Effect of Fiber Content and Temperature on the Dielectric Properties of Kenaf Fiber-filled Rigid Polyurethane Foam

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Kenaf fiber-filled polyurethane foams were prepared using the free rising method. The dielectric constants and the loss tangents of the composites were studied as functions of fiber content (0, 5, 10, and 15 parts per hundred of polyols by weight), temperature (from 30 to 200 °C), and electric field frequency (from 20 Hz to 2 MHz). The dielectric constant and the loss tangent increased with increasing fiber content. The dielectric constant was very high in the range of 10^1 to 10^2 Hz and varied little in the range of 10^3 to 10^6 Hz, but decreased rapidly above 10^6 Hz. The loss tangent decreased as the frequency increased. The effect of frequency on the loss tangent value was greater at frequencies below 10^2 Hz. Higher temperatures led to a higher dielectric constant and loss tangent. When the temperature was above approximately 120 °C, the loss tangent dramatically increased. The incorporation of kenaf fiber can improve the growth rate of the dielectric constant with increasing temperature. The dielectric constant and the loss tangent increased with increasing fiber content, indicating that both the dielectric capability and energy dissipation ability of the composites were improved.

Keywords: Kenaf fiber; Polyurethane foam; Composites; Dielectric Properties

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INTRODUCTION

Polyurethane foams (PUF) are typically produced through interaction between polyols and polyisocyanates *via* addition polymerization. Other additives, such as catalysts, surfactants, and foaming agents (Yuan and Shi 2009), are frequently used. PUF can be rigid, semi-rigid, or flexible, depending on the raw material used. The incorporation of fillers can result in improvement of certain properties of PUF composites, such as their mechanical, thermal, and acoustic properties. Natural fibers are relatively cheap and are considered renewable, sustainable resources. The densities of natural fibers are on the order of those of plastics and are only 40 to 50% of that of glass fibers (Bledzki *et al.* 2001). Plastics can therefore be reinforced or filled with fiber without there being a significant effect on the density of the resulting polymer composites. Further, the hydroxyl groups (-OH) on the surfaces of lignocellulosic fibers can interact with isocyanate groups, leading to excellent interfacial bonding between fibers and the polyurethane (Mosiewicki *et al.* 2009).

Wood flour has been used as a filler for PUF composites. Yuan and Shi (2009) developed wood-polyurethane hybrid composites containing up to 20 parts wood flour per hundred polyols by weight (php). Incorporation of wood flour improved the compressive properties of PUF but diminished its tensile and flexibility properties. The thermal stability of the composites was improved with the addition of wood powders. Racz *et al.* (2009) focused on the production and characterization of lightweight PU composites reinforced with pine wood flour and showed that the strength, modulus, and storage modulus of the composites increased as the filler content increased. Mosiewicki *et al.* (2009) used wood flour as a filler in rigid polyurethane and showed that the chemical reaction between wood flour and isocyanate strongly affected the composites' response to thermogravimetric tests. The compression modulus and yield strength of the PU composites decreased as the wood flour content increased. Aranguren *et al.* (2012) developed a tung oil-based polyurethane and wood flour composite and found that incorporating wood flour could influence the biodegradation of polyurethane foam composites.

Bast fiber has also been used as reinforcement in PUF composites. Bledzki *et al.* (2001) prepared reinforced polyurethane-based composites with woven flax and jute fabrics. The composites had an evenly distributed micro-void foam structure. Increasing the fiber content improved the shear modulus and impact strength. The woven flax fiber resulted in composites with better mechanical strength than that of the woven jute fiber composites. Kuranska *et al.* (2012; 2013) fabricated rigid polyurethane foams modified with flax and hemp fibers. Incorporating natural fibers improved the mechanical and thermal properties of the final products.

Cotton and bamboo have been used as reinforcements in polyurethane matrices to improve the sound absorption and thermal conductivity properties of the resulting composites (Büyükakinci *et al.* 2011). Tea-leaf fibers (Celebi and Kucuk 2012) have been used as reinforcements in different polyurethane matrices to improve the sound absorption properties of the composites. Adding tea-leaf fibers to rigid polyurethane foam yielded little contribution to the sound absorption properties of the composites, but gave a significant improvement in sound absorption if added to the soft foam.

