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The effect of graphene nanoplatelets filler size on the electrical and mechanical fatigue properties of conductive epoxy composites

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ABSTRACT

Graphene nanoplatelets (GNPs) have exceptional electrical and mechanical properties that can be used as a filler for conductive polymer. However, the size of the GNP can affect the conductivity of the conductive polymer as well as its reliability, especially when it is subjected to a different type of loading during its applications. This study is conducted to showcase the effect of particle sizes of GNP as a filler on its conductivity and the reliability of the conductive polymer composites when subjected to mechanical fatigue stress through the bending test. In this work, two types of GNP filler sizes are considered, these being the 5µm (5M) and 15µm (15M) with an epoxy binder. The initial results show that 5M GNP-filled conductive polymer composites has 92.54% and 96.28% higher in bulk and sheet resistivity than 15M GNP conductive polymer. Following the cyclic bending test, the results show that the resistivity increases as the number of cycles increases due to cracks' formation. Other than that, it was found that the rate at which the resistivity increases within the 5000 cycles of bending for 5M conductive polymer is much lesser compared to that of 15M conductive polymer. The increment in bulk and sheet resistivity is 22.70% and 17.68%, respectively, for 5M, while 15M was found to be as much as 55.90% and 36.33%. The stability on the conductivity of the smaller size particle was discussed to be due to its area of surface contact after being bent through the cycles.

Keywords: Bending, Conductive Polymer Composite, Cyclic Fatigue, Filler Size, Graphene Nanoplatelets.

1. INTRODUCTION

Recently, there is an increase in growth in the research of flexible and stretchable electronics technologies, which is influence by the rise in demand for flexible and stretchable electronic devices in the industry, such as flexible display technology based on organic light-emitting diodes (OLEDs), organic thin-film transistors, supercapacitors, and radiofrequency recognition tag sensors (RFID)[1, 2]. These flexible electronics are commonly manufactured using printing methods such as screen printing, gravure printing flexography, lithography, and inkjet printing, allowing printing circuits with custom shapes and sizes [1, 3]. Even though the printing process is very well designed to meet printing requirements, its functionality and reliability are still being investigated by many researchers and are open for improvement. One of the main aspects that need to be considered for the functionality and reliability of printed electronics is its stability towards load bending, folding, and torsion, which can significantly affect the capability of printed electronics towards its intended use [4]. The most popular conductive filler used in conductive ink or conductive polymer for the production of flexible electronics is silver, owing to its excellent electrical conductivity and used in electronic devices such as RFID [5]. However, due to the high cost of silver filler, researchers have been turning to carbon base filler which has a lower price when compared to silver filler while still being competitive as far as the conductivity is concerned [6].

The nanofiller for the conductive polymer composite considered in this study is the Graphene Nanoplatelets (GNPs). Graphene is an allotrope of carbon discovered by 2010 Nobel Prize winners Novoselov and Geim [7]. Graphene is a 2D shape nanomaterial consisting of monolayer carbon atoms arranged in a honeycomb shape [8]. The hexagonal array has high electron mobility (¬10,000 cm2 V-1 s-1 at room temperature), contributing to high electrical conductivity. It also has high mechanical strength, flexibility, and optical properties [9, 10]. Other noteworthy properties of graphene include its wide surface area, which in theory can reduce the resistance in a conductive composite by increasing the surface contact area between filler particles [11, 12]. Graphene also has a high young's modulus (50GPa) and a thermal conductivity of 5000 W/m.K at 27°C [12]. All these properties are the reasons why researchers are attracted to use graphene as a conductive filler for conductive ink or a conductive polymer.

For this study, the type of graphene that was used was graphene nanoplatelets (GNP). GNP is a graphite thin sheets which have the thickness of lesser than 100 nm. As stated before, it has very good mechanical, electrical and thermal properties and the most common way of using its extraordinary properties is to disperse it into various material matrices such as polymers [13]. The substrate that was used is polyethylene terephthalate (PET). PET is semi-crystalline material which means that it will remain solid until a certain amount of heat is absorbed, after which it will quickly transform into a low viscosity liquid. PET also have a high strength and transparency [14]. The reason of using PET as a substrate for this study is due to its flexibility to use in the cyclic bending test.

In this study, the GNP fillers are varied in size to study the difference in resistivity and the dispersion of the fillers in an epoxy binder. As reported in past researches work by Banfield [11] and Jasmee [15], the filler size can play a role in the electrical resistivity of the conductive polymer due to the contact area between the fillers. In the studies conducted by Kim and Moon [16] and Ye [17], different fillers' sizes were considered in their conductive ink. It was found that the difference in size affects the resistivity of the conductive ink. Kim and Moon [16] argued that the bigger particle size has a higher resistivity. Contrary to that, Ye [17] claimed that larger particle size has lower resistivity. As we can see, both of this study used a silver filler. There is a lack of study on the effect of filler particle sizes using graphene nanoplatelets. Thus, this study will look more towards the characteristic of the different GNP sizes in a polymer composite.

The preliminary stage of the current research is focused on the effect of different graphene filler particle size on the resistivity of the conductive polymer composites. Following this, the functionality of the samples subjected to cyclic loading (cyclic bending) will be characterised. As mentioned in the introduction section, a flexible electronic main requirement is stability towards bending, stretching, and torsion. This cyclic bending test can prove the stability of a device and its potential use in a flexible system by comparing the resistivity of the sample before and after the cyclic bending test [18]. Here, the samples are subjected to bending stress up to 5000 cycles to evaluate the change in resistivity.

Therefore, this study aims to evaluate the effect of the GNP filler size on the electrical conductivity of the conductive polymer composites subjected to mechanical fatigue stress. In addition, the wettability of the samples was measured before and after the cyclic test, using a contact angle test where the angle between a drop of distilled water and the surface of the sample is measured [19]. The importance of measuring the wettability of a conductive polymer is to know if it has a high moisture absorption which can cause the degradation of the conductive polymer [20].

