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Utilization of natural zeolite in aerated concrete production

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ABSTRACT

In this study, natural zeolite (clinoptilolite) was used as an aggregate and bubble-generating agent in autoclaved aerated concrete (AAC) production. The crushed and grinded samples were classified into two different particle sizes: 100 μm (fine-ZF) and 0.5–1 mm (coarse-ZC) before using in AAC mixtures. The effects of particle size, replacement amount (25%, 50%, 75% and 100% against quartz) and curing time on the AAC properties were experimentally investigated. It was found that usage of natural zeolite, especially with a coarser particle size, has beneficial effect on the physical and mechanical properties of AAC. The optimum replacement amount was determined as 50% and at this rate the compressive strength, unit weight and thermal conductivity of AAC were measured as 3.25 MPa, 0.553 kg/dm^3 and 0.1913 W/mK, respectively. Scanning electron microscopy analysis also confirmed the above findings. Denser C–S–H structures were obtained up to a replacement amount of 50%. Finally, the test results demonstrated that calcined zeolite acts as both an aggregate and a bubble-generating agent, and that AAC with a compressive strength of 4.6 MPa and unit weight of 0.930 kg/dm^3 can be produced without aluminum powder usage.

1. Introduction

Lightweight concrete (LWC) has been widely used in buildings as masonry blocks, wall panels, roof decks and precast concrete units. LWC offers design flexibility and substantial cost savings as a consequence of its lower unit weight. The required air pores for the production of LWC can be obtained by using expanded lightweight aggregate (perlite, blast furnace slag, volcanic ash, etc.) or the air pores can be formed in cement paste by the addition of gas-generating agents such as zinc or aluminum powder [1,2]. Although different types of lightweight aggregates have commercially been introduced into the market, the use of these aggregates is limited [3].

Autoclaved aerated concrete (AAC) is a well-known lightweight concrete and consists of a mixture of sand, lime, cement, gypsum, water and an expanding agent. AAC can be molded and cut into precisely dimensioned units and cured in an autoclave. During the production process, the ingredients combine to form the calcium silicate hydrate gels that establish the special properties of the finished product. Air is entrapped artificially by the addition of metallic powders, such as Al and Zn, or foaming agents. The chemical reaction caused by the addition of aluminum makes the mixture expand to about twice its volume, resulting in a highly porous structure. Curing it in an autoclave under pressure considerably reduces drying shrinkage and water movements [4]. Consequently the final products with an average compressive strength of 2.5–7.5 MPa and an oven-dry unit weight of 400–600 kg/m^3 (G2–G6 according to TS 453) offer considerable advantages over other construction materials, such as improved high-thermal and sound insulation, excellent fire resistance, high resource and energy efficiency and outstanding structural performance [5]. AAC products were first developed in Europe in the early 1920s as an alternative building material to lumber. A Swedish architect, Axel Johanson, introduced the product to Europe. Since then, AAC has become widely used in building construction throughout the world. As in the United States, China, India and the EU, the share of AAC in the total domestic concrete market has also progressively increased in Turkey over the last few decades. In 2008, the total amount of production capacity was reported as 2.1 million m^3, which made Turkey second biggest AAC producer in the EU. In recent years, the use of pozzolanic materials for the preparation of lightweight concrete, e.g., natural zeolite, silica fume, coal fly ash and slags, has gained attention owing to stringent environmental directives to recycle waste material.

Zeolites are crystalline alumina silicates with uniform pores, channels and cavities. They possess special properties such as ion exchange, molecular sieves, a large surface area and catalytic activity, which makes them a preferable material for tremendous industrial applications [6]. About 40 natural zeolites have been

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identified during the past 200 years; the most common ones are analcime, chabazite, clinoptilolite, mordenite and philipsite. Worldwide production of natural zeolite was estimated at about 3–4 M ton on the basis of recorded production and production estimates [7]. Rather than the known application areas, a considerable amount of research [8–13] concerning the use of natural zeolite, especially clinoptilolite in concrete applications as pozzolanic cement, lightweight aggregates and dimension stone has been conducted in recent years. However, its use in AAC as an aggregate and foaming agent is still limited. Furthermore, it should be considered that the physicochemical properties of raw materials, mixture formulation and employed methods are significantly affected by the final properties of concrete. Zeolite surface can be activated to have a high energy after calcination at temperatures greater than 400 °C. When subsequently immersed in water, the surface generates a large amount of air and heat due to adsorption. This heat increases the temperature of the air in the pores, or it is absorbed on the surface of zeolite particles. The expansion of the air volume results in the foaming and volume expansion of concrete during mixing and pre-storage periods. Turkey has 50 billon tons of natural zeolite, mainly clinoptilolite reserve [14]. Only 40,000 tons of this reserve was mined and consumed in 2005 mainly as soil conditioner and feed additives. This necessitates finding new alternative usage areas to increase this lower utilization ratio.

