Utilization and biodegradation of wastes

UDC 66.043.1:[666.198+661.635.2] DOI: 10.25514/CHS.2020.1.17006

Fiber heat insulation waste recycling in laminated high-temperature composites with phosphate binder

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Received: June 2, 2020, Revised: June 16, 2020, Accepted: June 18, 2020

Abstract – A recycling procedure is described for utilization of waste derived from spent fibrous refractory materials such as mullite-siliceous fibers, traditionally used for thermal insulation of high-temperature furnaces and other equipment. It is proposed to use the spent thermal insulation for manufacturing a composite material of the sandwich type, in which an intermediate layer consisting of the recycled fibers and a phosphate binder is located between two outer layers of an industrial roll material. In the case of using spent fibrous thermal insulation containing a binder the waste has been previously deagglomerated. The optimal composition of the intermediate layer containing a layered fibrous material of the mullite-siliceous fiber type (MKRP-130 or MKRKH-150) and an alumina-boron-phosphate/alumina-chromium-phosphate binder has been established, and the characteristics of the obtained composite materials have been determined.

Keywords: mullite-siliceous fibers, fiber waste, alumina-boron-phosphate binder, alumina-chromium-phosphate binder, laminated material.

Утилизация отходов волокнистой теплоизоляции в составе слоистых высокотемпературных композитов на фосфатных связующих

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Поступила в редакцию: 02.06.2020 г., после доработки:16.06.2020 г., принята в печать: 18.06.2020 г.

Аннотация – Описан способ утилизации отходов волокнистых огнеупорных материалов типа муллитокремнеземистых волокон (МКВ), традиционно применяемых для теплоизоляции высокотемпературных печей и другого оборудования. Предложено использовать отработанную теплоизоляцию для изготовления композиционного материала типа сэндвич, в котором между двумя слоями заводского рулонного материала находится промежуточный слой, состоящий из переработанных волокон и фосфатного связующего. Утилизированная волокнистая теплоизоляция, содержащая связующее, предварительно была дезагломерирована. Установлен оптимальный состав промежуточного слоя, содержащего слоистый волокнистый материал типа МКВ (МКРР-130 или МКРРХ-150) и алюмоборфосфатной/алюмохромфосфатное связующее; определены свойства этих материалов.

Ключевые слова: муллитокремнеземистые волокна, отходы волокон, алюмоборфосфатное связующее, алюмохромфосфатное связующее, слоистый материал.

INTRODUCTION

Fiber-based thermal insulation materials provide a very effective heat protection due to a useful combination of low values of thermal conductivity and volume weight with high resistance to thermal shock, etc. Insulation produced from such refractory materials improves the productive capacity of heat aggregates which results in lowering cost of the cladding and reducing fuel consumption [1, 2].

After long-term operation of fibrous materials at high temperatures, their gradual disintegration is observed, which leads to dismantling and replacement of the spent thermal insulation. The gradual degradation of the structure of the worked out constructions results in the release of fine particles that are hazardous for respiratory tracts of humans and animals. In addition, the heat insulation layer replacement is accompanied by the formation of a waste which could contain bound fibers.

Consequently, despite a considerable interest of researchers and industry in fibrous refractories [3–5], a problem of utilization of spent fiber heat insulation still remains unresolved issue except for several disparate attempts [6–11].

We offer a utilization procedure of fibrous refractory waste by using it for producing a sandwich type composite material with an intermediate layer consisting of recycled refractory fibers and a phosphate binder, which is imbedded between two outer layers of a factory roll material. Thus, an interlayer is a composition of spent heat insulation fibers and alumina-boron-phosphate binder (ABPB) and/or alumina-chromium-phosphate binder (ACPB).

