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Selection of Chemically Cured Molding Sands' with Inorganic Binders Dedicated to 3D Sand Printing

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Abstract

Due to the high technological potential, 3D printing technologies in the foundry industry are developing very dynamically. Binder jetting technology is most commonly used for the production of sand molds and cores with 3D printing. The binding materials used in foundry practice are organic resins modified with furfuryl alcohol. These materials are characterized by excellent technological properties, but at the same time, they are harmful to the environment. Environmentally friendly inorganic binders are an alternative to the organic binders used for the production of molds and cores, and this is the subject of research carried out at various research centers.

This work determines the influence of molding sands' with different inorganic binders composition on their chosen properties. The molding sands with 3 commercial inorganic binders used in traditional mold and core production technologies were tested as well as the molding sands dedicated to 3D printing with binders based on them. Four types of hardeners were used for chemical curing.

The technological (strength, permeability, abrasion) and thermophysical (thermal deformation) tests carried out on molding sands and the physicochemical tests on binders (viscosity, wettability of the quartz substrate) have shown that inorganic binders elaborated on the basis of commercial binders can be used in 3D printing technology. The selected sands' compositions were chosen for further research.

Keywords: 3D sand printing, Molding sand, Resin, Inorganic binder, Hardener

1. Introduction

Binder jetting - 3D printing technology is finding increasing application in the foundry industry in the production of sand molds and cores [1-2]. 3D printed molds and cores allow for the achievement of very good surface quality of castings, which is a basic criterion for assessing their quality. The binders used in foundry practice to bind the matrix grains in 3D printing technology are organic binders - mainly resins modified with furfuryl alcohol and cured using no-bake technology [3-5]. The

classic no-bake technology is widely used in foundry practice for the production of molds and cores. It enables the production of castings with high dimensional accuracy and complex shapes, bonding takes place at ambient temperature, the process of mold making and removing the casting from the used mold does not require a lot of labor, a low binder content is used, and thanks to the use of an organic binder, the molding sand is characterized by good knock-out properties and mechanical reclamation capacity. The main disadvantages of this technology include the high toxicity of the gases emitted, mainly during the pouring of the liquid alloy [6-7].



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Inorganic binders are an alternative to organic binders. They are more difficult to process, have poor knock-out properties and, are difficult to regenerate mechanically, but they are environmentally friendly. It is the advantage of inorganic binders that has led to research being conducted for several decades in many Polish and foreign centers to improve the properties of the molding sands with these binders. Similar to the no-bake technology with organic binders, compounds with inorganic binders can be cured using self-curing technology. This is the so-called ester technology using hardeners based on esters of acetic acid, carbonic acid, or their mixtures.

As part of this work, molding sands with inorganic binders cured in ester technology will be investigated. The possibility of developing environmentally friendly inorganic binders for 3D printing using printers designed for organic systems will be demonstrated. The authors will prove that binders can be developed based on commercial inorganic binders intended for the production of molds and cores using conventional methods.

2. Methodology and materials

2.1. Materials

The following materials were used for the tests:

– matrix

Table 1.

Compositions of tested molding sands

Sym.	Sand matrix	Amount [weight part]	Binder	Amount [weight part]	Hardener	Amount per binder content [%]
MA1	Quartz sand	100	A	2.5	Flodur 5	10
MA2	Quartz sand	100	A _{3D}	2.5	Flodur 5	10
MA3	Quartz sand	100	A _{3D}	2.5	Flodur 5	7.35
MA4	Quartz sand	100	A	2.5	Ixional	10
MA5	Quartz sand	100	A _{3D}	2.5	Ixional	10
MA6	Quartz sand	100	A _{3D}	2.5	Ixional	7.35
MA7	Quartz sand	100	A	2.5	Mach 1	10
MA8	Quartz sand	100	A _{3D}	2.5	Mach 1	10
MA9	Quartz sand	100	A _{3D}	2.5	Mach 1	7.35
MB1	Quartz sand	100	B	2.5	Flodur 5	10
MB2	Quartz sand	100	B _{3D}	2.5	Flodur 5	7.35
MB3	Quartz sand	100	B	2.5	Ixional	10
MB4	Quartz sand	100	B _{3D}	2.5	Ixional	7.35
MB5	Quartz sand	100	B	2.5	Mach 1	10
MB6	Quartz sand	100	B _{3D}	2.5	Mach 1	7.35
MC1	Quartz sand	100	C	2.5	hardener	10
MC2	Quartz sand	100	C _{3D}	2.5	hardener	10
MC3	Quartz sand	100	C _{3D}	2.5	hardener	7.35

Sibelco quartz sand was used for the tests. Sieve analysis showed that the main fraction was collected on sieves numbered 0.200/0.160/0.100. According to the Polish standard PN-85/H-11001, this classifies the tested material as fine-grained sand. In the tested matrix, the main fraction value is 92%, which classifies the tested sand as homogeneous, while the average grain size is 0.225 mm.

