Effect Of Nanosilica Content On Longitudinal And Transverse Tensile Properties Of Unidirectional Kenaf Composite
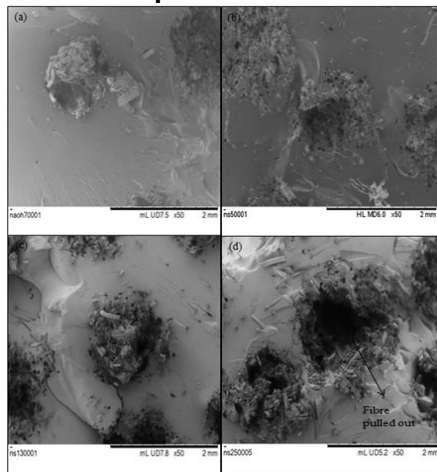
Norhashidah Manap, Aidah Jumahat*, Napisah Sapiai

Faculty of Mechanical Engineering, Universiti Teknologi MARA, 40450 Shah Alam, Selangor, Malaysia

*Corresponding author
aidahjumahat@salam.uitm.edu.my

Graphical abstract

Abstract

Nowadays, continuous natural fibre reinforced polymer nanocomposites have attracted substantial attention among researchers due to various benefits possesses by the natural fibres. Kenaf fibre has become one of the high potential candidates to replace synthetic fibres in polymer composite. Kenaf fibre exhibits good strength and modulus properties, low density, non-abrasive during processing and biodegradable. This study is aimed to evaluate the effect of nanosilica on longitudinal and transverse tensile properties of unidirectional (UD) kenaf composite. The UD kenaf composite samples were prepared based on three different nanosilica content; i.e. 5, 13 and 25 wt.%. The samples were prepared using filament winding and vacuum bagging techniques. The 0° and 90° tensile tests were conducted in accordance to ASTM standard D3039 in order to obtain longitudinal and transverse tensile properties of unmodified and nanosilica-modified kenaf composites. The fracture surfaces of the specimens were observed using scanning electron microscope in order to identify fracture mechanisms involved during tension. The results showed that the addition of nanosilica reduced longitudinal tensile Young's modulus, strength and failure strain of the kenaf composite. SEM micrographs revealed incomplete resin wetting and fibre pull-out mechanism at high nanosilica content that contributed to premature failure of the kenaf composites. However, it was found that the addition of nanosilica improved transverse tensile properties of kenaf composites since these properties were mostly governed by the properties of the matrix. A stiffer matrix improved the transverse tensile modulus and strength of kenaf composites.

Keywords: Natural fibres, kenaf composites, nanosilica, polymer composite, tensile properties

1.0 INTRODUCTION

Recently, continuous fibre reinforced polymer nanocomposites have extensively been studied by many researchers. Nevertheless, the properties induced by the nanometric scale of fillers still not fully understood. The benefits of nanofillers as reinforcement material to fibre composites remain as an interesting area to be explored. This is due to the fact that the properties of fibre reinforced polymer nanocomposites depend on various factors; especially the degree of dispersion of nanofillers in the polymer matrix, the interfacial bonding in between nanofillers, matrix and continuous fibre, size and shape of the nanofillers, filtering effect and others.

Previous research has shown that silane treated nanosilica spherical particles have a very good dispersion in epoxy polymer matrix [1]. Therefore, this contributed to an improvement in mechanical properties of cured polymer. Jumahat et. al. [1] reported that nanocomposites offer higher tensile

stiffness and strength when compared to the neat polymer without sacrificing the failure strain of the material[1]. It is expected that the properties of fibre reinforced polymer composites will also be improved by adding this nanosilica particles. A study on the effect of nanosilica on carbon fibre reinforced polymer (CFRP) composites was conducted in [2]. The results showed that the presence of spherical silica nanoparticles stiffened the epoxy matrix and offered a better lateral support to the carbon fibre. Therefore, the mechanical properties were improved significantly in comparison with the unmodified CFRP system.

