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Hydrophobisation of mortars containing waste polyurethane foam

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Abstract. This paper investigates the physical and mechanical properties of mortars modified with waste polyurethane foam (WPF) and the influence of hydrophobising agents on the physical characteristics. The sand was substituted with WPF (2-4 mm grain size) in the mortars which consisted of 5, 10 and 15% of foam, respectively. The contact angle of lightweight mortars was determined (θ_w) in the function of time, prior to and after the frost resistance test. The surface free energy characterizing the wettability and adhesion of mortars under normal conditions and following frost erosion was calculated with Neumann's method based on the obtained data. The structure of mortars, adhesion of lightweight aggregate to cement paste and the structure of thin hydrophobic film were demonstrated by means of scanning electron microscopy. The mortars subjected to hydrophobisation process revealed a slight mass change caused by freezing and thawing processes: 0.1% for methyl silicone resin, 2.1% for alkyl-alkoxy-silane, and 9.2% for the samples which were not hydrophobized. On average, the contact angle of the standard mortars was 3 times lower than the one of hydrophobic material. The best results illustrating the efficiency of hydrophobisation were obtained for methyl silicone resin.

1 Introduction

Due to a growing problem of construction waste utilization, engineers are seeking possibilities of their management. One of them is recycling of energy saving waste in construction materials. A proof of the effectiveness of this solution is more common application of heat insulating mortars with the use of waste [1-3]. An example of artificial aggregate achieved from material recycling can be waste polyurethane foam (WPF) used in lightweight mortars [4-6] and lightweight concrete [7,8]. Materials containing this type of lightweight aggregate have smaller mass and strength but bigger absorbability and thermal insulation [4-8]. The application of waste polyurethane foam in heat insulating mortars considerably reduces heat transmission coefficient by the mortar. However, waste polyurethane foam can be a reason for increased wettability of mortars, hence after they have been prepared, a surface hydrophobisation should be considered. The purpose of our further research [3, 9-11] was the evaluation of the possibility

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of applying hydrophobising preparations based on silica-organic compounds in the protection of lightweight mortars and concrete with construction waste, as well as water and wastewater management.

The authors would like to call attention not only to the problem of construction waste recycling, including polyurethane foam (WPF), but also to their low resistance to water and frost. The purpose of these studies is to determine physical reference properties of hydrophobised insulating mortars containing recycled foam, because the literature lacks information on this subject. Owing to its unique, pitted structure after having been grated, the foam has good sorption properties and can cumulate the sufficient amounts of ice and salt. The authors have confirmed in their research that the application of different hydrophobising agents causes a significant reduction of wettability and the increase of frost resistance of lightweight mortars.

2 Materials and methods

2.1 Materials

The content of 3 mineral mixtures of lightweight mortars with WPF has been prepared. In the mortars the sand was partially substituted with the foam of 0-4 mm granulation. WPF came from waste obtained during the construction of a single family house. The waste was grated in a way to achieve grains of size not bigger than 4 mm. The samples were marked as follows: M5, M10, M15 – mortars containing 5, 10, 15 % WPF respectively. Table 1 presents the percentage content of mortars with addition of utilised foam in proportion by capacity.

Table 1. Composition of mortars with polyurethane foam, proportion by capacity, vol. %.

Components	**M5**	**M10**	**M15**
Portland cement CEM I 32.5 R	25	25	25
Polyurethane foam 0/4 mm	5	10	15
Quartz sand 0/4 mm	15	10	5
Hydrated lime	25	25	25
Water	30	30	30
Vinyl acetate copolymer	0.15	0.15	0.15

The hydrated lime parameters were in line with the requirements of PN-EN 459-1:2015-06 [26] and the value of its apparent density was 390 kg·m^{-3}. The composition of the lime, in terms of oxide, was as follows: CaO – 95.5%, CO_2 – 2.1%, MgO – 0.5%, SO_3 – 0.1%, free water – 1.5%. The Portland cement CEM I 32.5R was developed according to the Polish standards PN EN 197-1:2012 [27] and PN-B-19707:2013-10 [28]. CEM I 32.5R that was used had the following parameters: a specific surface of 3985 cm^2·g^{-1}, density of 3.05 g·cm^3, the initial setting time of 248 min, compressive strength after 2 days – 17.6 MPa and after 28 days – 43.2 MPa, loss on ignition of 5.0% by weight cement, volume stability < 10 mm.