– binders

Three different inorganic binders available on the market were used for the tests: two binders based on hydrated sodium silicate, designated as A and B in the study, and an aluminosilicate binder – C. As part of the study, binders with modified viscosity were developed for 3D printing technology: A_{3D}, B_{3D}, C_{3D}.

– hardeners

The following hardeners were used for the tests: a classic hardener for hardening compounds with hydrated sodium silicate in Floster technology – Flodur 5 (based on esters of acetic acid); Ixional hardener (based on esters of carbonic acid); Mach 1 hardener (commercial hardener based on the mixture of acetic and carbonic acid esters) and a hardener dedicated to the chemical hardening of compounds with aluminosilicate binder.

The compositions of the tested compounds are presented in Table 1.

The molding sands were prepared in a paddle mixer; the mixing time of the matrix with the hardener was 1 min and with binder – additional 1 min. The molding sands were compacted by vibration using a WADAP LUZ device; compaction time was 9 s.

2.2. Research Methodology

The basis for the development of new inorganic binders for 3D printing was research into the viscosity of a commercial organic binder for 3D printing [8-9] and the technical parameters of the Kocel AJS 300A 3D printer [10]. The new binders were developed by changing the viscosity of commercial inorganic binders intended for the production of molds and cores using conventional methods. The viscosity was changed by adding an appropriate amount of solvent to the binders.

The dynamic viscosity of the binder depends on the shear stress and shear rate in a linear manner. It is the slope of the line formed by the relationship between shear stresses and shear rates. The viscosity was tested using a rheometer modified by Jota. To carry out the test, 25 ml of binder was subjected to increasing and decreasing speeds of the S₁ measuring roller, which generated shear stresses. The value of the parameter was read from equation 1 [11]

$$y = ax + b \quad (1)$$

where:

a – viscosity.

In the next stage of the research, the effect of viscosity change on the wettability of quartz by new binders was determined. The research was conducted using a high-temperature microscope produced by Leitz at the High-Temperature Microscopy Laboratory of the Faculty of Materials Science and Ceramics at AGH University of Krakow. The contact angle was determined using the static method at room temperature (22°C). A drop was applied to the quartz substrate, and the sample was kept on a substrate until it reached its final shape. The wetting angle determined in this way is the equilibrium angle. It should be emphasized that the wetting angle is not a material constant but characterizes a given test system. The wetting angle values for the binder on different substrates, such as corundum or steel, can vary significantly. In addition, the smoothness of the substrate surface also impacts the wetting angle value [12].

Due to the change in the viscosity of the binders, the change in the number of grain matrix layers penetrated by the new binders was determined. This is very important from the point of view of 3D printing technology. Based on the height of the print head of the Kocel AJS 300A printer and mathematical models [13] of liquid penetration into porous material, a test setup was developed, the diagram of which is shown in Fig. 1. According to the developed methodology, a drop of binder from a burette is dripped onto a sand loose sand bed with a grain size of less than 0.20 mm. After 1 minute, the depth of binder penetration into the substrate is measured. A tape measure was attached to each of the measuring cuvettes and images were taken at a fixed distance with a camera placed on a tripod. ImageJ software was used to measure the depth.

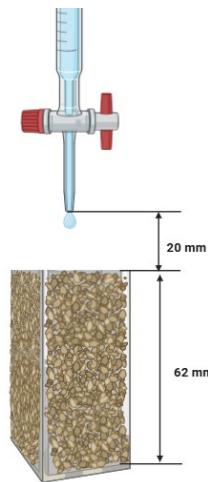


Fig. 1. Diagram of a measuring station for determining the depth of penetration of a binder into the matrix

Then, the influence of the used binder type on the curing kinetics of the tested molding sands was determined. The tests were carried out using the ultrasonic method [14]. Chemically self-hardening sands are classified as rheologically unstable materials. During the first phase of hardening, they have the characteristics of a viscoelastic body, and as the hardening time progresses, they acquire the characteristics of an elastoplastic body with decreasing elasticity, so that at the end they acquire the characteristics of a non-linearly elastic body. This change in the rheological properties of the molding sand has a significant impact on the propagation of ultrasonic waves during their hardening process. The conditions in the bonding chamber are similar to conditions near the casting pattern in the mold [15, 16].

Bending strength and tensile strength tests of molding compounds were performed after 2, 4, and 24 hours of curing on a universal device for determining strength properties, MultiSerw Morek LRu-2e. Permeability tests of the compounds were performed on a WADAP LPiR1 device. The measurement method involves placing a cylindrical fitting in a removable sleeve of the device and placing it in an apparatus for rapid determination of permeability - the apparatus was operated for approximately 30 seconds. Standard cylindrical fittings (50x50 mm) were used to test permeability and friability, while standard longitudinal fittings (22.36x22.36x172 mm) were used to test the bending strength and standard "dog bone" fittings (with a cross-section of 22.36x22.36 mm) were used to test tensile strength. The falling shot method was used to measure the friability of the molding sands. The test was carried out on standard cylindrical fittings after 24 hours of curing, using the Huta Stalowa Wola device. Shot with a diameter of approximately 1 mm fell from a height of 0.307 m onto a fitting rotating at a speed of 1 rpm. The amount of shot used was 1.75 kg.

