

Theoretical Analysis on the Thermal and Electrical Properties of Fiber Reinforced Laminates Modified with CNTs

Fatih DARICIK^{*1}, Alparslan TOPCU²

¹Alanya Alaaddin Keykubat University, Rafet Kayış Engineering Faculty, Mechanical Engineering Department, Antalya

²Adana Alparslan Türkeş Science and Technology University, Engineering Faculty, Mechanical Engineering Department, Adana

Geliş tarihi: 17.11.2020

Kabul tarihi: 30.12.2020

Abstract

In the present study, the effect of the multi-walled carbon nanotubes (MWCNTs) fillers weight fraction on the mechanical, electrical, and thermal properties of the epoxy was calculated analytically. The results were then compared and it was found out that the MWCNTs has a significant effect on the electrical conductivity of the epoxy. The MWCNT modified epoxy composites were considered as the matrix material to design quasi-isotropic carbon fibre/epoxy composite. The change of the weight fraction of the MWCNTs on the mechanical, electrical, and thermal properties of the carbon fibre/epoxy laminates was also calculated. Finally, the hygrothermal load and the bending load response of the laminated composites were researched. MWCNTs fix the mismatch between the hygrothermal properties of the epoxy matrix and the carbon fiber.

Keywords: Nanoparticle, Modified epoxy, Composite laminate, Hygrothermal response

Karbon Nanotüp ile Modifiye Edilmiş Fiber Takviyeli Laminelerin Isıl ve Elektriksel Özelliklerinin Teorik Analizi

Öz

Bu çalışmada, çoğul duvarlı karbon nanotüplerin (MWCNT) epoksinin mekanik, elektrik ve ısı özellikleri üzerindeki etkisi analitik olarak hesaplanmıştır. MWCNT'nin epoksinin elektriksel iletkenliği üzerinde önemli bir etkiye sahip olduğu bulunmuştur. MWCNT modifiyeli epoksi malzeme ile karbon fiber/epoksi tabakalı kompozit malzemeler tasarlanmıştır. MWCNT'lerin ağırlıkça katkısının karbon fiber/epoksi kompozitlerin mekanik, elektriksel ve termal özellikleri üzerindeki etkisi de hesaplanmıştır. Son olarak, MWCNT takviyeli tabakalı kompozitlerin higrotermal yük ve eğilme yükü altındaki tepkileri araştırılmıştır. MWCNT'lerin, epoksi matris ve karbon fiberin ısı ve neme bağlı özellikleri arasındaki uyumsuzluğu azalttığı sonucuna varılmıştır.

Anahtar Kelimeler: Nanopartikül, Modifiye epoksi, Kompozit lamine, Isı ve neme bağlı tepki

*Sorumlu yazar (Corresponding author): Fatih DARICIK, fatih.daricik@alanya.edu.tr

1. INTRODUCTION

Carbon fibre reinforced composite laminates have increasing importance for engineering applications due to their excellent properties. Owing to high corrosion and chemical resistance makes carbon/epoxy laminates a good alternative for the durability needed applications such as bipolar plates (BPs) that are used in the fuel cells. On the contrary, the poor electrical conductivity of the carbon/polymer laminated composites restricts their use as BP in the fuel cells. To overcome this deficiency, it is a good solution to modify the insulating constituent of the carbon/polymer composites with nano-sized particles [1–8]. It is also well known that the functionalization and the surface modification of nanoparticles improve the dispersion of the particles in the resins by preventing agglomeration [9,10]. The fracture toughness, the mechanical and thermal properties of polymeric materials filled with functionalized particles are better than the neat polymers [9–13].

In the past two decades, especially carbon nanotubes have come to the fore as the filler materials for polymers. The carbon nanotubes (CNTs) which have excellent conductivity, high aspect ratio, and good bonding with the polymers, offer improved electrical, thermal and interfacial properties. The unmodified CNTs can enhance the electrical conductivity of the epoxy resin better than the surface modified CNTs [6]. In fact, the electrical conductivity of the CNT modified polymers is directly depending on the used functional groups, the functionalization method, and the parameters of the nanoparticles [14,15]. Type of the CNT is also another fact that the multi-walled CNT (MWCNT) particles have a higher increasing effect on the electrical conductivity of the epoxy than that of the single-walled CNT (SWCNT) particles [16]. Chen et al. [3] reported that the addition of electrospun carbon nanofibres into the matrix of the carbon fibre/epoxy laminated composite can increase the in-plane and the out-of-plane electrical conductivities of the laminate up to 150% and 20% respectively according to the pristine laminate. Costa et al. [6] found out that both the glass fiber reinforced and the carbon fiber reinforced laminates with CNT modified matrix

have doubled through-thickness electrical conductivity according to the neat laminates and the modified matrix does not affect the in-plane electrical conductivity of the laminates. Moisala et al. [16] presented that a very small amount of MWCNT or SWCNT can bring significant electrical conductivity to the epoxy resin and the thermal conductivity is negatively influenced by SWCNT particles while MWCNT particles increase the thermal conductivity. Jarali et al. [17] offered analytical equations to determine the hygro-thermal-electrical properties of the CNT-modified polymer composites. On the other hand, the ambient temperature and the amount of the existing moisture also affect the electrical, thermal, and interfacial properties of the CNT modified composites [18].

MWCNT particles are preferred as a filler material for composite bipolar plates (BPs) owing to their superior electrical properties in the last decade. BPs are responsible for gas distribution to the flow channels and collecting the current that occurred in the cell. Therefore, BPs should have high electrical conductivity, and mechanical strength as well as corrosion resistance [19]. In this respect, the MWCNT effect was evaluated as BP application for PEM fuel cells in many studies [20-26]. Bairan et al. [20] constituted composite BP using polypropylene (20%), carbon black (25%), graphite (47-52%), and MWCNT (3-8%) materials. The electrical conductivity and flexural strength were increased by the addition of the MWCNTs up to a limit weight ratio of 6%. In the rates above this limit value, there was a decrease in the properties. According to another study of the researchers [23], 158.32 S/cm electrical conductivity and 30 MPa flexural strength were obtained by the addition of 6% MWCNT into the composite BP. Similar results were obtained by other researchers, too [21,22].