While the mechanical, thermal, and acoustic properties of natural fiber-filled PUF composites have been researched, little is known about how incorporating natural fibers affects the dielectric properties of PUF composites. When a dielectric material is placed in an electric field, electrical charges do not flow through the material as they do in a conductor. Rather, the electrical charges only slightly shift from their average equilibrium positions, causing dielectric polarization. Due to dielectric polarization, positive charges shift toward the field and negative charges shift in the opposite direction. This creates an internal electric field that reduces the strength of the overall electric field within the dielectric material itself. If a dielectric material is composed of weakly-bonded molecules, those molecules not only become polarized, but also become re-oriented such that their axis of symmetry aligns with the direction of the electric field.

The study of dielectric properties is concerned with the storage and dissipation of electric and magnetic energy in materials (von Hippel 1954). Dielectric properties are important in materials used for insulation, packaging, and capacitors. Developing an efficient and light-weight dielectric material from sustainable resources, such as kenaf fiber, is quite attractive from both application and environmental point of view. The dielectric properties of a composite are related to the polarizability of its constituents (matrix and filler) and are functions of interfacial, dipole, atomic, and electronic polarizations (George *et al.* 2013). Polyurethane foam has been shown to be an applicable dielectric material (Argin and Karady 2008). The -OH groups in the lignocellulosic fiber contribute to the polarization. The atomic and electronic polarizations are instantaneous and do not affect the dielectric constants within the applied frequencies (Jose *et al.* 2010; George *et al.* 2013; Pethrick and Hayward 2002). The dipole orientation and interfacial polarizability of the kenaf fiber-filled PUF composites are the main contributors towards the net polarizability. The objective of this study was to investigate the dielectric properties of kenaf fiber-filled, rigid PUF composites as affected by differing fiber content and temperature.

EXPERIMENTAL

Materials

The raw materials used to prepare the foam were obtained from Fiber Glast Developments Corporation. The two parts, Part A (#24) and Part B (#25), were mixed in equal proportions, and a 32 kg/m^3 closed-cell foam was formed by free rising method. The kenaf fibers were obtained from Kengro Incorporation in Charleston, Mississippi. The length of the fibers was in the range of 1 to 3 mm. The fibers were dried to constant weight before being mixed into the PU matrix.

Composite Preparation

The PUF samples were prepared *via* the free rising method. The appropriate weight of Part B was added to a 200-mL disposable plastic cup. The kenaf fibers were added and mixed at 3000 rpm for 10 to 15 s with a mechanical stirrer. After the mixture was degassed for 120 s, Part A was added. Stirring was continued for the next 10 to 15 s at the same rotational speed. Foaming began less than 45 s after Parts A and B were mixed, and continued for a few minutes. The foam expanded to approximately 30 times its liquid volume, then cured. The foam rose and set at room temperature. The fiber contents were 0, 5, 10, and 15 php. Dielectric measurements were carried out using prepared samples of 3-mm thickness and 25-mm diameter. Three replicate samples were used for each measurement and the average value was reported.

Methods Dielectric Measurements

Dielectric measurements were carried out using a Dielectric thermal analyzer (DETA 2980, TA Instruments). The measurements were conducted over a temperature range from room temperature to 600 °C. The frequency interval ranged from 1 to 1000 kHz. During the dielectric analysis, the sample was placed between two parallel, metal electrodes. A sinusoidal voltage was applied to create an alternating electric field, which polarized the sample, with poles oscillating at the same frequency as the electric field with a phase angle shift denoted θ. The phase angle shift was measured by comparing the applied voltage to the measured current, which was separated into capacitive and conductive components. The measured capacitance and conductance were used to calculate the following properties:

1. The real part of permittivity (apparent permittivity), ε', which is proportional to the capacitance and measures the alignment of dipoles;
2. The imaginary part of permittivity (loss factor), ε'', which is proportional to the conductance and represents the energy required to align dipoles and move ions;
3. The loss tangent (or dissipation factor), $\tan\theta = \varepsilon''/\varepsilon'$; and
4. The dielectric constant, $\varepsilon = Cd/\varepsilon_0 A$, where C is the capacitance of the measured

sample in Farad, d is the sample thickness in meters, A is the cross-sectional area of the sample, and ε_0 is the permittivity of vacuum, 8.85×10^{-12} F/m.

