Article

Effect of Electrospun Nanofiber Additive on Selected Mechanical Properties of Hardened Cement Paste

Tri N.M. Nguyen ¹ , Do Hyung Lee ² and Jung J. Kim ^{1,*} ¹ Department of Civil Engineering, Kyungnam University, Changwon-si 51767, Korea; nnmtri@utc2.edu.vn² Department of Civil, Railroad and Unmanned Systems Engineering, PaiChai University, 155-40 Baejaero, Seo-gu, Daejeon 35345, Korea; dohlee@pcu.ac.kr

* Correspondence: jungkim@kyungnam.ac.kr

Received: 4 October 2020; Accepted: 23 October 2020; Published: 26 October 2020

Abstract: This study presents an estimation of the mechanical property enhancing mechanism of electrospun-nanofiber-blended cementitious materials. Four types of electrospun nanofibers (NFs) were introduced into Portland cement: nylon 66 nanofibers, tetraethyl orthosilicate/polyvinylpyrrolidone nanofibers (TEOS/PVP), hybrid nanofibers containing carbon nanotubes (CNTs) and nylon 66, and hybrid nanofibers containing carbon nanotubes and TEOS/PVP (CNTs-TEOS/PVP NFs). Due to the mechanical strength results, there was an increase of 27.3% and 33.4% in compressive strength when adding TEOS/PVP nanofibers and the hybrid nanofibers containing carbon nanotubes and TEOS/PVP into the pastes, respectively. In addition, there was an increase of 25.7% and 54.3% in tensile strength when adding nylon 66 nanofibers and the hybrid nanofibers containing carbon nanotubes and nylon 66 into the pastes, respectively. The highest toughness of 61.7% was obtained by the paste blended with CNTs-TEOS/PVP NFs. The results observed from scanning electron microscopy, transmission electron microscopy, and thermogravimetric analysis clarified the change in the microstructure of the modified pastes, as well as the mechanical property enhancing mechanism of the electrospun-nanofiber-blended cementitious materials.

Keywords: TEOS/PVP nanofibers; nylon 66 nanofibers; cement; electrospinning; carbon nanotubes; mechanical strength

1. Introduction

Electrospinning has been viewed as a strong technique in nanomaterials science due to its simplicity and flexibility [1,2]. The basic principle of this technique is the electrostatic interaction between two electrodes [3]. Electrospun nanofibers have many promising properties, such as their high strength, rough surface, high specific surface area, good functional abilities, and so forth. Therefore, the feasibility of the industrial use of electrospun nanofibers has been clarified through numerous works. According to a literature review, various types of composite fiber membranes show a good ability for use in wastewater treatment, with applications in oil/water mixture separation [4,5], oil filtration [6], and heavy metal ion adsorption [7]. In the biomedical industry, nanofibers also have impressive applications as treatments of skin carcinoma [8], tissue regeneration [9], and wound healing [10]. In addition, the tremendous application of electrospun nanofibers has been recognized in other industries such as the textile industry [11–13], the battery industry [14–16], and so forth. However, it seems that fewer works have focused on the application of electrospun nanofibers to increase the mechanical properties of construction materials, especially of cementitious materials. From the literature review, it is worth mentioning that the application of nanosized short fibers to enhance the mechanical properties of cement has been conducted and has gained a considerable amount of attention in the research community. For instance, Azevedo and Cleize [17] reported an increase in

the mechanical characteristics of Portland cement paste that included silicon carbide nanowhiskers (SiC NWS): 25% in compressive strength and 75% in flexural strength when using a silicon carbide nanowhisser proportion of 0.25–1.5 wt %. They observed the bridging phenomenon of SiC NWS across the pores and cracks inside the cement paste, showing good linkage between them. Barbhuiya and Chow [18] showed an increase in compressive strength of cement when using 0.2 wt % of carbon nanofibers (CNFs) in the paste. They found the crack bridging effect of CNFs in the microscale structure, as well as an increase in the high-density calcium–silicate–hydrate (CSH) gel. Li et al. [19] pointed out an enhancement of up to 19% in compressive strength and up to 25% flexural strength when introducing carbon nanotubes into cementitious materials. They found that CSH layers wrap tightly around the fibers in the microstructure of the paste, indicating good bonding between cement hydration products and the nanosized fibers. Rocha et al. [20] reported an increase of 45% in tensile strength and 46% in flexural strength when adding 0.1% of carbon nanotubes (CNTs) into the pastes. From the micrographs, they found that CNT filaments were embedded and well-adhered to cement paste. However, increasing the mechanical strength of cement by blending with electrospun nanofibers has not been widely studied. Therefore, this present work aimed to evaluate the feasibility of using electrospun nanofibers to enhance the mechanical properties of Portland cement paste, as well as the enhancing mechanism of electrospun nanofibers in the cement matrix.