Considering the aforesaid effects of the CNTs on mechanical, hygrothermal, and electrical properties of the polymeric materials, it is inevitable to see similar changes on the polymer matrix composites reinforced with continuous fibres. The stress state and the deformation of the CNT modified

laminated composites under the thermal and the moisture loads will certainly differ according to the unmodified composites. However, it is well known that the effect of the mechanical properties of the continuous fibre and the matrix on the mechanical properties of the laminate can be calculated by analytical approaches. In this study, initially, the hygrothermal, electrical, and mechanical properties of the MWCNT modified epoxy was calculated. Then the properties of the layers with the carbon fibre and the modified epoxy layers were analytically determined according to the rule of the mixture and the effect of the MWCNTs was compared. Finally, MWCNT modified quasi-isotropic carbon fibre/epoxy laminates were designed to investigate the response of the material to the hygrothermal and the bending loads which are generally applied on BPs.

2. METHOD

For the calculations, the MWCNT/epoxy composite resin was considered by using the properties of the constituents (Table 1). The addition of the MWCNTs with a high weight ratio promotes the agglomeration of additives in epoxy resin [27]. Thus, the weight ratio of the MWCNTs in the epoxy resin was assumed as differed into a range of 0.1% to 2% by taking previous researches into account [4,28,29]. Modified resins were designated due to the weight ratio of MWCNTs they contain, such as M_05 contains 0.5% MWCNT in a weight ratio, etc. The agglomeration of the MWCNTs was ignored and the mixture of the MWCNTs and the epoxy resin was assumed as homogenous. Effects of the MWCNTs on the mechanical, electrical, and thermal properties on the epoxy resin were calculated by using a series of Equations 1-10 [17].

Table 1. Properties of MWCNT and epoxy resin

Properties	MWCNT	Epoxy resin	Fibre	
			Longitudinal	Transverse
Density (ρ), gr/cm ³	2.10	1.18	1.84	
Elastic modulus (E), GPa	450	2.47	260	0.8
Shear modulus (G), GPa	173.1	0.88	110	
Poisson's ratio (ν)	0.3	0.3	0.2	
Coefficient of thermal expansion (α), C ⁻¹	4.00*10 ⁻⁶	4.50*10 ⁻⁵	-3.80*10 ⁻⁶	2.00*10 ⁻⁵
Coefficient of moisture expansion (β),	0	0.06	0	
Thermal conductivity	-	0.196	-	-
Electrical conductivity (κ), S/m	1.00*10 ⁵	1.67x10 ⁻¹³	106	

$$V=V^{\Omega}+V^e \tag{1}$$

$$1=\frac{V^{\Omega}}{V}+\frac{V^e}{V}=f^{\Omega}+f^e \tag{2}$$

$$E=\frac{3}{8}(E^{\Omega}f^{\Omega}+E^e f^e)+\frac{5}{8}\frac{E^{\Omega}+E^e}{(E^{\Omega}f^e+E^e f^{\Omega})} \tag{3}$$

$$G^m=\frac{E^m}{2(1+\nu^m)} \tag{4}$$

$$\alpha_a=\frac{(E^{\Omega} \alpha^{\Omega} f^{\Omega}+(1-f^{\Omega})E^e \alpha^e)}{E^{\Omega} f^e+E^e f^{\Omega}} \tag{5}$$

$$\alpha=\frac{1}{8}\left[\frac{3(E^{\Omega} \alpha^{\Omega} f^{\Omega}+E^e \alpha^e f^e)}{E^{\Omega} f^e+E^e f^{\Omega}}+5([\alpha^{\Omega}+(\alpha^{\Omega}-\alpha_a)\nu^{\Omega}]f^{\Omega}+[\alpha^e+(\alpha^e-\alpha_a)\nu^e]f^e)\right] \tag{6}$$

$$\beta_{11}=\frac{(E^e f^e \beta^e)}{[E^{\Omega} f^e+E^e f^{\Omega}]} \tag{7}$$

$$\kappa^*=\kappa^{\Omega}\left(10^{0.085}\left\{\log\left(\frac{f^{\Omega}}{d^{\Omega}}\right)-1\right\}\right) \tag{8}$$

$$f^*=\left(\frac{f^{\Omega}}{d^{\Omega}}\right)^{-1.1\pm} \tag{9}$$

$$\kappa^m=\kappa^*(f^{\Omega}-f^*)^t \tag{10}$$

The MWCNT modified resin with its calculated properties was regarded as the matrix for the newly designed carbon fiber reinforced composite lamina. The direction of the yarns of the unidirectional carbon fabric was assigned as the lamina principle axis 1 and the orthogonal of the axis 1 was axis 2. By considering the common knowledge of the volume ratios of the fibre and the matrix constituents ($V_f=0.55$, $V_m=0.45$) in hand layed up laminated composites, the mechanical and the hygrothermal properties of the carbon/fibre composite lamina were predicted. Calculations were performed due to the rule of mixtures (Equations 10-19) [30].

$$E_1 = V^f E_1^f + V^m E^m \quad (11)$$

$$E_2 = \frac{E_2^f E^m}{V^m E_2^f + V^f E^m} \quad (12)$$

$$G_{12} = \frac{G^f G^m}{V^m G^f + V^f G^m} \quad (13)$$

$$\nu_{12} = V^f \nu^f + V^m \nu^m \quad (14)$$

$$\alpha_1 = \frac{V^f E_1^f \alpha^f + V^m E^m \alpha^m}{V^f E_1^f + V^m E^m} \quad (15)$$

$$\alpha_2 = V^f \alpha^f (1 + \nu^f) + V^m \alpha^m (1 + \nu^m) - (V^f \nu^f + V^m \nu^m) \alpha_1 \quad (16)$$

$$\beta_1 = \frac{V^f E_1^f \beta^f + V^m E^m \beta^m}{V^f E_1^f + V^m E^m} \quad (17)$$

$$\beta_2 = V^f \beta^f (1 + \nu^f) + V^m \beta^m (1 + \nu^m) - (V^f \nu^f + V^m \nu^m) \beta_1 \quad (18)$$

$$k^c = k^m \left[\frac{(V^f)^{\frac{2}{3}} + \left(\frac{k^m}{k^f}\right) \left(1 - (V^f)^{\frac{2}{3}}\right)}{(V^f)^{\frac{2}{3}} \cdot V^f + \left(\frac{k^m}{k^f}\right) \left(1 + V^f \cdot (V^f)^{\frac{2}{3}}\right)} \right] \quad (19)$$

