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THE EFFECT OF CARBON BLACK LOADINGS ON THE ELECTRICAL PERCOLATION THRESHOLD OF LINEAR LOW-DENSITY POLYETHYLENE (LLDPE) AND LIQUID SILICONE RUBBER (LSR) CONDUCTIVE POLYMER COMPOSITES

The recent advancement in the flexible electronic devices have led to a growing interest around conductive polymer composites (CPCs). In this present study, a Linear Low-Density Polyethylene (LLDPE)/Carbon Black (CB) conductive polymer composites and Liquid Silicone Rubber (LSR)/CB conductive polymer composites were made by melt compounding and mixing technique, respectively. The CB serves as conductive filler enhances the performance of LLDPE and LSR by improving their electrical conductivity in the composite systems. In this study, various CB loadings (2 wt.%, 4 wt.%, 6 wt.%, 8 wt.%, and 10 wt.%) had been incorporated within LLDPE and LSR matrices respectively. The CPCs were melt blended using an internal mixer for LLDPE/CB composite, whereas, for LSR/CB composite, a stir mixing technique was employed. In general, additions of CB within LLDPE and LSR matrices had affected the tensile properties of the composites. Nevertheless, it was found that the electrical conductivity for both CPCs were increased with increasing of CB loading. The SEM micrographs revealed the morphology of a fractured CPCs samples. Formation of a network path was believed to be the primary reasons for the increased in electrical conductivity of both composites systems and it was anticipated that the percolation threshold for both CPCs was at 6 wt.% loading.

Keywords: Mechanical Properties; Conductive Polymer Composite; Percolation Threshold; Flexible composite

1. Introduction

CPCs have a broad research field and have piqued the interest of many researchers and professionals. Conductive polymer composites were introduced as an intriguing new class of electronic materials in 1977, and enormous research has been conducted in the subsequent several decades, resulting in diverse applications in various conducting materials. Polymers are becoming a prominent area of scientific interest for scientists. CPCs are appealing due to their electrical characteristics, which allow for a wide range of electrical conductivity to be achieved through doping while maintaining mechanical and thermal stability. CPCs have numerous advantages over non-conducting polymers, owing principally to their electrical characteristics [1]. In recent years, CPCs were employed in a variety of applications such as electronic devices, energy storage, and sensors.

It is the most essential principle of percolation to regard the conductor unit as a single point with no structure and resistors connecting each point. The percolation threshold is the filler concentration at which an extrinsically conducting polymer composite system changes from an insulating to a conducting condition. The type, shape, and size of filler particles, as well as their dispersion and distribution within the composite, all influence electrical percolation in extrinsically conductive composites [2]. The development of CPCs is tempting owing to their electrical features, which allow for a wide range of electrical conductivity to be obtained by doping while preserving mechanical and thermal stability, flexibility may not be attained. Nonetheless, due to their unique electrical properties, CPCs have various benefits over non conducting polymers.

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2. Methodology

2.1. Material

The main material is LLDPE, LSR and CB. The LLDPE was obtained from the Chemical Titan (M) Sdn. Bhd. The LSR was obtained from the Clay Art Sdn. Bhd. The conductive filler CB powder (Carbon Black Acetylene, 100% compressed), used in this study has an average particle size of 86 µm and supplied by Alfa Aesar, France.

2.2. Composite preparation

The LLDPE/CB CPCs were prepared through melt blending technique using Z-blade internal mixer. The LLDPE and CB were dry mixed prior melt blended in the internal mixer at 140°C for 15 minutes with a rotor speed was at 20 rpm. The compounded LLDPE/CB CPCs were then hot pressed by using compression moulding at temperature of 140°C to produce compressed CPC samples. In the case of LSR/CB CPCs, the composites were prepared by stir mixing of silicone rubber resin with conductive filler at room temperature. The resulting mix were then poured into a mould and left in oven for 30 minutes at 85°C for curing process to complete. Both LLDPE/CB and LSR CB CPCs were then cut into suitable size and shape for testing and characterisations using a dumbbell cutter. The formulations used for composite preparations of both CPCs are as tabulated in TABLE 1.

