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# Acoustic and Dielectric Properties of Polypropylene-Lignocellulosic Materials Composites

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## 1. Introduction

In recent years, one can observe the tendency to replace the thermoplastic polymers by the composite materials in several branches of industry, e.g. automotive and building engineering (Peijs, 2003), aviation and packaging industry (Bledzki & Gassan, 1999). The composite materials obtained by reinforcement of the polypropylene with lignocellulosic fillers are known to show improved mechanical and physical properties in comparison with the pure propylene (Averous & Le Digabel, 2006; Bhattacharyya et al., 2003; Mohanty et al., 2000). Composites made from polypropylene and wood fibre are characterized by significantly higher stiffness than unreinforced polypropylene (Bhattacharyya et al., 2003). The loading of the polypropylene with rice husk powder increases Young's modulus and flexural modulus of the composite, compared with those of the polypropylene (Hattotuwa et al., 2002). The studies of the fire behavior (Borysiak et al., 2006) revealed a significant decrease of such an essential parameter as the heat release rate (HRR) peak, especially low value of HRRmax in comparison with those of the polypropylene. Moreover, very important feature of the composites with lignocellulosic materials is their partial biodegradability as the filler materials come from natural resource. In this chapter we would like to point out to improved acoustic and dielectric properties of the polypropylene-lignocellulosic materials composites in comparison with the pure polypropylene based on our measurement results.

Nowadays, technical progress in manufacturing the modern equipment, generating higher sound pressure, implies the need to search for new sound absorbing materials to improve the human comfort today. The ascending requirements related to the construction materials absorbing the undesired noise occur mainly in the automotive industry and building. The commonly used and unporous materials as ceramic tile, concrete, cement, fiberboard and playwood are characterized by weak sound absorption properties with the sound absorption coefficient bellow 5% in the frequency range from 125 Hz to 8000 Hz (Tiwari et al., 2004; Yang et al., 2003). The sound absorption capacity of the environment is usually corrected by the sound absorbance systems made from glass wool, foam (metals and polyurethanes), rubber, mineral fibres and their composites. Although the sound absorption can be significantly increased due to installation of these traditionally applied materials, they cause environmental pollution and pose danger to human health. Recent tendency towards the environmental protection stimulates the utilization of natural materials as sound absorbers, e. g. random cut rice straw (Yang et al., 2003), coconut coir (Nor et al., 2004), bamboo (Liu & Hu, 2008) and tea-leaf-fibre (Ersoy & Kucuk, 2009). We propose the use of composites made from polypropylene and lignocellulosic material derived from hemp, flax, beech wood and rapeseed straw as promising sound absorbers (Markiewicz et al., 2009).

Combination of the polymer and the lignocellulosic material results in new dielectric properties of the composite. The proper formation of the dielectric properties of the composites is very important in the field of their application, particularly when they are designed as electronic packaging. In this case, the electrical parameters of the microelectronic devices, such as signal attenuation, propagation velocity, and cross talk, are influenced by the dielectric permittivity value $\varepsilon'$, dielectric losses $\varepsilon''$ and their temperature stability in a wide frequency range (Pecht et al., 1999; Chung, 1995; Subodh et al., 2007). On one hand the permittivity $\varepsilon'$ should not be low because of the demand for the miniaturization of the device but on the other hand it cannot be too large in order to enable the high signal propagation speed. The signal delay $T_d$ propagated through the metal embedded in the packaging material is determined by the dielectric permittivity $\varepsilon'$ according to the formula (Tummala, 1991):

$$T_d = \sqrt{\frac{\varepsilon'}{c}} \tag{1}$$

where $c$ is the elastic coefficient. It is evident that high dielectric permittivity $\varepsilon'$ reduces the signal propagation speed. Similarly, in the case of application of the composite as the substrate in sensor of acoustic surface waves the signal propagation speed $v$ is reduced due to high dielectric permittivity $\varepsilon'$ in accordance with the relationship:

$$v = \sqrt{\frac{c + \frac{e^2}{\varepsilon'}}{\rho}} \cdot \sqrt{1 - \beta^2}\ , \tag{2}$$

where $e$ denotes the piezoelectric coefficient, $\rho$ stands for the density and $\beta$ is the factor of decreasing the signal amplitude inside the substrate material (Soluch, 1980). In this chapter, the relationship between the dielectric permittivity $\varepsilon'$ of the composite and the volume fraction of the lignocellulosic material is established. The effect of temperature variation from 150 to 450 K on the dielectric spectrum of polypropylene and the composites was investigated in the frequency range 100 Hz to 1MHz.

## 2. Methods of sample preparation

Polypropylene-lignocellulosic materials were prepared from the following materials:

- isotactic polypropylene (PP) type Malen F-401 (melt flow rate $MFR_{230/2.16}$ = 2.4 -3.2g/10 min, isotacticity 95 %), produced by Basell Orlen Polyolefins (Poland) was used as a matrix for preparation of the composites;

- lignocellulosic materials derived from hemp, flax, beech, pine, rapeseed straw were used as filling materials.

| Number | Kind of material | Density [kg/m$^3$] |
|---|---|---|
| No. 1 | Polypropylene (PP) | 881.8 |
| No. 2 | PP+40% of crumble hemp plant | 872.8 |
| No. 3 | PP+40% of long hemp fibres | 927.9 |
| No. 4 | PP+40% of long flax fibres | 934.6 |
| No. 5 | PP+40% of rapeseed straw Kaszub | 918.8 |
| No. 6 | PP+40% of crude beech wood | 803.5 |
| No. 7 | PP+20% of crude beech wood | 921.0 |
| No. 8 | PP+20% of beech modified with succinic anhydride | 920.7 |
| No. 9 | PP+20% of crude pine wood | 850.9 |
| No. 10 | PP+20% of pine modified with succinic anhydride | 974.1 |
| No. 11 | PP+30% of crude rapeseed straw Kaszub | 889.6 |
| No. 12 | PP+30% of rapeseed straw Kaszub modified with acetic anhydride | 904.0 |
| No. 13 | PP+30% of crude rapeseed straw Californium | 924.8 |
| No. 14 | PP+30% of rapeseed straw Californium modified with acetic anhydride | 969.9 |
| No. 15 | PP+25% of short hemp fibres | 862.5 |
| No. 16 | PP+25% of hemp shivers | 911.3 |
| No. 17 | PP+25% of short flax fibres | 883.0 |
| No. 18 | PP+25% of flax shivers | 943.1 |
| No. 19 | PP+30% of crude beech wood | 922.1 |
| No. 20 | PP+30% of mercerized beech wood | 922.0 |
| No. 21 | PP+30% of beech modified with maleic anhydride | 921.9 |
| No. 22 | PP+30% of crude pine wood | 861.1 |
| No. 23 | PP+30% of mercerized pine wood | 895.4 |
| No. 24 | PP+30% of pine modified with maleic anhydride | 901.3 |
| No. 25 | PP+30% of mercerized rapeseed straw Kaszub | 902.5 |
| No. 26 | PP+30% of mercerized rapeseed straw Californium | 950.7 |

Table 1. Specification of the samples investigated

Two different methods were used to make the composites. The first one consisted in mixing crumble lignocellulosic materials with polypropylene granulate in different proportion (20 – 40 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 (*Polish Patent* 186577, 2004). The composite granulates were melted in mould between heating plates at the temperature of 200°C under load of 3000 kG to obtain the samples required for the experiments.

The composites containing the long hemp and flax fibres were produced in a different way. A technique of hydraulic pressing at temperature 200°C under load of 3000 kG (*Polish Patent* 190405, 2005).

Finally, the samples took the shape of discs. Table 1 specifies all the samples prepared.

## 3. Acoustic properties of the polypropylene reinforced with lignocellulosic

### 3.1 Effect of reinforcement of polypropylene with lignocellulosic materials on the acoustic properties

The sound absorptive power of a given material sample is characterized by the sound absorption coefficient $\alpha$ defined as the ratio of the acoustic wave energy $E_a$ absorbed by the sample to the total energy $E_i$ incident on the sample:

$$\alpha = \frac{E_a}{E_i} . \tag{3}$$

Generally, the composites are known to exhibit better sound absorption than the homogenous materials. The fact results from the additivity of all kinds of acoustic energy losses (Epstein & Carhart, 1953; Vinogradov, 2004). The sound wave propagated through the inhomogeneous medium interacts with a great number of suspended particles, which differ by the density, compressibility and thermophysical parameters from the matrix. This leads to the additional acoustic energy losses compared to that in the matrix. The property of the additivity allows to express the sound absorption coefficient of the composite $\alpha$ as a sum of four components:

$$\alpha = \alpha_0 + \alpha_F + \alpha_H + \alpha_S , \tag{4}$$

where: $\alpha_0$ - the coefficient of the matrix, $\alpha_F$ - the coefficient due to friction between filler particles and the matrix, $\alpha_H$ - the coefficient related to the heat exchange between the particles and the matrix and $\alpha_S$ - the coefficient caused by the decay of the acoustic wave in forward direction due to scattering by the particles. The results of the experimental studies by I. S. Kol'tsova et al. (Vinogradov, 2004) show that the sound absorption due to the scattering play an important role when the particle sizes are comparable or larger than the sound wave length. Thus, in the case of interaction between an acoustic waves of low frequency and the particles of micrometer/millimetre dimensions, the losses in acoustic energy due to friction and interfacial heat exchange play the main role. The different densities of the particles and the matrix are the reason for which the sound wave induced motions of both compounds can be considered as the separate ones with the friction existing between them. In real media, viscous forces arise balancing the motions of the particles and the matrix and giving rise to the sound absorption. When the heating coefficients on compression of both components are different, the effect of the variable sound pressure on the composite results in heat exchange between the components. At a macroscale, the process of compression and expansion proceeds adiabatically. However, at a microscale, i. e. in the scale of particle sizes, the process is nonadiabatic with the degree of heat exchange dependent on the frequency. At low frequencies, the temperature difference between the particles and the matrix has time to balance and the process is microscopically isothermal. In the higher frequency range, the process follows adiabatically at a microscale because balancing does not occur. The heat transfer through the filler particle - matrix interface is the reason for acoustic energy absorption (Vinogradov, 2004). Taking into account the above considerations, it can be stated that the increased sound absorption of the composites results from friction and interfacial heat exchange. This is an isothermal process, since the filler particles are of several millimetres in length and bellow millimetre in width.

