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Cement-Based Materials Modified with Nanoscale Additives

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Abstract. The most common and reliable material without which modern construction is indispensable is concrete. The development of construction production is pushing for new solutions to improve the quality of concrete mix and concrete. The most demanded and significant indicators of a concrete mixture are the compressive strength and mobility of the concrete mixture. Every year, the volume of research on nanomaterials as modifying components of concrete is significantly increasing, and the results indicate the prospects for their use. Nanoparticles with a large specific surface are distinguished by chemical activity, can accelerate hydration and increase strength characteristics due to nucleation and subsequent formation of C–S–H and compaction of the material microstructure. Sol of nanosilica, which can be used instead of microsilica from industrial enterprises, and carbon nanomaterial have a wide reproduction base. This paper presents studies of these types of nanomaterials and the results of their application in cement concrete. Studies have shown that the effect is also observed with the introduction of an additive containing only one type of nanoparticles. The dependence of the obtained characteristics of cement concretes on the content of these nanomaterials has been established. It has been found that the best results were obtained with an additive in which the above-mentioned nanomaterials were used together. Compressive strength of heavy concrete samples, improved by the complex nanodispersed system, was 78.7 MPa, which exceeds the strength of the sample containing the CNT additive in a pair with a super-plasticizer by 37 %. The paper proposes the mechanism for action of the presented complex additive.

Keywords: water sol of nanosilica, carbon nanotubes, super-plasticizer, heavy concrete, strength

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Материалы на основе цемента, модифицированные наноразмерными добавками

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Реферат. Самым распространенным и надежным материалом, без которого не обходится современное строительство, является бетон. Развитие строительного производства подталкивает к новым решениям в улучшении качества бетонной смеси и бетона. Наиболее востребованные и значимые показатели бетонной смеси — прочность при сжатии и подвижность бетонной смеси. С каждым годом исследований наноматериалов в качестве модифицирующих компонентов бетона становится больше, а результаты указывают на перспективность их применения. Наночастицы, обладающие большой удельной поверхностью, отличаются химической активностью, могут ускорять гидратацию и повышать прочностные показатели за счет зародышеобразования и последующего формирования С–S–H, уплотнения микроструктуры материала. Широкую базу воспроизводства имеют золь нанокремнезема, который может быть использован взамен микрокремнезема с индустриальных предприятий, и углеродный наноматериал. В статье представлены исследования данных видов наноматериалов и результаты их применения в цементных бетонах. Исследования показали, что эффект наблюдается также при введении добавки, содержащей только один вид наночастиц. Установлена зависимость получаемых характеристик цементных бетонов от количества данных наноматериалов. Выявлено, что наилучшие результаты были получены с добавкой, в которой совместно использовались вышеперечисленные наноматериалы. Прочность на сжатие образцов тяжелого бетона, улучшенная комплексной

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Наука итехника. Т. 20, № 3 (2021) нанодисперсной системой, составила 78,7 МПа, что превышает прочность образца, содержащего добавку углеродных нанотрубок в паре с суперпластификатором, на 37 %. Изучен механизм действия представленной комплексной добавки.

Ключевые слова: водный золь нанокремнезема, углеродные нанотрубки, суперпластификатор, тяжелый бетон, прочность

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Introduction

Currently, nanoscale materials have found their application in industry, as modifying components. Having very large values of the specific surface, nanosized particles are capable of affecting the physicomechanical characteristics of the new material. Depending on the type of raw material and the production method, the characteristics of the obtained nanomaterials vary widely [1–8]. All this affects the results obtained when using the same concrete mix formulations.

An analysis of the literature shows nanomaterials, such as carbon nanotubes and silica fume, were initially used only as a means to increase the strength characteristics of modified concrete. A little later it became known that the use of these nanomaterials together with plasticizing additives provide an effective reduction in water and an increase in the durability (frost resistance) of the created concrete [9–25]. However, questions regarding the compaction of the structure and the filling of voids between cement particles and even larger sand particles remain open at the moment. The working hypothesis of the study is that a complex nanodispersed system consisting of nanosilica, multilayer carbon nanotubes (MCNT) and a superplasticizer (SP) will allow to obtain a synergistic effect, i. e. summation of the positive properties and qualities of each of the components.

