Effect of Water Absorption on the Low-Frequency Dielectric Properties of Biodegradable PLA/Flax PCB Substrates

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Abstract— The dielectric properties of insulating materials are affected by moisture; hence, applying new PCB substrates requires knowledge of their dielectric behavior under different environmental conditions. This study investigated the dielectric loss of PLA/Flax biodegradable PCB boards in the 500 Hz…500 kHz (low frequency) range. The samples were measured under different environmental conditions, and the absorbed moisture content was determined by weight measurement. The results show that the dissipation factor of the samples increased with absorbed water. Since the measurements were executed at various temperatures, the loss factor increment of the samples is the combination of dipolar loss due to the increased number of water molecules and the increased conductivity of the samples.

Keywords— Biodegradable PCB, sustainable electronics, dielectric properties, water absorption, dissipation factor

I. Introduction

The economic progress of modern society is mainly based on communication and information technology solutions, requiring electronic and electrical equipment. Nevertheless, these technologies produce an increasing amount of e-waste, concerning environmental and health issues [1]. This increasing amount of e-waste leads to growing obsolete and unusable printed circuit boards (PCBs); the estimated portion of waste PCB is around 10% of the total e-waste [2]. Thanks to the intensive development of biodegradable polymers, applying these materials as substrates can significantly reduce the mass of waste PCB [3]. Nowadays, intensive research work is underway worldwide with several biodegradable materials to replace the classical glass fiber reinforced substrates such as silk protein, cellulose nanofibres (CNF), polylactic acid (PLA), flax, and poly (vinyl alcohol) (PVA), gelatin, and wood in minimizing PCB waste [4]. The PLA's electrical and mechanical properties are relatively close to those of thermoset resins, and practical application as a PCB substrate was experimentally demonstrated [3]. The performance as a substrate material of PLA can be improved by reinforcement, enabling compatibility with current subtractive PCB fabrication and classical surface mounting technologies (SMT) [5]. Nevertheless, the practical application requires adding several additives to the PLA matrix, e.g., for flame retardancy, affecting the electrical properties of the material [6]. These various additives make the polymer more complex, which means that the near frequency-independent permittivity and loss factor values measured on PLA will show a strong frequency dependence, especially at lower frequencies [7, 8].

This study investigates the dielectric properties of the PLA/flax substrate in the low-frequency range under various environmental parameters, changing the temperature and relative humidity.

II. Materials and Methods

Two groups (denoted as Group A and Group B) of samples containing three samples were prepared for the experiments. Both groups have the same core, comprising four layers of flax reinforcement and PLA as the matrices in the composite structure. All of the components were prepared with added flame retardants. Group A consists of a one-sided, 1.55 mm-thick board, which has a 35 µm Cu layer (HTE-ED-copper, electrodeposited, IPC-4562-compliant, from MSC Polymer, Germany) laminated on top with PLA foil acting as a bonding layer between the core and the conductor. Group B samples were prepared with an additional polypropylene (PP) layer to improve adhesion under the same initial conditions. The two board types (and the common cores) are the same as presented in the previous studies [5, 6, 8, 9].

The capacitance and dissipation factor of the samples were measured by a Wayne-Kerr 6430A impedance analyzer with a circular electrode system with a guarding ring. The guard ring eliminates stray capacitances and surface current, making the field more homogeneous inside the samples between the measuring electrodes, having a 10 cm^2 area. The measurement connection is shown in Fig. 1.

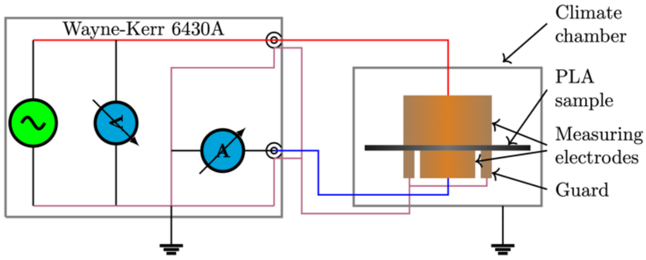

Fig. 1. The measurement setup

The measuring electrode system was placed in a Binder climate chamber to ensure constant temperature and humidity during the measurement. The climate chamber was connected to the ground to reduce electromagnetic interference. The capacitance and dissipation factor of samples were measured from 500 Hz to 500 kHz, applying a 5 V test voltage, and the real part of the relative permittivity of samples was calculated based on electrode area.

