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TENSILE PROPERTIES AND ELECTRICAL CONDUCTIVITY OF LINEAR LOW-DENSITY POLYETHYLENE (LLDPE)/CARBON BLACK CONDUCTIVE POLYMER COMPOSITES (CPCS) : EFFECT OF COMPOUNDING PARAMETERS

In recent years, the research and development in conductive polymer composites (CPCs) had gained considerable interests in both industry and academia as potential materials for electronic interconnects. These composites require to have the ability to conduct electric while maintaining sufficient flexibility while withstanding the bending, twisting, or stretching during service. To achieve the desired composite properties, the processing method and the parameters involved plays important role and ought to be investigated. In this study, the effect compounding parameters on the preparation of linear-low density polyethylene/carbon black (LLDPE/CB) polymer composite were carried out. Factors namely filler loadings, screw speed and maximum barrel temperatures were selected and their effects on the tensile properties and conductivity were analyzed in this research. It was observed that the increasing of filler loadings from 5 wt.% to 10 wt.% has increased the electrical conductivity from 1.11×10^{-2} S/m to 1.46×10^{-2} S/m. The pareto chart shows that the filler loading was important factors to the result of composite conductivity. Moreover, the main effect plot shows that the filler loading has the highest mean effect on conductivity as it is important for the formation of conducting path in composite. It was also established that the pareto chart also shows that filler loading and barrel temperature have the highest significant effect on LLDPE/CB polymer composite tensile properties. The changes in the combinations of factors affect the tensile properties as revealed by the main effect plots for LLDPE/CB CPCs.

Keywords: Mechanical Properties; Conductive filler; Conductive Polymer Composite; Polymer Processing; Compounding

1. Introduction

Polymer composites are polymer materials with a reinforcement, wherein the polymer acts as a matrix that penetrates the reinforcement bundles and bonds to the reinforcement. Composites offer properties and overall performance characteristics, together with better stiffness, strength, impact resistance, heat resistance, abrasion, and put on resistance. The gas barrier cannot be attained by using the matrix polymer in the absence of inclusions [1]. Conductive polymer composites (CPCs) are mainly composed of conductive fillers with high conductivity and insulating polymer matrices. Recently, potential conductive polymer composites have been suggested because of benefits such as presenting metal-like electrical and semiconductor-like optical characteristics. Many different things were done with the CPCs, such as in biomedicine and computing, as well as with electronics, energy storage, and sensors [2].

Conductive fillers are embedded to the polymer matrix to enhance the electrical conductivity of the composites. Nevertheless, due to their rigidity or stiffness and strong adhesion with the polymeric matrix, filler tends to increase the modulus of the matrix [3]. LLDPE (Linear-Low Density Polyethylene) is a thermoplastic polymer which has a lot of short branches. Moreover, due to these fewer and shorter branches, the chains may move against one other during elongation without getting entangled, in comparison with the LPDE, which has longer branched chains that can become entangled. LLDPE has higher tensile strength and high impact and puncture resistance than LDPE. The LLDPE and LDPE have a density of 0.91-0.94 g/cm^3. The only polymers with lower specific gravity than water is LLDPE and LDPE [4].

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Carbon black (CB) is a group of small, primarily amorphous carbon particles that have been formed into aggregates of different sizes and shapes. It comes in various commercial grades with variable primary particle sizes, aggregate size and shape, porosity, surface area, and chemistry. The process type and parameters may accurately control the properties [5-7].

This research aims to use a Design of Experiment (DOE) to the study the effect processing parameters on the mechanical and electrical properties of the CPSs. A statistical software is used in this study to investigate the design of different parameters. This software helps investigate factor variable's effect on output responses [8]. The polymer matrix and carbon black will be compounded using an internal mixer and resulting composites were subject to characterization and testing such as tensile test and four-point probe. The effect of parameters was analysed using pareto charts, main effect plots and interaction plot as the outcome of the present work.

2. Methodology

2.1. Materials

The polymer matrix used in this study wa a linear-low density polyethylene manufactured and provided by PT. Lotte Chemical Titan Nusantara, SDS Code: PE-002, Grade Name: LL0209SR. The carbon black powder (Carbon Black Acetylene, 100% compressed, 99.9% S.A, 75 m^2/g, bulk density 170-230 g/L) was supplied by Alfa Aesar, France.