It should be pointed out that the foam is a very compressible material in comparison to other components of mortar such as cement, lime, water and sand. Its density can thus vary depending on exerted pressure. Absorptivity (vol. %) of WPF was determined after it has been immersed in water for 24 hours. Compressive strength on three cubic rigid WPF samples (50x50x50) was measured on a Controls 50 kN universal testing machine at 20 °C. The test was conducted to deformation of 10% and the speed of crosshead movement was 5 mm/min. The composition of WPF was determined based on EDS. Table 2 presents the main physical properties of WPS on the basis of authors' research.

Table 2. Composition and properties of waste polyurethane foam.

Density (kg/m^3)	Absorp-tivity (%)	Total porosity (%)	Compressive strength (MPa)	C	O	N	H	Ca	others
				weight (%)					
22	15.1	95	0.170	64.55	20	7.2	6.4	1.0	1.03

The following hydrophobic agents were used in the research:

- The methyl silicone resin i.e. undiluted bicarbonate agent which can be used to hydrophobize porous materials and cement-lime plasters. The parameters of this agent are as follows: viscosity of 2.846 $Pa{\cdot}s{\cdot}10^{-3}$, density of 0.82 g/cm^3, surface tension of 24.30 $N/m{\cdot}10^{-3}$ and the active ingredient concentration is 11% [3,15].
- Micromolecular alkyl-alkoxy-silane with aliphatic hydrocarbons as carriers, $R_n{-}Si{-}(OR')_{4-n}$, where R_n is alkyl-alkoxy-silane derivative which was used as a hydrophobising preparation for porous building materials of mineral origin, sand-lime bricks, mineral renders. It has the following parameters: the viscosity of 1.79 $Pa{\cdot}s{\cdot}10^{-3}$ at 20˚C, the density of 0.80 g/cm^3 and siloxane content reaching approximately 7 % by mass [3, 15].

2.2 Methods

The porosity, density and bulk density were determined in line with EN 1015-10:2001 [16] standard. The absorptivity of mortars was investigated after 1 and 14 days, according to BS 1881-122, 2011 standard [17]. Before the testing, six samples of 40×40×160 mm were prepared and dried. In order to determine the thermal conductivity coefficient λ, plate apparatus FOX 314, TA Instruments, New Castle, USA with 300×300×50 mm plates was used. Three plates were prepared for each mortar. The samples were subjected to drying at the temperature of 20°C and 0°C for a heating and cooling plate, respectively, in order to obtain a constant weight. Water vapour permeability of mortars was determined on the basis of EN ISO 7783:2012 regulation [18]. Cylindrical samples 2 cm in thickness and 10 cm diameter were made of mortars. Three layers of hydrophobising preparation were applied with a brush on the surface of one of the samples. The measurements were carried out three times in constant temperature of 23°C and relative humidity, 93% on one side of the sample and 50% on the opposite side. The duration of the test was 8 – 9 days depending on diffusion through the surface of the material. The analysis of compressive strength was conducted in accordance with EN 1015-11:2001 standard [19], following sample curing for the duration of 28 days. The direct method, established in EN 12012:2007 [20] standards was implemented in the calculation of frost resistance. The dimensions of the utilized samples were 40×40×160 mm. All samples were subject to 50 cycles of freezing and thawing processes. However, the resistance has not been determined after the frost resistance test, but the samples were dried to constant mass, and mass loss was determined. The surface free energy (SFE) was established by measuring the contact angle of the considered mortars [3, 9, 11]. For that purpose, a test stand consisting of a goniometer and a camera for capturing the images of 2 mm^3 drops placed on the sample surface with a micropipette [3, 9] was used [15]. Adopting the Neumann model required the application of distilled water. The test involved placing 6 drops on each sample. The contact angle was analyzed before and after the frost resistance test. The SFE was calculated with the Neumann model using the following equation [15, 21]:

$$\cos\theta_w = \left[e^{-0.000125(\gamma_s-\gamma_w)^2} 2\sqrt{\frac{\gamma_s}{\gamma_w}} - 1 \right] \tag{1}$$

where: γ_w – SFE of water, θ_w – water contact angle. The value γ_w = 72.8 mJ/m^2 was assumed for distilled water [9, 15].

