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The Influence of Modified Inorganic Binders Intended for 3D Printing on Selected Properties of Thermally Cured Moulding Sands – Conventionally and with Microwaves

Dawid M. Halejcio[a], Katarzyna Major-Gabryś[a*]

[a] AGH University of Krakow, Faculty of Foundry Engineering, 30 Mickiewicza Av., 30-059 Krakow, Poland
[*] *email: katmg@agh.edu.pl*

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Abstract

This study determined the impact of thermal curing on the basic properties of moulding compounds made with commercial inorganic binders and binders based on them, modified for use in 3D printing technology (Binder Jetting). Two inorganic binders based on sodium silicate and a binder based on aluminosilicates were tested. As part of the work, the parameters for thermal curing of the mixtures were selected: for curing in a dryer, the best properties were obtained for mixtures containing 2.0 p.p.w. of binder cured for 10 min at 160°C. In the case of microwave curing, the best properties were obtained for moulding sands containing 2.0 p.p.w. of binder cured for 6 min at a device power of 800 W. The tests showed that the basic properties of moulding compounds with binders developed on the basis of commercial binders for use in 3D printing technology, thermally cured in a dryer, do not differ significantly from the properties of compounds with commercial binders. In the case of microwave curing, a reduction in the strength of compounds with new binders was observed in relation to compounds with classic binders. Thermal deformation tests of compounds with classic and modified binders confirmed the typical behavior observed for inorganic systems. It was proven that new, modified inorganic binders developed for 3D printing of moulds and cores using Binder Jetting technology can be used as binding materials in thermally cured moulding sands. Both thermal curing methods were assessed as suitable for curing moulding compounds with new binders.

Keywords:

moulding sands, sand 3D printing, inorganic binders, thermal curing, microwaves

1. INTRODUCTION

Strict environmental protection requirements have led to intensive work over the last few decades to develop environmentally friendly moulding sands with inorganic binding systems as an alternative to organic-bonded sands. These changes are currently being implemented in foundry practice, but the use of organic binders is still predominant in the production of castings in disposable moulds made of II generation moulding sands (bonded with binders) and in moulds using cores made of II generation compounds. This is the reason justifying further development of inorganic systems and the possibilities of adapting them for modern technologies aligned with Foundry 4.0 assumptions, such as additive manufacturing.

In foundry moulding sands, three main types of inorganic binders are used: sodium silicates, phosphate-based binders, and geopolymer binders. Sodium silicate binders are commonly used in foundry practice due to their good water solubility, compliance with environmental standards, and ability to achieve adequate mechanical strength through the curing process [1–3]. Geopolymer binders, known as inorganic polymers, are aluminosilicate materials. Their potential for use in foundry practice stems mainly from their natural origin and their ability to undergo controlled polymerization of aluminosilicate monomers [4, 5]. Compounds with phosphate binders are not currently used in foundry practice and are not the subject of research conducted as part of this work.

Curing (dehydration) of compounds with inorganic binders can be carried out at ambient temperature by using curing agents (chemical curing) – self-curing moulding sands (ester curing agents), and CO_2 degassing of moulds and cores, as well as at elevated temperatures (physical curing) – conventional thermal curing and microwave curing. The choice of moulding sand's hardening technology directly affects the properties of the moulds and cores made from them and may indirectly affect the quality of the castings obtained. Unlike chemical curing, thermal curing allows the binder to be cured by controlled temperature increase, which results in water evaporation and the formation of strong bonds that bind the sand matrix grains. The strength of thermally cured hydrated sodium silicate mixture is approximately 10 times higher than that of chemically cured mixtures with a similar composition. Table 1 compares the effect of the curing method of hydrated sodium silicate sands on their strengths [1, 2].

Table 1
Comparison of compressive strength obtained for hydrated sodium silicate sands prepared using different curing technologies [1, 2]

Moulding sands' components, amount, p.p.w.	Curing technology	Compressive strength, MPa
Quartz sand, 100 Hydrated sodium silicate (M = 2.2), 4.0	thermal (physical)	10.0–11.0
	CO_2 (chemical)	1.0–1.2
	ester/4 h (chemical)	0.8–1.3

The differences in moulding sands' strength values are caused by different mechanisms of water removal from the binding systems, as shown in Figure 1 [3, 4].

Chemical hardening, which occurs in the case of ester technology and CO_2 gassing technology, causes slow dehydration of the moulding sand – the time required to completely remove water from the binder can be as long as 24 hours (Fig. 1a). Convection drying (Fig. 1b), which depends on the thermal conductivity of the moulding sand, is a process in which water is gradually removed from successive layers of the sand during heating. Due to energy losses during heating, mainly related to the efficiency of drying equipment and the method of heat transfer, convection drying is a time-consuming and energy-intensive process. In the traditional thermal hardening method, a temperature range of 140°C to 180°C is used [6]. In the case of hydrated sodium silicate-based binders, the curing process consists of several distinct phases: elimination of free water, removal of bound water, and formation of silicate bridges as the result of dehydration reactions. The energy transfer mechanism is mainly based on conduction, which means that heating starts at the surface and gradually progresses into the material [7]. The use of heat accelerates the evaporation of water from the binder solution, which leads to an increase in viscosity and promotes the formation of stable bridges between sand grains [1, 8]. Microwave curing, which provides a different means of energy transfer than traditional heating methods (Fig. 1c), may be more efficient in the curing process of moulding sands with hydrated sodium silicate. Electromagnetic radiation at a frequency of 2.45 GHz rapidly heats the moulding sand by rotating water molecules, resulting in faster and more uniform temperature distribution throughout the material. Through high-frequency vibrations, the energy of the electromagnetic wave is converted into the thermal energy of the binder, intensifying the process of forming a dehydrated layer of glassy sodium silicate. This mechanism significantly reduces the curing time and can also improve the quality of the bonds between sand grains. The microwave curing process combines the advantages of traditional curing methods, such as process speed and reduction in binder quantity, while maintaining good mechanical and technological properties. The higher strength of the moulding sand allows for a reduction in wall thickness or a reduction in the volume of large cores. This can result in a reduction in the amount of sand used, and thus a reduction in production costs and an improvement in knockout efficiency. In addition, during hardening, the moulding sand is heated to lower temperatures (even twice as low) compared to conventional methods [5, 6]. The higher strength of the sand also allows for a reduction in the amount of binder in the sand, which has a positive effect on knockout and mechanical regeneration, i.e., the properties of silicate-bonded sands, which are well known to pose a technological challenge [1].

