The Influence of Compounding Parameters on the Electrical Conductivity of LDPE/Cu Conductive Polymer Composites (CPCs)

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Abstract. Low-linear density (LDPE) and copper (Cu) were used as main polymer matrix and conductive filler in order to produce electrically conductive polymer composites (CPC). The selection of the matrix and conductive filler were based on their due to their excellence properties, resistance to corrosion, low cost and electrically conductive. This research works is aimed to establish the effect of compounding parameter on the electrical conductivity of LDPE/Cu composites utilising the design of experiments (DOE). The CPCs was compounded using an internal mixer where all formulations were designed by statistical software. The scanning electron micrograph (SEM) revealed that the Cu conductive filler had a flake-like shape, and the electrical conductivity was found to be increased with increasing filler loading as measured using the four-point probe technique. The conductivity data obtained were then analysed by using the statistical software to establish the relationship between the compounding parameters and electrical conductivity where it was found based that the compounding parameters have had an effect on the conductivity of the CPC.

1 Introduction

Polymer composite is a material that made up from two or more constituent polymer matrix which will form a new composite that have better and unique properties by chemically or physically method. Composite material is one of the most widely used because of their adaptability to different situations and easy combination with other materials to achieve specific goals and exhibit desirable properties. Polymeric material are known as insulator material with the electrical conductivity between 10−15 to 10−4 S/cm [1]. However, polymer material can be conductive by utilizing it with conductive filler such as carbon black, carbon fiber and metallic particles and form conductive polymer composites (CPCs) [2]. CPCs has attracted significant academic and industrial interest such as electronic, chemical, space and energy industries [3] for several decades due to their unique physical properties, electrical properties, inexpensive, resistance to corrosion, excellence mechanical properties and easy to process. CPCs can be used in manufacturing of supercapacitors, solar cells, electrochromic devices, electronic interconnect application, sensors, and biomedical applications [4-5]. In CPCs, conductivity was influenced by many factors such as dispersion of conductive filler particles, aggregate structure, amount of conductive filler, structure of fillers, aspect ratio of fillers and filler orientation [6-7]. Other than that, percolation threshold also plays important role in electrical conductivity of CPCs. Percolation thresholds are known as the critical concentration of conductive filler content to establish conductive network path where it can promote the electrical conductivity of the CPCs [8-9]. H. Pang et. al [10] studied on conductive polymer composites with segregated structures. They claimed that the promising strategy to get the low percolation threshold is by forming segregated structure in a conductive polymer composites material.

In a study by Poblete et. al [11], they studied the microstructure, electrical behaviour, and percolation threshold as a function of metal filler concentration on copper-PMMMA nanocomposites. The copper-PMMMA composite were prepared by mixing copper powders of micron size and nanometre size using hot compression moulding. At highest filler volume fractions which is 35 vol%, the composite conductivity rises approximately to 1.5 × 10−3 S/m in Cu-PMMMA (78 nm) and 1.2 × 10−4 S/m in Cu-PMMMA (3.25-4.75 μm).

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Usually low-density polyethylene (LDPE) is used in engineering applications due to the properties of the LDPE that low weight and excellent mechanical strength [12]. In a study by X. Xu et. al they found that addition of carbon black (CB) more than 9 wt.% into LDPE/EVA the resistivity of the composite was subtle and it shows that the conductivity of the LDPE/EVA/CB also has little difference [3].

Design of experiments or known as DOE is refer to the process of planning, designing and analyzing the experiments to ensure that valid and objectives conclusions can be drawn effectively and efficiently. In DOE, randomization, blocking, and replication are the key concept in designing an experiment. It also a computational method that used to explain the value of different processing or material variables and how to monitor the parameter to maximize system performance while optimizing properties. By using this method, the property performance of products can be predicted under all possible conditions within the limits selected for the experimental design.

2 Methodology

2.1 Materials

Low-density polyethylene (LDPE) was used as polymer matrix and copper (Cu) powder was used as conductive filler. LDPE with the grade Titanlene®, LDI300YY from Lotte Chemical Titan (M) Sdn. Bhd., Pasir Gudang, Johor, Malaysia was purchased from Adv. Dynamic Sdn, Bhd., Kulim, Kedah, Malaysia while the Cu powder from Sigma Aldrich was purchased from AR Alatan Sains (K) Sdn. Bhd., Alor Setar, Kedah, Malaysia.

