



Original Research**Morphology Controlled Fabrication of Nickel Encapsulated MWCNTs on Carbon Fibers using Electroless Plating and Ammonia Assisted CVD**

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Abstract

The direct growth of carbon nanotubes on carbon fibers provides a promising route for constructing hierarchical carbon structures with multifunctional applications. In this study, nickel-associated MWCNTs were fabricated directly on carbon-fiber substrates via a sequential process of electroless nickel deposition followed by ammonia-assisted thermal CVD. The effect of nickel deposition level, CVD temperature, gas composition, and growth duration have been investigated for the resulting nanotube morphology. Electroless deposition provided nickel catalyst sites on the carbon-fiber surface, while subsequent CVD promoted MWCNT growth from the deposited catalyst. The use of NH₃ during CVD was associated with reduced visible amorphous-carbon deposition and improved nanotube orientation. FESEM observations revealed substantial changes in catalyst coverage and CNT density with increasing nickel deposition. Higher nickel loading and longer growth durations produced denser and more continuous CNT architectures. TEM observations revealed multi-walled nanotubes with high-contrast catalyst-associated particles at or near nanotube termini, consistent with a catalyst-assisted tip growth configuration. The electroless-plating/CVD strategy provides a relatively simple route for controlling the hierarchical morphology of MWCNT/carbon-fiber structures. The fabricated architecture provides a promising platform for subsequent development of electrically conductive, electromagnetic, and structural composite materials; however, quantitative application-specific properties such as electrical conductivity, electromagnetic-interference shielding effectiveness, and mechanical reinforcement require direct experimental evaluation.

Keywords— Carbon fiber; Multi-walled carbon nanotubes; Electroless nickel plating; Chemical vapor deposition; Ammonia assisted CVD; Nickel catalyst; Tip growth; Hierarchical carbon structures

Introduction

Carbon nanotubes (CNT) have attracted considerable interest due to their combination of high aspect ratio, mechanical strength, electrical conductivity, and thermal stability, plus a large specific surface area. These characteristics have encouraged their integration with carbon-fiber substrates to construct hierarchical structures in which nanoscale carbon structures are directly connected to a macroscale reinforcing framework. Such structures are attractive for multifunctional composite systems since the nanotube layer can modify the surface structure and interfacial properties of carbon fibers, while retaining the intrinsic advantages of the fiber substrate [1-4].

Direct growth of CNT on carbon fibers is particularly attractive compared to subsequent CNT coating or physical blending since the nanotubes can be formed directly at the fiber surface. Previous studies have demonstrated that CVD-grown CNT layers on carbon fibers can modify interfacial adhesion and, depending on the growth conditions, improve mechanical and electrical characteristics of the resulting composites [2-4]. For example, direct CNT growth on carbon-fiber yarns has been reported to enhance the interfacial adhesion between carbon fibers and epoxy matrices, while systematic studies have shown that CVD temperature and catalyst/gas conditions strongly influence both CNT formation and the integrity of the carbon-fiber substrate [2,3]. More recently, CNT-coated carbon-fiber architectures have also been investigated for electromagnetic-wave absorption and shielding applications [4].

Among available CNT synthesis routes, catalytic chemical vapor deposition (CVD) is particularly attractive since catalyst composition, loading, temperature, carbon-source concentration, gas composition, and growth time can be independently adjusted. Transition metals such as Fe, Co, and Ni are widely-used catalysts since they are able to facilitate hydrocarbon decomposition and carbon precipitation. Nickel is especially relevant to this study since it can catalyze CNT growth and can be deposited on carbon-fiber substrates using relatively simple electroless-plating procedures [5-8]. The preparation and distribution of the catalyst on carbon fibers are critical factors controlling CNT nucleation. Electroless nickel deposition provides a solution-based approach for introducing Ni onto nonmetallic substrates without the requirement for the substrate to function as an electrode. Recent studies have demonstrated that electroless nickel plating can produce relatively uniform Ni coatings on carbon fibers and modify their surface properties, mechanical behavior, wettability, and subsequent processing characteristics [5-7]. In particular, electroless Ni plating has been successfully applied to woven carbon fibers, where bath chemistry and processing conditions strongly influenced coating uniformity and interfacial characteristics [6]. These observations indicate that catalyst deposition can be used as an important process variable for controlling subsequent nanocarbon growth.

The present study also incorporates ammonia into the CVD atmosphere. The role of NH₃ in CNT growth has been investigated since ammonia can modify catalyst surface chemistry and influence carbon deposition and nanotube nucleation. Molecular-level analysis has shown that ammonia can participate in carbon removal and affect nucleation and growth kinetics,

providing a mechanistic basis for its use as a growth-control component [8]. Thus, the combination of controlled Ni deposition and NH₃-assisted CVD provides an opportunity to investigate the relationship between catalyst coverage and nanotube morphology.

The novelty of the present work does not arise from the use of electroless nickel plating or CVD, both of which are established techniques. Rather, the contribution of this study lies in their sequential integration to establish a practical process-structure relationship between electroless Ni catalyst deposition, ammonia-assisted CVD conditions, and MWCNT morphology directly on carbon fibers. In the present experimental design, the nickel-bath concentration was varied prior to CVD, while the CVD temperature, gas composition, and growth duration were systematically controlled. This approach enables the influence of catalyst coverage on CNT density, orientation, and surface architecture to be examined within a single fabrication route. Previous studies have established several routes for growing CNT on carbon fibers. Nickel nanoparticles deposited by sputtering, surface oxidation followed by catalyst deposition, and other catalyst-coating procedures have been successfully employed [2,3]. While these approaches can provide high-quality CNT growth, some require vacuum deposition, additional surface treatment, or more complex catalyst preparation. Electroless nickel deposition provides a comparatively simple wet-chemical alternative for introducing Ni catalyst onto carbon-fiber surfaces. Recent work has demonstrated the feasibility of electroless Ni deposition on carbon fibers and its influence on fiber properties [5-7].

