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THE CONDUCTIVITY AND MECHANICAL PROPERTIES OF HYBRID CARBON BLACK/COPPER FILLED LINEAR LOW-DENSITY POLYETHYLENE (LLDPE) AND LIQUID SILICONE RUBBER (LSR) FLEXIBLE CONDUCTIVE POLYMER COMPOSITES FOR ELECTRONIC INTERCONNECT APPLICATIONS

The advancement in the electronics industry have led to the emerging trends towards miniaturization and flexible electronic devices. The ability to be stretched, twisted, and bent have made Flexible Conductive Polymer Composites (FCPCs) one of the attractive materials in electronic interconnect applications. In this work, the combination of Carbon Black/Copper (CB/Cu) as hybrid fillers within linear low-density polyethylene (LLDPE) and liquid silicone rubber (LSR) matrices were investigated to overcome the limitation of single filler addition such as poor dispersion with the aim to further enhance the electrical and mechanical properties of the FCPCs. The FCPCs prepared in this study were characterized and tested using various techniques such as 4-point probe, tensile testing, hardness test and electron microscopy. Composites of LLDPE/CB/Cu and LSR/CB/Cu were prepared with varying filler loadings such 3 wt.%, 6 wt.%, 9 wt.%, 12 wt.% and 15 wt.%. The ratio of copper to carbon black are 1:1. The LLDPE and LSR exhibit increased tensile strength with up to 15 wt.% CB and Cu loadings. LLDPE shows enhanced elongation at break (55.93%) with 15 wt.% filler, while LSR declines beyond 12 wt.%. The modulus of elasticity is inversely proportional to elongation at break for both materials. Electrical conductivity increases in LLDPE and LSR with fillers up to 9 wt.%, forming a conductive network that decreases at higher loadings. Shore hardness indicates increased hardness with greater CB and Cu content. SEM analysis reveals filler dispersion and distribution within the polymer matrices, providing insights into the composites' microstructure.

Keywords: Hybrid fillers; Conductivity; Mechanical Properties; Flexible Conductive Polymer Composites; Electronic Interconnect

1. Introduction

Polymers are widely used in chemistry, materials science, engineering, and biology due to their versatility, affordability, and ease of shaping [1]. However, they have limitations. Composite materials, with superior properties like lightweight and high strength, are designed to replace conventional materials such as metals, woods, and ceramics [2]. It can be composed of a polymer, metal, ceramic, or any other material that imparts strength, durability, and desired characteristics to the composite [3].

Conductive polymer composites (CPCs) are formed by adding conductive fillers such as carbon black (CB), carbon nanotubes (CNT), metal particles, or conducting polymers to an insulating polymer matrix. In the context of LLDPE and LSR composites, an increasing trend in elongation at break suggests improvements in the material's flexibility and resistance to fracture, make them more ductile and flexible [4,5].

These fillers align to create a conductive path, forming a network that allows electrical current flow through the composite material [6]. This network path is created when the percolation threshold is achieved. Percolation threshold is the minimum filler quantity required to establish a continuous conducting network and create CPCs [7,8]. The volume fraction, size, shape, and structure of the fillers determine the electrical properties of CPCs. Optimization of processing parameters can help minimize agglomeration [9].

These days metal have been widely use as conductive filler in industries and research area. Carbon black and copper are

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popular conductive materials due to their chain-like structures, especially carbon black, which has a higher propensity for forming conductive networks compared to other metal powder [10-12]. It possesses excellent electrical and thermal conductivity [13-14]. This research investigates the effects of carbon black and copper as conductive hybrid fillers on the properties of linear low-density polyethylene (LLDPE) and liquid silicone rubber (LSR).

2. Methodology

2.1. Materials

Linear low-density polyethylene (LLDPE) with grade LL0209SR and density of 920 kg/m³ and liquid silicone rubber (LSR) was provided by Malaysia Clay Art used as polymer matrix in this research. In LSR, there were two components such as catalytic solution (A) and hardener (B) with same ratio, 1:1. Carbon black (CB) with N330 grade and copper (Cu) powder with 8.92 g/cm^3 was purchased from Alfa Aesar Avocado Research Chemistry, Ltd. was used as conductive filler in this experiment.

