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Application-oriented mix design optimization and characterization of zeolite-based geopolymer mortars

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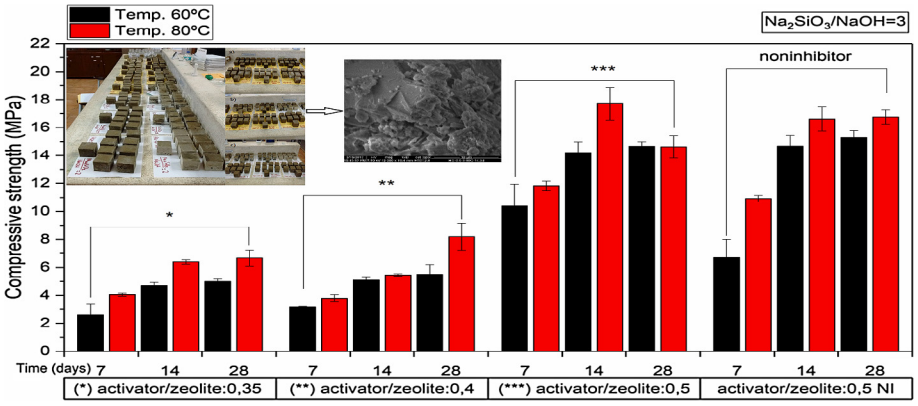
HIGHLIGHTS

- Zeolite-based geopolymer mortars were prepared for the first time.
- The best mixture and curing temperature, corresponding to: activator/zeolite: 0.5, Na_2SiO_3/NaOH: 3, and 60 °C.
- The compressive strengths of the specimens were evaluated for 7, 14 and 28 days.
- The use of corrosion inhibitor, MCI-2005 NS favored the compressive strength in ZGM specimens up to 14 days.

GRAPHICAL ABSTRACT



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ABSTRACT

In the present study, geopolymer mortar samples (ZGM) were prepared based on natural zeolites in which the effect of the addition of the MCI-2005 NS migratory type corrosion inhibitor was also evaluated through the compressive strength. Additionally, the design of the mixture was optimized using; two alkaline activators (NaOH 14 M and Na_2SiO_3) in different proportions, river sand as fine aggregate, curing time and temperature. All ZGM samples were characterized using quantitative X-ray diffraction, SEM-EDS, simultaneous TGA-DSC analysis, FTIR spectrometry and compressive strength. The best mixture and curing temperature, corresponding to activator/zeolite: 0.5, Na_2SiO_3/NaOH: 3, and 60 °C, was identified by the highest mechanical strength obtained after 28 days of aging of the synthesized specimens, which reached 17 MPa. The zeolite-based geopolymer mortars prepared can be used in real life applications due to acceptable compressive strength obtained, and the use of MCI-2005 NS favors the compressive strength in ZGM specimens at an early age up to 14 days.

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1. Introduction

Studies on geopolymers represent a growing field in environmental-friendly building materials because Ordinary Portland cement (OPC) production accounts for about 8% of annual global carbon dioxide emissions [1–3]. The substantial environmental impact of OPC production has led to increasing interest for such new cementing materials to substitute OPC partially. In addition, recent trends in geopolymers have led to a proliferation of studies that use different raw materials for geopolymerization synthesis such as solid industrial waste like fly ash [4,5], and blast furnace slag [3,6]; heat-treated biowaste like rice husk ash [7,8]; heat-treated clays like metakaolin [8–10]; or natural aluminosilicates like natural zeolites [2,11–14], etc. Consequently, this makes possible the usage on a large scale throughout the world and tackles different environmental concerns.

Geopolymers are synthesized by polycondensation reaction from solid precursors and alkali polysilicates [15]. The geopolymerization process involves a substantially rapid chemical reaction of silicate and aluminate minerals under highly alkaline conditions, thus resulting of three-dimensional inorganic polymeric chains in a microstructure contain in which exist covalent Si—O and Al—O bonds [2]. Previous studies have suggested that the chemical composition of this geopolymer materials is similar to that of natural zeolite materials, but instead, geopolymers present an amorphous structure [16,17].

Various studies on geopolymers have provided a substantial amount of knowledge about the alkaline activation of solid precursor materials rich in aluminosilicates, the most widespread method for geopolymerization synthesis. According to Palomo et al. [16], the type of activator plays a vital role in the process of geopolymerization. The reactions occur at a high rate when the alkaline activator contains soluble silicate, either sodium or potassium, compared to the use of only alkali hydroxides. On the other hand, the solid precursor must first meet the silica-alumina requirement that allows geopolymerization reaction proceeds. Xu and Van Deventer [17] confirmed that the addition of sodium silicate solution to the sodium hydroxide solution as the alkaline activator increased the reaction between Al—Si minerals and the solution. After studying the geopolymerization of sixteen natural minerals of Al—Si, they found that generally, the NaOH solution caused a higher mineral dissolution than the KOH solution on aluminosilicate minerals.

