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Effect of Silicone Rubber on the Properties of Epoxy/Recovered Carbon Black (rCB) Conductive Materials

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ABSTRACT

The primary focus of this study is to investigate the effect of silicone rubber (SR) content on the mechanical, thermal, electrical conductivity, and morphological properties of epoxy/recovered carbon black (rCB) conductive material. The conductive material is used to produce the electrostatic discharge (ESD) tray for the electronic packaging industry. This study investigated the effect of silicone rubber content (0, 5, 10, 15, and 20 vol.%) on the properties of epoxy/SR/rCB conductive materials, with the rCB content fixed at 15 vol.% for its optimum electrical conductivity. The silicone rubber acts as a toughening agent for epoxy. Through the fracture toughness result, it can be identified that silicone rubber plays a role in improving the toughness properties of the epoxy/SR/rCB conductive material. The optimum results for mechanical properties were recorded at 5 vol.% SR. The addition of SR to the epoxy matrix enhances the electrical properties of the epoxy/SR/rCB conductive material. The effect of thermal aging on epoxy/SR/rCB conductive materials was also studied to determine the properties of the conductive material materials at high temperatures for a long period of time. After thermal aging, the mechanical, thermal, electrical conductivity, and morphological properties of the epoxy/SR/rCB conductive material were slightly reduced.

Keywords: *Conductive materials, Epoxy, Recovered carbon black, Silicone rubber, Toughness*

1. INTRODUCTION

Nowadays, many conventional materials have been replaced by polymers in advanced applications. Due to good properties of polymer, the conductive polymer material is produced by adding the conductive filler to the polymer matrix for application in electronic field. The addition of conductive filler to the polymer matrix results in the creation of the desired qualities including high electrical conductivity, good mechanical properties, corrosion resistance, and light weight [1]-[2].

Epoxy has been widely used as a polymer matrix and has a wide range of applications because of its advantageous properties such as high thermal stability, good moisture resistance, and corrosion resistance [3]. However, due to the formation of high cross-link densities during curing, the epoxy resin may become brittle [3].

With purposely addressing to this issue, one of the successful approaching to toughening epoxy resins is by addition of small content of secondary thermoplastics [4] or rubbery [5] phase into the brittle epoxy resins.

In the study by Pakaya et al. [6], room-temperature vulcanised (RTV) silicone rubber was blended with epoxy to improve the properties of the epoxy. The findings indicate that while maintaining the high temperature resistance of epoxy resin, RTV silicone rubber may successfully improve the impact toughness of epoxy resin. Moreover, silicone rubber (SR) was used as a toughness agent to improve the impact strength and toughness of poly (lactic acid) (PLA) in the study of Yildiz et al. in 2014 [7].

The objective of this study is to investigate the effect of SR on the mechanical, thermal, electrical conductivity, and morphology properties of epoxy/SR/recovered carbon black (rCB) conductive materials with and without thermal aging. The study was focused on the effect of SR content (vol.%) in epoxy resin to determine the toughness of epoxy/SR/rCB conductive material. The performances of SR content which acted as toughening agent with fixed rCB content which acted as conductive filler were assessed by flexural, fracture toughness, electrical conductivity measurement, and thermogravimetry analysis.

2. MATERIAL AND METHODS

2.1. Materials

The epoxy used is diglycidyl ether of bisphenol A (DGEBA), grade DER 331, was purchased from Euro Chemo-Pharma Sdn. Bhd. The hardener, diethyltoluendiamine (DETDA) was supplied by Shandong Aonuo New Material Company Ltd. The rCB powder with size of 1500 mesh (around 8 μm) was obtained from Eco Power Synergy Sdn Bhd. The SR powder (KMP-594) with a 5 μm average particle size, which is manufactured by Shin-Etsu Chemical Co., Ltd., was used as toughening agent when added into epoxy resin.

2.2. Sample Preparation

Epoxy/SR/rCB conductive materials were prepared with solution mixing, in which a weighted amount of SR powder was added to epoxy resin and compounded with fixed rCB, which was obtained from a previous study. Different amounts of the SR powder were added, including 0, 5, 10, 15, and 20 vol.% with epoxy resin, and mixed for 5 minutes at 50 rpm using a mechanical stirrer. After that, 15 vol.% of rCB was slowly added to epoxy/SR by stirring with a mechanical stirrer for about 10 minutes. The DETDA was then added to the epoxy/SR/rCB mixture and mixed with a magnetic stirrer for 10 minutes at room temperature. After that, the epoxy/SR/rCB mixture was cast into a mould and degassed to eliminate any air bubbles present in the mixture for 10 minutes. The samples were then subsequently precured at 100 °C for 2 hours and post cured at 100°C for 4 hours.

