The effect of graphene addition on the Young’s modulus and tensile strength of kenaf fibre composites

Nabilah Afiqah Mohd Radzuan¹, Mihiressen Gunasegran¹ and Nisa Naima Khalid¹

¹Department of Mechanical and Manufacturing Engineering, Faculty of Engineering & Built Environment, Universiti Kebangsaan Malaysia, Malaysia

Abstract. Natural fibres such as kenaf fibres may be used to reinforce polymer matrices such as polypropylene (PP). Such composite materials are in demand for the production of interior components for automotive vehicles due to their low density (1.2 g/cm³) and good mechanical properties (Young’s modulus of 11 GPa and tensile strength of 780 MPa). The main objective of this study is to determine the effect of the addition of graphene fillers to the tensile strength and Young’s modulus of kenaf fibre composites. For this kenaf/graphene/polypropylene (PP) composites are to be studied via computer simulations using Abaqus CAE software and to compare the experimental data of Young’s modulus and tensile strength of the composite from previous researchers with the results of the simulations. For the Abaqus CAE simulations, general static and dynamic explicit analyses were conducted and set at 20 wt. % kenaf fibres, 0, 1, 3, to 5 wt. % graphene. The highest value of Young’s modulus was 1 600 MPa achieved with the material with composition of PP/kenaf/graphene 5 wt. % while the highest value of tensile strength was achieved by the composition of PP/kenaf/graphene 1 wt. % at 23.07 MPa. On the other hand, the values of tensile strength increase with the addition of graphene (1 wt. %) at first due to the improvement of interface adhesion between the polymer matrix and non-organic particles. Further addition of graphene content (3 – 5 wt. %) results in a decrease of tensile strength due to the presence of agglomeration and defects such as voids and fibre pull-outs which weakens the adhesion between the fibres and matrix. The addition of graphene to PP/kenaf composites was proved to improve the Young’s modulus and tensile strength of the materials. This improves the suitability of the materials in the production of interior components for automotive vehicles.

Keywords: Natural Fibre Composites; Kenaf; Graphene; Young’s Modulus; Tensile Strength.

1. Introduction
Pollution can be defined as the presence or introduction of pollutants (solid, liquid, or gaseous) or energy (heat, light, radiation, or sound) into an environment, be it directly or indirectly caused by human activities or natural causes [1]. This includes pollution of the air, water and land, which may cause changes to the climate and weather systems, which in turn may cause disastrous effects such as famine, drought floods and erratic weather patterns [2,3]. With the worsening of such a phenomenon, there could be significant negative effects on human life, agricultural yield, and the economy in the long term [4]. Everyday occurrences such as spills, accidents, negligence, and vandalism all contribute to pollution that leads to harmful effects to our health and environment. Although, human activities have inadvertently been causing pollution for thousands of years, this condition has worsened due to the relatively recent human modernisation and industrialisation, advancement of transportation technology, and increase in population since the 20th century [5].

Various efforts have been taken to overcome the problem of pollution due to human activities. One of the efforts pertains to the processing of composite materials used in daily life. A composite material is made up of two or more different materials: a matrix component and the reinforcing components, usually in the form of fibres [6]. The applications of composite materials include medicine, aerospace,
natural fibres such as jute, sisal, and kenaf are gaining more attention for this application. The benefits of using natural fibres in composites are they are available in large quantities, have low cost, and have mechanical properties up to par with synthetic fibres. Most importantly, these fibres are biodegradable and do not contain toxic substances [9]. Specifically, kenaf fibres are favourable as composite fillers as they have good tensile strength of around 780 MPa, tensile strength of 11 GPa, and a low density; approximately 1.2 g/cm³ [8,10]. An application of a kenaf composite, such as a kenaf/polypropylene (PP) composite would be to fabricate interior components for automotive vehicles. European car manufacturers such as BMW and Audi have already begun fabricating interior parts from natural fibre composites [11]. The low density material would help in reducing the vehicle’s mass, fuel consumption, and emissions, while having sufficient mechanical properties and being environmentally friendly [12].

