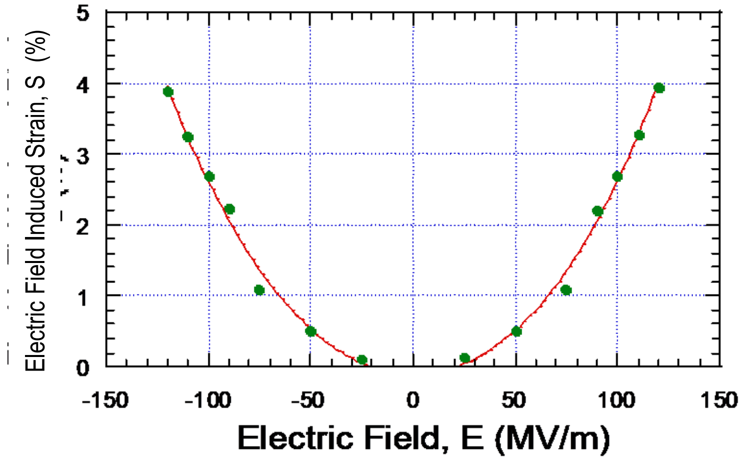
Functions
(a) to be the main contributor to three-dimensional elastomeric molecular network
(b) to provide space for dimensional change and adjust stiffness of the 3-D network

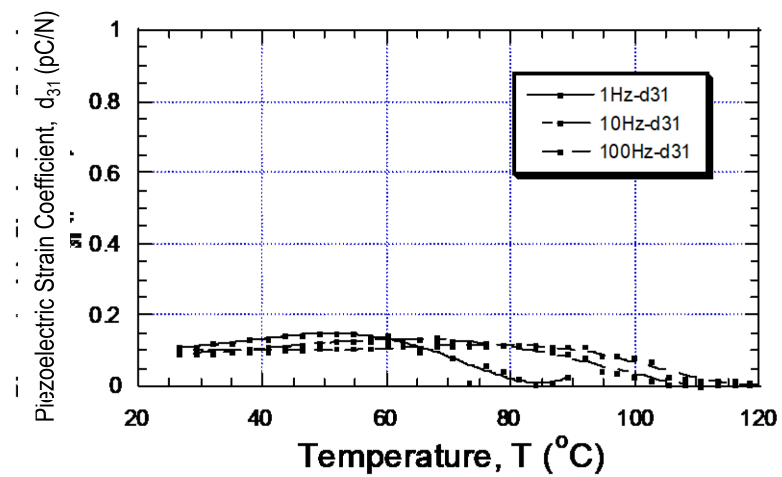


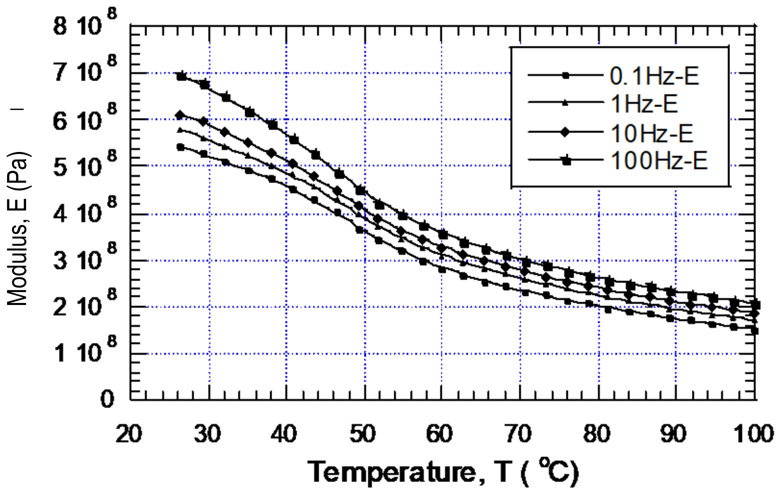
Grafted Polymer
trifluoroethylene-vinylidene flouride (VDF-TrFE) copolymer
Nature: **crystallizable and polar**

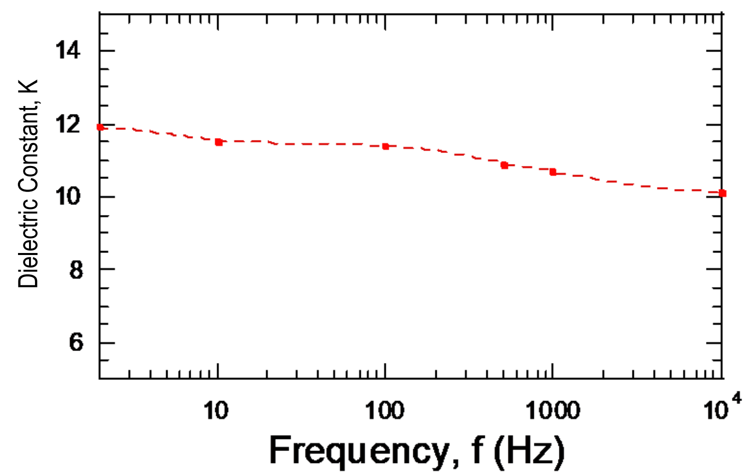
Functions
(a) to form physical cross-linking sites for elastomeric three-dimensional network
(b) to provide polar domains that are electric-field responsive and drive dimensional change

Important Properties of the Electrostrictive Graft Elastomers









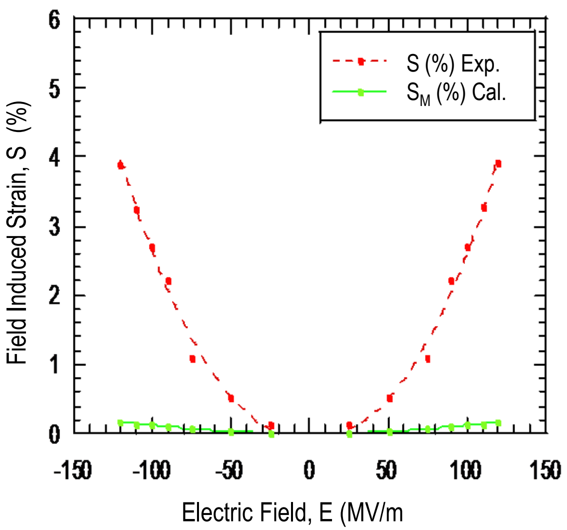
Mechanisms of Electric Field-induced Strain in Elastomers

Mechanisms

Electrostriction	$S_E = -Qe_o^2 (K-1)^2 E^2$
Maxwell effects	$S_M = -se_oKE^2 /2$

Contributions of Different Mechanisms

Materials	Maxwell Effect	Electrostriction
Graft elastomers	<5	>95
Polyurethane	35	65
Dielectric elastomers	100	0



Advantages of Electrostriction Mechanism

Increase both the field-induced-strain and the mechanical modulus simultaneously

Morphology-Property Control of Electrostrictive G-Elastomers

Molecular selection of flexible backbone and crystalized graft polymers

(dipole moment, crystal unit and crystal size as well as flexibility of the backbone polymers).

Fraction of the two constitutes (dielectric constant, mechanical properties, and electromechanical properties).

Processing of materials utilizing thermal, mechanical, and electrical processing techniques (morphology control and optimization of electric, mechanical, and electromechanical).