To determine the structure of mortars and the interfacial transition zone between the paste and aggregates, a scanning electron microscopy SEM was used. Observations were conducted by means of Quanta FEG 250 microscope, by FEI, Hillsboro, USA.

The surface hydrophobisation was performed by applying the preparation with a brush for three times. Next, all the samples were seasoned for the period of 7 days in laboratory conditions in order to start hydrolytic polycondensation.

The considered samples included:
M5, M10, M15 – control mortars without a hydrophobising layer;
M5.R, M10.R, M15.R - mortars with a methyl silicone resin solution;
M5.A M10.A, M15.A - mortars with alkyl-alkoxy-siloxane.

3 Results and discussion

The mechanical and physical properties of the considered heat-insulating mortars without a hydrophobising agent surface are shown in Table 3.

Table 3. Properties of heat-insulating mortars with WPF.

Descriptive statistics	Apparent density (kg/m^3)	Density (kg/m^3)	Open porosity (%)	Total porosity (%)	Thermal conductivity (W/m^{2}K)	Compressive strength (MPa)
M5						
Mean	1.47	2.41	25.1	38.07	0.441	19.4
SD	0.2	0.23	0.24	0.5	0.09	0.9
M10						
Mean	1.41	2.39	28.02	39.0	0.395	17.3
SD	0.1	0.3	0.75	0.75	0.05	0.41
M15						
Mean	1.38	2.38	30.1	41.1	0.302	16.7
SD	0.32	0.87	1.32	1.54	0.02	0.95

*Standard deviation

The achieved properties are characteristic of lightweight mortars. The value of heat transfer coefficient λ enables to classify the mortar as a heat-insulating one. This mortar demonstrates high porosity (38.07-41.1%), which is related to the increase of absorptivity in relation to the traditional mortars. However, despite high porosity and low density, good compression strength (16.7-19.4MPa) was also obtained. The increased quantity by 10 % of WPF addition causes the reduction of bulk density M15 by 6 % in comparison to M5. This amount of WPF addition in M15 has also resulted in the abatement of its compression strength by 14% and thermal conductivity coefficient by 31.5%, which raises its insulating properties in relation to M5. This is connected with the increase of open porosity by about 17%. Numerous studies have demonstrated that the application of lightweight aggregate often caused a substantial decline of mechanical properties of materials [2, 6, 9, 22]. However, in the analysed mortars this decline was not considerable as an admixture of vinyl acetate copolymer was applied. Polymers are commonly used in the modification of mortars and concrete due to their significant role in general enhancement of the efficiency of cement based materials, which was described by Wang et. al [23] and Wiliński et. al [24].

The results of research on the effectiveness of hydrophobisation of mortars with WPF in terms of absorptivity, contact angle, water vapour permeability and frost resistance has been presented in Table 4.

Table 4. Influence of hydrophobisation on the properties of heat-insulating mortars.

Mortars	Absorptivity 1 day (%)	Absorptivity 14 days (%)	Water vapour permeability (10^{-12} kg/m·s·Pa)	Contact angle (°)	Contact angle after frost test (°)	Mass loss of samples (%)
M5	4.4	24.1	5.7	27.5	17.6	6.2
M5.R	0.1	10.3	2.3	102.5	96.1	1.0
M5.A	0.4	15.7	4.3	100.3	82.0	1.2
M10	6.6	27.3	9.6	21.7	15.7	10.0
M10.R	0.1	11.1	3.0	91.5	69.6	2.2
M10.A	0.3	16.4	4.1	87.2	61.3	3.4
M15	8.1	29.7	12.3	15.3	7.3	14.7
M15.R	0.2	15.4	5.2	90.1	60.5	2.6
M15.A	0.5	18.2	7.3	80.1	54.9	3.8

