EWA MARKIEWICZ[1)*)], SŁAWOMIR BORYSIAK[2)], DOMINIK PAUKSZTA[2)]

Polypropylene-lignocellulosic material composites as promising sound absorbing materials

Summary — Composites made from polypropylene and lignocellulosic materials derived from plants were subjected to acoustic and mechanical investigations. Sound absorption coefficients for these composites were measured using the acoustic standing wave method in the frequency range from 1000 to 6500 Hz. This work shows that the frequency dependence of the sound absorption coefficient obtained for pure polypropylene can be modified by proper choice of the filler added to the polypropylene matrix. Fillers derived from hemp plant cause significant increase in the absorption coefficient starting from the frequency above 3000 Hz. The fillers obtained from rapeseed straw, beech and flax are recognized to suppress the sound in the frequency range from 3000 to 4000 Hz. The investigated composites can be recommended for application in the automotive industry and building as the construction materials absorbing the undesired noise.
Key words*:* composites, lignocellulosic fillers, sound absorption coefficient, sound absorbers.

KOMPOZYTY POLIPROPYLENU Z NAPEŁNIACZAMI LIGNOCELULOZOWYMI JAKO MATERIAŁY POCHŁANIAJĄCE DŹWIĘK

Streszczenie — W pracy przedstawiono wyniki badań właściwości akustycznych i mechanicznych kompozytów złożonych z matrycy polipropylenowej i materiałów lignocelulozowych uzyskanych z roślin konopi, lnu, rzepaku i drzewa bukowego. Współczynnik pochłaniania dźwięku wyznaczono metodą akustycznej fali stojącej w zakresie częstotliwości od 1000 do 6500 Hz w układzie pomiarowym Standing Wave Apparatus 4002 firmy Brüel&Kjaer (rys. 1). Zależność częstotliwościowa współczynnika pochłaniania dźwięku α dla czystego polipropylenu może być modyfikowana przez odpowiedni dobór napełniacza lignocelulozowego. Napełniacze uzyskane z rośliny konopi powodują znaczny wzrost absorpcji dźwięku w zakresie częstotliwości powyżej 3000 Hz (rys. 2). Współczynnik pochłaniania dźwięku kompozytów z tymi napełniaczami wzrasta do ~25 % i utrzymuje się na tym poziomie wraz ze wzrostem częstotliwości. Natomiast dodanie napełniaczy otrzymanych z lnu, słomy rzepakowej i drewna bukowego do matrycy polipropylenowej przyczynia się do bardziej rezonansowej charakterystyki pochłaniania dźwięku z wyraźnym maksimum absorpcji (α powyżej 20 %) w zakresie częstotliwości od 3000 do 4000 Hz (rys. 3). Wykonane badania potwierdzają możliwość zastosowania kompozytów zawierających materiały lignocelulozowe w budownictwie oraz przemyśle samochodowym z uwagi na korzystną właściwość pochłaniania dźwięku.
Słowa kluczowe: kompozyty, napełniacze lignocelulozowe, współczynnik pochłaniania dźwięku, absorbery dźwięku.

Composites made of thermoplastic polymers and natural lignocellulosic materials are known to exhibit many advantages [1—6]. They are characterized by very good mechanical resistance [7—11] and can be ecological alternatives [12, 13] to the composites reinforced with glass fibres or other mineral fillers. The composites with natural fillers are environmentally friendly during production and processing as well, as they can be subjected to the energy and material recycling. The addition of lignocellulosic materials resulted in a significant decrease in such important parameters such as heat release rate (*HRR*) peak and mass loss rate (*MLR*) [14, 15].