TABLE 1

The LLDPE/CB and LSR/CB composites formulations

Sample No.	LLDPE/CB CPCs		LSR/CB CPCs	
	LLDPE (wt.%)	CB (wt.%)	LSR (wt.%)	CB (wt.%)
1	100	0	100	0
2	98	2	98	2
3	96	4	96	4
4	94	6	94	6
5	92	8	92	8
6	90	10	90	10

2.3. Testing and characterisations

2.3.1. Mechanical analysis

The tensile testing for LLDPE/CB and LSR/CB CPCs was carried out on Instron 5569 in accordance to ASTM D638 and ASTM D412 [3]. A total of 5 samples for each batch of composites were tested at a crosshead speed of 10 mm/min for LLDPE/CB composite, whereas, for LSR/CB CPCs, the crosshead speed of 500 mm/min was used.

2.3.2. Electrical Analysis

LLDPE/CB composite and LSR/CB composite were cut into several test samples in the shape of a 20 mm diameter with 1 mm thickness. The electrical conductivity of CPCs was measured using a four-point probes namely Keithley 6221 AC/DC source and Keithley 2182A nanovoltmeter at 10 V of voltage.

2.3.3. Morphology Analysis

A Jeol JSM-6460LA SEM was used to investigate the morphology of the CPCs by means of scanning electron microscope. The samples were prepared in 1 mm × 1 mm dimensions and coated with platinum using a sputter coater to prevent charging on the polymeric sample surfaces. The sample microstructures were examined using magnifications of 500× and 1000× at 20 kV voltage.

3. Results and discussion

Fig. 1 shows the tensile strength of LLDPE/CB and LSR/CB composites with different CB loadings. It can be observed that the addition of CB resulted have an effect on the tensile strength for both CPCs. In general, additions of CB in LLDPE matrix resulted in an increase of tensile strength by up to 23.73% as can be observed for sample with 8 wt.% CB loading. Nevertheless, this was not the case for LSR/CB CPCs. It was found that the tensile strength of the LSR/CB composites decreases as the amount of CB loadings increases. The difference observed in the tensile strength for both polymer matrices could probably attributed to the structure of the polymer matrices. It is known that LLDPE is a thermoplastic polymer with a semi crystalline structure, whereas; LSR is a thermoset silicone rubber elastomer with a crosslink structure may explain the tensile behaviour ob-

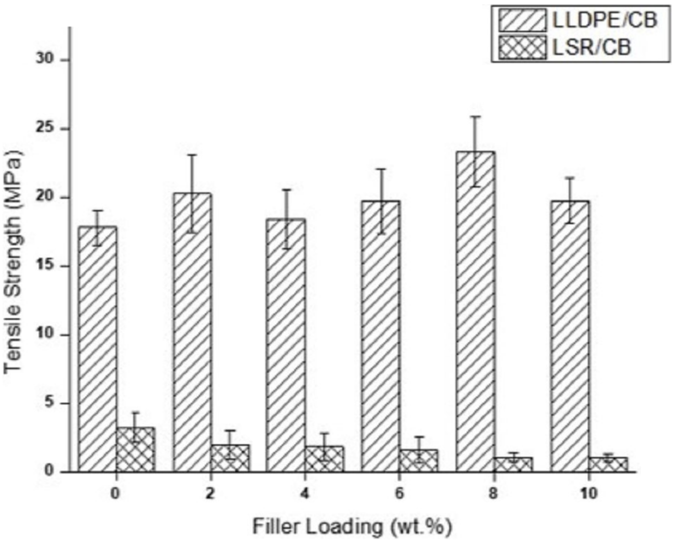

Fig. 1. The effect of CB loadings on the tensile strength of LLDPE/CB and LSR/CB composite

served. Furthermore, additions of stiff, agglomerated particles provide concentrated stress sites, reducing stress transmission from polymer chains to carbon particles which can result in reduction in the tensile strength of the composite samples [4]. In the present work, it was believed due to poor dispersion and agglomerations of conductive fillers during processing had affect the tensile properties of the polymer composites.

The elongation at break for LLDPE/CB and LSR/CB composites is illustrated in Fig. 2. Clearly, the trends recorded showed that elongation at break was affected due to CB additions. It can be observed that the elongation at break was decreased as the loading of CB increases. In the case of LLDPE/CB composite, although there was a slight difference in the elongation at break between 2 wt.% to 6 wt.%, there were noticeable decrease in elongation at break for sample with 8 wt.% and 10 wt.% CB loading. Nonetheless, there was a significant drop observed for LSR/CB composites. It can be seen at 4 wt.% CB loading, the elongation at break was decreased by up to 32% compared to pure LSR. Additions of CB had affected the toughness properties for both composite systems. This can be primarily attributed to the weak interaction between the filler and the polymer matrices that led to a decrease in the elongation at break observed. Moreover, the conductive filler may hinder the mobility of the polymer chains which can affect the mobility or deformation of the polymer the matrices [5,6].

