Influence of Carbon Nanotubes (CNTs) in the Cement Composites

Ba Rahma Ahmed*, Al-Jaberi Hussein, Dahi Saleh and Raizal S. M. Rashid

Civil Engineering Department, Universiti Putra Malaysia (UPM), Selangor, 43400, Malaysia

*ahmed.78288@gmail.com

Abstract: Throughout the construction life, nanoparticles can change and modify the properties and functions of cement-based materials. The traditional cement materials have weak sustainability characteristics insufficient to develop high quality performance structures. Besides that, modifications of compressive and tensile strength and durability durations of cement composites are required. Consequently, a suitable range of Carbon nanotubes (CNTs) concentrations were applied to improve these certain properties. This review presents analysis of the earlier techniques of adding CNTs to cement composites and its influence to mechanical properties and durability. The detailed interactions of CNTs with cement composites, dispersion of CNTs, durability and mechanical performance measurements are addressed from several researches. CNTs addition contributes in adjustment of cement composite microstructures, hence improving the functional components and mechanical properties. If a proper dispersion of CNTs achieved, hydrate products of cement including C-S-H and Ca(OH)2 interact with active CNTs, resulting in decrement of porosity of Portland cement composites which filled up by CNTs proportions leading to increase the mechanical strength. Therefore, this paper present state of the art regarding applications of the additive CNTs in cement composite, the reviewing results of CNTs-cements shows a positive influence on mechanical strength and durability.

1. Introduction

The next scientific revolution will be nanotechnology which is nowadays widely involved in several construction and structural industries. Nanotechnology play a key role for developing many ideas of manipulating and controlling structural matters to satisfy the need of people in the future construction projects. Nanotechnology can easily modify the structural materials in addition to its ability to analysis and characterize the materials at Nano scale [1].

In the field of cement composites there are some nanoparticles incorporated into cementitious materials whereas these nanoparticles are contributed one way or another to enhance the properties and the functions of cement composites. Carbon nanotubes (CNTs) are one of the nanoparticles additions that have effects and changes on cementitious composites which eventually lead to improvement in construction industry.

CNTs mainly consists of graphene sheets rolled up as cylindrical tubes where it can be single or multiple depending on the number of internal layers. Single-walled carbon nanotubes (SWCNT) contains only one of graphene sheets with outer diameter of 5–20 nm, while multi-walled carbon nanotubes (MWCNT) have more graphene sheets with approximate outer diameter up to 100 nm [2].
CNTs graphene sheets structure have exceptional extraordinary characteristics which allow them to be considered as important for developing the cement composites. CNTs possess many desirable thermal, mechanical, and physical properties which making it to be likely close to reinforcing materials [2]. Despite the CNTs density is so much lower than steel or glass fiber, the elastic modulus of CNTs is measured in tera pascal ranges and the yield strength approximately in the range of 20–60 GPa. Moreover, CNTs give a high thermal conductivity [3].

Cement composites containing CNTs shows very good response when subjected to stress, whereas some other nanoparticles change with respect of stress levels [3]. Adding CNTs into cement composites enhance and increase the compressive strength as well as the flexural strength comparing to the normal compressive and flexural strength that obtained from plain cement composites.

CNTs-cement composites have an obvious improvement in the microstructure of the composites, where the smaller size of CNTs acting as filler, therefore the bonding between the hydration products and the surface of the CNTs is much improved. Thus, the composites will experience low porosity and then will lead to delay the microcracks initiation [4].

The demand for smart structures able to give a strong response for any external effects is growing every year, where many researchers focused on developing multifunctional materials having high structural properties and combining some functional applications [1].

As it is known, cement composites provide functional properties which play a potential role in construction application. However, Modification is required for properties enhancement including compressive strength, flexural strength and microstructures as well as durability.

Carbon nanotubes (CNT) are one of the constructive additions which can be added to cement composites in order to create essential materials. The addition of CNTs to cement matrix is a new line to obtain good quality materials with high efficiency and wider application area [5].

