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Physical and Chemical Surface Modifiers of Carbon Nanotubes on the Mechanical and Physical Properties of Concrete

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ABSTRACT

This study utilizes nanofilament-based nanotechnology, such as carbon nanotubes (CNTs), to improve the mechanical characteristics of concrete as a building material. One of the important and effective factors in adding carbon nanotubes to cement composites is their proper dispersion in the mixture. Therefore, in this research, different methods of dispersing carbon nanotubes in cement mortar have been investigated. They were first functionalized through covalent bonding to prevent carbon nanotubes from accumulating in the water. To achieve the appropriate distribution of carbon nanotubes in water and maintain the stability of their dispersion, different surfactants including Polycarboxylate (PCE), Sodium dodecyl sulfate (SDS), Cetyltrimethylammonium bromide (CTAB), Polyethylene glycol (PEG) were employed. At that point, utilizing visual perception and UV test, PCE/PEG surfactant was presented as a reasonable surfactant for the scattering of carbon nanotubes. Once the most suitable dispersal technique was identified, the impact of incorporating multi-walled carbon nanotubes into concrete was investigated with varying weight proportions. The study revealed that even at low concentrations (0.015%, 0.030%, and 0.045%) relative to the cement weight, the inclusion of carbon nanotubes decreased the porosity of the concrete nanocomposite, resulting in a denser and more compact composite. As a result, it increases the concrete samples' compressive, tensile, and bending strength.

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1. Introduction

Concrete is a mixture of aggregate, cement, and water in proper proportions. Concrete is one of the most essential building materials for civil engineering projects worldwide. In any case, nanoscale cracks and pores in concrete create significant problems and diminish the mechanical performance and strength of concrete. Recently, nanomaterials have been used to increment the durability of concrete in concrete structures [1,2]. Nanomaterials have been used in various fields, including medicine, construction, the automobile industry, etc. Still, the construction field is one of nanomaterials' main and most beneficial uses. The use of these materials in concrete, steel, and glass can improve their properties and reduce environmental pollution [3]. It has been proved that

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carbon nanotubes (CNT) have better desirable properties among nanoparticles due to their resistance and high elastic modulus and are a more promising choice [1,2]. Carbon nanotubes are thin and long cylinders of carbon first discovered by Iijima in 1991 [3]. Carbon nanotubes possess a tubular configuration comprising coiled hexagonal lattice structures of graphene with tubes having nanometer-scale diameters. These tubes are categorized into two groups: single-walled (SWCNT) and multi-walled (MWCNT) [4]. With Young's modulus TPA of 1 and outstanding tensile strength in the range of 20-150 GPA, carbon nanotubes are considered the best candidates for cement and concrete reinforcement as they can prevent the propagation of small microcracks [5–7]. Carbon nanotubes (CNT) are increasingly being employed in concrete due to their high mechanical and electrical conductivity. Carbon nanotube concrete (CNTC) has been the subject of extensive research globally. Notably, they have shown that carbon nanotubes can bridge framework cracks, fill interior pores, and stimulate cement hydration responses in concrete. CNTs improve durability by optimizing the inner pore structure and increasing the interfacial transition zone (ITZ) [8,9]. Ahmed et al. and his colleagues discovered that nanomaterials such as CNT can make significant changes in cement composites, increasing cement quality and strength while decreasing the porosity [10]. Specifically, Shang and Song specified that the inclusion of CNT can increase the compressive and ductile strengths of concrete by 30.8% and 46%, respectively [11]. Kramer and colleagues examined the influence of incorporating 0.05 and 0.075% carbon nanotubes (CNT) in foam concrete. They noticed that the alteration in flexural strength after 28 days and the compressive strength of the concrete after 28 days were 40% and 7.5%, correspondingly [12]. In recent times, the influence of CNT in cement and concrete composites has been extensively studied., but there are contradictions in the reported results. Some results confirmed the improvement of mechanical properties in cement composites [13,14], while some did not show any improvement and in some other cases the mechanical properties even decreased [15–17]. The strengthening of concrete and cement matrices containing carbon nanotubes depends on various factors, including the type of carbon nanotubes, dispersion quality, interaction, and strength of their bond with the cement matrix [18,19]. According to Zijian Su, there are two primary obstacles for incorporating carbon nanotubes in cement mixtures: one is to standardize the distribution of CNTs within the cement mixture, and the other is to control the interaction of CNTs with substances produced during cement hydration [20]. It is challenging to evenly distribute CNTs throughout the cement matrix; Conversely, in order for CNTs to positively affect the mechanical characteristics of cement-based composites, proper dispersion of CNTs is necessary [21–23]. CNT aggregation occurs for several reasons. Due to their nanoscale diameter, the surface area of carbon nanotubes is high, as well as the interfacial attraction between CNTs. Dispersion problems can also arise from the tendency of CNTs to form bundles that are tightly bound by high van der Waals forces. In addition, the high aspect ratio of CNTs (length/diameter) combined with their flexibility allows for the formation of highly entangled agglomerates in the liquid phase. The incorporation of CNTs into cement also faces two major challenges: bonding and dispersion. Strong bonding between CNTs and the cement matrix as well as effective dispersion of CNTs are essential for effective load transfer to CNTs, which may act as reinforcements to improve the mechanical properties of cement composites. It is also believed that CNTs can provide significant mechanical reinforcement if the bonding between CNTs and the cement matrix can be controlled [23]. The proper dispersion of carbon nanotubes delays the growth of micro-cracks and reduces pores in nano-size, thus increasing the performance of concrete [24,25]. Therefore, an executive solution should be sought for this important matter. One of the available methods for proper dispersion is to use an additive in an aqueous solution and then in the concrete mixture. Many researchers have considered the use of different surfactants with different concentrations to disperse MWCNTs in cement materials [26,27]. A combination of physical (sonic) and chemical (surfactant-based) dispersion methods is the most recommended method for exfoliation of CNT agglomerates in water-mixed cement composites. Surfactants not only help in exfoliation of CNT bundles, but may also play a role in bonding with the cement matrix. Li et al. [4] claim that the surface interactions between surface-modified CNTs and hydration products provide high bond strength and enhance the efficiency of charge transfer from the cement matrix to the reinforcement [28]. Jang et al. studied the mechanical properties of MWCNT-reinforced cement pastes and their dispersion in cement pastes [29]. They used a water-reducing additive (ADVA Cast 575) as a surfactant and observed that MWCNTs tend to aggregate and precipitate immediately in distilled water without a surfactant. On the other hand, the presence of surfactants in MWCNTs improves their dispersibility. Additionally, Wang et al. employed gum Arabic as a surfactant and acknowledged its superiority as the optimal selection [30].

