Chapter 2
Electrically Aligned Epoxy-Based Nanocomposites: Processing and Characterization

Rahman Julkarnyne M. Habibur and Gururaja Suhasini

Abstract This study examines the electric-field alignment of amine-functionalized multi-walled carbon nanotubes (MWCNTs) within an epoxy matrix, aiming to enhance their mechanical, electrical, and thermal performance. Amine-functionalized MWCNTs were dispersed within the epoxy resin using high-shear mixing to achieve a homogenous nanocomposite. 0.1, 0.2, 0.3 wt% of nanomaterials are used to prepare the samples. A key feature of the research involved the application of an external AC electric field before curing to align MWCNTs in the desired orientation. The voltage and the distance of electrodes were varied to examine the effect of the AC electric field on the alignment time. Response surface methodology (RSM), along with the central composite design (CCD), has been used to analyze the influence of those process parameters. Analysis of variance (ANOVA) was performed to identify the significant parameters. A voltage of 1000V and a distance of 50 mm between the electrodes was the optimal solution to minimize the alignment time while aligning MWCNTs in the epoxy matrix.

Keywords Nanocomposite · Electric field alignment · ANOVA · RSM · Optimization

Introduction

Polymer matrix composites (PMCs) are the combinations of different organic polymers with reinforced fibers. Manufacturers can produce PMCs from polymers such as thermoplastics and thermosets. Because of their excellent mechanical properties, thermal stability, and chemical resistance, people widely employ epoxy resins—the most commonly used thermoset—in the aerospace, automotive, and electronics industries [1]. Nanofillers, such as carbon nanotubes, can further enhance the performance of epoxy resins. The efficacy of these epoxy-nanocomposites hinges on various factors such as the dispersion quality [2], [3], weight fraction [2], and alignment [4], [5], [6], [7] of nanofillers in the epoxy matrix.

Alignment is generally done by applying external forces such as an electric field [8], magnetic means [6], high-shear mixing, etc. Electric field-assisted alignment has emerged as a versatile technique for orienting conductive MWCNTs, achievable via alternating (AC) or direct (DC) potential fields. Pothnis et al. [9] used the non-uniform AC electric field to align the CNTs, and Khan et al. [10] and Oliva et al. [11] used the DC electric field to align the CNTs in the matrix. The alignment time mostly depends on the rotation and migration of the MWCNTs in the epoxy matrix [12]. Viscosity plays a major role in the alignment as the lower viscosity facilitates the alignment [13]. Factors such as voltage and electrode distance significantly influence this alignment by determining the electric field strength. During the alignment process, the applied electric field generates a torque needed to overcome the epoxy matrix's drag force, enabling CNT rotation and subsequent migration towards an electrode [12] Y. Minimizing the alignment time is an important factor to facilitate the large-scale fabrication of these aligned nanocomposites [14]. However, literature reports a wide disparity in these parameters—ranging from 10 V to 1000 V and distances of 1 mm to 50 mm—owing to variations in MWCNT type, epoxy viscosity, and system geometry [10], [11], [15]. While recent studies have leveraged RSM with ANOVA to optimize shear rate effects in MWCNT nanofluids along elongated surfaces [16] and employed Taguchi design for nitrogen-doped CNT synthesis [17], few have targeted the optimization of electric field parameters for minimizing CNT alignment time in epoxy matrix. This oversight limits the scalability and reproducibility of CNT/epoxy nanocomposite fabrication.

Rahman Julkarnyne M. Habibur · Gururaja Suhasini
Department of Aerospace Engineering, Auburn University, Auburn, AL 36849
e-mail: jzr0114@auburn.edu; suhasini.gururaja@auburn.edu

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In this work, a design of experiments approach integrating RSM and ANOVA has been adopted to optimize voltage and electrode distance for MWCNT alignment in the epoxy matrix. A CCD was utilized within the RSM framework to construct a systematic test matrix, incorporating critical process parameters: applied voltage, electrode distance, and MWCNT weight percentage. The response variable, alignment time, was experimentally determined for each condition outlined in the test matrix. Linear regression models were subsequently developed to map the relationship between input parameters and alignment time, enabling the identification of optimal process conditions. Additionally, ANOVA was performed to assess each parameter's statistical significance and interactions, elucidating their individual and combined effects on the alignment process.