The numerical calculation related to the elastic wave propagation in anisotropic media was initiated by Biot in 1955 (Biot, 1955, 1956a, 1956b). Biot used Lagrange's equations to derive a set of differential equations that govern the separated motions of a porous solid and a compressible viscous fluid confined to it. In 1962 Biot extended the acoustic propagation theory in a wider context of the dynamics of anisotropic media (Biot,1962a, 1962b). The theory is applied to the materials where fluid and solid are of comparable densities. As follows from Table 1, the densities of the composites differ from that of the polypropylene no more than 10% and the criterion of the applicability of the Biot theory is fulfilled. The plots of frequency dependence of sound absorption were derived by Biot for different combinations of elastic constants and densities of the porous solid and that of fluid taking into account the additional apparent mass due to inertia coupling. The theoretical curves (Biot, 1956b) exhibit a maximum value of the absorption at a characteristic frequency which depends on the kinematic viscosity of the fluid and pore diameter. The maxima are very pronounced in the case of fluid - saturated porous solids characterized by the elastic properties and densities far from the "compatibility condition":

$$z_1 = \frac{V_1^2}{V_c^2} \cong 1 , \tag{5}$$

where $V_1$ stands for the velocity of the stress wave in a real anisotropic solid and $V_c$ represents this velocity when the relative motion between fluid and solid is prevented in some way. The less the fraction $z_1$ the more enhanced are the maxima.

Acoustic standing wave method (Markiewicz et al., 2009) is the most popular way to determine the sound absorptive power of the material sample subjected to the plane acoustic wave. In this method, plane acoustic waves are generated by a loudspeaker placed at one end of a tube while the other end is terminated by the material sample. Due to the reflections from the sample, standing wave is produced in the tube as the superposition of the incident wave with the amplitude $A$ and the reflected one with the amplitude $B$. The reflected wave is characterized by lower amplitude and shifted phase in comparison to the incident one. The probe microphone, moved inside the tube, receives the alternating acoustic pressure of minimum amplitude $p_{min}=A-B$ at the distance of $\lambda/4$ ($\lambda$ – wavelength) from the sample followed by the pressure of maximum value $p_{max}=A+B$ at $\lambda/2$. As the acoustic wave energy is proportional to the square of the sound pressure, the equation (3) can be written:

$$\alpha = \frac{E_i - E_r}{E_i} = 1 - \left[\frac{B}{A}\right]^2 = 1 - \left[\frac{p_{\max} - p_{\min}}{p_{\max} + p_{\min}}\right]^2, \tag{6}$$

where $E_r$ denotes the energy of the reflected wave. The equation (6) shows that the absorption coefficient $\alpha$ can be easily determined by means of the measurement of $p_{min}$ and $p_{max}$ amplitudes of the sound pressure inside the tube.

Figs. 1 and 2 show the results of the measurement of sound absorption coefficient $\alpha$ for pure polypropylene and the composites with lignocellulosic materials. The values of coefficient $\alpha$ were measured at the frequencies: 1000, 1800, 3000, 4000, 5000 and 6300 Hz according to the method mentioned above. As follows from the figures, the polypropylene is characterized by the relatively weak sound absorption. The values of coefficient $\alpha$ vary between 2 and 13% with the tendency to slightly decrease with increasing frequency. Fig. 1 shows the effect of the addition of 40 wt. % of hemp fillers on the absorption spectrum. The effect is dominant in the range of higher frequencies. Starting from the frequency of 3000Hz, the value of the coefficient $\alpha$ increases rapidly up to about 25% and maintains at this level up to 6300 Hz. Below the critical frequency 3000 Hz the effect of the addition of the hemp fillers is inconsiderable. One can even notice a small decrease of the coefficient $\alpha$ at 1800 Hz. Taking into account the fact that two different methods were used to prepare the samples No. 2 and No. 3 (extrusion and hydraulic pressing), one can conclude that the manufacturing procedure does not influence the sound absorption in the case of the composites with hemp filler. The composites containing long flax fibres, crumble rapeseed and crumble beech wood exhibit also better sound absorption in comparison with pure polypropylene but the frequency dependence of the coefficient $\alpha$ is quite different (Fig. 2). For these materials the maxima of $\alpha$ coefficient are observed at the frequencies of 3000 Hz or 4000 Hz. The differences in sound absorption by composites containing hemp and the ones based on fillers: flax, rapeseed straw, beech wood can be explained taking into account the Biot theory. The composites with hemp fillers seem to be nearest the "compatibility condition" among the investigated materials. The maxima of the absorption are not noticeable in the frequency range of the measurement, on the contrary to the remaining fillers. The composition of polypropylene and hemp results in such a combination of elastic constants and densities that the relative motion between filler and matrix is prevented. The discrepancies in sound absorption characteristics can result also from the filler morphology and its chemical composition. The width of hemp fibres (30 μm) is larger in comparison with flax fibres (20 μm). Moreover, hemp plant is known to have the dimensions of the anatomic cells larger than the remaining plants under examination. Hemp is distinguished for the highest contents of cellulose (75 wt. %) (Averous & Le Digabel, 2006). Flax contains 71 wt. % of cellulose. Beech and rapeseed are characterized by smaller contents of cellulose: beech – 42 wt. % (Nik–Azar et al., 1997) and rapeseed – from 35 to 40 wt. % (Paukszta, 2005, 2006). The contents of lignin in hemp (4 wt. %) is twice that of flax (2 wt. %) (Averous & Le Digabel, 2006). However, beech and rapeseed are known to have relatively large amount of lignin (~20 wt. %) (Paukszta, 2006). Flax is characterized by the contents of pectins (2 wt. %), fats (2 wt. %) and waxes (2 wt. %) which are twice those of hemp. It can be concluded that the higher contents of the cellulose in the hemp probably enables the sound absorption in the relatively wide frequency range.

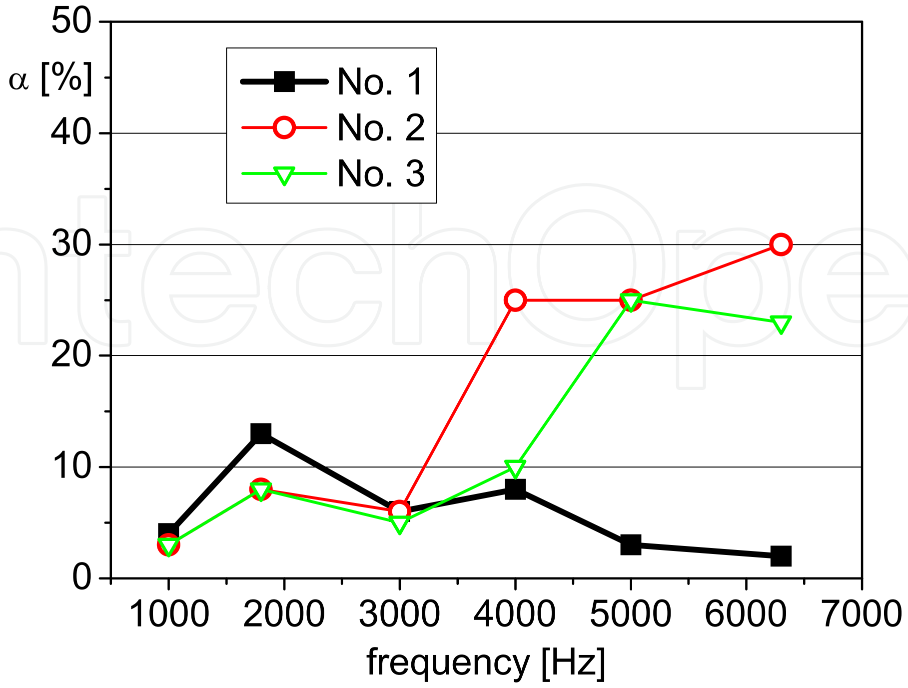

Fig. 1. Frequency dependences of sound absorption coefficient *α* for the samples: No. 1 - PP, No. 2 - PP + 40 wt. % of crumble hemp plant, No. 3 – PP + 40 wt. % of long hemp fibres

