

Improvement of actuation performance of dielectric elastomers by barium titanate and carbon black fillers

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ABSTRACT: Dielectric elastomers are promising materials for actuators resembling human muscle. Among elastomers, acrylic rubbers (ACM) have shown good actuation performance but its use is limited by the high operating voltages required. The present work demonstrates that simultaneous incorporation of nanostructured carbon black and dielectric fillers offers an increase in a dielectric permittivity and a suitable modulus of the elastomers matrix, enabling an improved electro-mechanical actuation performance at low voltages. By the use of reinforcing carbon black and barium titanate in an acrylic elastomer matrix a sixfold increase in the dielectric permittivity was realized. A fine tuning of the actuation stress and, consequently, actuation strain can be done by a judicious selection of the different filler concentrations in the soft rubber matrix. Finally, a synergistic effect of the fillers was observed in the improved actuation performance of the developed materials. This work may pave the way to design dielectric elastomers for actuator fabrication. © 2016 Wiley Periodicals, Inc. *J. Appl. Polym. Sci.* **2016**, *133*, 44116.

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INTRODUCTION

Field-activated dielectric elastomers (DE) are known as a promising class of materials for an actuator application. DEs are insulators that become polarized in applied electric field and then subjected to an active electrostatic pressure between electrodes.^{1,2} Compared with the other materials used to obtain muscle-like action, dielectric elastomers most resemble the natural muscles in strain, efficiency and response speed. Among other electroactive polymers (EAPs), dielectric elastomers offer good overall performance, high strains and decent cost.³ However, for their operation most EAPs, including dielectric elastomeric actuators (DEAs), require very high electric field that can reach 100 kV/mm,^{2,4} thus limiting their applications.^{5,6} Despite of the decades of productive research in the field of DEAs, no effective solution involving unmodified materials traditional for the rubber technology has been found to reduce working voltage of such devices.

Among currently applicable material modification approaches, addition of ceramic particles with high dielectric constant, conductive fillers and their combinations into DE matrix has attracted much attention from the researchers.⁷ Such modification should yield dielectric composites with improved electro-mechanical response that can be expressed by Maxwell pressure

(σ) generated between the compliant electrodes of an actuator due to the action of electric field (E):

$$\sigma = \epsilon' \cdot \epsilon_0 \cdot E^2, \quad (1)$$

where ϵ' is dielectric constant of the material and ϵ_0 is dielectric permittivity of vacuum. Although Maxwell pressure theory is valid only for ideal dielectric elastomers,⁸ it can still be applied to elastomers at low crosslink densities and/or at small deformations.^{9,10} Moreover, compressional strain (s_z) equation can give an estimation of electromechanical response of the actuator based on the Maxwell pressure and Young's modulus (Y) of the dielectric material:

$$s_z = -\frac{\sigma}{Y} = -\frac{\epsilon' \cdot \epsilon_0 \cdot E^2}{Y}. \quad (2)$$

Thus, increasing the dielectric constant while retaining control on the elastic modulus of a material is one of the possible methods to lower the operating voltage,^{1,7} and such approach is applied in the current work.

Barium titanate (BaTiO₃, BT) is widely used among high dielectric constant fillers in dielectric composite material with improved actuation properties.^{1,2,11,12} However, BT concentration needs to be around 15 vol %¹ in order to see improvements in relative dielectric permittivity. This increases the elastic modulus of rubber and thus the advantages obtained

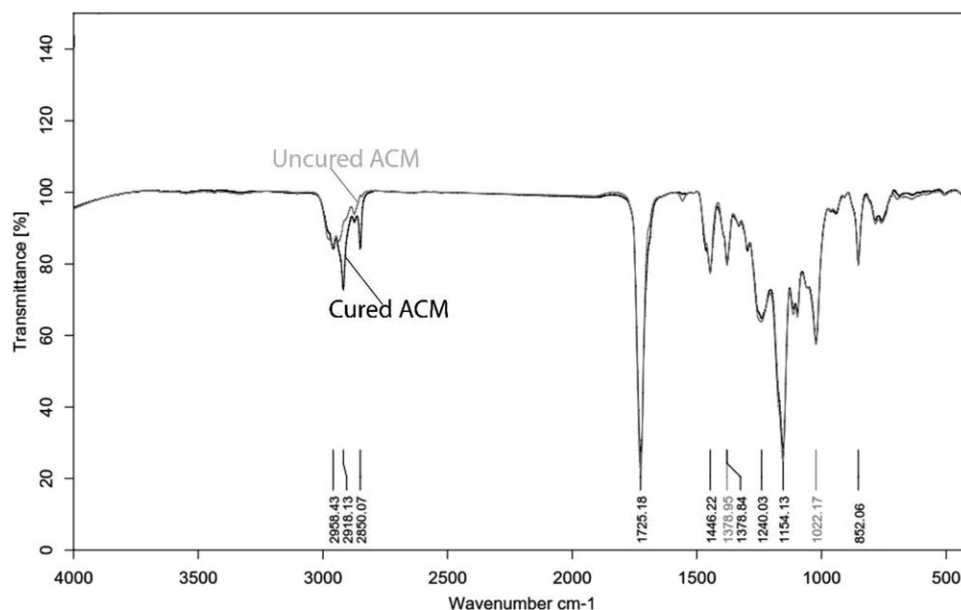


Figure 1. FT-IR Spectra of neat ACM and cured ACM.

