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Methods of introduction of glyoxal-containing additives into foam concrete mixture

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Abstract. The present paper investigates a cement based foam concrete with glyoxal-containing additives which are introduced into the mixture in different ways. The relevance of the given study is conditioned by the necessity to provide the required quality parameters of mixture for transportation and laying the formwork, as well as providing strength and physical characteristics of wall structures for housing construction. Four ways of introducing additives into the foam concrete mixture have been proposed: spraying with 40% glyoxal aqueous solution; joint grinding of sand with crystalline glyoxal in the amount of 0.01% by weight of cement; introduction of crystalline glyoxal into the foam concrete mixture; introduction of a 40% aqueous solution of glyoxal to the foam concrete mixture. The study was conducted in the Laboratory of Tomsk State University of Architecture and Building which is accredited in accordance with the national standards requirements. Glyoxal-containing additives applied within the technology of cement foam concrete of natural hardening contributes to ensuring increased aggregative stability of mixtures and compressive strength at the age of 28 days. The elaborated composition and technology of foam concrete production with glyoxal-containing additives is intended for use in the walls structures as well in monolithic construction as in pre-cast production.

1. Introduction

In accordance with the concept of Russia's development up to 2020, great attention is paid to building materials based on local raw materials with the required performance characteristics, low energy intensity and cost. Such materials include porous wall materials based on cement binders. Porous concrete (foam concrete) can be effectively used for the manufacture of wall blocks, constructing monolithic wall structures in low-rise construction, mainly individual housing. The construction of low-rise housing with the use of foam concrete in enclosing structures is a favorable object for small businesses.

Great attention is paid to reducing thermal conductivity and improving the strength, uniformity of quality parameters, and the rational and economical use of raw materials in the production of foam concrete of natural hardening. Effective modifying additives for binders and concretes [1-3] are widely used for managing the quality of cement based concretes. In the Russian building sector mainly used imported additives. Therefore, the development and use of domestic modifying additives is relevant. This is possible with the development of innovative technologies in the chemical industry. The Tomsk region (Russia) developed a technology for the synthesis of oxalic dialdehyde, glyoxal, which has a

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number of unique properties, which has a number of unique properties that can be effectively used in the pharmaceutical industry, construction, refining, textile, leather, paint and varnish, glue and metallurgical industries, etc.

Glyoxal can be attributed to surface-active substances. The surface activity value of 0.045 N•m^2/kg is provided by the concentration of glyoxal in an aqueous solution of 10 g/l. The dependence of surface activity on the concentration of glyoxal in solution is close to logarithmic. The maximum value of surface activity is observed at low concentrations. The manifestation of the surface-active properties of glyoxal in aqueous solution allows us to predict the plasticizing effect when it is use as additive to composite cement based building materials.

The pH value of the cement paste reaches 12.5 after cement interacts with water. Conditions are created for the Cannizzaro reaction to proceed after glyoxal is introduced into cement paste. Assuming the pH of the cement paste is 12, the introduction of glyoxal into the cement paste leads to the Cannizzaro reaction (Figure 1). Presumably, the interaction of glyoxal with calcium hydroxide (the product of the hydrolysis of alita in the cement paste), as a result of the Cannizzaro reaction, calcium glycolate is formed [4-7]. An increased heat release is observed in the cement paste after adding water to the mixture. The cement paste expands due to the intensive increase in the volume of newly formations (HO CH_2-$COO)_2Ca$ during the cement hydration. In addition, it was found that the resulting calcium glycolate has great strength.