2.2. Sample Preparation

In this experiment, LLDPE and LSR composites were formulated with different loadings of hybrid filler, ranging from 3 wt.% to 15 wt.%, which shows in TABLE 1 and TABLE 2. The LLDPE/CB/Cu composites were prepared by compounding the materials in a Z-blade mixer at 15 rpm. The compounded mixture was then compression molded using a hot press machine at 150°C. The molding process involved 7 minutes of preheating, 10 minutes of heating, and 3 minutes of cooling process to obtain a 1 mm thick sample. In the case of LSR/CB/Cu composites, the raw materials were mixed and cast into a 1mm thick glass mold. The composites were then cured in an oven at 80°C for 15 minutes. Then, the samples were cut into dumbbell shapes for tensile test. For electrical properties testing, spherical samples with a diameter of 2.3 cm. Additionally, 1 cm × 1 cm samples were prepared for morphology characterization.

TABLE 1

Formulation for LLDPE/CB/Cu

Sample	Composition (wt.%)	LLDPE (g)	CB (g)	Cu (g)
1	Pure LLDPE	150.00	0.00	0.00
2	LLDPE+CU/CB (3 wt.%)	145.40	2.30	2.30
3	LLDPE+CU/CB (6 wt.%)	141.00	4.50	4.50
4	LLDPE+CU/CB (9 wt.%)	136.40	6.80	6.80
5	LLDPE+CU/CB (12 wt.%)	132.00	9.00	9.00
6	LLDPE+CU/CB (15 wt.%)	127.40	11.30	11.30

TABLE 2

Formulation for LSR/CB/Cu

Sample	Composition (wt.%)	LSR (g)	CB (g)	Cu (g)
1	Pure LSR	50.00	0.00	0.00
2	LSR+CU/CB (3 wt.%)	48.40	0.80	0.80
3	LSR+CU/CB (6 wt.%)	47.00	1.50	1.50
4	LSR+CU/CB (9 wt.%)	45.40	2.30	2.30
5	LSR+CU/CB (12 wt.%)	41.00	3.00	3.00
6	LSR+CU/CB (15 wt.%)	41.40	4.30	4.30

2.3. Testing

The tensile properties of LLDPE and LSR composites were tested by using Instron 5569 tensile machine. ASTM D638 was used to assess the tensile properties of LLDPE/CB/Cu, while ASTM D412 was specific to LSR/CB/Cu. In this study, dumbbell shaped LLDPE and LSR specimens were tested at different crosshead speeds: 70 mm/min for LLDPE and 500 mm/min for LSR. Shore D hardness was used to measure the hardness of LLDPE/CB/Cu composites, while Shore A hardness was tested for LSR/CB/Cu composites. For four-point probe testing, 3 measurements were taken for each sample at different scanning points. Eq. (1) was used to calculate the conductivity of sample. The morphology of the samples was analysed using scanning electron microscopy (SEM), at specific magnifications of ×100, ×300, and ×500 with an accelerating voltage of 10 kV.

$$\sigma = \frac{L}{R \times A} \tag{1}$$

where: L – thicknessofsample, R – resistance of composite and A – area of sample.

3. Results and discussion

3.1. Mechanical properties

The mechanical properties of the LLDPE and LSR composites was analysed and discussed based on the Figs. 1 and 2. Fig. 1 illustrates the tensile strength of LLDPE and LSR composites where it shows that as the loading of CB and Cu increases, the tensile strength of the composites also increases. Tensile strength of pure LLDPE was 15.06 MPa, which it increased approximately by 3.4% to 15.58 MPa with the addition of 3 wt.% carbon black and copper compared to pure LLDPE. The tensile strength further improved up to 15 wt.%, showing a significant 40.11% increase compared to pure LLDPE.The addition of fillers to LLDPE modifies the microstructure of material and enhances its mechanical properties. Similarly, pure LSR was measured at 2.05 MPa. When 3 wt.% of carbon black and copper were introduced, the tensile strength decreased by 7.5% which the strength were recorded at 1.91 MPa. The initial decrease in tensile strength can be attributed to the weak interaction between the fillers and LSR [15]. However, as the hybrid filler content increases, the tensile strength of LSR also increases [16]. The addition of 15 wt.% CB and Cu achieves the maximum tensile

strength at 2.955 MPa. LLDPE has higher tensile strength due to stronger intermolecular interaction and entanglement of its linear chains, while LSR has lower tensile strength with a more flexible and less interconnected structure [17].

