

Improved electromechanical response in acrylic rubber by different carbon-based fillers

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ABSTRACT

Dielectric elastomers are materials often utilized for the fabrication of electroactive actuators. Acrylic rubber (ACM) is very widely used in dielectric elastomer actuators (DEAs). However, its overall good performance is limited by the high operating electric field required. In the present work, we compare the effect of different types of conventionally used carbon black (CB) as well as other carbon-based fillers on the dielectric and actuation properties of ACM in order to show that performance of DEAs can be improved by the development of ACM composites. Indeed, addition of CB, carbon nanotubes (CNTs) and synthetic graphite leads to an increase in the relative dielectric permittivity of elastomeric material. Moreover, incorporation of nanodiamonds results in reduction of dielectric losses. Finally, actuation stress is remarkably improved by CNTs and different grades of CB.

INTRODUCTION

During recent years, the feasibility of different conventional elastomers for dielectric elastomer actuators (DEAs) has been studied widely. These developments emphasized the materials most resembling those of natural muscles in terms of strain, actuation pressure, energy density, efficiency and response speed [1]. Dielectric elastomers are good candidates for actuators due to their ability to become polarized in applied electric field. In addition, due to low elastic modulus and high elasticity of elastomers they are able to deform with relative low external energy. However, many DEAs require very high operating electric field that can reach 100 kV/mm, which is the main drawback of such actuators [2,3]. This makes their utilization in real applications difficult and complicated. [4,5].

Silicone and polyacrylic rubbers (ACM) are the most widely used polymers in DEAs [6]. One of the approaches to improve their actuation performance is addition of electrically conductive fillers such as carbon black (CB) [7,8], carbon nanotubes (CNTs) [9-12] and graphene [13-15], while graphite is sometimes used in conductive rubbers for electrodes [16]. Such an approach is focused on utilizing conventional and readily available fillers and elastomers without any chemical modification and allows increasing total relative dielectric permittivity of the material. High dielectric constant i.e. relative permittivity is a key property in actuators as expressed by Maxwell's pressure (σ) equation [17,18]:

$$\sigma = \varepsilon' \cdot \varepsilon_0 \cdot E^2, \quad (1)$$

where ε' is dielectric constant of the material, ε_0 is dielectric permittivity of an empty space (8.854×10^{-12} F/m), and E is electric field. The Maxwell's pressure describes the stress of an ideal dielectric elastomer actuator generated in an electric field [19], but it is applicable for elastomers having low crosslink densities and/or small deformations [20,21]. Moreover, actuation behavior can

be estimated by calculating compressional strain (s_z) that is the Maxwell pressure divided by Young's modulus (Y) [17,18]:

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

Therefore, apart from high relative permittivity, a low elastic modulus of a material is strived for actuators that are able to work at lower operating voltages [22,23], and such approach is applied in the current work as well. As mentioned earlier, CB and CNTs are used to increase relative permittivity of elastomer, but such improvement in permittivity is usually accompanied with increased dielectric losses through the current leak [8]. It happens due to the induced Maxwell-Wagner polarization originating from the charge built up at the filler-matrix interface because of significant difference in their conductivities. Charge accumulated at the material interfaces can move through the material in electric field thus leading to the increase in dielectric losses and electrical conductivity. [24] Moreover, CB and CNTs typically increase elastic modulus because of their reinforcing nature, and thus the advantages obtained with higher dielectric permittivity are partly lost by increased material stiffness and energy dissipation. Therefore, other carbon-based fillers need to be studied for their ability to affect the dielectric properties of elastomers and electromechanical performance of the resulting composites. Graphite (G) [25] and detonation produced nanodiamonds (NDs), as less studied fillers for the application in electroactive elastomer composites, were chosen for this purpose. At ambient conditions bulk conductivity can reach 900 S/cm for graphite [26] and only about 10^{-11} S/cm for NDs [27], while bulk conductivity of carbon black is up to 0.1 – 100 S/cm [28] and up to 100 S/cm for multi-walled carbon nanotubes [26].

The present research focuses on actuation performance of plasticized ACM filled with different types of carbon-based fillers in order to obtain high-performance elastomer composite capable of deformations in response to an applied moderate electric field. Here, dielectric properties and

electrical conductivity of materials are studied with respect of CB particle size. Finally, effect of low loads of graphite, CNTs and NDs on the properties of electroactive elastomer composite are investigated and compared to that of carbon black.

EXPERIMENTAL

Materials.

Acrylic rubber (HyTemp 4051EP, Zeon Chemicals L.P.) was used as matrix because ACM is a well-studied polymer for actuator application. Curing system included sodium stearate (3 phr, parts per hundred rubber), sulphur (0.3 phr), and stearic acid (0.5 phr). Additionally, 2 phr of dioctyl adipate (DOA) plasticizer was added to improve dispersion of fillers and to decrease elastic modulus of compounds. The DOA is known to improve the actuation properties of acrylonitrile rubber (NBR)[29]. Therefore, due to polar nature of both NBR and ACM, it is expected that using DOA as plasticizer improves also the actuation properties of ACM.

Three different types of CB fillers were studied: N-234, N-375 and N-550 from Evonik at 5 and 20 phr loadings. In addition, low concentrations of graphite (Timrex KS44, Timcal Graphite & Carbon), CNTs (Baytubes C 150 P, Bayer) and two nanodiamond powders - with carboxylated surface (uDiamond Molto Vox, Carbodeon) and with hydrogen terminated surface with some residual amine groups (uDiamond Molto Nuevo, Carbodeon) were compared and designated carb.ND and hydr.ND respectively. Filler properties, provided by the suppliers, are presented in Table 1.

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. All the samples used in the study were prepared according to the

compounding scheme presented in Table 2. The sample names are designated as “Filler type abbreviation”_”phr amount”, if not mentioned otherwise. Mixed compounds were formed to 0.5 and 1 mm sheets and cured at 175°C for 15 minutes.

Characterizations

Tensile tests of the samples were carried out with a Messphysik Midi 10-20 universal tester according to ISO 37 using dumb-bell test specimen type 3 at 200 mm/min rate. Three specimens of each sample were tested, and the average value was calculated.

Dielectric permittivity, dielectric loss and electrical conductivity were measured with a Novocontrol Alpha-A dielectric analyzer. The frequency sweep was done from 1 MHz to 0.7 Hz. Five specimens of each samples were tested, and the average value was calculated.

For the scanning electron microscopy (SEM, model Zeiss ULTRApplus) imaging, 3 kV and 10 kV acceleration voltage was used for the samples containing 1 phr of fillers and the other samples respectively. The samples were cryo-fractured under liquid nitrogen and the fracture surfaces were coated with thin evaporated carbon layer to ensure good conductivity.