This study is extended to evaluate the effect of nanosilica on mechanical properties of natural fibres composites. Recent interest in environmental friendly materials has promoted the usage of natural fibres as reinforcement material for high performance polymer. Inexpensive inorganic substances are traditional ingredients in polymer industry. Natural fibres have promising properties to be used in future industries for structural and non-structural industries. Mechanical properties of natural fibre reinforced composite laminates have extensively been been studied by many researchers [3]–[8]. Kenaf is one type of natural fibre that has been used in polymer composites. Kenaf (Hibiscus cannabinus, L. family Malvaceae) is a herbaceous annual plant that can be grown under a wide range of weather condition; for example, it can grows to more than 3m within 3 months even in moderate ambient conditions with stem diameter of 25-51mm. It is an annual cane-like crop originating in Asia and Africa. Kenaf is traditionally used as rope, canvas, and sacking [9]-[10]. The ability of kenaf to absorb nitrogen and phosphorus and accumulate carbon dioxide at a clearly high rate have become an attractive feature for kenaf cultivation in preventing global warming. Nishino et al. [11] reported that kenaf exhibits good specific strength and modulus properties, low density, non-abrasive during processing and biodegradable. Therefore, this study is aimed to evaluate the effect of nanosilica on mechanical properties of continuous kenaf fibre reinforced polymer composites.

2.0 EXPERIMENTAL

2.1 Materials

The epoxy resin and hardener used for this study were supplied by Miracon (M) Sdn Bhd. The spherical silica nanoparticles (Nanopox F400) were supplied as a colloidal sol (40 wt% nanosilica) in epoxy by nanoresin AG, Geesthacht, Germany. The yarn kenaf fibres were supplied by Innovative Pultrusion Sdn. Bhd.

2.2 Fabrication Of Nanosilica Filled Kenaf Fibre Reinforced Polymer Composites

Pure polymer matrix used was an epoxy resin which consists of Miracast 1517 A/B DGEBA epoxy resin and amine-curing agent (hardener). The ratio of epoxy and hardener used was 100:30. A series of nanosilica (5wt%, 13wt% and 25wt%) was mixed with epoxy resin using mechanical stirrer at 400 rpm for 1 hour. Then, the mixtures were degassed under high vacuum machine for 1 hour, followed by adding the harderner at ratio 100:30 (epoxy:harderner). Degasification process was employed to remove entrapped air during the composite fabrication. Kenaf fibres, which were treated using 7 wt% sodium hydroxide (NaOH), were wound on aluminium frame of 430 x 300 mm in one direction. The wound kenaf were impregnated with epoxy resin. The impregnated fibres were vacuumed and left to cure at room temperature and 515Pa pressure for 24 hours. Finally, the samples were post cured at 60°C for 2 hours, followed by 80°C for 2 hours, 100°C for 2 hours and 120°C for 2 hours. After curing, the nanosilica modified and unmodified kenaf reinforced polymer (KFRP) composite plate was obtained by cutting-off the frame.

2.3 Longitudinal And Transverse Tensile Tests

The unmodified and nanosilica modified KFRP composite plates were cut into two different dimensions for londitudinal tensile test at 0° fibre direction and transverse tensile test at 90° fibre direction. Longitudinal tensile tests were conducted on rectangular specimens of 15 mm width x 250 mm overall length (with 56 mm tab length) and 4.5 mm average thickness. Transverse tensile tests were conducted on rectangular specimens of 25 mm width x 175 mm overall length (with 25 mm tab length) and 4.5 mm average thickness. The tests were conducted in accordance to ASTM Standard D3039 using an Instron Universal Tester machine and the BlueHill data acquisition software. Tensile tests were conducted on unidirectional (UD) laminates at 0° and 90° fibre directions to determine longitudinal and transverse tensile stress, tensile strain, maximum load, extension at maximum load and Young's modulus. A 100 kN load cell and 25 mm gauge length clip-on extensometer were used to record the applied load and elongation data. The tensile tests were conducted at a crosshead speed of 2 mm/min.

2.4 Observation On Fibre And Fractured Surface

The Field Emission Scanning Electron Microscope (FESEM), SUPRA 40 VP (Carl Zeiss) was used to observe the morphology of fibre surface and to identify the fracture mechanicsms involved during tensile test of KFRP composite specimens. The fracture surfaces perpendicular to the loading direction or the longitudinal splits were observed by sectioning the area of interest and mounted onto suitable holder.

Sections were mounted onto aluminum stubs using carbon adhesive patch. The specimens were then coated with thin layer of platinum using sputter coater.

