



Research into the properties of concrete modified with natural zeolite addition



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HIGHLIGHTS

- Zeolites are among the most promising of concrete components in the development of new building materials.
- Replacement of cement with natural can be increased concrete density, compressive strength, freeze-thaw resistance.
- Natural zeolite can be used for modification of cementitious systems.

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ABSTRACT

Concrete mixture used for the tests consisted of cement CEM I 42.5 R, natural zeolite (clinoptilolite), 0/4 fr. sand as fine aggregate, and 4/16 fr. gravel as coarse aggregate. Active components of the zeolite were determined by means of X-ray diffraction analysis, particle form (plate-shaped) and size (11.82 μm) were determined by means of SEM. 5 batches of specimens were made with zeolite content ranging from 0 to 10% (the binding material was substituted with natural zeolite). The following parameters of modified concrete were tested: compressive strength, water absorption, density, ultrasonic pulse velocity, porosity (open and closed), predicted freeze-thaw resistance. Substitution of up to 10% of cement with natural zeolite increases the compressive strength of concrete by 15%, reduces water absorption 2.3 times, increases density and ultrasonic pulse velocity. Substitution of up to 10% of cement with natural zeolite increases closed porosity and subsequently improved the freeze-thaw resistance of concrete. The predicted freeze-thaw resistance calculations revealed 3.3 times higher resistance of concrete modified with up to 10% of natural zeolite.

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1. Introduction

The term Zeolite (meaning boiling stone from Greek *zeo* (to boil) and *lithos* (stone)) was coined by the Swedish mineral researcher Alex Fredrik Cronstedt in 1756. Since then zeolites were classified as a separate class of minerals. The first attempts to synthesize zeolite by imitating natural geological conditions that enabled the formation of zeolites were made by Saint Claire de Ville in 1862. He continued the experiments until 1948, when M.R. Barrer announced that he had succeeded in obtaining a synthesized zeolite equivalent to the natural one.

Zeolites are crystalline aluminosilicates with open 3D framework structures built of $(\text{Al},\text{Si})\text{O}_4$ tetrahedra and are classified according to $\text{SiO}_2:\text{Al}_2\text{O}_3$ ratio. The advantageous properties of

zeolites are: regular structure, large inner specific surface area (approx. 600–800 m^2/g), uniform size pores, good thermal stability [1]. Aluminosilicate framework topology is the sole criterion that enables to identify the structure of zeolites. In theory there may be many framework structures, although only 32 have been identified yet. Crystal structures of clinoptilolite of Fig. 1 [2].

Zeolites are found in nature, however, not all zeolites can be synthesized and not all synthesized zeolites have natural equivalents.

Research have shown that zeolites have properties of molecular sieves, high absorption capacity at low pressures and low relationship with temperature in the range of 10–150 °C [3].

The specific structure of zeolites make them beneficial in many applications. They are used to adsorb different substances from gas mixtures and solutions; they are used as molecular sieves because they let through only molecules of certain size; cation exchange properties of zeolites enable to replace cations with weak bonds by other cations on the surfaces of internal cavities and channels;

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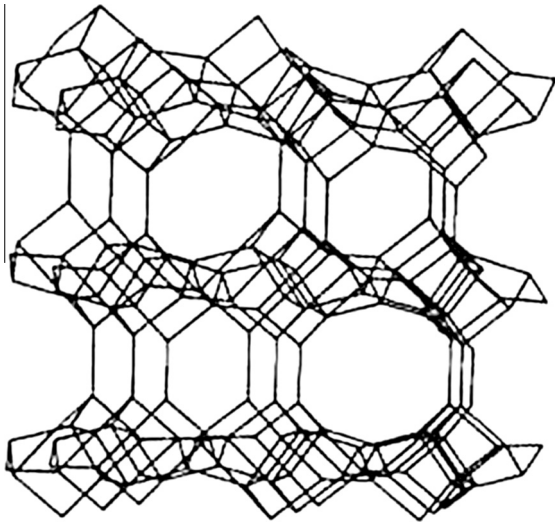


Fig. 1. Crystal structure of clinoptilolite [2].

the catalytic properties in certain reactions reduce the activation energy of molecules in the pores of crystal cells. Zeolites are environmentally clean, inert and non-toxic material suitable for many industrial, agricultural, household, and environmental applications [4,5].

There are many hypotheses explaining the adsorption properties of zeolites. R. Barrer claims that molecules of adsorbed substances dissipate in the cavities and channels of dehydrated zeolites, retain their mobility and produce incorporated solutions. Three-dimensional cells of thermally resistant zeolites adsorb the gas volume equivalent to water molecules. Dehydrated zeolites have adsorbing cavities where gas or liquid molecules are adsorbed. Zeolites can adsorb not only water, but also alcohol, benzene, chloroform, carbon disulfide. Metal zeolite cations are highly migrant and can participate in ion exchange reactions [6].