In the current article, different studies on the addition of CNTs into cement composites and the influence and changes in the properties and behavior of CNTs-cement based composites in terms of compressive, flexural strength, microstructure, and durability have been investigated.

2. Properties of carbon nanotubes

2.1 Electrical properties
Previous experiments have shown that the carbon nano-tubes have unique electrical properties, where their electrical resistance is very low [6]. These unique properties are largely resulted from (1-D) character and a graphite, which has a strange microelectronic structure. There are defects in the structure of the material resulted by various reasons, such as corn vibration around itself in the nucleus of the material or a defect in the main structure of the crystal (atom of impurity). Therefore, the resistance result when an electrical cargo in the atom collide with these defects. Electrons deviate from their pathway due to these collisions. On the other hand, the electrons that located in the CNT aren't appreciably dispersed due to the tiny diameter of electrons and the ratio of their length to a very large diameter of up to thousands or even more [7].

2.2 Thermal properties
Carbon nano-tubes have unique thermal properties, which have a cold texture like the metal along the tube, but at the same time, one side of the horizontal axis of the nanotubes is heat-insulating as a wood. The earlier experiments have shown that the single walled carbon nano-tubes have ability to absorb and transport the heat at room temperature. Generally, CNT considered a good conductor of heat even better than diamond [8]. Phonons in nanotubes can specify the strength of the thermal conductivity for the material [9]. The main information about the class of transporters and conductivity techniques can get it by measuring of the thermo-electric power of CNT.

2.3 Mechanical and strength properties
One of the most powerful fibers and materials that had been discovered in terms of elasticity, high hardness and tensile-strength are carbon nano-tubes. This power is due to the strength of internal bonds between carbon atoms (C-C, sp2) [10]. So, the mechanical properties such as flexible response, yield strength and breaking for the carbon nano-tubes need further scientific investigation. The CNT have the elongation percentage between twenty to thirty percent before reaching to the failure mode, with a Young coefficient of 1.39 TPa and with a tensile-strength of 100 gigapascals (GPa) or even higher. In contrast, 200 GPa and 1-2 GPa are the Young’s modulus and tensile-strength of steel respectively. Also, the effective strength of carbon nano-tubes bundles (multi-walled) have small mitigation of GPa’s, due to poor shear interactions between both of tubes and contiguous shells, even though the single shells of the CNT have high strength [11]. The high power of electron radiation has applied to the CNT, which led to intertwines between tubes and interior shells, therefore the force of multi walled CNT and double walled CNT bundles increased to 60 and 17 gigapascal consecutively [12].

3. Dispersion of CNTs
One of the challenges that can strongly affect the properties of CNTs cement composites is CNTs dispersion. The presence of the attractive forces which known as van der Waals allow CNTs to agglomerate and afford a strong tendency to combine together, these forces are gained from the extended polarizable p-electron structure. The result of this agglomeration and inadequate linkage of CNTs to cement composites restrained the usage of this system. In addition, some materials have hydrophobic tendency which hinders content incorporation and thus affect the identical distribution of CNTs in cement matrix. A few studies reported that this challenge of poor or insufficient dispersion of CNTs will result in no improvement within the properties of cement composites with addition of CNTs. Ibarra et al. [13] highlighted that unequal distribution of CNTs content in the matrix result in regression of the mechanical properties. Konsta-Gdoutos et al. [14] mentioned that not using any surfactant while spreading of CNTs in matrix, MWCNTs showed poor distribution and dispersion where high agglomerates and batches of CNTs were observed. The process of dividing agglomerates of CNTs in the matrix is very tough and relies on some factors such as attractive force, CNTs length, fraction volume, viscosity and density of matrix. Some approaches are used to overcome the agglomeration and dispersion of CNTs to achieve homogeneous composites such as surfactants, acids functionalizing, functionalized with noncovalent, microwave, sonication and grinding [15].