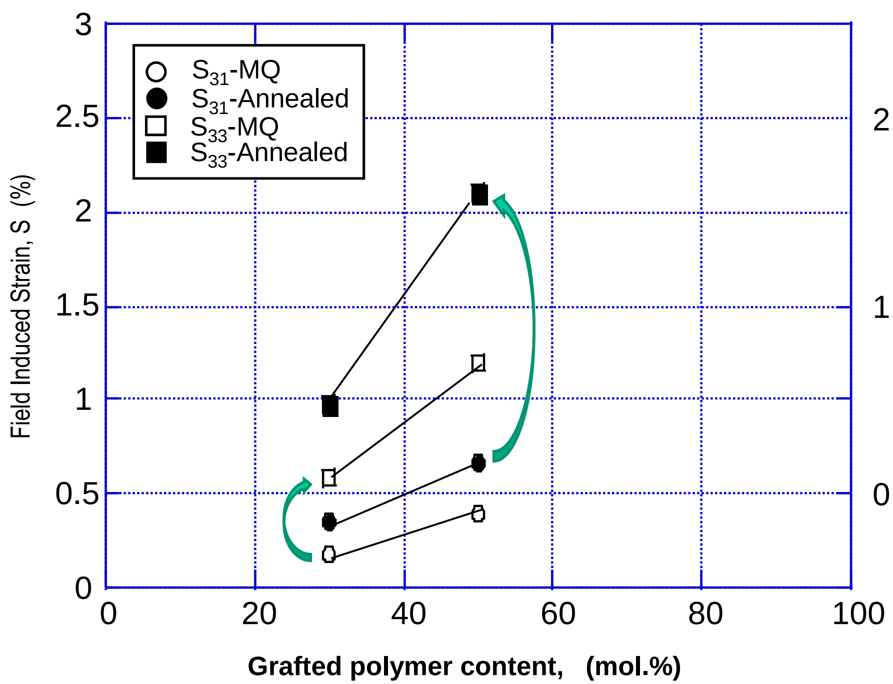
-The electromechanical properties of the electrostrictive G-elastomers depend on the fraction of the grafted polymer and the crystallinity formed by the polar grafted polymers.

-The fraction of the grafted polar polymers can be controlled through synthesis and the crystallinity can be controlled by thermal treatment and other processing.

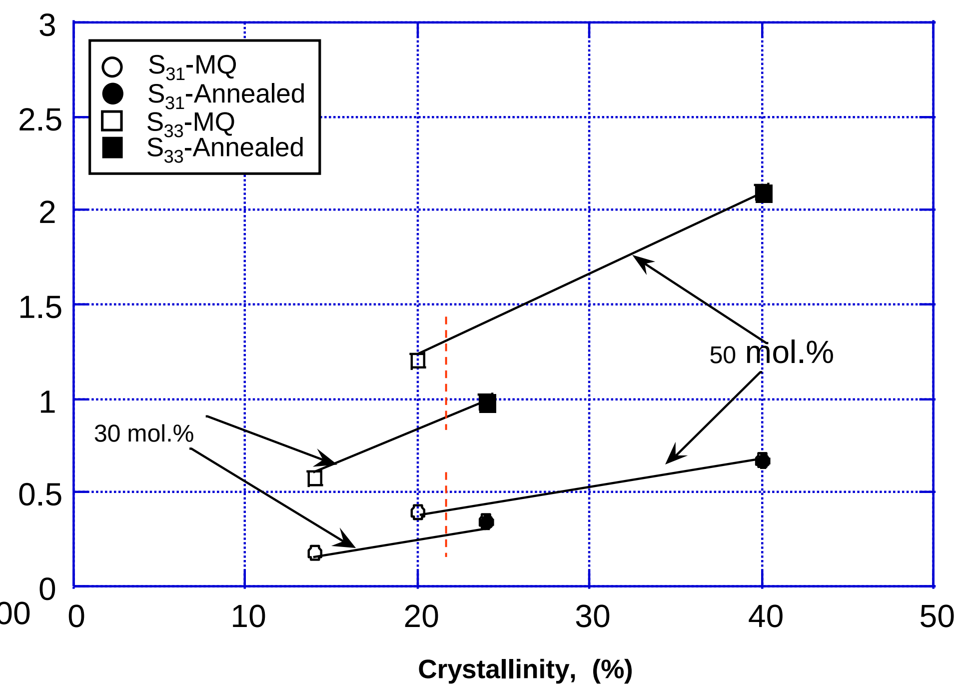
-Molecular engineering and morphological control can be employed to tailor and optimize the electromechanical properties of electrostrictive graft elastomers.

Effects of the Content and Crystallinity of the Grafted Polymer on Field-induced Strain of Electrostrictive G-Elastomers

Effects of the Content



Effects of Crystallinity



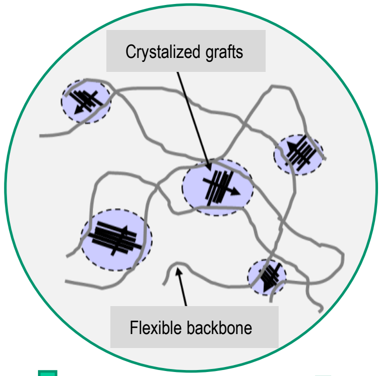
Comparison of Electromechanical Properties of Some Representative Electroactive Materials

Materials	Strain (s) (%)	Modulus (Y) (MPa)	Output Force (YS) (Mpa)	Salient Features
LaRC G-elastomer	4	580	23	conformable, high strain, lightweight
Polyurethane	4	20	0.8	high strain, low force
PVDF	0.3	1600	4.8	low strain
PZT	0.3	6400	192	brittle, heavy

Some Advantages of Electrostrictive G-Elastomers

-Large electric field-induced strain.
-Light weight and high output power density.
-Two constituent system with adjustable composition and morphology to optimize mechanical, electrical, and electromechanical properties.
-Excellent process-ability for low cost fabrication and device requirements in shape complexity.
-Excellent compatibility with other electroactive polymers for hybrid molecular systems offering multi-functions.

Research on Electrostrictive G-elastomers



Developed electrostrictive G-elastomer-based sensing-actuating dual functional polymers

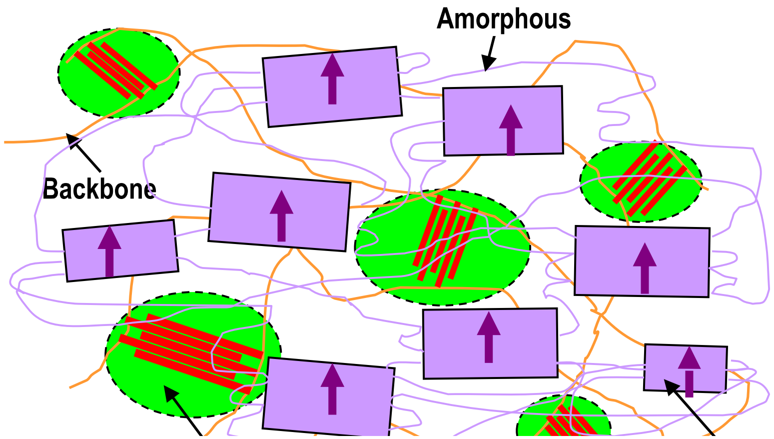
Developed electrostrictive G-elastomer-based nano-fibers (mats and oriented fibers)

