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MATERIALE COMPOZITE PE BAZĂ DE IPSOS CU CONȚINUT DE DEȘEURI INDUSTRIALE

COMPOSITE MATERIALS BASED ON GYPSUM PLASTER AND INDUSTRIAL WASTES

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The valorization of industrial wastes in the manufacture of composite materials is topical in construction industry.

The main objective of this study is to improve the thermal insulation properties of some gypsum plaster composites by it's partial substitution (5 and 30%wt.) with three types of industrial wastes i.e. polyurethane, rubber and chopped electric cables; the paper present also the influence of wastes on the main properties specific for thermal insulation materials (thermal conductivity, compressive strength, short-term water absorption by partial immersion).

The use of the above mentioned industrial wastes in the composition of gypsum plaster determines an improvement of thermal insulation properties (thermal conductivity decreases with 21-55%) correlated with a decrease of compressive strength comprised between 68-87%.

The manufacture of this type of insulation materials can contribute to solving environment issues, decreasing the amount of natural raw material used and valorization of industrial wastes as sustainable sources of alternative raw materials.

Valorificarea deșeurilor industriale în producerea materialelor de construcție reprezintă o temă de studiu de mare actualitate.

Prezentul studiu își propune îmbunătățirea proprietăților de izolare termică ale unor compozite pe bază de ipsos, prin substituirea parțială (5% și 30% procente gravimetrice) a acestuia cu deșeuri industriale (poliuretan, cauciuc și cabluri electrice tocate). De asemenea, s-a urmărit și influența pe care aceste deșeuri o exercită asupra unor proprietăți specifice pentru materialele de izolație termică (conductivitate termică, rezistența la compresiune și absorbția apei prin imersie parțială de scurtă durată).

S-a constatat că utilizarea acestor deșeuri industriale în compoziția plăcilor de ipsos determină o îmbunătățire a proprietăților de izolare termică (conductivitatea termică scade cu 21-55%) corelată însă cu o scădere a rezistenței mecanice cuprinsă între 68-87%.

Producerea acestor materiale de izolație poate contribui la rezolvarea unor probleme de mediu, prin reducerea cantității de materii prime naturale folosite și utilizarea unor deșeuri ca materii prime alternative.

Keywords: *thermal insulation materials, gypsum plaster, industrial waste, thermal conductivity, properties*

1. Introduction

For a sustainable future, the environmental requirements of the European and Romanian legislation impose the valorization of industrial and municipal wastes. To reduce the negative impact of various types of industrial wastes on the environment, many studies were performed in order to find new ways to capitalize them in the manufacture of construction materials.

Numerous types of inorganic waste were incorporated in gypsum plaster. Magallanes-Rivera *et al.* [1] showed that low amounts (10-25%wt.) of granulated blast furnace slag (GBFS) and pozzolanas (silica fume or fly ash) could increase the compressive strength of plain plaster cured under water. Badanoiu *et al.* [2] studied the substitution of plaster with small amounts (10-20%wt.) of thermally treated fine fraction (TTFF) of concrete rubble. TTFF, is the fine fraction (below 0.2 mm) which results in the thermal and mechanical treatment of concrete rubble [3]; this treatment is applied to improve concrete rubble properties in order to use it as recycled aggregate in concrete manufacture. TTFF is not used in new concrete manufacture and is generally regarded as a waste. The obtained results show an improvement of the compressive strength of plasters with TTFF cured in water up to 90 days, as compared with plain plaster.

Rio Merino *et. al.* [4] used bricks and extruded polystyrene waste in order to reduce the amount of raw materials used in the production of

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gypsum plaster and to improve its characteristics (superficial hardness values and water capillarity absorption).

Organic waste such as rubber (resulted from mechanical grinding of end-of-life tires) was also used in gypsum plaster manufacture in the attempt to improve thermal and acoustical performances [5]. The addition of recycled rubber improves the thermal insulation properties but decreases the bending and compressive strengths of composites.

