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Interfacial effects on the electrical behavior of elastomer nanoparticulate composites

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ABSTRACT

Polymer nanocomposites exhibit unique effective properties that do not follow conventional effective media approaches. The nanoparticle-polymer interphase has been shown to strongly influence the nanocomposites behavior due to its significant volume when the particles are nano-sized, affording an opportunity to tune the dielectric response of the resulting nanocomposite. In this study, we investigate the effects of TiO₂ nanoparticles on the electrical properties and the charges distribution and transport in polydimethylsiloxane (PDMS) nanocomposites. Impedance spectroscopy shows suppression of interfacial Maxwell-Wagner-Sillars (MWS) polarization accompanied by a reduction in the low frequency dielectric permittivity and loss at high temperatures in the presence of the TiO₂ nanoparticles. Thermally stimulated discharge current measurements confirm that the suppression of the interfacial polarization relaxations happens by redistributing or depleting the charges through the composite and hindering their mobility, potentially resulting in lower electrical conduction and higher breakdown strength. Although the model materials investigated here are TiO₂ nanoparticles and Sylgard 184 PDMS, our findings can be extended to other nanoparticulate-filled elastomer composites to design lightweight dielectrics, actuators and sensors with improved capabilities.

Keywords: Interfacial effects, elastomer nanocomposites, electrical properties, thermally stimulated depolarization currents

1 INTRODUCTION

Polymer nanocomposites have been extensively studied due to their unique effective properties that do not follow conventional effective media approaches. These unique properties are governed by the nanoparticle-polymer interphase. As the particles size decreases, the interfacial surface area exponentially increases. Elastomer nanocomposites are of special interest due to their low dielectric losses, high mechanical strains and potential electromechanical coupling properties. In our previous study, we investigated the mechanical properties of polydimethylsiloxane (PDMS) at low loadings of TiO₂ nanoparticles.[1] That study showed that PDMS in the nanocomposites behaved differently than in the control PDMS films, and we observed three different PDMS “phases” (1) “occluded” PDMS, where the chains are constrained within the nanoparticle agglomerates, (2) “interfacial” PDMS, where the elastomer chains surround the agglomerates, forming a layer much stiffer than those chains further from the particles, and (3) “bulk” PDMS where chains have higher mobility. These three “phases” were shown to behave differently not only compared to the unfilled

PDMS, but also in the series of nanocomposites we observed. Similarly, a number of studies in the open literature have looked at the impact of fillers on the behavior of elastomers. Klonos et al. investigated the effects of silica, titania and other types of particles on the segmental mobility of a semi-crystalline PDMS.[2]–[6] Using thermal and dielectric techniques, they found that the presence of the nanoparticles suppressed the crystalline phase formation and reduced the mobility of the elastomer chains by constraining the chains surrounding the fillers.[3], [4] The interfacial effects on the elastomer chains mobility were also probed by studying silica-PDMS core-shell particles with two types of fumed silica having different porosities.[6] The experimental results showed that there was strong interaction between the PDMS chains and the silica and that the silica morphology affects the fraction of elastomer chains confined in the silica pores and therefore the overall composites properties and behavior.

As seen in the literature and our previous study, oxide nanoparticles affect the elastomer by mechanical confinement and there is evidence that electrical properties are impacted too. In this study, we investigate how TiO₂ nanoparticles impact the electrical performance of an elastomer, polydimethylsiloxane (PDMS), by employing dielectric spectroscopy and thermally stimulated depolarization current (TSDC) techniques. Two nanoparticles volume contents, 1 and 4vol%, are studied and compared to the control elastomer. Scanning electron microscopy is used to assess the dispersion of the particles and their interaction with the elastomer matrix.

2 MATERIALS AND EXPERIMENTS

2.1 Nanocomposite fabrication

Sylgard 184 was used as the elastomer in this study, and it was purchased from Dow Corning. It consists of two parts, base and hardner used at a weight ratio of 9:1. Anatase TiO₂ nanoparticles, 15 nm diameter, were acquired from NanoAmor. N-heptane, purchased from Sigma Aldrich, was used as a solvent, at a base to solvent ratio of 5:1 by weight, to reduce the solutions' viscosity for a better dispersion of the nanoparticles. The fabrication of the nanocomposites started by mixing the Sylgard 184 base with the solvent, then the nanoparticles were added. The solution was mixed using a mechanical stirrer at 250rpm for 6 hours. The hardner was then stirred into the solution before degassing it to remove any bubbles present. The films were then cast using a doctor blade, cured at 120°C for a total of 12 hours, where vacuum was applied during the last 6 hours. Two nanocomposites were fabricated at 1 and 4 volume percent (vol%). The control PDMS film was fabricated using the same solvent and hardner ratios as the composites and cured following the same temperature profile. The thickness of the resulting films (2 for each case for a total of 6 films) was uniform but varied from 230 to 270 μm for the different films.

