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Tensile properties of Graphene Epoxy Nanocomposite
(Sifat Ketegangan Nanokompisit Grafin Epoksi)

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ABSTRACT

In this research, tensile test was conducted to reveal the notable enhancement of Young Modulus and tensile stress composites. Epoxy resin is a cross-linked polymer, thus it has major influence on the properties of polymers generally in hardness, strength, stiffness, brittleness, and better dimensional stability. Epoxy resin plays many roles in industry especially in adhesion process in automotive and aerospace composites. Discovered in 2004, graphene has revealed that it has tremendous mechanical properties and thermal properties. On top of that, addition of graphene to epoxy can lead to innovate in various adhesion industry and structural materials. In this project, an experiment was conducted in various phases which varying with graphene loading ratio, stirring speed, and stirring time. Outcome from this experiment showed that 1.0% of graphene loading with 500 rpm stirring speed and 30 minutes stirring time shows the optimum condition to enhance the measured properties. The enhancement showed are 53.03 % of Young Modulus and 51.19 % for tensile test result compared to the preliminary result. This study clearly stated the enhancement of mechanical properties of epoxy composites by merely adding 1 wt.% graphene.

Keywords: Graphene; Young Modulus; Tensile Stress; Epoxy Nanocomposites

ABSTRAK

Dalam penyelidikan ini, ujian tegangan dilakukan untuk mengungkapkan peningkatan ketara komposit Modulus Muda dan tegangan. Resin epoksi adalah polimer bersilang silang, oleh itu ia mempunyai pengaruh besar terhadap sifat polimer secara amnya dalam kekerasan, kekuatan, kekakuan, kerapuhan, dan kestabilan dimensi yang lebih baik. Resin epoksi memainkan banyak peranan dalam industri terutamanya dalam proses lekatan dalam komposit automotif dan aeroangkasa. Ditemui pada tahun 2004, grafena telah menyatakan bahawa ia mempunyai sifat mekanikal dan sifat termal yang luar biasa. Selain itu, penambahan grafena ke epoksi boleh menyebabkan inovasi dalam pelbagai industri lekatan dan bahan struktur di alam semesta ini. Dalam projek ini, eksperimen dijalankan dalam pelbagai fasa yang berbeza-beza dengan nisbah pemuatan graphene, kelajuan pengadukan dan masa pengadukan. Hasil dari eksperimen ini menunjukkan bahawa 1.0% pemuatan grafena dengan kelajuan pengadukan 500 rpm dan masa pengadukan 30 minit menunjukkan keadaan optimum untuk meningkatkan nanokomposit. Peningkatan yang ditunjukkan adalah 53.03% Modulus Muda dan 51.19% untuk hasil ujian tegangan dibandingkan dengan keputusan awal. Kajian ini dengan jelas menyatakan peningkatan sifat mekanik komposit epoksi dengan hanya menambahkan 1 wt.% grafena.

Kata kunci: Grafena; Modulus Young; Tekanan tegangan; Nanokomposit Epoksi

INTRODUCTION

Epoxy resins are a significant class of polymeric materials and one of the most versatile classes of polymers. Epoxy resins are used widely in industrial use such as electrical insulation materials, aerospace composites, and automotive primer (Sankaramoorthy et al. 2020). This epoxy resins are formed by transforming liquid polyether into infusible solids through a specific curing process.

Graphene is the allotrope of carbon in the structure of a plane of sp^2 which are arranged in a hexagonal honeycomb structure (Ghaemi et al. 2018). The bonded atoms are separated by a distance of 1.4 angstroms and the bonding energy is approximately 5.9 eV (Pop, Varshney & Roy 2012). Graphene has such unique and extraordinary mechanical properties. This material is the 2D atomic crystals and known as thinnest object ever obtained but also strongest material ever (Ahmadi et al. 2021). The mechanical properties of graphene are influenced by layer numbers and temperature (Zhang & Gu 2013). Previous theoretical work has found to study the mechanical properties of graphene and armchair graphene. The result shows that critical mechanical loads for failure and buckling of armchair ribbons are smaller than zigzag ribbons. Strength of monolayer graphene were up to 130 GPa (Liu et al. 2012). Another test found that shear strength of graphene sheet is 367 MPa, with Atomic Force Microscopy measurement of Young's Modulus of 1.0 TPa (Huang et al. 2012) .