A review of the technical literature shows that only a few researchers have succeeded in properly dispersing carbon nanotubes in water [31]. In this study, to achieve the optimal scattering of oxidized multi-walled carbon nanotubes in distilled water, through a modification technique, the influence of employing various surfactants to enhance the scattering of carbon nanotubes and consequently enhancing the structural characteristics of concrete was investigated and ultimately, a novel surfactant was introduced. For this purpose, various surfactants such as CTAB, sodium dodecyl sulfate (SDS), polyethylene glycol (PEG), and polycarboxylate (PCE) were used. Among the surfactants tested, combining polyethylene glycol and polycarboxylate provided good carbon nanotube dispersion and improved concrete mechanical properties. We have introduced a variety of polyethylene glycol and polycarboxylate as a new surfactant. This is because these two surfactants are compatible with the cement paste. Also, the dispersion of carbon nanotubes was more stable.

2. Materials and Methods

For concrete sample preparation, fine-grained and coarse-grained stones were used according to standard grading curves based on Iranian National Concrete Batching Plant. The ratio of fine grain used to the total fine grain and coarse grain used was calculated as 56%. The ratio of water to cement was chosen to be constant and equal to 0.48 for all samples. The characteristic compressive strength of the concrete was considered as 25 MPa. In this research, Portland type 2 Jovin cement with a strength class of 315 kg/cm^2 has been used. All the samples were processed in the same condition. The proportions and concrete mixing plants are given in Table 1. The samples containing carbon nanotubes with values of 0.015%, 0.030%, and 0.045% relative to the weight of cement (weight fraction) are named CNT0%, CNT0.015%, CNT 0.030%, and CNT0.045%. The used carbon nanotube is a multi-walled tube. Its length is 10-30 micrometers, and it is black. Carbon nanotubes are from Fine Nano company and made by US Nano. The complete information on multi-layered carbon nanotubes can be found in Table 2. Carbon nanotubes exist in a powdered state, initially undergoing acid modification to generate the functional group COOH. A novel surfactant, comprising polyethylene glycol and polycarboxylate, was employed to disperse the carbon nanotube powder in water. Polyethylene glycol is a polymer compound made from ethylene glycol (EG) that exhibits varying physical and chemical characteristics based on the quantity of EG units present in its molecular structure. These polymers are the most important commercial type of polyether and increase their hydrophilic properties. Additionally, a super lubricant based on polycarboxylate (PCE) obtained from Rezin Concrete Company has been utilized to enhance the distribution of carbon nanotubes in distilled water and cement paste, in order to achieve the desired consistency and flow...

Table 1.

Weight ratio of materials used in making concrete samples (For making 135 kg Concrete)

Sample	**1**	**2**	**3**	**4**
sample name	0	0.015% cnt	0.030% cnt	0.045% cnt
Cement (Kg)	27	27	27	27
Water (Kg)	12.5	12.5	12.5	12.5
Fine-grained agg. (Kg)	54.5	54.5	54.5	54.5
Coarse-grained agg. type 1 (Kg)	16	16	16	16
Coarse-grained type 2 (Kg)	25	25	25	25
carbon nanotube (gr)	0	4.05	8.1	12.1
Carbon nanotube /cement ratio%	0	0.015	0.030	0.045
amount of super lubricant in concrete production (gr) (0.5 percent of cement weight)	135	135	135	135
Amount of superlubricant in carbon nanotubes solution (gr) (Ratio of CNT to super lubricant 1:5)	0	20.25	40.5	60.5
Amount of polyethylene glycol in carbon nanotube solution (gr) (Ratio of CNT to PEG 1:5)	0	20.25	40.5	60.5

Table 2

Characteristics of carbon nanotubes used.

A type of carbon nanotube	Purity	Outer diameter (nm)	Inner diameter (nm)	length (micrometer)	specific surface $(\frac{m^2}{gr})$	True density $(\frac{gr}{cm^3})$
mutiwalled	>95	10-20	5-10	10-30	>200	~2.1

3. Functionalization of carbon nanotubes

To create carbon nanotubes hydrophilic, they are surface-modified using nitric acid (HNO3) and sulfuric acid (H2SO4) [32]. In order to alter and oxidize the exterior of multi-layered carbon nanotubes, the carbon exterior must initially be coated with nitric acid (HNO3, which causes oxidation of the exterior) and sulfuric acid (H2SO4, which creates a jagged exterior) [33]. Surface roughening of nanofibers is a procedure where carbon-carbon connections are fractured and imperfection sites are formed. This permits the nitric acid to generate a functional group on the nanofilaments. Then, according to Figure 1, a MWCNT solution paraffin bath with nitric acid and sulfuric acid was installed under the laboratory hood. Then, a balloon containing 30 ml of nitric acid, 90 ml of sulfuric acid, and 5 g of carbon nanotubes was placed inside the beaker along with a magnetic magnet. In this research, the ratio of sulfuric acid to nitric acid was 3:1 [34]. The ratio of nitric acid and sulfuric acid is crucial as altering the ratio of nitric acid to sulfuric acid either enhances or diminishes the extent of functionalization. Also, increasing the degree of functionalization disrupts the performance of carbon nanotubes and they are cut into smaller lengths. The duration of surface modification was considered as 3 h. Temperature is one of the important factors for oxidizing carbon nanotubes, because if the temperature exceeds a certain limit, the carbon nanotubes will shorten, and if it is lower than a certain limit, the carbon nanotubes will not be oxidized. The solution containing nitric acid, sulfuric acid, and carbon nanotubes was washed three times with Whatman paper with a diameter of 9 cm, which is connected to a vacuum pump. The pH of the solution is between 6.5 and 7, so the oxidized carbons are not acidic and don't cause the excessive arrangement of ettringite due to excess sulfate particles in the cement paste. Then, the carbon nanotubes placed on Whatman paper were placed in an oven set at 48 degrees for a day to dry. Thus, carboxyl functional groups are formed on the surface of carbon nanotubes, which are utilized as MWCNTCOOH. The carboxyl group is represented as COOH. Among the carboxyl, hydroxyl and carbonyl functional groups, the carboxyl functional group, due to its special conditions, causes a proper connection and bonding between the nanotubes and the concrete structure. Cement hydration is a complex process of interactions between water and the chemical phases of cement, tricalcium silicate (3CaO.SiO2 or C3S), dicalcium silicate (CaO.SiO2 or C2S), tricalcium aluminate (CaO.Al2O3) and tetracalcium alumino ferrite (CaO.Al2O3Fe2O3 or C4AF) which are hydraulic and occur as a result of interaction with water. Therefore, a range of hydrated phases are formed as a result of these interactions. Ca (OH) 2 and C-S-H are mainly produced as hydrated cement products. C-S-H gel and Ca(OH)2 are the two main products of the reaction of C3S and C2S with water. Therefore, in the structure of the hydration reaction products, namely calcium silicate hydrate and calcium hydroxide, there is a hydroxyl functional group, so through the carboxyl group, the nanotubes and the hydration reaction products are connected to each other [35].