2.2 Characterization

The nanoparticles dispersion was assessed using a FEI NovaSEM 630 scanning electron microscope where samples were freeze-fractured in liquid nitrogen, then 4 nm layer of iridium was sputtered on the cross-sections prior to imaging.

For the electrical measurements, a 2-cm diameter electrode was applied to both sides of each sample using gold leaves. Impedance spectroscopy was carried out using a Modulab XM MTS system at frequencies ranging from 10mHz to 100kHz. The temperature was varied from 25 to 150°C at a step of 25°C. Three samples were tested for each film. The dielectric permittivity, ϵ' , was calculated from the capacitance C as:

$$\epsilon' = \frac{C * A}{\epsilon_o * t} \quad Eq (1)$$

Where A and t are the electrode area and sample's thickness, respectively.

The electrical conductivity was extracted from the dielectric permittivity and loss using Eq (2):

$$\sigma = 2\pi f \epsilon_o \epsilon' \tan(\delta) \quad Eq(2)$$

Where f is the frequency. DC conductivity is extracted from the conductivity plateau at low frequencies.

Thermally stimulated depolarization current (TSDC) technique is a powerful technique to probe the material's polarization relaxations at frequencies typically equivalent to 100 μ Hz-10mHz. This technique works by applying a DC electric field to the sample at a temperature, T_p , higher than the polymer's glass transition temperature (T_g) for a time t . Then while the electric field is still applied, the temperature is quickly dropped to below T_p . Once the temperature is stabilized, the electric field is turned off and the temperature is increased at a selected rate, β , while the depolarization current is recorded. The most commonly used theory to analyze TSDC thermograms is the Bucci-Fieschi theory, that assumes a Debye single relaxation behavior.[7]–[9] Common dielectric materials investigated using TSDC have multiple relaxations, and in order to use the Bucci-Fieschi model and extract activation energies for the different polarizations, the depolarization current density as a function of temperature, $J(T)$, is decomposed into single peaks and each peak is fitted to Eq (3):

$$J(T) = \frac{P_o}{\tau_o} \exp\left(-\frac{E_a}{kT}\right) \exp\left[-\frac{1}{\beta\tau_o} \frac{kT^2}{E_a} \exp\left(-\frac{E_a}{kT}\right)\right] \quad Eq(3)$$

Where T is in Kelvin, P_o is the equilibrium polarization reached at the poling temperature, E_a is the activation energy, k is the Boltzmann constant, β is the heating rate, and τ_o is the relaxation time at infinite temperature, which is calculated using Eq (4):

$$\tau_o = \frac{kT_m^2}{\beta E_a} \exp\left(-\frac{E_a}{kT_m}\right) \quad Eq(4)$$

T_m is the temperature at which the current density reaches the maximum.

For this study, TSDC tests were performed using a computer-controlled home-built system consisting of a 4140B pA meter / DC voltage source, a Trek amplifier, a Keithly multimeter, an oven and a K-type thermometer. Two electric field magnitudes, 7.5 or 15MV/m, were used. The sample was first poled at 55°C for 5 minutes. Then the temperature was decreased to -140°C at a cooling rate of 15°C/min. After 2 minutes, the electric field was turned off, the sample was short-circuited for about 20 seconds to dissipate the stray charges, then the temperature was increased at a 7°C/min rate up to 100°C, and the discharged current was measured. Four samples were tested for each film to ensure measurements repeatability.

