



Waste-based geopolymeric mortars with very high moisture buffering capacity

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HIGHLIGHTS

- Construction wastes were used to produce high moisture buffering capacity.
- Addition of H₂O₂ dramatically enhanced the mortars' moisture buffer value.
- Fly ash-based mortars presented low thermal conductivity.

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ABSTRACT

In this study, lightweight waste-based geopolymeric mortars were evaluated for the first time regarding their potential to passively adjust indoor relative humidity (RH) levels. Geopolymer mortars were prepared using a mixture of fly ash (FA) and metakaolin (MK) as a binder, in a proportion of 75:25 wt% (FA:MK), construction and demolition waste as the fine aggregate and a pore forming agent in varying amounts. The results showed that the addition of a pore-forming agent to the compositions considerably increased the moisture buffer value (MBV) of the mortars, that is, from 0.80 (reference mortar) to 5.61 g/m² Δ%RH (mortar with highest porosity). The moisture buffering capacity shown by these eco-friendly mortars is higher than values reported for other binder materials and can be classified as excellent (MBV > 2 g/m² Δ%RH). The porous FA-based mortars also presented low thermal conductivity (as low as 0.19 W/m·K), which suggests that these innovative binders could be simultaneously used for indoor moisture buffering and as low thermal conductivity materials.

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1. Introduction

Interest in the development of novel building materials with improved moisture buffering capacity is related to the increasing energy consumption associated with mechanical heating, ventilation and air-conditioning (HVAC) systems in buildings [1]. Besides the economic aspects, high indoor humidity levels can cause condensation on interior surfaces, material defacement and the proliferation of microorganisms with negative effects on human comfort and health [1,2]. Thus, the use of alternative materials, devices and approaches to minimize the use of HVAC systems and consequently to reduce the energy demand in buildings has been recently proposed. One promising option is the use of novel materials to control the indoor hygrothermal conditions passively [3].

The mechanism of moisture diffusion in hygroscopic materials is dependent on the moisture capacity and water vapor or liquid permeability. The moisture buffer value (MBV) is used as an unequivocal measure to characterize this property of building materials. This is a direct measurement of the amount of water vapor adsorbed or desorbed by a hygroscopic material when it is exposed to a periodic wave in daily cycles [4]. A hypothetical example of such cycles could be 12 h of higher relative humidity (RH = 75%) followed by 12 h of lower relative humidity (RH = 50%). It has been reported that the energy saving increases with increasing MBV values [1].

Different studies have shown that materials commonly used in building and construction (wood and wood-based components [5–7], modified mortars [8–10], and cellulose insulation [11]) or furnishings (textiles, wood and paper) [12], can be used for indoor moisture buffering. Modified mortars with enhanced moisture buffering can be produced using a porogenic additive (aluminum powder, sodium olefin sulfonate or superabsorbent polymers)

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[8–10,13] or an aggregate (sand, zeolite, perlite, biomass waste) [14,15]. Although the MBV of these modified mortars is improved, all of these materials are classified as “moderate” ($0.5 < MBV < 1.0$) or “good” ($1.0 < MBV < 2.0$) [4] in terms of their moisture buffering capacity. Moreover, these mortars contain Portland cement, which has excellent binder properties but its use is considered unsustainable due to the high level of CO₂ emissions arising from its production [16]. One eco-friendly alternative to Portland cement is the use of geopolymers. Geopolymers are synthesized by mixing solid aluminosilicates with alkaline activators at low temperatures (below 100 °C) [17,18]. This exciting technology also allows the use of various waste streams as raw materials [19,20] which further decreases the production cost and carbon footprint. However, despite the promising properties of these innovative binder systems the possibility of using geopolymer mortars as moisture buffer materials remains unexplored.

In this study, lightweight waste-based geopolymeric mortars were prepared using varying amounts of hydrogen peroxide (pore-forming agent) to produce very high moisture buffering eco-friendly materials, able to provide an efficient control of indoor humidity levels.

2. Experimental section

2.1. Raw materials

Geopolymers were prepared using a mixture of biomass waste (fly ash – FA) and metakaolin (MK, Argical M 1200 S, Univar) as a source of reactive silica and alumina (binder). The FA provided the main aluminosilicate source (75 wt%), while MK was used in lower amounts (25 wt%) to balance the molar ratio of the compositions.

The FA (particle size of $d_{50} = 32.63 \mu\text{m}$ and $d_{90} = 86.91 \mu\text{m}$) was derived from biomass (eucalyptus) burning in the bubbling fluidized combustor of a paper pulp plant in Portugal. The SiO₂/Al₂O₃ molar ratio of the binder was 4.56.

Construction and demolition waste (CDW) were used as the fine aggregate in the production of the mortars, and the binder:aggregate ratio was 1:1 (by weight). The CDW was collected from a typical source, according to LER 1701 [21] and Directive 2008/98/EC [22]. The residues were dried at 110 °C for 24 h and prepared via a double comminution process: jaw crushing (BB2 Retsch) followed by hammer milling (5657 Retsch). The grain size distribution curve of the fine aggregate is given in Fig. S1. After milling, a CDW fraction of 0.5–1.0 mm was selected by sieving.

For the alkaline activation, a mixture of sodium silicate (9.13 wt% Na₂O, 28.77 wt% SiO₂, 62.1 wt% H₂O; Quimiamel) and sodium hydroxide (97 wt% purity, Sigma Aldrich) was used with a weight ratio of Na₂SiO₃:NaOH = 1. The NaOH solution (10 M) was prepared by the dissolution of sodium hydroxide beads in distilled water. Hydrogen peroxide, H₂O₂ (reagent grade 30 wt%, Analar Normapur), was used as a porogenic agent, with contents of 0.15, 0.30 and 0.45 wt%.

