Investigation of electric properties of flax reinforced polypropylene/strontium titanate composite for multilayer dielectric applications

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Abstract. Electrical properties such as dielectric constant, dissipation factor, capacitance, and conductivity are some of the properties that are required to be investigated when a novel composite is fabricated. This research aims to achieve a low dielectric constant (less than 4.2), lowered dissipation factor, lowered conductivity by fabricating a novel natural fiber reinforced polymer composite and investigating its properties. In this research, polypropylene with 14 wt% of strontium titanate, 8 wt% of treated flax fibers was fabricated using compression molding and tested for their dielectric and conductive properties. The capacitance and dielectric constant were significantly higher (43 percent) than that of pure PP or PP-flax, and a gradual decrease was observed as the frequency was increased. The dissipation factor and conductivity were found to be higher for pure PP. However, the full composite exhibited moderate conductivity showing that delays in charge losses could occur, making it an ideal composite for dielectric multilayer applications.

Keywords: Natural fiber-reinforced polymer composites, flax fibers, ceramic, dielectric constant, conductivity

1. Introduction

The application of natural fibers has become increasingly popular in many industries due to the technological advancements in many standards of manufacturing, tooling, and fabrication procedures. This aspect opens up numerous opportunities for making composites with tailored properties for selective applications and, as a result, using natural fibers in many applications to replace synthetic fiber (which is not sustainable) inclusion into a composite. Also, the addition of natural fibers in a composite increases renewability, recyclability and offers more biodegradable options [1].

In their raw form, natural fibers could exhibit physical properties that can be slightly inferior in terms of mechanical and electrical strength. This can be due to the presence of impurities and hydrophilic components possessed by the natural fibers. However, the right treatment method and curing procedures will alter the natural fibers such that hydrophobicity is introduced and susceptibility to
degradation is significantly lowered. Subsequently, the physical properties can be increased, and a superior product can be produced. Natural fibers are incorporated in the making of a composite because materials that can accept the fiber inclusion and act homogeneously as a composite. One such material is polymers, which are usually the right candidate for being a matrix where treated natural fibers can be added. Besides, polymers offer a lot of advantages in comparison to ceramic products. The main issue addressed by polymer composites includes mechanical properties, cost of manufacture, and sustainable feasibility [2][3].

Keeping that in mind, some of the primary properties that are assessed for any novel composite material are their electrical properties, which include dielectric strength, conductivity, and charge dissipation. These properties are investigated to find if the novel material can be applied in electrical and energy storage applications. This allows the material to be manufactured with pre-discovered physical constraints or conditions, which will yield a higher quality product with superior electrical properties. In this research, such a novel composite material was fabricated, and electrical properties such as dielectric permittivity, dissipation factor, conductivity, and capacitance if the composite was investigated. The results yielded from the composite are comprehensively explained, and application in multilayer dielectric structures has been discussed [4].

1.1 Theory of Dielectrics

A material is defined as a dielectric when it can be polarized by applying an external electric field. Most dielectric materials fall under insulators, where most dielectric materials are put to fair use in electrical insulation products. When these materials are subjected to an electric field, the polarization phenomenon occurs due to the presence of positive and negative charges aligned to the electric field's direction, otherwise called an electric dipole. With the company of different materials, the polarization phenomenon can vary, which causes storage of energy such as that of a capacitor. The applied electric field is found to be directly proportional to that of the charge density possessed by material and is equated to a constant called the dielectric permittivity, \( \varepsilon^* \).

\[
\varepsilon^* = \varepsilon' + \varepsilon''
\]  \hspace{1cm} (1)

\[
\delta = \tan^{-1} \frac{\varepsilon''}{\varepsilon'}
\]  \hspace{1cm} (2)

\[
\tan \delta = \frac{I_L}{I_C} = \frac{\varepsilon''}{\varepsilon'}
\]  \hspace{1cm} (3)

Equation 1 shown above represents dielectric permittivity, otherwise called the relative dielectric constant, where it splits into the real dielectric and complex dielectric constant. The complex dielectric constant is otherwise called dielectric loss, which occurs in the material. Dielectric losses happen in a separate phase with the application of electric fields and are affected by the electric field strengths. This loss phase attributes to charge loss, where charge dissipation occurs at a gradual period. Also, the magnitude through which this loss occurs in a material is on par with the dielectric constant, which is shown in equation 2 as dissipation factor, delta, \( \delta \) is the ratio of complex dielectric permittivity to the real dielectric permittivity.
1.2 Charge Loss in Dielectric Materials