The resulting carbon-fiber/MWCNT architecture is also relevant to several emerging applications. CNT-grown carbon fibers have been investigated for improved interfacial adhesion, electrical transport, thermal management, microwave absorption, and electromagnetic-interference shielding [3,4]. For example, hierarchical CNT-coated carbon fibers demonstrated strong microwave absorption when incorporated into polymer matrices, while CNT-modified carbon-fiber/epoxy systems have exhibited improvements in mechanical and electrical properties [4,10]. However, such application-specific performance cannot be assumed solely from morphology. Direct measurements are required to establish electrical conductivity, EMI shielding effectiveness, mechanical reinforcement, or thermal conductivity.

Therefore, the objective of this study was to develop a relatively simple electroless-plating/CVD route for fabricating Ni-catalyzed MWCNT structures directly on carbon fibers and to investigate how nickel deposition and ammonia-assisted CVD conditions influence the resulting morphology. Particular attention was given to catalyst distribution, CNT density, nanotube orientation, and catalyst-associated particles observed by electron microscopy. The study thus provides a framework for subsequent development of carbon-fiber-supported CNT materials for multifunctional composite applications.

Materials and Methods

Materials

Commercial polyacrylonitrile (PAN) based continuous carbon fiber fabrics (Toray TGP-H-030, 30 cm × 30 cm) were obtained from Nano Getters Co. (Korea) and used as substrates for

catalyst deposition and CNT synthesis. Electroless plating solutions were prepared using analytical grade reagents nickel sulfate hexahydrate ($\text{NiSO}_4 \cdot 6\text{H}_2\text{O}$) as the Ni source, trisodium citrate dihydrate ($\text{Na}_3\text{C}_6\text{H}_5\text{O}_7 \cdot 2\text{H}_2\text{O}$) as the chelating agent, sodium hypophosphite monohydrate ($\text{NaH}_2\text{PO}_2 \cdot \text{H}_2\text{O}$) as the reducing agent, and NH_4Cl to stabilize the bath. Aqueous ammonia (25%) was added to adjust and maintain the bath pH near 9.0. Surface activation was performed using aqueous solutions of SnCl_2 , 10 g/L and PdCl_2 , 1 g/L. Process gases used during CVD included high purity H_2 , C_2H_2 , N_2 , NH_3 , and Ar. All materials were used as received without further purification.

Surface Pre treatment of Carbon Fibers

To remove surface impurities and improve catalyst adhesion, carbon fiber fabrics were immersed in acetone at room temperature for 60 minutes. Fibers were then rinsed with deionized (DI) water and dried in a convection oven at 80 °C for 1 hour. Catalytic activation was performed by sequential immersion in SnCl_2 (10 g/L) and PdCl_2 (1 g/L) aqueous solutions for 5 minutes each. The Sn^{2+} ions sensitized the surface, facilitating subsequent adsorption of Pd^{2+} , which served as nucleation centers for nickel deposition.

Electroless Nickel Plating

Electroless plating was carried out to deposit nickel nanoparticles uniformly onto the activated carbon fibers. The bath composition is listed in Table 1. The solution was preheated and maintained at 60 °C with the pH adjusted to ~9.0 using ammonia. Fibers were immersed for 5 seconds per sample, enabling controlled nanoparticle deposition. Nickel concentrations were varied at 0.5%, 1%, 30%, and 100% relative to the stock bath. After plating, the samples were rinsed with DI water and dried at room temperature. Variation of the plating bath concentration was used as a process variable to modify the surface coverage and distribution of Ni catalyst prior to subsequent CNT growth.

Table 1. Composition of electroless nickel plating and activation solutions.

Component	Concentration
$\text{NiSO}_4 \cdot 6\text{H}_2\text{O}$	25 g/L
$\text{Na}_3\text{C}_6\text{H}_5\text{O}_7 \cdot 2\text{H}_2\text{O}$	50 g/L
$\text{NaH}_2\text{PO}_2 \cdot \text{H}_2\text{O}$	25 g/L
Ammonia Solution (25%)	pH ~9.0
SnCl_2 (Activation)	10 g/L
PdCl_2 (Activation)	1 g/L

Carbon Nanotube Growth using Thermal CVD

CNTs were synthesized using a horizontal quartz tube furnace (inner diameter: 5 cm; heated length: 80 cm) under atmospheric pressure. Ni coated carbon fiber substrates were placed in the central heating zone.

Annealing: Fibers were heated to 500 °C under a 200 sccm hydrogen flow for 20 minutes to reduce surface oxides and activate the nickel.

Growth: Temperature was ramped to 550–750 °C under H₂ (150 sccm) and Ar (50 sccm). CNTs were grown by introducing acetylene (C₂H₂) at 10 or 20 sccm. Two gas mixtures were tested:

Set A: C₂H₂ (10 sccm), H₂ (150 sccm), Ar (500 sccm), N₂ (50 sccm).

Set B: C₂H₂ (10 sccm), NH₃ (100 sccm) promoting vertical CNT alignment via carbon etching.

Duration: Synthesis was performed for 2, 5, 10, and 20 minutes to assess time dependent growth.

Cooling: Samples were cooled naturally under inert gas (Ar/N₂, 450/50 sccm) to avoid oxidation and preserve CNT structure.