2. MATERIAL AND METHODS

This section presents detailed information regarding the formulation and characterisation of graphene-filled conductive polymer composites with different particle sizes. The conductive

polymers are then printed onto a polyethene terephthalate (PET) substrate to test the cyclic bending test. The first observation of this study is to see the difference in conductivity between different conductive filler sizes. The second is the stability of the samples under cyclic loads for 5000 cycles.

2.1 Raw Material

Graphene Nanoplatelets (GNPs) used was purchased from Sigma Aldrich, with the detailed physical properties as shown in Table 1, with two-particle sizes; these being the 5 µm (5M) and 15 µm (15M). Meanwhile, an epoxy resin named Araldite 506, supplied by Sigma Aldrich, is considered as the binder. It has a density of 1.168 g/cm3 with a molar mass of 178.5 epoxy equivalent. For the curing agent, JEFFAMINE D-230 polyester amine with a density of 0.948 g/cm3 was purchased from Huntsman Singapore Pte Ltd and has a molar mass of 60 g/mol equivalent to amine hydrogen. The substrate used is polyethene terephthalate (PET) with a thickness of 100 µm was purchased from Katco Lothmann.

Table 1 Graphene Nanoplatelets specifications

Particle Size, µm	Surface Area, m^2/g	Average Thickness, nm
5	120-150	6-8
15	120-150	6-8

2.2 Sample Preparation

This study involves multiple filler sizes with the same mixing method. The fillers were set at 15 wt.% of the total weight of the mixture, while the curing/binder mixture was set at a 3:1 ratio. The filler and epoxy were first weighed by using an analytical balance Mettler Toledo. The mixture was mixed using a centrifugal mixer, a Thinky Mixer Model ARE-310 at 2000 rpm for 5 min. Then, the curing agent was added with a specified weight percentage and mixed using the Thinky Mixer for 2 min at 2000 rpm. Here, the uncured conductive polymer was printed onto a 120 mm x 40 mm Polyethylene terephthalate (PET) with the parameter shown in Figure 1 via the stencil printing method. Three points were marked onto the PET substrate with 10 mm between each point. The point is used during the four-point probe test to find the resistivity at each point. After a set of 3 samples per filler size was obtained, the sample was cured using a Memmert UF55 oven at 130℃ at 3 hours.

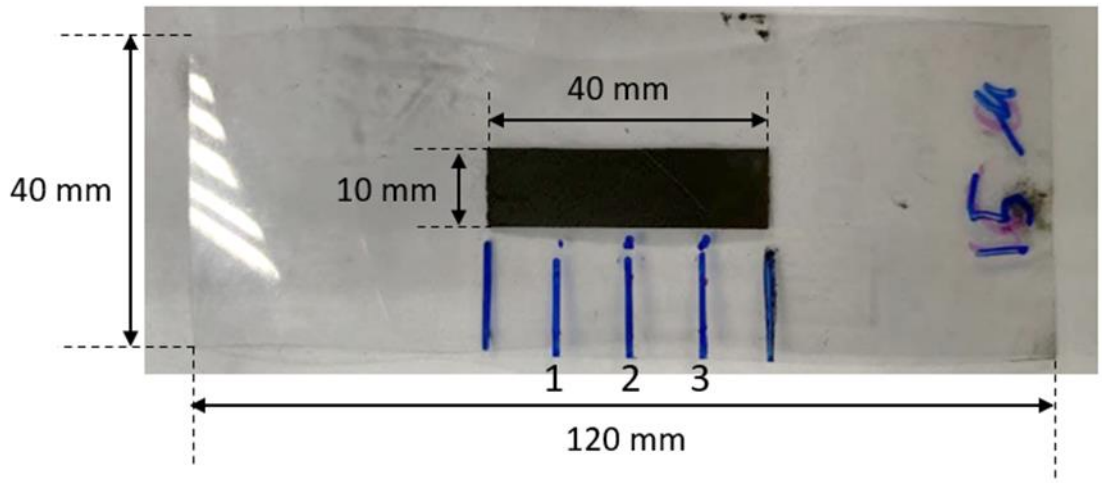


Figure 1. Sample for cyclic fatigue test (5M and15M)

2.3 Cyclic Bending Test

This test is to study the condition of the conductive polymer under cyclic fatigue. The rig that was used for this test is from the Advance Academia-Industry Collaboration Lab (AiCL). The dynamic loading was applied to each of the samples of the study in a cyclic bending form. The sample is first fixed onto a holder and set flat before running the test. The counter for the test rig is reset to zero, and the power supply is connected to the motor. After the setup is finished, the power supply is turned on. The sample was bent at a rate of 1 cycle per second and bent for 180º. The samples were bended up to 5000 cycles, and the resistivity was taken before the start of the test and for every 1000 cycles. Following this, the sample's morphology and the wettability study was examined before and after 5000 cycles.

2.4 Electrical Characterisation

Both sheet and bulk resistivities were examined to demonstrate the difference in electrical resistivity for the varied filler size and the aftereffect of a dynamic cyclic loading onto the samples. Sheet resistivity was analysed using a four-point probe machine since it is independent of a relatively low resistivity of thin-film [5]. This test used an incline four-point probe (Jandel RM3000 + Test Unit with an input range of 10 μA to 100 mA). The sample was first place below and was set in place using tape so that the sample will not move during the testing. The probe pin was lowered slowly onto the surface of the conductive polymer until there is a reading. After the setting up process was finished, the sample was repositioning so that the pin will detect the resistivity at the points in Figure 1. For each point, 3 readings were taken, which in total 9 readings for each sample and the average was calculated.

Bulk resistivity was measured using a digital multi-meter. This method used a two-point terminal where the resistivity was measured across the entire length of the conductive polymer. The negative terminal was first placed at the end of the thin film, and the positive terminal was placed at the opposite end of the conductive polymer. The terminal was placed at the mid-section of the width of the conductive polymer. The reading was taken three times to gain the average value.

2.5 Morphology Characterisation

Selected samples were examined under a scanning electron microscope (SEM) to understand the GNPs filler distribution and dispersion based on different particle sizes and the effect of the cyclic bending test on the sample. The findings were correlated with its electrical properties as well as the wettability of the sample.