Four types of electrospun nanofibers were utilized for combination with Portland cement powder in this study: nylon 66 nanofibers (N66 NFs), tetraethyl orthosilicate/polyvinylpyrrolidone nanofibers (TEOS/PVP NFs), hybrid nanofibers containing carbon nanotubes and nylon 66 (CNTs-N66 NFs), and hybrid nanofibers containing carbon nanotubes and TEOS/PVP (CNTs-TEOS/PVP NFs). The combination process was conducted by means of an improved collector, details of which were published in previous works [21–23]. The observations from the compressive strength test and the tensile strength test showed the effectiveness of the electrospun nanofibers on the mechanical properties of the hardened cement pastes. In addition, the enhancing mechanism of the mechanical properties of nanofiber-blended cementitious materials was found from the results of transmission electron microscopy (TEM), scanning electron microscopy (SEM), as well as thermogravimetric analysis (TGA).

2. Materials and Experimental Work

2.1. Characteristics of the Cement Used

The ordinary Portland cement (OPC) used in this work was purchased from Ssangyong Co., Korea—a type I cement that complies with the specifications of ASTM C150 [24]. The chemical composition and physical properties of the cement are shown in Table 1.

Table 1. Chemical composition and physical properties of the cement.

CaO	Al ₂ O ₃	SiO ₂	SO ₃	MgO	Fe ₂ O ₃	Ig. Loss	Specific Surface Area (cm ² /g)	Compressive Strength, 28 Days (MPa)
61.33	6.40	21.01	2.30	3.02	3.12	1.40	2800	36

2.2. A Method of Obtaining a Blend of Electrospun Nanofibers and Cement

The chemicals and precursors used in this study include poly(hexamethylene adipamide) (Nylon 66, Sigma-Aldrich, Louis, Missouri, USA, density: 1.14 g/mL), formic acid (Daejung, Siheung-si, Gyeonggi-do, Korea, density: 1.22 g/mL), chloroform (Daejung, Korea, density: 1.492 g/mL), tetraethyl orthosilicate (TEOS, Alfa Aesar, Heysham, Lancashire, UK, density: 0.934 g/mL), polyvinylpyrrolidone (PVP, Sigma-Aldrich, USA, density: 1.2 g/mL), butanol (Sigma-Aldrich, USA, density: 0.81 g/mL), and multiwalled carbon nanotubes (97% purity, 10 nm outer diameter, 30 µm length, Hanos, Jung-gu, Seoul, Korea). All chemicals were used as received.

Four polymer solutions were prepared in this study. To prepare the nylon 66 polymer solution, first of all, formic acid was diluted by chloroform with the volume proportion of 4:1 for forming the solvent. Then, nylon 66 was dispersed in this solvent with a weight proportion of 10%. To prepare the TEOS/PVP polymer solution, firstly, a solution was made by stirring TEOS and butanol with the volume proportion of 5:3, then merging PVP in this solution to yield a 10% polymer solution. The polymer solution containing CNTs and nylon 66 as well as the one containing CNTs and TEOS/PVP were prepared by the following steps: First, the solutions were prepared by adding CNTs into the presolution containing formic acid and chloroform, as well as the presolution containing TEOS and butanol, as mentioned above. It is worth mentioning that the added content of CNTs had to compensate for the content of nylon 66 or PVP that was taken out of the above solutions. In this work, the added CNT proportion of 0.4 wt % was chosen for the polymer solution. The polymer solution proportions are shown in Table 2. However, dispersing CNTs in a liquid solution is a complicated process due to its high aspect ratio, and the agglomeration phenomenon tends to occur under the strong van der Waals forces between CNT molecules [25]. Therefore, at this period, to obtain homogeneous solvents, an ultrasonication process was conducted to break the connection between CNT molecules [26–28]. Finally, nylon 66 and PVP were added into each solution with a consistent proportion of 9.6 wt % to obtain the final polymer solution containing CNTs and nylon 66, as well as the one containing CNTs and TEOS/PVP.