One important feature of biocomposites is their sound absorption capacity. The pure and unporous materials applied in the industry are characterized by weak sound absorption properties. The sound absorption coefficient (α) of the materials commonly used in the construction: ceramic tile, concrete and cement is below 5 % in the frequency range from 125 to 8000 Hz [16, 17]. The wood-based materials such as fiberboard, plywood as well as wooden floor show α also about 5 % [17, 18]. Synthetic polymers such as polyethylene, poly(vinyl chloride) are good reflective shields for sound waves,

[1)] Institute of Molecular Physics, Polish Academy of Sciences, ul. Smoluchowskiego 17, 60-179 Poznań, Poland.
[2)] Institute of Technology and Chemical Engineering, Poznan University of Technology, ul. M. Sklodowskiej-Curie 2, 60-965 Poznan, Poland.
[*)] Corresponding author; e-mail: ewamar@ifmpan.poznan.pl

particularly in the high frequency range [19]. Their impedance rapidly raises with the frequency resulting in a decrease in α value below 1 % for the frequencies above 1000 Hz. Better acoustic absorption properties are exhibited by porous materials used in construction and also in the automotive industry. Their sound absorption usually increases with the frequency and their coefficients can reach values from 25 to 80 % in the frequency range from 1000 to 5000 Hz in case of aluminum foam, but the frequency dependence is characterized by the resonance peak [20]. For comparison, traditionally used polyurethane foams show lower sound absorption performance because their coefficients vary from 15 to 40 %. Sound absorbers made of rubber particle layers are known to have good properties in the lower frequency range. Their α in the frequency range from 200 to 1600 Hz is relatively high (30—40 %) [18]. Excellent sound absorption properties are shown by glass wool. Its α reaches about 90 % in the frequency range from 2000 to 5000 Hz [21].

The composite materials, developed in recent years, provide good sound absorption over a wide frequency spectrum. Their sound absorption capacity can be significantly increased in comparison to that of the initial matrix. For example, α of rice straw-wood particle system boards can be higher in the middle and high frequency range (increases from 15 % at 1000 Hz to 70 % at 8000 Hz) than that of commercial wood-based materials, such as particleboard (varies from 15 % at 1000 Hz to 50 % at 8000 Hz), fiberboard ($\alpha < 5\%$) or plywood ($\alpha < 5\%$) [16]. Sound absorption of cement concrete with addition of hollow ceramic-micro balloons can be enhanced up to 30 % in comparison to ~10 % for the matrix material [16].

The aim of the work is to measure and compare the sound absorption capacity of composites made of thermoplastic polymer with or without the addition of different lignocellulosic materials. For estimation of acoustic properties, the sound absorption coefficient was adopted. The electrical and mechanical properties of composites made of thermoplastic polymers reinforced with natural fibres are well known [7—11, 22—24], however no information is available on the acoustic properties of the composites.

EXPERIMENTAL

Materials

In this work the following materials were used:

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

— lignocellulosic materials: hemp and flax (long fibres), hemp plant, hemp shivers, rapeseed straw, beech wood (crumble-materials) were used as filling materials.

Sample preparation

Two different methods were used to make the composites. The first one consisted in mixing of crumble-lignocellulosic materials with polypropylene granulate in different proportions (25—40 wt. % of natural component). After that, the extrusion was carried out using a "Fairex" (McNell Akron Repiquetn, France) single-screw extruder with L/D = 25. The composite material was obtained in a granulated form [25].

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 acoustic experiments.

The composites containing the long hemp or flax fibres were produced in a different way. A technique of hydraulic pressing at temp. 200 °C under load of 3000 kG, described in our patent [26], was used instead.

Finally, the samples took the shape of discs. Two tubes are used in measuring setup for acoustic investigations, therefore two series of samples of different diameters (28.8 mm or 98.8 mm) and the same thickness (5 mm) were made.

Table 1 specifies all the samples prepared.