3.0 RESULTS AND DISCUSSION

3.1 Longitudinal Tensile Properties Of Nanosilica Filled Kenaf Fibre Reinforced Polymer Composites

Figure 1 shows the effect of 5wt%, 13wt% and 25wt% nanosilica content on longitudinal tensile modulus of NaOH treated kenaf composite. The longitudinal tensile modulus of unmodified kenaf composites (denoted by 7Na/K) is 5.6 GPa. The addition of 5 wt.% nanosilica improved the elastic modulus of kenaf composites (denoted by 7Na/K/5si). The longitudinal tensile modulus of 7Na/K/5si is 7.265GPa which is 29.73% higher than the unmodified kenaf composites 7Na/K. It is expected that the value of Young's modulus will keep improving with further increase in the amount of nanosilica. This is due to the fact that the Young's modulus of nanosilia is about 70GPa, therefore it will improve the Young's modulus of kenaf composites (about 5.6GPa). However, Figure 1 shows that the value of tensile modulus decreased with further addition of nanosilica. The addition of 13 wt.% and 25 wt.% nanosilica reduced the value of longitudinal tensile modulus to 5.605GPa and 5.608GPa, respectively. This will be explained later based on the observation on fracture surface of nanomodified-kenaf composites using FESEM.
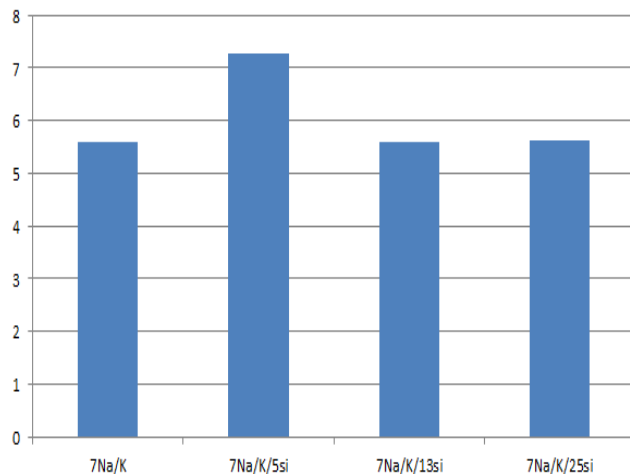

Figure 1 Longitudinal tensile modulus of 5wt%, 13wt% and 25wt% nanosilica modified NaOH treated kenaf composites compared to unmodified kenaf composite.

Figure 2 shows the longitudinal tensile strength of unmodified kenaf composite (7Na/K) and 5wt%, 13wt% and 25wt% nanosilica modified kenaf composites (7Na/K/5si, 7Na/K/13si and 7Na/K/25si). Longitudinal tensile strength of unmodified kenaf composite 7Na/K is 79.18MPa. Longitudinal tensile strength of 7Na/K/5si is 53.04MPa which is 33.01%

lower than the pure system of 7Na/K. Tensile strength of 7Na/K/13si and 7Na/K/25si are 58.25MPa and 75.58MPa, respectively. These show that the addition of nanosilica reduced the longitudinal tensile strength of kenaf composites.

Figure 3 shows the longitudinal tensile failure strain for kenaf composites. It can be seen that the addition of nanosilica also cause a significant reduction in failure strain of kenaf composites. Longitudinal tensile failure strain of composites as shown in Figure 3 has a similar trend to tensile strength as shown in Figure 2 where the addition of nanosilica caused premature failure of the kenaf composite. The longitudinal tensile failure strain of 7Na/K is 2.313%. Figure 3 shows that the longitudinal tensile strain of 7Na/K/5si reduced of about 1.3% strain when compared to 7Na/K. The longitudinal tensile failure strain of 7Na/K/13si and 7Na/K/13si is 0.6% and 0.8% strain, respectively, higher than the longitudinal tensile strain of 7Na/K/5si.
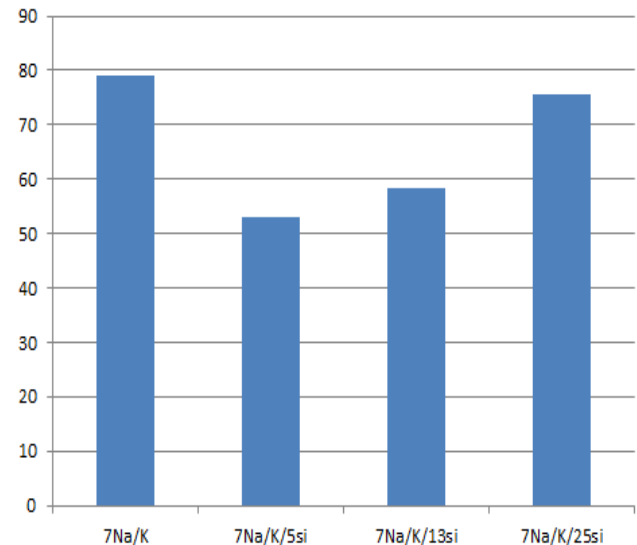