Table 2. Component proportions of polymer solutions by weight percent (%).

Polymer Solution	Presolution	N66	PVP	CNTs
N66 polymer solution	90 (Formic acid + Chloroform)	10	-	-
CNTs-N66 polymer solution	90 (Formic acid + Chloroform)	9.6	-	0.4
TEOS/PVP polymer solution	90 (TEOS + Butanol)	-	10	-
CNTs-TEOS/PVP polymer solution	90 (TEOS + Butanol)	-	9.6	0.4

N66: nylon 66; CNT: carbon nanotube; TEOS: tetraethyl orthosilicate; PVP: polyvinylpyrrolidone

The electrospinning process for fabricating nanofibers, as well as combining nanofibers with cement powders, was conducted according to the procedure presented in previous works [21–23]. The whole procedure is shown in Figure 1.

Figure 1. The setup of the electrospinning process. OPC: ordinary Portland cement.

The input parameters for the electrospinning process were set consistently for the four types of nanofiber-blended cementitious materials, as summarized in Table 3. To obtain good dispersion and

avoid the gathering phenomenon of electrospun nanofibers in cement, the proportion of polymer solution of 5 wt % was chosen for the composite cementitious materials. Summarily, CNTs were utilized with a proportion of 0.4% of the polymer solution. Then, the electrospinning processes were conducted using these polymer solutions with a proportion of 5 wt % of the blended binder. Consequently, the proportion of CNTs in the blended binder was 0.02 wt %. It is worth noting that, for analyzing the morphological properties of nanofibers by TEM, the nanofibers were electrospun directly onto the copper grids that were fixed on the plate collector.

Table 3. Input factors of the electrospinning process.

Applied Voltage (kV)	Syringe's Volume (mL)	Taylor Cone Gauge	Needle-to-Collector Distance (mm)	Pumping Speed ($\mu\text{L}/\text{min}$)
12	12	20	60	30

2.3. Sample Preparation and Testing Methods

As is known, research on the mechanical properties of hardened cement pastes is very difficult to conduct, which is due to the changes in their volume and the difficulty of controlling spontaneous cracking during the maturation process. For this reason, cement paste samples usually have very small dimensions. Figure 2 shows the briquette and cubic samples used for the tensile strength test and the compressive strength test in compliance with the specifications of ASTM C307-03 (2012) [29] and ASTM C109/109M-16a [30], respectively. A set of three briquettes and three cubic samples of each composite cementitious material as well as of the plain cement paste was prepared with the constant water/binder ratio of 0.5. The water/binder ratio was chosen for consistency with previous works as well as for comparison with the literature [21,22]. All of the samples were matured for 28 days in water under laboratory conditions, namely, an ambient temperature of $23 \pm 2^\circ\text{C}$ and relative humidity of 50%. The SEM samples were prepared by using a cross section obtained from the failure sample from the tensile strength test. SEM samples had a square section with a polish surface and dimensions of $5 \times 5\text{ mm}$. For thermogravimetric analyses, five pulverized samples were prepared.

Figure 2. Samples for mechanical strength tests: (a) briquette samples; (b) cubic samples.

In this study, the tensile strength tests were conducted by means of a mortar tensile strength test device with a capacity of 5 kN, in compliance with ASTM C307-03 (2012) [29]. The compressive strength tests were conducted by means of a hydraulic universal testing machine with a capacity of 1000 kN according to ASTM C109/109M-16a [30]. The SEM analyses were conducted using the Zeiss Merlin Compact system, with input parameters that were set up as follows: an accelerating voltage of 3–5 kV and a working distance of 7.1–7.9 mm. To obtain the highest resolution of SEM results, the samples were coated with a 5 Å platinum layer. The TEM analyses were done using the FEI Tecnai F30 Twin system, where the input parameters were set up as follows: an acceleration voltage of 300 kV. In addition, the TGA analysis was conducted using a TA Instruments SDT-Q600, and the input parameters were set as follows: nitrogen atmosphere condition, a flow rate of 100 mL/min, heating velocity of 10 °C/min, and a temperature range from room temperature to 1000 °C.