Actuation measurements were carried out in equipment prepared in-house. The actuator (20x50 mm) was from 0.5 mm thick elastomer film (Fig. 1a). Electrodes were fabricated by spreading carbon black paste (carbon black type N-234 mixed with toluene) on both sides of elastomer and drying at ambient conditions. DC voltage from 0 to 5 kV was applied to the electrode via conducting copper plates from high voltage amplifier (TREK model 10/10B-HS) in 0.5 kV steps. The scheme of the assembly is shown in Fig. 1b, and the actual equipment – in Fig. 1c. The measurement was controlled via LabVIEW 2012. The change in force during electric actuation was measured with a 10 N load cell (LTS-1KA, Kyowa).

RESULTS AND DISCUSSION

Effect of different types of carbon black

Relative dielectric permittivity describing material's ability to polarize in an electric field is one of the key properties in DEAs as described in Eq. 1. Electrical conductive fillers are known to induce strong Maxwell-Wagner polarization i.e., interfacial polarization that arises due to the significant difference in permittivity and electrical conductivity of a matrix and a filler and increases both the relative dielectric permittivity and loss of the composite due to the charge accumulation at the interface. Use of CB in DEAs is especially interesting because it is the conventional filler for the rubber industry. The effect of different CB types on the relative permittivity of the ACM is presented in Fig. 2. It is evident from the Fig 2a that all CB types increase the relative permittivity of ACM remarkably already at 5 phr concentrations. This is due to higher polarization at the interfaces of fillers and matrix as well as due to increased electrical conductivity. The Maxwell-Wagner effect is much stronger for composites containing higher loads of conductive filler as can be seen from Fig 2b. Addition of 20 phr conductive CB particles into ACM matrix results in increase of relative permittivity up to 1490 at 1 Hz for N234 which is more than 200 times higher than the permittivity of the matrix. At the same time, N550 increases the relative permittivity of the ACM composite only 7-fold. From the shape of the N234 curve it can be assumed that increase in permittivity is mostly due to the transport of the charges through the CB particles, as a dramatic increase in dielectric permittivity at low frequencies normally indicates the interfacial effect and the propagation of mobile charge carriers.

Similar trends and observations, as for relative permittivity, are valid for the electrical conductivity plots of the ACM-CB compounds presented in Fig. 3. At 5 phr CB loadings, the electrical conductivity of the composites is a bit higher than that of the neat ACM, as such amount of conductive particles

is not enough for percolation. At higher filler content, the electrical conductivity of the compounds increases significantly and differences between various CB types can be observed. The compound containing carbon black N234 has the highest conductivity (6.3×10^{-9} S/cm at 1 Hz for 20 phr CB load) in the group and it is rather semiconductor than dielectric material. At the same time, conductivity of the composites containing 20 phr of N375 and N550 is 1.4×10^{-10} S/cm and 2.9×10^{-11} S/cm at 1 Hz, respectively. Changes in electrical conductivity can be explained by different particle sizes and surface areas of the CBs. N234 has the smallest density and thus its volume fraction is higher than the other carbon blacks when the weight of the filler is same in all the compounds. Therefore, filler-filler interaction is stronger in the compound containing N234 and, thus, it is able to form conductive path, while addition of N375 and N550 to the matrix leads to poor network formation.

The negative aspect in using electrically conductive fillers in DEAs is high dielectric losses due to the energy dissipation by current leak. High dielectric losses reduce lifetime of materials, increase energy consumption and reduce functionality of the composite. The loss factor, or tan delta, is a ratio of dissipated energy to energy stored in cycle. The loss factors of ACM-CB compounds as a function of frequency are presented in Fig. 4. Compounds containing 5 phr of CBs have similar dielectric losses, but, compared to the unfilled material, have steeper slope of the tan delta curve due to small increase in electrical conductivity and the effect of electrode polarization. It can be seen that the compound containing 20 phr N234 has much higher losses than the other compounds in the whole frequency range that is associated with significantly increased electrical conductivity, as well as macroscopic polarization effect. Other CB types have clearly lower loss factor. Therefore, the elastomers filled with 20 phr of N550 and N375 carbon black types would resist sinusoidal stress in DEAs longer than the N234 and perform better as less energy is dissipated during the actuation cycle.

Although the dispersion of the samples and solely filler size seem to have little effect on the dielectric permittivity already at 5 wt.% of filler, as discussed in the work of Yang et al.[30], CB N234 having smaller particle size with larger surface area had stronger effect on dielectric properties. As no significant differences in CB dispersion were seen for both 5 and 20 phr of filler (Fig. 5), the dispersion of fillers was assumed to have no effect on the dielectric properties of the materials. Finally, it should be noted that the fillers considered here were incorporated into the rubber with respect to their weight, more particularly, parts per hundreds of rubber and the fillers have different density. The higher the volume fraction of CB and the larger the surface area is, the higher the permittivity of the ACM-CB compounds can be due to enlarged matrix-filler interface and thus stronger Maxwell-Wagner polarization and possibility to accumulate more electric charges.

In order to enable movement of polymer chains during actuation and ease the thickness contraction of the material the elastic modulus should be low. CB is reinforcing filler that typically has strong interaction with polymer. This increases the elastic modulus often referred to as Young's modulus. Here, tensile stress at 75% elongation was used to characterize the mechanical stiffness of the materials because the samples were pre-stretched to 75% in the actuation tests. The corresponding stress values are presented in Fig. 6 and will be further referred to as elastic modulus of the material. It can be seen that the addition of all CBs increases elastic modulus and this increase is more pronounced with the addition of CB N234 that has the smallest particle size and the highest volume fraction in the compound and thus better mechanical reinforcement due to increased filler-filler interaction.

The actuation results of ACM-CB compounds are presented in Fig. 7. The curves show the stress reactions obtained from the samples at certain electric field starting from the lowest stress valued detected. It can be seen that all CB-filled compounds have much stronger electromechanical

response than neat ACM at relatively low electric fields. The compound containing N234 has stronger actuation effect than the compounds containing other CB types at the same load, but the difference is not very pronounced between different grades. Although the compound containing 20 phr N234 first shows higher induced stress it is subjected to the early failure at only about 3.5 kV/mm (Fig. 7b) possibly due to the conductive paths formation within the material and thus due to decrease in breakdown strength.

Effect of carbon nanotubes

Multiwall carbon nanotube consists of tubular graphene sheet layers. CNTs are electrically conductive and their electrical conductivity is higher compared to CBs. Due to a very small particle size and high aspect ratio it is able to form a conductive path at low filler concentrations and should be able to increase permittivity of the rubber strongly [31]. The Fig. 8a presents the relative permittivity values of CNT-filled ACM compared to unfilled ACM. It can be seen that relative dielectric permittivity increases already at very small filler concentrations. Addition of up to 1 phr of CNTs results in less than one-unit increase in permittivity. Although dielectric losses are also increased by addition of carbon nanotubes, as can be seen from Fig. 8b, results showed no notable dependence of loss factor on the concentration of CNTs.