2.3. Characterizations

The viscosity of mixture was measured by Brookfield viscometer under ASTM D4016 with a rotation speed ranging from 2.5 rpm to 50 rpm. Electrical bulk conductivity measurement was performed on a circular shaped specimen at room temperature. The measurement was carried out in accordance with ASTM D257 by using multimeters. The flexural test under three-point bending method was performed using the Instron Universal Testing Machine 5569 by following the standard procedure stated in ASTM D790. The crosshead speed was set at 2.38 mm/min. The standard that used in fracture test under tensile mode was followed the procedure as stated in ASTM D638 and was performed using Instron machine, Model 5569 at crosshead speed of 1 mm/min. The scanning electron microscopy (SEM) study is performed on a JEOL JSM-6460 LA at activation voltages of 10 kV and 50 kV, to observe the surface morphology of epoxy/SR/rCB conductive material materials. Thermogravimetry analysis was conducted by using TA Instruments Q 500. The analysis is conducted by placing 5 mg of the specimens in a platinum pan and heating it at a rate of 10 °C/min under nitrogen purging (50 mL/min) in a heating chamber with temperature ranges of 50 °C to 500 °C. Thermal aging was conducted by referred to ASTM D3045 by putting the specimens into an oven at 100˚C for 72 hours.

3. RESULTS AND DISCUSSION

3.1. Viscosity

Figure 1 shows the effect of rotor speed from 2.5 rpm to 50 rpm and different SR content on the viscosity of the epoxy/rCB mixture. As rotor speed increases, bonding between epoxy and filler breaks, resulting in decreased viscosity. The same observation was observed for the epoxy/SR/rCB mixture, which varied at different silicone rubber (SR) contents as polymer materials have non-Newtonian behaviour [8], [9].

Figure 2 shows the effect of silicone rubber content on the viscosity of the epoxy/SR/rCB mixture at 15 vol.% of rCB content. The viscosity of the epoxy/SR/rCB mixture increases with increasing SR content due to the interaction between the polymer chain and particles. The epoxide group in epoxy resin reacts with the siloxane group in silicone rubber, causing particles to contact closely, as shown in Figure 3. The nucleophilic oxygen atom of the siloxane attacks one of the carbon atoms in the epoxide ring, breaking the C-O bond and leading to the formation of epoxy/SR. This interaction results in an increase in viscosity as the SR content increases. The addition of SR content with a long back bond chain contributes to the increased molecular weight of the mixture, further increasing the viscosity of the mixture [10].

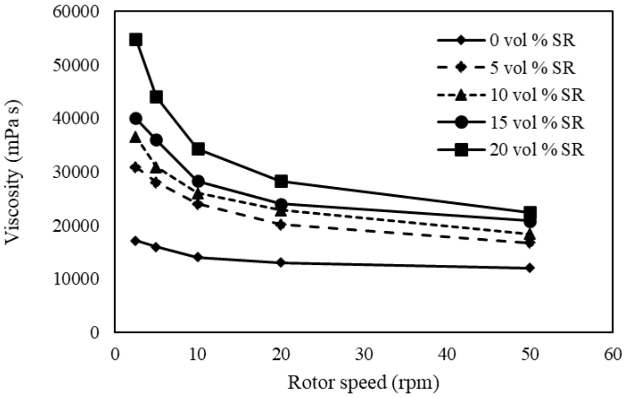

Figure 1 Effect of rotor speed on the viscosity of epoxy/SR/rCB mixture with different SR content

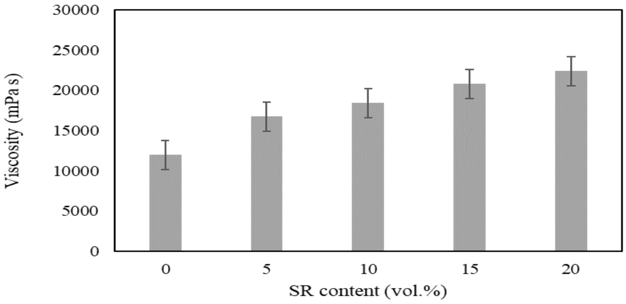

Figure 2 Effect of SR content on the viscosity of epoxy/SR/rCB mixture with 15 vol.% of rCB content at rotor speed of 50 rpm

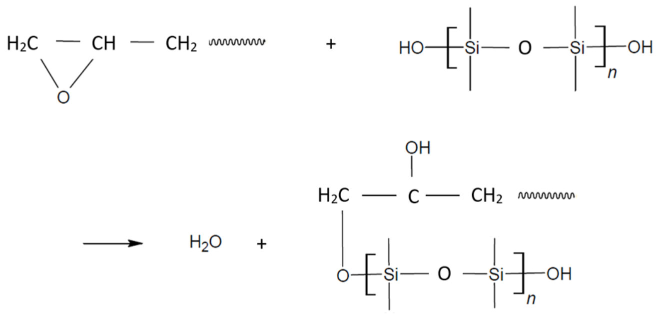

Figure 3 Chemical reaction between epoxide and siloxane groups [10]

3.2. Electrical Conductivity

Figure 4 illustrates the effect of SR content on the bulk electrical conductivity of epoxy/SR/rCB conductive material at a fixed 15 vol.% rCB content with and without thermal aging. The bulk conductivity of the epoxy/SR/rCB conductive material increased with increasing SR content. This is because the SR particle aligned the rCB particle in the epoxy matrix to form the conductive path easily, enabling the epoxy/SR/rCB conductive material to achieve higher bulk electrical conductivity.