Developed electrostrictive G-elastomer-based devices: bending actuators and hybrid actuators

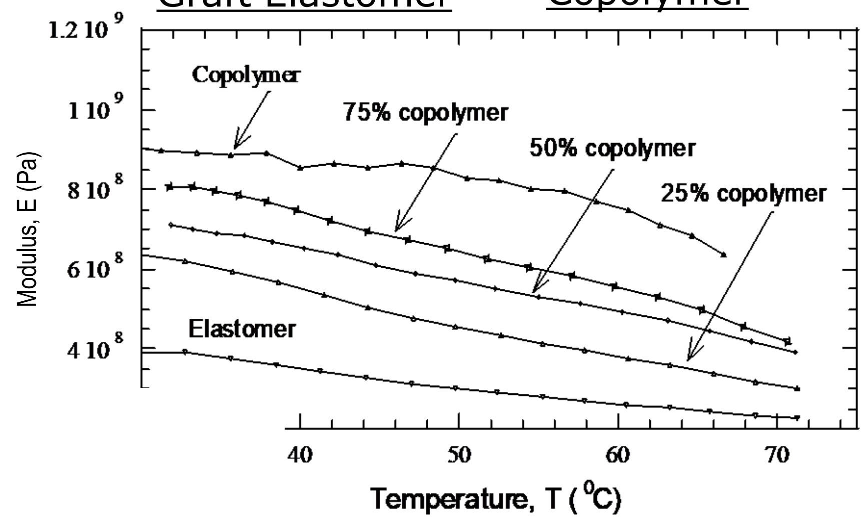
Applied hybrid concept proven by the hybrid ctuators in development of a series of high performance actuators, transducers, and energy harvesters

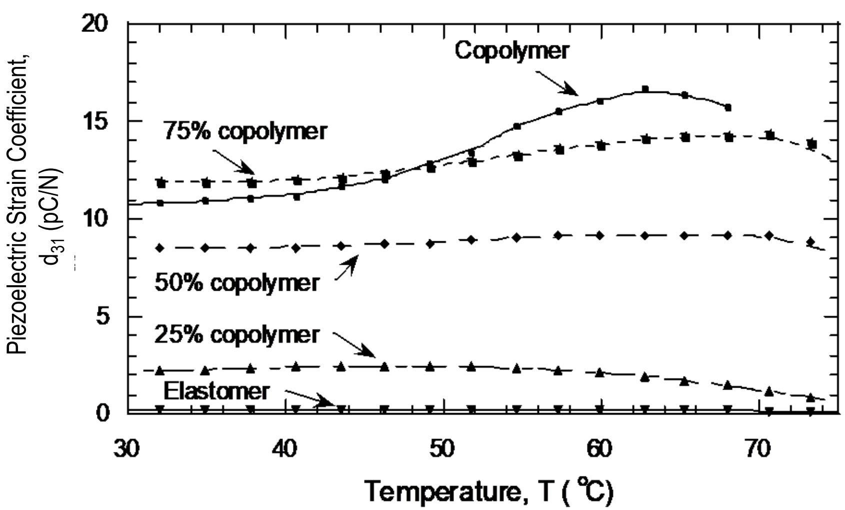
Conducted computational study on mechanism of electrostrictive G-elastomer and guidance for optimizing electrostrictive G-elastomers via molecular engineering route

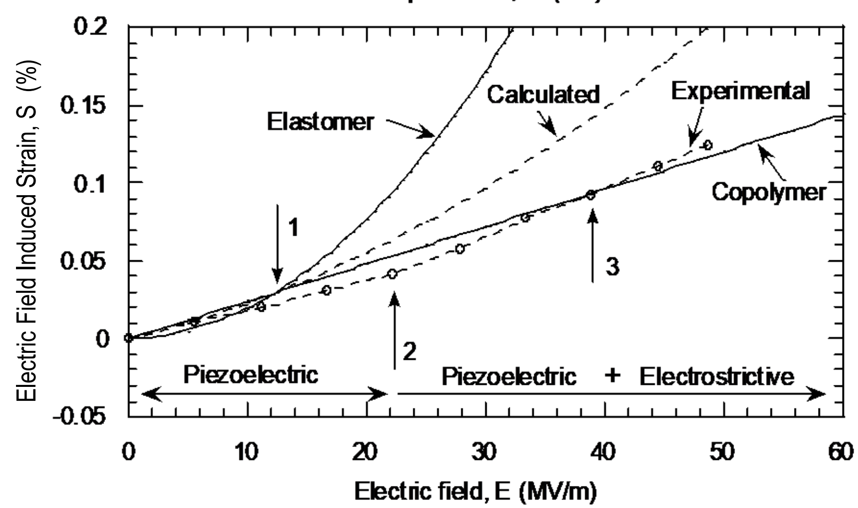
Sensing-Actuating Dual Functional Polymer Blends