Geopolymer binders, similarly to sodium silicate-based binders, can be cured chemically and thermally. After adding the hardener, polymerization and gelation (cross-linking) occur, resulting in the formation of an inorganic polymer that binds the sand matrix grains [9]. During thermal curing, the binder also undergoes polymerization, leading to the formation of three-dimensional aluminosilicate networks (N-A-S-H gels, C-A-S-H gels) – the formation of an inorganic polymer with appropriate mechanical properties and stability [10, 11].

When comparing the presented physical curing methods, it should be noted that thermal curing requires heating the entire working chamber of the furnace, which is associated with significant energy losses to the environment and high unit energy consumption, especially in the production of large-size moulds [12]. In the case of microwave curing, during which energy is selectively supplied to the material, mainly to the water contained in the binder, it is possible to shorten the process time and reduce unit costs, despite losses associated with the conversion of electricity into microwaves [13, 14].

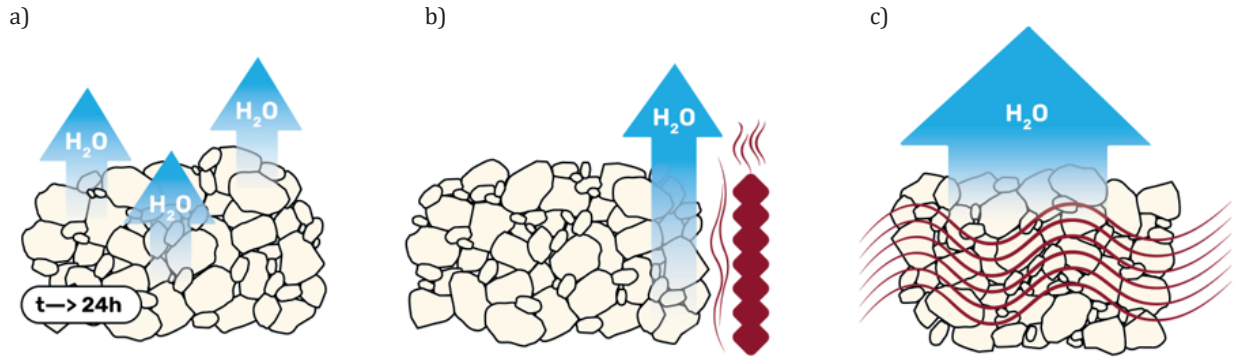

Fig. 1. Schematic diagrams of water removal during the curing of moulding sands with hydrated sodium silicate: a) using chemical methods; b) by heating in a convector; c) by microwave curing [3, 4]

Currently, in foundry practice, conventional or microwave thermal hardening of moulds and cores made using Binder Jetting technology is not used. However, these processes are used in the production of ceramic printed components ($BaTiO_3$, $CaSO_4 \cdot ½H_2O$), in which postprocess thermal treatment (heating, firing) is necessary to give the finished components the desired properties [15, 16]. Considering the benefits of the thermal curing of moulding compounds, it seems entirely justified to conduct research on the development of moulding compounds with thermally cured inorganic binders intended for 3D printing of sand moulds and cores. Furthermore, 3D printing of sand moulds using inorganic binders is an advanced and forward-looking technology that ensures high dimensional accuracy, repeatability, and process efficiency, making it an excellent solution for modern foundry engineering.

Binders used in Binder Jetting technology should ensure the highest possible print quality. One of the key parameters is the appropriate viscosity of the binder, which allows for its precise jetting using a print head. Another important aspect is the degree to which the substrate layers are saturated with the binder. Under ideal conditions, the binder should cover only a single layer of the substrate; however, in practice, the drop spreads across the material grains, and its excess penetrates deeper into the substrate. This is the reason for the varying properties observed across different cross-sections in the printed parts. The final important parameter is the interaction between the binder and the substrate. The binder used should be selected to suit the material so that it adequately wets the substrate, allowing a thin layer to form on the surface of the grains and subsequently enabling the formation of bonding bridges [17–20].