2.2. Composite Preparations

In this research, the LLDPE and carbon black powder were weighted according to the formulations. The materials were then dry mixed to ensure homogeneous mixing. A DOE study was conducted for the compounding process using an internal mixer where the factors of interest namely screw speed, barrel temperature and filler loading were selected. A total of nine runs (including a middle run) were formulated as listed in TABLE 1. The polymer composites were then compression mould to obtain samples suitable to cut into the desired test samples. The compression moulding was carried out at 15 Pa, 140°C and 3 minutes preheating time and 7 minutes for full compression.

2.3. Characterization and Testing

A 4-Point probe, model Keithley 6221 AC/DC source and Keithley 2182A nanovoltmeter four-point probes was used to measure the conductivity and resistivity value of the polymer composite. A total of five samples for each formulation were tested at a 50 mm/min crosshead speed in accordance to ASTM D638 tensile test performed using an Instron 5569 tensile machine.

TABLE 1

DOE formulations of LLDPE/CB CPCs (Full fraction factorial design)

LLDPE/CB Filler Loading	Formulations (CB loading, Blade speed, Processing Temperature)
S1	5% 20S 120T
S2	5% 30S 120T
S3	5% 20S 140T
S4	5% 30S 140T
S5	10% 20S 120T
S6	10% 30S 120T
S7	10% 20S 140T
S8	10% 30S 140T
S9	7.5% 25S 130T

3. Result and discussion

3.1. Electrical conductivity of LLDPE/CB CPCs

TABLE 2 tabulated the electrical conductivity data for LLDPE/CB measured using 4-Point probe. In general, it was found that the electrical conductivity of the composite increased with increasing CB content. With reference to Fig. 1, the S7 sample showed the highest conductivity value of 1.46×10^{-2} S/cm whereas

TABLE 2

The electrical conductivity of LLDPE/CB composites

LLDPE/CB Filler Loading	Electrical Conductivity, S/cm
S1	1.10×10^{-2}
S2	1.23×10^{-2}
S3	1.30×10^{-2}
S4	1.33×10^{-2}
S5	1.38×10^{-2}
S6	1.40×10^{-2}
S7	1.46×10^{-2}
S8	1.43×10^{-2}
S9	1.37×10^{-2}

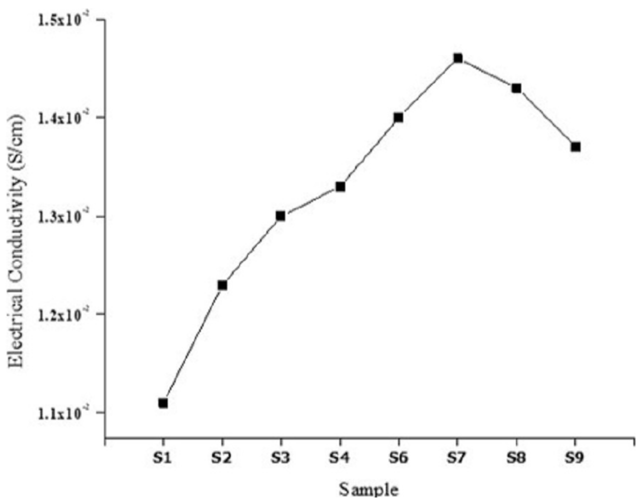

Fig. 1. The effect of CB loading on the electrical conductivity of LLDPE/CB polymer composites

for formulation S1, the lowest conductivity value of 1.11×10^{-2} S/cm was recorded. The conductivity increases from 1.11×10^{-2} S/cm to 1.46×10^{-2} S/cm when carbon black content increases from 5 wt.% to 10 wt.%. It was believed that the increase in conductivity of the CPCs was primarily attributed to the uniform dispersion of CB within the LLDPE matrix. Moreover, the dispersion of CB particles created a conductive network path that permits electrons to flow between particles and become significant as the filler loading increased. The findings of the electrical conductivity in this study were in agreement with reported by other researcher where a uniform dispersion and distribution of carbon black created continuous pathways in the LLDPE matrix that resulted in an increase of electrical conductivity of the CPCs [9].