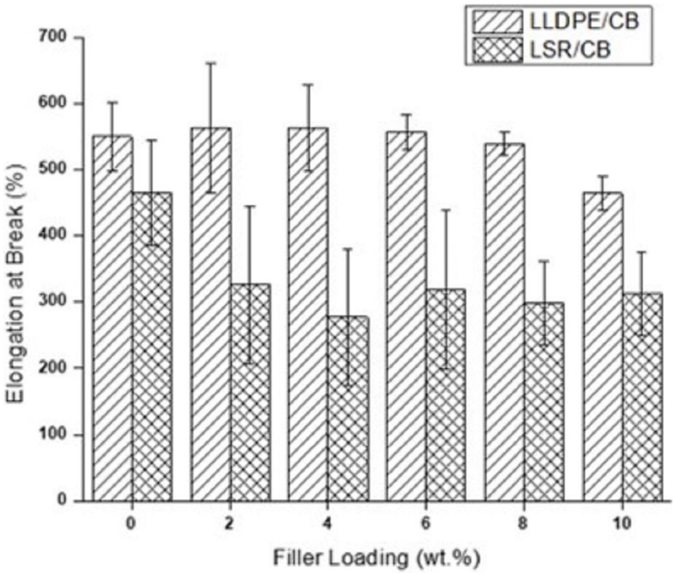

Fig. 2. The effect of CB loadings on the elongation at break of LLDPE/CB and LSR/CB composite

Fig. 3 shows the modulus of elasticity or young's modulus of LLDPE/CB composites with varying CB loading. The modulus for LLDPE/CB CPCs was significantly increased due to CB additions. Similarly, this trend can also be observed for LSR/CB composite with filler loading up to 4 wt.%. Beyond 4 wt.% of CB loading, the modulus was found to decrease and it was believed due to filler agglomeration and poor interaction between filler and LSR matrix. In general, it is widely accepted that addition of stiffer filler into a soft polymer matrix will increase the stiffness of the matrices. The increasing stiffness as can be observed for composites samples will also affect the toughness properties of the CPCs.

Fig. 4 shows the electrical conductivity findings of filler loading in LLDPE and LSR composites. From Fig. 4, it shows that as the CB filler loading increase, the electrical conductivity also increased. The addition of CB at low loading had increased the electrical conductivity approximately by 4% compared to pure LLDPE. As the CB loading was increased up to 10 wt.%, LLDPE/CB composite electrical conductivity was increased from 1.65×10^{-2} S/cm to 1.98×10^{-2} S/cm. For pure LSR, the electrical conductivity was recorded at 2.27×10^{-2} S/cm. The electrical conductivity was increased at 2 wt.% and 4 wt.% approximately by 9% and 11%, respectively. However, addition of CB at 6wt.% shows that the electrical was slightly decreased from 2.53×10^{-2} S/cm to 2.51×10^{-2} S/cm compared to LLDPE with 4 wt.% of CB. However, addition at highest CB loading which was at 10 wt.% showed that the electrical conductivity was increased by 18% compared to pure LLDPE. As CB loading increases, the resultant resistance decreases, followed by a large