The value of absorptivity of reference mortar after 1 day ranged between 4.4 and 8.1%. After hydrophobisation, absorptivity was at the level of merely 0.1 and 0.5%. However, after 14 days of the test, the differences between the reference samples and hydrophobised ones decreased considerably and amounted to 29.7% for M15 and 24.1% for M5. Another parameter that confirms high wettability of the material is contact angle before hydrophobisation which was 3.7, 4.2 and 5.9 times higher for M5, M10 and M15 mortars, respectively. This indicates reduced adhesive properties and hydrophobicity of porous mortar surface. Silica-organic compounds in aliphatic hydrocarbons modified the surface properties of the mortars with WPF. The decrease of water vapour permeability of mortars with WPF after the hydrophobisation has been observed, which especially in the case of methyl-silicon resin (R) reduces the material ability to diffusion cumulated inside the moisture even from 58% to 69% for M15 and M10, respectively. However, mortars with lightweight aggregate have higher permeability than the ones made of normal materials, which was presented in the papers of Sugiyama et al. [25], (1.7–1.9 times higher). This increase of permeability in reference mortars can be caused by the fact that WPF is a highly porous material meaning it has relatively high diameter of capillaries, which makes the water vapour flow easier [26]. Additionally, the foam is highly netted which means the presence of ducts or capillaries that are placed in the whole mortar and by connecting with each other they make a thin net enabling vapour to easily flow through.

Despite the decrease of water vapour permeability, agent R was the best at protecting the surface of mortars from humidity and frost corrosion. The frost resistance of the mortars can be formulated with only moderate confidence level, because the analysis only involved the mass loss of samples following the frost resistance test and the changes in SFE. No studies reflecting the changes in compression strength after freezing and thawing cycles were conducted. However, on the basis of the performed research, the authors obtained preliminary, but sufficient information on the changes in wettability and frost resistance of the material.

Methyl-silicon resin (R) increased the frost resistance by 77-84%, while alkyl-alkoxy-siloxane (A) raised it by 66-80% in relation to the reference samples. The layer of polisiloxane resin protected the surface against the infiltration of water, and consequently, against the destructiveness of ice [3,9-11].

When the surface of the mortar has to be resistant to corrosive environment, the preparation with the lowest possible content of SFE should be used. Figure 1 shows the alteration of SFE due to hydrophobisation of mortars, as well as SFE before and after the frost resistance test.

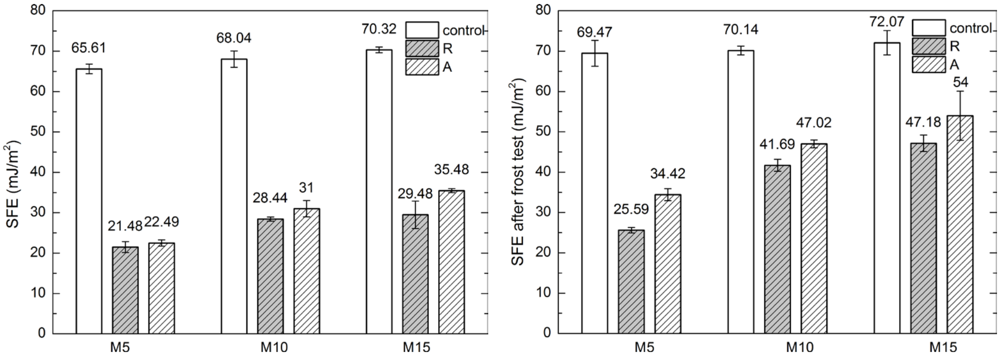

Fig. 1. SFE of mortars with WPF before and after the frost resistance test.