The bulk conductivity of the epoxy/SR/rCB conductive material increases from 0 vol.% to 10 vol.% of SR content, indicating that high SR content may affect the conductive material's conductivity. At 10 vol.%, the conductive material has the highest conductivity of 1.54×10^{-3} S/m, reaching the maximum percolation threshold. However, as SR content increases, it creates an insulating effect, diluting the conductive network and reducing the conductive material's conductivity. The conductivity of the epoxy/SR/rCB conductive material drops at 15 vol.% and increases at 20 vol.% of SR content due to agglomeration, poor distribution, and dispersion of filler in the epoxy matrix. Agglomeration causes uneven distribution, leading to the formation of insulating regions within the conductive material. At 20 vol.% SR, conductivity slightly increases due to more agglomerations, giving higher conducting phases. However, these changes are still considered insignificant, falling under the same magnitude at $\times 10^{-3}$.

Figure 4 also shows the effect of thermal aging with different SR content on the electric conductivity of epoxy/SR/rCB conductive material at fixed rCB content. The bulk conductivity of epoxy/SR/rCB conductive materials that undergo thermal aging is lower as compared to those without thermal aging from 0 vol.% to 15 vol.% SR. This is due to further crosslinking, which reduces charge carrier mobility and decreases conductivity [12]. At 20 vol.% of SR content, the conductivity of conductive materials with thermal aging is almost similar to that of those without thermal aging. Thermal aging can remove impurities, enhance rCB dispersion, and promote better interfacial interactions between components [13]–[16], resulting in a more efficient conductive network and higher conductivity compared to the non-aged state. However, the overall results show that the electrical conductivity without and with thermally aged samples is falling at the same magnitude ($\times 10^{-3}$) and the electrical conductivity is not influenced much by the effect of thermal aging at 100 °C for 72 hours.

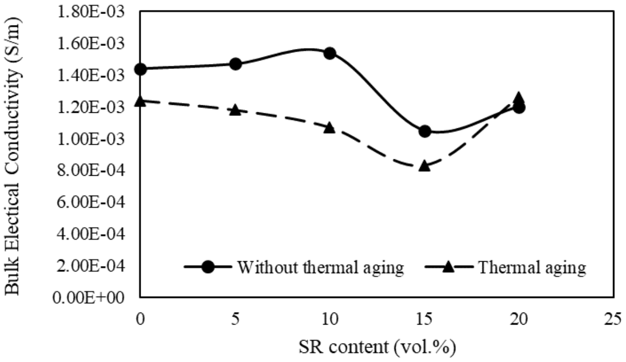

Figure 4 Effect of SR content on the bulk electrical conductivity of epoxy/SR/rCB conductive material at fixed 15 vol.% rCB content with and without thermal aging

3.3. Flexural Properties

Figure 5 shows the effect of silicone rubber content on the flexural strength of epoxy/SR/rCB conductive material material with and without thermal aging. The higher the SR content, the lower the flexural strength, as SR is highly tough and elastic compared to epoxy [17]. At 5 vol.% of SR content, the flexural strength decreased to 82.51 MPa, 27.79% lower than unfilled SR (109.14 MPa). At 15 vol.% SR content, the flexural strength slightly increased by 2.02%, resulting in a synergistic effect between SR and epoxy. This synergy improves load distribution and resistance to crack propagation, leading to a slight increase in flexural strength. However, with 20 vol.% SR, the flexural strength decreased to 57.53 MPa. The compressibility of SR can result in internal voids or microcracks during curing or loading, acting as stress concentration points and reducing the material's flexural strength [13].

Thermal aging of epoxy/SR/rCB conductive materials results in a decrease in flexural strength due to the deterioration of epoxy resin [18]. When exposed to high temperatures, cross-linking or chain scission occurred and lead to the deterioration and loss of desirable properties of the epoxy matrix, affecting the overall integrity and mechanical properties of the conductive material. At 5 vol.% SR, the flexural strength of epoxy/SR/rCB with thermal aging increases by 47.90% due to the addition of high elasticity and toughness SR. However, as SR content increases from 5 vol.% to 20 vol.%, the flexural strength of epoxy/SR/rCB with thermal aging decreases. This is due to an increased rubber phase volume fraction, resulting in phase separation between silicone rubber and epoxy phases. This separation, more pronounced during thermal aging, creates voids and weak regions, reducing the load-bearing capacity and resulting in lower flexural strength [18].

Figure 6 shows the effect of SR content on the flexural modulus of epoxy/SR/rCB conductive materials with and without thermal aging. The flexural modulus decreased with increasing SR content due to the long and flexible chain of SR [19], which contributed to the lower modulus in conductive materials without thermal aging. Additionally, SR's chemical nature and lower compatibility with epoxy make it less effective for bonding rCB particles to the epoxy matrix.

The effect of thermal aging on the flexural modulus of epoxy/SR/rCB conductive materials with and without thermal aging is also evident. Thermal aging may result in the formation of carbonyl groups, which can cause brittleness in silicone rubber near the conductive material's surface [14]. As can be seen in Figure 7, the epoxy/SR/rCB with thermal aging showed fewer fracture lines, indicating more brittle behaviour compared to the conductive materials without thermal aging.