To further improve the suitability of the kenaf composite, its mechanical properties can be improved, as the automotive components must be able to withstand loads in use [13]. One way to achieve this would be to add fillers to increase the Young’s modulus and tensile strength of the composite. Previous research has shown that the addition of graphene as a filler to kenaf composites is able to improve the mechanical properties [14]. However, as of yet, there is not many comprehensive studies regarding the effect of graphene addition on the mechanical properties of PP/kenaf composites, as well as a lack of computer simulations that could aid in making estimations regarding the material’s properties. Thus, the main objective of this study is to find the effect of the addition of graphene fillers to the tensile strength and Young’s modulus of kenaf/PP composites at room temperature.

2. Methodology
The research was started with preparation of the raw materials. Polypropylene (PP) powder of grade HM20/70P with particles less than 90 micrometres in size was used as the composite matrix. C-500 graphene nanoplatelets were used as fillers, with surface areas of 500 m²/gram. Micron sized kenaf fibres (size 20 mesh) were firstly treated with sodium hydroxide solution with a concentration of 6 wt. %. The fibres were soaked for three hours in the alkali solution before being rinsed with distilled water until it free from the alkali substance. This was determined by testing the fibres with litmus paper. The fibres were then dried in a heating oven for 24 hours at 80 °C. The graphene nanoplatelets and PP powder were physically mixed by hand for 5 minutes, followed by ball milling at 200 rpm for one hour using the Fritsch Pulverisette 6 machine. Next, the materials were mixed together using a Sigma Blade process at 190 °C and 45 rpm. The composition of the composite samples were 40 wt. % kenaf, with graphene content of 0, 1, 3, and 5 wt. % and the PP powder making up the rest of the composition (55, 57, 59 wt. %). The PP/graphene mixtures were poured into the machine compartment for 5 minutes to allow the polymer to melt evenly, followed by the kenaf fibres for another 10 minutes. The materials were removed and allowed to cool and harden. The hardened materials were crushed into a coarse powder using a crushing machine after that. The materials were then placed onto a mould with dimensions 175 mm × 175 mm × 2 mm. The material was then shaped with a hot press machine at 190 °C and 5 MPa for a 5-minute holding time. The moulds were then removed and allowed to cool for three hours before the composite material plate was removed. The samples were then shaped using a hand saw according to specifications of ASTM D 3039 for tensile tests, which is 250 mm × 25 mm × 3 mm. This shaped was repeated do for all the compositions of samples.

After the sample is drawn, new material should be produced in the ‘Property’ module. Values for mechanical properties (Young modulus, tensile strength, Poisson ratio) and physical properties of the sample (density). It should be noted that the program does not use units for those values, and the values entered must be consistent throughout the simulation process. After that, a new part needs to be produced with the material, and the part is assigned to the drawn component (Assign Section). Next to the Assembly module, the sample component given the material must be selected (Create Instance). After that, in the Step module, a new step needs to be created, by selecting Static, General. The time period is
reduced from one second to 1.0 seconds to shorten the simulation time, and ‘Nlgeom’ needs to be turned on (On). Thereafter, in the Contrain module, a new ‘coupling’ type barrier is set at the top of the sample. Next, a mesh for the sample will be generated on the Mesh module. The net size used is 2 mm was chosen because the results of the simulation will be more accurate. However, a size that is too small will take a lot of time to complete the analysis. The type of mesh chosen is tetrahedral type, as other types of mesh do not fit such components, and many steps need to be taken to use them. The number of elements is 1240 and the number of nodes is 2079. For explicit dynamic analysis, the values for the material should be included in the ‘Property’ module. Values for mechanical properties such as Young modulus and Poisson ratio are the same as in general static analysis. Values for plastic deformation are taken from previous studies on the strength of PP / kenaf composite mechanical properties. The value for ductile damage is (Ductile Damage) taken from previous studies as well. The same steps for the Assembly are taken, followed by the Step module. The Explicit Dynamic Type is selected, and the following values are entered for Mass Scaling with the reference point is also set at 2 cm from the top center of the sample. In the interaction module, the constraint (Constrain) type Coupling (coupling) is selected. The same steps for the Mesh and Job modules as in general static analysis are taken. Results will be generated later in the Visualization module.