Thermal deformation tests (hot distortion parameter) of molding compounds were carried out on a DMA device manufactured by Multiserw Morek [17]. In accordance with the methodology, the tested samples with dimensions of 114x25.4x6.3 mm after 24 hours of curing are heated by two 500 W halogen lamps to a maximum temperature of 900°C [18].

3. Results and discussion

3.1. Dynamic viscosity, contact angle & penetration depth of tested binders

The flow curves obtained for the new binders, together with their dynamic viscosity values in relation to commercial base binders, are shown in Figs. 2-3.

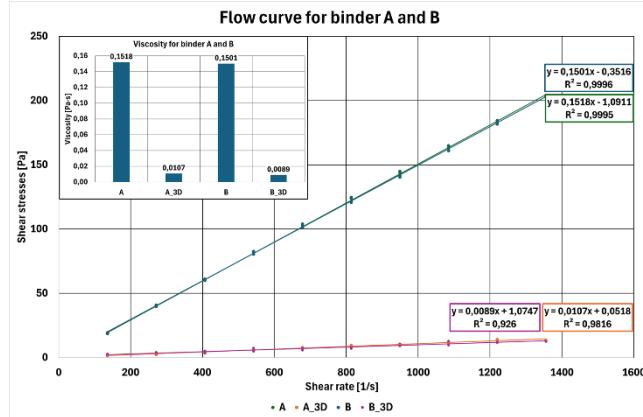


Fig. 2. Flow curves and dynamic viscosity of binders based on hydrated sodium silicate (A and B)

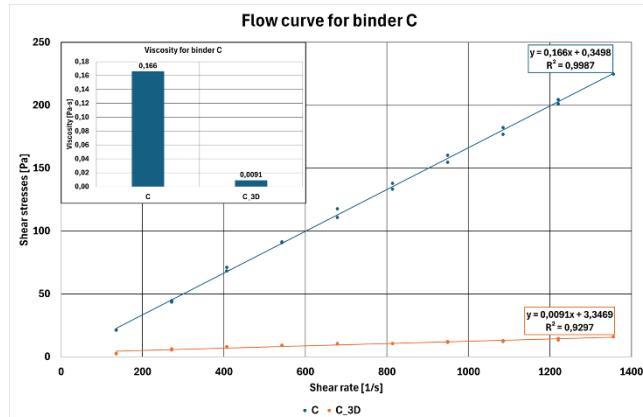


Fig. 3. Flow curves and dynamic viscosity of binders C

In the case of binder A, the base viscosity (before modification) is 0.1518 Pa·s, and after modification of its physicochemical properties, it was reduced to the value of 0.0107 Pa·s, which is 7% of the viscosity of the base binder. In the case of binder B, a reduction in viscosity to 6.52% of the value obtained for the base binder B is observed. In the case of binder C, to 5.5% reduce in viscosity compared to the viscosity of the base binder C is observed. A reduction in viscosity to the specified level is required due to adapting the new inorganic system to the currently used 3D printers. Commercial inorganic binders are characterized by approximately twice the viscosity of organic binders used in conventional mold and core production methods.

The effect of viscosity change on the wettability of quartz by new binders is shown in Fig. 4.

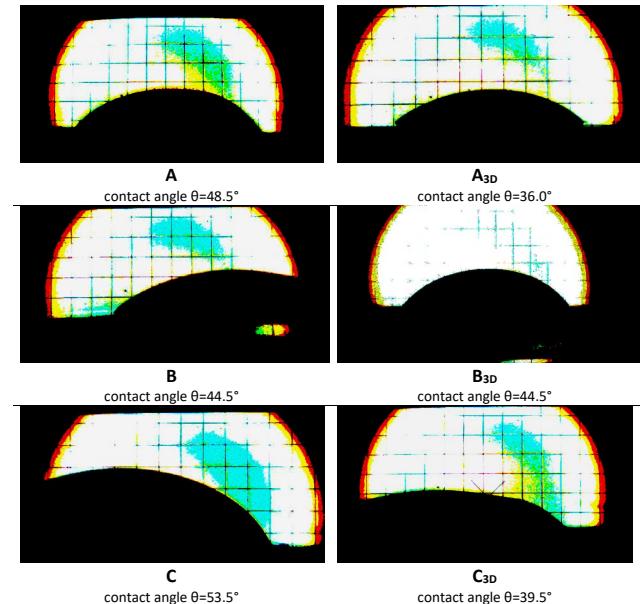


Fig. 4. Wettability of the tested binders on a quartz plate

The tests showed that all the tested binders wet quartz substrate well – the contact angle θ is below 90°. For modified binders A_{3D} and C_{3D}, a decrease in the contact angle was observed compared to the base binders A and C, which means better wettability. This wasn't seen with binder B_{3D}.

Fig. 5 shows the effect of reduced binder viscosity on the depth of their penetration into the matrix layers.