Li et al. [16] stated some development of reaction with CSH and CNTs dispensability within the composites by the interaction with SiO2 in the process of sol-gel. Stynoski et al. [17] stated that when SiO2 covered CNTs in the procedure of mixing, the dispersion of CNTs could be strongly improved in the matrix. The interaction of CNTs with SiO2 allows them to act as seeds of nucleation where the hydration is quite speedy resulting in strong effect on the C3S hydration when CNTs are completely coated with reactive SiO2. This cover of SiO2 could improve the inclusion of CNTs in the composites as highlighted by Butters et al. [18]. Sanchez [19] stated that some elements can be used to assist dispersion of SWCNTs in the matrix. He used sonication or ethanol in the dispersion process of SWCNTs which results in some improvement. MWCNTs are different form where the dispersion of it needs some extra procedure. Musso et al. [20] reported that the dispersion of MWCNTs can be achieved by minimizing the aggregate size of MWCNTs where some elements of ultrasonic probe used for dispersing of MWCNTs in acetone. The result was some improvement on the dispersion of MWCNTs in the cement matrix.

4. Compressive Strength
Compressive strength can be defined as the ability of material to sustain loads capacity from different resources, while ultimate compressive strength can be achieved when material is completely failed under uniaxial high stresses [21]. The means of compressive test in experiments are usually used to obtain the compressive strength where huge loads are applied to materials until their failure.
From many available references related on the influence of CNTs in cement composites, different results were obtained [22]. Some of them stated increase in the mechanical properties of cement, while other studies showed decreases or no effect. Indeed, more and different percentages of CNTs content are required for further and accurate investigation which stated as following:

4.1 CNTs levels up to 0.15%
According to Xu et al. [21] the addition of few amount of MWCNTs about 0.01% to the weight of cement mortars, where this MWCNTs interact with H2SO4 & HNO3, and two dispersion substances used which are carbonyl and aqueous dispersions to facilitate the process. The outcomes after 59 days showed using aqueous dispersion in MWCNTs increased compressive strength by 8.4 %, while using carbonyl dispersion in MWCNTs increased the compressive strength by 15.9 %.

Hamzaoui et al [22] showed around 21% improvement in the 90 days compressive strength of cement composites by adding 0.01% of MWCNTs by weight of cement. However, cement mortars having 2.1% wet hemp fibers showed improvement by 24.44% of the 90 days compressive strength while adding 0.01% of MWCNTs.

Yazdani and Mohanam [23] reported that addition of 0.1% and 0.2% of CNTs by weight of cement mortars with different amounts of w/c. The dispersion methods sonication and surfactant are used in this process. The 28 days results showed that usage of 0.1% of CNTs at w/ c ratio of 0.35, 0.4, 0.45 increased the compressive strength by 54%, 12% and 14% respectively. While usage of 0.2% of CNTs increased the compressive strength by 13 %, 27%, 21% and 10%, respectively with w/c ratios.

4.2 CNTs levels up to 0.5%
Li et al. [24] reported addition of around 0.5% of MWCNTs which has been treated with HNO3 and H2SO4. The influence of this addition was enhancement of the compressive strength by 18.86% at the 28 days.

Musso et al. [20] used different methods of functionalization to cement mortars then added 0.5% of MWCNTs. These methods were described as pristine (as grown) method (dia.40-80nm), annealed method (dia.40-80nm) and carbonyl (dia. 10-20nm). Methods of dispersion of MWCNTs were acetone and ultrasonic. After 28 days, the results showed increase of the compressive strength by 10.58% and 17.31% by the addition of pristine and annealed respectively, whilst the compressive strength decreased by 85.57% when adding carbonyl.

Manzur et al. [25] used MWCNTs of 0.05-0.5% by weight of cement composite (dia. 10-20nm, length 10-30nm). The MWCNTs were dispersed by ultra-sonication with interact of plasticizer. After 3,7,28 and 100 days the results showed improvement of the compressive strength by adding 0.05%, 0.1% ,0.2% and 0.3% of MWCNTs. At the same time, the compressive strength decreased by adding 0.5% of MWCNTs. Manzur et al. showed the highest compressive strength achieved, which are 8.81%, 23.43%, 17.38%, and 10.36% at 3,7,28 and 100 days respectively, by the addition of 0.3% of MWCNTs.