2. OWN RESEARCH

As part of this work, moulding compounds with thermally cured inorganic binders were tested. Three commercial binders (A, B, C) and their equivalents developed for 3D printing Binder Jetting technology were used [12, 13]. In the first stage of the research the curing process parameters were selected. Then, the influence of the binder type on selected properties of the compounds was determined. The aim of this article is to evaluate selected properties of molding compounds containing modified inorganic binders, designed for Binder Jetting technology, which are thermally cured using both conventional heating (drying) and microwave treatment. The study includes a comparison of test results obtained for molding sands using commercially available binders and their counterparts modified for 3D printing. As part of the study, an analysis of the effect of curing parameters on selected properties of the mixtures was also conducted.

2.1. Selection of process parameters

Moulding compounds intended for microwave curing were prepared using a paddle mixer, in which the matrix and binder were mixed for 60 s. Standard test specimens were compacted by vibration using a WADAP LUZ device (vibration time 9 s). The moulding sands were cured in an 800 W microwave oven at a frequency of 2.45 GHz. Both the amount of binder and then the curing time (for moulding sand A binder – 2.0 p.p.w.) were selected. The strength of the compounds was determined within a curing time range of 3–8 min. Figure 2 shows the research results. It has been demonstrated that compounds cured by microwaves for 3 min exhibit good strength properties from the perspective of foundry practice and extending the time of curing improves it. On the other hand, extending the curing time results in higher energy consumption. Therefore, it is necessary to select process parameters that meet production requirements. Based on the results obtained, the following parameters were selected as optimal for microwave hardened compounds: a curing time of 6 min and 2.0 p.p.w. of binder.

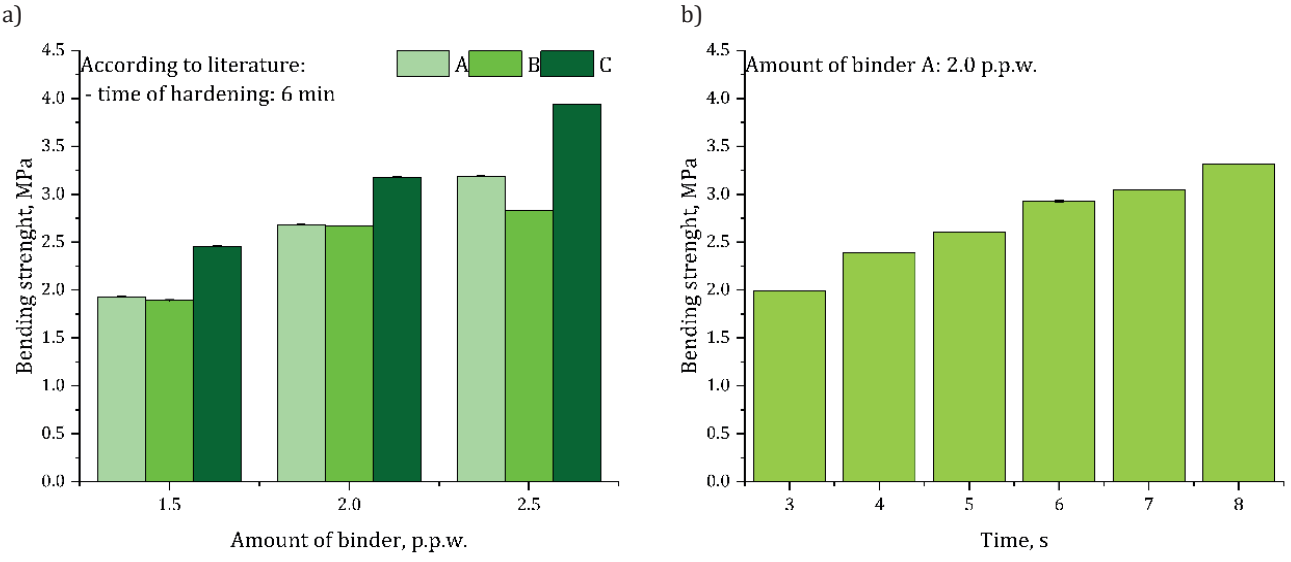

Fig. 2. Selection of parameters of the microwave curing process: a) binder amount; b) time of hardening

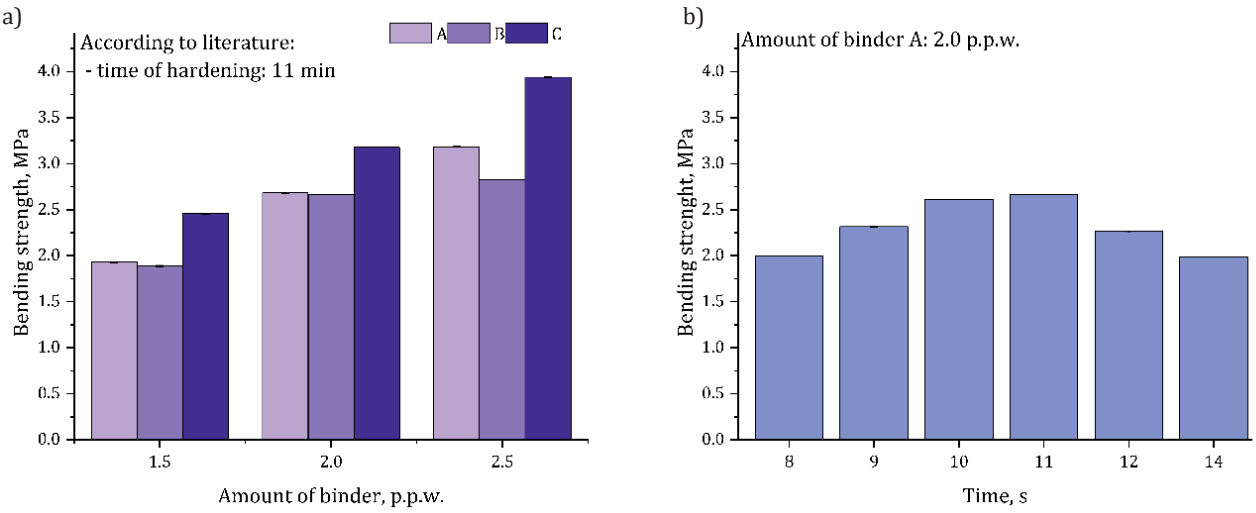

Fig. 3. Selection of parameters of the thermal (in a dryer) curing process: a) binder amount; b) time of hardening

