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## ACCEPTED MANUSCRIPT

#### A combination of silicone dielectric elastomer and graphene nanosheets for the preparation of bending flexible transducers

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**Abstract:** A newly developed dielectric elastomer (DE) bending transducer composed of a functionalized graphene-silicone DE composite sandwiched between two graphene flexible electrode layers had been investigated. This bending transducer displayed high actuation displacement (5.3 mm) under 2500 V direct-current electric field. Compared with pure silicone DE, the silicone DE filled with silane coupling agent functionalized graphene nanofillers showed enhanced dielectric constant, and low loss modulus. The silane coupling agent functionalization method is simple, green, and easy to control, and can be used as a general strategy for improving the dielectric and electric actuated strain of DE.

Keywords: Polymeric composites; Smart materials; Graphene; Electrical properties; Sensors and actuators.

#### 1. Introduction

As their fantastic characteristics of light, soft, and achievable strain and stress similar to natural muscles, electro-active polymers, such as ionic polymer-metal composite [1], ferroelectric polymer [2], conducting polymer [3], carbon nanotube [4], gaphene [5, 6], and dielectric elastomer (DE) [7-10], have attracted wide attention for lots of biomimetic applications including bionic flying insects or robots, haptics for portable consumer devices, and dynamic sensors. Among the different electro-active polymers, DE has been intensively studied in past decades and emerged as promising flexible transducer material candidates. DE transducers consist of thin electrically insulating elastomeric membranes coated on both sides with compliant electrodes. Recently, Verdejo et al. summarized the methods of increasing the performance of DE transducers from the materials perspective [11]. The approaches to enhance the actuation performance of DE transducers are by: (1) reducing the DE film thickness, (2) reducing the DE mechanical stiffness, (3) increasing the DE dielectric constant, and/or, (4) developing highly compliant electrodes.

In previous studies as to increasing the dielectric constant of DE, commonly adopted methods were modifying DE by adding high dielectric constant ceramic fillers [12-13], or conductive fillers [14-16]. Both two kinds of fillers have advantages and shortages. High dielectric constant ceramic particles are dielectric in nature and will not induce high dielectric loss, but the large volumetric concentration of ceramic fillers will increase the mechanical stiffness and damage the flexibility of DE. Although only by using a very little mount amout of conductive fillers can dramatically increase the dielectric constant of the DE matrix, the DE composite will suffer from high dielectric loss, resulting in limited applications. Very recently, graphene nanosheets have shown superior physical characteristics and have been considered to be an ideal filler candidate [15]. Graphene has a two dimension layered structure with a large aspect ratio, and thus it is easier to form a large number of parallel micro-capacitors within DE matrix. However, the incorporation of graphene nanosheets into a polymer matrix can cause several problems, such as the aggregation or restacking of

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