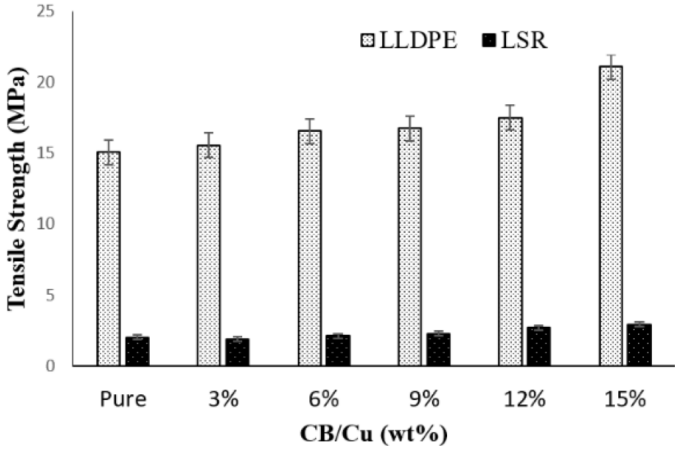

Fig. 1. Tensile strength of LLDPE and LSR composites

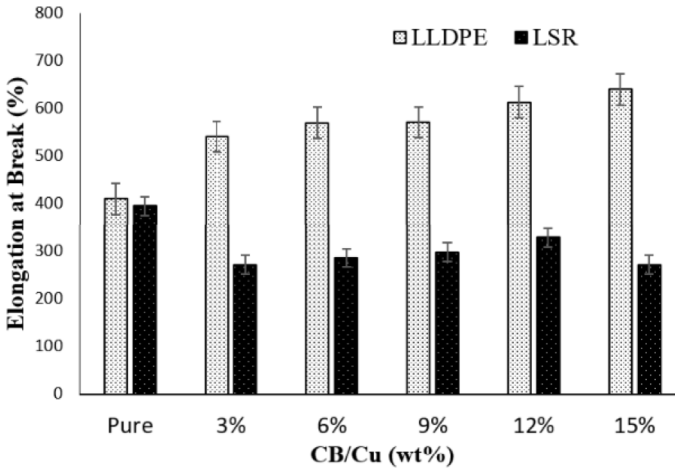

Fig. 2. Elongation at break of LLDPE and LSR composites

Fig. 2 shows the elongation at break of LLDPE and LSR composites. Pure LLDPE has an elongation at break of 410.50%, which rises to 540.90% with 3 wt.% CB and Cu. The highest elongation at break of 640.10% is achieved with a 15 wt.% hybrid filler composition, indicating an improvement of approximately 55.93% compared to pure LLDPE. For LSR, the elongation at break initially increases with filler addition up to 12 wt.% but decreases for higher filler content. Pure LSR has an elongation at break of 395.83%, while LSR with 3 wt.% CB and Cu has an elongation at break of 270.83%. The decrease in elongation at break at higher filler content is due to filler agglomeration. The elongation of LLDPE/CB/Cu is higher than LSR/CB/Cu, thus it becomes stiffer.

Fig. 3 shows the modulus of elasticity of LLDPE and LSR composites. The addition of CB and Cu to LLDPE and LSR significantly increases their modulus of elasticity. LLDPE achieves its highest modulus at 3 wt.% filler loading. However, beyond this loading level, the modulus decreases. The fillers create a network structure, enhancing interfacial interactions and improving stiffness (Ramesh et al., 2022). The pure LSR has a relatively low modulus of elasticity at 136 MPa, but was higher than that of LLDPE due to its cross-linked structure. The addition of CB and Cu increases the modulus, reaching its highest value at 6 wt.% loading with a recorded value of 187.7 MPa, representing a 38% increment compared to pure LSR. However, the modulus decreases when the filler loading exceeds 6 wt.%.