Thermally cured compounds (in a dryer) were prepared in the same way as microwaved cured compounds – in a paddle mixer with a mixing time of 60 s. The moulds were also compacted in the same way as the moulds made of compounds intended for microwave curing – by vibration with a vibration time of 9 s. Curing was carried out in a laboratory dryer at a temperature of 160°C. The selection of the curing time and the amount of binder were also studied (Fig. 3). In order to select the curing time, a moulding compound with 2.0 p.p.w. of binder A was tested, based on its good strength properties. Although a mixture containing 2.5 p.p.w. of binder exhibits better properties, increasing the amount of inorganic binder in the moulding sand results in poorer knockout properties and a reduced capacity for mechanical reclamation. Based on the results presented, a mixture containing 2.0 p.p.w. of binder cured at 160°C for 10 min was selected for further tests of moulding sands thermally hardened in a dryer.

2.2. Materials and methodology

Table 1 presents the compositions of the tested mixtures along with the curing parameters selected on the basis of the tests presented in Section 2.1. The moulding compounds were prepared in a paddle mixer; the mixing time for sand and binder was 60 s. The test specimens (standard: longitudinal, cylindrical, and dog bone standard test specimens) were prepared using a WADAP LUZ vibratory compacting device with vibration time of 9 s. Sibelco quartz sand with a main fraction of 0.20/0.16/0.10 and a main fraction content of 92% was used as the matrix. The standard test specimens were thermally cured according to the method specified in Table 2. A laboratory dryer SA-P-2 (750 W, max temp. around 200°C) was used for conventional thermal curing, and a Whirlpool 800 W microwave oven was used for microwave curing. The binders used were hydrated sodium silicate-based binders (binders A and B) [21, 22] and aluminosilicate binder C [23]. Each of the tested binders underwent physicochemical modification on the basis of Kocel AJS300 3D printer requirements to adapt their parameters for use in 3D printing (A3D, B3D, C3D).

The tests were conducted "cold" – 1 hour after the end of thermal curing. The time was selected experimentally so that the standard test specimens reached ambient temperature throughout their entire volume. The bending and tensile strength tests of the moulding compounds were performed using a universal testing machine (MultiSerw Morek LRu-2e). Permeability was tested using a WADAP LPiR1 apparatus. Standard cylindrical standard test specimens (50 mm × 50 mm) were used to assess apparent density, permeability and friability, while standard-longitudinal standard test specimens (22.36 mm × 22.36 mm × 172 mm) were used to test bending strength. Tensile strength was determined using dog bone standard test specimens with a cross-section of 22.36 mm × 22.36 mm. The abrasion resistance of moulding compounds was tested using a method involving shot falling onto a cylindrical standard test specimen placed in the jaws of an apparatus manufactured by Huta Stalowa Wola. In accordance with the test methodology, steel shot (1.75 kg) with a diameter of approximately 1 mm falls freely from a height of 0.307 m onto a mould rotating at a speed of 1 rpm [24].

The purpose of testing the thermal deformation (by hot distortion parameter) of moulding compounds is to determine the thermal stability of casting cores. When liquid metal is poured into the mould the temperature rises and the metal level rises, leading to intense heating of the cores. The one-sided nature of heating in the initial phase of the process promotes the formation of thermal distortions in the cores, resulting from their expansion and contraction. These phenomena can lead to damage to the cores and the formation of casting defects, affecting its geometry, dimensional accuracy, and surface quality. The hot distortion test was performed using a DMA device manufactured by Multiserw Morek. In accordance with the methodology used, standard test specimens with dimensions of 114 mm × 25.4 mm × 6.3 mm were heated using two halogen lamps with a total power of 500 W to a maximum temperature of 900°C [25, 26].

Table 2
The compositions of the tested moulding sands with the processes curing parameters

Symbol	Matrix, amount, p.p.w.	Binder	Amount of binder, p.p.w.	Method of hardening	Time of hardening, min	Temperature or power
MA1	quartz sand, 100	A*	2.0	in a dryer	10	180°C
MA2		A3D	2.0	in a dryer	10	180°C
MA3		A	2.0	microwave	6	800 W
MA4		A3D	2.0	microwave	6	800 W
MB1		B**	2.0	in a dryer	10	180°C
MB2		B3D	2.0	in a dryer	10	180°C
MB3		B	2.0	microwave	6	800 W
MB4		B3D	2.0	Microwave	6	800 W
MC1		C***	2.0	in a dryer	10	180°C
MC2		C3D	2.0	in a dryer	10	180°C
MC3		C	2.0	microwave	6	800 W
MC4		C3D	2.0	microwave	6	800 W

* A – commercial inorganic binder;
** B – commercial inorganic binder with improved knockout properties;
*** C – commercial inorganic binder based on aluminosilicates;
A3D, B3D, C3D – modified binders elaborated within own research for use in 3D printing technology

2.3. Results and discussion

The results of the tests are presented in Figures 4–10.