3.2. DOE Analysis – Effect of compounding parameters on CPC conductivity

Based on Fig. 2(a), the Pareto chart shows the effects of compounding parameters on the electrical conductivity of LLDPE/CB composites. The purpose of the Pareto chart is to study the significant effect and the interaction between factors with the samples. From Fig. 2, the filler loading factor A has the significant effect on the conductivity of the CPCs followed by factor B namely maximum barrel temperature. Fig. 2(b) shows the main effects plot for the CPCs. It is worth to note that the main effect plot measures the interdependent of a variable on other process variables has on the response being measured. The graph shown in Fig. 2(b) shows that the filler loading has a larger main effect than screw speed and maximum barrel temperature. The higher the filler loading, the higher the mean effect for conductivity. Furthermore, it can be observed that by changing each factors affects the conductivity of the CPC samples. The interaction plot in Fig. 2(c) shows the interaction plot for LLDPE/CB CPCs. These plots show the interaction effect of two factors on the response and compare to the relative strength of the effects. Clearly, it can be seen the interaction between screw speed and maximum barrel temperature plot showed a significant combination of a two-way interactions that affect the composite conductivity. This was believed due to that screw speed determines how well the CB filler dispersed in LLDPE matrix whereas, the maximum barrel temperature effects to flowability of the polymer matrix melts which also facilitate the dispersion of conductive filler during compounding.

3.3. Tensile Properties of LLDPE/CB CPCs

Fig. 3(a) shows the effect of CB loading on the tensile strength of LLDPE/CB polymer composite with different CB loading and compounding parameters. The tensile strength of LLDPE with the addition of 5 wt.% CB recorded maximum value is 30.67 MPa for sample compounded with 20 rpm screw speed and 140°C barrel temperature as the parameter, decreases when

