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Porous Magnetorheological Nanocomposites: Dielectric and Electrical Properties

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ABSTRACT

This paper presents a brief experimental study on electrical and dielectric properties of porous film-shaped magnetorheological nanocomposites (MRNCs) based on room temperature vulcanized (RTV) silicone rubber and nano-sized carbonyl iron particles (CIPs). The electrical and dielectric properties of porous MRNCs were measured at five different filler concentrations. Several experiments were performed to measure the volume resistivity, dielectric constant and dielectric loss. The MRNCs dielectric properties were analysed with respect to the parameters like frequency and CIPs loadings. The electrical conductivity was studied in terms of volume resistivity. The distribution of internal charge was also measured for each sample using pulsed- electro-acoustic (PEA) method, under a DC electric field. The investigation's resultssuggest the porous MRNCs for smart, sensitive and light-weighted structures benefit from a lower electrical property,charge density and dielectric constants.

1. Introduction

Composites are the combination of two or moreMagneto-Rheological Nanocomposites (MRNCs) are the new category of smart materials which are sensitive to applied magnetic fields. MRNCs consisting of nano-sized CIPs embedded in a silicone rubber matrix, display a variety of interesting properties to design the flexible actuators [1- 5].The Author's previous works [3-5] were focused to fabricate a light-weight and flexible nonporous MRNCs for miniature gripper applications using laser ablated nanopowder of CIPs. The magnetic and mechanical property values of manufactured MRNCs have been determined and the different fabrication technologies have been tested as hot press, chemical vacuum vaporization and laser beam moulding to study the property changes which benefiting the operational condition of miniature actuators.

The Author's recent study [6] is concerned to report the manufacturing of porous MRNCs and its mechanical and magnetic properties variations. The work discusses the fabrication and characterization aspects of porous MRNCs with detail.

As the literature indicates there has been no reference research reported directly to the electrical and dielectric properties of MRNCs yet and the available published articles are concerning to the part of MR family composites as MR elastomers and silicone rubbers.

According to the literature, Enhanced electrical conductivity is observed in the modified multi-walled CNTs/methlyvinyl silicone rubber nanocomposites which is 7-order of magnitude larger than that of silicone rubber host [7].

In silicone rubber composites filled with CIPs, the variation of dielectric constant and loss values are dependent on the frequency increase and mostly the decreasing trends are appeared during the frequency cycle [8].The electro-ceramic fillers such as titanium dioxide can increase the dielectric constant and losses of a silicone rubber composite. Also, there will be a considerable boost in the dielectric constant of the oriented composite compared with randomly-distributed Titania particles [9].

It is examined that the values of relative dielectric constant increase with the increasing frequency and content of titanium diboride (TiB_2) nanoparticles in a silicone rubber matrix [10] at a frequency range of 1-12 GHz. The electrical conductivity of the as-prepared CNTs/ silicone rubber nanocomposites is increased with the increase of CNTs loading and through further increase in CNTs loading, more conductive paths are formed [11].

In a silicone rubber composites based on ferrite and CIPs, the dielectric loss is increased in some desired range of frequency [12]. In a comparative study of silicone rubber and epoxy based composites filled with CIPs[13], it has been declared that real permittivity of silicone rubber based composite is lower than epoxy based composites whilst the imaginary permittivity of silicone rubber based composite is higher than epoxy based composites regarding a frequency range of 26-40 GHz. The silicone rubber nanocomposites filled with titanium dioxide and barium titanate synthesized over a frequency range from 100Hz-10MHz [14] and its findings show that the dielectric permittivity increases whereas dielectric losses decreases with increasing filler concentration.

A mathematical relation for magnetorheological resistance was introduced to measure the electrical resistivity as a function of applied forces on the surface of samples [15] considering the fixed values of transverse magnetic field strength. The work summarizes that MREs become electro-conductive for volume concentrations of magnetizable phase greater than 40%.

Dependency of the dielectric permittivity on mass fraction of carbon black was reported on silicone rubber/carbon black nanocomposites incorporated with $BaTiO_3$ [16] and claimed that the dielectric loss of the nanocomposites increased with increasing mass fraction of carbon black.

Recently, the measurement of dielectric strength, dielectric constant, volume and surface resistivity of EPDM/Silicone rubber nanocomposites incorporated of organically modified montmorillonite (OMMT) nano-clay are discussed[17] and it is concluded that the presence of OMMT in EPDM/Silicone rubber nanocomposites improves the dielectric strength and volume resistivity.