with higher dielectric permittivity are partly lost by increased stiffness of the elastomers. At the same time, conductive fillers at concentrations below the percolation threshold are known to increase dielectric permittivity of polymeric materials significantly at relatively low filler concentrations.^{13–15} Such effect can be related to the Maxwell–Wagner polarization arising at the interface between dielectric matrix and conductive filler particle at low frequencies. The idea of combining different types of fillers is not new in rubber technology,¹⁶ and specifically in the field of DEAs.^{17,18} However, addition of conductive filler particles often leads to an increase in dielectric losses of the material and current leakage, or does not result in the expected improvement of the relative dielectric permittivity due to poor distribution and other reasons.^{11,12,18–20} Although several studies have proved that these problems can be successfully overcome,^{18,20} this does not solve the problem of the high working electric field and make such actuators quite costly thus preventing their use in large-scale industrial applications.

The role of dielectric matrix is very important in providing proper insulation and reducing filler-induced high dielectric losses.²¹ Acrylic rubber (ACM) is a well-studied polymer with Young's modulus of 1–2 MPa and elongation at break up to 600%²² that already showed good actuation performance. Dielectric properties of ACM, like of any other rubber, depend on the chemical structure and its polarity, exact composition of the compound, presence of plasticizers and softeners, and many other aspects. Nevertheless, most of the experiments have been conducted with commercial adhesive tape VHB 4910 by 3M company, and just few studies involved preparation of polyacrylate composition and its vulcanization. Being most studied and reported in multiple articles, acrylic VHB 4910 tape has dielectric permittivity of $\epsilon' = 4.2 - 4.8$ at 1 kHz,^{2,23,24} although some researchers report higher value $\epsilon' = 7$.²⁵

The main goals of the current research is to utilize readily available, affordable, and well-known materials in order to obtain

high-performance material capable of significant deformations in response to an applied moderate electric field. The combination of conductive carbon black (CB) conventionally used in rubber technology and ferroelectric BT is promising from this perspective. At certain concentrations conductive particles lead to significant increase in dielectric permittivity and ceramic particles interrupt the possible percolation paths thus reducing leakage current. Moreover, combination of non-reinforcing barium titanate with reinforcing carbon black results in comparably low elastic modulus.

EXPERIMENTAL

Materials

Acrylic rubber consisting of acrylic esters and a small amount of chlorine containing cure-site monomer (HyTemp AR 715) used in the study was produced by Zeon Chemicals L.P. Curing system included sodium stearate (NaSt, 3 phr, parts per hundred rubber), sulfur (S, 0.3 phr), and stearic acid (St.acid, 0.5 phr). CB type N-234 (single particle size about 19 nm) from Evonik was used as a conductive filler and BT (particle size <2 μm) was delivered by Sigma-Aldrich. Dioctyl adipate (DOA) was used as plasticizer (2 phr) to decrease elastic modulus and improve dispersion and distribution of filler particles.

Soap/sulfur curing system was chosen for the chlorine cure-site ACM, as being more recommended in the literature, with the metal soap/sulfur combination in 10:1 ratio widely used. Soap has a function of curing agent, while elemental sulfur is an activator.^{26,27} Polysulphide vulcanization bonds are formed.²⁸ Moreover, it is mentioned that sodium stearate serves as acceptor of the released chlorine. As the curing systems used with ACM is basic in nature, stearic acid, used in the present formulation, is a retarder, and also a processing aid.^{26,29} Fourier transform infrared (FT-IR) spectra of the uncured and cured ACM measured by Bruker Optics Tensor 27 (Diamond ATR (Attenuated total reflection)) are presented in Figure 1.

Table I. Mixing Order for the Rubber Compounds

| Sample | Ingredient addition time (min) | | | | | | | Total mixing time (min) |
|------------------------|--------------------------------|-----|------|---------|-----|----|----|-------------------------|
| | ACM | S | NaSt | St.acid | DOA | CB | BT | |
| ACM | 0 | 1.5 | 1.5 | 1.5 | 1 | — | — | 3.5 |
| BT0 | 0 | 1.5 | 1.5 | 1.5 | — | — | — | 3.5 |
| BT5/10/20/30/50/70/140 | 0 | 3 | 3 | 3 | — | — | 1 | 5 |
| CB5/10/15/20 | 0 | 4 | 4 | 4 | 2 | 1 | — | 6 |
| CB20-BT2/5/10 | 0 | 4 | 4 | 4 | 2 | 1 | — | 6 |
| CB5/10/15-BT15/10/5 | 0 | 4 | 4 | 4 | 2 | 1 | — | 6 |

VHB 4910 acrylic tape produced by 3M company is the often used material for DEAs and, therefore, the actuation performance of the studied materials is compared with it.

Preparation of the Samples

ACM and ingredients were compounded in a laboratory scale mixer (Brabender® W 50 driven by Brabender Plasti-Corder®) with tangential rotors. The rotor speed of the mixer was 60 rpm and the starting temperature was 20 °C. The mixing order of the compounds is presented in Table I. After the first mixing step presented in Table I, the compound was taken out from the mixer, feeded back, and mixed two more minutes to guarantee a good dispersion of fillers. Mixed compounds were formed to 0.5 and 1 mm sheets and cured at 175 °C for 15 minutes. The compounds were named according to the filler type abbreviation and amounts of those in phr.

