Dielectric Properties of Wood-Polymer Composites

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Composite materials of different composition were obtained by utilizing either a polyethylene or polypropylene matrix and a wood flour filling. In order to characterize these materials, a series of experiments were conducted to determine density, electric strength, complex dielectric constant components ε_r' and ε_r'' and dielectric loss factor tgδ for various electric field frequencies and at different temperatures. The results show that the dielectric properties are heavily influenced by the filler content with very high registered values in the case of low frequency electric fields and high temperatures. It can be ascertained that samples having a polypropylene matrix have superior electric properties than those having a polyethylene matrix. Finally, the opportunity for utilization within low voltage and industrial frequency electrical engineering applications of the tested composite materials is shown.

Keywords: wood-polymer composites, dielectric properties, dielectric spectroscopy

Composite materials are made up of two or more constituents with significantly different physical or/and chemical properties which remain separate and distinct on a macroscopic level within the finished structure. At least one of them serves as the matrix, whereas the other/others - the reinforcement. A wide variety of materials are employed both for the matrix (polymers, metals, ceramics etc.) and for the reinforcements (carbon, glass and organic fibers, silicon carbide, alumina, zirconia etc.) [1].

The various properties of composite materials are essentially the result of the chemical nature and physical characteristics of their constituents, as well as the size and shape of the particles which make up the filler material [3]. In the case of a fiber-shaped reinforcement, the composites are anisotropic (or, more exactly, ortotropic), whereas in the case of spherical particles, the composites are isotropic. Generally, composite materials imply achieving superior properties (thermal, mechanical, electrical etc.) than those of their components [2-4].

The matrix of polymer composites is a polymer thermoplastic or thermorigid, the most utilized being polyesters, vinyl esters, epoxy and phenol resins, polyimides, polyamides, polyethylenes etc. In the case of wood-polymer composites, the matrix usually consists of a thermoplastic polymer (polyethylene, polypropylene, polyvinyl chloride etc.) and the filler - an wood flour or a sawdust [2].

As both, the filling and the matrix can be obtained from industrial waste, wood-polymer composites are relatively cheap to produce and are advantageous with respect to effective recycling. In order to allow for proper utilization within the electrical engineering – particularly in the case of low voltage applications - their behaviour in electrical fields of various intensities and frequencies and at different temperatures must be known.

If an electric field of magnitude E_0 is applied to a dielectric at any time, t_0 and this field remains constant for $t \geq t_0$, the polarization $P(t)$ is the time domain response of the dielectric and is given by the equation:

$$P(t) = \varepsilon_0 \chi(t)\, 1(t) E_0, \qquad (1)$$

where:

$\chi(t)$ is the dielectric susceptivity,

ε_0 – the permittivity of vacuum;

$1(t)$ - the unit step for the electric field E_0.

$P(t)$ and $\chi(t)$ represent step response functions of the dielectric.

For a time dependent excitation $E(t)$, the time dependent polarization $P(t)$, is given (using Duhamel's Integral) by the following equation:

$$P(t) = \varepsilon_0 \chi_\infty E(t) + \varepsilon_0 \int_{-\infty}^{t} f(t-\tau) E(\tau)\, d\tau, \qquad (2)$$

where $f(t)$ is the so-called dielectric response function (monotonously decreasing function and inherent parameter of the dielectric being investigated).

Considering that an AC voltage of pulsation ω is applied to a condenser, that the polarization processes are instantaneous, and $\underline{F}(\omega)$ is the Fourier Transform of the dielectric response function $f(t)$ (the complex susceptivity $\chi(\omega) = \chi'(\omega) - j\chi''(\omega)$), it results:

$$\underline{F}(\omega) = \underline{\chi}(\omega) = \int_0^{\infty} f(t) \exp(-j\omega t)\, dt, \qquad (3)$$

$$\underline{\varepsilon}_r(\omega) = 1 + \underline{\chi}(\omega) = \varepsilon_r' - j\varepsilon_r'', \qquad (4)$$

$$\text{tg}\delta = \frac{\varepsilon_r'' + \dfrac{\sigma}{\varepsilon_0 \omega}}{\varepsilon_r'}, \qquad (5)$$

$$\varepsilon_r' = 1 + \int_0^{\infty} f(t) \cos \omega t\, dt, \qquad (6)$$

$$\varepsilon_r'' = 1 + \int_0^{\infty} f(t) \sin \omega t\, dt, \qquad (7)$$

where:

$\underline{\varepsilon}_r$ is the complex relative permitivity;

χ', ε_r, χ'' and ε_r''- real and imaginary parts of the quantities χ and, respectively $\underline{\varepsilon}_r$, tg$\delta$ - the loss factor;

σ - the electrical conductivity of the dielectric [4].

