The influence of foundry wastes on the quality of autoclaved sand-lime materials

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Abstract

The paper presents the results of research on the possibility of reusing selected types of foundry waste, i.e. used molding and core sands and dust obtained in the process of their regeneration, as alternative or supplementary materials in relation to the quartz sand originally used in manufacturing of sand-lime bricks. In the production technology of sand-lime products, quartz sand is used as aggregate. A rational factor in favor of such a technological solution is the high content of crystalline silica in the waste molding and core sands, in which the mineral matrix is composed of good quality quartz sands.

The research concept covers production of a series of samples of autoclaved silicate materials formed by pressing, from raw material mixtures involving the discussed waste materials. In the laboratory tests conducted for the preparation of autoclaved materials of the sand-lime brick type, apart from traditional raw materials in the form of natural quartz sand and burnt lime, different molding waste and/or core sand materials and post-regenerative dusts were used for composing raw material mixtures. Foundry industry wastes were introduced into the basic raw material based on the gradually increasing substitution of quartz sand, in the amount within the range of 0 - 100% (wt. %).

The assessment of the possibility of using the discussed waste materials in the indicated direction was based on the result of a comparative analysis, covering the main functional characteristics of two types of materials, i.e. reference material, obtained from raw material mixture, containing no waste and series of experimental materials, produced with various quantitative and qualitative contributions of waste materials. The characteristics of the obtained autoclaved materials are also supplemented by the results of the leaching tests, for heavy metal elements and the analysis of selected aspects of the microstructure carried out by the SEM and EDAX method.

The results of the research show that it is possible to use casting waste compounds after their processing, in the production of autoclaved sand-lime products. This processing of wastes is two-stage operation. The purpose of the first stage is to restore the primary graining of quartz sand used to obtain foundry moulds and cores, while the second stage is to remove the organic binder residues that appear on the quartz sand grains in the form of thin layers hindering the reaction with burnt lime.

Keywords: calcium silicate brick, moulding and core waste, organic binder residue, heavy metal leaching

Kulcsszavak: kalcium-szilikát téglá, öntési és maghulladék, szerves kötőanyag maradványok, nehézfémek kioldódása

1. Introduction

Foundry and core molds are in most cases single-use casting molds. In practice, this means that after the process of casting metals or their respective alloys has been completed and the castings are removed from the molds, they are treated as a waste. Thus, they are intended for storage on various types of landfills, with virtually no possibility of their reuse in the foundry process. Considering the fact that the used foundry molds contain a large amount of silica in the form of β -quartz, there are rational premises for their secondary use in non-foundry areas. A good example here can be the building materials industry, and more precisely those manufacturing technologies that require the use of large quantities of quartz sand [1-6]. The assessment of the possibility of reusing quartz sand obtained from the recycling of used foundry molds was analyzed on the example of the autoclaved building materials

technology based on natural sand and burnt lime as basic raw materials. In the conducted tests, it was assumed that the silica material obtained as a result of processing of used casting molds [7] will act as aggregate in raw material mixtures intended for obtaining autoclaved materials. Therefore, it will be a partial or complete substitute (replacement) of natural quartz sand, commonly used as aggregate for the production of pressed silicate products. The usefulness of these quartz sands, derived from the recycling of used foundry and core molds, for obtaining this type of products was determined based on the results of comparative analysis. The scope of this analysis included the basic functional characteristics of autoclaved materials obtained with this type of sands (materials with experimental compositions) in relation to the properties of reference materials. On the other hand, in order to confirm or eliminate the potentially negative impact on the natural environment regarding the release of heavy metals from

silicate products obtained with the used of cast foundry sands, which process can take place throughout their entire lifetime, leachability tests were carried out [8-14]. The results obtained during the research are presented in this paper.

2. Materials

During the implementation of this research work, traditional and alternative raw materials were used to obtain silicate product. Industrial raw materials were used as the basic raw materials, while the alternative to quartz sand of natural origin raw material was silica material recycled from the used foundry masses showing different properties, determined mainly by the type of binder used to prepare them. The group of basic raw materials therefore comprised:

- quartz sand of natural origin (symbol QS-LU),
- highly reactive ground burnt lime, non-slaked lime obtained in industrial conditions (symbol LBU-HR),
- distilled water (symbol DW).