Materials and methods of experimental studies

The following materials were used in the work as nanomaterials: water sol of nanosilica SiO_2 , obtained from hydrothermal fluids, with characteristics of a pH = 9.2, density ρ = 1075 g/dm³, solids content of nanosilica SiO_2 = 120 g/dm³, total salinity equal to 1720 mg/dm³ [26–28] and carbon nanotubes obtained in a plasma of a high-voltage atmospheric pressure discharge with the optimal composition of the CH₄ gas mixture: air = 1:(2.4–2.5), followed by chemical treatment with a mass fraction of solids of not less than 37 %, pH = 7.0, ρ = 1.1 g/cm³ [29–36]. Superplasticizer from a series of polycarboxylates highly effective in water-reducing ability in the form of an aqueous solution.

Portland cement CEM I 42.5N was used as a binder as a fine aggregate, quarry sand with a fineness modulus $M_f = 3.8$ with a true density $\rho = 2572$ kg/m³ and bulk density $\rho = 1540$ kg/m³ was used. As a coarse aggregate, the fraction is 5–20 mm thick, bulk density $\rho = 1460$ kg/m³.

The particle size distribution of water sols of nanosilica was determined using the Analysette 22 laser diffraction device, which is built on the principle of "Inverse Fourier Optics" or a converging laser beam (Fig. 1). Reverse Fourier Optics also allow extremely high resolution measurements. By automatically moving the measuring cell supported by the computer in a converging beam in the NanoTec model, it is possible to create a supermatrix with a max 520 measuring channels for calculating. In this case, the measurement range is from 0.01 to $1000~\mu m$.

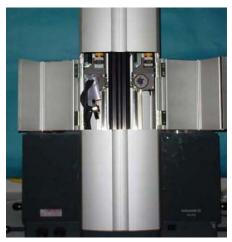


Fig. 1. Laser analyzer Analysette 22 NanoTec

The results of determining the particle size of nanosilica were given the following results: the size of the solid particles of nanosilica contained in aqueous ash is less than 10 nm, which is explained by the resolution of the device (from 10 nm to $1000 \mu m$).

The concentration and solids content of nanosilica SiO_2 in the sol was determined by evaporation of the initial water sol with a mass of $m_{initial} = 100 \text{ cm}^3$ in an oven at a temperature of $t = 150 \,^{\circ}\text{C}$.

Weight reduction according to studies:

1 day $- m_1 = 233 \text{ ml/g}$;

2 days $-m_2 = 166 \text{ ml/g}$;

3 days $-m_3 = 146$ ml/g; 4 days $-m_4 = 145$ ml/g;

 $8 \text{ days} - m_8 = 145 \text{ ml/g}.$

This indicates that the $SiO_2 = 120$ g/dm³ approximating value for the indicated solid particle content is solid; the mass from the 4th day remained unchanged due to the room humidity. Upon evaporation, the sol particles stuck together.

The results of measuring the particle size distribution of the dispersed composition of carbon nanomaterial are presented in Fig. 2.

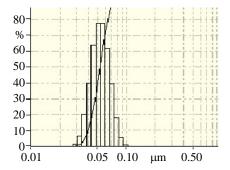


Fig. 2. The size distribution of nanostructured carbon particles

In order to study the effect of nanomaterials on the basic properties of cement concrete, an additive was made and a series of laboratory studies was carried out. The amount of added additive in all studied compositions was 0.8 % by weight of cement.

To determine the effect of an aqueous sol of SiO_2 (NS) silica on the strength characteristics of concrete, it was preliminarily mixed with mixing water, uniformly distributed throughout the volume, after which water was introduced into the concrete mixture. The mixture filled the cube-shaped nests (100×100 mm), which was mounted on a vibrating table and compacted. After manufacture, the samples were unformed and stored in bathtubs with water until they reached a certain

age. Testing the samples for compressive strength was carried out on the 1st, 7th, 14th and 28th days.