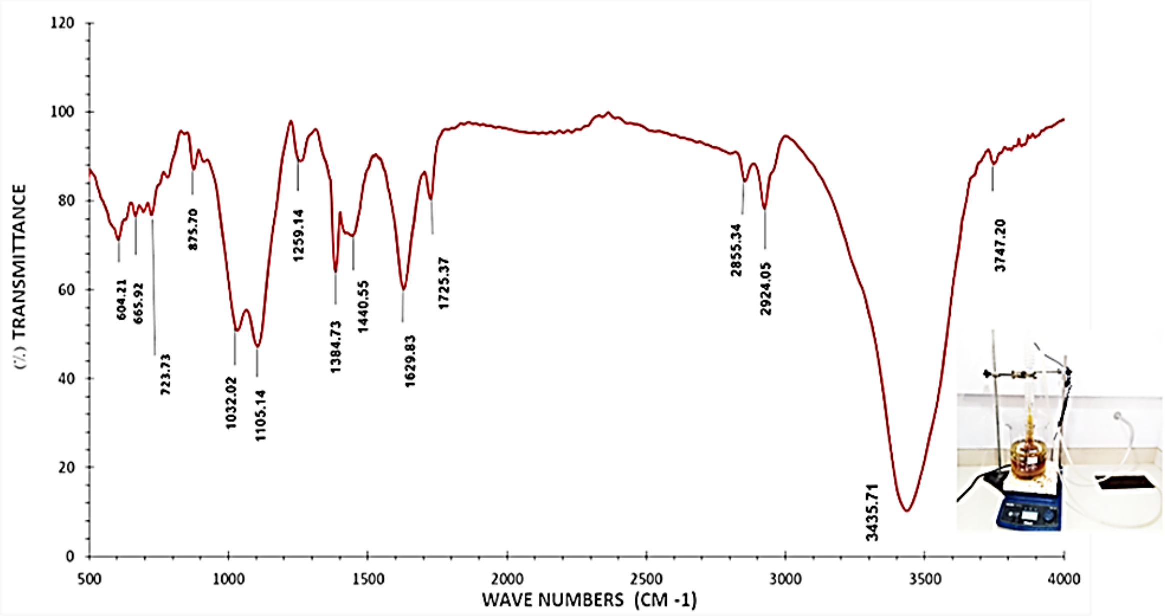


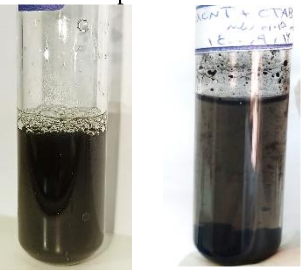
Figure 1: FTIR diagram of oxidized multi-walled carbon nanotubes

FTIR spectroscopy analyses were carried out on oxidized carbon nanotubes to qualitatively determine the hydroxyl and carboxyl groups linked to the surface of the carbon nanotubes and their level of functionalization [36], the findings of which are illustrated in Figure 1. In the mentioned diagram, each peak corresponds to a functional group and the intensity of each peak indicates that functional group in the carbon nanotube. Among the detectable absorption spectra in modified carbon nanotubes, the spectrum centered at wave number 3400 is observed, which indicates oxygen groups [37]. Additionally, the peaks associated with fluctuations in C=O stretching and C-O functional groups can be seen within the range of 1629.83, 1105.14, and 1384.73, respectively. These peaks are indicative of the existence of COOH-carboxyl groups. The results of FTIR spectroscopy clearly show that hydrophilic groups such as hydroxyl and carboxyl are well placed on the surface of functionalized CNTs.

