

Facile growth of carbon nanotubes coated with carbon nanoparticles: A potential low-cost hybrid nanoadditive for improved mechanical, electrical, microstructural and crystalline properties of cement mortar matrix



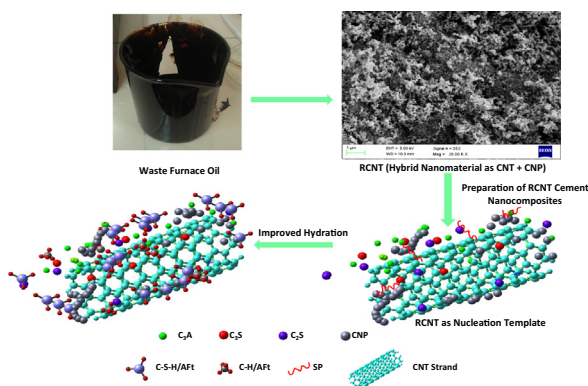
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HIGHLIGHTS

- Cost effective synthesis of RCNT using a petroleum refinery waste has been reported.
- Characterization of RCNT confirmed presence of CNTs and CNPs with diameter of 20 nm and 100 nm respectively.
- Optimized concentration of 0.50 wt% RCNT in cement matrix produced maximum strength enhancement.
- Cement matrix with 0.50 wt% RCNT had improved electrical behavior.
- Improved mechanical and electrical behaviors were attributed to good dispersion and crack-bridging effect of RCNT.

GRAPHICAL ABSTRACT



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ABSTRACT

A low-cost synthesis of Raw Carbon Nanotubes (RCNT) from petroleum refinery waste is presented. RCNT was incorporated in cement matrix at 0.125%, 0.25%, 0.50% and 1.00% by wt. of cement. Mechanical, electrical, microstructural and crystalline properties of Raw Carbon Nanotube-Cement Nanocomposites (RCNT-CNCs) were evaluated after 28 days, and compared with Pure Carbon Nanotube-Cement Nanocomposites (PCNT-CNCs) and Pure Carbon Nanoparticle-Cement Nanocomposites (PCNP-CNCs). Nanocomposites with 0.50 wt% RCNT, 1.00 wt% PCNP and 0.25 wt% PCNT respectively exhibited rise of 18%, 8% and 37% in compressive strength and 34%, 22% and 57% in flexural strength.

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1. Introduction

Since the discovery of Carbon Nanotube (CNT) in 1991 [1], the material has been applied extensively in different fields of engineering [2–7]. The CNTs exhibit exceptional mechanical properties

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such as high Young's modulus, high tensile strength, high flexibility and high bulk modulus [8–12]. The unique mechanical properties of CNT have motivated the researchers to reinforce various kinds of matrices [13–23]. Although CNTs have been extensively studied in polymeric composites [19–25], their use in cementitious matrix (ceramic matrix) has remained limited. Due to the quasi-brittle behavior of cementitious materials, they are quite susceptible to cracking phenomenon. The main reason for the high brittleness of cement composites originates from the hardened cement paste which consists of hydration products such as ettringite (AFt), monosulfonate (AFm), calcium hydroxide (C-H) and calcium silicate hydrate (C-S-H) gel. Out of these, C-H, AFt and AFm usually exhibit rodlike and needle-like crystals [26]. Different reinforcements such as polymeric fibers, glass fibers and carbon fibers have been used to overcome the brittleness of cement matrix [27]. These materials exhibit their reinforcement either at macro- or micro-level and therefore cannot arrest the cracks originating at nanoscale. This consequently affects the strength of cementitious matrix.

These issues have been further overcome with the incorporation of Carbon Nanotubes (CNTs) and carbon nanofibers (CNFs) in the cementitious matrices [28–30]. Obtained results have shown remarkable improvements in strength due to crack-bridging effects of CNTs or CNFs at nanoscale and their tendency to act as nucleation sites for formation of cement hydration products [31–36]. Besides mechanical properties, improvements in electrical properties of nanocomposites have also been reported when CNTs are sufficiently dispersed [29,37,38].