The dielectric property measurements were conducted using isothermal runs at various fixed temperatures (30, 50, 100, 150, and 200 °C) and scanning frequencies (20 Hz to 2 MHz).

RESULTS AND DISCUSSION

Effect of Fiber Content on Dielectric Properties

Figure 1 shows that at a given frequency, the dielectric constant increased with increasing fiber content. The incorporation of -OH-rich kenaf fiber into the PUF increased the total number of polar groups inside the composites, leading to dipole orientation or orientational polarization. Interfacial polarization also effectively contributes to an increase in the dielectric constant (George *et al.* 2013).

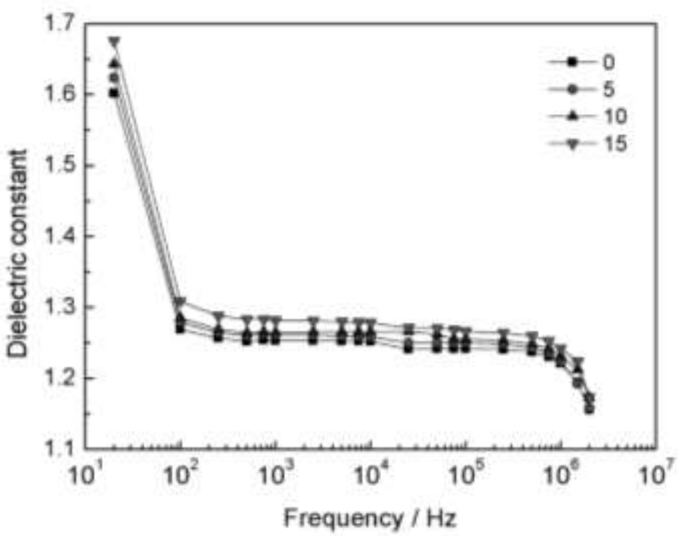

Fig. 1. Effect of fiber content on the dielectric constant of composites as a function of frequency at 100 °C (fiber content varied from 0 to 15 php)

Figure 1 also shows that the dielectric constants were higher at lower frequencies (roughly 10^1 to 10^2 Hz) than in those above 10^2 Hz. At low frequencies, the complete orientation of the dipoles could lead to a high dielectric constant. As the frequency was increased, molecular vibrations became more intense, so the complete orientation of dipoles did not take place. Therefore, the dielectric constant of the kenaf-filled, rigid PUF varied little in the range of 10^3 to 10^6 Hz. However, when the frequency was above 10^6 Hz, the intensity of molecular vibrations dramatically increased. The intense molecular vibration resulted in most of the dipoles being randomly arranged, such that a rapid reduction in orientational polarization occurred, causing a decrease in the overall dielectric constant. The same effect of the frequency on the dielectric constant was observed at temperatures of 30, 50, 150, and 200 °C. Comparing the dielectric constant of the neat PUF with that of the 15 php fiber-filled PUF, the average improvements in the dielectric constant were approximately 2.2%, 2.7%, 3.0%, 4.3%, and 6.7% at temperatures of 30, 50, 100, 150, and 200 °C, respectively.

Figure 2 shows that at a given frequency, the higher the fiber content, the higher the resulting loss tangent. The incorporation of -OH-rich kenaf fibers into the PUF matrix increased both the orientational polarization and the interfacial polarization, which resulted in an increase in the loss tangent value.