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Table 1
WOOD FLOUR CHARACTERISTICS

No.	Feature	Admissible value
1	Structure	Unfibrous
2	Color	Uniform
3	Smell	Specific
4	Usual impurities (abrasive particles)	Inadmissible
5	Ferrous impurities, mg/kg	Max. 0.2
6	Density, kg/dm^3	0.250+/-0.025
7	Humidity, %	Max. 10
8	Ash, %	Max. 1.5

Table 2
GENERAL CHARACTERISTICS OF MATRIX MATERIALS

No.	Feature	Material		
		PEID	PP	PVC
1	Density, g/cm^3	0.961-0.965	0.902-0.910	1.16-1.35
2	Breaking stress, MPa	Min. 14	28-37	48-70
3	Breaking elongation %	220-385	200-700	200-459
4	Vicat softening point, °C	130-140	121-160	66-93
5	Volume resistivity ρ_v, Ωm	Max. 1.6x10^{13}	Max. 5x10^{14}	10^{14}
6	Dielectric constant ε_r	Max. 2.5	2.2-2.6	2.8-7
7	Loss factor tgδ	Max. 2.5x10^{-4}	Max. 5x10^{-4}	Max. 2x10^{-2}
8	Electric strength, kV/mm	Max. 40	20-27	40-50
9	Water absorbtion (24 hour immersion,3mm thick film), %	0	0.01	0.15-0.75

Equation (3) represents the link between time and frequency domains: the complex susceptivity $\chi(\omega)$ and its real and imaginary parts can be converted to the dielectric response function $f(t)$ and vice versa [4].

This paper presents a part of the results obtained with various wood-polymer composite materials with regards to the dependence of electrical complex permittivity parts and loss factor on the electric field frequency and the temperature.

Experimental part

The experiments were performed on wood-polymer composites with a wood particle (wood flour), embedded within a thermoplastic polymer matrix (polyethylene, polypropylene). The wood flour (reinfort) was obtained by the grinding of the sawdust resulting from hardwood processing, with a medium particle size of 0.4 mm (STAS 6318/1-88) (table 1).

High-density polyethylene (HDPE) and polypropylene (PP) powders were utilized for the matrix (table 2).

The compounding process used in order to obtain the samples for the experiments involved the following operations:

- pre-treating the wood flour: mixing it with vinyl acetate in a mixer, at 170-180°C for 3 min;
- compounding process itself: adding the polymer into the mixer and continuing the process for 5 min for PE and 10 min for PP;
- removal of the mixer contents and compressing them: 3 mm thick plane boards are obtained;
- grinding the cooled boards in a BELMOS-PEEBLES equipment and obtaining pellets of sizes smaller than 0.5 mm;
- compressing the pellets at relatively-high temperature (using a SIEMPELKAMP press with 600 mm x 600 mm turntables, with cooling) and obtaining g = 2 - 5 mm thick boards. First, a higher temperature compression takes place (for the temperature T = 185-200 °C, pressure p = 75 atm, and duration t = 2 min), followed by a lower temperature (for T = 80 °C, p = 2-3 atm, and t = 4-5 min).

40x40 mm^2 samples were taken from these boards in order to determine their dielectric properties.

Several types of samples with various polymer and wood flour contents were tested (table 3).

Table 3
WOOD-POLYMER SAMPLE CONTENTS AND DENSITY

No.	Sample	Polymer	Polymer content, %	Wood flour content, %	Density, g/cm^3
1	PE0	PE	100	0	0.961-0.965
2	PE1	PE	70	30	0.990
3	PE2	PE	60	40	1.020
4	PE3	PE	50	50	1.040
5	PE4	PE	40	60	1.060
6	PE5	PE	30	70	1.130
7	PP0	PP	100	0	0.902-0.910
8	PP1	PP	70	30	1.030
9	PP2	PP	60	40	1.060
10	PP3	PP	50	50	1.100
11	PP4	PP	40	60	1.110
12	PP5	PP	30	70	1.150

Experiments

In order to characterize the wood-polymer composite materials, a series of tests concerning structure (electronic microscopy [2]), density, breaking stress and elongation [2], Vicat softening temperature, water absorption and electrical properties (permittivity, loss factor, electric strength) were made. The following refers strictly to density and dielectric properties tests.