The dielectric parameters were measured at 30°C, 40°C, and 50°C temperatures under dry and moistened conditions. The test temperatures were below the PLA's glass transition temperature [10]. Before the experiments, the samples were dried at 50°C for 24 hours in the climate chamber. The relative humidity (RH) was 6.9%. The samples' weights were measured using a KERN PCB 100-3 scale, and then the dielectric measurements were executed under these conditions. These weight and dielectric measurement results were considered the reference values under dry conditions. Then, two pots filled with water were inserted into the climate chamber to increase the moisture content. After 24 h of conditioning, the samples' weight and dielectric parameters were measured again at 50°C and 7.9% RH. Then the temperature decreased to 40°C (18.0% RH) and 30°C (21.4% RH). The weights and dielectric parameters were measured at both temperatures after 24 h conditioning. The measurement arrangement in the laboratory can be seen in Fig. 2.

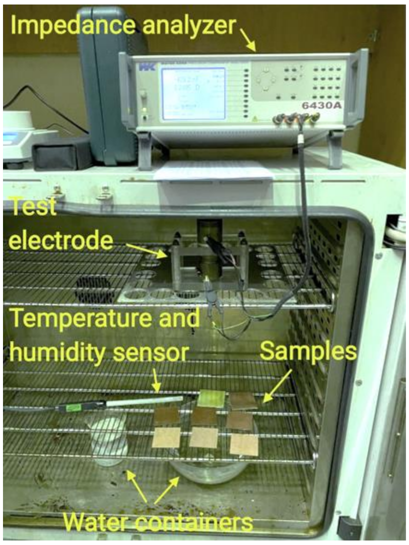

Fig. 2. The experimental setup in a climate chamber

III. Results

A. *Weigh Measurement*

Fig. 3 shows the samples' weight increment results under wet conditions, considering the 50°C dry measurement as the reference. The diagram shows the relative moisture content of the samples in wt.%, assuming the total weight increment is due to moisture absorption. The results show that Group B samples absorbed less water under the same conditions, probably due to the additional PP layer. The additional PP layer decreases the water absorption probably acting as a barrier from the circuit surface sides (top/bottom).

B. *Dielectric Measurements*

The real part of permittivity can be calculated from the capacitance results of the measurement using the electrode geometry. The results are depicted in Figs. 4 and 5. Fig. 4 shows the values of Group A samples, while Fig. 5 represents those of Group B samples.