The mechanical and the hygrothermal behavior of the quasi-isotropic laminates were calculated according to Classical Laminate Theory. The hygrothermal stress-strain relationship for unidirectional carbon/epoxy layer without mechanical load was given in Equation 20. Thermally and moisture-induced strains can also be determined by using the changes of the temperature and the moisture by Equations 21,22, respectively. ΔT is the temperature change and the ΔM is the absorbed moisture amount per unit weight. After determining the elastic properties of reinforced composite plies which contain MWCNTs in different weight ratios, quasi-isotropic composite laminates were designed with $[-45_2/45_2/90_2/0_2]_s$ oriented layers (Figure 1a). The stacking sequence of the plies was kept constant for each laminate. Reference ambient during designing was assumed with attributes of $T=20^\circ\text{C}$ and $w\%=0.95$ for all laminates. Inevitably, the change of the elastic properties of the layers owing to MWCNTs alters the response of the laminates to external loads. Especially in fuel cell applications, MWCNT modified composite laminates are subjected to the bending load and the interlaminar shear response of the laminate is crucial under the bending load. The effect of the weight ratio of the MWCNTs on the plane stress and the strain of the composites under hygrothermal load was determined (Equation 23).

Therefore, MWCNT modified composite short beams were also examined for stresses caused by the bending load (Figure 1b).

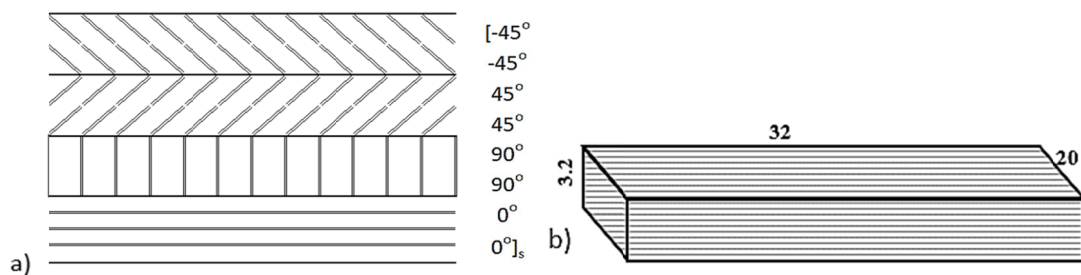


Figure 1. a) Stacking sequence of the laminates and b) the dimensions of the model for bending analysis

$$\begin{bmatrix} \sigma_1 \\ \sigma_2 \\ \tau_{12} \end{bmatrix} = \begin{bmatrix} Q_{11} & Q_{12} & 0 \\ Q_{12} & Q_{22} & Q_{26} \\ 0 & 0 & Q_{66} \end{bmatrix} \begin{bmatrix} \varepsilon_1 - \varepsilon_1^T - \varepsilon_1^M \\ \varepsilon_2 - \varepsilon_2^T - \varepsilon_2^M \\ \gamma_{12} \end{bmatrix} \quad (20)$$

$$\begin{bmatrix} \varepsilon_1^T \\ \varepsilon_2^T \\ 0 \end{bmatrix} = \Delta T \begin{bmatrix} \alpha_1 \\ \alpha_2 \\ 0 \end{bmatrix} \quad (21)$$

$$\begin{bmatrix} \varepsilon_1^M \\ \varepsilon_2^M \\ 0 \end{bmatrix} = \Delta M \begin{bmatrix} \beta_1 \\ \beta_2 \\ 0 \end{bmatrix} \quad (22)$$

$$\begin{bmatrix} \sigma_1 \\ \sigma_2 \\ \tau_{12} \end{bmatrix}_k = \begin{bmatrix} Q_{11} & Q_{12} & Q_{16} \\ Q_{12} & Q_{22} & Q_{26} \\ Q_{16} & Q_{26} & Q_{66} \end{bmatrix}_k \begin{bmatrix} \varepsilon_1^C \\ \varepsilon_2^C \\ \gamma_{12}^C \end{bmatrix}_k \quad (23)$$

3. RESULTS

The determined effect of the MWCNT particles on the mechanical, electrical, and hygrothermal properties of the epoxy was presented in Figures 2, 3, and 4. The effect of the MWCNT particles on the properties of the epoxy is evident. Theoretically, it is possible to increase the modulus of elasticity of the epoxy by 94.49%. Compared to the insulating structure of the epoxy, it is possible to add electrical conductivity to the epoxy by aid of MWCNTs. On the other hand, MWCNTs significantly decrease the hygrothermal expansion coefficients of the epoxy. By adding MWCNT in a weight ratio of %3, the thermal expansion coefficient and moisture expansion coefficient of the epoxy decreased by 29.69% and 181.37%, respectively.

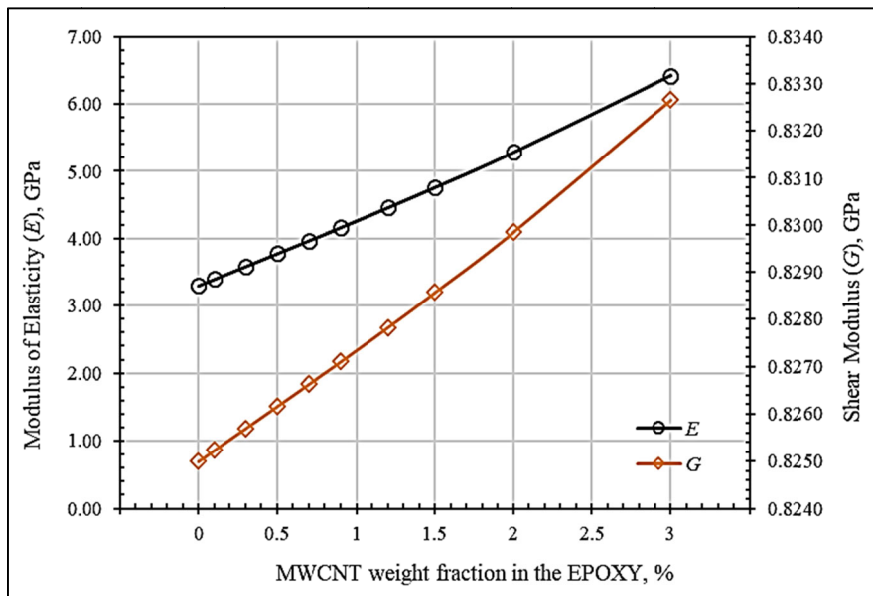


Figure 2. Change of the elastic modulus and shear modulus of the epoxy according to MWCNT ratio