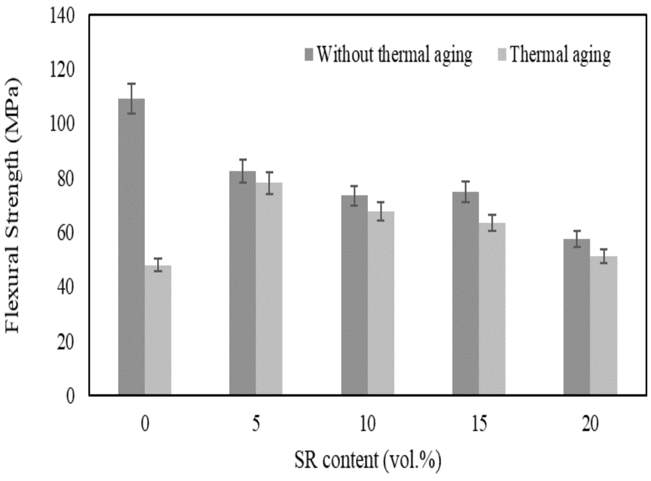

Figure 5 Effect of SR content on the flexural strength of epoxy/SR/rCB conductive material at fixed 15 vol.% rCB content with and without thermal aging

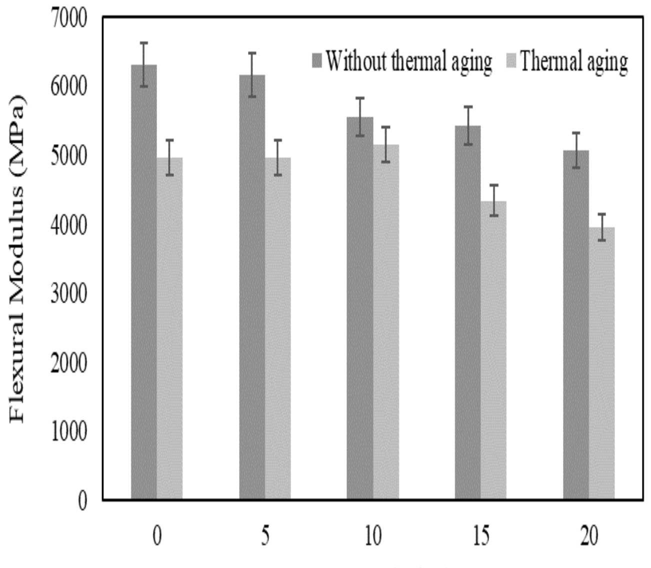

Figure 6 Effect of SR content on the flexural modulus of epoxy/SR/rCB conductive material at fixed 15 vol.% rCB content with and without thermal aging

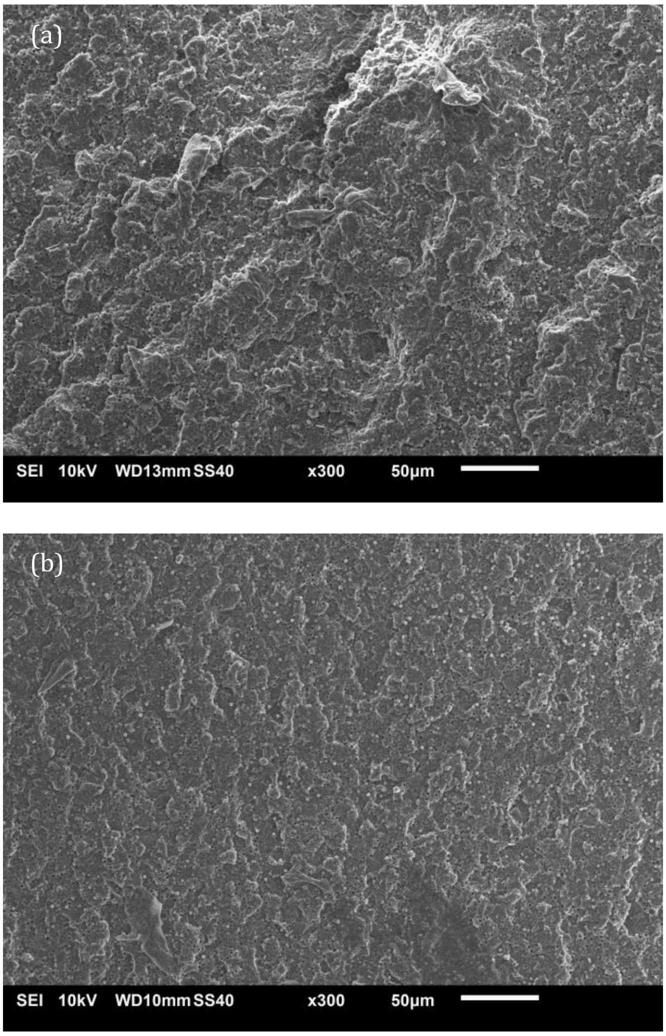

Figure 7 The SEM micrographs of epoxy/SR/rCB composites at 5 vol.% of SR (a) without and (b) with thermal aging

3.4. Fracture Toughness

Figure 8 shows the effect of different SR content on the fracture toughness of epoxy/SR/rCB conductive materials with and without thermal aging. At a fixed 15 vol.% of rCB content, the fracture toughness of the epoxy/SR/rCB conductive material increases by 35.85% at 5 vol.% of SR content, compared to the filled conductive material with 0 vol.% of SR content. This is due to the high toughness properties provided by the SR. The study by Kam et al. [20] also found that adding a small volume fraction of NR content enhances the toughness properties of pure epoxy. However, the fracture toughness decreases with increasing SR content, from 5 vol.% to 20 vol.%. Agglomeration of rCB particles in the presence of SR can lead to stress concentrations and reduced fracture toughness. Additionally, interfacial bonding between the epoxy and SR phases weakens as SR content increases, creating sites for crack initiation and propagation, resulting in reduced fracture toughness.