It is known that porous polymer results in materials with good flexibility, lower electrical conductivity and lower dielectric losses. To get the light-weighted MRNCs with a better MR effect, higher flexibility, lower electrical and dielectric properties; two sets of fabricated MRNCs based on silicon rubber matrix are selected to investigate the dielectric and electrical properties. The MRNCs samples are provided in five different weight percentage categories as 10%, 20%, 30 %, 40 % and 50 % of nano-sized CIPs .

The contribution of current study is concerning to the first time experimental study of porous MRNCs through space charge destribution, dielectric and electrical properties measurements and an analysis to reach the design point of flexible magneto-dielectric actuators and soft micro structures.The sensitive behavior of porous MRNCs during charging and discharging process is a start data-point for pressure sensitive devices in a wide range of industrial production. The sensivity achived from PEA analysis can be used to optimize the sensor based application ranges of porous MRNCs for pressure measurements.

2. Experimental Procedure

2.1Dielectric property measurement

Disk shaped specimens of 0.8 mm thickness were made of film-shaped MRNCs samples. The specimens were prepared using a sensitive electrical cutter. The surfaces of the test specimens were covered with copper thin films. The test samples were fixed between two electrodes and kept inside the sample holder. All the measurements were performed at a temperature of 27^0C and at a relative humidity of 20%. Besides that, the measurement environment is maintained constant so that they do not influence the results and each measurement cycle is repeated three times under the same condition to represent theaverage datavia diagrams.

The capacitance and dielectric loss measurements in this study were performed using a LCR HiTESTER (3532-50, HIOKI-JAPAN). The measurements were carried out in the frequency range of 100 Hz – 0.1 MHz.The dielectric constant (ε') was calculated from the capacitance using the following equation [18-21]

$$\varepsilon' = Ct/\varepsilon.A \quad (1)$$

whereε' is the dielectric constant of the material, ε_0 is the permittivity of air (8.85 X 10-12 F /m), C is the capacitance,A is the area of cross section of the sample, and t is the thickness of the sample.The loss tangent is estimatedusing theequation

$$Tan\delta = \frac{\varepsilon'}{\varepsilon''} \quad (2)$$

Whereε'' is referred as loss factor of material.

2.2 Electrical property measurement

For the DC volume resistivity measurements, a Keithley electrometer (Model 8009) was used and the readings were obtained under an applied voltage of 250 V DC. The test is repeated three times for each MRNCs sample and the reported figures represents the average values of repeated experiments.

Through measuring the sample resistivity (R) the volume resistivity can be determined as

$$\rho = \frac{R \times A}{t} \quad (3)$$

Where A is the cross-sectional area of the sample and t represents the sample thickness between the two electrodes.

The electrical conductivity of sample is defined by the available volume resistivity values as

$$\sigma = \frac{1}{\rho} = \frac{t}{R \times A} \quad (4)$$

2.3 Charge density measurements

The pulsed electro acoustic (PEA) study was conducted to assess the internal charge distribution [22-23] in the film-shaped MRNCs samples.The charge distribution in MRNCs samples was measured using a homemade DC sources (0-5kV) with a pulse voltage of 0-1kV and pulse width of 25 ns. The diameter of copper electrode was 20 mm. The thin aluminum sheet was located on two sides of 0.8 mm thickness MRNCs samples as an electrode. The silicone oil was applied as an acoustic coupling agent to provide a proper acoustic contact between the MRNCs sample electrode and measuring electrode.

All the PEA measurements were performed at lab room temperature (27^{0}C) and each measurement was repeated five times considering the same condition to achieve the average data for graph representation. The charge density can be calculated using relation [24],

$$\sigma = KV_m \tau S_\upsilon \quad (5)$$

Where, Vm is the height of the waveform peak, τ is the half-width, $S\upsilon$is the sound velocity and K is the sensitivity factor defined as,

$$K = \varepsilon_0 \varepsilon' V_{DC}/(dV_m \tau S_\upsilon) \quad (6)$$

Here, d is the thickness of the sample.

3. Results and Discussion

3.1 Dielectric property

The variations of dielectric constant and loss with respect to frequency for the porous MRNCs samples at four different CIPs concentrations are shown in figures 1 and 2 respectively. It can be seen from Figures that the dielectric loss and constant of porous MRNCs samples decrease with increasing frequency and keep the frequency dependent behaviour up to the end of the process.

The experimentalmeasurements of dielectric properties of porous MRNCs show that porous MRNCs have the lower dielectric loss and constants. Probably, this is due to the absence of a large number of charge carriers in the bulk of porous MRNCs which are free to migrate under an applied electric.Besides that the further addition of the CIPs into the nonporous silicone rubber increases the dielectric constant since the amount of interface increases in the composite.

