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THE LIGNOCELLULOSIS AND VERMICULITE COMPOSITE AS FIRE BARRIERS

Lignocellulosic boards are one of the most popular materials widely used in the building industry for outfitting (furniture) and for interior decorating (wall and ceiling panelling). This type of boards possesses considerable advantages such as high mechanical strength, thermal insulation properties and possibility of different type of finishing application. Due to their unfavourable behaviour under fire conditions however their use has often been limited particularly in tall and public buildings. Investigation of the Institute of Natural Fibres concerning the flame-retardant particleboard's with application both organic and mineral binders and mineral geopolymers were carried. Technology of three-layer composite board manufacturing in which the inner layer is made of lignocellulosic particles while the outer layers of expended vermiculite as fire barrier was developed. The surface finishing of these composite boards can be made by covering them by veneer, foils (Al), laminates, paints and intumescent fire barrier. The influence of different ratio of the mineral particles to the lignocellulosic particles and different fractions (granulation's) both natural and expanded vermiculite on flammability and physic-mechanical properties of one- and three-layer particleboard are presented.

KOMPOZYTY Z CZĄSTEK LIGNOCELULOZOWYCH I WERMIKULITU JAKO BARIERY OGNIOWE

Płyty lignocelulozowe są jednym z najbardziej powszechnych materiałów stosowanych w przemyśle budowlanym, wyposażeniowym (meble) oraz dekoracji wnętrz (panele ścienne i sufitowe). Jednakże niekorzystne zachowanie w warunkach pożaru ogranicza zakres ich wykorzystania. Obecnie stosowane metody i środki obniżają, do pożądanego stopnia, palność płyt lignocelulozowych. Jedną z metod jest dodanie wypełniaczy mineralnych w procesie wytwarzania płyt. Wypełniacze mineralne, dodawane do płyt w postaci cząstek o różnej granulacji, oddzielają materiał łatwo palny, jakim są cząstki lignocelulozowe zawarte w płytach, oraz obniżają przewodnictwo cieplne, co łącznie daje efekt ognioodporności płyt. Najpopularniejszymi wypełniaczami mineralnymi stosowanymi w tym celu są wermikulit i perlit.

Do wytwarzania niniejszego kompozytu zastosowano paździerze lniane, wióry drzewne, wermikulit oraz termoplastyczne substancje zaklejające na bazie żywic syntetycznych: żywica 112E, Silekol M i Silekol W-1. W przeprowadzonych próbach laboratoryjnych w pierwszym cyklu badań wytwarzano płyty jednorodne, a następnie płyty trójwarstwowe, stosując na warstwy zewnętrzne wermikulit, a na warstwę wewnętrzną paździerze lniane lub wióry drzewne. Wermikulit stosowano w postaci niespęcznionej oraz spęcznionej w różnym udziale procentowym w stosunku do cząstek zaklejanych. Celem prób było uzyskanie odpowiedzi, w jakiej postaci wermikulit jest najbardziej przydatny do produkcji płyt, mając na uwadze względy ekonomiczne i jakościowe.

Przy wytwarzaniu płyt jednorodnych z zastosowaniem wermikulitu surowego udział wermikulitu kształtował się na poziomie 50÷80%, a dla postaci spęcznionej 80÷100%. W przypadku wytwarzania płyt warstwowych udział wermikulitu surowego i spęcznionego wynosił 40÷60%. Dla płyt jednorodnych z wermikulitem surowym efekt zabezpieczenia ogniochronnego w stopniu niezapalnym uzyskiwano, dodając do cząstek lignocelulozowych o ok. 30÷40% mniej wermikulitu niż w postaci spęcznionej. W przypadku wytwarzania płyt jednorodnych zauważalny wpływ na ilość wypełniacza miał również rodzaj cząstek lignocelulozowych (wióry, paździerze). Najlepsze rezultaty badań nad optymalną ilością wypełniacza uzyskano, wytwarzając płyty trójwarstwowe przy zastosowaniu wermikulitu spęcznionego na warstwy zewnętrzne (jako barierę ogniową), a paździerzy lub wiórów drzewnych na warstwę wewnętrzną. Rezultatem prowadzonych prac jest technologia wytwarzania trójwarstwowego niezapalnego kompozytu płytowego, w którym warstwę wewnętrzną stanowią cząstki lignocelulozowe, a warstwy zewnętrzne wermikulit spęczniony, stanowiący barierę ogniową. Właściwości opracowanego kompozytu, w porównaniu z typową płytą paździerzową, przedstawiono w tabelach 1 i 2 oraz na rysunku 2. Palność opracowanego kompozytu (tab. 1) określono, stosując dwa testy palności. Pierwszym z nich była Polska Norma PN-B-02874, będąca modyfikacją Normy Francuskiej NF P 92-501 dla materiałów budowlanych, zaś drugim był pomiar na kalorymetrze stożkowym przeprowadzony zgodnie z ISO 5660. Opracowany kompozyt płytowy stanowi dobry materiał płytowy, który, zgodnie z polskimi przepisami budowlanymi, należy do klasy materiałów niezapalnych. Wyniki otrzymane podczas testów w kalorymetrze stożkowym wskazują, że trójwarstwowe płyty kompozytowe nie zapalają się w strumieniu cieplnym 30 kW/m^2, a przy ekspozycji na działanie intensywniejszego strumienia cieplnego równego 50 kW/m^2 ogólna ilość wydzielonego ciepła jest trzykrotnie mniejsza niż w przypadku typowej płyty paździerzowej. Technologia produkcji płyt kompozytowych oraz ich właściwości fizyczne i mechaniczne są podobne do typowych płyt paździerzowych i wiórowo-paździerzowych. Płyty te mogą być stosowane w: wyposażeniu wnętrz, przemyśle budowlanym, budownictwie okrętowym, transporcie kolejowym.