Due to this, graphene is often added into various polymers as an attempt to increase the mechanical properties of the composite. The polymers or matrix include epoxy resin, polymethyl methacrylate (PMMA), polypropylene (PP); just to name a few. The values of Young's Modulus, tensile strength, and flexural strength of the composites were seen to increase, depending on the graphene loading as well as the matrix/polymer used (Kobyliukh et al. 2020). As reviewed by (Kobyliukh et al. 2020), the values of Young's Modulus in epoxy based polymer is observed to increase by 15%. For PMMA based polymer, incorporating graphene nanoplatelet (GNP) resulted and increase in 133% for Young's Modulus. The tensile strength is also reported to increase at 200% for linear low-density polyethylene (LLDPE). A complete review on enhanced mechanical properties values of various polymer matrix is available in the literature such as (Zhao et al. 2020) and many more.

Although there are many papers that reported the enhancement of epoxy composites by incorporating GNP, there are still relatively few that focuses on the effects of composites processing in influencing the values of mechanical properties. As such, the objective of this paper is to analyse the effects of graphene loading, stirring rate, and stirring time on the mechanical properties of graphene epoxy nanocomposites. The results will also be compared with rules of mixture and series model.

EXPERIMENTAL

Diglycidyl ether of bisphenol-A (epoxy) CP 812P were used in this experiment with the corresponding hardener CP 812P, as supplied by Oriental Option Sdn. Bhd., a local manufacturer here in Malaysia. Graphene Nanoplatetes (GNP) were purchased from TOB Group (China), specialised for thermal conductivity enhancement.

The objectives of this research is to study the effects of three important parameters such as graphene loading, stirring rate, and stirring time. Due to this, the values of stirring rate and stirring time were kept constant at 500 rpm and 30 minutes, respectively for the first stage. Graphene were first added to the epoxy at various loadings of wt.% set to 0.1 %, 0.3 %, 1.0 %, and 1.5 %. Next, hardener will be added to the mixture accordingly, at weight ratio of 2:1. The mixture was further stirred at 200 rpm for 15 minutes. Next, the mixture was poured into the 12 cm plastic mould with 1 cm diameter. The samples were left in the fume cupboard for 24 hours in the fume cupboard to ensure the samples was cured properly.

The purpose of the next stage is to evaluate the effects of stirring rate on the values of mechanical properties, by keeping other parameters constant. As such, the next experiment was conducted by varying the stirring rate to 200 rpm, 750 rpm, and 1000 rpm while keeping the stirring time at 30 minutes. The graphene loading was set to 1.0 wt.% as this was the loading that yielded the highest Tensile Stress and Young's Modulus.

For the final stage of the experiment, the stirring time was varied at 15 minutes, 60 minutes, and 90 minutes. The graphene loading and the stirring rate were set to 1.0 wt.% and 500 rpm, respectively; which was decided based on the highest Tensile Stress and Young's Modulus.

After cured, mould of the samples was cut using a cutter to take out the cured samples and further tested using Instron Tensile Test Machine. Samples have to be clamped properly to make sure tensile testing done accordingly. Extensometer were fixed at the gauge area of samples to measure the samples' elongation due to tensile testing. After installing the samples and extensometer properly, tensile machine must be in fine position to ensure the jaw does not become loose while conducting the test. Control panel of the machine were adjusted for the machine to be in fine position. Before starting the tensile test, load and strain reading need to be balanced and to be reset to give zero reading. Then, tensile test can be conducted and wait for samples to fracture and all data and information were recorded and tabulated.