Materials

Epon-862 (diglycidyl ether of bisphenol F) and EPIKURE curing agent W (bisphenol-F epoxy resin and an aromatic amine) purchased from Miller-Stephenson were used as the resin and hardener following manufacturers recommended ratio of 100:26.4. Amine-functionalized MWCNTs (length: 10∼30 μm, outside dia: 10∼20 nm) were sourced from MKnano. The MWCNT dimensions were confirmed via SEM. All the materials were used as received.

Nanocomposite Fabrication with Electric Field Alignment

The nanocomposite is fabricated following a systematic approach to ensure proper dispersion. Initially, CNTs were preheated at 105°C for 10 minutes to eliminate moisture. The required amount of preheated nanofillers was then added to the epoxy resin, and dispersion was performed using an overhead stirrer (ONiLAB Electric Overhead Stirrer) equipped with a propeller-shaped blade. The stirring process was conducted at approximately 1000 rpm for 4.5–5 hours at 80°C to achieve effective dispersion. Afterward, the curing agent (Epikure W) was introduced into the mixture, followed by additional stirring for 7 minutes. The mixture was then vacuum-degassed for 10 minutes to remove the entrapped air. Following the dispersion process, the nanocomposite mixture was maintained at 80°C in an oven before being transferred into a preheated silicone mold. The mold was rectangular, with dimensions of 180 mm × 40 mm (L × W), and was initially preheated in the oven to ensure uniform thermal conditions. Subsequently, the mold was placed on a hotplate set to 125°C, ensuring the mixture temperature remained around 80°C to maintain low viscosity during the alignment process. Two copper electrodes were positioned at a fixed distance and immersed in the nanocomposite mixture. An alternating current (AC) voltage was applied via a function generator coupled with a power amplifier to induce the alignment of carbon nanotubes (CNTs). The current was monitored with a digital multimeter throughout the alignment process to assess the progression of CNT movement. The voltage supply was discontinued after a predetermined duration, which depended on the applied voltage and electrode spacing. The mixture was then allowed to cool for 25 minutes before being transferred to an oven for curing. The curing process was conducted at 121°C for 2 hours, followed by post-curing at 177°C for 2 hours. Using this methodology, CNT/epoxy nanocomposites were fabricated with CNT weight fractions of 0.1, 0.2, and 0.3 wt.%. Figure 1 depicts the process flowchart.

Experimental Procedure

Three voltage levels of 500 V, 870 V, and 1000 V were used for these experiments. Distances of 50 mm, 75 mm, and 100 mm were used and combined with the mentioned voltage level to find the minimum alignment time. The frequency was fixed at 10 kHz. An RSM approach employing CCD was utilized to assess alignment time across various parameter combinations. Key process parameters—voltage, electrode distance, and CNT weight percentage (wt%)—served as experimental inputs (Table 1). The objective was to identify the optimal parameters yielding the minimum alignment time. The DOE approach reduced the experimental trials required while enabling analysis of voltage and distance effects on alignment time.

Table 1 Process parameters

Parameters	Code	Level of parameters		
Voltage (V)	A	500	870	1000
Distance (mm)	B	50	75	100
Weight percentage (wt%)	C	0.1	0.2	0.3

