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MECHANISMS OF ACTION OF FINELY DISPERSED AGGREGATES IN CEMENT SYSTEMS

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Abstract: The article details the physicochemical mechanisms of the influence of finely dispersed aggregates on cement stone structure formation processes. Three main effects have been described: filling, nucleation, and putzolan. Their synergistic effect on the formation of a dense and strong microstructure has been shown.

Introduction. The effectiveness of using finely dispersed aggregates in cement systems is due to their complex effect on hydration and hardening processes. Their role goes beyond simply diluting cement. Understanding the mechanisms of this influence is key to purposeful design of high-quality concrete compositions.

- **Physical and nucleation effects.** The primary effect of fillers is physical.
- **• Filling Effect (Microfiller Effect):** The smallest aggregate particles, which are significantly smaller than cement particles, fill the intergranular space. This leads to the optimization of the granulometric composition of the mixture, its compaction, and the displacement of excess water into larger pores. The result is a decrease in the water demand of the mixture and an increase in the density of the cement stone structure at the early stages of hardening.Эффект центра кристаллизации (Нуклеация):

• The surface of aggregate particles serves as additional active centers for the precipitation and growth of cement hydration products (calcium hydrosilicates, C-S-H) crystals. This accelerates hydration, contributes to the formation of a more homogeneous and fine-crystalline structure, which positively affects strength [1].

Pozzolan reaction. For active fillers (microsilica, metakaolin, fly ash), the key is the chemical mechanism - the pozzolan reaction. It occurs in the late stages of hardening (7 days and beyond) and consists of the following:

1. Dissolution: In an alkaline environment of cement paste (pH>13), amorphous silica (SiO_2) dissolves from the surface of the aggregate particles.

2. Interaction: The dissolved silica reacts with calcium hydroxide (Ca $(OH)_2$) - a byproduct of cement hydration, which has low strength and is a vulnerable part of the structure.

3. Gel formation: As a result of the reaction, additional low-basic calcium hydrosilicates (C-S-H phases) are formed - the main carriers of cement stone strength. This process is called "self-compaction," because the volume of solid reaction products exceeds the volume of initial reactants. This leads to the filling of pores and capillaries, a sharp decrease in permeability, and an increase in the strength and durability of the material.

Synergistic effect. The greatest efficiency is achieved when all mechanisms act synergistically. The filler works as a "physical filler" and "nucleator" in the early stages, and then, in the case of its putzolan activity, it enters into a chemical reaction, ultimately compressing and strengthening the matrix. The combination of these processes transforms the microstructure: the total porosity decreases, the pore size decreases, the proportion of strong gel phases increases, and the content of large-crystal Ca $(OH)_2$ decreases [2].

The modern technology of cement composites has undergone a profound evolution, finally moving beyond the traditional three-component system of cement, water, and aggregate. Currently, the development of cement system compositions is a complex scientific and engineering process aimed at creating multicomponent binders with predetermined operational characteristics. This transition is necessitated not only by increasing the strength and durability of materials but also by addressing environmental, resource-saving, and economic challenges related to the decarbonization of the construction industry.

One of the most promising directions for the development of materials science is the integrated use of finely dispersed mineral fillers of various nature and functional purpose. In recent decades, cement composite design technologies have evolved from monosystems to binary, triple, and more complex multicomponent systems. This approach allows for achieving a synergistic effect where the combined result of the interaction of components significantly exceeds the sum of their individual impacts [3].

The combined use of several fillers with different mechanisms of action contributes to the mutual elimination of their shortcomings and the enhancement of the positive properties of each of them. Thus, the combined use of finely ground carbonate and active silica additives creates optimal conditions for the formation of a complex set of operational characteristics. Limestone, due to its pronounced filling effect and ability to accelerate the hydration of aluminate phases, provides an intensive early strength gain, which is especially important in the first stages of cement stone hardening. Simultaneously, active additives, such as microsilica, react with calcium hydroxide in a pozzolan reaction, forming additional C-S-H phases, which increase the strength and durability of the material in the later stages of hardening.

Based on these theoretical provisions, the hypothesis of the need for targeted management of the microstructure of cement composites through the rational combination of finely dispersed mineral fillers, differing in their chemical nature and surface activity level, is scientifically substantiated. This approach allows for the formation of a multi-mode structure where particles of different sizes and reactivity perform complementary functions.

According to the postulates of the theory of composite building materials of V.I. Salomatov, the synergistic effect of multicomponent systems is realized through the combination of two interconnected processes [4].

Firstly, the creation of crystallization centers and acceleration of hydration are achieved by introducing fillers whose surface activity is equal to or exceeds the activity of the binder (Fn ≥ Fv). Microsilicon and metakaolin particles act as additional nucleation centers, contributing to the intensive growth of hydrate phases and the formation of a dense, homogeneous structure.

Secondly, the ordering of the structure and the reduction of deformation stresses are carried out by introducing fillers with a lower surface activity (Fн < Fв), such as finely ground limestone or quartz sand. These materials contribute to the compaction of the phase boundaries, the reduction of internal stresses, and the formation of a more stable microstructure, which minimizes the risk of microcracks and increases the durability of the composite.