However, high production cost of conventional pure CNT (PCNT) is one of the key challenges that makes its incorporation in cement matrix to be uneconomical for practical applications. Another challenge is its effective dispersion in cement matrix because of strong Vander Waals attractive forces leading to formation of CNT bundles. Synthesis of CNT has always been carried out using processed hydrocarbon precursors like Methane [39], Ethane [40,41], Acetylene [42] and light distillates such as LPG [43] using highly sophisticated equipments. Little work has been reported on mass production of Carbon Nanotubes from heavy petroleum fractions such as Furnace Oil (F.O) which is waste oil in petroleum refineries. F.O is cheapest residual oil, but has adverse affect on burners and causes blockage of filters or refractories. Therefore, it is unsuitable for its original purpose due to the presence of impurities or loss of original properties and requires proper recycling or disposal in order to avoid environmental problems. Combustion practices of this carbon-rich waste generates enormous amount of carbon soot, which is a secondary waste and needs proper disposal. Presence of a useful material like CNT in the soot produced from a waste would not only solve the issues of environmental pollution but also reduce its production cost. Synthesis of CNT is usually accompanied with formation of carbonaceous particles which are removed prior to incorporation in any matrix, consequently eliminating their chances of possible benefits when used as such. Since as-grown CNTs have simultaneous production of impurities of nano-sized carbon particles, the material can be called as Raw Carbon Nanotubes (RCNTs). Replacement of PCNT with the low-cost RCNT can surmount the obstacle of high cost of conventional CNT.

Our previous research reported preliminary utilization of RCNT in cementitious matrix at 0.1 wt% and 0.125 wt%, but various influential factors were ignored in previous study such as efficacy of RCNT in comparison to PCNT and effects of higher concentrations of RCNT [44]. Therefore, the novelty of current investigation is to compare the mechanical and electrical performance of RCNT in comparison to costly PCNT as nanoadditive in cement matrix, and other significant parameters such as influence of higher doses of RCNT in matrix, its effects on flexural strength, changes in microstructure and crystalline phase composition of nanocom-

posites as a function of type and concentration of Carbon Nanomaterial (CNM). A comparative study of these properties was carried out with PCNT-CNCs and PCNP-CNCs to check the effectiveness of low-cost RCNT.

It is proposed that the environmental effects of synthesis of RCNT using current approach will be lower than those of PCNT, because the material can be used in its native form in cement matrix without carrying out additional purification steps. This would ensure the reduction in toxic gases or chemicals in the environment. Besides, the disposal issues of waste oil from petroleum refineries would also be solved.

2. Experimental program

2.1. Materials and chemicals

Ordinary Portland Cement (Grade 43) was procured from ACC Ltd. (India) and used as a binder material. The material had a specific gravity of 3.18 and specific surface area of 234 m²/kg. Chemical properties of OPC are shown in Table 1. Indian standard sand (Grade I, II and III) following IS 650:1991, with average specific gravity of 2.64 was obtained from Tamin, Ennore (India). Equal proportion of each grade was used in the study. The specifications of the sand used are given in Table 2. The particle size distribution of standard sand used is shown in Fig. 1. A polycarboxylate ether

Table 1
Chemical properties of OPC.

Contents	Test value (%)
CaO	64.90
SiO ₂	21.49
SO ₃	0.70
Al ₂ O ₃	4.21
Fe ₂ O ₃	3.50
MgO	2.90
Insoluble residue	1.10
Total loss on ignition	1.20

Table 2
Specifications of Indian standard sand.

Grade	Type of standard sand	Particle size range (mm)	Amount used in study (%)
Grade I	Coarse	2–1 mm	33.33%
Grade II	Medium	1–0.5 mm	33.33%
Grade III	Fine	0.5–0.09 mm	33.33%

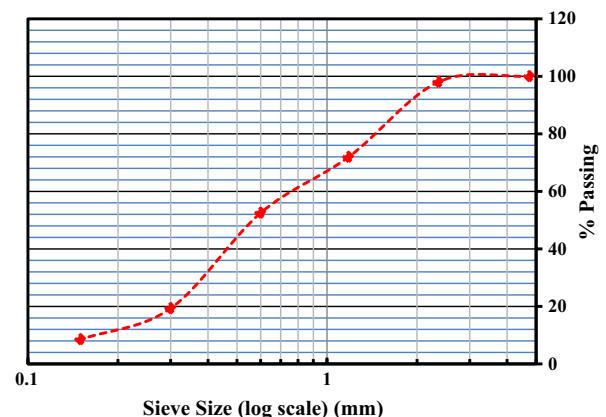


Fig. 1. Sieve analysis of fine aggregates (equal proportion of Grade I, II and III) showing particle size distribution.

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