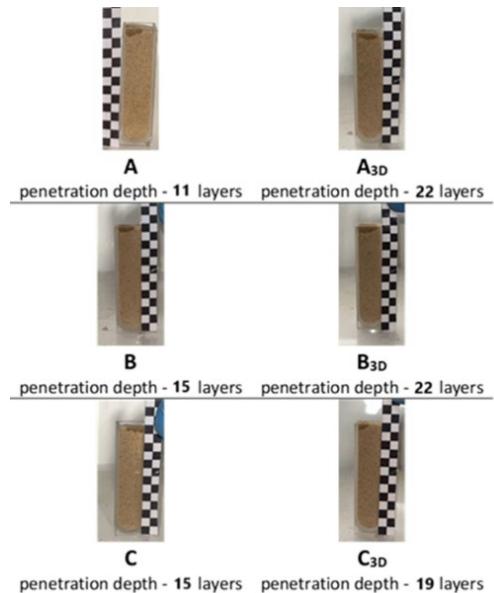


Fig. 5. The influence of binders' viscosity on penetration depth

The tests conducted have shown that reducing the viscosity of the binder increases the depth of their penetration between successive layers of the grain matrix. For binder A, the penetration depth is 11 layers, and for binder A_{3D}, it is 22 layers. For binder B, the penetration depth is 15 layers, and for binder B_{3D}, it is 22 layers. In the case of binder C, the penetration depth is 15 layers, and in the case of binder C_{3D}, it is 19 layers. For comparison, tests were carried out on an organic binder dedicated to 3D printing, which showed its penetration into the sand substrate to a depth of 17 layers. This means that the new binders have better penetration than base binders. However, further research should consider the possibility of using appropriate additives to the new binders to reduce their penetration depth. From the point of view of 3D printing, and especially the use of self-hardening sands technology, this is a very important observation. The penetration of the binder to a depth of too many layers can affect its uneven distribution in the bonded layers of the mold or core. Excessive penetration depth of the binder when using self-curing technologies in 3D printing may necessitate the use of highly reactive hardeners. Additionally, with this technology, it will be necessary to test the penetration of the binder into the substrate, which is a mixture of the matrix and the hardener.

3.2. Kinetics of hardening

The MA3, MA6, MA9, MB2, MB4, MB6, and MC3 compounds with compositions presented in Table 1 were selected for the curing kinetics tests. The aim of the study was to determine the effect of the hardener used on the curing kinetics of compounds with new binders. The results of the tests are presented in Figures 6-8.

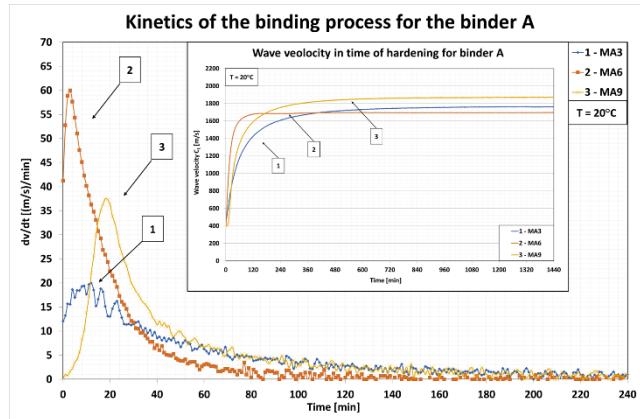


Fig. 6. The course of curing kinetics of tested molding compounds with binder A and various hardeners

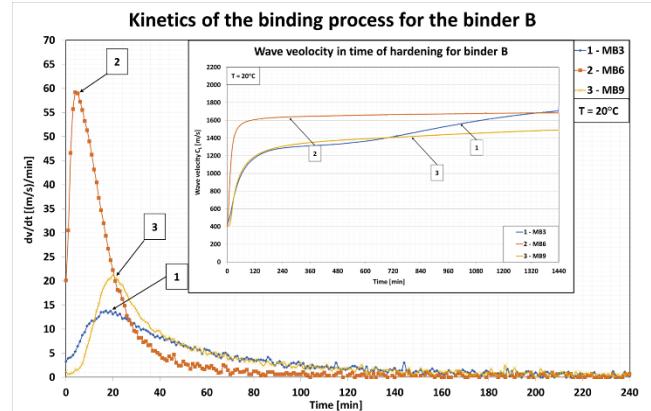


Fig. 7. The course of curing kinetics of tested molding compounds with binder B and various hardeners

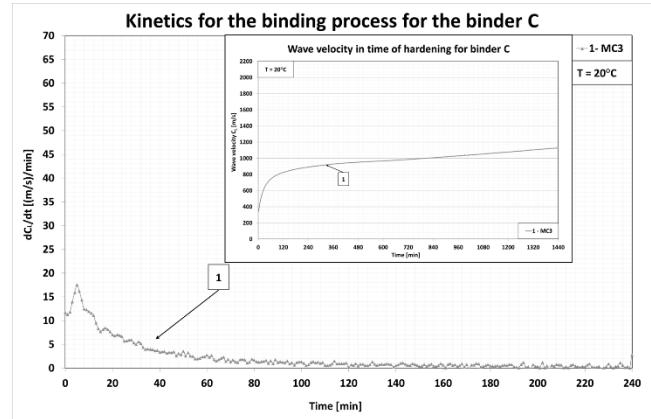


Fig. 8. The course of curing kinetics of tested molding compounds with binder C and a dedicated hardener

