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A soft and conductive PDMS-PEG block copolymer as a compliant electrode for dielectric elastomers

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Annual Polymer Day 2015
Motivation

Principle of dielectric elastomer (DE) as an actuator:

Requirement of compliant electrodes: 1) Inherently soft 2) conductivity
Stereotypes of electrodes

1) A conductive material is generally non-stretchable.

2) A stretchable material is usually non-conductive.

Our goal: soft-conductive polymer
Conventional electrodes for DEs

1) Losse carbon black
   - Samuel Rosset (EPFL)
   - Helmut Schlaak (University of Darmstadt)

2) Carbon grease
   - Samuel Rosset (EPFL)

Alternative electrodes:
1) Ionic conductor (hydrogel)
2) Silver nanowires
3) Conductive rubber
PDMS3-PEG copolymer

1. Hydrosilylation reaction of PDMS-PEG copolymer:

\[
\begin{align*}
\text{PDMS3-PEG} & \quad \rightarrow \quad \text{Stiff} \\
\text{at } 60^\circ \text{C} \\
\end{align*}
\]

2. Conductivity (PDMS-PEG copolymers)\(^1\)

PDMS3-PEG \rightarrow \text{high conductivity (}10^{-8}\text{ S/cm)}

3. Linear viscoelasticity-LVE (PDMS-PEG copolymers)\(^1\)

\[\text{Storage modulus, } G'(\text{Pa})\]

\[\text{Loss modulus, } G''(\text{Pa})\]

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Compliant electrodes | PDMS-PEG | MWCNTs | Dielectric properties | Rheology | Stress-strain

5 DTU Chemical Engineering, Technical University of Denmark 30 June 2016
Chain-extended PDMS3-PEG copolymer

1. To obtain a soft-conductive polymer → Chain extended PDMS-PEG copolymer

2. Crosslinked copolymer: Chain-extended PDMS-PEG copolymer + 15-functional vinyl crosslinker + 30 ppm Pt catalyst
Multi-walled carbon nanotubes (MWCNTs)

1. \(\downarrow\) conductivity (PDMS3-PEG) \(\rightarrow\) add conductive nanofillers (MWCNTs)

2. Obstacle \(\rightarrow\) MWCNTs entangle

![SEM image of pure MWCNTs showing entanglements.](image)

3. Dispersion methods:

<table>
<thead>
<tr>
<th>Chemical</th>
<th>Mechanical</th>
</tr>
</thead>
<tbody>
<tr>
<td>Oxidation process by acid e.g. (\text{HNO}_3) &amp; solution of (\text{H}_2\text{O}_2/\text{NH}_4\text{OH})</td>
<td>1) Probe sonicator 2) Ball milling</td>
</tr>
<tr>
<td>Drawback: intrinsic properties of MWCNTs are destroyed due to structural defects</td>
<td>Drawback: rupture MWCNTs into smaller lengths</td>
</tr>
</tbody>
</table>

4. Non-covalent physical treatment

![Mechanism of flocculation of CNTs via surfactant molecules.](image)

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Multi-walled carbon nanotubes (MWCNTs)

- Dispersion of MWCNTs → Rastogi et al.¹, Geng et al.² and Goswami et al.³

1. Dispersion of MWCNTs using surfactants. J Colloid Interface Sci 328:421–428


3. Dielectric properties of ultraviolet cured poly(dimethyl siloxane) sub-percolative composites containing percolative amounts of multi-walled carbon nanotubes. RSC Adv 5:12792–12799

Stability versus time for a reference method (MWCNT/NMP/Triton X-100) dispersed by a mechanical shaker at 23 °C: a) Immediately b) 5 min c) 30 min d) 60 min.

Stability versus time for MWCNT/NMP/Triton X-100 dispersed by water-bath ultrasonication at 23 °C for 6 hours: a) Immediately b) 5 min c) 30 min d) 60 min.

Optical microscope image of this film containing MWCNTs (0.07 phr) in PDMS-PEG matrix.
Conductivity & permittivity

Fig. 1

Conductivity (S/cm) vs. Frequency (Hz)

- 0CNT Si3PEG_H25
- 1CNT Si3PEG_H25
- 2CNT Si3PEG_H25
- 3CNT Si3PEG_H25
- 4CNT Si3PEG_H25
- LR 3162

Retest with normal force = 10N
Modulus

![Graph showing storage modulus and modulus loss factor vs frequency for different samples.](image)

- **Storage modulus (Pa):**
  - 0CNT Si3PEG_H25
  - 1CNT Si3PEG_H25
  - 2CNT Si3PEG_H25
  - 3CNT Si3PEG_H25
  - 4CNT Si3PEG_H25
  - LR 3162

- **Modulus loss factor:**
  - Same samples as above

**X-axis:** Frequency (Hz)
**Y-axis:**
- Storage modulus (Pa)
- Modulus loss factor
Stress-strain plots

![Stress-strain plots diagram](image)

- 0CNT Si3PEG_H25
- 1CNT Si3PEG_H25
- 2CNT Si3PEG_H25
- 3CNT Si3PEG_H25
- 4CNT Si3PEG_H25
- LR 3162

Y = 0.23 MPa
Y = 0.47 MPa
Y = 0.92 MPa
Y = 0.70 MPa
Y = 0.26 MPa
Y = 1.17 MPa
Conclusion

• The cross-linked conductive PDMS-PEG copolymers were successfully prepared with addition of different MWCNT concentrations.
• The conductivity of the chain-extended elastomers increases nearly to $10^{-3} \text{ S/cm}$;
  - $< \text{LR3162} = 10^{-1} \text{ S/cm}$
• The mechanical properties of chain-extended PDMS-PEG copolymers with MWCNTs ($< 3$ phr) indicate soft networks with low modulus losses.
• Future work:
  - The conductivity can be improved by adding silver nanoparticles in the system if properly designed.
  - Measure the conductivity of samples in “stretch” mode.
Acknowledgement