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# Electrical and Mechanical Properties of Silicone Electrically Conductive Adhesive (ECAs) Filled Graphene-Carbon Black (GR-CB)

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## ABSTRACT

ECAs were successfully introduced to replace hazardous lead-based solder, but their electrical conductivity and mechanical properties are limited. Hence, the incorporation of carbon black (CB) and Graphene (GR) filled silicone based ECAs with various ratios of GR and CB (0:0, 1:0, 0:1, 10:5, 5:10, 5:5, and 3:5) via solvent casting technique was investigated. The characterization of the conductive adhesive film using Fourier Transform Infrared Spectroscopy (FTIR) and electrochemical impedance spectroscopy (EIS) was investigated using the Nyquist plot and conductivity of silicone film. Meanwhile, the mechanical properties were measured using tensile and hardness tests. The appearance of a sharp peak in the FTIR spectrum confirmed that the interaction between the silicone matrix and GR-CB occurred (Si-$CH_3$). The optimum ratio with the highest conductivity value, 7.98 $10^{-8}$ Ω $cm^{-1}$, was obtained by the synergistic effect of conductive fillers, which is GR with CB at 10:5 GR-CB ratio, which might be attributed to the synergistic effect from the filler. At the same time, tensile and hardness tests on silicone ECAs samples exhibited that the optimum ratio of synergistic GR-CB was at 5:10 with the highest tensile strength, young modulus, elongation at break, and hardness with values of 1.16 MPa, 2.02 MPa, 25.64 mm, and 47.56, respectively due to the inherent stiffness of CB and GR. Finally, the electrical and mechanical properties revealed that adding GR and CB improved the conductivity, strength, and toughness of silicone ECAs composites, making way for electronic applications.

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## INTRODUCTION

Soldering plays an important role in electric and electronic fields, assembling different components by melting solder made of tin and lead. However, due to environmental and health issues, the electronic industry now uses electrically conductive adhesives (ECAs) as alternatives to the traditional soldering process. ECAs act as glue primarily used to assemble parts of electronic devices. The advantages of ECAs over soldering are that they are environmentally friendly, have lower processing temperatures, have fewer steps in processing that reduce the processing cost, and have increased capacity of fine performance due to small-sized conductive fillers [1].

ECAs comprise binders such as polymeric resin and a conductive metal filler. The most widely used polymeric binders are epoxy, polyurethane, and silicone. In this research, silicone-based ECAs were used as the main composition in matrices of the ECAs system due to their good adhesion, high impact strength, thermal and flexible electrical contact, which can maintain flexibility above 150°C, and stretchable electronic due to their unique elasticity combination, moisture resistance, thermal stability, and flexibility [2]. Also, silicone binders are becoming increasingly popular due to their radiation resistance, excellent electrical insulation, corrosion resistance, enhanced fatigue resistance at high temperatures, high fracture, and toughness. Aside from that, silicones are high-value polymers with a Si-O-Si-O chain instead of a C-chain. The higher Si-O bond energy (110 kcal/mol) than the C bond (80 kcal/mol) gives improved stability and durability in the presence of UV radiation and high temperatures [3]. However, its lower electrical conductivity prevents widespread use in the electronic industry. As a result, many researchers studied conductive fillers like carbon black, graphene, and micron or nano-sized metal particles like silver, nickel, copper, or aluminum to improve conductivity and mechanical properties [2].

The conductive filler is one of the most important ECAs components since it creates a conductive conduit for electric current flow, while the binder phase functions to hold the conductive filler together and install the electronic component onto the board [2]. As an alternative, Graphene (GR) and Carbon black (CB) were recommended due to their incredible electrical conductivity and mechanical properties, which might improve the properties of conductivity and mechanical properties towards polymer matrices [3]. Therefore, a synergistic conductive filler of GR with CB has been studied as another alternative. It is believed that introducing the synergism of GR-CB into the system increases the electrical conductivity of the silicone ECA system. It also increases the stability of the ECA system and helps it withstand room temperature [4]. The effects of the synergistic are prepared using the film casting method and testing using some methods on electrical and mechanical properties.

The silicone ECAs filled GR-CB in various ratios [0:0, 1:0, 0:1, 5:10, 10:5, 5:5, 3:5]. It was agitated with a sonicator to produce a film using the film casting method, which was then cured in an oven. After curing, the samples were subjected to Fourier-transform infrared spectroscopy (FTIR) to characterize the GR-CB interaction and Electrical impedance spectroscopy (EIS) for electrical conductivity. Furthermore, mechanical testing is carried out using the Hardness and Tensile tests.