Phosphate compounds are widely used in various application fields, such as food additives, detergents, fertilizers, etc. Their benefits and drawbacks are well-known as well as safety precautions for handling the binders. The phosphate binder usage does not require a preliminary high temperature treatment. Besides, the obtained constructions are characterized by low shrinkage. Phosphate binders are homogeneous metastable aqueous solutions of phosphate compounds which undergo spontaneous solidification resulting from their transformation under heating, pH change, hardener adding or dehydration [3]. At the same time, this transformation is accompanied by the formation of newly generated colloidal particles on a fiber surface, which are gradually transformed into crystal form which provides a several-fold increase in the resulting composite strength. A more detailed description of mullite-siliceous fibers and phosphate binders is given in [12, 13].

EXPERIMENTAL

Industrially produced roll materials based on the following high-temperature fibers were used, namely:

- mullite-siliceous roll material of trademark MKRR-130 (GOST 23619-79), produced by the refractory plant “Sukhoy Log” (Russian Federation) with the following characteristics: Al_2O_3 content is 51.0 wt% (hereinafter, the composition of all materials is given in wt.%), Al_2O_3 + SiO_2 = 99.4%, loss on ignition is 0.1%, average fiber diameter is 2–3 μm, content of non-fibrous inclusions (beads) with a size of more than 0.5 mm is 0.7–1.9%, apparent density is 80–99 kg/m^3 [14];
- chromium-mullite-siliceous roll material of trademark MKRRC-150 (GOST 23619-79), produced by the refractory plant “Sukhoy Log” (Russian Federation) with the following characteristics: Al_2O_3 content is 51.3%, Al_2O_3 + SiO_2 = 95.8%, Cr_2O_3 content is 3.14%, loss on ignition is 0.44%, average fiber diameter is 2 μm, non-fibrous inclusions (beads) with a size of 1.9–3.0%, apparent density is 103–134 kg/m^3 [15].

Alternatively, wastes of high-temperature insulation fibers containing binder residues were used. The following binders were chosen:

- alumina-boron-phosphate binder (ABPB), produced by Biysk chemical plant (Russian Federation) (according to technical specifications 113-08-606-87) with the following composition: P_2O_5 36–39%, Al_2O_3 7.5–9.5%, B_2O_3 1.0–2.0%, density is 1578 kg/m^3, pH $\geq$ 1 [16];
- alumina-chromium-phosphate binder (ACPB), produced by Biysk chemical plant (Russian Federation) (according to technical specifications 6-18-166-83) with the following composition: P_2O_5 35–39%, Al_2O_3 6,5–9%, Cr_2O_3 3.5–4.5%, SO_4^{2-} $\geq$ 0.5%, formaldehyde $\geq$ 0.2%, loss on ignition 47–55%, density is 1470 kg/m^3, pH $\geq$ 1 [17].

Micrographs were obtained by means of a binocular microscope MICROMED (Optical Instruments LLC, Russian Federation) with a digital video ocular (resolution is 1.3 Megapixel, general magnification x1000).

Thermal conductivity was detected by means of an installation MIT-1 (probe method) (INTERPRIBOR, Russian Federation).

Procedure for fabrication of laminated sandwich type composite material

It seemed rational to use for recycling fiber waste without a binder by repulping the waste in water in a vessel with a magnetic stirrer for 3 min followed by squeezing an excess water until the ratio *fiber waste : water* is equal to 1 : 4. The obtained moist mass was mixed with a predetermined quantity of ABPB/ACPB binder. This suspension acted as a binder for the targeted composite material. Then it was uniformly distributed between two outer layers of the industrial roll material. A cantledge was placed onto the surface of the prepared sandwich type composition (under the pressure of 120 Pa), then it was put into a muffle furnace (300°C) or into a microwave oven (2.45 GHz, 6 kWt). Afterwards, a set of technical characteristics of the composite (volume weight, flexural strength, thermal conductivity) was determined.

Fiber waste containing some quantity of a binder were preliminarily deagglomerated mechanically in a mill (planetary or ball-ring).

RESULTS AND DISCUSSION

The optimal conditions for preparing the laminated composite based constructions along with their composition ratio were determined. The upper and lower layers of the sandwich construction consisted of the industrial roll material (MKRR-130/MKRRC-150). The interlayer of the sandwich material consisted of a phosphate binder as a matrix and disintegrated fibrous waste as a filling agent.

