Synthetic Strategies for High Dielectric Constant Silicone Elastomers

Dielectric electroactive polymers (DEAPs) are a new and promising transducer technology and are often referred to as 'artificial muscles', due to their ability to undergo large deformations when stimulated by electric fields. DEAPs consist of a soft and thin elastomeric film (an elastomer) sandwiched between compliant electrodes and have many interesting properties such as a simple working principle, large achievable strains, high-energy densities and the fact that they are lightweight and low in cost. Some issues, however, limit the current commercial viability of the technology, as high driving voltages (several kV's), for instance, are necessary to activate the material. Driving voltage can be lowered by increasing the energy density of the DEAP elastomer film, achieved by creating elastomers with high dielectric constants, which is a material's ability to store electrical energy. Two synthetic strategies were developed in this Ph.D. thesis, in order to create silicone elastomers with high dielectric constants and thereby higher energy densities. The work focused on maintaining important properties such as dielectric loss, electrical breakdown strength and elastic modulus. The methodology therefore involved chemically grafting high dielectric constant chemical groups onto the elastomer network, as this would potentially provide a stable elastomer system upon continued activation of the material. The first synthetic strategy involved the synthesis of a new type of cross-linker for silicone polymer networks. The silicone compatible cross-linker allowed for copper-catalysed azide-alkyne cycloadditions (CuAAC) and thereby the attachment of functional groups to the network crosslinking point. The functional groups were very well-distributed in the silicone elastomer matrix, and various functional groups provided a number of elastomers with diverse properties. High dielectric constant molecules, such as the dipolar 4-nitrobenzene and nitroazobenzene, resulted in elastomers with an approximately 20% increase in dielectric constant at low concentrations of dipolar species (~0.5 wt%). The second synthetic strategy was to create elastomers with high concentrations of functional groups and thereby even higher dielectric constants through the synthesis of novel copolymers. Two different routes were followed to accomplish this aim. One route involved the synthesis of a so-called 'chain extender' that allowed for chemical modifications such as Cu- AAC. This route was promising for one-pot elastomer preparation and as a high dielectric constant additive to commercial silicone systems. The second approach used the borane-catalysed Piers-Rubinsztajn reaction to form spatially well-distributed copolymers where functional groups could be attached along the polymer backbone. The functional copolymers contained vinyl or allyl end groups that allowed for elastomer synthesis. The dielectric properties of the formed elastomers were found to increase significantly and an optimum concentration of functional groups was identified. At a concentration of 5.6 wt% of a nitrobenzene functional group the dielectric permittivity increased 70% while at this loading important properties such as electrical breakdown strength, elastic modulus and dielectric loss were not significantly compromised. The developed synthetic strategies facilitate new ways of functionalising elastomers in general and dielectric elastomers in particular.

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