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## Original Research Article

# Mechanical and durability properties of concretes incorporating natural zeolite



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## ABSTRACT

This paper investigates the application of natural zeolite as a substitute of cement in concrete. It studies the mechanical and durability properties of concretes containing 10% of natural zeolite and superplasticizer, as well as 10% of natural zeolite, superplasticizer and an air-entraining agent in comparison to concretes without natural zeolite. The study shows that fresh concrete mixture containing zeolite due to its high surface area demands a higher dosage of superplasticizer to achieve the targeted slump flow. While concretes containing natural zeolite characterize a lower compressive strength until 90 days of hardening, the compressive strength exceeds the strength of the concretes without zeolite after 180 days. The results also reveal the considerable effectiveness of using a superplasticizer and an air-entraining agent in zeolite incorporating concretes on water penetration, drying shrinkage and freeze–thaw resistance of concretes.

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## 1. Introduction

Concrete is a fundamental component of modern infrastructure, which is why its strength and durability play an important role. The properties of concrete in most cases depend on Portland cement, which remains the main hydraulic binder in use today. It is known that a partial substitution of cement by pozzolans results in an increase in durability of the concrete and, as a result, the service life of concrete structures [1,2].

Among the most common natural pozzolanic materials such as fly ash and silica fume is zeolite, which is used in some regions of the world, because of its lower cost and accessibility

[3–5]. Although natural zeolites are crystalline, they show proper pozzolanic activity [6]. According to Uzal and Turanlı [7] a lime reactivity of the clinoptilolite zeolite is comparable to silica fume, higher than fly ash and a non-zeolitic natural pozzolan. Therefore calcium hydroxide as a cement hydration product combines with natural zeolite consisting of reactive SiO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub> to form calcium hydrosilicates [7,8]. It was confirmed by many researchers [1,6–8], that the pozzolanic reaction in pastes containing zeolite of clinoptilolite type considerably decreased the calcium hydroxide content. The reduction of free Ca(OH)<sub>2</sub> in the hardened concrete improves the durability and concrete becomes more resistant to lime leaching by flowing waters and to expansion due to sulfate attack [8,9]. Chan and Ji [10] concluded that zeolite is

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more effective than pulverized fuel ash but worse than silica fume in improving the compressive strength. A number of studies show that the optimal amount of zeolite replacement level is approximately 10% [11–13]. A higher amount of zeolite replacing cement in concrete causes reduction of compressive strength.

Najimi et al. [6] concluded that concretes incorporating zeolite are characterized by the reduction of the heat of hydration and consequently of thermal cracking and improved durability properties such as chloride ion penetration, corrosion rate, drying shrinkage and water penetration. Sabet et al. [14] and Ahmadi and Shekarchi [15] also reported that the incorporation of natural zeolite as a mineral admixture in concrete enhanced its durability properties. However, various types, structures and purities of natural zeolites influence concrete strength and durability in different ways and can lead in some cases to contradictory results in experimental studies. Moreover, properties of concretes with the addition of different types of zeolite located in various regions of the world are already partly researched, but there is very low researches in particular mechanical and durability properties of concretes relating to the zeolite tuffs from fields located in Ukraine.

The aim of this paper is to study the effect of natural zeolite coming from Sokyrnytsia (Ukraine) on the mechanical and durability properties of concrete. These properties include water absorption, water penetration, drying shrinkage, freeze and thaw resistance.

## 2. Materials and methods

The materials used in this study include a clinoptilolite type of natural zeolite and commercially available Portland cement CEM I 42.5R. The chemical composition of the natural zeolite and Portland cement are presented in Table 1. It was determined by X-ray spectrometer ARL 9800 XP. Zeolite in comparison with Portland cement is characterized by a higher amount of silica, alumina and iron oxides, which are responsible for the pozzolanic activity of zeolite.

The tests of Portland cement properties were carried out according to EN 196 [16]. The specific surface of Portland cement used in the study is 425 m<sup>2</sup>/kg. Water demand attains 27%, initial and final setting time are 200 and 260 min, respectively. The compressive strength of Portland cement CEM I 42.5R after 2 and 28 days is 30.5 and 55.0 MPa.

The mineral composition of zeolitic tuff was determined by means of a powder X-ray diffraction method, using a PANalytical X'pert APD diffractometer (with a PW 3020 goniometer), a Cu lamp and a graphite monochromator. The analysis was performed at the angle range of 5–65° 2 $\theta$ . Philips X'Pert Highscore software was used to process the diffraction data. The identification of mineral phases was based on the PDF-2 release 2010 database formalized by the ICDD.

Powder X-ray diffraction patterns of the zeolite showed that clinoptilolite is the major crystalline phase. Zeolitic tuff also contains quartz as a minor impurity (Fig. 1a). The main components of the natural zeolite are identified as clinoptilolite, accompanied by other components.

The morphology of clinoptilolite and chemical composition in the field of main mineral components of the investigated materials was examined by scanning electron microscope SEM FEI Quanta 250 FEG, equipped with EDS. Clinoptilolite occurs in forms of thin plates in the range 10–30  $\mu$ m, sometimes of distinct hexagonal shapes (Fig. 1b).

According to the thermal analysis, in the first thermolysis phase of the zeolitic tuff, sample mass loss takes place in the temperature range of 20–200 °C (Fig. 2). It is associated with the release of physically-absorbed water. The second thermolysis phase of the sample occurs in the temperature range of 200–440 °C and corresponds to the emission of chemically bonded water, which is coordinatively and chemically linked to the crystal lattice of the mineral and forms aqua complexes with metal cations that enter the structure of the zeolite.

A shallow endothermic effect is observed in the temperature range of 440–580 °C, which relates to the dehydroxylation process of surface due to splitting off of OH– groups, which corresponds to a small weight loss in the TG curve.

In the high-temperature range of 580–1000 °C, the appearance of an endothermic effect in the DTA curve can be explained by a combination of several processes: a deeper surface dehydroxylation, destruction and a depreciation of the crystal structure of zeolite under high temperatures.

Mercury intrusion porosimetry (MIP) was used for determining the pore size distribution of natural zeolite. The tests were performed for pressures ranging from 0.1 to 200 MPa or pore radii from 10.0 to 3.8  $\times 10^{-3}$   $\mu$ m. The volume of mercury  $V$  [m<sup>3</sup>/kg] intruded at a given pressure  $P$  [Pa] gave the pore volume that can be accessed. The intrusion pressure was translated on equivalent pore radius  $R$  [m] following the Washburn equation:

**Table 1 – Chemical and mineralogical phase composition of the cement and zeolite.**

Portland cement (CEM I 42.5R)				Natural zeolite			
Chemistry	wt.%	Mineralogy	wt.%	Chemistry	wt.%	Mineralogy	wt.%
SiO <sub>2</sub>	20.17	C3S	59.76	SiO <sub>2</sub>	75.34	Clinoptilolite	60
Al <sub>2</sub> O <sub>3</sub>	4.41	$\beta$ -C2S	18.12	Al <sub>2</sub> O <sub>3</sub>	8.77	Quartz	30
Fe <sub>2</sub> O <sub>3</sub>	2.39	C3A	6.76	Fe <sub>2</sub> O <sub>3</sub>	1.30	Other phases	10
CaO	63.45	C4AF	10.91	CaO	4.6		
MgO	2.41			MgO	0.55		
SO <sub>3</sub>	3.21			SO <sub>3</sub>	0.05		
Na <sub>2</sub> O	0.17			Na <sub>2</sub> O	1.22		
K <sub>2</sub> O	1.02			K <sub>2</sub> O	2.41		

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