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Electrically conductive cement mortar: Incorporating rice husk-derived high-surface-area graphene

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HIGHLIGHTS

- Electrical properties of GRH with High SSA from rice husks were evaluated.
- GRH mortar showed a moderate conductivity among other carbon nanomaterials.
- Mortar with 1D carbon inclusions showed much better conductivity than those of 2Ds.

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ABSTRACT

This study examines the electrical properties of cement mortar using rice husk-derived graphenes (GRHs). It investigates enhanced types of graphene-like materials synthesized by extracting rice husk from agricultural waste. Control specimens in this study were chosen by scale and shape of their nano component to compare the performance of GRH inside cement mortar. This group encompassed with multi-walled carbon nanotubes (MWCNTs), MWCNTs decorated with COOH, carbon nanofibers with relatively higher aspect ratio because of their 1D structure. The other members of the comparison group were xGnP M15, xGnP C650, and GRH, all of which exhibited 2D planar or corrugated-planar structures. The electrical conductivity was proven to be better in 1D rather than in 2D structured inclusions. The electrical performance of the GRH composite, measured by change in volume resistivity vs. stress and strain, exhibited a moderate range similar to that of carbon fiber.

1. Introduction

When the mortar composite is deformed or stressed by an external loading condition, the conductive network inside might be changed accordingly. This change reflects variation in electrical resistance, capacitance and impedance of the mortar composite. Strain, stress and cracking under static and possibly dynamic loading conditions can thus be detected. In the past two decades, many different kinds of functional fillers for mortar and concrete have been composited in order to independently sense a perturbation of electrical properties such as carbon fiber (ozone-treated, metal-coated), carbon-coated nylon fiber, carbon black, carbon nanotubes, steel fiber and graphite powder [19]. Since these functional fillers have relatively high material costs and some environmental concerns, a cheap process using an abundant resource for reducing the price of graphene has been widely sought. Recently, green production methods that use environmentally friendly biomass precursors (*e.g.*, sugar, chitosan, alfalfa plants, and other edibles) [7–12] have been suggested. Muramatsu et al. [18] proposed an alternative method for making graphene from rice husk. Their study, using ordinary synthetic apparatus and abundant agricultural waste, suggested that low-cost graphene materials could be easily and cheaply synthesized on an industrial scale. Due to its abundance, rice husk has already received much attention as a starting material for generating high-value-added materials such as silica and porous carbon. This finding may lead to a reduction in the cost of graphene. One of the previously executed mechanical properties (compressive strength enhancement of cement mortar composite mixed with graphene oxide [16,22] and rice husk derived graphene [21]) showed ample enhancement with the aid of silica fume and superplasticizer [3] compared to ordinary cement mortar. Past research has examined electrical properties of cement mortar composites [1,2] and using graphene oxide and graphene nanoplatelet [15–17] and compared the mixture to carbon nanotube (CNT) mortar composite [13,14,20]. In this paper,

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we attempt to explore the enhancement of electrical properties of cementitious mortar using rice husk-derived graphene (GRH) and then propose a novel manufacturing process using agricultural waste (rice husks). The study focuses on the feasibility test of GRH on the electrical properties of cement composites by incorporating silica fume and polycarboxylate superplasticizer comparing the product to commercially available functional fillers.

2. Rice husk-derived graphene materials

The feasibility of producing bulk amounts of a new type of highly specific surface area corrugated paper-like graphene by activating rice husk ash using potassium hydroxide (KOH) at 550–700 °C has been demonstrated [21]. Because the end planes of GRHs (edges) with a high specific surface area were expected to provide a better bonding environment with cement molecules, the synthetic condition of producing GRH was calibrated in more detail to increase both the specific surface area and the number of the clean edges by changing the quantity of KOH and the activation temperature, and by manipulating the time taken to complete the process (Fig. 1 (a)). Ten grams of raw rice husk was thermally treated at various pretreatment temperatures ranging from 400 to 600 °C with a heating rate of 10 °C/min to obtain rice husk ash (RHA) (the yield of RHA from raw rice husk is 30%). Then 2.5 g of RHA was then mixed with 10 g of KOH (4 times the amount of RHA) and placed in an alumina pot that was fully covered with ceramic fiber wool. To chemically activate the RHA, the alumina pot was encased in a SiC melting pot and thermally treated at 850 °C for 2 h with a heating rate of 10 °C/min. The chemically activated RHA (ca. 1 g) was washed several times for 6–7 h and dried for 24 h. To evaluate the number of defects in the synthesized graphene, we measured Raman spectra using a 532 nm laser line (Nanobase, Korea). As shown in Fig. 1 (b), we observed a strong peak around 1583 cm^{-1} (E_{2g2} mode), a relatively broad peak around 1353 cm^{-1} (defect induced band, D band), and a relatively sharp peak around 2702 cm^{-1} (2D or G' band) [6]. The clear observation of the relatively strong peak around 2702 cm^{-1} compared to the intensity of the G band (see $I_{G'}/I_G$ in Table 1) signifies that the sample we produced consists of the two or three layered graphenes. The chemically activated sample exhibited particle-like morphology with large sized holes (Fig. 2 (a)). Interestingly, the basic building block consisted of corrugated-paper like graphenes (Fig. 2 (b)). In addition, we found the GRH to be a multi-layered graphene with clean edges, as previously demonstrated in Muramatsu et al. and Rhee et al. [18,21].

