

# Effect of Various Conductive Filler Additions on the Percolations Threshold of LLDPE Conductive Polymer Composites

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Polymeric materials are known to have insulating properties in general. Nevertheless, the insulating nature of polymers can be turned into electrically conductive by adding conductive fillers subjected to their critical filler loading or percolation threshold. In this study, the effects of various conductive fillers additions, namely copper, silver, and carbon black, on the percolation threshold of linear low-density polyethylene conductive polymer composites were investigated. The mechanical properties were determined using the tensile test, and the electrical conductivity was determined using the four-point probe. The incorporation of conductive fillers generally had an impact on the tensile strength and elongation at the break of the linear low-density polyethylene conductive polymer composites. Nonetheless, it was found that the electrical conductivity of all composites increases where the percolation threshold is estimated for carbon black at 2 wt% and for Ag and Cu at 6 wt% of filler additions.

topics: conductive polymer composites, percolation threshold, linear low-density polyethylene (LLDPE)

## 1. Introduction

Polymers are known to be insulating materials. However, they can become conductive by adding a conductive filler, such as carbon black (CB), alumina (Al), copper (Cu), zinc (Zn), and silver (Ag). Such polymers are known as conductive polymer composites (CPCs). CPCs have fascinating characteristics, such as light weight, resistance to corrosion, high electrical conductivity, low cost, resistance to water, low density, and good mechanical properties [1–3]. Due to their interesting properties, they have been used in many applications, such as sensors, batteries, fuel cells, and anti-static.

The composites become conductive due to the formation of the conductive network pathways inside the composite. This network path is created when a fixed conductive filler loading, called the percolation threshold, is achieved [3]. In a study

by X. Cui et al. [4], percolated conductive network was formed in conductive polymer chlorinated poly (propylene carbonate) (CPPC)/multi-walled carbon nanotubes (MWCNTs) where the composites can obtain insulator-conductor transition.

The aim of this paper is to study the effect of various types of conductive filler additions on the mechanical properties and electrical conductivity percolation threshold of LLDPE CPCs.

## 2. Experimental details

In this study, a linear low-density polyethylene (LLDPE) was used as the main matrix, whereas copper (Cu), silver (Ag), and carbon black (CB) were used as the conductive filler. The composition of Cu, Ag, and CB within LLDPE varied between 2–8 wt%. Next, the mixture was fed into an internal mixer with the speed of the rotor and temperature

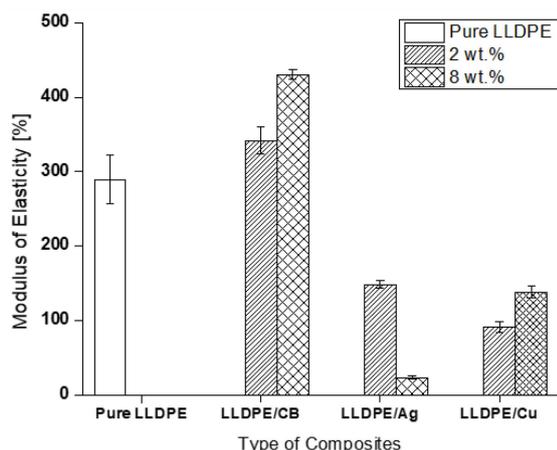


Fig. 1. Modulus of elasticity of different types of composites.

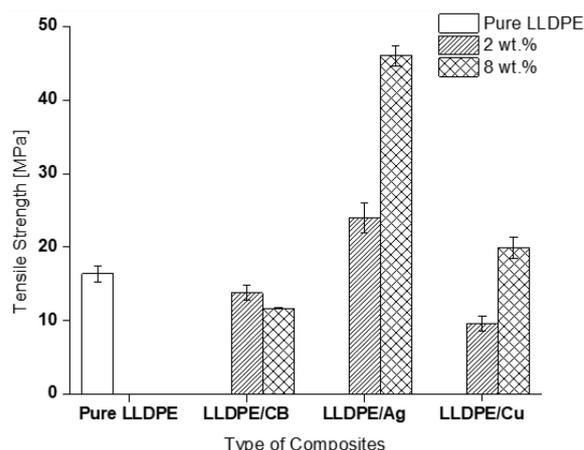


Fig. 2. Tensile strength of different types of composites.

of the compounding set to 15 rpm and 180°C, respectively. The composites were then hot-pressed into a rectangular shape mould with 1 mm thickness using a pressure of 15 MPa at 180°C.