Based on the analysis of the curves shown in Fig. 6, it can be concluded that in the case of binder A_{3D} and hardener Ixional, there is a clear maximum (MA6) – the curve in this case is most similar to the curve obtained for resins used in 3D printing [9]. The maximum also occurs on the curve obtained for the compound with Mach 1 hardener (MA9). In the case of the MA3 compound – cured with Flodur 5 – no clear maximum is observed on the wave velocity curve. In the case of the B_{3D} binder, a similar characteristic of the bonding kinetics curves can be observed as in the case of compounds made with the A_{3D} binder (Fig. 7). In the case of curing compounds with A_{3D} and B_{3D} binders, the kinetics curve obtained for the Mach 1 hardener shows that the reaction starts with a slight delay, which may be disadvantageous when making molds and cores in 3D printing technology.

The curing kinetics of the MC3 compound is characterized by a distinct peak, while the reaction rate is similar to that of the MA3 and MB9 compounds. The course of curing kinetics is similar to MA6 and MB6, but the value is almost three times lower.

3.3. Molding sands' chosen technological properties

As part of the work, bending and tensile strength, permeability, and friability tests were carried out. The results of the tests are presented in Figures 9-20.

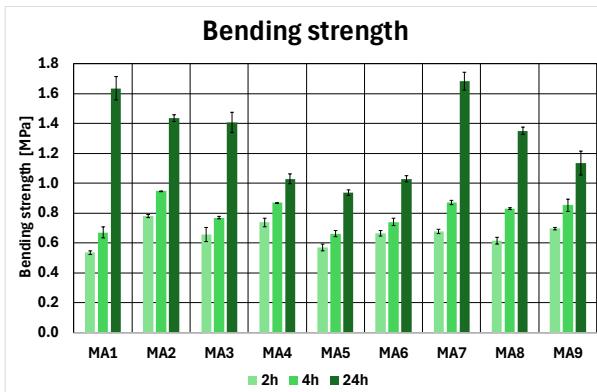


Fig. 9. The effect of binder A modification and hardener type on sands' bending strength after 2, 4, and 24 hours of curing

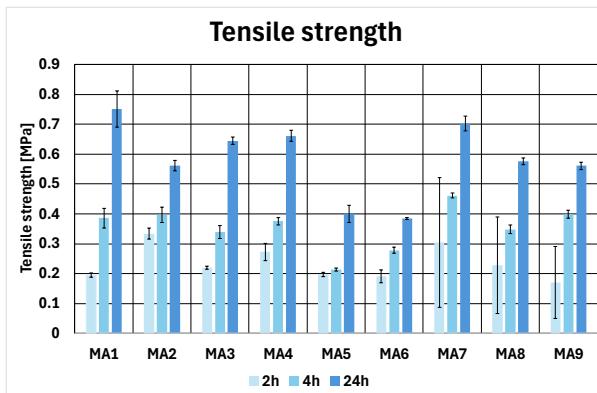


Fig. 10. The effect of binder A modification and hardener type on sands' tensile strength after 2, 4, and 24 hours of curing

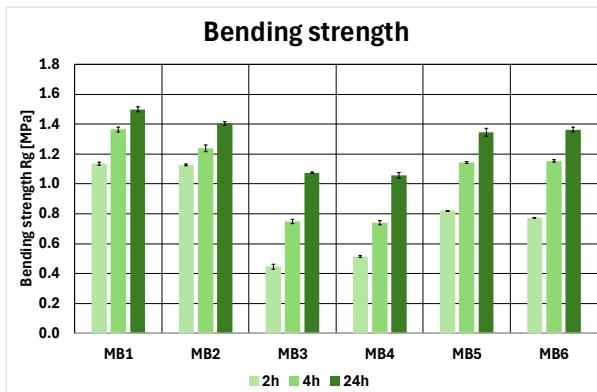


Fig. 11. The effect of binder B modification and hardener type on sands' bending strength after 2, 4, and 24 hours of curing

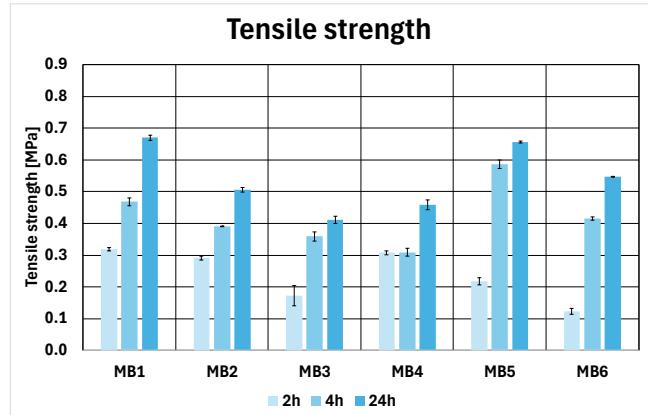


Fig. 12. The effect of binder B modification and hardener type on sands' tensile strength after 2, 4, and 24 hours of curing