T a b l e 1. Specification of the samples investigated

Number of sample	Kind of material	Density kg/m^3
1	PP	881.8
2	PP + 40 % of crumbled hemp plant	872.8
3	PP + 40 % of long hemp fibres	927.9
4	PP + 40 % of long flax fibres	934.6
5	PP + 40 % of crumbled rapeseed straw	918.8
6	PP + 40 % of crumbled beech wood	803.5
7	PP + 25 % of crumbled hemp harls	911.3

Determination of the sound absorption coefficient

Theoretical background

The sound absorption coefficients (α) of the investigated materials were determined by the standing wave method. 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 sample. Due to the reflections from the sample, standing wave is produced in the tube as the superposition of the incident and reflected waves. 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 maximum amplitude (p_{max}) followed by the pressure of minimum value (p_{min}) with increasing the distance from the sample. The acoustic pressure measurement at the distances determined by p_{max} and p_{min} allows to calculate the standing wave ratio (n) which means the relationship between the maximum and the minimum sound pressure in the tube [27]

$$n = \frac{p_{\max}}{p_{\min}} \quad (1)$$

In turn, the ratio n is known to be involved into the expression for the absolute value of the reflection factor (r) [25]

$$r = \frac{n-1}{n+1} \quad (2)$$

The r^2 value characterizes the ratio of the reflected acoustic energy (E_r) to the incident energy (E_i) according to the formula

$$r^2 = \frac{E_r}{E_i} \quad (3)$$

The α coefficient, defined as the ratio of the energy absorbed by the sample (E_a) to the total energy incident on the sample (E_i), can be obtained from the expression

$$\alpha = \frac{E_a}{E_i} = \frac{E_i - E_r}{E_i} = 1 - \left(\frac{n-1}{n+1}\right)^2 \quad (4)$$

The eq. (4) shows that α coefficient can be easily determined by means of the measurement of p_{min} and p_{max} amplitudes of the sound pressure inside the tube.

Measuring setup

The measuring setup used for determination of the absorption coefficients is shown in Figure 1 [28]. All the equipment is made by the acoustic firm Brüel&Kjaer. The acoustic wave is propagated along the tube of the Standing Wave Apparatus 4002. The enlargement in Fig. 1 shows the placement of the sample in the holder fastened to one end of the tube.

The loudspeaker, mounted in the box screwed to the opposite end, is excited by the Sine Wave Generator 1023, covering the frequency range from 10 to 20 kHz. The output voltage from the probe microphone is conducted to the Measuring Amplifier 2606 equipped with Heterodyne Slave Filter 2020. The measurement scale of the amplifier enables the α coefficient to be read out directly in percentage. Although the frequency range of the generator as well as the amplifier is very broad the frequency range of the standing wave method is limited. As follows from the theoretical background, the necessity of the positioning of the probe microphone at the distances related to p_{min} and p_{max} implies the condition that the length of the tube must be greater than 0.5 of the wavelength. Whereas the highest frequency is determined by the diameter of the sample (internal diameter of the tube) that should be less than 0.5 of the wavelength to avoid the transverse resonances with the tube. In accordance with the above assumptions we made the measurement α coefficient in the frequency range from 1000 to 6500 Hz using two measuring tubes of different diameters: the largest one (diameter = 99 mm, length = 1 m) designed to be usable in the frequency range from 90 to 1800 Hz and the smaller one (diameter = 29 mm, length = 280 mm) covering the range from 800 to 6500 Hz.