Figure 2 Longitudinal tensile strength of 5wt%, 13wt% and 25wt% nanosilica modified NaOH treated kenaf composites compared to unmodified kenaf composite.
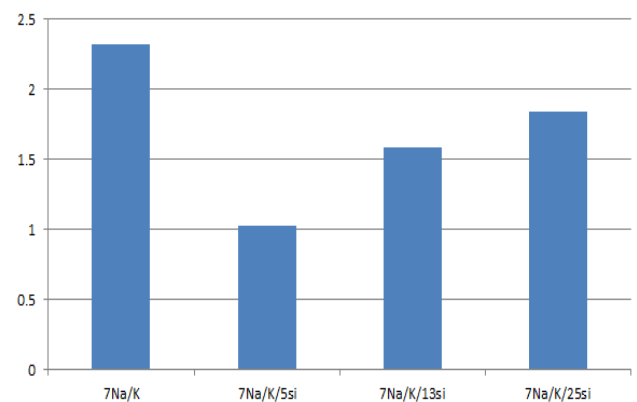

Figure 3 Longitudinal tensile failure strain of 5wt%, 13wt% and 25wt% nanosilica modified NaOH treated kenaf composites compared to unmodified kenaf composite.

3.2 Transverse Tensile Properties Of Nanosilica Filled Kenaf Fibre Reinforced Polymer Composites

Transverse tensile modulus of kenaf composite is shown in Figure 4. The transverse tensile properties of nanosilica filled epoxy kenaf composite shows increasing trend in Young's modulus. The unmodified
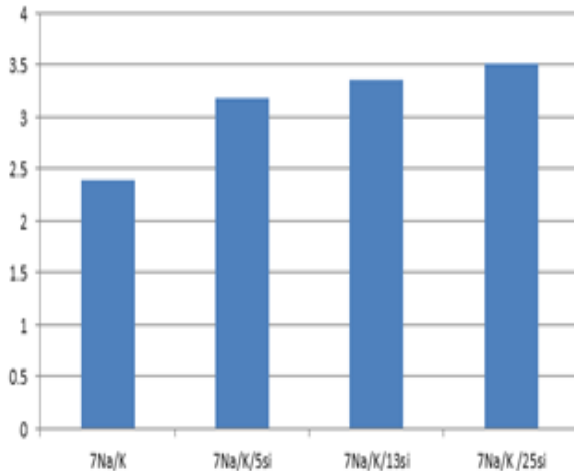

Figure 4 Transverse tensile modulus of 5wt%, 13wt% and 25wt% nanosilica modified NaOH treated kenaf composites compared to unmodified kenaf composite.

kenaf composite has transverse tensile modulus of 2.38GPa. The addition of 5 wt.% nanosilica in epoxy improved the transverse tensile modulus by 33.44%. The transverse tensile modulus of 7Na/K/13si and 7Na/K/25si are 3.357GPa and 3.507GPa, respectively. The addition of nanosilica improved the transverse tensile modulus of kenaf composites. This improvement is caused by the properties of the nanosilica. The high Young's modulus of nanosilica stiffened the matrix. Therefore this contributes to a higher transverse tensile modulus of kenaf composites.

A summary of transverse tensile strength of kenaf composite is shown in Figure 5. The transverse tensile strength of kenaf fibre without addition of nanosilica is 22.193MPa. The transverse tensile strength of the composite increases when added with 5 wt.% nanosilica which is 24.44MPa. However the strength decreases with addition of 13 wt.% nanosilica in epoxy. This phenomenon occurs maybe because of formation of nano sized voids in epoxy matrix. The high viscosity of epoxy matrix when added with nanosilica causes difficulty in removing the entrapped bubble during fabrication process. The transverse tensile strength of 25 wt.% of nanosilica in epoxy shows the highest strength which is 26.36 MPa. The purpose of addition of nanosilica can be seen in transverse tensile properties since the transverse tensile properties usually dependent on the properties of the matrix.