Electrostrictive Graft Elastomer + Piezoelectric Copolymer

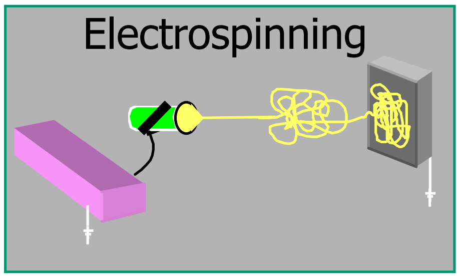


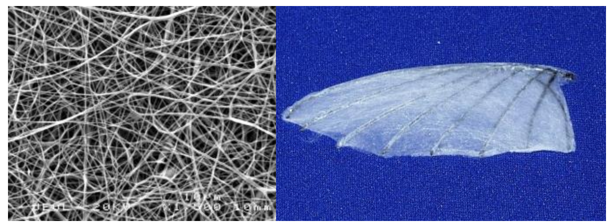


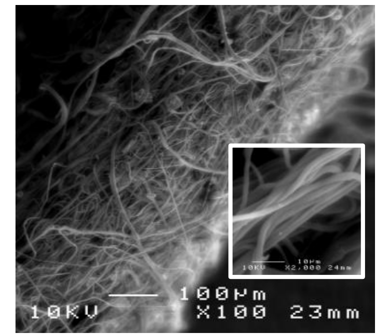


Electrostrictive G-elastomer Nano-fibers

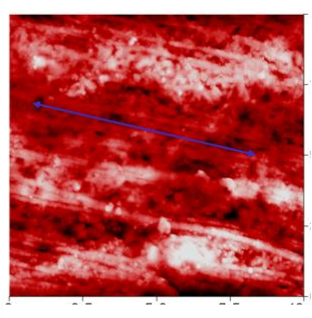
Nano-fibers and Mats by Electrospinning

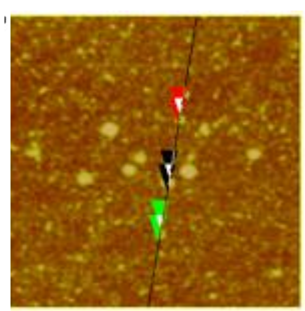


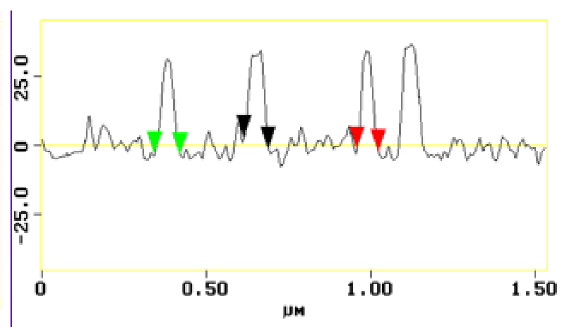


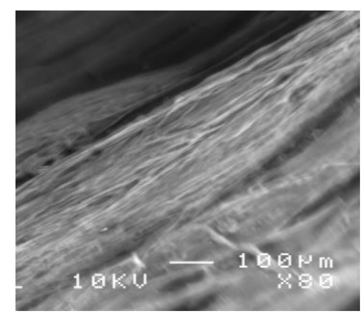


Highly Aligned Nano-fibers by Dynamic Precipitation



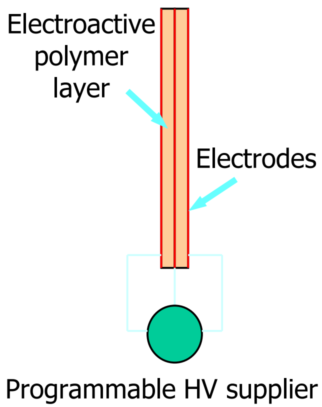


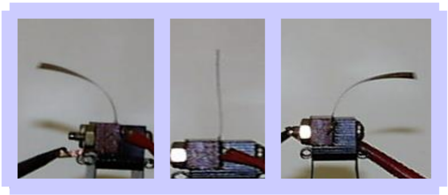


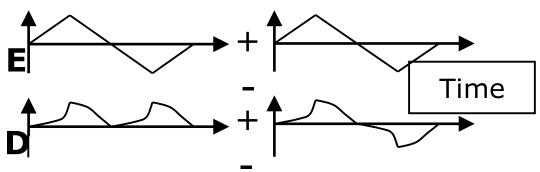


Actuators Using Electrostrictive G-Elastomer

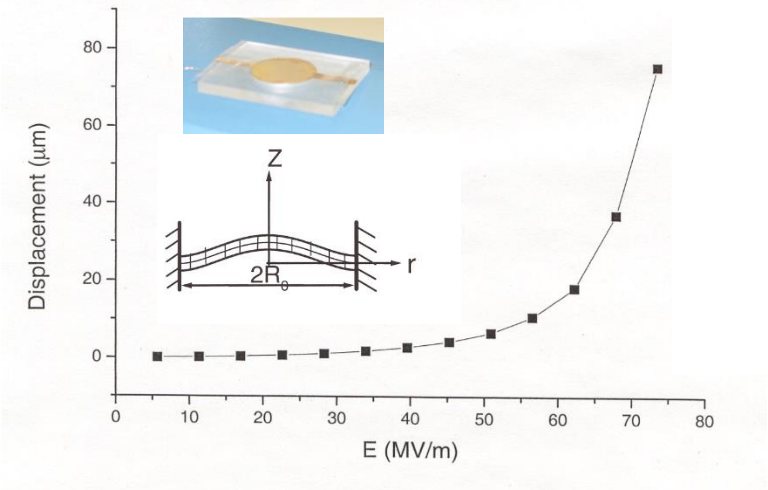
Two Direction Bending Actuator



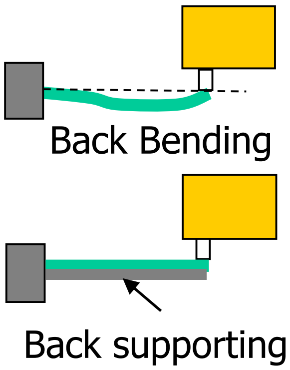


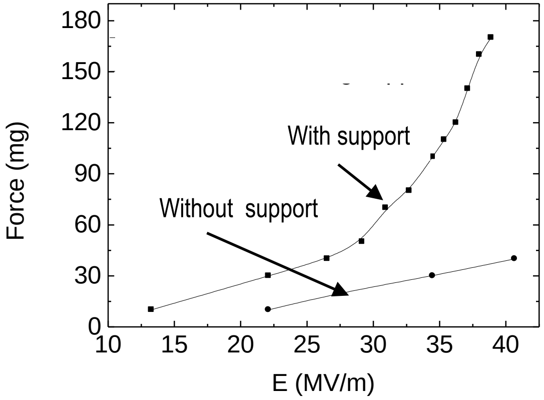


Circular Actuator



Backed Bending Actuator

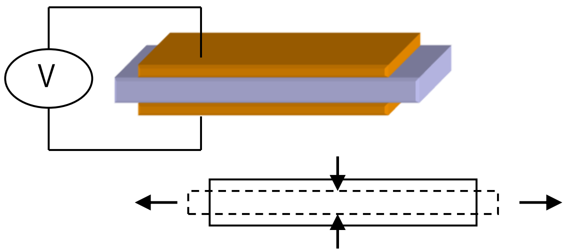




Hybrid Electromechanical Actuators

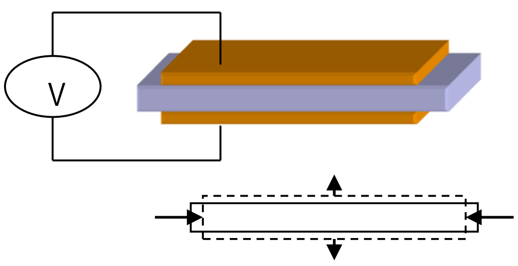
Electric Field-induced Strain

Electroactive polymer



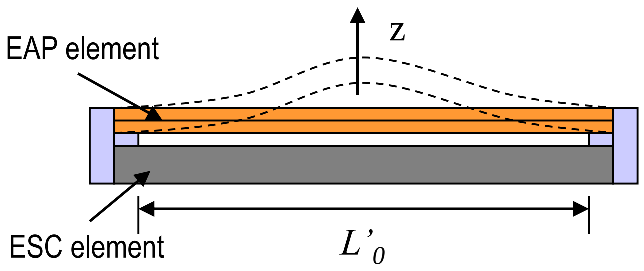
Longitudinal strain: **Negative**
Transverse strain: **Positive**

Electroactive Ceramics



Longitudinal strain: **Positive**
Transverse strain: **Negative**

Hybrid Actuator



EAP: Electrostrictive G-elastomer
ESC: Electroactive single crystal

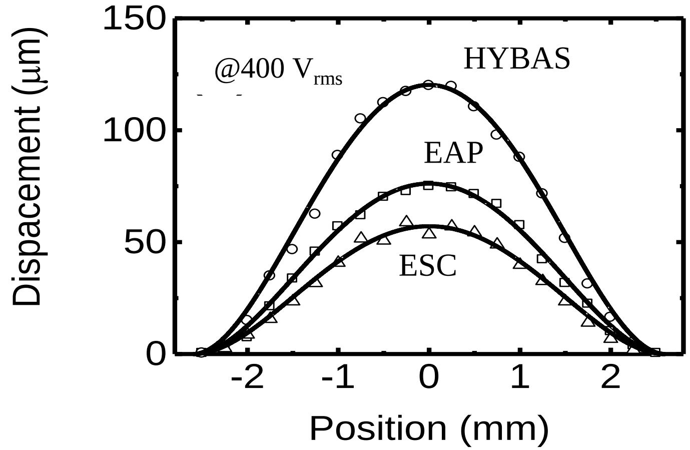
The HYBrid Actuation System (HYBAS) utilizes the characteristics of the electromechanical response of the two electroactive constituents cooperatively to achieve significantly increased displacement for actuation under one electric excitement source.

