

CERCETĂRI PRIVIND PROPRIETĂȚILE FONOABSORBANTE ALE UNOR NOI MATERIALE COMPOZITE CU DEȘEURI RESEARCH ON THE SOUND-ABSORBING PROPERTIES OF NEW COMPOSITE MATERIALS WITH SOME WASTES

**MIHAI BRATU¹, OVIDIU DUMITRESCU^{2*}, OVIDIU VASILE³,
ALINA CONSTANTIN (CRISTEA)^{1,2}, MARCELA MUNTEAN²**

¹ Institutul Național de Cercetare – Dezvoltare pentru Ecologie Industrială – ECOIND, Str. Drumul Podu Dâmboviței Nr. 71-73, Sector 6, 060652, București, România

² Universitatea Politehnică București, Facultatea de Chimie Aplicată și Știința Materialelor, Departamentul Știința și Ingineria Materialelor Oxidice și Nanomateriale Str. Gheorghe Polizu, Nr. 1-7, Sector 1, 011061, București, România

³ Universitatea POLITEHNICA București, Departamentul Inginerie Mecanică, Splaiul Independenței nr. 313, București, România

Noise is a 'non-periodic sounds' complex which affects biological and psychological state of people and other natural organisms. The present paper is dedicated to some new polymeric composites with sound-absorbing properties. The originality consists in obtaining new polymeric composites from formaldehyde resin and various waste types used as reinforcing agents. The sound-absorbing capacity of the new composites varies depending on the proportion of waste used. The absorbing coefficient is presented for each sample; it depends on the porosity of the composite material obtained at the interface between the matrix and reinforcing agent.

Zgomotul este un complex de sunete fără un caracter periodic, care afectează starea psihologică și biologică a oamenilor și a altor organisme din natură. În această lucrare sunt prezentate cercetări privind proprietățile fonoabsorbante ale unor noi tipuri de materiale compozite ecologice, care au la bază utilizarea ca material de armare a unor diverse deșeuri, care pot afecta mediul înconjurător. Capacitatea de absorbție a sunetului pentru noile compozite variază în funcție de proporția deșeurilor utilizate. Este prezentat coeficientul de absorbție pentru fiecare probă, care depinde de porozitatea materialului compozit obținut, datorat interfeței dintre matrice și agentul de armare.

Keywords: sound-absorbing properties, wastes, absorbing coefficients, porosity, interface .

1. Introduction

Widespread use of mechanical and electrical equipments at home and in industry has created serious concerns related to noise reduction. It can be reduced by using various materials that decrease the sound pressure level by absorbing or mitigating the sound waves [1].

As a result, a series of measures have been taken in order to reduce the noise, both in the environment and at the workplace. Being an EU member, our country is required to align and adapt its relevant legal provisions with a series of measures intended to limit noise pollution [2].

In contrast with the traditional materials it is tried to obtain new types of composites incorporating various wastes that can affect the environment. A composite material is made when two or more materials, in combination, are leading to a product with superior properties [3,4].

New composite materials are obtained, for

which the porosity is a key characteristic in absorbing sound waves, analysing a possible correlation between the amount of resin used as matrix and the reinforcement material. This paper aims to present the obtaining process of new composite materials with sound-absorbing properties. These properties depend on the porosity of the obtained composite material, on the open spaces formed at the interface between the matrix and the reinforcing agent, which gives to the resulted composite material the property of absorption or attenuation of sound waves emitted by a source.

2. Experimental part

2.1. Materials used for samples preparation

Samples were made from various types of composite materials using as polymeric matrix a formaldehyde resin containing a small amount of free formaldehyde and good storage stability.

* Autor corespondent/Corresponding author,
E-mail: ovidiu_d_dumitrescu@yahoo.fr

The used resin is obtained by batch process, having low formaldehyde emission limits, in compliance with the law. It is known from the literature that formaldehyde resins are obtained via continuous or discontinuous processes in two or more steps, by condensation of urea with formaldehyde [5]. It was also identified that during the technological process of obtaining the composite based on the formaldehyde resin, during pressing and after that, there is an emission of formaldehyde. The emission of formaldehyde is influenced by the content of the free formaldehyde adhesive manufactured: the emission is lower when the content of free formaldehyde is small [6]. Various waste types were used as reinforcing agents; wastes resulted from various industrial processes that can affect the environment when discarded. The wastes we used are: crushed glass, wood and polypropylene [7, 8].