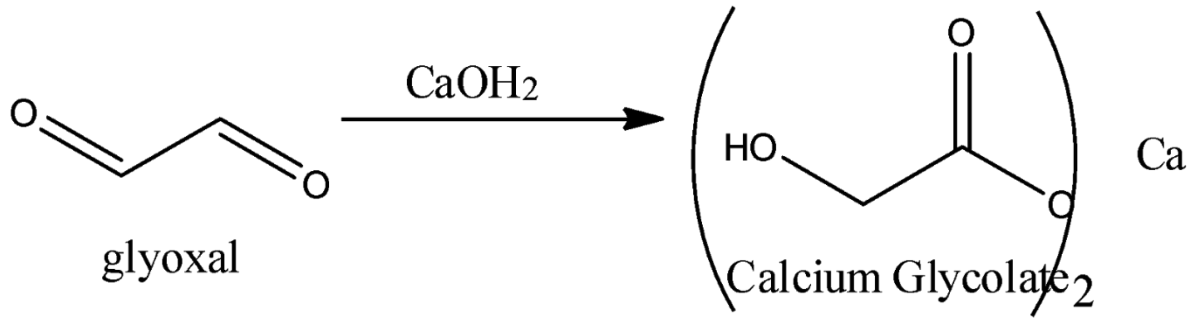

Figure 1. Cannizzaro reaction

The heme-diol and dimeric forms of glyoxal remain for some time resistant to the alkaline environment of concrete, since aldehyde groups are involved in the Cannizzaro disproportionation of aldehydes, and not heme-diol groups, it takes time to shift the balance to the aldehyde form. Since water is weakly bound to heme-diol forms, the amount of water decreases during the hydration process, and it starts to be released from these complexes. This provides prerequisites for the scientific substantiation of the choice of glyoxal as a modifying additive in the production of building materials. The ability to plasticize, retain water and give it over time into the volume of the composition allows the forming of a reserve of structure formation in cement compositions [8,9]. Glyoxal is capable of forming structures cross-linked with silicates during the hardening process of the hardened cement paste.

The above properties of glyoxal in cement systems allow us to recommend it to control the structure formation of cement based building compositions of various functional purposes. The use of glyoxal in the technology of building materials has recently been investigated by A.S. Knyazev, V.V. Strokova, Yu.S. Sarkisov, N.P. Gorlenko, A.I. Kudyakov. Based on the results of the analysis of security documents and published articles about the use of glyoxal in the building sector, the development of research and the acquisition of new knowledge in order to develop new building materials using Russian-made glyoxal is relevant.

2. Materials and methods

In carrying out experimental research were used raw materials that meet the requirements of national standards of Russia. As binders Portland cement of Topki Cement Factory (Kemerovo Region, Russia)

CEM I 42.5H (Russian State Standard GOST 30515-2013) was used, sand of Kudrovskoe deposit of Tomsk Region (Russia) with fineness modulus 1.86 (Russian State Standards GOST 8736-2014 and GOST 26633-2012), water (Russian State Standard GOST 23732-2011) and foam agent PB-2000.

A 40% aqueous solution of glyoxal and crystalline glyoxal (TU 2633-003-67017122-2011), manufactured by TD Novokhim (Tomsk, Russia) were used as a structure formation accelerator for foam concrete mixtures. Technical characteristics of glyoxal-containing additives are presented in table 1.

Table 1. Technical characteristics of glyoxal-containing additives

Indicator	Crystalline glyoxal	40% aqueous solution of glyoxal
Appearance	white to yellowish beige powder	colorless / pale yellow liquid
Mass fraction of the main substance,%	80	40±1
The rate of activity of hydrogen ions H * (pH)	-	2-3.5
Density, g /cm^3	1.0	1.20-1.35
Mix	water, ethanol, methanol, isopropanol	ethyl alcohol, acetone
Volatility	-	low

Determination of compressive strength, shrinkage and average density of foam concrete was carried out in accordance with standard techniques, adapted to the requirements of European standards.

The selection of the composition of the foam concrete mixture was carried out taking into account the requirements of SP 277-80 “Instructions for the production of cellular concrete products”. Taking into account the results of optimization of technological modes of preparation of foam concrete mixture in a laboratory mixer, the basic composition of foam concrete was developed, which is given in table 2.