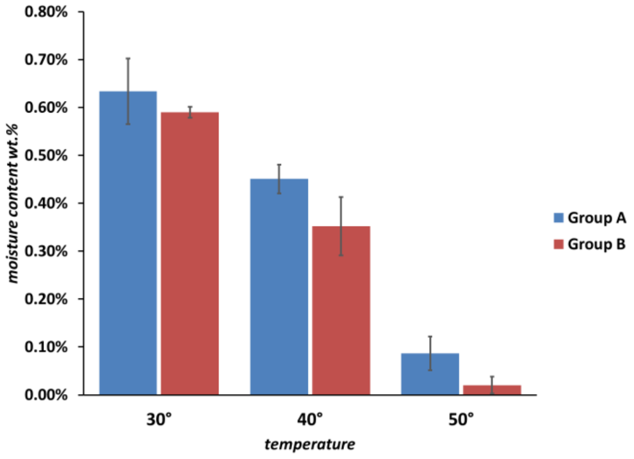

Fig. 3. Comparison of water absorption of Group A and B samples, wt.%

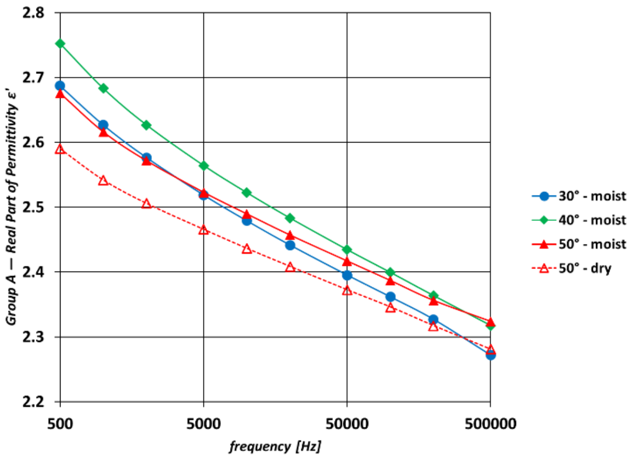

Fig. 4. The real part of the permittivity of Group A samples

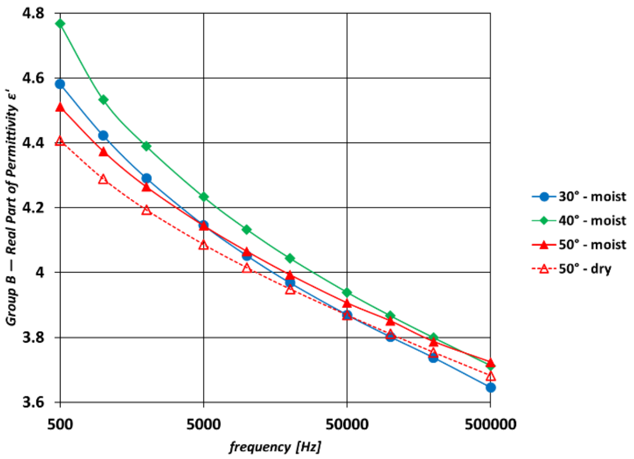

Fig. 5. The real part of the permittivity of Group A samples

The measurement results show that the real part of the permittivity decreases with frequency, which is a general behavior of dielectric materials. Comparing the ε' values of both groups, it can be observed that the real part of the permittivity of Group B samples is greater than that of Group A samples in the entire investigated frequency range. This observation can be explained by the additional PP layer, which fills acts as a barrier on the surface. Since the PP has a higher permittivity than the air (2.2 vs. ~1), the Group B samples have an elevated permittivity. An interesting behavior can be seen by observing the dependence of the real part of the permittivities on temperature and moisture content. At 50°C, the permittivity values of the moistened samples are greater than those of the dried samples. Suggesting that the absorbed water molecules increase the permittivity. Nevertheless, the permittivity at 500 kHz of the moistened samples at 30°C exhibited the lowest real part of permittivity values in both groups of samples. In contrast, the moistened samples at 50°C have the highest ε' values. At the lowest (500 Hz) frequency, the dry samples at 50°C have the lowest real part of permittivities, while the highest values were measured on moistened samples at 40°C. Figs. 6 and 7 show the dissipation factor measurement results of A and B samples, respectively.