In zeolites zeolitic water is removed gradually and reversibly, without changing its structure. Zeolites have a well-organized structure of internal pores. Adsorption processes on their surfaces take place along reversible isotherms. Material adsorption and desorption isotherms coincide. Zeolites have strong adsorption capacity due to the pores, channels and voids. Molecules and cations of zeolitic water are distributed along the channels of zeolite framework. Zeolites cannot adsorb molecules (0.3–1.0 nm) larger than their pores and channels. Dehydrated zeolites are good molecular sieves. The internal surface of channels and voids inside the zeolite framework can adsorb molecules selectively. The diffusing molecules correspond to the diameter of the channel. Some molecules can, while others cannot pass the channel. The adsorption and selection depends on the correspondence of molecule size and form with those of the channels inside the zeolite framework; zeolites also separate molecules based on their polarity [5–8].

Both, natural zeolites, which are available in 50 types (clinoptilolite, mordenite, phillipsite, erionite, chabazite et al.), and specially synthesized zeolites (A, X, Y, L, ZSM-5 et al.) [9], are widely applied.

Construction materials with zeolites are developed in the following directions: binding materials, concretes, non-autoclaved silicate products [10]. Synthetic zeolites can be used as adsorbents or traditional water softening agents, detergents [11,12].

Zeolites are rich in active SiO_2 and Al_2O_3 . Silica dioxide and ash, like other pozzolanic materials, can improve concrete strength by means of $\text{Ca}(\text{OH})_2$ reaction with pozzolans. On the one hand, zeolites, like other pozzolanic materials, better than cement improve the strength of concrete stronger. On the other hand, zeolites pro-

mote the formation of side products, such as alkali and other complex compounds. Researchers have found that zeolites of different modifications act in concrete as pozzolanic additive [13].

Zeolites are one of the most prospective components of concrete in the development of new construction materials, such as high-performance concretes, specially designed concretes to absorb heavy metals or suppress radiation. The addition of zeolites enable to produce lighter structural elements without deteriorating the strength properties. Zeolites are widely used in finishing materials to achieve even surfaces and better drying of the mortar. Zeolite additives accelerate cement hydration process, change physical and mechanical properties of cement [14].

According to the studies by Yun-Sheng S., Chen-Lin H., the addition of up to 15% zeolite in the cement mixture improves the early compressive strength. Zeolite additives also shorten the hydration time [15].

Turkish researchers made tests with concrete specimens containing 5, 10, 20 and 40% of zeolite. Compressive strength was measured after 1, 2, 7 and 28 days of curing. The results showed that after 24 h of curing concrete specimens with zeolite additive had lower compressive strength compared with the control specimen. The same trend was observed after 2 and 7 days of curing. After 28 days of curing the compressive strength of specimens containing 5% of zeolite increased 6.8% compared with the control specimens; in other specimens the compressive strength increased as follows: 15.9% in specimens containing 10% of zeolite, 22.3% in specimens containing 20% of zeolite, 4.1% in specimens containing 40% of zeolite [16].

F. Canpolat with colleagues researched into concrete containing zeolite used as a mineral additive. Cement was substituted with zeolite added at 5–35%. They also tested the strength properties of concrete containing zeolite and fly ash additives. The fly ash was added at 5% by cement weight. The compressive strength was measured after 2, 7, 28 and 90 days. According to the test results the highest compressive strength was achieved after 28 days in specimens containing 20% of zeolite. In the case of adding zeolite and fly ash, the optimal limits were 10–25% of zeolite and 5% of fly ash [17].

Researchers B. Ahmadi and M. Shekarchi studied the effect of zeolite on mechanical properties and durability of concrete compared to other cement additives. Experimental testing proved good pozzolanic activity of zeolite, although zeolite reacted with portlandite in a different way than SiO_2 microparticles. Besides, the researchers found that compressive strength, water absorption, oxygen and chloride permeability, and electric resistance in concrete modified with different content of zeolite improved, were similar or even better compared to concrete containing SiO_2 microparticles [18].

Researchers have tested zeolite additives in cement systems. The best results of compressive strength (increase by 10%) were found in specimens where 5% of portland cement was substituted with zeolite [19].

Porosity is a very important parameter of concrete in the evaluation of its durability. Capillary porosity and entrained air content have the highest effect on freeze-thaw resistance of concrete. These parameters can be controlled in concrete production process.

The porosity system in hardened cement paste is made of four types of pores: gel pores, capillary pores 5–5000 μm , macro pores produced by entrained air and macro pores resulting insufficient compaction. Gel pores have no negative effect on the strength of concrete. Capillary pores and macro pores reduce the strength of concrete [20].

It is known that low w/c ratio and good setting conditions are important factors in the manufacturing of freeze-thaw resistant products [21–24].

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