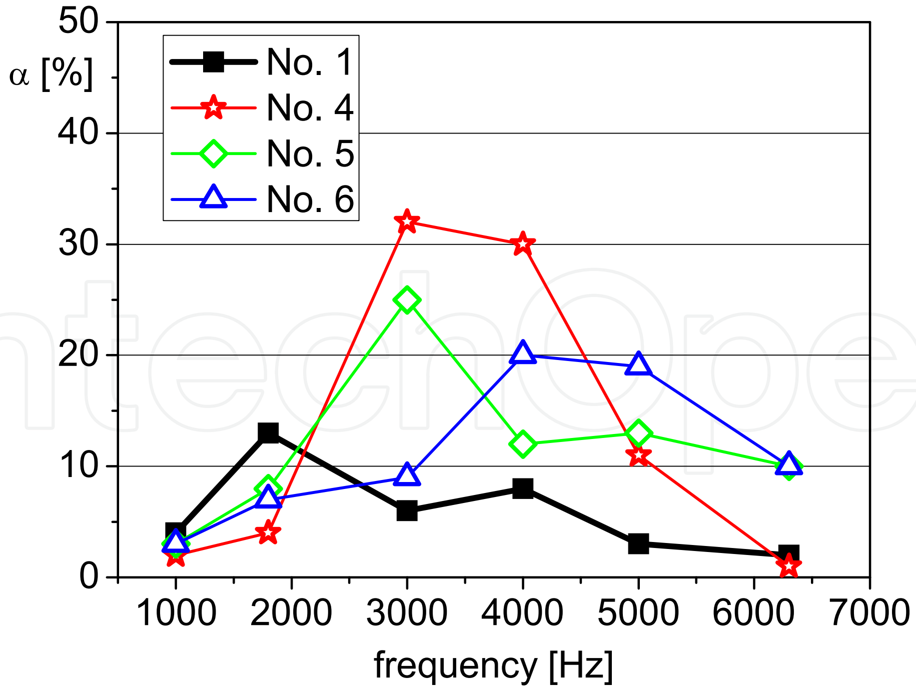

Fig. 2. Frequency dependences of sound absorption coefficient *α* for the samples: No. 1 – PP, No. 4 - PP + 40 wt. % of long flax fibres, No. 5 - PP + 40 wt. % of crumble rapeseed, No. 6 – PP + 40 wt. % of crumble beech wood

### 3.2 Effect of chemical treatment of lignocellulosic fillers on the acoustic properties of the composites

Chemical treatment of lignocellulosic filler as mercerization and modification with anhydride is often necessary to improve the mechanical properties of the composite due to better adhesion between the hydrophilic lignocellulosic filler and the hydrophobic polymer matrix (Borysiak & Garbarczyk, 2003; Borysiak & Doczekalska, 2005; Liu & Hu, 2008). Chemical treatment of the filler surface results in positive changes of the mechanical parameters of the composite as tensile strength, flexural strength and elongation at break (Bledzki et al., 2005; S.J. Kim et al., 2008; Mahlberg et al., 2001; Nachtigall et al., 2007; Yang et al., 2006). The interfacial region is the most vulnerable location to mechanical fracture. When subjected to the stress, it should show the ability to transmit the acoustic wave – induced tension from one phase to the other. Chemical treatment of the lignocellulosic filler is often necessary to get better the mechanical properties of the composite. Thus, the information about the influence of the modification on the acoustic properties is also very useful from the application point of view. In this chapter, the results of acoustic measurements for composites with crude and modified fillers are presented.

Figs. 3 – 6 show the values of sound absorption coefficient $\alpha$ measured for the composite samples No. 7 – 14, specified in Table 1. All composite samples, crude and modified, are compared with the pure polypropylene. The effect of the addition of the lignocellulosic filler to the polypropylene matrix is predominant in the frequency range above 3000 Hz where one can observe the improvement in the sound absorption of the order of more than ten percentage. All the investigated composite samples show the resonance characteristics of sound absorption with the maximum in the frequency range from 4000 to 6000 Hz. This behaviour can be ascribed to the combination of elastic constants and densities of both materials which makes possible the comparatively big relative motion between matrix and filler according to the Biot theory. The elastic constants and densities of the filler material result from its chemical composition. Our previous investigations, described in chapter 2.1, performed on composites with flax, hemp, rapeseed and beech fillers, showed that the increased sound absorption in a narrow frequency range can be probably related to the relatively small content of the cellulose. Now, we can confirm this presumption. The lignocellulosic materials used in this experiment are characterized by lower contents of cellulose: beech – 42 wt. % (Nik–Azar et al., 1997), pine – 41 wt. % (Gosselink et al., 2004), rapeseed Kaszub – 38 wt. % (Paukszta, 2005, 2006) and rape Californium - 37 wt. % (Paukszta, 2006) in comparison with hemp and flax which contain above 70 wt. % of cellulose (Averous & Le Digabel, 2006).

The effect of the modification of the fibre surface consists in the shift of the sound absorption maximum towards higher frequency range and is accompanied by the decrease of the sound absorption coefficient $\alpha$. The reduction of the coefficient $\alpha$ is not large. It amounts 2.5 % in the case of composites with pine wood and rapeseed straw Californium. For the samples with beech wood filler the coefficient $\alpha$ remains unaffected after modification. The exception is the composite containing the rapeseed straw Kaszub as a filler that shows the decreasing in the sound absorption of 7%. The reduction of the $\alpha$ coefficient value at the frequency related to the maximum of the sound absorption can be associated with the increase in the density of the composite after modification (Table 1) due to better adhesion between filler particles and polypropylene matrix. The fact implies that the specific acoustic impedance of

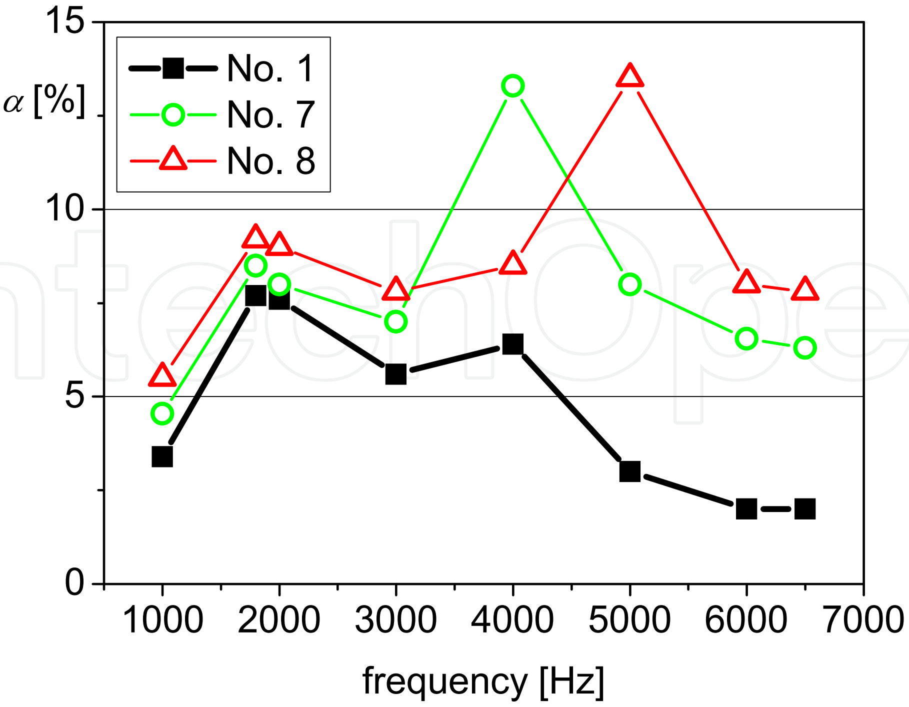

Fig. 3. Frequency dependences of sound absorption coefficient $\alpha$ for the samples: No. 1 – PP, No. 7 – PP + 20 wt. % of crude beech, No. 8 - PP + 20 wt. % of beech modified with succinic anhydride

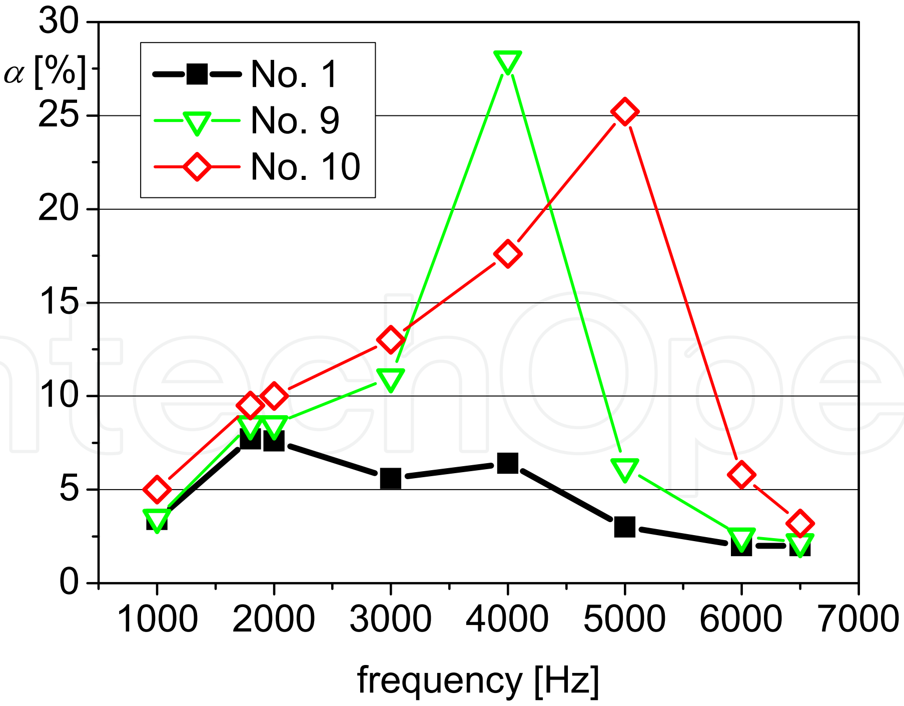

Fig. 4. Frequency dependences of sound absorption coefficient $\alpha$ for the samples: No. 1 – PP, No. 9 – PP + 20 wt. % of crude pine, No. 10 – PP + 20 wt. % of pine modified with succinic anhydride

