

Sustainability of Carbon Nanotube-Reinforced Concrete

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Abstract—Concrete, despite being one of the most produced materials in the world, still has weaknesses and drawbacks. Significant concern of the cementitious materials in structural applications is their quasi-brittle behavior, which causes the material to crack and lose its durability. One of the very recently proposed mitigations for this problem is the implementation of nanotechnology in the concrete mix by adding carbon nanotubes (CNTs) to it. CNTs can enhance the critical mechanical properties of concrete as a structural material. Thus, this paper demonstrates a state-of-the-art review of reinforcing concrete with CNTs, emphasizing on the structural performance. It also goes over the properties of CNTs alone, the present methods and costs associated with producing them, the possible special applications of concretes reinforced with CNTs, the key challenges and drawbacks that this new technology still encounters, and the most reliable practices and methodologies to produce CNT-reinforced concrete in the lab. This work has shown that the addition of CNTs to the concrete mix in percentages as low as 0.25% weight of cement could increase the flexural strength and toughness of concrete by more than 45% and 25%, respectively, and enhance other durability-related properties, given that an effective dispersion of CNTs in the cementitious mix is achieved. Since nano reinforcement for cementitious materials is a new technology, many challenges have to be tackled before it becomes practiced at the mass level.

Keywords—Sustainability, carbon nanotube, microsilica, concrete.

I. INTRODUCTION

CONCRETE, and other cementitious composites, are usually described as quasi-brittle materials. This, in simple terms, means that such materials are subject to cracking when loaded [1]. Since the development and propagation of cracks is the main drawback and the biggest concern when using cementitious materials for structural purposes, scientists have recently been studying using fibers in order to inhibit and limit the propagation of cracks in these materials. The main concept behind this trend was to allow for a more scattered distribution of stresses in the material under loading, rather than depending on the reinforcing steel bars (which cannot stop cracks at the micro level) alone, trying to control the development of cracks at the micro level [2].

Earlier studies have shown that the behaviors of cementitious materials are significantly dependent on their configurations at the nano-level. Nanotechnology was first announced by Richard P. Feynman in one of his seminars at the California Institute of Technology in 1959. This

technology consists of the “re-engineering” of a certain object at the atomic level, in an attempt to enhance its properties [3]. Thus, adjustments done to cementitious materials at the nano-level can have great impact on the characteristics that are usually targeted by engineers, such as: “Strength, ductility, creep and shrinkage, fracture behavior and durability” [1].

Based on that understanding, several recent attempts have been done to incorporate CNTs in concrete mixes, in order to take advantage of the different superior characteristics these nano-scale materials have. These attempts have led to the conclusion that this new technology is very promising in the construction field. However, no major implementation of CNTs has yet been witnessed in the construction industry because of many reasons including the necessary decrease in the market prices of CNTs before they could be implemented in such a competitive environment and the lack of information and enough understanding of the behaviors of CNTs-reinforced cementitious materials and the different factors that could influence these behaviors [4]. In an attempt to explore more details, this work represents a review on the status quo of the implementation of CNTs as reinforcement for concrete, taking structural enhancement as the main focus.

II. LITERATURE REVIEW

A. CNTs & Material Strength

The effect of dispersing CNTs in the concrete mix can be seen and studied at the micro level. After looking at the micro-structure of a concrete mix without CNT reinforcement, it was noted that crystal calcium hydro-silicate existed at the contact area between the aggregates’ surfaces and the cement particles, which made this area structurally weak (see Fig. 1 (a)) [5].

On the other hand, the dispersion of CNTs in a concrete mix results in creating a strong covering (with thickness of 1 to 5 μm) for the solid particles including cement and fillers, which causes these particles to be more interlocked with the aggregates’ surfaces (Fig. 1 (b)) [5].

Based on the observation mentioned above, it could be easily expected that dispersing CNTs in a cement concrete mix would increase the overall strength of concrete (since the interlock between aggregates and the binder is made stronger after adding CNTs to the mix). In contrast to macro or micro-fibers, CNTs interrupt the creation of cracks at the nano-level and inhibit their growth and spread to the micro-stage [2].