The electric strength was determined using an ac automatic setup with voltages of U = 0…60 kV and frequency f = 50 Hz. The tests were done in air on 300x300 mm^2 samples.

The complex permittivity components ε'_r and ε''_r and loss factor tgδ were determined using a NOVOCONTROL dielectric spectrometer, for frequencies ranging from 5 .10^{-4} to 10^6 Hz, on 40x 40 mm^2 samples.

The measurements were done on groups of 5 samples when determining electric strength, and groups of 3 samples, when determining other dielectric properties.

Results and discussions

Density

The density of the composites increases with the content of wood flour (table 3). This is due to the higher density of wood flour (approx. 1.1g/cm^3) than that of polyethylene (0.961-0.965 g/cm^3) or that of polypropylene (0.902-0.910 g/cm^3). Nevertheless, although polypropylene has a lower density than polyethylene, for the same wood flour content, polypropylene-based composites have a higher density. This is probable due to the superior “compactness” of wood flour particles when utilizing the polypropylene matrix.

Electric strength

The presence of wood flour within the composite leads to a decrease in electric strength value E_{br} as compared to that of the pure polymer. Consequently, in the case of polyethylene matrix, E_{br} decreases from 40 kV/mm to 9.21 kV/mm as a result of mixing in 30 % wood flour and decreases even further, to 6.4 kV/mm, if the wood flour content is 40 %. Noteworthy is the fact that the electric strength decreases to a lesser extent in the case of polypropylene: E_{br} decreases from 20-25 kV/mm (for pure polypropylene) to 10 kV/mm by mixing in 30 % wood flour.

On the other hand, E_{br} increases when utilizing a polypropylene matrix. This effect should be a consequence of the lower number of pores associated with polypropylene composites (greater density values, table 3) and, therefore, of a decrease in partial discharge level and of an increase of inception voltage of partial discharges.

Electric permittivity and loss factor

In figures 1- 7 the variations of the real and imaginary components of the complex permittivity ε'_r and ε''_r and of the loss factor tgδ with the frequency of electric field f are presented.

Figure 1 presents the variations of the real component of the permittivity ε'_r with frequency (for f = 2x10^{-4} …10^5 Hz) and temperature (T = 30, 50, 70 and 90 °C). It can be seen that ε'_r values increase with the decrease of frequency regardless of the nature of the matrix or filler concentration. This is, firstly, a result of the re-alignment of the permanent dipoles of polar molecules contained within the wood-polymer composite (H_2O etc.) and of the polar parts of the matrix and polymer molecules (–OH, -CH_3, -C_2H_5, etc radicals). Secondly, reducing of the frequency facilitates ion migration and their binding to the matrix-filler discontinuity interfaces. Consequently, the inhomogeneous polarization is intensified, which leads to an increase in the value of the real component of complex permittivity [5].

