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Dielectric properties of lignocellulosic materials–polypropylene composites

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Composites were made from isotactic polypropylene and lignocellulosic materials derived from hemp and flax plants using moulding and hydraulic pressing techniques. Frequency dependences of the electric permittivity ε' and the ac conductivity σ_{ac} of the composite samples were measured in the frequency range from 100 Hz to 13 MHz at room temperature. The linear relationship between the reciprocal relative permittivity ($1/\varepsilon'$) and the volume fraction of the lignocellulosic material was established. The loading of the polypropylene with the lignocellulosic materials increases the electric permittivity ε' and improves the electrical conductivity σ_{ac}. The effect of temperature (150–450 K) on the dielectric spectrum of polypropylene and the composites was investigated in the frequency range from 100 Hz to 1 MHz. Four dielectric relaxation processes were found in the spectra of the composites: β – ascribed to the local motion of the main polymer chain, β_{wet} – associated with the presence of water, δ – related to the motion of the end groups in the branched polymers and σ – due to the conductivity effect. The dielectric spectrum of polypropylene does not exhibit anomalies.

Key words: *polymer matrix composites; lignocellulosic material; electrical properties*

1. Introduction

Composites of thermoplastic polymers based on lignocellulosic fillers are widely used in several branches of industry, e.g. automotive and building engineering [1], aviation and the packaging industry [2]. Mechanical and physical properties of a polymer can be improved by addition of a lignocellulosic filler [3–5]. Composites made from polypropylene (PP) and wood fibre are known to exhibit significantly higher stiffness than unreinforced polypropylene [6]. The loading of the polypropylene

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with rice husk powder increases Young's modulus and the flexural modulus of the composite, compared with those of the polypropylene [7]. Cellulose fibres incorporated into the polyethylene matrix also enhance the mechanical performance of the composite [8–10]. The most interesting feature of the biocomposites is their partial biodegradability as the filler materials come from a natural resource. The addition of lignocellulosic materials results in a significant decrease of such essential parameters as heat release rate (HRR) peak and mass loss rate (MLR) [11, 12] which are very important in terms of the resistance against the fire expansion. The nonabrasive nature of lignocellulosic fillers allows one to treat biocomposites as an ecological alternative [1] to composites reinforced with glass fibre and other mineral fillers. An additional advantage of the polymer composite based on the lignocellulosic filler is their sound absorption capacity due to the anatomic structure of the tubiform lignocellulosic components [13–16]. The hollow structure is responsible for the insulation against heat and noise.

Combination of the polymer and the lignocellulosic materials results in new dielectric properties of the composite. The possibility of designing new materials with predictable properties is very promising from the application point of view. The materials characterized by a low electric permittivity are applied as insulators to cut off the energy flow in alternating and direct electric current circuits. However, to achieve good energy accumulation in the presence of an electric field, materials having high electric permittivity are particularly suitable. In the latter application, the temperature insensitivity and the dispersion of the electric permittivity are of great importance because the materials are often used for thermal compensation of the electric circuits as well as they work in high frequency range [17]. Recently, new composite materials with non-standard dielectric properties were designed. The composites containing hollow keratin fibres are characterized by unusually low value of electric permittivity due to air present inside [18]. They can replace dielectrics in microchips and printed circuit boards used in the electronic industry. Some composites were designed with the aim of application as antistatic materials to dissipate static charges. They show typical behaviour which can be concluded: the electric permittivity, the dissipation factor and the electrical conductivity increase steadily with increasing fibre loading whereas the volume resistivity decreases with increase of fibre content. Sisal fibre reinforced LDPE [19, 20], sisal–oil palm hybrid biofibre reinforced natural rubber [18] and epoxy resin reinforced with sisal fibre [21] belong to this kind of biocomposites.

In recent years, the composites of polypropylene reinforced with hemp [22] and flax [23] were developed. The thermal behaviours and thermal decompositions of both composites were examined by the differential scanning calorimetry (DSC) and thermogravimetry (TGA), in addition to microscopy analyses. The mechanical properties (tensile modulus, stress and elongation at break) of polypropylene–hemp composites were also determined [22]. However, dielectric properties of polypropylene composites reinforced with hemp and flax have not been investigated to date. The aim of this work is to show the influence of the lignocellulosic material loading on the electric permittivity ε' and on the ac conductivity σ_{ac} of the composites. The temperature dependences of the electric permittivity ε' and the loss factor ε'' have also been studied.