Figure 4 shows a comparison of the apparent density of the tested moulding sands. Based on the analysis of the density test results, it was found that the apparent density of thermally hardened moulding sand at 160°C ranges from 1.267 g/cm^3 to 1.376 g/cm^3. The highest density was found for the MC1 mixture (1.376 g/cm^3), while the lowest value was obtained for the MB2 mixture (1.267 g/cm^3) (Fig. 4a). The apparent density of microwave-cured moulding sands ranges from 1.283 g/cm^3 to 1.361 g/cm^3. The highest value was obtained for sand MC3 (1.449 g/cm^3), and the lowest for MB4 (1.283 g/cm^3) (Fig. 4b). In the case of both thermal curing methods, the highest density – the highest degree of compaction – is characteristic of compounds with aluminosilicate binders: a classic one (C) (MC1, MC3) and intended for 3D printing (C3D) (MC2 and MC4).

Figures 5 and 6 show the bending and tensile strength values of the tested moulding sands. Based on the analysis of the bending strength of thermally hardened mixtures using a dryer (Fig. 5a), it can be concluded that the highest results are obtained by moulding mixtures made with sodium silicate binders A and A3D, reaching 2.542 MPa and 2.464 MPa, respectively. For mixtures with modified sodium silicate binder (B) and aluminosilicate binder (C) and their modified equivalents for 3D printing, the strength values do not differ significantly and fluctuate around 2.000 MPa. In the case of microwave-cured sand (Fig. 5b), the obtained bending strength values allow us to conclude that the use of a modified binder for 3D printing leads to a reduction in strength compared to classic binders. The highest value, 3.122 MPa, was achieved for the MC3 compound with binder C. The lowest strength was observed for the MB3 and MB4 compounds, with the MB4 compound using binder B modified for 3D printing, achieving a result 0.582 MPa lower.

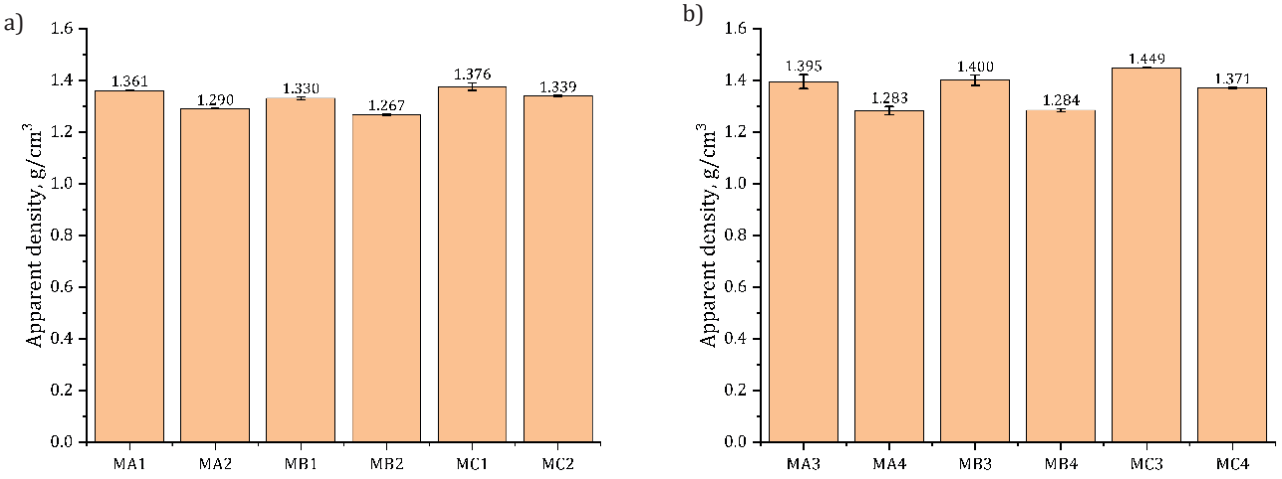

Fig. 4. The influence of binder type on the apparent density of tested moulding sands: a) thermal hardening in a dryer; b) thermal hardening by microwaves

