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Research paper

Fabrication, microstructural and mechanical characterization of Luffa Cylindrical Fibre - Reinforced geopolymer composite

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ABSTRACT

This study reports on the preparation, microstructure, density and mechanical properties of new geopolymer composites (LG-composite) that are unidirectionally and randomly reinforced with 10 vol% natural Luffa Cylindical fibres (LCF). The geopolymer matrix was synthesized from metakaolin activated with sodium silicate and sodium hydroxide solutions. A greater amount of geopolymer gel was formed after introducing the LCF into the geopolymer matrix. As a result of the strong alkali setting reactions (geopolymerization), the hemicelluloses and lignin present in the LCF were extracted, leaving a rough LCF surface. In this way the hydrophobicity of the rough LCF increased and helped strengthen the bonding between fibre and geopolymer matrix. Two main morphological types of crystalline objects were observed in the LG-composite due to the incorporation of the extracted constituents of LCF: fibre-like crystals, or whiskers, and fine crystals of cubic shape. These crystals assist in crack healing while increasing the tensile strength and toughness of the composite. In terms of mechanical properties, it is found that by introducing LCF as reinforcement, the compressive and flexural strengths of the end geopolymeric products respectively increase from 13 MP and 3.4 MPa up to 31 MPa and 14.2 MPa. The LCF-reinforced geopolymer composite exhibited ductile-like failure with a strain hardening Modulus of 72 MPa, unlike the brittle matrix. In addition, the bulk density decreases from 1.5 g/cm^3 to 1.38 g/cm^3. A preliminary aging study has demonstrated that the LG-composite shows no significant deterioration in mechanical performance over a duration of 20 months.

1. Introduction

Composite materials with high mechanical performance have received much attention in recent years (Alomayri et al., 2014). These materials cannot always be synthesized by conventional monolithic brittle ceramics or cement (MacKenzie and Welter, 2014) because they have poor tensile and flexural properties. One solution to this problem is to use an inorganic matrix, such as a ceramic or cement, in combination with fibres to overcome the problem of brittle failure and produce a more acceptable failure mode (Nematollahi et al., 2014; Assaedi et al., 2015). Since ceramics require high processing temperatures, they can be prepared only in combination with inorganic fibres. Furthermore, the high cost and the special requirements needed for ceramic processing are valid reasons for proposing other similar materials involving low cost processing and simple preparation methods (Hung et al., 2008; El-Eswed et al., 2017; Komnitsas et al., 2013).

Although ordinary cements have good mechanical properties, there may be limitations regarding their adequacy for some applications in the construction field. For example, the surface deterioration of concrete due to the chemical attack of some liquids causes a serious problem for its durability (Pernica et al., 2010). Moreover, increasing awareness of greenhouse emissions resulting from Portland cement manufacturing makes it necessary to look for an alternative "green" material —around 5% of global CO_2 emissions arise from Portland cement production (Huntzinger and Eatmon, 2009), and studies (Roy, 1999; Alzeer and MacKenzie, 2013) indicate that producing 1 ton of

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ordinary Portland cement entails emitting approximately one ton of CO_2.

A new class of alternative matrix materials that are free of such drawbacks can be found in geopolymers (Roy, 1999; Barbosa et al., 2000; Huntzinger and Eatmon, 2009; Alzeer and MacKenzie, 2013). The curing, setting and hardening of these inorganic compounds occur at low temperature. Subsequently, a low-temperature ceramic-like matrix is formed with the typical temperature resistance and strength of ceramics (Komnitsas et al., 2007; Alshaaer, 2013; Hajjaji et al., 2013; Alshaaer et al., 2014). Processing geopolymers at low temperatures makes them suitable as matrices for a wide range of fibres, including organic fibres (Kriven et al., 2003). At the same time, since their production entails much lower CO_2 emission —approximately 62%–66% of that of Portland cement (Davidovits, 2015)— they are considered green cements (Duxson et al., 2007; Essaidi et al., 2014).

The geopolymer microstructure comprises an amorphous, three-dimensional network resulting from the reaction of either geological mineral or industrial wastes with an alkali silicate solution. Several studies (Davidovits, 1982, 2005; El- Eswed et al., 2009; Zaharaki et al., 2010) have explored the kinetics of this reaction and identify the solution chemistry, and a reaction pathway for geopolymerization involving polycondensation of hypothetical monomers, i.e. orthosialate ions, has been proposed by Davidovits (2005). Hard and stable materials with three-dimensional networks, similar to hydroxysodalite, feldspathoid or zeolite, are formed due to these reactions (Alshaaer et al., 2002). The amorphous or microcrystalline materials produced are composed of SiO_4 and AlO_4 tetrahedra linked alternately by sharing all the oxygen atoms. When aluminum is four coordinated to oxygen atoms, a negative charge is created; therefore, the presence of cations such as Na^+ and K^+ is essential to balance the negative charge of Al in the fourfold coordination (Davidovits, 1982). Despite the promising mechanical properties of geopolymers, the material's matrix is characterized by brittle failure readily taking place under applied force. Therefore, applications of this material in construction technology will benefit significantly from an enhancement of the mechanical properties such as flexural strength and toughness. This goal may be accomplished by developing "environment-friendly materials" through the use of natural fibres in a fibre-reinforced geopolymer composite (Nematollahi et al., 2014).