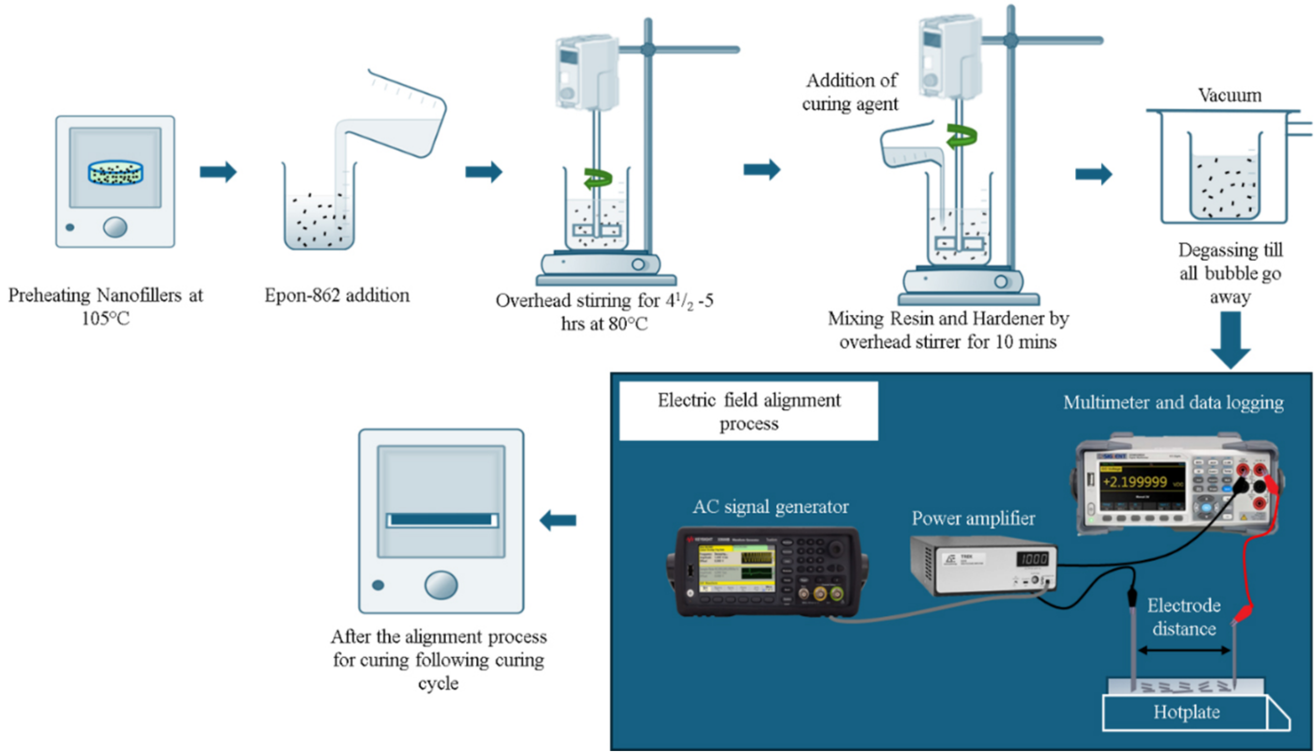

Fig. 1 Nanocomposite fabrication process with electric field alignment process

Results and Discussion

Using Stat-Ease 360 software, DOE tables were generated based on these inputs. Two experimental cases were considered for this approach. The first case involved only 0.1 wt.% CNT/epoxy, while the second case included 0.1, 0.2, and 0.3 wt.% CNT/epoxy. Table 2 presents the design matrix and corresponding output responses for 11 experimental runs in the first case, where voltage and distance were varied. Similarly, Table 3 details the design matrix and output responses for 15 experiments conducted under the second case based on the selected process parameters. In all these experiments, alignment time was monitored by measuring current (A) data from the multimeter. An empirical model was developed for both cases utilizing RSM. Following RSM implementation and identification of significant input parameters, ANOVA was performed to ensure the responses' statistical accuracy and validate the proposed model's suitability for predicting alignment time.

Table 2 Experiment matrix and obtained results for 0.1 CNT/Epoxy

	Factor 1	Factor 2	Response 1
Run	A: voltage (V)	B: Distance (mm)	Alignment time (min)
1	500	75	15
2	870	50	6
3	870	100	16
4	870	75	10
5	500	100	18
6	1000	50	3
7	1000	100	15
8	870	75	10
9	870	75	10
10	500	50	8
11	1000	75	12

Table 3 Experiment matrix and obtained results for different weight percentages (0.1, 0.2, 0.3 wt%)

	Factor 1	Factor 2	Factor 3	Response 1
Run	A: Voltage (V)	B: Distance (mm)	C: Weight percentage (wt%)	Alignment time(min)
1	870	100	0.1	16
2	870	50	0.1	6
3	500	75	0.1	15
4	870	75	0.1	10
5	1000	75	0.3	9
6	500	50	0.2	10
7	500	75	0.3	10
8	500	100	0.2	18
9	1000	100	0.2	10
10	500	50	0.1	8
11	870	50	0.3	10
12	1000	75	0.1	12
13	870	75	0.2	11
14	1000	50	0.1	3
15	870	100	0.3	14