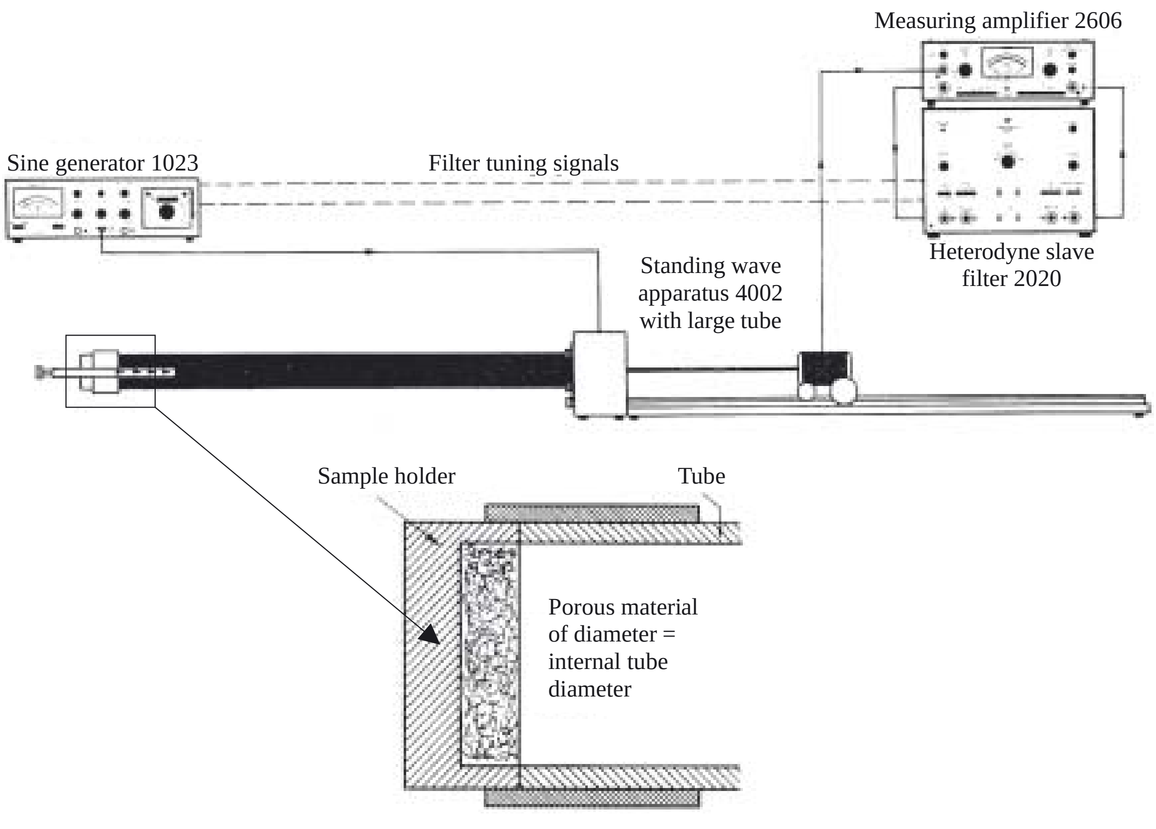

Fig. 1. Measuring setup (Standing Wave Apparatus 4002, Brüel&Kjaer) for determination of the absorption coefficient (α)

RESULTS AND DISCUSSION

Figures 2—4 show the results of α coefficient measurements for the frequencies: 1000, 1800, 3000, 4000, 5000 and 6500 Hz. Pure polypropylene was chosen as the reference material. Its sound absorption coefficient tends to slightly decrease with increasing frequency (curve 1 in the Figs. 2—4). The values of coefficient α range from 2 to 13 %.

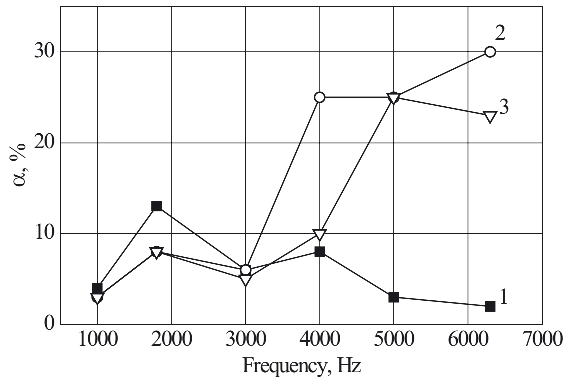

Fig. 2. Frequency dependence of sound absorption coefficient for pure polypropylene and polypropylene composites with 40 wt. % of hemp filler; description of curves correspond to numbers of samples defined in Table 1