3 RESULTS AND DISCUSSIONS

3.1 Scanning electron microscopy

Figure 1 shows representative SEM images of the 1 and 4vol% nanocomposite cross-sections taken at low and high magnifications. The SEM images show that the particles are not individually dispersed, rather they formed agglomerations, with sizes ranging from tens to hundreds of nanometers, uniformly dispersed through the nanocomposites. It can be seen from the micrograph taken at higher magnifications that the agglomerations consist of nanoparticles bonded together by an elastomer layer. This observation was confirmed by transmission electron microscopy in our previous work.[1] These SEMs also show strong interaction between the particles and the elastomer matrix with the agglomerations being embedded in the matrix and no voids were detected during imaging.

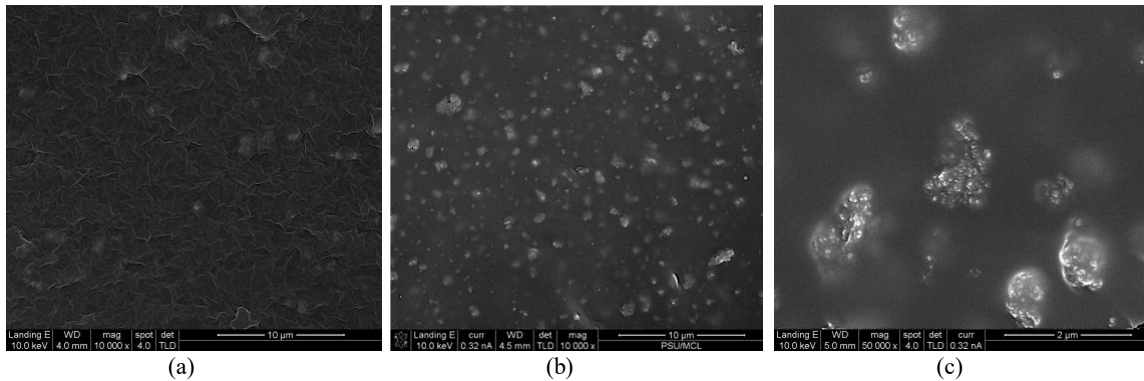


Figure 1. SEM images of cross-sections of (a) 1 and (b and c) 4vol% showing uniform dispersion with presence of agglomerations. b and c are taken at lower and higher magnifications, respectively.

3.2 Impedance spectroscopy

Figure 2 shows the dielectric permittivity, ϵ' , and loss, $\tan(\delta)$, as a function of frequency at 25 and 150°C for 1 and 4 vol% nanocomposites, compared to control PDMS. The data plotted is the average value of three measurements for each composition. At room temperature, the PDMS exhibited an increase in dielectric permittivity and loss at low frequencies, attributed to interfacial polarization. At 1 vol% TiO₂ loading, there was a slight increase in dielectric permittivity between 10Hz and 10mHz, however this increase is within the measurements' standard deviation. As the TiO₂ loading was increased to 4 vol%, the permittivity was higher than that of the control PDMS through the frequency range. The highest increase was present at the lower frequencies, where ϵ' increased from 2.8 for the control PDMS to 3.8 at 10mHz. Both 1 and 4 vol% nanocomposites had lower dielectric losses compared to the control PDMS below 100mHz, as shown in Figure 2(b). Figure 2(c) compares the dielectric permittivity of the control PDMS and the nanocomposites at 150°C. For the control PDMS, the dielectric permittivity remained around 2.4 between 100kHz and 10Hz, and then it increased as the frequency decreased reaching 85 at 10mHz. A linear dependence of ϵ' to $\log(f)$, f being the frequency, in the low frequency range indicates electrodes/sample interfacial polarizations.[10] To probe the dielectric permittivity dependence on $\log(f)$, the ϵ' values at 150°C, between 100mHz and 10mHz, were linearly fitted as $\epsilon' = a \cdot \log(f) + b$ for the control PDMS, 1 and 4vol% TiO₂ nanocomposites resulting and the fitting parameters are summarized in Table 1.