3. Result and discussion

3.1. PP/Kenaf/Graphene composite samples

After remove the mould from the hot press machine, the material was left to be cooled for 3 hours to avoid defects such as cracks and warping before being removed from the mould and shaped into test samples for tensile testing as per the standards of ASTM D3039 using a hand saw. It had found that the addition of graphene changes the physical appearance of the composites, causing them to become considerably darker in shade, even with only 1 wt. % graphene addition. The samples were shown in Figure 1 (a) – (d). found that the PP/kenaf sample could not be formed as the composite material plate formed by the hot press process shattered upon opening the mould. This may be due to weaker adhesion between the PP matrix and kenaf fibres, causing the material to have lower mechanical properties. Previous studies show that the composite comprised of just PP and kenaf have a lower values of both Young’s modulus and tensile strength compared to PP/kenaf/graphene composites [14]. Besides that, the defect may have been due to insufficient amounts of wax applied on the moulds, causing the composite material to adhere strongly to the mould upon opening [15]. Furthermore, it is possible that the material did not have enough time to cool down and harden completely. Premature opening while the material was still in 2 phases (solid and semi-solid) prevented the plate from staying in one piece [16].

From FESEM (Field Emission Scanning Electron Microscopy) analyses from previous research, excellent dispersion of materials with no sign of agglomeration was observed for the PP/kenaf/graphene 1 wt. % sample (Idumah & Hassan 2016). The same was observed for the PP/kenaf/graphene 3 wt. % sample, which also showed the intact, embedded graphene nanoplatelet sheets protruding from the fracture surface studied. The increase in mechanical properties (tensile strength from 17.5 MPa to 21.3 MPa) reported for the sample with the 3 wt. % graphene content is attributed to the homogenous distribution of the fillers within the polymer matrix. On the other hand, the PP/kenaf/graphene 5 wt. % sample shows a decrease in tensile strength to 17.3 MPa. the cause for the drop in tensile strength was attributed to the presence of agglomerates and fibre pull-outs, and defects such as voids in the PP matrix.
3.2. Computer simulations: General static analysis

For the general static analysis, the tensile test was set up in a fashion similar to a physical test following the ASTM D3039 standards, with a sample of dimensions 250 mm × 25 mm × 3 mm, with the sample clamped at both ends. For the PP/kenaf composite without graphene addition, the highest values of stress and strain during tensile testing were 22.955 MPa and 0.02404 respectively. The values of Young’s modulus for the samples was determined by calculating the gradient of the stress-strain graphs generated by the software for each simulation and the value of tensile strength was the highest stress obtained from the stress-strain graphs. For the PP/kenaf composite, the Young’s modulus was determined to be 954 MPa while the tensile strength was 22.95 MPa. For the PP/kenaf/graphene 1 wt. % sample, the highest value of stress obtained was 23.07 MPa, corresponding to the material’s tensile strength, while the highest value for its strain was 0.021165. The Young’s modulus value obtained was 1090 MPa. This shows a 0.12 MPa or 0.5 % increase in tensile strength compared to the PP/kenaf sample, while the Young’s modulus increased by 135.34 MPa or 14.18 %.

![Figure 1](image1.png)

**Figure 1.** (a). Tensile testing sample: PP/kenaf composite, (b). Tensile testing sample: PP/kenaf/graphene 1 wt. % composite, (c). Tensile testing sample: PP/kenaf/graphene 3 wt. % composite, (d). Tensile testing sample: PP/kenaf/graphene 5 wt. % composite.