Characterization

Tensile tests for determining moduli of the samples were carried out with a Messphysik Midi 10–20 universal tester using dumb-bell test sample type three from ISO 37. The test rate was 200 mm/min and three parallel measurements were conducted and the average value was calculated. Dielectric permittivity, dielectric loss and electrical conductivity were measured with a Novocontrol Alpha-A at the frequencies from 1 MHz to 0.75 Hz at ambient conditions. Samples were placed tightly between 2 mm thick rigid gold-plated electrodes 20 mm in diameter to ensure good electrical conductivity and avoid any possible actuation. Thickness of each specimen was measured with an outside micrometer after the material was placed into the sample holder. Five specimens of each sample were tested, and the average value was calculated.

Actuation measurements were carried out in equipment prepared in-house. The sample actuator was prepared by cutting 20 × 50 mm piece of 0.5 mm thick elastomer film. Two to three samples per material was tested. Electrodes made by carbon black (N-234)-toluene mixture (ratio 1:10) were spread on both sides of elastomer and then toluene was fully evaporated by drying the sample at room temperature for 24 hours. DC voltage from 0 to 5 kV was applied to the electrode in 0.5 kV steps via conducting copper plates from high voltage amplifier (TREK model 10/10B-HS). During the measurements, changes in force, when voltage was applied, were measured with a 10N

load cell (LTS-1KA, Kyowa). The measurement was controlled via LabVIEW 2012.

RESULTS AND DISCUSSION

BT is known to have high relative permittivity. The relative permittivity of BT can vary from 1000 to 2500,¹¹ thus it is expected that the addition of BT increases relative permittivity of rubbers. However, it was reported that high amounts of BT is required to get improvements in permittivity values of rubbers.³⁰ Figure 2(a) shows the relative permittivity of ACM at various BT concentrations in the frequency range from 1 MHz

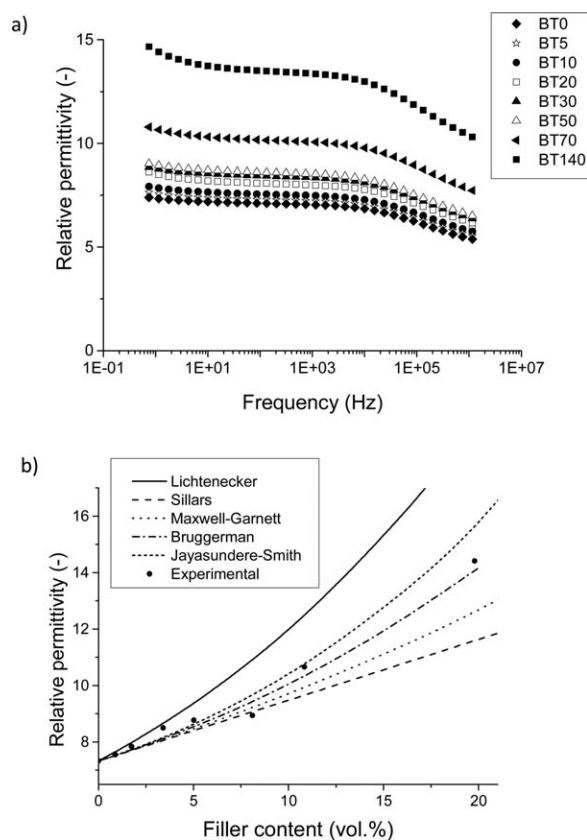


Figure 2. (a) The relative permittivity of the ACM-BT compounds; (b) Prediction of relative permittivity at 1 Hz frequency depending on the BT content by classical dielectric mixing rules.

to 1 Hz. It can be seen that 30 phr BT is required to get 1 unit increase in relative permittivity of ACM. A twofold increase of the permittivity is observed when the BT concentration is 140 phr (20 vol %).

To predict relative permittivity of composites as a function of volume fraction of spherical particles as the fillers in the polymer matrix, several theories, called as classic dielectric mixing rules, have been developed. Classical mixing rules consider isotropic matrix with permittivity ϵ_m and volume fraction v_m filled with particles of permittivity ϵ_f and volume fraction $v_f = 1 - v_m$. It is assumed that both components have no dielectric losses in the described frequency region. In general, the permittivity of the obtained composite lies between extremes $\epsilon_{c,\min}$ and $\epsilon_{c,\max}$ that are obtained from equivalent series connection and parallel circuit, respectively:

$$\epsilon_{c,\min} = \frac{\epsilon_m \epsilon_f}{\epsilon_m v_f + \epsilon_f v_m} \quad (3)$$

$$\epsilon_{c,\max} = \epsilon_m v_m + \epsilon_f v_f \quad (4)$$

Intermediate form was proposed by Lichtenecker in logarithmic form:

$$\ln \epsilon_c = v_m \ln \epsilon_m + v_f \ln \epsilon_f \quad (5)$$

Sillars mixing rule is based on the consideration that at filler volume fraction lower than 0.1, distribution of filler in dielectric matrix gives rise to the same electric potential that the hypothetical larger spheres with dielectric constant ϵ_c :

$$\epsilon_c = \epsilon_m \left[1 + \frac{3v_f(\epsilon_f - \epsilon_m)}{2\epsilon_m + \epsilon_f} \right] \quad (6)$$