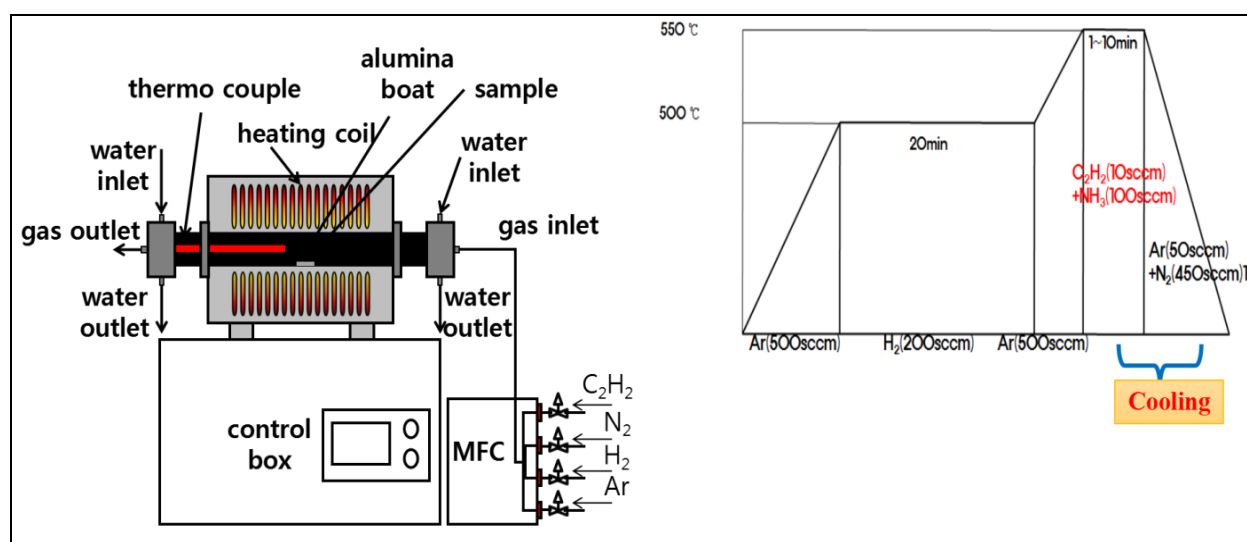


Figure 1. Schematic of the horizontal thermal CVD setup and gas/temperature profile for CNT growth on Ni coated carbon fibers. Growth was performed at 550 - 750° C using a C₂H₂/NH₃ mixture. Gas flow rates and thermal stages are indicated for each step of the process.

Consider Figure 1 that shows the horizontal thermal CVD deposition configuration and a process profile of gases and temperature to grow CNT on nickel coated carbon fibers. Its growth process was done between 550-750°C temperatures in a combination of C₂H₂ and NH₃. The description given to each step in the process includes the flow rates of gases and temperatures. Figure 1 shows a schematic plot of the thermal CVD experimental configuration including the temperature and gas flow profile that was used in the synthesis of the CNT. Horizontal quartz tube furnace was a programmable type of furnace which included individual gas inlet and programable temperature controls. First the materials were annealed in hydrogen at 500° C in 20 minutes. C₂H₂ and NH₃ were injected afterward and used as feed gases at temperatures of 550 to 750° C in a time range of 1 to 10 minutes. The NH₃ addition was essential to obtain a selective etching of the amorphous carbon thus able to preserve the activity of the catalyst during CNT growth. The system was then cooled using Ar/N₂ to safeguard CNT structure and avoid oxidation that a material may have gone through after being grown.

2.5 Characterization

The morphology of the electroless Ni-coated carbon fibers and subsequent growth of CNT structures was investigated using field-emission scanning electron microscopy (FESEM; JEOL JSM-7001F). FESEM was used to investigate catalyst coverage, surface morphology, CNT density, nanotube orientation, and the overall hierarchical architecture. Transmission electron microscopy (TEM; JEOL JEM-2100, operated at 200 kV) was used to investigate the nanotube wall structure, tube diameter, and catalyst-associated particles at or near the nanotube termini. TEM observations were used to evaluate the morphological characteristics of the catalyst-assisted CNT growth process. Since electron-microscopy contrast alone cannot establish the elemental identity of a catalyst particle, elemental analysis by energy-dispersive X-ray spectroscopy (EDS/EDX) should be used as complementary evidence for Ni identification. Where available, X-ray photoelectron spectroscopy (XPS) should additionally be employed to determine the surface elemental composition and chemical states of Ni, C, and O. XPS analysis should include survey, Ni 2p, C 1s, and O 1s spectra. Raman spectroscopy may also be used to evaluate the graphitic structure and defect characteristics of the CNT layer through the D and G bands and the corresponding ID/IG ratio.

Result and Discussions

Catalyst Morphology and Distribution Analyzed by FESEM

Nickel catalyst layers were structured and deposited onto carbon fibers and the carbon fibers were coated via electroless plating, but this changed quite a bit based on the concentrations of the plating bath. Figure 2 demonstrates that micrographs at high magnification FESEM (x100,000) reveal distinguishing changes in the sizes, coverage and span of nanoparticles as the concentration of nickel varies. At low concentration of Ni (0.5%) the carbon fiber substrates exhibited a random and sparse distribution of nanoscale nickel clusters that tended to have diameters of less than 10 nm. The fact that these small particles were found to accumulate at the fibers ridges, defects and micro cracks that are high energy sites, which are preferable for heterogeneous nucleation process, is in agreement with the published works on electroless plating on textured surfaces [1,2,4]. This uneven patchy, low density catalyst pattern caused uneven coverage, which constrained the number of active nucleation sites to CNT growth. Although isolated particles are capable of initiating nanotube growth, a low surface density would allow only difficulty to construct bulk CNT networks and vertical alignment during CVD synthesis [9,14]. The concentration of plating was then increased until 1% when landscape of nanoparticles was highly favorable. Nickel nanoparticle dispersion as shown in Figure 2b was more uniform giving almost a single layer of nanoparticles on the surface of the fiber even where the surface was rough or curved. This improvement follows past related studies that have shown that an optimized chelation and bath stability in electroless plating may increase the distribution and adhesion of nanoparticles [3,5,11]. There were some drastic morphological changes observed at intermediate (30%) and high (100%) nickel concentration levels. Just look at Figure 2c: the 30% Ni sample denotes a composite structure, and different nanoparticles are scattered with emerging metallic patches. This would indicate that localised coalescence occurred in the process of deposition. As we