As can be seen from a micrograph of the fiber waste, it is characterized by a nonuniform structure (Fig. 1) consisting of long and short fibers, beads and other fragments of irregular shape and size.

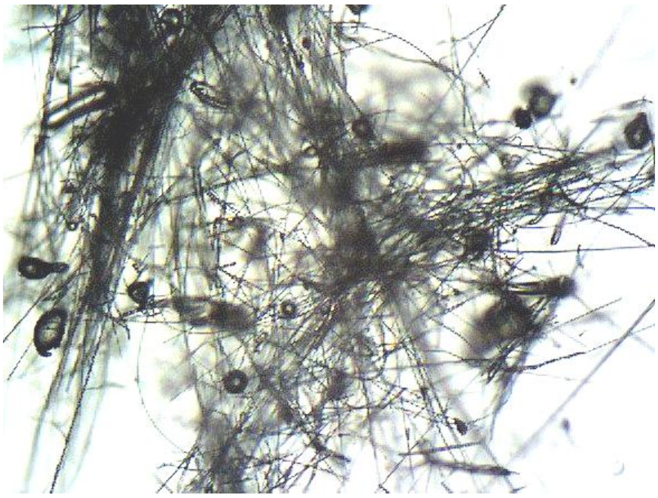


Fig. 1. Micrograph of MKRR-130-based waste after disintegration (x100).

Fiber waste after repulping in water kept its elasticity and a high resistance to flexural strain. Being mixed with a phosphate binder solution, these fibers formed a complex binder agent.

One of the key aspects of the research was to provide a balance between an adherence, an adequate strength, a low volume weight and the maximum possible volume of the utilizable waste. In order to determine the interlayer composition of the sandwich we varied the binder content range. The quality of the prepared samples was evaluated visually followed by testing their properties.

Samples with low level of binder had slightly curved form and delaminated segments in layer's junctions. An increase of the phosphate binder content in the interlayer resulted in retaining the composite plasticity, and the samples sagged under tension without any loss of integrity.

As expected, a binder content increase yielded a simultaneous growth of the material density and strength. A too high increase in fiber content resulted in reducing the composite strength due to the poor damping of the fiber. Consequently, fiber content increase was unfavorable as fibers were badly covered by the phosphate binder. As a result, it led to an insufficient adhesion of the matrix with fibers.

Considering the above, the following composition ratios were chosen as optimal for composite derived from the fiber waste and the binder:

– fibrous waste of MKKR type : ABPB = 1 : 3;
– fibrous waste of MKKR type : ACPB = 1 : 2;
– fibrous waste of MKKRC type : ABPB/ACPB = 1 : 3.

Figure 2 illustrates a typical appearance of the prepared laminated sample.

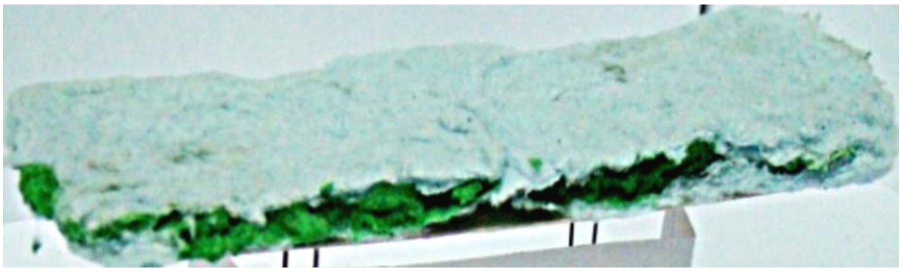


Fig. 2. Laminated composite sample produced from MKRRC-150 type fiber waste and alumina-chromium phosphate binder (waste : binder ratio = 1 : 3).