Thermal aging significantly impacts the fracture toughness of epoxy/SR/rCB conductive materials. At 0 vol.% SR, higher fracture toughness was observed. This is due to the curing and crosslinking reactions of epoxy resin, resulting in increased polymer chain entanglement and improved intermolecular bonding [18]. This enhanced crosslinking leads to a more rigid and robust matrix, increasing the fracture toughness of the epoxy/SR/rCB conductive material. However, the conductive material with 5 vol.% to 20 vol.% of SR decreases after thermal aging due to degradation of the SR component, resulting in a decrease in mechanical properties [6]. The main contributor to SR aging is the breakdown of molecular structure in a high-temperature environment, causing the conductive material insulator's SR to deteriorate, leading to the rubber's seal failing, moisture entering, and eventually deteriorating the conductive material rod [16].

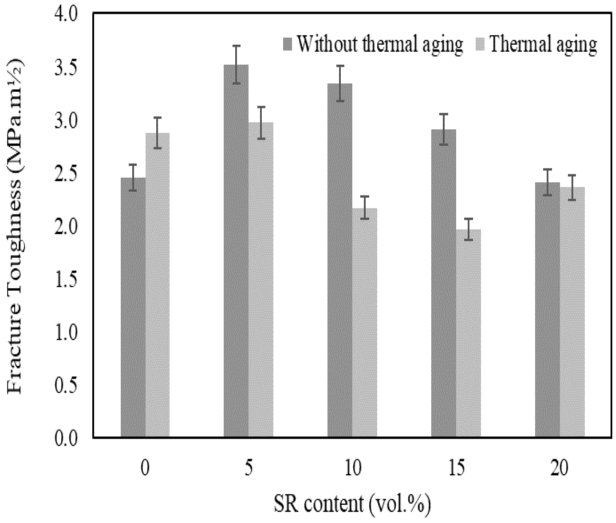

Figure 8 Effect of SR content on the fracture toughness of epoxy/SR/rCB conductive material at fixed 15 vol.% rCB content with and without thermal aging.

3.5. Scanning Electron Microscopy (SEM)

Figure 9 shows the SEM micrographs show flexural fractured surfaces of epoxy/SR/rCB conductive materials at 0 vol.% SR with thermal aging. As discussed earlier (Figure 7), the flexural fracture surface of an epoxy/SR/rCB conductive material with 0 vol.% SR content without thermal aging has a rough surface with a corrugated crack line. As illustrated in Figure 9, thermal aging accelerates interlayer delamination, causing cracking and fracture. High temperature aging damages inner layers, leading to extensive delamination and degradation [13].

Figure 10 shows that the morphological surface of the epoxy/5 vol.% SR/rCB conductive material was rougher and had more matrix rip lines than the epoxy/0 vol.% SR/rCB conductive material. This suggests that the excellent interfacial interaction between the epoxy matrix and the SR hindered flexuring, resulting in rough surfaces [21]. Figure 10 (b) clearly demonstrates the presence of two different phases, continuous epoxy matrix and spherical rubber phases, on the flexural fractured surfaces of epoxy/5 vol.% SR/rCB conductive materials. The smaller particle sizes of SR (5 μm) phases contributed to the larger surface area to interact with the surrounding epoxy matrix, allowing SR phases to more efficiently absorb and transmit tension. Additionally, epoxy/SR/rCB conductive materials filled with 5 vol.% SR had finer fracture propagation, indicating SR's larger potential as an effective energy dissipation centre [22]. Based on Figure 10 (a) and (c), the epoxy/5 vol.% SR/rCB with thermal aging showed fewer fracture lines, indicating more brittle behaviour. As observed in Figure 10 (b) and (d), which are with higher magnification, the surface roughness of the epoxy/5 vol.% SR/rCB conductive material with thermal aging showed smoother surfaces, indicating brittle behaviour.