3. Results and Discussion

3.1. Mechanical Strength

The influence of electrospun nanofibers on the 28-day mechanical properties is shown in Figure 3 (where “MCP” means “modified cement paste”). From an overall perspective, it is evident that the mechanical strength rose significantly in most cases of electrospun-nanofiber-modified cement pastes. As is illustrated in Figure 3a,b, the increase in compressive strength of the hardened pastes containing TEOS/PVP-based nanofibers was stronger than that of the pastes containing nylon-66-based nanofibers. For instance, compared with the compressive strength of the hardened plain cement paste, the compressive strength of the hardened pastes containing TEOS/PVP NFs and the hybrid CNTs-TEOS/PVP NFs increased 27.3% and 33.4%, respectively, and that of the hardened pastes containing N66 NFs and the hybrid CNTs-N66 NFs increased 6.9% and 14.8%, respectively. These observations show the effectiveness of TEOS/PVP-based nanofibers at increasing the compressive strength of cement materials. Figure 3c shows the toughness results calculated from the stress–strain curves in Figure 3a. Theoretically, the material toughness can be calculated by the area under the stress–strain curve [31]. The observation from Figure 3c shows a significant increase in the toughness of 16.0%, 18.2%, 47.3%, and 61.7% of the pastes containing N66 NFs, CNTs-N66 NFs, TEOS/PVP NFs, and CNTs-TEOS/PVP NFs, in that order, compared with the toughness of the hardened plain cement paste. In addition, Young’s modulus can be observed from the slope of the stress–strain curve. It is worth mentioning that despite changing the composition of the material, Young’s modulus remained unchanged, as shown in Figure 3a. Furthermore, there was an increase in the ductile deformation of the nanofiber-blended cement pastes compared with that of the plain paste, namely, increases of 10%, 13.3%, 26.7%, and 30% when introducing N66 NFs, CNTs-N66 NFs, TEOS/PVP NFs, and CNTs-TEOS/PVP NFs into the cement pastes, respectively. Table 4 summarizes the ductile deformation ratio of the blended paste to the plain paste. In contrast, the observations from the tensile strength tests show the effectiveness of nylon-66-based nanofibers at enhancing the tensile characteristics of Portland cement (see Figure 3d). There was a sharp increase in tensile strength in the hardened cement pastes containing N66 NFs and CNTs-N66 NFs of 25.7% and 54.3%, respectively, compared with that of the plain paste, while the tensile strength of the hardened cement pastes containing TEOS/PVP NFs and CNTs-TEOS/PVP NFs rose slightly, with an average increase of 5.7% and 17.1%, respectively. These observations show an important role of nylon-based nanofibers in increasing the tensile strength of Portland cement. In addition, the role of CNTs in reinforcing the nanofibers is clarified through these results.

Figure 3. The 28-day mechanical properties: (a) the 28-day stress–strain curves; (b) the 28-day compressive strength results; (c) the 28-day toughness results.; (d) the 28-day tensile strength results.

Table 4. Ductile deformation ratio.

	Fracture Strain	Ductile Deformation Ratio
Plain paste	0.003	1.00
N66 NFs MCP	0.0033	1.10
CNT-N66 NFs MCP	0.0034	1.13
TEOS/PVP NFs MCP	0.0038	1.27
CNTs-TEOS/PVP NFs MCP	0.0039	1.30

MCP: modified cement paste; NFs: nanofibers.

As mentioned above, the proportion of CNTs can be relatively estimated to be 0.02 wt % of the blended cementitious materials. In the literature, Li et al. [19] reported an increase of 19% in compressive strength when adding 0.5 wt % treated CNTs into the paste, and Rocha et al. [20] reported an increase of 45% in tensile strength when adding 0.1 wt % CNTs into the paste. This present work reports an increase of 33.4% in compressive strength and 54.3% in tensile strength when adding CNTs-TEOS/PVP NFs and CNTs-N66 NFs into the paste (0.02 wt % CNTs), respectively, which shows the feasibility of an indirect approach of using CNTs in enhancing the mechanical properties of cementitious materials.

3.2. SEM and TEM Results

Figure 4 presents the cross-section morphologies of the modified hardened cement pastes including electrospun nanofibers. As is illustrated, some common cement hydration products can be observed, such as calcium hydroxide (CH) and calcium–silicate–hydrate (CSH), with the morphologies of large prismatic crystals and small fibers, respectively [32]. The electrospun nanofibers with a diameter of around 200–300 nm could interleave easily in the space from 10 nm to 1 μm of the capillary cavities or voids, which were created when the initial spaces occupied by water could not become totally filled by the hydrated products such as CH or CSH [32]. Therefore, these electrospun nanofibers not only act as the bridging agents, linking the hydrated products physically, but also act as the filling agents, filling up the spaces among the hydrated products. As a result, the existence of electrospun nanofibers inside the cement matrix can enhance the mechanical properties of this material. Based on the model of the well-hydrated Portland cement paste from [32], the bridging effect as well as the filling effect of nanofibers in the matrix can be summarized schematically (Figure 5).