## EXPERIMENTAL

### Materials

Silicone (Polydimethylsiloxane-PDMS)-Sylgard 184 Silicone Elastomer Kit and with Sylgard 184 Silicone Elastomer Curing Agent was purchased from Dow Chemical. A carbon black with an average particle size of 69 nm was bought from Birla Carbon (Thailand) Public Co., Ltd. Graphene nanoplatelets with a surface area of 500m 2/g and an average particle size of < 2µm were obtained from Sigma Aldrich.

### Preparation of GR-CB Filled Silicone Film.

7.80 ml of silicone solution (Polydimethylsiloxane) was mixed with different loading of GR-CB ratio (0:0, 1:0, 0:1, 5:10, 10:5, 5:5, 3:5) in beaker/vial using sonicated for 30 minutes then cured at 110°C temperature for 1 to 2 hours to form thin films. The samples were conditioned at room temperature.

### Characterization and Testing.

The FTIR analysis was conducted using Perkin Elmer FTIR Spectroscopy analysis at room temperature at 4000-500 $cm^{-1}$. The hardness tests were carried out with a hand-held Durometer (Shore Instruments, Type A-2) following the ASTM D2240-05 (2010) type A standard. The tensile test was performed via an Instron Universal testing machine according to DIN EN 53504, and the tensile velocity was set at 25mm/min. The EIS was performed with an Autolab Potentiostat set to $10^3$ to $10^6$ hertz, and the results were analyzed with the Nova 1.11. The results of the conductivity test were acquired from the data following the equation 1:

$$\sigma = t/R_bA \qquad (1)$$

$\sigma$ = electric conductivity ($\Omega\ cm^{-1}$)
t = thickness of the film (cm)
Rb = the bulk resistance ($\Omega$)
A = area of the electrode ($cm^2$)

## RESULTS AND DISCUSSION

### Fourier Transform Spectroscopy (FTIR) analysis.

Figure 1 shows the presence of assorted peaks of silicone film for the control sample, which is the sample without fillers with a ratio of 0:0 of GR-CB. Assignment at (i) 1010 $cm^{-1}$ was present in all samples and corresponds to Si-O-Si from the principal chain of PDMS. It shows that the silicone has a distinctive silicon-oxygen (Si–O) siloxane backbone. This is also supported by Du et al. (2014), who also stated that strong connections between Si–O–Si bonds are responsible for the exceptional heat resistance exhibited by silicone resin and, at the same time, affect mechanical properties such as adhesion, tensile strength, impact strength, and flexibility [5].

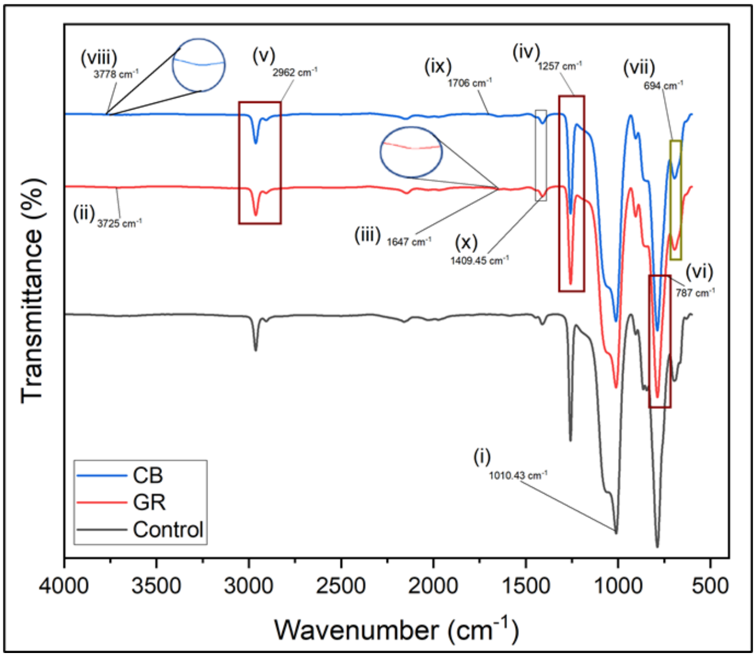

Fig. 1. FTIR spectrum peak for every control sample with a ratio of control (0:0), GR (1:0), and CB (0:1) GR-CB.