Due to high conductivity of CNTs it was expected that electrical conductivity of ACM compounds would increase strongly after addition of CNTs but only minor increase in conductivity is observed (Fig. 8c). This is probably due to poor dispersion of CNTs: They stay in big agglomerates and CNT network is not formed. Despite of that and the double increase in dielectric loss factor at low frequencies, addition of 1 phr of CNTs leads to threefold increase in actuation stress at 4.5 kV/mm electric field (Fig. 8d) compared to neat ACM. However, addition of 0.1 phr of CNTs resulted in worse

actuation properties compared to unfilled ACM. Small increase in relative permittivity as well as twofold increase in dielectric losses without notable change in relative permittivity may be responsible for that.

Effect of graphite and nanodiamond

Graphite and diamond are allotropes of carbon that are available as nano-sized particles. Synthetic graphite is fabricated at very high temperatures from unstructured carbon. Graphite is highly crystalline form of carbon and it is known to have good thermal and electrical conductivity. [32] Unlike graphite, nanodiamonds are poor conductors of electricity. Detonation nanodiamonds (NDs) are carbon-based spherical particles about 4-6 nm in diameter obtained by blasting-induced decomposition of the combination of explosives [33]. Nanodiamonds have an active surface that can be modified in order to get desired properties. With the right polymer choice, the use of small concentrations of such fillers can lead to an increase in relative permittivity due to the interface effects but not facilitate energy dissipation even when composite is stretched. However, ND particles form strong aggregates and agglomerates that are very difficult to disintegrate without use of surfactants, surface modifications or special techniques. [34] This affects the dielectric and actuation performance of the composites to some extent.

From Fig. 9a it is seen that addition of low amounts of graphite into the ACM leads to moderate increase in relative permittivity while dielectric losses are maintained at reasonable level. Energy dissipation at low frequencies is almost unaltered for all three graphite-filled compounds and increase in total electrical conductivity of composite can be neglected (Fig. 9b and Fig. 9c). This leads to an assumption that here electrical conductivity of graphite plays no significant role and does not give rise to the notable Maxwell-Wagner polarization. Possible explanation can be found in the

structure of the graphite filler, that is not exfoliated, and formation of particle agglomerates that reduce the active filler surface thus decreasing an interface between matrix and filler.

All the studied ACM-graphite compounds showed very similar behavior to the composites containing up to 1 phr of CNTs in terms of relative permittivity, losses and electrical conductivity. However, ACM-CNT composite containing 1 phr of filler showed much better actuation behavior compared to ACM-graphite composite with 1 phr and even 5 phr of filler (Fig. 9d). Nevertheless, material mechanically reacted to the lower applied electric field than the unfilled ACM, but otherwise addition of up to 5 phr of graphite had no significant effect on actuation stress.

Nanodiamonds are known to have little reinforcing effect on elastomeric materials and they are dielectrics [35]. Some authors report increase in both tensile strength and dielectric permittivity after addition of ND into the silicone matrix [36]. Dielectric and electrical properties of NDs depend to some extent on its surface functionalization. Relative dielectric permittivity of NDs is about 3.5, but it can be increased owing to the absorbed water [27,37,38]. Carboxylated NDs (carb.ND) utilized in the current study have hydrophilic surface that may absorb water vapors under ambient conditions. Hydrogen terminated nanodiamond powder (hydr.ND) with some residual amine groups is less prone to absorb air humidity. The relative permittivities of the ACM compounds containing 1 phr of different nanodiamond powders are presented in Fig. 10a and dissipation factors are shown in Fig. 10b. It can be observed that addition of carboxylated NDs leads to a small decrease in dielectric permittivity at low frequencies, while other ND filler cause no significant changes in dielectric permittivity of the composite. No increase in dielectric loss factor is noted for all ACM-ND compounds, and ACM-hydr.ND compound even show reduced dielectric energy dissipation at low frequencies. It can possibly be explained by the surface chemistry of such NDs and that addition of hydrogen terminated NDs resulted in increased electrical resistance of the composite shown in Fig.

10c. Carboxylated nanodiamonds do not induce any notable changes to the electrical conductivity of ACM.

It was first expected that more polar carboxylic groups on the ND surface would lead to higher relative permittivity and electromechanical actuation. However, among the ACM-ND composites, ACM-hydr.ND shows the best actuation results compared to the neat ACM rubber as seen in Fig. 10d that is possibly due to the reduced dielectric losses of the material and higher electrical resistivity.

The effect of mentioned various carbon-based fillers can be seen more clear when material properties are compared at 1 phr. Figures 11a-c show the frequency dependences of relative dielectric permittivity, loss factor and conductivity respectively on the type of filler used. It is clear that more conventional fillers like graphite, CB and CTNs lead to more significant increase in relative permittivity, but they also increase dielectric loss factor and electrical conductivity of the compounds. Addition of hydroxyl-terminated ND is advantageous in terms of reduced dielectric losses, but carboxylated ND powder does not modify loss properties of ACM much. The observed changes are not related to the differences in the filler dispersion, as similar filler agglomerates were detected from all the samples (Fig.12). Elastic moduli of the ACM composites containing 1 phr of carbon-based fillers are shown in Fig. 13. Addition of 1 phr of fillers, except for graphite, leads to minor increase in the elastic moduli of the composites.

Actuated stress versus applied electric field for the composites discussed above is presented in Fig. 11d. It can be seen that addition of fillers resulted in the improved electromechanical actuation, Among the studied fillers, compounds containing 1 phr of CNTs and CB are clearly improving the actuation properties of ACM the most, regardless of the small increase in elastic moduli. Such,

actuation stress at 5 kV/mm can be improved almost threefold by addition of 1 phr of CNTs and more than twofold by addition of 1 phr of CB N234 type.

As addition of NDs and graphite also leads to an increase in actuation stress, such fillers in larger amounts could have possibility to be used in DEA applications. Moreover, the future research can take into account an original combination of ND and graphitic structures known as diamond blend or diamond soot. Diamond blend is a primary product of a detonation process, and is a precursor for obtaining purified surface-modified NDs. Finally, diamond blend may show synergetic effect as the mixture of carbon allotropes [34], similar to what was studied previously for other filler combination, for example, in the work of Zhao et al. [8], Dang et al. [39] and our previous work [40].

CONCLUSIONS

Addition of conductive CB particles into ACM matrix results in significant increase of relative permittivity up to 1490 at low frequencies. Such improvement is related to the filler size and its developed surface. However, such raise in permittivity level is accompanied with very high losses and an increase in electrical conductivity of the material thus meaning that one and the same filler at different loads can make a composite unsuitable for DEA application despite of its high relative permittivity. Next, ACM compositions with different carbon-based fillers showed promising results for DEA applications, such as addition of 1 phr of CNTs led to over threefold increase in actuation stress at 5 kV/mm compared to neat ACM. Such improvement is similar to the results obtained with 20 phr of CB. Instead, addition of up to 5 phr graphite resulted in very moderate increase of actuation stress and carboxylated NDs showed even less improvement. At the same time addition of hydrogen terminated NDs resulted in better actuation compared to the same amount of graphite,

and reduced dielectric loss factor of ACM twice at 1 Hz. Thus, nanodiamond powders could be utilized in dielectric elastomer composites that may extend the area of their application.

