RSC Advances

PAPER

View Article Online
View Journal | View Issue

Cite this: *RSC Adv.*, 2015, **5**, 58428

Polar–nonpolar interconnected elastic networks with increased permittivity and high breakdown fields for dielectric elastomer transducers†

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Received 16th April 2015
Accepted 30th June 2015

DOI: 10.1039/c5ra06865g

www.rsc.org/advances

Elastic materials with increased permittivity (ε') were obtained in a three-step process starting from a hydroxyl end-functionalized polydimethylsiloxane (PDMS) of a high molecular weight (M_w = 139 kDa), trimethylsilyl end-blocked silicones that carry hydrosilane, cyanopropyl and hexyl groups $\mathbf{P}_x$ (where x represents the mol% of cyanopropyl groups), and tetraethoxysilane (TEOS). The hydrosilane groups of $\mathbf{P}_x$ were first hydrolyzed and the formed hydroxyl groups were subsequently reacted with partially hydrolyzed TEOS and further used as high ε' components, cross-linkers, and reinforcing agents for the PDMS matrix. A high wt% of the polar component $\mathbf{P}_x$ was incorporated into the nonpolar PDMS matrix by forming interconnected networks. Thermal (DSC, DMA) and morphological investigations (SEM) show the biphasic morphology of the networks. The dielectric, mechanical, and electromechanical properties of the films were investigated. Materials with good elastic properties, increased ε', and a high breakdown field (E_b) were obtained. The best material has an elastic modulus of 800 kPa at 10% strain, an $\varepsilon' = 4.5$, and a maximum actuation strain of 8% at $E_b = 56$ V μm^{-1}.

Introduction

Dielectric elastomer transducers (DET) are devices able to convert electrical energy to mechanical energy or *vice versa*.[1] DET are finding applications in robotics[2–6] artificial muscles[7,8] medical devices,[9–13] replacements of space engines[14,15] or in energy harvesting from different sources like wind,[16] human motion,[17,18] or waves.[19–21] As dielectrics, several elastomers such as silicones, polynitrile, polyacrylate, and natural rubbers as well as composites thereof have been used.[22,23]

The transducer performance is directly influenced by the stiffness and the permittivity (ε') of the dielectric. Large actuation strains are expected for materials that have a high ε' and a low elastic modulus. However, in terms of stress, a low elastic modulus (Y) value is not always of advantage, since the maximum stress attainable in an actuator increases for stiff materials.[21]

Silicones are characterized by a high flexibility of the Si–O bond,[24,25] which is responsible for the large achievable strains. Additionally, they are biocompatible, resistant to weathering, and have stable properties over a wide range of temperatures and frequencies.[26,27] Furthermore, silicones have good dielectric strength typically over 100 V μm^{-1} and maintain their specific energy over a wide frequency domain.[28] Despite of these great properties, silicones have a low ε' of less than 3.

The application potential of dielectric silicone materials would increase significantly if ε' could be increased while keeping the breakdown field strength and the elastic moduli at reasonable values. Attempts to increase the ε' of silicones include blending it with polarizable particles or with a polar component, chemical modification with polar side groups, or using a polar cross-linker.[29–32]

When polar cross-linkers are used, homogenous silicone elastomers form, but unfortunately the increase in the ε' is low due to the limited amount of polar groups that can be incorporated.[32]

A larger degree of functionalization of silicones can be achieved when the Si–O repeating units comprise a polar group.[33,34] Cyanopropyl-modified silicones were recently used as fillers in a silicone elastomer matrix[31] cross-linked by hydrosilylation. Composites with polar filler content up to 39 wt%, that reached ε' values as high as 7 were reported. However, the breakdown field (E_b) decreased significantly with increasing the ε'. Blending allows formation of materials with a polar phase physically dispersed into a nonpolar matrix. However, when the filler is

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† Electronic supplementary information (ESI) available: ^{29}Si and ^{1}H NMR spectra, FT-IR spectra, images of the films, SEM image of a blend with $\mathbf{P}_x$ particles, DSC curves, DMA, fatigue resistance tests, calculated actuation, mechanical and electromechanical properties of the optimized materials. See DOI: 10.1039/c5ra06865g