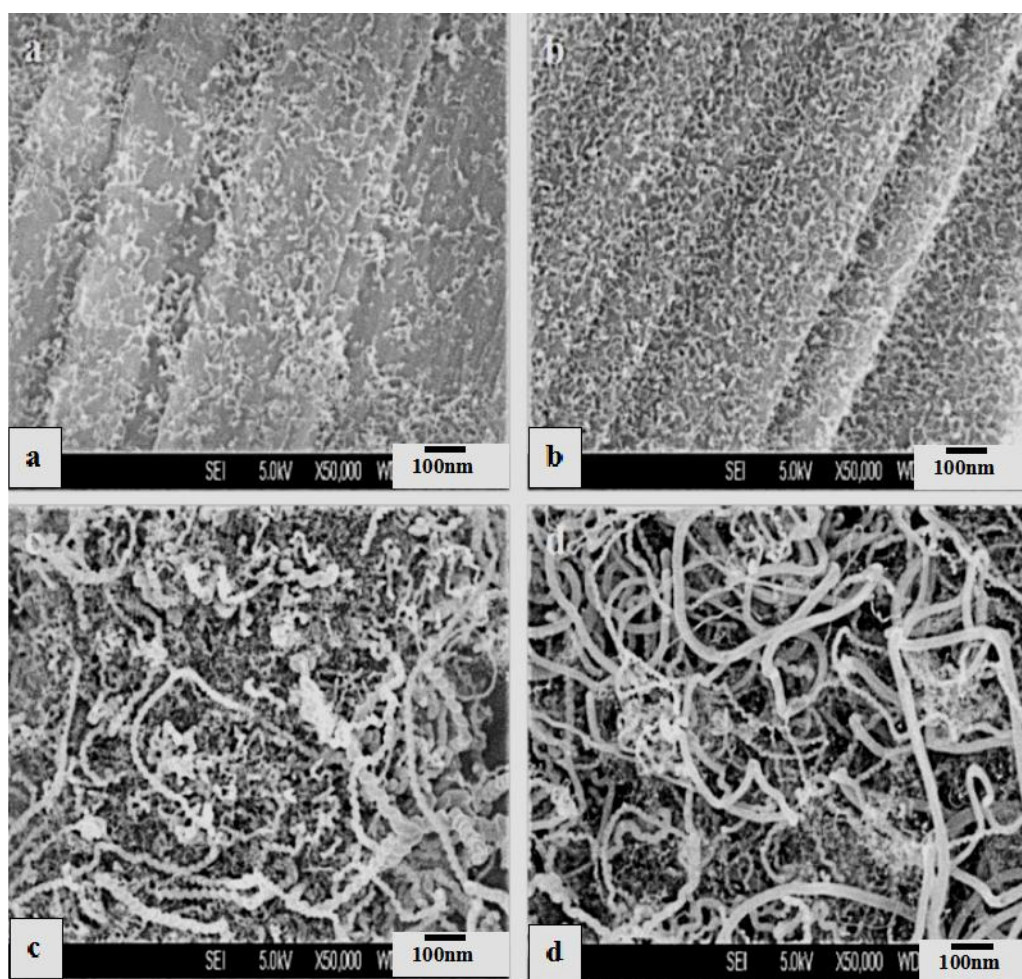


Figure 2. FESEM images of Ni-coated carbon fiber surfaces under different electroless plating concentrations. Ni concentrations: (a) 0.5%, (b) 1%, (c) 30%, (d) 100%.

increased to 100 per cent Ni concentration (Figure 1 d), it resulted in almost homogeneous layer of nickel, which not only covered completely the fiber but even gave the fiber a metallic shell. Although such thick nickel films may well appear to be a means of increasing the catalytic surface area, they do have some drawbacks. To begin with, the clustering of particles restricts the occurrence of the discrete nucleations events and, hence, there are reduced numbers of active growth centers. Moreover, such repeated layers of metals can prevent gaseous precursors access to catalyst substrate interface thus causing difficulty in diffusing carbon uniformly. Besides, excess of nickel may cause a sintering during the application of CVD temperature thereby forming oversized clusters which may destabilize Carbon nanotube (CNT) formation and may induce misalignment of the nanotubes or even just random entangling of nanotubes [3,5,19]. The results actually highlight the importance of catalyst concentration in determining nanoscale morphology which in itself has a direct impact on the growth behavior of CNTs. The observations indicate that intermediate catalyst loading provides a different balance between surface coverage and catalyst particle separation compared with the lowest and highest deposition conditions. The resulting catalyst distribution appears to influence subsequent CNT nucleation and growth, although

quantitative determination of catalyst particle density and size distribution would be required to establish an optimum catalyst loading. This balance promotes a well regulated nucleation, uniform tube orientations and reproducibility in the substrate factors that are required in the production of high-performance, hierarchical CNT carbon fiber composites [4,5,17].

Carbon Nanotube Growth Behavior

We examined the method of controlling the growth of CNTs on carbon fibers with electroless nickel plated in great detail and tested various CVD deposition conditions through thermal CVD properties such as the concentration of catalyst, the type of gasses used, and the duration of the process in detail. As depicted in Figure 3, the C_2H_2/NH_3 gas mixture at 750 °C does affect the vertical alignment, structural uniformity, and purity of the CNTs significantly when we introduced it to our furnace and left it hanging in there, until it settled (20min). These findings were directly related with the initial distribution of the nickel catalyst in the conduct of the electroless deposition process [1,3]. A 30 wt.% loading of nickel catalyst showed that CNTs produced semi-aligned patches with an intermediate packing density. In spite of a certain vertical alignment, we also found an uneven surface coverage, probably because the catalyst was unevenly dispersed. In contrast, the dense CNT forest with vertical alignment could be obtained by using 100 % concentration of nickel solution and thoroughly covered the surface of carbon fiber.