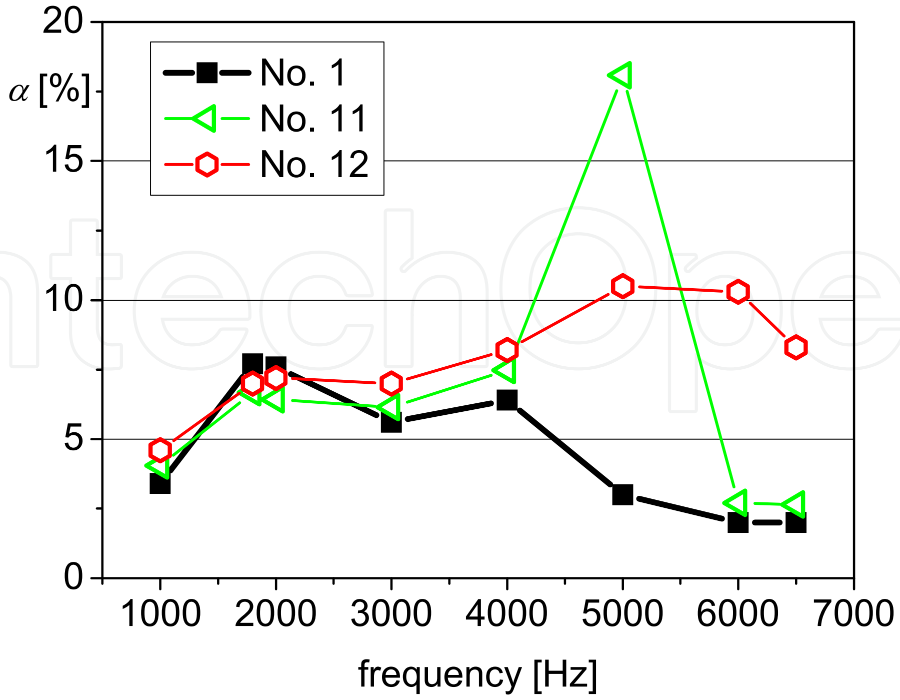

Fig. 5. Frequency dependences of sound absorption coefficient *α* for the samples: No. 1 – PP, No. 11 – PP + 30 wt. % of crude rapeseed straw Kaszub, No. 12 - PP + 30 wt. % of rapeseed straw Kaszub modified with acetic anhydride

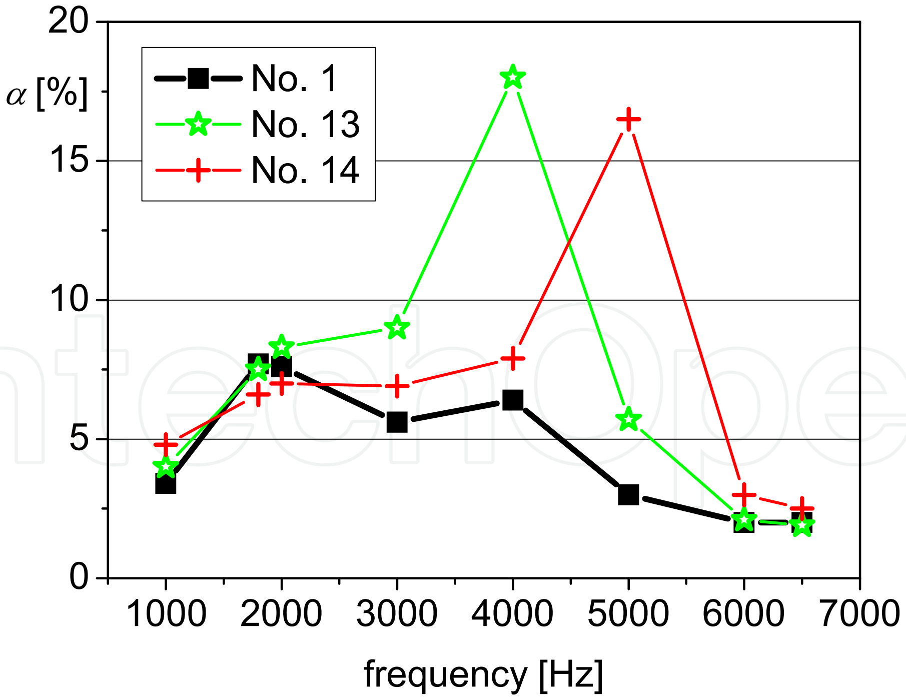

Fig. 6. Frequency dependences of sound absorption coefficient *α* for the samples: No. 1 – PP, No. 13 – PP + 30 wt. % of crude rapeseed straw Californium pure polypropylene (No. 1) and polypropylene composites with 30 wt. % of crude rapeseed straw Californium, No. 14 – PP + 30 wt. % of rapeseed straw Californium modified with acetic anhydride

the material, defined as the product of the density and the sound velocity (Lee & Chen, 2001), is increased. In turn, the higher the acoustic impedance the more acoustic energy is reflected from the material surface and the less of it can be absorbed. Better adhesion between polymer matrix and lignocellulosic filler leads to the better ability to transmit the acoustic wave – induced tension from one phase to the other. The process of the transmission in the composites with the modified fillers can be more rapid in comparison to that observed in non-modified composites and it can follow with higher frequency being a reason of a shift of the sound absorption band of about 1000 Hz.

## 4. Dielectric properties of the polypropylene reinforced with lignocellulosic materials

### 4.1 Effect of reinforcement of polypropylene with lignocellulosic materials on the dielectric properties

The dielectric properties of a material are determined by the polarizability of its molecules. There are three primary contributions to the electric polarization of a dielectrics: electronic, ionic and dipole reorientation – related (Uchino, 2000). The intensity with which each mechanism occurs depends on the frequency of applied electric field. The electronic polarization causes a displacement of the electrons with respect to the atomic nuclei and can follow alternating field with the frequencies up to $10^{12}$ – $10^{15}$ Hz. The ionic polarization relies on a displacement of the atomic nuclei relative to one another and responds up to $10^{9}$ – $10^{12}$ Hz. Both mentioned polarization mechanisms are related to the non-polar molecules. The third mechanism associated with the dipole reorientation is valid only in the case of polar molecules. It can follow with the frequency of alternating electric field up to $10^{6}$ – $10^{9}$ Hz. The dielectric permittivity $\varepsilon'$ of a material represents the ratio of the capacitance of a plane condenser filled with the dielectric to that of the same condenser under vacuum and is to calculate from the expression:

$$\varepsilon' = \frac{C \cdot d}{\varepsilon_0 \cdot S}, \tag{7}$$

where: $C$ is the capacitance of the condenser with the dielectric, $S$ stands for the area of the sample covered by the electrode, $d$ relates to the thickness of the sample and $\varepsilon_0 = 8.85 \cdot 10^{-12}$ F/m is the dielectric constant of the vacuum. The alternating current conductivity $\sigma_{a.c.}$ is described by the relationship:

$$\sigma_{a.c.} = \varepsilon_0 \cdot \omega \cdot \varepsilon' \cdot tg\delta , \tag{8}$$

where $\omega$ stands for the angular frequency.

The frequency dependencies of the dielectric constant $\varepsilon'$ measured at room temperature for the polypropylene and its composites with various lignocellulosic materials derived from hemp and flax are presented in Figs. 7 and 8. The effect of the reinforcement of the polypropylene with a lignocellulosic material consists in the increase of the dielectric permittivity $\varepsilon'$ over the whole measurement frequency range. The effect is predominant at lower frequencies. Pure polypropylene is a non-polar hydrophobic material which shows only instantaneous ionic and electronic polarization. Its dielectric permittivity $\varepsilon'$ holds