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Table 1. Material properties of different carbon-based fillers

Filler	Carbon black			CNTs	Graphite	NDs
	N234	N375	N550			
Particle size	19 nm	27 nm	47 nm	13 nm x 1 μ m	13 μ m	4-6 nm
Nitrogen adsorption specific surface area, 10 ³ m ² /kg	~126	~100	~46	210	9	300
Bulk density, kg/m ³	320	345	360	130-150	190	500

Table 2. Mixing order for the rubber compounds.

Compounding ingredient	ACM	CB*	G	CNT	ND**	DOA	Sulfur	Sodium stearate	Stearic acid	Total mixing time, min
Compound	Ingredient addition time, min									
ACM (pure)	0	-	-	-	-	1	1.5	1.5	1.5	3.5
ACM-Carbon black	0	1	-	-	-	1.5	3.5	3.5	3.5	5.5
ACM-Graphite	0	-	1	-	-	1.5	3.5	3.5	3.5	5.5
ACM-CNT	0	-	-	1.5	-	2	4	4	4	6
ACM-ND	0	-	-	-	1	1.5	3.5	3.5	3.5	5

*type N-234, N-375 or N-550

** Hydrogenated or Carboxylated

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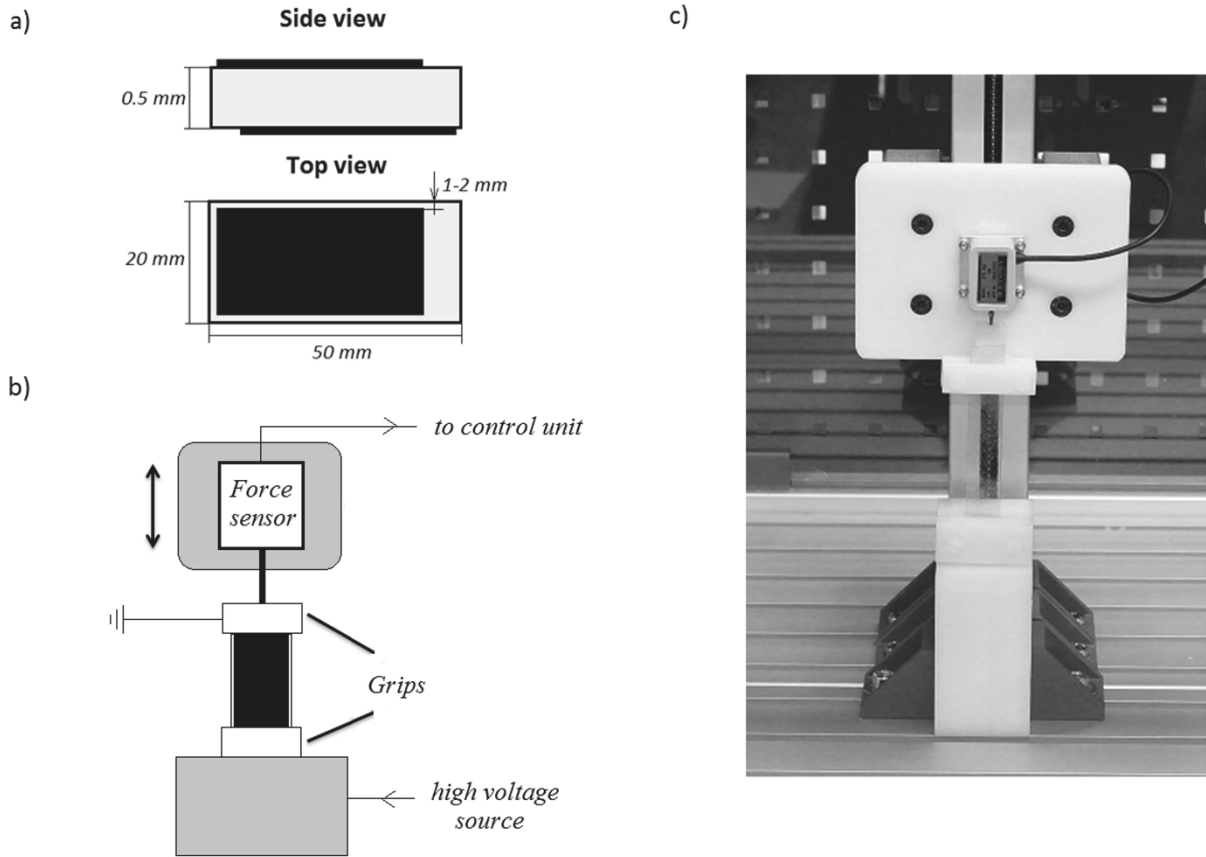


Figure 1. a) Electrode fabrication pattern and the dimensions of a rubber sample; b) placement of the actuator in the sample holder; and c) sample holder of the testing equipment.

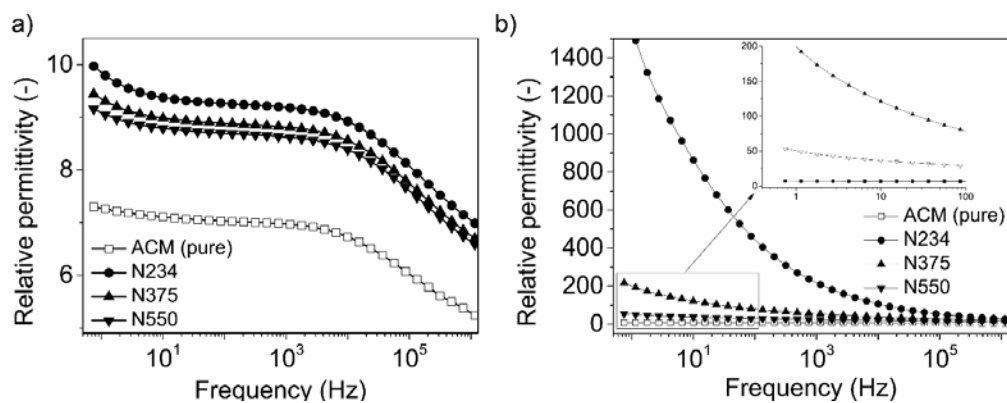


Figure 2. Relative permittivity of different acrylic rubber (ACM) samples containing a) 5 phr; b) 20 phr carbon black. Insert is added for better resolution of relative permittivity of the materials at low frequency.

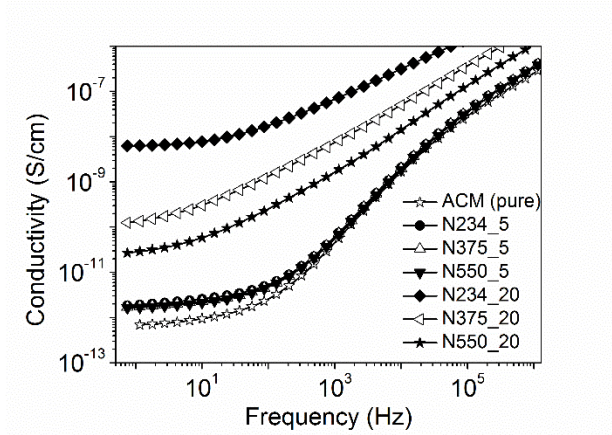


Figure 3. Electrical conductivity of different carbon black (CB)-filled acrylic rubber (ACM).