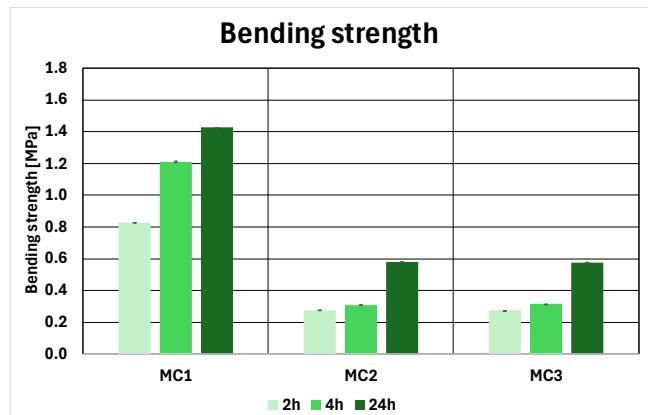


Fig. 13. The effect of binder C modification on sands' bending strength after 2, 4, and 24 hours of curing

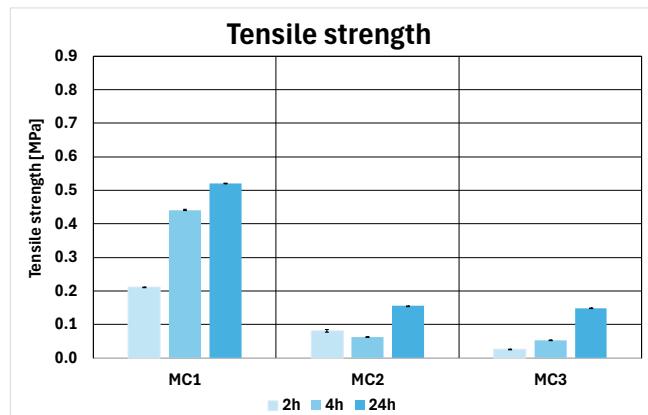


Fig. 14. The effect of binder C modification on sands' tensile strength after 2, 4, and 24 hours of curing

Based on the analysis of the strength test results, it can be seen that the best strength properties are achieved by molding sands using binder A. Compounds with binder A_{3D} cured with Flodur 5 hardener, show the lowest percentage decrease in strength compared to other hardeners – about 13%. The bending strength of

MA2 and MA3 is approx. 1.4 MPa, which gives satisfactory strength properties (Fig. 9). Similar results can be observed for tensile strength (Fig. 10).

In the case of binders B and B_{3D}, the lowest strength values are achieved by molding sands MB3 and MB4 cured with Ixional hardener, reaching approximately 1.1 MPa in bending strength and 0.45 MPa in tensile strength. The strength values for the other hardeners are similar to each other, but slightly lower than those for sands made with binder A (Fig. 11-12).

Based on the analysis of the strength test results for molding sands with binder C, it can be seen that the modification of the binder in this case does not bring satisfactory results. The bending strength values of sands made with binder C_{3D} are 42.8% lower than those of sands with unmodified binder (Fig. 13-14).

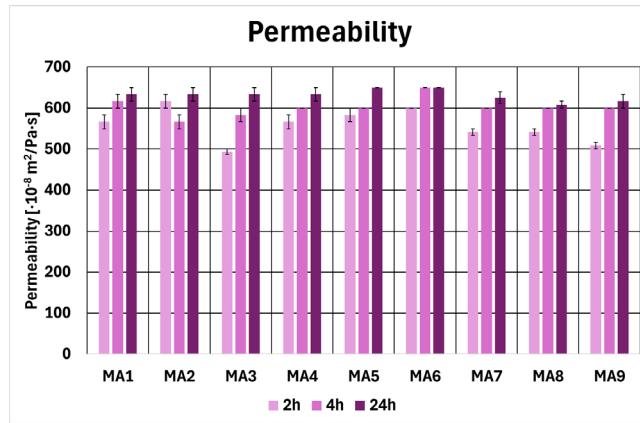


Fig. 15. The effect of binder A modification and hardener type on sands' permeability after 2, 4, and 24 hours of curing

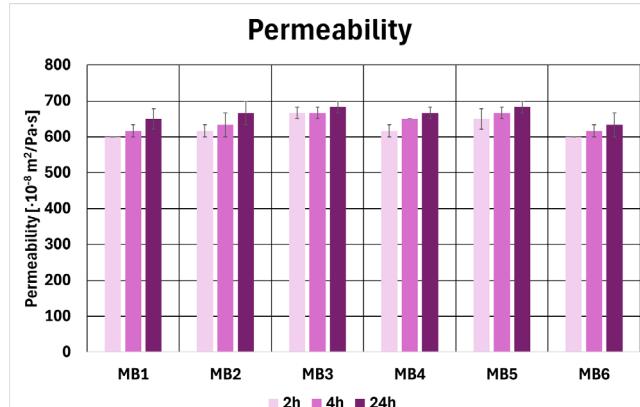


Fig. 16. The effect of binder B modification and hardener type on sands' permeability after 2, 4, and 24 hours of curing

