5

Optimization of Moulding Parameters on the Electrical Conductivity of Carbon Black/Graphite/Epoxy Composite for Bipolar Application

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Optimization of the moulding parameters on the carbon black/graphite/epoxy (CB/G/EP) composite for bipolar plate application using the Taguchi method were carrying out. Moulding parameters of the compression moulding process such as moulding temperature, moulding pressure and moulding time were measured. Analysis of variance (ANOVA) shows that, the most significant moulding parameter is moulding time with percentage contribution of 59.98%.The confirmation experiment using additive model shows that, the electrical conductivity of CB/G/EP composites was 168.50 S/cm. The electrical conductivity of CB/G/EP composite obtained was improved 65.72 % compare with the initial trial. The results show that Taguchi method is an effective approach to obtain the optimal moulding parameters of the CB/G/EP composites.

Keywords Electrical conductivity, bipolar plate, moulding parameters, Taguchi method

INTRODUCTION

Moulding parameters of the compression moulding process was affected to produce the electrical conductivity of the composite materials [1, 2]. A composite material has been produced from the incorporation of matrix as a binder and filler as conductive or reinforcement. Composite that has electrical conductivity property known as conductive polymer composites (CPCs). The U.S. Department of Energy (DOE) has already stated that the electrical conductivity must be ≥100 S/cm for bipolar plate application [3]. Generally, manufacturing process method such as compression moulding and injection moulding were used. Conductive fillers such as carbon nanotubes (CNTs), carbon black (CB), graphite (G) carbon fiber (CF) with different sizes and shapes were used to enhance the overall performance of composite materials [2, 4]. Although a significant improvement in the electrical conductivity had achieved, higher filler loadings concentrations are still needed to get the highest electrical conductivity as a prerequisite by U.S. DOE [2]. Wolf and Porda [5] were developed a liquid crystal polymer composite bipolar plate using compression moulding. The conductive filler used were the incorporation of CB and CF. Volume loading concentration of the conductive fillers was below 40 vol. %. They obtained the electrical conductivity of the composite bipolar plate was less than 6 S/cm, although the bipolar plate had excellent mechanical and gas barrier properties. Dweiri and Sahari [6] investigated CB/G/polypropylene (PP) composite bipolar plates using compression moulding method. They reported that the electrical conductivity obtained was only around 35 S/cm, at conductive fillers loading concentration as high as 80 wt. %. Similar work had also done by Yin et al. [7]. High fillers loading concentration of 85 wt. % G powder was required to achieve 100 S/cm of the electrical conductivity. Suherman et al. [8] also reported high filler loading concentration with different sizes of the conductive filler were needed to achieve the high electrical conductivity.

This paper thus describe Taguchi method for optimising the moulding parameters of CB/G/EP composite that consist of moulding temperature, moulding pressure and moulding time. These parameters affected the electrical conductivity of CB/G/EP composites produced. The Taguchi method was used to obtain the optimal moulding parameters. This method had already used in manufacturing processes that consist of: turning [9, 10], milling [11, 12], casting [13, 14] and conductive polymer composites (CPCs) [15- 18]. Although the moulding parameters are the important factor on the resulting high electrical conductivity of the CB/G/EP composite, the optimization of the moulding parameters has been given less attention in the literature, specially in manufacturing CB/G/EP composite.

In this study, the optimization of moulding parameters on the electrical conductivity of CB/G/EP composite using Taguchi method was investigated.

EXPERIMENTAL PROCEDURE

Materials

Carbon black (CB) and graphite (G) were used as conductive fillers. The particle sizes of CB and G were 30 nm and 74 µm, respectively. The surface area of CB is 250 m^2/gr, while G is 1.5 m^2/gr. Both of conductive fillers were supplied by Insutex Industries Sdn. Bhd., Malaysia as the local agent for Asbury Carbons Inc., U.S. The epoxy resin is a bisphenol-A based epoxy resin, with viscosity 6 Poise and curing agent, 4-Aminophenylsulphone were obtained from Mid Western Lab Suppliers., Malaysia as the local agent for U.S composite.

Moulding Parameters of the CB/G/EP Composites

Based on the previous research we found that the moulding parameters are significantly affected the electrical conductivity of the CB/G/EP composite [2]. The moulding parameters are moulding temperature, moulding pressure and moulding time. Table 1 shows that moulding parameters and their levels used in this study. The parameters are moulding temperature (A) = 110, 130, 150 ^{0}C, moulding pressure (B) = 20, 25, 30 MPa and moulding time (C) = 60, 75, 90 minutes.