The peak at (ii) 3725 cm$^{-1}$ assigned to O-H groups from the GR compound is visible in the GR spectrum. The wavelength at (iii) 1647 cm$^{-1}$ and the sharp peak at (iv) 1257 cm$^{-1}$ are signified to C=O and C-O stretching due to carbonyl groups in the composite structure. Peaks at (v) 2962 cm$^{-1}$ indicated sp3 C-H bonding or C-H stretching, indicating the presence of alkene in the structure corresponding to PDMS [6]. The band at (vi) 787 cm$^{-1}$ and (vii) 694 cm$^{-1}$ reveals the Si-C and Si-CH$_3$, which indicates the stretching of silicone groups with graphene, which is supported by funding from Surekha et al. (2020). This indicates that the GR interacts with the silicone matrix [7]. The original PDMS consists of Si-O-Si in the structure, which causes a peak in the control sample. However, the peak red-shifted when the GR and CB were observed due to the disruption of filler and changes in the intensity of the ECAs. It is suggested that Si-O-C is formed due to the interaction between PDMS, GR, and CB [8]. Changes in the transmittance intensity were detected due to the darker sample color, which indicates the mixing of CB and GR in the PDMS. The interaction of CB with silicone was shown at the peak (vi) 787 cm$^{-1}$ and (vii) 594 cm$^{-1,}$ representing Si-C and Si-CH$_3$, respectively. CB comprises 90-99% elemental carbon, with oxygen and hydrogen making up the other major constituents. These other significant constituents can be arranged throughout the silicone matrix in various organic functional groups [9]. With the addition of CB into silicone PDMS, spectra show some short peaks dispersed between the frequency 2000-1000 cm$^{-1}$. According to Wu et al. (2022), this is due to various groups from the CB molecular structure, including the C-O at (iv) 1257 cm$^{-1}$[10].

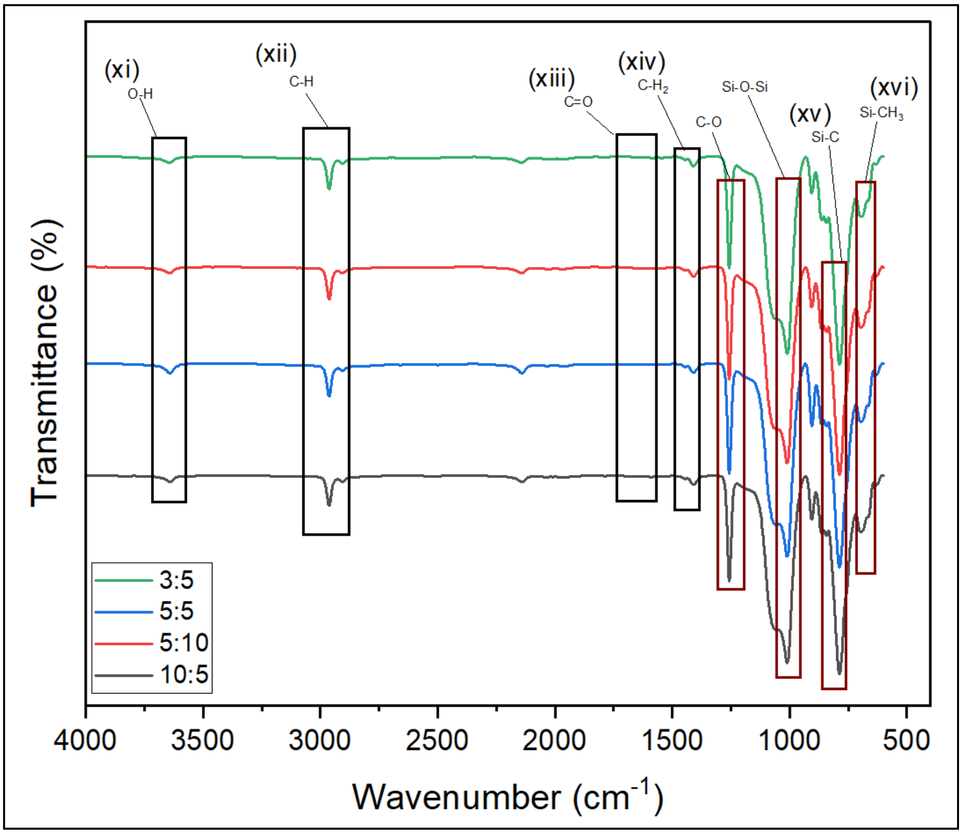

Fig. 2. FTIR spectrum peak for every ratio of synergistic graphene and carbon black GR-CB (3:5, 5:5, 5:10 and 10:5) samples.

