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# Review of nanocarbon-engineered multifunctional cementitious composites

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#### ABSTRACT

As structural materials, cementitious materials are quasi-brittle and susceptible to cracking, and have no functional properties. Nanotechnology is introduced into cementitious materials to address these issues. Nano materials, especially nano carbon materials (NCMs) were found to be able to improve/modify the mechanical property, durability and functional properties of cementitious materials due to their excellent intrinsic properties and composite effects. Here, this review focuses on the recent progress of fabrication, properties, and structural applications of high-performance and multifunctional cementitious composites with NCMs including carbon nanofibers, carbon nanotubes and nano graphite platelets. The improvement/modification mechanisms of these NCMs to composites are also discussed.

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#### 1. Introduction

Cementitious materials are the most commonly and widely used construction materials for various types of infrastructures. However, cementitious materials are in general very brittle, and characterized by low tensile strength and low strain to failure [1]. Since macroscopic steel bars were used to reinforce the cementitious materials, the size of the reinforcing fillers has diminished from macro scale to micro even nano scale with the development of nanotechnology. The addition of these tiny fillers not only improves mechanical properties and durability of cementitious materials, but also endues them with such functionalities as electrical, thermal and electromagnetic properties [2,3]. For example, the fracture toughness of cementitious composites can be enhanced by 400% when nano-ZrO<sub>2</sub> is used as fillers [4]. An addition of 5% nano-Al<sub>2</sub>O<sub>3</sub> can increase the elasticity modulus of cementitious composites by 143% [5]. The electrical resistance of cementitious composites can be decreased by 45% with 5% nano-Fe<sub>2</sub>O<sub>3</sub> [5]. Nano-TiO<sub>2</sub> can endow cementitious materials with the photocatalytic effect to decompose both organic pollutants and oxides such as NO, NO<sub>2</sub> and SO<sub>2</sub> [6]. Moreover, due to their remarkable mechanical, electrical and thermal properties, excellent nanoscale effects, low density, and excellent chemical and thermal stability, nano carbon materials (NCMs) offer the possibility to develop a new generation of tailored, high-performance, and multifunctional cementitious composites[5,7–9]. Extensive research endeavors demonstrated the potential of various NCMs including carbon nanofibres (CNFs), carbon nanotubes (CNTs) and nano graphite platelets (NGPs) for enhancing/modifying cementitious materials [10–15]. For example, Li et al. observed that an addition of 0.5 wt.% CNTs can improve the flexural, compressive strength and the failure strain of the composites by 25%, 19% and 27%, respectively [11]. Raki et al. reported that CNTs can improve the Vivtorinox hardness of the early hydration of composites by 600%, the Young modulus by 227% and the flexural strength by 40% [7]. Gao et al. found that the compressive strength of composites with 0.16 wt.% CNFs is 42.7% higher than that of the plain cementitious materials [12]. Huang obtained a maximum increase of 82% in the flexural strength by adding NGPs into cementitious materials [13]. NCMs achieve the enhancement effect by nucleation, increasing the amount of C-S-H gel of high hardness, improving pore structures, controlling nanoscale cracks, improving the early strain capacity and reducing autogenous shrinkage of cementitious composites [5-14]. These mechanisms would also improve the durability of the cementitious materials [15]. In addition, due to their excellent electrical, thermal and electromagnetic properties, NCMs can impart electrical, thermal, electromagnetic and sensing properties to cementitious materials [13,14].

During the past decade, considerable research effort has been directed towards the development of cementitious composites with NCMs, and many innovative achievements have been gained in both development and application of the NCMs filled cementitious composites (NFCC). The aim of this paper is to provide a systematical review on major progresses and advances in the fabrication, properties and mechanism, and structural applications of NFCC.

### 2. Brief introduction to three types of NCMs used for fabricating NFCC

CNTs are allotropes of carbon with a hollowly cylindrical nanostructure. They are generally a few nanometers in diameter and several microns in length. According to the number of rolled layers of graphene, CNTs are categorized as single-walled CNTs (SWCNTs) and multi-walled CNTs (MWCNTs) [16]. Like CNTs, CNFs are also the guasi-one-dimensional carbon material, and their diameters are between those of CNTs and carbon fibers (CFs). The representative properties of CNTs and CNFs are listed in Table 1. NGPs are carbon-based conductive nano-particles made from graphite. Graphene oxide (GO) is a single layer of NGPs that have been oxidized to intersperse the carbon layers with oxygen molecules. The structures of NGPs are two-dimensional platelets consisting of a few to several graphene layers with an overall thickness in nanometer scale and the particle diameter ranging from submicron up to 100 µm [13,17,18]. Typical properties of NGPs are also given in Table 1.

#### 3. Dispersion of NCMs in cementitious composites

Because the strong Van der Waals forces cause the agglomeration of the nano-particles, a key issue in fabricating high-quality NFCC is to homogeneously disperse NCMs in cementitious materials. Poor dispersion will lead to the formation of defects in the matrix and limit the nano-enhancement/modification effect [27]. A lot of research work has been done to improve the dispersion of NCMs in the cementitious composites. Commonly, there are two types of methods used to disperse NCMs. One method is to mechanically separate the NCMs by adopting ultrasonic [28– 39,41–55,57–63,65–72,74–76], ball milling [56,77]or high-shear mixing technologies. The other method chemically alters the surface of the NCMs by using covalent [11,36,59,61,74]or noncovalent modification approaches [35,36,40–52,54,55,58–73,75,76]. Moreover, the mechanical methods are often used in combination with the chemical methods [10,14,29–34].

Recently, researchers have proposed some novel approaches to solve the dispersion issue of NCMs. The primary approach is to adopt commonly used water reducing admixtures (including plasticizers and superplasticizers) as surfactants. The research at the National Research Council Canada has shown that a small amount of CNTs can be dispersed by ultrasonication in the water containing 5% superplasticizer [14]. Shah et al. also achieved an effective dispersion of MWCNTs with different lengths and concentrations in cementitious materials by applying polycarboxylate-based superplasticizers [37–39]. Yazdanbakhsh et al. performed a three-dimensional simulation study, and stated that only when

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Properties of NCMs	[1 13 17-26]

Property	CNTs		CNFs	NGPs
	MWCNTs	SWCNTs		
Elastic modulus/TPa	0.3-1	1	0.4-0.6	1 (in-plane)
Strength/GPa	10-60	50-500	2.7-7.0	10-20
Electrical resistivity $/\mu\Omega$ cm	5-50		55	50 (in-plane)
Dimensions	Diameter: 2-30 nm	Diameter: 0.75-3 nm	Diameter: 50-200 nm	Diameter: 1–20 µm
	Length: 0.1–50 μm	Length: 1–50 µm	Length: 50–100 µm	Thickness: ~30 nm
Surface area/m <sup>2</sup> /g	>400	- ·	~200	~2630
Aspect ratio	~1000		100-500	50-300

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