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Effect of Undensified Silica Fume on the Dispersion of Carbon Nanotubes within a Cementitious Composite

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Abstract. The synergistic effect of multi-walled carbon nanotubes (MWCNTs) and Undensified Silica Fume (USF) on the microstructure of cementitious composites has been studied. In the current work, USF was used to enhance the dispersion of nanotubes throughout the composite and prevent the re-agglomeration of nanotubes by providing a physical barrier of particles of small size. Ultrasonication was employed to disperse MWCNTs in water in the presence of polycarboxylate-based superplasticizer (PCE) as a dispersion agent. The results indicate that incorporation of USF considerably improves the dispersion of nanotubes in the composites, with subsequent enhancement of composite packing density. This enhancement can be attributed to the synergistic effect of MWCNTs and USF in reducing the volume of pores through the cementitious composites.

Keywords: Cementitious composites, multiwall Carbon nanotubes (MWCNTs), Undensified Microsilica (USF).

1. Introduction

Since the study published Iijima in 1991 [1], carbon nanotubes (CNTs) are emerging as a promising candidate for many applications, however the agglomeration tendency of CNT material presents a considerable barrier to realising the potential offered by these nanostructures [2]. The outstanding properties of carbon nanotubes (CNTs) make them promising candidates as a reinforcing element in cementitious composites [3, 4]. Based on their mechanical and physical properties, CNTs are one of the strongest and stiffest materials known to date. Compared to mild steel, the most widely used reinforcing material for concrete [5], CNTs are about 100 times stronger, 6 times lighter, with a Young's modulus about 5 times greater, and elongation at break up to 60 times greater [5-7].

Similar to other application, dispersion of CNTs for cementitious application is however a highly challenging task, significantly affecting the mechanical properties when added to a composite. The poor wettability of nanotubes and the van der Waals interaction forces between nanotubes create bundles and agglomerates, which hinders their dispersion [8, 9]. Poor dispersion of CNTs in cementitious composites can lead to reduction of mechanical performance, as sufficient dispersion is a prerequisite for their reinforcing and crack-arresting properties [10-12]. In recent works, various approaches including mechanical and physical methods have been proposed to achieve uniform dispersion. The two main dispersion techniques are (i) CNTs added to the cement as a powder and mixed using rotary mixer [13, 14], (ii) ultrasonication technique via covalent/non-covalent bond in combination with a dispersion agent to disperse the nanotubes in aqueous liquid [11, 15]. Moreover, efforts to achieve a proper dispersion through the nano composites were extended to use the pozzolanic admixtures [16-18]. Microsilica has been extensively used as partial substitution of portland cement in cementitious composites, to improve the durability and mechanical properties of the composites[19]. Utilization of microsilica in nano cementitious composites have also been proposed as a means to improve the dispersion of nanotubes in composites. Their great effect on dispersion and densification of cementitious composite comes from their pozolanic reaction and relatevely small particle size (compared to cement grain size) [20, 21].

Although extensive efforts have been made to obtain well dispersed nanotubes in water, it has been found that when incorporated with cementitious composite, a uniform distribution of nanoparticles is may be not found due to the re-agglomeration of CNT. This paper investigates the synergistic effect of usdensified silica fume (a class of microsilica that exhibits extremely fine particle size in a range of 10-200 nm) and MWCNTs on the microsturcure of cementitious composite. Also, this study contributes to a further understanding of the mechanisms of silica particles in the prevention of re-agglomeration of MWCNTs through the composite.

2. Materials and methods

2.1. Materials

The cementitious materials used were ordinary Portland cement (OPC) meeting the requirements of British Standard BS EN 197-1[22], and undensified silica fume 940 U (with ~200 nm mean particle size, ~15m^2/g specific surface area, and >105% 7-day pozzolanic activity index). Polycarboxylate-based superplasticizer (PCE), conforming to ASTM C494 Type F[23], used to impart high workability of the mixture. Silica sand (>99.5 wt.% SiO2, ball milled and sieved to two particle size categories: 0.1–0.18mm and 0.18–0.5 mm). Commercially multiwall carbon nanotube (Cheap Tubes Inc, cat# sku-030102, Brattleboro, VT) with 8–15nm diameter, 10–50 µm length was used in this study. Multiwall carbon nanotubes as provided and undensified Silica fume are shown in Fig.1. Table 1 describes the mix proportions of the two mixes of cement mortar composites in this study.