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INTRODUCTION

Lignocellulosic boards are one of the most popular materials used in the building industry, for outfitting (furniture), and for interior decoration (wall and ceiling paneling). However, because of their unfavorable behavior under fire conditions, their use has often been limited, particularly in high-rise and public buildings.

There are methods and means which make possible to decrease, to a significant degree, the flammability of these boards. From the economic point of view, however, the best solution to this problem is the manufacture of ready-to-use fire retardant boards.

The ready-made fire retardant product can be obtained by the following methods [1-3]:

- implementation of fire retardant during production process,
- addition of mineral particles at the stage of board manufacture,
- application of non-flammable binders,
- modification (fire retardant treatment) of lignocellulosic particles.

These methods can be applied alternatively or jointly.

Addition of mineral fillers in the form of particles of different size brings about separation of flammable material (i.e. lignocellulosic particles present in a board) and reduction in thermal conductivity which results in acquiring fire retardant properties. Most commonly used mineral fillers are vermiculite and perlite, which belong to so-called „geopolymers" [4]. The one worth special attention is vermiculite. It is commonly known that vermiculite is a material displaying excellent thermoinsulating properties, which make it suitable for many applications, including manufacture of various fire retardant materials, also boards [5, 6]. Vermiculite has the structure of trioctahedral layers of mica separated with water molecules. When heated up to 500°C, it looses water and increases its volume considerably [7]. After cooling down, it can adsorb two layers of water again. Heating above 550°C causes reduction in vermiculite ability to rehydrate and calcination at about 700°C results in its complete disappearance.

EXPERIMENTAL

Flax shives, wood chips (shavingt) and vermiculite were used for the manufacture of non-ignitable particle boards.

Flax shive

Flax shives make an excellent raw material for the manufacture of lignocellulosic boards, particularly fireproofed ones, because in comparison with other materials used for the production of such boards, e.g. wood chips, they are characterized by a low content of substances which are easy-to-decompose and easy-to--ignite. It is due to the elimination of degradable matter from shives during processing aimed at obtaining flax fiber and as a result of this, the main components of flax shives are lignin and cellulose.

Bulk density of shives is about 120÷150 kg/m^3.

Wood chips

Depending on wood properties and techniques used, wood chips are obtained in different sizes, shapes, fraction content and bulk density. For the manufacture of composite boards in our laboratory, wood chips of bulk density 148 kg/m^3 were used.

Mineral particles - vermiculite

Unexfoliated vermiculite has a form of gold-and--brown or greenish flakes and can be classified into six grades. Its thermogravimetric curve, recorded on a thermal analyzer Shimadzu TGA-50H at temperature increase rate of 5°C/min, is shown in Figure 1.