Table 1. Linear fitting parameters of low frequency dielectric permittivity ($\epsilon' = a \cdot \log(f) + b$) and fitting R²

	a	b	R ²
Control PDMS	-69.81	-57.14	0.99
1vol% TiO ₂	-0.03	2.45	0.86
4vol% TiO ₂	-0.67	2.57	0.94

The linear fitting results show a clear difference in the frequency dependence of the dielectric permittivity at low frequencies: strong dependence for the control PDMS, with a slope of about -70 compared to close to 0 for the nanocomposites, indicating electrode interfacial polarization, while the dependence was very limited in the nanocomposites cases. Additionally, the dielectric permittivity of the nanocomposites did not significantly change with the temperature increase. As shown in Figure 2(d), at 150°C the dielectric loss of the control PDMS increased as the frequency decreased, reaching a plateau around 10Hz, which is possibly the relaxation related to interfacial polarization. The range of frequencies at which the dielectric loss of the nanocomposites was lower than that of the control PDMS widened as the temperature was increased, from 100mHz-10mHz at room temperature to 100Hz-10mHz at 150°C. These

results confirm that MWS polarization strongly contributes to the dielectric response of the control PDMS, while it was suppressed by the addition of the nanoparticles. We surmise that the lower dielectric permittivity and loss in the nanocomposites are due to charge redistribution throughout the composite, likely due to presence of the nanoparticles and the interphase surrounding them, therefore reducing their contributions to the electrode interfacial polarization. The electrode interfacial polarization suppression with the addition of nanoparticles has been reported by Nelson et al. when they compared the electric behavior of an epoxy nanocomposite to the unfilled epoxy and the microcomposite.[11] Additionally, as shown in Figure 2(d), the relaxation behavior of PDMS was affected by the addition of the nanoparticles as the $\tan(\delta)$ peaks in the nanocomposites, 5kHz and 100Hz for 1 and 4vol% nanocomposites, respectively, while it is at 100mHz for the control PDMS.