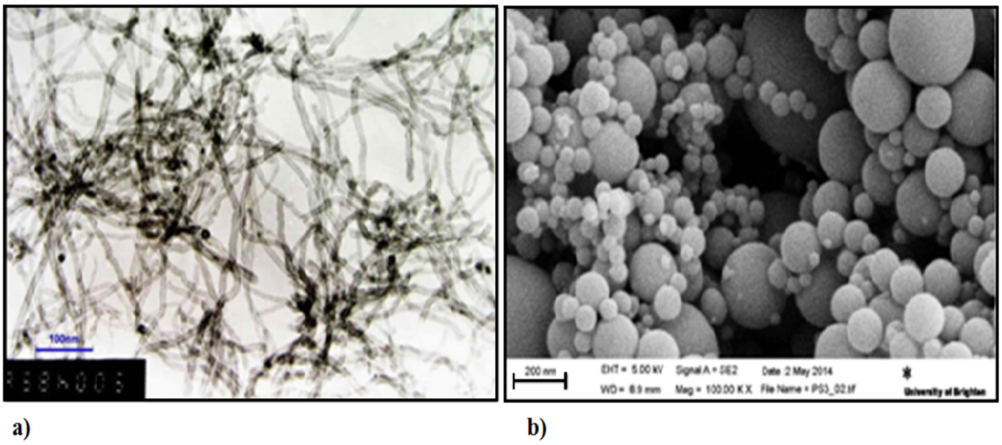

Fig. 1: Scanning electron microscope images of a)carbon nanotubes[24],and b) undensified silica fume.

Table 1 Mix proportion of cementitious composite.

Mix	Water/cement ratio	CNTs weight percentage in cement, %	Microsilica USF % of cement	Sand/ cement ratio	SP weight percentage in cement, %
PC_1	0.35	0	0	1.5	0.8
PC_2	0.35	0	0.1	1.5	0.8
CT_1	0.35	0.025	0	1.5	0.8
CT_2	0.35	0.025	0.1	1.5	0.8

2.2. MWCNTs dispersion in mixing water and Sample preparation

A polycarboxylate based superplasticiser (Molecular design of comb-shaped)[25] is an effective dispersant that typically has a charged backbone and uncharged side chains. It has been considered for maintaining composite workability, and can wrap around nanotubes in order to improve their dispersion in water and interaction with cementitious grains. MWCNTs were first dispersed in water in the presence of SP; SP-to-CNF weight ratio of 4.0:1.0 was considered[26].

CNTs were mixed with 25% of the mixing water of cementitious composite, then the suspension was sonicated using a tip ultrasonic probe (Sonic FB-705) set to deliver high intensity ultrasonic energy, which was set to 100% amplitude and operated in 20 s cycles over 5 minutes' duration, following[27].

Four mixes of cementitious martials were prepared for SEM testing: i) plain cement mortar (PC_1), ii) cement mortar with 10% of USF (PC_2), iii) cementitious composite with 0.025% of MWCNTs (CT_1), and iv) cementitious composite with 0.025% of MWCNTs and 10% of USF (CT_2). After dispersion of MWCNTs, dry ingredients including cement, USF, and silica sand were firstly mixed for 2 minutes in the Hobart planetary mixer (Hobart Model A200F) following the ASTM 305[28]. Then water and SP were added (remaining quantity) and after 2 minutes of mixing. The suspension was then added and mixed for a further 4 minutes. Cubes of 50mmx50mmx50mm, and dog-bone shaped briquette in accordance to AASHTO T132 [29] were cast in order to evaluate the compressive and tensile performance of the examined mixes (to be investigated in future work). From the fracture surfaces of cement pastes containing carbon nanotubes, typical samples were taken for microstructure characterisation. MWCNTs and Undensified microsilica characterisation