2.2. Preparation procedure

Glass shards are reused both in glass and (in some cases) ceramic factories, as an added material. In the present situation, when the glass factories have limited activity, there are very large reserves of glass shards, with various provenance and quality. Commercial glass, particularly glass bottles and construction glass (glass, glass tiles) can be used to obtain polymeric composites. It is important that these wastes present a well-controlled granulometric distribution, which insures the reproducibility of the obtained data. Glass shards were washed, dried and then grounded to appropriate size with the help of a mill using grinding balls [9], and then grain size distribution analysis was performed using a laser particle size analyzer, shown in Figure 1.

The wood waste is the result of obtaining wood boards with increased resistance at the wood processing plants, waste resulted during the log cutting process flow to obtain the sheets. For the sample preparation of composite material it was used wood waste with particles dimension between 0.1 ÷ 15 mm, particle size distribution achieved by means of a set of vibrating table screeners shown in Figure 2.

Polypropylene, polyethylene and polystyrene wastes are the most widespread plastic wastes. All these wastes are the result of the technological flow process in the packaging industry. In this paper in order to reinforce the polymeric matrix we used granules of polypropylene with 5 mm grain size. Wastes used as reinforcement material were subjected to a drying operation, thus determining the total humidity.

The total humidity W_t represents the sum between the soaked humidity W_s and the hygroscopic humidity W_h .

The total humidity values determined for the wastes used as reinforcement material in each sample are:

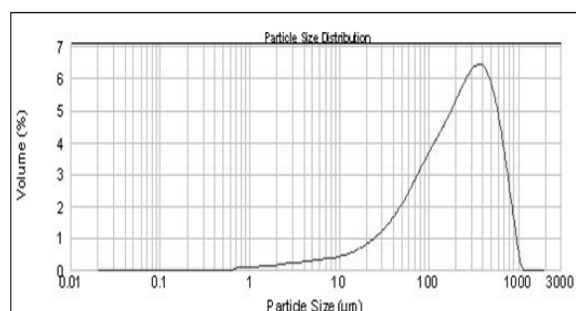


Fig.1 - Laser particle size analysis – Ground glass waste / Granulometrie cu laser – deșeu de sticlă măcinată.

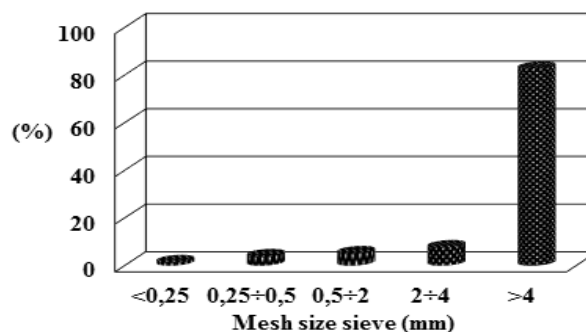


Fig.2 - Grain size distribution – Wood waste / Distribuția granulometrică – deșeu de lemn.

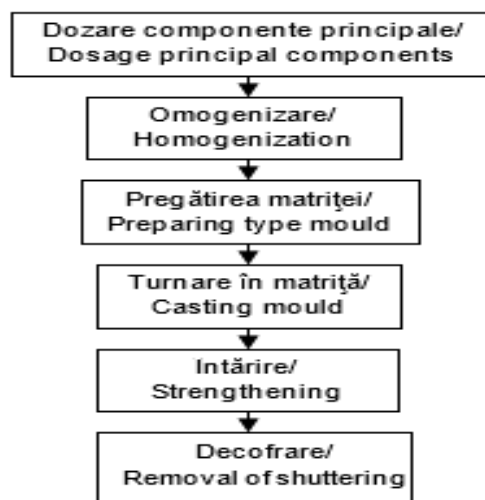


Fig.3 - Technological process scheme for obtaining polymer composites / Schema procesului tehnologic de obținere a compozitelor polimerice.

- For sample P1 – ground waste glass $W_t^i = 0.72\%$.
- For sample P2 – wood waste $W_t^i = 10.74\%$.
- For sample P3 – polypropylene waste $W_t^i = 0.55\%$.

Figure 3 shows the technological process scheme for obtaining new polymeric composites:

- Waste and resin dosage were made gravimetrically with an accuracy of $\pm 0.01g$;
- Homogenization was performed in a shock resistant container; Heidolph RZR 2021 shaker type with cross blades.