The electrical conductivity of the 3% MWCNT including epoxy was increased by 158.22% by introducing unidirectional carbon fibres into it. Electrical properties are matched with the literature. Bairan et al. reported that the electrical conductivity of the MWCNT reinforced composite BP was increased till 6% filler (158.32 S/cm) [20,23]. In another study, Suherman et al. [25]

fabricated CNT/G/EP (CNT/graphite/epoxy) nanocomposite BP and indicated that the electrical conductivity values were determined as 180 and 75 S/cm for in-plane and through-plane measurements, respectively (with 5% MWCNT). Also, it was noted that the pure composite (without MWCNT) material was unsatisfied in terms of conductivity targets by DOE [19]. As understood,

the MWCNT additive is increased the electrical conductivity, in general. After a certain rate, not only decrease the conductivity but also increase the BP cost because of the high price of the MWCNT.

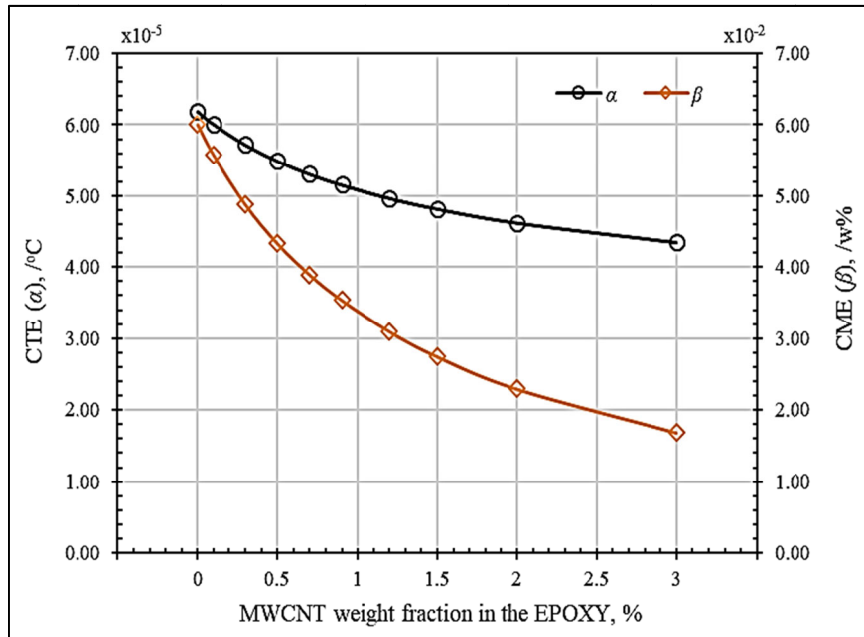


Figure 3. Change of the hydrothermal coefficients of the epoxy according to MWCNT ratio

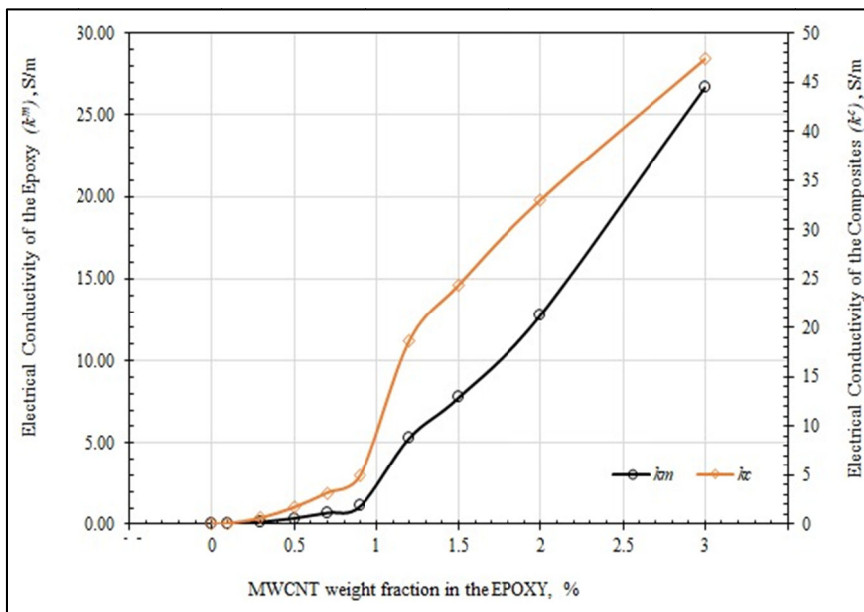


Figure 4. Change of the electrical conductivities of the epoxy and the composite layer according to MWCNT ratio

The mechanical and the hygrothermal properties of the designed unidirectional carbon/epoxy layers with different MWCNTs weight ratios were listed in Table 2 and the changes of the hygrothermal properties were also presented in Figures 4 and 5. Since the in-plane mechanical properties of the unidirectional fibre reinforced layer depend on the fibres, it was concluded that the effect of the MWCNTs on the composite layer is limited. The longitudinal elastic modulus of the layer with 3%

MWCNT increased only by 0.97% and the transverse elastic modulus increased by 8.74%. By introducing the carbon fibres into the 3% MWCNT modified epoxy, the coefficients of longitudinal and transverse thermal expansions were changed in ratios of 4.54% and -80.20%, respectively. On the other hand, the coefficients of longitudinal and transverse moisture expansions decreased in order of 45.80% and 71.98%.