The morphology of dispersed carbon nanotubes and USF were examined using transmission electron microscopy (TEM) (Hitachi-7100 Transmission Electron Microscope (TEM) operated at 125 kV). To characterize the morphology of MWCNTs, USF, and USF/MWCNTs using TEM, samples of 0.025% MWCNTs, 10% USF (by cement weight), and 10% USF and 0.025MWCNTs were prepared in 100 ml water. Drops of suspension were placed on a grid prior to imaging and allowed to dry in air. Characterisation of the hydrated composites was performed using a high resolution, scanning electron microscope (SEM, Zeiss; model of LEO 1455VP). Prior to SEM imaging fractured surfaces were gold plated and mounted on aluminium stub using copper tape.

3. Results and discussion

TEM images reveal the morphology of the dispersed MWCNT and USF, Fig 2(a, b, and c). The diameter of the individual nanotubes is around 25 nm, and the thickness of the adsorbed layer of SP on the surface of nanotubes is about 10 nm. Spherical particles of USF were found to possess a variety of diameters ranging from 10 nm to more than 200 nm. Fig. 2c (image of CNTs/USF) revealed the synergistic action of silica fume particles in the prevention of re-agglomeration of dispersed nanotubes through the composite, the very small particle size was found in between the neighbouring nanotubes and can act to prevent the re-agglomeration through the hardened phase.

Typical SEM images (Fig. 3 a-d) show the microstructures of cement mortar, and MWCNT/ cementitious composites in the presence and absence of USF. Fig.3 (a and b) represent the effect of USF on the structure of cement paste, USF act to fill the pores system and improve the binding quality of cement hydrate[30]. The small particle size fill up the macropores (more than 50nm) and micrpores(less than 50nm) and leading to refinement the porous structures to a nano scale[31]. (Fig. 3 c-d) revealed that the dispersion states of MWCNTs through the composite was strongly affected by the presence of small particle size of undensified silica fume. Fig. 3a represents composite containing only MWNCTs, although individual nanotubes were observed in suspension through TEM images, a large areas of agglomerated carbon nanotubes through the composites were observed (indicated by the arrows). The re-agglomeration phenomena was attributed to the effect of drying of free water from the mixture during early time of mixture hydration process [32]. When the mixing water (dispersion agent and the media) began to evaporate during cement hydration process, nanotubes move towards each other and form bundles as results of surface tension forces exerted through the evaporation process. During the re-agglomeration process, cement hydration products may not have developed to warp the individual nanotubes before the re-agglomeration of nanotubes occurs.

In case of the presence of USF, the availability of smaller particle size at interfaces between the neighboured nanotubes and cement matrix were effectively hinders the re-agglomeration of dispersed nanotubes. The nanotubes were found to individually embedded through the cement hydration products (Fig. 3b).

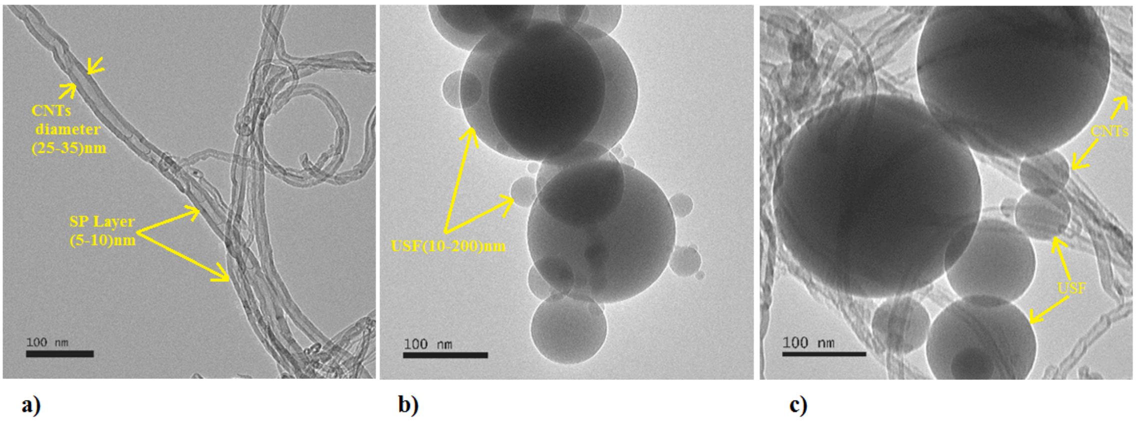

Fig.2 TEM images showing a) morphology of MWCNTs, b) diameter of USF, and c) the interaction between MWCNTs and MWCNTs.