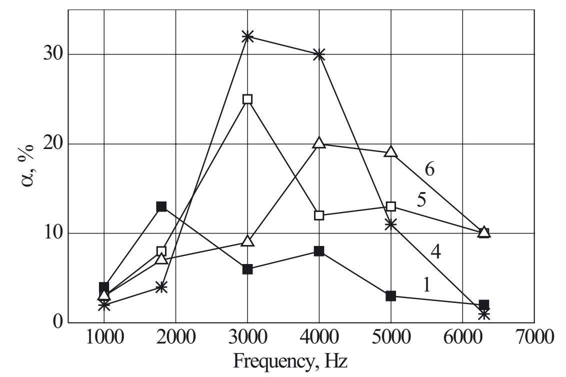

Fig. 3. Frequency dependence of sound absorption coefficient for pure polypropylene and polypropylene composites with 40 wt. % of flax, rapeseed or beech fillers; description of curves correspond to numbers of samples defined in Table 1

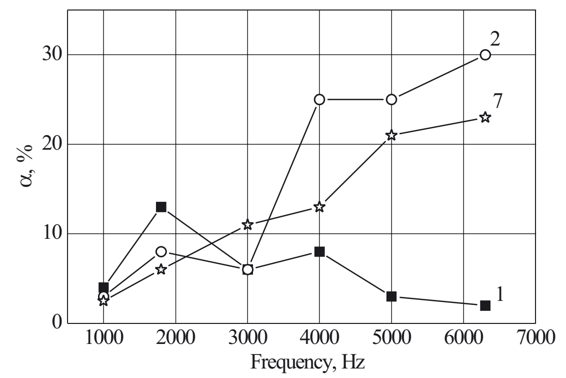

Fig. 4. Frequency dependence of sound absorption coefficient for polypropylene composites with hemp fillers; description of curves correspond to numbers of samples defined in Table 1

Fig. 2 shows the modification of the frequency dependence of the sound absorption coefficient due to the addition of 40 wt. % of hemp filler. One should notice that the effect of the filler appears above the frequency of 3000 Hz. Below this frequency the coefficient α remains nearly constant (about 7 %). Just above the frequency 3000 Hz, the value of the coefficient increases rapidly up to about 25 % and maintains at this level in the frequency range from 4000 to 6500 Hz. Probably, there is a critical frequency value from which the sound absorption in the polypropylene/hemp composite significantly grows. The manufacturing procedure does not influence the sound absorption in the case of the composite containing hemp. The quality of good sound absorption by polypropylene/hemp composite is likely due to the anatomic structure of the hemp. Both kinds of the hemp are characterized by the tubular structure.

Quite different properties in terms of sound absorption are exhibited by the composites made of polypropylene matrix and 40 wt. % of fillers obtained from long flax fibres, crumbled rapeseed straw or crumbled beech wood. For these materials the maxima of α coefficient were observed in the frequency range from 3000 to 4000 Hz (Fig. 3).

The largest value of the α coefficient (about 30 %) is shown by the composite made of polypropylene and long flax fibres. The maximum appears at the frequency of 3000 Hz. Also at the same frequency, the composite obtained from polypropylene and crumbled rapeseed straw shows the maximum (α = 25 %). The lowest value of α coefficient (α = 20 %) is exhibited by the composite made of polypropylene and crumbled beech wood but the maximum is at the frequency of 4000 Hz.

Figs. 2 and 3 show significant differences in sound absorption by composites containing hemp and the ones based on fillers: flax, rapeseed straw, beech wood. The differences can be explained taking into account the Biot theory of the sound propagation in a porous media [29—31]. The theory considers the propagation of stress waves in a porous elastic solid containing a compressible viscous fluid and 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 not more than 10 % and the criterion of the applicability of the Biot theory is fulfilled.

The theory is derived by considering the separate motion of the elastic solid and the fluid, induced by the sound wave. The sound absorption results from the energy loss due to friction existing between the solid and the fluid. The plots of frequency dependence of sound absorption were derived by Biot for different combinations of elastic and dynamic constants of the porous solid. The theoretical curves exhibit a maximum value of the absorption at a characteristic frequency which depends on the kinematic viscosity of the fluid and pore diameter [31]. The maxima are very pronounced in the case of fluid-saturated porous solids characterized by the elastic and dynamic properties far from the "compatibility condition". The measure of the enhancement of the maximum is fraction z_1

$$z_1 = \frac{V_1^2}{V_c^2} \cong 1 \qquad (6)$$

where: V_1 — *velocity of the stress wave in a real porous solid*, V_c — *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.

