



# THE IMPACT OF INTERNAL HYDROPHOBIZATION ON THE PROPERTIES OF THE CEMENT-BASED MATERIALS WITH MINERAL ADDITIVES

## WPŁYW HYDROFOBIZACJI OBJĘTOŚCIOWEJ NA WŁAŚCIWOŚCI MATERIAŁÓW CEMENTOWYCH Z DODATKAMI MINERALNYMI

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### Abstract

*The paper presents results regarding the possibility and effectiveness of carrying out the internal hydrophobization in cement-based materials with mineral additives such as granulated blast furnace slag, silica dust and silica fly ash. The obtained results indicate that effective internal hydrophobization by using triethoxyoctylsilane is achievable and provides protection against water by decreasing the capillary absorption of water in the material. However, it also affects the hydration process of the binder, which results in a reduction in the compressive strength of the material.*

**Keywords:** internal hydrophobization, triethoxyoctylsilane, granulated blast furnace slag, silica fly ash, silica fume

### Streszczenie

*W pracy przedstawiono wyniki dotyczące możliwości przeprowadzenia i skuteczności procesu hydrofobizacji objętościowej w materiałach cementowych z dodatkami mineralnymi, takimi jak: granulowany żużel wielkopiecowy, pył krzemionkowy oraz lotny popiół krzemionkowy. Otrzymane wyniki wskazują, że efektywna hydrofobizacja w masie wykonana przy pomocy trietoksyoktylosilanu jest osiągalna i zapewnia ochronę przed działaniem wody w postaci ograniczenia absorpcji kapilarnej w materiale. Jednakże wiąże się ona również z wpływem na proces hydratacji spoiwa, co skutkuje obniżeniem wytrzymałości na ściskanie materiału.*

**Słowa kluczowe:** hydrofobizacja objętościowa, trietoksyoktylosilan, granulowany żużel wielkopiecowy, mikrokrzemionka, popiół lotny

### 1. INTRODUCTION

The use of mineral additives in cement-based building materials, such as granulated blast furnace slag (ŻW), silica fly ash (PK) and silica fume (MK), can ensure many benefits and become more and more popular. Mentioned additives can improve the rheological properties of mixtures, prevent segregation of components and improve workability. Their use can provide develop in strength, impermeability and

corrosion resistance of the hardened material. The use of mineral additives allows for partial replacement of Portland cement CEM I, the production of which is associated with the emission of large amounts of carbon dioxide. As a result, the carbon footprint of the cement-based materials (composites) might be reduced and the negative impact on the environment is limited [1, 2]. Increasing the durability of cementitious building materials can also reduce their negative

impact on the environment. Water and moisture are one of the factors causing degradation and destruction of porous building materials. One of the methods of protection them from water and improving their durability is hydrophobization. This is a process by which material acquires hydrophobic properties, i.e. is difficult to wet by water. Hydrophobic surfaces are characterized by a wetting angle above  $90^\circ$ . In case of building materials, the process of hydrophobization might be implemented on the surface or in the whole volume of the material, which is called internal hydrophobization. Surface hydrophobization consists in applying a hydrophobic agent only to the surface of the material. Hydrophobic properties are obtained only by the surface layer, due to the limited depth of penetration of the hydrophobic agents [3]. Internal hydrophobization ensures hydrophobicity of the material in its entire volume. It is performed by adding appropriate agents (usually in the form of admixtures) during the production of the material together with the batch water or in the end of mixing. Organosilicon compounds such as silicone resins, siloxanes or silanes can be used for hydrophobization [3]. In particular the latter might provide a highly effective protection against water in cementitious materials, as shown, for example, in [4-7]. There are only few papers in the scientific literature considering the internal hydrophobization of cementitious materials with mineral additives, performed by organosilicon compounds. The presented paper concerns the issue of possibilities and difficulties in performing internal hydrophobization by using triethoxyoctylsilane in cementitious materials with the addition of ground, granulated blast furnace slag, silica fume and silica fly ash. The paper presents results concerning the influence of the organosilicon hydrophobic admixture on the hydration of selected binders. The impact on the strength of the tested material after 2, 28, 56 and 90 days of curing was also determined. The effectiveness of the internal hydrophobization was examined by the capillary water absorption test.