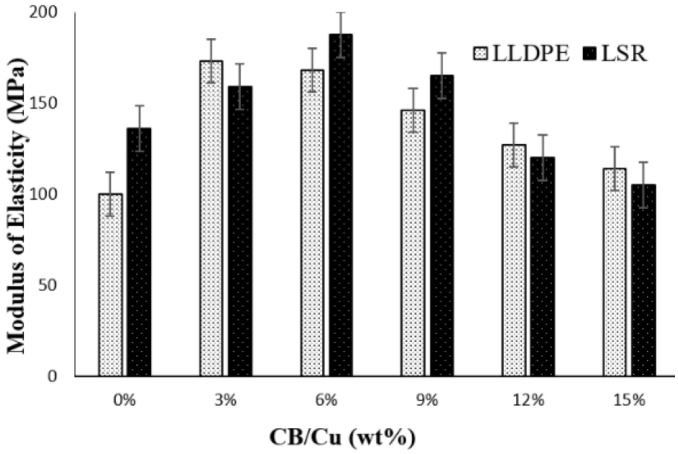

Fig. 3. Modulus of elasticity of LLDPE and LSR composites

3.2. Hardness

The hardness of LLDPE and LSR composites, as shown in Fig. 4, increases linearly with the addition of carbon black (CB) and copper (Cu). Shore D was used for LLDPE composites, while Shore A was used for LSR composites. Adding 3 wt.% fillers to LLDPE resulted in a 2.1% increase in hardness compared to pure LLDPE (with a hardness value of 47.2). The hardness continued to increase, reaching 55.6 with the addition of 15 wt.% fillers. In contrast, the hardness of LSR composites increased with increasing filler loading. The hardness of pure LSR was 32, while the maximum hardness of 44.4 was achieved with 15 wt.% CB and Cu fillers, representing an increase of approximately

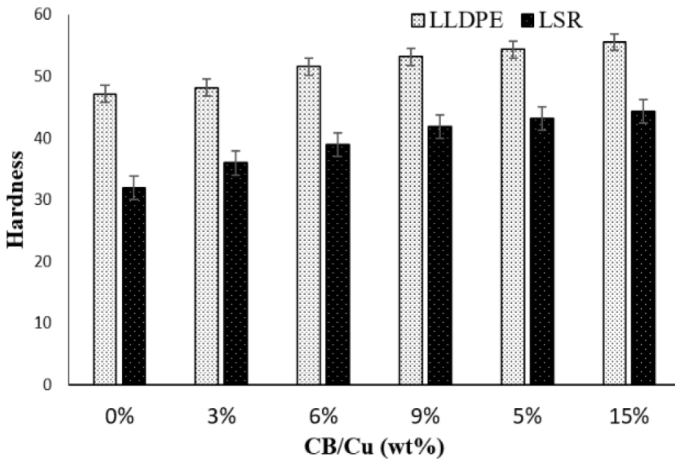

Fig. 4. Hardness of LLDPE and LSR composites

38.8% compared to pure LSR. The increase in crosslink density contributed to the increase in hardness and modulus [18]. It can be said that LLDPE composites harder than LSR composites.

3.3. Electrical conductivity

The electrical conductivity calculated from the obtained resistivity was plotted in Fig. 5(a). The electrical conductivity of pure LLDPE was found at 6.719×10^{-4} S/cm. The addition of 3 wt.% fillers resulted in a conductivity increase of approximately 16.37% for LLDPE, reaching a value of 7.820×10^{-4} S/cm. At carbon black and copper loadings of 9 wt.%, the amount of hybrid filler reached saturation, resulting in the highest electrical conductivity (8.607×10^{-4} S/cm) in LLDPE. However, a reduction in conductivity beyond this point, the greater composition of filled CB and Cu particles finding in a lower electrical conductivity. When the amount of hybrid filler increases until 15 wt.%, the conductivity drops to 8.422×10^{-4} S/cm. Besides, Fig. 5(b) demonstrates the conductivity of LSR after incorporating copper and carbon black. pure LSR was recorded at 7.416×10^{-4} S/cm, while the addition of 3 wt.% fillers increased the conductivity by 14.58%. The conductivity continued to rise until reaching a saturation point at 9 wt.% fillers, resulting in the highest conductivity of 9.235×10^{-4} S/cm, with an increment 24.53% compared to pure LSR. However, conductivity decreased when the filler loading exceeded 9 wt.%. Agglomeration or clustering of fillers within the polymer matrix created insulating barriers, impeding electron movement and reducing conductivity [12].