J. Su and T-B. Xu, US patent 7,394,181 "High Performance, High Energy Efficiency Hybrid Actuator Systems (HYBAS)"
J. Su, T. Xu, S Zhang, T. Short, & Q.M. Zhang, Appl. Phys. Lett. 85 (6), 1045 (2004)

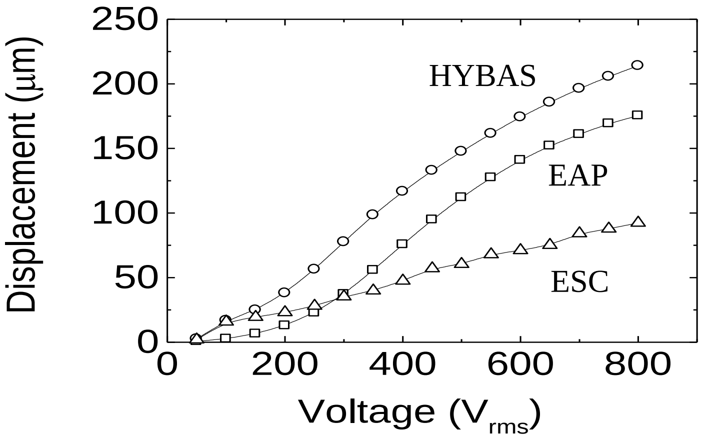
Performance of the Hybrid Electromechanical Actuators

Significantly increased displacement

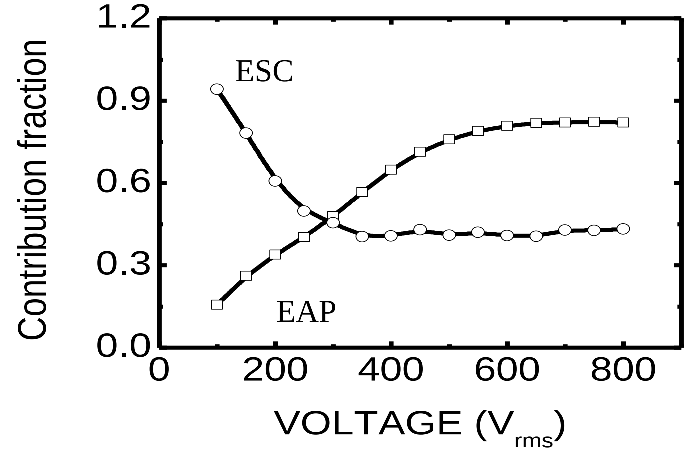
Proven hybrid concept
for advanced electromechanical devices



Comparison of Displacement of HYBAS and Its Constituents (at center)

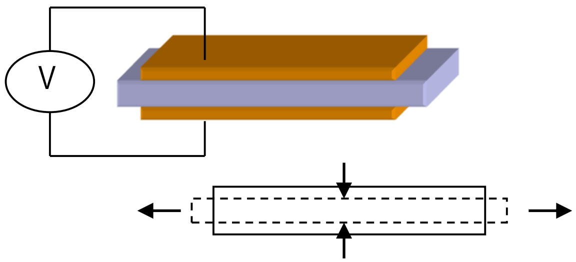


Voltage Dependence of Contributions of Each Constituent



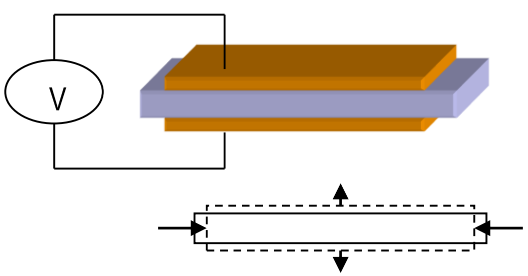
Generalization of Hybrid Concept and Its Extended Application

Active Constituent 1



Longitudinal strain: **Negative**
Transverse strain: **Positive**

Active Constituent 2



Longitudinal strain: **Positive**
Transverse strain: **Negative**

Design and Selection of Active Constituents

Purpose: to make the two active constituents respond in opposite directions when a driving electric field is applied.

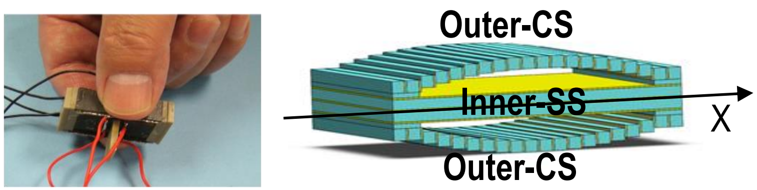
Single Material:
ceramics, polymers, single crystals, etc.

Composites:
ceramics/polymer, polymer/polymers, single crystal/ceramics, single crystal/polymers, etc.

Multilayer/stacks:
single material, composites, single material-based laminates, stacks, etc.

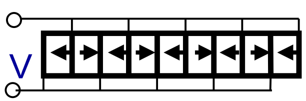
A Successful Sample: HYbrid Piezoelectric Energy Harvesting Transducer System (HYPEHT)