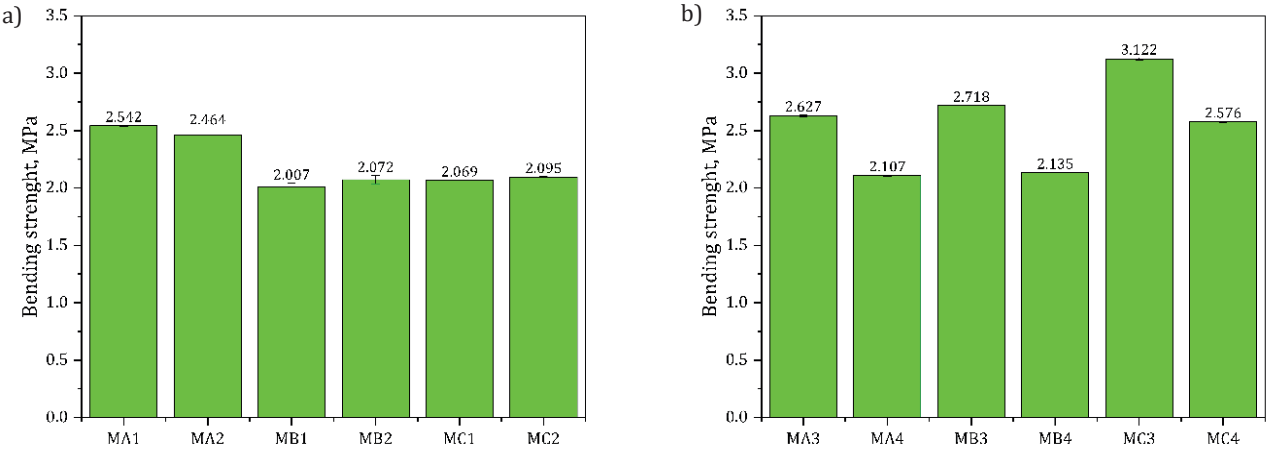

Fig. 5. The influence of binder type on bending strength of tested moulding sands: a) thermal hardening in a dryer; b) thermal hardening by microwaves

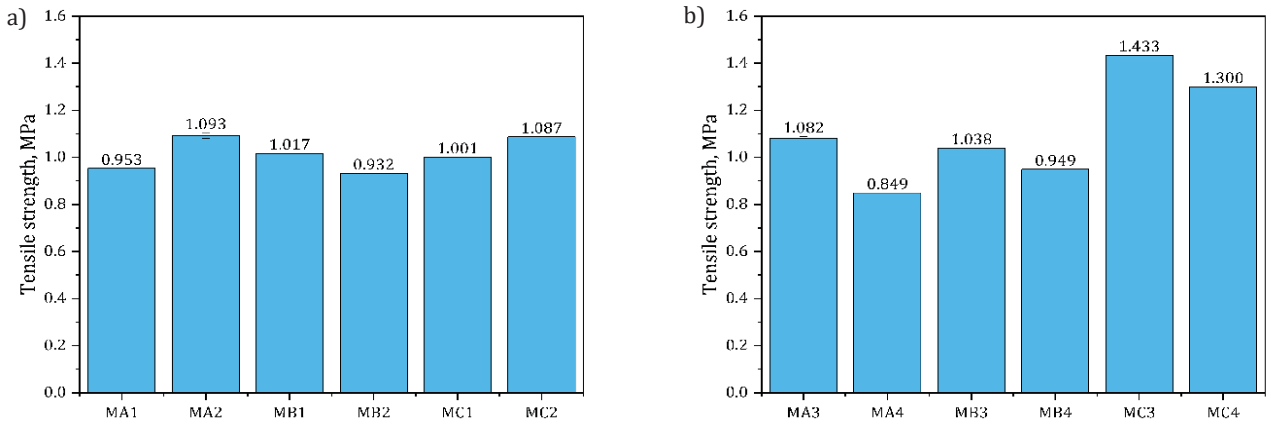

Fig. 6. The influence of binder type on tensile strength of tested moulding sands: a) thermal hardening in a dryer; b) thermal hardening by microwaves

In terms of tensile strength (Fig. 6a), the highest results were obtained for the MA2 compound – 1.093 MPa. This compound is characterized by better tensile strength than its counterpart with a classic binder. In the case of the compound with binder B3D – MB2, a slight decrease in strength was observed compared to its classic counterpart – MB1. In the case of the compound with binder C modified for use in 3D printing (C3D) – MC2, a slight increase in tensile strength was observed compared to the compound with a classic binder – MC2. In the case of microwave-cured mixtures, it was observed that all tested moulding sands using modified binders (MA4, MB4, MC4) are characterized by lower tensile strength values compared to mixtures with classic binders (MA3, MB3, MC3) (Fig. 6b).

Figure 7 shows a comparison of the permeability of the tested moulding sands. Analysis of the obtained test results showed slight differences between the tested moulding compounds thermally cured in a dryer (Fig. 7a). The permeability values range 583.33–608.33 · 10^{-8} m^2/(Pa·s). Mixtures with classic binders, sodium silicate A (MA1) and aluminosilicate C (MC1), are characterized by the best permeability – 608.33 · 10^{-8} m^2/(Pa·s). The lowest permeability value was observed for the mixture with modified sodium silicate binder MB1 – 583.33 · 10^{-8} m^2/(Pa·s). When comparing the permeability values of moulding compounds with a classic binder and one developed for use in 3D printing, it can be seen that the modification of binders does not significantly affect the permeability of the tested compounds. In the case of microwave-cured compounds (Fig. 7b), the permeability values are not significantly higher than in the case of thermal curing in a dryer. The permeability values range 600.00–633.33 · 10^{-8} m^2/(Pa·s). The highest value was obtained for the MA4 mixture with a modified binder (A3D) – 633.33 · 10^{-8} m^2/(Pa·s).

Figure 8 shows a comparison of the friability of the tested moulding compounds. Analysis of the test results showed that thermally cured mixtures in the dryer achieve friability values ranging from 3.007% to 5.328% (Fig. 8a). The lowest friability was observed in the compound with aluminosilicate binder C (MC1) – 3.007%, and the highest in the compound with modified sodium silicate binder B developed for use in 3D printing B3D (MB2) – 5.328%. Modification of commercial binders for use in 3D printing results in the following: in the case of the thermal curing of sands in

a dryer, modified silicate B and aluminosilicate C increases the friability tendency of the tested compounds. As a consequence, this may cause casting defects. Microwave-cured sands are characterized by similar friability in the range of 4.453–4.594% (Fig. 8b). The differences in friability values between compounds made with classic binders (MA3, MB3, MC3) and compounds made with binders developed for 3D printing (MA4, MB4, MC4) are minimal and do not exceed 0.150 percentage point.