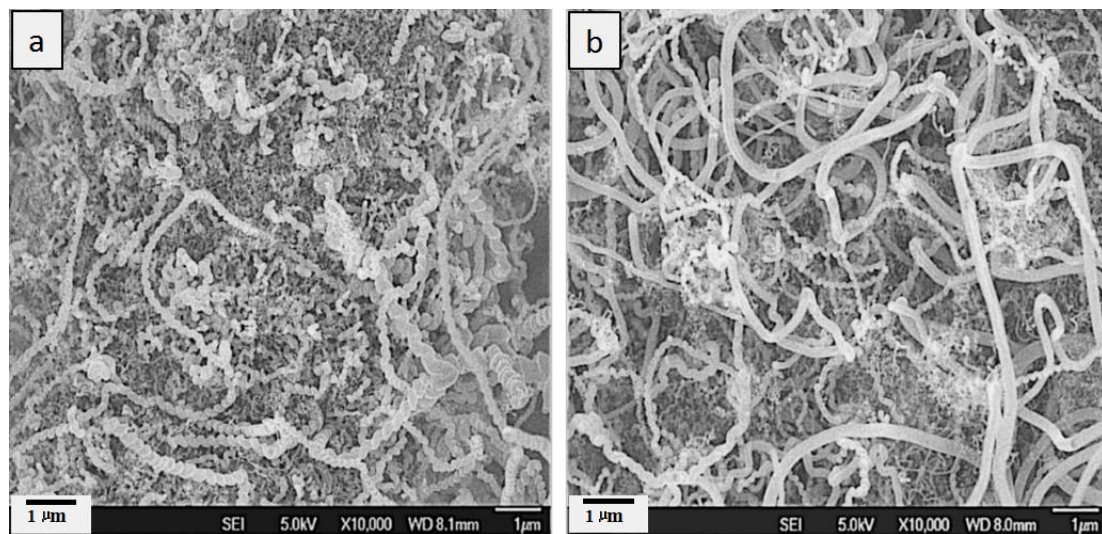


Figure 3. FESEM images of CNTs grown on carbon fibers at 750 °C using a C_2H_2/NH_3 gas mixture: (a) 30% Ni catalyst, (b) 100% Ni catalyst.

It has been concluded that this observation supports other literature findings indicating that elevated catalyst density increases the quantity of active nucleation sites leading to improved alignment, tube density and morphology [2,6,9]. The use of gaseous ammonia on the CVD treatment was important as far as the improvement of the CNT alignment was concerned. It generated atomic hydrogen, and this effectively anneals out amorphous carbon by performing in situ etching.

This is an etching process that will make it difficult to deactivate the catalyst and also assist in the carbon diffusion that is continuous thus helping in a more graphitic strand on the nanotube walls. Similar increase, as yet recorded in many studies, in the CNT structural quality and purity as a result of ammonia [4,10,12] exists. CNTs grown through such critically optimized conditions grew as a conductive, tangled mesh between and upon the carbon fibers, demonstrating vertical orientation and a nanoscale interconnectivity. Such networks have a huge electric conductivity and assist with the detachment of mechanical loads, which makes them wonderful prospects in multifunctional composite use [6,11].

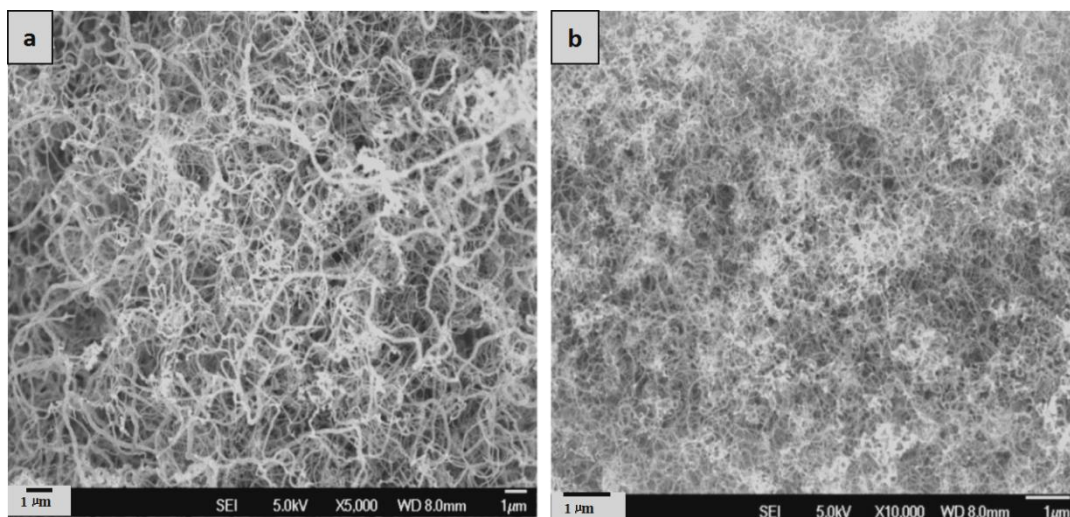


Figure 4. FESEM images of CNTs grown at 750 °C for 20 minutes using a C_2H_2/NH_3 gas mixture. (a) At 30% Ni, CNT growth appears moderate and irregular. (b) At 100% Ni, CNTs show dense and uniform coverage.