4. Dispersion of carbon nanotubes

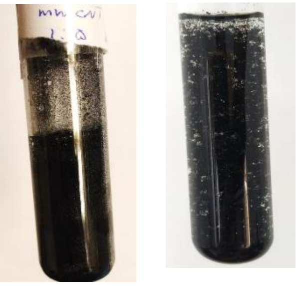
The dispersion of carbon nanotubes plays a crucial role in enhancing the mechanical characteristics of concrete. To prepare a homogeneous and uniform solution to have a favorable effect on concrete properties, carbon nanotubes should be well dispersed in water [38]. Modifications using covalent or ionic methods have been reported to disrupt the electronic band structure and intrinsic crystal structure of CNTs. Furthermore, CNT ropes or fragments can be observed in CNT-containing composites. Hence, physical adsorption of surfactants with long chains is commonly used in the dispersion of carbon nanotubes [39]. In this research, after the carbon nanotubes were surface modified using sulfuric acid and nitric acid and became hydrophilic, the modified carbon nanotubes were dispersed in a certain amount of water (30% of the total water required to make the samples) along with various surfactants (CTAB, SDS, polyethylene glycol (PEG) and polycarboxylate (PCE)). In this research, we functionalized carbon nanotubes with hydroxyl functional groups and the carbon nanotubes became hydrophilic, which functional group created in carbon nanotubes was confirmed in the FTIR test. Therefore, carbon nanotubes do not agglomerate and are easily dispersed in water. In this research, the carbon nanotubes hydrophilic were made using a hydroxyl functional group. It is worth mentioning that the carbon nanotubes with the functional group created in them, which was also confirmed in the FTIR test, are easily dispersed in water and do not agglomerate. However, in order for the dispersion of the water solution and carbon nanotubes to be durable and uniform, various surfactants have been used. It is important to note that the remaining blending water (70%) was utilized to saturate the aggregates and create concrete. In the first step, SDS was used as a surfactant, so that the ratio of SDS to carbon nanotubes was considered equal to 4:1, but no proper distribution and dispersion were observed. In the second step, CTAB (with a ratio of 1:1 compared to carbon nanotubes) was used as a surfactant. After 0.01g of carbon nanotubes and 0.01g of CTAB were poured into a certain amount of water and after 15 minutes of ultrasonic time, it was observed (immediately after ultrasonication) that the carbon nanotubes easily settled and separated from the solution. As can be seen in Figure 2, no sedimentation was observed until 90 minutes, indicating that the dispersion was improved; But as seen in Figure 2b, after one month, the solution containing CTAB settled, which shows that the dispersion of the solution containing CTAB surfactant was unstable. In the third step, polyethylene glycol (PEG), which is a hydrophilic polymer, increases the hydrophilicity of oxidized carbon nanotubes (the ratio of oxidized carbon nanotubes to PEG is 1:1, 1:3, and 1:5). be) was used. Ultrasonic was considered equal to 15 minutes. Observations indicate that by increasing the ratio of carbon nanotubes to PEG, the dispersity improves., but after 20 minutes, as seen in Figure 3, carbon nanotubes settled and agglomerated. In the fourth step, a solution containing oxidized carbon nanotubes, PEG, and polycarboxylate was prepared with a ratio of 1:5:5. The duration of ultrasonication was considered equal to 15 minutes and it was observed that a completely uniform solution was obtained. To observe the consistency of the solution, the solution was diluted at a 1:100 ratio, and no settling was observed again, indicating that the solution is very stable. As depicted in Figure 4, the solution remains consistent even after two months with no settling. Thus, the combination of polyethylene glycol and polycarboxylate is introduced as a novel surfactant. Generally, CNTs are dispersed using different surfactants, such as polycarboxylate-based superplasticizers, polyacrylic acid polymer, SDS, sodium dodecylbenzene sulfonate (SDBS or SDDBS), polyvinyl pyrrolidone (PVP), gum arabic (AG or GA), Pluronic F-127 and sodium deoxycholate (NaDC)[40]. CNTs adsorb the hydrophobic part of the surfactant on the side walls or ends of the tubes through van der Waals forces, π-π bonding or CH-π bonding, while the aqueous solubility and electrostatic repulsion are provided by the hydrophilic part of the surfactant [26]. The extent and strength of the electrostatic repulsion are determined by the density of polymers on the CNT surface and the length of the polymer chains. In the construction industry, polycarboxylate (PC)-based superplasticizers (SP) have been widely used to improve the workability and rheological properties of cement mixtures and reduce the amount of mixing water for cement. Polycarboxylate ester (PCE) is an example that has been successfully used for cement-based materials, and can also be used as a surfactant for CNT dispersion due to its better adsorption ability to CNT compared to polycarboxylate (PC) superplasticizers. However, although PCE is commercially available and is highly compatible with cementitious materials, a well-dispersed CNT solution can be obtained with relatively higher PCE values. Polyethylene glycol (PEG) is a non-ionic hydrophilic polyether that is widely used to modify polymer membranes due to its hydrophilicity. Studies show that PEG-modified polymer membranes can effectively reduce membrane fouling. Compatibility between surfactants and the cement matrix is an important issue. Parvin et al. reported that Pluronic F-127 surfactants can reduce porosity and improve microstructure density [40]. These surfactants, containing polyethylene oxide side chains, act similarly to polycarboxylate-based superplasticizers, which can disperse cement particles and improve mortar fluidity [41].

The dispersion of carbon nanotubes was also examined using UV-VIS spectrophotometry. UV-VIS spectroscopy was employed to establish the correlation between the quality of CNT dispersion, the duration of sonication, and the concentration of surfactant. This method is based on the Beer-Lambert law. The UV-Vis range is used to allow comparison of dispersion methods using absorbance values recorded for different suspensions. .



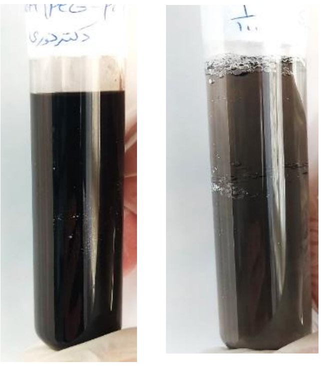
(a) (b)

Figure 2. Dispersion of carbon nanotube oxidized with CTAB: (a) solution containing MWCNTCOOH and CTAB 90 minutes after ultrasonication (b) solution containing MWCNTCOOH and CTAB one month after ultrasonication



(a) (b)

Figure 3. Dispersion of carbon nanotubes oxidized with PEG with a ratio of 1:5 (a) solution containing MWCNTCOOH and PEG immediately after ultrasonication (b) solution containing MWCNTCOOH and PEG 20 minutes after ultrasonication



(a) (b)

Figure 4. Dispersion of oxidized carbon nanotubes with PEG/PCE (a) solution containing MWCNTCOOH and PEG/PCE two months after ultrasonication (b) solution containing MWCNTCOOH and PEG/PCE two months after ultrasonication in a diluted form to better observe the dispersion.