On the other hand, the group of alternative silica raw materials used as a partial or complete substitute for natural quartz sand were quartz sands obtained from the recycling of used foundry masses. These masses represented various types of waste foundry or core molds occurring alone or in the form of appropriate compositions. Used foundry sands included in the research program were of the following types:

- casting mass with an organic binder in the form of furan resin (symbol WSK-FU and MP-FU),
- casting mass from Cold Box technology (symbol WSK-CB),
- a mixture of different types of used foundry masses (symbol WSK-M),
- water-glass casting mass (symbol MO-WG),
- alkaline-phenol casting mass (symbol MO-AF),
- regenerative dusts obtained as a result from regeneration of casting mass with furan resin (symbol MP-P).

At the same time, it is worth noting that the abovementioned used foundry sands were derived from the same type of quartz sand from the "Grudzeń Las" mine (symbol QS-GL) used to obtain foundry and core masses.

The raw material mixtures intended for the preparation of individual series of samples were characterized by a constant quantitative composition, while a variable qualitative composition. The constant quantitative composition of these mixtures was ensured by the same shares of aggregate, binder and water in them, which were 83.6%, 8.0% and 8.4% (wt. %) respectively. The given shares of burnt lime and water resulted from the adopted design assumptions regarding the constant of activity "A" and humidity "w" of the raw material mixtures. The variability of the qualitative composition of the raw material mixtures, intended for the preparation of experimental materials, was obtained as a result of gradual (25, 50 and 100 wt. %) substitution of quartz sand QS-LU by the sands obtained from recycling, diversified in terms of properties and source of used foundry masses. In addition,

it was assumed that the degree of processing of used casting sands has a significant impact on the quality of the silicate materials obtained. In connection with the above, in the first stage of research, waste casting sands were subjected to an initial processing process, aimed only at restoring the original granulation of the quartz matrix. On the other hand, in the second stage of the study, casting sands were subjected to additional mechanical treatment consisting of a short-term "dry" milling process, carried out to remove the residue of organic binders from the quartz matrix grains surface. It is worth noting that in the second stage of testing, silicate samples were obtained from raw material mixtures, characterized by a constant share of foundry waste, constituting 25% of the mass of natural quartz sand.

Individual series of samples of silicate materials prepared from raw material mixtures of specified compositions were obtained each time in a fixed and repeatable manner. Their activity and humidity were monitored all the time. Required amounts of individual components, resulting from the adopted quantitative composition, were subjected to a two-stage homogenization process; at the beginning without the participation of water, and then with its participation. Distilled water was added in the amount necessary for the total hydration of quicklime and obtaining 6% of the mixture moisture. In order to carry out the lime slaking process, the raw material mixture was placed in a sealed glass vessel and kept for about 1 hour in a laboratory dryer at a temperature of 65 °C. After this period of time, the raw material mixture was cooled to ambient temperature and subjected to final homogenization. From the mass prepared in this way, cylindrical samples with dimensions of diameter and height equal to 25 mm were formed using a fold-out metal mold. Sample formation was carried out using a hydraulic press method of two-sided and two-stage pressing with interstage venting, at pressures of 10 and 20 MPa, respectively. 12 samples were formed in the same way for each mass composition. Then the samples were placed in Teflon crucibles, which in turn were inserted into the chambers of steel pressure cylinders acting as laboratory autoclaves. Stainless-steel cylinders, containing samples and distilled water located in its lower part, were inserted into the heater and subjected to the heating process according to a specific regime. In this way the process of hydrothermal treatment of samples under laboratory conditions was carried out. The conditions used in the sample treatment, reflected the conditions for processing of sand-lime products in industrial autoclaves and were as follows:

- saturated steam pressure - 1.002 MPa
- steam temperature - 180 °C
- total autoclaving time - 9.5 hours

3. Methods

The samples obtained in hydrothermal conditions were tested for the determination of their basic performance characteristics. The tests of physical properties were carried out in accordance with the scope and procedures given in the product standard PN-EN 771-2: 2004 [15] and the standards referred to selected

properties, in particular concerning: compressive strength - PN-EN 772- 1: 2001 [16] and dry density - PN-EN 772-13: 2001 [17]. According to the reference documents provided, the scope of testing of these materials included the following performance parameters:

- compressive strength f_b ,
- dry density (in parallel with two methods): $\rho_{n,u}^0$ and $\rho_{n,u}^1$,
- c_w water absorption.