2. Experimental

Materials. Isotactic polypropylene Malen F-401 (Melt Flow Index 230/2.16 = 2.4 –3.2 g/10 min, isotacticity 95%), produced by Orlen Płock (Poland) was used as a matrix for fabrication of the composites. Lignocellulosic materials: long and short flax fibres, long and short hemp fibres as well as flax and hemp shivers were used as filling materials.

Sample preparation. The composites were made by two different methods. The former one consisted in mixing crumble lignocellulosic materials with polypropylene granulate in proportion of 25 wt. % of natural component. After that, the extrusion was carried out using a "Fairex" (McNell Akron Repiquetn, France) single-screw extruder, *L*/*D* = 25. The composite material was obtained in a granulate form [24]. To obtain samples for investigations, the granulate of the composite was melted in a mould between heating plates at 200 °C under the load of 3000 kG. The composites containing long fibres of hemp and flax (40 wt. %) were fabricated using only the technique of hydraulic pressing at 200 °C under the load of 3000 kG, described elsewhere [25]. Finally, all the samples assumed the shapes of disks, typically having the diameter of 10 mm and the thickness of 1.5 mm. In Table 1, all the fabricated samples are specified.

Table 1. Specification of the investigated samples

No.	Material	Density [kg/m^3]	Volume fraction of the lignocellulosic material
1	Polypropylene (PP)	881.8	0
2	PP+25% of short hemp fibres	862.5	0.27
3	PP+25% of hemp shivers	911.3	0.28
4	PP+40% of long hemp fibres	927.9	0.37
5	PP+25% of short flax fibres	883.0	0.25
6	PP+25% of flax shivers	943.1	0.20
7	PP+40% of long flax fibres	934.6	0.37

Measurement. The plane surfaces of the samples (Table 1) were covered with gold electrodes using the BALTEC SCD 050 sputter coater and fixed to the sample holder of the measurement device. Frequency dependences of the electric permittivity ε' and the ac conductivity σ_{ac} of the samples were measured by means of a computer aided (program WinDETA V4.0) HP 4192 LF impedance analyzer operating in the frequency range from 100 Hz to 13 MHz at room temperature.

The effect of temperature variation on the dielectric spectrum of PP as well as the of the composites was investigated in the frequency range from 100 Hz to 1 MHz using a computer-aided (program HP VEE) HP-4284A precision LCR meter. The samples were mounted in a CF 1204 Oxford Instruments cryostat equipped with an ITC 4 temperature controller. The measurements were performed during heating from 150 K to 450 K at the rate of 1 K/min.

The relative electric permittivity ε' was calculated from the expression:

$$\varepsilon' = \frac{Cd}{\varepsilon_0 S} \tag{1}$$

where: C is the capacitance of the condenser with the dielectric, S stands for the area of the sample covered by the electrode, d means the thickness of the sample and ε_0 equal to $8.85 \cdot 10^{-12}$ F/m is the electric permittivity of the vacuum.

The ac conductivity σ_{ac} was determined by using the following equation:

$$\sigma_{ac} = \varepsilon_0 \omega \varepsilon' \tan\delta \tag{2}$$

where $\tan\delta$ denotes the dielectric dissipation factor and ω stands for the angular frequency.

3. Results and discussion

The frequency dependences of the electric permittivity ε' measured at room temperature for the polypropylene composites reinforced with various lignocellulosic materials derived from hemp and flax are presented in Figs. 1 and 2. The effect of loading with the lignocellulosic material consists in the increase of the electric permittivity ε' over the whole measurement frequency range. The effect is predominant at lower frequencies. This result is very similar to that obtained by Paul et al. [19] for sisal fibre–LDPE composites.

Pure polypropylene is a non-polar hydrophobic material which shows only instantaneous ionic and electronic polarization. Its electric permittivity ε' remains nearly constant over the whole frequency range with a slender increase below 10^3 Hz. The addition of a hydrophilic lignocellulose material to the polypropylene entails the insertion of polar groups into the non-polar material, explaining why the polarization, dependent of the dipole reorientation, increases. Moreover, the presence of the hydroxyl groups –OH in the cellulose, the hemicellulose and the lignin extends the moisture absorption, due to the interaction of –OH groups and water molecules. The overall polarization of the composite, being the sum of three contributions: electronic, ionic and dipole reorientation-related ones, exhibits the maximum values at low frequencies and decreases with increasing frequency. The same behaviour shows the electric permittivity of the composites ε’.