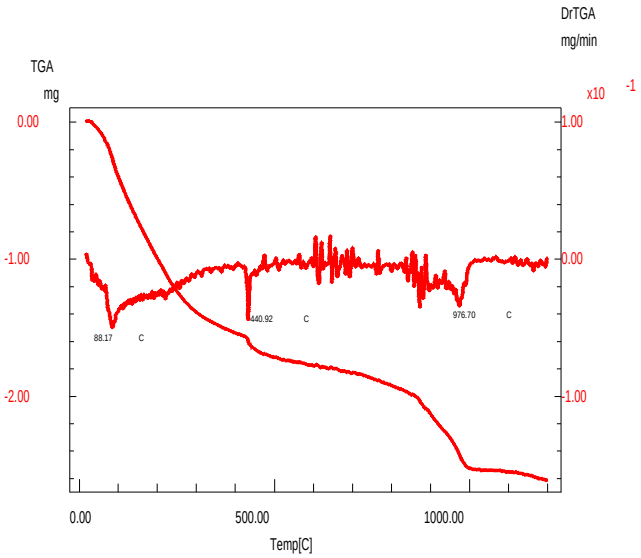


Fig. 1. Thermogravimetric analysis curve (TGA) and its derivative (DrTGA) recorded for unexfoliated vermiculite

Rys. 1. Krzywe termograwimetryczne wermikulitu surowego (TGA i DrTGA)

There are three peaks on DrTGA curve of unexfoliated vermiculite. Peaks with extremes at 88 and 441°C are associated with water removal which occurs in two stages (total mass loss up to 441°C is 10.5%) and the third peak with extreme at 977°C originates from unidentified impurity (total mass loss up to 977°C is 16%).

Such a conclusion can be drawn from comparison of our results with differential thermal analysis curves of vermiculites reported by MacKenzie [8] which shows that peaks in Figure 1, associated with water removal, occur

in the temperature range typical of vermiculites, whereas the origin of the peak at 977°C is certainly different from that recorded by MacKenzie at about 900°C. The latter originates from recrystallization into enstatite (phase transformation occurring without change in mass) and could appear on DTA curve, but not on thermogravimetric curve. Therefore the peak at 977°C recorded for our sample of unexfoliated vermiculite is caused by decomposition of an impurity accompanied by mass loss. It is worth to add that the removal of water from vermiculite is not limited to the temperature range in which the first two peaks present in Figure 1 are observed. According to MacKenzie, dehydroxylation of vermiculite samples proceeds in a continuous way between 450 and 850°C [8], but mass loss is too slow to be reflected by a peak. Continuous descent of TGA curve in Figure 1 points to the occurrence of such a slow process.

In our studies we have used both unexfoliated and exfoliated vermiculite of 2 mm grain size (grade Fine-2). Bulk density of vermiculite of the above grain size was 620 and 130 kg/m^3 in the case of unexfoliated and exfoliated vermiculite, respectively.

Glues

The following thermosetting glues made of synthetic resins were applied to the production of non-ignitable boards: resin 112E, Silekol M and Silekol W-1. These resins were characterized by low formaldehyde content due to the addition of amino-magnesium scavenger of formaldehyde which makes possible to obtain boards of E 1.0-E 0.5 hygiene class.

RESULTS OF BENCH SCALE EXPERIMENTS IN THE MANUFACTURE OF BOARDS

In the laboratory tests, conducted at the first stage of the study, homogeneous boards and then three-layer boards with vermiculite in the face layers and flax shives or wood chips in the core layer were manufactured [11]. Vermiculite was used in unexfoliated and exfoliated form in different proportions to the bonded particles. The tests were aimed at establishing which form of vermiculite is the best for the production of boards taking into account the aspects of both economy and quality.

Results of previous studies of the manufacture of fireproofed composite boards [9, 10] showed that proper method of raw material bonding is of very high importance. The method of bonding was established experimentally by gradual gluing of lignocellulosic particles with particles of mineral filler in different pro-portions of bonding agent-containing mixture. The opti-mal method of bonding was selected for the tests.

While selecting technological parameters, the guideline was to maintain pressing parameters, especially time and temperature of pressing, on a level enabling to obtain boards with parameters meeting at least requirements for boards of E-1 hygiene class.

In the case of the manufacture of homogeneous boards by using unexfoliated vermiculite, its content ranged from 50÷80%, whereas in the case of exfoliated vermiculite - from 80÷100%. In three-layer boards vermiculite was used at the amount of 40÷60%.

The amount of unexfoliated vermiculite necessary to obtain non-ignitable homogeneous boards was by 30÷40% lower than that required to obtain such boards with exfoliated vermiculite. In the case of the manufacture of homogeneous boards the nature of lignocellulosic particles (wood chips, shives) also influenced the amount of mineral filler.

As it was already mentioned, boards made of shives show a better performance in flammability tests than those based on wood chips. It was confirmed by results of our studies because non-ignitable boards were obtained at vermiculite content of 50%, i.e. by 5% less than in the case of wood chip-based boards.