Figure 6 shows the transverse tensile failure strain for kenaf composites. The transverse tensile failure strain of nanosilica filled epoxy kenaf composite decreased with increasing nanosilica content. The transverse tensile strain of kenaf epoxy without the addition of nanosilica is 1.727%. The transverse tensile strain of 5 wt.% of nanosilica in kenaf composite is 1.11% and 1.031% for 13 wt.% of nanosilica filled epoxy in kenaf composite.

The transverse tensile strain of 7Na/K/25si shows the lowest value which is 1.023. These results show that the addition of nanosilica in kenaf composite decreases the ductility of composite. F
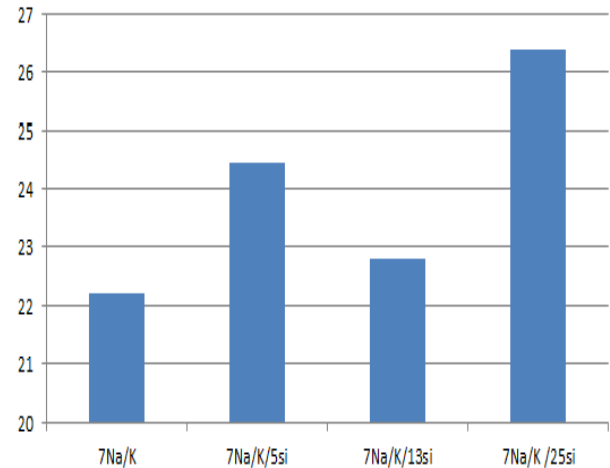

Figure 5 Transverse tensile strength of 5wt%, 13wt% and 25wt% nanosilica modified NaOH treated kenaf composites compared to unmodified kenaf composite.
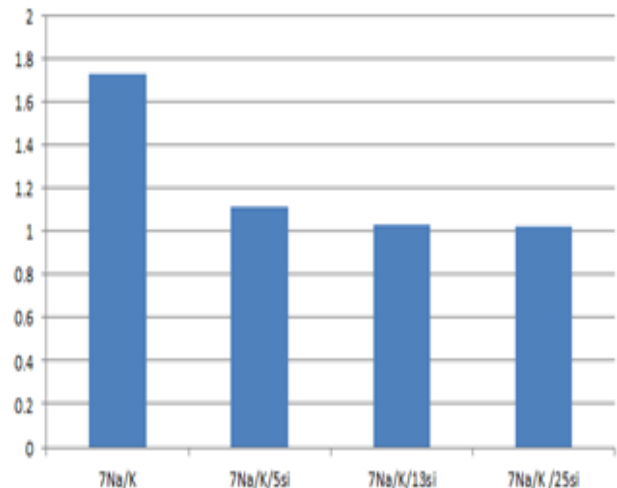

Figure 6 Transverse tensile failure strain of 5wt%, 13wt% and 25wt% nanosilica modified NaOH treated kenaf composites compared to unmodified kenaf composite.

Table 1 illustrates the summary of longitudinal tensile properties of unmodified and nanomodified kenaf composites, while Table 2 shows summary of transverse tensile properties of unmodified and

nanomodified kenaf composites. The tables show the percentage of error of each average data. The data were taken based on a minimum of 5 test samples of each composite system.

3.3 Scanning Electron Microscope

Figure 7 shows the fracture surface of longitudinal kenaf composite observed under scanning electron microscope at 50x magnification. Figure 7(a) shows fracture surface of unmodified unidirectional kenaf composites 7Na/K. The fracture surface of 7Na/K/5si (Figure 7(b)) shows a smoother surface when compared to the fracture surface of unmodified composite, 7Na/K. A smoother surface fibre indicates low interfacial bonding strength where mechanical interlocking mechanism does not occur during loading. When the amount of nanosilica content increases, the fracture surface of longitudinal tensile load of 7Na/K/13si (see Figure 7(c)) and 7Na/K/25si (see Figure 7(d)), show larger gap between the fibre and matrix interface. This indicates the composite fails by fibre debonding mechanism. This fracture surface shows that the addition of nanosilica results in a fibre wetting problem. The presence of nanosilica reduces the wetting process of fibre in polymer matrix. This cause uneven distribution of polymer matrix especially at the middle of fibre bundle. This results in improper interfacial bonding in between fibre and polymer matrix and this limits the transferring load capability of the fibre. This then causes fibre debonding when the longitudinal fibre kenaf composite is loaded in tension and also premature failure of the system. The micron sized voids also appear at the matrix, surrounding and within the kenaf yarn. These answers the reduction in longitudinal Young's modulus, strength and failure strain of the kenaf composites when the systems were added with nanosilica. This is further proven by the presence of fibre pull out mechanism at high nanosilica content in the kenaf composites