Table 1. Moulding parameters and their levels

Symbols	Moulding parameters	Level 1	Level 2	Level 3
A	Moulding temperature (^{0}C)	110	130	150
B	Moulding pressure (MPa)	20	25	30
C	Moulding time (min)	60	75	90

Orthogonal Array

The orthogonal array (OA) can provide an efficient way to perform the experiments with the minimum number of trials. The arrangement of OA is based on the total degree of freedom (DOF_T) of the objective purpose. Since there are three moulding parameters and three levels, the DOF is given as eight. The performance comparisons between various combinations of moulding parameters can be obtained which selected OA having at least nine experimental trials (DOF + 1). For that reason, the standard L9 (3^3) OA is selected for the study.

Fabrication of CB/G/EP composite

Fabrication of CB/G/EP composite consists of three stages. In the firs stage, the CB and G as conductive fillers were mixed in a ball mill to get a homogenous mixture. A 20 Stainless steel balls (10 mm in diameter) at rotating speed of 200 rpm for one hour was used. In the second stage, EP and curing agent were mixed using a high speed mechanical mixer (RW 20-KIKA-WERK) for 40 seconds at 1200 rpm. In the third stage, CB/G/EP was mixed using an internal mixer (Haake Reomix) at 30°C. The rotational speed and the mixing time of the internal mixer were set at 25 rpm and 10 minutes [16]. The composition of CB/G/EP composite was fixed at 10/65/25 in vol. % [2].

The electrical conductivity of CB/G/EP composites

Jandel four-point probe combined with RM3 test unit were used to measure the electrical conductivity of CB/G/EP composites. It has a constant current sources and digital voltmeter. This equipment can be used to measure volume resistivity in range from 10^{-3} to 10^6 Ω-cm and 1 mΩ/cm up to 5 × 10^8 Ω/cm, respectively.

Data analysis

There are three quality characteristics in the Taguchi method which are the larger-the- better (LTB), the smaller-the-better (STB) and the nominal-the-best (NTB). LTB was chosen as data analysis tools because the objective of this study was to obtain the highest electrical conductivity of CB/G/EP composites. Signal-to-noise (S/N) ratio was also used to represent the quality characteristics. Signal means an expected output quality characteristics of the moulding parameters process, while noise means an unexpected quality characteristics. The quality characteristic (η) for the LTB is define as:

$$\eta = -10 \log (MSD) \quad (1)$$

where MSD is the mean-square-deviation of the objective purpose, and given as

$$MSD = \frac{1}{n}\sum_{i=1}^{n} 1/yi^2 \quad (2)$$

where n is the number of repeated experiments for each combination of moulding parameters and y_i is the measured electrical conductivity of the CB/G/EP composite at its repeating experiments.

RESULTS AND DISCUSSION

The electrical conductivity of CB/G/EP composites

Table 2 shows the experiment results of the electrical conductivity based on L9 (3^3) OA with implementation of the moulding parameters on the fabrication of CB/G/EP composite. The results consist of the electrical conductivity and S/N ratio for every various moulding parameters. Three measurements were performed for every combination of moulding parameters. The average maximum value of the electrical conductivity obtained was 143.73 S/cm. The results showed that the electrical conductivity of CB/G/EP composite increased from 101.68 S/cm (trial no.1) to 143.73 at trial no.5. It caused of the increasing of moulding parameters such as moulding temperature, moulding pressure and moulding time has a significant effect on the electrical conductivity of CB/G/EP composites produced [1, 2].

Table 2. Experimental results of the electrical conductivity based on L9 (3^3) OA.

Run no.	Moulding parameters			Electrical conductivity (S/cm)	S/N ratio (dB)
	Moulding temperature (^{0}C)	Moulding pressure (MPa)	Moulding time (minutes)	Averages	
1	110	20	60	101.68	40.13
2	110	25	75	107.85	40.49
3	110	30	90	114.04	41.10
4	130	20	75	116.60	41.32
5	130	25	90	143.73	43.15
6	130	30	60	104.47	40.29
7	150	20	90	131.43	42.37
8	150	25	60	93.89	39.42
9	150	30	75	85.32	38.61

Table 3. Effect of the moulding parameters on the S/N ratio (larger is better) of CB/G/EP composites

Level	Moulding parameters		
	A (Moulding temperature, ^{0}C)	B (Moulding pressure, MPa)	C (Moulding time, min)
1	40.57	41.27	39.95
2	41.59	41.02	40.14
3	40.14	40.00	42.21

The experimental design use in this study is an OA. The advantage of OA is possible to separate the effects of the moulding parameters at different various levels. Table 3. show the effect of moulding parameters on the electrical conductivity and S/N ratio (LTB), respectively. The optimal moulding parameter obtained were A_2 B_1 C_3, i.e., A_1 = 130 ^{0}C, B_1 = 20 MPa, C_3 = 90 minutes.

