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THERMAL INSULATING MATERIALS BASED ON MAGNESIUM-CONTAINING TECHNOGENIC RAW MATERIALS

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ABSTRACT

The article considers the possibility of replacing traditional mineral raw materials (dolomite and asbestos) to obtain thermal insulating material – sovelite – with technogenic raw materials, i.e. tails of polymetallic ores (dolomite-barium tails) and synthetic wollastonite, obtained on the basis of phosphoric slag. The research results allowed us to establish that the dolomite-barium "tails" can serve as a technogenic raw material base to obtain thermal insulating material – sovelite. The advantages of the proposed technology to obtain thermal insulating material –sovelite– in comparison with the traditional one allow for reducing energy costs for production due to the absence of stages of raw material extraction, crushing, and grinding, a reduced firing temperature, the absence of the stage of grinding the fired product since during firing there is waste dispersion, natural dolomite saving, replacement of expensive natural reinforcing material – chrysotile asbestos–with synthetic wollastonite, obtained on the basis of phosphoric slag. The use of synthetic wollastonite significantly improves the quality characteristics and increases the service life of the products.

Keywords: Dolomite-Barium "Tails", Firing, Synthetic Wollastonite, Chrysotile Asbestos, Natural Wollastonite, Thermal Insulating Material-Sovelite

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INTRODUCTION

The development and improvement of the production of building materials and an increase in their economic efficiency at the present stage will largely be determined by the rationality of the use of raw materials, and the completeness of involvement in the production of wastes from various industries.[1-10] In the industrially developed regions of the Republic of Kazakhstan, the volumes of accumulated technogenic formations are so significant, and the possibilities for their use are so diverse that they require proper and immediate accounting of them along with natural mineral resources.[3-12] Mining and processing production in the Republic of Kazakhstan, as well as in other countries, is still characterized by a high level of output of industrial wastes in the form of rock refuses, stripping and mining faces, tails of concentration processing, metallurgical slags. In fact, for each ton of mined ore, there are up to 0.15-0.25 tons of associated rock and up to 0.8 tons of mine refuse.[9-20] To date, mining and processing enterprises are faced with the need to process their own technogenic dumps, especially tails, and out-of-balance ores for the following reasons:

- The service life of tailing dumps is limited, the filling of many has already been completed or is coming to an end in the coming years. The trend towards the accumulation of dumps is growing due to the need to introduce complex ores into processing.

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- Dumps occupy vast territories and are a source of environmental risk for the areas of operation of mining and processing complexes, fines are growing.
- Dumps are huge amounts of preserved raw materials both for the needs of non-ferrous metallurgy and for the production of building materials. Therefore, the relevance of the complex processing of technogenic deposits with the maximum extraction of metals, utilization of the nonmetallic part, and improvement of the ecological situation is of equal ecological and economic importance.

In this regard, the technogenic dumps of the Khantagy tailings dump in the form of tailings of polymetallic ores of JSC "Achpolymetal", whose reserves amount to more than a hundred million tons, are of considerable interest. The choice of these wastes as an object of study to obtain thermal insulating material – sovelite – is due to the fact that in the process of concentration of polymetallic ores, the homogeneity of the resulting wastes significantly increases, both in chemical and mineral composition.[20-25] The main thermal insulating materials that are widely used today are mineral cotton and polystyrene foam, which, with all their advantages, have obvious disadvantages. Mineral cotton degrades over time during operation – shrinks, forming spaces unprotected from heat leakage. Polystyrene foam is a combustible material. In this regard, the search for new types of raw materials and the organization of the production of thermal insulating materials of good quality from industrial wastes is an urgent and ecologically expedient task. Thermal insulating materials are materials and products used for the thermal insulation of buildings, structures, technological equipment, etc. They are characterized by low thermal conductivity, high porosity, low bulk density, and strength. The main purpose of using such materials is to reduce energy consumption for heating a building. In addition, the use of thermal insulation in building construction can significantly reduce the mass of structures, and reduce the consumption of wall-building materials (cement, metal, wood, ceramics, etc.). The use of thermal insulating materials is very effective in energy, metallurgy, mechanical engineering, chemical, and environmental processes.