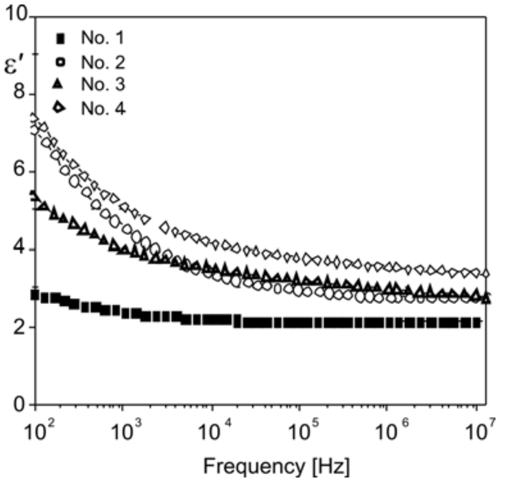

Fig. 1. Frequency dependences of electric permittivity ε' obtained for PP and the composites with hemp: No. 1 – PP, No. 2 – PP + 25 wt. % of short hemp fibres, No. 3 – PP + 25 wt. % of hemp shivers, No. 4 – PP + 40 wt. % of long hemp fibres

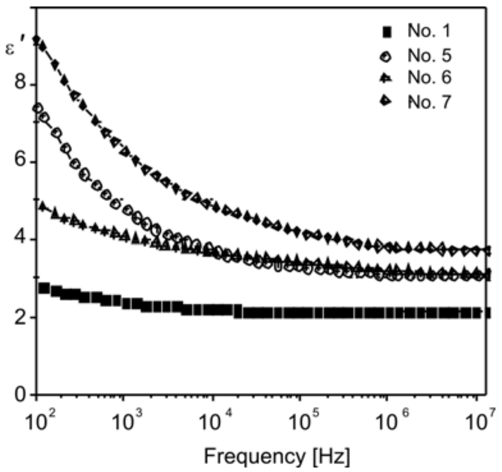

Fig. 2. Frequency dependences of electric permittivity ε' obtained for PP and the composites with flax: No. 1 – PP, No. 5 – PP + 25 wt. % of short flax fibres, No. 6 – PP + 25 wt. % of flax shivers, No. 7 – PP + 40 wt. % of long flax fibres

The relative electric permittivity ε' increases as the content of the lignocellulosic material increases. In the higher frequency range, i.e. above 10^6 Hz, the value of the relative electric permittivity ε' tends to the constant value fixed by squared refractive index. Moreover, the values for composites with the same content (25 wt. %) of different lignocellulosic materials converge. In the case of hemp–polypropylene composites (Fig. 1), one can notice that the values of ε' for PP + 25 wt. % of short hemp fibres (No. 2) and for PP + 25 wt. % of hemp shivers (No. 3) are ca. 2.7. For the flax –polypropylene composites it can be seen (Fig. 2) that the values of ε' obtained for PP + 25 wt. % of short flax fibres (No. 5) and for PP + 25 wt. % of flax shivers (No. 6) tend to 2.8 as the frequency increases. In the frequency range between 10^3 and 10^4 Hz, one can notice a characteristic point below which the crucial modification in the frequency dependences of the relative permittivity ε' appears. Thus, the differences in the electric permittivity ε' are most significant in the low frequency range, and they are observed also for the composites having the same content of lignocellulosic materials derived from different parts of hemp and flax. The composites containing the shivers derived from hemp as well as from flax (25 wt. %) exhibit lower ε' than the ones comprising short fibres (also 25 wt. %). The shivers are the lignified parts of stems, separated from the fibres, and they show lower capacity of moisture absorption. This fact can be indicative of a smaller number of polar groups and lower polarization related to dipole reorientation.

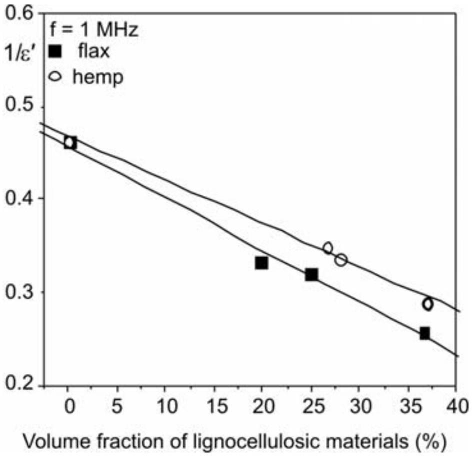

Fig. 3. Reciprocal electric permittivity $1/\varepsilon'$ in function of volume fraction of lignocellulosic material derived from hemp and flax

Figure 3 displays the plots of the reciprocal relative electric permittivity $1/\varepsilon'$ vs. the volume fraction of lingocellulosic material derived from hemp and flax at the frequency of 1 MHz. The obtained results are in agreement with the theoretical model of the electric permittivity for natural rubber biocomposites reinforced with sisal oil palm hybrid biofibre, as presented by Jacob et al. [18]. The relationship between the reciprocal of the relative electric permittivity ε' and the volume fraction of the lignocellulosic material is linear for the applied contents of the filler. The dielectric impermeability of the composite decreases as the filler content increases.