As for the structural performance of CNT-reinforced concretes, Hunashyal, Tippa, Quadri, and Banapurmath [6] reported an experimental study that has proven that the addition of 0.25% weight of cement of Multi-walled Carbon

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Nanotubes (MWCNTs) to beams made of cement has only resulted in major improvements in the toughness and flexural capacity of these beams. Compared to beams casted from cement alone, CNT-reinforced cement beams (with 0.25% weight of cement MWCNTs) have had flexural capacity that is, on average, 47% higher and toughness that is increased by 25%. Further similar experiments against a plain-cement reference beam have shown that even the addition of CNT with percentages as low as 0.025% weight of cement to the concrete mix could have remarkable effects on the overall flexural performance of the reinforced beams [6].

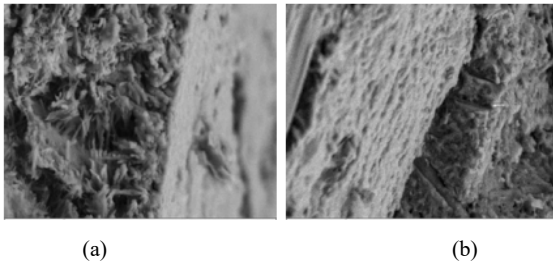


Fig. 1 The creation of coverings for solid particles: (a) no CNTs, (b) CNTs added [5]

Yakovlev et al. [5] observed similar trends of flexural strength increase after using MWCNT as reinforcement for cement concrete, in a project involving the production of pre-stressed poles for HV power lines, where the increase in flexural strength has reached up to 46%.

In an experimental study reported by Ferro et al. [7], prisms (40 x 40 x 160 mm³) of cementitious mixes (mortars) reinforced with 0.5% weight of cement of MWCNTs, as well as without any CNT, were tested for flexural strength. In this study, the effect of CNTs on the flexural strength of the material was reported against the duration of curing provided (Table I).

TABLE I
THE EFFECT OF CNT-REINFORCEMENT AND/OR CURING TIME ON THE FLEXURAL CAPACITY OF MORTAR PRISMS [7]

Curing Time (days)	Mortar as such (MPa)	Mortar +0.5wt/% MWCNTs (MPa)
1	2.49	3.81
7	6.86	7.79
28	7.73	10.08

In order to find the effect of adding CNTs to the concrete mix, the percentage difference in the flexural capacity between plain mortar and CNT-reinforced mortar at each curing time can be calculated. For one day, seven days, and 28 days of curing, the percentage difference happens to be 53%, 13%, and 30%, respectively. Thus, the biggest effect of CNT reinforcement to the mortar appears when the curing time is as low as one day. But since curing is usually done for 28 days, 30% increase in flexural capacity can be taken as the most useful figure from this experiment.

In addition to flexural capacity, tensile strength could be a valid measure of the strength of the cementitious materials,

despite not being very useful for structural evaluation. In the same study reported by Ferro et al. [7], cylindrical concrete samples (after curing for 7 days) with 2% weight of cement addition of CNTs, as well as samples with no CNTs addition, were passed through the Brazilian tensile strength test. The results of these tests indicated almost no effect on the tensile strength of the concrete (Fig. 2). The proposed explanation for such results is provided in the Challenges and Drawbacks section.

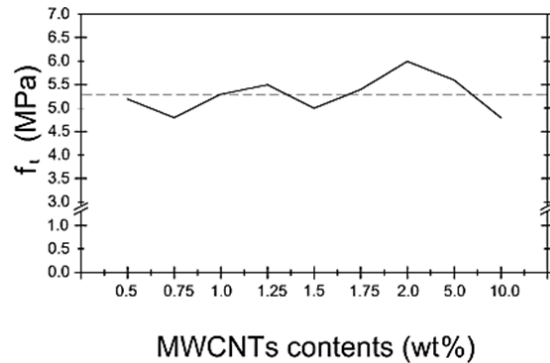


Fig. 2 Tensile strength of mortar cylinders cured for 7 days against CNT content (dashed line: samples without CNTs) [7]

Another measure of structural strength of cementitious materials comes from estimating the internal damage caused by the applied load on them. The internal damage in the material could be represented by the “dissipated energy density”, which could be calculated using a mathematical expression or simply by finding the area under the stress-strain curve of the tested sample [8].

