Dielectric characterization of thermoplastic natural rubber/strontium titanate composites at 10 Hz to 1 MHz

Jumiah Hassan[a], Deventharan a/l Rajaram[a], Mansor Hashim[a], Azizan Ahmad[b] and Zulkifly Abbas[a]
[a]*Department of Physics, Faculty of Science, Universiti Putra Malaysi, 43400 UPM Serdang, Selangor, Malaysia*
[b]*Institute of Microengineering and Nanoelectronics (IMEN), Universiti Kebangsaan Malaysia, 43600 Bangi, Selangor, Malaysia*

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The dielectric properties of Thermoplastic Natural Rubber (TPNR)/Strontium Titanate ($SrTiO_3$) composites was determined at room temperature, 27^{o}C using the Impedance Analyzer at frequencies 10 Hz to 1 MHz. TPNR is formed as the matrix while $SrTiO_3$ acts as the filler. Six samples were prepared at percentages of fillers from 0 to 50% at 10% intervals. The results indicate that the dielectric constant is almost independent of frequency from 100 Hz to 1 MHz, but the dielectric loss factor decreased with increasing frequency. This could probably be due to interfacial polarization and ionic losses. The dielectric properties are also affected by the composition of the filler, $SrTiO_3$.

I. INTRODUCTION

Natural rubber which has the ability to stick to itself and to other materials makes it easy to fabricate into rubber products. There are a large number of engineering applications in almost every industrial sector. These include components like radiator hose, windscreen wipers, tyres, engine mounts, suspension mounts, etc. Thermo plastic natural rubber is used as a matrix in the preparation of the samples, with strontium titanate as the filler, is a combination of natural rubber and linear low density poly-ethylene. Generally, polymers have low dielectric constant. However, dielectric ceramics have high dielectric constant but require high processing temperatures. Strontium based ceramics have been widely used to fabricate some electronic components such as grain boundary layer capacitors, varistors and sensors. Demand is increasing for thinner capacitor material to as low as 10 microns [1]. Reports have been made on polymer-ceramic composite materials such as dielectric materials in capacitors because ceramics have high dielectric performance and polymers are low cost materials and easily processed [2]. One important application for these polymer-ceramic materials is for usage in microelectronics packaging [3]. Dielectric permittivity or relative complex permittivity can be expressed as $\varepsilon^* = \varepsilon' - j\varepsilon''$ where ε' is the dielectric constant, which is the ability of the material to be polarized whereas the imaginary part, ε'' is the dielectric loss factor, which is a measure of energy dissipated usually in the form of heat.

II. MATERIAL AND METHODS

Three major steps are involved in the preparation of TPNR/$SrTiO_3$ composites which include filler, matrix and composite preparation. The conventional method was used to prepare $SrTiO_3$ filler where appropriate compositions of strontium carbonate and iron oxide powders were sintered at 1300^{o}C. The matrix TPNR was prepared by blending 40% natural rubber and 40% linear-low-density polyethylene using an internal mixer machine, Brabender Plasticorder PL2000. The final step is the preparation of TPNR/$SrTiO_3$ composites using HAAKE Plasticorder. TPNR was cut into small pieces and loaded into a heated mixer chamber stabilized at 150^{o}C. TPNR was heated for 2 minutes at rotor speed 50 revolution per minute and the required amount of ceramic was then added in small portions into the soft mixture and mixing was continued for another 13 minutes. Compression moulding was carried out by preheating the compound at 150^{o}C for 3 minutes and full pressing at the same temperature and pressure of 110 kg/cm^2 for 5 minutes using the hot and cold pressing machine. The compound was then cold pressed at 20^{o}C for 3 minutes with the same pressure to produce the composite sample sheets. The weight percentage of TPNR/$SrTiO_3$ composites is tabulated in Table I. The samples were then measured for its dielectric properties using the Impedance Analyzer (HP 4192A) at frequencies 10 Hz to 1 MHz at room temperature, 27^{o}C.

TABLE I. Weight percentage of TPNR/$SrTiO_3$ composites.