## 2. MATERIALS AND METHODS

The ordinary Portland cement CEM I 42.5R with the following additives were used: granulated, ground blast furnace slag (ŻW), silica fume (MK) and silica fly ash (PK). Cement pastes and mortars with a water-binder ratio equal to 0.5 were prepared. River sand of fraction 0/2 mm was used. In pastes and mortars with the addition of granulated blast furnace slag, 50% of

the ordinary Portland cement was replaced by the above-mentioned mineral additive in accordance with the recommendations of the PN-EN 15167-1 standard [8]. In the case of silica fume (microsilica) 10% of the ordinary Portland cement was replaced according to the PN-EN 13263-1 standard [9]. In the material with silica fly ash the share of the additive was 25% in accordance with the recommendations of the PN-EN 450-1 standard [10]. Cement with the appropriate mineral additive was combined before putting into the batch water and mixed manually until the binders combined. Mortars were prepared in accordance with the PN-EN 196-1 standard [11]. In order to determine and analyze the possibility and effectiveness of internal hydrophobization by means of organosilicon compounds, a silicon-based hydrophobic admixture (DH) was used, the main component of which was triethoxyoctylsilane (Fig. 1). The properties and principles of use of the admixture are presented in Table 1. The dosage of 0.8% of the binder mass is recommended by the product manufacturer. The admixture was dosed to the mixing water. The applied water-binder ratio (0.5) allowed for the preparation of materials without the addition of a plasticizer.

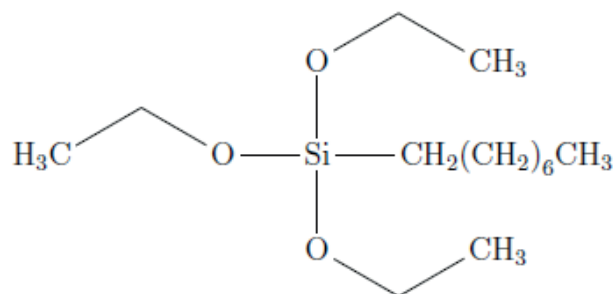


Fig. 1. Schematic chemical structure of triethoxyoctylsilane

Table 1. Principles of use and properties of a hydrophobic admixture

Main component	Dosage (%bm)	Solvent	Type of dispersion	Dosing method
Triethoxyoctylsilane	0.8%	Water	Emulsion	Batch water

Reference (REF\_ŻW, REF\_MK, REF\_PK) and internal hydrophobic samples (DH\_ŻW, DH\_MK, DH\_PK) were prepared. In order to determine the effect of the hydrophobic admixture on the binders hydration, a calorimetric tests were carried out in the TAM Air isothermal calorimeter at  $20^\circ\text{C}$  for 7 days according to the ASTM C 1679-08 standard

[12]. The test of capillary water absorption in the mortars was carried out in accordance with the PN-EN 1015-18 standard [13]. The compressive strength of hardened mortars after 2, 28, 56 and 90 days of curing was carried out according to the PN-EN 1015-11 standard [14]. Mortars samples with dimensions of 40 mm × 40 mm × 160 mm were demolded after 24 hours and stored in water for the first 14 days after demolding, and then in air-dry conditions ( $T = 20^{\circ}\text{C} \pm 2^{\circ}\text{C}$ ,  $\text{RH} = 55\% \pm 5\%$ ) until the test was performed. The implemented method of sample conditioning was aimed for activating the hydrophobic admixture [15]. Compressive strength after 2 days of mortars curing was determined on moist samples.

### 3. RESULTS AND DISCUSSION

#### 3.1. Isothermal calorimetry

The influence of the used hydrophobic organosilicon admixture on the hydration of the cement with mineral additives was determined by isothermal calorimetry. The rate of heat release is presented in Figure 2. Two samples of each pastes were tested. In general organosilicon compounds affect the hydration of binders by slowing down their hydration [16-19]. Triethoxyoctylsilane attaches to the surface of binder grains, thus hindering the access of water to the binder. Moreover, the organic, hydrophobic part of the compound, i.e. the octyl group, also repels water molecules. Organosilicon compounds might create a steric hindrance around the binder grains [2, 20], which is why they affect the amount and rate of heat released during the hydration process. The addition of a hydrophobic admixture to the mixing water caused changes in the heat release rate regardless of the used mineral additive, as shown in Figure 2. The silane-based admixture also caused a decrease in the amount of heat released after 7 days of testing. In the pastes with the addition of granulated blast furnace slag, the amount of heat released in the reference sample REF\_ŻW was 180.6 J/g. In the internal hydrophobic sample, it was reduced by 4.1% (to 173.3 J/g for DH\_ŻW). In the pastes with the addition of microsilica, the amount of heat released in the reference sample REF\_MK was 229.0 J/g. In the internal hydrophobic sample, it was reduced by 5.2% (to 225.4 J/g for DH\_MK). In the pastes with the addition of silica fly ash, the amount of heat released in the reference sample REF\_PK was 270.4 J/g. In the internal hydrophobic sample it was reduced by 1.6% (to 256.3 J/g for DH\_PK).