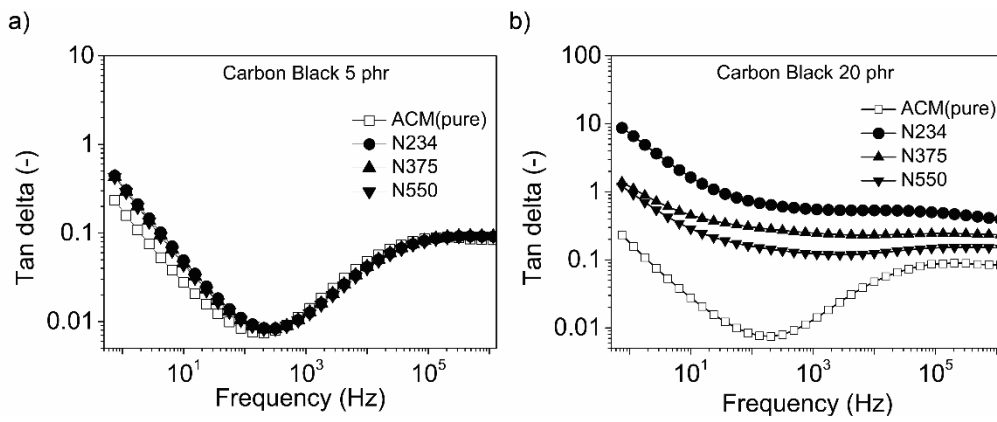


Figure 4. Dielectric loss factor of acrylic rubber compounds with a) 5 phr and b) 20 phr CB load.

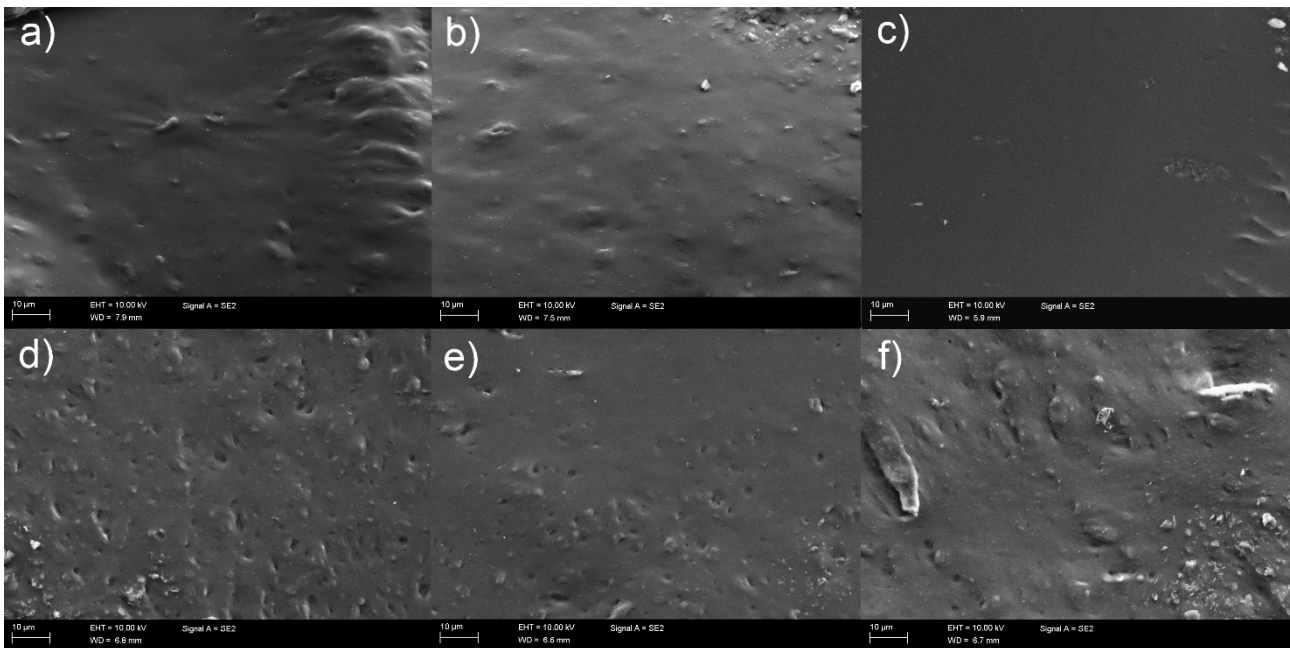


Figure 5. SEM images of ACM filled with carbon black: a) 5 phr of N-234; b) 5 phr of N-375; c) 5 phr of N-550; d) 20 phr of N-234; e) 20 phr of N-735; and f) 20 phr of N-550 (scale bar is 10 μm).

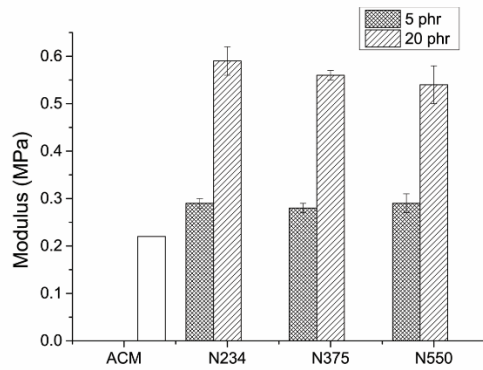


Figure 6. Elastic modulus (with standard deviations) of the compounds.

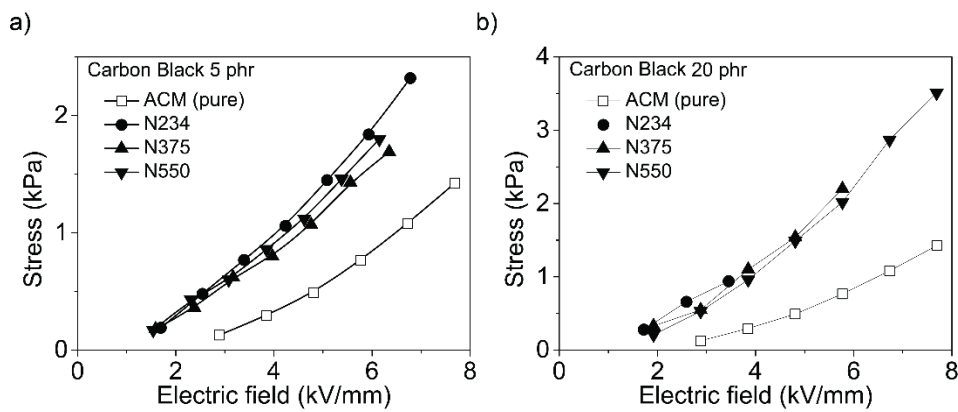


Figure 7. Actuation stress of the different CBs at a) 5 phr concentration; and b) 20 phr concentration.