Table 1

Composite samples obtained / Probele de materiale compozite obținute			
No. sample Nr. probă	Formaldehyde resin content Conținut rășină formaldehidică [%]	Type waste Tip deșeu	Waste content Conținut deșeu [%]
P1	80	Ground glass waste Deșeu sticlă măcinată	20
P2	20	Wood waste Deșeu lemn	80
P3	50	Polypropylene waste Deșeu polipropilenă	50

- The casting mould was subject to a preliminary preparation (cleaning, followed by lubricating with a temperature resistant silicone grease); the mixture obtained was put into a circular shape stencil, without any pressing operation.
- Strengthening – the heating took place at a temperature of 120°C in oven, for 4 hours, followed by cooling/ strengthening for another 4 hours at room temperature.
- The shuttering removal is the final operation: the sample of obtained composite material is removed from the mould.

In Table 1 are presented samples of the obtained composite materials.

Tests were performed on samples of circular form with a diameter of 63.5 mm and a height of about 20 mm, which were made in cylindrical molds. [10]

The composite materials samples are presented : in Figure 4.a sample P1 formaldehyde resin reinforced with ground glass; in Figure 4.b sample P2 formaldehyde resin reinforced with wood waste; Figure 4.c sample P3 formaldehyde resin reinforced with propylene waste;



Fig.4.a - P1 – Formaldehyde resin – ground glass waste / P1 – Rășină formaldehidică – deșeu sticlă măcinată.



Fig.4.b - P2 – Formaldehyde resin – wood waste / P2 – Rășină formaldehidică – deșeu lemn.



Fig.4.c - P3 – Formaldehyde resin – polypropylene waste / P3 – Rășină formaldehidică – deșeu polipropilenă.

Acoustic interferometer method (Kundt tube) was used to determine the absorbing coefficient. The test equipment consists of a type 4206-A tube acoustic interferometer, shown in Figure 5 and a system acquiring signal simultaneously on five channels with signal generator – multianalyzer PULSE type 3560-B-030; the determination method is in accordance with the current applicable standards [11,12].

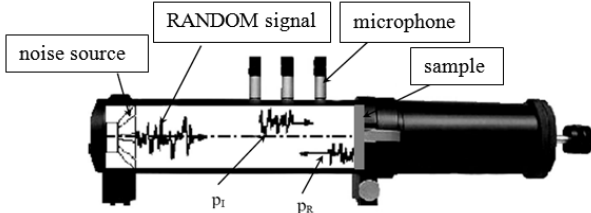


Fig.5 - Equipment for determining the absorbing coefficient
Echipament pentru determinarea coeficientului de absorbție.

The measurement method for the acoustic absorbing coefficient (using an impedance tube) is based on the fact that the acoustic reflection coefficient at normal incidence r can be determined from the transfer function H_{12} measured between two positions, with a microphone placed in front of the material under test [13].

The acoustic pressures for the incidence wave p_I and for the reflected wave p_R are:

$$p_I = \hat{p}_I \cdot e^{jk \cdot x} \quad (1)$$

$$p_R = \hat{p}_R \cdot e^{-jk \cdot x} \quad (2)$$

Where: \hat{p}_I and \hat{p}_R - are the values for p_I and p_R in the reference plan ($x=0$);

k – is a complex wave number.

The acoustic pressures p_1 and p_2 between the two microphone positions are:

$$p_1 = \hat{p}_I \cdot e^{jk \cdot x_1} + \hat{p}_R \cdot e^{-jk \cdot x_1} \quad (3)$$

$$p_2 = \hat{p}_I \cdot e^{jk \cdot x_2} + \hat{p}_R \cdot e^{-jk \cdot x_2} \quad (4)$$

The transfer equation for the incidence wave H_I is:

$$H_I = \frac{P_{2I}}{P_{1I}} = e^{-jk \cdot (x_1 - x_2)} = e^{-jk \cdot s} \quad (5)$$

Where: $s = x_1 - x_2$ represents the distances between the two microphones.

