DIELECTRIC PROPERTIES OF POLYSTYRENE/ALUMINA COMPOSITES FOR MICROELECTRONIC DEVICES

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Abstract

The availability/lightness of electronic equipments in automotive, telecommunication and computer sectors have great importance in terms of the cost decrease and usability in high speed applications. Printed circuit boards (PCBs) are the main components of these systems and composed of two basic elements: substrate and passive electronic devices. Microelectronic segments having low dielectric permittivity (dielectric constant) and loss tangent minimize the capacitive coupling effects and decrease the signal attenuation. Ceramic particle filled polymer composites combine the desired dielectric properties and have been employed in electronic industry. In this study, 5%, 15% and 30% wt. alumina (Al_2O_3) loaded polystyrene (PS) composites were produced. The size of the PS granules was reduced below 100 microns by ball milling process. The Al_2O_3 and PS powders were mixed by vibratory disc mill and subjected to injection molding for manufacturing the circular specimens. The dielectric permittivity (real and imaginary parts) and loss tangent parameters of the composites were measured via dielectric analyzer up to 1 MHz at room temperature. Based on dielectric measurements, there was no pronounced difference among the samples' real part permittivities along the frequency variation. As expected, the ceramic content increase led to the increase of permittivity and 30%Al_2O_3 wt. loaded structure showed the maximum values. This behavior was attributed to the effect of polarization which appears in heterogeneous media consisting of phases with different dielectric constant and conductivity.

1. Introduction

The development of information technology leads to remarkable increase for the usage of electronic components in telecommunication, automotive, defense and energy sectors. The printed circuit boards (PCBs) consist of substrate and passive electronic devices (capacitors, transistors, diodes and resistors) are widely employed in the applications mentioned above. The weight reduction of available electronic components and PCBs for the communication operations requiring high speed has been a very critical problem. Therefore the researchers focus on alternative hybrid systems instead of monolithic materials that possess superior electrical, thermal and mechanical properties.

In PCBs, low dielectric permittivity (dielectric constant-ε_r) is expected for the substrate component in order to reduce the signal propagation delay while the capacitor components generally possess higher dielectric constant to store energy. Dielectric materials are electrically insulated, yet susceptible to polarization in the existence of an electric field. This polarization phenomenon accounts for the ability

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of the dielectrics to increase the charge storing capability of capacitors, the efficiency of which is defined in terms of a dielectric constant [1-2].
The permittivity is a specific material property that expressed as the permittivity of the dielectric material to the permittivity of a vacuum and shown as in Eq. (1) and Eq. (2) [3].

$$\varepsilon_r = \frac{\varepsilon}{\varepsilon_o} \tag{1}$$

$$\varepsilon_r = \frac{C.d}{\varepsilon_o.A} \tag{2}$$

The parameter *"ε_0"* is the permittivity of vacuum, "*C*" is the symbol of capacitance, "*d*" represents the thickness and "*A*" is the surface area of the material. The dielectric constant can be also displayed in the complex form as in Eq. (3). In this formulation, ε^* represents the complex permittivity, ε' and ε'' describe the real part permittivity (dielectric constant) and imaginary part permittivity (dielectric loss), respectively.

$$\varepsilon^* = \varepsilon' - j\varepsilon'' \tag{3}$$

The dielectric loss (loss tangent) is a measure of energy loss in the dielectric during AC operation which is a material feature and resulted from distortional, dipolar, interfacial, and conduction loss [4]. This property is usually expressed as "$tan\delta$"and formulated as in Eq. (4). In the ideal case, it is desired to have $tan\delta$ values as close to 0 as possible [4-5]