Figure 4. The microstructure of hardened cement paste containing electrospun nanofibers: (a) the paste including N66 NFs; (b) the paste including CNTs-N66 NFs; (c) the paste including TEOS/PVP NFs; (d) the paste including CNTs-TEOS/PVP NFs.

From the tensile strength test results presented above, the tensile strength of the group containing nylon-66-based nanofibers was higher than that of the group containing TEOS/PVP-based nanofibers. This observation can be explained due to the structure of the electrospun nanofibers, as presented in TEM images (see Figure 6). As shown in Figure 6, the structure of nylon-66-based nanofibers presented a solid and dense state [33], in contrast with the porous and hollow structure of TEOS/PVP-based nanofibers [34]. As a result, the density of the structure of nylon 66 nanofibers resulted in the higher tensile strength among the nanofiber-modified cement pastes. In addition, the existence of CNTs inside

the electrospun nanofibers was observed from the TEM images, as shown in Figure 6. The nanofibers with a diameter of around 200 nm were strengthened by CNTs with a diameter of around 10 nm. As a result, the tensile strength of the hardened cement pastes containing the hybrid electrospun nanofibers was higher than that of the pastes containing the raw electrospun nanofibers. Besides the bridging effect, nanofibers also formed among the hydrates and interleaved among them as mentioned above. Therefore, the porous structure of the cement matrix can be filled up by these nanofibers. Consequently, more compacted structures of hardened cement pastes were formed, and greater compressive strength was observed.

Figure 5. The bridging and filling mechanism of electrospun nanofibers in the hydrated cement paste.

Figure 6. TEM images of electrospun nanofibers: (a) the hybrid nanofiber containing CNTs and nylon 66; (b) the hybrid nanofiber containing CNTs and TEOS/PVP.

3.3. Thermogravimetric Analysis Results

Figures 7 and 8 present the thermogravimetric analysis (TGA) and the derivative thermogravimetric (DTG) results of the five samples in this study. As shown by the DTG curves in Figure 7, the boundaries of various phases or groups of phases in the hardened cement paste samples were determined and these results also agreed with the findings of many works in the literature [35–37]. For instance, the corresponding decomposition temperature range of each phase or group can be ascribed as follows: the temperature range until 200 °C was attributed to the dehydration of CSH, C_2SH_8 , ettringite, and AFm; from 200 to 400 °C was attributed to the dehydration of C_2AH_8 and C_2AH_6 ; from 400 to 470 °C was the dehydration of CH; finally, from 550 to 900 °C was the decarbonation of calcite. In addition, according to [33,38] and [39,40], the decomposition temperature of nylon-66-based electrospun nanofibers is from around 400 to 500 °C, and this parameter of TEOS/PVP-based electrospun nanofibers is from around 350 to 470 °C. Therefore, the peaks from around 350 to 500 °C in Figure 7 can be attributed to the decomposition of both CH and the electrospun nanofibers in the cement pastes. It is worth mentioning that the weight loss before 145 °C was due to the evaporation of the arbitrary water that was included in each paste [41]; therefore, the comparison of TGA results was made with 0% weight loss at 145 °C, as shown in Figure 8. In this manner, the content of hydration products observed from the cement paste containing CNTs-TEOS/PVP NFs was highest, followed by that of the paste containing TEOS/PVP NFs, then the pastes containing CNTs-N66 NFs and N66 NFs, compared with the hydration product contents of the plain paste. From the inset plot in Figure 8, at 200 °C, there were different weight losses of the TGA results from the pastes containing TEOS/PVP-based nanofibers compared with that from others, and the lowest belonged to the TGA result of the plain paste. Therefore, the contents of the CSH phase in the pastes containing TEOS/PVP-based nanofibers were higher than those in the others pastes. In addition, according to [32], the CSH phase makes up around 60% of the solid volume of the hydrated products and plays a major role in forming the strength and durability of cementitious materials. Consequently, the TGA results explain the order of the increase of compressive strength in this study.

Figure 7. Derivative thermogravimetric (DTG) curves of the hydrated cement pastes.

Figure 8. TGA curves of the hydrated cement pastes.