The transfer equation for the reflected wave H_R is:

$$H_R = \frac{P_{2R}}{P_{1R}} = e^{-jk \cdot (x_1 - x_2)} = e^{-jk \cdot s} \quad (6)$$

The transfer equation H_{12} for the total

acoustic field is achieved using the relation (4) and

(5) and noting that $\hat{p}_R = r \cdot \hat{p}_I$, with the relation:

$$H_{12} = \frac{p_2}{p_1} = \frac{e^{jk \cdot x_2} + r e^{-jk \cdot x_2}}{e^{jk \cdot x_1} + r e^{-jk \cdot x_1}} \quad (7)$$

The reflexion coefficient r is obtained from the relations (6) and (7):

$$r = \frac{H_{12} - H_I}{H_R - H_{12}} e^{2jk \cdot x_1} \quad (8)$$

The acoustic absorbing coefficient, α at normal incidence is calculated using the relation:

$$\alpha = 1 - |r|^2 \quad (9)$$

The composites structure and especially the matrix-agent interface have an essential role for some properties of the composite materials. The quality of the interface determines the structural integrity, the answer to environmental action and the physical-mechanical properties of the obtained composite material. The interface refers to the contact surface between the polymer matrix and the reinforcement materials. Their connection is determined by the structural characteristics of the surface of the reinforcement agent mainly by the roughness, porosity and specific surface area of the particle size distribution. The adhesion between the polymer and the reinforcement material is based on two surface phenomena: absorption and watering [14]. The absorbing capacity and the mechanical properties for the new composite materials are depending on the porosity and open spaces formed at the interface between the matrix and the reinforcement material.

To highlight the correlations between the interface matrix–reinforcement agent and physical and mechanical properties of newly developed composite materials, SEM electron microscopy was performed. The apparent porosity was also determined, as were the compressive strength and the water absorbing (stability towards water) for the newly obtained composite materials. Physical, mechanical and acoustic measurements on a number of 4 samples of each sample were performed and so the final result is an average of four determinations.

Humidity, size distribution, porosity and determination of water absorbing of the reinforcing material were performed according to standards [15 – 17].

3. Results and interpretations

As was discussed within the above paragraphs, new types of composite materials made from formaldehyde resin and reinforcement material consisting of waste with sound-absorbing properties were obtained. The apparent porosity was determined and the results are shown in Figure 6.

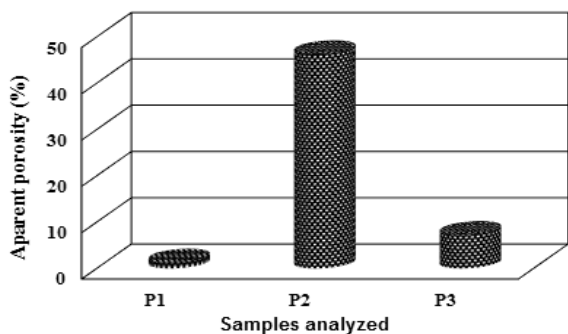


Fig.6 - Apparent porosity for composite samples obtained
Porozitatea aparentă pentru probele de material compozit obținute.

A higher porosity value can be observed for sample P2 (46.28%) which depends on the reinforcement material used (wood waste), thus the structure of the composite material obtained gives it the highest porosity. Next sample is P3, with a porosity of 7.24% and then there is the lowest porosity sample P1, with 1.28%.

A good absorbance for a material is given by the value of $\alpha = 1$ or close to 1 and an absorbing plateau at this α value with a wider frequency range.

Such materials are classified by acoustic absorbing classes [18], function of α , and are presented in Table 2.

Table 2

Absorbing class according to the absorbing coefficient
Clase de absorbție acustică funcție de coeficientul de absorbție

Acoustic absorbing class <i>Clasă de absorbție acustică</i>	α
A	0.90; 0.95; 1.00
B	0.80; 0.85
C	0.60; 0.65; 0.70; 0.75
D	0.30; 0.35; 0.40; 0.45; 0.50; 0.55
E	0.15; 0.20; 0.25
Without class <i>Fără clasă</i>	0.00; 0.05; 0.10

In the figures 7 ÷ 9 are presented the results of the analysis of the absorbing coefficient for the prepared composite samples (the frequency at which measurements were made is 16 ÷ 3150 Hz) and also presents the electron microscopy SEM (for segments 1 mm and 500 μ m) for the prepared samples in which are presented the interface resin-reinforcement agent and the presence of pores.