In terms of electrical current, the dissipation factor can be represented as the ratio of loss current, \(I_L\), over the charging current, \(I_C\), with the current flown from an A.C power source (Refer Equation 3) [5][6]. The charge build-up and charge loss phases are adjusted by the polarization process in varying frequencies at a given time and investigated how a material can cope with these phenomena. Charge build-up allows lags between energy transfers and storage, allowing the material to act as an insulator with minor conduction levels amid charge losses. A material is currently defined as possessing good dielectric properties when it has very low dielectric losses at mid-range frequencies (100 kHz to 1 MHz) corresponding to the dielectric permittivity at the same frequencies. As for the conductivity occurring at a loss of current phases, the magnitude of conductivity in alternating current is defined as the product of phase-frequency and dielectric loss of the material [7][8].

\[
\sigma_{A.C} = \varepsilon_0 \omega \varepsilon''
\]  

(4)

Where \(\sigma_{A.C}\) is A.C conductivity, \(\varepsilon_0\) is the permittivity of free space (8.854187 x 10^{-12}) respectively. The composite fabricated in this research will be assessed for conductivity and all the properties mentioned above to understand the polarization processes and the results that are yielded from it [9].

2. Experimental data analysis

2.1. Fiber Preparation

Long stranded flax fibers were immersed in 5 wt % of sodium hydroxide solution for 24 hours until the fibers’ discoloration was observed. The fibers were later removed and dried in an oven for 24 hours at a temperature of 75 °C until they are dried thoroughly. The fiber was then chopped into 1 mm wide fine fibers, and a sieve was used to remove wider fibers and prepared for fabrication [10].

2.2. Sample Fabrication

The polymer matrix used in this research is polypropylene (PP) supplied by Sigma Aldrich as pellets with a shrinkage factor of 2.0. The ceramic compound in this research is strontium titanate powder supplied by Sigma Aldrich, where the particle sizing was found to 60 micrometers. Both polypropylene and strontium titanate are physically mixed and placed on an ASTM D150-98 (50 mm diameter and 5 mm thickness) dielectric mold. The pelletized PP and strontium titanate mixture is placed like a layer surrounding the mold, and the fibers were placed as layers on top of the PP-SrTiO\textsubscript{3} matrix layer. Compression molding was performed at a temperature of 190 °C with a pressure of 10 tons for a heating time of 45 minutes [11]. The samples were cooled in convectional air for 6 hours, and the samples were then removed from the mold plates. With these samples, the dielectric measurements were performed. Weights of 14 percent were used for strontium titanate, and flax fibers of 8 wt % were used to achieve the optimum fabrication method and yield good electrical properties simultaneously.

2.3. Dielectric Measurements

Dielectric Testing was performed using the Agilent 16541 B Dielectric Test Fixture made from Keysight Technologies\textsuperscript{TM}. The machine has an in-built micrometer, which allows the electrodes to be fixed at the 5 mm thickness mark and holds the sample in place. The device applies the desired voltage, which instantly provides the readings of capacitances and dissipation factor of the material under test (MUT) on the user interface. Samples were measured at 1 kHz to 2 MHz, respectively. The
capacitance values that were acquired was used to find the relative dielectric permittivity using the equation shown below:

\[ \varepsilon' = \frac{C_p}{C_0} \]  

(5)

\( C_p \) is the capacitance of the sample and \( C_0 \) is the capacitance of air that is occupied between the electrodes where \( C_p \) can be represented in another way.

\[ C_p = \frac{\varepsilon_0 A}{t} \]  

(6)

\[ \varepsilon' = \frac{C_p t}{\varepsilon_0 A} \]  

(7)

\( \varepsilon_0 \) is the permittivity of free space, \( t \) is the sample’s thickness between the electrodes, and \( A \) is the area of contact between the electrodes and the sample. The recorded capacitances were used to calculate the dielectric permittivity of the samples. For the dielectric loss, the dissipation factor is used. The dielectric loss, \( \varepsilon'' \) is the product of the dissipation factor and dielectric permittivity of the sample. Hence, the equation is written as such:

\[ \varepsilon'' = \varepsilon' \tan \delta \]  

(8)

\( \tan \delta \) in the dissipation factor. The dielectric loss is evaluated as the product of permittivity and dissipation factor [8]. A total of 40 samples were fabricated and tested. The samples included Pure PP, PP/Flax, PP/SrTiO\(_3\), PP/SrTiO\(_3\)/Flax [12].