The structure of boards manufactured by using unexfoliated vermiculite was not as homogeneous as that of exfoliated vermiculite-containing boards because the content (by volume) of unexfoliated vermiculite was 18-fold lower than that of exfoliated one. For this reason non-flammable particles of vermiculite did not coat flammable lignocellulosic particles as closely as in case of exfoliated vermiculite, however, it was enough to obtain non-ignitable class material. Supposedly, this is associated with evaporation of water from unexfoliated vermiculite at elevated temperature of board pressing.

Best results with optimal amount of filler were obtained when manufacturing three-layer boards with exfoliated vermiculite in the face layers as fire barrier and shives in the core layer. This recipe was taken as a starting point for further research aimed at developing production technology of composite non-ignitable boards based on lignocellulosic particles and exfoliated vermiculite.

Research carried out at the Institute of Natural Fibres resulted in the development of production technology of non-ignitable three-layer boards based of lignocellulosic particles making core layer and exfoliated vermiculite making fire barrier.

PROPERTIES OF COMPOSITE FIRE RETARDANT NON-TOXIC LIGNOCELLULOSIC BOARDS

Flammability

Fire characteristics of composite three-layer boards were determined in comparison with typical shive particleboards of similar density in accordance with two fire

test procedures. First of them was the Polish Standard PN-B-02874, which is a modification of the French Standard NF P 92-501 for building materials. The second method was Cone Calorimeter in acc. with ISO 5660. The latter method enables to measure a set of thermal parameters based on oxygen consumption theory and to determine such characteristics of importance to the evaluation of material behavior during a real fire as: time to ignition (IT), heat release rate (HRR), total heat released (THR), peak HRR, mass loss rate (MLR), effective heat of combustion (HOC), and smoke generation expressed as extinction coefficient and specific extinction area (SEA). Results of flammability tests are shown in Table 1 and Figure 2.

Physical and mechanical properties

Physical and mechanical properties of the composite boards in comparison with standard shive boards are presented in Table 2.

TABLE 2. **Properties of three-layered composite boards compared to properties of shive boards**

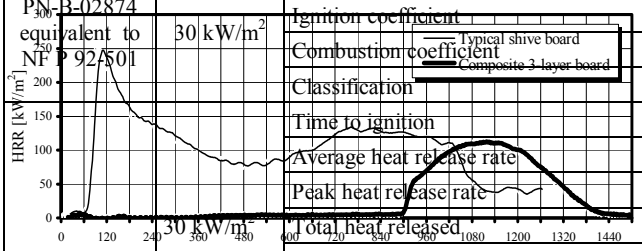
TABELA 2. **Właściwości trójwarstwowych płyt kompozytowych w porównaniu z płytami paździerzowymi**

Parameter	Unit	Type of board	
		shive	composite
Density	kg/m^3	600	640
Swelling in thickness after 24 h, no less than	%	20	6.4
Water absorption after 24 h, no less than	%	90	32.7
Static bending strength, no less than	MPa	15.0	14.8
Resistance to stretching in perpendicular direction, no less than	MPa	0.34	0.33
Ability to hold screws perpendicularly, no less than	N/mm	55	56
Ability to hold screws in parallel, no less than	N/mm	40	49
Heat conduction	W/(m · K)	0.100÷0.106	0.124
Formaldehyde content	mg/100 g	≤ 10	4.3

TABLE 1. **Results of flammability tests of typical shive board and composite three-layer board based on shives and vermiculite**

TABELA 1. **Wyniki testów palności typowej płyty paździerzowej i kompozytowej płyty trójwarstwowej opartej na paździerzach i wermikulicie**

Standard used while testing	Heat flux	Parameter	Unit	Typical shive particleboard	Composite three-layered board
PN-B-02874 equivalent to NF P 92-501	30 kW/m^2	Time to ignition	s	43.04	not ignited
		Ignition coefficient	-	1.65	0.00
		Combustion coefficient	-	2.84	0.03
		Classification	-	easy-to-ignite	non-ignitable
ISO 5660 Cone Calorimeter	30 kW/m^2	Time to ignition	s	89.29	
		Average heat release rate	kW/m^2	71.47	
		Peak heat release rate	kW/m^2	260.61	
		Total heat released	MJ/m^2	122.73	not ignited
		Average heat of combustion	MJ/kg	15.46	
		Average mass loss rate	g/s · m^2	7.59	
		Average specific extinction area	m^2/kg	30.71	
	50 kW/m^2	Time to ignition	s	33.28	865.24
		Average heat release rate	kW/m^2	109.85	73.52
		Peak heat release rate	kW/m^2	254.43	118.03
		Total heat released	MJ/m^2	121.93	41.64
		Average heat of combustion	MJ/kg	11.85	5.94
		Average mass loss rate	g/s · m^2	10.12	5.89
		Average specific extinction area	m^2/kg	40.34	94.30