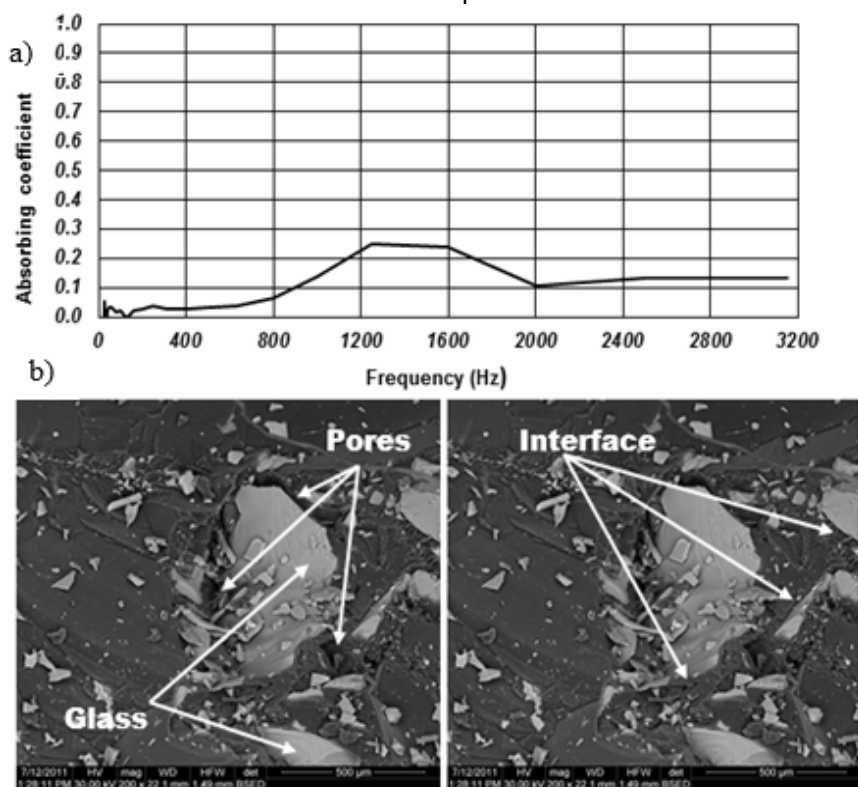


Fig.7 - Sample1 (formaldehyde resin 80% + glass waste 20%)
Proba 1 (rășină formaldehidică 80% + deșeu de sticlă 20%)
a) Absorbing coefficient / *Coeficientul de absorbție.*
b) Electron microscopy SEM / *Microscopia electronică SEM.*

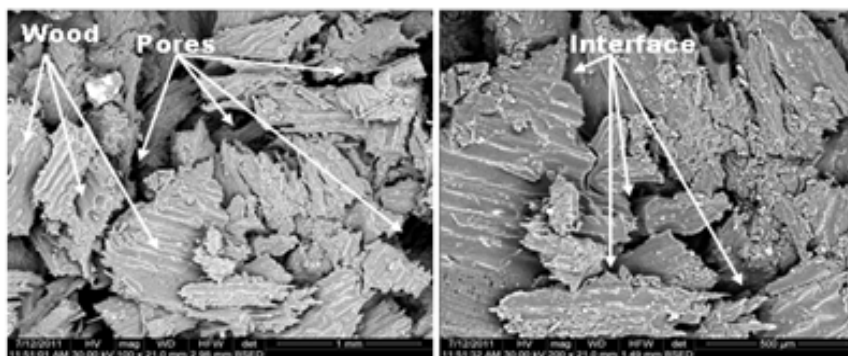
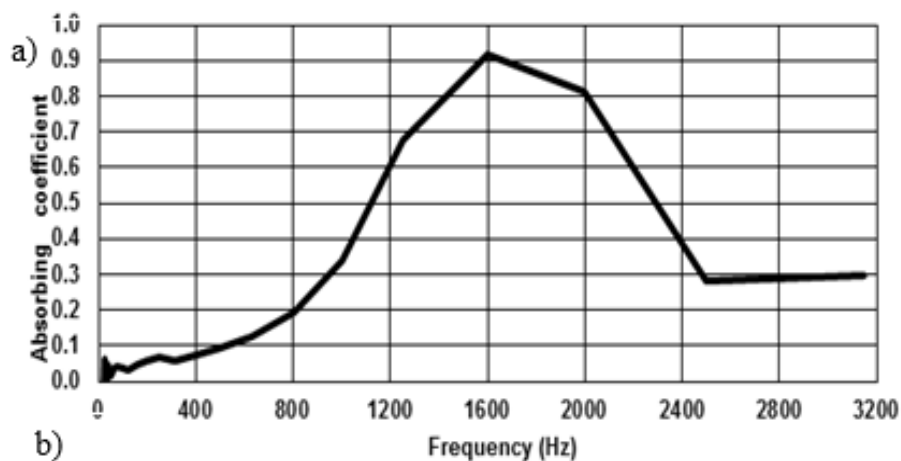


