

# Electrophoretic Deposition of Carbon Nanotubes onto Carbon Fiber Laminated Composites: Effect of Suspension Medium, Deposition Voltage, and Time

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# Electrophoretic Deposition of Carbon Nanotubes onto Carbon Fiber Laminated Composites: Effect of Suspension Medium, Deposition Voltage, and Time

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### DECLARATION

I declare that the work in this thesis was carried out in accordance with the regulations of Universiti Malaysia Sarawak. Except where due acknowledgements have been made, the work is that of the author alone. The thesis has not been accepted for any degree and is not concurrently submitted in candidature of any other degree.

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### ABSTRACT

Multiwalled carbon nanotubes-Carbon Fiber (MWCNT-CF/) epoxy laminated composites are widely used in many applications, and electrophoretic deposition (EPD) is a hybridisation method that is often used. This research focuses on improving the technique by optimising the input parameters to obtain composites with enhanced tensile properties. Various studies have utilized the EPD method, but there hardly any study utilized the watermethanol mixture as the medium. This study showed that the input parameters (volume ratio of suspension medium, deposition voltage, and time) influenced the responses of the research (tensile strength and Young's modulus). Firstly, the optical observation showed good distribution of MWCNTs throughout the medium. Secondly, the analyses of Fourier Transform Infra-Red (FTIR), Scanning Electron Microscopy (SEM), and tensile properties demonstrated that the input factors directly influenced the composites. Thirdly, the ideal factors that correspond to the desired responses were obtained through the optimization. For the first design of experiment (DoE) (0% water, 100% methanol and 100% water,0% methanol), the optimum conditions were a volume ratio of 99.99% water, a voltage of 20V, and time of 8.88 minutes, producing maximum tensile strength and young's modulus of 7.983 N/mm<sup>2</sup> and 268.558 N/mm<sup>2</sup>, respectively. For the second DoE (20% water,80% methanol and 80% water, 20% methanol), tensile strength and young's modulus of 7.2766 N/mm<sup>2</sup> and 266.78 N/mm<sup>2</sup>, respectively, were achieved when the ideal conditions were: volume ratio of 79.99 % water, voltage of 20V, and time of 5.22 minutes.

Keywords: MWCNTs, CF, epoxy, EPD, tensile properties

## Pemendapan Electroforetik Karbon Nanotiub pada Komposit Berlapis Gentian Karbon: Kesan Media Ampaian, Voltan dan Masa Pemendapan

#### **ABSTRAK**

"Multiwalled "Karbon Nanotiub-Gentian Karbon (MWCNT-CF) epoksi komposit berlamina digunakan dalam banyak aplikasi, dan pemendapan elekrtoforetik (EPD) ialah kaedah hibridisasi yang kerap digunakan. Kajian ini berfokuskan peningkatan teknik dengan mengoptimum parameter input bagi mendapatkan komposit dengan sifat tegangan dipertingkat. Pelbagai kajian telah menggunakan EPD, tetapi hampir tiada kajian menggunakan campuran air-metanol sebagai media. Kajian ini menunjukkan bahawa parameter input (nisbah isipadu medium ampaian, voltan dan masa pemendapan) mempengaruhi respon kajian (kekuatan tegangan dan modulus "Young"). Pertama, pemerhatian optikal menunjukkan pengedaran MWCNTs yang bagus di keseluruhan medium. Kedua, analisis spektroskopi inframerah fourier transformasi (FTIR), pengimbasan mikroskop electron (SEM) dan sifat tegangan menunjukkan bahawa faktor input mempengaruhi komposit secara langsung. Ketiga, faktor ideal yang sepadan dengan respon diingini diperolehi daripada pengoptimuman. Bagi desain eksperimen (DoE) pertama (0% air, 100% methanol dan 100% air, 0% metanol), kondisi optimum ialah nisbah isipadu 99.99% air, voltan 20V, dan masa 8.88 minit, menghasilkan kekuatan tegangan dan modulus "Young" maksima 7.983N/mm<sup>2</sup> dan 268.558N/mm<sup>2</sup>, masing-masing, Bagi DoE kedua (20% air,80% methanol dan 80% air,20% metanol), kekuatan tegangan dan modulus "Young" 7.2766N/mm<sup>2</sup>dan 266.78 N/mm<sup>2</sup>, masing-masing, diperolehi apabila keadaan ideal: nisbah isipadu 79.99% air, voltan 20V, dan masa 5.22 minit.