In addition, open porosity P_0 was determined after the obtained materials by hydrostatic weighing [18].

In case of each prepared mixtures intended for obtaining a given series of samples, its molding moisture “w” and activity “A” were monitored [19, 20]. The moisture content was determined by the drying method as the average of two samples. On the other hand, the activity of raw material mixtures was determined by a chemical method consisting of titration a specified amount of mass taken in a wet state with 1M HCl in the presence of a 1% alcohol solution of phenolphthalein. The amount of acid needed to neutralize calcium hydroxide, i.e. the disappearance of the pink color of the suspension, was the basis for calculating the percentage of CaO in the dry form and thus its activity (Table 1).

Determination of the leaching level of selected heavy metals in case of silicate samples containing different categories of waste molding and/or core sands was carried out in accordance with the procedure given in the standard PN-EN 12457-2 [21]. The subject of the analysis were eluates prepared in accordance with the guidelines given in this standard, while the determination of the content of selected heavy metals and the interpretation of the obtained test results was made according to the criteria given in the PN-EN ISO 11885 standard [22]. Determination of the content of heavy metal elements in the prepared eluates was performed by the method of atomic emission induction spectrometry (ICP-AES), using the ICP emission spectrometer of the Perkin-Elmer company (model Plasma 400). The measurements included determination of the content of the following elements: chromium, copper, cobalt, molybdenum, selenium, vanadium, zinc, tellurium, lead, cadmium, nickel, tin, antimony, arsenic, boron and barium.

The microstructure examination of the sand-lime materials containing the used casting sands was carried out using the NOVA_{NANO} SEM 200 scanning electron microscope equipped with EDAX microanalyzer, FEI COMPANY.

4. Results

The results concerning the examination of the technological properties of the masses regarding their humidity and activity, as well as the basic functional properties of the sand-lime materials obtained with the use of used casting sands are presented in the Table 1.

The results of the microstructure elements examination of selected silicate materials obtained are presented in Fig. 1.

The results of the heavy metal leaching tests are presented in Table 2.

a) PK-LU sample (reference sample of the I stage of the research)

b) Sample WSK-FU₂₅ (experimental sample of the I stage of the research)

c) mPK-LU₂₅ sample (reference sample of the II stage of the research)

d) mWSK-FU₂₅ sample (experimental sample of the II stage of the research)

Fig. 1 Microstructure of autoclaved sand-lime samples.

1. ábra Autoklávozott homok-mész minták mikrostruktúrája

Mass symbol // Sample symbol	Technological properties of masses		Functional properties of silicate materials samples				
	w, [%]	A, [%]	$\rho_{n,u}^0$ [g/cm ³]	$\rho_{n,u}^1$ [g/cm ³]	f _b , [MPa]	c _w , [%]	P _o , [%]
I stage of research							
PK-LU	6.2	7.6	1.81	1.85	25.6	12.7	23.4
WSK-FU ₂₅	4.4	7.5	1.76	1.81	23.5	14.0	25.4
WSK-FU ₅₀	6.4	6.9	1.74	1.78	15.7	15.1	26.8
WSK-FU ₁₀₀	4.6	7.6	1.70	1.76	10.3	14.5	25.4
WSK-CB ₂₅	4.2	7.6	1.77	1.81	23.0	14.6	26.4
WSK-CB ₅₀	4.6	7.5	1.75	1.79	16.7	15.5	27.8
WSK-CB ₁₀₀	4.6	7.5	1.69	1.75	8.3	16.3	28.5
WSK-M ₂₅	5.7	7.7	1.86	1.85	19.2	13.5	25.0
WSK-M ₅₀	5.7	7.6	1.80	1.84	19.6	14.3	26.2
WSK-M ₁₀₀	5.6	7.3	1.75	1.75	20.2	17.0	29.7
MO-WG ₂₅	5.5	7.7	1.79	1.82	15.4	13.5	24.7
MO-WG ₅₀	5.3	8.0	1.76	1.81	11.8	13.4	24.3
MO-WG ₁₀₀	4.8	7.3	1.70	1.78	8.0	13.8	24.5
MO-AF ₂₅	4.1	7.5	1.79	1.83	22.5	14.0	25.8
MO-AF ₅₀	5.1	7.4	1.77	1.81	15.5	15.4	28.0
MO-AF ₁₀₀	6.5	7.4	1.76	1.81	10.5	15.9	28.8
MP-FU ₂₅	4.7	7.5	1.78	1.82	9.9	15.4	27.9
MP-FU ₅₀	5.1	7.4	1.76	1.79	4.1	16.9	30.2
MP-FU ₁₀₀	6.4	7.4	1.71	1.70	1.1	19.8	33.7
MP-P ₂₅	6.1	7.4	1.76	1.79	13.7	15.3	27.3
MP-P ₅₀	6.8	7.0	1.65	1.65	7.2	19.5	32.2
MP-P ₁₀₀	6.2	6.6	1.38	1.35	4.9	31.1	42.0
II stage of research							
mPK-LU ₂₅	6.2	7.5	1.82	1.85	30.7	12.4	22.9
mWSK-FU ₂₅	5.9	7.5	1.81	1.84	30.0	12.7	23.4
mWSK-CB ₂₅	5.7	6.5	1.79	1.82	28.0	13.5	24.7
mWSK-M ₂₅	6.2	7.0	1.84	1.85	16.6	13.7	25.4
mMO-WG ₂₅	6.4	7.4	1.80	1.84	19.3	12.4	22.8
mMO-AF ₂₅	7.3	7.4	1.82	1.85	27.7	13.1	24.3
mMP-FU ₂₅	6.3	7.4	1.82	1.85	11.4	14.1	26.2