4. Conclusions

In this study, the effectiveness of electrospun nanofibers at enhancing the mechanical as well as microstructural properties of Portland cement was estimated. The mechanical properties increased significantly depending on different types of electrospun nanofibers. For instance, the increase in compressive strength of the cement paste was observed when blending TEOS/PVP nanofibers as well as the hybrid nanofibers containing carbon nanotubes and TEOS/PVP with OPC. In addition, an increase in tensile strength was observed when incorporating nylon 66 nanofibers as well as the hybrid nanofibers containing carbon nanotubes and nylon 66 with Portland cement. Further, the toughness property also showed a significant increase when blending the electrospun nanofibers with cementitious materials. In addition, based on the observations from SEM, TEM images, and TGA–DTG results, the bridging effect as well as the filling effect were found. Therefore, the enhancing mechanism of the mechanical properties of the blended cementitious materials has been confirmed.

Above all, this work is an initial step toward a new approach to strengthening cementitious materials. More high-strength electrospun nanofibers should be involved to estimate their effect on cement paste. For practical applications, further work should consider the setting time, the workability, as well as the adhesion with aggregate of blended cement.

Author Contributions: Conceptualization, J.J.K.; Data curation, T.N.M.N. and J.J.K.; Formal analysis, T.N.M.N., D.H.L. and J.J.K.; Funding acquisition, J.J.K.; Investigation, T.N.M.N. and J.J.K.; Methodology, D.H.L. and J.J.K.; Project administration, J.J.K.; Supervision, D.H.L. and J.J.K.; Validation, D.H.L. and J.J.K.; Visualization, T.N.M.N.; Writing—original draft, T.N.M.N. and J.J.K.; Writing—review & editing, T.N.M.N., D.H.L. and J.J.K. All authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by National Research Foundation of Korea: 2020R1A2B5B01001821