More accurate Maxwell–Garnett equation is applicable for filler volume fraction lower than 0.1 with no restrictions on electrical resistivity of the components:

$$\epsilon_c = \epsilon_m \left[1 + \frac{3v_f(\epsilon_f - \epsilon_m)}{(1 - v_f)(\epsilon_f - \epsilon_m) + 3\epsilon_m} \right] \quad (7)$$

Bruggeman's equation is valid up to $v_f = 0.5$ or for disordered systems on the stipulation that the dispersed particles do not form percolative paths:

$$\frac{\epsilon_f - \epsilon_c}{\epsilon_c^{1/3}} = \frac{(1 - v_f)(\epsilon_f - \epsilon_m)}{\epsilon_m^{1/3}} \quad (8)$$

The formula provided by Jayasundere and Smith takes into consideration the polarization of the neighboring filler particles:

$$\epsilon_c = \frac{\epsilon_m v_m + \epsilon_f v_f \frac{3\epsilon_m}{(2\epsilon_m + \epsilon_f)} \left[1 + 3v_f \frac{\epsilon_f - \epsilon_m}{2\epsilon_m + \epsilon_f} \right]}{v_m + v_f \frac{3\epsilon_m}{(2\epsilon_m + \epsilon_f)} \left[1 + 3v_f \frac{\epsilon_f - \epsilon_m}{2\epsilon_m + \epsilon_f} \right]} \quad (9)$$

Lichtenecker [eq. (5)], Sillars [eq. (6)], and Maxwell–Garnett [eq. (7)] equations are valid for filler volume fraction lower than 0.1 and at higher filler loads such models tend to underestimate the effective permittivity. At small filler loads there is no significant difference between the models.^{7,31} The measured permittivity values of the ACM-BT composites were compared with the classic mixing rules [Figure 2(b)] and it was found that at low filler content Lichtenecker model describes the results best, while at

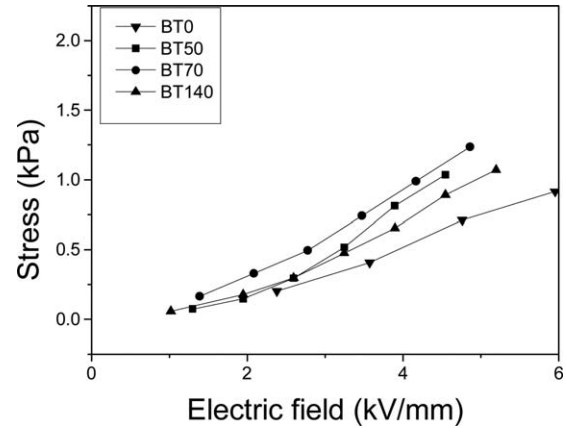


Figure 3. Actuation performance of the ACM-BT compounds.

high filler load the results are closer to Bruggeman [eq. (8)] and Jayasundere–Smith [eq. (9)] formulas.

The main drawback of the incorporation of a substantial amount of dielectric filler is that such a high amount of fillers makes material stiffer due to the hydrodynamic effect and increases Young's modulus that plays very important role in actuation performance [in eq. (2)] and thus should be maintained at the low level. The actuation results of the acrylic rubber-barium titanate composites (ACM-BT) are presented in Figure 3, where actuation stress is plotted against applied electric field. It can be seen that actuation improves when the amount of BT increases up to 70 phr. The compound containing 140 phr BT has clearly poorer actuation due to increased Young's modulus.

Electrically conductive fillers can increase relative permittivity of rubbers at generally lower concentrations, particularly, below the percolation threshold. Elastomeric polymers are known to be insulators, but incorporation of electrically conductive fillers can make rubber electrically conductive. Thus, for dielectric elastomer actuators, the content of electrically conductive fillers may not exceed percolation threshold. Therefore, it is important to find out the maximum filler loading that can be used in DEAs. To be dielectric, electrical conductivity of the material may not be higher than approximately 10^{-8} S/cm.³² The electrical conductivity of CB-filled ACM is presented in Figure 4(a). As expected, the incorporation of CB into ACM increases electrical conductivity of rubber significantly. After addition of 20 phr CB the conductivity is approximately 10^{-9} S/cm at 1 Hz whereas, the rubber without any filler has electrical conductivity approximately 10^{-12} S/cm. However, the ACM-CB 20 phr compound shows still frequency dependent behavior. Generally, the composite is frequency independent at low frequencies if the filler content is close to or above percolation threshold as the current is able to flow through the filler network while at higher frequencies the prevailing factor is the capacitor admittance of the insulating matrix.³³

For dielectric actuators it is beneficial that the conducting network of the CB is broken by the increment of the nanoscopic gap in between two neighboring conducting CB particles. In the current study, non-conductive BT is added to interrupt