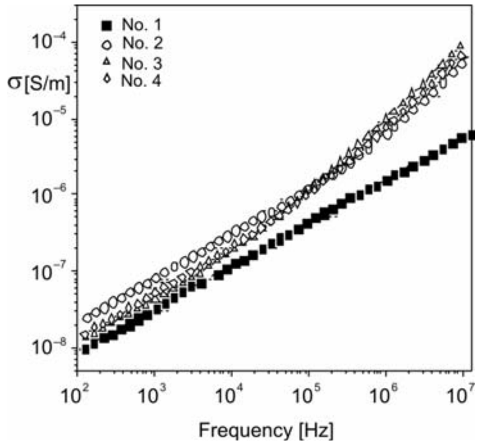

Fig. 4. Frequency dependences of ac conductivity for the samples: No. 1 – PP, No. 2 – PP + 25 wt. % of short hemp fibres, No. 3 – PP + 25 wt. % of hemp shivers, No. 4 – PP + 40 wt. % of long hemp fibres

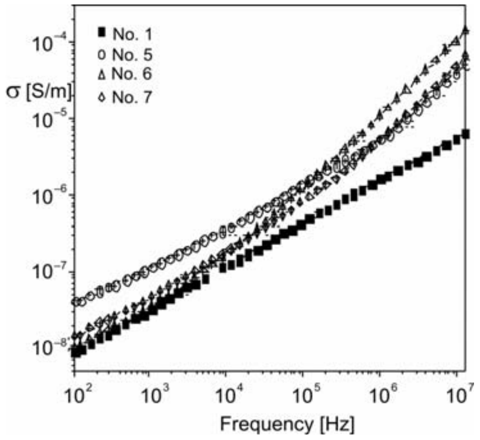

Fig. 5. Frequency dependences of ac conductivity obtained for the samples: No. 1 – PP, No. 5 – PP + 25 wt. % of short flax fibres, No. 6 – PP + 25 wt. % of flax shivers, No. 7 – PP + 40 wt. % of long flax fibres

The variations of the ac conductivity in function of the frequency measured for the polypropylene and all composite samples at room temperature are shown in Figs. 4

and 5. The reinforcement of the polypropylene with the lignocellulosic material increases the conductivity. The frequency dependence of the electrical conductivity is described by the expression [26]:

$$\sigma(\omega) \propto \omega^n \tag{3}$$

The exponent n is close to 0.5 for pure polypropylene and points to diffusive carrier transport. For the composites, n changes from ca. 0.5 at low frequencies to ca. 1 at high frequencies. This fact proves the existence of diffusive as well as hopping carrier transport. The lack of any frequency-independent conductivity at low frequencies indicates that no ohmic conduction occurs at room temperature in any of the samples under investigation.

One can state the influence of the distribution of the lignocellulosic fillers in the matrix on the frequency dependence of ac conductivity. The incorporation of the shivers (No. 3 Fig. 4, and No. 6, Fig. 5) as well as long fibres (No. 4, Fig. 4, and No. 7, Fig. 5) in the polypropylene causes a relatively small increase in the conductivity at lower frequencies. A comparatively high increase in the conductivity at lower frequencies is shown by the composites with the short fibres derived from hemp (No. 2, Fig. 4) as well as from flax (No. 5, Fig. 5). This fact is probably due to the different distribution of the filler. Short fibres are randomly distributed in the matrix, and the electric field is applied at different angles with respect to the axes of the fibres. The random distribution enables rearrangement of the short fibres in a chain structure which ensures better carrier mobility in the presence of electric field. However, the manufacturing method of the composites with long fibres implies the perpendicular orientation of the electric field with respect to the fibre axis. It is difficult to arrange the fibres so that they form a chain structure. These results demonstrate that the composites having short, randomly distributed fibres provide the best conductivity among the investigated samples.

The variations of the relative electric permittivity ε' with the temperature investigated for the polypropylene as well as the composites with hemp and flax are presented in Figs. 6 and 7. The value of ε' measured for polypropylene is almost independent of temperature, up to the melting point at 438 K [27]. The dielectric response of the composites is dominated by the relaxation ascribed to an orientational motion of mixed phase of both lignocellulosic material and water. This phenomenon, characteristic of cellulose and its derivatives, has been reported by many authors [28–31]. The electric permittivity ε' of the composites increases with the temperature up to the maximum associated with traces of water, and then decreases. The position of the maximum is determined by the contents of chemically bound water which cannot be removed during the preparation. The maximum is shifted towards higher temperatures in the case of higher water contents [21]. In the vicinity of the melting point of the polypropylene, a steep fall is visible. The relative electric permittivity ε' decreases as the frequency increases, as is seen for two frequencies: 100 Hz and 1 MHz.