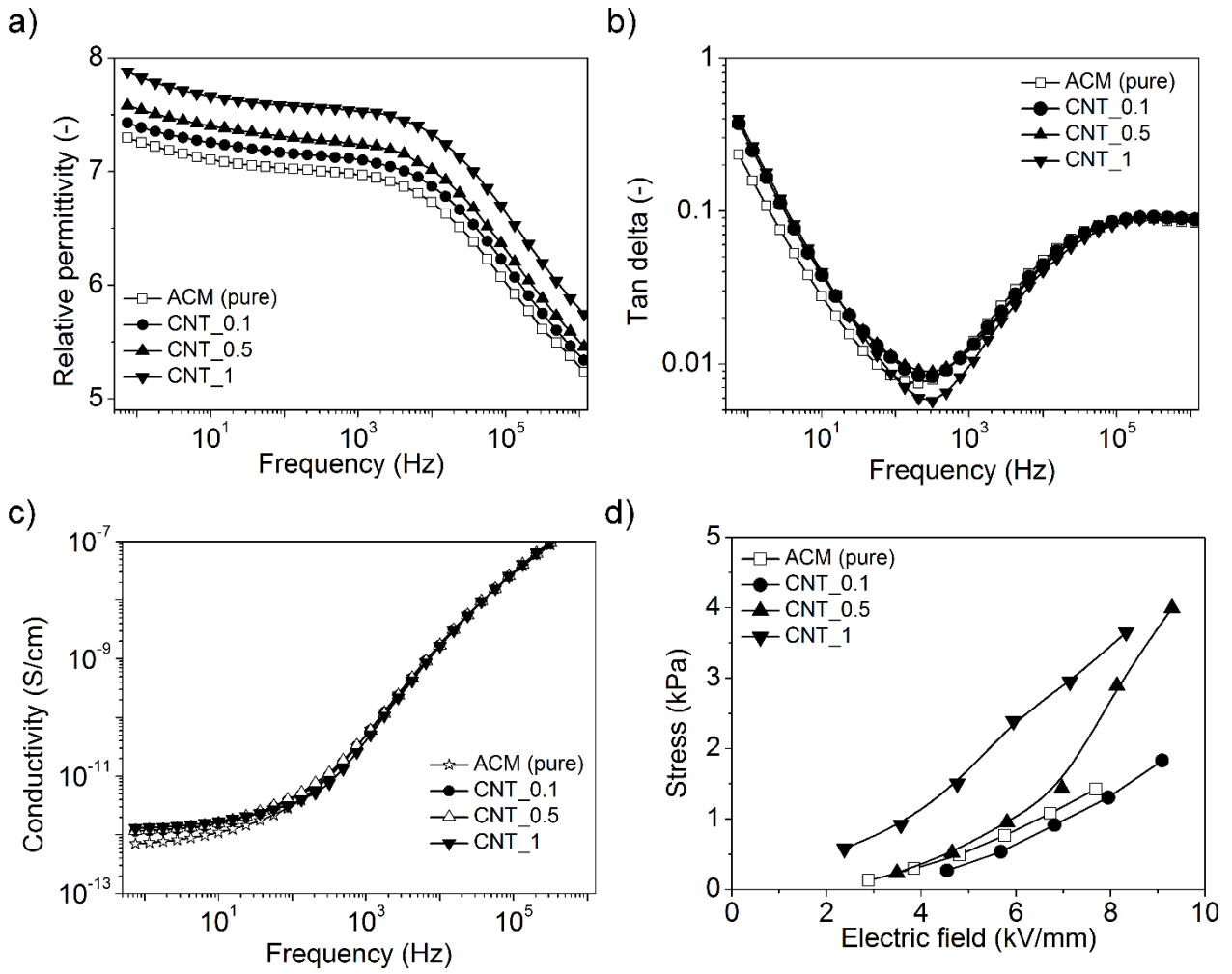


Figure 8. Effect of CNT load (0.1, 0.5 or 1 phr) on a) relative dielectric permittivity; b) loss factor; c) electrical conductivity; and d) actuated stress of ACM compounds.