Analysis of variance

Table 4. shows the experiment moulding parameters based on L9 (3^3) OA. The percentage contribution of each control factor is employed to measure the corresponding effect on the electrical conductivity of CB/G/EP composites. It can be seen that factor C (moulding time) put on the biggest percentage contribution (59.98 %) to the electrical conductivity of CB/G/EP composite, followed by factor A (moulding temperature) 17.54 % and factor B (moulding pressure) 13.96 % respectively.

Moulding time has the biggest contribution in obtain high electrical conductivity of CB/G/EP composites because of the mixture of the composites required enough time to solidified (cross-link). This argument also supported by the data in Table 2, where the electrical conductivity of CB/G/EP composites increased gradually from moulding time 60 to 90 minutes.

Table 4. Experimental results of moulding parameters based on L9 (3^3) OA

Variance Sources	Sum of square (SS)	Degree of freedom (DoF)	Variance (V)	Pure SS (SS')	Contribution (ρ)
A	1599.86	2	799.93	1542.27	17.54
B	1286.31	2	642.65	1227.72	13.96
C	5331.60	2	2665.8	5274.01	59.98
Error (*e*)	575.91	20	28.79		8.51
Total Variance (*T*)	8792.68	26			100.00

Higher moulding temperature is required to reduce the viscosity of the mixture to obtain good dispersion and distribution of the conductive fillers within polymer matrix, while a suitable moulding pressure is also required to reduce the contact distance between CB and G within polymer matrix. Good dispersion, distribution and contact distance between conductive fillers within polymer matrix can be increase the electrical conductivity of CB/G/EP composites. Percentage contribution of the combined error moulding parameters of CB/G/EP composites based on ANOVA was 8.51 %. It means that moulding parameters selected were covered 91.49 % to the electrical conductivity of CB/G/EP composite.

Confirmation Experiment of CB/G/EP Composite

In Taguchi method, confirmation experiment is a very important stage to confirm the electrical conductivity of CB/G/EP composite produced. Table 3 show that the optimal moulding parameters obtained were as A_2, B_1, and C_3. Previous results reported by Suherman et al. [17] and Liu et al. [18] used experiment and additive model (equation 3) to confirm the optimum parameters. They found that the value was nearly with the experimental results [17, 18]. Therefore, this study was only used additive model. Since all factors of the moulding parameters were significantly, $\eta_{predict}$ (optimum parameters) can be calculated as:

$$\eta_{predict} = \mathcal{T} + (A_2 - \mathcal{T}) + (B_1 - \mathcal{T}) + (C_3 - \mathcal{T}) \quad (3)$$

$$= 168.50 \text{ S/cm}$$

where $\mathcal{T}$ is mean value of total the electrical conductivity, and A_2, B_1, C_3 are the electrical conductivity of CB/G/EP composites on each significant level. $\eta_{predict}$ value (168.50 S/cm).

Table 5. shows the results of the electrical conductivity and S/N ratio of CB/G/EP composites for initial combination ($A_1B_1C_1$) are 101.68 S/cm and 40.14 dB, respectively. While the optimal combination using additive model ($A_2B_1C_3$), the electrical conductivity and S/N ratio of CB/G/EP composite higher than at initial combination namely, 168.50 and 52.45, respectively. The results showed that the electrical conductivity of CB/G/EP composite at optimal combination can be successfully increasing the electrical conductivity as 66.82%.

Table 5. Comparisons of moulding parameters on the electrical conductivity of CB/G/EP composites

	Level	Electrical conductivity (S/cm)	S/N Ratio (dB)
Initial combination	$A_1B_1C_1$	101.68	40.14
Optimal combination using additive model	$A_2B_1C_3$	168.50	52.45

CONCLUSION

The optimizations of moulding parameters using the Taguchi method for CB/G/EP composites were performed. The main results are as follows:

1. The moulding parameters such us moulding temperature (A), moulding pressure (B), and moulding time (C) were selected as control factors. The experimental run based on the L9 (3^3) OA were carried out.
2. ANOVA results show that the most significant factor in affecting the electrical conductivity of the CB/G/EP composite was moulding time (C) with percentage contribution of 59.98%.
3. The electrical conductivity of CB/G/EP composites obtained was more than 100 S/cm. The value passes to requirement of the U.S DOE target for bipolar plate application.

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