$$tan\,\delta = \frac{\varepsilon''}{\varepsilon'} \tag{4}$$

In the literature, a few ceramics such as silicates and aluminates are extensively used for substrate and packaging applications due to their convenient dielectric, thermal and mechanical properties. However ceramic materials are brittle and requires high process temperatures. In order to overcome these difficulties, the polymers that exhibit flexibility, lower cost and easy machining features are integrated with ceramics. The most effective polymer used as the composite component is the polytetrafluoroethylene (PTFE) owing to its excellent dielectric properties such as low dielectric constant and extraordinary low tangent loss at various frequency range. In addition to that epoxy, polyethylene (PE), polymethylmethacrylate (PMMA) and polyetheretherketone (PEEK) are the other plastics used for composing polymer/ceramic hybrid systems [6-7]. Thomas et al. (2009) investigated the dielectric and thermomechanical features of $Sm_2Si_2O_7$ reinforced polyethylene (PE) and polystyrene (PS) composites. The effect of filler content (10, 20, 30, 40 and 50%) on the dielectric behavior of the composites was determined at 1 MHz and 8 GHz frequency. The ceramic content increase resulted in the increase of tangent loss and density of the composites at both frequencies. The permittivity values significantly decreased for the PS and PE based composites after 40% $Sm_2Si_2O_7$ powder introduction [8]. George et al. (2010) concentrated on the $Ca[(Li_{1/3}\,Nb_{2/3})_{0.8}Ti_{0.2}]O_{3-\delta}$ or CLNT filled composite structures. In that study, polyethylene and epoxy were used as the matrix materials their dielectric characteristics at 9 GHz were revealed. Relative permittivity of PE/CLNT and epoxy/CLNT composites became higher with the enhancement of ceramic addition where the dielectric loss of the same composites showed diferent paths. The $tan\delta$ value of epoxy/CLNT composite firstly increased up to 10% filler content and then showed a regular decrease. However, PE/CLNT composite displayed a stable increase for the same parameter [9].Yoon et al. (2013) studied about the frequency dependent dielectric properties of $ZnNb_2O_6$/epoxy composites. The composites having more ceramic content exhibited higher permittivity but lower tangent loss [10]. The fiber reinforced polymer composites (FRPs) are also widely used as substrates in PCBs. Their commercial productions are very common and show very successfull dielectric properties [6-7].
The main aim of this work is to fabricate alternative polymer/ceramic composites which can be

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employed as PCB substrate material. The PS/Al_2O_3 composites' dielectric features were investigated with respect to Al_2O_3 content at 1 MHz frequency.

2. Experimental

The PS granules and Al_2O_3 ceramic powder used in this study were supplied from AMG-PLAST and CTS3000SG-ALMATIS, respectively. The granules of PS were ground via ball mill to homogeneously mix with Al_2O_3 filler. The schematic views of PS size reduction process is shown in Figure 1.

(a) **(b)** **(c)**

Figure 1. The size reduction process of PS granules (a) as-received PS granules, (b) during ball mill process and (c) powder form PS

When the PS particle size decreased below 100 microns, the plastic component and ceramic phase were blended by using vibratory disc mill at 700 rpm for 5 minutes. The Al_2O_3 contents in the composites were determined as 5%, 15% and 30% wt. and the mixtures were exposed to injection moulding process (Figure 2-a). The special aparattus was manufactured to produce circular specimens for dielectric characterization (Figure 2-b). Before molding, the composite blends were pre-heated at 80°C for 2 hours to eliminate the moisture effect. In addition to that the dies were left in the oven to easily remove the composites after injection. The fabricated PS/Al_2O_3 samples are displayed in Figure 2-c according to their ceramic content.

Before dielectrical measurements, the density values of the structures were also determined by means of Archimedes technique. The permittivity properties of PS/Al_2O_3 composite structures were determined via Novocontrol Alpha-N High Resolution Dielectric Analyzer. The real and imaginary components of permittivity and loss tangent values of composites were obtained by means of this device.As exhibited in Figure 3, there are two suspectible electrodes in sample cell of analyzer which do not contaminate the structures. Before analysis, the specimens were positioned between upper and lower electrodes and then compressed gently. It should be noted here that all the analysis were conducted at room temperature.

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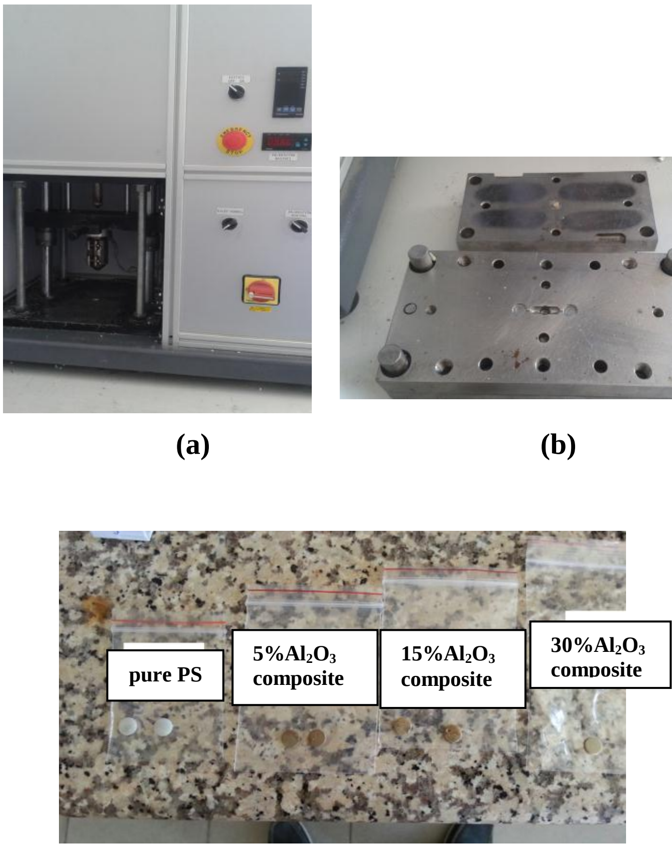

(c)

Figure 2. (a) Injection molding equipment, (b) dies used for producing circular specimens and (c) manufactured specimens with respect to their powder content.