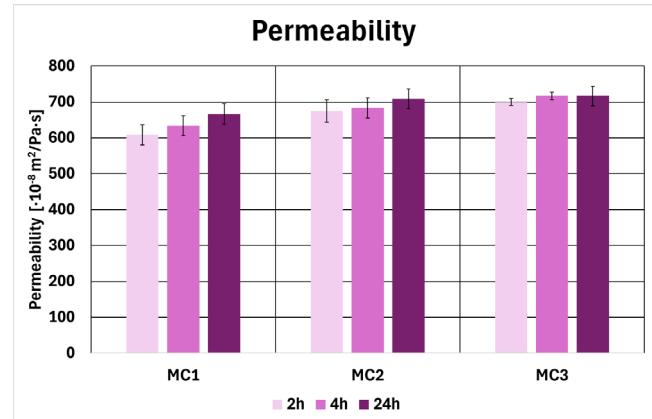


Fig. 17. The effect of binder C modification on sands' permeability after 2, 4, and 24 hours of curing

The permeability tests carried out showed that all the tested molding sands have similar permeability. This parameter increases with curing time. The values after 24 hours of curing range from 625 to 700 10⁻⁸ m²/(Pa·s) (Fig. 15-17), which indicates good sands' permeability.

In the case of compounds cured with Flodur 5 hardener (MA1, MA2, MA3, MB1, and MB2), friability is comparable. The highest mass loss during abrasion testing was observed for MA5 cured with Ixional hardener (Fig. 18-19). In the case of binder C, an increase in friability of molding sands with the new binder compared to sands with the base binder of approx. 50% is observed (Fig. 20).

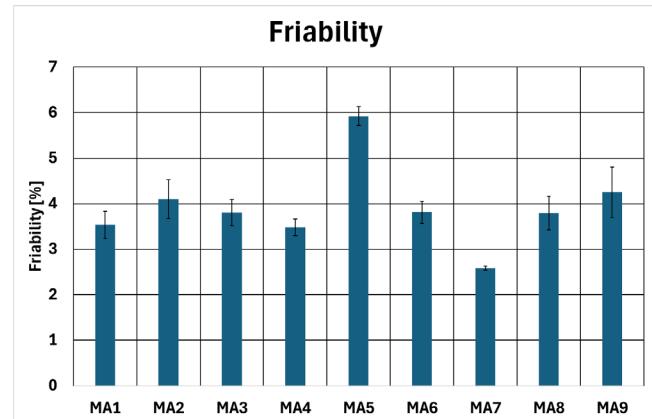


Fig. 18. The effect of binder A modification and hardener type on sands' friability after 2, 4, and 24 hours of curing

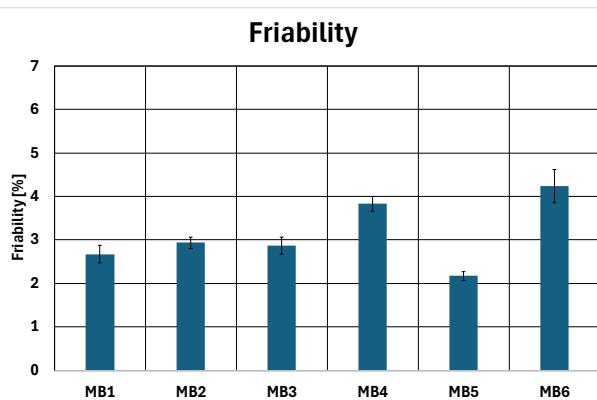


Fig. 19. The effect of binder B modification and hardener type on sands' friability after 2, 4, and 24 hours of curing

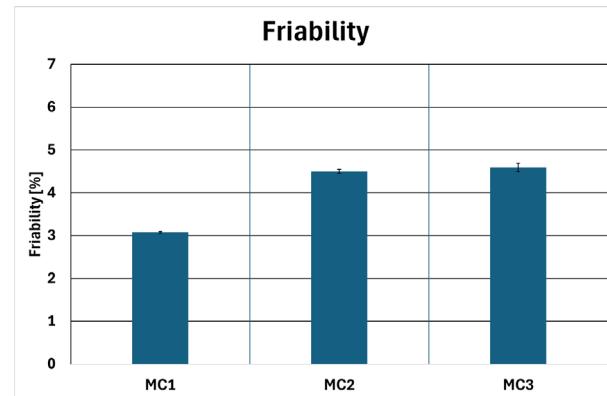


Fig. 20. The effect of binder C modification on sands' friability after 2, 4, and 24 hours of curing

3.4. Molding sands' thermophysical properties - hot distortion parameter

Thermal deformation tests were carried out on mixtures with binders A, B, and new: A_{3D}, B_{3D}. Due to the low strength of compounds with chemically hardened binder C_{3D}, it was not possible to produce fittings for their hot distortion tests. The results of the tests are presented in Fig. 21-22.