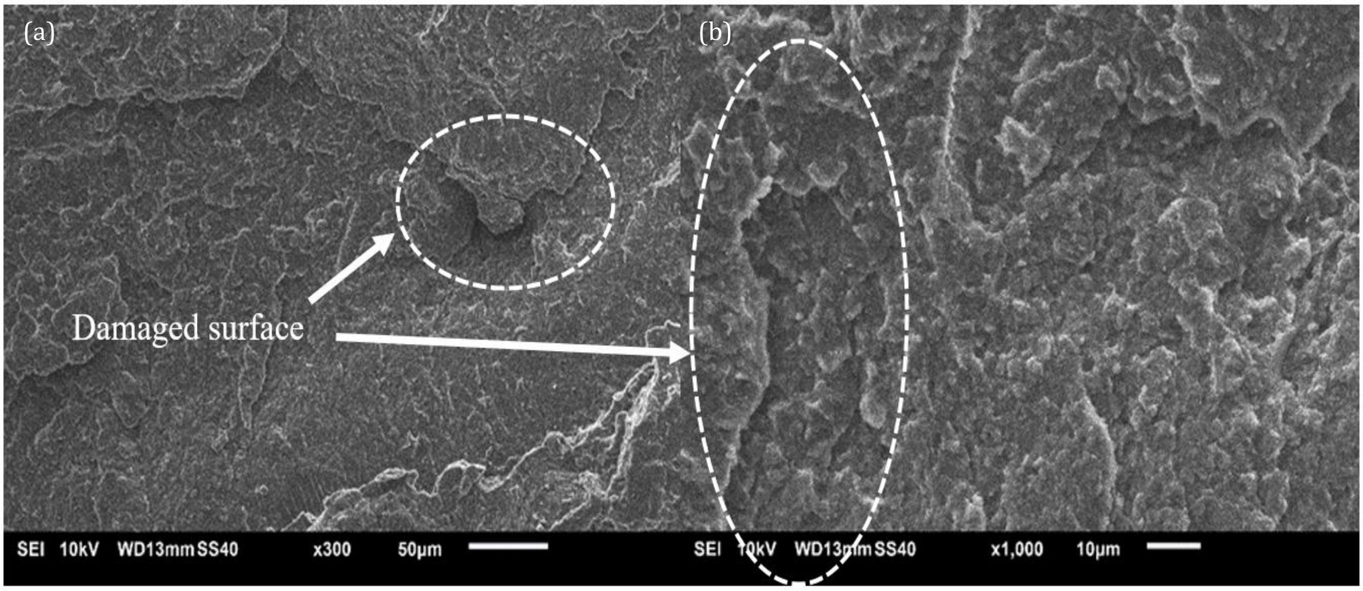

Figure 9 The SEM micrographs of the flexural fractured surfaces of epoxy/SR/rCB composites at 0 vol.% of SR with thermal aging at (a)×300 and (b)×1000 magnification, respectively