Scrap tires and crumb rubber can be also valorized in the production of bituminous asphalt for pavement [6] as well as portland cement concrete and mortars with good thermal insulation properties [7-9].

Another organic waste used in the production of lightweight aggregate concrete and mortar based on portland cement is rigid polyurethane (PUR); the incorporation of PUR waste decreased the thermal conductivity as well as compressive strength compared with reference [10,11].The reduction of dry density (up to 36%) i.e. increase of porosity has, as expected, a negative effect on the durability of this type of materials [12].

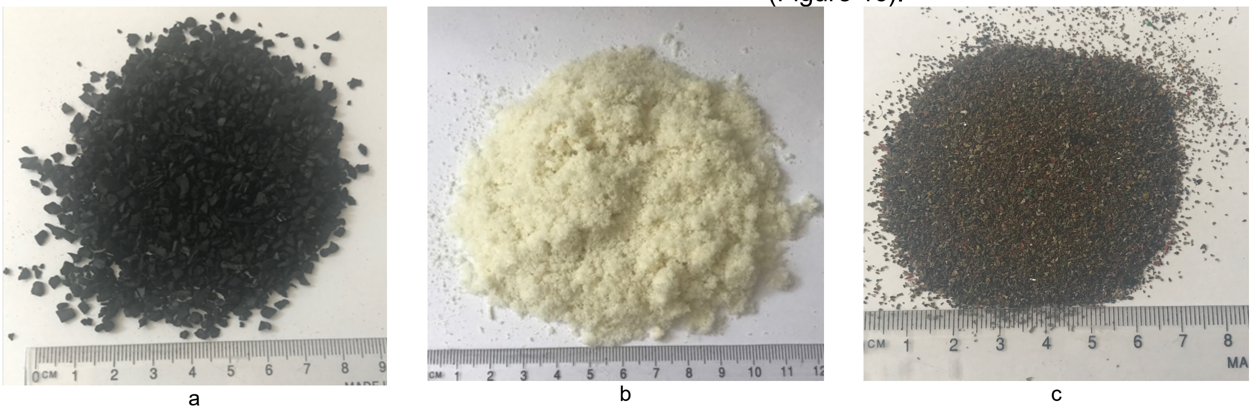
Based on these information and considering the importance of using insulation materials in buildings (buildings and construction sector consumes about 40% of world's energy [13]), the aim of the research presented in this paper is to obtain cost-effective and environmentally friendly gypsum composites; the paper presents the influence of the substitution of gypsum plaster with various types of waste (rubber, polyurethane and chopped electric cables) on mechanical and thermal insulation properties of resulted composites.

2. Materials and methods

2.1. Materials

The materials used in this study were:

- ***gypsum binder (I)*** for building purposes, in conformity with EN 13279-1 [14], with the following properties [15]: i) reaction to fire: Class A1; ii) fineness: 99% below 315 μm; iii) content of calcium sulphate: >50%; iv) initial setting time: 10-15 min.
- ***rubber particles waste (C)*** from recycling of scrap tires, with grain sizes comprised between 2-6 mm (Figure 1a). In order to improve the adhesion of rubber particles to the binding matrix [16], C was immersed in 5M NaOH solution for various periods of time (from 15 up to 45 minutes); after this treatment the waste was washed with water and dried in air; this material was noted with C_t.
- ***polyurethane foam waste (P)*** obtained from the shredding of insulation panels, with grain sizes comprised between 0.1- 2 mm (Figure 1b);
- ***chopped electric cables waste (E)*** obtained from the chopping of electrical cables with jacket insulation, with grain sizes comprised between 0.1-1.5 mm (Figure 1c).



a b c

Fig.1 – Waste rubber (a), polyurethane foam waste (b) and chopped electrical cable (c) / *Deșeu de cauciuc (a), deșeu de spumă poliuretanică (b) si deșeu de cabluri electrice tocate (c).*

Table 1

Compositions of gypsum composites with waste content/*Compoziții ale materialelor compozite pe bază de ghips cu diferite deșeuri*