4.3 CNTs levels up to 10%
As stated by Chaipanich et al. [26] the addition of 0.5% and 1% of CNTs (dia. Less than 100nm) to weight of mixture matrix of (80% cement and 20% FA). The dispersion method of CNTs was ultrasonic. All results showed enhancement of the compressive strength which are increasing by 9%, 8% and 8% at 7, 20 and 60 days respectively at the addition of 0.5% CNTs. Although the compressive strength increased by 9%,8% and 8% by the addition of 1% of CNTs.

Morsy et al. [27] reported that addition of 0.5%,2%,5% and 10% of MWCNTs showed different compressive strength results. Cement mortar were having cement-replacement material which is Nano-metakaolin by 6% of mortar weight. NH4Cl used as a dispersion solution of MWCNTs in the process. The 28 days compressive strength results showed improvement by the addition of 0.5%, 2% and 5% of MWCNTs, whilst the strength decreased by the 10% addition of MWCNTs.
5. Flexural Strength

Flexural strength is defined as the ability of material to resist the deformation caused by applied load. Most of materials have less tensile stress, therefore they fail by tension before compression. The maximum resistance of tensile stress before failure is called flexural strength [28].

There are several experiments and investigations taken place to study the influence of adding CNTs into cement composites. Konsta-Gdoutos et al. [29] conducted experiment on adding CNTs with the same size, but different in length and concentration. At age of 28 days, the result showed that there are increasing in flexural strength comparing to the plain cement composite. Different length and concentration of CNTs all enhance and improve the flexural strength. This indicates that increasing the content of CNTs lead to increase the flexural strength without any interface of the length of CNTs [30] as shown in Figure 1.

![Figure 1. Effect of different types (short and long) of MWCNTs and concentration on the flexural strength [25].](image1)

The concentration of CNTs into cement composite is studied to distinguish the influence of different concentration of CNTs on flexural strength. Figure 2 shows the result as an average flexural strength of three different concentration of 0.025 %, 0.48 % and 0.08 % which taken at different ages of 3, 7 and 28 days. Composites with higher CNTs content of 0.08% exhibit higher result of flexural strength, while if the sample contains higher than 0.08 % CNTs then will exhibit lower result. Therefore, it is indicated that dispersion of short length CNTs with concentration more than 0.08 % is difficult to be obtained. Low concentration of CNTs below 0.08 % is not that enough to resist the micro cracks. [29] [31] [32]

![Figure 2. Fracture mechanics test results of the flexural strength of Nano-composites [26].](image2)

Addition of CNTs has an improvement in maximum load and deflection, where CNTs cement composites show increasing in maximum load before failure comparing to composite without CNTs. In turn there is no increasing in the deformation due to the increasing of the maximum load. This is
because of CNTs act as a reinforcing material to cement composite which result in increasing flexural strength. [28] [33] [34]

Elastic modulus or tensile modulus is also affected by addition of CNTs into cement composites. Elastic modulus is determined during tensile test from the slope of stress-strain curve. The result obtained from the sample including CNTs shows higher elastic modulus comparing to plain samples regardless of the concentration of the CNTs in the sample. [35]

6. Microstructure

Microstructure is considered as surface obtained from materials such as cement composite. This microstructure can determine the properties of materials which reflect and control the large scale of material to be applied in industrial practice. [34]

As it is well known that the cement components are hydrated when mixing with water to produce the hydration products included C-S-H and Ca(OH)$_2$. There is attraction between the hydration products and CNTs which leads to improvement in microstructure of cement composites. For better understanding of the improvement occur because of CNTs, micrograph of cement composite contains CNTs are checked using Scanning Electron Microscopy (SEM) analysis.

Due to large surface area of CNTs, there is high potential for CNTs to attract hydration products then these hydration products coat the surface of CNTs. Therefore, at early stage CNTs are easily to be recognized by SEM [36].