The greater the level of scattering of MWCNTs in water, the greater the absorbance value measured. The maximum absorption intensity of individual her MWCNTs was reported by previous researchers to be 200–300 nm [42]. Because the amount of light absorbed is directly proportional to the concentration of carbon nanotubes, the level of dispersion is quantified using the Beer-Lambert law [43]. The gathering of MWCNTs diminishes the strength of the absorption band. This conduct may subsequently be connected to the level of scattering and presents the potential for ascertaining the ideal scattering circumstances. The solutions were ultrasonicated for 10 minutes before scanning with a UV-Vis spectrophotometer. Considering that Lambert Beer's law is established with good accuracy only at lower concentrations, the solution was mixed with purified water at a ratio of 1:100 [43]. Absorption spectra for all three solutions are shown in Figures 5. The absorption peak intensity is different in different surfactants, so the highest absorption was observed in the solution containing polycarboxylate surfactant and polyethylene glycol, which indicates the best dispersion. It is evident that the highest point of absorption at a range of 200-300 nm can be observed in the solution containing multi-walled carbon nanotubes, polyethylene glycol, and polycarboxylate.

5. Preparation and mixing of concrete samples:

Before making concrete samples, a carbon nanotube dispersed solution must be prepared firstly. In this research, two surfactants CTAB and PEG/PCE were used to make concrete samples with carbon nanotubes because these two surfactants had the best dispersion and the most stability among the surfactants used. The procedure of blending substances to create all the examinations is as follows: Initially, a specific quantity of components is blended with

60% of the total blending liquid for three minutes, then cement is introduced and subsequently, the mixture prepared in the prior step (comprising 30% of total water with carbon nanotubes or lacking carbon nanotubes) is incorporated. After 8 min, the specified amount of polycarboxylate-based superplasticizer was mixed in 10% total water and added to the mortar. It is important to note that carbon nanotubes soak up a significant quantity of water because of their immense specific surface area (which is responsible for many of their notable characteristics). This can cause the concrete mix to dry out, and therefore the use of superplasticizers is essential. Consequently, the utilization of superplasticizers leads to a more consistent and simpler dispersion of the solution of carbon nanotubes in cement mortar. A total mixing time of 10 minutes was assumed to produce the concrete. The concrete was subsequently poured into metal molds in three phases and compressed with 25 strokes of a unique rod (16 mm in diameter, 60 cm in length). The occupied molds were shaken for 1 min and then their exterior was made even. Specimens were removed from the molds after 24 hours and treated in water for 7, 14, and 28 hours.

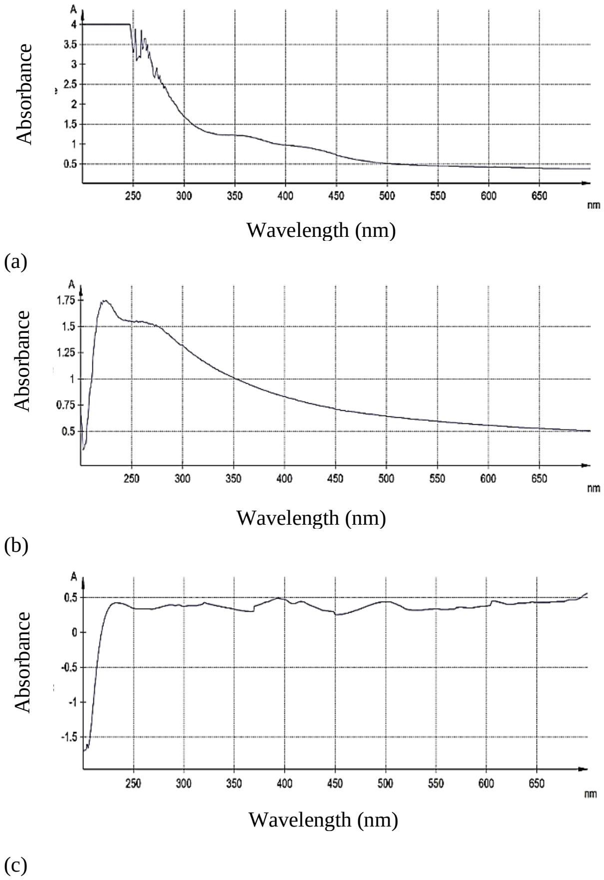


Figure 5: UV-vis test results:(a) for PEG/PCE (b) for CTAB (c) for PEG

6. Results and Discussion

6.1. Effect of carbon nanotube on compressive strength of concrete

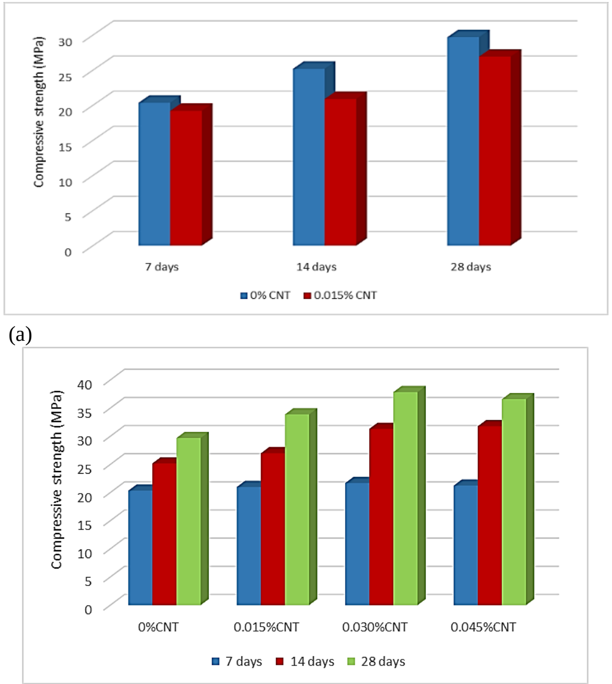
This examination is the most prevalent examination that is conducted on the strength of concrete. The examination was conducted in accordance with the guidelines of ASTM C39 at the durations of 7, 14, and 28 days using a hydraulic jack and with a rate of loading of 1 mm/min. As seen in Figure 6, the load is uniformly applied to the sample by two heavy hydraulic jaws. To conduct the experiment, standard cubic samples (15x15x15 cm) were used and the effect of adding multi-walled carbon nanotubes with different proportions of 0.015%, 0.030%, and 0.045% to the weight of cement was investigated.