Code	Gypsum plaster / *Ipsos* [%]	Waste/*Deșeu*				Water/plaster *apă/ liant*
		Rubber/ *Cauciuc* [%]	Chemically treated rubber/ *Cauciuc tratat* [%]	Polyurethane foam / *Spumă poliuretanică* [%]	Electric cable *Cabluri electrice* [%]	
I	100	-	-	-	-	0.6
IC	70	30	-	-	-	0.6
IC_t	70	-	30	-	-	0.6
IE	70	-	-	-	30	0.6
IP	95	-	-	5	-	0.8

2.2. Specimens preparation

In order to assess the influence of wastes on the main properties of gypsum composites, the mixtures presented in Table 1, were prepared and tested.

The water to gypsum ratio was 0.6 for all compositions except for the one with 5% polyurethane foam waste – in this case it was necessary to increase the ratio to 0.8.

2.3. Methods

The *grain size distribution* of waste rubber (C) was determined according to the method presented in EN 933-1 standard [17] and for chopped electric cables (E) was determined according to the method presented in EN 1015-1:2001/A1 standard [18].

The density of wastes was determined with a helium pycnometer (Pycnomatic model).

The following properties were assessed on gypsum composites, hardened for 7 days at 23 ± 2°C and 50 ± 5% R.H.:

- *apparent (geometrical) density*, assessed, on cubs (20x20x20 mm).
- *compressive strength* was determined according to the method presented in European Standard EN 13279-2 for gypsum binders and gypsum plasters [19].
- *water absorption coefficient* due to capillary action was determined with the method presented in European Standard EN 1015-18 [20].
- *thermal conductivity* was determined with an HESTO-Lambda-CONTROL A90 – equipment; this equipment measures the heat flow through a specimen placed between two plates with different temperatures; the measurement accuracy is ± 3% in accordance with EN 12667 [21].

3. Results and discussion

3.1. Materials characterization

The grain size distribution of waste rubber particles (C) and chopped electric cables (E) is presented in Figure 2.

It can be noticed the presence of a high amount of rubber particles with sizes comprised between 3 mm and 6 mm and for the chopped electrical cables between 0.2- 1 mm.

Due to the low density of polyurethane waste (less than 1g/cm^3) and specific shape (flake-like), it was not possible to assess its grain size distribution by this method.

As previously mentioned, the presence of rubber waste in cement mortar or concrete, determines an important decrease of the mechanical strengths [9,16]; one possible method to reduce this negative effect is to treat the rubber waste, prior to its incorporation in cement composite, with NaOH solution in order to improve the hydrophilicity of rubber particles thus improving the adhesion to cement matrix [16].

In this study we assessed also the influence of NaOH treatment of rubber particles on the mechanical strengths of plaster composites. The SEM analyses of the surface of rubber particles, treated with 5M NaOH solution for 15, 30 and 45 minutes is presented in Figure 3. It can be noticed the increase of the surface roughness after the NaOH treatment up to 30 minutes; the further increase of treatment time (up to 45 minutes) does not lead to an important modification of surface texture, therefore in future experiments we've used rubber particles treated with NaOH solution for 30 minutes.