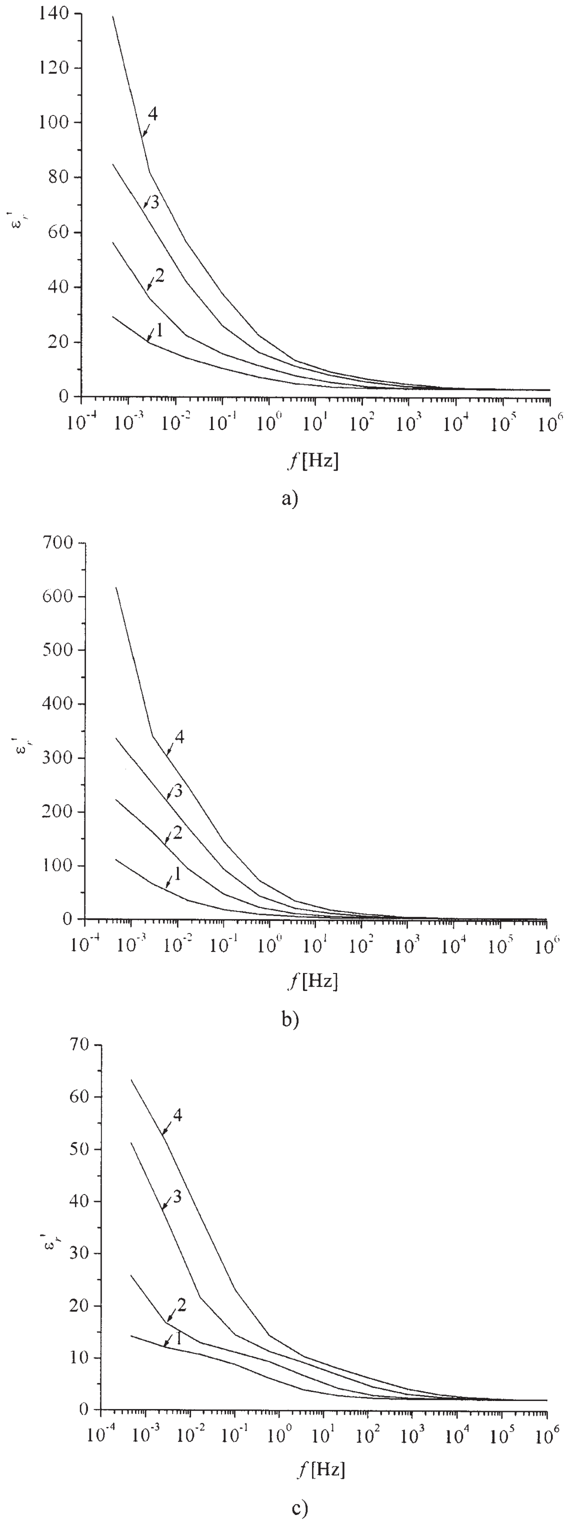
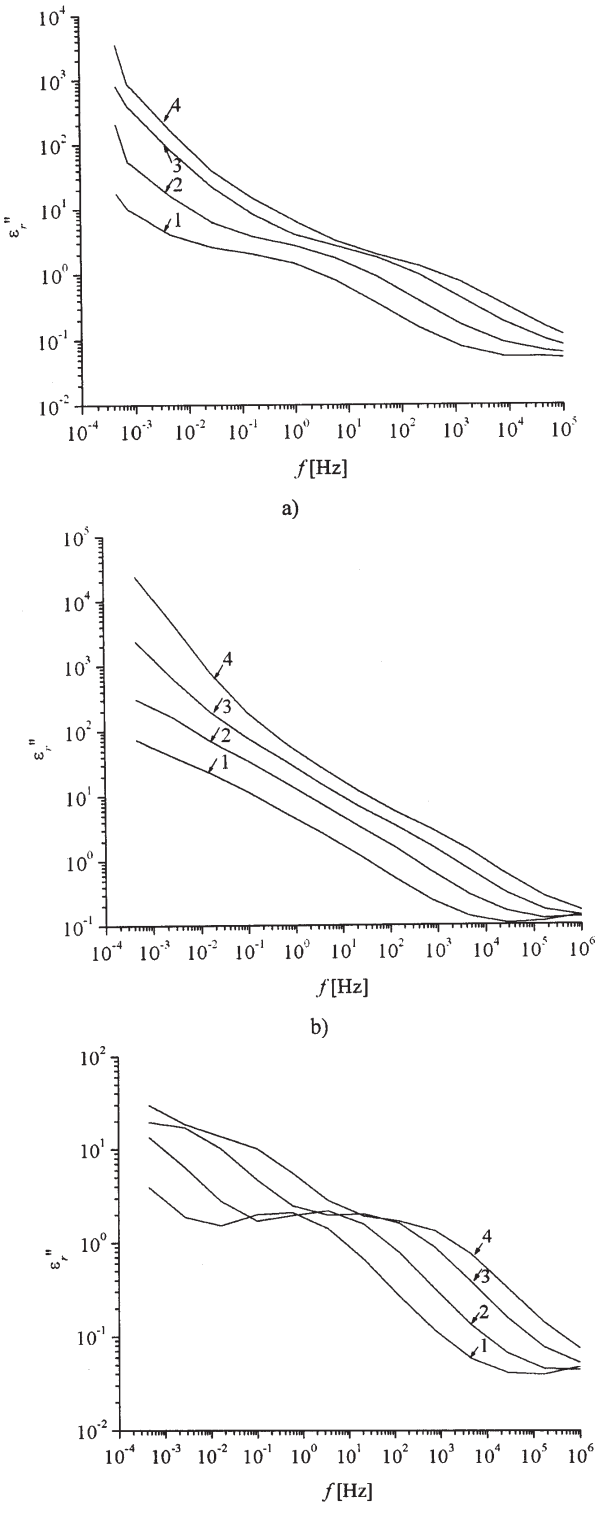