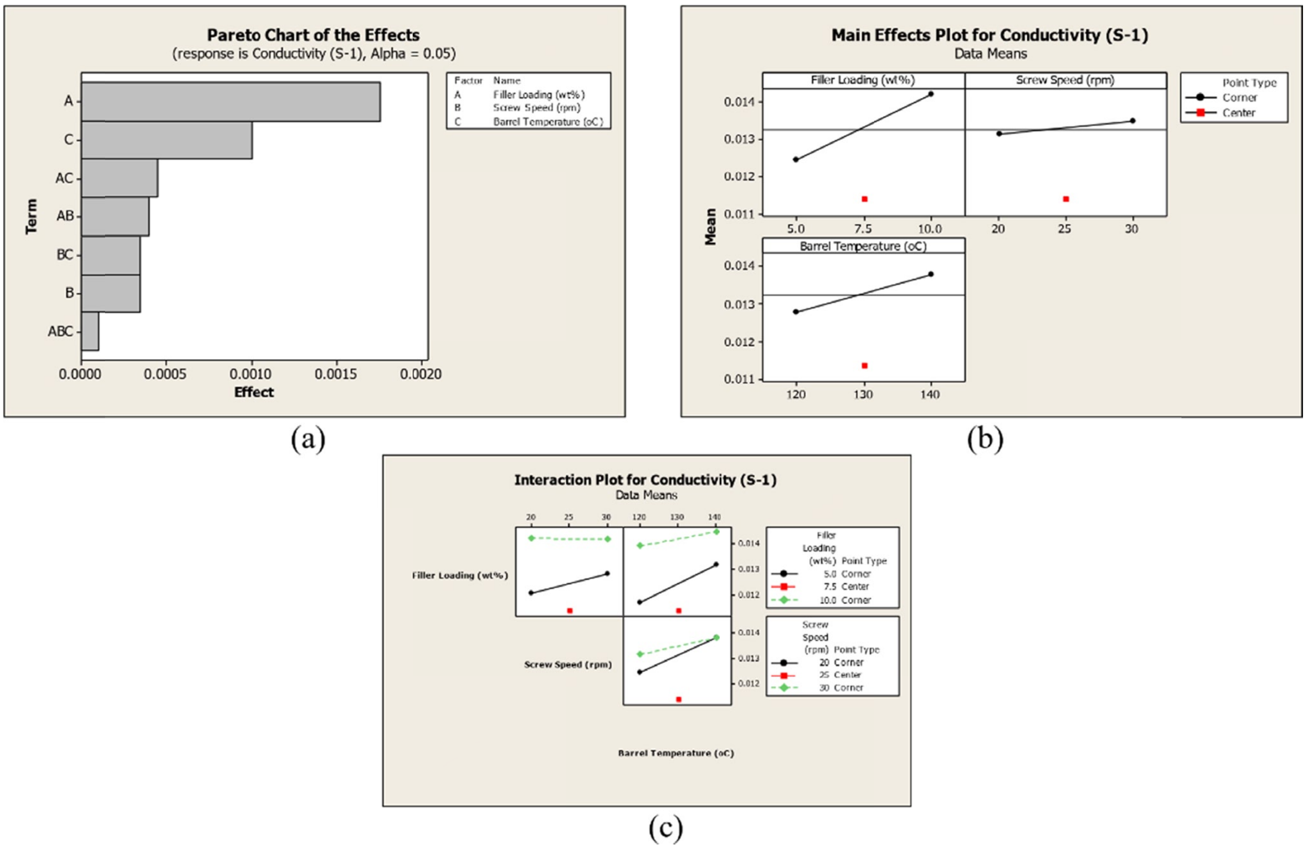

Fig. 2. (a) The pareto chart of electrical conductivity for LLDPE/CB polymer composites, (b) The main effect plots of conductivity for LLDPE/CB and (c) Interaction plot for conductivity of the LLDPE/CB CPCs

CB content is more than 5 wt.%. This indicates that the optimum carbon black loading in LLDPE was at 5 wt.% CB loading. The degree of reinforcement of LLDPE composites increases with filler loading and the extent of polymer-filler interaction. The reduction of tensile strength beyond 5 wt.% loadings can be explained by particle agglomeration at higher CB content [10,11]. Nevertheless, as the CB loading increases to 7.5 wt.%, it can be observed that there were decreased in tensile strength to 22.20 MPa. Further increase in CB loading to 10 wt.% resulted in reduction of the tensile strength of the CPCs. The changes in the screw speed and maximum barrel tempatures hat detrimental effect toward the tensile strength of the CPCs. It was believed when filler loading increases to 10 wt.%, a combination of filler agglomerations as well as the weak interfacial area between filler and matrix were the primary reason for the reduction in the tensile strength observed [12,13]. Fig. 3(b) shows the influence of CB loading on the modulus of elasticity of all sample LLDPE/CB polymer composite. In general, addition resulted in an increase of the CPC tensile modulus. A change of distortion performance from ductile to more brittle occurred at higher CB loadings. As the loading of CB increases in the LLDPE matrix, some portions of the polymer are trapped inside the filler network, which increases the effective volume ratio of the solid particles in the composite [14,15]. The modulus of elasticity with the addition of 7.5 wt.% CB, had increased the modulus to 192.8 MPa. At higher CB loadings, the 10 wt.% CB filler additions caused the modulus of elasticity of the polymer composite rose up to 201.06 MPa. It was believed that the tensile modulus of the polymer composite increases due to CB is stiffer than LLDPE. Young modulus is an indication of the relative stiffness of composites. The filler enhances the composites' rigidity and reduces the elongation at break [16,17]. Fig. 3(c) shows that elongation at break of the LLDPE/CB CPC was a critical factor explaining the ductility of the composite samples. The graph shows that the CB loading at 5 wt.% recorded an elongation at the break value of 806%. A further increase in the CB loading to 7.5 wt.% CB caused the elongation at break of the composite to decreased to 453%. A further increase of filler loadings also resulted in the decrease of the toughness properties of the composite samples. The decrease in the elongation at break can be primarily attributed to the filler agglomeration within the polymer matrix.

3.4. DOE Analysis – Effect of compounding parameters on CPC tensile properties

The Pareto chart illustrated in Fig. 4 shows the standardized effects for tensile properties for LLDPE/CB CPCs that determines the magnitude and the important of the effects. The significant factors for tensile strength and elongation at break were the combinations between filler loadings and maximum barrel temperatures. In the case of tensile modulus, the com-