3.2 Influence of waste addition on the main properties of gypsum composites

The visual aspect of gypsum composites

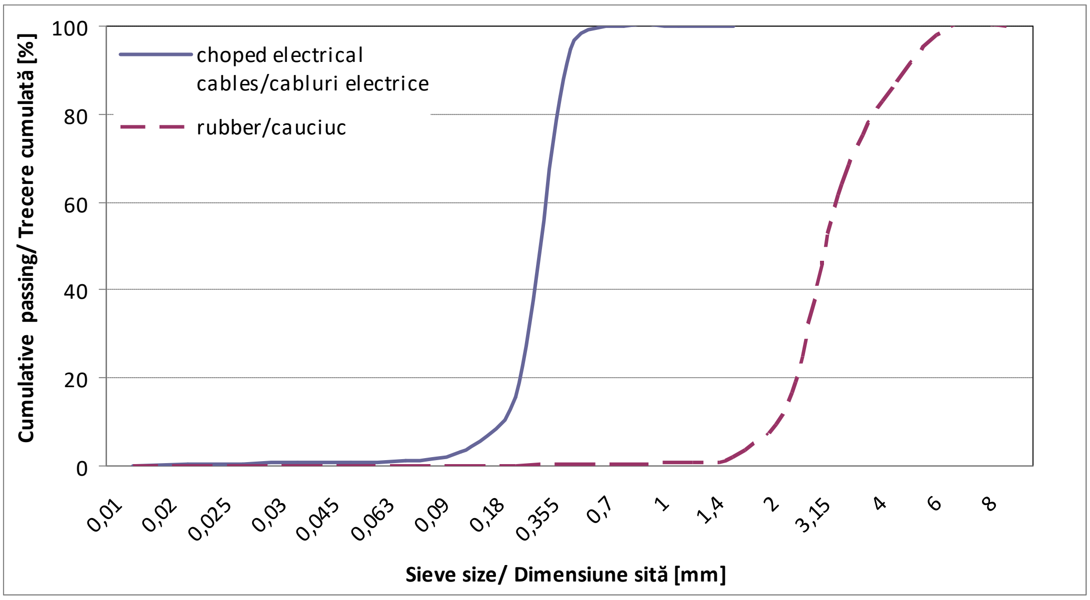
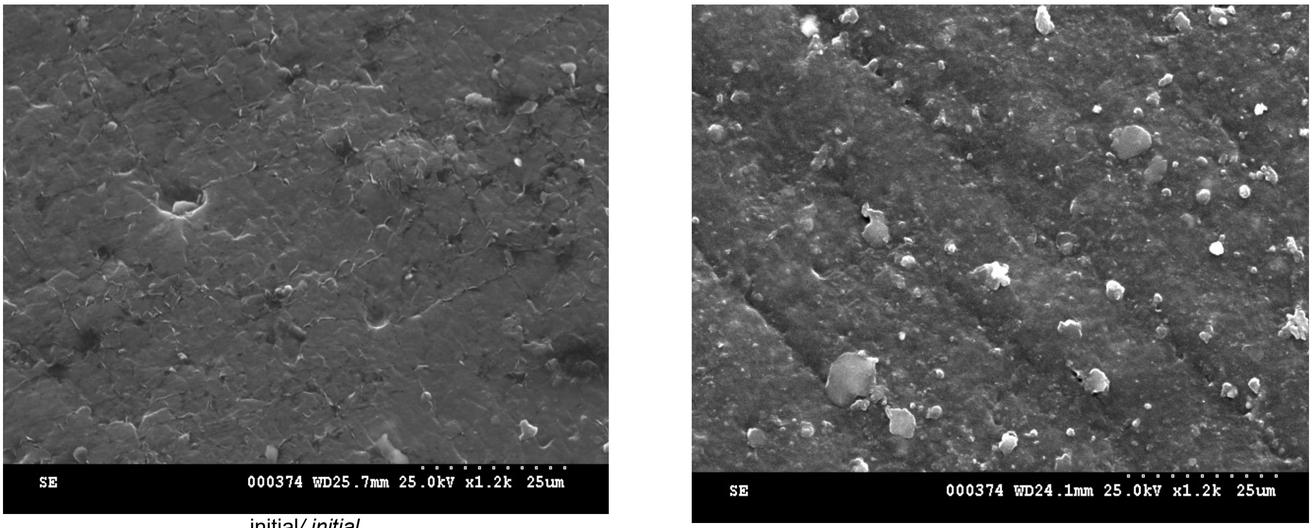
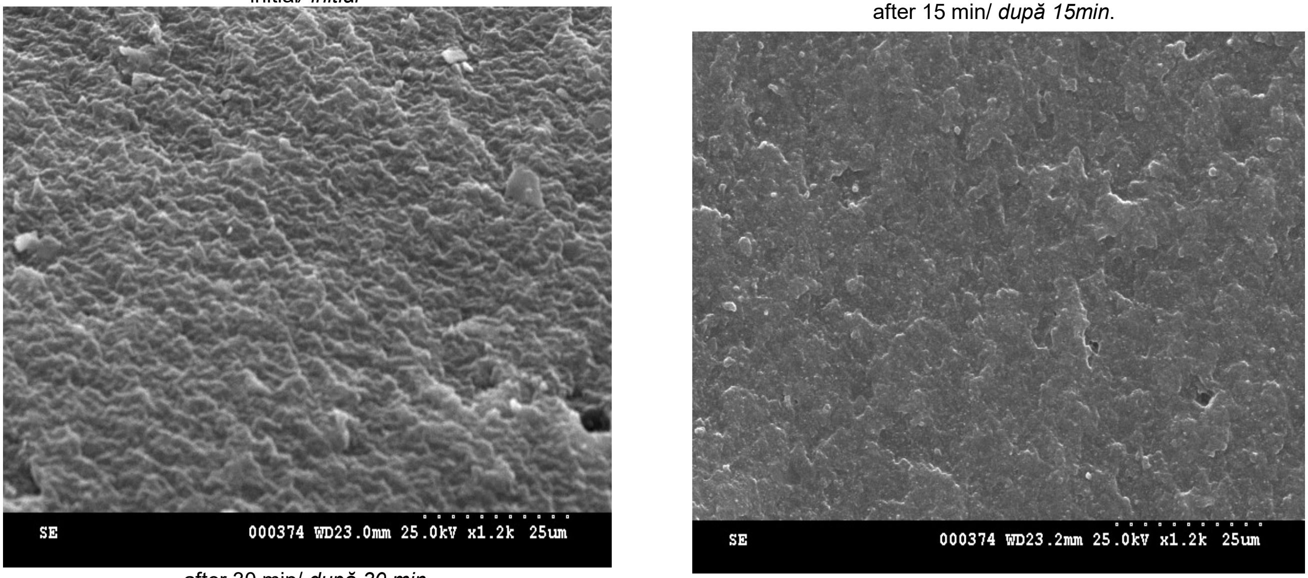