Referring to the measurement results presented in Figs. 2 and 3 one can state that the hemp fillers seem to be nearest the "compatibility condition" among the investigated materials. The maxima of the absorption are not noticeable in the measurement frequency range, on the contrary to the remaining fillers. In the other words, the composition of polypropylene and hemp results in such a combination of elastic and dynamic constants that the relative motion between fluid and solid is prevented.

The observed discrepancies between sound absorption characteristics can be ascribed not only to the mechanical properties of the filler but also to the filler morphology and its chemical composition. The width of hemp fibres (30 μm) is larger than of flax fibres (20 μm). Moreover, hemp plant is known to have the dimensions of the anatomic cells larger than the remaining plants under examination. From comparison of the chemical composition, it follows that hemp is distinguished for the highest contents of cellulose (75 wt. %) [32, 33]. Flax contains 71 wt. % of cellulose. Beech and rapeseed are characterized by smaller contents of cellulose: beech — 42 wt. % [32] and rapeseed — from 35 to 40 wt. % [35, 36]. The contents of lignin in hemp (4 wt. %) is twice that of flax (2 wt. %) [32, 33]. However, beech and rapeseed are known to have relatively large amount of lignin (~20 wt. %) [36]. 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 cellulose and lignin in the hemp probably enables the sound absorption in the relatively wide frequency range.

Fig. 4 shows the effect of the concentration of the lignocellulosic materials on the sound absorption of the composite. The results were obtained for the composites containing 25 wt. % of hemp harls and 40 wt. % of crumbled hemp plant. It is obvious that higher contents of the hemp filler improves the sound absorption in the frequency range above 4000 Hz. The low amount of hemp results in other frequency dependence of the sound absorption than that of the composites with the remaining hemp fillers. The almost linear increase with the frequency was observed. The results let conclude that the content of lignocellulosic filler is a constitutive factor which influences the frequency characteristic of the sound absorbing material.

CONCLUSION

The investigations of the frequency dependence of the sound absorption coefficients of the composites made of polypropylene/lignocellulosic material proved great suitability of these material to be applied as sound-absorbing substances. Addition of lignocellulosic material to the pure polypropylene results in significant increase in the sound absorption coefficient in the frequency range above 3000 Hz. The frequency dependence of the absorption coefficient can be shaped by the proper choice of the lignocellulose filler added to the polypropylene matrix. The fillers composed of tubular fibers derived from different parts of the hemp plant cause increase in the absorptive power of the composites above the frequency of 3000 Hz and the sound absorption coefficients remain at constant level (about 25 %) in the frequency range from 4000 to 6500 Hz. However, the fillers obtained from long flax fibers, crumbled rapeseed straw, crumbled beech wood were found to intensify the absorptive power of the composites in the frequency range from 3000 to 4000 Hz. The composites can be recommended for applications in the automotive industry and building as the construction materials absorbing the undesired noise: roof and wall sheathing, interior surfaces for walls and ceilings, insulation strips. Since the materials commonly used in building: concrete, wooden floor, ceramic tiles, fiberboard and plywood are known to have the sound absorption coefficient below 5 % in the frequency range from 125 to 8000 Hz they can be well replaced with the proposed new composites. Moreover, the composites proposed in this paper can be applied as the completion of the absorbers made of polyurethane foam which exhibits good properties in the low frequency range with a resonance peak in the vicinity of 1000 Hz.

ACKNOWLEDGMENTS

This research was supported by University Grant of Poznan University of Technology 32-171/08-DS.

The authors is grateful to Professor Dr. Józef Garbarczyk for inspiration and valuable discussions of the material presented in this manuscript.