EXPERIMENTAL

During the research, the following materials were used: tails of polymetallic ores (Khantagy, Kazakhstan); chrysotile asbestos of Dzhetigary deposit (Kazakhstan); natural wollastonite of Verkhne-Badam deposit (Kazakhstan); synthetic (artificial) wollastonite. Physicochemical methods of analysis were used as analytical methods for the research, in particular: chemical, electron microscopic, thermal engineering, X-ray phase, and physicomechanical.[1,4,6-10,13,20-22]

RESULTS AND DISCUSSION

The tails of polymetallic ores are a finely ground product that does not require additional grinding before use. The main minerals in the tails are dolomite 50-60%; limestone 10-15%; barite 10-20%; clay substances 5-8%; ore minerals 2-3%. Ore minerals are represented by sulfides of iron, lead, and non-ferrous metals. Since the tails of polymetallic ores consist mainly of dolomite, calcite, barite, and clay substances, it is advisable to use both unfired and fired wastes in the building materials industry.[19-25] The chemical composition of the tails of polymetallic ores is characterized by stability and is presented in Table-1. The wastes contain catalytic and modifying elements, mass percent: Zn 0.01-0.05; Cu 0.002-0.004; Ti 0.03-0.05; Cd 0.002-0.003. The absence of toxic emissions, low activity of radionuclides 53-55Bq/m^2, and low volatility of heavy metals testify to the radiation and environmental safety of wastes.[1,4,6,26-36]

Table-1: The Chemical Composition of the Tails of Polymetallic Ores

Content of oxides, mass percent										
SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	$BaSO_4$	FeS_2	$PbSO_4$	$PbCO_3$	PbS	Loss on ignition
4.34-6	0.98-1.2	2.86-3.5	27.79-29	14.03	12.7-13.5	1.39-1.5	0.03-0.05	0.09-1.2	0.14-0.2	35.25-37

Chrysotile asbestos of Dzhetigary deposit, used as a reinforcing additive in the production of thermally insulating material–sovelite, is a hydrous magnesium silicate, the theoretical composition of which corresponds to the formula $3MgO \cdot 2SiO_2 \cdot 2H_2O$, which corresponds to the mass ratio of MgO – 42.4%, SiO_2 – 44.5% and H_2O – 13.04%. In the presence of impurities, the amount of MgO and SiO_2 of

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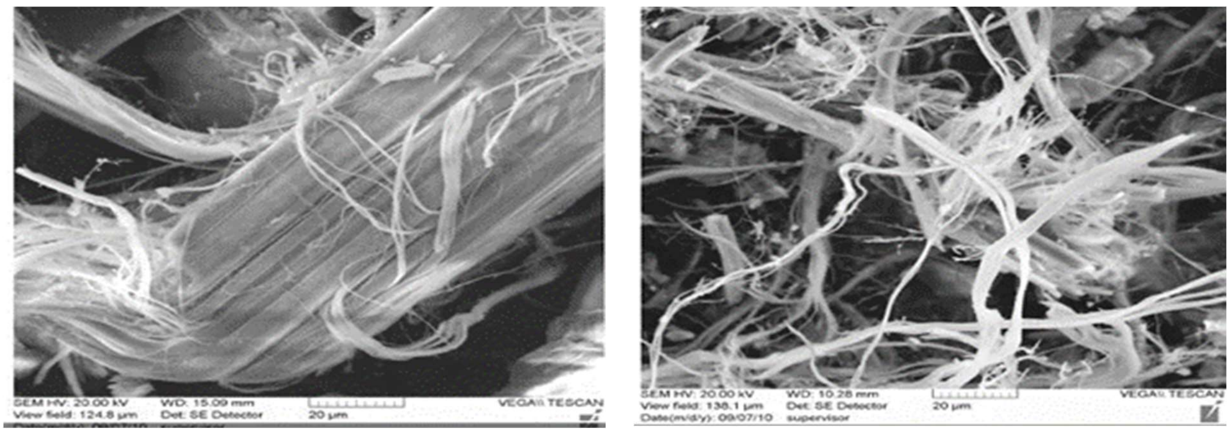
chrysotile is usually reduced to 40% or less, the content of constitutional water also fluctuates, sometimes increasing to 14.5-15%, then falling to 11.5-12%.

The chemical composition of chrysotile asbestos is presented in Table-2, a – photo of chrysotile-asbestos and b – its microstructure in Fig.-1.