Fig.2 – The grain size distribution of rubber and electrical cables wastes / *Distributia granulometrică a deşeurilor de cauciuc şi cabluri electrice tocate.*



initial/ *initial*

after 15 min/ *după 15min*.



after 30 min/ *după 30 min*

after 45 min/ *după 45 min.*

Fig.3 – SEM images of the surface of waste rubber particle after NaOH treatment / *Micrografii SEM ale suprafeței granulei de cauciuc după tratamentul cu NaOH.*

with waste content (fracture surface), hardened for 7 days, is presented in Figure 4.

For the composites with rubber waste can be noticed the poor adhesion of the gypsum matrix to the rubber grains (see arrow showing the numerous gaps from which the rubber grains were pulled out). The NaOH treatment of rubber grains does not seem to improve the matrix adhesion to the surface of the rubber grains, contrary to the effect reported for cement matrices. This can be explained by the important differences of the microstructure and porosity of interfacial transition zone (ITZ) between rubber particles and binding matrix. Si *et al.* [16] reported the decrease of the porosity of portland cement composites containing NaOH treated rubber particles compared with the as-received rubber particles. This suggests a better adhesion (adherence) of the hydrates formed by cement hydration (mainly calcium silicates hydrates with low crystallinity at early ages) on the rough (coarse) surface of treated rubber particles.

On the opposite side, the gypsum crystals, formed by the plaster hydration, have a specific needle-like shape (see Figure 5). These crystals do not adhere at the surface of the rubber particle (C) but surround and include them in a “felt” like matrix (Figure 5b); therefore, the increase of rubber particle roughness does not lead to the reduction of ITZ porosity (like in cement case).

The visual aspects of the gypsum composite with chopped electric cables and polyurethane wastes (Figure 4) suggest a higher homogeneity of these composites.