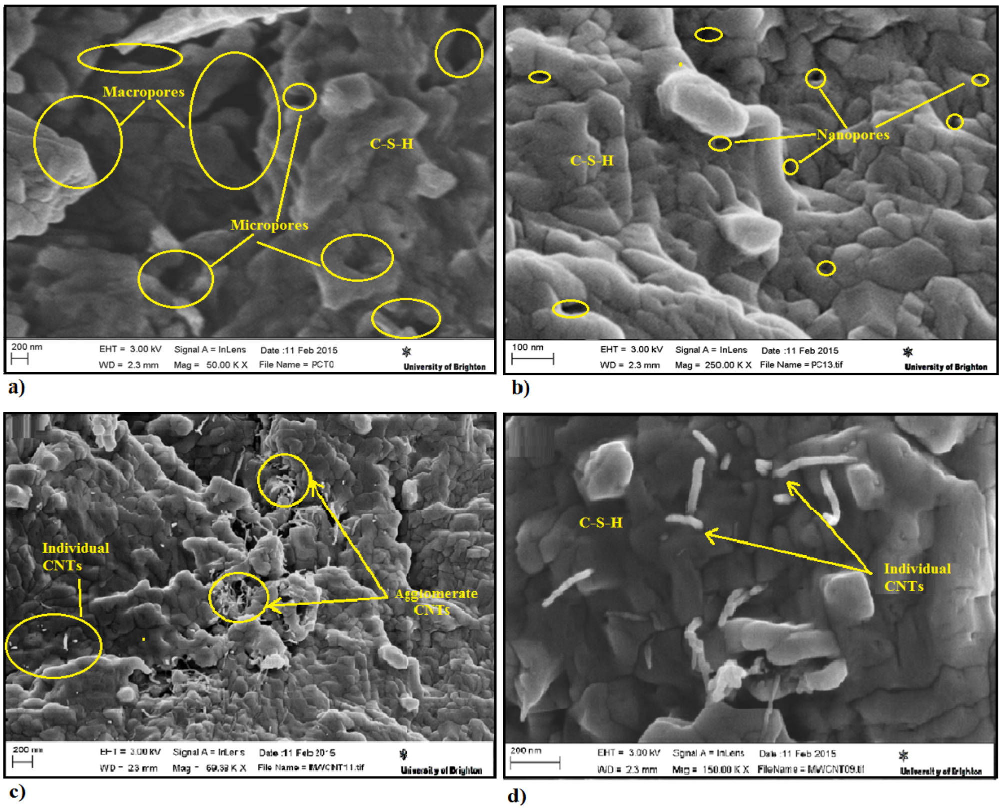

Fig 3 SEM images showing the effect of USF on the structure of plain cement mortar, and the synergistic effect of USF on dispersion of nanotubes in the cementitious matrix a) plain cement

mortar, b) cement mortar with 10% of USF, b) composite without USF, c) composite containing USF and MWCNTs.

4. Conclusion

The effect of USF on the dispersion of MWCNTs through the cementitious composite has been studied. The microstructure analysis revealed that USF particles were found to facilitate the dispersion of CNTs and improve the interfacial interaction between carbon nanotubes and the surrounding matrix. These can be attributed to the pozzolanic reaction and small particle size relative to the size of cement grain. The observed small particle size of silica fume (in range from 10 nm) was found to act as a physical barrier reducing the possibility of nanotube re-agglomeration within the cementitious composite. This should lead to an improvement in the physical performance of the composite.

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