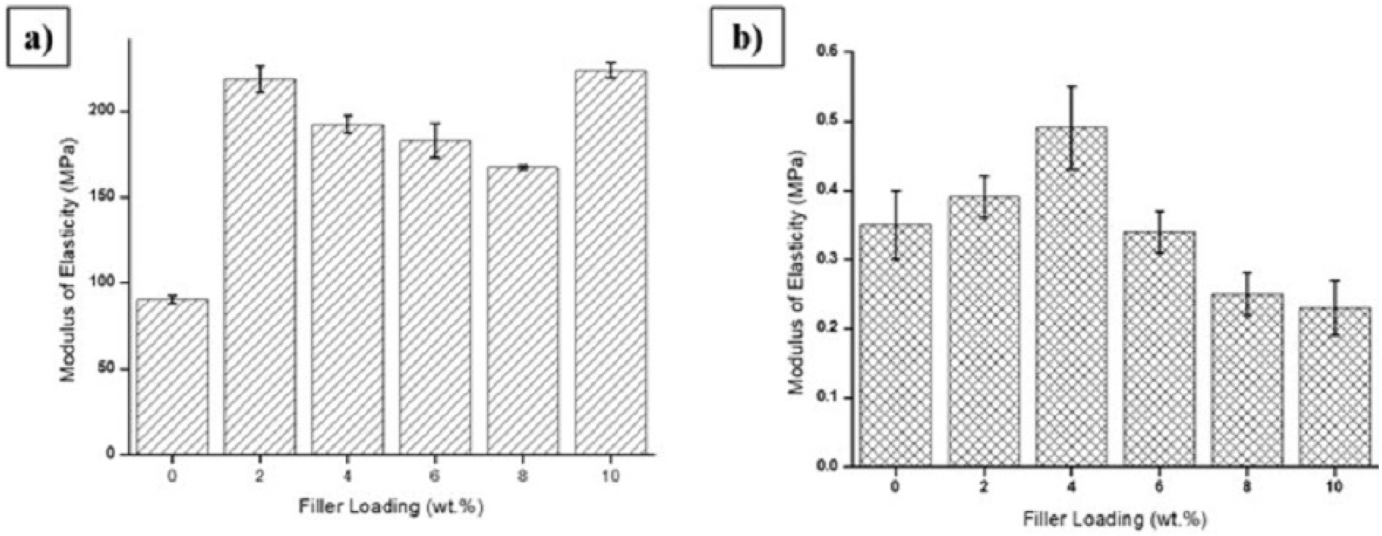

Fig. 3. The effect of CB loading on the Young's modulus of (a) LLDPE/CB and (b) LSR/CB composite

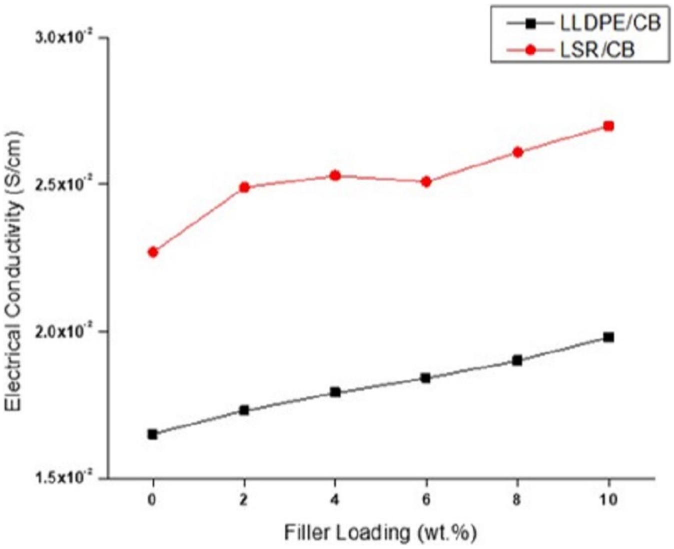

Fig. 4. The effect of CB loading on the electrical conductivity of LLDPE/CB and LSR/CB composites

increase in conductive pathways dominated by electron hopping. The electrical conductivity was very low at low CB loadings and gradually increases as the conductive network route was formed with increasing CB concentration. The electrical conductivity CB content may be influenced by agglomeration of the conductive fillers [7-9]. It was believed that the increase in conductivity can be probably attributed to formation of network path within the CPCs where percolation threshold for both LLDPE and LSR CPCs was at the range of 6 wt.% CB loading [10-12].

Fig. 5 shows the SEM micrographs of LLDPE/CB and LSR/CB at 2 wt.% and 10 wt.%. It can be observed that the CB conductive filler was well dispersed within both LLDPE and LSR matrices. Nevertheless, there was also presence of larger CB particles presents as highlighted in the micrographs which was believed due to agglomeration of CB particles. The presence of CB particles alters the morphology of the composites which affect the mechanical and electrical conductivity as discussed previously [13-15].