The aim of the present study is to test the use of natural zeolite in AAC production as an aggregate in place of silica sand and as a bubble-generating agent. Natural and calcined forms of clinoptilolite specimens were used as aggregates with different replacement amounts to determine the effects of clinoptilolite usage on the final properties of the products. The compressive strength, flexural strength and thermal conductivities of cured concretes and microstructural properties of the raw and final products were examined.

2. Experimental study

2.1. Materials and method

The zeolite-aerated concrete (ZAC) mixture was produced by using ordinary Portland cement (CEM I 42.5R), limestone, quartz sand, zeolite and water. The zeolite, clinoptilolite rich tuff (CLN), sample used in the AAC mixture was supplied by the Enli Mining Company, Gördes region, Turkey. The average clinoptilolite content of this material has been reported as nearly 80% by an earlier publication [15]. The grainy raw materials were mixed and ground in a ceramic ball mill to ensure the required homogenization and reduce the particle size of mixed material near 100 μm. The crushed and grinded samples were classified into two different particle sizes, 100 μm (fine-ZF) and 0.5–1 mm (coarse-ZC), before using in AAC mixtures to determine the particle size effect of zeolite on the concrete properties. Aluminum powder, obtained from Ytong aerated concrete factory, Turkey, was used as a foaming agent. The main oxide compositions of zeolite were determined by using X-ray fluorescence (XRF) analysis and the other ingredients used in concrete production are given in Table 1.

Representative AAC compositions were prepared by progressive incorporation of zeolite in place of quartz sand (25, 50, 75 and 100 wt.%). These compositions were designated as reference, ZAC-25, ZAC-50, ZAC-75 and ZAC-100, respectively. The mixture compositions of the test specimens are summarized in Table 2. Weighed materials according to the mix proportions were dry mixed for 1 min, then water was added to the mixture and then it was mixed again for 2 min. Aluminum powder was introduced into the mixture, following the addition of water to increase the volume of the slurry.

Compressive strength, flexural strength and thermal conductivity tests were performed on 100 mm cubic, 40 × 40 × 160 mm prismatic and 20 × 50 × 100 mm prismatic specimens, respectively. Mixtures were molded in steel molds, which were preheated in a drying oven under 40 °C to prevent heat loss of the slurry. After molding, specimens were first heated in a furnace at 40 °C for 2 h to achieve the settling and volume stability of the specimens. Then autoclave curing was conducted under 12 atm and 180–200 °C steam pressure for 8 and 18 h.

In the second part of the study, natural zeolite was also considered as both a gas-generating agent and an aggregate in the concrete production. Zeolite samples were first calcined for 2 h at 550 °C in a furnace to activate the zeolite surface according to previous study performed by Fu et al., 1996 as an optimum condition [12]. The activated specimens were cooled in desiccators and then used for the preparation of five different types of concrete mixtures. The specimens were then subjected to 18 h of autoclave curing at 180–200 °C.

2.2. Characterization techniques

The unit weights were determined on 100 × 100 × 100 mm ZAC specimens dried at 105 ± 5 °C for 24 h in accordance with TS 453 [5]. The ultrasound pulse velocity tests (UPV) were carried out by a non-destructive ultrasonic testing utility. This test is based on the theory of ultrasound transfer inside the material and is generally used to provide information about the porosity of the material in concrete applications [16,17]. Chemical analyses of the raw materials were carried out by using an XRF unit (Spectro X-Lab 2000). The mineralogical analyses of zeolite samples were conducted by using an X-ray diffractometer (XRD) (Rigaku, Rint 2200 with a nickel filtered Cu $K\alpha$). The microstructural evolution of bodies was observed by using versatile, analytical, ultra high-

Table 1
Properties of natural zeolite and cement.

Chemical composition	Zeolite[a] (%)	Cement (%)	Lime (%)	Cement	
				Physical and mechanical properties	
SiO_2	77.07	20.96	1.62	Blaine fineness (cm^2/g)	3345
Al_2O_3	13.56	5.58	0.61	Specific gravity	3.15
Fe_2O_3	1.59	3.69	0.23	Initial setting (h:min)	3:00
CaO	2.36	63.97	96.08	Final setting (h:min)	4:00
MgO	1.45	1.69	1.02	Le Chatelier soundness (mm)	3
K_2O	3.86	–	–	2 day Comp. strength (MPa)	21.9
Na_2O	0.11	–	–	7 day Comp. strength (MPa)	38.3
SO_3	–	2.84	0.015	28 day Comp. strength (MPa)	45.1
Cl	–	0.008	–		
LOI	–	1.15	0.55		

[a] Semi quantitative X-ray spectrometer results.