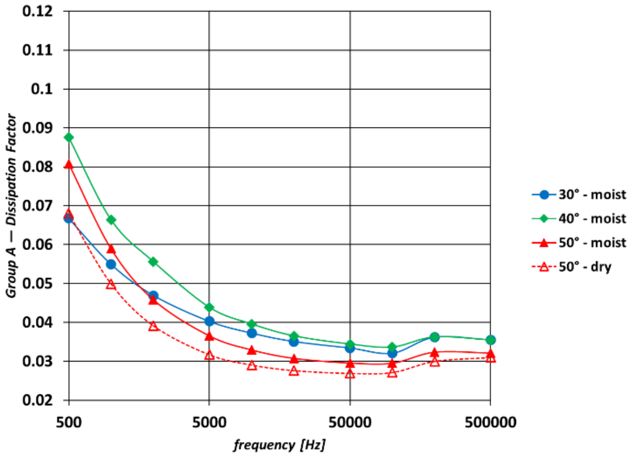

Fig. 6. Tan delta of Group A samples

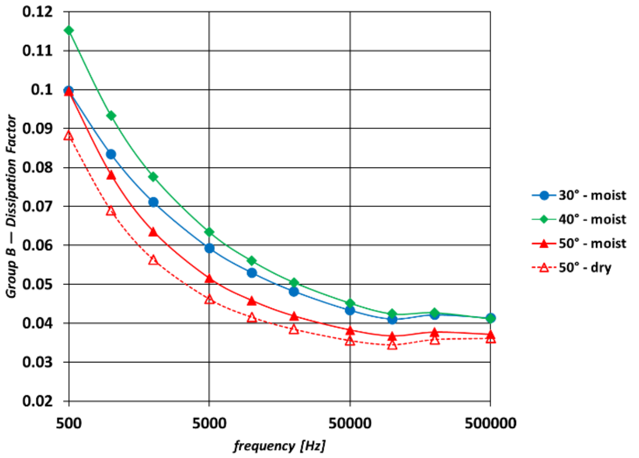

Fig. 7. Tan delta of Group B samples

When comparing the dissipation factor of samples, Group B samples have a higher dissipation factor than Group A samples in all cases. The PP layer can explain this since this component can cause additional loss.

IV. Discussion

The relative increment of ε' due to the moisture content was calculated from the 50°C measurement results to investigate the effect of moisture on the samples' permittivity. Since the capacitance was measured on dry and moistened samples at 50°C, only these results were compared (Fig. 8).

Fig. 8 shows that the permittivity of Group A samples increased more with moisture than that of Group B samples under the same conditions. This phenomenon is in accordance with the higher water absorbance of Group A samples (Fig. 3). Because the water molecules are polar ones, and if there is a greater number of these dipolar species in a unit volume of the material, it results in an elevated permittivity because of the increase in the dipole moment of the unit volume [11]. The relatively higher increment of permittivity at lower frequencies can be observed in both sample groups. This phenomenon can be due to the increased conductivity of the samples due to water content because the low-frequency range of polarization is the range of space charge polarization [12]. The increased conductivity can result from greater charge accumulation in the interfacial regions of the compounds. As shown in the figures, the temperature, humidity, and frequency-dependent behavior of the samples' ε' is significantly heterogeneous; hence, the relative change in the real part of permittivity compared to the results of the permittivity measurement of dry samples at 50°C can be calculated (Fig. 9).