The experimental result obtained in this experiment would be compared to the theoretical results. The equations that involved were the rule of mixture and the series model which represents the theoretical values of the mechanical properties. The rule of mixture was assumed to have perfect contact between particles in a fully percolating network which maximizes the contribution of the conductive phase. Otherwise, the basic series model was assumed to have no contact between particles and thus the contribution of particles is confined to the region of matrix embedding the particle (Han & Fina 2011). The equation for rule of mixture is

$$E_c = fE_f + (1\text{-}f)\, E_m \qquad (1)$$

where E_c = material property of the composite, E_f = material property of fibers, E_m = material property of matrix, f = volume fraction of the fibers. This is known as upper limit modulus in the case of elastic modulus and corresponds to loading parallel to the fibres. The inverse rule of formula then states that in the direction perpendicular to the fibers, the elastic modulus of a composite can be as low in this equation (Das & Prusty 2013). On the other hand, the equation for series model is

$$E_c = KfE_f + (1\text{-}f)\, E_m \qquad (2)$$

with K=1/5. This is called lower limit modulus and corresponds to a transverse loading. In this experiment, as long as the data collected lies between these two limits, the result will be considered as acceptable. Both of the equations presented here represents the upper bound and the lower bound for Young's Modulus.

RESULTS AND DISCUSSION

EFFECTS OF GRAPHENE LOADING

In this stage, the experiments were carried out to determine tensile strength at maximum load and Young's Modulus of the composite with various graphene loading ratio. Figure 1 and Figure 2 show the graph of tensile strength at maximum load and Young's Modulus versus loading ratio. In this phase, the stirring speed is fixed at 500 rpm with 30 minutes stirring time for different loading ratio. From this experiment, we can deduce that 1.0% graphene loading yields the highest tensile stress at maximum load which are 29.83 and 27.01 MPa. Furthermore 1.0% loading yields highest Young Modulus results which are 2413.94 and 2298.55 MPa.

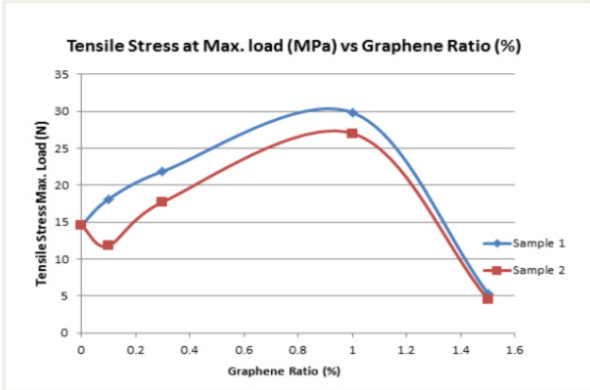


FIGURE 1. Tensile stress at maximum loading of the epoxy composites as a function of weight loading

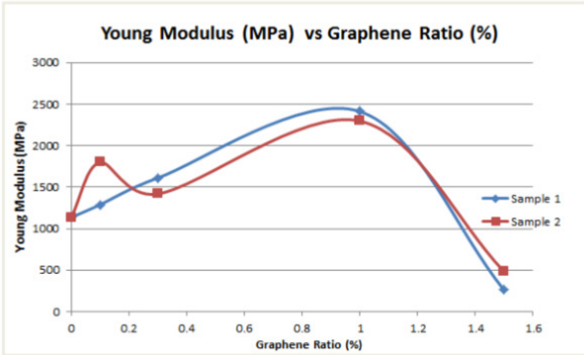
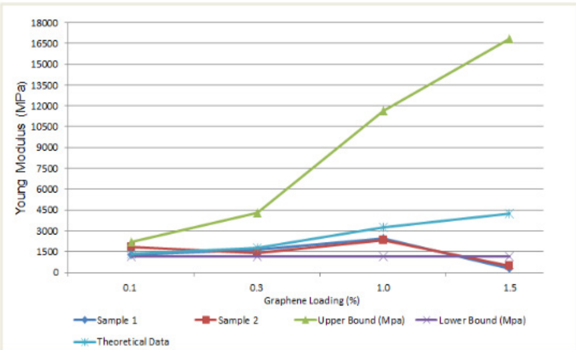


FIGURE 2. Youngs Modulus of the epoxy composites as a function of weight loading

Based on the calculations, all graphene loading from 0.1 to 1.0% loading lies between boundaries for both samples. Graphene loading 1.5% shows data lower than lower boundaries. From all data and results, it can be proven that all data are acceptable and verified. From results tabulated, 1.0% of graphene loading are chosen to the next phase of experiment which is stirring speed variables.