There is an experiment done by Li GY et al. [37] comparing the cement composite without and with CNTs. The micrograph of plane cement composite excluding CNTs shows the hydration products of C-S-H gel and Ca(OH)$_2$ and there are pores and cracks observed as well. Whereas in the composites containing CNTs the microstructure showed less cracks and less pores as well as homogenous hydration product.

Porosity affects the mechanical properties of cement composites as well as the durability, where large size of pores reduces the mechanical properties and durability [38]. Porosity and pores size decrease with addition of CNTs into cement composites [39]. The small size of CNTs which is measured in Nano has ability to fill the pores hence decrease the porosity of cement composites. Different percentage of addition of CNTs also control the decrease of porosity, where found that a higher amount of CNTs the lower the porosity [40].

Properties of microstructure is investigated by adding 1% of CNTs into cement composite using SEM analysis. The micrograph showed that CNTs present good interaction with the hydration product of cement which indicates that CNTs act as a filler to produce dense microstructure [31]. Chaipanich et al. study CNTs-fly ash- cement composite by using SEM analysis and found that addition of 0.5% and 1% of CNTs helped filling up the pores, hence mechanical properties of the composite much improved. From the micrograph also, it can be noticed that there is good interaction between CNTs and fly ash-cement composite [41].

Furthermore, addition of CNTs is studied by Shilang et al. [30] with different content of CNTs from low to high. The high energy of CNTs surfaces enhance the hydration process of the cement. The microstructure showed the CNTs surrounding the cracks where these cracks only appeared on the areas that not reinforced with CNTs. This occurred in the sample that contains low content of CNTs which is 0.025%. Second sample that contains 0.1 % of CNTs showed intensive and well distribution of CNTs in cement composite. There was no agglomeration noticed on the samples of low and medium content of CNTs. The last sample contains high content of CNTs which is 0.2 % is showed a good connect between the hydration products. However, there are agglomeration of CNTs seen only in the middle region of the microstructure. The agglomerated CNTs can cause some weak zone which presented as pores. Therefore, excessive CNTs has no effect on the enhancement of the properties of cement composite, however sometimes it may lead to deterioration.

7. Durability
According to recent research, carbon nanotubes have been shown to have positive effects when mixed with the basic cement structure by showing unique properties for cement such as high durability. Moreover, the CNT helps to reduce self-contraction (shrinkage) and control the expansion of the crack by reducing the capillary porosity in cement. This shows an immediate impact on durability, because there is a decrease in the quantity of trajectories for agents of aggression to matrix [42]. Durability of cement structures have been studied by using many of the conventional mechanisms, where that mechanisms applied on nano-composites of cement. So, the important parameters to evaluate are penetration of chloride, impact of acid, cycles of ice-freezing, ASR, sulfate propagation, and capillary water absorption. Unluckily, there is not too much information about the mechanisms of CNT in cement composites and what is the effect of these applied techniques on it. So, a lot of experiment in this field have been specified in the last century, not exclusive only on CNT’s, but for nano-particles of other materials which can use in cement compounds [42].

**Table 1. Summary of technique used to study durability of CNTs-cement composites [38]**

| Parameter                                    | Technique                                                                 | Reference |
|----------------------------------------------|---------------------------------------------------------------------------|-----------|
| Thermal stability                            | Thermogravimetric analysis and dilatometry                                | [9]       |
| Transport properties                         | Water permeability, water sorptivity, gas permeability                    | [44]      |
|                                              | Chloride penetration                                                     | [43]      |
|                                              | Mercury intrusion porosimetry                                            | [47]      |
| Electrochemical properties of reinforcement  | Carbonation of cement matrix and corrosion by polarization resistance of reinforcement rebar | [46,48]  |
|                                              | Chloride penetration of cement matrix and corrosion by polarization resistance of reinforcement rebar | [46]      |