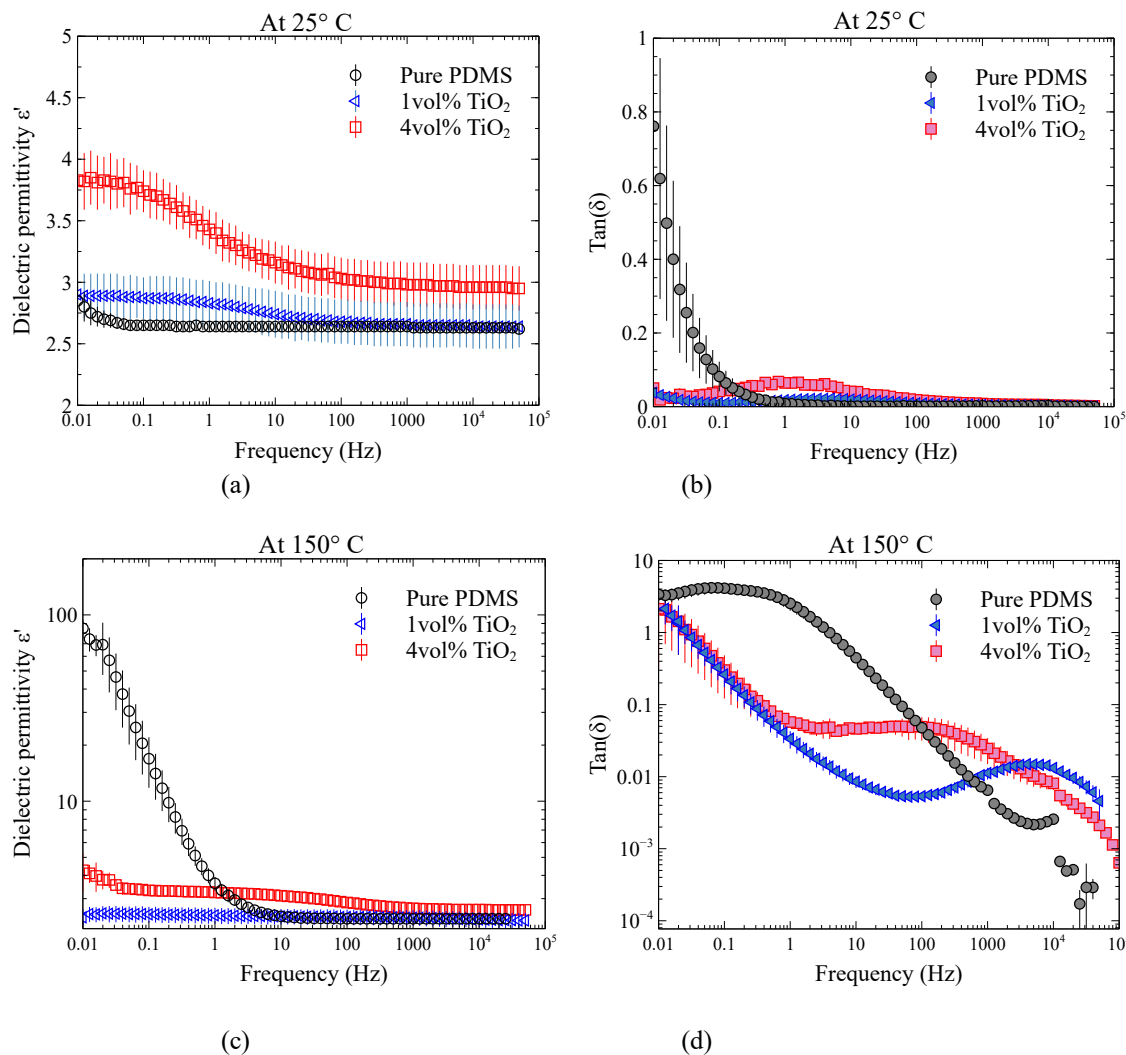


Figure 2. Dielectric permittivity and loss ($\tan(\delta)$) as a function of frequency at (a & b) 25°C and (c & d) 150°C. y-axes are in log scale in 150°C plots.

As discussed above, the addition of the nanoparticles to PDMS affected the dielectric behavior especially at high temperatures by suppressing the electrode interfacial polarization, therefore reducing the dielectric permittivity and loss at low frequencies. Electrical conductivity is another way to show the nanoparticles effects on the dielectric behavior, and as discussed in section 2.2., the DC conductivity was extracted from dielectric loss at low frequencies following

Eq(2). The extracted DC conductivity of the nanocomposites and the control PDMS are plotted as a function of temperature in Figure 3. The addition of TiO₂ nanoparticles led to 1 to 2 orders of magnitude decrease in DC conductivity as compared to the control PDMS in the temperature range tested. The higher drop occurs as the temperature increases, confirming that the particles and interphases are interfering with the electrical charges and limiting their mobility within the material, therefore reducing the electrical conductivity.

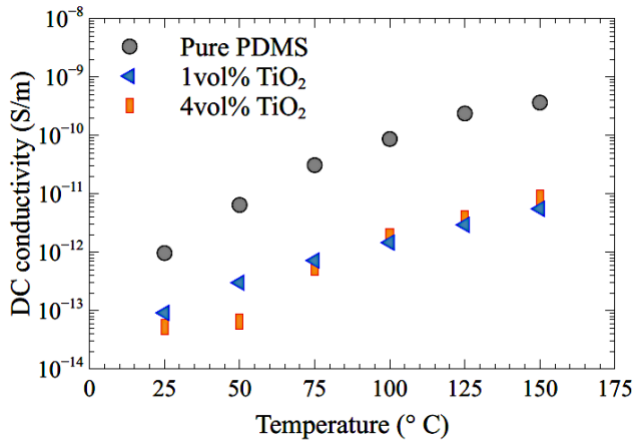


Figure 3. DC conductivity as a function of temperature of the nanocomposites is 1 to 2 orders of magnitude lower than that of the control PDMS.

3.3 Thermally stimulated depolarization currents

The impedance spectroscopy measurements showed that the nanoparticles addition leads to suppression of the electrode interfacial polarization in the elastomer nanocomposites. Thermally stimulated depolarization currents technique is a powerful tool to further probe the effects of the nanoparticles on the charge carriers' distribution and behavior in the elastomer. First, the effects of the poling electric field on control PDMS depolarizing currents are investigated after poling the samples at 7.5MV/m and 15MV/m and the TSDC thermograms are shown in Figure 4.

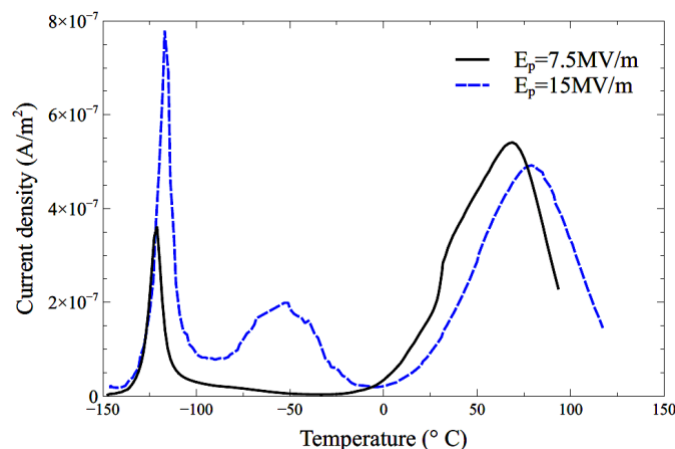


Figure 4. Effect of electric field magnitude on depolarization currents in PDMS films