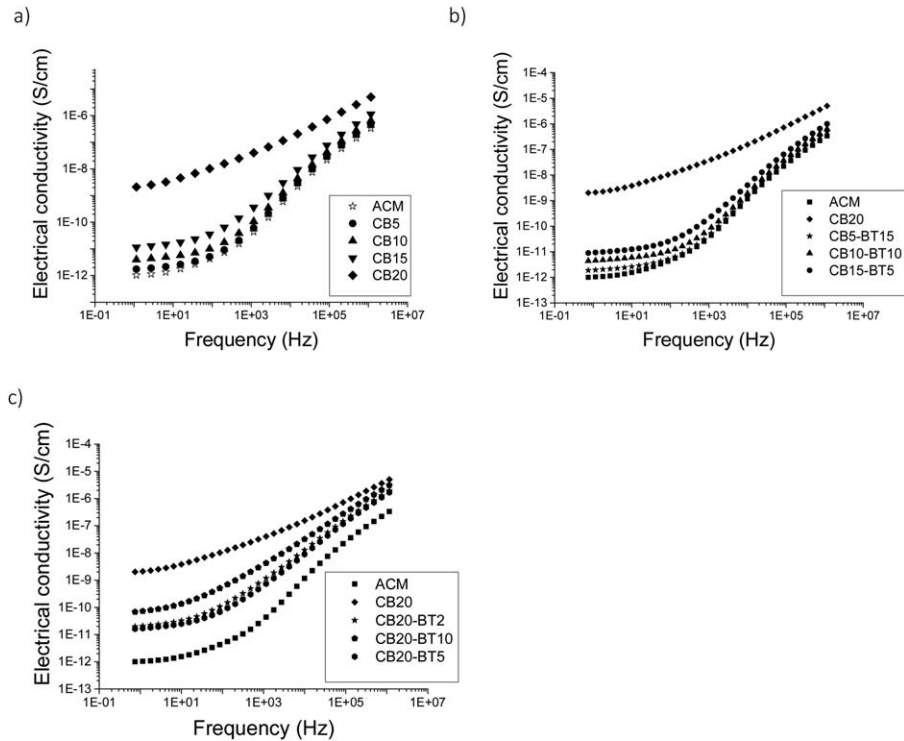


Figure 4. Electrical conductivities of (a) ACM-CB compounds; and (b) ACM-CB-BT compounds containing 20 phr of fillers; (c) the ACM compounds with 20 phr CB and different amounts of BT.

formation of CB network and thus avoid percolation. The electrical conductivities of the ACM-CB compounds are compared with the electrical conductivities of the ACM-CB-BT

compounds with total filler amount of 20 phr in Figure 4(b,c), respectively. It was found that higher amount of CB leads to a higher conductivity of the composite. When the CB content is

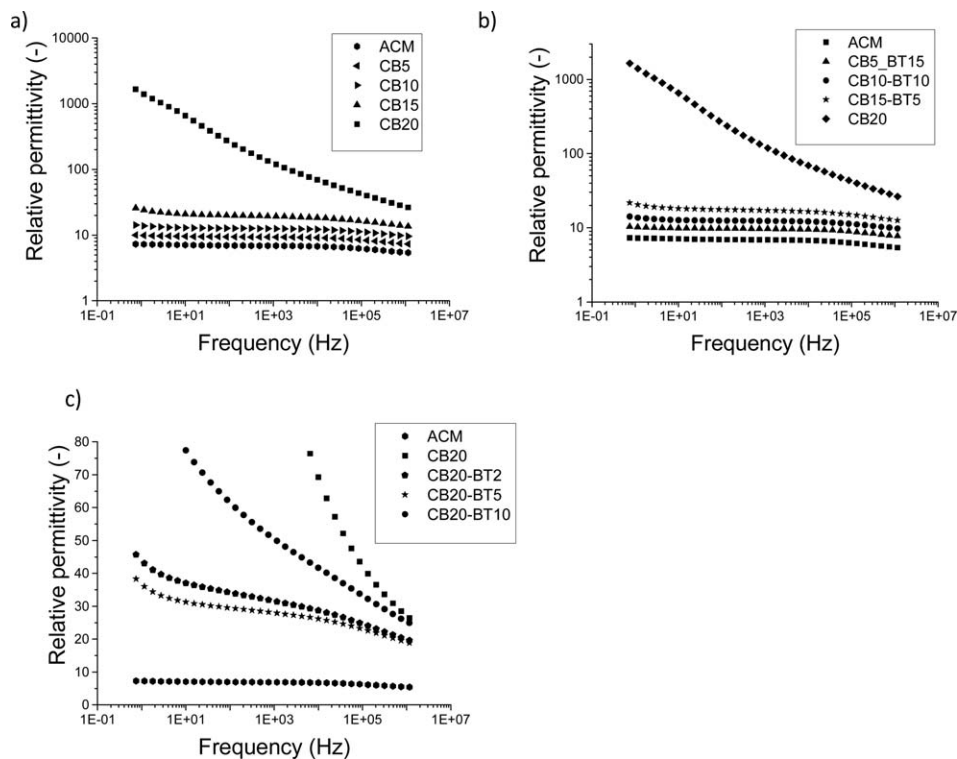


Figure 5. Relative dielectric permittivities of (a) ACM-CB and (b) ACM-CB-BT compounds; (c) The effect of BT concentration on the relative permittivity of the ACM-CB-BT compounds.