Ferro et al. [7] have used one of the remaining parts of each of the prisms after the flexural test for compressive strength test. Again, these prisms consisted of cementitious mixes (mortars) reinforced with 0.5% weight of cement of MWCNTs, and others without any CNT addition. Based on the Stress vs. Strain graphs obtained from the compression tests, the dissipated energy densities were calculated and plotted versus the days of curing, as shown in Fig. 3. It can be clearly noted that reinforcing the mortar with 0.5% weight of cement of MWCNTs has caused the dissipated energy density to increase by around 50%, at any given duration of curing, allowing for more internal damage (utilization of material capacity) before failure.

In the same HV power lines project mentioned earlier, MWCNT-reinforced concrete (that was used for electricity poles) has also demonstrated improvements in two other characteristics that are related to the durability of the material. The first characteristic is the water resistance, which was improved from W6 to W14. The other characteristic is the frost resistance of concrete, which was also increased by more than double (F150 to F400) [5]. The micro-structure of the CNT-reinforced concrete after the test for frost resistance is shown in Fig. 4.

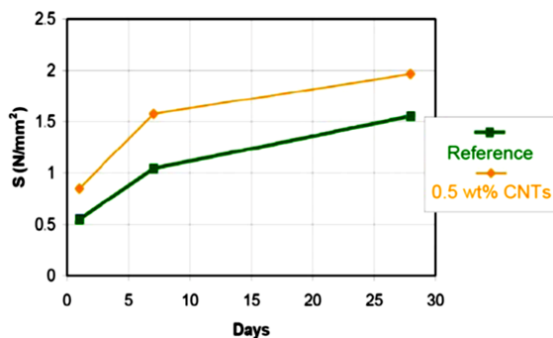
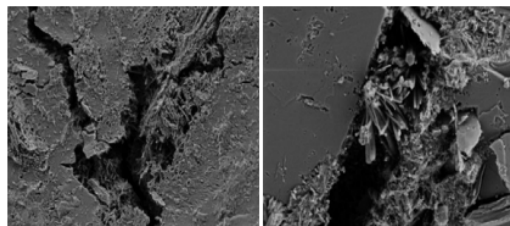


Fig. 3 Dissipated Energy Density vs. days of curing [7]

B. CNTs & Material Durability



(a)

(b)

Fig. 4 Microstructure of concrete after frost resistance test: (a) No CNTs (reference sample with frost resistance of F200), (b) CNT-reinforced sample (F400) [5]

As mentioned earlier, CNTs inhibit crack development and growth starting from the nano-level, rather than the micro-level. Sasmal et al. [1] optimistically state that CNTs, because of the observation mentioned lately, will enable civil engineers to create “crack-free” concrete someday. Of course, having structural concrete that does not crack at all will mean a lot in terms of the durability and life expectancy of the whole structure. For instance, if CNT reinforcement is done along with traditional steel reinforcement, and given that the concrete will not cracked under the designed load, then we are talking about almost no corrosion to steel (one of the most common reasons behind durability depreciation of steel-reinforced concrete).

C. Author's Critique

Looking at what has been done in the past few years in the literature regarding reinforcing cementitious materials with CNTs makes one learn that the attempt of utilizing nanotechnology in the field of construction materials was a very successful one. This is because, despite not having deep research done on them yet, CNTs have enhanced many of the most important properties in concrete.

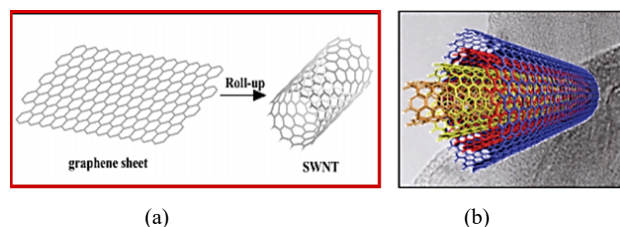
What is important for this paper is that CNTs-reinforcement for concrete was proven to have positive impacts on the structural performance of concrete. However, there exists some kind of inconsistency in the results, which allows some question marks to arise. To be more specific, Hunashyal et al. [6] have found 47% increase in flexural strength after reinforcing their concrete with 0.25% weight of cement MWCNTs. Similarly, Yakovlev et al. [5] have reported 46%

increase in the flexural strength of their concrete that was reinforced with MWCNTs, but they did not mention the percentage of MWCNTs used in the mix. Surprisingly, Ferro et al. [7] have shown results that indicate only a 30% increase in the flexural strength of the concrete that was reinforced with 0.5% weight of cement MWCNTs and cured for 28 days. Is it the difference in curing time provided for the samples that caused this decrease in the improvement, despite increasing the dosage of MWCNTs? Is there an “optimum” dosage of MWCNTs to be added, and after which the addition of CNTs starts to have reverse impacts on concrete? Or is it nothing more than experimental differences between the reported studies, owing to the unavailability of a standardized procedure for reinforcing concrete with CNTs yet?