Table 2. Calculated elastic properties of unmodified and MWCNT modified carbon/epoxy layers

Layer Code	Weight Ratio of MWCNTs %	Elastic Modulus, GPa		Shear Modulus (G_{12}), GPa	Thermal Expansion $\times 10^{-6}$		Moisture Expansion $\times 10^{-4}$	
		Longitudinal (E_1)	Transverse (E_2)		Longitudinal (α_1)	Transverse (α_2)	Longitudinal (β_1)	Transverse (β_2)
L_00	0	144.49	1.21	1.82	-3.30	24.75	6.17	349.49
L_01	0.1	144.53	1.22	1.82	-3.32	16.84	5.89	324.51
L_03	0.3	144.61	1.23	1.82	-3.34	14.70	5.44	283.80
L_05	0.5	144.70	1.24	1.82	-3.37	13.02	5.09	252.03
L_07	0.7	144.79	1.25	1.82	-3.38	11.68	4.80	226.56
L_09	0.9	144.87	1.26	1.82	-3.40	10.58	4.57	205.77
L_12	1.2	145.01	1.27	1.82	-3.41	9.23	4.28	180.04
L_15	1.5	145.14	1.28	1.82	-3.42	8.15	4.05	159.66
L_20	2.0	145.38	1.29	1.83	-3.44	6.77	3.75	133.40
L_30	3.0	145.89	1.32	1.83	-3.45	4.90	3.34	97.94

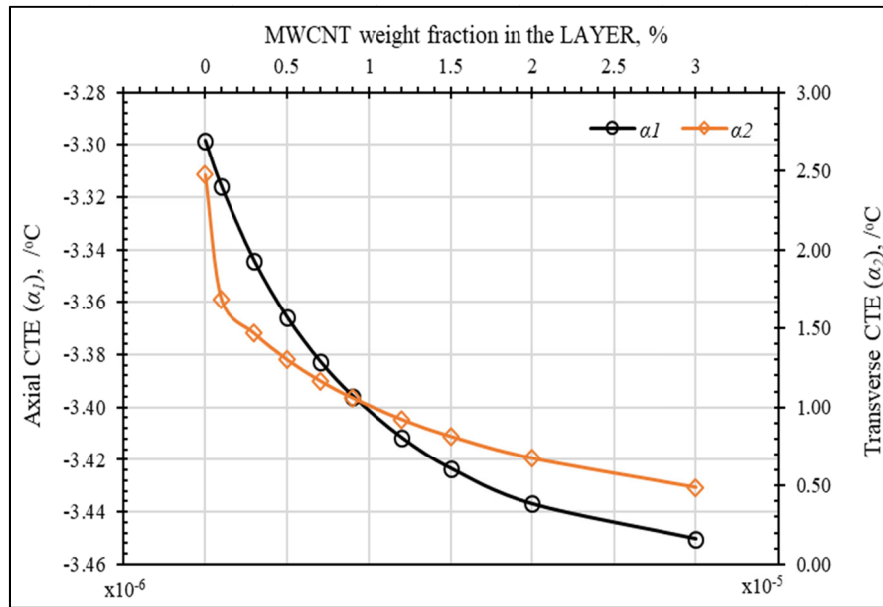


Figure 4. Change of the longitudinal and transverse thermal expansion coefficients of the layers according to MWCNT ratio

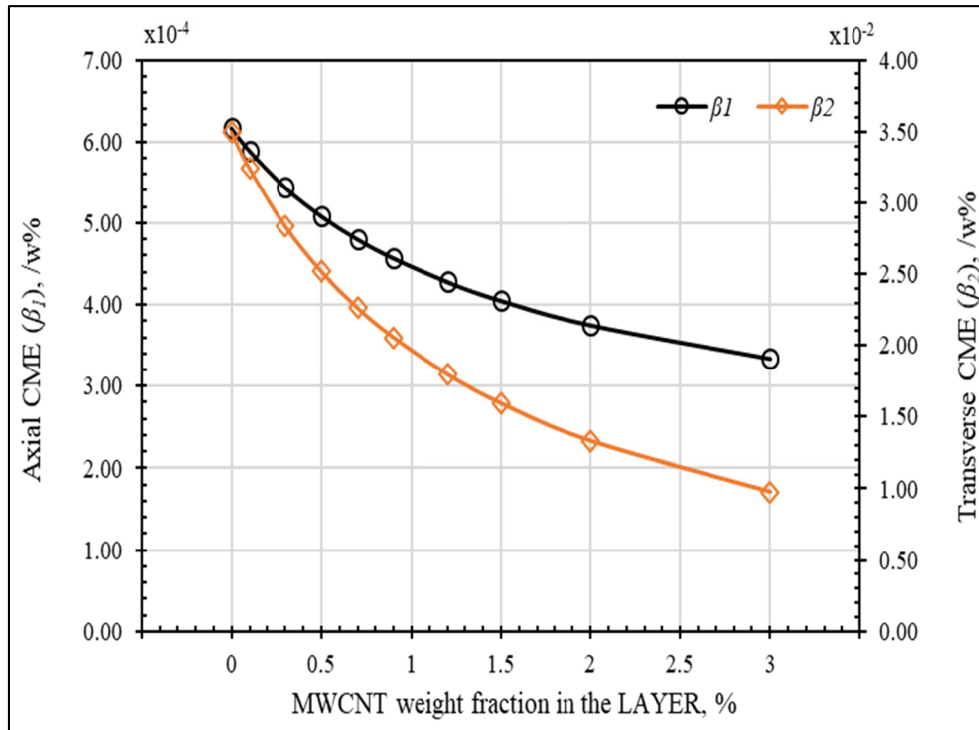


Figure 5. Change of the longitudinal and transverse moisture expansion coefficients of the layers according to MWCNT ratio

Hygrothermal load-induced plane stresses and strains in the composite laminates were also calculated and given in Figure 6. Since the laminates were designed as balanced and quasi-isotropic, stresses and strains along longitudinal and transverse directions came out as equal. However, it was obviously seen that the MWCNT particles improve the stability of the fiber-reinforced laminates under hygrothermal loads, and the improvement of the MWCNTs decreases by the increasing of the weight ratio of the nanotubes in the composite. In fact, because of the viscosity reducing effect and curing delaying effect of the nanotubes on the epoxy, it is very difficult to produce a composite laminate containing more than 1.5% particles by weight.

