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Modelling mechanical behavior of cementitious material incorporating CNTs using design of experiments

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HIGHLIGHTS

- Effect of dispersion protocol when adding CNTs to cementitious materials is assessed.
- Design of experiments is used to perform the study and examine mechanical properties of blocks.
- Factors examined include wet mixing, sonication exposure time, whisking and CNTs concentration.
- Response surface methodology is used to model the experimentally determined compressive strength.
- The model can be used to determine the optimum methodology to disperse CNTs in cement composites.

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ABSTRACT

One of the primary factors affecting the effectiveness of nanomaterials in enhancing cement composites properties is its ability to disperse efficiently without aggregation. To-date a number of methods have been proposed to increase the dispersion of CNTs in the mixes. Studies have shown that the response of cement-based materials to CNT addition can vary depending on the dispersion mechanism employed. Despite the need, there is no standardized methodology used to disperse CNTs, and consequently, it is hard to model the behavior. This study focused on using design of experiments (DOE) to assess the effect of dispersion protocol on the characteristics of cement-based composites. Experiments were carried out with different procedures to compare the effect of wet and dry mixing of CNTs, sonication exposure time, whisking and CNTs concentration on the mechanical properties of mortar blocks. Response Surface Methodology (RSM) was used to model the experimentally determined compressive strength. The model was further validated by comparing its predictions with other experimental scenarios. The model can be used to determine the optimum methodology to disperse CNTs in cement composites. To compare experimental data, there is a need to provide the exact details of all steps involved in a dispersion protocol that is captured by the proposed model. Following the suggested protocol for specimen preparation can ensure achieving a high level of dispersion and superior performance.

1. Introduction

On a yearly basis, the world is witnessing an ever-growing trend in the construction of mega structures, be it taller buildings, or deeper and larger tunnels. In all these mega structures the need for stronger building materials is vital to their existence and durability. The most common of these building materials has long been the use of concrete, the properties of which depend on the mix design, materials used in the concrete and method used to prepare the concrete. Providing the required large quantities of concrete require high quality control during production and following definite protocols. Concrete has been reinforced with different materials to increase its tensile and flexural strengths, from ancient civilizations using straw in sun-dried mud bricks [24] to the use of steel in most present buildings. Although these reinforcing materials provide good crack resistance that can increase strength and life of the concrete, the cracks are reduced in the micrometer range (or larger) and does little in the nanometer-size (<100 nm) range [4,11].

In recent times, the potential use of nanoparticles in the preparation of concrete to reduce nano-sized cracks has gained significant attention [8]. The introduction of nanomaterials has been

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shown to increase strength and durability of concrete and prolong the service life [16,13]. Despite the promising improvements in concrete quality, there has been mixed results regarding nanotube-induced-concrete properties in the literature. One of the fundamental requirements, but with the largest difficulty, for improving concrete quality through the use of nanomaterials, such as carbon nanotube (CNT) or nanoparticles, is proper dispersion of the nanomaterials. At the nano-size level, materials tend to agglomerate due to the high attractive forces between them, the high length to diameter ratio and the large surface area [2] resulting in poor dispersion. Ineffectively dispersing CNTs in cement-based materials has been shown to cause the formation of defect sites and reduce the efficiency of CNT-added cement materials. Although there are number of methods commonly used to disperse the CNTs during their use in concrete preparation, such as sonification, addition of surfactants and other admixtures, chemical functionalization of the CNTs, grinding of the CNTs to name a few, each method tends to provide varying ranges of concrete properties [8,19,22]. Collins et al. [3] found that the addition of polycarboxylate admixture increased the compressive strength of their CNT-OPC paste by 25% when compared to without the admixture, concluding that the addition of the admixture was responsible for improved CNT dispersion in the paste, and subsequently an increase in the compressive strength. A study by Peyvandi et al. [16] found that the addition of carbon nano-fibers improved the physical properties of concrete; however, their carbon nanofiber preparation and dispersion in the mixing water comprised of elaborate steps that included the addition of polyacrylic acid, sonification, microwave radiation, stirring and the addition of superplastisizer to the nanofiber mix. Parveen et al. [15] determined the effects of CNT-reinforced cement composites with the use of a dispersing agent and found that proper dispersion increased mortar flexural modulus by 72% and 28 days compressive strength by 19%. Siddique and Mehta [17] presented an extensive review on the effects of carbon nanotubes in the properties of cement mortars. The authors concluded that based on information in the literature, the compressive strength of mortars were the highest with CNT content of 1% by weight; however, higher flexural strength was increased at a higher CNT content. In addition to the structural strength of the mortars, the porosity and total pore volume of concrete decreased with the addition of CNTs which resulted in lowering shrinkage values [17]. In their research Liew et al. [7] used an aromatic modified polyethylene glycol ether and polyvinylpyrrolidone (PVP) to disperse 0.1% wt CNTs and found increases in compressive and flexural strengths of CNT/cement composites as high as 17.3% and 16.3%, respectively, when PVP was used; the addition of modified polyethylene glycol ether only provided limited increase in strength.