The variations of dielectric constants in figure 1 show that there is an effect of the CIPs concentration percentages. In a CIPs filled porous MRNCs sample, with increasing the nano-sized CIPs, the porous MRNCs dielectric constant increases.

Experimentally, the dielectric properties of the MRNCs depend on the concentration level of CIPs and the interfacial properties of the MRNCs.

According to the experiments, the dynamic model of beam-shaped actuator's performance is directly influenced by the stiffness and dielectric constant of a silicone based nanocomposites as MRNCs. In terms of actuator strain induced by magnetic field, lower values of the dielectric constant are desirable. Accordingly, the dielectric constant should be minimized to give the desired performance of actuator. So, the pores will cause decrement of the dielectric constant of MRNCsAs examined experimentally, the decreased dielectric constant provides increased strain of the MRNCs actuator (i.e. Grippers).

It also should be mentioned that the morphology of the layered nano-sized CIPs determines the dielectric constant of MRNCs. When CIPs are homogeneously dispersed (exfoliated) in a silicone rubber matrix , the dielectric constant is decreased.

3.2 Electrical property

Figure 3 depicts the variation of volume resistivity against nano-sized CIPs loadings in porous MRNCs samples.From the figure, it is noticed that the volume resistivity decreases with increasing of CIPs loadings due to the interaction of nano-sized CIPs with silicone rubber matrix.It can be concluded from figure 3 that porous MRNCs show a higher value of higher resistivity as compared to the nonporous MRNCs filled with CIPs for the same filler loadings.

Basically, the motion of charge carriers contributing to the conductivity primarily occurs along the silicone rubber chains. A barrier to the charge transport in silicone rubbers (causing reduction in electrical conductivity) can occur due to defects, moisture content, inter-chain charge transport, interfaces related transport and porosity.

Probably, the presence of a large number of interfaces and silicone rubber chain entanglements inhibit the motion of charges in the nanocomposites, which in turn causes a reduction in the electrical conductivity (hence a lower tan delta value).

It also can be recorded that an increase in the weight percentage of nano-sized CIPs loadings causes increase in conductivity as well (refer totable 1).

3.3 Charge density

Figures 4-5 show typical PEA measurements profiles for porous MRNCs samples measured with the electrical field applied, after DC stresses of 1, 2, 3 and 4 kV/mm DC during 30 sec - 30 minutes.

Figure 4 shows the charge distribution for porous MRNCs samples during the charging process, whereas. Figure 5 illustrates the charge distribution for porous MRNCs samples during the discharging processes.

Regarding the charge process, the density charge in the central of porous MRNCs sample is much more uniform and hetero-charge is built up in front of the both electrodes. It is noticed that the charge levels in the porous MRNCs samples are considerably reduced.

In the porous MRNCs sample, immediately the voltage is applied, homo-charge is immediately injected into electrode, and moves into the bulk MRNCs sample toward the opposite electrode. The injected positive and negative charges are observed in the bulk of MRNCs sample with the peaks close to the electrodes.

It can be observed that the charge density's variation is visible in the surfaces of the porous MRNCs sample during the charge process while the charge density's variation isnotable in the middle part of sample thickness during the discharge process.

The observed behavior of porous MRNCs during charging and discharging process will promot the applications of porous MRNCs as for pressure sensitive devices in a wide range of industrial productions. The sensivity achived from PEA analysis can be used to design the sensor for pressure measurement based on porous MRNCs.

Table 1. Electrical conductivity of porous MRNCs samples

Samples	Conductivity (S/cm)
MRNCs 10 %	1.96481E-09
MRNCs 20%	2.60659E-09
MRNCs 30%	3.33995E-09
MRNCs 40%	4.15720E-09
MRNCs 50%	4.90156E-09