2.6 Wettability Test

In the wettability study, the contact angle between the sample's surface and the liquid is measured. The contact angle will indicate the degree of wetting of the sample. The sample is first put on a stage in front of a digital microscope connected to a computer. The image is observed using the Digital Viewer software. After focusing the digital microscope, a 5μL water droplet was placed onto the sample. Here, the image was captured, and the step was repeated to place water droplets at different spots on top of the sample, with a total no of 5 times to find the average contact angle.

3. RESULTS AND DISCUSSION

3.1 Before Fatigue Test

Based on the experimental work conducted in this study, the effect of the GNP filler size on the electrical conductivity of the conductive polymer composites subjected to mechanical fatigue stress is studied. First, we investigated the filler distribution inside both conductive polymers using morphological characterisation. For both samples, as shown in Figure 2, it is evident that the 5M filler size sample has a more uniform distribution while the 15M filler size has more varied sizes.

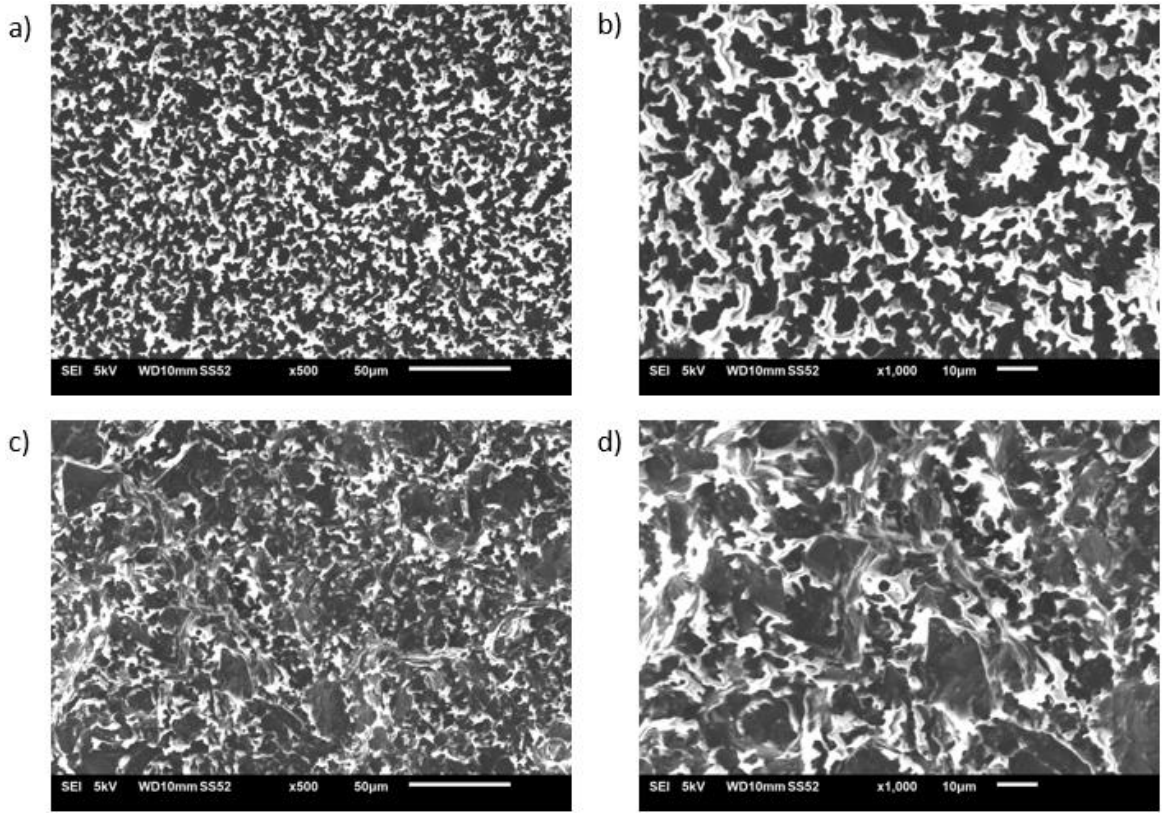


Figure 2. Surface morphology of sample: a) 5M under 500 times magnification, b) 5M under 1000 times magnification, c) 15M under 500 times magnification, d) 15M under 1000 times magnification

The GNP filler dispersion in the conductive polymer composites using two-particle size is evaluated using a simple statistical approach. The initial sheet resistivity was taken at the different points shown in Figure 1 and was used to calculate the standard deviation. An expression as shown in Eq. (1) used is given below:

$$S_x = \sqrt{\frac{\sum_{i=1}^{n}(X_i - \overline{X})^2}{n-1}} \quad (1)$$

where S_x is the standard deviation, n is the number of data points, X_i is the value of each data, and $\overline{X}$ is the mean of X_i. From Eq. (1), we obtained the standard deviation of particle size 5M and 15M, which is 119.13 and 141.30, respectively shown in Table 2. Based on the standard deviation, we can say that even though the conductivity for 5M is lower compared to 15M before the load is applied, the low standard deviation in 5M particle has provided consistency on all the surface compared to 15M. It can be suggested that the particle distribution in 5M GNP conductive polymer is more even and consistent but has a lower stacking probability of the filler particles. It is a short-range order resulting in a high resistivity value.

Table 2 Statistical Analysis

Particle Size, µm	5M	15M
Average Sheet Resistivity	14039.93	683.45
Minimum Sheet Resistivity	13889.09	521.63
Maximum Resistivity	14249.87	828.93
Standard Deviation	119.13	141.30