Furthermore, Figure 2 shows the spectrum for synergistic GR-CB samples with different ratios of GR-CB, which is 10:5, 5:10, 5:5, and 3:5. In functional group region, there is stretching by hydroxyl O-H bond at (xi) 3719 cm$^{-1}$, C-H at (xii) 2140 cm$^{-1}$, carboxyl (C=O) stretching at (xiii) 1706 cm$^{-1}$, and C-H extending of sp3 carbon at (xiv) 1410 cm$^{-1}$ for 10:5 synergistic GR-CB ratio. In the fingerprint region, the interaction of silicone PDMS with GR and CB at typical Si-C and Si-CH$_3$ bands at (xv) 787 cm$^{-1}$ and (xvi) 696 cm$^{-1}$, respectively, which is in good agreement with Surekha et al. [7].

## Electrochemical Impedance Spectroscopy (EIS)

Figure 3 shows the complex impedance behavior of different ratios GR-CB for the control sample (0:0) and with the addition of GR (1:0) and CB (0:1) samples in the frequency range of 1 MHz to 1 kHz. The Warburg impedance resulted from high to low frequency, as presented in Fig. 3. It suggests a Warburg diffusion-limited process of electrochemical reaction or ion transport that is regulated by the diffusion of species in the electrolyte, which is consistent with the research conducted by Zhu et al. [11]. It also reveals that with the addition of GR and CB fillers, the tangent shifted to the right compared to the control sample, indicating that the film's resistivity decreased. At 1:0 of GR-CB, Warburg impedance formation is higher than CB at a ratio (0:1), which matches the conductivity measurements in Figure 5. The diffusion in the low-frequency domain is no longer noticeable due to the low contributions from dipolar polarization and free charge migration. The semi-infinite linear diffusion at low frequencies may imply that the interfacial impedance results from many processes, possibly connected to the polymeric film and double-layer alterations [12].

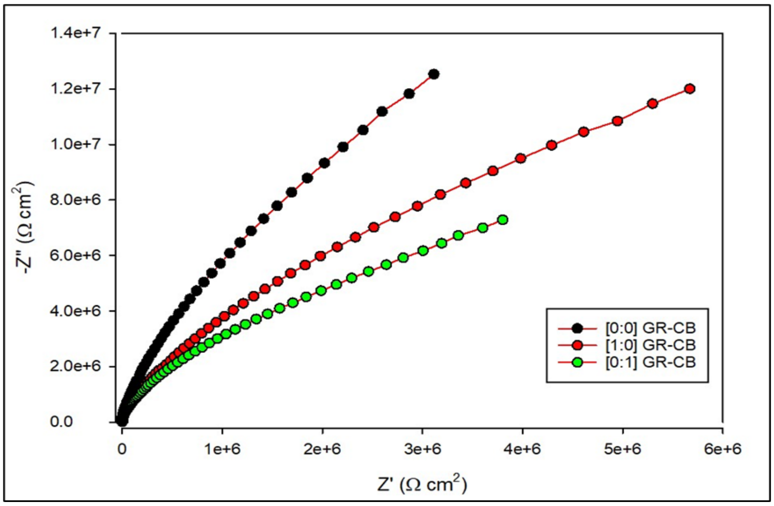

Fig. 3. Nyquist plot of the control, GR, and CB with different ratios (0:0), (1:0), and (0:1) of GR-CB.

Figure 4 demonstrates the Nyquist plots representing the semi-infinite linear diffusion for synergistic ratio (10:5), (5:10), (5:5), and (3:5) of GR-CB film. It follows Warburg impedance at 45-degree, and the physical interpretation is related to the nonuniform diffusion or multiple paths existing in the system: GR, CB, and silicone [12]. It reveals that with the addition of GR-CB, the Nyquist plot is dominated by a linear tail or unformed semicircle, reflecting a mixed kinetic and charge-transfer process consistent with [11,13,14]. The 45-degree Warburg impedance diffusion limited is present for the optimum synergistic GR-CB at a ratio (10:5), giving the lowest resistivity semi-infinite linear among the other GR-CB silicone ratios of film samples. This indicates the decrement of resistivity and conductivity increment with the higher loading of GR-CB filled the silicone ECAs structure.