Figure 7(a) displays the outcomes of compressive strength examination of concrete specimens incorporating carbon nanotubes and CTAB surfactant. The results indicate that the addition of 0.015% carbon nanotube with CTAB surfactant reduces the compressive strength by -5%, -17%, and -9% for 7, 14, and 28-day samples, respectively. This reduction can be attributed to the irregular effect of the CTAB agent on cement hydration. This surface-active agent can increase the quantity of air trapped in the cement mixture, thereby reducing the strength. Additionally, these varieties of surface-active agents experience reactions with water-reducing additives that could impede the process of hydration and lead to reformation, further reducing the strength. This surface-active agent can increase the quantity of air trapped in the cement mixture. Additionally, these varieties of surface-active agents experience reactions with water-reducing additives that could impede the process of hydration and lead to reformation. Figure 7(b) shows the compressive strength of concrete samples containing varying proportions of carbon nanotubes, polyethylene glycol, and polycarboxylate surfactants. Regarding the 7-day concrete samples, a rise in compressive strength was solely noticed in samples encompassing 0.030% of carbon nanotubes. This increase is marginal and seems to be due to insufficient distribution of the carbon nanotubes. The escalation in the compressive strength of 14-day concrete samples encompassing carbon nanotubes with proportions of 0.015%, 0.030%, and 0.045% is 7.14%, 24.3%, and 26.19%, correspondingly. This increase is thought to be due to the effective dispersion of carbon nanotubes. It is thought that this rise in power is attributed to the effective distribution of carbon nanotubes, which, alongside occupying the pores, reduce the porosity of the framework and assist in creating more compact cement-based composites. The findings of Hawreen and Bogas demonstrate that the surge in compressive strength in concrete specimens containing CNT in comparison to standard concrete is linked to the filling characteristics of CNT and their interconnection and bridging [44].

Figure 6. Compressive strength test on cubic sample

The findings also indicate that the highest rise in compressive strength in 28-day concrete samples is linked to samples containing 0.030% carbon nanotubes, thus resulting in a surge of approximately 27.4%. Importantly, concrete samples containing 0.015% and 0.045% carbon nanotubes exhibited a 14.14% and 23.4% boost in compressive strength at the 28-day mark, respectively. In simpler terms, as the quantity of carbon nanotubes increased, the upward trend in strength improvement ceased, and the optimal value of the 28-day compressive strength was observed in the samples containing 0.030 carbon nanotubes. It is important to note that excessively increasing the amount of carbon nanotubes may halt the process of increasing compressive strength, as high CNT concentrations may lead to the re-accumulation of carbon nanotubes. In other words, it can be said that smaller amounts of carbon nanotubes may disperse better and thus stop the propagation of microcracks and reduce nano-sized holes [45]. The decrease in strength of CTAB-containing concrete can be attributed to the incompatible effect of CTAB on cement hydration, which leads to a delay in the cement hydration process. This surfactant can increase the amount of air entrapped in the cement paste to the extent seen in the fresh mix. In addition, these types of surfactants undergo reactions with water-reducing admixtures that may delay hydration and cause re-agglomeration. As previously mentioned, the instability of the CTAB-containing solution resulted in the lack of proper dispersion of carbon nanotubes in water and cement mortar, resulting in a lack of improvement in the mechanical properties of concrete.

The higher strength achieved is believed to be due to the physical contribution of well-dispersed carbon nanotubes, which act as pore fillers and reduce matrix porosity, contributing to denser cementitious composites. The bridging effects of nanotubes can also enhance the ability of the materials to delay microcrack formation and increase the load-bearing capacity of the cement matrix by bridging adjacent hydration products. There are four main mechanisms by which carbon nanotubes strengthen cementitious matrices, the first of which is the crack-bridging mechanism mediated by carbon nanotubes. Carbon nanotubes can bridge nano- and micro-sized pores or gaps in the range of 10–103 nm in hydration products. This bridging effect of carbon nanotubes provides efficient charge transfer from the matrix to the carbon nanotubes within the composites, thereby delaying the growth of nano-sized pores or cracks and preventing the formation of micro-sized pores or cracks. Carbon nanotubes reduce the nano-porosity of the cement paste by filling the pores and cracks between the hydrate gel, thus CNTs enhance the mechanical properties [40]. In addition, carbon nanotubes can act as nucleating agents for C-S-H gels. The hydrophilic nature of the CNT structure due to the functional groups (or surfactants) allows water to be more effectively adsorbed onto it, thereby producing hydration products. There are two mechanisms of nucleating agents for enhancing mechanical properties. One is that such nucleating agents can improve the hydration process, leading to a denser and stronger microstructure. Furthermore, the C-S-H gel coated with CNT bundles leads to a much larger effective contact area between the CNT and the matrix, resulting in an extremely strong bond between them [46].



(a)

(b)

Figure 7. Compressive strength of concrete containing (a)carbon nanotubes and CTAB surfactant at different ages (b) Carbon Nanotubes, Polyethylene Glycol, and Carboxylic Acid Poly surfactant at different ages

6.2. The effect of carbon nanotube on the tensile strength of concrete

In this investigation, a technique of dividing shaped or cored cylindrical samples was employed to assess the pulling power of concrete. As depicted in Figure 8, applying a diagonal compressive force on 28-day cylindrical concrete samples (diameter 15 cm and height 30 cm) containing different amounts of carbon nanotubes, which are placed horizontally between two plates of the test device and the tensile strength is determined and compared.

Figure 9(a) shows the tensile strength of concrete samples containing varying amounts of carbon nanotubes with PEG and polycarboxylate surfactants at 28 days. It is evident that the tensile strength rises of the specimens with carbon nanotube concentrations of 0.015%, 0.030%, and 0.045% are 16.7%, 28.5%, and 19.7% correspondingly. As can be seen, the increasing trend in tensile strength stops for the samples containing carbon nanotubes as high as 0.045%. This is due to the redistribution of carbon nanotubes in the cement matrix. Table 3 presents a comparison between the ideal quantity of carbon nanotubes in this research and the ideal quantity of carbon nanotubes in earlier studies. It is evident that the optimal amount of carbon nanotubes in the current investigation is lower than the figure in the prior study., the reason can be the proper performance of the new combined surfactant in this research, which has led to a more favorable dispersion of carbon nanotubes. Figure 9(b) compares the tensile strength of concrete samples containing carbon nanotubes with CTAB surfactant, polyethylene glycol, and polycarboxylate surfactants. It is apparent that the pulling power of concrete with CTAB surfactant diminished by -4.17% in comparison to the standard sample.