Composite board was not ignited at the heat flux of 30 kW/m^2, irrespectively of the testing method used and therefore it can be classified as non-ignitable material according to the Polish Building Code. In the same conditions a typical particleboard was ignited. Under heat flux of 50 kW/m^2, composite particleboard was ignited after about 14 minutes and all parameters, except for smoke release, were significantly reduced in comparison with a typical shive particleboard.

Fig. 2. Heat release rate (HRR) from composite board in comparison with typical shive board in acc. with ISO 5660 at heat flux of 50 kW/m^2

Rys. 2. Szybkość wydzielania ciepła (HRR) z płyty kompozytowej w porównaniu z typową płytą paździerzową, oznaczona w kalorymetrze stożkowym (zgodnie z ISO 5660) w strumieniu cieplnym 50 kW/m^2

It results from Table 2 that physical and mechanical properties of composite boards are similar to those of shive boards. Some parameters, e.g. swelling in thickness and water absorption, are more advantageous. It is worth to add that non-ignitable composite boards are strong, durable, able to ensure tough mechanical joints and easy to decorative finishing. They can be shaped with typical wood processing devices.

CONCLUSIONS

1. Composite three-layer boards make a good board material included in the class of non-ignitable materials according to the Polish Building Code classification. This was confirmed by results obtained during cone calorimeter tests which showed that composite three-layer boards did not ignite at heat flux of 30 kW/m^2, and at higher heat flux of 50 kW/m^2 the total amount of heat released was three times lower than in the case of typical shive boards.
2. Production technology of boards as well as their physical and mechanical properties are similar to those of typical shive and wood chip-shive particleboards.
3. Composite non-ignitable boards are easy to decorative finishing, strong, durable and ensure tough mechanical joints.
4. The boards can find application to: interior fittings, building industry, shipbuilding, railway engineering.

REFERENCES

[1] Moslemi A.A., Particleboard, vol. 1-2, Southern Illinois University Press, Carbondale 1974.

[2] Maloney T.M., Modern Particleboard and Dry-Process Fibreboard Manufacturing, Miller Freeman Publications, Inc., San Francisco 1977.

[3] Lewin M., Atlas S.M., Pearce E.M., Flame-Retardant Polymeric Materials, Chapter 2: Lewin M., Sello S.B., Technology and Test Methods of Flameproofing of Cellulosics, Plenum Press, New York - London 1975.

[4] Zielinska K., Niepalne płyty krzemianowo-wapniowe do biernej ochrony przeciwpożarowej, Non-flammable Calcium-
-Silicate Boards for Fire Protection, Materiały Budowlane (Building Materials) (245):1, 13-15.

[5] Stoch L., Minerały ilaste (Silty Minerals), Geological Publishers, Warsaw.

[6] Lis J., Sylwestrzak H., Minerały Dolnego Śląska (Minerals of Lower Silesia), Geological Publishers, Warsaw.

[7] Görlich E., Chemia krzemianów (Silicate Chemistry), Geological Publishers, Warsaw 1957.

[8] Mackenzie R.C., Differential Thermal Analysis, vol. 1, Academic Press, London & New York 1970, 515.

[9] Kozlowski R., Helwig M., Przepiera A., Properties of Fire-Retardant Particleboards Glued with Adhesives on Basis of Polycondensation Products of Polyborates and Polyphosphates of Urea with Silicates, Proceedings of Technical Forum Presentation of 23rd International Particleboards/ Composite Materials Symposium, Pullman, USA 1989, 11.

[10] Kozlowski R., Helwig M., Przepiera A., Miciukiewicz A., Flame Retardant Composite Particleboards Based on By-products of Fibrous Plants and Other Materials, Pacific Rim Bio-Based Composites Symposium, Rotorua, New Zealand, FRI Bulletin 1992, No. 117, 320-325.

[11] Kozlowski R., Mieleniak B., Przepiera A., Plant Residues as Raw Materials for Particleboards, Proceedings of the 28th International Particleboard/Composite Materials Symposium, W.S.U., Pullman, USA 1994, 181-197.

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