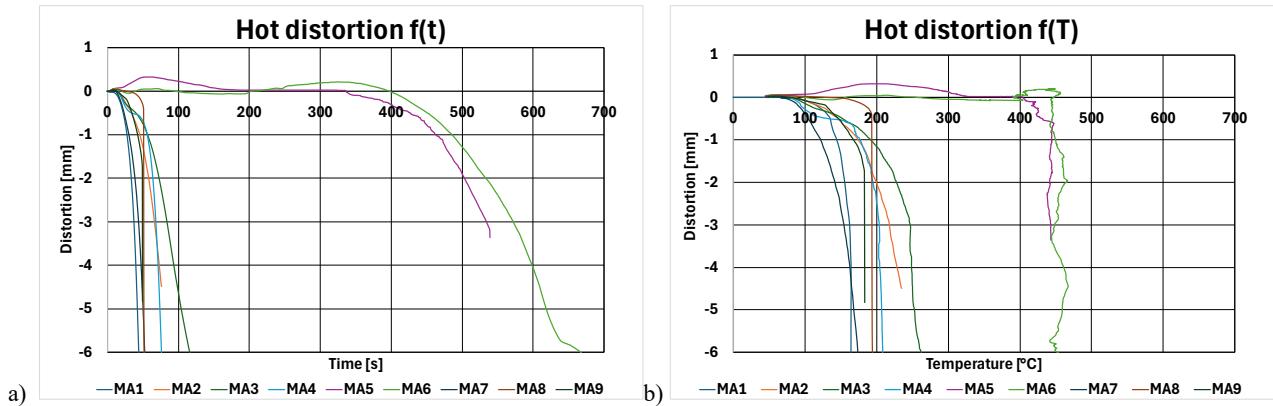


Fig. 21. The effect of binder A modification and hardener type on the thermal deformation of tested molding sands; as a function of heating a) time; b) temperature

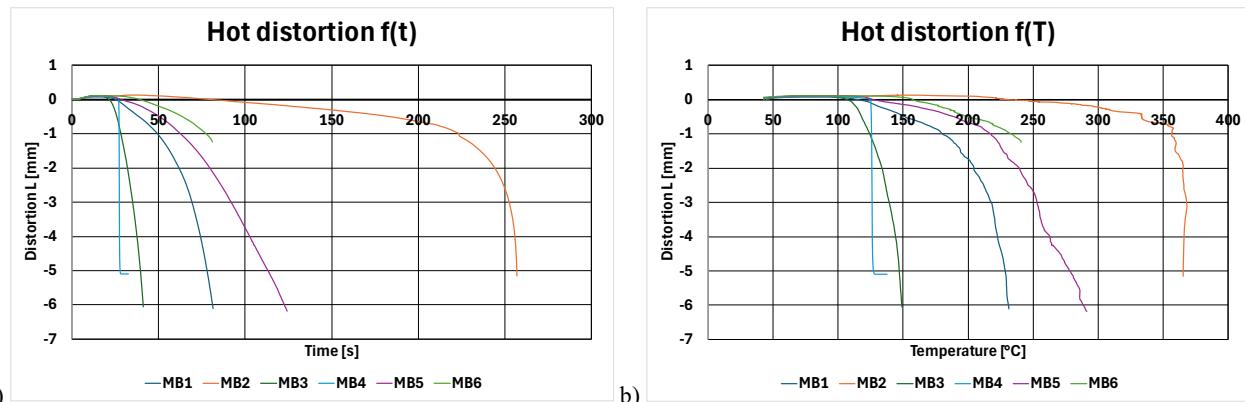


Fig. 22. The effect of binder B modification and hardener type on the thermal deformation of tested molding sands; as a function of heating a) time; b) temperature

The deformation process of the tested molding compounds is typical for molding compounds made with an inorganic binder. Thermal stability is observed in the initial stage of the test, with mild deformation of the fitting until its destruction. In the case of molding compounds made with binders A and A_{3D}, the longest time and highest thermal stability temperature are achieved by compounds hardened with Ixional hardener (MA5 & MA6) – app. 350 s and 400°C. The other tested compounds are characterized by thermal stability for up to approximately 100 s and at temperatures ranging from 150 to 220°C.

The tested compounds using binders B and B_{3D} are characterized by a shorter time and lower temperature range of thermal stability. The best results were obtained for compound MB2, which is characterized by thermal stability for up to 256 s and a temperature of 356°C.

4. Conclusions

Based on the conducted research, the following conclusions were drawn:

- the physicochemical properties of commercial inorganic binders can be adjusted to the parameters of binders intended for 3D printing;
- the new A_{3D} and B_{3D} binders, cured with commercial hardeners, can be used for chemical bonding of sands during 3D printing of molds and cores;
- molding sands cured with Flodur 5 hardener have the best technological properties;
- the use of Ixional hardener accelerates the curing kinetics of sands, which may be beneficial for printed molds and cores;
- molding sands with the new C_{3D} chemically cured binder have too low properties for 3D printing. This does not mean that this binder will be eliminated from further research.

Further research will be focused on the possibility of using other sands' curing technologies with new inorganic binders designed for 3D printing technology.

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