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Lightweight hybrid organic-inorganic geopolymers obtained using polyurethane waste

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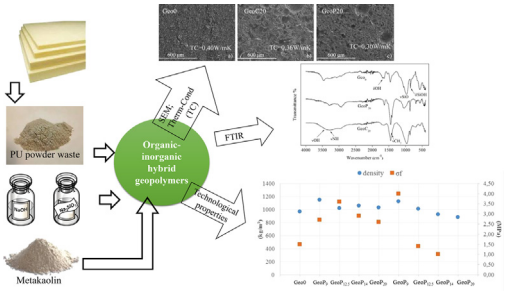
HIGHLIGHTS

- Recycling of polyurethane wastes is proposed.
- New lightweight hybrid organic-geopolymers are developed.
- Metakaolin-based geopolymers are obtained by adding polyurethane foam wastes.
- The hybrid geopolymer composites show good mechanical and thermal properties.
- Lightweight hybrid geopolymer composites can conveniently proposed as insulation building materials.

GRAPHICAL ABSTRACT



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ABSTRACT

Lightweight geopolymers from wastes with reduced thermal conductivity have become an important trend for green buildings sector. In this work, organic-inorganic hybrid geopolymers were prepared from polyurethane powders wastes (polyurethane foam and polyisocianurate foam). Chemical, physical and mechanical properties of geopolymers, manufactured with different percentages of polyurethane wastes are reported and discussed. The FTIR spectroscopy allowed to verify the influence of both polyurethane powders on the framework structure revealing H-bonds between polyurethane and inorganic phase. Flexural and compressive strengths showed that content and typology of the used polyurethane wastes affected the material behavior. The strength of hybrid geopolymers was maximized with polyisocianurate foam waste addition in a concentration lower than 14% in weight. Thermal conductivity was carried out on appropriate samples, showing that the hybrid geopolymers exhibit satisfactory isolation properties. The new materials proposed in this work represents an innovative solution, enhancing the thermal resistance of the buildings by using a waste intended for landfill.

1. Introduction

Polyurethanes (PUs) represent an important class of both thermoplastic and thermoset polymers, characterized by the urethane group as a common feature. Their mechanical, thermal and chemical properties can be tailored by the reaction of various polyols and polyisocyanates [1]. They can be considered as copolymers constituted by alternating hard and soft segments. Diisocyanate and chain extender are the hard segments while the polyether or polyester polyol flexible chain the soft segments [2].

Polyurethanes are known for their versatility with a wide range of applications in different industries: building, automotive, sports, naval and furniture [3]. In particular, rigid polyurethane foams are the most industrially used insulators thanks to their low thermal conductivity, high strength-to-weight ratio and low cost [4].

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PUs, mainly in foams form, represent about the 7.5% (15 Mt/year) of the total plastics used worldwide [5]. Due to the environmental growing concern to landfill waste accumulation and to the tight legislation on treatment and disposal, PU wastes from district heating, end-of-life vehicles and many other sources are receiving great attention for the development of new technologies to recycling this material [6–7].

During the process of rigid polyurethane foam products, 15% of wastes is generally generated, the technology disposal of which can be landfill, incineration, and recycling [8]. Landfill and incineration are not recommended except when it cannot use recycling method [1,9]. The polyurethane waste recycling methods, comprehend energy recovery, chemical recycling and mechanical recycling [10–12]. A common recycling method for PUs foam wastes is the re-bonding process: foam wastes are milled into fine flakes, blended with a powerful binder and then formed into boards or moldings under heat and pressure [1,8]. The resulting products are generally used for insulation boards in sound proofing applications. The problem of this solution is the low mechanical and physical performances of the obtained sample. Several studies reported the application of PU wastes as fillers in the construction industry [13–14]. PU foam powder can be directly added to concrete to improve adiabatic effects, or to realize roof heat insulating layer [8]. PU foam powder has been used for the production of lightweight aggregate concrete composites [15–16]. Ben Fraj et al. [17] reported the reduction of concrete density and the increasing of porosity by addition of rigid polyurethane (PU) foam waste as coarse aggregates (8–20 mm).

To the author's knowledge no scientific study to date focuses on using PU waste to manufacture lightweight geopolymer bricks. However, knowing their potential as alternative binders, several attempts have been made on the use of geopolymers to produce mortars and concrete [18–20]. Geopolymers have received increasing interest worldwide as an alternative to ordinary Portland cement concrete due to cost increases in energy supply, requirements to reduce CO_2 emission, limited reserve of limestones and limited manufacturing growth of cement [21]. The additional motivation for exploring this alternative is attributed to its high early compressive strength, low drying shrinkage, superior durability in aggressive environment compared to Portland cement concrete, etc. [22]. Moreover, from the environmental point of view, development and production of geopolymer materials from wastes has been attracted growing importance in the last years [23]. The use of granulated blast-furnace slag and fly ash in geopolymers has been reported by many authors showing excellent mechanical and physical properties [24]. Bayer liquor and fly ash have been utilised by E.J. Jamieson and coworker [25] to obtain a new class of ambient-cured construction geopolymers with compressive strength higher than 30 MPa associated to a very low carbon footprint. An increase of ductility property of geopolymers has been obtained by T.A. Kua et al. [26] by adding various mix designs of spent coffee grounds, fly ash and slag.