Analysis for Alignment time

ANOVA for minimized alignment time has been represented in Tables 4 to 5, respectively. The F-value is a critical metric for identifying factors that significantly influence the physical quantity of interest. Specifically, a large F-value indicates that variations in a particular factor have a substantial impact on the observed output [17]. Table 4 shows that the model F-value of 54.93 indicates significant model performance, with only a 0.01% probability that such a high F-value is due to noise. P-values below 0.0500 denote significant model terms [18], specifically A and B, in this case.

Table 4 ANOVA for alignment time for 0.1 CNT/Epoxy

Source	**Sum of Squares (SS)**	**df**	**Mean Square**	**F-value**	**p-value**	**Remarks & Percentage of Contribution (%)**
Model	209.64	2	104.82	54.93	< 0.0001	
A-Voltage	28.14	1	28.14	14.75	0.0049	12.51 (significant)
B-Distance	181.50	1	181.50	95.12	< 0.0001	80.70
Residual	15.27	8	1.91			
Cor Total	224.91	10				
Std. Dev.	1.38			$\mathbf{R}^2$	0.9321	
Mean	11.09			Adjusted $\mathbf{R}^2$	0.9152	
Coefficient of variance (%)	12.45			Predicted $\mathbf{R}^2$	0.8652	
				Adequate precision	20.9609	

In Table 5, the Model F-value of 9.45 implies the model is significant, and there is only a 0.22% chance that an F-value this large could occur due to noise. Additionally, P-values less than 0.0500 indicate that model terms are significant. A (voltage) and B (distance) are significant model terms valid at 95% confidence in both cases. Additionally, the signal-to-noise ratio is evaluated using adequate precision measures, with a ratio exceeding four deemed desirable [16]. In Table 4, the adequate precision is 20.96, and in Table 5, it is 9.6, signifying both models are desirable and can be used to navigate the design space.

Figure 2 represents the normal plot of the residual for the alignment time. The residuals align closely along the normal line, indicating that the errors follow a normal distribution [18]. In both cases, the empirical model obtained for the output

Table 5 ANOVA for alignment time for different weight percentages of CNT/Epoxy

Source	Sum of Squares (SS)	df	Mean Square	F-value	p-value	Remarks& Percentage of Contribution (%)
Model	160.60	3	53.53	9.45	0.0022	
A-Voltage	40.65	1	40.65	7.17	0.0215	18.23
B-Distance	131.81	1	131.81	23.26	0.0005	59.13
C-Weight percentage	0.1124	1	0.1124	0.0198	0.8906	0.05
Residual	62.33	11	5.67			
Cor Total	222.93	14				
Std. Dev.	2.38			$\mathbf{R}^2$	0.7204	
Mean	10.73			Adjusted $\mathbf{R}^2$	0.6442	
Coefficient of variance (%)	22.18			Predicted $\mathbf{R}^2$	0.4162	
				Adequate precision	9.6048	