![Figure 2](image2.png)

**Figure 2.** (a) Stress-strain graphs of PP/kenaf/graphene samples, (b) Young’s modulus values of samples (general static analysis).
Next, the PP/kenaf/graphene 3 wt. % sample had shown a tensile strength value of 22.878 MPa, with a strain value of 0.019066, and Young’s modulus of 1200 MPa; a decrease of 0.076 MPa (0.33 %) in tensile strength and increase of 245.15 MPa (25.67 %) in Young’s modulus when compared to the PP/kenaf composition. Lastly, the PP/kenaf/graphene 5 wt. % sample had a tensile strength of 22.846 MPa, highest strain value of 0.014279, and Young’s modulus value of 1600 MPa. This corresponds with a 0.109 MPa (0.47 %) decrease in tensile strength and 645.16 MPa (67.57 %) increase in Young’s modulus value compared to the sample without graphene addition. The stress-strain curves for all the sample compositions is displayed on Figure 2. It can be seen that increasing graphene content in the sample composition causes steeper gradients in the graphs. This indicates that the higher the graphene content, the higher the Young’s modulus of the sample. The values of Young’s modulus are plotted and compared on a bar chart in Figure 2.

From Figure 2(b), it is clear to see that the values of Young’s modulus increase with the addition of graphene fillers. This may be attributed to a uniform and homogenous distribution of the graphene filler in the PP matrix [17]. Graphene has very high mechanical properties and stiffness; its Young’s modulus is up to 1 TPa and maximum tensile strength is 130 GPa, and it contributes to the increase of mechanical properties of the composites [18]. Furthermore, the addition of graphene may have improved the adhesion between the PP matrix and kenaf fibres. This reduces and restricts the deformation of the matrix during the elastic region of tensile testing phase, thus increasing the Young’s modulus. Other than that, the possible alignment of the graphene nanoplatelets parallel to the direction of flow of the polymer matrix during the hot press process further improves the composites’ Young’s moduli. Moreover, the dispersion of inorganic materials, such as graphene, within the polymer material plays a role in the structure of the matrix. The movement of molecular chains will be restricted as cross-links between the graphene and PP matrix are formed. This increases the composite system’s stiffness. As PP is a crystalline polymer, the addition of graphene as a filler will have an effect on the heterogenous nucleation, which leads to an increase in the degree of crystallinity or may even alter the crystal structure. This also increases the stiffness of the composites.

The tensile strength of the composites is laid out and compared using a bar chart as seen in Figure 3. Generally, the values do not undergo any significant changes, with a range of only 0.228 MPa between the lowest value of 22.85 MPa for the PP/kenaf/graphene 5 wt. % sample and the highest value of 23.08 MPa for the PP/kenaf/graphene 1 wt. % sample. It can be seen that the trend of tensile strength is an increase in the value from 22.95 MPa (PP/kenaf sample) to 23.08 MPa (PP/kenaf/graphene 1 wt. % sample), before gradually decreasing to 22.88 MPa (PP/kenaf/graphene 3 wt. % sample), to 22.85 MPa (PP/kenaf/graphene 5 wt. % sample). The initial increase in tensile strength can be attributed to the interface adhesion between the inorganic particles and the polymer matrix. For particles in the form of thin sheets, the effect of adhesion depends significantly on the surface area of the particle, whereby a particle with higher specific surface area results in a composite with higher mechanical properties due to better interface adhesion [19]. Graphene is a 2-dimensional material with a high surface area relative to its weight (500 m²/g surface area and 12.01 g/mol molecular weight for C-500 graphene nanoplatelets) and is capable of increasing the interface adhesion with the PP matrix [18]. The subsequent decline in tensile strength could be due to filler-filler interactions (that of the graphene and kenaf fibres) caused by agglomeration which increases with the increase in filler content. The effect of agglomeration will weaken the adhesion between the fillers and matrix, and thus weakening the whole composite system [17]. Besides that, the presence of other defects such as pores and voids, non-uniform dispersion of fillers, and fibre pull-outs weakens the composite system by causing stress concentrations and a non-homogenous sample [20].
The strains and extensions for the composite samples after tensile testing are displayed on bar charts on Figure 4 and Figure 5 respectively. The values of strain show a consistent decrease with increase in graphene content, from 0.024 for the PP/kenaf sample to 0.014 for the PP/kenaf/graphene 5 wt. % sample as seen in Figure 4. This results in a decrease of 0.01 (mm/mm) or 41.67 %. This indicates that the increase in graphene content increases the brittleness of the composite material. The same decreasing trend is observed for the extension of the samples as shown in Figure 5; from 1.80 mm for the PP/kenaf sample to 1.06 mm for the PP/kenaf/graphene 5 wt. % sample, which is a 41.33 % decrease (0.76 mm). These decreases in strain and extension are possibly due to the graphene particles initiating β-PP nucleation, or the incompatibility between the matrix and the fillers. Besides that, the graphene addition increases the composites’ densities and causes them to become more brittle [17]. The consistent decrease in strain values justifies the increase in Young’s modulus with increasing graphene content for the samples, even though there is a decrease in the tensile strength of the samples. The value of Young’s modulus is obtained by calculating the gradient of the stress-strain graphs for each composition that is to divide the highest value of stress with the highest value of strain on the straight-line graph. Since the
strain values decrease more drastically (41.67 %) compared with the decrease in tensile strength values (0.47 %), the values of Young’s modulus increase with the increase of graphene content.