Fig.8 - Sample2 (formaldehyde resin 20% + wood waste 80%)
 Proba 2 (rășină formaldehidică 20% + deșeu de lemn 80%)

- c) Absorbing coefficient / Coeficientul de absorbție.
- d) Electron microscopy SEM / Microscopia electronică SEM.

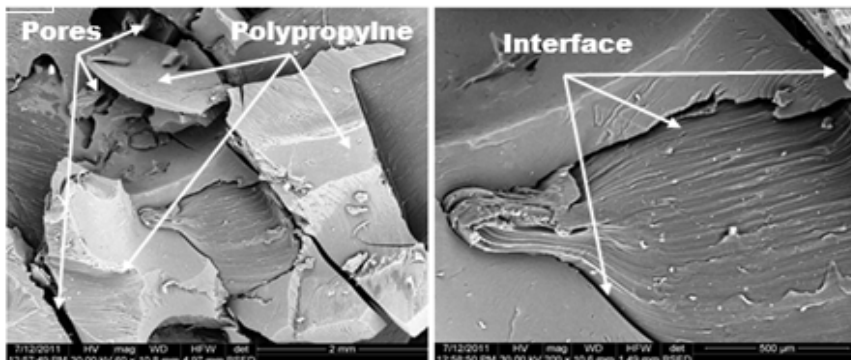
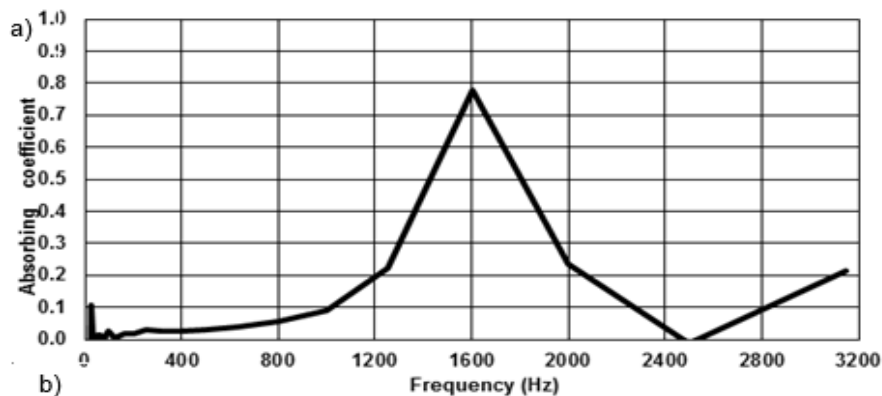


Fig.9 - Sample 3 (formaldehyde resin 50% + polypropylene waste 50%)
 Proba 3 (rășină formaldehidică 50% + deșeu polipropilenă 50%)

- a) Absorbing coefficient / Coeficientul de absorbție.
- b) Electron microscopy SEM / Microscopia electronică SEM .

The sample P1 shows an absorbing coefficient which reaches an $\alpha = 0.25$ value. An increase of the absorbing coefficient from 800 Hz to 1000 Hz was observed, and a plateau of absorption for sound waves but with a low absorbing coefficient between 1000 Hz and 1600 Hz. It is a very small value in terms of sound-absorbing, because this sample is a compact one. This composite material was made of small glass shards that have been completely or partially wet by the polymeric matrix. The proportion of resin and reinforcing agent used in preparing the samples is influencing the degree of wetting of the reinforcement agent at the interface between these two components. In sample 1 were used a ratio of 80% by weight of formaldehyde resin and 20% ground waste glass as a reinforcing agent resulting in a compact sample with a very small porosity, as shown in the graph in Figure 6. The low absorption of sound waves is due to the low porosity that is observed in SEM images.

The interface resin-reinforcement agent influences the compactness of the sample and also the absence of the pores and low values of the absorbing coefficient.