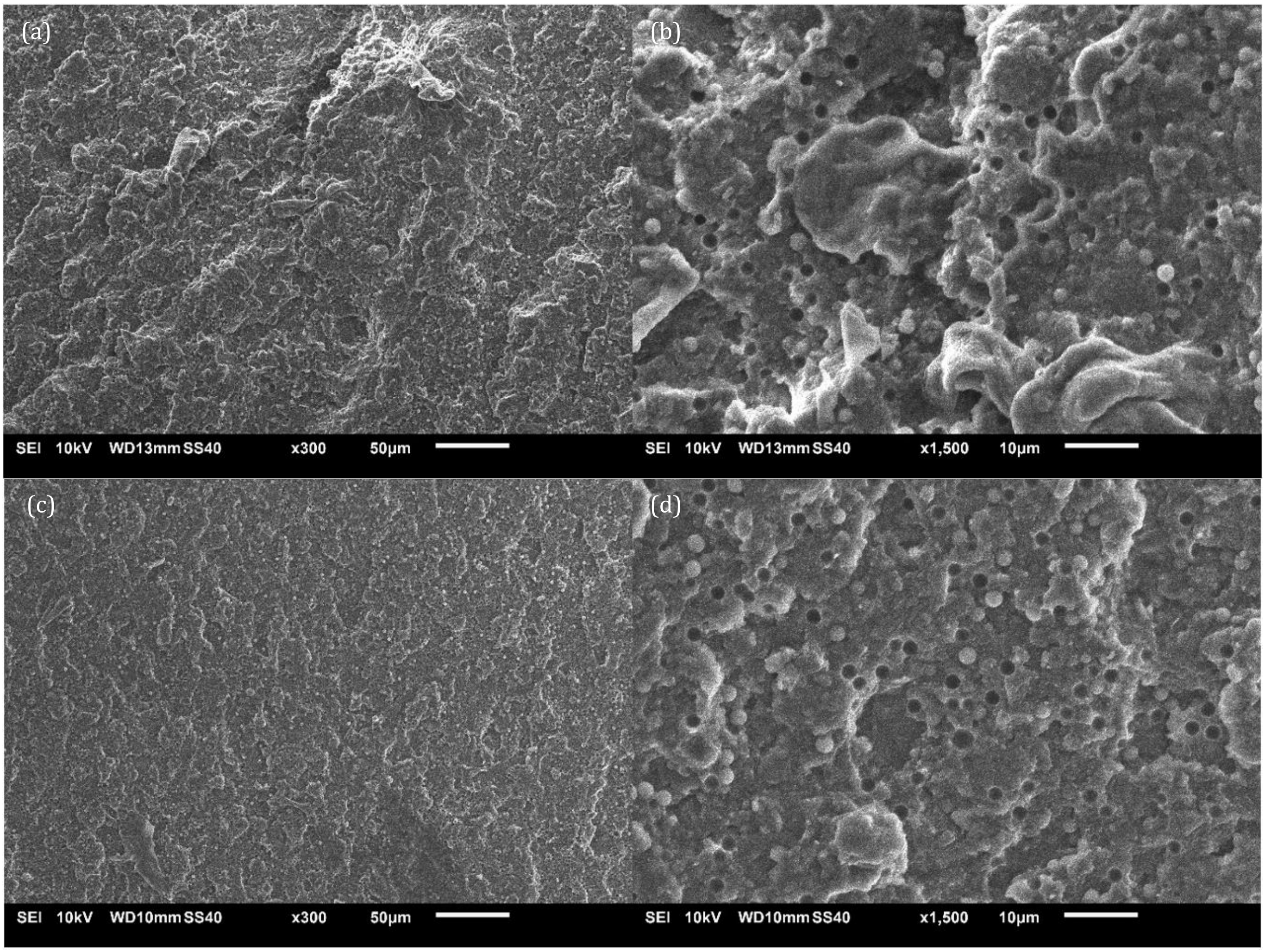

Figure 10 The SEM micrographs of the flexural fractured surfaces of epoxy/SR/rCB composites at 5 vol.% of SR (a, b) without and (c, d) with thermal aging under ×300 and ×1500 magnification

3.6. Thermogravimetry Analysis (TGA)

Thermogravimetry Analysis (TGA) was done in order to analyse the thermal stability characteristics of the epoxy/SR/rCB conductive material. The specimen with 5 vol% of SR content was selected for discussion due to its good electrical conductivity and mechanical properties. Figure 11 and Figure 12 show thermogram and differential thermogravimetry (DTG) curves, respectively. Based on Figure 11, as expected, all epoxy/SR/rCB conductive materials showed similar single-stage thermal decomposition. This indicates that the epoxy matrix's thermal degradation mechanisms did not change significantly due to the addition of SR and the induced thermal aging. The thermal decomposition temperature of epoxy/SR/rCB conductive material with 5 vol.% SR at T_5 and T_{max} was higher than that of epoxy/SR/rCB conductive material with 0 vol.% SR. This is because the siloxane bond –Si–O– in the epoxy resins has a higher binding energy than the carbon-carbon bond –C–C– [11]. This indicates that with the addition of SR, the thermal stability of epoxy/SR/rCB increases as the thermal decomposition temperature increases.

Table 1 shows that epoxy/SR/rCB conductive materials with thermal aging at 0 vol.% SR have a higher initial thermal decomposition temperature compared to conductive materials without thermal aging. This is due to the presence of crosslink precursors, which cause poor dispersion of rCB particles within the epoxy matrix. This results in less heat energy required to overcome the crosslinking [21]. However, the maximum decomposition temperature of the epoxy/0 vol.% SR/rCB conductive material with thermal aging decreases slightly. This is due to the degradation of amorphous regions in the epoxy matrix during thermal aging which leads to a decrease in the T_{max} [18].

The initial decomposition temperature of the epoxy/SR/rCB conductive materials with thermal aging at 5 vol.% SR is slightly lower as compared to the epoxy/0 vol.% SR/rCB conductive material with thermal aging. During thermal aging, the silicone rubber may undergo oxidation reactions, leading to the formation of reactive species such as free radicals [16]. These radicals can initiate chain scission or crosslinking reactions, altering the material's decomposition behaviour. On the other hand, epoxy/SR/rCB conductive materials with thermal aging at 5 vol.% SR have a higher maximum decomposition temperature. This may be due to the interaction between the hydroxyl (-OH) and carboxyl (-COOH) groups in the rCB and the polar -Si-O- bond, which increases the stiffness of siloxane chains and slows the breakdown of the epoxy matrix molecular chain [6].

Hence, the epoxy/SR/rCB conductive materials with 5 vol.% SR had more thermal stability after undergoing thermal aging as the maximum decomposition temperature was the highest, as illustrated in Table 1

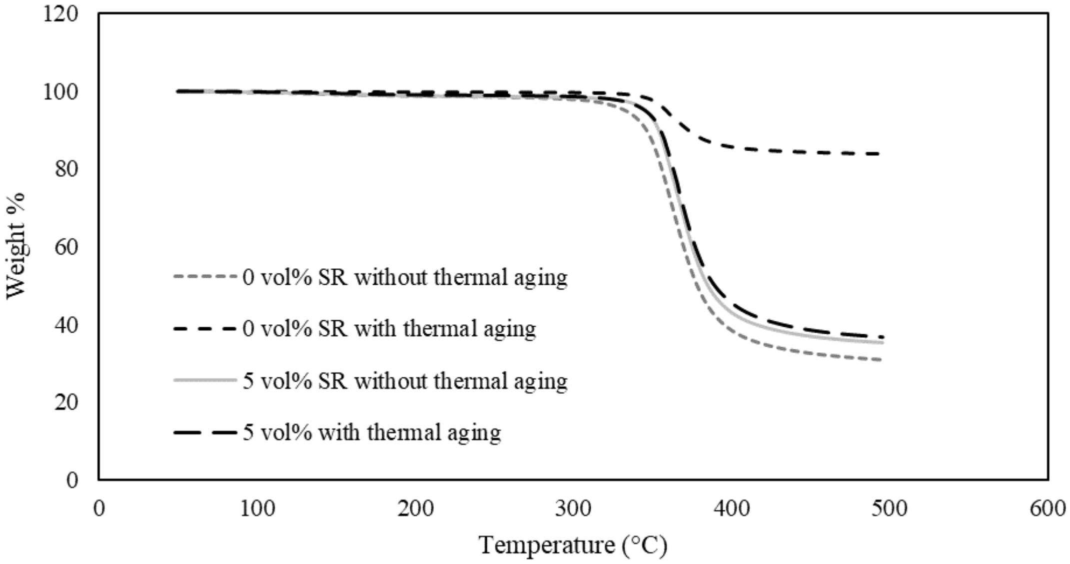

Figure 11. TGA thermogram curves of epoxy/SR/rCB at 0, 5 vol.% SR with and without thermal aging.