Conflicts of Interest: The authors declare no conflict of interest

References

1. Xue, J.; Xie, J.; Liu, W.; Xia, Y. Electrospun Nanofibers: New Concepts, Materials, and Applications. *Acc. Chem. Res.* **2017**, *50*, 1976–1987. [\[CrossRef\]](#) [\[PubMed\]](#)
2. Thenmozhi, S.; Dharmaraj, N.; Kadirvelu, K.; Kim, H.Y. Electrospun nanofibers: New generation materials for advanced applications. *Mater. Sci. Eng. B* **2017**, *217*, 36–48. [\[CrossRef\]](#)
3. Arinstein, A. *Electrospun Polymer Nanofibers*; Pan Stanford Publishing Pte. Ltd.: Temasek Boulevard, Singapore, 2018.
4. Zhu, L.; Wang, Z.; Shu, Q.; Takala, J.; Hiltunen, E.; Feng, P.; Yuan, Z. Nutrient removal and biodiesel production by integration of freshwater algae cultivation with piggery wastewater treatment. *Water Res.* **2013**, *47*, 4294–4302. [\[CrossRef\]](#) [\[PubMed\]](#)
5. Gimenez, R.; Berli, C.L.A.; Bellino, M.G. Oil Recovery from Nanoporous Media via Water Condensation. *Adv. Mater. Interfaces* **2019**, 1900250. [\[CrossRef\]](#)
6. Ma, W.; Ding, Y.; Zhang, M.; Gao, S.; Li, Y.; Huang, C.; Fu, G. Nature-inspired chemistry toward hierarchical superhydrophobic, antibacterial and biocompatible nanofibrous membranes for effective UV-shielding, self-cleaning and oil-water separation. *J. Hazard. Mater.* **2020**, *384*, 121476. [\[CrossRef\]](#) [\[PubMed\]](#)
7. Erdem, E.; Karapinar, N.; Donat, R. The removal of heavy metal cations by natural zeolites. *J. Colloid Interface Sci.* **2004**, *280*, 309–314. [\[CrossRef\]](#)
8. Garrett, R.; Niiyama, E.; Kotsuchibashi, Y.; Uto, K.; Ebara, M. Biodegradable nanofiber for delivery of immunomodulating agent in the treatment of basal cell carcinoma. *Fibers* **2015**, *3*, 478–490. [\[CrossRef\]](#)
9. Shin, S.-H.; Purevdorj, O.; Castano, O.; Planell, J.A.; Kim, H.-W. A Short Review: Recent Advances in Electrospinning for Bone Tissue Regeneration. *J. Tissue Eng.* **2012**, *3*. [\[CrossRef\]](#)
10. Wang, X.; Lv, F.; Li, T.; Han, Y.; Yi, Z.; Liu, M.; Chang, J.; Wu, C. Electrospun Micropatterned Nanocomposites Incorporated With Cu₂S Nanoflowers for Skin Tumor Therapy and Wound Healing. *ASC Nano* **2017**, *11*, 11337–11349. [\[CrossRef\]](#)
11. Lu, Y.; Xiao, X.; Fu, J.; Huan, C.; Qi, S.; Zhan, Y.; Zhu, Y.; Xu, G. Novel smart textile with phase change materials encapsulated core-sheath structure fabricated by coaxial electrospinning. *Chem. Eng. J.* **2019**, *355*, 532–539. [\[CrossRef\]](#)
12. Mirjalili, M.; Zohoori, S. Review for application of electrospinning and electrospun nanofibers technology in textile industry. *J. Nanostructure Chem.* **2016**, *6*, 207–213. [\[CrossRef\]](#)
13. Baji, A.; Agarwal, K.; Oopath, S.V. Emerging Developments in the Use of Electrospun Fibers and Membranes for Protective Clothing Applications. *Polymers* **2020**, *12*, 492. [\[CrossRef\]](#) [\[PubMed\]](#)
14. Li, Y.; Li, Q.; Tan, Z. A review of electrospun nanofiber-based separators for rechargeable lithium-ion batteries. *J. Power Sources* **2019**, 443. [\[CrossRef\]](#)
15. Liang, Z.; Zhao, Y.; Li, Y. Electrospun Core-Shell Nanofiber as Separator for Lithium-Ion Batteries with High Performance and Improved Safety. *Energies* **2019**, *12*. [\[CrossRef\]](#)
16. Santangelo, S. Electrospun Nanomaterials for Energy Applications: Recent Advances. *Appl. Sci.* **2019**, *9*. [\[CrossRef\]](#)
17. Azevedo, N.H.d.; Gleize, P.J.P. Effect of silicon carbide nanowhiskers on hydration and mechanical properties of a Portland cement paste. *Constr. Build. Mater.* **2018**, *169*, 388–395. [\[CrossRef\]](#)
18. Barbhuiya, S.; Chow, P. Nanoscaled Mechanical Properties of Cement Composites Reinforced with Carbon Nanofibers. *Materials* **2017**, *10*, 662. [\[CrossRef\]](#)
19. Li, G.Y.; Wang, P.M.; Zhao, X. Mechanical behavior and microstructure of cement composites incorporating surface-treated multi-walled carbon nanotubes. *Carbon* **2005**, *43*, 1239–1245. [\[CrossRef\]](#)
20. Rocha, V.V.; Ludvig, P.; Trindade, A.C.C.; Silva, F.d. The influence of carbon nanotubes on the fracture energy, flexural and tensile behavior of cement based composites. *Constr. Build. Mater.* **2019**, *209*, 1–8. [\[CrossRef\]](#)
21. Nguyen, T.N.M.; Yoo, D.; Kim, J.J. Cementitious Material Reinforced by Carbon Nanotube-Nylon 66 Hybrid Nanofibers: Mechanical strength and microstructure analysis. *Mater. Today Commun.* **2020**, *23*. [\[CrossRef\]](#)
22. Nguyen, T.N.M.; Moon, J.; Kim, J.J. Microstructure and mechanical properties of hardened cement paste including Nylon 66 nanofibers. *Constr. Build. Mater.* **2020**, *232*, 117–132. [\[CrossRef\]](#)
23. Kim, J.J.; Nguyen, T.N.M.; Nguyen, X.T.; Ta, D.H. Reinforcing cementitious material using singlewalled carbon nanotube-nylon 66 nanofibers. *Transp. Commun. Sci. J.* **2020**, *71*, 46–55. [\[CrossRef\]](#)
24. C150-18, A. *Standard Specification for Portland Cement*; ASTM International: West Conshohocken, PA, USA, 2018.