Table-2: The Chemical Composition of Chrysotile Asbestos

Content of oxides, mass percent

SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	K_2O	Na_2O	H_2O+105°	H_2O+105°
42.60	0.65	1.04	0.03	40.77	traces		13.46	0.95



a b

Fig.-1: Photo of Asbestos (a) and its Microstructure (b)

The natural wollastonite of the Verkhne-Badam deposit was also used as a micro-reinforcing additive in the production of sovelite. The wollastonite ore of the Verkhne-Badam deposit contains wollastonite – 49.4%, calcite – 20.6%, quartz – 20.2%, and granite – 5.1%. Table-3 shows the chemical composition of the natural wollastonite of the Verkhne-Badam deposit, and Fig.-2 shows a photo of the natural wollastonite rock and its microstructure. The needle-like form of wollastonite determines the main direction of its use as a micro-reinforcing filler. The synthetic wollastonite used in this work was obtained by thermal treatment of a mixture of CaO-, SiO_2-containing components in the presence of an alkaline reagent, while granular slags of electrothermal production of phosphorus were used as the CaO-containing component, quartz sand was used as the SiO_2-containing component, and sodium sulfate, with a ratio of components in the CaO/SiO_2 mixture equal to 0.75-0.9, and Na_2SO_4 / SiO_2 equal to 0.12-0.16, was used as the alkaline reagent.

Table-3: The Chemical Composition of the Wollastonite Ores Content of oxides, mass percent

SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	TiO_2	K_2O	Na_2O	MnO	Loss on ignition
50.4	0.3	14.87	31.51	0.2	0.15	0.3	0.87	0.03	0.02

a b

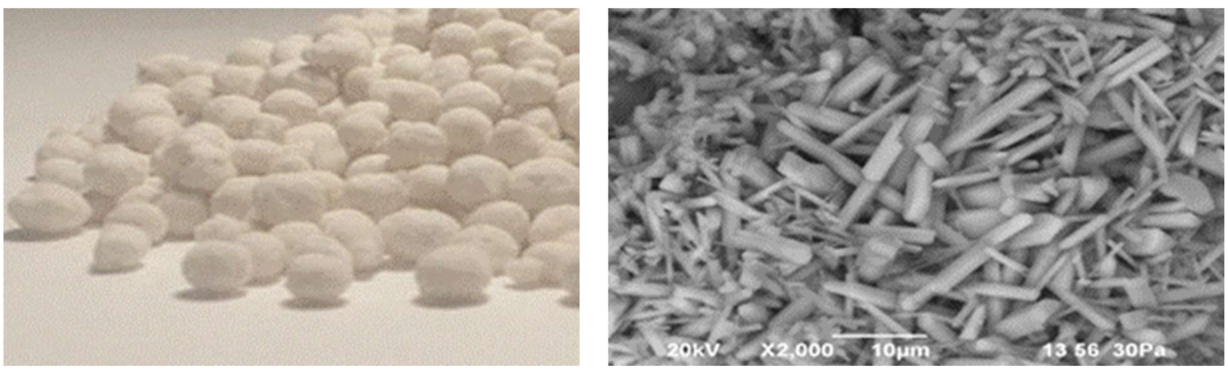
Fig.-2: Photo of the Natural Wollastonite Rock (a) and its Microstructure (b)

The mixture was prepared by joint dry grinding of the components and the reagent, followed by granulation of the mixture by obtaining rounded particles while moistening the charge. Thermal treatment of raw granules (pellets) from the charge was carried out at 1000-1050°C for 40-60 minutes.[37-57] Table-4 shows the chemical composition of the synthetic wollastonite.