III. RESULTS AND DISCUSSION

Figs. 1 and 2 illustrate the dielectric constant, ε' and dielectric loss factor, ε'' of TPNR/$SrTiO_3$ composites with respect to frequency, at weight percentages 0–50%, respectively. The dielectric constant shows a rapid decrease until it reaches frequency stability above 100 Hz. The dielectric loss factor also decreases with increasing frequency where it becomes almost constant beyond 1 KHz with very small losses. The values of ε' and ε'' at selected frequencies are tabulated in Table II.

Samples	$BaTiO_3$ Weight Percentage (%)	Weight of TPNR (g)	Weight of $BaTiO_3$ (g)
TPNR/$BaTiO_3$ composites	0	44	0
	10	39.6	4.4
	20	35.2	8.8
	30	30.8	13.2
	40	26.4	17.6
	50	22	22

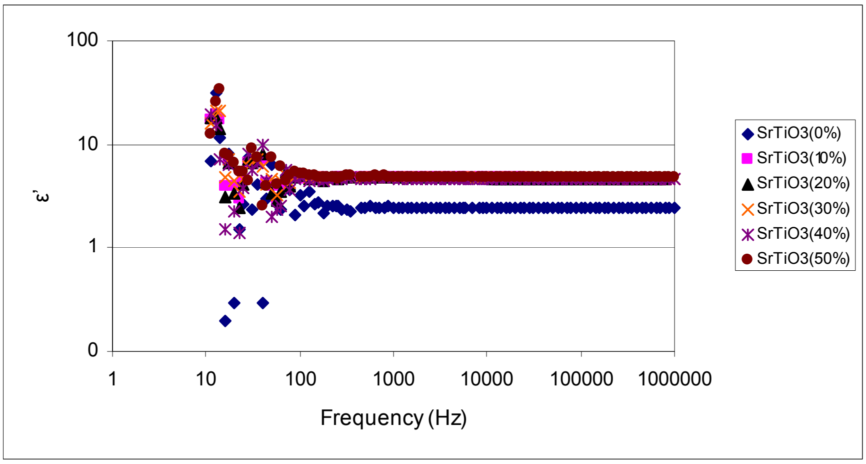

FIG. 1. Frequency dependence of the dielectric constant of TPNR/$SrTiO_3$ composites at various weight percentages of strontium titanate.

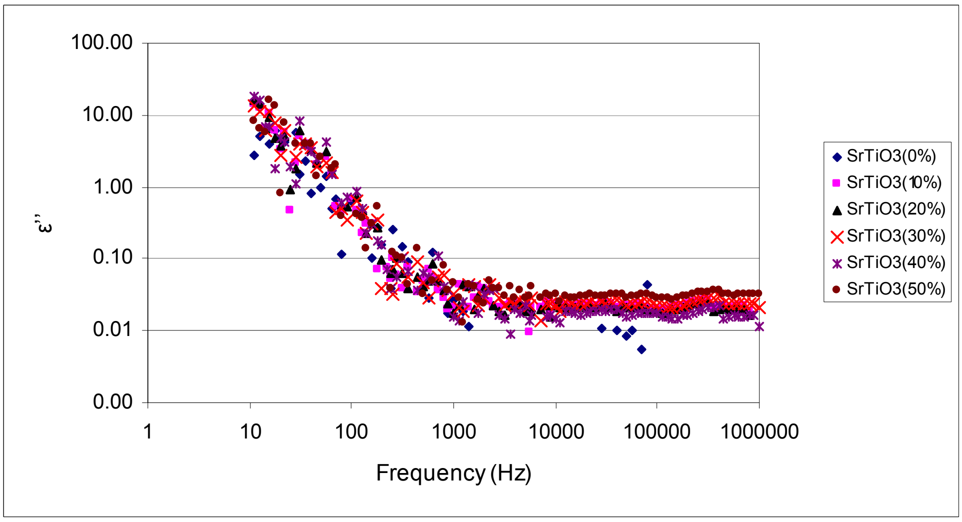

FIG. 2. Frequency dependence of the dielectric loss factor of TPNR/$SrTiO_3$ composites at various weight percentages of strontium titanate.