Parallel testing of two kinds of the thermal treatment procedures of the laminated composites was carried out: i) in a muffle furnace (at 300°C), and ii) in a microwave oven. It was determined that samples treatment in a muffle furnace during ~20 min was equal to 5 min of heating in the microwave field in terms of flexural strength of the composite samples (Table 1).

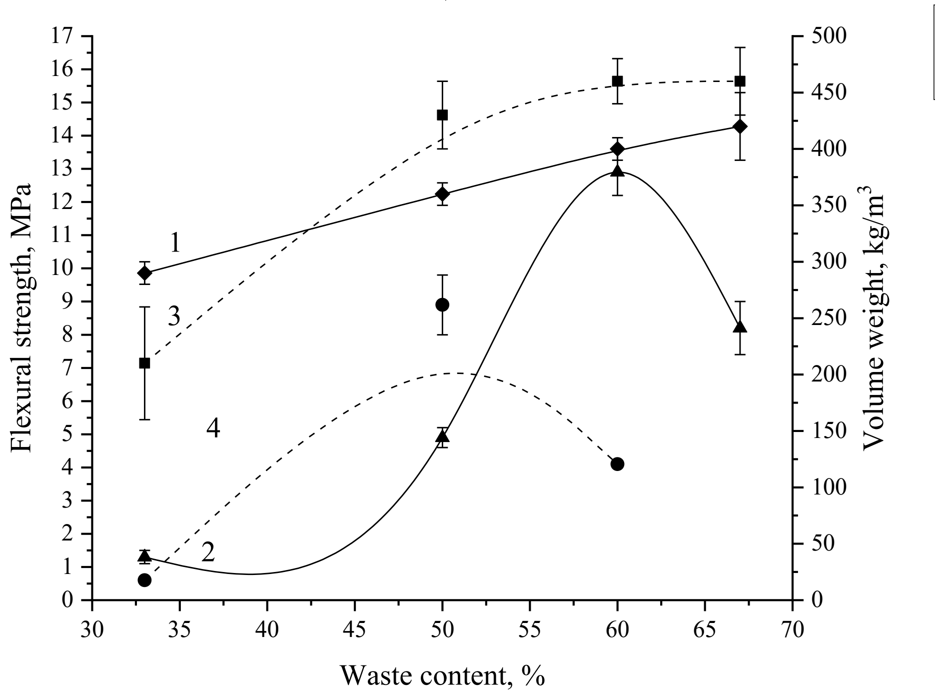
Table 1. Effect of thermal treatment mode on laminated material strength

Thermal treatment mode	Flexural strength, MPa	
	MKRR-130	MKRRC-150
Muffle furnace (300°C), 20 min	1.1 ± 0.1	2.2 ± 0.1
Microwave oven, 5 min	0.9 ± 0.2	2.7 ± 0.2

Alternatively, we used binder-containing fiber heat insulation wastes which are known to be badly repulped in water. So, it was difficult to obtain a homogeneous composite binder from this type of waste. Thereby, an effect of preliminary mechanical treatment of the waste (attrition- or impact-assisted treatment) on morphological peculiarities of the obtained materials was studied. Preliminarily dispersed fibers formed fragments with a high content of powder fraction; an attrition mediated treatment provided a higher quantity of fine-dispersed particles. As a result, the fiber was uniformly distributed in the binder volume and the laminated composite had more deeply impregnated fiber layers which resulted in the flexural strength increase of the end products.

From ecological point of view, it is desirable to utilize the greater volume of waste, so it would be preferable to achieve the maximum interlayer thickness in the laminated composite. However, an increase of the composite interlayer part impairs the fencing function of the outer roll material which is responsible for the strength and stability under bending loads. The experiments on varying waste content in the interlayer composition revealed the retention of high strength level up to ~60–67% of the fiber (Fig. 3). The further growth of the waste content was undesirable because of the strong composite embrittlement.

a)



b)