3. Basic electrical properties of rice husk-derived graphene

A strip consisting of GRH (40 mm × 15 mm × 0.56 mm, L × H × T) was prepared for cyclic voltammetry measurement [5] in order to evaluate both conductivity and specific capacitance (Fig. 3 (a, b)). The GHR powders were mixed with 5 wt% of polytetrafluoroethylene binder. Then, the mixture was fabricated into strip. The strip was cut into a disc-shape with a diameter of 5 mm, and then they were directly attached to the surface of glassy carbon using 5 µl of Nafion solution (0.5 wt%) (Fig. 3 (c)). The method for evaluating the electrical properties of the GRH strip was carried out by applying cyclic voltage from initial voltage, V_i to target voltage, V_λ with varying scan rates (Fig. 3 (d)). First, the target voltage was fixed at 1.0 V and then varied from 10 to 100 mV/s (Fig. 3 (d)). A two-electrode system was used to acquire electrochemical reactions by using a pair of L-shaped nickel plates as electrodes and the GRH strip sample sandwiched between the electrodes in the electrolyte. The electrochemical reactions (*redox* potentials) were then measured under different voltage scan rates so that a voltammogram could be drawn. From current–voltage (CV) curves (Fig. 3 (e)), the specific capacitances were obtained by calculating their area using the following Eq. (1).

$$C_s = \int idV/(2 \times m \times \Delta V \times SR) \quad (1)$$

$\int idV$ is the integrated area of the CV curve, m is the mass of the GRH strip, dV is the potential range and SR is the scan rate.

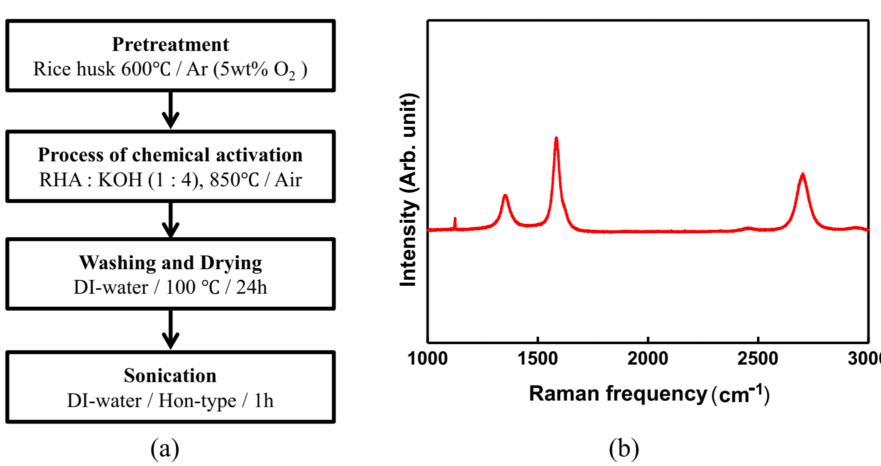

Fig. 1. (a) The experimental procedure of synthesizing graphene from rice husk and (b) Raman spectrum of rice husk-derived graphene using a 532 nm laser line.

Table 1
Summarized Raman factors of rice husk derived graphene using a 532 nm laser line.

	D-band (cm^{-1})		G-band (cm^{-1})		D′-band (cm^{-1})		G′-band (cm^{-1})		*R* value	
	υ	HWHM	υ	HWHM	υ	HWHM	υ	HWHM	(I_D/I_G)	($I_{G'}/I_G$)
GRH	1353	49.5	1583	35	1625	23.2	2702	64.6	0.66	1.21