The bending load caused ply-wise longitudinal stresses and shear stresses were also studied for the unmodified and the MWCNT modified laminates (Table 3). In the bending analysis performed with some of the designed models, it was seen that the

MWCNTs did not affect the stresses due to bending load. Therefore, the analyzes made on half of the models (C_00, C_05, C_10, C_15, C_30) were found sufficient. As indicated in Table 2, MWCNTs have no significant effect on the in-plane elastic properties of the layers and so on laminates. Thence, the bending load response of the unmodified and the MWCNT modified laminates are similar and MWCNTs have no sense on the bending resistance of the composite laminates. It is clearly stated in the literature that the MWCNT modify has no significant increase in the bending or flexural strength [22-25]. Also, it was reported that the flexural strength was decreased in low quantities with the MWCNT reinforce by Yao et al. [21]. Similarly, the flexural strength was decreased with the 3%, 5% and 7% MWCNT modified carbon-felt-reinforced polypropylene-polyethylene composite [22]. In addition, it was noted that the MWCNT reinforce was decreased area specific resistance of the composite structure.

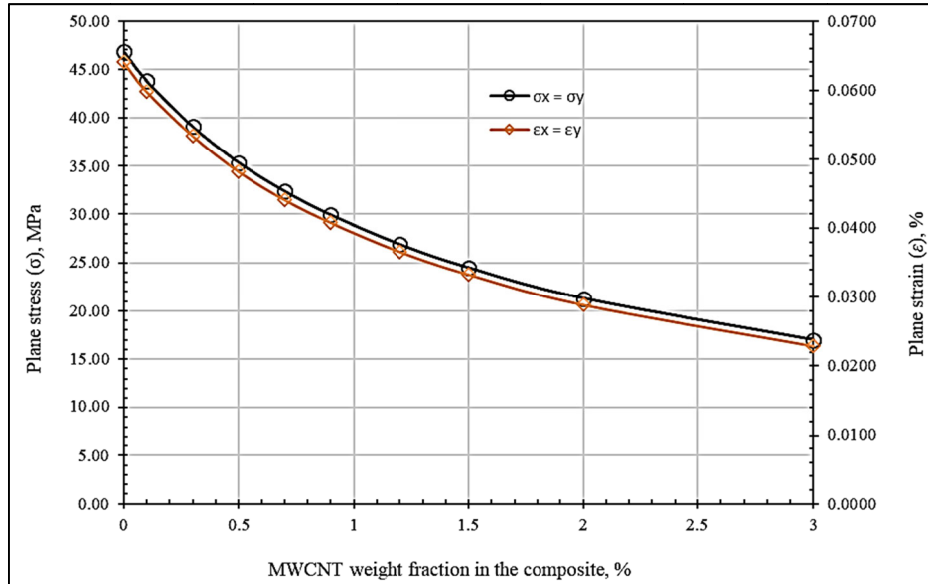


Figure 6. Change of the plane stresses and the strains in the quasi-isotropic laminates according to MWCNT ratio

Table 3. Bending load induced ply-wise stresses in the unmodified and MWCNT modified carbon/epoxy composites

Layer	C 00		C 05		C 15		C 30	
	σ_1	τ_{12}	σ_1	τ_{12}	σ_1	τ_{12}	σ_1	τ_{12}
-45	96.0054	20.5484	95.8848	20.5455	96.0231	20.5407	96.2445	20.5245
-45	86.9047	19.0831	86.7405	19.0779	86.8185	19.0698	86.9704	19.0509
45	77.8040	17.6179	77.5962	17.6104	77.6140	17.5990	77.6962	17.5773
45	77.7993	17.6179	77.5915	17.6104	77.6092	17.5990	77.6914	17.5773
90	71.8273	16.2199	71.5951	16.2106	71.5845	16.1969	71.6396	16.1735
90	65.8554	14.8219	65.5988	14.8109	65.5597	14.7949	65.5877	14.7697
0	108.7764	-14.8219	108.5710	-14.8109	108.6042	-14.7949	108.7415	-14.7697
0	97.9490	-13.4414	97.7100	-13.4308	97.6980	-13.4148	97.7806	-13.3900
0	87.1217	-12.0609	86.8490	-12.0507	86.7918	-12.0347	86.8198	-12.0102
0	87.1098	-12.0609	86.8371	-12.0507	86.7797	-12.0347	86.8076	-12.0102
90	78.7479	-10.7558	78.4654	-10.7463	78.3981	-10.7312	78.4162	-10.7082
90	70.3861	-9.4506	70.0938	-9.4418	70.0165	-9.4277	70.0248	-9.4061
45	-85.0970	-0.6910	-85.4919	-0.6917	-85.8083	-0.6909	-86.2221	-0.6885
45	-71.6758	-0.5686	-72.0778	-0.5701	-72.3673	-0.5703	-72.7188	-0.5692
-45	-58.2546	-0.4462	-58.6637	-0.4485	-58.9264	-0.4498	-59.2155	-0.4499
-45	-58.2664	-0.4462	-58.6756	-0.4485	-58.9384	-0.4498	-59.2277	-0.4499

4. CONCLUSION

In the present study, carbon fibre/MWCNT modified epoxy composite laminates were

investigated by means of electrical, mechanical, and hygrothermal properties, theoretically. Properties of the layers with the carbon fibre and the modified epoxy layers were analytically determined and the effect of the MWCNT content

was compared. The ratio of the MWCNTs in the epoxy was evaluated in the range of 0.1% to 3% by weight considering the existing literature when preparing epoxy/MWCNT mixture. MWCNTs significantly decrease the hygrothermal expansion coefficients of the epoxy. By adding MWCNT in a weight ratio of %3, the thermal expansion coefficient and moisture expansion coefficient of the epoxy decreased by 29.69% and 181.37%, respectively.