Away from all these questions that remain un-answered until today, reinforcing cementitious materials with MWCNTs does, at least, allow us to utilize the capacity of the material more than usual. This was proven by studying the internal damage of the concrete samples by Ferro, Tulliani and Musso [7] (see Fig. 3).

D. Characteristics of CNTs & CNTs-Reinforced Concrete

CNTs, as the name implies, have tabular shapes and are hollow. They can consist of one wall of graphene (SWCNT) or more (MWCNT) (Fig. 5). The outer diameter of SWCNT ranges between 0.4 nm to 10 nm, while that of the MWCNT ranges between 4 nm to 100 nm. What is notable is that the length of the CNTs varies and can reach up to few millimeters in length, which gives them the critical crack-bridging property when used in the concrete mix [7]. According to Madhavi et al. [9], the carbon purity of MWCNTs exceeds 90%, their densities range between 0.15 g/cm³ to 0.35 g/cm³, their surface areas equal 350 m²/g, the number of walls in them can reach up to 15, and their diameters range between 20 nm to 40 nm.



(a)

(b)

Fig. 5 (a) SWCNT, (b) MWCNT [7]

CNTs have extraordinary mechanical properties as well. These tiny particles can have strain up to 12%, a tensile strength of 60 GPa, and a modulus of elasticity of 1 TPa. This means that CNTs compete steel (as materials) by having 60 times higher maximum achievable strain, 100 times higher tensile strength, and a modulus of elasticity that is five times bigger [2].

After reinforcing concrete with CNTs, a unique characteristic in the hardened concrete was noticed: piezoresistivity. This characteristic means that CNT-reinforced concretes possess an electric resistance that is related, to some extent, to the compressive load applied on

them. In fact, a lot of studies were focused on understanding this behavior of CNT-reinforced concretes and finding the different factors that could influence it. The results from these studies show that there is an inverse relationship between the electric resistance of the CNT-reinforced concretes cured in dry atmospheres and the applied compressive loads. Also, using carboxyl to increase the efficiency of CNTs increases the piezoresistive behavior of the CNT-reinforced concrete. Similarly, reducing the water content in the concrete mix increases the piezoresistivity. In fact, concretes with water to cement ratios exceeding 0.5, in addition to moist-cured concretes, did not exhibit any piezoresistivity. This observation has led to the conclusion that water to cement ratio is the most important factor influencing the piezoresistive behavior of CNT-reinforced concretes [10].

E. Synthesis & Cost of CNTs

Due to their superior mechanical properties and promising future, CNTs have been the focus of many scientists as well as businessmen. Many attempts have been made with the aim of the mass production of CNTs, keeping the quality uncompromised [11].

According to De Volder et al. [12], the most commonly used technique for bulk production of CNTs is the "Chemical vapor deposition (CVD)". This technique mainly utilizes a fluidized bed reactor in order to pass a certain gas, in addition to heat, to the metal catalyst nanoparticles. Fig. 6 illustrates the CVD process. Other older techniques include:

- The pulsed laser vaporization of a heated carbon target (with Ni/Co catalyst).
- An electric-arc method (with a Ni/Y catalyst)
- And more suitably for large-scale production, the improved floating catalyst technique [11].