Interestingly directional electrical conduction and higher dielectric loss can be used to advantage vertically aligned CNTs with enhanced EMI shielding properties [7,8,14]. The above observation emphasizes the importance of the fact that in order to fully optimize the CNT performance we must have a precise control over not only the morphology of the catalyst but also the dynamics of the CVD gas phase. The use of 100 % Ni, electroless deposition combined with NH_3 -rich gas atmospheres, resulted in very high density, vertically aligned CNT arrays with minimal amorphous carbon. All these features are essential in the development of the future of CNT carbon fiber hybrids with enhanced electrical, thermal and EMI shielding effects [4,6,8]. As shown in Figure 4a-b the FESEM micrographs clearly reveal compact and oriented CNT array on the fiber surface uniformly evenly distributed on them. Enhancement of the nickel loading and length of the synthesis up to 10-20 minutes greatly increased the quality of the CNTs. At larger magnifications, the CNTs were shown to be highly entangled inter-tube and uniformly covered to form a robust and three dimensional network over the carbon surface. The presence of NH_3 during CVD was associated with a cleaner CNTs morphology and reduced visible amorphous-carbon deposition under the investigated conditions. This observation is consistent with previous mechanistic studies showing that ammonia can alter catalyst surface chemistry and carbon removal during CNT nucleation and growth [8]. However, because the present study did not directly monitor the

gas surface reaction pathway, the precise chemical mechanism responsible for the observed morphological change cannot be established solely from the present data.

Comparative Analysis of Ni Catalyst Concentration

In order to comprehensively evaluate the impact of distribution of catalysts to the morphology of CNTs, we made a comparative analysis of the impact by having carbon fiber substrates which had been coated by electroless nickel plating at the concentration of 30 %

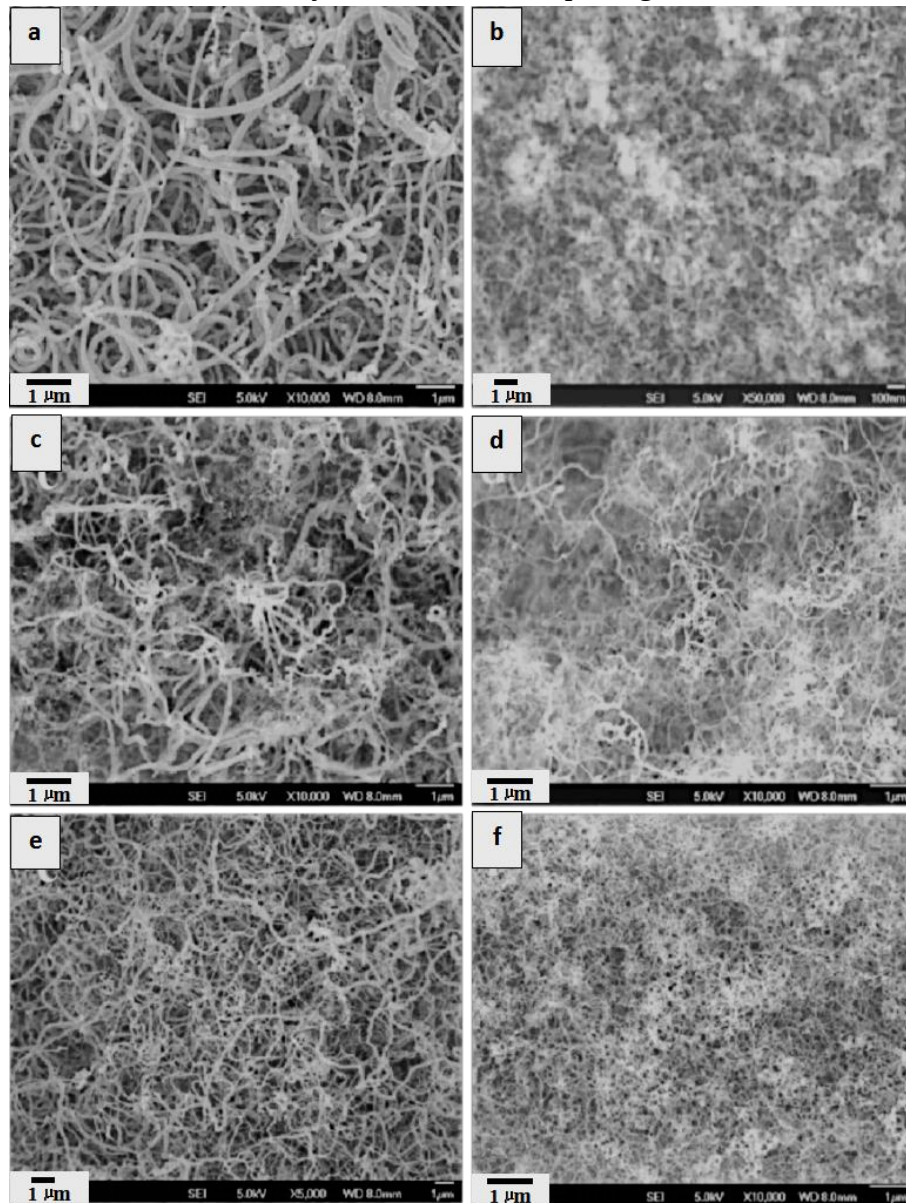


Figure 5. FESEM images illustrating CNT morphology based on nickel concentration and plating time. (a, b) 5 min at 30% and 100% Ni; (c, d) 10 min; (e, f) 20 min.