The MWCNT modified resin was approved as the matrix for carbon fiber reinforced composite lamina. The mechanical and the hygrothermal behavior of the quasi-isotropic laminates (with $[-45_2/45_2/90_2/0_2]_s$ oriented layers) were calculated and the stacking sequence of the plies was kept constant for each laminate. Ambient temperature and humidity values were assigned as 20 °C and 0.95, respectively. Moreover, the bending characteristics of MWCNT modified composite laminates were performed which is one of the most important operating characteristics for bipolar plates used in fuel cells. The main conclusions from this investigation can be drawn as follows:

- First of all, the electrical properties of the epoxy were improved with the MWCNT contribution. The electrical conductivity of the 3% MWCNT including epoxy was determined as 47.39 S/m and the conductivity value was increased by 158.22% with introducing carbon fibres into it. These conductivity values satisfy the electrical conductivity of composite bipolar plate requirements of the Department of Energy (DOE) [19].
- The effect of the MWCNTs on the mechanical features of composite layers are limited due to the mechanical properties of the unidirectional fibre reinforced layer depend on the fibres. Even so, MWCNT is contributed to mechanical characteristics. 0.97% and 8.74% increase were calculated for the longitudinal and transverse elastic modulus, respectively with the 3% MWCNT additives.
- The coefficients of longitudinal and transverse thermal expansions were changed in ratios of

4.54% and -80.20%, respectively for the 3% MWCNT/epoxy-carbon fibre composite. On the other hand, the coefficients of longitudinal and transverse moisture expansions decreased in order of 45.80% and 71.98%.

- It was understood that MWCNT has no important effect on the bending strength of composite laminates. Similar bending results were obtained for unmodified and MWCNT modified composites.

Nomenclature

f	volume fraction
V	volume of constituents
E	elastic modulus
G	shear modulus
ν	poisson's ratio
α	coefficient of thermal expansion
β	coefficient of moisture expansion
K	electrical conductivity

The superscripts are as follows;

e	epoxy polymer
Ω	MWCNT
m	MWCNT modified epoxy matrix
f	carbon fibre
c	carbon fibre/epoxy composite
T	thermal
M	moisture

The subscripts are as follows;

1	longitudinal direction of reinforcements
2	transverse direction of reinforcements

5. REFERENCES

1. Gojny, F.H., Wichmann, M.H.G., Fiedler, B., Bauhofer, W., Schulte, K., 2005. Influence of Nano-Modification on the Mechanical and Electrical Properties of Conventional Fibre-Reinforced Composites, Compos. Part A Appl. Sci. Manuf., 36, 1525–1535. doi:10.1016/j.compositesa.2005.02.007.
2. Wichmann, M.H.G., Sumfleth, J., Gojny, F.H., 2006. Glass-fibre-reinforced Composites with Enhanced Mechanical and Electrical Properties

- Benefits and Limitations of a Nanoparticle Modified Matrix, *Eng. Fract. Mech.*, 73(16), 2346–2359. doi:10.1016/j.engfracmech.2006.05.015.
3. Chen, Q., Zhang, L., Rahman, A., Zhou, Z., Wu, X., Fong, H., 2011. Hybrid Multi-scale Epoxy Composite Made of Conventional Carbon Fiber Fabrics with Interlaminar Regions Containing Electrospun Carbon Nanofiber Mats, *Compos. Part A*, 42, 2036–2042. doi:10.1016/j.compositesa.2011.09.010.
 4. Ashrafi, B., Diez-Pascual, A.M., Johnson, L., Genest, M., Hind, S., Martinez-Rubi, Y., González-Domínguez, J.M., Martínez, M.T., Simard, B., Gómez-Fatou, M.A., Johnston, A., 2012. Processing and Properties of PEEK/Glass Fiber Laminates: Effect of Addition of Single-Walled Carbon Nanotubes. *Compos. Part A*, 43, 1267–1279. doi:10.1016/j.compositesa.2012.02.022.
 5. Ashrafi, B., Naffakh, M., Di, A.M., Gonza, M., Johnston, A., Simard, B., Martínez, M.T., Gómez-Fatou, M.A., 2011. Influence of Carbon Nanotubes on the Thermal, Electrical and Mechanical Properties of Poly (Ether Ether Ketone)/Glass Fiber Laminates, *Carbon*, 49(8), 2817–2833. doi:10.1016/j.carbon.2011.03.011.
 6. da Costa, E.F.R., Skordos, A.A., Partridge, I.K., Rezai, A., 2012. RTM Processing and Electrical Performance of Carbon Nanotube Modified Epoxy/fibre Composites, *Compos. Part A Appl. Sci. Manuf.*, 43, 593–602. doi:10.1016/j.compositesa.2011.12.019.
 7. Socher, R., Krause, B., Boldt, R., Hermasch, S., Wursche, R., Pötschke, P., 2011. Melt Mixed Nano Composites of PA12 with MWNTs: Influence of MWNT and Matrix Properties on Macrodispersion and Electrical Properties. *Compos. Sci. Technol.*, 71, 306–314. doi:10.1016/j.compscitech.2010.11.015.
 8. Han, S., Meng, Q., Araby, S., Liu, T., Demiral, M., 2019. Mechanical and Electrical Properties of Graphene and Carbon Nanotube Reinforced Epoxy Adhesives: Experimental and Numerical Analysis. *Compos. Part A Appl. Sci. Manuf.*, 120, 116–126. doi:10.1016/j.compositesa.2019.02.027.
 9. Ma, P.C., Siddiqui, N.A., Marom, G., Kim, J.K., 2010. Dispersion and Functionalization of Carbon Nanotubes for Polymer-Based Nanocomposites: A Review, *Compos. Part A Appl. Sci. Manuf.*, 41, 1345–1367. doi:10.1016/j.compositesa.2010.07.003.
 10. Eskizeybek, V., Avci, A., Gülce, A., 2014. The Mode I Interlaminar Fracture Toughness of Chemically Carbon Nanotube Grafted Glass Fabric/epoxy Multi-scale Composite Structures, *Compos. Part A Appl. Sci. Manuf.*, 63, 94–102. doi:10.1016/j.compositesa.2014.04.013.
 11. Ghislandi, M., de A. Prado, L.A.S., Barros-Timmons, K.S.A., 2013. Effect of Filler Functionalization on Thermo-mechanical Properties of Polyamide-12/Carbon Nanofibers Composites: A Study of Filler–Matrix Molecular Interactions, *J. Mater. Sci.*, 48, 8427–8437. doi:10.1007/s10853-013-7655-4.
 12. Zhu, Y., Bakis, C.E., Adair, J.H., 2012. Effects of Carbon Nanofiller Functionalization and Distribution on Interlaminar Fracture Toughness of Multi-scale Reinforced Polymer Composites, *Carbon*, 50(3), 1316–1331. doi:10.1016/j.carbon.2011.11.001.
 13. Chen, X., Wang, J., Lin, M., Zhong, W., Feng, T., Chen, X., Chen, J., Xue, F., 2008. Mechanical and Thermal Properties of Epoxy Nanocomposites Reinforced with Amino-functionalized Multi-walled Carbon Nanotubes *Mater. Sci. Eng. A*, 492, 236–242. doi:10.1016/j.msea.2008.04.044.
 14. Kim, Y.J., Shin, T.S., Choi, H.D., Kwon, J.H., Chung, Y.C., Yoon, H.G., 2005. Electrical Conductivity of Chemically Modified Multiwalled Carbon Nanotube/epoxy Composites, *Carbon*, 43, 23–30. doi:10.1016/j.carbon.2004.08.015.
 15. Sagar, R., Petrova, R.S., Somenath, M., 2018. Effect of Carbon Nanotube (CNT) Functionalization in Epoxy-CNT Composites, *Nanotechnol. Rev.*, 7, 475–485. doi:10.1016/j.physbeh.2017.03.040.
 16. Moisala, A., Li, Q., Kinloch, I.A., Windle, A.H., 2006. Thermal and Electrical Conductivity of Single- and Multi-walled Carbon Nanotube-epoxy Composites, *Compos.*