Fig.1. Variations of the real component of the electrical permittivity ε'_r with frequency f, for PE1 (a), PE2 (b) and PP2 (c) samples, at 30 °C (1), 50 °C (2), 70 °C (3) and 90 °C (4)

Furthermore, electrode polarization, a very significant phenomenon at low frequencies, contributes, too, to sample polarization and increasing the value of ε'_r [7].

In low frequency situations, a very sharp increase in the values of both ε''_r and tgδ can be observed (figs. 2-3). This is, firstly, an effect of the increase in ion conduction corresponding to the ion space charge: the mobility of ion carriers may increase at low frequencies as a result of a greater possible duration in which ions may hop (duration



a)

b)

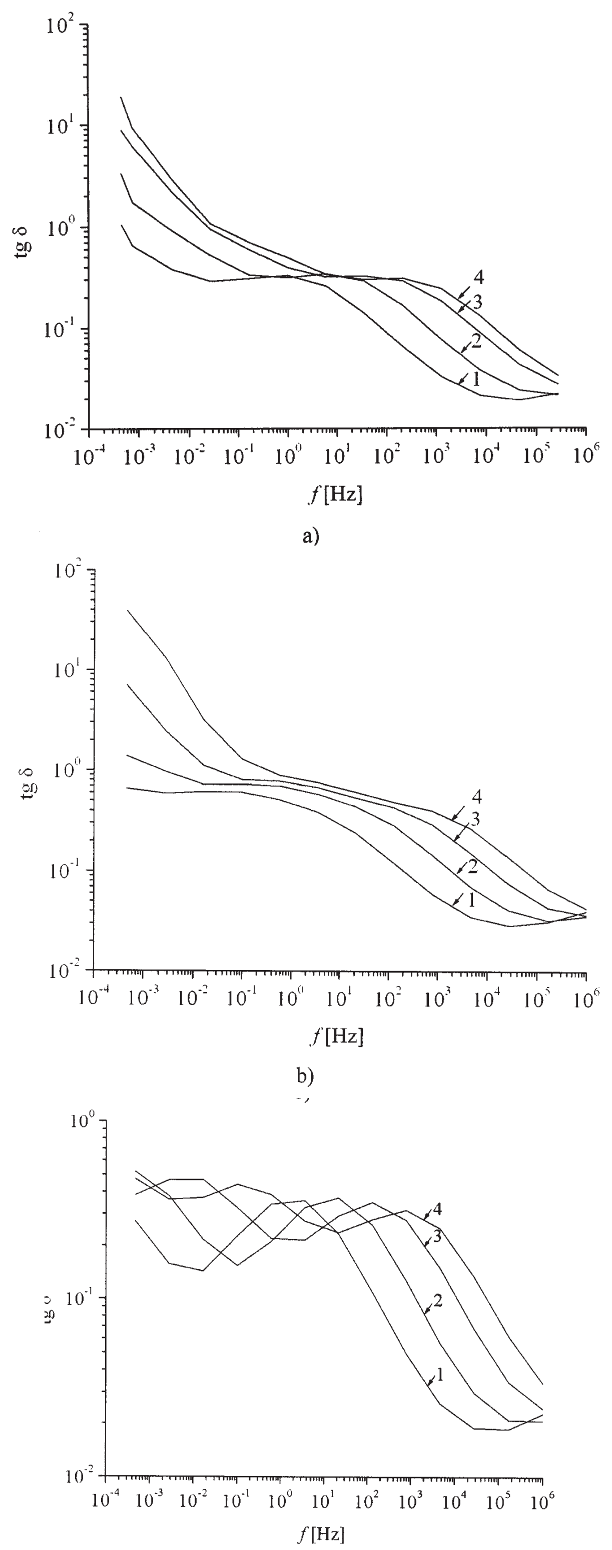
c)

Fig. 2. Variations of the imaginary component of the electrical permittivity ε''_r with frequency f, for PE1 (a), PE2 (b) and PP2 (c) samples, at 30 °C (1), 50 °C (2), 70 °C (3) and 90 °C (4)

in which the electric field maintains its orientation) [6]. The rise of ion conductivity determines a corresponding increase both in ε_r'' and in tgδ (5).