The Energy Harvester

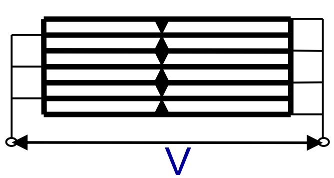


Dimensions: 30 mm (l) × 15 mm (w) × 11 mm (h)
Weight: 21 gram

Active Constituents



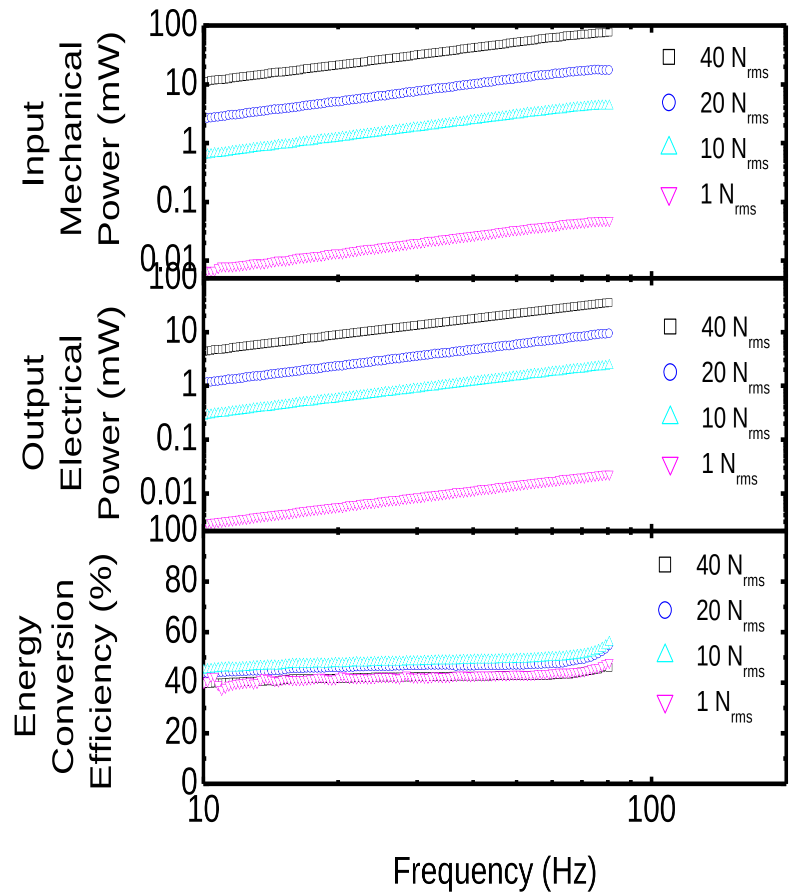
Outer Constituent:
Multi-stacked
d_{33} effect in x-direction



Inner Constituent:
Multi-layered
d_{31} effect in x-direction

Conversion Efficiency: >40%

Performance



- U.S. Patent 7,446,459 B2, "Hybrid Piezoelectric Energy Harvesting Transducer System," T-B Xu, X. Jiang, J. Su, P. W. Rehrig, and W. S. Hackenberger.

Recognition of the HYPEHT Energy Harvesting Technology

- A proven hybrid concept for advanced eletromechanical devices has been successfully applied to both actuation and broadband energy harvesting transducers.
- Innovation through multidisciplinary approaches

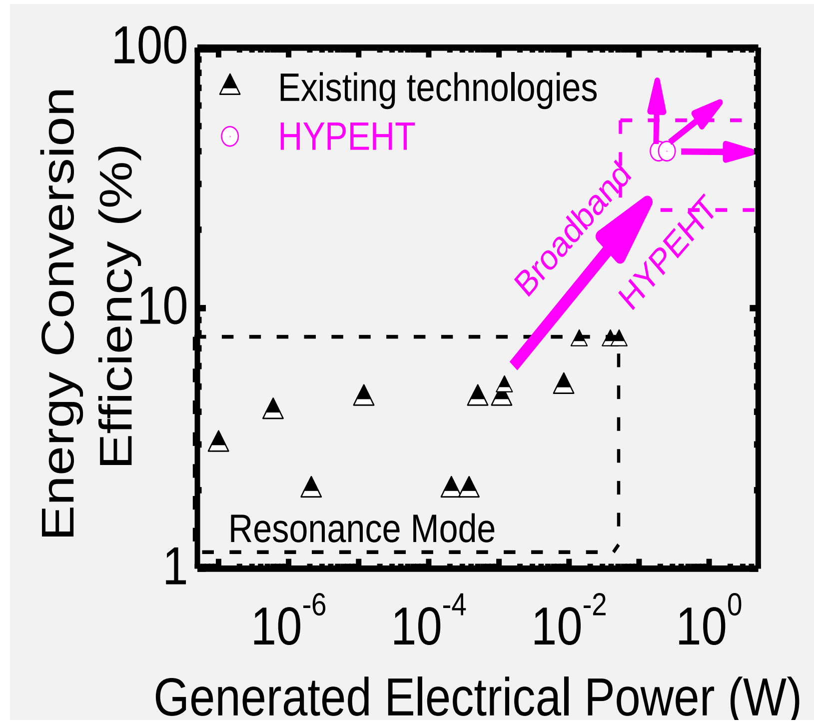
  1) to capture one order of magnitude more mechanical energy into piezoelectric structure

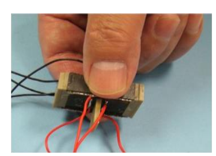
  2) to increase energy conversion efficiency

  3) to enhance energy charging/storage performance

More than **40%** mechanical to electrical energy conversion efficiency can be achieved.

- HYPEHT is a broadband harvester at both off-resonance frequencies and resonance frequency.





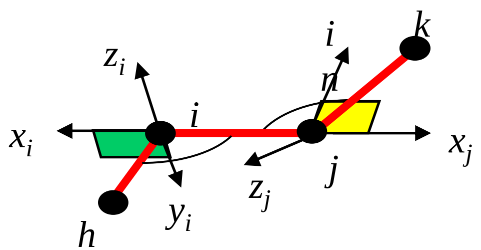
ENERGY HARVESTING & STORAGE USA
WIRELESS SENSOR NETWORKS & RTLS USA

IDTechEx
Winner 2011
Best Technology Development of Energy Harvesting

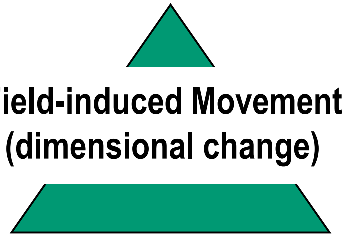
Computational Study of Mechanisms of G-elastomers