Kata kunci: MWCNTs, CF, Epoksi, EPD, Sifat Tegangan

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# LIST OF ABBREVIATIONS

2D	Two dimensional
3D	Three dimensional
AC	Alternating Current
ANOVA	Analysis of Variance
AP	Adequate Precision
ASTM	American Society for Testing and Materials
BA	Bioactive Glass
BBD	Box-Behnken Design
CCC	Circumscribed Central Composite Design
CCD	Central Composite Design
CCF	Face-centred Central Composite Design
CCI	Inscribed Central Composite Design
CF	Carbon Fiber
CFRP	Carbon Fiber Reinforced Polymer
СМС	Ceramic Matrix Composite
CNT	Carbon Nanotube
CNT-CF	Carbon Nanotube-Carbon Fiber
CO <sub>2</sub>	Carbon Dioxide
C.V.	Coefficient of Variance
CVD	Chemical Vapor Deposition
DC	Direct Current
DE	Design Expert
DoE	Design of Experiment

DOM	Methodology of Desirability	
E	Young's modulus	
EM	Electrophoretic Mobility	
EPD	Electrophoretic Deposition	
FFD	Full Factorial Design	
FRP	Fiber Reinforced Polymer	
FTIR	Fourier Transform Infrared	
HA	Hydroxyapatite	
IR	Infra-Red	
LED	Light-Emmiting Diode	
LIBs	Lithium Ion Batteries	
LOF	Lack-of-Fit	
МеОН	Methanol	
MFC	Microbial Fuel Cells	
MWCNT	Multiwalled Carbon Nanotube	
MWCNT-CF	Multiwalled Carbon Nanotube-Carbon Fiber	
NHST	Null Hypothesis Significance Testing	
PA	Polyamide	
PANI	Polyaniline	
PE	Polyethylene	
PEDOT	Poly-(3,4-ethylenedioxythiophene)	
PEEK	Polyetheretherketone	
PEI	Polyetherimide	
рН	Potential of Hydrogen	
PMC	Polymer matrix composite	

PP	Polypropylene
PP	Polypyrrole
PRESS	Prediction error sums of squares
PVA	Polyvinyl alcohol
$R^2_{Adj}$	Adjusted R <sup>2</sup>
R <sup>2</sup> <sub>Pred</sub>	Predicted R <sup>2</sup>
RSM	Response Surface Methodology
SEM	Scanning Electron Microscopy
SSA	Specific Surface Area
Std. Dev.	Standard Deviation
SWCNT	Single-walled Carbon Nanotubes
TAI	Turkish Aerospace Industries
TiO <sub>2</sub>	Titanium Dioxide
TS	Tensile Strength
UPR	Unsaturated Polymer Resin
VE	Vinyl Ester
W	Water

### **CHAPTER 1**

### **INTRODUCTION**

### **1.1 Background of study**

Carbon fiber (CF)/epoxy composite, also known as carbon fiber-reinforced polymer composites (CFRPs), has been used extensively in fields such as automotive, aerospace, marine, and energy to replace conventional metal materials. High strength and stiffness combined with moderately low density have increased the demand for CFRPs in the industries (Rodríguez-González & Rubio-González, 2019). The interfacial interactions of the CFs and epoxy matrix greatly influenced the performance of CFRPs, especially their mechanical properties (Park & Park, 2020). However, interaction between the matrix and fiber for CF and resin epoxy is not favored as the CF surface is non-polar while the epoxy resins are polar (Keyte et al., 2019). Additionally, CF surfaces that are chemically inert, hydrophobic, and intrinsically smooth have caused low interfacial bonding strength between the CF and the polymer matrix (Yao et al., 2018). Hence, the modification must be done at the fibre surface to overcome the CF's inertness and obtain strong fiber/matrix interfacial adhesion (Salahuddin et al., 2021).

Studies have shown that the CFRPs' interfacial properties can be improved by introducing carbon nanotubes (CNTs) onto the reinforcing CF (Sheth et al., 2020). Owing to their excellent mechanical properties, the addition of CNT may effectively enhance the CF–matrix interaction (Moaseri et al., 2016). In addition, the CFs' surface roughness is improved with the presence of CNT and subsequently increases the CF–matrix interfacial adhesion (Zakaria et al., 2020a). Not only taking advantage of the excellent properties of

CNTs, but the reinforced FRP material also maintained the superiority of the conventional fiber reinforcements (J. Li et al., 2021).

CNT-CF hybrids can be fabricated via several methods such as spray coating, chemical vapor deposition (CVD), electrophoretic deposition (EPD), dip coating, etc. Then, the nanoparticles are successfully deposited or attached to the CF surface. Among these methods, EPD has several advantages compared to the other techniques, which allowed this method to be used for CF surface modification (Yao et al., 2018). It is essential to use stable suspension media throughout the EPD process to ensure that the particles are dispersed stably (Chavez-Valdez et al., 2013). Water is commonly used in the EPD process for several reasons: cost effective, low requirement of electric field, easy to regulate throughout the process, and environmentally compatible (Ervina et al., 2019). Despite that, water electrolysis may occur when high voltage is applied in the process, which compromises the depositions' quality. Organic solvents such as alcohol are used to overcome the issue. However, a high applied voltage is needed for pure organic solvents. Apart from that, their particle mobility is low due to the little electric charge on the particles. It can be overcome by combining organic solvent and water as the solvent for the EPD process (Ouedraogo & Savadogo, 2013).