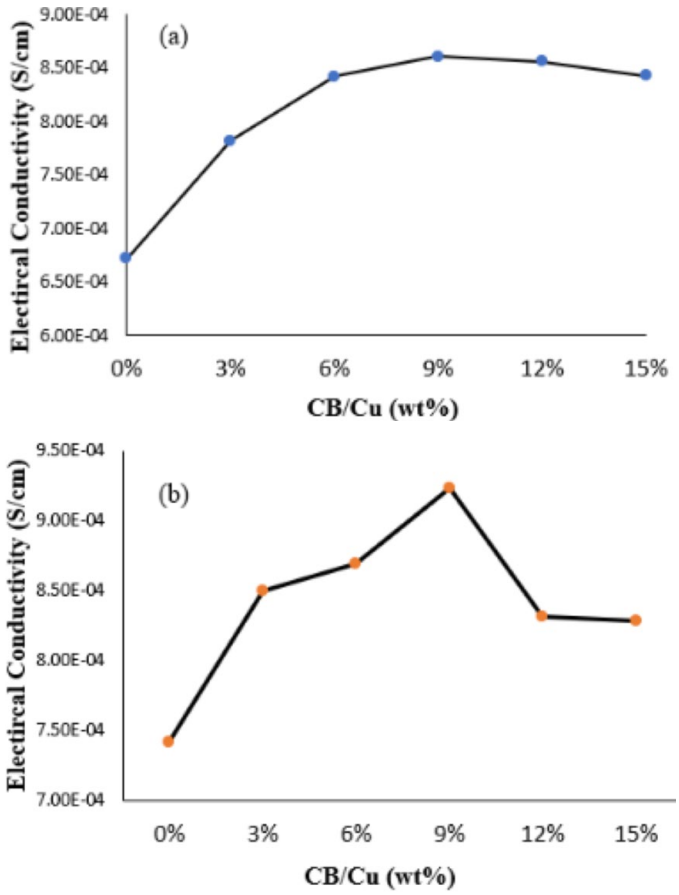

Fig. 5. Electrical conductivity of (a) LLDPE composites (b) LSR composites

3.4. Morphology

Fig. 6 (a) and (b) illustrate the morphologies of the tensile fracture surfaces of LLDPE/CB/Cu composites with filler loadings of 3 wt.% and 12 wt.%, respectively. In both figures, the copper filler displayed a particle shape resembling flakes, whereas the CB exhibited a typical morphology of aggregated spherical particles. Void formation and partial agglomeration of carbon black (CB) and copper (Cu) fillers within the LLDPE matrix can be observed. As the CB content increases, the particles tend to merge, forming larger aggregates due to attractive van der Waals forces, resulting in loosely assembled agglomerates [19,20]. Increasing the filler content to 12 wt.% significantly reduced the presence of voids in the LLDPE composite.