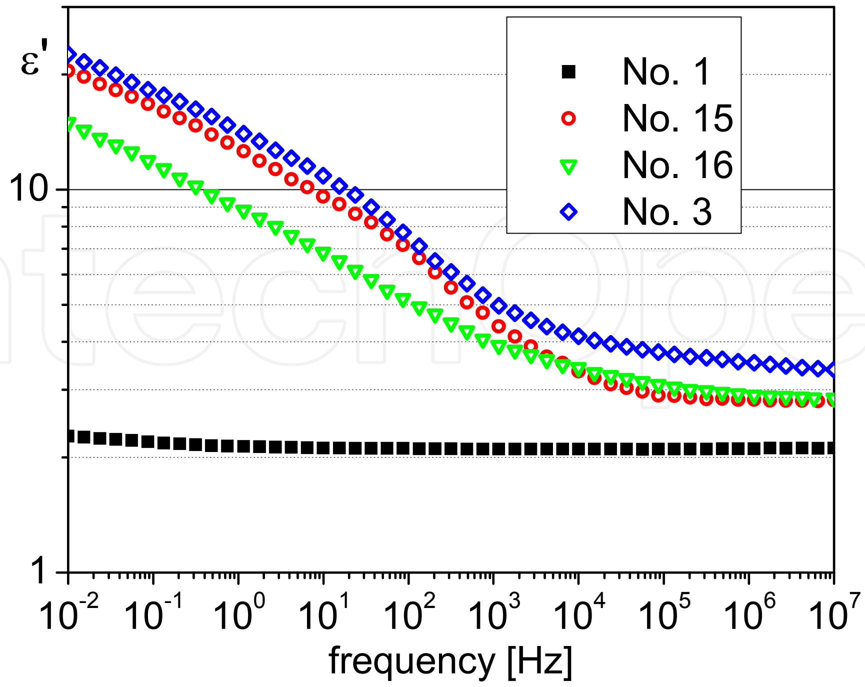

Fig. 7. Frequency dependences of dielectric permittivity $\varepsilon'$ for the samples: No. 1 – PP, No. 15 - PP+25 wt. % of short hemp fibres, No. 16 - PP + 25 wt. % of hemp shivers, No.3 – PP + 40 wt. % of long hemp

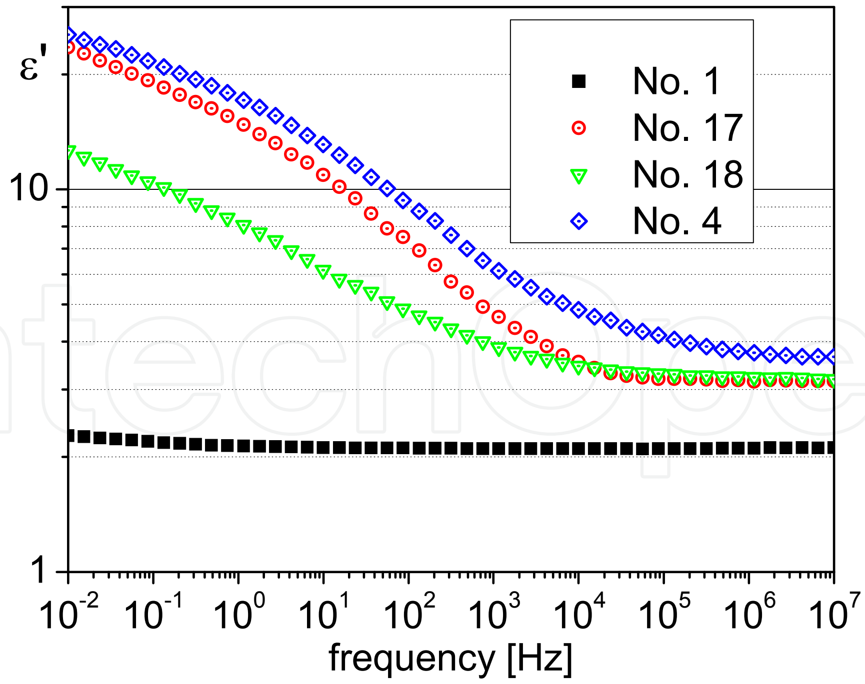

Fig. 8. Frequency dependences of dielectric permittivity $\varepsilon'$ for the samples: No. 1 – PP, No. 17 – PP + 25 wt. % of short flax fibres, No. 18. – PP + 25 wt. % of flax shivers, No. 4 - PP + 40 wt. % of long flax fibres

nearly constant value in the whole frequency range with a slender increase bellow $10^3$ Hz. The addition of a hydrophilic lignocellulosic material to the polypropylene entails the insertion of polar groups into the non-polar material giving the reason for rising the polarization related to the dipole reorientation. 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 dielectric permittivity of the composites $\varepsilon'$. The value of the dielectric permittivity $\varepsilon'$ increases with the content of the lignocellulosic material. In the higher frequency range, i.e. above $10^6$ Hz, the value of the relative dielectric permittivity $\varepsilon'$ 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. The differences in the dielectric permittivity $\varepsilon'$ values are most significant in the low frequency range, and they are observed also for the composites comprising 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 $\varepsilon'$ than the ones comprising short fibres (also 25 wt.%). The shivers are the lignified parts of the stems, separated from the fibres and they show lower capacity of moisture absorption. This fact can indicate a smaller number of polar groups and lower polarization related to dipole reorientation.

Plots of the reciprocal of dielectric permittivity $1/\varepsilon'$ versus volume fraction of lignocellulosic material derived from hemp and flax at the frequency of 1 MHz (Fig. 9) are linear for the

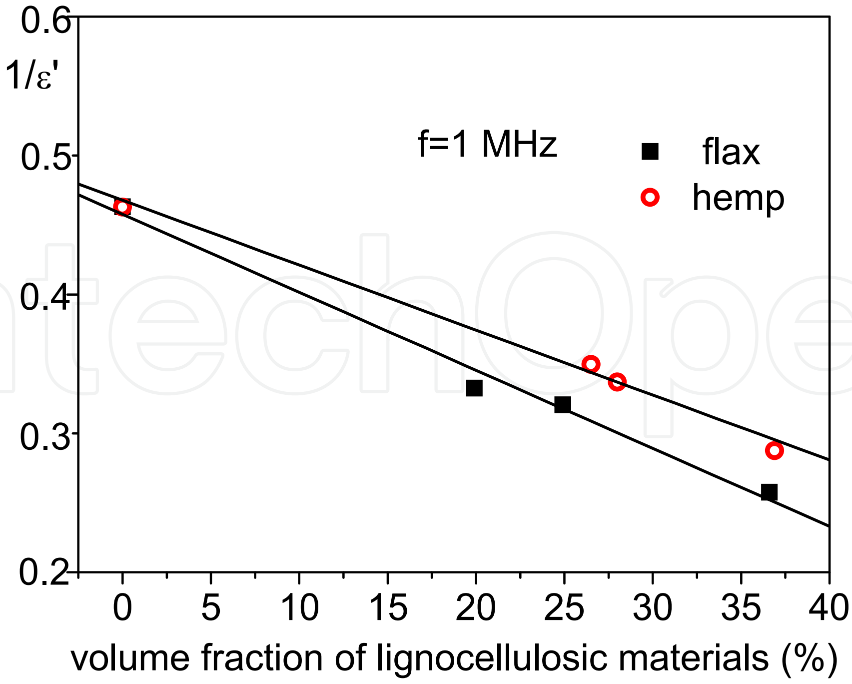

Fig. 9. Reciprocal dielectric permittivity $1/\varepsilon'$ versus volume fraction of lignocellulosic material derived from hemp and flax (Markiewicz et al., 2009)

applied contents of filler. A number of numerical relations as Lichtenecker, Maxwell Garnet, Jayasundere, Poon-Shin equations or Effective Medium Theory were developed by researchers to predict the effective dielectric constant of the composites (Subodh et al., 2007). All the mentioned models differ at higher volume fraction of the filler and they can be replaced by the linear fit at lower filler content, when the permittivity contrast between matrix and filler is low, particularly. For our investigated samples with the volume fraction less than 0.4 the dependence of the reciprocal of the dielectric permittivity on the volume fraction can be approximated by the linear fit (Sareni et al., 1997). The obtained results are in agreement with those presented by Jacob M. at al. (Jacob, 2006) for sisal-oil palm hybrid biofibre reinforced natural rubber biocomposites.

The reinforcement of the polypropylene with the lignocellulosic material results in the increase of ac conductivity (Figs. 10 and 11). The random distribution of the lignocellulosic fillers in the polypropylene matrix enables rearrangement of the fibres in a chain structure which ensures better carrier mobility in the presence of electric field. The frequency dependence of the electrical conductivity is described by the expression (Jonsher, 1997):

$$\sigma(\omega) \propto \omega^n . \tag{9}$$

The exponent $n$ is close to 0.5 for pure polypropylene and points to diffusive carrier transport. For the composites, $n$ changes from ~ about 0.5 at the low frequencies to ~ 1 at the high frequencies. This fact proves the existence of diffusive as well as hopping carrier transport. In the lowest frequency range the composite samples show the frequency-independent behavior pointing to the ohmic conduction. This property makes them to be better antistatic material than pure propylene.