Figure 8. Testing the tensile strength of the cylindrical sample

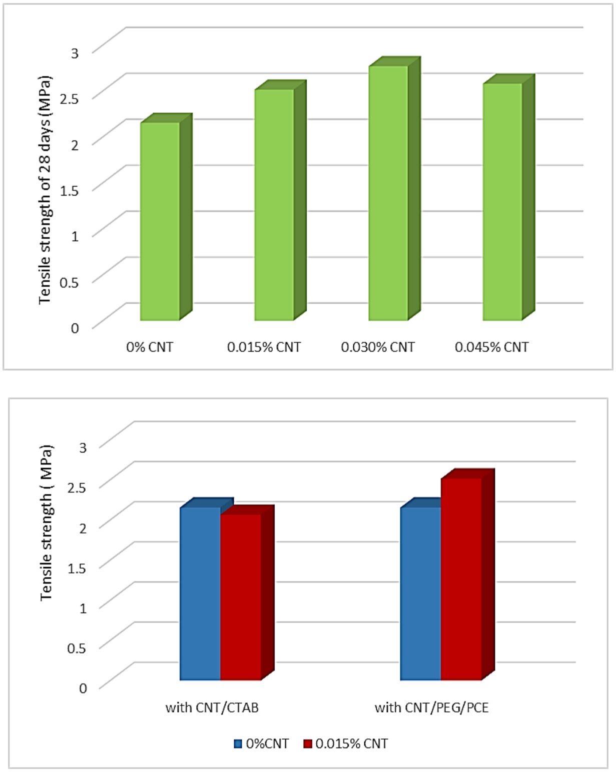


Figure 9. Tensile strength of concrete containing (a) different amounts of carbon nanotubes and polyethylene glycol and polycarboxylate surfactants (b) carbon nanotubes and CTAB surfactant with concrete containing carbon nanotubes and polyethylene glycol and polycarboxylate surfactants

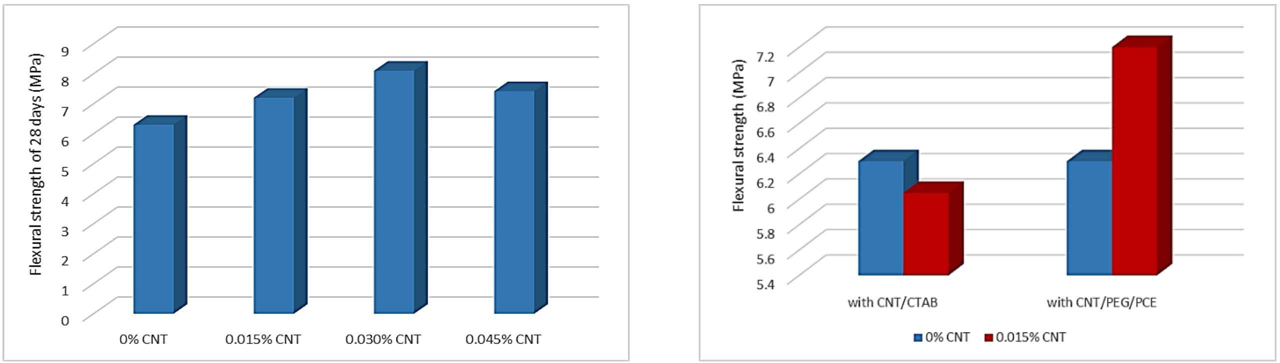
Figure 10. Flexural strength test of prismatic beam

6.3. Effect of carbon nanotube on flexural strength of concrete

As depicted in Figure 10, the bending strength test is performed on a rectangular concrete cube of 100 x 100 x 500 mm for 28 days. This research test was performed based on the ASTM C293 standard using the three-point method.

Table 3
Comparison of optimal values of carbon nanotubes in different studies

The type of surfactant	Measured mechanical properties	Percentages of CNTs used (%)	Measured strength values (MPa)			Optimum percentage of CNT (%)	Reference
super lubricant	flexural strength	0.030% 0.08% 0.15% 0.25%	2.9 2.6 4 4.25			0.25%	[47]
super lubricant	compressive strength	0.06% 0.12%	31 25			0.06%	[21]
-	compressive strength Tensile strength	0.01% 0.02% 0.03%	compressive strength		Tensile strength	0.03%	[48]
			31.2 33 35		3.3 3.5 3.6		
Polyethylene glycol and polycarboxylate surfactant	compressive strength flexural strength Tensile strength	0.015% 0.030% 0.045%	compressive strength	flexural strength	Tensile strength	0.03%	current article
			33.6 37.86 36.6	7.2 8.1 7.42	2.51 2.77 2.58		



(a) (b)

Figure 11. Flexural strength of concrete (a) containing different amounts of carbon nanotubes and polyethylene glycol and polycarboxylate surfactants (b) carbon nanotubes and CTAB surfactant with concrete containing carbon nanotubes and polyethylene glycol and polycarboxylate surfactants.

Figure 11(a) shows the flexural strength of concrete samples containing varying amounts of carbon nanotubes, polyethylene glycol, and polycarboxylate surfactants for 28 days. It has been noticed that augmenting the quantity of carbon nanotubes in concrete enhances the bending strength of the concrete. This increase in bending strength for samples containing 0.015%, 0.030%, and 0.045% of carbon nanotubes is equal to 14%, 28.5%, and 17.7%, respectively. These results confirm the findings of previous researchers. Put differently, approximately 70% or higher of prior research has indicated that the incorporation of carbon nanotubes enhances flexural strength by 50% or greater [49]. Current findings suggest that the impact of carbon nanotubes on enhancing the bending resistance of the cement structure is more significant compared to its influence on compressive strength. Sedagatdoost and Behfarnia have linked the improved bending resistance of the carbon nanotube mixture to its spatial arrangement, which involves filling the gaps between C-S-H and decreasing the porosity of the cement structure [50]. The rise in flexural potency is thought to be caused not just by the filling of moistened pores but also by the linking impact of carbon nanotubes and the sealing of tiny fractures. This fortifies the framework and adds to the enhancement of bending strength [51]. The optimal amount of carbon nanotubes to produce maximum bending strength is 0.030% relative to the weight of the cement, and increasing amounts of carbon nanotubes stop the trend of increasing bending strength. Figure 11(b) displays the flexural strength of concrete samples incorporating carbon nanotubes and CTAB surfactant over a period of 28 days. It is observed that the inclusion of carbon nanotubes in conjunction with the CTAB surfactant resulted in a decrease of 3.9% in the flexural strength at the end of the 28-day period. Conversely, the utilization of polyethylene glycol and polycarboxylate surfactants led to an increase of approximately 16% in the 28-day flexural strength of concrete samples containing carbon nanotubes.