(7Na/K/25si). It clearly can be seen that the fracture surface of 7Na/K/25si (see Figure 7d) shows the kenaf fibre was pulled-out and left hole in the epoxy matrix. There are also micron sized voids within the matrix and surrounding the fibre. The surface fracture shows that the fibre is easily pull out from the matrix and leave holes which indicate that the weak adhesion between kenaf fibre and matrix. It can be seen that the kenaf strand yarn is not fully attached to the matrix causing it to be easily pulled out form the composite under tensile loading.

Figure 8 shows the fracture surfaces of transverse fibre direction kenaf composite which were observed using scanning electron microscope at 50x magnification. Figure 8 presents the damage view of (a) 7 Na/K (b) 7Na/K/5si, (c) 7Na/K/13si and (d) 7Na/K/25si for transverse fibre direction under tensile loading. Fracture surfaces of nanosilica filled kenaf composite show that the fracture happens at the interface between kenaf-epoxy matrix. This shows that the strength of the transverse tensile properties not only depends on the properties of epoxy but also the interfacial strength of kenaf-epoxy. The elastic modulus increases with increasing percentage of nanosilica. This is due to the fact that the matrix become stiffer when added with nanosilica. However, the presence of micron sized void at high percentage nanosilica content limits the modulus of elasticity, strength and failure strain of the kenaf composites. The presence of void in the polymer is very difficult to eliminate due to high viscosity of the nanomodified resin where the removal of entrapped bubbles is difficult during fabrication process. The void absolutely will give adverse effects on mechanical properties of the composite. Figure 8c shows the presence of micron sized void in the polymer matrix and fibre composites. This may be the reason why a slight reduction on tensile properties were observed at 13wt% nanosilica content in kenaf composites.

Table 1 Summary of longitudinal tensile properties of kenaf composite

Material properties	7Na/K	7Na/K/5si	7Na/K/13si	7Na/K/25si
Tensile modulus, (GPa)	5.6±0.2	7.265±0.2	5.605±0.37	5.608±0.17
Tensile strength, (MPa)	79.18±1.69	53.04±2.93	58.25±2.29	75.58±1.16
Tensile strain at break, (%)	2.313±0.45	1.02±0.027	1.576±0.22	1.83±0.08