- Sci. Technol., 66, 1285–1288. doi:10.1016/j.compscitech.2005.10.016.
17. Jarali, C.S., Patil, S.F., Pilli, S.C., 2015. Hygrothermo-electric Properties of Carbon Nanotube Epoxy Nanocomposites with Agglomeration Effects, *Mech. Adv. Mater. Struct.*, 22, 428–439. doi:10.1080/15376494.2013.769654.
 18. Zhang, Y.C., Wang, X., 2006. Hygrothermal Effects on Interfacial Stress Transfer Characteristics of Carbon, *J. Reinf. Plast. Compos.*, 25(1), 71-88. doi:10.1177/0731684406055456.
 19. Antunes, R.A., de Oliveira, M.C.L., Ett, G., Ett, V., 2011. Carbon Materials in Composite Bipolar Plates for Polymer Electrolyte Membrane Fuel Cells: A Review of the Main Challenges to Improve Electrical Performance, *J. Power Sources*, 196, 2945-2961. doi.org/10.1016/j.jpowsour.2010.12.041.
 20. Bairan, A., Selamat, M.Z., Sahadan, S.N., Malingam, S., Mohamad, N., 2018. Effect of CNTs on the Electrical and Mechanical Properties of Polymeric Composite as PEM Fuel Cell Bipolar Plate, *J. Teknol. Sci. Eng.*, 80(6), 115-122.
 21. Yao, K., Adams, D., Hao, A., Zheng, J.P., Liang, Z., Nguyen, N., 2017. Highly Conductive and Strong Graphite-Phenolic Resin Composite for Bipolar Plate Applications, *Energy Fuels*, 31, 14320-14331. doi:10.1021/acs.energyfuels.7b02678.
 22. Lee, H.E., Chung, Y.S., Kim, S.S., 2017. Feasibility Study on Carbon-Felt-Reinforced Thermoplastic Composite Materials for PEMFC Bipolar Plates, *Compos. Struct.*, 180, 378-385. doi.org/10.1016/j.compstruct.2017.08.037.
 23. Bairan, K.A., Selamat, M.Z., Sahadan, S.N., Malingam, S.D., Mohamad, N., 2016. Effect of Carbon Nanotubes Loading in Multifiller Polymer Composite as Bipolar Plate for PEM Fuel Cell, *Proced. Chem.*, 19, 91-97. doi:10.1016/j.proche.2016.03.120.
 24. Chaiwan, P., Pumchusak, J., 2015. Wet vs. Dry Dispersion Methods for Multiwall Carbon Nanotubes in the High Graphite Content Phenolic Resin Composites for use as Bipolar Plate Application, *Electrochim. Acta*, 158, 1-6. doi.org/10.1016/j.electacta.2015.01.101.
 25. Suherman, H., Sulong, A.B., Sahari, J., 2013. Effect of Compression Molding Parameters on the In-Plane and Through-Plane Conductivity of Carbon Nanotubes/Graphite/Epoxy Nanocomposites as Bipolar Plate Material for a Polymer Electrolyte Membrane Fuel Cell, *Ceram. Int.*, 39, 1277-1284. doi.org/10.1016/j.ceramint.2012.07.059.
 26. Liao, S.H., Yen, C.Y., Weng, C.C., Lin, Y.F., Ma, C.C.M., Yang, C.H., Tsai, M.C., Yen, M.Y., Hsiao, M.C., Lee, S.J., Xie, X.F., Hsiao, Y.H., 2008. Preparation and Properties of Carbon Nanotube/Polypropylene Nanocomposite Bipolar Plates for Polymer Electrolyte Membrane Fuel Cells, *J. Power Sources*, 185, 1225-1232. doi:10.1016/j.jpowsour.2008.06.097.
 27. Davé, R., Gupta, R., Pfeffer, R., Sundaresan, S., Tomassone, M.S., 2006. Deagglomeration and Mixing of Nanoparticles, NSF Nanoscale Science and Engineering Grantees Conference, Grant#: 0506722, 2006, Dec 4-6.
 28. Ashrafi, B., Guan, J., Mirjalili, V., Zhang, Y., Chun, L., Hubert, P., Simard, B., Kingston, C.T., Bourne, O., Johnston, A., 2011. Enhancement of Mechanical Performance of Epoxy/carbon Fiber Laminate Composites Using Single-walled Carbon Nanotubes, *Compos. Sci. Technol.*, 71, 1569–1578. doi:10.1016/j.compscitech.2011.06.015.
 29. Mirjalili, V., Ramachandramoorthy, R., Hubert, P., 2014. Enhancement of Fracture Toughness of Carbon Fiber Laminated Composites Using Multi Wall Carbon Nanotubes. *Carbon*, 79, 413–423. doi:10.1016/j.carbon.2014.07.084.
 30. Jones, M.R., 1999. *Mechanics of Composite Materials*, 2nd Ed., Taylor & Francis, Inc. PA, 19106.