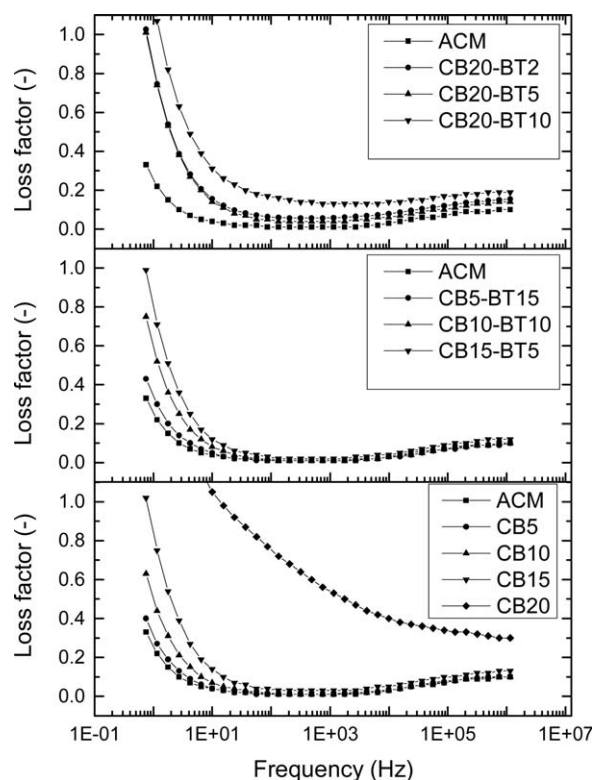


Figure 6. Dielectric loss factors of ACM-CB and ACM-CB-BT compounds and the effect of BT concentration on the dielectric loss factor of the ACM-CB-BT compounds.

maintained at constant level at 20 phr and very small amount BT particles (2–5 phr) are added into the compounds, it is observed that the conductivity of the composite is decreased at lower frequency region. However, after further addition of BT particles the conductivity is increased again. Probably, at higher concentration of BT the CB particles are forming more compact network thus enhancing the electrical conductivity of the composite.³⁴

It would be now interesting to discuss about the relative dielectric permittivity of the composites. It can be seen that after addition of small amount of CB the permittivity of the composites increases in the whole frequency region [Figure 5(a)]. At 20 phr filler loading the sample shows a strong frequency dependent behavior, particularly at lower frequencies. When CB content is close to the percolation threshold, the very strong Maxwell–Wagner polarization can be observed and dielectric permittivity of the composite increases tremendously. Maxwell–Wagner, or interfacial, polarization originates from the charge built up and accumulation at the filler–matrix interface due to the significant difference in their conductivities. The accumulated charge can move through the material in electric field thus leading to the increase in electrical conductivity discussed previously. Therefore, the higher the CB concentration, the stronger is the polarization and thus higher the relative permittivity, electrical conductivity and dielectric loss.

When CB is partially replaced by BT, some changes in dielectric permittivity values can be observed in Figure 5(b). Basically, the

relative permittivity values follow the values of the compounds containing only CB as filler. At low CB contents, the added BT has only minor effect on relative permittivity of the elastomer composite containing the same amount CB. However, when the concentrations of CB and BT are 15 and 5 phr, respectively, a decrease in relative permittivity is observed due to reduced Maxwell–Wagner polarization. The value of dielectric permittivity of the composite containing only 15 phr CB is approximately 25 whereas the composite containing additional 5 phr BT has relative dielectric permittivity approximately 20.

The addition of different amounts of BT into the ACM containing 20 phr CB decreases relative dielectric permittivity remarkably [Figure 5(c)]. However, the relative permittivity is still much higher than that of pure ACM and thus the CB-BT filler combination improves actuation capability of the material. It is interesting to note that the addition of 2 and 5 phr BT decreases the relative permittivity more than the addition of 10 phr of BT, following the same trend as described for electrical conductivity of the studied composites.

Typical drawback of electrically conductive fillers is high dielectric losses (ϵ'') and consequently high loss (dissipation) factor that describes the amount of energy is lost, for example, transferred into heat build-up, compared with the energy stored in cycle.³⁵ Dielectric loss factors of ACM-CB compounds as a function of frequency are presented in Figure 6. It is evident from the figure that ACM with 20 phr of CB has very high dielectric losses especially at low frequencies due to the strong

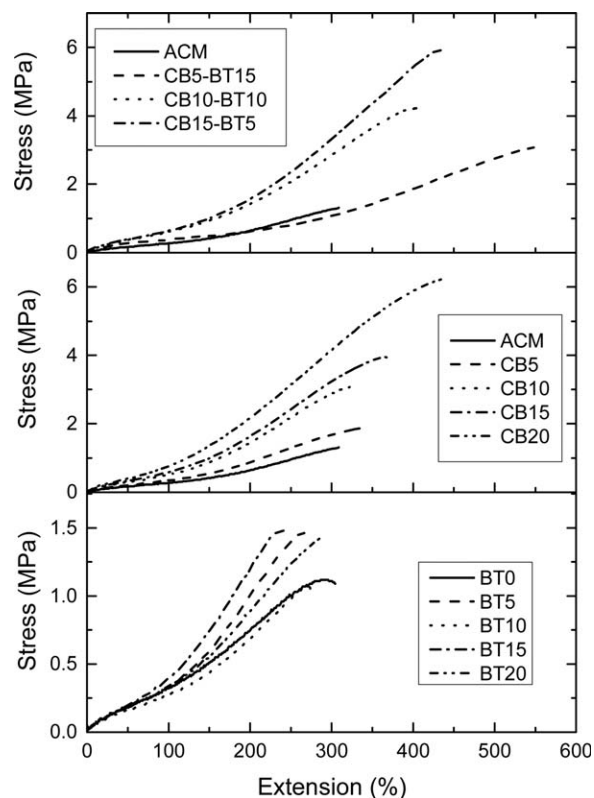


Figure 7. Comparison of tensile stress-strain curves of CB-BT-filled, CB-filled, and BT-filled compounds.