2.2 Compounding process

The LDPE and Cu powder were weighted according to formulations design by the Minitab statistical software. The formulations and factors selected for this work are namely filler loading, rotor speed and maximum barrel temperature as listed in Table 1.

| Code     | Cu loading (wt.%) | Rotor Speed (rpm) | Max Temperature (°C) |
|----------|-------------------|-------------------|----------------------|
| 5%10S120T | 5                 | 10                | 120                  |
| 5%10S140T | 5                 | 10                | 140                  |
| 5%30S120T | 5                 | 30                | 120                  |
| 5%30S140T | 5                 | 30                | 140                  |
| 10%20S130T | 10               | 20                | 130                  |
| 15%10S120T | 15               | 10                | 120                  |
| 15%10S140T | 15               | 10                | 140                  |
| 15%30S120T | 15               | 30                | 120                  |
| 15%30S140T | 15               | 30                | 140                  |

All nine formulations including a middle run were dry mixed prior to compounding process to ensure homogeneous dispersion of filler within the LDPE matrix. This to ensure that both materials can be mixed well and homogeneous during compounding process in an internal mixer. Subsequently, the compounded material will be compression moulded by using hot press machines to produce specimen suitable to be cut according to specific test sample dimension.

2.3 Morphology of conductive filler

Scanning electron microscopy (SEM) was used to determine the particle shape and size of the copper powder. The copper powder was spread on the duct tape with the dimension 1cm × 1cm and coated with a thin layer of palladium to enable and improve the imaging of sample and avoid charging effect. The magnification was set between ×500 to ×2000 with the acceleration voltage at 20kV.

2.4 Electrical conductivity of LDPE/Cu composites

Electrical conductivity of the composites was measured by using 4-point probe machine. The sample was cut into round shape with the diameter and thickness of the sample was 2.3 cm and 0.1 cm.
3 Result and Discussions

Fig. 1. shows the particle shape of the copper (Cu) in flaky-like shape. The particle size of Cu was calculated by using Image J software and from the calculation, the particle size of the Cu particle was 51.05µm. In a work carried out by Yaman et. al [13], they used dendritic shape Cu powder with 75 µm average size as conductive filler and unsaturated polyester resin as matrix. They found that the dendritic shape particle structure has higher surface contact area than spherical and flake structure by SEM images. The conductivity results obtained shows that the conductivity increases as the Cu concentration increased. Nevertheless, the particle size of the flake-like shape Cu used in this work was smaller therefore it was anticipated that the flake-like shape of filler will give better dispersion due to large contact surface area.

Table 2 shows the result for conductivity of LDPE/Cu measured by using 4-point probe. Initially, the virgin LDPE with conductivity of 1.06×10⁻³ S/cm was recorded. Addition of 15 wt% copper within the LDPE matrix in batch 15%30S120T, a higher conductivity was observed where the conductivity measured was 1.484×10⁻¹ S/cm. Nonetheless, the lowest conductivity was recorded for batch 5%10S120T with a value of 7.997×10⁻² S/cm.

| LDPE/Cu       | Electrical conductivity, S/cm |
|---------------|-------------------------------|
| Pure LDPE     | 1.06×10⁻³                     |
| 5%10S120T     | 7.997×10⁻²                    |
| 5%10S140T     | 8.717×10⁻²                    |
| 5%30S120T     | 9.578×10⁻²                    |
| 5%30S140T     | 8.591×10⁻²                    |
| 10%20S130T    | 1.031×10⁻¹                    |
| 15%10S120T    | 1.237×10⁻¹                    |
| 15%10S140T    | 7.388×10⁻²                    |
| 15%30S120T    | 1.484×10⁻¹                    |
| 15%30S140T    | 9.058×10⁻²                    |
Fig. 2. shows the trend of the conductivity for selected LDPE/Cu CPC formulations with different copper loading. The increasing filler loading had resulted in an increased in the electrical conductivity of the composite. The conductivity increases from $7.997 \times 10^{-2} \text{ S/cm}$ to $1.484 \times 10^{-1} \text{ S/cm}$ when copper content increases from 5 wt.% to 15 wt.%. When copper content exceeds 10 wt.%, there was significant increase in the composite conductivity. It was believed that the significant increase was observed due to the dispersion of the Cu particle within the LDPE maybe surpass the percolation threshold thus resulting in a significant increase in electrical conductivity of the CPCs. Percolation thresholds are known as the critical amount filler content added into the matrix where at this point the conductive network path formed inside the composite and it promote the conductivity of the composites. After this point, the conductivity will increase rapidly because of Cu start to form a conductive path through the LDPE matrix.