4. Conclusion
The effect of addition of graphene to the Young’s modulus and tensile strength of PP/kenaf composites has been investigated throughout this research. Test samples of the composite materials were successfully fabricated using hot press method according to ASTM D3039 specifications for tensile testing. Static general and dynamic explicit computer simulations using Abaqus CAE software were conducted to study the aforementioned mechanical properties of the composite materials. From the general static analysis, the highest value of Young’s modulus was 1 600 MPa achieved with the material with composition of PP/kenaf/graphene 5 wt. % while the highest value of tensile strength was achieved by the composition of PP/kenaf/graphene 1 wt. % at 23.07 MPa compared to a Young’s modulus of 954 MPa and tensile strength of 22.95 MPa for the PP/kenaf composite without addition of graphene. The values of Young’s modulus were similar when compared to previously conducted studies, while the values of tensile strength and the effect of graphene addition on the properties of the composites were congruent to the previous studies as well. It can be concluded, with the additional of graphene in PP/Kenaf have shown in decreasing of their tensile strength. Meanwhile, addition of graphene in PP/Kenaf have resulted increases in their Young’s Modulus.

Acknowledgements
The authors wish to gratefully thank and acknowledge the Center for Research and Instrumentation Management (CRIM), Universiti Kebangsaan Malaysia for their financial support to complete this study under grant number GGPM-2020-002.

References
[1] W. Wang, Y. Zhang, S. Wang, C.Q. Fan, H. Xu, Distributions of phthalic esters carried by total suspended particulates in Nanjing, China, Environ. Monit. Assess. 184 (2012) 6789–6798. https://doi.org/10.1007/s10661-011-2458-z.
[2] T.E. Lovejoy, L. Hannah, Biodiversity and climate change: Transforming the biosphere, 2019.
[3] C. Lu, S. Grundy, Urban Agriculture and Vertical Farming, Elsevier, 2017. https://doi.org/10.1016/B978-0-12-409548-9.10184-8.
[4] J. Bell, L. Paula, T. Dodd, S. Németh, C. Nanou, V. Mega, P. Campos, EU ambition to build the world’s leading bioeconomy—Uncertain times demand innovative and sustainable solutions, N. Biotechnol. 40 (2018) 25–30. https://doi.org/10.1016/j.nbt.2017.06.010.
[5] Y. Zare, K.Y. Rhee, Effects of carbon nanotubes and interphase properties on the interfacial conductivity and electrical conductivity of polymer nanocomposites, Polym. Int. 69 (2020) 413–422. https://doi.org/10.1002/pi.5969.
[6] R. Taherian, M.J. Hadianfard, A.N. Golikand, Manufacture of a polymer-based carbon nanocomposite as bipolar plate of proton exchange membrane fuel cells, Mater. Des. 49 (2013) 242–251. https://doi.org/http://dx.doi.org/10.1016/j.matdes.2013.01.058.
[7] M.H. Cetin, B. Ozcelik, E. Kuram, E. Demirbas, Evaluation of vegetable based cutting fluids with extreme pressure and cutting parameters in turning of AISI 304L by Taguchi method, J. Clean. Prod. 19 (2011) 2049–2056. https://doi.org/10.1016/j.jclepro.2011.07.013.
[8] M. Ramesh, Kenaf (Hibiscus cannabinus L.) fibre based bio-materials: A review on processing and properties, Prog. Mater. Sci. 78 (2016) 1–92. https://doi.org/http://dx.doi.org/10.1016/j.pmatsci.2015.11.001.
[9] R.A. Ilyas, S.M. Sapuan, M.R. Ishak, E.S. Zainudin, Development and characterization of sugar palm nanocrystalline cellulose reinforced sugar palm starch bionanocomposites, Carbohydr. Polym. 202 (2018) 186–202. https://doi.org/10.1016/j.carbpol.2018.09.002.
[10] H.M. Akil, M.F. Omar, A.A.M. Mazuki, S. Safiee, Z.A.M. Ishak, A. Abu Bakar, Kenaf fiber reinforced composites: A review, Mater. Des. 32 (2011) 4107–4121.
[11] J.A. García Sánchez, J.M. López Martínez, J. Lumbreras Martín, M.N. Flores Holgado, H. Aguilar Morales, Impact of Spanish electricity mix, over the period 2008-2030, on the Life Cycle energy consumption and GHG emissions of Electric, Hybrid Diesel-Electric, Fuel Cell Hybrid and Diesel Bus of the Madrid Transportation System, Energy Convers. Manag. 74 (2013) 332–343. https://doi.org/10.1016/j.enconman.2013.05.023.