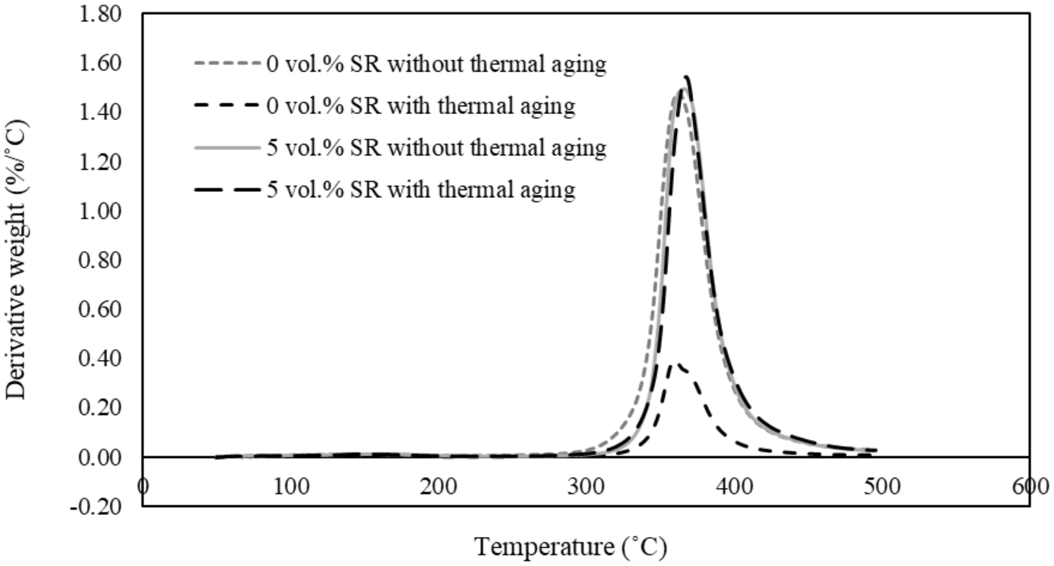

Figure 12. DTG curves of epoxy/SR/rCB at 0, 5 vol.% SR with and without thermal aging.

Table 1 Effect of SR content on the initial decomposition temperature (T_5) and maximum decomposition temperature (T_{max}) of epoxy/SR/rCB conductive material with and without thermal aging.

SR content	T_5 (˚C)	T_{max} (˚C)
0	333.07	362.41
5	345.95	366.28
0 (with thermal aging)	358.82	359.40
5 (with thermal aging)	345.53	367.40

4. CONCLUSION

In this study, various contents of silicone rubber powder were added to the epoxy/SR/rCB conductive material with a fixed rCB content of 15%. Silicon rubber powder acts as a toughening agent, which is believed to improve the electrical conductivity and mechanical properties of the material. Based on the results obtained, the electrical conductivity properties of the epoxy/SR/rCB conductive material increased as the SR was added to the conductive material. The dispersion and interaction between the rCB particles within the conductive material were enhanced by the addition of SR. At a fixed rCB content, as the SR content in the epoxy/SR/rCB conductive material increased, the flexural strength reported a decreasing trend. Besides, the addition of SR recorded an increase in the fracture toughness of the epoxy/SR/rCB conductive material. The highest value of fracture toughness of epoxy/SR/rCB was reported at 5 vol.% SR. The addition of 5 vol.% of SR to the epoxy/SR/rCB conductive material showed higher maximum decomposition temperature as compared to the epoxy/rCB conductive material at 0 vol.%. The epoxy/SR/rCB conductive material reported higher initial and maximum degradation temperatures around 345.95 °C and 366.28 °C at 5 vol.% SR.

On the other hand, the properties of the epoxy/SR/rCB conductive material with thermal aging were also obtained. The bulk conductivity of epoxy/SR/rCB conductive materials that undergo thermal aging are lower as compared to those without thermal aging. During thermal aging, the epoxy/SR/rCB conductive material may undergo further crosslinking, reducing the mobility of the charge carrier and leading to decreased conductivity. The mechanical properties of the epoxy/SR/rCB conductive material with thermal aging are lower as compared to those without thermal aging. During thermal aging, the phase separation between the silicone rubber and epoxy phases within the conductive material becomes more pronounced, and the SR component may undergo degradation, resulting in a decrease in its mechanical properties. Moreover, the epoxy/SR/rCB conductive materials with thermal aging at 5 vol.% SR has a highest maximum decomposition temperature. This behaviour may be a result of the interaction between the hydroxyl (-OH) and carboxyl (-COOH) groups in the rCB and the polar -Si-O- bond during thermal aging.

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