Table II. Young's Modulus and Calculated Electrical Breakdown Strength of the Compounds

| | Young's modulus Y (Mpa) | Permittivity ^a /modulus ϵ'/Y | Calculated breakdown strength E_b (V/mm) | Breakdown during the test ^b |
|---------------|----------------------------|---|---|---|
| ACM | 0.87 | 8.4 | 49,200 | Pass |
| ACM-BT5 | 0.86 | 8.6 | 48,600 | Pass |
| ACM-BT10 | 0.79 | 9.9 | 45,400 | Pass |
| ACM-BT30 | 0.84 | 10.2 | 44,600 | Pass |
| ACM-BT50 | 0.94 | 10 | 45,100 | Pass |
| ACM-BT140 | 1.33 | 10.2 | 44,600 | Pass |
| ACM-CB5 | 1.15 | 8.6 | 48,700 | Pass |
| ACN-CB10 | 1.68 | 8.2 | 49,700 | Pass |
| ACM-CB15 | 1.70 | 12.4 | 40,500 | 4545 V/mm |
| ACM-CB20 | 2.33 | 596.6 | 5840 | 2941 V/mm |
| ACM-CB5-BT15 | 2.11 | 9.7 | 45,900 | Pass |
| ACM-CB10-BT10 | 1.42 | 9.6 | 45,900 | Pass |
| ACM-CB15-BT5 | 1.04 | 9.8 | 45,500 | 7377 V/mm |
| ACM-CB20-BT2 | 1.98 | 21.7 | 30,600 | N/A |
| ACM-CB20-BT5 | 1.94 | 18.6 | 33,100 | N/A |
| ACM-CB20-BT10 | 2.62 | 40.5 | 22,400 | N/A |

^aThe permittivity values were taken at 1 Hz frequency.

^bBreakdown value is mentioned if at least one specimen failed during the actuation test = electric field at failure. Test is marked as "Pass" if the maximum applied voltage of 4 kV did not lead to the dielectric breakdown of the sample, and "N/A," if the test was stopped at 1.5 kV due to the sudden rise in leak current.

interfacial polarization, when the charge is built up at the filler–matrix interface and then moved in electric field, thus, leading to a current leak. Besides that, the high dielectric losses will cause heating of the material under electrical energy reducing the materials life time. Material with high loss factor is not suitable for electromechanical applications as also more energy is required to get it actuated and thus materials with low dielectric losses are preferred. The addition of small amount of BT decreases dielectric losses to more acceptable level especially above 10 Hz frequencies. At higher BT content, the synergistic effect of the fillers discussed previously leads to an increase in dielectric losses (Figure 6). When the ACM compounds that are having filler content well below percolation threshold are studied, it can be observed that the addition of BT has only minor effect on dielectric losses of CB-BT compounds when compared with CB only filled materials.

Another key parameter in DEAs is the elastic modulus that should be low. CB is traditionally used as reinforcing filler in rubbers due to strong polymer–filler interaction causing significant increase in tensile strength and elongation at break compared with the same amount of BT (Figure 7). Moreover, this explains why CB increases modulus of rubber much more than non-reinforcing BT filler (Table II). The compressional strain can be estimated by the eq. (2). According to this equation, dielectric permittivity and Young's modulus are determining factors for actuation performance in a constant electric field. Thus, the actuation behavior of rubber can be estimated by ratio of dielectric permittivity and Young's modulus, as applied by some researchers.³⁶ According to this estimation CB filled compounds should give better actuation strain than BT filled

ones (Table I). However, this formula does not take dielectric losses into account and thus should not be applied for high-loss compounds. Furthermore, rubbers do not show linear deformation and due to that Hooke's law is not valid for them. Thus, the determination and use of Young's modulus can be challenging for rubbers and will not necessarily give realistic values as the modulus of elastomeric actuators is dependent, for example, on pre-strain used. In the published articles many different ways to determine Young's modulus has been used and the most of the articles do not give details on how the Young's modulus has been determined. In the present article, the actuation measurements have been carried out with the 75% pre-strain, thus, for actuation measurements, the modulus at 75% elongation has been determined.