[12] N.A.M. Radzuan, A.B. Sulong, M. Rao Somalu, Electrical properties of extruded milled carbon fibre and polypropylene, J. Compos. Mater. 51 (2017) 3187–3195. https://doi.org/10.1177/0021998316688075.

[13] P. Wambua, J. Ivens, I. Verpoest, Natural fibres: Can they replace glass in fibre reinforced plastics?, Compos. Sci. Technol. 63 (2003) 1259–1264. https://doi.org/10.1016/S0266-3538(03)00096-4.

[14] C.I. Idumah, J.E. Ogbu, J.U. Ndem, V. Obiana, Influence of chemical modification of kenaf fiber on xGNP-PP nano-biocomposites, SN Appl. Sci. 1 (2019) 1–11. https://doi.org/10.1007/s42452-019-1319-1.

[15] M.K.F. MdRadzi, A.B. Sulong, N. Muhamad, M.A. MohdLatiff, N.F. Ismail, Effect of Filler Loading and NaOH Addition on Mechanical Properties of Moulded Kenaf/Polypropylene Composite, Pertanika J. Trop. Agric. Sci. 38 (2015) 583–590.

[16] C.G. Kang, J.S. Choi, K.H. Kim, Effect of strain rate on macroscopic behavior in the compression forming of semi-solid aluminum alloy, J. Mater. Process. Technol. 88 (1999) 159–168. https://doi.org/10.1016/S0924-0136(98)00383-5.

[17] N. Afiqah, M. Radzuan, A.B. Sulong, M. Irwan, M. Firdaus, T. Husaini, E.H. Majlan, Fabrication of multi-filler MCF / MWCNT / SG-based bipolar plates, Ceram. Int. 45 (2019) 7413–7418. https://doi.org/10.1016/j.ceramint.2019.01.028.

[18] R.B. Mathur, S.R. Dhakate, D.K. Gupta, T.L. Dhami, R.K. Aggarwal, Effect of different carbon fillers on the properties of graphite composite bipolar plates, J. Mater. Process. Technol. 203 (2008) 184–192. https://doi.org/http://dx.doi.org/10.1016/j.jmatprot.2007.10.044.

[19] W.F. Santos, M. Quattrone, V.M. John, S.C. Angulo, Roughness, wettability and water absorption of water repellent treated recycled aggregates, Constr. Build. Mater. 146 (2017) 502–513. https://doi.org/10.1016/j.conbuildmat.2017.04.012.

[20] N.A.M. Radzuan, A.B. Sulong, M.R. Somalu, Influence the filler orientation on the performance of bipolar plate, Sains Malaysiana. 48 (2019) 669–676. https://doi.org/10.17576/jsm-2019-4803-21.