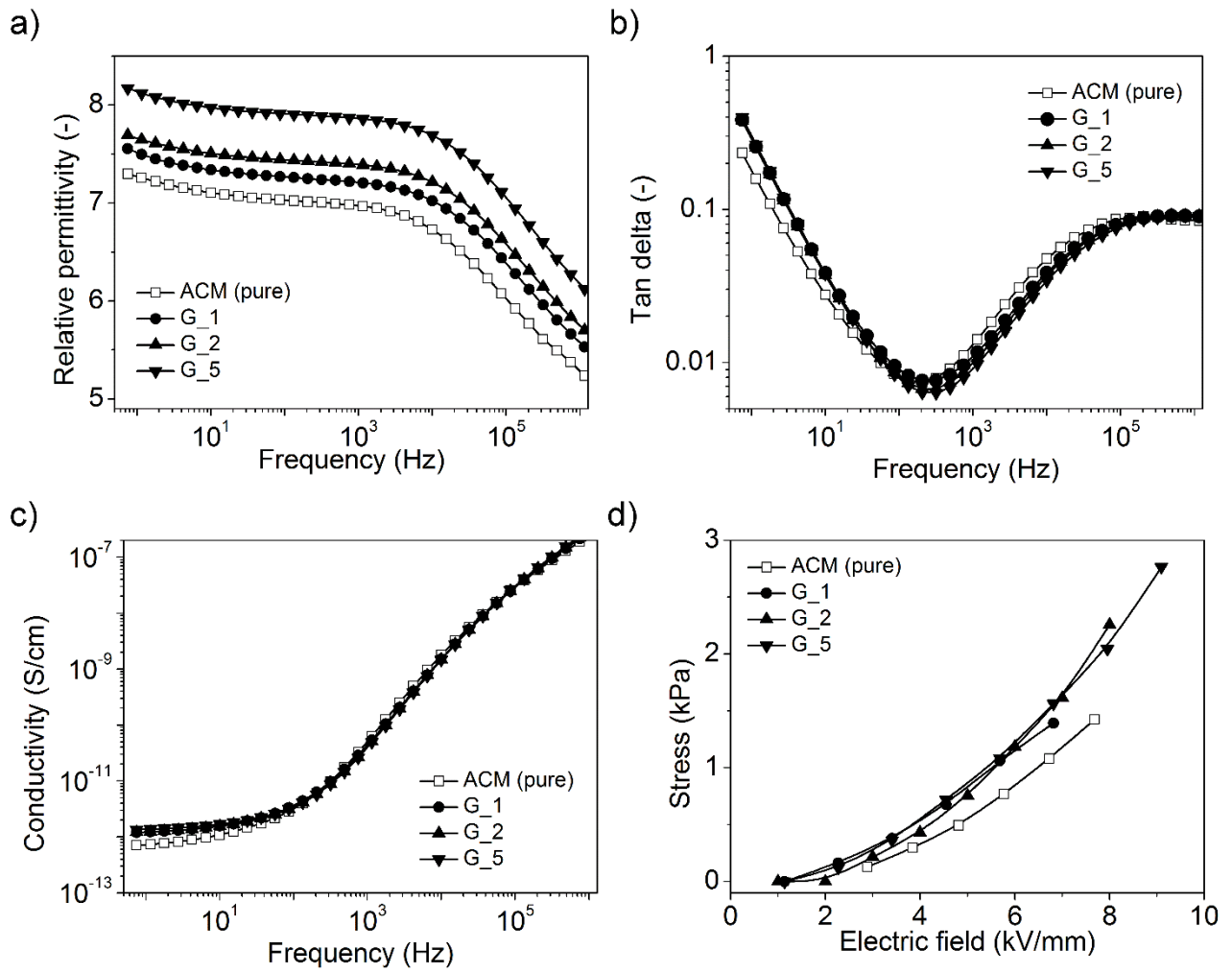


Figure 9. Effect of graphite load (1, 2 or 5 phr) on a) relative dielectric permittivity; b) loss factor; c) electrical conductivity and d) actuated stress of ACM compounds.

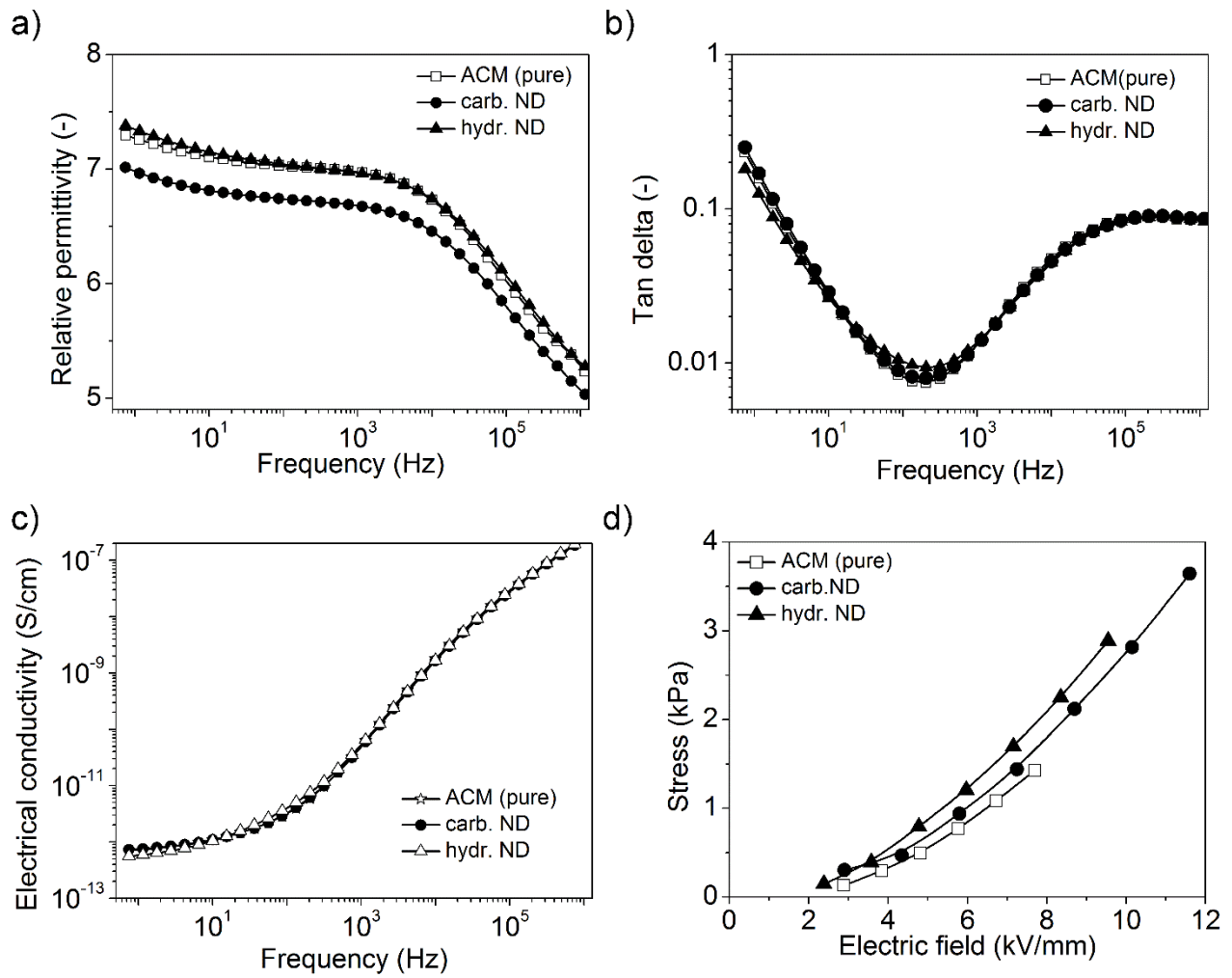


Figure 10. Effect of 1 phr nanodiamond powders on a) relative dielectric permittivity; b) loss factor; c) electrical conductivity; and d) actuated stress of ACM compounds.