Aside from relative permittivity and elastic modulus, another key property of DEAs is electrical breakdown strength E_b , which determines the maximum electric field a material can withstand. Although E_b can be estimated for random composites with the following equation⁷:

$$E_b = e^{-1/2} \sqrt{\frac{Y}{2\epsilon_0\epsilon'}} \cong 0.6 \sqrt{\frac{Y}{2\epsilon_0\epsilon'}} \quad (10)$$

it is not precise as it does not take into account important factor such as, for example, pre-strain of the material, type, size, shape, and conductivity of fillers and other compound ingredients. Moreover, such equation cannot include the effect of filler agglomerates that cause the distortion and enhancement of local electric fields.³⁶

The calculated E_b values are presented in Table II. It can be seen that, as expected, the use of fillers decreases the dielectric

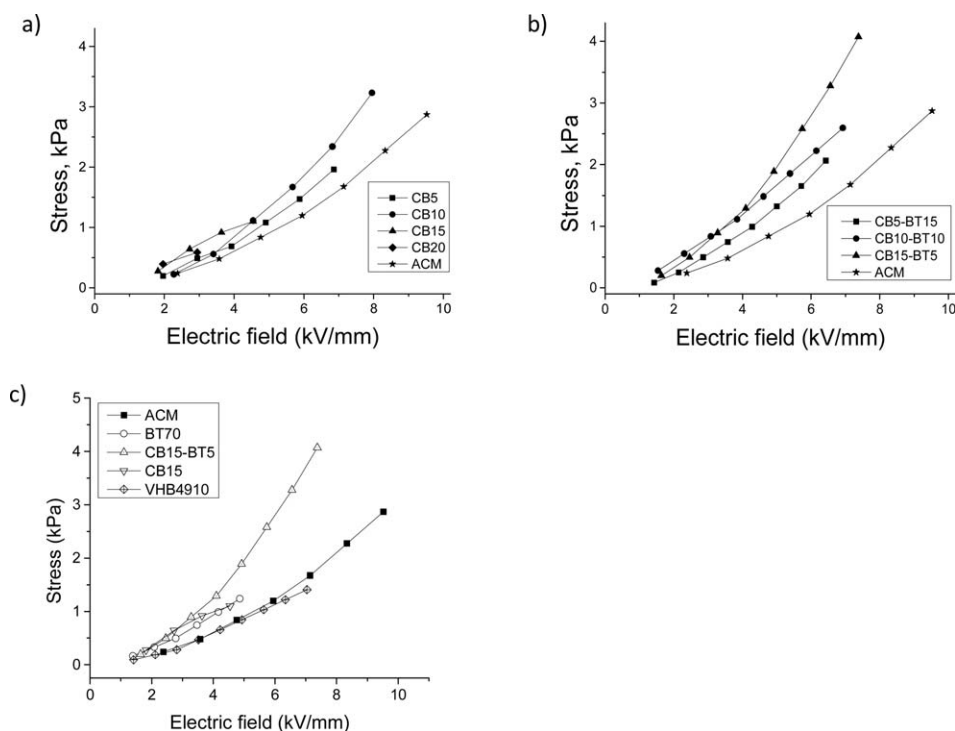


Figure 8. Actuation stress of (a) ACM-CB and (b) ACM-CB-BT compounds depending on the applied electric field, and (c) the comparison of actuation stress to the VHB4910 acrylic adhesive by 3M.

strength especially when the amount of the electrically conductive CB is close to the percolation threshold (20 phr). According to the Gyure and Beale this is due to regional short-circuits in the composite causing higher electric field locally.³⁷ However, it is interesting to note that according to the eq. (10), the addition of small amount of BT increases E_b of ACM-CB20 compounds due to increased interparticle distance of CB particles making the material more suitable for DEAs. It is worth to mention, that dielectric breakdown strength is a complex phenomenon that is affected by multiple factors, including the type of electrode, sample thickness, duration of the test, heat build-up, etc., and is a statistical event. The work of Carpi *et al.*³⁸ points out that the standard method of measuring electrical breakdown properties of soft dielectric elastomers concerning their use in DEA is not available. Some authors,^{39,40} however, present the electric field at which the actuator failed during the test as the dielectric breakdown value. In the present work, the similar approach is applied and the results are shown in Table II. Nevertheless, it should be noted, that these values are dependent on the test set-up and cannot be compared with the results obtained with other test set-ups and sample specifications.

The actuation results of ACM-CB compounds are presented in Figure 8. It can be seen that after incorporation of the BT particles in carbon black containing rubber the actuation stress is greatly improved. It can be noted that compounds containing 15 and 20 phr of CB are limited in their working range to much lower electric fields due to their very low dielectric breakdown strength. When BT is added into the compounds, the dielectric strength of the samples increases and the actuators can be used at higher electric fields as BT prevents formation of

CB network. All the compounds with fillers show higher actuation stress at a given electric field compared with the unfilled ACM. If the actuation stress values of the sample CB 10 is compared with the pure rubber at 7 kV/mm applied field a nearly twofold increase of the stress values can be seen.

Finally, the studied rubber compounds is compared with VHB 4910 acrylic tape produced by 3M company [Figure 8(c)] as it is the most frequently used material for DEAs. It is seen that pure ACM has similar actuation stress with the VHB 4910 tape whereas all other compounds show higher actuation performance than the tape.

CONCLUSIONS

The effect of traditional high dielectric filler and electrically conductive filler on the actuation behavior of ACM was studied. It was found that carbon black increase relative permittivity of ACM substantially more than BT leading to higher electromechanical response under applied electric field. Therefore, small carbon black concentrations below the percolation threshold can be used as filler in DEAs. Moreover, the formation of conductive path of CBs can be interrupted by adding small amounts of BT into the rubber compound. The synergistic effect of fillers further increase actuation performance and also decreases dielectric losses and increase electrical breakdown strength making the rubber compounds more suitable for the DEAs. The developed rubber compounds had higher actuation stress than the often used VHB 4910 tape indicating their potential for the DEAs.

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