Looking back at the initial experimental result for the resistivity between the particle sizes, for both bulk and sheet resistivity, the larger filler particle size (15M) has a lower reading when compared to the smaller filler particle size (5M). The larger particle size sample has a bulk and sheet resistivity of 13.81 Ω.cm and 523.16 Ω/sq respectively, whereas the smaller particle size has a bulk and sheet resistivity of 185.00 14075.14 Ω/sq respectively. When comparing the differences of both bulk and sheet resistivity between the particle sizes, we see that for bulk resistivity, there is a 92.54% decrease in resistivity from 5M to 15M. For sheet resistivity, there is a decrease of 96.28% in resistivity from 5M to 15M. Here we can see that the larger particle size has a lower resistivity when compared to the smaller particle size. It is due to several factors which affect the resistivity in the conductive polymer. Firstly, when a larger filler particle is used, as shown in Figure 3, it has a more extensive contact area between the filler particles when compared to the smaller filler particle. As the GNP filler nest together, the 15M GNP filler particle will have much more surface interaction between each particle and more path for the electrons to travel from particle to particle compared to the 5M GNP filler particles reducing the resistivity of the conductive polymer. Other than that, using a larger filler particle will reduce the gap between the conductive fillers in the polymer matrix. It will allow the electron to jump from one filler particle to another through the polymer matrix at a shorter distance, which reduces the resistance of the conductive polymer [11]. Next, we will investigate the mean free pathway of electrons. The mean free pathway is the distance for the electrons to travel before it is scattered, changing its kinetic energy and direction. The larger the mean free path, the lower the resistivity of the conductor. When an electron travels through a smaller GNP particle, more particle boundaries will result in a high scattering of electrons, thus restricting its mobility [21]. By reducing the mobility of the electron, it will directly affect the electrical conductivity of the material, thus increase its resistivity [22]. The study which was conducted by Ye [17], shows the same result where the larger silver filler particles have smaller resistivity compared with the smaller size silver filler particle.

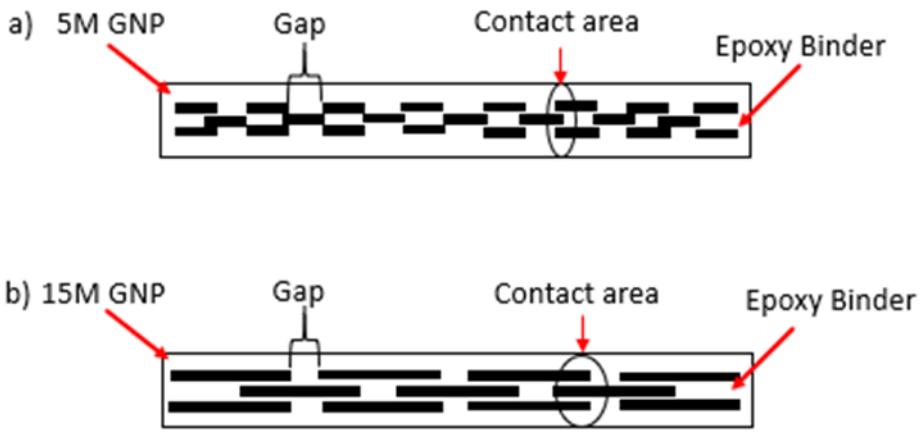


Figure 3. a) 5M GNP, b) 15M GNP conductive polymer

That being said, to decrease the resistivity of the conductive polymer with 5M GNP filler, we need to increase the weight percentage of the filler to increase the contact area between the filler particles, thus making it easier for an electron to travel through the conductive polymer [11]. In

short, it can be suggested that particle size does impact the resistivity of a composite. Larger particle size has a more extensive contact area between the particles, a smaller gap between particles to promote electron transmission, and a more significant electron mean free path, contributing to the decrease in resistivity.

3.2 After Fatigue Test

The conductive polymers were subjected to loading to observe how well the two-particle sizes contribute to the mechanical loading since it will be necessary for its applications, such as the wearability of the conductive polymer. To better understand what happened to the samples after the cyclic fatigue test, we investigate the morphology of the samples. Referring to Figure 4, we can see the formation of bent mark and cracks along the two samples caused by the cyclic bending test [23].

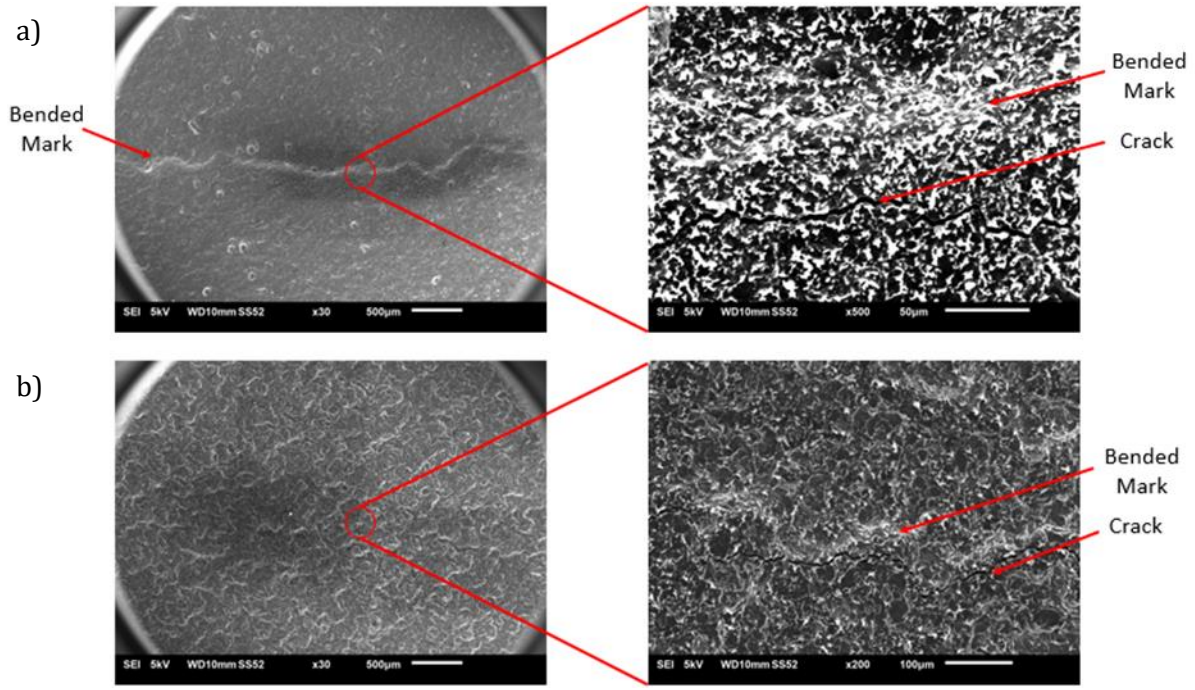


Figure 4. Morphology of: a) 5M GNP conductive polymer, b) 15M GNP conductive polymer after 5000 cycles of cyclic bending test