REFERENCES

1. Rudawska A., Zajchowski S.: *Polimery* 2007, **521**, 453.
2. Bledzki A. K., Faruk O., Kirschling H., Kühn J., Jaszkiewicz A.: *Polimery* 2007, **52**, 3
3. Garbarczyk J., Borysiak S.: *Polimery* 2004, **49**, 541.
4. Kaczmarek K., Zielińska A., Sionkowski G., Bajer K., Stasiek A., Dzwonkowski J.: *Polimery* 2008, **53**, 60.
5. Bledzki A. K., Faruk O., Mamun A.: *Polimery* 2008, **53**, 121.

6. Kaczmarek H., Bajer K.: *Polimery* 2008, **53**, 631.
7. Abdelmouleh M., Boufi S., Belgacem M. N., Dufresne A.: *Compos. Sci. Technol.* 2007, **67**, 1627.
8. Bhattacharyya D., Bowis M., Jayaraman K.: *Compos. Sci. Technol.* 2003, **63**, 353.
9. Hattotuwa G. B., Premalal, Ismail H., Baharin A.: *Polym. Test.* 2002, **21**, 833.
10. Herrera-Franco P. J., Valadez-Gonzalez A.: *Compos. Part B.* 2005, **36**, 597.
11. Błędzki A., Jaszkiewicz A.: *Polimery* 2008, **53**, 564.
12. Peijs T.: *Materials Today* 2003, **6**, 30.
13. Kijeński J., Polaczek J.: *Polimery* 2004, **49**, 669.
14. Borysiak S., Paukszta D., Helwig M.: *Polym. Degrad. Stab.* 2006, **91**, 3339.
15. Helwig M., Paukszta D.: *Mol. Cryst. Liq. Cryst.* 2000, **354**, 373.
16. Tiwari V., Shukla A., Bose A.: *Appl. Acoust* 2004, **65**, 263.
17. Yang H.-S., Kim D.-J., Lee Y.-K., Kim H.-J., Jeon J.-Y., Kang Ch.-W.: *Bioresour. Technol.* 2004, **95**, 61.
18. Yang H.-S., Kim D.-J., Lee Y.-K., Kim H.-J.: *Bioresour. Technol.* 2003, **86**, 117.
19. Voronina N.: *Appl. Acoust.* 1996, **49**, 127.
20. Zhou H., Li B., Huang G., He J.: *J. Sound Vib.* 2007, **304**, 400.
21. Kosuge K., Takayasu A., Hori T.: *J. Mater. Sci.* 2005, **40**, 5399.
22. Augustine P., Kuruvilla J., Sabu T.: *Compos. Sci. Technol.* 1997, **57**, 67.
23. Chand N., Jain D.: *Compos. Part A.* 2005, **36**, 594.
24. Li Y., Mai Y.-W., Ye L.: *Compos. Sci. Technol.* 2000, **60**, 2037.
25. *Polish Pat.* 186577 (2004).
26. *Polish Pat.* 190405 (2005).
27. Żyszkowski Z.: "Miernictwo akustyczne", Wydawnictwa Naukowo-Techniczne, Warszawa 1987, p. 86—133.
28. Brüel&Kjaer, Standing Wave Apparatus Type 4002, Instruction Manual 1974.
29. Biot M. A.: *J. Appl. Phys.* 1955, **26**, 182.
30. Biot M. A.: *J. Acoust. Soc. Am.* 1956, **28**, 168.
31. Biot M. A.: *J. Acoust. Soc. Am.* 1956, **28**, 179.
32. Averous L., Le Digabel F.: *Carbohydr. Polym.* 2006, **66**, 480.
33. Le Digabel F.: "Incorporation de co-produits de paille de ble dans des matrices thermoplastiques. Approche de la compatibile charge-matrice et proprieties des composites", PhD Thesis URCA Reims (France) 2004, p. 198.
34. Nik-Azar M., Hajaligol M. R., Sohrabi M., Dabir B.: *Fuel Process. Technol.* 1997, **51**, 7.
35. Paukszta D.: *Fibres and Textiles in East. Eur.* 2005, **13**, 90.
36. Paukszta D.: *Oilseed Crops.* 2006, (006) XXVII, 151.

Received 16 VII 2008.