The values of compressive strength of gypsum composites (Figure 6) confirm the above mentioned results. The lower values of compressive strengths are recorder for the composites with rubber waste (IC and IC_t); gypsum composites with chopped electric cables (IE) and polyurethane (IP) have higher compressive strengths as compared with C and C_t; the higher compressive strength of IP as compared with IC

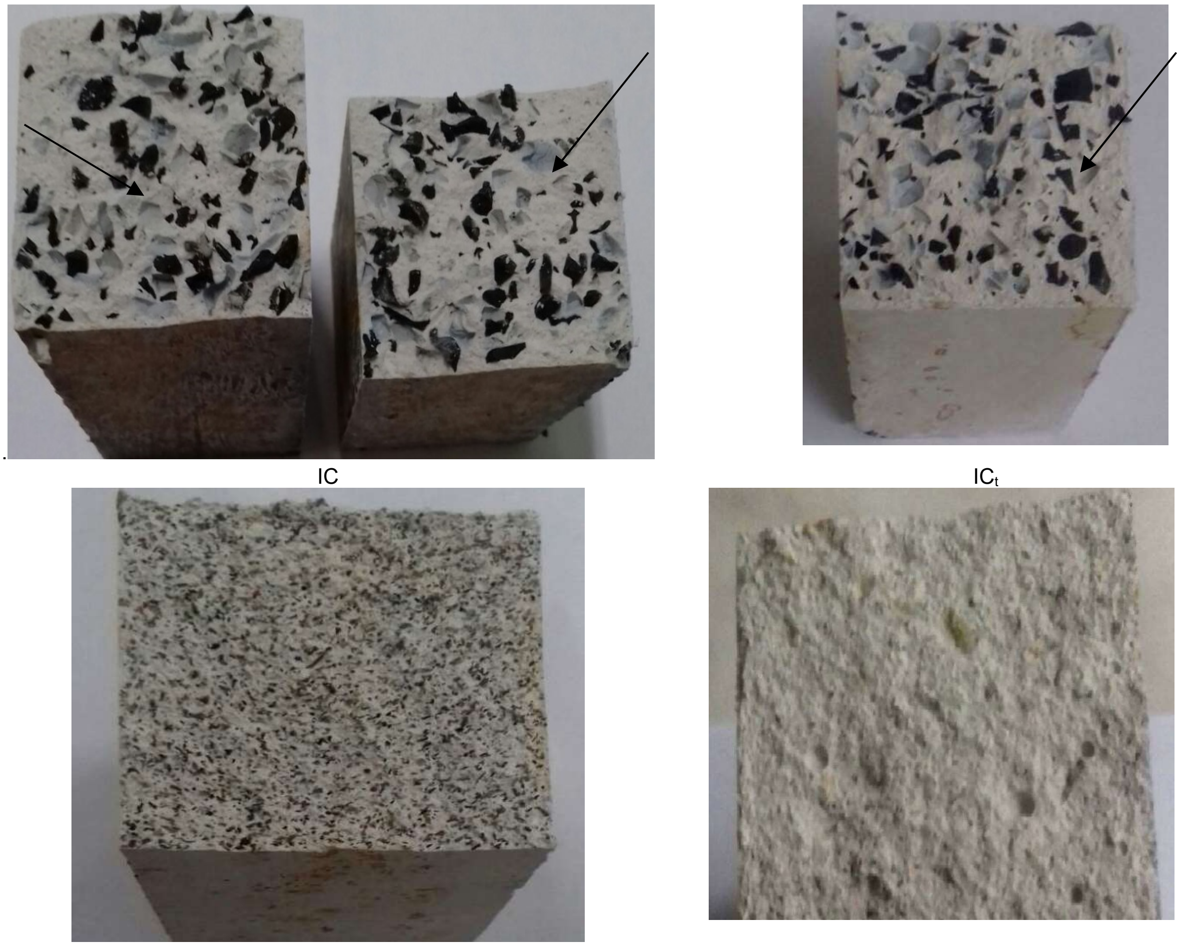
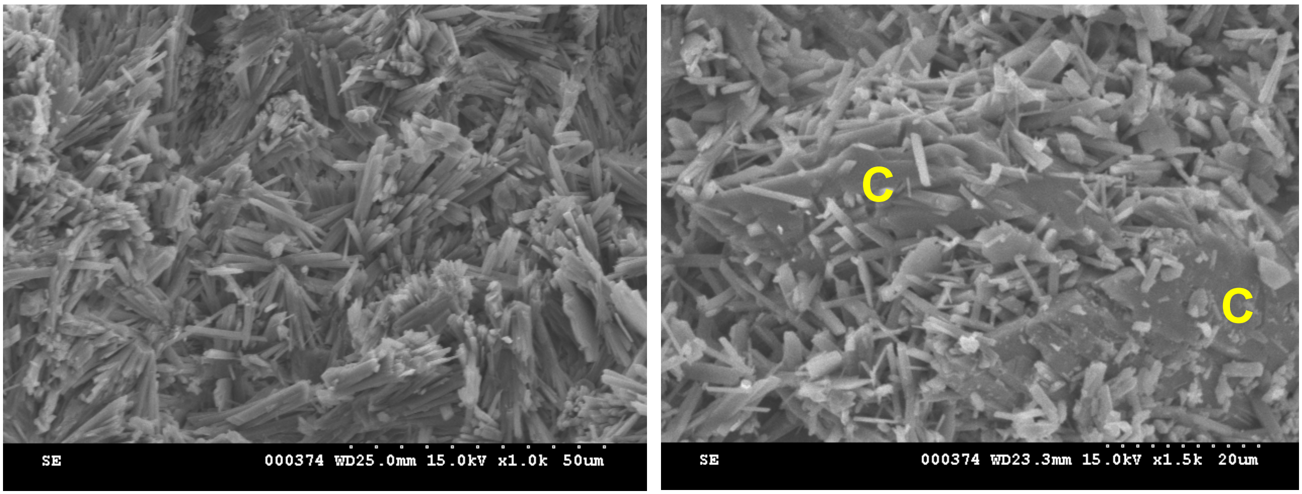