When the load is applied, as shown in Figure 5, deformation took place the most at the maximum point of stress. For the 5M GNP filler, as we know earlier from the standard deviation, it has a low stacking probability due to its small size. Thus, during the bending, we can see that there is some misalignment of filler particles, but there is no apparent displacement between the particles and remain in contact. For the 15 M GNP fillers, the misalignment is more noticeable. It is because of the stacking between the filler particles, making the fillers push and pull between each other more. As this displacement occurs, the filler particles will move away from each other, resulting in an increase in stress on the epoxy binder around them. It will contribute to the formation of cracks, thus the possibility of larger particle size in polymer composites having a higher tendency to form cracks which will be discussed later. As the loading continues, the crack will then start propagating and change the surface microstructure. It will then increase surface roughness which will cause the drop of contact angle, which will be discussed later. The formation of cracks in the polymer composite will increase proportionally to the number of cycles [24]. As the formation of

cracks increases, the resistivity will also increase [5]. It is due to the destruction of the electron transmission path or the increase in the distance of the electron transmission path [25].

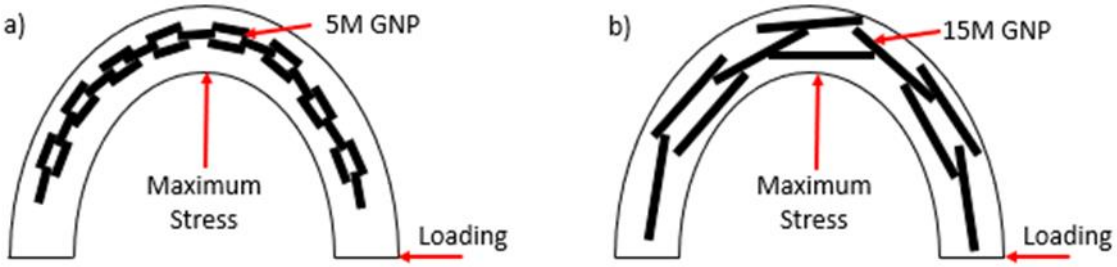
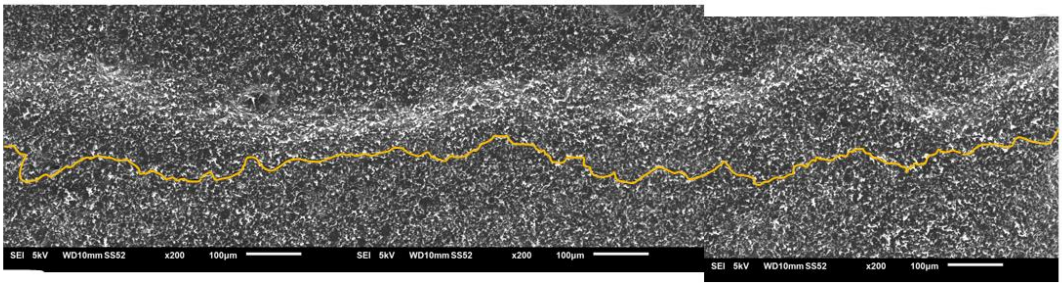


Figure 5. a) 5M GNP, b) 15M GNP conductive polymer under load

Here, it can be suggested that the cracks tend to propagates while avoiding the particles present in a composite, as reported in past research by Chen and Tokaji [26]. Meaning that, the crack grows along the conductive polymer composites between the GNP filler particles and the epoxy matrix. The assumption is confirmed from SEM analysis, as evident in the micrographs shown in Figure 6. The 5M GNP conductive polymer has a much smoother crack propagation than the 15M GNP conductive polymer, which has a much more fluctuating crack propagation. This is due to the particle size in the composites. It has a smaller particle size for a 5M GNP conductive polymer; thus, the crack will propagate along with the interface between the particle and polymer matrix, resulting in a much smoother crack formation than the 15M conductive polymer, a fluctuating crack formation. Figure 7 shows how the cracks propagate due to the filler particle sizes. We can see that, due to the particle size, the crack propagation differs, as the smaller particle size has a straighter line compared to the larger particle size. For the 15M GNP filler size, there are also branching and multiple cracks forming on the surface sample under a high load; the larger particle size has a lower crack initiation resistance [26].

To sum up, it can be implied that crack tends to propagate while avoiding particles inside a composite. The crack propagates much smoother in the 5M GNP conductive polymer than in the 15M GNP conductive polymer. Besides, larger particle size inside a composite under high load tends to form crack and multiple crack branches due to its lower crack initiation resistance.

a)



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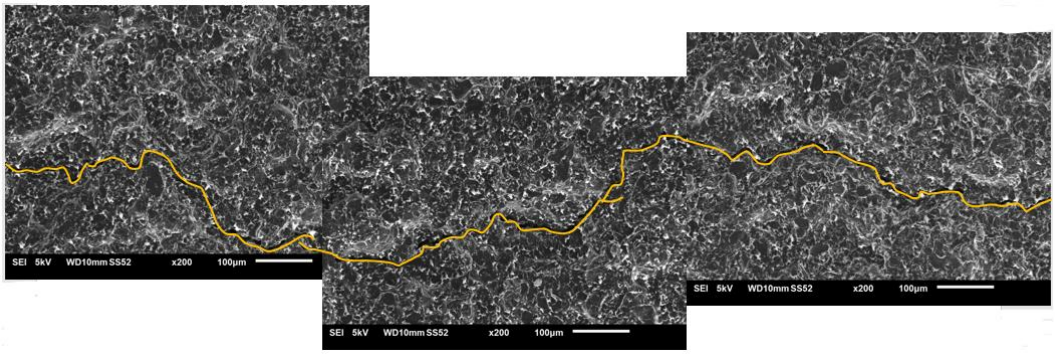

