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Electromechanical response of muscle-like actuators enhanced by carbon fillers

Alexandra Shakun, Minna Poikelispää, Amit Das, and Jyrki Vuorinen

The actuation behavior of an acrylic dielectric elastomer is improved by the addition of different carbon fillers, such as carbon blacks, carbon nanotubes, graphite, and nanodiamonds.

Actuators that are inspired by the operation of biological muscle (i.e., muscle-like actuators) are useful for a variety of applications, including braille displays, thin flexible loudspeakers, microvalves, and robotics. Among the materials suitable for use in muscle-like actuators, electroactive dielectric elastomers exhibit a number of desirable properties (e.g., a low elastic modulus, high recoverable deformation, and electric polarizability).The working principle of such dielectric elastomer actuators (DEAs) is illustrated in Figure 1. Although DEAs exhibit a number of attractive properties, including a low elastic modulus and low dielectric losses, some DEAs require very high fields for their operation. The use of DEAs in real applications is therefore extremely challenging, or even impossible.[1]

One approach to achieving actuation at lower electric-field values is to increase the relative dielectric permittivity of the elastomer. In turn, this improves the polarizability of the material in the applied field. This can be realized by, for example, adding electrically conductive fillers.[2] Such materials often have high dielectric losses, however, which reduces the usability of the actuator.[3] Moreover, because previously reported experiments were not standardized, comparing the effect of different fillers on the electromechanical properties of DEAs is very complicated.

In our work, we therefore focused on obtaining direct comparisons between a number of carbon-based fillers with different electrical conductivities, including two that have not previously been studied for application in DEAs (i.e., nanodiamonds, NDs, and graphite). To this end, we have investigated the actuation performance of alkyl acrylate copolymer (ACM)—an acrylic polymer that is the most popular material for use in DEAs—filled with different carbon-based fillers.[4] The fillers we used included a number of types of carbon black (CB: N-234, N-375, and N-550 grades), carbon nanotubes (CNTs), graphite, and detonation-produced NDs. To prepare the ACM composites, we mixed each filler with ACM and other compounding ingredients (i.e., sodium stearate, stearic acid, sulfur, and dioctyl adipate) in a laboratory-scale Brabender W50 mixer. After compounding, we formed rubber sheets, with thicknesses of 0.5 and 1mm, in a press and vulcanized them for 15 minutes at 175°C. We then studied the dielectric properties and measured the elastic modulus of the resultant ACM composites.

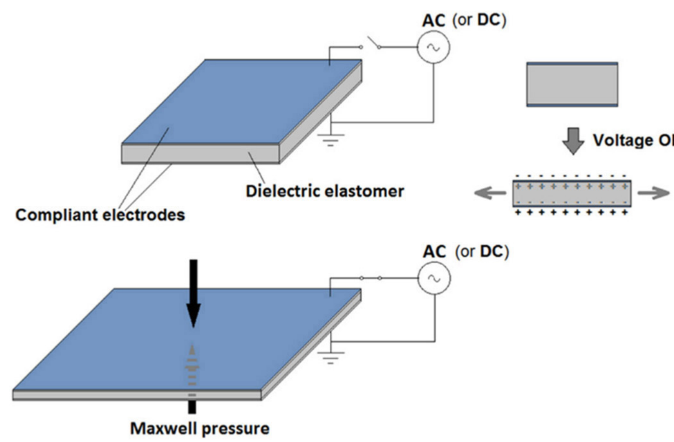

Figure 1. *Illustration of the working principle of a dielectric elastomer actuator (DEA), i.e., in which dielectric elastomers are designed to act as muscle-like actuators. When a voltage is applied to the DEA, it becomes polarized and charge builds up on the electrodes. The attraction between these opposite charges creates pressure (the Maxwell pressure) across the thickness of the elastomer, which leads to an expansion of the actuator area.[8] When the voltage is turned off, the material returns to its original dimensions. AC: Alternating current. DC: Direct current.*

Our results show that, with the exception of NDs, the addition of the fillers increased the relative permittivity of our ACM-based composites. This increase was more pronounced for fillers with high

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electrical conductivities and small particle sizes and, thus, a higher filler volume fraction. For example, incorporation of 20phr (parts per hundred rubber) of N-234 CB increased the relative permittivity of ACM by up to 1490 times at 1Hz. Such an increase in dielectric permittivity, however, is often associated with high dielectric losses because of increased electrical conductivity. Composites with higher filler loadings may therefore be unsuitable for use in DEAs and it is essential to keep the filler content below the percolation threshold. We also found that the addition of surface-carboxylated NDs gave rise to a marginal decrease in the dielectric permittivity of ACM and had no effect on dielectric losses. In contrast, NDs with a hydrogenated surface had no deteriorating effect on relative permittivity, but reduced dielectric losses of the material twofold at 1Hz. The differences in these results are mainly caused by the strongly dielectric nature of NDs and differences in the surface chemistry.