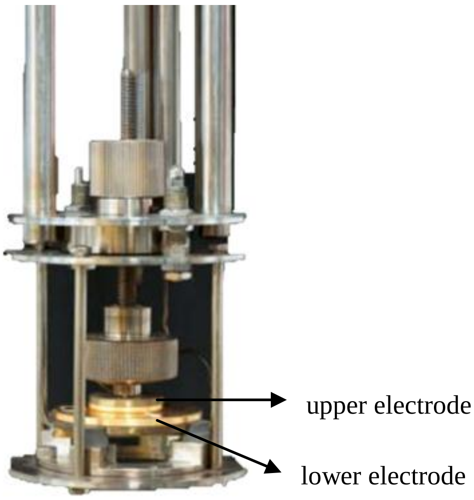

Figure 3. Illustration sample cell of Novocontrol Alpha-N High Resolution Dielectric Analyzer

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3. Results and Discussions

3.1. Density Measurements

The calculated densities of PS/Al_2O_3 composites and pure PS are shown in Table 1. As expected, the densities of the samples increase with the increase of alumina content. The maximum density in the table is calculated as 1.35 g/cm^3 for the 30% wt.Al_2O_3 composite where the pure PS powder has 1.06 g/cm^3 value.

Table 1. Density values of pure PS powder and PS/Al_2O_3 composites

Specimen Type	Density (g/cm^3)
Pure PS	1.06
5% Al_2O_3 composite	1.11
15% Al_2O_3 composite	1.20
30% Al_2O_3 composite	1.35

3.2. Dielectric Behavior Characterization

The thermoplastic PS used in this study shows an amorphous structure with low dielectric loss and permittivity. Alumina, which is extensively employed in various applications because of its high mechanic strength and chemical resistence, was selected as the filler component in this work. The Al_2O_3 has relatively low dielectric constant (9 at 1 MHz) and tangent loss (0.0001 at 100 MHz) as compared to perovskite ceramics such as $BaTiO_3$, $SrTiO_3$ and TiO_2 [1]. Therefore it is a very suitable candidate for the substrate constituent of PCBs.

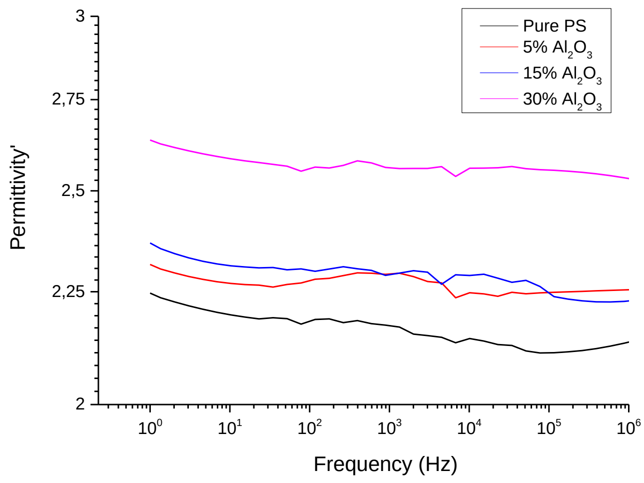

Figure 4. Variation of real part of permittivity as a function of frequency.

The real part permittivity (ε') versus frequency graph of pure PS and composites is given in Figure 4. These results were estimated from the measured data of capacitance, area and thickness of the samples. Based on Figure 4, there is no pronounced difference among the samples except 30% wt. Al_2O_3 composite at 1 Hz frequency which has 2.61 real part permittivity.The ε' value of pure PS is 2.25 at the same frequency and the composites including 5% and 15% wt.Al_2O_3 show slightly higher

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values. The real part permittivity parameter generally does not exhibit a significant variation with frequency increase both for pure PS and PS/Al_2O_3 composites.The minor ε' change among the specimens is attributed to the dipolar polarization. This suggests that pure PS and composites are not affected considerably by the variation of frequency.