Figures 9 and 10 show the effect of the type of binder used on the thermal deformation of the tested compounds, determined on the basis of the hot distortion parameter tests.

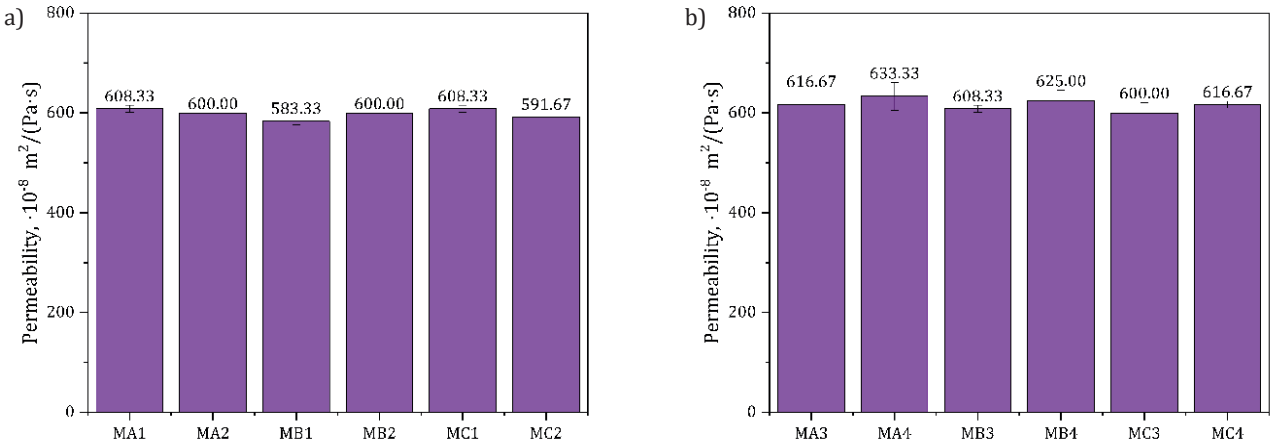

Fig. 7. The influence of binder type on permeability of tested moulding sands: a) thermal hardening in a dryer; b) thermal hardening by microwaves

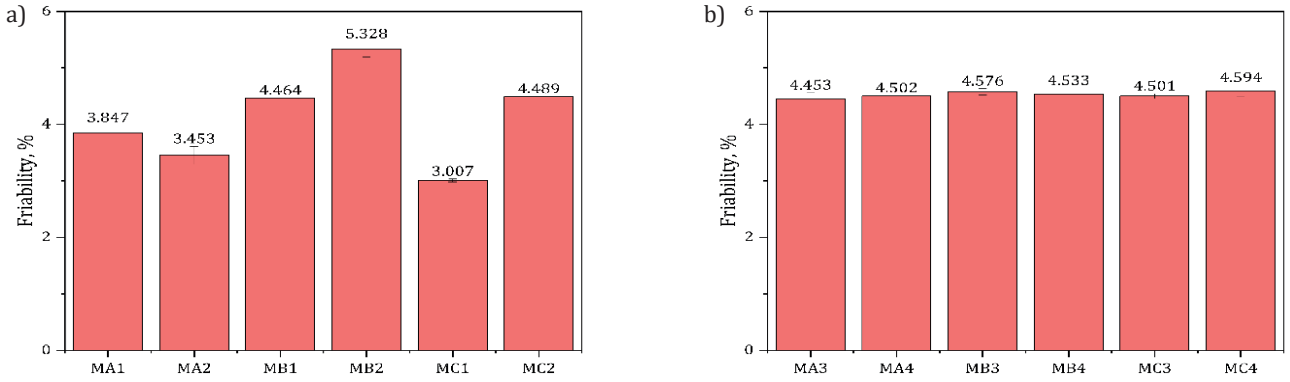

Fig. 8. The influence of binder type on friability of tested moulding sands: a) thermal hardening in a dryer; b) thermal hardening by microwaves