Table 1 Summary of the results obtained for raw material mixtures and autoclaved materials

1. táblázat A nyersanyag-keverékekkel és autoklávozott anyagokkal kapott eredmények összefoglalása

Analyzed component	Sample designation								Admissible critical leaching levels liquid/solid phase = 10 L/kg for:			Reference data according to [14, 22, 25]
	PK-LU	MO-SW ₅₀	MO-AF ₅₀	MP-FU ₅₀	WSK-CB ₅₀	WSK-FU ₅₀	WSK-M ₅₀	MP-P ₅₀	Neutral wastes	Other than neutral and hazardous wastes	Hazardous wastes	
Leaching of the given component mg/kg of dry mass												mg/L
Chromium _(total)	0.04	0.17	0.08	0.16	0.10	0.09	0.19	0.37	0.5	10	70	0.027
Copper	0.05	0.03	0.02	<0.01	0.09	0.23	<0.01	0.02	2	50	100	0.148
Cobalt	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	- (*)	- (*)	- (*)	0.003
Molybdenum	0.24	0.11	0.11	<0.01	0.11	0.09	0.23	0.11	0.5	10	30	- (**)
Selenium	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	0.1	0.5	7	- (**)
Vanadium	0.21	<0.01	0.14	<0.01	0.22	0.08	0.21	0.09	1 (*)	- (*)	- (*)	- (**)
Zinc	0.10	0.31	0.05	0.38	0.16	0.07	0.09	0.05	4	50	200	0.380
Tellurium	<0.03	<0.03	<0.03	<0.03	<0.03	<0.03	<0.03	<0.03	- (*)	- (*)	- (*)	- (**)
Lead	<0.01	0.21	<0.01	<0.01	<0.01	0.24	0.11	0.35	0.5	10	50	0.063
Cadmium	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	0.04	1	5	0.003
Nickel	0.05	0.05	0.06	0.07	0.03	0.10	0.06	0.04	0.4	10	40	0.048
Tin	<0.06	<0.06	<0.06	<0.06	<0.06	<0.06	<0.06	<0.06	- (*)	- (*)	- (*)	- (**)
Antimony	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	0.06	0.7	5	- (**)
Arsenic	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	0.5	2	25	0.047
Boron	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	- (*)	- (*)	- (*)	- (**)
Barium	2.26	3.96	1.94	3.78	1.70	2.02	1.97	2.03	20	100	300	0.707

(*) - no reference data (**) - no data available

Table 2 Leaching of heavy metals from samples containing used molding and core sand

2. táblázat Nehézfémetek kimosódása öntvényt és maghomokot tartalmazó mintákból

5. Discussion

Analyzing the obtained test results presented in *Table 1* it should be stated that the numerical values of the tested parameters of the obtained materials are clearly influenced by both the share of used foundry masses in the raw material mixture, expressed by the level of substitution of natural quartz sand, as well as their type and degree of processing. Regardless of the type of foundry waste mass, along with the increase in their share in the raw material mixture, we observe a gradual deterioration of the tested parameters characterizing a given material. However, in relation to the strength parameters, the method of processing of used casting sands has a very beneficial effect. In particular, this applies to the processing method associated with the removal of binder residue from quartz matrix surfaces, especially organic origin, present on them in the form of thin layers. Removing this layer of binder mechanically leads to the activation of the quartz grain surface. As a result, a chemical reaction with the binder is easier, during which the desired mineral phases are formed in quantities that improve the mechanical properties of the obtained materials.