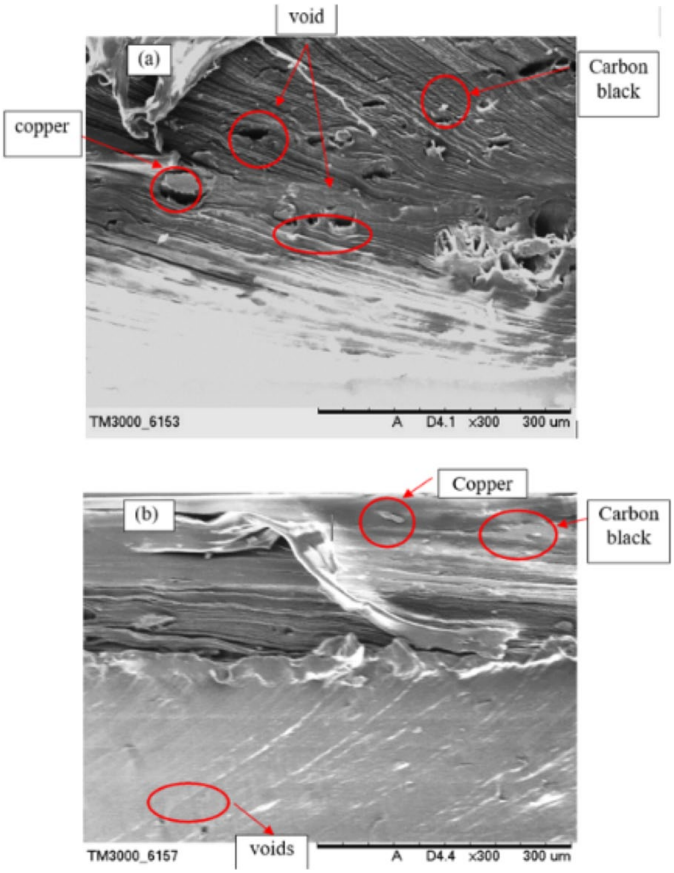

Fig. 6. SEM micrographs of LLDPE/CB/Cu composite with various filler contents at 300× magnification: (a) tensile fracture surface of 3 wt.% CB and Cu (b) tensile fracture surface of 12 wt.% CB and Cu

Fig. 7(a) demonstrate the dispersion of carbon black (CB) and copper (Cu) fillers at 3 wt.% and 12 wt.% in the LSR matrix. The interface between the fillers and the matrix played a crucial role in determining the mechanical and electrical properties of the

composite material. Poor dispersion led to agglomeration of LSR, compromising its mechanical and electrical properties [21]. As a result, cracks formed on the fracture surface after the tensile test. Fig. 7(b) reveals the presence of pores and agglomeration between carbon black (CB) and copper (Cu) fillers within the LSR matrix. These fillers tend to aggregate due to the high density of copper and uneven distribution, resulting in voids. This agglomeration and the presence of voids significantly affect the roughness of the fracture surface in the LSR material.

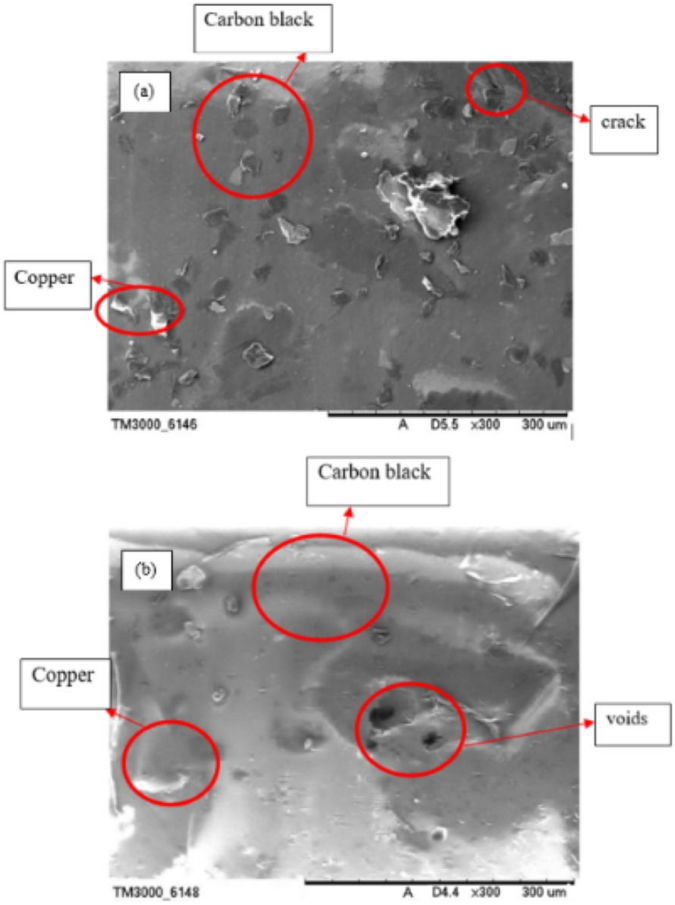

Fig. 7. SEM micrographs of LSR/CB/Cu composite with various filler contents at 300× magnification: (a) tensile fracture surface of 3 wt.% CB and Cu (b) tensile fracture surface of 12 wt.% CB and Cu