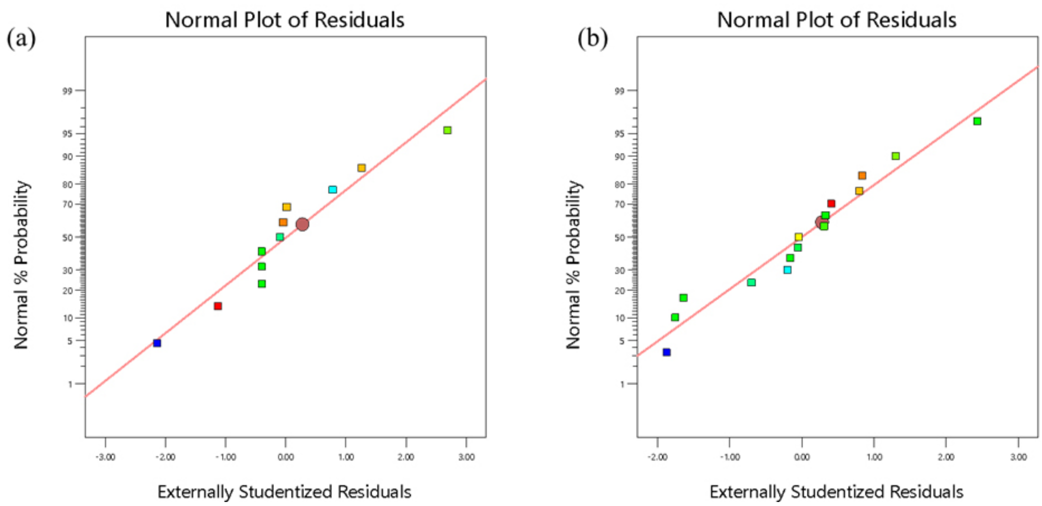

Fig. 2 Normal probabilities of residuals for (a) Alignment time using only 0.1 wt% CNT/Epoxy and (b) alignment time when different weight percentages of CNT used

was deemed effective at a 95% confidence level. The regression model for both cases factoring voltage and distance for the alignment time is presented in equations(1) and (2).

When the weight percentage is fixed at 0.1 wt% CNT, then the regression model of minimum alignment time is represented in equation (1)

$$\text{Alignment time} = 11.540 - 2.061^{*}\text{A} + 5.5^{*}\text{B} \quad \text{(Equation 1)}$$

When the weight percentage is varied between 0.1, 0.2, and 0.3 wt% CNT, then the regression model of minimum alignment time is represented in equation (2)

$$\text{Alignment time} = 11.22 - 2.017^{*}\text{A} + 3.939^{*}\text{B} - 0.106^{*}\text{C} \quad \text{(Equation 2)}$$

Where A represents the voltage, B represents the distance, and C represents the weight percentage of the CNT.

Multi-response optimization

The desirability function approach was employed for multi-response optimization to determine the optimal input process parameters. This method is widely utilized for optimizing both single- and multi-response problems. In this approach, each output response is converted into a dimensionless desirability index (D), ranging from 0 (least desirable) to 1 (most desirable). The optimal parameter set is identified when the corresponding solution achieves the highest overall desirability, ensuring the best possible outcome across multiple responses [18], [19]. In this study, the optimal values of voltage and distance are achieved after establishing the goal to minimize the alignment time. *Figure 3* represents the optimized bar histogram for desirability. Tables 6 and 7 present the constraints for the input parameters and the corresponding optimal solutions for minimizing alignment time. In both cases, the predicted optimal parameters were determined to be 1000 V and 50 mm, achieving the highest desirability score of 0.8. Furthermore, Table 8 compares the predicted and observed values of the process parameters to validate the model's accuracy. The average deviation between observed and predicted values is

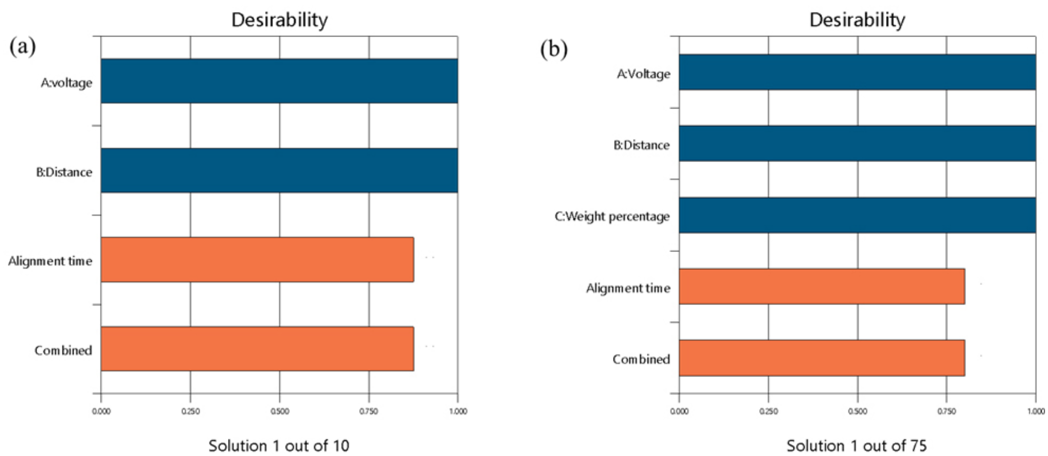

Fig. 3 Bar histogram demonstration for desirability (a) Alignment time using only 0.1 wt% CNT/Epoxy (b) alignment time when different weight percentages of CNT used