Figure 2 also illustrates that the loss tangent value decreased with increasing frequency. The effect of frequency on the loss tangent value was more dramatic at lower frequencies, especially below 10^2 Hz. This is because orientational polarization at low frequencies (below 10^2 Hz) increases, leading to a notably high loss tangent value. When the frequency was high, complete orientational polarization of polar groups did not take place and a lower loss tangent was obtained.

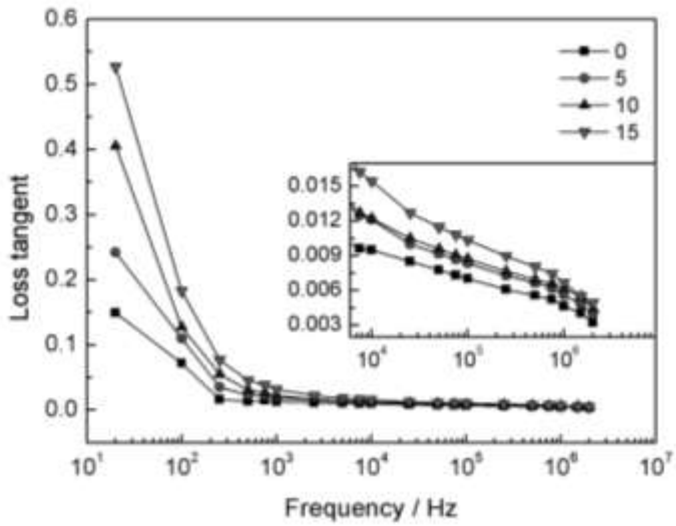

Fig. 2. Effect of fiber content on the loss tangent values of composites as a function of frequency at 150 °C (fiber content varied from 0 to 15 php)

Effect of Temperature on Dielectric Properties

As shown in Fig. 3, at all tested fiber contents, the dielectric constant increased as the temperature increased.

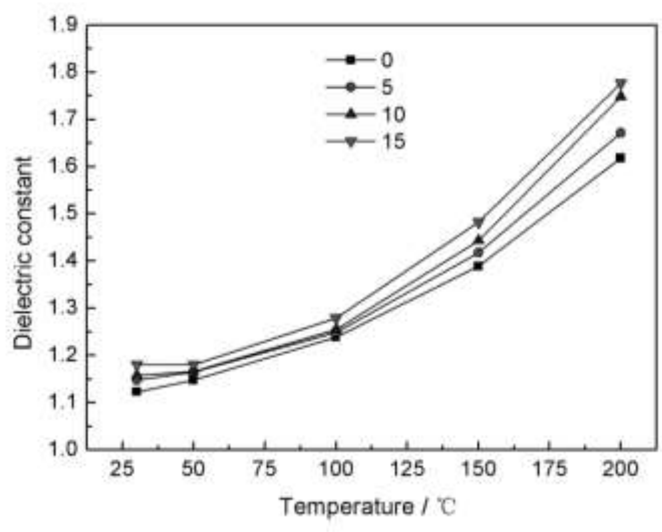

Fig. 3. Effect of temperature on the dielectric constant at a frequency of 100 Hz

The polarizability of a composite depends on the electronic, atomic, interfacial, and orientational polarizability, but only orientational polarizability depends directly on temperature (George *et al.* 2013). In natural fiber-filled composites, orientational and interfacial polarizability are the main contributors to the polarizability, and as such, the temperature plays an important role in determining the dielectric properties. When an electric field is applied to a polar material, the potential energies associated with individual dipoles change. Those dipoles aligned with the field have a lower potential energy than those aligned against the field. In other words, less energy is required for a dipole to align with the applied electric field (*i.e.*, orientational polarization) and more is required for it to align against the applied field (Zaman *et al.* 2010). At a temperature of 30 °C, only some molecules will have the required energy to align themselves with the applied field. When the temperature increases from 30 to 200 °C, the energy in the system is enough to increase the potential energy of the remaining orientations to that critical value where molecules get themselves aligned with the applied electric field. This results in a high orientational polarization and an increase in the dielectric constant.