The M5.R mortar after hydrophobisation is characterized by the lowest SFE value which is about 3 times lower than the value of the referential mortar. Other mortars have been effectively protected to a similar extent (1.7-2.2 times). Lower SFE results in lower adhesive properties, higher resistance of the material to the infiltration of water and corrosive compounds, as well as higher resistance to freezing and thawing cycles, as evident from frost resistance tests. Calculations of SFE after frost resistance tests demonstrated a decrease in the efficiency of hydrophobisation. The contact angle of reference mortars following 50 cycles of freezing and thawing was smaller by 36% and 52% for M5 and M15 mortars, respectively. SFE raised by about 16% for M5-15.R and 35% for M5-15, respectively, which signifies weaker properties of the layer of polisiloxane resin.

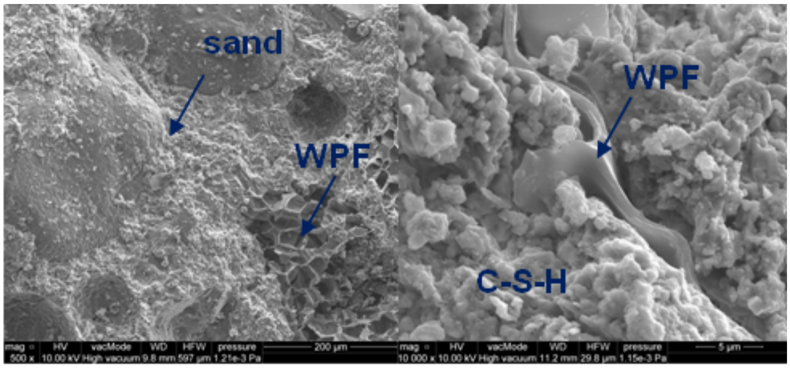

Fig. 2. Structure of M15 mortar: (a) 500x, (b) 10000x.

The macromolecular resin (R) hermetically covered the structure of mortars with WPF resulting in a slightly better efficiency of hydrophobisation than the agent A with smaller molecules. In the case of absorbent, more absorptive mortars with bigger amount of WPF, the number of preparation layers that are applied with a brush should be increased or a low-pressured spray should be applied in order to enhance the effectiveness of the agent A with smaller molecules.

Figure 2 presents the structure of M15 mortar with the highest content of WPF. In reality, mechanical properties can be explained with the presence of WPF, as well as the quality of interfacial zone between cement paste and aggregate, which significantly affects its strength. Cement paste is dense while WPF aggregate is highly porous (Fig. 2a). A very good adhesion of an aggregate grain to coarse pellets of grated WPF has been noticed (Fig. 2a,b). In the contact zone there are no cracks or scratches. Figure 3b shows crystallized C-S-H phase.

All of these properties are the reason for a very good strength of cement paste, which has been confirmed by the analysis of mechanical properties. On the other hand, it has been observed that interfacial transition zone is slightly higher in the case of recycled aggregates than in the case of sand.

This is related to high water absorption by porous WPF aggregate, which has also been described by Rodríguez et al.[27] in his research.

4 Conclusions

The following main conclusions can be drawn:

- The increase of the amount of WPF has led to about 16% higher porosity of cement paste which makes the exchange of humidity with the ambient environment easier, water vapour permeability (up to 54%) and reduces thermal conductivity coefficient by about 35%.
- The increase of the amount of foam by 10% caused a significant increase in weight loss after freezing and thawing cycles by as much as 51% while the reduction of compressive strength was 16%.
- Contact angle after hydrophobisation was 3.7-5.9 times lower than for control mortars, which proves the effectiveness of hydrophobisation.
- Hydrophobisation by methyl-silicon resin (R) increased frost protection by 77-84%, while hydrophobisation by alkyl-alkoxy-siloxane (A) raised it by 66-80% in relation to the reference samples.
- The weight loss after freezing and thawing cycles is closely dependent on water absorptivity and hydrophobisation of the surface explicated by means of SFE.
- The highest hydrophobisation efficiency of lightweight mortars with WPF was obtained by applying methyl silicone resin in an organic solvent.

These results reinforce the idea of using waste polyurethane foam in the production of lightweight insulating aggregates with the application of hydrophobisation as an effective protection against humidity and frost.

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