All specimens were subjected to a tensile test using an Instron machine following the ASTM D3039. For the electrical conductivity test, the four-point probe was used to measure the conductivity of the specimen. A total of 5 sample points were taken during measurements to get the average electrical conductivity of the sample.

### 3. Results and discussion

Figure 1 illustrates the modulus of elasticity of LLDPE/CB composite at various loading of CB. As can be seen, the addition of CB of 2 wt% in LLDPE increased the modulus by 14% compared to pure LLDPE. Furthermore, the modulus of this CPC shows a significant increase at 8 wt% — by 44% more than of pure LLDPE. This improvement also was related to the addition of CB, which constrains the chains from free movement [5].

The modulus of elasticity of pure LLDPE was recorded at 289.55 MPa. At low loading, the modulus of elasticity of the composite was decreased by 49% due to additions of Ag within the LLDPE matrix. Increasing the Ag loading within LLDPE to 8 wt% decreased the modulus by 91% — from 289.55 to 23.63 MPa.

The effect of the addition of Cu into LLDPE on the modulus of elasticity is shown in Fig. 1. The addition of the Cu of 2 wt% had decreased the modulus by 68%, compared to pure LLDPE. At higher loading of the Cu, which is 8 wt%, the modulus of elasticity decreased by 52%.

Tensile strength of 16.40 MPa was recorded for pure LLDPE as determined from tensile test data. Figure 2 shows that at low loading of CB, which is 2 wt%, the tensile strength of the composites was decreased by 5% more than that of pure LLDPE.

At higher CB loading, a decrease in tensile strength of up to 20% compared to pure LLDPE can be observed. As stated by V. Sangeetha et al. [6], they found that with the addition of halloysite nanotubes (HNTs) of over 5 %, the HNTs had acted as obstacles due to agglomeration in the LLDPE/HNTs composites.

The addition of Ag at low loading has increased the tensile strength by 46% compared to pure LLDPE. The addition of Ag at higher loading, which is 8 wt%, has increased the strength significantly — by 151%. The increase in the tensile strength may be due to the interaction that may exist between Ag particles with the LLDPE matrix. This is a similar trend to that of the GO–Ag and PVA/ $\beta$ -CD study [7].

Additions of Cu of 2 wt% had decreased the strength of the composites by 41% compared to pure LLDPE. However, with 8 wt% of Cu loading, the tensile strength of the composites was also increased by 21%.

Figure 3 shows that the elongation at break of the composite had decreased by 2% when CB of 2 wt% was added. The most significant decrease of elongation at break can be seen at 8 wt%, where it decreases by 90% more compared to pure LLDPE. The decrease in the elongation at break may probably be due to the poor interactions between the filler and the matrix.

The effect of the addition of Ag into the LLDPE composite shows that there is an increase of the elongation at break at 2 wt% — by 30% more than in pure LLDPE. However, as the Ag powder addition was increased up to 8 wt%, the elongation at break decreased approximately by 141%. According to the works by F. Ren et al. [8], poor interaction between the LLDPE and silicon carbide (SiC) decreased the elongation at break of the composites due to the presence of voids around SiC particles.

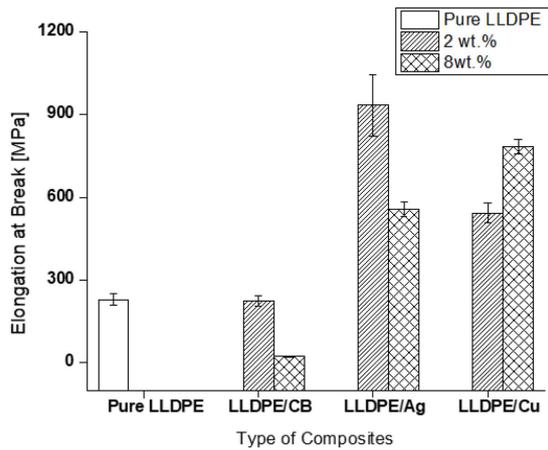


Fig. 3. Elongation at break of different types of composites.

The elongation at break was increased by 108% at low loading of Cu — at 2 wt% of LLDPE composites. The addition of Cu at higher loading, in particular at 8 wt%, had significantly higher elongation at break, by 200% compared to pure LLDPE.