Figure 6. Crack propagation for: a) 5M GNP, b)15M GNP conductive filler

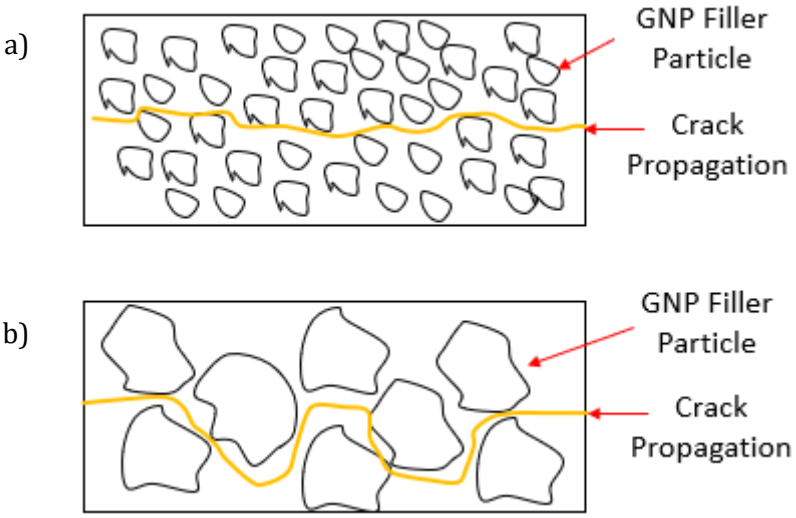

Figure 7. Drawn Crack propagation for a) 5M GNP b)15M GNP conductive filler

Looking at the crack sizes shown in Figure 8, when comparing both samples with different particle sizes filler with one another under the same magnification, the 5M GNP conductive polymer has a smaller crack size. This is due to the particle size of the polymer composite. Barbosa [27] stated that the larger the filler particle size in an epoxy binder, the lesser the fracture toughness. Fracture toughness is defined as the ability of the material to contain a crack and resist fracture [28]. It indicates that the larger particle size filler is more brittle and more prone towards fracture and will have a more extensive crack formation which supports the statement that was made earlier. Thus, the findings suggest that a more significant filler in a polymer binder particle has a lower fracture toughness, resulting in a more significant and higher number of cracks.

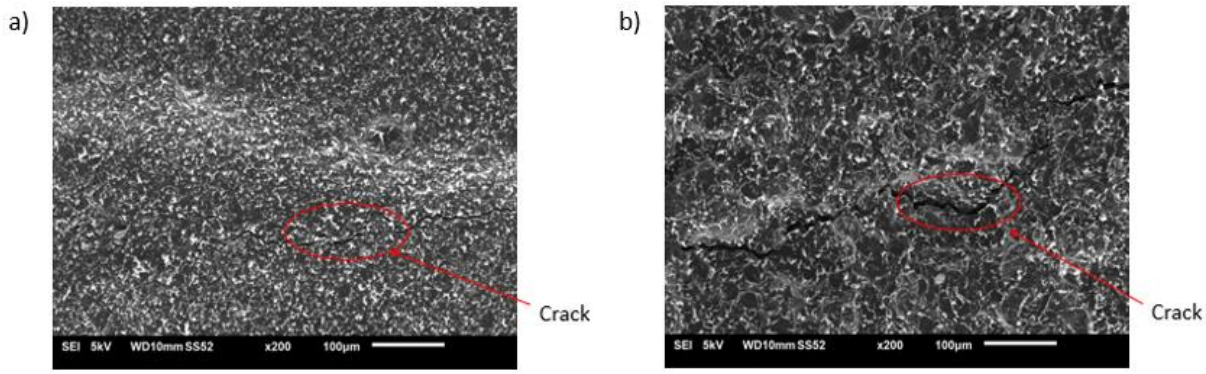

Figure 8. Crack size for a) 5M GNP b) 15M GNP conductive polymer

After we analyse the morphology of both samples, we look into the effect of those cracks on the resistivity of the conductive polymer. As shown in Figure 9, both conductive polymer composites with 5M and 15M GNP fillers increase resistance due to fatigue loading. Figure 9 shows the relationship between the sheet and bulk resistivity with the number of cycles for both particle sizes. For the conductive polymer composites with a particle size of 5M, the increase in sheet resistivity and bulk resistivity are 17.68% and 22.70%, respectively. Meanwhile, there is an increase in sheet resistivity and bulk resistivity with 36.66% and 55.90%, respectively, for the composites with 15M particle size. Thus, it implies that the resistance of the conductive polymers increases as the number of cycles increase. It is because of the formation of cracks that disturb the movement of the electron by destroying the electron transmission path or in the rise in the distance of the electron transmission path.