IC ICt
IE IP

Fig.4. – Visual aspect of gypsum composites with different types of waste / *Aspectul materialelor compozite pe bază de ipsos cu diferite tipuri de deșeu.*



a b

Fig.5. – SEM image of plaster (a) and composite with waste rubber (b) hardened for 7 days / *Micrografii SEM ale ipsosului (a) și ale materialul compozit cu deșeu de cauciuc (b) după 7 zile de întărire.*

and IC_t can be explained by the higher plaster content in this formulation (95% as compared with 70%). However, the compressive strength of IP and IE are much smaller as compared with reference (almost 70% strength loss). The high internal porosity of these wastes (especially polyurethane) contributes to the decrease of mechanical strength.

The substitution of plaster with the studied waste determines, as expected, the reduction of apparent (geometrical) density (Table 2). The highest decrease of the apparent density was

Table 2

Density and thermal conductivity of composites / *Densitatea și conductivitatea termică a compozitelor*

Sample/ *cod probă*	Density/ *Densitate* (kg/m^3)	Thermal conductivity 10°C/ *Conductivitate termică la 10°*C (W/m.K)
I	1253	0.2112
IC	1032.9	0.1410
ICt	1038.3	0.1658
IE	1015.1	0.0951
IP	890.6	0.1612

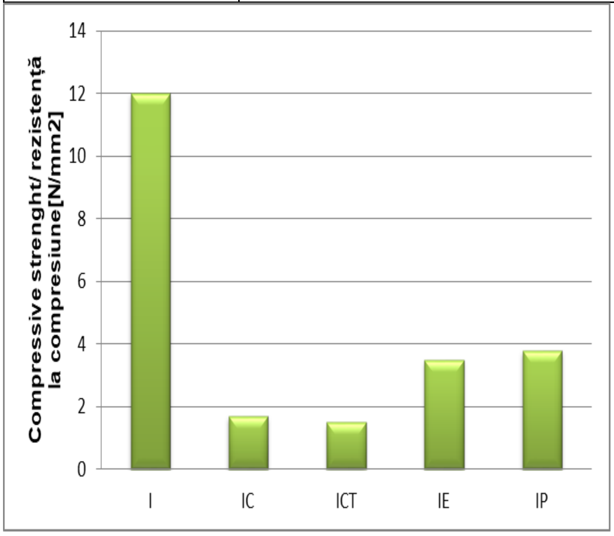


Fig.6. – Compressive strength of gypsum composites after 7 days of hardening/ *Rezistența la compresiune a compozitelor pe bază de ghips după 7 zile de întărire [N/mm^2].*

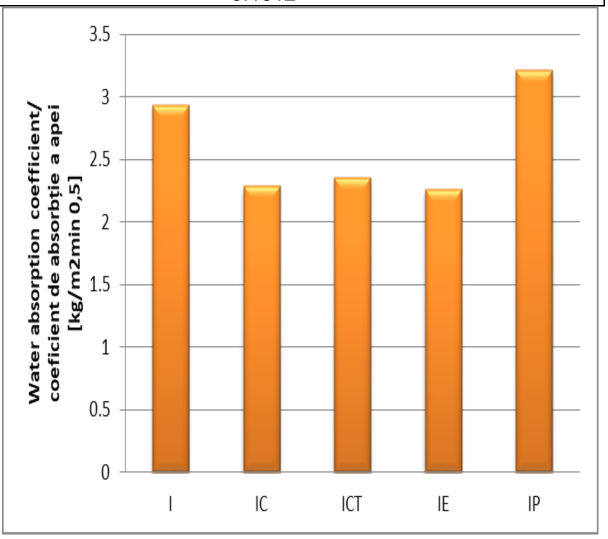


Fig.7 - Water absorption coefficient of gypsum composites with waste content/*Coeficientul de absorbție a apei determinat pe compozitele pe bază de ghips cu conținut de deșeuri.*

recorded for the composites with polyurethane in good correlation with the low density of this waste; the density assessed with helium pycnometer was 0.94 g/cm^3 for polyurethane waste (P) and 1.17g/cm^3 for rubber particles (C).

As expected, the decrease of apparent density (increase of the porosity) determines also the decrease of thermal conductivity (table 4).

Thermal performance of gypsum composites increases with 21-55%, after partial substitution with different types of waste, thereby improving the thermal insulation properties.

The values of water absorption coefficient due to capillary action are presented in Figure 7; these values depend on the pore microstructure and connectivity of hardened gypsum composites, as well as the water absorption specific for the waste. The highest value of water absorption coefficient was obtained for the composite with polyurethane waste, in good correlation with the high porosity (low density) of this material. On the opposite side, the plaster substitution with rubber waste and chopped electric cables determined a decrease of water absorption coefficient; these data suggest a higher impact of waste porosity on the values of water absorption coefficient compared with the porosity of gypsum matrix.

4.Conclusions

The main conclusions of this study are:

- The plaster substitution with organic wastes (rubber, polyurethane and chopped electric cables) determines an important decrease of compressive strength; the NaOH treatment of rubber waste does not improve the compressive strengths, opposite to its effect in composites based on Portland cement; this is due to the specific microstructure of ITZ in gypsum composites i.e. rubber particles are surrounded by a "felt"-like matrix consisting from needle-like gypsum crystals.
- The density of gypsum composites with waste content decreases due to the partial substitution of plaster with organic wastes with a lower density (high intrinsic porosity).
- The increase of porosity of gypsum composites determines the decrease of the thermal conductivity i.e. improves the thermal properties of these materials as compared with plain gypsum.
- The water absorption of these composites depends to a larger extent on the intrinsic characteristics of waste.
- The results presented in this paper highlight the possibility to valorise three types of wastes (rubber particles, chopped electric cables

and polyurethane foam) in the manufacture of cost effective and environmentally friendly thermal insulation materials; the best results i.e. the lower thermal conductivity and adequate compressive strength, were obtained for the plaster composites with chopped electric cables.

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