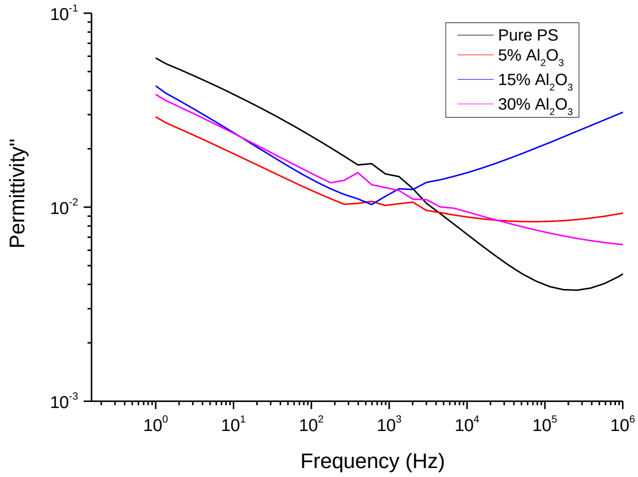

Figure 5. Variation of imaginary part of permittivity as a function of frequency.

Figure 5 depicts the variation of imaginary part of permittivity versus frequency for pure PS and Al_2O_3/PS composites at room temperature. It is observed that as the frequency decreases the ε'' values of all samples increase. Because of the dipolar orientation and interfacial polarization of surface charges between the electrode and PS/Al_2O_3 composite surface, the imaginary part of permittivity occurs [11]. Furthermore, the charge carriers undergo dipole polarization and the trend changes at particular mid frequency region of 1 kHz for all the composites.

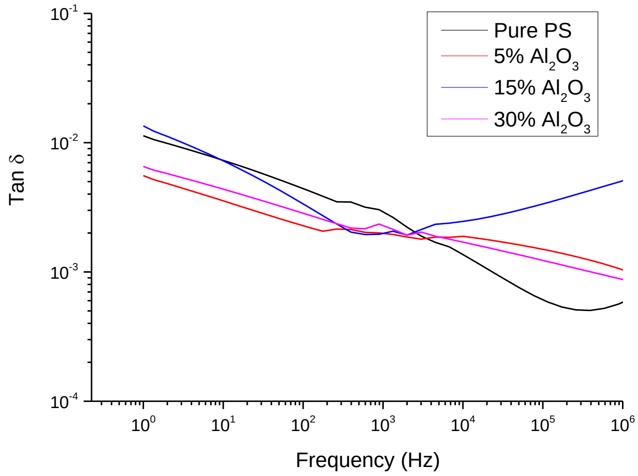

Figure 6. Variation of dielectric loss as a function of frequency

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The dielectric loss (*tanδ*) versus frequency graph is given in Figure 6. Before 100 Hz, the dielectric loss values of all structures including pure PS decrease with frequency increase. Between 100 Hz and 1 kHz frequency range, the tangent loss of composites' are almost same and show a stable value. However, the pure PS continues to exhibit a falling path through the rest frequencies. The composites including 5% and 30% wt. Al_2O_3 show a regular *tanδ* decrease at all frequency values while the 15% wt. Al_2O_3 sample follows an opposite way after 1 kHz. Generally, the dielectric loss increases with increasing ceramic content because of the interfacial polarization. Nevertheless, the composites with low loss ceramics may show a decreasing *tanδ* with increasing powder amount due to the ceramics' very low tangent loss [10]. In our study, the composite specimens correspond to this case except 15% wt. Al_2O_3 loaded one which may undergo to the non-activated charge motion such as tunneling [8].

4. Conclusions

In this study, the dielectric properties of pure PS and PS/Al_2O_3 composites were investigated. The PS granules were ground and decreased below 100 microns. The homogeneous blending of ceramic and polymer powders were performed by using a vibratory disc mill. The composite mixtures were subjected to injection molding and 5%, 15% and 30% wt.Al_2O_3 added composites were manufactured. The dielectric measurements were carried out between 1 Hz to1 MHz range by using a dielectric analyzer. Based on the graphical results, the real part permittivity (ε') of structures including pure PS exhibit a reduction trend at all frequencies although some minor deviations were observed. The 30% wt.Al_2O_3 showed the maximum ε' value among all the specimens. It is concluded that pure PS and composites were not affected significantly by the variation of frequency. For the imaginary part of the permittivity (ε''), it is observed that as the frequency decreased, the ε'' values of all samples increased. Both pure PS and the composites displayed a reduction in terms of dielectric loss (*tanδ*) up to 1 kHz where the 15% wt. Al_2O_3 loaded sample followed a different path after this frequency. The increase of tangent loss in 15% wt.Al_2O_3 composite after 1 kHz might be due to the non-activated charge motion.
The fabricated PS/Al_2O_3 composites may be a potential candidate for the PCB substrates instead of fiber reinforced polymer laminates. However the thermal and mechanical features of these composites' should be also revealed to accurately determine their performances.

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