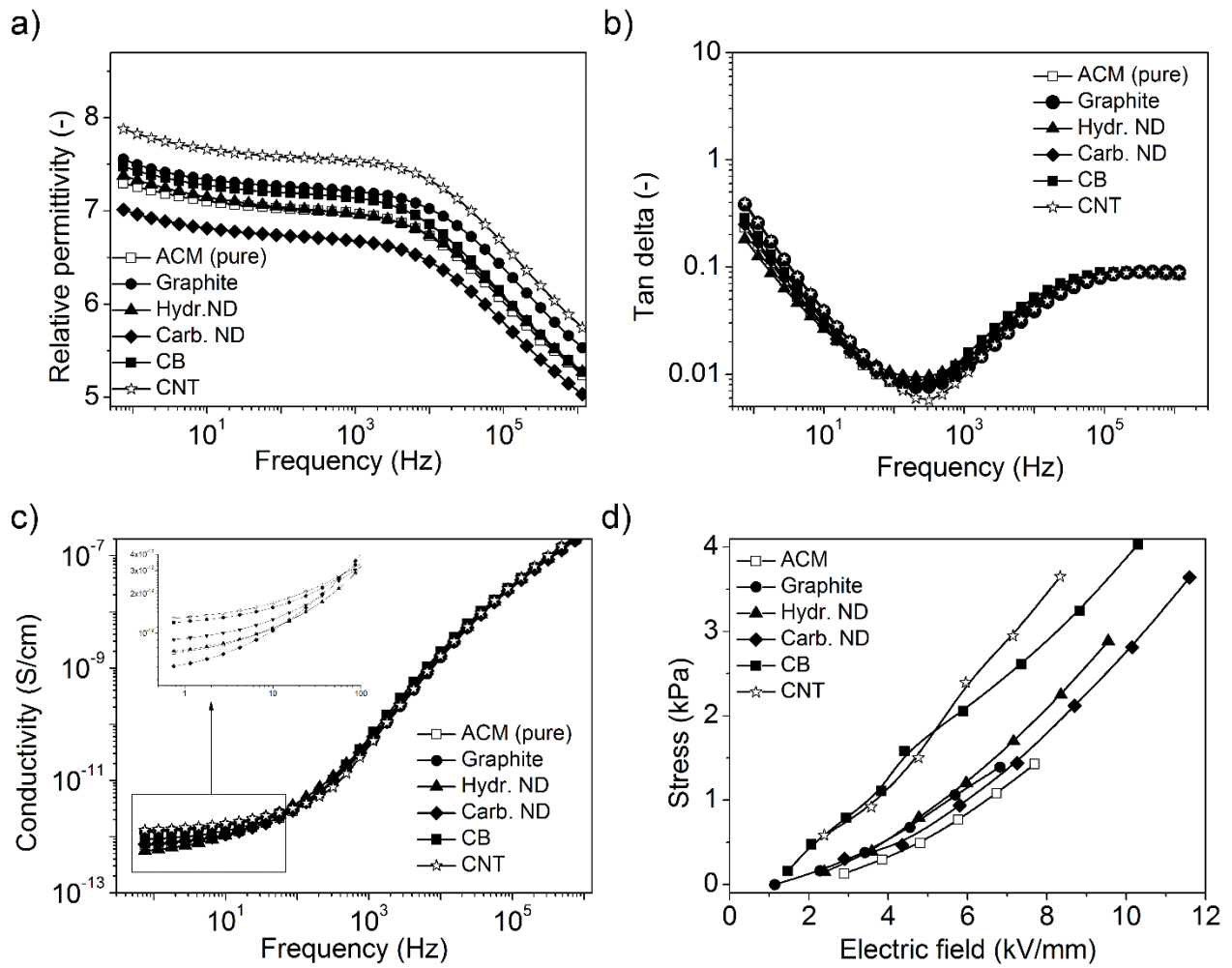


Figure 11. Effect of 1 phr of carbon-based fillers on a) relative dielectric permittivity; b) loss factor; c) electrical conductivity with the insert for better resolution of electrical conductivity at low frequency; and d) actuation stress of ACM compounds.

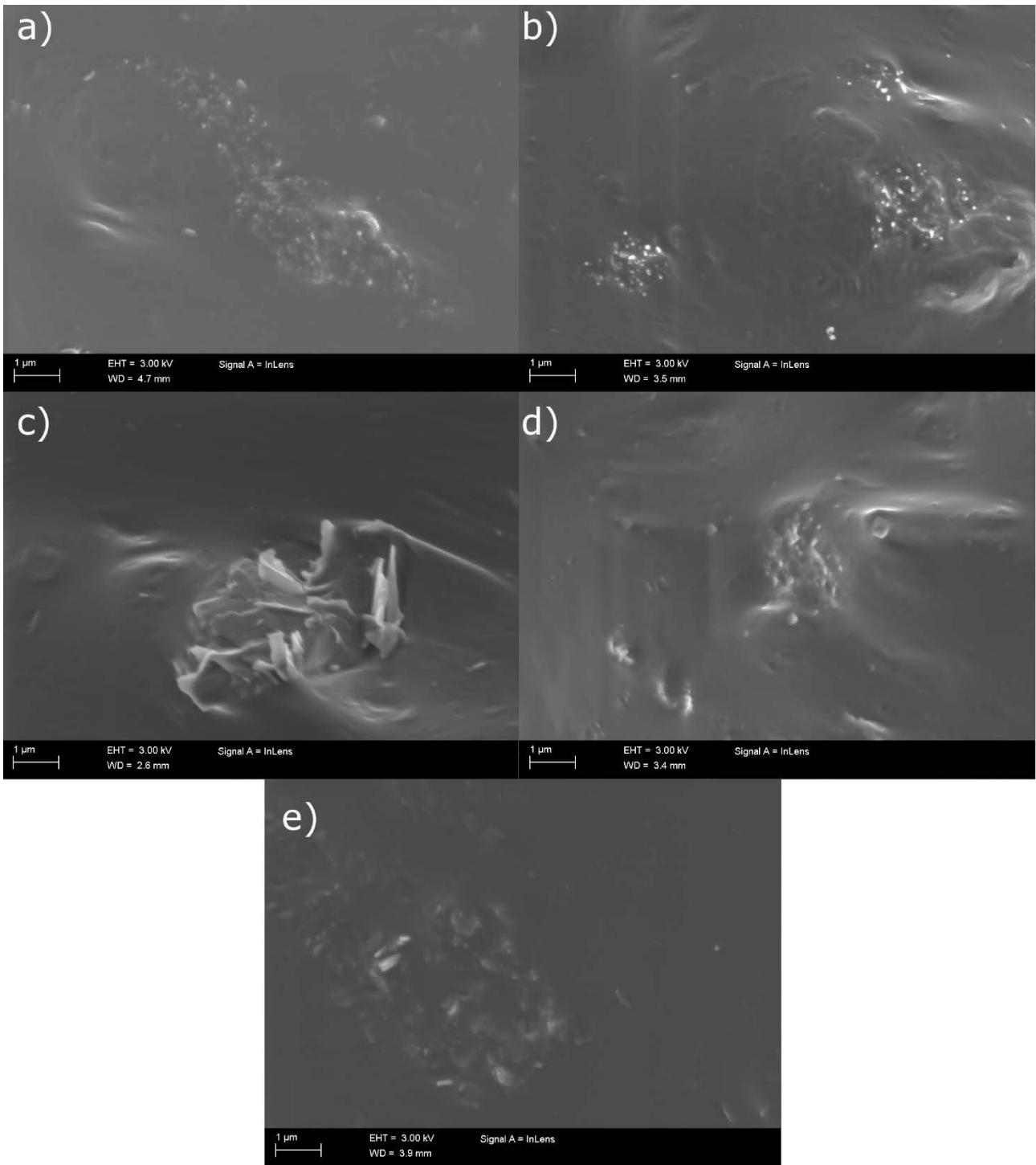


Figure 12. SEM images of ACM filled with 1 phr of a) carbon black N-234; b) CNTs; c) graphite; d) hydrogenated NDs; and e) carboxylated NDs (scale bar is 1 μm).

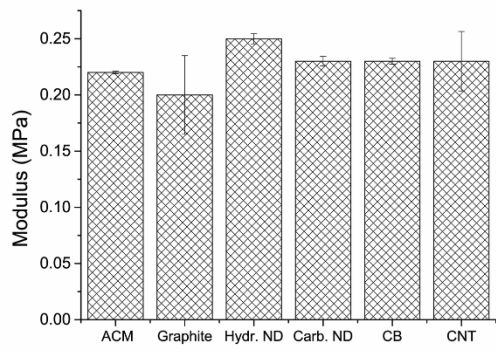


Figure 13. Elastic modulus (with standard deviations) of the ACM compounds containing 1 phr of carbon-based fillers.