Table 2 Summary of longitudinal tensile properties of kenaf composite

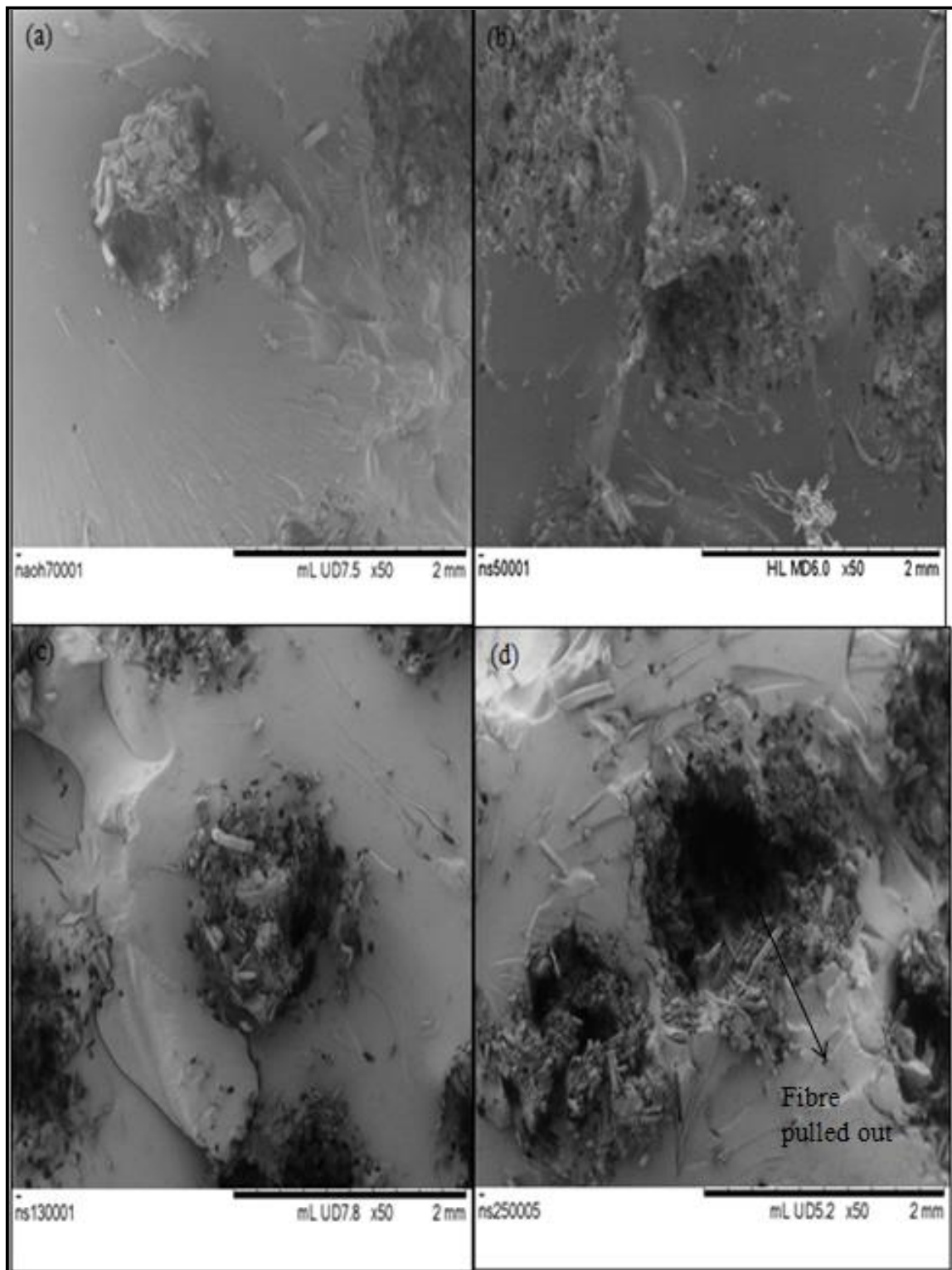| Material properties	7Na/K	7Na/K/5si	7Na/K/13si	7Na/K/25si
Tensile modulus, (GPa)	2.38±0.2	3.176±0.1	3.357±0.12	3.507±0.15
Tensile strength, (MPa)	22.193±2.23	24.437±2.3	22.781±3.1	26.36±2.1
Tensile strain at break, (%)	1.727±0.103	1.11±0.09	1.031±0.11	1.023±0.09