In recent times, studies have focused on one of the most important factors that has a direct impact to the cement compounds which is durability, where the researches on CNT have mentioned three main coefficients namely thermal stability, characteristics of cement matrix transmission and erosion of reinforcing rebars. Concerning thermal stability, a research for a mixed CNT’s with the liquid cement composite has shown a growth in crystallinity for the cement composites that mixed with CNT’s, therefore, a development in thermal stability happen to the hydration material and reduction in shrinkage because of heating, thus this prove the positive effect for CNT’s [9]. Concerning the characteristics of cement matrix transmission, permeability of water, water absorption, chloride penetration [43] and gas permeability [44] are the parameters presently examined. In all situations, the micro and nano materials got an improvement in porosities at their cement composites structures, where the durability became stronger [45]. Concerning erosion of reinforcement in the cement matrix composites that included CNT in their component, the studies discovered that the adding of CNT into cement would help to prevent the value of the corrosion levels to increase in emergency situations [46]. Because of that, the resistivity of the cement matrix that had an additional CNT would become lower than the portland-cement, thus the corrosion has been distributed to the cement matrix cell and that would create a pair of galvanic between the CNT’s and reinforcement [46]. The outline of the mechanisms that discussed in this part is shown in Table 1.

The researches didn't study deeply the deformation and chemical stability for cement matrix, therefore there are not too much reports and results that describe all parameters for CNT's cement durability. Nanotechnology investigations have shown a good improvement for cement in durability side, due to adjustment that occurred in the nano-scale, especially in Young’s coefficient of (CASAH) [49]. Due to the advanced properties in the cement matrix, the unique chemical durability had been
generated in the CNT's cement composites. The CNT’s-cement composites need to be investigated more, where the important tests such as freeze-thaw, alkali-silica reaction, carbonation, acid and sulfate attack must be examined widely.

8. Conclusion
This review paper targets to preview the previous researches achieved on the electrical, thermal, mechanical and strength properties of CNTs, also dispersion, durability, microstructure, compressive and flexural strength for the CNTs in traditional cementitious materials. The important points of this review-paper can be concluded as following:

1. The results showed that CNTs has distinctive properties that have a positive effect when added to cement compounds. Electrical conductivity properties and fire resistance of that cementitious matrix have been increased significantly by adding CNTs, but the opposite happen for the electrical resistance. The fillers of CNTs have shown an obvious influence on the mechanical properties where the SEM micro-graphs showed a uniform distribution for the CNTs in the matrix of cement compounds. High strength has been discovered, due to the perfect interactions between the CNTs (fillers) and the cement matrix.

2. To obtain positive benefits of CNTs in cement composite, an appropriate dispersion of CNTs is required. In this review paper, different dispersion methods were investigated such as surfactant and sonication, where enhancement of the mechanical properties achieved. However, no significant improvement observed when poor dispersion of CNTs occurred.

3. The addition of CNTs with different influence in cement composites compressive strength has been observed. Some certain content of CNTs addition stated increase in the compressive strength, whilst other content of CNTs addition showed drop in the compressive strength. Overall, adding particular proportion of CNTs with appropriate dispersion method leads to high compressive strength production materials.

4. Addition of CNTs enhance and increase the flexural strength of cement composites depending on the concentration of CNTs. Regardless of the length of the CNTs, optimum concentration can result in higher flexural strength, whereas some excessive in concentration may lead to reduction in the strength. Moreover, Elastic modulus showed increment when CNTs is added.

5. CNTs in cement composites acts as a filler which can fill the pores and reduce the porosity as well as control the micro-cracks. Micrographs showed that the microstructure including CNTs is denser and well packed compared to the plain cement composite without CNTs.

6. The blended traditional cementitious materials with CNTs have shown positive impacts through increase the durability and ductility in cement compounds, due to the connections between the pozzolanic microparticles, where micro-fibers of CNT's strongly unite with cement matrix. According to the reviewed studies, it can be concluded that addition of CNTs helped to improve the cement composite in terms of durability and mechanical properties including compressive and flexural strength as well as microstructure.

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