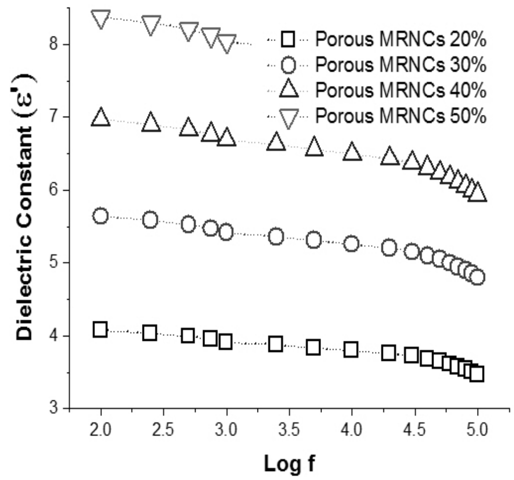

Fig.1. Dielectric constants for porous MRNCs samples

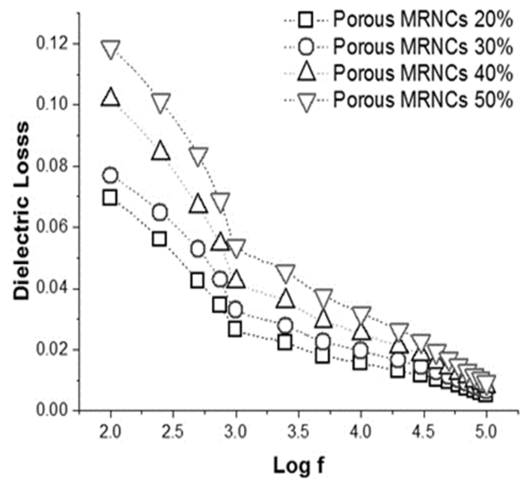

Fig.2.Dielectric loss for porous MRNCs samples

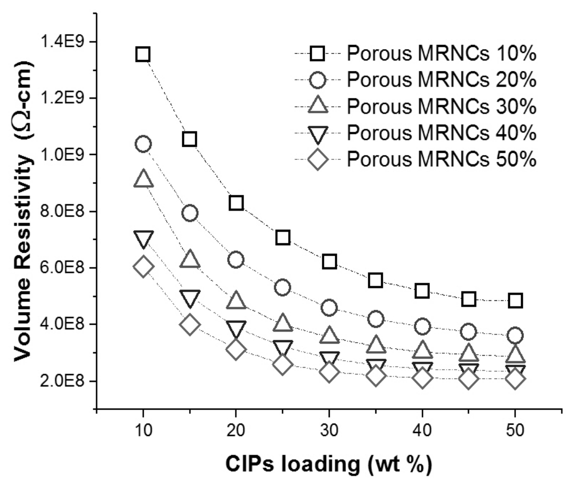

Fig.3.Volume resistivity profiles for porous MRNCs samples

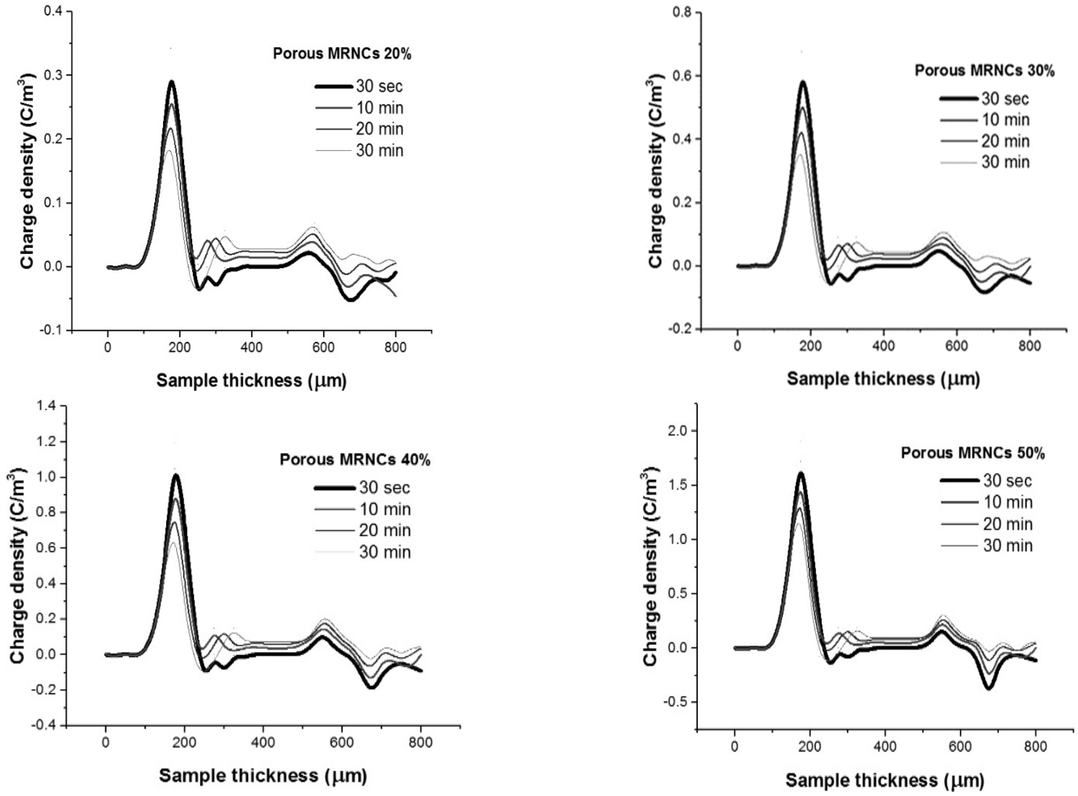

Fig.4.Charge density profiles for porous MRNCs samples during the charging process