It follows that this type of composite material falls within the class E of acoustic absorption according to Table 2.

The sample P2 shows an absorption coefficient which exceeds the value $\alpha = 0.9$. There is a progressive increase of the graph in the frequency range between 400 ÷ 2400 Hz and then linearity in the frequency range between 2400 ÷ 3150 Hz.

It is a good absorption of sound waves for this type of material, absorption on a wide range of frequencies, and this absorption of sound waves is influenced by the wetting degree of the agent reinforcement's particles. In this case in the sample preparation it was used a lower ratio by weight of formaldehyde resin 20% in comparison with the wood waste which was 80%. Thus, at the interface between resin and reinforcement agent voids are formed as shown in SEM images resulting in a material with high porosity according to the graph shown in Figure 6. A higher percentage of porosity due to the interface between the formaldehyde resin, which is the matrix, and the wood waste as reinforcing agent, causes a good absorption of the sound waves.

Thus this type of composite material falls within the class A acoustic absorption.

For sample P3, the graph obtained with the equipment for determining the absorbance coefficient is in form of a peak with a maximum $\alpha = 0.78$ and results that this type of composite, according to Table 2, falls in Class B sound-absorbing.

There is a progressive increase in absorbance from 800 Hz with a maximum at 1600

Hz, but increasing the frequency, the absorbance decreases sharply up to 2500 Hz at higher frequencies, where there is a slow growth up to 3200 Hz. This composite material was made of equal proportions of formaldehyde resin and also polypropylene waste, so the sample obtained had a lower porosity than sample P2, according to the graph in Figure 6. The presence of voids at the interface between the resin and the reinforcing agent is less, as shown in SEM images, also the degree of wetting of the reinforcing agent is higher, because a higher percentage was used by weight of formaldehyde resin.

Because this sample has a lower porosity compared with samples P1 and P2 being a more compact material, because of the reinforcing agent and to its interface with the formaldehyde resin, resulting in lower absorbance of sound waves and thus framing this composite material in a lower class in terms of acoustic absorbance.

Due to the fact that the sound absorption capacity is depending on the composite material porosity, the pores dimension is influenced by the dimensions of particles used as filling material. The pores content (the total pores volume) is determined by the filling volume ratio within the composite material. The interface between the resin and reinforcement agent, the wetting degree of the reinforcement agent particle depends on the particle size and is influencing the sound absorbance. Moreover, the proportion between the resin and the reinforcement agent and also the reinforcement of the agent's particle's shape are influencing the formation of holes in the homogenization and molding technological phases [14].

An analyzing of the previous cases regarding the interface between the polymeric matrix – the reinforcing agent and the apparent porosity of the obtained composite material, influencing the sound waves absorbing coefficient was presented in the chart shown in Figure 10.

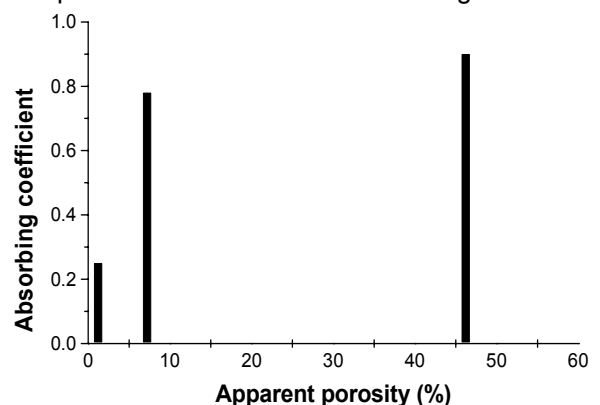


Fig.10 - Sound waves absorbing coefficient depending on the apparent porosity of the obtained composite material / Coeficientul de absorbție a undelor sonore funcție de porozitatea aparentă a materialelor compozite obținute.

This graph shows that the presence of pores at the interface between the resin and the reinforcement agent affects the ability to absorb the sound waves for each composite sample obtained. The proportion between the resin and the waste and the reinforcement agent's granulometric distribution influence the degree of wetting and, respectively, coating of the particles which form the reinforcement agent in the polymeric matrix.

Correlating the apparent porosity and the absorbance coefficient, we can say that sample 1 presents the lowest value of apparent porosity and a low absorption coefficient. The porosity increase is determining a better sound absorption, consequently the sample 2 presents the highest value of apparent porosity and the best absorption coefficient.