The results of analysis of elements belonging to the heavy metals group in case of silicate material samples obtained in individual series are presented in the *Table 2*. The results were obtained as an average of three determinations. *Table 2*, apart from the results of measurements, also contains the leaching limit values for individual elements given in RMGiP [23] and recommended values [14, 24, 25] for selected types of building materials. These values relate to the criteria and procedures for the admission to deposit in outdoor landfills, three categories of waste, i.e. inert, other than hazardous or neutral and hazardous. The obtained results prove that the level of leachability of the tested elements only slightly, in individual cases, exceeds the leaching limit values for the inert waste category, while these values are definitely lower than the permissible limit values applicable for the other two categories of waste, i.e. other than inert and hazardous and dangerous.

Only selected samples from all prepared series were subjected to microstructure tests. The basic criterion for selecting samples were their final properties. In accordance with the above, the samples with the most favorable parameters, which are equivalents due to the type of casting mass used and its share in the raw material mixture, materials obtained in the first and second stage of research, were assigned for microstructure tests. Samples of reference materials were also analysed for comparative purposes. The most characteristic images of the microstructures of the analyzed samples are shown in *Fig. 1a)* and *1b)* show the images of the reference and experimental materials respectively. Presented figures show microstructure of the samples obtained as part of the first stage of research, with a magnification enabling observation of its individual elements. Therefore, these images show the aggregate grain size and pore distribution. In turn, *Figs 1c)* and *1d)* show images of microstructures of reference and experimental materials respectively, but originating from the second stage of research and covering areas of the surface of sand grains and the products of synthesis formed on this surface under

hydrothermal conditions. The basic observation regarding the morphology of these products is that the morphology usually consist of structural elements in the form of needles or fibers, which is a positive phenomenon, but to a much lesser extent in the form of platelets or elongated ribbons.

6. Conclusions

Based on all the preliminary results obtained in this paper, the following conclusions can be made:

1. Used casting sands obtained on the basis of quartz matrix are a potential source of silica raw material, which may be a partial substitute for natural quartz sand used in the production of sand-lime products.
2. The said silica material derived from the recycling of used foundry sands, is obtained as a result of appropriate processing of these sands, aimed primarily at crushing the agglomerates of grains (lumps) and restoring the original granulation, and then to remove the residue of the organic binder used for quartz warp grains preparing fresh molding and core sands.
3. The process of processing used foundry sands consisting in mechanical removal of hardened binder residues from the surface of grains has a very beneficial effect on functional properties, including mainly strength, of sand-lime materials obtained with their participation.
4. Durability, in terms of resistance to low temperatures of sand-lime products containing properly processed spent casting masses, does not deteriorate significantly compared to this type of materials obtained on the basis of traditional raw materials.
5. The presence of used casting sands in raw material mixtures intended for the preparation of sand-lime materials, regardless of their degree of processing, adversely affects the color of these products.
6. There are no visible differences in the microstructure of autoclaved materials obtained with the use of foundry sands, in relation to this type of materials obtained from traditional raw materials.
7. The level of heavy metal leaching from the obtained silicate materials is lower than the recommended values.

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SCIENTIFIC SOCIETY OF THE SILICATE INDUSTRY

The mission of the Scientific Society of the Silicate Industry is to promote the technical, scientific and economical progress of the silicate industry, to support the professional development and public activity of the technical and economic experts of the industry.

- > We represent the silicate industry in activities improving legal, technical and economic systems
- > We establish professional connections with organizations, universities and companies abroad
- > We help the young generation's professional education and their participation in public professional activities
- > We ensure the continuous development of experts from the silicate industry by organizing professional courses
- > We promote the research and technological development in the silicate industry
- > We organize scientific conferences to help the communication within the industry

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