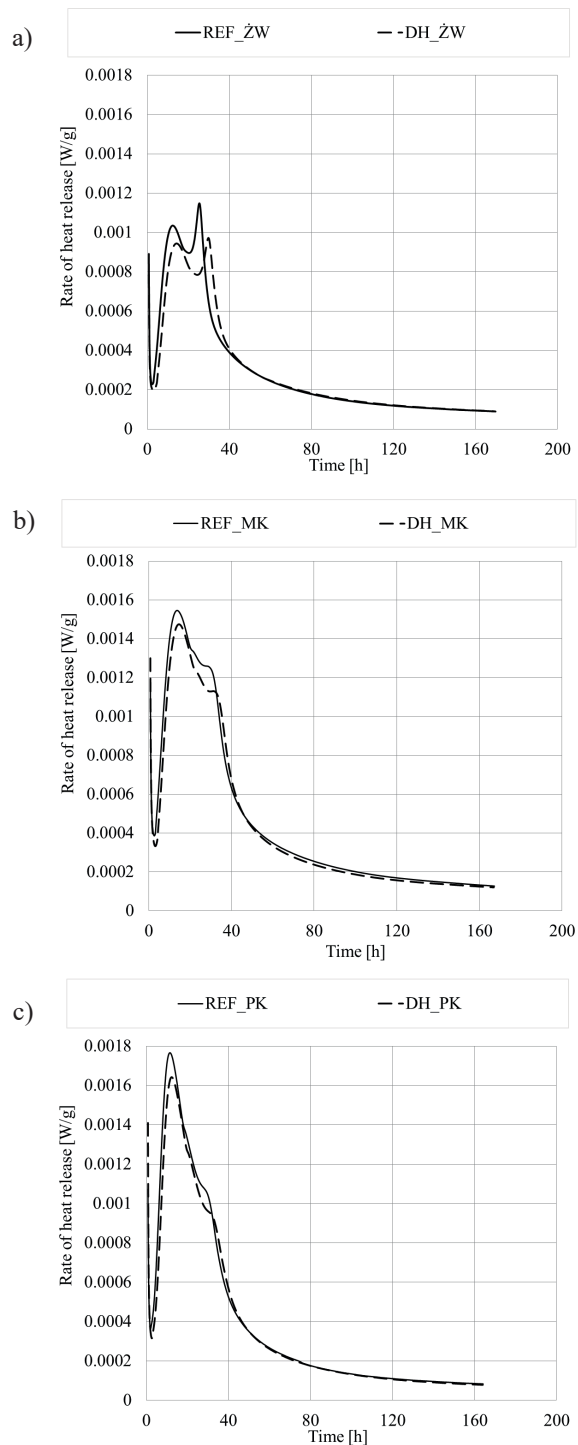


Fig. 2. The rate of heat release in pastes with a) granulated blast furnace slag, b) silica fume, and c) silica fly ash

#### 3.2. Capillary water absorption

The capillary water absorption test for mortars was carried out on 6 samples with dimensions of 40 mm × 40 mm × 80 mm. The side walls of the samples were covered by a silicone sealant to prevent water evaporation. The mortars were placed in water to