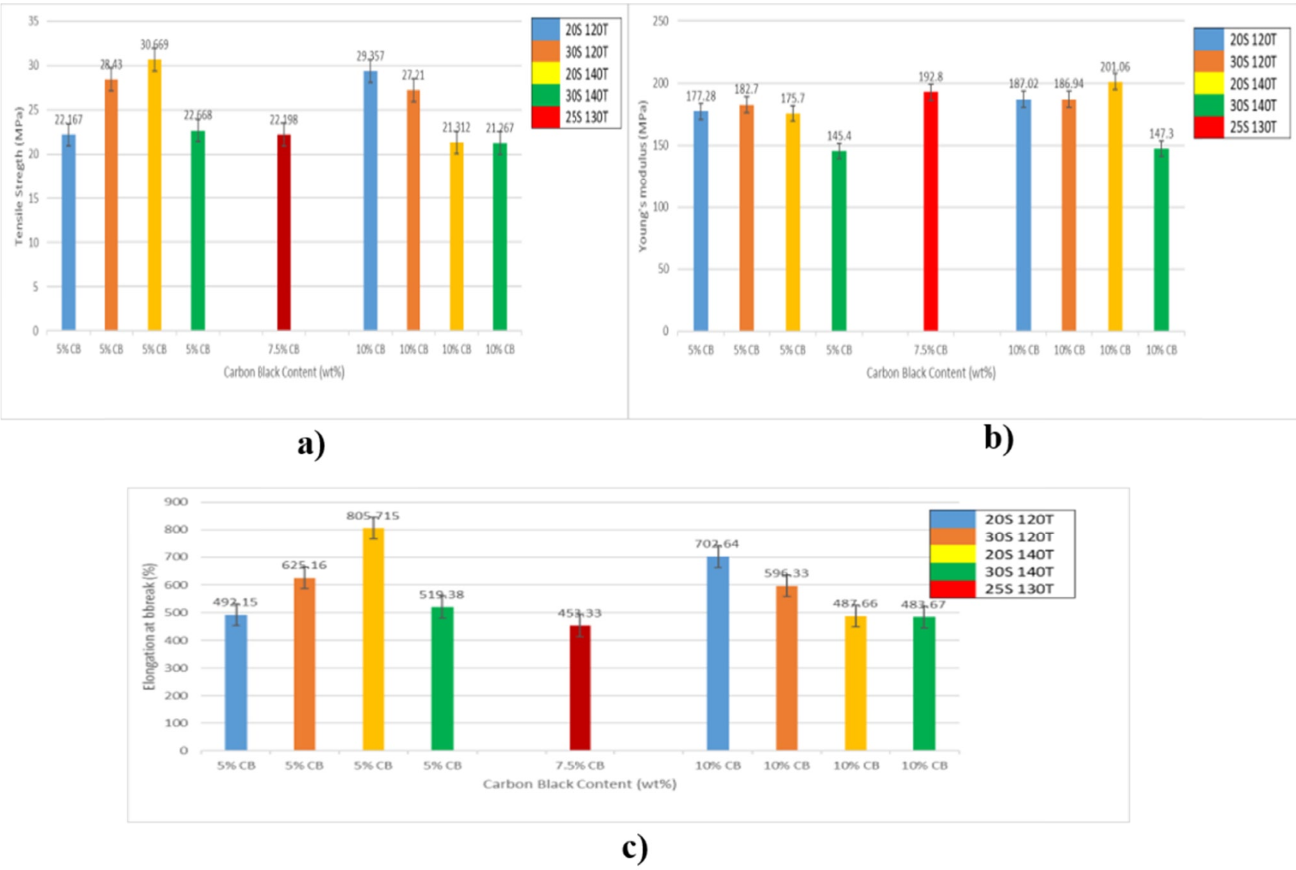

Fig. 3. The effect of CB within LLDPE on the (a) tensile strength, (b) Young's modulus and (c) elongation at break at different CB loading and compounding parameters