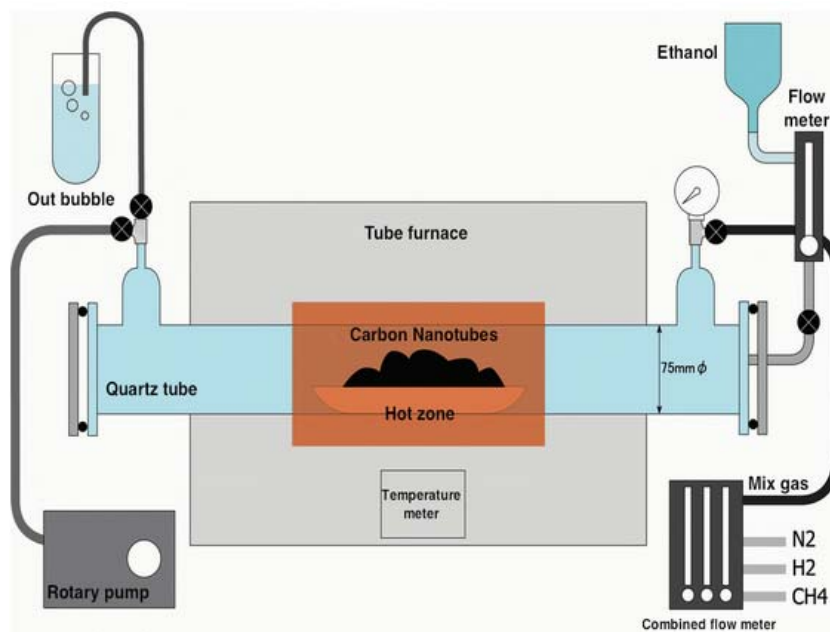


Fig. 6 CVD [13]

Using the CVD technique has allowed for a significant drop in the prices of CNTs. This is because in this technique, production volume is increased, cheaper raw materials are used, higher profit margins are achieved, less energy is needed, and less waste is produced. Nevertheless, this method produces CNTs with certain impurities that may lower the CNTs' quality and affect their targeted characteristics. Fixing this problem (i.e. removing these impurities) is usually expensive and could cause damage to the resulting CNTs' outer surfaces and/or length. Today, MWCNTs are available at prices less than \$100/ kg, while carbon fibers are sold at prices up to 10 times lower than this rate [12]. One should, however, keep in mind the difference in the percentage/amount needed from each of the two materials to be used as reinforcement in a cementitious mix. As mentioned

earlier, from an experimental study, CNT-reinforced cement beams with 0.25% weight of cement MWCNTs have had 47% increase in flexural capacity and 25% increase in the toughness, compared against a plain-cement beam [6].

It is also worth mentioning that the prices of SWCNTs are higher than those of MWCNTs. This is due to the fact that manufacturing SWCNTs necessitates more accurate quality control than manufacturing MWCNTs and because a lot of costs are associated with studying and improving the manufacturing process of SWCNTs [12].

F. Applications of CNTs-Reinforced Concrete

Nanotechnology has already made breakthroughs in many fields of applied science. Unfortunately, civil engineering has not yet benefited enough from this new technology, despite

the fact that a lot of potential applications of nanotechnology in general and CNTs in specific do exist. Nanotechnology can help us develop innovative construction materials that have special characteristics, including: increased strength and decreased weight, fire and water resistance, sound insulation/damping, decreased maintenance requirements, protection against UV light, and air filtering [3]. In fact, the use of nano-scale reinforcement for concrete has demonstrated major enhancements in both the fresh and the hard properties of the mix, and CNTs happened to be the brightest nano-fiber reinforcement done so far due to their attention-grabbing mechanical, thermal, and electrical characteristics.

CNT-reinforced concrete has been proven to have higher workability when it is fresh and higher compressive and tensile strengths when it is hard. Also, the usage of CNTs has helped in reducing the required time for the concrete mix to set. CNTs can also be used to lower the percentage of water absorption in the concrete. In a recent experiment, more than 15% decrease in the water absorption has occurred after adding 0.045% of CNTs to the concrete mix [9].

In addition to these applications, CNT-reinforced concretes were successfully utilized as self-sensing materials, in order to closely monitor the health of structural members. In fact, CNTs were found to have a linear piezoresistive response that is also reversible. That is, the electrical conductivity of CNTs changes when loading is applied on them and this change is, to some extent, predictable by empirical mathematical equations. This interesting characteristic in CNTs have opened the eyes of scientists and engineers to explore new applications for CNT-reinforced concretes including measuring stresses in concrete under loading, spotting unnoticeable defects in concrete, and sensing vehicle flow in roads and highways [14]. Furthermore, the addition of CNTs to cementitious mixes as means of reinforcement has been proven to increase the early-age strain and reduce the autogenous shrinkage. This leads to enhancing the transport properties of the mixes significantly [7].