From experiment conducted and data collection from phase 1, the optimum loading for graphene to enhance the mechanical properties when added with epoxy resin is 1.0% loading ratio. Data taken from tensile machine shows that 1.0% loading shows highest maximum tensile strength and Young Modulus compared then other loading ratio. Thus, 1.0% graphene loading is an optimum loading ratio and to be selected to the next phase of experiment. Throughout analysis by using rule of mixture, the values of 1.0 wt.% loading of graphene showed that the values of Young's Modulus were found between upper and lower boundaries and hence are acceptable. It can be seen that by increasing the loading, it will increase the mechanical properties of the composites. However, there were some point that increasing the loading ratio of graphene will cause the graphene to agglomerate between graphene particles and lead to decreasing of mechanical properties of the composites

Agglomeration was caused by poor adhesion between epoxy matrix and graphene due to poor dispersion. Hence, the dispersion between graphene and epoxy resin will be not uniformly disperse. Agglomeration cause brittle behavior and reduces the tensile strength significantly (Ghaemi et al. 2018). Figure 3 shows the graph of Young Modulus against graphene loading including upper and lower boundaries and theoretical values. From analysis by using the rule of mixture, loading ratio that relies between upper and lower boundaries are verified and acceptable but the values that exceed or are lower than the boundaries be considered neglected and there might be some error that will occur when conducting the experiment.



FIFIGURE 3. Comparison of experimental values of Young's Modulus of the epoxy composites with rule of mixture and series model

EFFECTS OF STIRRING RATE

This set of experiment is performed to determine the effects of graphene addition to the epoxy on various stirring speed. Figure 4 and Figure 5 show the graph that shows the relation of the mechanical properties obtained varies with stirring speed. In this phase, loading ratio is fixed at 1.0% with varies stirring speed with epoxy and 15 minutes stirring speed with hardener. From the results depicted, it can be concluded that 500 rpm stirring speed shows the highest tensile stress at maximum load which are 29.83 and 27.01 MPa. In addition, 500 rpm stirring time yields highest Young Modulus results which are 2413.94 and 2298.55 MPa.

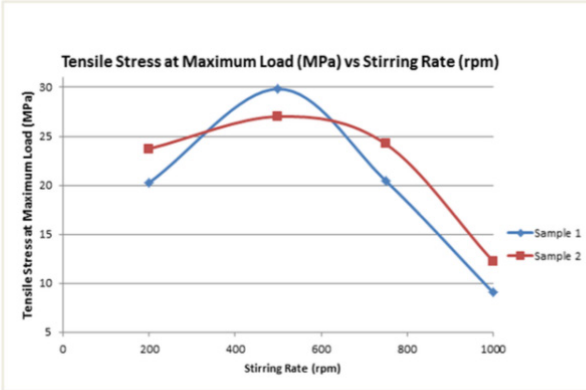


FIGURE 4. Tensile stress at maximum load as a function of stirring rate (rpm)

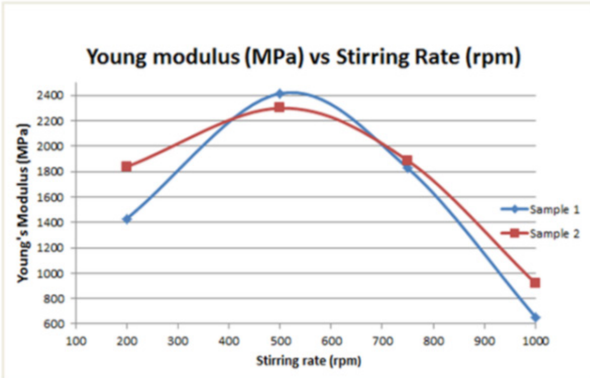


FIGURE 5. Youngs Modulus of the epoxy composites as a function of rate (rpm)

An amount of 1.0 wt. % graphene were selected to undergo tensile test that varies with stirring speed. Variation of stirring speed are 200, 500, 750, and 1000 rpm. From data obtained and analysed, 500 rpm stirring speed shows the highest mechanical properties of maximum stress and Young Modulus among other stirring speed variables. Thus, 500 rpm is an optimum stirring speed in order to enhance the mechanical properties of graphene when added with epoxy resin. When stirred at 500 rpm, it makes the dispersion of graphene and epoxy resin uniform and the particles between graphene and epoxy resin were regularly bonded. Increasing stirring time will also increase the mechanical properties of the composites but as time increases, the shear force from stirrer will cut the bonding between graphene and epoxy resin particles and lead to decreasing of mechanical properties.