Natural zeolites are aluminosilicate minerals and have been used as the solid precursor in geopolymerization synthesis. In case of geopolymers pastes, i.e., no sand is involved in mix design, Villa et al. [18], obtained compressive strengths of about 30 MPa in geopolymer samples based on natural zeolites (clinoptilolite) activated with a mixture of NaOH and Na_2SiO_3 in different proportions and cured at different temperatures. They concluded that an optimum temperature range is 40–80 °C. On the other hand, Baykara et al. [12], studied the effect of NaOH concentration and calcium hydroxide ($Ca(OH)_2$) and Na_2SiO_3 proportions on geopolymer specimens, based on natural zeolites (mordenite) and cured at different temperatures. The authors showed geopolymers of approximately 10 MPa of compressive strength after curing for 24 h at 60 °C. In case of geopolymers mortar, i.e., sand is involved in the mix design, Nikolov et al. [2], prepared geopolymer based natural zeolite (clinoptilolite) and quartz sand for the synthesis of mortar using three different alkaline activators: sodium hydroxide, sodium silicate and sodium carbonate, resulting in geopolymers mortars whose maximum compressive strength was of 3.7 MPa after 28 days.

The temperature of geopolymerization synthesis also affects the performance of geopolymers. Recently researchers concluded that an increase in temperature causes adverse effects on mechanical properties [19]. Even chemical additions have been incorporated into the mix design to improve the compressive strength. Geopolymers based on natural zeolites with the addition of MCI-2005 NS migratory type corrosion inhibitors, cured at 40 °C/24 h and with 6-day of aging, showed that the addition of MCI-2005 NS favored the early compression strength of such geopolymers [11]. Regardless these studies, what is not yet understood is the relative effect of temperature and mix design on mechanical properties of zeolite-based geopolymers mortars.

The present work is the first attempt to prepare Ecuadorian natural zeolites-based geopolymer mortars (ZGM) as potential building materials and aims to evaluate the influence of the addition of MCI-2005 NS corrosion inhibitor on mechanical properties of ZGM specimens. To do this, compressive strength, mineralogical composition, and microstructure were investigated using characterization techniques such as quantitative X-ray diffraction, scanning electron microscope, simultaneous thermal analysis, Fourier transform infrared spectroscopy for both the raw material (zeolite) and the corresponding mortars prepared by the activation of zeolite using two alkaline activators (NaOH and Na_2SiO_3) in three different proportions (14 M NaOH, Na_2SiO_3/NaOH: 2, 2.5 and 3), using river sand as fine aggregate, a sand/zeolite: 1.5 ratio, curing time and temperature.

2. Methods and materials

2.1. Raw materials

In this research study, natural zeolite was used as raw material, due to its high availability and abundance in Ecuador [20,21]. The natural zeolites used in this investigation was provided by INDAMI S.A. The samples, with a particle size of ~200 μm, were dried in an oven at 100 °C for 24 h. Then, particle size was reduced in a disk mill until it could pass through a No. 325 mesh. The passed zeolite powder was used without any additional treatment.

River sand, used as the only aggregate for the ZGM synthesis, was manually collected from the banks of the Pastaza river located in the Penipe canton of the city of Riobamba-Ecuador. River sand was removed from the waste stream and dried in an oven at 100 °C for 24 h. After the drying period, the river sand was passed through a 2-mm sieve (No. 10) and retained in a sieve of 0.475-mm (No. 40) [5,22], which satisfied and complied with the standard ASTM D 2487 [23].

Alkali activator solutions, for zeolite mortar preparation, were selected from publications with high mechanical strength results [5,11,12,16–18,24–26]. NaOH (Merck-Millipore, 99% purity) pellets with high purity (99.0%) and Na_2SiO_3 (Proquiandinos S.A., Specifications: 51 °Bé a 25 °C, 14.13% Na_2O, 31.19% SiO_2) were used for the preparation of three different alkali activator solutions.

The migratory type corrosion inhibitor based on the amine carboxylate technology, MCI-2005 NS was kindly supplied by Cortec® Corporation. MCI-2005 NS is a liquid additive that protects reinforcing steel, carbon steel, galvanized steel and other metals embedded in concrete from corrosion induced by carbonation and chlorides. This inhibitor has the following specifications: pH: 11–12 (1% solution), non-volatile content: 25–30% and density: 9.9–10 lb/gal.

2.2. Geopolymer mortar preparation

For the preparation of the ZGM, the previously homogenized mixture (sand + zeolite) was activated with 14 M solutions of NaOH and Na_2SiO_3. These solutions were used in proportions of Na_2SiO_3/NaOH: 2, 2.5 and 3. The components were mixed by adding 1 l/m^3 of MCI-2005 NS, according to product specifications and literary recommendations [11]. ZGM specimens were prepared at three different liquid to solid ratios (activator/zeolite: 0.35, 0.4, 0.5), with a sand/zeolite constant ratio of 3:2. To obtain the ZGM blend, a HOBART model N-50 mechanical mixer with mixing speeds and stirrer type with 4.73 L capacity, was used. This machine complied with the standard for mixtures of pastes and cement mortars, ASTM C305 [27]. The wet ZGM paste was poured into 50 × 50 × 50 mm cubic molds. Cubic samples for strength test were prepared in agreement with standard ASTM C109/C109 M-16a [28]. Samples were cured in the mold for 24 h at two different temperatures of 60 and 80 °C. For this, cubes were covered with plastic packing to prevent dehydration, excessive moisture loss and thermal stress in the structure of the geopolymer mortar [2,11,29]. Mortar cubic samples were demolded and put to rest at room temperature until completing 7, 14 and 28 days for the compression strength testing (see Fig. 1). Besides, ZGM samples without MCI-2005 NS were prepared for comparison. By the parameters mentioned above, three replicates were