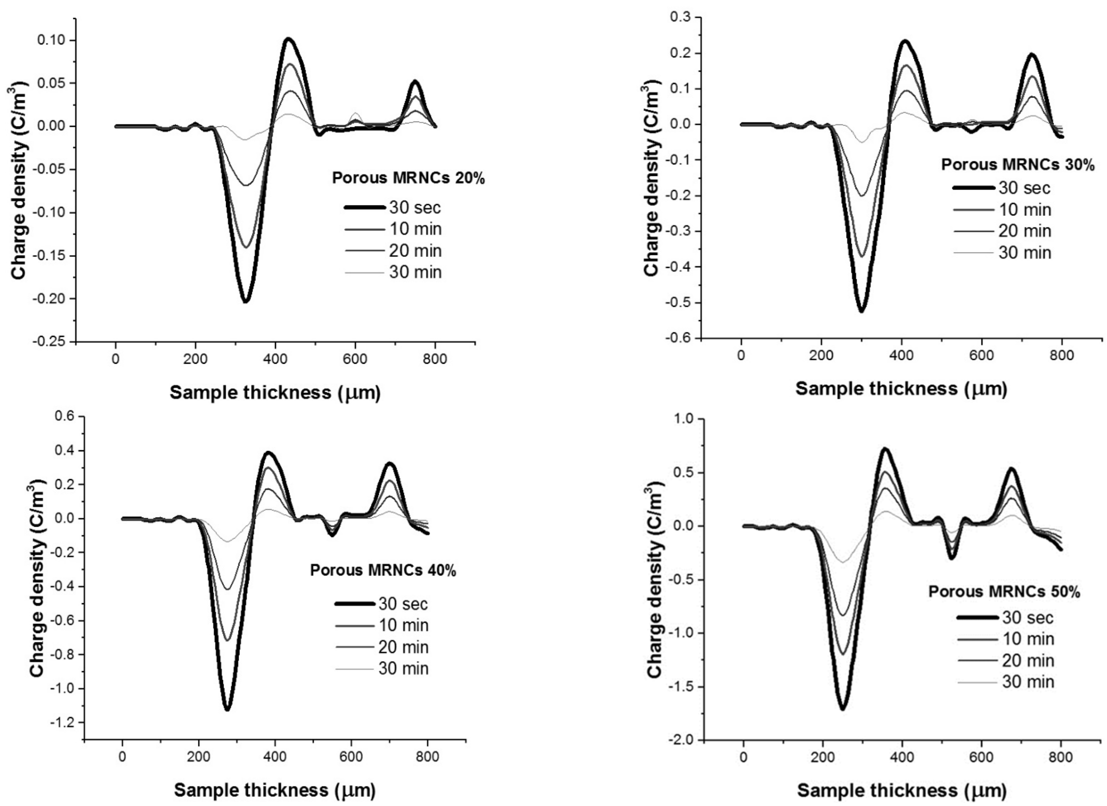

Fig.4.Charge density profiles for porous MRNCs samples during the charging process

4. Conclusions

In this study, the dielectric and electrical properties porous MRNCs based on silicone rubber were studied and analysed using experimental data. A series of desired dielectric and electrical property experiments were conducted to study the dielectric and electrical behaviours of porous MRNCs samples. It is observed that the porosity effects on dielectric and electrical properties of MRNCs samples. The MRNCs with nano-sized CIPs fillers display the advantageous dielectric behaviours at higher CIPs loadings. The dielectric constant and loss values in porous MRNCs are found to be lower than that of Nonporous MRNCs. For the porous MRNCs, The dielectric constants and dielectric losses are decreasing with increase of frequency and related to the CIPs loadings. The results of show that porosity play an interesting role to the electrical characteristics changes of film shaped MRNCs. It also can be summarized that the charge density's variation iscountable in the surfaces of the MRNCs sample during the charge process while the middle parts of sample thickness endure the higher variation of charge density during the discharge process.

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