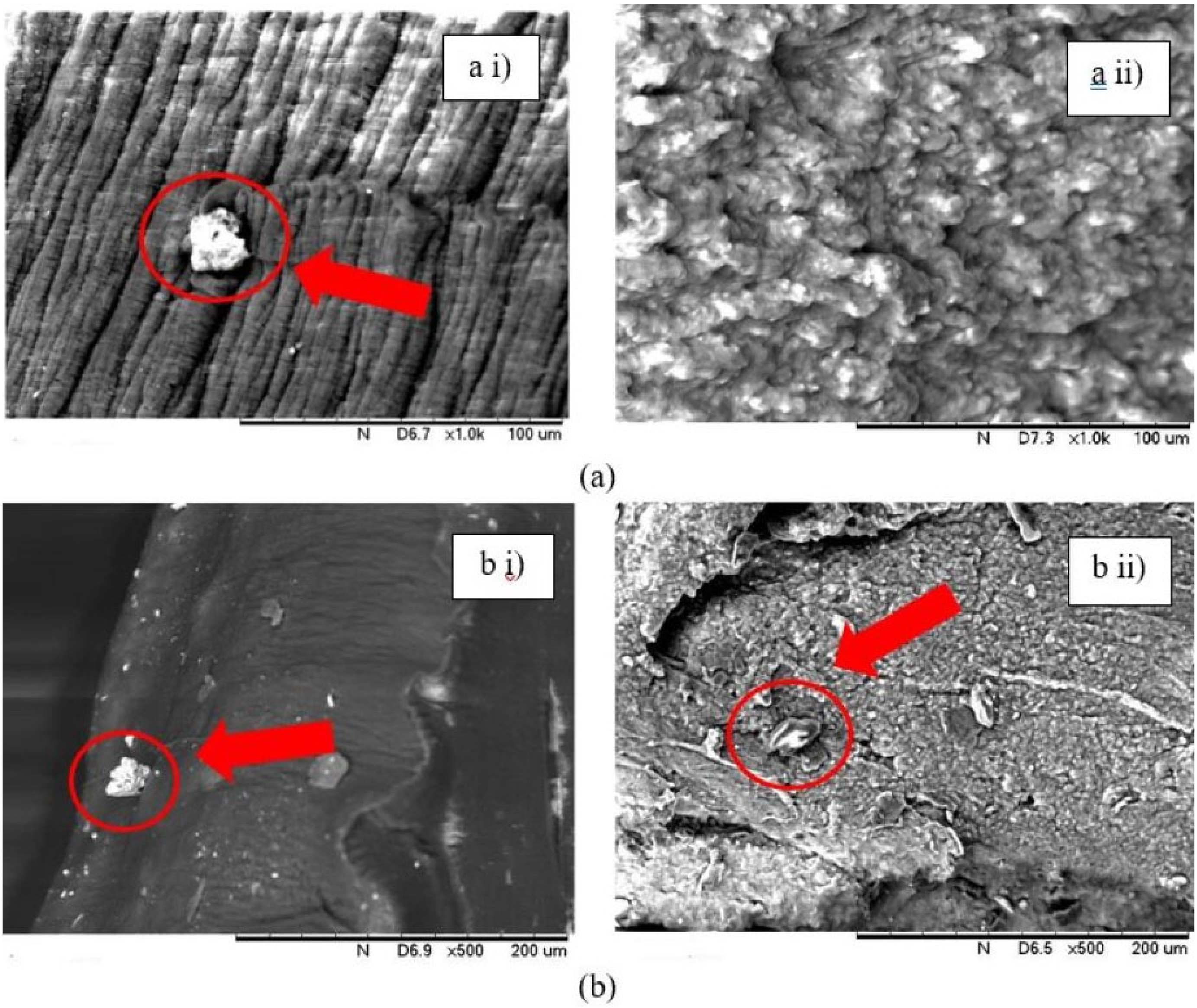

Fig. 5. Surface and fracture morphology of (a) i) LLDPE/CB ii) LSR/CB at 2 wt.% CB loading (1.0 Kx, (b)) i) LLDPE/CB ii) LSR/CB at 10 wt.% CB loading (500×)

4. Conclusion

The present study investigated the effect of CB additions on the percolation threshold of LLDPE/CB and LSR CPCs. Furthermore, the mechanical properties namely tensile properties were also investigated at various CB loadings. In the case of mechanical properties, additions of CB resulted in an increase of tensile strength by up to 23.73% as can be observed for sample with 8 wt.% CB loading. However, the elongation at break were decreased significantly at higher CB loadings. For LSR/CB composites, the tensile strength and elongation at break were significantly affected due to the presence of CB filler. In terms of electrical conductivity, presence of CB at various loadings resulted in an increase of the electrical conductivity for both LLDPE/CB and LSR/CB CPCs. The improvement in conductivity was believed due to the formation of network path where it was anticipated that the percolation threshold was at 6 wt.% loading.

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