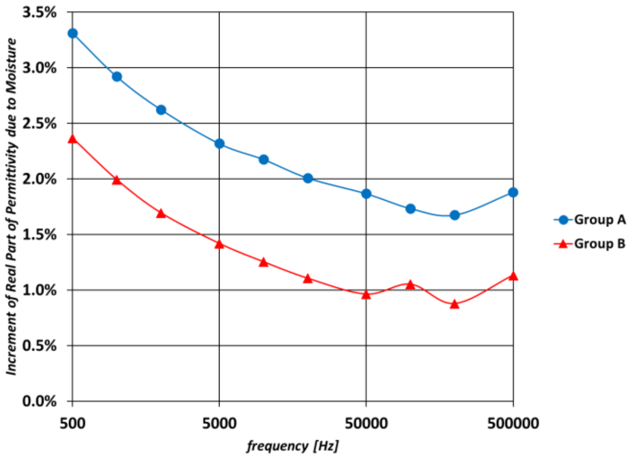

Fig. 8. ε' increment due to moisture absorption at 50°C

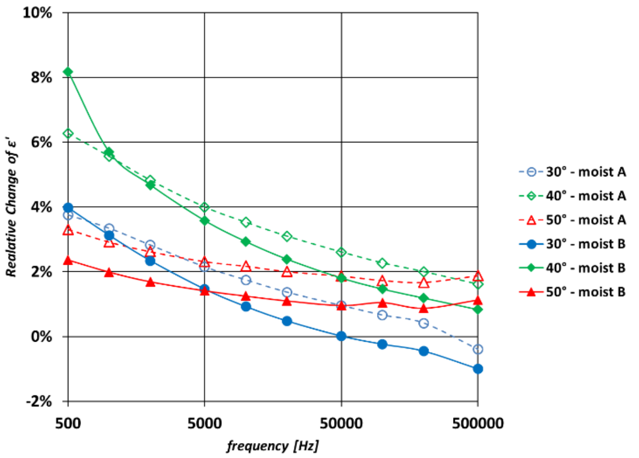

Fig. 9. Relative change in ε' of samples compared to the ε' values at 50°C

The results show that the relative change in the real part of permittivity is higher for Group A samples at higher frequencies. Still, the ε' change is greater for Group B samples at the lowest, 500 Hz frequency, except at 50°C. At 50°C the Group A samples have higher relative ε' shift in the entire investigated frequency range (see Fig. 8). Surprisingly, the real part of permittivity measured at 30°C is lower than that measured at 50°C on dry samples above around 300 kHz for Group A samples and above 50 kHz for Group B samples. It is an interesting phenomenon because, at 30°C, the moisture content is much higher. Therefore, more dipolar molecules are inside the samples, which should result in greater permittivity. Nevertheless, a portion of absorbed water can be bound to the polymers' groups, reducing the intensity of the β relaxation process [12, 13]. Comparing the real part of the permittivities of moistened samples measured at 30°C and 40°C to the values measured at 50°C, it can be observed that at lower temperatures, the permittivity increments are higher at lower frequencies. At the higher frequency ranges, the ε' increment is most significant at 50°C. In the case of 30°C measurements, the threshold frequency, where the ε' increment is greater at 50°C, is 5 kHz for both sample groups, while this frequency is around 300 kHz in the case of 40°C samples.

The increment of the dissipation factor can also be observed due to the absorbed moisture. Since the lowest dissipation factor data were measured on the dry samples at 50°C, the relative changes of dissipation factor values were calculated compared to values measured at 50°C on dry samples. The results are depicted in Fig. 10. It can be observed that the dissipation factor increment, similar to the permittivity increment, is greater for Group A than for Group B due to the higher water absorption, as the weight measurement showed.

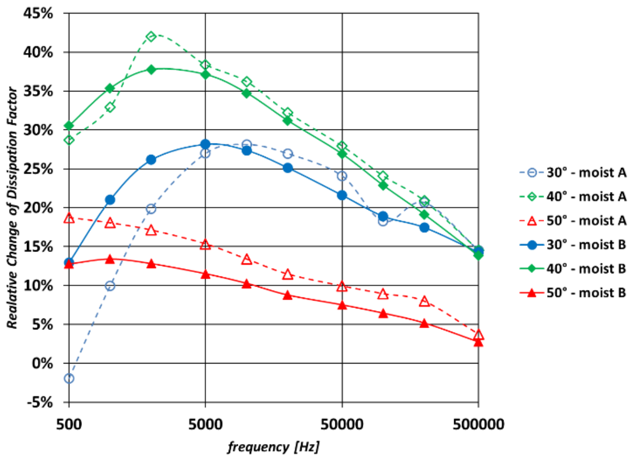

Fig. 10. Relative change in dissipation factor of samples compared to the dissipation factor values measured at 50°C

The figures show that the dissipation factor increased with the absorbed water in both sample groups. This is due to the dielectric loss of absorbed water, i.e., a higher amount of water results in a higher dielectric loss. However, in all cases, the Group B samples' loss factor was higher than that of Group A. Interestingly, the highest tan delta data was measured at 40°C, while the absorbed water was higher at 30°C. This phenomenon can result from two opposing processes: At lower temperatures, more moisture is absorbed, increasing the dielectric loss. Nevertheless, the dissipation factor and electrical conductivity are lower in insulating materials at lower temperatures, which decreases the loss factor. Probably, the second effect becomes more dominant at 30°C.

V. Conclusion

Two groups of PLA/Flax PCB substrate samples were prepared to investigate the effects of temperature and humidity on the dielectric parameters. One of them contained an additional PP layer. According to the weight measurement results, the PP layer reduced the water sorption of the samples, but it increased their permittivity and dissipation factor. The absorbed water also resulted in increased permittivity and dielectric loss. However, these dielectric parameters showed strong frequency and temperature dependence. The highest permittivity was at 40°C below 50 kHz, while the dissipation factor was the highest in the investigated frequency range. At 30°C, the permittivity and the dielectric loss were lower than at 40°C. This phenomenon is probably the result of two opposing effects: the absorbed moisture is higher at lower temperatures, but the loss factor and conductivity decrease with the temperature decrease. A deeper understanding of this behavior requires more investigation, which is a research path for the future.

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