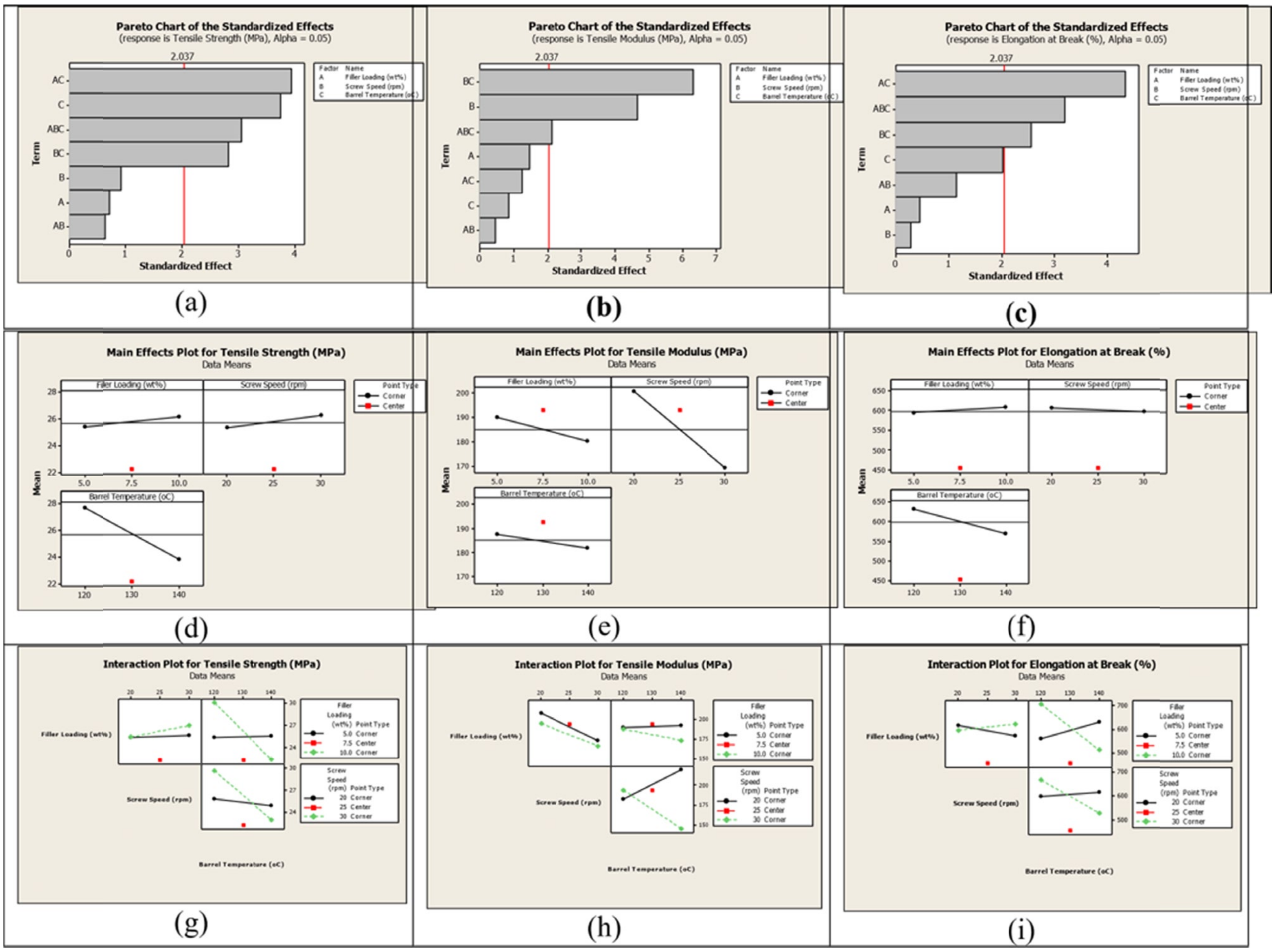

Fig. 4. (a-c) The pareto chart of tensile properties for LLDPE/CB polymer composite: (d-f) The main effect plot of tensile properties for LLDPE/CB: (g-i) Interaction plot for tensile properties of LLDPE/CB CPCs

bination of factors between screw speed and maximum barrel temperatures were found to be significant. Regarding the main effect plots, the changes in the factors have had significant effects on the tensile properties. As can be observed in Fig. 4(b) although increasing the filler loading and screw speed will result in an increase in tensile strength, it was not the case for the maximum barrel temperature. The increase in barrel temperature resulted in the decrease of tensile strength. Similar trends were found on main effect plots for elongation at break of LLDPE/CB CPCs. Furthermore, the interaction plots obtained from the DOE analysis for tensile properties shows that there were interactions occurred between all factors that affect the mechanical properties of the polymer composites.