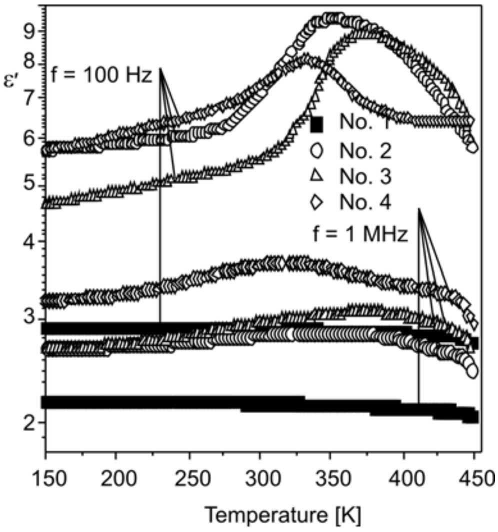

Fig. 6. Temperature dependences of electric permittivity ε' obtained at frequencies 100 kHz and 1 MHz for the samples: No. 1 – PP, No. 2 – PP + 25 wt. % of short hemp fibres, No. 3 – PP + 25 wt. % of hemp shivers, No. 4 – PP + 40 wt. % of long hemp fibres

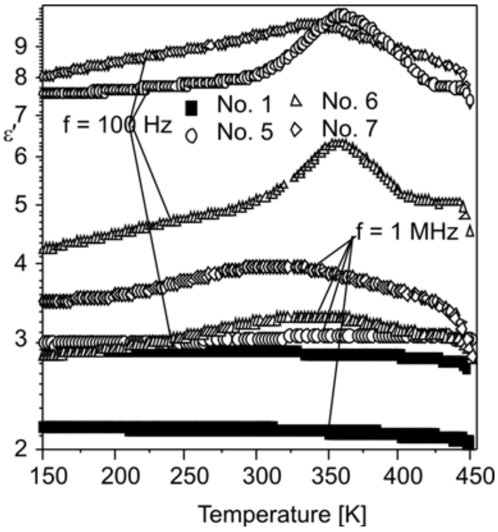

Fig. 7. Temperature dependences of electric permittivity ε' obtained at the frequencies 100 kHz and 1 MHz for the samples: No. 1 – PP, No. 5 – PP + 25% of short flax fibres, No. 6 – PP + 25% of flax shivers, No. 7 – PP + 40% of long flax fibres

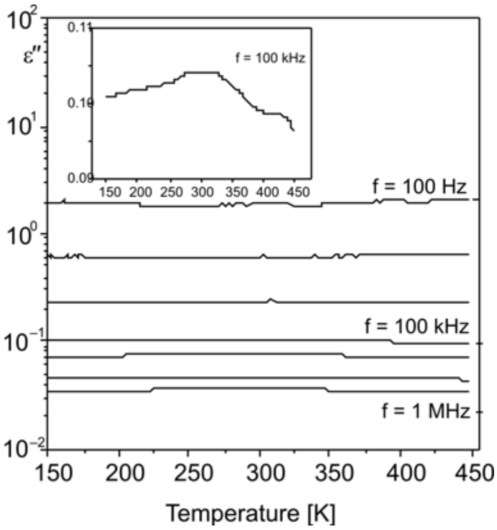

Fig. 8. Temperature dependence of dielectric loss factor ε'' for polypropylene. Inset: the dependence at the frequency of 100 kHz. Note an expanded ε'' scale

The temperature dependence of dielectric losses of the composites is strongly influenced by the contribution of the lignocellulosic materials. The dielectric loss factor ε'' of pure PP in function of temperature is presented in Fig. 8. Pure PP is known to exhibit two characteristic features [32]: a glass relaxation peak around 263 K, and a high-temperature (ca. 323 K) shoulder associated with chain relaxation in the crystalline phase. These features cannot be exactly detected by the dielectric relaxation spectroscopy (DRS) method without special modification of the polymer structure or introducing polar groups in the structure [33] because of the non-polarity of the polypropylene. The absence of polar groups is the reason why the temperature dependence of the dielectric losses does not show any anomalies (Fig. 8). In the inset, the two shoulders are hardly visible. The former one starts to increase above 263 K and is ascribed to the glass transition. The other one appears around 314 K and is caused by the chain relaxation in the crystalline α-phase. These trace relaxation processes in the spectrum of pure polypropylene result from the presence of a small number of polar groups, due to undesired phenomena (thermal degradation of chains and oxidation) during processing of the polymer.