By using the rule of mixture analysis, all experimental values relied on upper and lower boundaries except for 1 000 rpm which is lower than boundaries. This might be because of some errors that occurred during conducting of the experiment. The mechanical properties of reinforced composite does not depend on the properties of the composite but also depends on the alignment of particles disperse in the composites (Ghaemi et al. 2018).

EFFECTS OF STIRRING TIME

This set of experiment is performed to determine the effect of graphene addition to the epoxy on various stirring time. Figure 6 and Figure 7 show the graph that shows the relation of the mechanical properties obtained varies with stirring time. In this phase, loading ratio is fixed at 1.0% with 500 rpm stirring speed and varies with stirring time. From the results depicted, it can be concluded that 30 minutes stirring speed shows the highest tensile stress at maximum load which are 29.83 and 27.01 MPa. Moreover, 500 rpm stirring time yields the highest Young Modulus results which are 2413.94 and 2298.55 MPa.

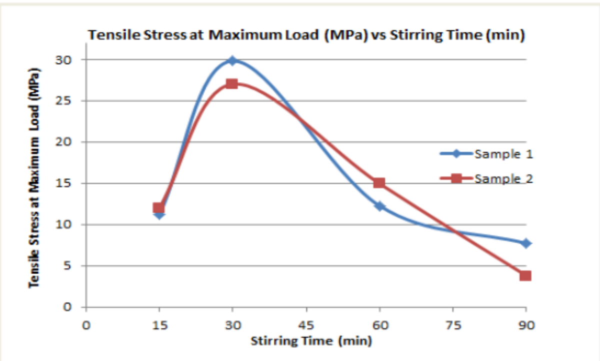


FIGURE 6. Tensile Stress at maximum load as a function of stirring time (min)

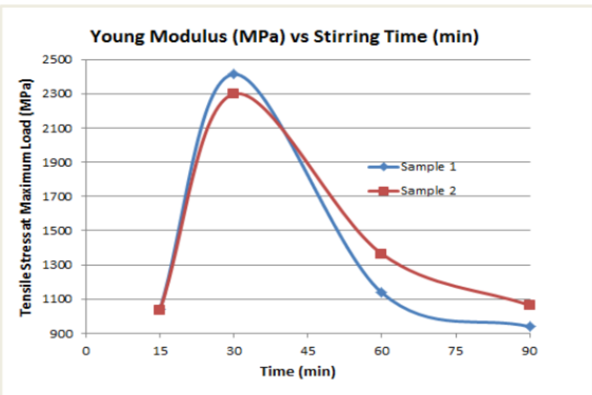


FIGURE 7. Youngs Modulus of the epoxy composites as a function of stirring time (min)

From data obtained, it can be seen that 60 minutes stirring time shows the highest mechanical properties of maximum stress and Young Modulus compared to other stirring time variables. Thus, 60 minutes stirring time is an optimum time that can enhance the mechanical properties of graphene when added with epoxy resin. From tensile test data, increasing of stirring time will also increase the mechanical properties of the composite. But when stirring time exceeds the optimum stirring time, the mechanical properties of the composites will decrease. This is because, increasing of stirring time leads to increasing of temperature during the stirring process. Increasing of temperature makes the composite undergo heat treatment in curing process since this experiment doesn't need heat treatment in curing process. Furthermore, increasing of stirring time also leads to vitrification on the composites. Vitrification is an exothermic reaction that produces heat along the process that causes the composites to become frozen since the composite undergoes curing process. Analyzing from the rule of mixture, stirring time of 15 minutes and 90 minutes does not rely in upper and lower boundaries. Both data shows lower than lower boundaries. This might be errors that occur during experiment that lead to decreasing of mechanical properties than lower boundaries data.

