

Preparation and Characterization of Functional Rubber Materials Based on Iontronic Pressure Sensing Mechanism: Postprint

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Abstract

To enable silicone materials to play a greater role in flexible sensors rather than merely serving as dielectric or substrate materials, polydimethylsiloxane (PDMS) was modified through effective methods—incorporating organic electrolyte and doping with silica—to impart iontronic pressure sensing characteristics, and the microstructure of the material was observed. The mechanical and electrical properties were investigated through comparisons of organic electrolyte content and surface roughness. The experiment successfully prepared an ionic rubber functional material with a mass ratio of PDMS:organic electrolyte:silica of 2:1:0.3, a corresponding Young's modulus of 1.1 MPa, and a pressure sensor sensitivity of 1.46 nF/(kPa · cm²).

Full Text

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Methods

To enable silicone materials to serve not merely as dielectric or substrate components in flexible sensors, but to impart iontronic pressure sensing capabilities through effective modification methods, thereby expanding their functional role in flexible sensing applications, we developed a strategy incorporating organic electrolyte into polydimethylsiloxane (PDMS). This approach yields iontronic pressure sensing characteristics while allowing systematic investigation of material microstructure, mechanical properties, and sensor sensitivity through controlled variation of organic electrolyte content.

Material Preparation and Processing

The ionic rubber functional material was prepared by incorporating organic electrolyte into PDMS with silica doping. The optimal composition ratio of PDMS:organic electrolyte:silica was determined to be 2:1:0.3, yielding a Young's modulus of [value] and sensor sensitivity of [value] nF/(kPa · cm). The preparation process involved dissolving lithium bis(trifluoromethanesulfonyl)imide (Li[TFSI]) in tributyl citrate (TBC) to form the organic electrolyte phase. This electrolyte was then blended with PDMS prepolymer and curing agent, followed by the gradual incorporation of silica particles under vacuum degassing to ensure uniform dispersion. The mixture was cast onto metal molds lined with sandpaper to create controlled surface microstructures, then thermally cured at [temperature] to form the functional ionic rubber.

Iontronic sensing operates through the formation of an electric double layer (EDL) at the ion-electrode interface, with capacitance changes resulting from variations in contact area under pressure. While liquid electrolytes inherently possess ionic characteristics suitable for EDL formation, flexible sensing applications require mechanically stable yet deformable materials. Therefore, elastomeric substrates such as polyurethane (PU), fluorinated rubber, and thermoplastic elastomers are commonly employed [11-20]. These substrates can be processed from commercial films or prepolymer kits, with the latter offering casting capabilities that simplify manufacturing. PDMS is particularly advantageous due to its excellent optical transparency, biocompatibility, and tunable mechanical properties. However, its inherently non-polar nature necessitates compatibility enhancement with polar ionic electrolytes. Silica doping effectively improves this compatibility while forming an ionic conductive network within the rubber matrix.

Surface Structuring

To enhance sensor sensitivity, controlled surface roughness was introduced by adhering sandpaper to metal mold surfaces prior to casting. This created microstructured surfaces with feature sizes determined by sandpaper grit (ranging from 000 to 7000 grit), providing tunable surface topography for optimized electromechanical performance.

Results and Discussion

Microstructural Characterization

Scanning electron microscopy (SEM) analysis revealed significant differences in cross-sectional morphology between pristine and silica-doped ionic rubber samples (Figure 2(a), (b)). In undoped samples, the organic electrolyte appeared as dispersed phase-separated domains. Conversely, silica-doped samples exhibited a continuous network structure with silica particles adsorbing the electrolyte, forming interconnected ionic conductive pathways throughout the PDMS matrix. This microstructure is critical for achieving consistent electromechanical response.

Surface topography analysis (Figure 2(c)) demonstrated that sandpaper-treated molds successfully replicated microscale roughness onto the ionic rubber surface. The resulting uneven, textured morphology substantially increases the effective contact area with electrodes under compression, thereby enhancing sensor sensitivity compared to smooth surfaces.

Mechanical Performance

Mechanical characterization focused on the relationship between organic electrolyte content and tensile properties. Samples were prepared with PDMS-to-electrolyte mass ratios of 1:2, 1:1, 2:1, and 4:1 at fixed silica content. Young's modulus values varied significantly across these compositions, enabling material selection based on application-specific requirements. Stress-strain measurements confirmed that all formulations maintained excellent stretchability and elastic recovery, with the ionic rubber demonstrating good extensibility even after fracture testing. The material's castability facilitates fabrication into diverse geometries, highlighting its potential for customizable soft device applications.

Electrical Performance

The ionic rubber was integrated with flexible printed circuit electrodes to form a capacitive pressure sensor configuration. Capacitance-pressure characterization revealed that increasing organic electrolyte content enhanced sensor capacitance at equivalent pressure levels. The 2:1 PDMS-to-electrolyte ratio exhibited optimal performance, achieving a sensitivity of [value] nF/(kPa · cm) within the pressure range of [range]. While higher electrolyte concentrations increased capacitance, they concurrently reduced mechanical robustness and stretchability. Conversely, lower electrolyte contents yielded insufficient sensitivity despite improved mechanical properties. The selected 2:1 formulation provided a balanced compromise, maintaining nanofarad-level sensitivity while preserving adequate mechanical performance and measurement repeatability.

Effect of Surface Roughness

Comparative analysis of surface roughness effects demonstrated that rough surfaces exhibit lower initial contact area and baseline capacitance, but undergo more dramatic contact area expansion under pressure stimuli. Sensors fabricated with 7000-grit treated surfaces showed superior sensitivity of [value] nF/(kPa · cm), while smoother surfaces (000-grit) exhibited progressively reduced sensitivity values below [value] nF/(kPa · cm). This confirms that increased surface roughness directly enhances pressure sensor sensitivity through amplified contact area modulation.

Conclusion

Based on iontronic pressure sensing principles, we successfully prepared functional ionic rubber by incorporating Li[TFSI]-based organic electrolyte and silica dopants into PDMS. Systematic investigation of electrolyte content effects on mechanical and electrical properties identified an optimal composition (PDMS:electrolyte:silica = 2:1:0.3) achieving a Young' s modulus of [value] and sensor sensitivity of [value] nF/(kPa · cm). The material' s inherent castability enables fabrication of complex geometries, providing a foundation for advanced electronic skin applications in subsequent research.

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