Tab.-4: The Chemical Composition of the Synthetic Wollastonite

Content of oxides, mass percent										
SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	P_2O_2	R_2O	F	SO_3	Loss on ignition	Σ
46.7	1.26	0.35	43.55	3.36	1.35	1.09	1.52	0.87	-	100

Figure-3 shows photos of granules and microstructure of the obtained synthetic wollastonite. The simulation of the process of obtaining sovelite in laboratory conditions was carried out as follows. The waste sample was subjected to thermal treatment in a muffle furnace in the temperature range of 500-900°C every 50°C in order to establish the optimal decarbonization temperature of magnesium and calcium carbonates. It was found that with an increase in the firing temperature, the specific surface area of the wastes increases. The dynamics of the decarbonization process were determined by the value of losses on ignition. It was found that the decarbonization process of the dolomite fraction contained in the tails of polymetallic ores completely ends at a temperature of 800°C.



a b

Fig.-3: Photos of the Synthetic Wollastonite Pellets (a) and its Microstructure (b)

Then the calcined samples were quenched with water to determine the completeness of the transition of magnesium and calcium oxides to the hydrated state by the reaction:

$$CaO+H_2O \rightarrow Ca(OH)_2; \quad (1)$$

$$MgO + H_2O \rightarrow Mg(OH)_2. \quad (2)$$

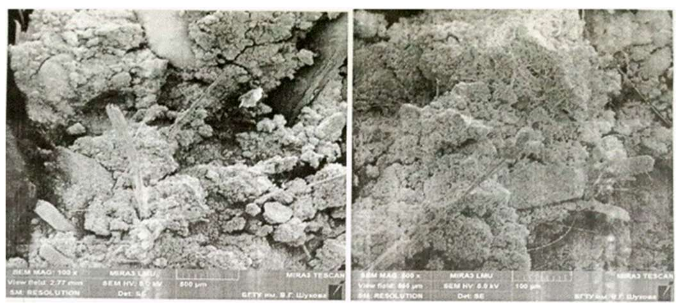
During the research, the masses of the test samples were taken in the same amount, equal to 20 g. Then the calcined sample was placed in a glass and poured with water in an amount of 500 ml, kept with stirring for a day, filtered and the residue on the filter was washed in 500 ml of water. The determination of magnesium and calcium in the solution was carried out complexometrically. The results obtained showed that with an increase in the calcination temperature, the output of active calcium into the solution increases proportionally. As for magnesium, in the range of calcination temperatures from 500 to 600°C its transition to an aqueous solution slightly increases, from 600°C to 800°C it slows down, and above 800°C it increases again. Subsequently, the obtained dolomite milk was subjected to carbonization by passing through a solution of carbon dioxide:

$$Ca(OH)_2+ 2Mg(OH)_2+3CO_2+ nH_2O \rightarrow CaCO_3 + 2[MgCO_3 \cdot H_2O]+ (n+1) H_2O \quad (3)$$

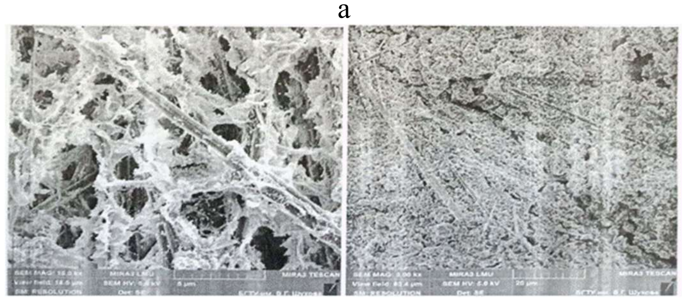
As a result, a mixture of finely dispersed precipitate $CaCO_3$ and needle-like crystals $2MgCO_3 \cdot 3H_2O$ was formed. The reaction continued until all of the magnesium oxide hydrates were completely converted to magnesium carbonate trihydrate. Then the obtained magnesium carbonate trihydrate was subjected to heating with direct steam at a temperature of 95°C for recrystallization into tetra hydrate basic magnesium carbonate according to the reaction

$$2[MgCO_3 \cdot 3H_2O] \rightarrow MgCO_3 \cdot Mg(OH)_2 \cdot 4H_2O + H_2O \qquad (4)$$

Thus, the sovelite base was obtained, which consisted of tetrahydrate basic magnesium carbonate and freshly precipitated calcium carbonate. A reinforcing component (ground natural and synthetic wollastonite) was introduced into the resulting base in an amount of 20% of the total mass at a ratio of ground wollastonite: fluffed asbestos 1:0; 1:0.5, and 1:1 and stirred thoroughly for uniform distribution throughout the volume. The resulting mass was filtered to form a paste with a moisture content of up to 50-60% and shaped beams of 4x4x16 cm. The results of the experimental work are shown in Table-5. The characteristics given in the table show that the use of synthetic wollastonite obtained according to the technologies developed by the authors allows for improved such properties as thermal conductivity and flexural strength.[58-64] It should be noted that the use of ground synthetic wollastonite as a reinforcing component gives the formed mass increased plasticity and improves the formability of the wet mixture. The research results showed that the thermal and physic-mechanical characteristics of the samples obtained meet the requirements of GOST 6788-74. Figure- 4 shows electron micrographs obtained from chips of the samples of the thermal insulating material – sovelite.