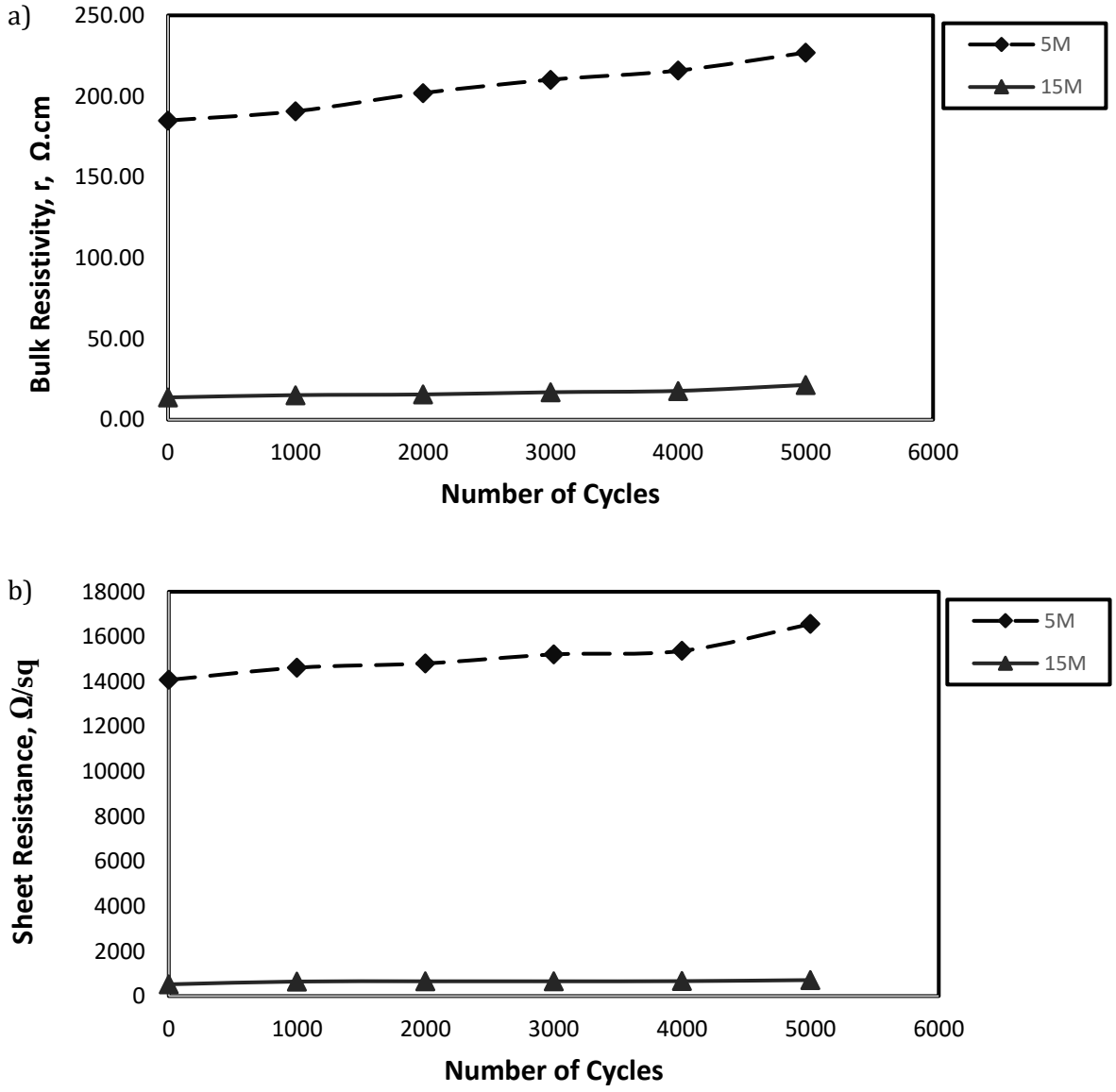

Figure 9. Graph of: a) Bulk resistivity against the number of cycles, b) Sheet resistivity against the number of cycles

In contrary to Figure 9 which shows only the difference in resistivity between two filler sizes, this section will look into the rate at which the resistivity will increase as the number of cycle increases. The rate of increase in electrical resistivity is important for the reliability of the conductive polymer throughout its use. As the conductive polymer is used in real world application, it will undergo a lot of fatigue. Thus, by understanding the rate of increase in resistivity for the conductive polymer between two filler particle sizes, we can determine which size is better for a specific application. As stated before, the bigger the size of fillers, the larger the size of cracks and the higher the number of cracks formed on the surface and the inside of the sample. Due to this, the electron pathway inside the 15M GNP conductive polymer will obstruct more due to the above. As more and larger cracks formed due to the cyclic loading, the rate at which resistance increase will also increase. To know the rate of increase in resistivity, graph of relative resistivity against number of cycles was plotted for both bulk and sheet resistivity. The calculation of relative bulk and sheet resistivity is shown as below:

$$Reltive\ Bulk\ Resistivity = \frac{\rho_b}{\rho_o} \tag{2}$$

$$Reltive\ Sheet\ Resistivity = \frac{R_s}{R_o} \tag{3}$$

where Eq. (2) and Eq. (3) is for bulk and sheet resistivity respectively. For Eq. (2), $\rho_b$is the current bulk resistivity and ρ_o is the initial bulk resistivity. For Eq. (3), $R_s$is the current sheet resistivity and R_o is the initial sheet resistivity. As shown in Figure 10, we can see that the rate of resistivity for both bulk and sheet resistivity for 15M GNP conductive polymer is higher than the 5M GNP conductive polymer. It can result in a decrease in the functionality of the 15M GNP conductive polymer over time. For conductive polymer with a particle size of 5M, both bulk and sheet resistivity increase as much as 22.70% and 17.68%, respectively. For the particle size 15M, the increase in resistivity is as much as 55.90% and 36.33% for both bulk and sheet resistivity. We can see that the larger particle size has a more significant increase in resistivity. Thus, it implies that the rate of increase in resistance for 15M GNP conductive polymer is higher than 5M GNP conductive polymer due to the larger particle size having less resistance towards the crack, thus producing a higher amount and more significant cracks, which disturbs the electron transmission path hence increasing the resistivity of the conductive polymer. Thus, even though 15M GNP conductive polymer composites have a lower resistivity overall when compare to 5M GNP conductive polymer composites, the rate of increase of resistivity for 15M GNP conductive polymer composites is higher when compare to 5M GNP conductive polymer composites. This can result in the decrease in reliability of 15M GNP conductive polymer composites over a period of time under load.