Another three impedances are shifted upward from the GR-CB ratio of 5:5, 5:10 to 3:5, which shows higher resistivity than the 10:5 GR-CB ratio. This signifies the higher resistivity and lower conductivity for those three GR-CB ratios, which also indicate that the lowest conductivity among the synergism samples was found at a 3:5 GR-CB ratio, as supported by the conductivity value in Fig. 5. Nevertheless, the sample's conductivity value is the highest when filler GR-CB is added in the ratio of (10:5), as opposed to other ratios shown in Fig. 5. It is believed, the incorporation of GR-CB content reaches the percolation threshold, electrons flow through the GR-CB via the tunnel effect and subsequently conduct electricity. When both conductive fillers efficiently interact, electrons move through the surface of the conductive filler by interface contact and tunneling effect, creating a conductive network [15]. However, with the utilization of GR-CB further increased beyond the percolation threshold, the GR-CB has a high tendency to stack together and reduce the conductivity as shown in (5:5) and (3:5) ratios at 7.1 x $10^{-8}$ $\Omega^{-1}$ $cm^{-1}$ and 6.07 x $10^{-8}$ $\Omega^{-1}$ $cm^{-1}$ respectively.

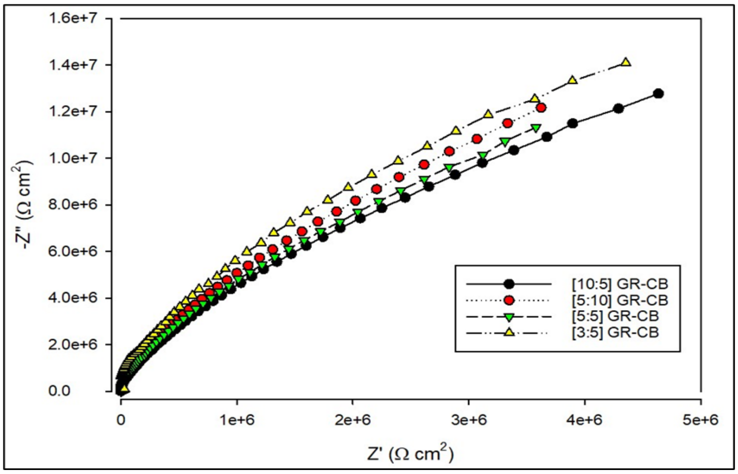

Fig. 4. Nyquist plot of the synergistic GR and CB ratio (10:5), (5:10), (5:5) and (3:5) of GR-CB.

Figure 5 also displays the conductivity of various ratio graphene to carbon black (GR: CB) ECAs. Specifically, both fillers (GR and CB) contribute to low resistance due to their enhanced electrical conductivity and robust mechanical properties attributed to their purely rigid material, making them better suited to withstand physical stress with optimum conductivity [17,18].

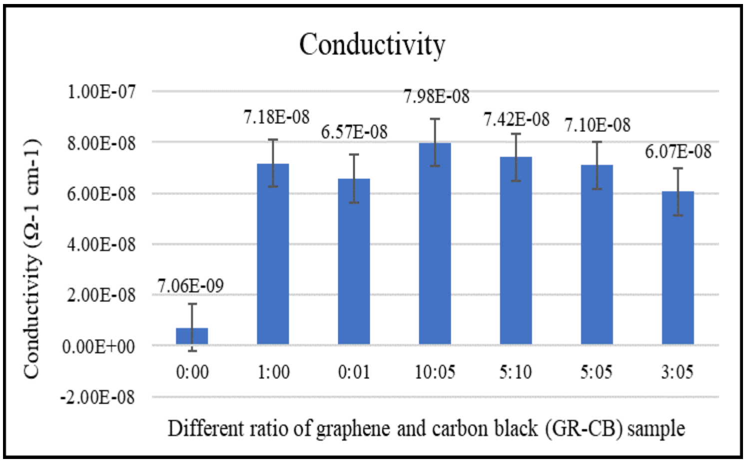

Fig. 5. Conductivity of different ratio graphene and carbon black (GR: CB) ECAs.