a



b

a – obtained by traditional technology, b – with the addition of synthetic wollastonite

Fig.-4: Electron Micrographs of the Thermal Insulating Materia –Sovelite, Obtained from the Chips of the Samples

Table-5: Technical Characteristics of Sovelite Products

Indicators	Standards for brand products		Indicators of the obtained products			
			natural		synthetic	
			wollastonite: asbestos			
			1:0	1:0	1:0,5	1:1
Bulk weight, kg/m^3	350	400	390	395	393	392
Thermal conductivity W/(m$^{\circ}$C), no more at medium temperature, $^{\circ}$C						
25±5	0.079	0.082	0.082	0.083	0.083	0.082
125±5	0.091	0.094	0.094	0.095	0.095	0.094
Flexural strength, MPa	0.20	0.22	0.22	0.27	0.25	0.24

Figure-5 shows the spectrogram of the non-carbonate part of the “tails”.

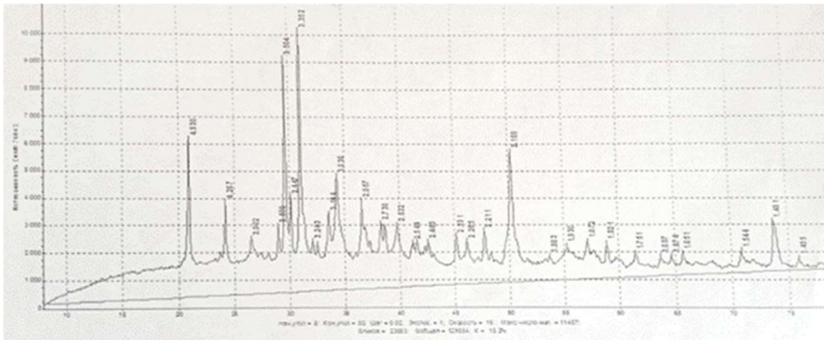


Fig.-5: The Spectrogram of the Non-Carbonate Part of the "Tails"

The spectrogram of the non-carbonate part of the tails identified the following: PbS cd/n =3.447; 2.331; 1.872; 1.544 Å; FeS_2 cd/n= 2.109; 1.491 Å; $BaSO_4$ cd/n = 3.580; 3.036; 2.730; 2.109; 1.751 Å; $PbSO_4$ cd/n=4.267; 2.730; 2.403; 1.872; 1.651 Å. The spectral analysis of the filter residue showed the following components in %: aluminum 2.7; iron 3; zinc 3; lead > 3; barium >3. The energy and resource-saving technology developed by us for the production of thermally insulating material – sovelite – on the basis of the tails of polymetallic ores and the synthetic wollastonite significantly improves the quality characteristics and increases the service life of products, and also gives a significant economic effect: cost reduction while improving quality (the cost of synthetic wollastonite is lower than the cost of asbestos); reduction of energy consumption; replacement of expensive fillers, etc.

CONCLUSION

The research results allowed us to establish that the dolomite-barium tails of polymetallic ores can serve as a technogenic raw material base to obtain the thermal insulating material – sovelite – from the dolomite component of the wastes, as well as to extract metals from its enriched part, i.e. to practically completely process the wastes into useful products. The non-carbonate part of the dolomite-barium tails, formed during the production of sovelite, can serve as a raw material base for the extraction of non-ferrous ones. It was found that the thermal insulating material – sovelite, obtained by resource-and energy-saving technology, meets modern requirements – increased construction and technical properties and reduced the cost of the final product.

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CONFLICT OF INTERESTS

The authors declare that there is no conflict of interest.

AUTHOR CONTRIBUTIONS

All the authors contributed significantly to this manuscript, participated in reviewing/editing, and approved the final draft for publication. The research profile of the authors can be verified from their ORCID ids, given below:

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