6.4. Effect of nanotube carbon on specific weight of concrete

Figure 12 shows the specific weight of concrete samples containing different amounts of carbon nanotubes and polyethylene glycol and polycarboxylate surfactants in two states of hardened concrete and fresh concrete. It is evident that the incorporation of carbon nanotubes decreases the specific density of concrete in comparison to the control concrete sample. These results support the findings of other researchers. These results confirm the findings of other researchers. The findings of a study conducted by Hawreen and Bogas similarly demonstrated that incorporating carbon nanotubes at a concentration of 0.1% of the cement mass had a minimal impact on the performance and density of the concrete, suggesting that the dispersion of carbon nanotubes was unaffected [44]. This figure shows that carbon nanotubes' specific gravity reduction effect is more significant in hardened concrete than in fresh concrete. The largest reduction in specific weight is observed for hardened concrete containing 0.030% carbon nanotubes, which is about 10%, while fresh concrete containing the same amount of carbon nanotubes has a specific gravity reduction of about 3%. Note that increasing concrete strength is usually accompanied by increasing specific gravity; However, the unique properties of carbon nanotubes also positively affect the compressive strength and specific weight of concrete. As one of the critical problems in building design and implementation is the dead weight of the building, reducing the dead weight of the building and using concrete with lower the specific weight and higher compressive strength in concrete structures has been the focus of many design engineers. This fact is important because the seismic force acting on a structure is proportional to the mass of the system, and reducing the group of the design is the most critical factor in mitigating the impact of earthquakes on the form [52]. Therefore, carbon nanotubes solve this need and produce concrete with high compressive strength and low specific weight.

7. Conclusion

Laboratory experiments demonstrate that adequate scattering of carbon nanotubes enhances the structural characteristics of concrete. In this study, by means of the surface alteration technique, carbon nanotubes were oxidized and discovered to possess hydrophilic characteristics; Nevertheless, the ideal scattering and steadiness of carbon nanotube dispersion in water were insufficient. Therefore, different surfactants (such as CTAB, SDS, PEG, and PCE) were used. Finally, the

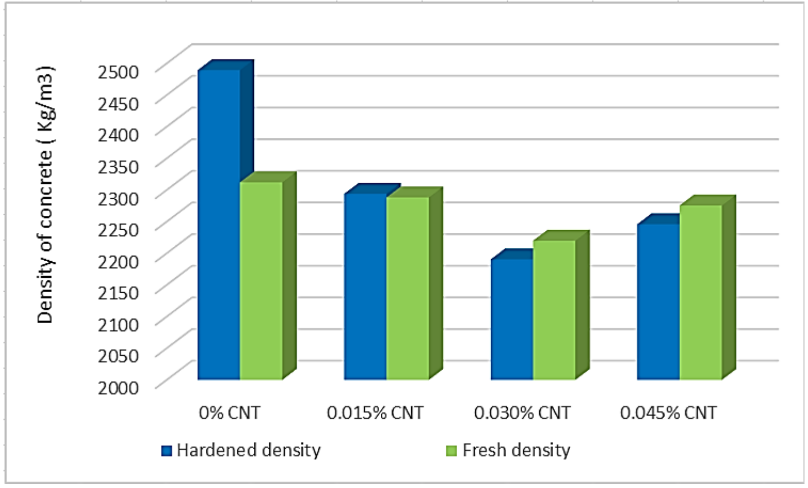


Figure 12. Specific weight of 28-day concrete samples containing different percentages of carbon nanotubes and polyethylene glycol and polycarboxylate surfactants

CTAB surfactants and PEG/PCE combination caused better dispersion and homogeneous distribution of carbon nanotubes. Observations showed that the solution containing PEG/PCE surfactant is more stable than the solution containing CTAB surfactant. In other words, the solution containing CTAB surfactant settled after one month, while the solution containing PEG/PCE surfactant did not settle after two months. Therefore, two surfactants, CTAB and PEG/PCE were used to make concrete samples due to their uniform distribution and optimal dispersion. The findings indicated that the compressive strength of concrete samples incorporating CTAB surfactant did not experience significant enhancement; the reason for this can be the incompatibility of the CTAB surfactant with the cement matrix, this slows down the hydration process and destabilizes the dispersion of solutions containing carbon nanotubes with CTAB surfactant, which results from the uneven dispersion of carbon nanotubes in cement mortar. Finally, a PEG/PCE surfactant combination was selected as a suitable surfactant for carbon nanotube dispersion to improve concrete's mechanical and physical properties. So, in summary, we can say that using different surfactants gives different results. The remedy with the ideal quantity of 0.030% of carbon nanotube and PCE/PEG surfactant enhanced the compressive, bending, and tensile resistance of concrete by 27.49%, 28.57%, and 28.53%, and the higher the amount of carbon nanotube, the increasing trend of compressive strength decreased. The carbon nanotubes also had a favorable effect on the specific gravity of concrete, decreasing the specific gravity of hardened concrete by about 10% and that of fresh concrete by about 3%. The suggestions for future research could include as: investigating the mechanism of chemical adsorption and the effect of different molecular weights of polycarboxylate and polyethylene glycol, adsorption on the outer walls of nanotubes and surfactant agents on nanotubes following high ultrasonic intensity on dispersion, investigating methods for improving the dispersion of nanotubes for large-scale cement applications (because the use of nanocomposites for large-scale structural applications is encouraging), and investigating the effect of carbon nanotubes on thawing and freezing, shrinkage, creep, corrosion, and chloride infiltration of concrete.

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