Formation of Element for the Model

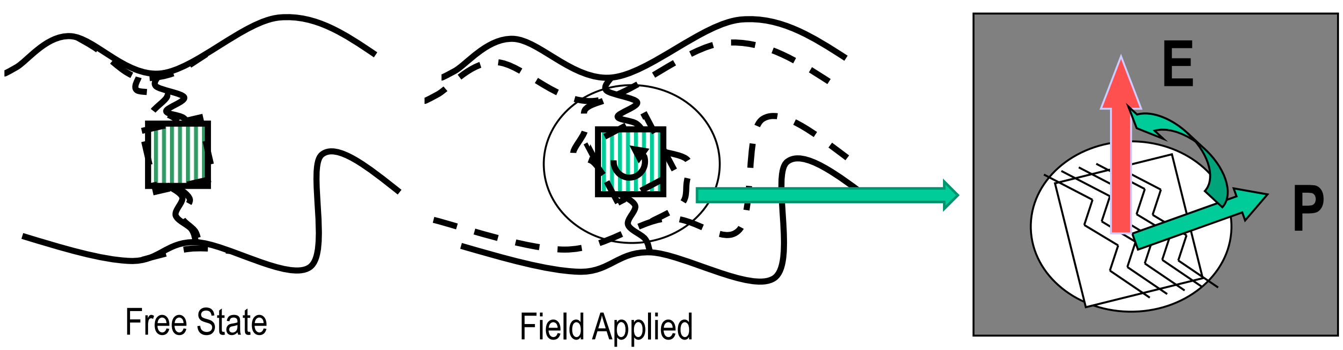
Chemical Bond Element



Key Parameters Considered



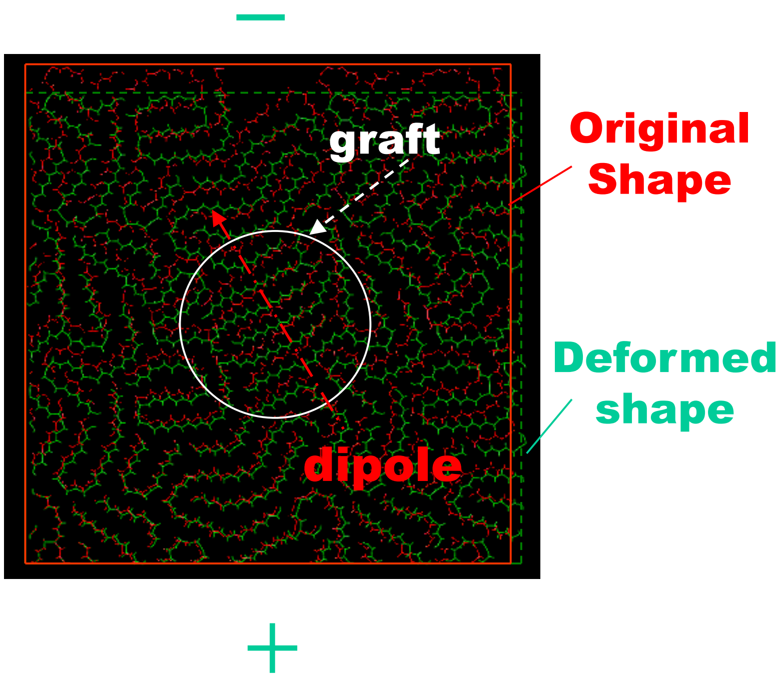
Deformation Mechanisms



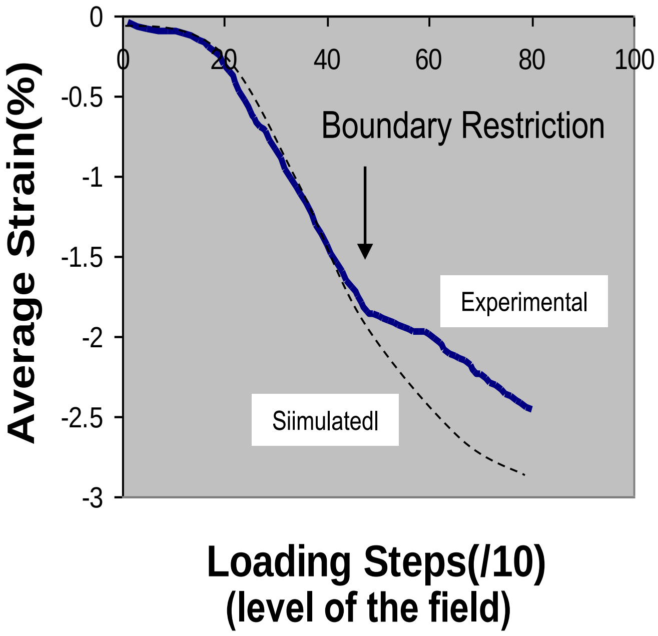
E: electric field
P: polarization

Comparison of Simulated Deformation of Electrostrictive G-elastomers and Experimental Results

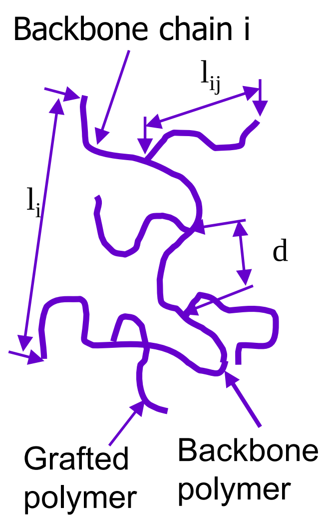
Simulated Deformation

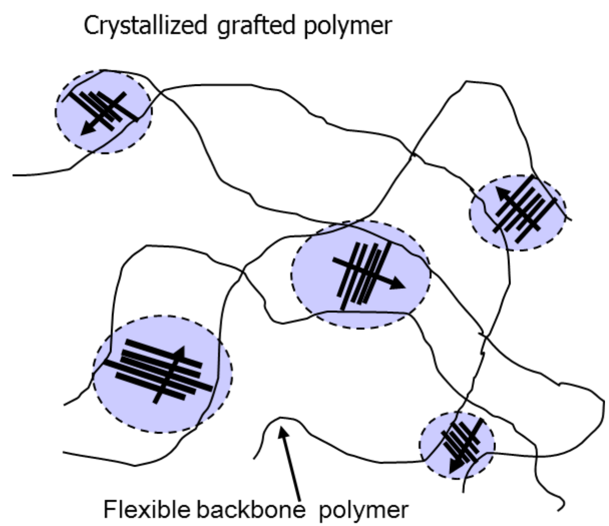


Comparison



Molecular Engineering for Property Control of Electrostrictive G-elastomers





Fraction of a grafted polymer
$F = S_iS_jl_{ij}/(S_il_i+\_S_iS_jl_{ij})$

Average length of a grafted polymers
$l = S_jl_{ij}/N_{ij}$

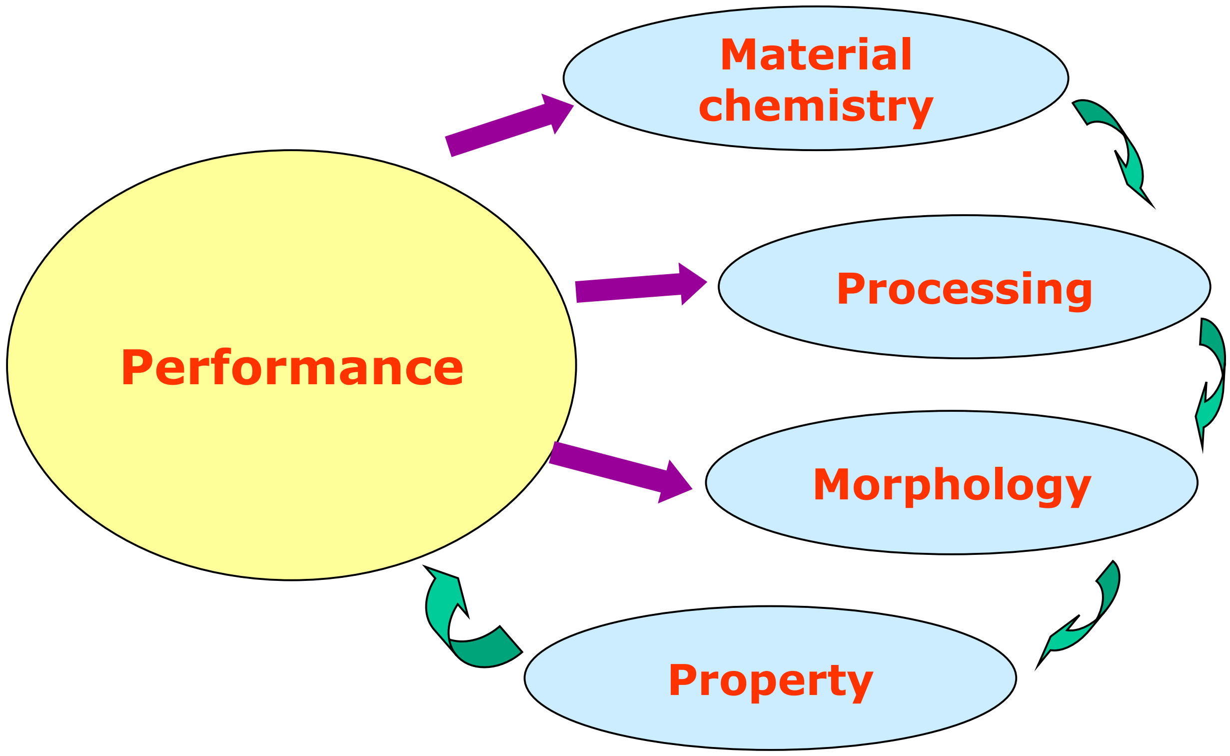
Average interval distance between the grafting sites
$d = l_{ij}/N_{ij}$

Total amount of field responsive grafts
- Fraction of grafted polymer $F = S_iS_jl_{ij}/(S_il_i+\_S_iS_jl_{ij})$

Crystal size and size distribution
- Average length of grafted polymers $l = S_jl_{ij}N_{ij}$

Cross-linking density and free volume
- Average interval between grafting sites $d = l_i/N_{ij}$

The Route to the Desired Electroactive Polymers



Machine Learning?