The standardization of methods to disperse CNTs for use in cement-based materials is of vital importance to the research community as it will allow researchers and laboratories to compare their work and results without bias. Tantra et al. [22] highlighted the importance of standardizing methods commonly used to disperse nanomaterials and also standardizing the analytical technique that will be used to determine the degree of dispersion. Although the work by Tantra et al. [22] was carried out using titanium dioxide, the same importance can be applied to all nanomaterials. Hartmann et al. [6] provided a review of different techniques currently being employed to disperse nanoparticles in water and suggested a step-wise approach towards developing a definite protocol in the future. In addition to mechanical dispersion, the use of surfactants to assist with CNT dispersion depends on a number of factors which also complicates standardization techniques and comparison of cement composite characteristics. In their study, Tourozzi et al. [23] emphasized the importance of different dispersion processes used, as each method is capable of producing CNTs of varying size distributions. These varying size distributions in CNTs will lead to different cement properties. Metaxa et al. [10] developed a methodology to produce highly concentrated CNTs using different processes which lead to a fivefold increase in the concentration of the CNT suspension. In addition to the complexities arising from the method of dispersion, standardization of analytical technique to assess dispersion quality is also vaguely defined in the literature, and varies between research groups. Some of the methods commonly used to describe degree of dispersion include particle size [14], imaging techniques such as using scanning electron microscope (SEM) [21] and non-imaging techniques [20]. Liew et al. [8] provides an extensive overview of carbon nanotubes use for reinforcing cementitious composites and discussed the impact of nanotubes on the workability, hydration process and autogenous shrinkage of cement paste.

This paper investigates the effects of different CNT dispersion mechanisms on the mortar properties to determine the most influential protocols/parameters during mixture preparation. The aim of this study is to model the influence of various dispersion protocols on the mechanical performance of the mortar using Design of Experiments (DOE) and Response Surface Methodology (RSM). From the results, an optimum procedure can be recommended for standardization. Recommended procedure will enhance the mechanical compressive strength and minimize the agglomeration of the CNTs in the mortar mixes.

2. Materials and methods

The concrete prepared and tested in this study was composed of Ordinary Portland cement (OPC), crushed sand and CNTs. OPC having a surface area of 355 m^2/kg on the Blaine test was used for the concrete. Crushed sands, with particle sizes in the range of 0.15–4.75 mm and specific gravity of 2.65 were used as fine aggregate. The crushed sand had water absorption of 5% based on the ASTM test. Industrial grade multiwall carbon nanotubes with an 88+% purity having an outside diameter between 20–40 nm, an inner diameter between 5 and 10 nm and length varying between 10 and 30 μm were used in the study. MWCNTs were purchased from Nanostructured and Amorphous Materials, Inc. The binder mix consisted of Type 1 Portland cement, crushed sand and water.

2.1. Design of experiments

Various samples were prepared using different methodologies based on experimental design to predict the effect of the dispersion protocol on the mechanical properties of the concrete mix. Experimental design is an important tool that can be used for product or process development and improvement [9]. In this study the effect of five control factors on the response, compressive strength of concrete, was investigated. All continuous variables were studied at 3 levels while categorical variables at two levels only. 3 out of 5 factors, CNT%, curing period and sonication time, are continuous while last two, wet mixing and whisking, are categorical. Control factors levels used in this designed experiments were: % of CNTs added (0%, 0.5%, 1%), length of curing period/age (7, 14, 28 days) and initial pre-wetting of CNTs versus dry mixing (Wet/Dry). In case of wet mixing, sonication exposure time (0, 5, 20 min) and whisking the CNT mixture or applying a pulsating electrical energy to initially disperse CNTs (Yes/No), were also examined. Table 1 below summarizes factors considered along with number of levels and level settings.

In this multifactor experiment, not all factors levels can be examined under all levels. Fig. 1 illustrates the DOE utilized. 39 sets of experiments were replicated 3 times totaling 117 runs. In the illustrated design some factors are arranged in a factorial layout while others are nested. In each complete trial or replication of the factorial experiment all possible combinations of the level of the factors should investigated. Since some factors levels depends on whether wet mixing is utilized, the levels of some factors are not identical for different levels of another factor. For those factors a nesting or hierarchical design was utilized. The levels of whisking factor, and sonication time were nested under the levels of wet mixing, whereas CNT%, sonication exposure time and wet mixing were arranged in a factorial layout. Thus a nested- factorial design was utilized. The interaction between whisking factor or sonication time and pre-wetting (Wet/Dry mix) cannot be captured due to this hierarchal dependency.

Minitab® 17.2.1 software is used create and design the DOE illustrated in Fig. 1. Table 2 summarizes the list of 39 sets of mixtures that were prepared. Experimental results along with design matrix are tabulated. The order in which the experiments were performed was selected at random so that this design is completely randomized. The compressive strength of each of the samples was measured and evaluated for each case as the response variable.