However, there are a few papers on the production of waste-based lightweight geopolymers [27–28]. In this direction, some studies have proposed lightweight geopolymers using rice husk ash and diatomaceous earth aggregates [29] or recycling lightweight block wastes mixed with lignite fly ash [30]. They obtained mean bulk density of 880 kg/m^3 and compressive strength of 1.5 MPa in the optimized condition. Arellano Aguilar et al. [31] obtained aerated geopolymer concrete by mixing metakaolin and fly ash. The density of concretes was reduced by aeration by adding aluminium powder and in some cases by adding lightweight aggregate derived from blast furnace slag. Densities were set at 600, 900, and 1200 kg/m^3 and maximum compressive strength of about 3.5, 8, and 16 MPa was reached with these densities, respectively.

In this scenario, this study is a preliminary investigation on the possibility of utilizing PU powdered waste as raw materials in the production of hybrid geopolymers. For this research, geopolymer and PU powders were combined together to study the effect on the geopolymers properties and the feasibility of using this waste as fillers in geopolymers.

2. Experimental

2.1. Materials

Kaolin ($Al_2Si_2O_5(OH)_4$), sodium hydroxide (NaOH) and sodium trisilicate ($Na_2O_3SiO_2$) were purchased by Sigma Aldrich. Two different powders, Corafoam® PIR50 (Polyisocyanurate foam) and Corafoam® CP125 (Polyurethane foam), were provided by DUNA Corradini (Italy). The density of Corafoam® PIR50 and Corafoam® CP125 were 0.110 and 0.085 g/cm^3, respectively.

2.2. Samples preparation

Metakaolin was obtained by calcination of kaolin at 550 °C for 10 h based on the work of Ilić et al. [32]. The alkaline solution was prepared by mixing sodium hydroxide solution (6M) and sodium silicate solution (2.5 M) in a ratio of 4:1.

The preparation of geopolymers paste samples was obtained by mixing metakaolin powder and alkaline activator solution with a mass ratio of 3:1 as suggested by Wang et al. [33]. The hybrid geopolymers were obtained adding different percentages of polyurethane PU powders (0, 9, 12.5, 14, and 20 wt% with respect metakaolin), to the metakaolin-based geopolymers matrix. Resulting slurries were stirred mechanically for about 10 min at 750 rpm to reach good homogenization and then were poured in rectangular and cylindrical silicon molds and vibrated for 2 min to remove the entrapped air. The molds were left at room temperature for 24 h and successively placed in an oven at 40 °C. Demolding was carried out after 24 h and the samples were stored in a desiccator for 28 days. The so obtained samples were then fully characterized.

Depending on the quantity and type of added PU (PIR50 or CP125), the samples were coded GeoX_y where, X is the kind of PU (P for PIR50 and C for CP125) and y is the percentages of PU with respect to metakaolin.

2.3. Sample characterization

Samples chemical features were studied by FT-IR analysis, recorded on Nicolet 5700 FT-IR Spectrophotometer (Thermo Electron Corp). 32 scans between 4000 cm^{-1} and 400 cm^{-1} were averaged for each spectrum at intervals of 1 cm^{-1}. FT-IR spectra were generated by pressing 500 mg of dried sample on KBr crystal.

XRD patterns were obtained by Bruker D2 PHASER X-ray diffractometer with 1-dimentional LYNXEYE detector, using Cu-Kα radiation at 40 kV and 10 mA, at 0.2°/s scan rate (in 2θ) in the range 0–80°. RT measurements were obtained with the powdered geopolymer samples spread on a conventional glass sample holder. Powdered silicon reflections were used for 2θ calibration. The identification of crystalline phases was made under comparison with data on the JCPDS files.

Morphological observations were performed on geopolymer cross-section by a Scanning Electron Microscope (SEM) JEOL 6400 equipped with Energy Dispersive X-Ray Microanalysis System (EDS) Oxford-INCA, with Si(Li) window-less detector.

Apparent density was measured by considering the weight and the dimension of specimens. The results were the mean of three measurements. The experimental errors in the apparent density evaluations was ±0.03 g/cm^3.

Flexural strength was measured by means of three-point-bending tests using a universal testing machine (MTS model DY3X/M with a 10 kN load cell). Tests were performed controlling the displacement of the actuator with a speed of 0.05 mm/min. Young's modulus was computed from the initial slope of the load – flexural displacement curve. Flexural displacements were measured using Digital Image Correlation (DIC), which permitted to observe and to remove possible rigid body displacements of the specimens. Specimens had nominal dimensions span × base × height equal to 50 mm × 10 mm × 5 mm.

Because of limitations of our laboratory production chain of geopolymers, specimen had a maximum thickness of 6 mm. This small value required small flexural specimens and did not permit to perform standard compression tests on cubes or cylinders (e.g. according to EN-1015-11:2007 [34]). For this reason, Double Punching Test (DPT) as specified by DIN 18555-9:1998 [35–36] were performed using the universal testing machine employed for bending tests, controlling the displacement. Tests were performed on disks of diameter 50 mm and thickness 6 mm. The steel punches had a diameter of 20 mm and the displacement speed was 0.45 mm/min. Punching strength σ_{DPT} was obtained by dividing the ultimate load by the area of the punch.

The thermal conductivity was measured by adopting a parameter estimation procedure [37] based on the solution of the inverse heat conduction problem within the sample. A thin copper plate, connected to a power supply, was used as a plane