4. Conclusion

The effect of hybrid CB/Cu additions within LLDPE and LSR matrices on the conductivity and mechanical properties of CPCs can be summarized as following:

1. The addition of carbon black (CB) and copper (Cu) fillers affects the mechanical and electrical properties of LLDPE and LSR composites. LLDPE and LSR show the tensile strength increase when up to 15 wt.% of CB and Cu added. The elongation at break increases in LLDPE, reaching its maximum value of 640.10% at a loading of 15 wt.%.
2. The modulus of LLDPE increase initially beyond 3 wt.% of fillers but decrease of 51.9% compared to the sample containing 15 wt.%. For LSR, the highest increment in modulus was observed at 6 wt% loading, but it decreases by 13.04% with 12 wt.% filler loading.
3. Both composites exhibit increased electrical conductivity with filler addition, reaching a peak at 9 wt.% before declining due to agglomeration. These fillers play a crucial role in creating pathways for electrical conduction within the composite material.
4. SEM analysis shows agglomeration and changes in surface morphology at higher filler loadings. Since the tensile strength, elongation at break, electrical conductivity and hardness of LLDPE/CB/Cu was higher than LSR/CB/Cu, LLDPE can be determined as the most suitable or better materials for application in electronic and electrical industries.

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REFERENCES

[1] Y. Lin, F. Yin, Y. Liu, L. Wang, Y. Zhao, M. Farzaneh, Effect of ultraviolet – A radiation on surface structure, thermal, and mechanical and electrical properties of liquid silicone rubber. Journal of Applied Polymer.
DOI: https://doi.org/10.1002/APP.47652

[2] T.-D. Ngo (ed.), Composite and Nanocomposite Materials – From Knowledge to Industrial Applications. IntechOpen (2020).
DOI: https://doi.org/10.5772/intechopen.80186

[3] K. K. Kar (ed.), Composite materials: Processing, applications, characterizations. Composite Materials: Processing, Applications, Characterizations, Springer (2016).
DOI: https://doi.org/10.1007/978-3-662-49514-8

[4] N.N. Azmi, M.N.A. Ab Patar, S.N.A. Mohd Noor, J. Mahmud, Testing standards assessment for silicone rubber. ISTMET 2014 – 1st International Symposium on Technology Management and Emerging Technologies, Proceedings, 332-336 (2014).
DOI: https://doi.org/10.1109/istmet.2014.6936529

[5] L.A. Bloomfield, Borosilicones and viscoelastic silicone rubbers: network liquids and network solids. arXiv preprint arXiv:1801.09253 (2018). DOI: http://arxiv.org/abs/1801.09253

[6] W.J. Wright, E. Celik, In situ electrical network activation and deactivation in short carbon fiber composites via 3D printing. Advanced Functional Materials **33** (40), 2303282 (2023).
DOI: https://doi.org/10.1002/adfm.202303282

[7] E. Chang, A. Ameli, A.R. Alian, L.H. Mark, K. Yu, S. Wang, C.B. Park, Percolation mechanism and effective conductivity of mechanically deformed 3-dimensional composite networks: Computational modeling and experimental verification. Composites Part B: Engineering **207**, 108552 (2021).
DOI: https://doi.org/10.1016/j.compositesb.2020.108552

[8] P.C. Dartora, R.M.C. Santana, A.C.F. Moreira, The influence of long chain branches of LLDPE on processability and physical properties. Polimeros **25** (6), 531-539 (2015). DOI: https://doi.org/10.1590/0104-1428.1732