**Table 1.1** lists some of the previous works in which EPD was used to deposit materials and their limitation in comparison to this research study. Based on the articles and previous works related to EPD, the number of investigations into the usage of mixture suspension medium, especially those involving the usage of methanol and water for the MWCNT deposition using the EPD method, is still relatively low. Therefore, this huge research gap in working with a methanol-water mixture suspension medium that had not yet been highlighted gives the researcher an opportunity to makes improvements in this area of study. Not only can the issue of the MWCNTs' stability in medium with various ratios be resolved, but the impact of the EPD parameters on the tensile properties of composites can also be resolved. Apart from that, the optimized values for the input parameters could also be obtained. This study investigate the effects of mixture suspension medium for the EPD process in enhancing the tensile properties of the composites.

This study is done in combination with the Response Surface Methodology (RSM) paired with the central composite design (CCD) to help optimize the obtained data. The interaction of the parameters used in the research was observed and optimized using RSM. Combining mathematical and statistical methods, RSM was able to build models by evaluating the effects of multiple independent variables to find the best value for each variable and get good results (Breig & Luti, 2021).

	Author (s)	Brief description of parameter studied in the article	Limitation/Gap compared to this study
1	J.Guo et al. (2012)	CNT/CF hybrid materials prepared using ultrasonically assisted EPD by using deionized water as medium. Parameter of EPD: 20V for 15 minutes	Only single solvent is used (Deionized water) Only single deposition voltage and time
2	Li et al. (2013)	Coating of two different CF with two types of functionalized MWCNTs using aqueous suspension deposition method in deionized water as medium. Parameter of deposition: 20 minutes; with and without additional of surfactant.	Single solvent used (Deionized water) Used different types of deposition (no electric field applied) Only one immersion time

**Table 1.1:** Previous works related to EPD, and their limitation compared to this study

## **Table 1.1**continued

3	Cordero-Arias et al (2013)	Chitosan composite coating using titania (n-TiO <sub>2</sub> ) nanoparticles by EPD using ethanol-water mixture as medium. Parameter of EPD: Voltage (2 to 50V), time (15s to 5 minutes)	Different types of materials of deposition Uses trial-and-error approach for parameter, i.e., ratio of medium, deposition voltage and time (wide range of parameter's value)
4	Moaseri et al. (2016)	Effect of electrostatic repulsion of MWCNT-CF hybrid epoxy composite on the mechanical properties using ethanol as medium Parameter of EPD: 10V for 20 minutes	Using single solvent (Ethanol) Fixed/ single deposition voltage and time Presence of electrostatic during molding
5	C.Xiao et al. (2018)	CF coated with MWCNTs using aqueous suspension deposition method in deionized water as medium. Parameter of deposition: 20 minutes	Single solvent used (Deionized water) Used different types of deposition (no electric field applied) Only one immersion time
6	Ervina et al. (2019)	EPD of MWCNT onto CF using deionized water, and testing of colloidal stability of MWCNT in medium (with and without presence of voltage) Parameter of EPD: Voltage (10 to 60V), time (3 to 30 minutes)	Only used single solvent (deionized water) Wide range of deposition voltage and time. More focus on the colloidal stability of suspension medium

Based on the table, it was proven that there is a huge gap in the study of the use of mixture suspension medium in depositing MWCNTs via the EPD process, providing opportunities for researchers to find out more about this area of study.

#### **1.2 Research Problems**

Firstly, the study involving the EPD process commonly utilized water as a single suspension medium, as shown in **Table 1.1**. However, water electrolysis might occur during the process when the voltage used is too high, which affects the deposition's quality (Ouedraogo & Savadogo, 2013). This will directly impact the properties of the composites produced. Hence, using a mixture of alcohol and water as an EPD medium is encouraged, as alcohol by itself requires a high deposition voltage (Ouedraogo & Savadogo, 2013). As a result, in this study, methanol and water are used as both a single suspension medium and a mixture suspension medium to compare the tensile properties of composites prepared using these mediums.

Secondly, when alcohol is used in the EPD process, ethanol is chosen as the medium. However, the ideal suspension medium had a low viscosity but a high dielectric constant, which is demonstrated by another type of alcohol, namely methanol. In a book chapter by Amrollahi et al., methanol was listed as having better properties as a suspension medium compared to ethanol (Amrollahi et al., 2016). Because of that, methanol was selected as the organic solvent for the study in order to find out its capability as an EPD suspension medium.

Thirdly, most of the previous studies, as shown in **Table 1.1**, are either using trialand-error approaches or using repetitive values for the parameters in the study. The tabulated data is commonly generated based on previous studies. For instance, in the study by Li et al. in 2013, Moaseri et al. in 2016, and Xiao et al. in 2018, all three studies used 20 minutes of deposition time. Meanwhile, a study in 2013 by Cordero-Arias et al. used the trial-and-error method for their selection of parameters values. Hence, for this reason, in the study, RSM is paired with CCD to give more targeted parameters values as well as save time in terms of the experimental runs to be done.