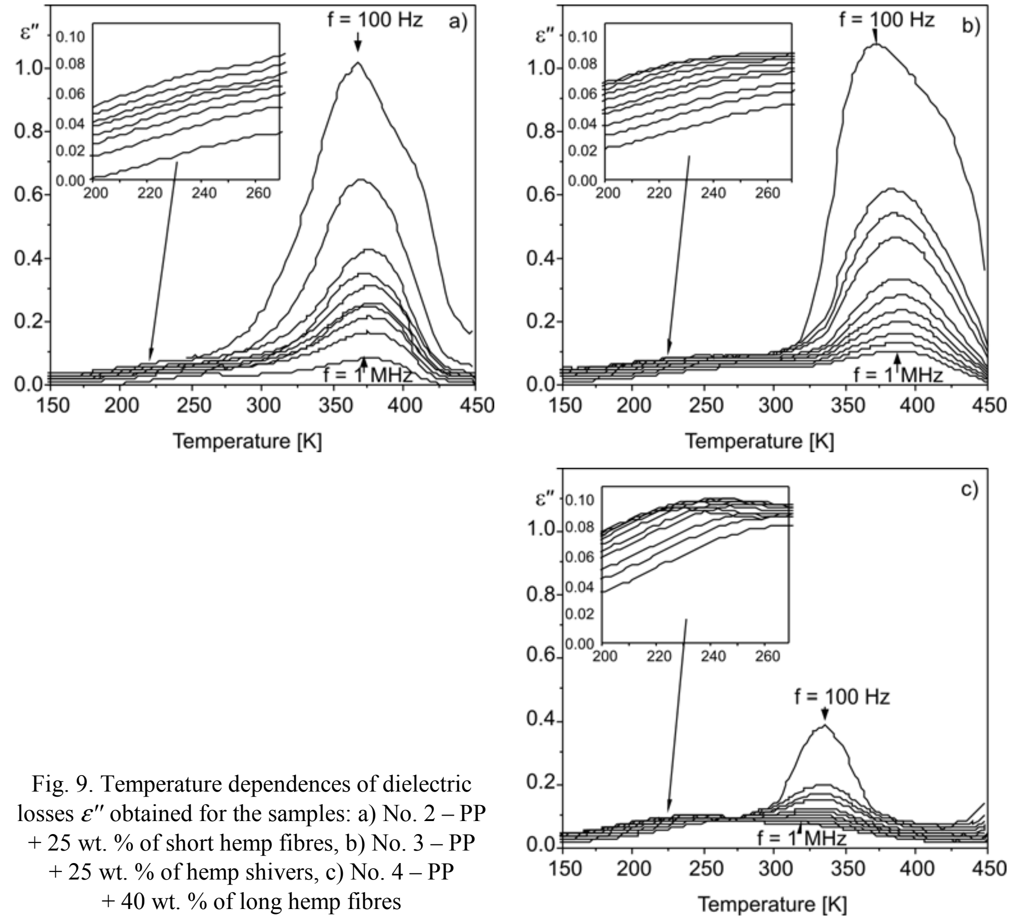

Fig. 9. Temperature dependences of dielectric losses ε'' obtained for the samples: a) No. 2 – PP + 25 wt. % of short hemp fibres, b) No. 3 – PP + 25 wt. % of hemp shivers, c) No. 4 – PP + 40 wt. % of long hemp fibres

Figures 9 and 10 show the dielectric loss factor ε'' in function of temperature measured for the polypropylene composites reinforced with the lignocellulosic material derived from hemp and flax, respectively. Both kinds of fillers modify the dielectric absorption spectrum in the same way. The anomalies due to polar groups present in the lignocellulosic material can be easily detected by means of DRS.