The relationship between the dielectric constant and the temperature can be described by the relative variation ratio of the dielectric constant with increasing temperature. The relative variation ratio, TK, is defined as (Zhao 2006),

$$\mathrm{TK} = \frac{\varepsilon_t - \varepsilon_s}{\varepsilon_s (T - T_s)} \tag{1}$$

where ε_t is the dielectric constant at the end of measurement, ε_s is the dielectric constant at the beginning of measurement, T is the ending temperature, and T_s is the starting temperature. From 30 to 200 °C, the relative variation ratios were about 0.0026, 0.0027, 0.0030, and 0.0031 at fiber contents of 0, 5, 10, and 15 php, respectively. This illustrates that the addition of kenaf fiber can improve the growth rate of the dielectric constant with increasing temperature.

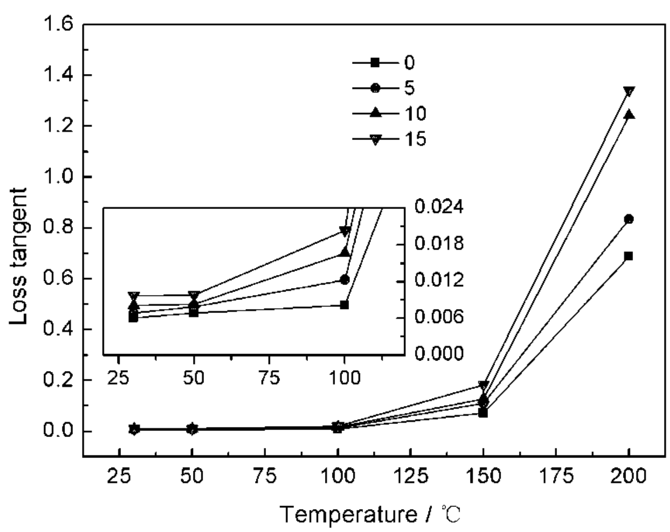

Fig. 4. Effect of temperature on the loss tangent values at a frequency of 100 Hz

The same trend (increasing temperature equates to higher loss tangent values) was observed in the case of the loss tangent value. The loss tangent value is also dependent on orientational polarization. Because the orientational polarization increases as a function of temperature, the loss tangent value changes accordingly. As can be seen in Fig. 4, at a given frequency, the loss tangent increases with increasing temperature. When the temperature is above approximately 120 °C, when the glass transition of PUF typically begins (Liang and Shi 2011), the loss tangent dramatically increases.

CONCLUSIONS

1. Fiber content had a positive impact on the dielectric constant and loss tangent of kenaf fiber-filled, rigid PUF. As the fiber content increased from 0 to 15 php, the dielectric constant and the loss tangent increased. Therefore, the dielectric capability and the energy dissipation ability of PUF can be improved by adding fiber.
2. The electric field frequency affected the dielectric constant and the loss tangent of kenaf fiber-filled, rigid PUF. The effects on the dielectric constant were different at different frequency ranges. The dielectric constant was higher at lower frequencies (roughly 10^1 to 10^2 Hz) than in those above 10^2 Hz, and varied little in the range of 10^3 to 10^6 Hz, but decreased rapidly above 10^6 Hz. The loss tangent decreased with increasing frequency. The effect of frequency on the loss tangent value was greater at frequencies below 10^2 Hz.
3. The dielectric constant and loss tangent of kenaf fiber-filled, rigid PUF increased with temperature from 30 to 200 °C. Comparing the dielectric constant of the neat PUF with that of the 15 php fiber-filled PUF, the average improvements in the dielectric constant were approximately 2.2%, 2.7%, 3.0%, 4.3%, and 6.7% at temperatures of 30, 50, 100, 150, and 200 °C, respectively.
4. Incorporating kenaf fiber can improve the growth rate of the dielectric constant as the temperature increases. From 30 to 200 °C, the relative variation ratios were about 0.0026, 0.0027, 0.0030, and 0.0031 at fiber contents of 0, 5, 10, and 15 php, respectively.

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