The conductivity of the composites depends strongly upon the fillers loading, as shown in Fig 5; the conductivity was steadily increased with the addition of fillers. This is supported by Gilleo K [19], who found that fillers utilized in ACAs improved electrical conductivity, oxidation resistance, moisture resistance, and filler dispersion. The finding is also supported by FTIR spectra showing the stretching of Si-C and Si-$CH_3$ at a range of 787 – 787 cm$^{-1}$ and 694 – 697 cm$^{-1,}$ respectively.

The control sample with a ratio of GR-CB 0:0 shows the lowest conductivity with $7.06\times10^{-9}$ Ω cm$^{-1}$ due to the silicone itself, which is highly insulating in nature. The inclusion of GR exhibits higher conductivity with $7.18\times10^{-8}$ $\Omega^{-1}$ $cm^{-1}$ as compared to CB addition, which is $6.57\times10^{-8}$ $\Omega^{-1}$ $cm^{-1}$. This is consistent with Zuliahani et al. (2021) [16], the conductivity was $1.55\times10^{-8}$ $\Omega$ $cm^{-1}$ for GR at 5% loading was higher as compared to 5% CB loading by Hasnol et al. [17] with $7.73\times10^{-9}$ $\Omega$ $cm^{-1}$. This is attributed to the high surface area and aspect ratio of GR, which can create an electrically conductive network in a polymer matrix with low filler loading [20]. CB also has strong electrical conductivities, but GR has extremely high conductivities due to its high surface-to-volume ratio or aspect ratio [10].

While the 10:5 synergistic GR and CB show the highest conductivity with the optimum value, $7.98\times10^{-8}$ $\Omega^{-1}$ $cm^{-1}$, this means the system has evolved to include more channels for electric current flow, and the electrically conductive networks have become more stable and continuous. The high quantity of conductive fillers creates an efficient conductive network in the silicone matrix, providing current and heat conduction routes [21]. Highly dispersed GR and CB among the silicone matrix formed three-dimensional conducting networks to improve the conductivity of ECAs. This is consistent with the results of Cao et al. in the synergistic filler of Carbon nanotube and CB inside Ag matrix ECAs [22]. The conductivity value for other ratios of GR-CB samples decreases after the optimum ratio, as shown in Figure 5. The GR-CB ratios of 5:10, 5:5, and 3:5 all decreased in conductivity at $7.42 \times 10^{-8}$ $\Omega^{-1}$ $cm^{-1}$, $7.10 \times 10^{-8}$ $\Omega^{-1}$ $cm^{-1}$, and $6.07 \times 10^{-8}$ $\Omega^{-1}$ $cm^{-1}$, respectively. Upon the addition of GR-CB content is less than the percolation threshold, a small number of conductive channels between conductive fillers causes the conductivity to be low. When the concentration of GR-CB particles reaches the percolation threshold, electrons flow through the GR-CB via the tunnel effect and conduct electricity. However, with the utilization of GR-CB further increased beyond the percolation threshold, the GR/CB powders overlap. When both conductive fillers efficiently make contact, electrons travel through the surface of the conductive filler through interface contact and tunneling effect, generating a conductive network [21].

## Mechanical Properties

### Tensile Test and Hardness

Figure 6 depicts the relationship between maximum tensile stress or tensile strength and Young's modulus. The addition of GR and CB filler, regardless of ratios, increased the tensile strength compared to the control sample. Stand-alone CB shows the highest tensile strength compared to stand-alone GR, with 0.24 and 0.91 MPa, respectively. Both showed an increase compared to the control sample's value at 0.12 MPa. According to previous research Zuliahani et al (2021) and Hasnol et al. (2022), the tensile strength of GR should be higher than that of CB at 5% filler loading, yielding 3.5 MPa and 0.03567 MPa, respectively [16,17]. However, GR filled ECAs films showed the lowest tensile strength. The raw material used, CB, was in nano size, which is smaller, whereas GR was in micron size, contributing to a lower aspect ratio [23]. The mixing technique is incapable of breaking the GR stackings. As a result, the crystalline nature does not manifest in the composite system.

While 10:5 GR-CB exhibits the lowest tensile strength compared to other GR-CB sample ratios at 0.52 MPa, this is attributed to the poor interaction and dispersion inside the matrix caused by combining GR and CB in one system. The ratio [5:10] of (GR-CB) produced the best tensile strength of 1.16 MPa, and the addition of GR-CB ratios 5:5 and 3:5 reduced the tensile strength to 0.96 and 0.88 MPa, respectively.