Fig. 2. Conductivity of LDPE/Cu polymer composite with different copper loading (5, 10, 15 wt%)

The conductivity data of the composites have been further analysed using the statistical software. With the use of the software, the Pareto charts of effects, main effect plot and interaction plot were constructed to establish the relationship between compounding parameters and the electrical conductivity of the LDPE/Cu CPC. The Pareto chart shows the standardized effects for electrical conductivity of LDPE/Cu CPCs. The purpose of the Pareto chart is to identify the magnitude and the important of the factor effects. In brief, any factor bars cross the line on the Pareto chart is considered as significant. It can be observed in Fig. 3., the significant factors were based on a two-way interaction that was a combination between filler loading and screw speed (AB) along with combination of screw speed and maximum barrel temperature (BC). Nevertheless, to combination of factors (AB) exhibited the highest and had a significant effect on the conductivity of the LDPE/Cu CPC compared to other combinations of factors.

Fig. 3. The pareto chart of standardize effect of conductivity for LDPE/Cu composite
The differences between the level means for one or more factors was examined with the aid of main effect plot. The main effect plot shows the different responses on electrical conductivity against different type of factors. As illustrated in Fig. 4, it shows the main effects plot for conductivity of conductive polymer composite (CPCs) materials where the filler loading, and screw speed have the larger main effect compared to the maximum barrel temperature. The higher the filler loading, and screw speed resulted in the higher the mean effect for conductivity. It can be observed that the changes in the factors will affect the conductivity of the LDPE/Cu composites. This finding was in an agreement to the work reported by A.S Luyt et. al [14] on the thermal, mechanical and electrical properties of Cu powder filled with LDPE where it was shown that increasing of copper loading had resulted in an increased in the conductivity of their composite samples.

![Main Effects Plot for Conductivity](image)

**Fig. 4.** The main effect plot for conductivity of LDPE/Cu composite.

Fig. 5. shows the interaction plot for conductivity of CPCs. The interaction plot shows the relationship between one factor and the responses of another factors. In brief, an interaction is present when the change in the response mean from the low to the high level of factor depends on the level of another factor. In or to visualise the interactions, if the lines are parallel to each other, there is no interaction present whilst if the lines are not parallel, there is possibility an interaction present. The interactions between combinations of filler loading and screw speed factors on the electrical conductivity of LDPE/Cu CPC have had significant interactions among other combinations of factors. Thus, this explains whereby the dispersion of filler can be optimised with the higher screw speed which will resulted in the better dispersions of filler loading within the LDPE matrix.
Fig. 5. Interaction plot for conductivity of the LDPE/Cu CPCs.

4 Conclusion

The recent works investigates the effect of compounding parameters on the electrical conductivity of LDPE/Cu CPCs. The CPCs were melted blended using an internal mixer according the formulations designed in the DOE studies. The SEM micrographs reveals a flake-like shape of Cu filler, and it was found that the conductivity of the LDPE/Cu CPC were increased as the loading of the filler increase. It can be observed that the optimum formulation with highest conductivity was achieved for sample 15%30S120T with the conductivity value of $1.484 \times 10^{-1}$ S/m whereas the lowest value observed is with 5%10S120T with conductivity $7.997 \times 10^{-2}$ S/m. Moreover, it was established from the DOE analysis where the Pareto chart of effects, Main effect plot and Interaction plot shows that the combinations of factors have had significant impact on the electrical conductivity of the LDPE/Cu composite.

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