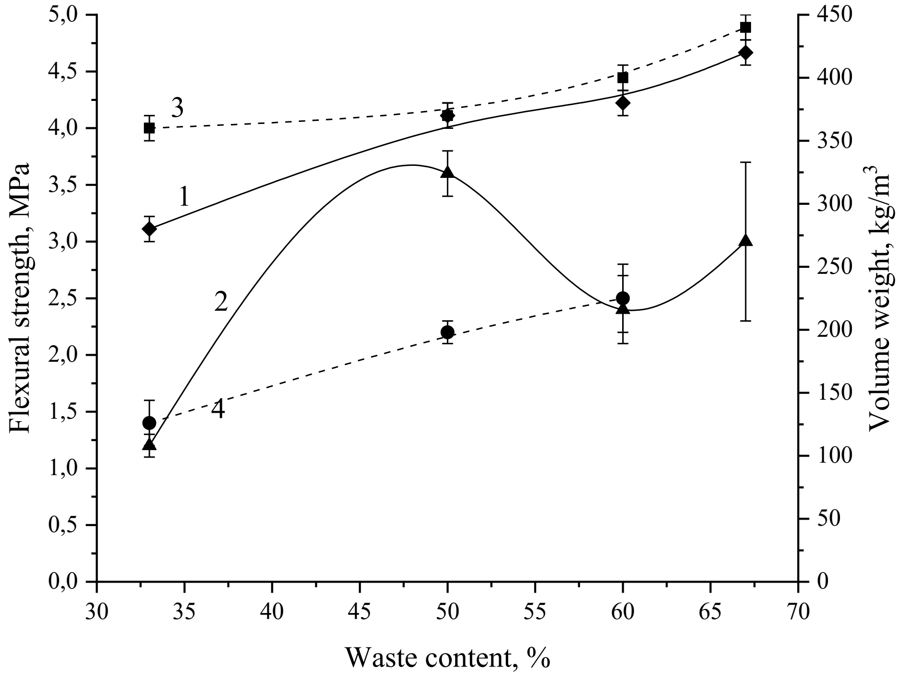


Fig. 3. Effect of waste content in fibrous materials upon laminated composite strength (curves 2, 4) and volume weight (curves 1, 3). Fiber waste type: MKRR-130 (*a*), MKRRC-150 (*b*). Binder: alumina-chromium-phosphate (solid lines), alumina-boron-phosphate (dotted lines).

The obtained laminated composites had high level of flexural strength (Fig. 4).

a) *b)*

Fig. 4. Loaded laminated constructions produced from of MKRR-130 fibers and ACPB (*a*); MKRR-130 fibers and ABPB (*b*). Load mass: 1.5 kg (*a*), 0.7 kg (*b*).

The manufacturing of profiled laminated products from the developed laminated composites based on the fibrous waste was described earlier [18].

It was determined that the thermal conductivity of the prepared laminated composite materials containing fiber waste varied in the range of 0.03–0.05 Wt/(m^2·K) for all the studied waste types. So, the thermal characteristics of the obtained composites are similar to that of the known composite material Triton Kaowool (England) based on alumina-silicate fibers, which is characterized by the thermal conductivity values in the range of 0.03–0.04 Wt/(m^2·K).

CONCLUSIONS

Summarizing the results, we have developed a laminated heat insulation material of sandwich type containing a composite binder produced from fiber waste and phosphate binder compounds. A possibility is demonstrated of using fibrous refractory waste as a component of the composite binder in laminated constructions with good thermal and mechanical characteristics.

An optimal composition of the complex binder based on mullite-siliceous fiber refractory waste and phosphate binding agents has been determined and the binder-fibrous waste ratios have been recommended.

References:

1. Kashcheev, I.D., Strelova, K.K., & Mamykin, P.S. (2000). *Refractories for industrial units and furnaces.* Reference edition in two books. Book 1. M.: Intermet Engineering (in Russ).
2. Mazur, V.L., Ryabov, A.I., & Mazur, V.V. (2005). The solution to the problem of heat conservation and thermal insulation in metallurgy. *Proceedings of the International Conference of refractory materials and metallurgists.* M.: New Refractories (pp. 51 - 56) (in Russ).