A similar trend in tensile strength was obtained for the Young’s Modulus as shown in Figure 6, which is the control samples (0:0) GR-CB ratios show the lowest value at 0.21 MPa as compared to GR-CB with higher tensile properties was [5:10] at 2.02 MPa. It provides good adherence between the filler and the matrix because graphene or carbon black and silicone have a high level of compatibility with one another. This contributes to the high rigidity and Young's modulus of the graphene-carbon black-silicone ECA. The GR ECAs film had a lower Young's modulus value than CB, with 0.4 and 1.65 MPa, respectively. Whilst the various GR-CB sample ratios other than 5:10 show a lower value for ratio 10:5 at 0.94 MPa, there is also a decrement in Young's modulus value as seen at ratios 5:5 and 3:5 at 1.71 and 1.59 MPa, respectively.

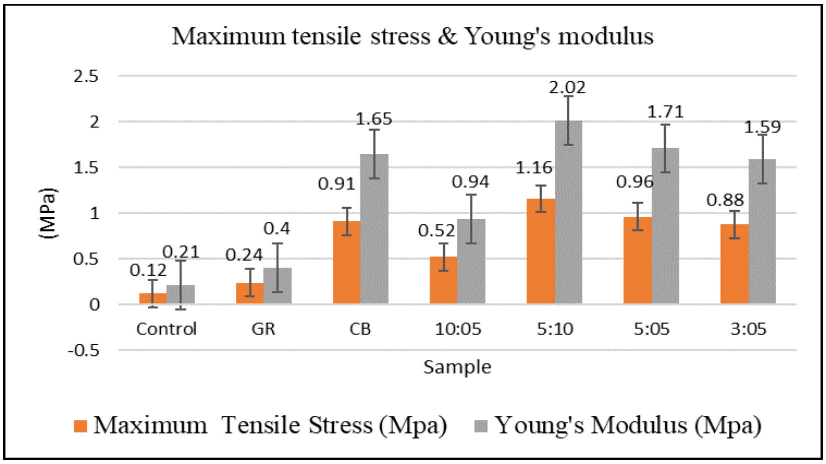

Fig. 6. Tensile strength/Maximum tensile stress and Young's modulus.

From the result illustrated in Figure 7, the filled silicone film also shows an increment from all samples compared to the control sample at 25.64 and 17.53, respectively. It is in line with Song et al. [24], the elongation at the break of composite increases with an increase of graphene nanoplatelets loading content. The higher the elongation at break percentage, the higher the ductility. The optimum elongation at break is shown at 5:10 of synergistic GR-CB PDMS film at 25.64 mm. For control GR and CB samples, CB has high elongation at break compared to GR, with 22.19 and 21.04, respectively, due to the dispersion of CB in the silicone matrix-producing absorption interfaces. The CB stress transferred at the interfaces and decreased the crack initiation rate, significantly increasing the composites' toughness [24]. Then, a different ratio of synergistic GR-CB (10:5) decreased to 21.19 mm. After the optimum ratio in the figure, the other two ratios show that the 5:5 decrease and 3:5 increase at 19.48 and 23.24, respectively.

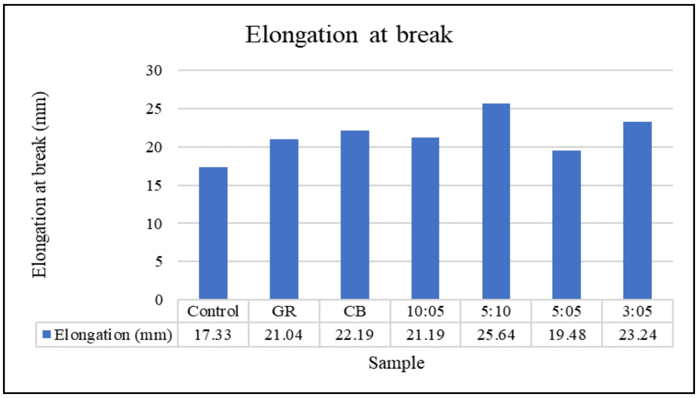

Fig. 7. Elongation at break.

As shown in Figure 8 below, the hardness result indicated that adding fillers increased the hardness value compared to the unfilled silicone PDMS ECAs film. Adding GR and CB increased the hardness compared to the control film, which is 16.74 for GR and 45.52 for CB. The composite film of PDMS filled with GR or CB makes the ECAs film stiffer and increases the hardness, aligning with tensile findings.