Both samples exhibit a peak between -122 and -117°C, which correspond to the PDMS chains motion in the glass transition region.[12], [13] The intensity of the glass transition relaxation almost doubles when the poling field is doubled, which is anticipated considering the linear dependence of the T_g relaxation intensity on the poling field magnitude.[14] A peak around -50°C appears only at the higher magnitude (15 MV/m), and can be associated with electron injection during poling. This peak likely does not appear at the lower field because electron injection is limited

and shallow, reducing its relaxation intensity to below the resolution of TSDC.[14] A third peak at higher temperatures, namely between 25 and 100°C, is present in both spectra, this peak is assigned to MWS electrode interfacial polarization relaxation.[4], [6], [8] This peak shifts to higher temperatures with the increase of the poling field, which is in agreement with what was reported in the literature for interfacial polarization relaxation mechanisms.[15], [16]

To compare the nanocomposites depolarizing currents to those of the control PDMS, the poling electric field was set to 7.5MV/m to avoid potential breakdown events in the presence of nanoparticles. Representative TSDC thermograms for the control PDMS compared to 1 and 4 vol% TiO₂-PDMS samples are shown in Figure 5. As discussed in section 2.2, the peaks were fitted using equations (3) and (4), and the fitting parameters: activation energy E_a, equilibrium polarization reached at the poling temperature P₀, and relaxation time at infinite temperature τ_∞, are summarized in Table 2. The comparative analysis of the fitting parameters will be discussed later in this section. All the samples exhibit a peak around -122°C, P₁, which correspond to the PDMS chains motion in the glass transition region, indicating that the TiO₂ nanoparticles do not seem to affect the glass transition temperature, T_g, of the bulk PDMS. And as seen in Table 2, the glass transition relaxation parameters are not significantly affected by the nanoparticles' presence.

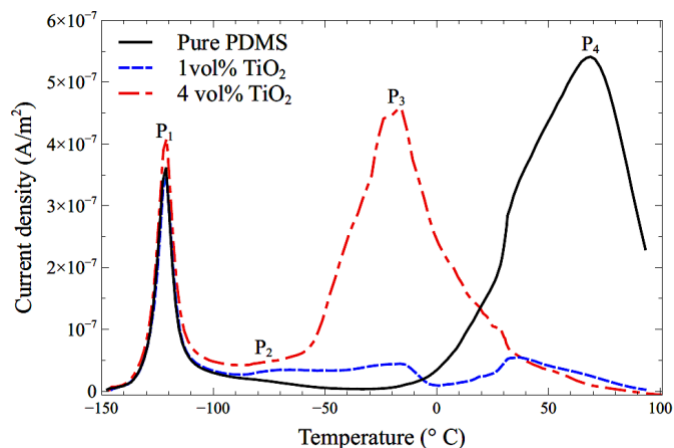


Figure 5. Thermally stimulated depolarization currents as a function of temperature for the control PDMS, 1 and 4vol% TiO₂-PDMS.

Table 2. Fitting results for TSDC relaxation peaks (glass transition, occluded, interfacial and MWS) for control PDMS, 1 and 4vol% TiO₂-PDMS

	Control PDMS			1vol% TiO ₂			4vol% TiO ₂		
	E _a (eV)	P ₀ (A.s/m ²)	τ (s)	E _a (eV)	P ₀ (A.s/m ²)	τ (s)	E _a (eV)	P ₀ (A.s/m ²)	τ (s)
P ₁	0.47	3.0*10 ⁻⁵	1.1*10 ⁻¹⁵	0.46	3.0*10 ⁻⁵	2.2*10 ⁻¹⁵	0.42	3.9*10 ⁻⁵	3.9*10 ⁻¹⁴
P ₂				0.15	1.9*10 ⁻⁵	2.7*10 ⁻²	0.15	3.6*10 ⁻⁵	5.9*10 ⁻²
P ₃				0.33	1.7*10 ⁻⁵	1.9*10 ⁻⁵	0.18	2.7*10 ⁻⁴	4.0*10 ⁻⁴
P ₄	0.34	3.4*10 ⁻⁴	1.1*10 ⁻³						