Analyzing the graphs presented in Figures 8 and 9 it can be observed for sample 2 (Figure 8) a sound waves absorption on a wider frequency domain, from 400 Hz to 3200 Hz, with a maximum absorbance $\alpha = 0.9$ at 1600 Hz. For sample 3 (figure 9) the sound absorption is lower, a significant value being in the domain 1200 Hz to 2000 Hz, with a maximum at 1600 Hz with $\alpha = 0.78$. A higher porosity, as presented by sample 2, determines a better sound absorption, as shown within figure 10.

The water absorbance (stability against water) shown in Figure 11, and the compressive strength (Figure 12) for samples obtained were also determined.

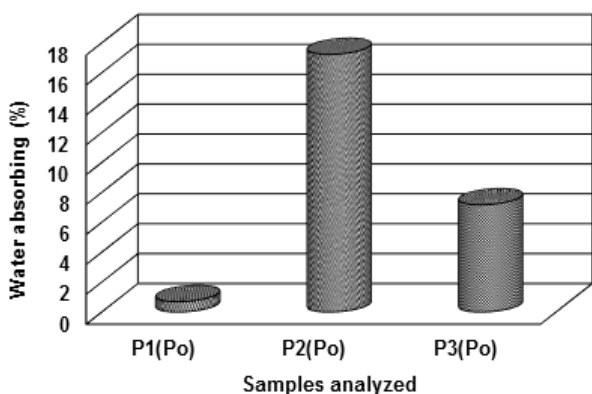


Fig. 11 - Water absorbing for samples analyzed / Absorbția apei pentru probele analizate.

We can observe that the sample with the highest water absorbance coefficient is the sample P2 (17.23%) and that this value depends on the reinforcing material (wood waste) used. The structure of this type of waste determines the value of water absorbance. The next sample is P3 with an absorbing coefficient of 7.17%, and the most stable sample is P1 with 0.69%. P1 is a compact sample and has a low porosity.

The highest value for the compression strength corresponding to the new composite materials is obtained with sample P1 (45.8 N/mm²)

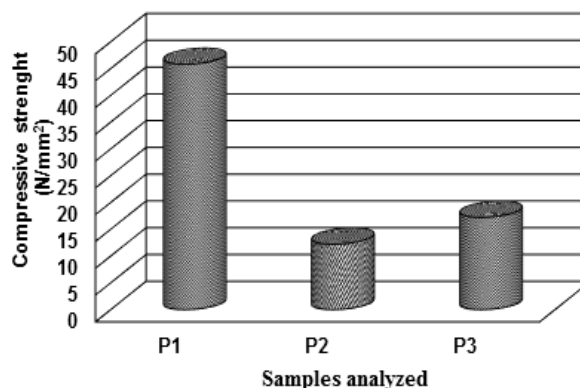


Fig. 12 - Compressive strength for samples analyzed / Rezistența la compresiune pentru probele analizate

because this is a compact sample. The other samples have smaller values: P3 – 17.21 N/mm² and then P2 – 12.21 N/mm². The compressive strength reduction depends on the agent used for reinforcing, on the proportion of the reinforcement agent and the resin used, and also on the granulometric distribution of the agent used for reinforcement.

4. Conclusions

In this paper we presented the results obtained when producing sound-absorbing composite materials, with the polymeric matrix of formaldehyde resin and as reinforcing agent materials from different wastes such as: ground glass waste, wood waste and polypropylene waste.

Correlations between the sound-absorbing properties of the developed composites and the interface between the resin and the reinforcing agent are presented

The best results in terms of sound-absorbing properties were obtained on samples P2 and P3, samples from composite material made from formaldehyde resin 80% and reinforcing agent 20% consisting of waste type: wood waste and polypropylene waste.

These materials were characterized by a sound-absorbing coefficient of $\alpha = 0.8 \div 0.9$ and can be used to manufacture soundproofing panels with applications in industry, road, rail or air transport. A good absorbance coefficient is given by a high porosity of the material, which depends on the interface between the reinforcing agent and the resin used.

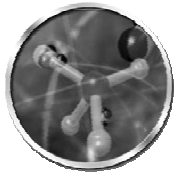
The sample P2 presents the highest water absorbance.

The sample P3 presents the highest resistance to compression.

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