The low elastic modulus of our ACM rubber composites facilitates the movement of the polymer during actuation. Although CB and CNTs are known to typically reinforce polymers by increasing the elastic modulus, NDs have little reinforcing effect on elastomeric materials.[5] Nevertheless, in our experiments the addition of the CB, CNT, and ND fillers at 1phr led to a small increase in the elastic moduli of the materials (because of the hydrodynamic effect). We observed no such increase, however, in the composites that contained graphite fillers.

In our actuation tests, we found that all CB types improved the electromechanical response of the ACM composites when they were subjected to relatively low electric fields (see Figure 2). In addition, our results for the relative permittivity of the fillers showed that N-234 had the highest actuation stress, yet the composite containing 20phr N-234 failed at about 3.5kV/mm. We believe this was caused by the formation of a conductive path within the material (leading to a short circuit). The 1phr CNT-filled ACM composite showed promising results. For instance, it exhibited a threefold increase in stress at 5kV/mm compared with pure ACM. The addition of NDs and graphite (at filler loadings of up to 5phr), however, did not cause any significant improvements in actuation behavior. Nevertheless, we achieved better actuation with the composites that were filled with hydrogenated NDs than those filled with graphite at the same weight. This may be related to the positive effect of reduced dielectric losses in the lower-frequency area. The range of applications for ND powders may thus be extended to include dielectric elastomer composites for use in electromechanical transducers.

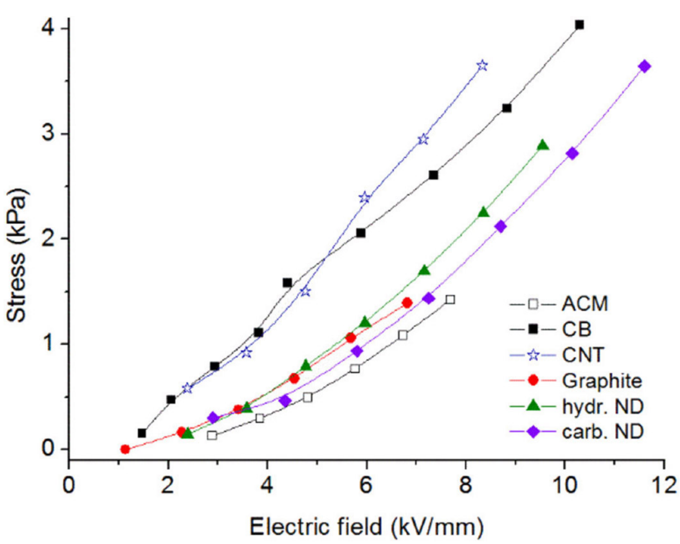

Figure 2. *Actuation performance of alkyl acrylate copolymer (ACM) containing different carbon-based fillers (at 1 part per hundred rubber). CB: Carbon black. CNT: Carbon nanotube. hydr. ND: Surface-hydrogenated nanodiamonds. carb. ND: Surface-carboxylated NDs.*

In summary, we have found that the incorporation of carbon-based fillers into an ACM compound improves the electrical actuation of the material. We also found that a lower electric field is required to achieve the same actuation pressure in these composites as in unfilled acrylic rubbers. Given that our addition of NDs and graphite caused an increased actuation stress, we plan to use a combination of ND and graphitic structures (i.e., 'diamond soot,' an original product obtained by a detonation process) in our future work. We expect this mixture of carbon allotropes to exhibit a synergetic effect[5] similar to that which arises when other filler combinations are used (e.g., carbon black and barium titanate[6,7]).

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Author Information

Alexandra Shakun, Minna Poikelispää, and Jyrki Vuorinen
Laboratory of Materials Science
Tampere University of Technology
Tampere, Finland

Alexandra Shakun is currently conducting her PhD research on the development of a latex-based elastomer nanocomposite for use in energy-harvesting applications. Her research interests include rubber technology, elastomer nanocomposites, dielectric elastomer transducers, nanodiamonds, and energy harvesting.

Minna Poikelispää is a PhD student. Her research areas include rubber technology, elastomer nanocomposites, and smart rubber applications.

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Jyrki Vuorinen is the Dean of the Faculty of Natural Sciences. He has worked as a professor of composite materials in the Laboratory of Materials Science since 2004, and was a professor of plastic technology between 2008 and 2016.

Amit Das
Leibniz Institute of Polymer Research
Dresden, Germany

Amit Das is a senior scientist at the Leibniz Institute of Polymer Research and an adjunct professor in the Laboratory of Materials Science, Tampere University of Technology. His research interests include self-healing and mechanoadaptive rubbers, artificial muscles, and the investigation of rubber chemistry.

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