Table 2. Basic composition (control) of foam concrete D500 per 1 m^3

Cement, kg	Sand, kg	Water, kg	Foaming agent, l
270	135	202	1.3

When establishing the regularities of the influence of additives on the properties of the foam concrete mixture and concrete foam, additives were introduced into the basic composition of the foam concrete without changing the number of components and then adjusting the recommended composition according to the values of the actual average density of the concrete mix. The preparation of the foam concrete mixture was carried out using a single-stage technology with laboratory foam concrete mixer.

The prospects of mechanical and chemical methods of activation are substantiated according to the results of the analysis of various methods of activation of the components of foam concrete, which are aimed at improving the contact interaction of the hardened cement paste with the aggregate.

The positive effect is achieved by controlling the structure of the foam concrete. Improvement of the performance characteristics of foam concrete is possible through the development of new technological methods regulating the physicochemical phenomena at the contact of the interacting components of the hardened cement paste-aggregate section [10]. Regardless of their state of aggregation, all raw materials are usually in an inactive position and with complex processing may acquire chemical activity as a result of interaction at the interface. Activation of the components of the foam concrete mixture can significantly improve the basic technical properties of foam concrete. And the aggregate should be modified to obtain the greatest effect from the action of additives. It has been established that surface treatment of the aggregate with aqueous solutions of acids leads to a significant increase in the microhardness of the hardened cement paste on contact with the aggregate grains and to an increase in the strength of the samples. The process of contact formation begins with the moment of contacting the surface of the grains of the aggregate, mineral particles and binder with liquid phase of cement paste. By controlling the surface properties of the contacting particles, it is possible to influence the processes of structure formation, primarily in the zone of contact of cement and sand.

We have proposed four ways of introducing glyoxal-containing additives into the foam concrete mixture:

- spraying of 40% glyoxal aqueous solution onto the aggregate surface;
- joint grinding of sand with crystalline glyoxal in the amount of 0.01% by weight of cement;
- introduction of crystalline glyoxal into the foam concrete mixture in the amount of 0.01% by weight of cement;
- introduction of a 40% aqueous solution of glyoxal to the foam concrete mixture in the amount of 0.01% by weight of cement.

Spraying with 40% glyoxal aqueous solution

The ways of chemical activation of the aggregate are considered, which leads to the enhancement of their contact interaction with the products of hydration of binders. In conducting studies quartz sand was treated with 40% aqueous glyoxal solution by spraying with a spray gun. Next, the foam concrete mixture was prepared using a single-stage method in a laboratory foam concrete mixer. The finished foam concrete mixture was placed in metal forms 15x15x15 cm. The foam concrete samples were kept at standard conditions at a temperature of 20±2°C for 24 hours. Next, the samples were solid cubes in a natural hardening chamber, at a temperature of 20±2°C and a relative air humidity of at least 90% for 27 days prior to testing.

Introduction of glyoxal into the foam concrete mixture

Pre-dosed water with a foaming agent was added to the foam concrete mixer and mixed for 1 minute. Then sand and cement were loaded into the mixer and the mixture was mixed for 2 minutes until a homogeneous state was obtained. It is very important to distribute the cement well in the sand. Then, 40% aqueous solution of glyoxal was added to the mixture and all components were mixed for another 4.5 minutes. Crystalline glyoxal was introduced using the same technology.

Joint grinding of sand with crystalline glyoxal

Sand sieved on a sieve before grinding. The dosed components are loaded into a laboratory mill and subjected to joint grinding. Activation occurs due to abrasion – simultaneous deformation of compression and shear. Preparation of a foam concrete mixture with activated aggregate was carried out by a single-stage method.

3. Results and discussion

The results of studies of the influence of glyoxal-containing additives and the method of their introduction on the rheological properties of the foam concrete mixture and the physicmechanical properties of foam concrete are given in table 3.