A number of changes with the addition of the nanoparticles are observed between -100 and 100°C. A small peak, P₂, between -80 and -50°C, appears in the nanocomposites. A new feature between -50 and 40°C occurs, P₃, which is more predominant and has a higher intensity with 4vol% loading compared to the 1 vol% nanocomposite. These new relaxation peaks, P₂ and P₃, a different charges distribution when the nanoparticles are incorporated in the elastomer and the intensities of these peaks are dependent on the nanoparticles content. In our previous study, we identified two interfacial regions in the presence of TiO₂ nanoparticles: (1) “occluded” PDMS where the PDMS chains are trapped within the TiO₂ agglomerations and (2) “interfacial” PDMS where PDMS chains are bonded and surrounded the TiO₂ agglomerations.[1] The volume fraction of the occluded PDMS in the nanocomposites is close to the particles volume fraction. The volume fraction of the interfacial PDMS is an order of magnitude higher than that of the particles.[1] The occluded PDMS chains

are constrained by the hydrogen bonds that form between the polymer's oxygens and the hydroxyl groups on the nanoparticles surface.[2] These bonds reduce the occluded PDMS mobility compared to the bulk PDMS, increasing the time scale of its relaxation, leading to a relaxation peak, P_2 , at higher temperatures.[3], [4] The occluded PDMS relaxations in both nanocomposites have similar activation energies E_a , equilibrium polarizations P_0 , and relaxation times τ_0 (see Table 1). At 4 vol% loading, the interfacial PDMS has a significant volume fraction in the composite and higher chain mobility compared to the 1 vol% nanocomposite due to the fact that there are more nanoparticles which would interfere with the elastomer chains crosslinking.[1] The presence of the interfacial PDMS in the nanocomposites results in the peak, P_3 , between -50 and 40°C. The peak P_3 has a higher intensity in the 4 vol% nanocomposite compared to the 1 vol% nanocomposite due to the increase in the phase's volume and chains mobility with the increase of the nanoparticles content. The peak P_3 fitting results show that the 4 vol% nanocomposite has an equilibrium polarization one order of magnitude higher than that of the 1 vol% nanocomposite, due to the higher intensity, and half the activation energy, relating to the higher chains mobility. The Maxwell-Wagner-Sillars electrode polarization relaxation, P_4 , that is present in the control PDMS is suppressed in the nanocomposites by charges carrier redistribution as surmised from the dielectric spectroscopy results. Overall, both nanocomposites have lower depolarized currents compared to the unfilled elastomer, with 1 vol% nanocomposite having the lowest depolarizing currents among the tested compositions.

4 CONCLUSIONS

In this study, the dielectric behavior of TiO₂-PDMS nanocomposites is investigated by impedance spectroscopy and thermally stimulated depolarization currents. Over a wide range of frequencies and temperatures, the dielectric permittivity of the nanocomposites has limited frequency and temperature dependence when compared with the unfilled PDMS. Additionally, the electrode/sample interfacial (MWS) polarization is suppressed and the low frequencies' dielectric permittivity and loss at high temperatures are reduced by the addition of the nanoparticles. Thermally stimulated depolarization currents measurements confirm the suppression of MWS interfacial polarizations. A peak at a temperature higher than the glass transition relaxation and an additional peak between -50 and 40°C appear in the nanocomposites TSDC thermograms. These two relaxations can be associated with the presence of interphases, namely occluded and interfacial PDMS. The interfacial PDMS relaxation has a significantly higher intensity in the 4vol% nanocomposite and lower activation energy compared to the nanocomposite with the lower loading. Additionally, the 1 vol% nanocomposite has the lowest depolarizing currents compared to the 4vol% nanocomposite and the control PDMS. These observations indicate that the particles content has a strong effect on the elastomer chains mobility, charges distributions, depletion and polarizations relaxations intensity. These findings based on the model materials investigated in this study, TiO₂ nanoparticles and Sylgard 184 PDMS, can be applied to other nanoparticulate-filled elastomer composites.

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