3. Experimental results and Discussion

3.1. Capacitance and Dielectric Constant of the composite

The capacitances of pure polypropylene and polypropylene-flax composite are lower than those of other composite mixtures, as observed from Fig.1. The capacitance values were almost constant between the frequencies of 250 kHz and 2 MHz, where values between 3.1 and 4.2 pF. On the other hand, for the capacitance of polypropylene-strontium titanate composite, it was as high as 12.8 pF at a frequency of 1 kHz. It followed a gradual decrease with increasing frequencies. The capacitance decreased by 60.1 percent from 1 kHz to 2 MHz. A similar trend can be observed for the full composite with the addition of flax fibers and strontium titanate with PP, where the magnitude of capacitance had significantly increased to 14.01 pF, and the percentage loss was found to be 53.12 percent where the capacitance at 2 MHz was found to be a lowered 6.55 pF. This decrease in capacitance can be related to the dielectric constant of the material, which can be observed from Fig.2 [13][14].

The dielectric constants of the full composite and polypropylene-strontium titanate mixture show a decreasing trend from 4.2 to 1.6 and 3.2 to 1.55. This same trend can be seen in Fig.1 for the capacitances as well. This decrease could be since when frequency increases, the polarization process decreases due to increased vibration in the composite. The increased vibrations can distort the dipole
moments present between the molecules. The increase in oscillations will also remove the polarization between interfaces of polypropylene, strontium titanate, and flax fiber surfaces, which would be the primary reason for the decrease in both capacitance and dielectric constant. As for the lowered capacitances and dielectric constants for pure polypropylene and PP-Flax composites, their values are generally lower due to fewer interfaces between the composite, hence the lowered values for charge storing constants [15].

![Capacitance Vs Frequency of the composite](image1.png)

**Figure 1. Capacitance comparison for the composite and its components**

![Dielectric Constant Vs Frequency of the composite](image2.png)

**Figure 2. Dielectric constant comparison for the composite and its components**

3.2. Dissipation factor and Conductivity of the composite

The dissipation factor shows a very similar trend to that dielectric constant and capacitances, where they were higher at 1 kHz and lowered as the frequency increased. The dissipation factor increases slightly for pure PP and decreases slightly for PP with flax reinforcement. The increase in dissipation factor could be caused due to repeated charge loss, which introduced conductivity when the frequency was increased from 1 kHz to 2.0 MHz. The decrease in loss factor in PP-flax composite could be due
to the fact that flax fibers require time to arrange dipole moments where charge storage occurs, and charge loss decreases eventually when the frequency was increased. However, from the observed graph for dissipation factors of polypropylene-strontium titanate, the values increase until 0.30 and decrease till 0.24 when the frequency reaches 2 MHz. The decrease can be explained by adding a ceramic compound, which allows more charges to be stored in the ceramic before releasing it immediately [16]. As for the full composite's dissipation factor, the values are very low, from 0.12, and showed a massive increase of 167 percent to 0.32. This increase is due to the high dielectric constants, where more charges are allowed to be lost due to the current losing phase [17].

As for a general trend, the conductivity increases as the frequency increases. The investigation of A.C conductivity for pure polypropylene is the highest since the dipole moments can lose charge immediately due to lesser interfaces to hold charges. In all the composite mixtures, there is an approximate 120 percent increase in conductivity when frequency increased. PP-SrTiO₃-Flax achieved the second-highest conductivity. The lowest conductivity was achieved for PP-flax, which
could be why two insulating compounds surround the interfaces, which does not allow much charge conduction within one end of the composite to the other end [18]. The increase in conductivity for the full composite is caused by losing the current phase where the dissipation factor is higher, causing conduction to increase charge transfer or loss.

4. Conclusion

This study found that the dielectric constant is below the low-k (< 4.1) dielectric properties, and the capacitances for the full composite mixtures prove that it can store and hold charges just like a capacitor. The introduction of conductivity using flax and strontium titanate will also add that charge losses can be delayed according to the application it suits. The dissipation factor that was found was comparatively lower and allowed ionic conduction to take place. The role of polarization processes in this composite would be electronic, dipole, and interfacial polarization, which cause an increase in dielectric constant when ceramic is introduced to the composite. Interfacial polarization will also occur when there are two or more interfaces present, which is the presence of strontium titanate and homogeneous dispersion of flax fiber into the polymer matrix. When a composite such as this is applied for circuit boards and multilayer dielectric resonators, it could offer good electrical properties that have been rendered. In conclusion, with the inclusion of natural fiber and a ceramic component, this composite can be used in sustainable ways of manufacturing products.

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