The data obtained showed that when the sol content of nanosilica SiO_2 in the amount of 0.1 wt. % in cement (NS 0.47) the increment in compressive strength compared to the control (Control) was +30% on the first day, +20% on the 28^{th} (Fig. 3).

It was found that an increase in the concentration of this nanosupplement NS 4.7 leads to a decrease in strength by 1.2 times, which indicates the advisability of using SP in this system.

A method of preparing a plasticizing additive used hereinafter consists in dispersing a superplasticizer and nanomaterial (nanosilica or nanocarbon) in a mixer for several minutes. After that, concrete samples were manufactured and tested according to the above methodology.

Tests of a plasticizing additive, including a nanosilica sol paired with a superplasticizer, were carried out at W/C values from 0.2 to 0.3 (Fig. 4).

An additive containing a nanosilica sol with a concentration of solid SiO_2 equal to 0.47 g/kg, introduced into the superplasticizer in amounts of 8, 4, and 1 g/t when determining the compressive strength, showed the best result of 73.7 MPa when solid SiO_2 4 g was added per 1 t of superplasticizer.

At the same time, the introduction of NS increased the strength of concrete to 30 % compared to concrete containing only SP and to 40 % compared to the control sample.

The effect of a complex additive containing MCNT (MLCNT), NS and SP on the strength characteristics of concrete are presented in Fig. 5.

The compressive strength of samples of heavy concrete, improved by an integrated nanodisperse system, was 78.7 MPa, which exceeds the strength of a sample containing an MLCNT additive paired with superplasticizer by 27 %.

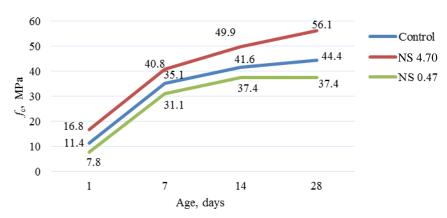


Fig. 3. The effect of nanosilica sol on the strength characteristics of heavy concrete

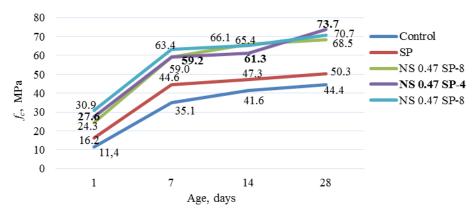


Fig. 4. The effect of nanosilica sol in combination with superplasticizer on the strength characteristics of heavy concrete

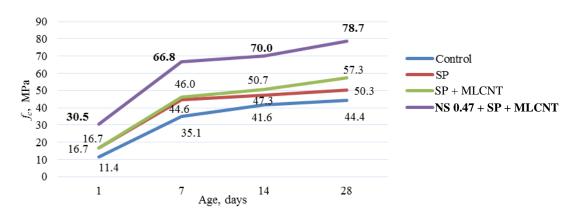


Fig. 5. The effect of complex additives on the strength characteristics of heavy concrete

It can be assumed that the mechanism of action of the presented complex additive is as follows: the introduction of a nanocarbon material into the cement system leads to the packing density of the particles of the system, and when an even smaller nanosilica is introduced, additional crystallization centers of hydrated neoplasms are created. Therefore, nanosilica acts as a nanofiller of this system, which more actively affects the concentration of Ca²⁺ and OH⁻ and reduces the number of pores.

CONCLUSION

Based on the analysis of the test results, it can be concluded that a complex nanodispersed system, including multilayer carbon nanotubes, nanosilica and superplasticizer, helps to bring together particles, densify the structure and form intergrowth contacts, which effectively affects the structure of heavy concrete. Thus, this effect is achieved by directed structural modification of the main components of the cement stone – calcium hydrosilicates relative to the composition and morphology of neoplasms.

List of accepted symbols and abbreviations

 f_c – compressive strength of concrete, MPa; $m_{1...8}$ – weight reduction, ml/g; pH – pH value; M_f – size module; ρ – density, kg/m³; indexes: c – compression; f – large-ness.

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