Shrinkage deformations of foam concrete, which lead to deformations and cracking, are of great importance for foam concrete building technologies throughout the entire life cycle of building management. Plastic shrinkage occurs in freshly molded products. Plastic shrinkage is a consequence of the rapid loss of water from the surface of the concrete, leading to the formation of negative capillary pressure in the micropores.

It was established that the introduction of additives into the foam concrete mixture leads to reduce plastic shrinkage by 29-69% (2.86-1.24 mm/m). The obtained data confirm the results of a study on the effect of modifying additives on the process of early structure formation of foam concrete [11]. Glyoxal additive accelerates the process of early structure formation, which allows stabilizing the porous structure of the foam concrete mixture and thereby reducing shrinkage deformation of the foam concrete mixture. The aggregative stability of the foam concrete mixes increases from the beginning to the end of the cement paste setting [11].

Table 3. Properties of foam concrete mixtures and foam concrete

Properties	**The compositions**				
	Control	Crystalline glyoxal 0.01%	40% glyoxal aqueous solution 0.01%	Sand treatment with 40% glyoxal water solution	Co-grinding sands with crystal glyoxal
Plastic shrinkage, mm/m	4.02	2.86	2.56	2.20	1.24
Drying shrinkage, mm/m	3.00	1.75	2.8	1.63	1.37
Spread, cm	11	11.5	14	14.4	11.9
Compressive strength, MPa	0.70	0.83	0.70	1.28	1.50
Average density in dry condition, kg/m^3	455	478	490	480	480

According to the experimental results presented in table 3, it was found that the introduction of glyoxal-containing additives into the mixture leads to reduce drying shrinkage of natural hardening foam concrete by 7-54%. The introduction of glyoxal contributes to the reduction of chemically unbound water in foam concrete, as a result, a decrease in shrinkage is observed during the drying of foam concrete [1].

During natural hardening of foam concrete, a significant decrease in shrinkage deformations allows predicting a reduced level of stress formation during structure formation in the volume of foam concrete and, as a result, increased operational reliability of products (structures) made of this material [13]. Processing sand by spraying with a 40% aqueous solution of glyoxal leads to increase in the compressive strength of foam concrete by 30%. The introduction of crystalline glyoxal in the amount of 0.01% by weight of cement, leads to a slight increase in the average density of foam concrete, while the compressive strength increases by 56% (0.83 MPa).

Joint grinding of sand with crystalline glyoxal in the amount of 0.01% by weight of cement leads to enhancing the compressive strength of foam concrete D500 up to 1.50 MPa. The positive effect of grinding sand with a glyoxal additive on the strength of foam concrete is explained by the mechanochemical activation of the surface of quartz sand, which leads to a decrease in plastic shrinkage and an increase in the contact strength of foam concrete . At joint grinding of quartz sand with crystalline glyoxal the class of foam concrete increases to B1, while maintaining the average density D500.

4. Conclusion

The introduction of modifying glyoxal-containing additives into the concrete mix during its preparation leads to reduce shrinkage deformations, to accelerate the process of structure formation and to increase the compressive strength of foam concrete.

1. Optimal content of 40% aqueous solution of glyoxal or crystalline glyoxal is 0.01% by weight of cement in foam concrete mixture based on Portland cement, sand, water and a foaming agent. Foam concrete mixture with 40% aqueous solution of glyoxal has better flowability of mixture from 11 to 14 cm and reduced plastic shrinkage of natural hardening foam concrete by 45 %.

2. Joint grinding of sand with crystalline glyoxal in the amount of 0.01% by weight of cement leads to enhancing the compressive strength of foam concrete D500 up to 1.50 MPa and also leads to reduce drying shrinkage by 54%.

3. Good agreement of the experimental results obtained by the authors was established during the elaboration of scientifically justified compositions of foam concrete with application of glyoxal-containing additives, and that agrees with the other private research results of other authors obtained during application of structure formation accelerator for foam concrete mixtures and given in independent sources. Thus, glyoxal-containing additives are an interesting object for obtaining new knowledge and developing innovative technologies for hardening cement compositions in construction.

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