and 100%. To each concentration, we used deposition of 5, 10, and 20 minutes and the parameters of thermal CVD were kept constant. As demonstrated in Figure 5, it is observable that there is direct correlation between the degree of the catalyst coverage and the density, uniformity and orientation of which the CNTs synthesised are. At lower periods of deposition (5 minutes) both 30% and 100% samples of Ni catalyst exhibited very little growth of CNTs,

which makes them highly sparse and randomly oriented nanotubes on the surfaces of the carbon fibers. Nonetheless, when we increased the plating duration to 10 and 20 minutes, our samples with 100% Ni started showing much denser and directional CNT networks with a much homogeneous coverage of fiber surfaces. Although it was also observed that the 30% Ni sample got better with time, the overall alignment and entanglement of CNT was not as high as that of the more concentrated samples. Because more nickel is now accessible, catalytic sites are more, and this helps decompose hydrocarbons and on-going addition of carbon. This undergoes a continuous vertical growth of CNTs and promotes the formation of interconnected networks which are much necessary when it comes to multifunctional composite applications and that require very high levels of electrical, and thermal properties [5,7,9]. The results indicate clearly that optimized electroless plating is wonderful in the adjustment of catalyst morphology and minimized undesired nanoparticle agglomeration. At large concentrations of the bath, nickel nanoparticles remained in the form of well-dispersed stable catalytic islands which retained their activity after long CVD exposure. Such uniformity enabled to grow MWCNTs reliably in well-aligned with minimal structural defects. The vertically aligned CNTs also enhance the electron conduction capability as well as reinforce the mechanical interlocking between fiber-matrix interface leading to the increased EMI shielding and the load bearing capacity. It is important that electroless plating parameters especially nickel bath concentration and deposition time be accurately controlled to tailor the architecture of the catalyst to allow uniform growth of CNTs. The increase in CNT density with increasing deposition/growth duration suggests that catalyst availability and exposure time jointly influence nanotube nucleation and growth. The 100% Ni condition produced a comparatively dense CNT network at longer growth durations, whereas the 30% Ni condition produced less extensive coverage under otherwise similar CVD conditions. These observations demonstrate that catalyst loading provides a practical parameter for tailoring CNT surface architecture. However, the relationship between CNT morphology and functional properties such as conductivity or thermal transport should be established experimentally rather than inferred solely from the observed morphology.

TEM Observation of Catalyst-Associated Particles

TEM examination revealed multi-walled nanotubes with graphitic walls and high-contrast nanoparticles at or near some nanotube termini. The nanotubes exhibited an internal diameter in the approximate range of 10-20 nm. The location of the high-contrast particles relative to the nanotube termini is morphologically consistent with a catalyst-associated tip-growth configuration. It is important to distinguish morphological observation from elemental identification. The high electron contrast observed in TEM is consistent with the presence of a relatively high-density catalyst particle, but TEM contrast alone cannot unambiguously establish that the particle is nickel. Therefore, elemental analysis by EDS/EDX is required to confirm the presence and spatial distribution of Ni. XPS can provide complementary information concerning the surface chemical state of nickel, including the distinction between metallic and oxidized Ni species.

The observed catalyst-associated particles therefore provide morphological support for a catalyst-assisted tip-growth mechanism, while definitive identification of the particles as Ni should be based on complementary elemental and surface-chemical characterization.

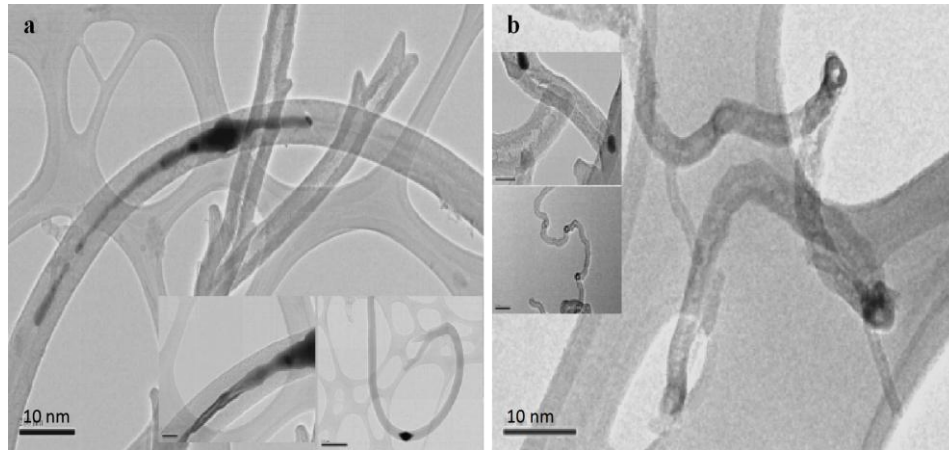


Figure 6. TEM images of CNTs: (a) Ni nanoparticle encapsulation at tube tips, (b) high-resolution lattice fringes confirming semi-graphitized walls.

Proposed Growth Mechanism

Figure 1 represents the diagrammatic view of CNT synthesis mechanism. Ni nanoparticles are important in the thermal CVD process in that they catalyze the decomposition process of carbon and the growth of the tips as demonstrated in Figure 7. Based on the FESEM and TEM observations, a catalyst-assisted tip-growth configuration is proposed for the formation of the MWCNTs.

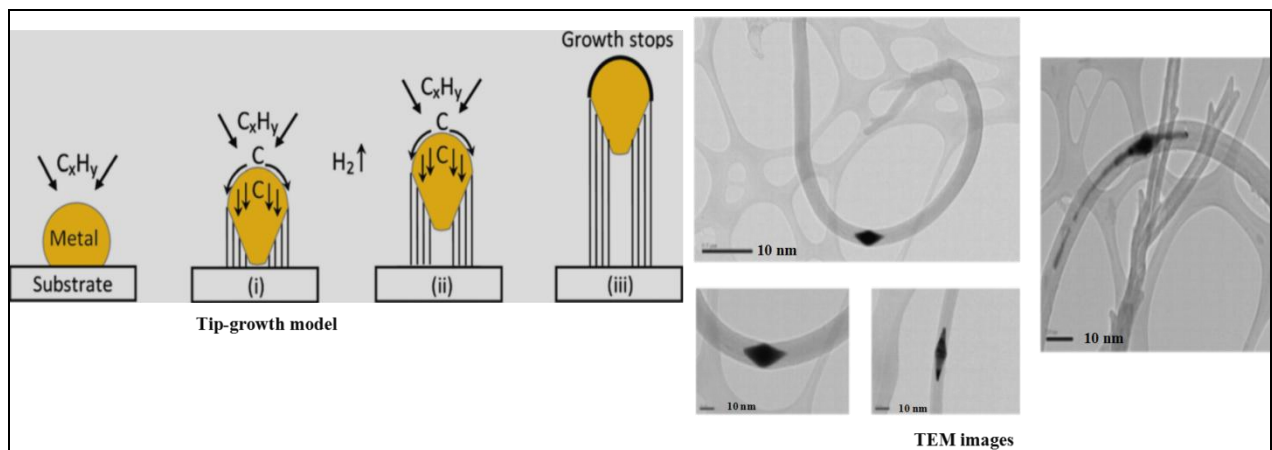


Figure 7. Schematic of the tip growth mechanism showing Ni nanoparticle-assisted CNT nucleation, elongation, and encapsulation during thermal CVD.

