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

A Razak, Aliff Hisyam

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

Aliff H. A Razak
Danish Polymer Centre (DPC)
Main supervisor: Anne Ladegaard Skov
Co-supervisor: Peter Szabo

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DTU Chemical Engineering
Department of Chemical and Biochemical Engineering
Motivation

Principle of dielectric elastomer (DE) as an actuator:

Requirement of compliant electrodes: 1) Inherently soft 2) high 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:

\[
\text{PDMS3-PEG} + \text{Pt}^2+ \xrightarrow{60 \degree C} \text{PDMS3-PEG} \rightarrow \text{high conductivity (10}^{-8} \text{ S/cm)}
\]

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

\[
\begin{align*}
\text{Conductivity, } \sigma \text{ (S/cm)} & \quad \text{Frequency (Hz)} \\
10^{-13} & \quad 10^{-12} & \quad 10^{-11} & \quad 10^{-10} & \quad 10^{-9} & \quad 10^{-8} & \quad 10^{-7} & \quad 10^{-6} \\
10^{-11} & \quad 10^{-10} & \quad 10^{-9} & \quad 10^{-8} & \quad 10^{-7} & \quad 10^{-6}
\end{align*}
\]

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

\[
\begin{align*}
\text{PDMS3-PEG} & \rightarrow \text{Stiff} \\
\text{Storage modulus, } G' \text{ (Pa)} & \quad \text{Loss modulus, } G'' \text{ (Pa)} \\
10^{1} & \quad 10^{2} & \quad 10^{3} & \quad 10^{4} & \quad 10^{5} & \quad 10^{6} & \quad 10^{7} & \quad 10^{8} & \quad 10^{9} & \quad 10^{10} & \quad 10^{11} & \quad 10^{12} & \quad 10^{13}
\end{align*}
\]

Chain-extended PDMS3-PEG copolymer

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

![Chemical structure](image)

PDMS-PEG (vinyl terminated)

PDMS232 (hydride terminated)

23 deg. C

Pt²⁺

(PDMS - PEG) - PDMS232 (hydride terminated)

Mn = 38 kg/mol

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

1. ↓ conductivity (PDMS3-PEG) → add conductive nanofillers (MWCNTs)

2. Obstacle → MWCNTs entangle

SEM image of pure MWCNTs showing entanglements.

3. Dispersion methods:

<table>
<thead>
<tr>
<th>Chemical</th>
<th>Mechanical</th>
</tr>
</thead>
<tbody>
<tr>
<td>Oxidation process by acid e.g. HNO₃ &amp; solution of H₂O₂/NH₄OH</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.¹

Multi-walled carbon nanotubes (MWCNTs)

- Dispersion of MWCNTs $\rightarrow$ Rastogi et al.\textsuperscript{1}, Geng et al.\textsuperscript{2} and Goswami et al.\textsuperscript{3}

1. 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.

2. 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.

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


Conductivity & permittivity

![Graph showing conductivity vs. frequency for different samples.]
Modulus

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

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

**Modulus loss factor**
- 0CNT Si3PEG_H25
- 1CNT Si3PEG_H25
- 2CNT Si3PEG_H25
- 3CNT Si3PEG_H25
- 4CNT Si3PEG_H25
- LR 3162

**Frequency (Hz)**
- $10^{-2}$
- $10^{-1}$
- $10^{0}$
- $10^{1}$
- $10^{2}$

**Modulus Loss Factor**
- $10^{-2}$
- $10^{-1}$
- $10^{0}$
- $10^{1}$
- $10^{2}$
Stress-strain plots

- Compliant electrodes
- PDMS-PEG
- MWCNTs
- Dielectric properties
- Rheology

**Stress** vs. **strain**

![Stress-strain plots](graph.png)

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

- Stress (MPa)
  - Y = 0.23 MPa
  - Y = 0.47 MPa
  - Y = 0.92 MPa
  - Y = 0.70 MPa
  - Y = 0.26 MPa
  - Y = 0.23 MPa

- Strain (%)

**Rheology**

**Dielectric properties**

**Compliant electrodes**

**PDMS-PEG**

**MWCNTs**

**DTU Chemical Engineering, Technical University of Denmark**

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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 $10^{-3}$ S/cm;
  - $< \text{LR3162} = 10^{-1}$ 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