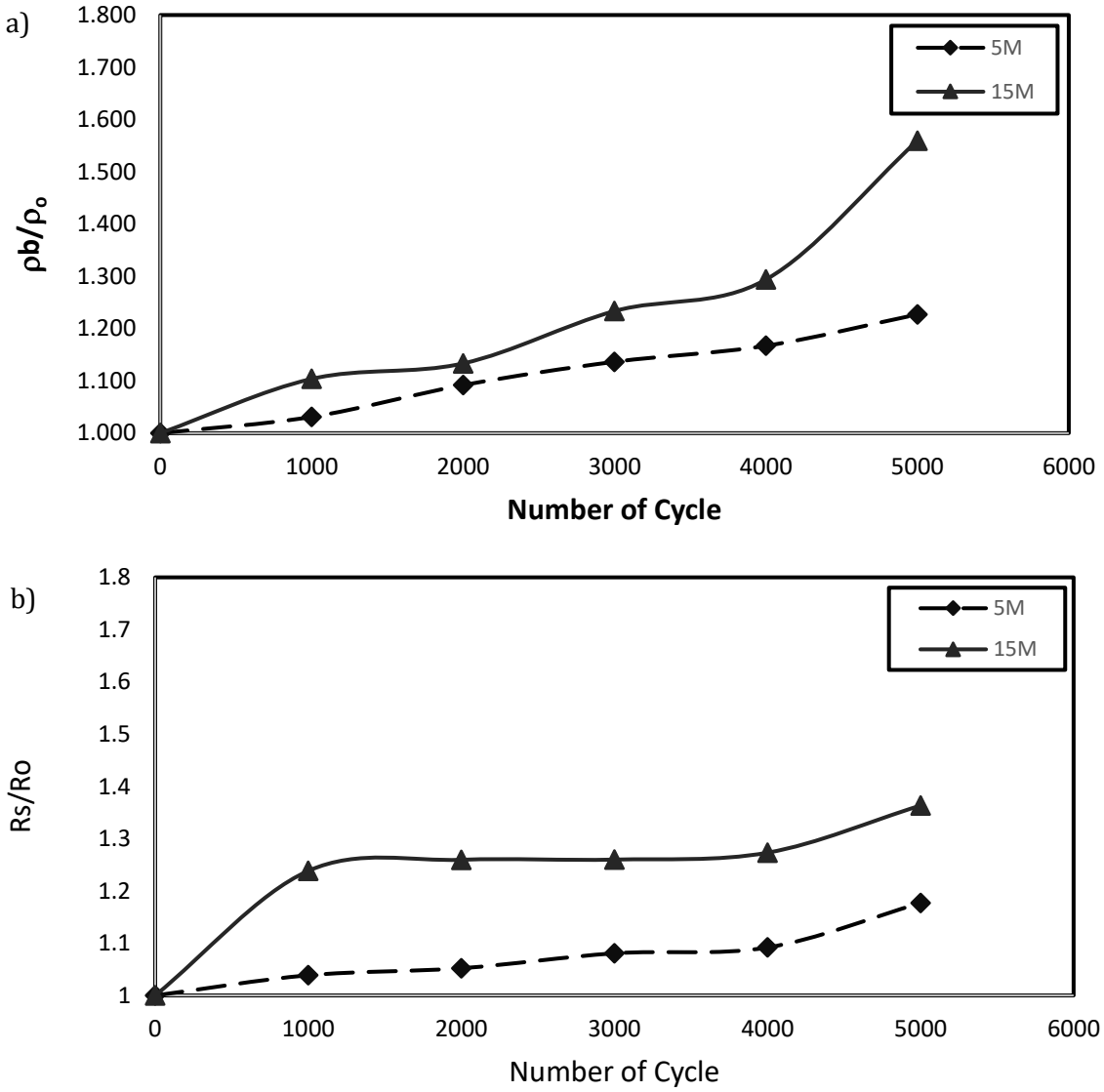

Figure 10. Graph of: a) Relative bulk resistivity against the number of cycle, b) Relative sheet resistivity against the number of cycles

The wettability test assessed the tendency of the sample to become more hydrophilic due to the cracks formed from the fatigue test. The more hydrophilic the sample is, the more moisture it can trap inside of itself. It will result in interface delamination, which will affect the conductive polymers' functionality [29]. According to the established theory on wetting, a sample is considered hydrophobic if the contact angle is above 90^0, while if it is below 90^0, it is considered hydrophilic. As the sample undergoes cyclic bending test, the sample becomes more hydrophilic; that is, it is more prone to moisture absorption [20]. As stated earlier, the crack formation will cause a drop in contact angle. Here, it is apparent that both samples with 5M and 15M GNP fillers are hydrophilic before and after the cyclic fatigue test. As listed in Table 3, the average contact angle before the cyclic bending test for the composites with GNP filler size of 5M and 15M are 70.50^0 and 80.43^0, respectively. Following the cyclic bending test, the average contact angle for the composites with GNP filler size of 5M and 15M is 61.72^0 and 72.83^0, respectively. For the 5M GNP-filled conductive polymer, there is a decrease of 12.45%, while the 15M GNP-filled conductive polymer composites have a reduction of 9.45%. Both samples are more prone to absorbing moisture after the cyclic bending test due to the cracks formed, which could yield sample failure if the samples are exposed to a more humid environment.

Table 3 Contact angle measurement before and after the cyclic bending test

5M GNP-Filled Conductive Polymer Composites		**15M GNP-Filled Conductive Polymer Composites**	
Before	**After**	**Before**	**After**
Contact angle (^{0})	Contact angle (^{0})	Contact angle (^{0})	Contact angle (^{0})
73.59	62.45	78.43	73.27
70.42	65.67	83.82	73.19
69.10	60.98	79.92	71.35
70.37	58.68	76.62	73.15
69.02	60.80	83.39	73.22

4. CONCLUSION

This study focuses on the effects of different graphene nanoparticle size on the resistivity of the conductive polymer composites before and after the cyclic bending test. Based on the experimental work attained from this study, several conclusions can be drawn: -

- Initial data prior to the cyclic bending test shows that both sheet and bulk resistivity for the conductive polymer composites with GNP filler size of 15M is lower than those of the 5M.

- The larger surface contact area for bigger particles provides a wider mean free path for the electrons to travel. Smaller gaps between particles reduce the distance for the electrons to jump, resulting in a lower resistivity value.

- However, following the bending test, the resistivity of both conductive polymers increased significantly due to the formation of cracks along their length, which destroys or increase the distance for electron transmission path.

- Between the 5M and 15M GNP-filled conductive polymer composites, the latter shows a higher resistivity since larger particles create a more extensive and higher number of cracks. Both samples also show that because of the multiple cracks formed due to fatigue, the wettability of the samples increases, which can lead to decreased functionality over time. With prolonged exposure to moisture, the samples could be delaminated, thus failing.

Therefore, it is concluded that even though the initial resistivity is higher for 5M GNP-filled conductive polymer composites relative to those of the 15M GNP-filled conductive polymer composites, the former is found to be more stable and reliable when subjected to mechanical fatigue stress.

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