In principle, the advantages of using natural fibres in composites include low density, high flexural strength, flexibility and high elastic modulus (Ferreira et al., 2010; Davidovits, 2011; Bohlooli et al., 2012; Assaedi et al., 2015). Further advantages derive from the biodegradable, renewable and recyclable nature of natural fibres (Hammell et al., 2000; Assaedi et al., 2015, 2016). Based on these characteristics, natural fibres become an attractive reinforcement for composite systems. For instance, cellulose, cotton fibres, bamboo and flax are utilized to improve the mechanical performance of various composite systems (Low et al., 2009; Rahman et al., 2011).

This study aims to develop LCF-geopolymer composites with attractive physical and mechanical properties. The sponge gourd, the fruit of LCF, has a ligneous netting system in which the fibrous cords are disposed in a multidirectional array, forming a natural mat. Previous studies by Gianpietro et al. (2000), NagarajaGanesh and Muralikannan (2016), Seki et al. (2012), and Akhtar et al. (2003) involved LCF composed of 60% cellulose, 30% hemicellulose and 10% lignin. In this study, XRD and SEM were used to investigate the chemical composition, morphology and microstructure of geopolymer/LCF composites.

2. Experimental

2.1. Materials

Geopolymer cement was prepared using kaolinitic soil from natural deposit, as well as sodium silicate (Na_2SiO_3) and sodium hydroxide. The kaolinitic soil sample was collected from a deposit in Riyadh region (Saudi Arabia) with the assistance of Saudi Ceramic Company. The estimated kaolinite mass percentage in the precursor is 92%.

Table 1
Chemical analysis of kaolinite.

Compound	Composition%
MnO	0.3
Cr_2O_3	0.4
CaO	1.1
K_2O	0.1
P_2O_5	0.9
Fe_2O_3	9.3
Al_2O_3	22.5
SiO_2	38.1
TiO_2	14.2
LOI	13.1

The powdered clay (grain size less than 60 µm) was heated at 750 °C for 4 h in a laboratory furnace to obtain its respective metakaolinite. The chemical composition of the sample was determined by X-ray Fluorescence (XRF) (Bruker System S4 Pioneer) and is given in Table 1.

Sodium silicate (Na_2SiO_3) and sodium hydroxide (NaOH) solutions were used as alkaline activators for the dissolution of aluminosilicate phases. The sodium silicate solution (Merck, Germany) contained 27% SiO_2 and 8% Na_2O. The hydroxide solution, at a concentration of 6 M, was prepared using sodium hydroxide (NaOH) (Merck) and distilled water.

The luffa sponge, Fig. 1A, was obtained from dried fruits of Luffa Cylindrical Fibre (LCF). Only the outer core was used as the natural fibre matting. The homogeneous outer core of the sponge was cut into layers of fibre. The obtained fibre mats were washed with distilled water and dried at 80 °C during 24 h. Each fibre mat has two different sides: one is made up of unidirectional fibres as shown in Fig. 1B, while random fibres exist on the other side of the same mat, as in Fig. 1C.

2.2. Preparation of geopolymer cement

The ratios used during the alkaline activation process were: SiO_2 (in sodium silicate solution)/Al_2O_3 (in metakaolinite) molar ratio of 1, and Na_2O (in sodium silicate and NaOH solutions)/Al_2O_3 (in metakaolinite) molar ratio of 1. The H_2O/Na_2O molar ratio was 13. The solution of Na_2SiO_3, NaOH and H2O was mechanically mixed for 1 min. Metakaolinite was mixed with this alkaline solution for 15 min. The mixture was poured into a polycarbonate mold (16 cm × 10 cm × 2 cm), and then the mold was sealed and cured at 40 °C for one day. The geopolymer specimen was cut into smaller pieces (16 cm × 2 cm × 2 cm) using a diamond saw.

2.3. Preparation of LCF-reinforced geopolymer composite (LG-composite)

Laminates in this work were fabricated by means of the hand lay-up technique, illustrated in Fig. 2. The dimensions of each laminate are 16 cm × 10 cm × 1 cm. The amount of geopolymer used per layer was 56 g/(16 cm × 10 cm). Ten LCF mats (2D) with a fibre density of 4 g/

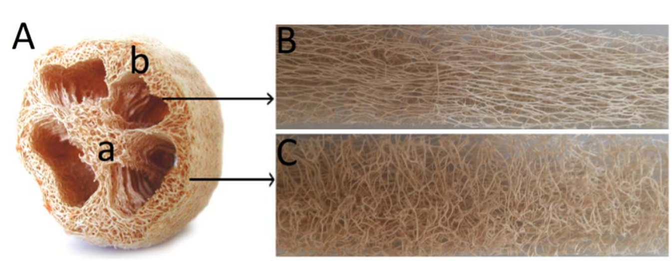

Fig. 1. The LCF fruit (A), the inner fibre core (a) and the outer core (b), unidirectional fibre (B), and random fibre (C).