The process begins with the formation of Ni-containing catalytic sites on the activated carbon-fiber surface during electroless deposition. During the initial hydrogen treatment, the

deposited catalyst undergoes surface activation and morphological restructuring. Upon introduction of acetylene at elevated temperature, catalytic decomposition of the carbon precursor occurs at the Ni surface. The resulting carbon species undergo surface diffusion and/or dissolution into the catalyst particle, followed by precipitation as graphitic carbon. Continued carbon incorporation leads to the formation and elongation of concentric graphitic layers. When the interaction between the catalyst particle and carbon-fiber substrate is sufficiently weak relative to the forces associated with nanotube growth, the catalyst can remain associated with the growing nanotube terminus, producing a morphology consistent with tip growth. The high-contrast particles observed near nanotube termini in the present TEM images are consistent with this model. However, the proposed mechanism is based on post-growth morphology and should therefore be regarded as a mechanistic interpretation rather than direct experimental observation of the dynamic growth process.

Application Outlook

The hierarchical Ni-MWCNT/carbon-fiber architecture produced in this study provides a potentially useful platform for multifunctional composite development. The direct growth of CNT on carbon fibers creates a nanoscale surface architecture that can potentially modify interfacial interactions, electrical pathways, and electromagnetic response. Previous studies have demonstrated that CNT-coated carbon fibers can be incorporated into polymer composites to improve interfacial and electrical properties [3,10]. Directly grown CNT forests on carbon fibers have also been investigated for electromagnetic-wave absorption and related microwave applications [4,11]. In addition, electroless Ni coatings on carbon fibers have been reported to influence electrical, mechanical, and surface properties [5-7]. The incorporation of Ni into CNT-containing architectures is particularly interesting since Ni can provide additional magnetic functionality. Ni-containing CNT structures have previously been investigated in catalytic and electrochemical applications, demonstrating that encapsulated Ni can remain functionally accessible depending on the architecture and surface structure [12,13]. Nevertheless, the present study was primarily focused on fabrication and morphological control. Quantitative electrical conductivity, magnetic permeability, EMI shielding effectiveness, microwave absorption, mechanical reinforcement, and thermal conductivity were not measured. Therefore, these properties should be considered potential application directions rather than experimentally demonstrated performance in this work. Future work should combine the optimized Ni-MWCNT/carbon-fiber architecture with application-specific measurements. Electrical conductivity and EMI shielding effectiveness would be particularly relevant because they can directly assess the functional consequences of the hierarchical conductive/magnetic architecture. Mechanical testing of carbon-fiber/epoxy laminates containing the CNT-grown fibers would similarly establish whether the nanotube layer improves interfacial load transfer. Zeta-potential analysis was not included because the investigated material consists of a CNT-functionalized continuous carbon-fiber substrate rather than a freely dispersed colloidal nanoparticle system. Conventional zeta-potential measurements require dispersion of the sample in a defined liquid medium, and the resulting value can depend strongly on pH, ionic strength, dispersion conditions, and sample

fragmentation. Zeta-potential analysis may nevertheless become relevant in future studies in which the CNT-containing material is processed as a dispersed filler.

Limitations of the study

This study has several limitations. First, although FESEM and TEM provided insights into MWCNT morphology and catalyst distribution, additional EDS/EDX and XPS analyses are needed to confirm the elemental composition and chemical states of the nickel catalysts. Second, the proposed tip-growth mechanism was inferred from morphological observations and requires further experimental validation. Third, while different deposition and CVD conditions were examined, a more systematic investigation is needed to understand their effects on MWCNT growth. Finally, the electrical, magnetic, mechanical, thermal, and EMI-shielding properties of the resulting structures were not evaluated. Future studies should address these limitations to confirm the growth mechanism and explore the practical applications of MWCNT/carbon-fiber composites.

Conclusion and Future Directions

Future studies should focus on optimizing electroless nickel deposition and ammonia-assisted thermal CVD conditions to achieve uniform catalyst distribution, controlled MWCNT growth, and improved reproducibility. Advanced characterization using EDS/EDX, XPS, high-resolution TEM, and Raman spectroscopy is recommended to better understand catalyst composition, nanotube structure, and growth mechanisms. Further investigations should evaluate the electrical, magnetic, mechanical, thermal, and EMI-shielding properties of MWCNT/carbon-fiber composites to assess their potential applications. Additionally, comparative studies with appropriate control samples and assessments of process stability and scalability are needed to support practical applications.

Authors Contributions

Nilay Kumar Dey contributed to experimental design, methodology, conceptualization, data analysis, and manuscript drafting. Ali Hossain, Md. Gazi Mobassir and Kazi Atahar Hossain conducted characterization and experimental work. Fahim Hossain, Aliya Akter, Md Sameul Islam, Rotan Chandra Roy and Md. Mamun Shekh contributed to data collection, analysis, and validation. Mahmudul Hasan provided supervision, resources, and manuscript review. Md. Shah Alam supervised the research, secured funding, and critically revised the manuscript. All authors reviewed and approved the final manuscript.

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Conflict of interest

The authors declare that they do not have any conflict of interest.

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