Nano-reinforcement for cementitious materials could become a competitive substitute or supplement to traditional fiber-reinforcement and could play a major role in developing future “high-performance” construction materials [2].

G. Challenges & Drawbacks

Despite the fact that CNTs have extraordinary characteristics and that they have been used successfully in strengthening polymers, enhancing the cementitious materials to the level of having extraordinary characteristics is not guaranteed, or at least not very predictable.

1. The Challenge of Dispersion

The effects of adding CNTs to a certain cementitious mix are significantly driven by two main issues: the dispersion of the CNTs in the mix and the adherence between CNTs and cement particles. These two issues arise from the strong Van Der Waals forces between and among the CNTs particles, which makes these particles strongly stick to one another. Such forces exist between all fine particles in general

(including cement itself), but since CNTs have very high surface area/ volume ratios, they are more affected by the Van Der Waals forces. For this reason, CNTs tend to accumulate and cluster within the mix in a very fast manner [2].

Discussing dispersion, it is also challenging to find the optimum percentage of CNTs to be added to the cementitious mix. This challenge arises from the fact that adding so much of CNTs (more than the optimum ratio, which is discussed in under Proposed Mitigations) could cause the connections between the CNTs and the cement particles to weaken, and thus, lowering, or even eliminating, the efficiency of the CNT-reinforcement. For instance, as mentioned earlier in this paper, in an experimental study reported by Ferro et al. [7], cylindrical concrete samples with 2% weight of cement addition of CNTs showed almost no increase in the tensile strength, and the observable CNTs after failure of the samples were plain and not attached to any of the materials that are produced while cement hydration.

2. Proposed Mitigations for Dispersion

In order to tackle the dispersion issue of CNTs in concrete mixes, Konsta-Gdoutos and Aza [14] report in their paper two effective methods that have been applied by other researchers. In the first method, ultrasonic energy is utilized in addition to a surfactant. The main observation from this method is the existence of an “optimum” surfactant to CNTs ratio, which is also a factor to determine the amount of ultrasonic energy needed. The other method consists of using a superplasticizer (polycarboxylate) to achieve the dispersion of CNTs in the concrete mix. This method, beyond achieving effective dispersion, aims to eliminate the potential negative effects of dispersants on the cement properties after it hardens. It was noted that using polycarboxylate has achieved very effective dispersion for both CNTs and cement particles. Furthermore, finding the optimum CNT content in the concrete mix was also addressed by researchers. Fig. 7 was developed in order to help determine the optimum amount of CNTs to be added to the cementitious mix, as well as the amount of superplasticizer required, based on the aspect ratio of the CNTs used [7].

TABLE II
CHARACTERISTICS OF MWCNTs USED

Property	MWCNTs
Average diameter [nm]	40-80
Length (average) [μ m]	400-1000
Carbon purity [% W/W]	>92
Metal oxide (impurity) [% W/W]	<6

3. Environmental Fears

Another challenge facing the mass production and the usage of CNTs in the construction industry comes from an environmental perspective. Jackson et al. [15], study the environmental impact that could potentially be a consequence of large-scale production of CNTs. The authors state that not much is known about the effects of the direct exposure to CNTs yet. They also claim that the toxicity of CNTs depends on many elements, including: gross area of the particles'

surfaces, chemistry of the surfaces, “functional groups”, coverings, polarity, solubility, particle shape, photochemistry, and the method by which CNTs were produced. The main findings from Jackson et al. [15] are as follows:

- CNTs do not pass through organic matters easily (negligible percentage only).
- Aquatic creatures are more sensitive to the toxicity of

CNTs than terrestrial ones.

- Invertebrates (bone-less creatures) are more sensitive to the toxicity of CNTs than vertebrates.
- SWCNTs are more toxic than MWCNTs.
- It is possible to find certain concentrations of CNTs that do not have any environmental effects, but more experimental works/data are needed.