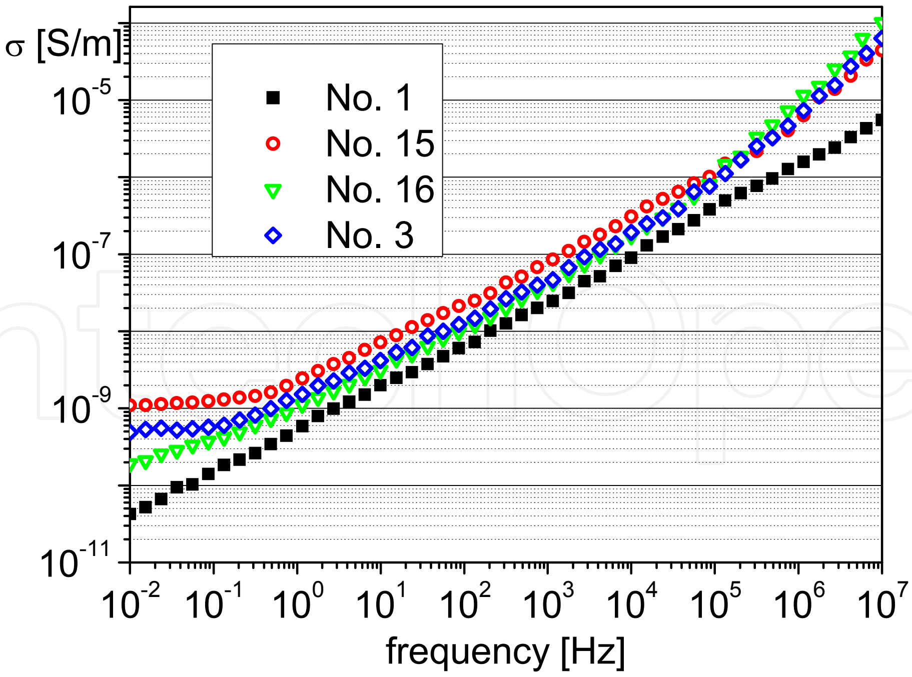

Fig. 10. Frequency dependences of ac conductivity $\sigma'$ for the samples: No. 1 – PP, No. 15 – PP + 25 wt. % of short hemp fibres, No. 16 – PP + 25 wt. % of hemp shivers, No. 3 - PP+40% of long hemp fibres

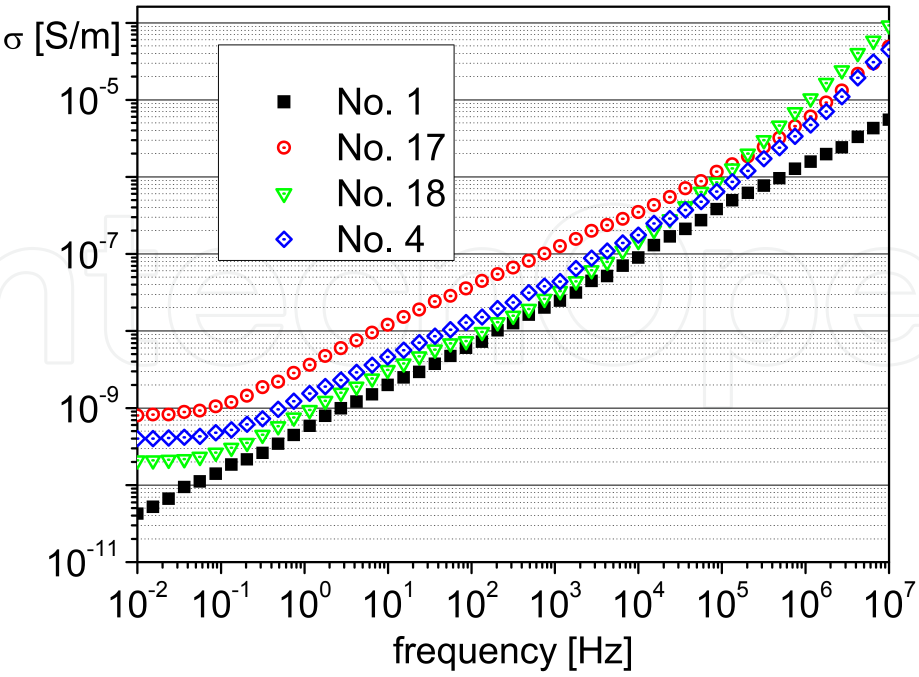

Fig. 11. Frequency dependences of ac conductivity $\sigma'$ for the samples: No. 1 – PP, No. 17 – PP + 25 wt, % of short flax fibres, No. 18 - PP+25 wt. % of flax shivers, No. 4 – PP + 40 wt. % of long flax fibres

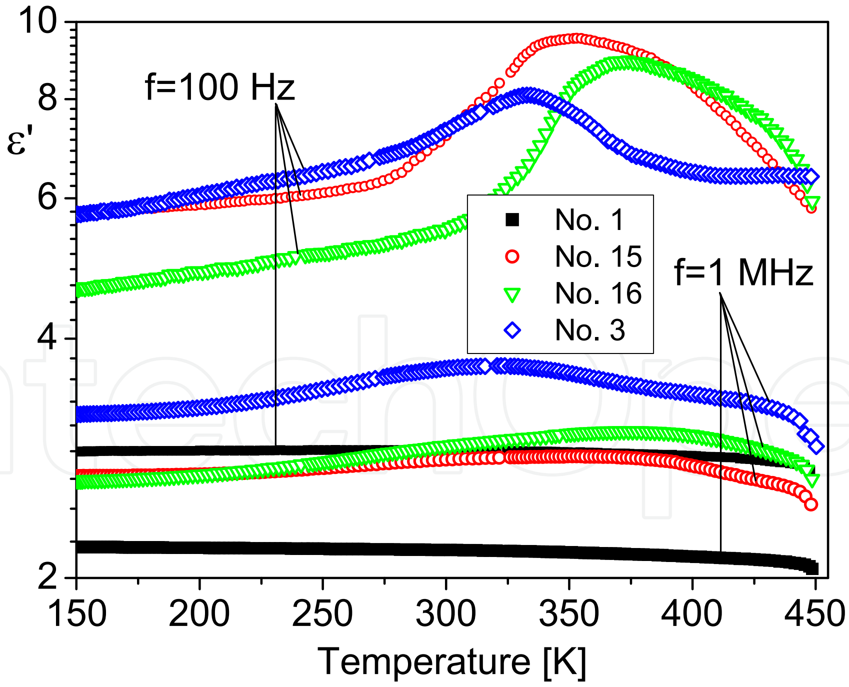

Fig. 12. Temperature dependences of dielectric permittivity $\varepsilon'$ obtained at frequencies 100 kHz and 1 MHz for the samples: No. 1- polypropylene PP; No. 15 - PP + 25 wt.% of short hemp fibres; No. 16 – PP + 25 wt.% of hemp shivers; No. 3 – PP + 40 wt.% of long hemp fibres

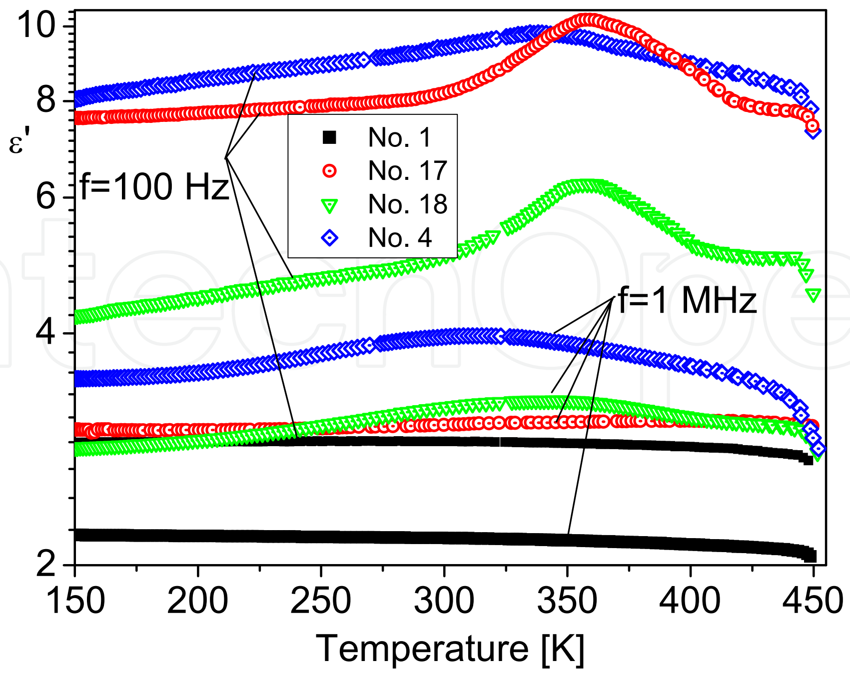

Fig. 13. Temperature dependences of dielectric permittivity $\varepsilon'$ obtained at frequencies 100 kHz and 1 MHz for the samples: No. 1 - polypropylene PP; No. 17 - PP + 25 wt.% of short flax fibres; No. 18 – PP + 25 wt.% of flax shivers; No. 4 – PP + 40 wt.% of long flax fibres

The temperature variations of the dielectric permittivity $\varepsilon'$ investigated for the polypropylene as well as the composite samples with hemp and flax are presented in Figs. 12 and 13. The value of $\varepsilon'$ measured for polypropylene is nearly independent on the temperature up to the melting point at 438 K (Doh, 2005). The dielectric permittivity $\varepsilon'$ of the composites increases with the temperature up to the maximum associated with the traces of water, and then decreases. The position of the maximum is determined by the contents of chemically bounded water which cannot be removed during the preparation. The maximum is shifted towards higher temperatures in the case of higher contents (Chand, 2005). As follows from Figs. 12 and 13, the technique of hydraulic pressing, applied for fabrication of composite samples with long fibers, implied the lowest content of water. In the vicinity of the melting point of the polypropylene, a rapid fall of the $\varepsilon'$ value is visible. The dielectric permittivity $\varepsilon'$ decreases with the increase of the frequency, as is seen for two frequencies: 100 Hz and 1 MHz. A weak dependency of dielectric permittivity $\varepsilon'$ on the temperature was observed for the frequency of 1 MHz, particularly in the case of polypropylene composites with short fibres. This feature is the evidence that the composites can be recommended for application in the high frequency range because of the stable dielectric permittivity $\varepsilon'$ value.