2.2. Geopolymeric mortar preparation

The alkaline activators were previously mixed by agitation at 60 rpm for 5 min, and all solid materials (FA, MK and CDW) were added to the reactor. The mixture was maintained under agitation (60 rpm) for 10 min until complete homogenization was achieved. The H₂O₂ was then added in the appropriate amount and the mixture was homogenized for 2 min at 95 rpm [23]. In the next step, the geopolymeric mortars were transferred to prismatic molds (40 mm × 40 mm × 160 mm), cylindrical molds (22 mm × 44 mm) and circular molds (12 mm × 94 mm) for the thermal conductivity, compressive strength and MBV measurements, respectively [4]. The samples were removed from the molds after 24 h and subsequently cured at ambient temperature (~20 °C) and humidity (~68%) for 28 days. Table 1 summarizes the composition of the samples prepared in this study.

Table 1
Mix design of geopolymeric mortars (wt%).

H ₂ O ₂	FA	MK	CDW	Na ₂ SiO ₃ :NaOH
0.00	25.05	8.35	33.3	33.3
0.15	25.01	8.34	33.25	33.25
0.30	24.98	8.32	33.2	33.2
0.45	24.94	8.31	33.15	33.15

2.3. Characterization of materials

The chemical compositions of FA and MK were determined by X-ray fluorescence (XRF, X'Pert PRO MPD Philips). The values for the loss on ignition (LOI) at 1000 °C were also obtained. The mineralogical compositions of MK, FA and pulverized geopolymer specimens (cured for 28 days) were determined by X-ray diffraction (XRD, D8 Advance, Bruker) with Cu K α radiation, at 5–80°, with 0.02° steps and 10 s/step. Phase identification was carried out using dedicated software (EVA Bruker).

The compressive strength of the mortars (cured for 28 days) was determined using a universal testing machine (LR 30 K, Lloyd) running at a displacement rate of 0.5 mm/min. Five cylindrical samples, which were polished flat, of each formulation (22 mm diameter and 44 mm length) were tested and the average data was reported.

Scanning electron microscopy (SEM, SU1510 Hitachi) was used to characterize the porous microstructure of the mortars. The Brunauer–Emmer–Teller (BET) surface area and pore properties were measured by a N₂ adsorption–desorption isotherm at liquid nitrogen temperature by using (Gemini V2, Micromeritics Instrument Corporation, Norcross, GA), and utilizing N₂ as adsorbate after drying of the monolith samples (~0.1 mg) at 200 °C. Mercury intrusion porosimetry was conducted using a porosimeter (Autopore IV 9500, Micromeritics Instrument Corp., Norcross, GA). Single-intrusion data were measured to provide information on the total porosity and the pore size.

The thermal conductivity (ASTM C518-04) [24] and water absorption coefficient (NP EN 1015-18) [25] were measured using cubic specimens (40 × 40 × 40 mm³). The bulk density was measured geometrically, considering the sample weight and dimensions. Three specimens were analyzed per composition.

2.4. Moisture buffering tests

The MBV was determined through the Nordtest method [4], using a climate chamber (Fitoclima 300 EDTU Aralab). The mass variation of cylindrical samples (diameter = 90 mm and height = 10 mm) was continuously determined and the data was recorded during the cyclic variation of the moisture according to ISO 24353:2008 [26] at a constant temperature of 23 °C.

The MBV was calculated through Eq. (1):

$$MBV = \frac{\Delta m}{A \times \Delta \%RH} \quad (1)$$

where Δm is the mass variation, A is the exposed surface of the sample, and $\Delta \%RH$ is the amplitude of the humidity variation. In this study, the specimens were first pre-conditioned at 63% relative humidity for 24 h and then the humidity levels inside the chamber fluctuated between 75% (12 h) and 50% (12 h), this corresponding to middle humidity levels according to ISO 24353:2008 [26]. The humidity changes were imposed four times in order to obtain four adsorption/desorption cycles.

3. Results and discussion

3.1. Evaluation of moisture buffering performance

The mass evolution of the mortars during the cyclic variation of the ambient moisture is shown in Fig. 1. All specimens show a slight increase in mass after each adsorption/desorption cycle. Nevertheless, the weight gain is moderate and the rate of increase tends to decrease with the number of cycles. For the mortar with the highest porosity (containing 0.45 wt% H₂O₂), the specimen mass increase was 0.8 wt% after the 1st cycle and decreased to 0.2 wt% after the 4th cycle. This result indicates that these samples do not have a strong tendency toward saturation.

The mass evolution for the least porous mortar (prepared without H₂O₂) indicates a poor moisture buffer ability. In fact, the moisture adsorption and desorption rates for this mortar, shown in Fig. 2, confirm that the specimen was not efficient in terms of promoting indoor humidity control. The compositions containing different amounts of the pore-forming agent displayed distinct behaviors, as clearly demonstrated by the adsorption/desorption curves (see Fig. 1). Thus, the additional porosity promoted by the oxygen release, due to the H₂O₂ decomposition in the alkaline medium, plays a vital role in the moisture buffer ability of the mortars, with the MBV increasing from 0.80 g/m²Δ%RH (reference mortar) to 5.61 g/m²Δ%RH (higher porosity mortar). Nevertheless, the results show that desorption is less efficient than adsorption, that is, the moisture removal/desorption during exposure at 50 %

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