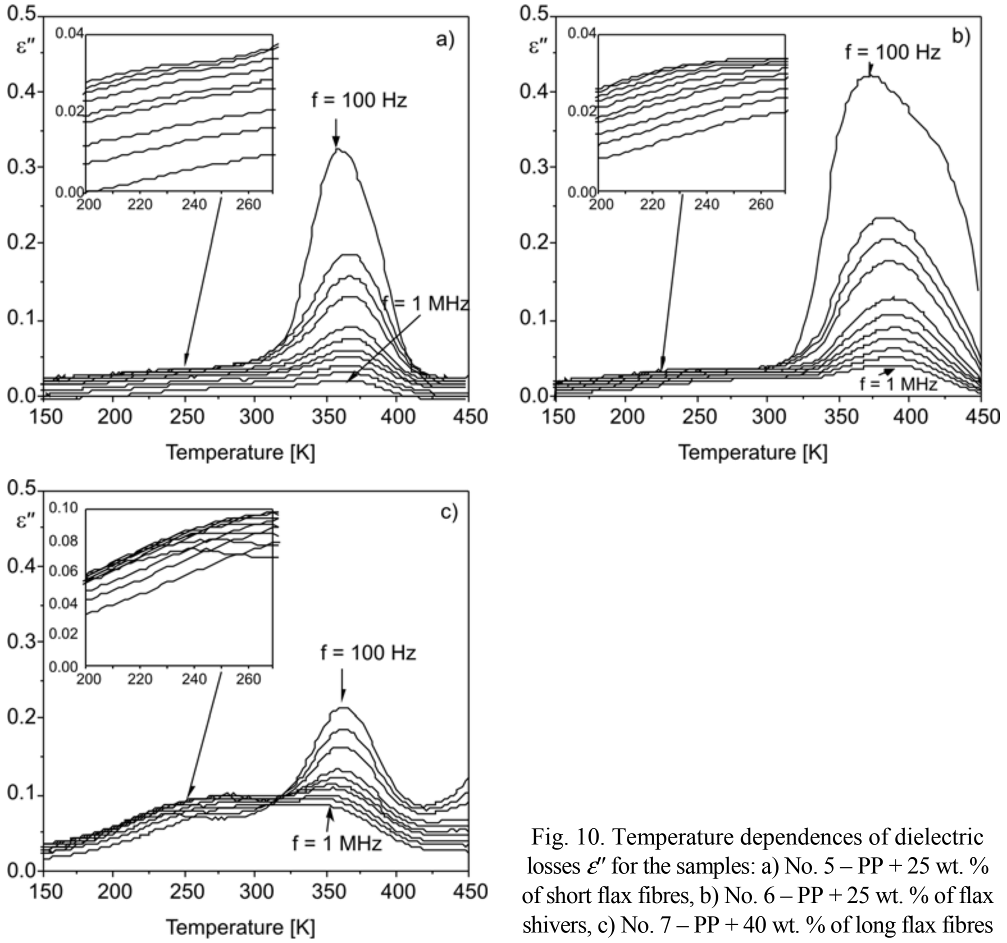

Fig. 10. Temperature dependences of dielectric losses ε'' for the samples: a) No. 5 – PP + 25 wt. % of short flax fibres, b) No. 6 – PP + 25 wt. % of flax shivers, c) No. 7 – PP + 40 wt. % of long flax fibres

In the low-temperature range (from 200 to 270 K) one can observe the maxima of ε'' ascribed to the β relaxation in cellulose, which is the main component of each lignocellulosic filler (71 wt. % in flax, and 75 wt. % in hemp fibres [3]). The β relaxation is interpreted as a local motion of chain segments via the glucosidic linkages [28]. Above room temperature (330–400 K) one can notice high relaxation peaks. From their shape, it can be deduced that two relaxation processes overlap in this temperature and frequency range: β_{wet} – the relaxation associated with the orientational motion of both cellulose and water [28, 29] and δ – the relaxation ascribed to the motion of the end groups in branched polymers [28] present in the lignocellulosic material (hemicellulose, pectin, lignin). Because the intensity of the δ relaxation is significantly lower than that of β_{wet} relaxation [28], one can state that in the polypropylene–lignocellulose material composites the β_{wet} relaxation is disturbed by the δ process. In the case of composites with long fibres derived from flax as well as hemp (Figs. 9c and 10c), higher dielectric losses were observed in the highest temperature range (above 420 K). This effect results from the electric conductivity and is called σ relaxation [28]. The losses due to the electric conductivity are ascribed to charge carrier hopping between localized sites in amorphous solids. The fact that the σ relaxation was observed only for the composites with long fibres confirms the conclusion [28] that the activation energy for the carrier hopping increases when the amount of water is reduced. The technique of hydraulic pressing, applied for fabrication of composite samples with long fibres implied low contents of water. The intensity of β_{wet} relaxation is proportional to the contents of water. The low intensity of β_{wet} relaxation in the composites with long fibres is a reason for a relatively high strength of β relaxation in comparison with that observed for other investigated samples where the β process is suppressed by β_{wet} relaxation.