25. Rao, A.M.; Chen, J.; Richter, E.; Schlecht, U.; Eklund, P.C.; Haddon, R.C.; Venkateswaran, U.D.; Kwon, Y.K.; Tománek, D. Effect of van der Waals interactions on the Raman modes in single walled carbon nanotubes. *Phys. Rev. Lett.* **2001**, *86*, 3895–3898. [[CrossRef](#)] [[PubMed](#)]
26. Huang, Y.Y.; Terentjev, E.M. Dispersion of Carbon Nanotubes: Mixing, Sonication, Stabilization, and Composite Properties. *Polymers* **2012**, *4*, 275–295. [[CrossRef](#)]
27. Yang, K.; Yi, Z.L.; Jing, Q.F.; Yue, R.L.; Jiang, W.; Lin, D.H. Sonication-assisted dispersion of carbon nanotubes in aqueous solutions of the anionic surfactant SDBS: The role of sonication energy. *Chin. Sci. Bull.* **2013**, *58*, 2082–2090. [[CrossRef](#)]
28. Sabet, S.M.; Mahfuz, H.; Hashemi, J.; Nezakat, M.; Szpunar, J.A. Effects of sonication energy on the dispersion of carbon nanotubes in a vinyl ester matrix and associated thermo-mechanical properties. *J. Mater. Sci.* **2015**, *50*, 4729–4740. [[CrossRef](#)]
29. C307-03(2012), A. *Standard Test. Method for Tensile Strength of Chemical-Resistant Mortar, Grouts, and Monolithic Surfacing*; ASTM International: West Conshohocken, PA, USA, 2012.
30. C109/C109M-16a, A. *Standard Test. Method for Compressive Strength of Hydraulic Cement Mortars (Using 2-in. or [50-mm] Cube Specimens)*; ASTM International: West Conshohocken, PA, USA, 2016.
31. Timoshenko, G. *Mechanics of Materials*; PWS-Kent Publishing: Boston, MA, USA, 1984.
32. Mehta, P.K.; Monteiro, P.J.M. *Concrete: Microstructure, Properties, and Materials*; The McGraw-Hill Companies, Inc: New York City, NY, USA, 2006.
33. Baji, A.; Mai, Y.W.; Wong, S.C.; Abtahi, M.; Du, X. Mechanical behavior of self-assembled carbon nanotube reinforced nylon 6,6 fibers. *Compos. Sci. Technol.* **2010**, *70*, 1401–1409. [[CrossRef](#)]
34. Patel, A.C.; Li, S.; Wang, C.; Zhang, W.; Wei, Y. Electrospinning of Porous Silica Nanofibers Containing Silver Nanoparticles for Catalytic Applications. *Chem. Mater.* **2007**, *19*, 1231–1238. [[CrossRef](#)]
35. Roychand, R.; Silva, S.D.; Law, D.; Setunge, S. High volume fly ash cement composite modified with nano silica, hydrated lime and set accelerator. *Mater. Struct.* **2016**, *49*, 1997–2008. [[CrossRef](#)]
36. Kim, J.J.; Foley, E.M.; Taha, M.M.R. Nano-mechanical characterization of synthetic calcium–silicate–hydrate (C–S–H) with varying CaO/SiO₂ mixture ratios. *Cement Concrete Compos.* **2013**, *36*, 65–70. [[CrossRef](#)]
37. Foley, E.M.; Kim, J.J.; Taha, M.M.R. Synthesis and nano-mechanical characterization of calcium-silicate-hydrate (C–S–H) made with 1.5 CaO/SiO₂ mixture. *Cem. Concr. Res.* **2012**, *42*, 1225–1232. [[CrossRef](#)]
38. Saeed, K.; Park, S.Y.; Haider, S.; Baek, J.B. In situ Polymerization of Multi-Walled Carbon Nanotube/Nylon-6 Nanocomposites and Their Electrospun Nanofibers. *Nanoscale Res. Lett.* **2009**, *4*, 39–46. [[CrossRef](#)] [[PubMed](#)]
39. Shahhosseini, M.; Bazgir, S.; Joupari, M.D. Fabrication and investigation of silica nanofibers via electrospinning. *Mater. Sci. Eng. C* **2017**, *91*, 502–511. [[CrossRef](#)] [[PubMed](#)]
40. Newsome, T.E.; Olesik, S.V. Electrospinning Silica/Polyvinylpyrrolidone Composite Nanofibers. *J. Appl. Polym. Sci.* **2014**, 40966–40975. [[CrossRef](#)]
41. Taylor, H.F.W. *Cement Chemistry*; Thomas Telford: New York, NY, USA, 1997.

Publisher’s Note: MDPI stays neutral with regard to jurisdictional claims in published maps and institutional affiliations.

© 2020 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (<http://creativecommons.org/licenses/by/4.0/>).