Summary, Challenges and Opportunities

Summary

-As a new class of electroactive polymers, **Electrostrictive G-elastomers** have demonstrated promising electromechanical properties with some advantages over other electroactive polymers.

-A device using the G-elastomer has proven a Hybrid Concept that provides a new route of developing high performance electromechanical devices.

-Computational study demonstrated the key factors that control the electromechanical properties of the G-elastomers

Challenges

Low voltage operation
Flexible, durable and low constraint electrode
High conversion efficiency
Low dielectric loss
Low mechanical loss
Thermal stability
Electric leaking

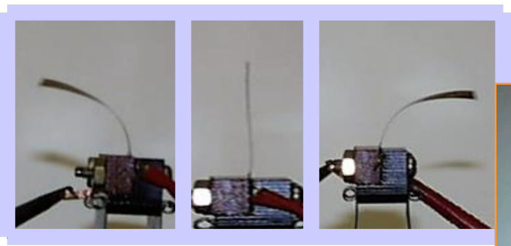
Challenges = Opportunities

In order to make EAPs that may be used in many possible applications, the electroactivities need to be **intrinsic**. Molecular Engineering should be one of the ways to achieve the goal.

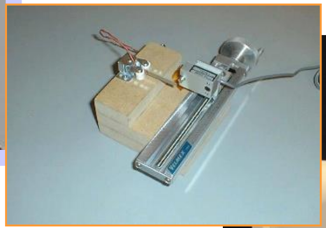
Some Applications in NASA Research Activities

Aerodynamic control
Deformable mirror array
Acoustic transducers
Passive optics
Tunable antennas

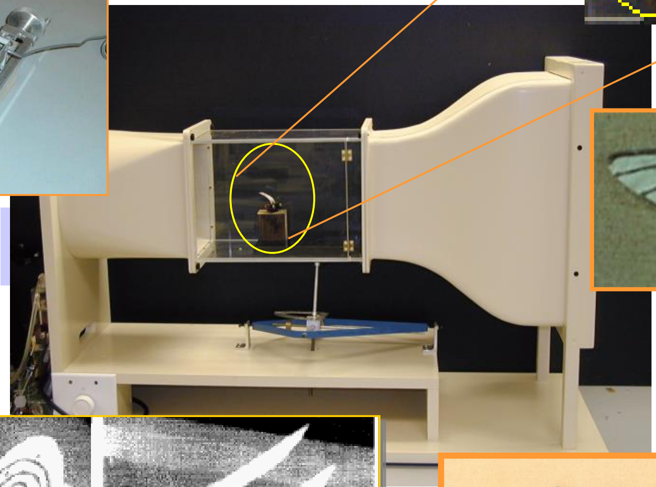
Bending actuators

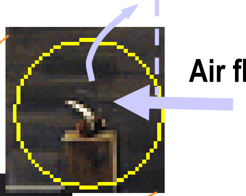


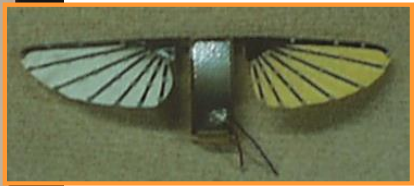
Force-test



Wind-tunnel test

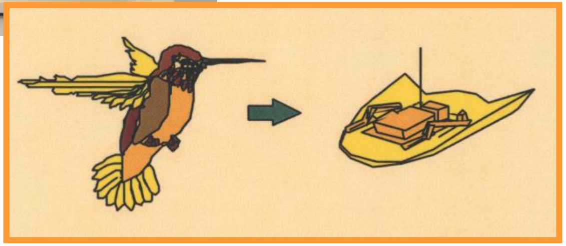




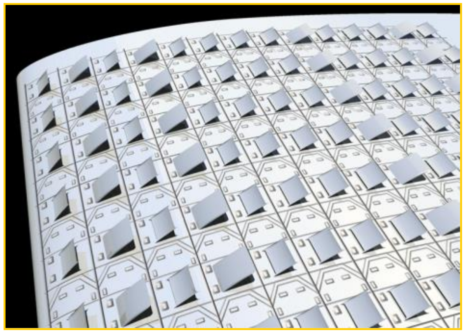


Biomimetic

Micro-air vehicle

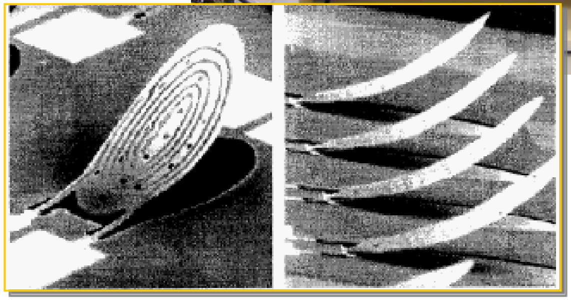


Sensor/actuator array



Self-sensing actuator array

(NASA-LaRC)



Acknowledgements

Dr. Tian-Bing Xu
National Institute of Aerospace (NIA)
Prof. Xiaoning Jiang and Laura Tolliver
NC State University
Prof. Lei Zuo and Dr. Wanlu Zhou
Virginia Tech
Drs. Shihai Zhang and Shan Wu
PolyK Technologies, LLC
Paul W. Rehrig
Saint-Gobain Innovative
Dr. Wesley S. Hackenberger
TRS Technologies
Dr. Muralidharan S. Nair
National Science Foundation

Dr. Qiming Zhang
Dr. Zoubeida Ounaies
Pennsylvania State University
Dr. Jerry I Scheinbeim
Rutgers University
Dr. Roy Kornbluh
SRI International
Dr. Joycelyn Harrison
Air Force Research Office
Dr. Yoseph Bar-Cohen
Jet Propulsion Laboratory
Dr. Peter X. Ma
University of Michigan

Dr. Peter Lellihi
Dr. William Cooks
Rheal Turcotte
Dr. Emilie Siochi
Dr. Kristin Pawloski
Dr. Terry St. Clair
NASA Langley Research Center
Ray Baughman
Lockhead Martin Corp
Dr. Changjie Sun
Dr. Chongjian Zhang
Dr. Youqi Wang
Kansas State University