The temperature dependence of the β relaxation time $\tau(T)$ follows an Arrhenius type behaviour, characterized by the activation energy E_a and a pre-exponential factor τ_0:

$$\tau(T) = \tau_0 \exp\left(\frac{E_a}{kT}\right) \tag{4}$$

where k is the Boltzmann constant. The activation energy is interpreted as the height of the potential barrier between two orientational conformation states which can be reached during the molecular motion [28]. The pre-exponential factor τ_0 represents the inverse oscillation frequency of the molecular group in a conformational potential minimum [28]. According to the Eyring rate theory [34], the factor τ_0 is an indicator of the cooperativity of the orientational motion within polar groups. A smaller value of τ_0 means higher cooperativity of the motion.

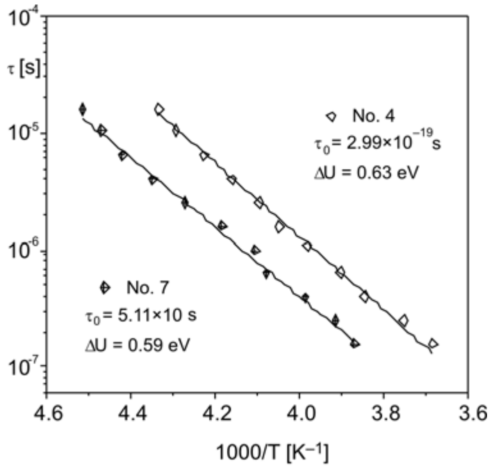

Fig. 11. β -relaxation time vs. reciprocal of temperature for the composite samples: No. 4 – PP + 40 wt. % of long hemp fibres and No. 7 – PP + 40 wt. % of long flax fibres

The Arrhenius plots of the relaxation times for the polypropylene composites with long fibres are presented in Fig. 11. The values of the activation energy E_a and the pre-exponential factor τ_0 resulting from fitting indicate better cooperativity of the motion than that of cellulose, which is usually characterized by the activation energy below 0.5 eV and pre-exponential factor of the order of 10^{-16} s [28]. The origin of the improved cooperativity might be the presence of branched polymers (hemicellulose, pectin, lignin) in the lignocellulosic materials. Branched polymers are known to have the capacity to form interchain hydrogen bonds, due to their high dimensions. In this manner, the local chain motion can be involved not only in the nearest repeating unit of the same chain but also in motion of another chains [35].

4. Conclusions

The dielectric properties (relative electric permittivity ε' and ac conductivity σ_{ac}) of polypropylene–lignocellulosic material composites were evaluated in function of frequency, filler contents and temperature. The relative electric permittivity ε' increases with loading of the polypropylene with the lignocellulosic material. In all the filler types considered, the relative permittivity ε' is inversely proportional to the volume fraction of the lignocellulosic material. The increase in the electric permittivity ε' is attributed to the increase in the orientation polarization of polar groups present in the lignocellulosic material. The effect is more predominant in the low frequency range. The incorporation of the lignocellulosic materials in the polypropylene matrix improves ac conductivity σ_{ac}.

The effect of temperature variation (150–450 K) on the dielectric spectrum of polypropylene and the composites was investigated in the frequency range from 100 Hz to 1 MHz. Four dielectric relaxation processes were found in the spectra of the composites: β – ascribed to the local motion of the main polymer chain, β_{wet} – associated with the presence of water, δ – related to the motion of the end groups in the branched polymers and σ – due to the conductivity effect. The dielectric spectrum of polypropylene does not show anomalies. The temperature dependence of dielectric losses of the composites is strongly influenced by the contribution of the lignocellulosic materials.

The obtained results point to the possibilities of practical application of the investigated composites. The increased electric permittivity, in comparison with the polypropylene, is preferable in the application for accumulation of the energy in electric field. The constant value of ε' at frequencies above 1 MHz is evidence that the composites can be used in the high frequency range. The temperature measurements reveal that the composites with short fibres show the best thermal stability of the relative electric permittivity ε' in the vicinity of the room temperature. The other investigated samples exhibit more pronounced maxima in the temperature dependence profile of the relative electric permittivity because of the contents of water. Moreover, the polypropylene composites reinforced with short fibres assure the best antistatic properties, because of their improved electrical conductivity over the whole measurement frequency range. The results presented point to the possibility of exploiting polypropylene composites reinforced with flax and hemp lignocellulosic material in the furniture, building and automotive industry in modern wood–plastic composite (WPC) designs [36]. Not only these materials have better antistatic properties in comparison with the polypropylene, but they also have good sound absorption [36]. Composites containing lignocellulosic fibres [13–15] will improve the quality of human life.

Acknowledgements

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