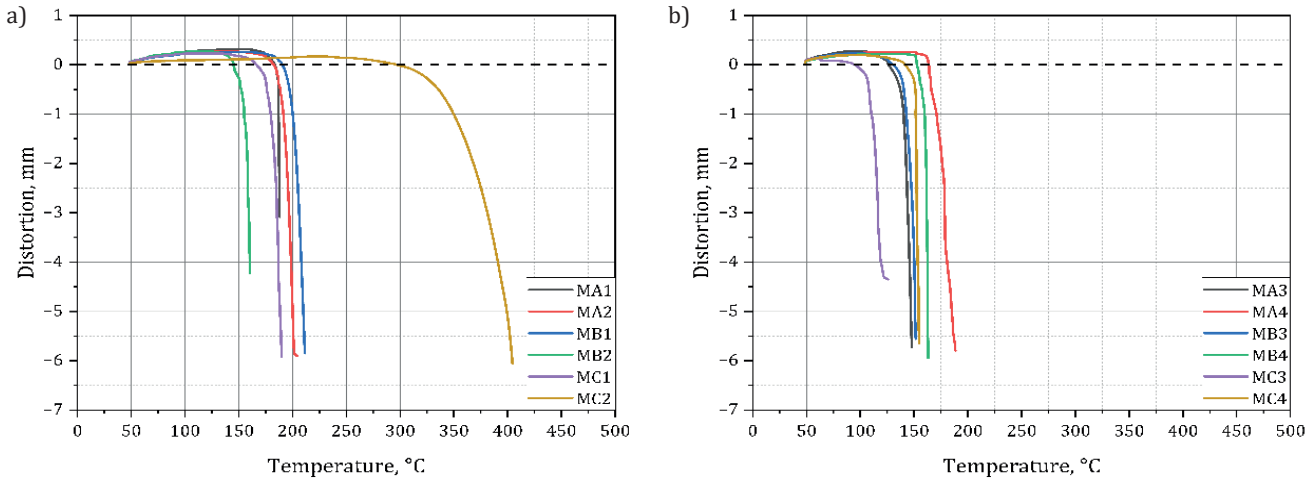

Fig. 9. The influence of binder type on hot distortion parameter in function of temperature: a) thermal hardening in a dryer; b) thermal hardening by microwaves

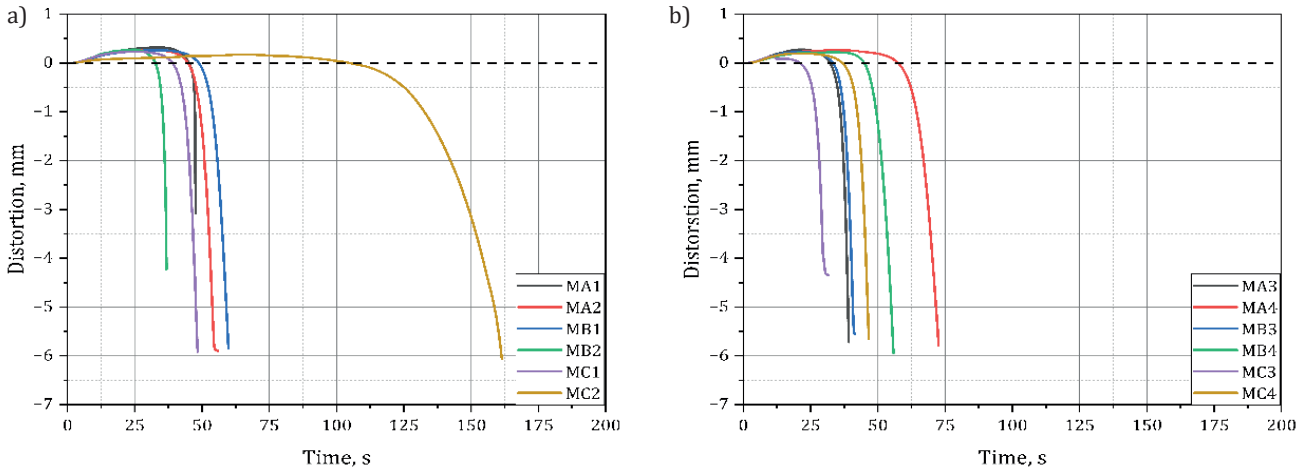

Fig. 10. The influence of binder type on hot distortion parameter in function of time: a) thermal hardening in a dryer; b) thermal hardening by microwaves

Analysis of the test results showed that all the moulding compounds tested exhibit thermal deformation typical for compounds with inorganic binders (Figs. 9 and 10). Most of the mixtures show thermal resistance for up to approximately 50 s. The exception is the MC2 mixture, which is thermally resistant for up to 161.2 s and at temperatures up to 404.9°C. The lowest thermal stability is characteristic of the MB2 mixture – the standard test specimen deformed after 36.8 s at a temperature of 124.6°C. In the case of microwave-cured compounds, the lowest thermal resistance is characteristic of the MC3 compound – the standard test specimen deformed after 31.8 s of heating at a temperature of 134.3°C. The best thermal stability is exhibited by the sand with sodium silicate binder A – MA3 – the standard test specimen deformed after 72.6 s of heating at a temperature of 188.4°C.

3. CONCLUSIONS

Based on the conducted research, the following conclusions can be drawn:

- New, modified inorganic binders developed for 3D printing of moulds and cores using Binder Jetting technology can be used as binding materials in thermally cured moulding sands.
- It is possible to use thermal curing carried out both in a dryer and in a microwave oven for the production of moulds and cores using inorganic binders developed for use in 3D printing technology (binder jetting).
- The best parameters for curing in a dryer were obtained for compounds containing 2.0 p.p.w. of binder cured at 160°C for 10 min.
- The best parameters for microwave curing in an 800 W microwave oven at a frequency of 2.45 GHz were obtained for compounds containing 2.0 p.p.w. during 6 minof curing.
- The chosen technological properties of moulding compounds with binders developed on the basis of commercial binders for use in 3D printing technology, thermally cured in a dryer, do not differ significantly from the properties of compounds with commercial binders. However, a decrease in the strength of microwave-cured mixtures with new binders was observed in comparison to mixtures with classic binders.
- The method of thermal curing and the modification of binders do not adversely affect the thermal stability of the tested compounds.
- The most important part of these results significantly shows that used materials and techniques have huge potential for use as inorganic binders in 3D printing Binder Jetting technology.
- In the next phase of the research, the feasibility of using new binding systems in semi-industrial and industrial settings will be evaluated, including their potential application in the production of casting molds and cores using the Binder Jetting method.

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