In this experiment, the value of K used in finding theoretical value for every volume fraction is fiber efficiency parameter (Rethwisch 2010). This means that the value of K takes into account the dispersion of graphene addition to the epoxy resin at any direction and orientation. The different fiber orientation resulting in different stress orientation. Taking fiber orientation is randomly and any direction of stress direction, value of K is 1/5 (Rethwisch 2010).

Table 1 below shows the comparison of mechanical properties of graphene epoxy nanocomposites from other research (Martin-Gallego et al. 2013) and result from this project. Graphene loading ratio used in other research were different from graphene loading ratio used in this project except for 1.5% graphene loading ratio but mechanical properties for both results showed resemblance trend where both Young Modulus and maximum tensile stress were increasing with the increase of graphene loading ratio. Since graphene loading ratio were different, but the effect of graphene loading ratio in determining the mechanical properties can be analyzed and compared although both samples for both result were prepared and cured in different method.

TABLE 1. Comparison of mechanical properties of epoxy composites

Results From Other Research			Result From This Project		
Graphene Loading Ratio (%)	Young Modulus (MPa)	Maximum Tensile Stress (MPa)	Graphene Loading Ratio (%)	Young Modulus (MPa)	Maximum Tensile Stress (MPa)
0.25	2163	65.2	0.1	1800.92	18.10
0.50	2296	64.4	0.3	1419.20	21.87
0.75	2393	68.1	1.0	2298.55	29.83
1.5	2466	68.5	1.5	485.80	5.32

Source: Martin-Gallego et al. (2013)

Results from other research (Martin-Gallego et al. 2013) does not show decreasing of mechanical properties with increasing of graphene loading ratio up to 1.5% graphene loading ratio. Result from this project showed different results where mechanical properties were increasing with graphene loading ratio but reaching to 1.5% graphene loading ratio, the results were decreasing tremendously. These differences of results are because of the curing process for this project were conducted in a fume chamber at room temperature whereas the curing process from other research were conducted with different curing temperature and process. Curing temperature for other research is at 60 minutes. Other than that, samples from other research were degassed in vacuum oven to eliminate the air bubbles within the samples. Then, dispersion of matrix and filler were examined at Transmission Electron Microscopy (TEM) in order to have good dispersion and interfacial bonds between matrix and filler.

From this difference of curing process from other research and this project, it shows that better curing process and samples preparation can lead to better results of tensile properties of graphene epoxy nanocomposites. Different stages in curing process and samples preparation were conducted to avoid agglomeration since interfacial bonds between matrix and filler were crucial for the samples to have good mechanical properties when tested although increasing of graphene loading ratio. Although results from other research still shows increasing of mechanical properties to 1.5% graphene loading ratio, the mechanical properties slightly increased from 1.0% graphene loading ratio.

This showed increasing of graphene loading ratio of more than 1.5% graphene loading ratio and more will tend to slightly increase and when it reaches a certain graphene loading ratio, the mechanical properties of graphene epoxy nanocomposites will decrease. Obtaining the same result from this project. Result from other research showed that by adding 3.0% graphene loading, the mechanical properties decreased. This is because agglomeration occured between matrix and the fillers although the samples undergo same sample preparation and curing process. Agglomeration is the condition where the filler particles were not bonded with matrix since the loading of filler were excessive to bond with matrix particles.

CONCLUSIONS

In this project, we had studied the mechanical properties of graphene addition to epoxy nanocomposites can be enhanced by various variables and parameters such as graphene loading, stirring speed, and stirring time. All variables are crucial in determining the mechanical properties of the composites in as much as the variables might change the dispersion and orientation of the graphene in epoxy. Through tensile test performed through cured samples had revealed notable enhancement of elastic moduli and tensile strength of the composites. Improvement of 53.03% of Young Modulus and 51.19 % of tensile stress pilot test result and experiment result showed that the objective of this project in enhancing mechanical properties of graphene epoxy nanocomposites is proven.

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DECLARATION OF COMPETING INTEREST

None

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