In general, additions of rigid conductive filler are expected to make the composites stiffer, which can result in poor toughness properties. Nevertheless, the addition of Ag and Cu fillers into the LLDPE matrix investigated in this study had resulted in elongation at break superior to that of pure LLDPE. This may be attributed to the filler–matrix interactions where the filler had bonded with the matrix through physical bonding, which made the toughness of the CPCs increase. Moreover, the filler aspect ratio, shape, and size, as well as the formation of smaller crystallites within the composites, can also be a factor contributing to the observed trends. Nevertheless, further investigations on the observed trends are essential to establish the relation between the filler–matrix interactions with the toughness of the CPCs, which will be carried out in future studies.

The results for the electrical conductivity of LLDPE/CB, LLDPE/Ag, and LLDPE/Cu are shown in Fig. 4. The electrical conductivity of  $4.82 \times 10^{-5}$  S/cm was recorded for pure LLDPE. In the case of LLDPE/CB CPCs, at 2 wt% of CB, the electrical conductivity was increased by 5% compared to pure LLDPE. The addition of CB of 8 wt% into LLDPE increased the electrical conductivity by 8% more than pure LLDPE. This is due to the conductive paths that have increased as the filler loading increased. A marginal increase in the conductivity can be seen at 6 wt% where the percolation threshold of the LLDPE/CB was achieved. Zhou et al. [9] found that the percolation threshold of addition CB into LLDPE was at 20 wt%, and the conductivities were affected due to the distribution of the CB in the composites.

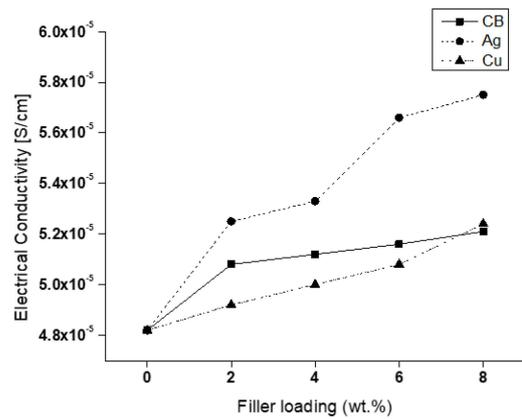


Fig. 4. Conductivity of LLDPE with different types of filler.

At the low loading of Ag, which is at 2 wt%, the conductivity has increased by 9% compared to pure LLDPE. Further increase in conductivity can be observed as the filler loading was increased, as can be seen for LLDPE/Ag composites between 4–8 wt% where the conductivity was increased by 10–19% compared to pure LLDPE. In a study carried out by C. Li et al. [10], the authors found that the electrical resistivity of treated Ag decreases, which indicates that the conductivity was higher compared to untreated Ag.

The addition of the Cu of 2 wt% increased the conductivity of the LLDPE/Cu composites by 2% more than pure LLDPE. At the higher loading of Cu, which is 8 wt%, the electrical conductivity was  $5.24 \times 10^{-5}$  S/cm, where it increased by 8% more than pure LLDPE.

The estimated percolation threshold for CB was the lowest compared to Ag and Cu. For CB, the estimated percolation threshold was at 2 wt% of CB loading. Moreover, for Ag and Cu, the percolation threshold was estimated at 6 wt% filler loading. Improvement in electrical conductivity was attained due to the conductive network that forms as soon as the filler load outreach the percolation threshold [11].

#### 4. Conclusions

In conclusion, the incorporation of different types of conductive fillers at various loading for LLDPE CPCs can be summarized as follows:

1. The mechanical properties of the composites were affected due to the presence of conductive fillers. The tensile strength and the elongation at break for LLDPE/CB were significantly decreased compared to LLDPE/Ag and LLDPE/Cu composites.
2. The addition of various conductive fillers into the LLDPE matrix increased the conductivity of the CPCs, and the highest electrical conductivity was recorded for CPCs with 8 wt%

Ag addition. This was attributed primarily to the formation of the network or conductive path within the composites.

3. The percolation threshold for CB was estimated at 2 wt%, in which the electrical conductivity was  $5.08 \times 10^{-5}$  S/cm, whereas the estimated percolation threshold for Ag and Cu was at 6 wt% filler loading, with the measured conductivity of  $5.66 \times 10^{-5}$  S/cm and  $5.08 \times 10^{-5}$  S/cm, respectively.

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