The dielectric loss factors $\varepsilon''$ of the pure polypropylene and the composites containing the lignocellulosic materials derived from hemp and flax are presented in Figs. 14 and 15 as a function of the temperature for the frequency of 1000 Hz. Pure polypropylene is known to exhibit two characteristic features (Kotek et al., 2005): a glass relaxation peak around 263 K and a high – temperature (~323 K) shoulder associated with chain relaxation in the crystalline phase. These features cannot be detected by the Dielectric Relaxation

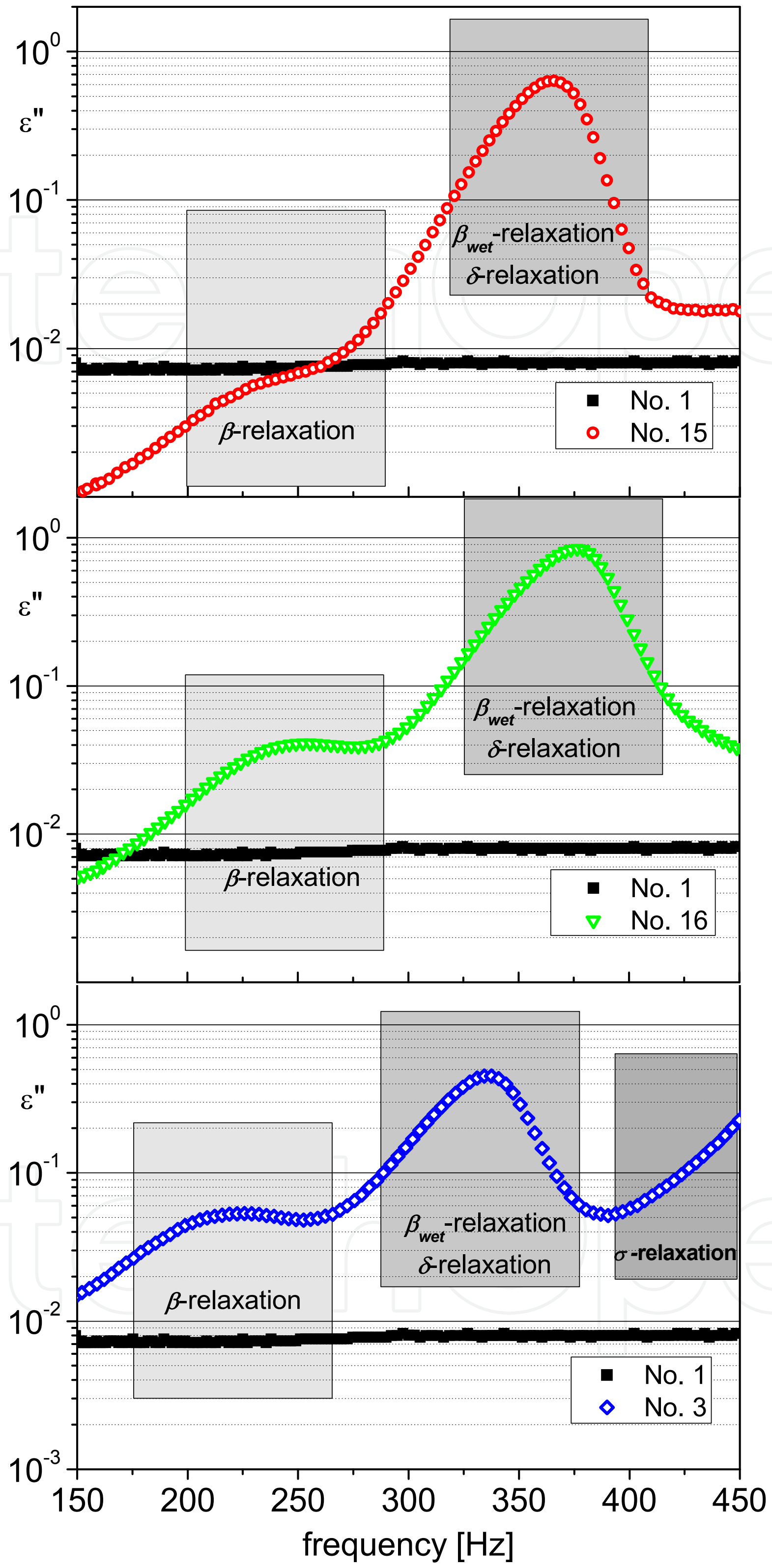

Fig. 14. Temperature dependences of dielectric losses $\varepsilon''$ obtained at frequency 1000 Hz for the samples: No. 1- polypropylene PP; No. 15 - PP + 25 wt.% of short hemp fibres; No. 16 – PP + 25 wt.% of hemp shivers; No. 3 – PP + 40 wt.% of long hemp fibres

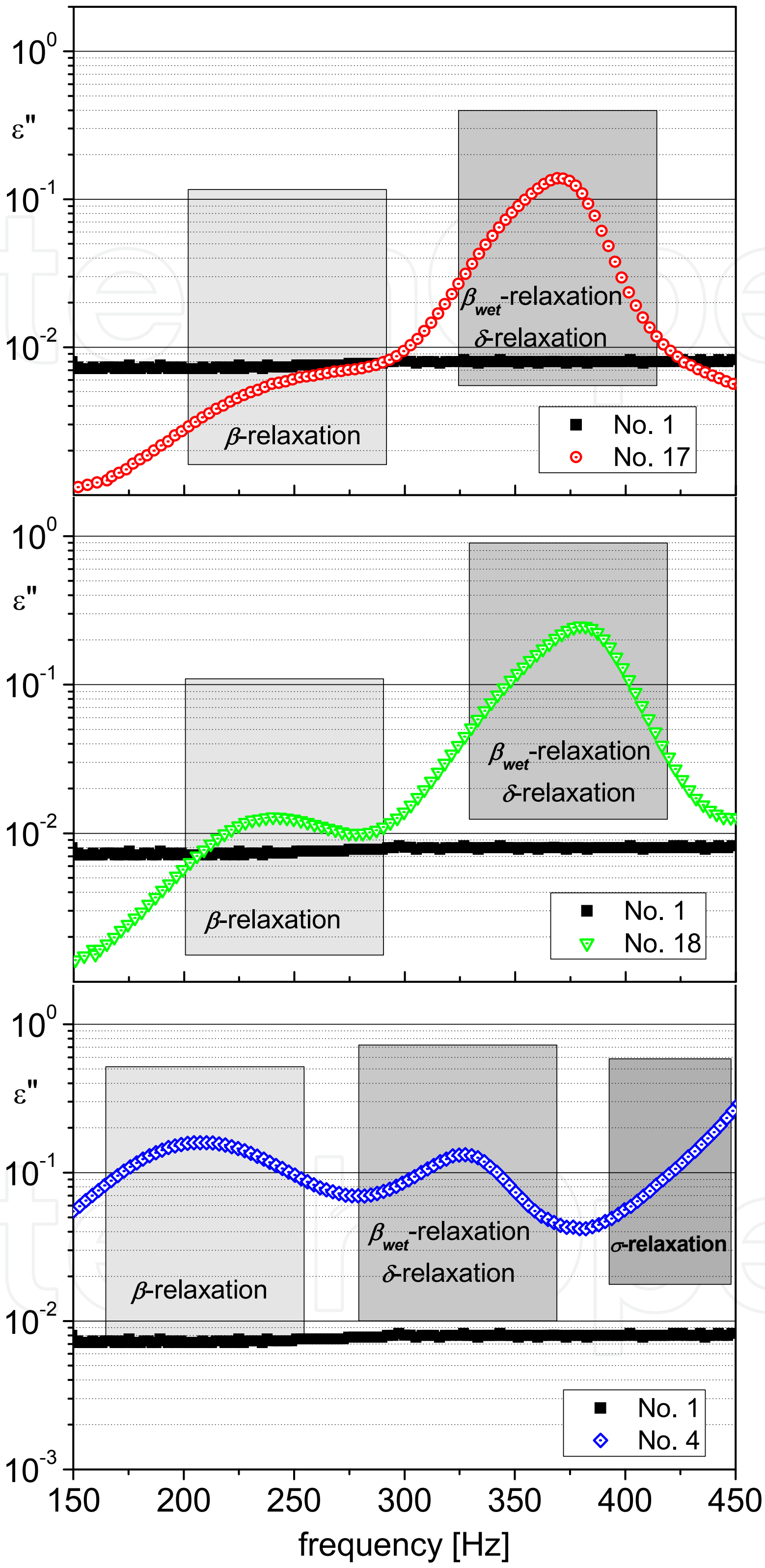

Fig. 15. Temperature dependences of dielectric losses $\varepsilon''$ obtained at frequency 1000 Hz for the samples: No. 1- polypropylene PP; No. 17 - PP + 25 wt.% of short flax fibres; No. 18 – PP + 25 wt.% of flax shivers; No. 3 – PP + 40 wt.% of long flax fibres