3. Bolden, J., Abu-Lebdeh, T., & Fini, E. (2013). Utilization of recycled and waste materials in various construction applications. *American Journal of Environmental Science, 9(1),* 14 - 24. https://doi.org/10.3844/ajessp.2013.14.24
4. Nayab-Ul-Hossain, A.K.M., Sela, S.K., & Sadeque, S.B. (2019). Recycling of dyed fiber waste to minimize resistance and to prepare electro thermal conductive bar. *Results in Engineering, 3*(100022). https://doi.org/10.1016/j.rineng.2019.100022
5. Mukherjee, B., Das, S.K., Ghosh, A., & Banerjee, G. (2003). Effect of sillimanite beach sand composition on mullitization and properties of Al_2O_3-SiO_2 system. *Bull. Mater. Sci., 26*(2), 217 - 220. https://doi.org/10.1007/BF02707794
6. Daigo, I., Kiyohara, Sh., Okada, T., Okamoto, D., & Goto, Y. (2018). Element-based optimization of waste ceramic materials and glasses recycling. *Resources, Conservation & Recycling, 133*, 375 - 384. https://doi.org/10.1016/j.resconrec.2017.11.012
7. Petropavlovskaya, V.B., Novichenkova, T.B., Bur'yanov, A.F., Solov'yev, V.N., & Petropavlovskiy, K.S. (2017). Utilization of mineral fiber waste in the production of gypsum products. *Vestnik MGSU = Bulletin of Moscow University of Civil Engineering, 12(*12*)*, 1392 - 1398 (in Russ.). https://doi.org/10.22227/1997-0935.2017.12.1392-1398
8. Bartl, A., Hackl, A., Mihalyi, B., Wistuba, M., & Marini, I. (2005). Recycling of fibre materials. *Process Safety and Environmental Protection, 83*(B4), 351 - 358. https://doi.org/10.1205/psep.04392
9. Wang, Y. (2010). Fiber and textile waste utilization. *Waste and Biomass Valorization, 1*, 135 - 143. https://doi.org/10.1007/s12649-009-9005-y
10. Feng, Yan-chao, Zhao, Feng-qing, & Xu, H. (2016). *Recycling and Utilization of Waste Glass Fiber Reinforced Plastics.* MATEC Web of Conferences, 67, 07012. DOI:10.1051/matecconf/20166707012
11. Hossain, S.K.S., & Roy, P.K. (2019). Fabrication of sustainable insulation refractory: Utilization of different wastes. *Bulletin of the Spanish Society of Ceramics and Glass, 58*, 115 - 125. https://doi.org/10.1016/j.bsecv.2018.09.002
12. Sudakas, L.G. (2008). *Phosphate binders.* SPb: RIA Kvintet (in Russ).
13. Pimkov, Yu., Filatova, N., & Kosenko, N. (2014). *Heat-insulating composites based on aluminosilicate fibers. Obtaining, properties, application.* LAP Lambert Academic Publishing (in Russ).
14. Filatova, N.V., Pimkov, Yu.V., & Kosenko, N.F. (2013). Development of the composition of the fibrous composite material using waste refractory insulation. *Izv. Vyssh. Uchebn. Zaved. Khim. Khim. Tekhnol. = Russian Journal of Chemistry and Chemical Technology*, *56*(12), 74 - 79 (in Russ).
15. GOST (State Standard) 23619-79. Refractory heat-insulating mullite-silica glass-fibrous materials and products (in Russ).
16. TU (Technical Specifications) 113-08-606-87. Aluminobrphosphate concentrate (in Russ).
17. TU (Technical Specifications) 6-18-166-83 Alumochromophosphate binder (in Russ).
18. Filatova, N.V., Pimkov, Yu.V., & Kosenko, N.F. (2014). *Heat insulation products of sandwich type from refractory fibrous waste.* Ecology and Industry of Russia, 4, 21 - 23 (in Russ).