The "low frequency dispersion" phenomenon [3] also contributes to the increase of both ε_r'' and tgδ, particularly in humid environments (wood has a high hygroscopicity) [11-14]. In this situation, movements which occur both in intra- and inter-clusters contribute to a greater energy absorption and an increase in the values of both ε_r'' and tgδ respectively [9].

On the other hand, because the samples contain both positive ions (A^+) as well as negative ions (B^-), depending



a)

b)

c)

Fig. 3. Variations of the loss factor tgδ with frequency f, for PE1 (a), PE2 (b) and PP2 (c) samples, at 30 °C (1), 50 °C (2), 70 °C (3) and 90 °C (4)

on the relative distances between them, the pairs A^+ and B^- form electric "dipoles", whose size and orientation vary with the movement of the ions under the influence of the electric field. When the ions pass one-another, they form a neutral compound (AB) for a certain time, according to the symbolic equation

$$A^+ + B^- \leftrightarrow AB. \qquad (8)$$

This process is capable of retaining a large amount of charge without creating any electric field, only a potential which is associated with the electromechanical potential

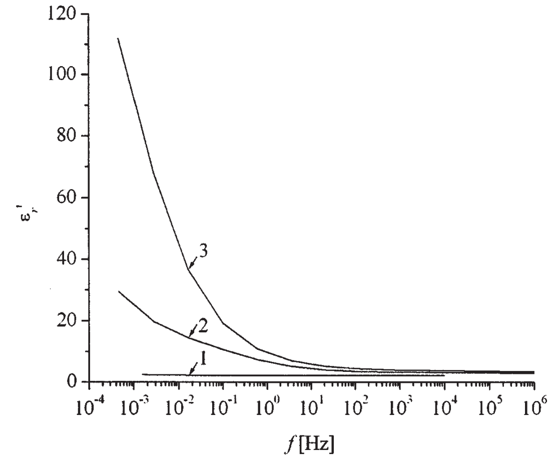


Fig.4. Variation of the real component of the electrical permittivity ε'_r with frequency *f*, for PE0 (1), PE1 (2) and PE2 (3) samples

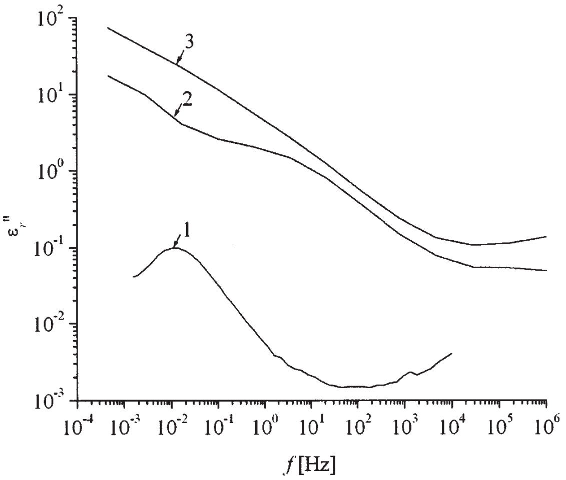


Fig. 5. Variation of the imaginary component of the electrical permittivity ε''_r with frequency for PE0 (1), PE1 (2) and PE2 (3) samples

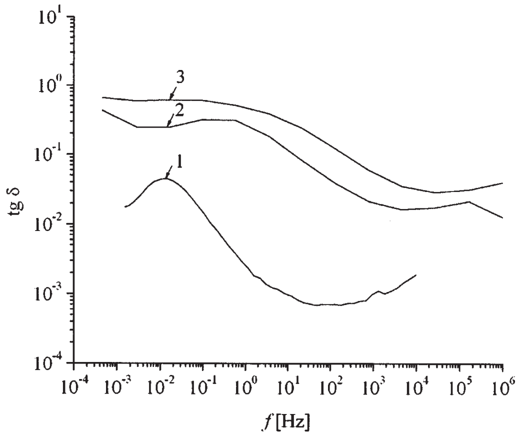


Fig. 6. Variation of the loss factor tgδ with frequency *f* for PE0 (1), PE1 (2) and PE2 (3) samples