Table 6 Constraints and optimal solutions when weight percentage is fixed at 0.1 wt%

Name	Goal	Lower Limit	Upper Limit	Lower Weight	Upper Weight	Importance	Optimal solutions
A: Voltage	is in range	500.0	1000.0	1.0	1.0	3	1000
B: Distance	is in range	50.0	100.0	1.0	1.0	3	50
Alignment time	minimize	2.0	18.0	1.0	1.0	3	3.980

Table 7 Constraints and optimal solutions when weight percentage is varied

Name	Goal	Lower Limit	Upper Limit	Lower Weight	Upper Weight	Importance	Optimal solutions
A: Voltage	is in range	500.0	1000.0	1.0	1.0	3	1000
B: Distance	is in range	50.0	100.0	1.0	1.0	3	50
C: Weight percentage	is in range	0.1	0.3	1.0	1.0	3	0.3
Alignment time	minimize	2.0	18.0	1.0	1.0	3	5.165

Table 8 Observed and predicted results at optimum values of process parameters

Voltage (V)	Distance (mm)	Weight percentage (wt%)	Alignment time (Predicted by model)	Alignment time (Experimental value)
870	75	0.1	10.35	10
500	50	0.1	9	8
1000	75	0.3	9.15	9
1000	100	0.2	13.15	15
500	100	0.2	17.18	18

approximately 0.83, demonstrating the model's reliability in predicting alignment time for different voltage and distance combinations.

Effect of Process Parameters on Alignment Time

From the ANOVA results (Table 4, Table 5), it can be observed that both voltage and distance play a significant role in minimizing alignment time. The percentage of contribution ratio (PCR) of each input parameter can quantify the effect and the interaction of those parameters. The PCR in both tables is calculated by dividing the sum of squares (SS) for each factor by the total SS [17]. As shown in Table 4, the electrode distance contributes 80.70% to minimizing the alignment time for 0.1 wt.% CNT/Epoxy, whereas the applied voltage accounts for only 12.51%. Similarly, Table 5 presents the percentage contribution ratios (PCRs) for varying CNT weight fractions, indicating that distance and voltage contribute 59.13% and 18.23%, respectively, to reduce alignment time. These findings highlight the dominant influence of electrode distance over voltage in optimizing the alignment process. The ANOVA results indicate that electrode distance is the primary factor influencing alignment time. This is attributed to its significant impact on the electric field strength, which plays a crucial role in facilitating the formation of an aligned conductive network under the applied AC electric field [9], [10]. With the decrease in distance and the increase in voltage, the electric field strength increased which eventually increased the torque that facilitates the alignment of the CNT in the electric field direction[20]. In addition to the ANOVA results, the initial test matrix (Tables 1 and 2) highlights that a lower electrode distance and higher applied voltage enhance the electric field strength, thereby influencing alignment time. However, excessively high voltages can induce CNT aggregation, leading to the formation of rope-like structures, which may adversely impact the nanocomposite's properties [13].

As shown in Table 5, the weight percentage of CNTs has a minimal effect on reducing alignment time, with a PCR of only 0.05%. However, in practice, CNT weight percentage does influence alignment time. A higher CNT concentration provides more conductive pathways, facilitating alignment under the applied voltage compared to lower concentrations [10].

Conclusions

This study demonstrates the efficacy of integrating RSM and ANOVA to optimize voltage and electrode distance for minimizing the MWCNT alignment time in the epoxy matrix.

- Higher voltage and shorter distance significantly enhance alignment efficiency.
- Future work will involve COMSOL simulations to further investigate the effects of viscosity and epoxy curing dynamics, refining alignment kinetics, and paving the way for industrial scalability.
- The study will be extended to optimize the voltage and distance for the GNP/Epoxy sample in the future.

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