Following that, the 10:5 GR-CB ratio has the lowest hardness value of the four ratios in this study, with 34.78. The optimum ratio was obtained at ratio [5:10] with a value of 47.56 for the various GR: CB ratios. This could be due to the system of synergistic ECAs, where the ratio of CB particles with a size of 69 nm to GR particles with a size less than 2 m is relatively high, which could account for these findings, which are also supported by Sreekumar et al (2012) [25]. The increment of hardness contributed by the decreasing particle size related to the nano-sized CB could provide the composite with superior mechanical properties because of good interfacial adhesion between the polymer matrix and fillers [26]. Hence, the hardness of the material is also proportional to the stiffness of the material. That is why the same trend as Young's modulus in Figure 6 was obtained. This indicates that a high stiffness value corresponds to a high material hardness.

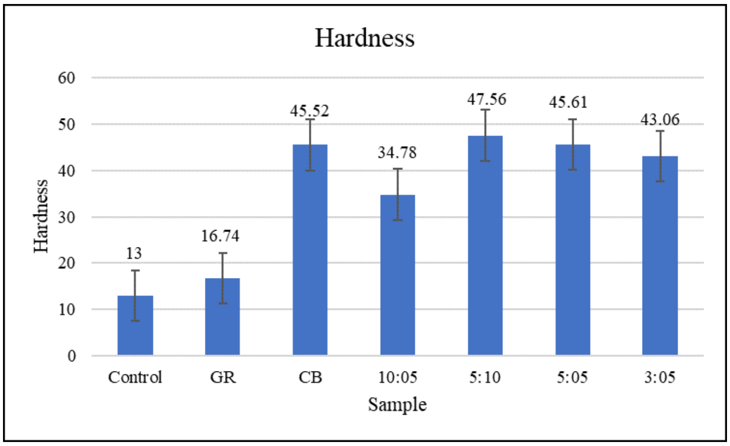

Fig. 8. Hardness of the ECAs

## CONCLUSION

In conclusion, the results of electrical and mechanical properties revealed that the incorporation of fillers, which are graphene (GR) and carbon black (CB), could simultaneously improve the conductivity, strength, and toughness of the silicone ECAs composites. The characterization of the silicone-filled GR-CB was done using FTIR spectroscopy. The spectrum confirms that the interaction between the silicone matrix with GR-CB happened. The IR spectra show a sharp peak at 687 cm$^{-1}$ (Si-C) and in the range 694 – 697 cm$^{-1}$ (Si-CH$_3$). From the electrical testing through electrochemical impedance spectroscopy (EIS), the optimum ratio with the highest conductivity value of $7.98\times10^{-8}$ $\Omega^{-1}$ cm$^{-1}$ was obtained by the synergistic effect of conductive fillers, which is GR with CB at 10:5 GR-CB ratio. When GR and CB were spread widely in the silicone matrix, they formed three-dimensional conducting networks that constructed extra conductive ECAs. The ratio of GR inside the synergistic system also has extraordinarily high conductivities due to its high surface-to-volume ratio or aspect ratio compared to CB. The mechanical properties of silicone ECAs samples through tensile and hardness tests show that the optimum ratio of synergistic GR-CB was at 5:10 ratio with the highest tensile strength, young modulus, elongation at break, and hardness with value 1.16 MPa, 2.02 MPa, 25.64 mm and 47.56 respectively. The system provides strong adhesion between the filler and the matrix due to the compatibility between graphene/carbon black and silicone. A high ratio of CB with a size of 69 nm in the system also contributes to superior mechanical properties because of good interfacial adhesion between the polymer matrix and fillers.

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## AUTHOR'S CONTRIBUTION

Muhammad Hanif Azhar carried out the research and wrote the article. Zuliahani Ahmad conceptualized the central research idea and provided the theoretical framework. Diana Nasuha Muhamad Kamsani, Adzrie Baharudin, Nur Raihan Mohamed, Sharifah Nafisah Syed Ismail, Suhaida Dila Safian, Pham Trung Kien and Muhamad Nabil Nazifi Mohd Ruhaizat had designed the research, supervised research progress and revised the article.

## CONFLICT OF INTEREST STATEMENT

The authors affirm that there are no competing interests regarding the publication of this paper.

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