4. Conclusion

In this work, the effect of compounding parameters on the tensile and electrical conductivity of LLDPE/CB CPCs was investigated and the conclusion is as follows:

1. Additions of CB conductive fillers resulted in an increase in the conductivity of the samples by up to 1.46×10^{-2} S/cm as can be observed for sample S7. The filler loading was determined as the primary factor that contributes to increase in electrical conductivity from the Pareto chart of the standardized effect. Changes in the filler loadings, screw speed and maximum barrel temperature affect the electrical conductivity of the composites as shown by the main effect plots.
2. At lower CB loadings, the tensile strength was found to increased, whereas, at higher loadings, the tensile strength was found to decrease. In the case of the tensile modulus, the modulus was increased as CB loading increases. Nevertheless, the elongation at break was severely affected with the additions of CB within LLDPE matrix.
3. Overall, it was shown that the combinations of factors and changes in the compounding parameters have had significant impact on the tensile properties of LLDPE/CB as determined by the DOE analysis.

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REFERENCES

[1] A. Malas, Rubber nanocomposites with graphene as the nanofiller. In Progress in Rubber Nanocompos., Woodhead Publishing, 179-229 (2017).

[2] A. Saad, K. Jlassi, M. Omastová, M.M. Chehimi, Clay/conductive polymer nanocomposites. Clay-Polym. Nanocompos, Elsevier, 199-237 (2017).

[3] S.Y. Fu, X.Q. Feng, B. Lauke, Y.W. Mai, Effects of particle size, particle/matrix interface adhesion and particle loading on mechanical properties of particulate-polymer composites. Composites Part B: Engineering **39** (6), 933-961 (2008).

[4] https://www.usplastic.com/knowledgebase/article.aspx?contentkey=508

[5] M.E. Spahr, R. Rothon, Carbon black as a polymer filler. Polymers and polymeric composites: a reference series, 2016 Springer, Berlin, Heidelberg.

[6] Z Borsos, P. Paun, I.C. Botez, C.M. Stoica, P. Vizureanu, M. Agop, Structural conductivity of carbon nanotubes. Revista de Chimie **59** (10), 1169-1171 (2008).

[7] P. Vizureanu, N. Cimpoesu, V. Radu, M. Agop, Investigations on thermal conductivity of carbon nanotubes reinforced composites. Experimental Heat Transfer **28** (1), 37-57 (2015).

[8] https://www.minitab.com/content/dam/www/en/uploadedfiles/documents/getting-started/MinitabGettingStarted_EN.pdf

[9] Q. Yuan, S.A. Bateman, D. Wu, Mechanical and conductive properties of carbon black-filled high-density polyethylene, low-density polyethylene, and linear low-density polyethylene. J. Thermoplast. Compos. Mater. **23** (4), 459-471 (2010).

[10] A. Chatterjee, B.L. Deopura, Thermal stability of polypropylene/carbon nanofiber composite. J. Appl. Polym. Sci. **100** (5), 3574-8 (2006).

[11] J.Z. Liang, Q.Q. Yang, Mechanical properties of carbon black-filled high-density polyethylene antistatic composites. J. Reinf. Plast. Compos. **28** (3), 295-304 (2009).

[12] M. Yasar, G. Bayram, H. Celebi, Effect of carbon black and/or elastomer on thermoplastic elastomer-based blends and composites. AIP Conf. Proc. **1664** (1), (2015).

[13] H.S. Yang, H.J. Kim, H.J. Park, B.J. Lee, T.S. Hwang, Effect of compatibilizing agents on rice-husk flour reinforced polypropylene composites. Compos. Struct. **77** (1), 45-55 (2007).

[14] H.F.M. Mohamed, H.G. Taha, H.B. Alaa, Electrical conductivity and mechanical properties, free volume, and γ-ray transmission of ethylene propylene diene monomer/butadiene rubber composites. Polym. Compos. **41** (4), 1405-17 (2020).

[15] A.Z. Vand, Development of Polymer Nanocomposite Films and Their Potential for Photovoltaic Cell Applications. Phd thesis, Université De Montréal (2014).

[16] Y. Kanbur, Z. Küçükyavuz, Electrical and mechanical properties of polypropylene/carbon black composites. J. Reinf. Plast. Compos. **28** (18), 2251-2260 (2009).

[17] H. Salmah, A. Romisuhani, H. Akmal, Properties of low-density polyethylene/palm kernel shell composites: Effect of polyethylene co-acrylic acid. J. Thermoplast. Compos. Mater. **26** (1), 3-15 (2013).