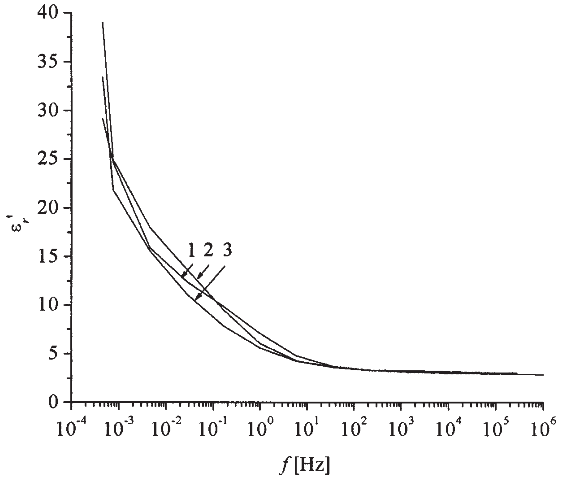


Fig. 7. Variation of the real component of the permittivity ε'_r with frequency *f*, for PE1 samples of thickness: 3.582 mm (1), 3.698 mm (2), and 3.703 mm (3) (*T* = 30°C)

of the system [8]. Likewise, some energy is supplied either in the formation of the compound or in its dissociation, so that either way, an irreversible energy loss will be incurred in every change in the orientation of the ionic “dipole”. Consequently, the values of ε_r” and tgδ will increase for very low frequencies.

Increasing wood flour content leads to an increase in all quantities ε_r’, ε_r” and tgδ (figs. 4-6). Therefore, in the case of polyethylene matrix, an increase in the mass concentration of wood flour from 30 to 40 % gives an increase of ε_r’ from 19.7 to 67.8, of ε_r” from 9.9 to 40, and of tgδ from 0.2 to 0.7 when the samples are subjected to an electric field with the frequency of 2.8 mHz. This is the result of the higher value of the electric permittivity of wood as compared to polyethylene on the one hand, and increasing the matrix-filler interface areas in which there are matrix polymer chain ends (they are more easily displaced under the action of the electric field) and wherein space charge separation occurs (which is responsible for increasing interfacial polarization, on the other hand.

Comparing results obtained on samples with the same wood flour content (for exemple, 40 %) but with different matrixes, it can be ascertained that samples with a polypropylene matrix have superior properties (lower values for ε_r’, ε_r” and tgδ). This is, probably, due to superior “compactness” in the case of a polypropylene matrix together with a more efficient interaction between the molecules of the matrix and the filler material and a lower material “free volume” .

Peaks can be noticed (one in the case of polyethylene matrix samples, two in the case of polypropylene matrix samples) in the frequency-dependant loss factor charts. (fig.3). Both the values of these peaks, as well as the frequency for which they are reached increase with temperature (this is especially apparent in the case of polypropylene matrix samples).

Reproductibility of the results is heavily influenced by the manner in which the samples are produced, specifically the temperature, mixture and compression durations of the constituents. A series of deviations were observed with respect to sample thickness (as high as 7%) which, of course, contributed to the relatively important differences between the values of the measured parameters (fig.7). Naturally, in order to raise result credibility, the number of tested samples should be increased (to 10-20 samples).

Presently, certain deviations have been noticed both regarding thickness, as well as the measured values of certain parameters (fig. 7). Furthermore, the number of used samples employed in order to properly characterize a composite material needs to be greater than 5 (10-20).

Conclusions

The technological process used to obtain wood flour-polymer composites is relatively simple and easy to control. Both the matrix materials, as well as the fillers can be obtained from industrial or commercial waste (polyethylene, sawdust etc.) which leads to low-priced composites.

The electric strength of all tested composites is higher than 6 kV/mm in the case of all samples with a wood flour content of 48% at most.

The polypropylene matrix leads to greater composite compactness and higher density and better dielectric properties. .

The electric permittivity and dielectric losses have relatively-close values to those of their matrix at medium and high frequencies, but these increase dramatically at low frequencies (under 50 Hz). Therefore, these materials can be utilized in electrical engineering for low voltage and industrial frequencies applications as terminal boards, spacers, insulating levers, insulating stands etc.

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