Thus, the modern concept of cement system design is based on the principles of multi-level structural regulation, where the main tool is the controlled interaction of components of varying dispersity and chemical activity. This allows for a shift from empirical composition selection to scientifically based engineering of new-generation cement composites with predictable properties and increased operational efficiency (Figure 1).

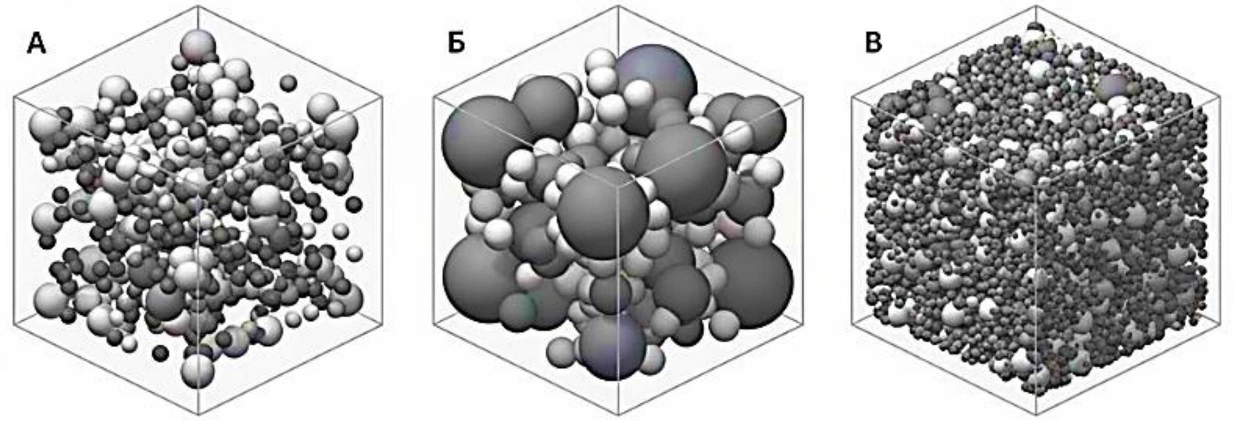

Fig. 1. Spatial-structural topology of a binder
A - Dispersity of mineral filler higher than cement dispersity; B - The dispersity of the mineral filler is significantly lower than the dispersity of cement; B - The dispersity of binary mineral filler is greater and less than the dispersity of cement (optimal packaging).

The phenomenon known as granulometric composition optimization represents one of the fundamental principles of modern materials science and underlies the rational design of cement composites. Its essence lies in achieving the maximum packing density of particles of various fractions, which ensures the minimization of pore space and, consequently, increases the strength, density, and durability of the material.

If we consider the structure of traditional concrete, it becomes clear that each aggregate fraction has a certain void. Thus, the void content of separately taken coarse aggregate (gravel) and fine aggregate (sand) can reach 45% or more. However, when they are combined, mutual compaction occurs: smaller sand grains effectively fill the intergranular space between large gravel fractions, which leads to a significant decrease in the total porosity of the system - on average up to 25% [5].

Further introduction of finely dispersed fraction represented by cement and especially mineral fillers allows for the filling of the remaining pores in the material structure. As a result, the total void capacity of the system can be reduced to 10% or less, which ensures the formation of a practically dense contact frame. Such a microstructure is characterized by minimal open pore volumes and a high degree of adhesion between the solid phases, which is directly reflected in the physical and mechanical properties of the cement stone.

The principle of dense packing of particles is the theoretical and practical basis for developing binary, triple, and multicomponent systems with optimized granulometric distribution. The use of fillers of various nature, morphology, and degree of dispersion contributes to the creation of a so-called multimodal structure, in which particles of different sizes and shapes effectively fill the intergranular space of each other. This ensures not only the compaction of the solid phase but also improves the entire complex of operational characteristics of cement composites [6].

The mechanism of action of the optimized granulometric system manifests itself in several interconnected effects:

Reduction of emptiness. Dense packing of solid particles leads to a decrease in pore space volumes, which is the main factor in increasing the strength, durability, and water resistance of cement stone.

Increasing material density. The reduction of voids contributes to an increase in bulk density and a decrease in permeability, which increases the concrete's resistance to external influences, including corrosion processes.

Improvement of rheological properties. Fine-dispersed particles of fillers, having a spherical or similar shape, perform the role of unique micropodшипners, facilitating the movement of particles in the fresh mixture. This contributes to increased workability, reduced water demand, and improved technological efficiency of concrete while maintaining its strength characteristics.

Thus, optimizing the granulometric composition by introducing binary and multicomponent mineral fillers ensures the formation of a denser, more homogeneous, and structurally stable cement stone matrix. This approach not only improves the physical and mechanical properties and operational reliability of the material but also contributes to the creation of energy-efficient, economical, and durable new generation cement composites.

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