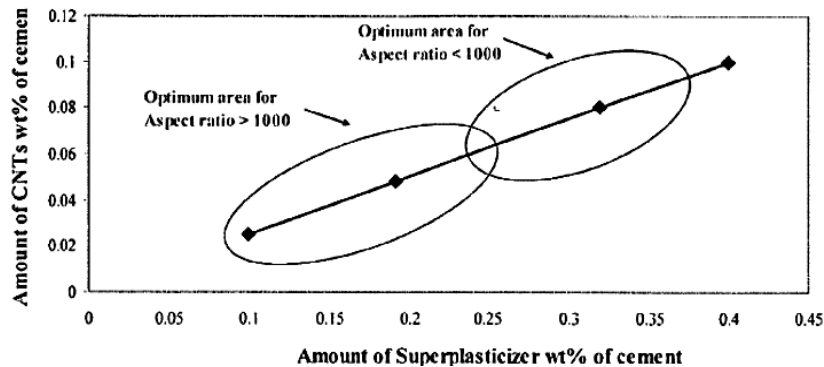


Fig. 7 Determination of the optimum amount of CNTs and the correspondingly required amount of superplasticizer depending on the aspect ratio of the CNTs used [7]



Fig. 8 Ultrasonic Dispersion of CNTs [16]

4. Experimental Methodology

Reinforcing concrete with CNTs requires the same set of materials that are required for regular concrete, in addition to CNTs and any other required materials/processes to achieve efficient dispersion of CNTs within the cement matrix. That is, Portland cement, sand, fine and coarse aggregates are needed in the first place (the specifications of each depend on the desired concrete grade) [9]. The other materials needed for achieving effective dispersion need to be designed for as well, because the addition of CNTs to a concrete mix will increase its durability and strength only if they are well spread in the mix.

In order to achieve efficient dispersion of CNTs within the cement matrix, ultrasonic energy is utilized. Ferro et al. [7] have inserted an ultrasonic probe in the water containing CNTs (with the characteristics shown in Table II) for four hours until CNTs are well dispersed in the water, before

adding cement and sand. Fig. 8 illustrates a typical ultrasonic probe set up.

Ferro et al. [7] have also, while stirring, used a superplasticizer and another chemical to adjust viscosity. The superplasticizer used was Mapei Dynamon SP1, while the viscosity-related chemical was VMA-Mapei Visco Fluid SCC/10. The amounts used of these two ingredients depended on the amount of CNTs used. For CNTs not more than 1.75% weight of cement, the amounts used were as recommended for self-compacting concrete. For higher contents of CNTs, extra superplasticizer was added. The addition of these two chemicals results in a more cohesive mix and prohibits bleeding. Table III shows the mix designs followed by Ferro et al. [7].

TABLE III
MIX DESIGN OF THE TESTED SAMPLES [7]

Components	Reference Mortar	Mortars + MWCNTs				
Cement: CEM II A-LL (EN 197/1 standard)						
Water to cement ratio (w/c): 0.50						
Sand: normalized (EN 196-1 standard)						
Superplasticizer (wt%/cement)	1.1	1.1	2	5	10	
VMA (wt%/cement)	0.5	0.5	0.5	0.5	1	
MWCNT (wt%/cement)	0	0.5, 0.75, 1, 1.25, 1.5 and 1.75	2	5	10	

III. CONCLUSIONS

To sum up, this paper studied the status quo of the utilization of CNTs as reinforcement for concrete, focusing mainly on the structural enhancement of concrete due to this reinforcement. It also covered the characteristics of CNTs, the current synthesis and cost of CNTs, the potential applications of CNTs-reinforced concrete, the main challenges facing this

new technology, and the experimental procedures that could be followed to reinforce concrete with CNTs in the lab.

In the recent literature, CNTs have been proven to be very effective reinforcement for cementitious mixes, in general, and concrete in particular. In fact, dispersing CNTs in the concrete mix in percentages as low as 0.25% weight of cement has shown an increase in flexural strength that exceeded 45%. It has also enhanced the durability of concrete through different aspects such as: water resistance, frost resistance, and inhibiting crack initiation and development at the nano-level. Today, we are certain that reinforcing cementitious materials with MWCNTs does, at least, allow us to utilize the capacity of the material more than usual. However, achieving an effective dispersion is, until today, a challenging task, and no one ideal method has been confirmed yet.

Since reinforcing cementitious materials with CNTs is a new technology, many challenges have to be considered before it becomes commonly practiced. As explored in this paper, the applications of CNTs are a lot, and thus, CNTs-reinforced cementitious materials are worth more extensive research.

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