The rapid decrease of ε' and ε'' could be due to the dipolar relaxation in the samples. Dipolar polarization occurs at lower frequencies and it can greatly affect the capacitive and insulative properties of ceramics in the low frequency applications. ε' and ε'' also exhibits a low frequency dispersion. This could be attributed to the increase in interfacial polarization with the increase in grain boundaries and interfaces between the matrix and the filler. The impurities could also affect the dielectric properties.

Fig. 3 exhibits ε' and ε'' with respect to the weight percentage of strontium titanate. At 1 KHz and at room temperature 27^{o}C, ε' indicates a sudden increase with the addition of the filler, strontium titanate but is almost independent of the weight percentage of the filler. ε'' shows an almost negligible losses. Similar results were obtained for epoxy/alumina composite where the loss tangent showed a flat response, independent of the filler volume percentage [4].

IV. CONCLUSION

The dielectric properties of TPNR/$SrTiO_3$ composites in the low frequency region, 10 Hz to 100 MHz, exhibit a frequency independent dielectric constant above 100 Hz and a negligible loss factor above 1 KHz. At 1 KHz, the dielectric constant also is independent of the weight percentage of strontium titanate filler showing an almost constant relationship with almost zero losses. TPNR/$SrTiO_3$ composites could be considered to be a stable and lossless material in this region.

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TABLE II. Dielectric constant and dielectric loss factor of TPNR/$SrTiO_3$ composites at various weight percentages of strontium titanate at selected frequencies.

Frequency (Hz)	0%		10%		20%		30%		40%		50%	
	ε'	ε''	ε'	ε''	ε'	ε''	ε'	ε''	ε'	ε''	ε'	ε''
1×10^{2}	3.18	6.34×10^{-1}	5.00	6.88×10^{-1}	4.98	-	5.08	-	4.88	-	5.27	-
1×10^{3}	2.47	2.75×10^{-2}	4.78	2.04×10^{-2}	4.75	1.76×10^{-2}	4.80	3.17×10^{-2}	4.74	1.58×10^{-2}	4.86	4.75×10^{-2}
1×10^{4}	2.48	-	4.73	2.31×10^{-2}	4.72	2.17×10^{-2}	4.76	2.30×10^{-2}	4.68	1.43×10^{-2}	4.83	3.17×10^{-2}
1×10^{5}	2.47	-	4.69	1.98×10^{-2}	4.68	1.82×10^{-2}	4.71	2.30×10^{-2}	4.64	1.74×10^{-2}	4.78	2.85×10^{-2}

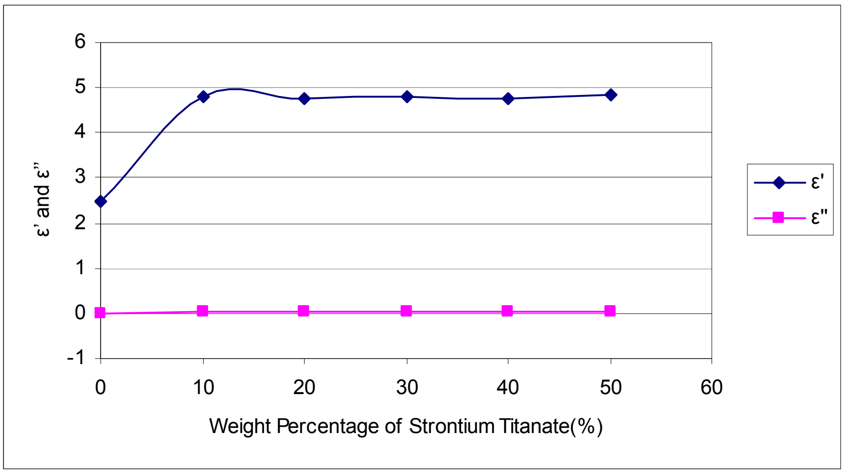

FIG. 3. Dielectric Constant and Dielectric Loss Factor with respect to Weight Percentage of Strontium Titanate at 1 KHz at 27^{o}C.

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