Spectroscopy (DRS) method without special modification of the polymer structure or introducing polar groups in the structure because polypropylene has no appreciable molecular dipoles and is not dielectrically active. The non-polar polypropylene does not show any anomalies in the dielectric loss spectrum. However, the temperature dependences of the dielectric loss factors $\varepsilon''$ measured for the composites are strongly influenced by the contribution of the lignocellulosic materials. Both kinds of the filler modify the dielectric absorption spectrum in the same way. In the low temperature range (from about 200 to 270 K) one can observe the maxima of $\varepsilon''$ ascribed to the $\beta$ - relaxation process in the cellulose which is the main component of each lignocellulosic material. The $\beta$ - relaxation is interpreted as a local motion of chain segments via the glucosidic linkages (Einfeldt et al., 2001). Above room temperature (from about 300 to 400 K) one can notice high relaxation peaks. Based on the shape of these peaks, one can deduce that two relaxation processes overlap in this temperature range: $\beta_{wet}$ – the relaxation associated with the orientational motion of both cellulose and water (Baranov et al., 2003; Einfeldt et al., 2001) and $\delta$ - the relaxation ascribed to the motion of the end groups in branched polymers (Einfeldt et al, 2001) present in the lignocellulosic material (hemicellulose, pectin, lignin). Because the intensity of the $\delta$-relaxation is significantly smaller than that of $\beta_{wet}$ - relaxation (Einfeldt et al., 2001), one can state that in the polypropylene – lignocellulosic materials composites the $\beta_{wet}$ - relaxation is disturbed by the $\delta$ - process. In the case of the composites with long fibres derived from flax as well as hemp, the increase in the dielectric losses was observed in the highest temperature range (above 420 K). The effect results from the electric conductivity and is called $\sigma$ - relaxation (Einfeldt et al., 2001). The losses due to the electric conductivity are ascribed to charge carrier hopping between localized sites in amorphous solids. The fact that the $\sigma$ - relaxation was observed only for the composites with long fibres confirms the conclusion from (Einfeldt et al., 2001) that the activation energy for the carrier hopping increases when the amount of water is reduced. The intensity of $\beta_{wet}$ – relaxation is proportional to the contents of water. The low intensity of $\beta_{wet}$ – relaxation in the composites with long fibers is a reason for relatively high strength of $\beta$ – relaxation in comparison with that observed for other investigated samples where the $\beta$ – process is suppressed by the $\beta_{wet}$ – relaxation and the position of high intensive $\beta_{wet}$ – relaxation peak in higher temperature range masks the $\sigma$ - process.

### 4.2 Effect of chemical treatment of lignocellulosic fillers on the dielectric properties of the composites

The effect of chemical treatment is dominant in the low frequency range, i. e. from $10^{-2}$ Hz to 1 kHz. It can be opposite for various kinds of lignocellulosic fillers. Figs. 16 and 17 show the frequency dependences of dielectric permittivity $\varepsilon'$ obtained at room temperature for the polypropylene composites containing crude, mercerized and modified lignocellulosic fillers derived from pine and beech wood as well as two kinds of rapeseed straw. The effect observed for the pine and beech wood (Fig. 16) consists in the increase of the dielectric permittivity $\varepsilon'$ value. The modification with maleic anhydride causes greater increase than the mercerization. In the case of both kinds of rapeseed straw: Kaszub and Californium (Fig. 17), the mercerization decreases the dielectric permittivity $\varepsilon'$ value several times and the modification with maleic anhydride reduces $\varepsilon'$ value to that measured for composites with crude pine and crude beech. One should take into account the location of the $\beta_{\mathrm{wet}}$ - relaxation in the vicinity of room temperature to explain the opposite influence of chemical treatment

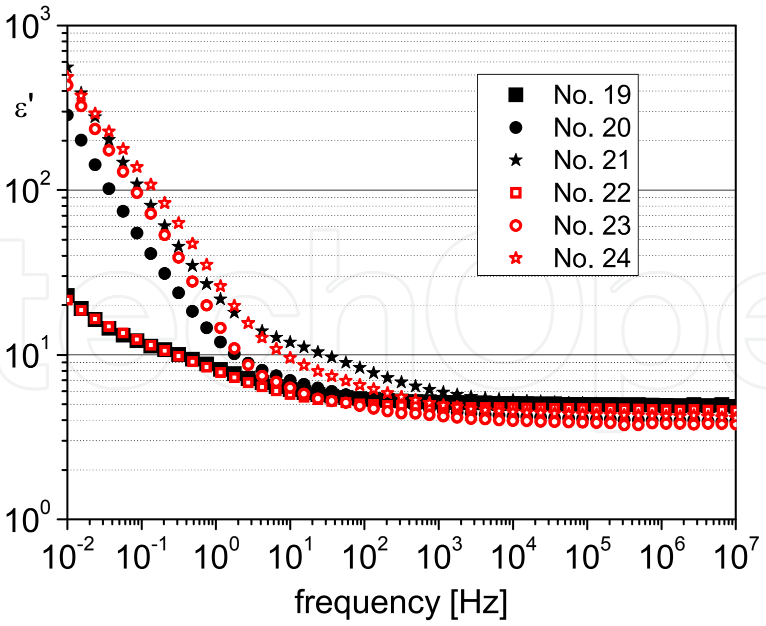

Fig. 16. Frequency dependences of dielectric permittivity *ε'* obtained for the samples: No. 19 – PP + 30 wt. % of crude beech; No. 20 – PP + 30 wt. % of mercerized beech; No. 21 – PP + 30 wt. % of beech modified with maleic anhydride; No. 22 – PP + 30 wt. % of crude pine; No. 23 – PP + 30 wt. % of mercerized pine; No. 24 – PP + wt. 30% of pine modified with maleic anhydride

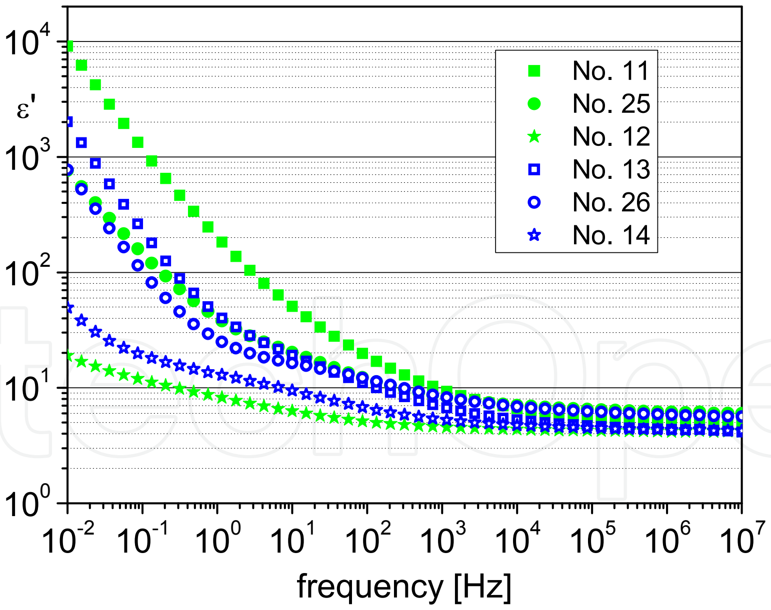

Fig. 17. Frequency dependences of dielectric permittivity *ε'* obtained for the samples: No. 11 – PP + 30 wt. % of crude rapeseed straw Kaszub; No. 25 – PP + 30 wt. % of mercerized rapeseed straw Kaszub; No. 12 – PP + 30 wt. % of rapeseed straw Kaszub modified with acetic anhydride; No. 13 – PP + 30 wt. % of crude rapeseed straw Californium; No. 26 – PP + 30 wt. % of mercerized rapeseed straw Californium; No. 14 – PP + 30 wt. % of rapeseed straw Californium modified with acetic anhydride

on the dielectric permittivity $\varepsilon'$ value at low frequencies. The effect is associated with the ability of moisture absorption. The swollen structure of crude rapeseed straw facilitates the moisture absorption and the $\varepsilon'$ value for this material is the biggest one. The mercerization and the modification with acetic anhydride make the structure more rigid and the moisture absorption is limited. However, in the case of beech and pine the crystalline structure of the cellulose confined to the fibres gets be swelled just after chemical treatment.

## 5. Conclusion

The investigated composite materials based on polypropylene matrix and lignocellulosic fillers can be recommended for application in building and automotive industry because of their good sound absorptive power as well as in packaging electronics due to their dielectric properties. Addition of lignocellulosic materials to the pure polypropylene increases the sound absorption coefficient by about 20% in the frequency range above 3000 Hz. The frequency dependence of the absorption coefficient can be shaped by the proper choice of the lignocellulosic filler. The shift of the sound absorption band due to chemical treatment of the lignocellulosic fibres seems to be suitable for manufacturing the composites with extended sound absorption frequency range as the effect of the adaptation of mixed filler containing the treated and untreated lignocellulosic material. The increased dielectric permittivity of the composites, in comparison with the polypropylene, is preferable in the application in the field of packaging industry. The polypropylene-lignocellulosic materials composites assure the thermal stability of the dielectric permittivity above 1 MHz and better antistatic properties than pure polypropylene.

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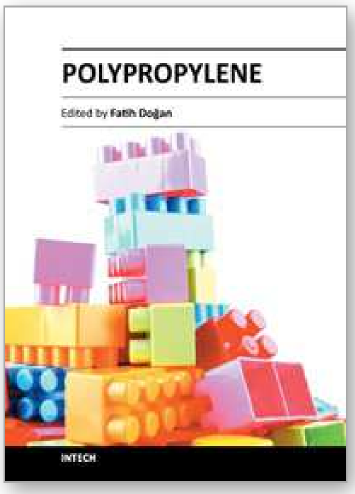

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