a depth of about 5 mm. The changes in the mass of the samples was measured after 10, 20, 30, 40, 50, 60, 90 minutes and 2, 3, 4, 5, 6 and 24 hours. The mass changes of the mortars are shown in Figure 3. The use of the silane-based hydrophobic admixture in the amount of 0.8% bm. ensured an effective reduction in water absorption in the mortars and a decrease in the capillary absorption coefficients. In case of mortar with the addition of granulated blast furnace slag, the capillary absorption coefficient of the reference sample (REF\_ŻW) was  $0.1063 \text{ kg/m}^2 \times \text{min}^{0.5}$ , and for the internal hydrophobic sample (DH\_ŻW) was reduced to  $0.0195 \text{ kg/m}^2 \times \text{min}^{0.5}$ , (by 81.6%). For mortar with the addition of microsilia, the capillary absorption coefficient was reduced by 67.0% (from  $0.0633 \text{ kg/m}^2 \times \text{min}^{0.5}$ , for REF\_MK to  $0.0208 \text{ kg/m}^2 \times \text{min}^{0.5}$ , for DH\_MK). In the reference mortar with the addition of silica fly ash (REF\_PK) the capillary absorption coefficient was  $0.0872 \text{ kg/m}^2 \times \text{min}^{0.5}$ , and it was reduced to  $0.0278 \text{ kg/m}^2 \times \text{min}^{0.5}$  (by 68.0%) for the hydrophobic sample DH\_PK.

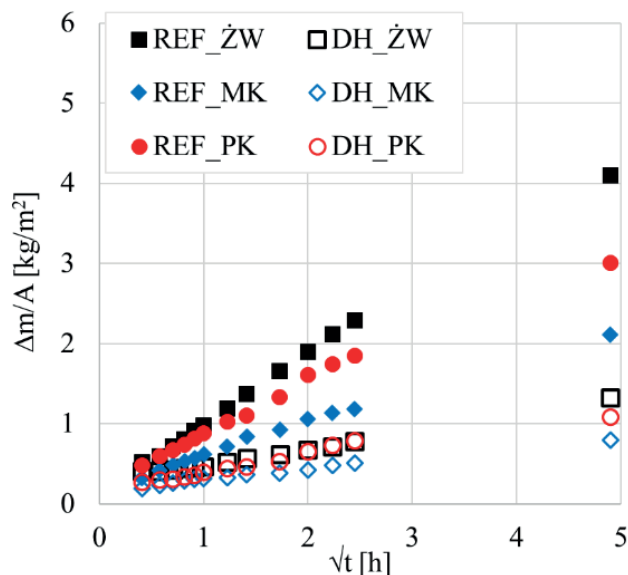


Fig. 3. Mass changes in mortars during capillary water absorption test

### 3.3. Compressive strength

Table 2 presents the results of compressive strength of mortars after 2, 28, 56 and 90 days of curing. Six samples of each type of mortar were tested. The use of the organosilicon hydrophobic admixture resulted in a decrease in compressive strength. For internal hydrophobic mortar with the addition of granulated blast furnace slag (DH\_ŻW), the strength was reduced by 7.0% after 2 days, by 4.3% after 28 days, by 9.6%

after 56 days and by 9.2% after 90 days of curing in relation to the reference samples (REF\_ŻW). In the case of hydrophobic mortar with the addition of silica fume (DH\_MK), the compressive strength was reduced by 25.7% after 2 days, by 27.1% after 28 days, by 26.6% after 56 days and by 25.0% after 90 days of curing in relation to the reference samples (REF\_MK). In internal hydrophobic mortar with silica fly ash added (DH\_PK), a decrease in strength by 5.2% after 2 days, by 23.9% after 28 days, by 8.5% after 56 days and by 10.7% after 90 days of curing was observed in relation to the reference samples (REF\_PK).

Table 2. Compressive strength of mortars after 2, 28, 56 and 90 days of curing

Sample/Test time	Compressive strength [MPa]			
	2 days	28 days	56 days	90 days
REF_ŻW	5.5	39.7	42.6	45.6
DH_ŻW	5.1	38.0	38.5	41.4
REF_MK	16.1	63.2	64.8	72.5
DH_MK	12.0	46.0	47.6	54.4
REF_PK	10.5	45.0	45.5	45.8
DH_PK	10.0	34.2	41.6	40.9

### 4. CONCLUSIONS

The presented paper investigated and analyzed the effectiveness of the internal hydrophobization and its impact on the basic properties of porous cementitious materials with mineral additives such as: granulated blast furnace slag, microsilia and silica fly ash. The obtained results indicate that internal hydrophobization of cementitious materials with mentioned mineral additives by means of triethoxyoctylsilane is possible and provides effective protection against water penetration into the material. However, dosing of the organosilicon hydrophobic admixture together with the mixing water affects the binder hydration process, which is visible as a reduction in the amount and rate of heat release. As a consequence, a decrease in the strength of the internal hydrophobic mortars is also observed even after 90 days of curing. The obtained results indicate that internal hydrophobization of cementitious materials with mineral additives is possible and effective, although this topic requires further research.

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