Oxidized thermally expanded graphite as a raw material for the production of cement composites

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Abstract. One of the current tasks is to improve the performance of cement composites. Effective ways to solve a problem is to use a new generation of carbon-containing modifiers. The regulation of the physical and mechanical and operational properties of cement composites must be carried out at the micro level, by regulating the structure formation of the cement matrix. One of the promising directions is the use of graphene as a carbon-containing modifier. The article represents research data on oxidized thermally expanding graphite. The results of microscopic and laser diffraction granulometric analyzes are represented. The performed studies allowed to characterize the properties of the research material. The analysis of particle size distribution was carried out on a Fritsch NanoTec laser particle size measuring device with a dispersion unit in a liquid space with a total measurement range from 0.01 to 2100 μm and showed a decrease in size from 574 μm to 22.2 μm. Microscopic research was carried out by using a scanning (raster) electron microscope TESCAN Mira 3 with a magnification range of x1 – x1 000,000. The experimental results presented in the present research prove the effectiveness of using oxidized thermally expanding graphite as a modifier of cement composites.

1. Introduction

Graphene is a two-dimensional allotropic form of carbon (figure 1), in which atoms combined into a hexagonal crystal lattice form a layer with a thickness of one atom [1,2]. Graphene has unique properties, which allows to use it in various fields. According to scientific research [3,4,5], the material is very durable, flexible and elastic, able to restore the crystalline structure in case of damage, has a high electrical conductivity of about 100 m / s, high electric capacity, which approaches to 65 kW · h / kg, very light, absorbs radioactive wastes [6,7].

Figure 1. The structure of graphene
According to the literature sources [8, 9], pure graphene is extremely rare. There are various methods for producing graphene [9, 10, 11, 12] from different types of graphite and its oxides with oxygen-containing functional groups and molecules attached at the edges or inside the carbon network.

An important direction in the production of heavy concrete is the transition to cement composites with improved physical, mechanical and operational properties [13,14,15]. To achieve this goal, an integrated approach to solving the problem is required. First of all, the optimization of the composition should be controlled at the micro level with the possibility of regulating the structure formation of the cement matrix. Promising are the studies on the use of carbon-containing components in the form of: carbon tubes, fibers and fullerenes in the production of high-strength concrete [16, 17]. The authors of the following papers [18, 19] consider it possible to obtain durable materials using graphene due to the micro-reinforcing effect. The introduction of carbon-containing modifiers into the composition makes it possible to increase compressive and bending strength, crack resistance, fire resistance, reduce water absorption and, as a result, increase frost resistance. The question still remains unanswered about the effective method of introducing and distributing carbon-containing additives and fibres in a cement composite in connection with low concentrations of the substance. Currently, the most actual task is to develop a technology for producing pure graphene in industrial scales and to reduce the production cost.

2. Methods

The analysis of particle size distribution was carried out on a laser device for measuring particle size with a dispersion unit in a liquid space and with a total measurement range of 0.01 - 2100 μm Fritsch NanoTec.

The structure of the obtained carbon-containing additive was studied using a TESCAN Mira 3 scanning electron microscope with a magnification range of up to 1 000 000 times.

3. Results and Comments

A powder of oxidized thermally expanding graphite was used as a sample material for producing a carbon-containing additive. This material is a grey crystalline powder with a characteristic metallic shine. Layering (scaly structure) due to the laying of the plates in the radial (flat) direction. A photograph of the source material is demonstrated in figure 2 (a, b, c).
Figure 2. A microphotograph of the sample graphite
The particle sizes of the initial powder vary from 4 to 1350 μm and are represented in figure 3.

Figure 3. Particle size distribution diagram of sample graphite
The particle-weighted average particle size D [4.3] is 574 μm, which is comparable with the fine sand fraction in the cement stone, therefore, in the initial state it cannot carry out the function of a modifier.

The carbon-containing additive was synthesized according to the method proposed by scientists of Rutgers University (USA) [21], by short-term exposure, not exceeding 10 seconds, of electromagnetic (microwave) radiation (frequency 2450 MHz), power 1000W.

As a result of processing procedure, there is a sharp thermal activation of the raw material with a hundred-fold volume increase of the material due to the removal of oxygen-containing groups. The rapid increase in pressure in the interlayer space leads to the splitting of the initial matrix, with the formation of elongated highly porous flakes (worm-shaped), characterized by a length of up to 20 mm and having a defective structure with a high content of voids. The morphology of the additive consists of
"crosslinked" graphene packs and contains up to 99.9% carbon without side liquids. A photograph of the microstructure is shown in figure 4 (a, b, c, d, e, f).

![Microstructure of thermally expanded graphite](image-url)

**Figure 4.** Microstructure of the synthesis of thermally expanded graphite
To assess the particle size distribution, the obtained material is dispersed in distilled water using ultrasonic treatment. The particle sizes of the obtained carbon-containing additives vary from 0.01 to 74 microns and are represented in figure 5.

![Figure 5. Particle size distribution diagram of carbon-containing additive](image)

The volume-weighted average particle size $D_{4.3}$ is 22.2 $\mu$m.

The granulometric composition of the additive corresponds to the cement dimension, the content of the additive fraction of up to 10 $\mu$m is 21%, which proves the effectiveness of the applied method and allows the additive to be used as a microfiller and modifier of cement composites.

4. Conclusions

As a result of studying the synthesized carbon-containing additive, an improvement in the characteristics of the material was shown compared with the initial oxidized thermally expanding graphite. Having examined the microstructure photographs, the presence of graphene packs with a dimension of 0.01 to 74 $\mu$m was diagnosed. Analyzing the data obtained during the particle size analysis, we can conclude that when exposed to electromagnetic radiation, the particle sizes are reduced from 574 microns to 22.2 microns, showing a decrease of about 26 times. A thine fraction of about 10 microns appears. This shows the possibility of introducing a graphite-containing additive into the composition of heavy concrete with a cement binder by optimizing the microstructure due to a denser packing of grains and the creation of additional centres for the growth of new materials.

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