[9] J. George, A.M. Poulose, A. Chandran, A.A. Somashekar, Influence of plasticizer on the dielectric properties of polypropylene/carbon black composites. Materials Today: Proceedings, (2023). DOI: https://doi.org/10.1016/j.matpR.2023.03.297

[10] I.M. Alarifi, Investigation the conductivity of carbon fiber composites focusing on measurement techniques under dynamic and static loads. Journal of Materials Research and Technology **8** (5), 4863-4893 (2019). DOI: https://doi.org/10.1016/j.jmrt.2019.08.019

[11] P. Vizureanu, C. Samoilă, D. Cotfas, Materials Processing Using Solar Energy. Environmental Engineering & Management Journal (EEMJ) **8** (2), (2009). DOI: https://doi.org/10.30638/eemj.2009.043

[12] T. Ding, L. Wang, P. Wang, Changes in electrical resistance of carbon-black-filled silicone rubber composite during compression. Journal of Polymer Science, Part B: Polymer Physics **45** (19), 2700-2706 (2007). DOI: https://doi.org/10.1002/polb.21272

[13] P. Vizureanu, M. Nabiałek, A.V. Sandu, B. Jeż, Investigation into the Effect of Thermal Treatment on the Obtaining of Magnetic Phases: Fe5Y, Fe23B6, Y2Fe14B and αFe within the Amorphous Matrix of Rapidly-Quenched Fe61+xCo10−xW1Y8B20 Alloys (Where x = 0, 1 or 2). Materials **13** (4), 835 (2020). DOI: https://doi.org/10.3390/ma13040835

[14] Y. Huang, S. Kormakov, X. He, X. Gao, X. Zheng, Y. Liu, J. Sun, D. Wu, Conductive Polymer Composites from Renewable Resources: An Overview of Preparation, Properties, and Applications. Polymers **11** (2), (2019). DOI: https://doi.org/10.3390/polym11020187

[15] G. Kaur, R. Adhikari, P. Cass, M. Bown, P. Gunatillake, Electrically conductive polymers and composites for biomedical applications. RSC Advances **5** (47), 37553-37567 (2015). DOI: https://doi.org/10.1039/c5ra01851j

[16] A.A. Baharum, A. Arman Alim, S.S. Mohammad Shirajuddin, F.H. Anuar, Blending of Low Density Polyethylene and Poly(Butylene Succinate) (LDPE/PBS) with Polyethylene–Graft Maleic Anhydride (PE–g–MA) as a Compatibilizer on the Phase Morphology, Mechanical and Thermal Properties. Polymers **15** (2), (2023). DOI: https://doi.org/10.3390/polym15020261

[17] M.A. Husnan, H. Ismail, R.K. Shuib, The effect of carbon black (CB) loading on curing characteristics and mechanical properties of virgin acrylonitrile butadiene rubber (Nbrv)/recycled acrylonitrile butadiene rubber (Nbrr) blends. IOP Conference Series: Materials Science and Engineering **309** (1), 012028 (2018). DOI: https://doi.org/10.1088/1757 899X/309/1/012028

[18] Y.J. Kwon, J. Bin Park, Y.P. Jeon, J.Y. Hong, H.S. Park, J.U. Lee, A Review of Polymer Composites Based on Carbon Fillers for Thermal Management Applications: Design, Preparation, and Properties. Polymers **13** (8), 2021). DOI: https://doi.org/10.3390/polym13081312

[19] M. Ramesh, L.N. Rajeshkumar, N. Srinivasan, D.V. Kumar, D. Balaji, Influence of filler material on properties of fiber-reinforced polymer composites: A review. E-Polymers **22** (1), 898-916 (2022). DOI: https://doi.org/10.1515/epoly-2022-0080

[20] C.W. Heng, P.L. Teh, N.A. Abdul Rahim, C.K. Yeoh, The influence of liquid silicone rubber on the properties of polyurethane elastomer/liquid silicone rubber/graphene nanoplatelets stretchable conductive materials. Progress in Rubber, Plastics and Recycling Technology **38** (4), 267-279 (2022). DOI: https://doi.org/10.1177/14777606221118659