Figure 7 Fracture surface of kenaf composite for longitudinal fibre direction under tensile loading.
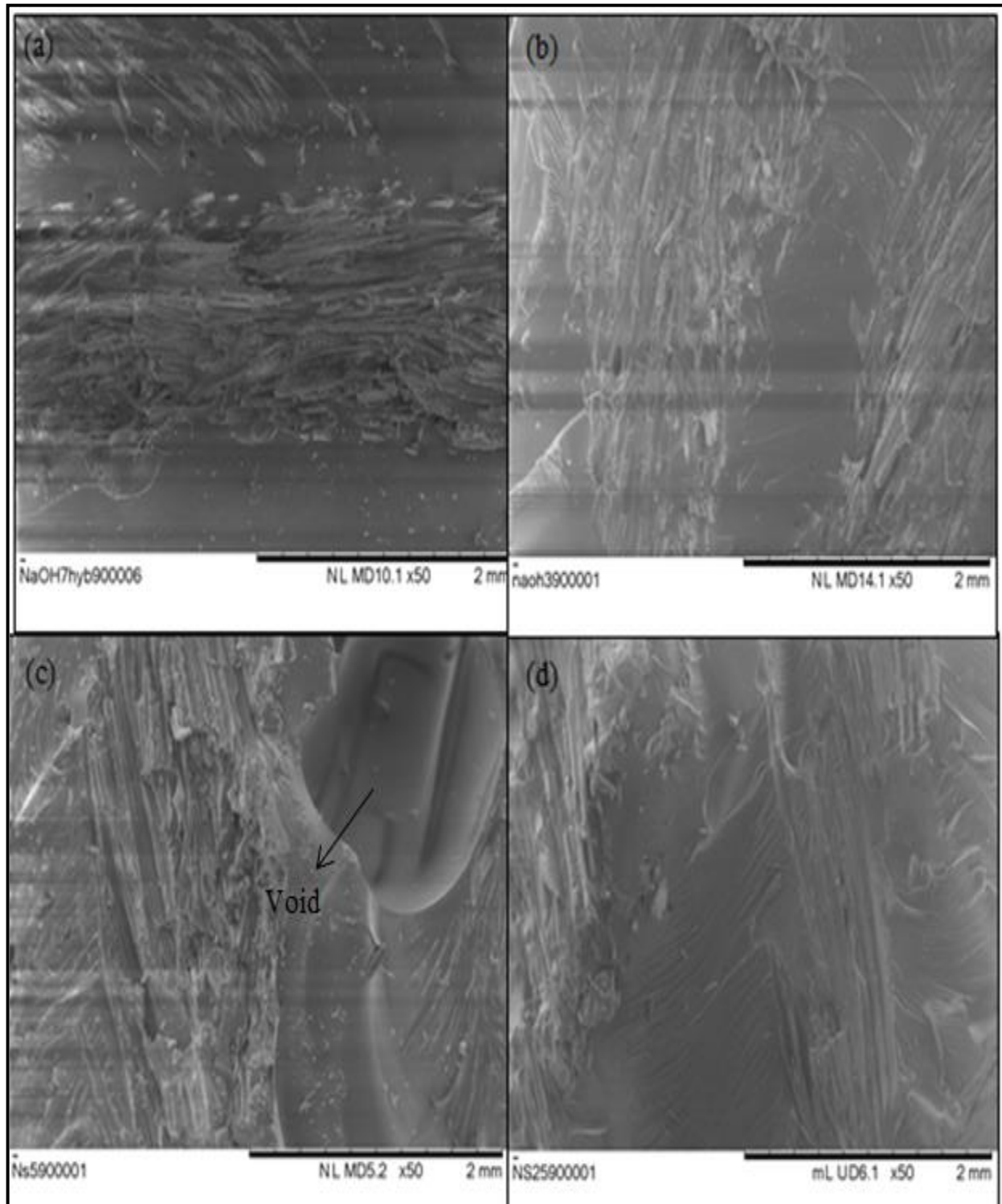
Figure 8 Fracture surface of kenaf composite for transverse fibre direction under tensile loading.

4.0 CONCLUSION

In general inclusion of nanosilica in kenaf composites gave adversed effect on longitudinal tensile properties. It can be observed that the addition of nanosilica reduced the Young's modulus, strength and failure strain of the kenaf composite. SEM micrographs revealed that high nanosilica content reduced the wetting capability of the epoxy on the fibre yarn. This wetting problem causes poor interfacial bonding in between fibre and the matrix. This causes reduction in load transfer capability, premature failure of the kenaf composites and fibre pull-out mechanism. Therefore this reduces the

longitudinal tensile properties of the kenaf composite. However, transverse tensile properties of the kenaf composites showed improvement since transverse tensile properties were mostly governed by the properties of the matrix and fibre-matrix interfacial bonding. The inclusion of nanosilica stiffened the matrix and this results in improved tensile modulus and tensile strength of kenaf composite loaded in transverse direction.

Acknowledgement

References

[1] Jumahat, A., Soutis, C., Abdullah, S. A. and Kasolang, S. 2012. Tensile Properties of Nanosilica/Epoxy Nanocomposites. *Procedia Engineering*. 41: 1634-1640.

[2] Jumahat, A., Soutis, C., Jones, F.R. and Hodzic, A. 2010. Improved compressive properties of a unidirectional CFRP laminate using nanosilica particles. *Advanced Composites Letters*. 19 (6): 218-221

[3] Sapiai, N., Jumahat, A., Mahmud, J. 2015. Flexural And Tensile Properties Of Kenaf/Glass Fibres Hybrid Composites Filled With Carbon Nanotubes. *Jurnal Teknologi*. 76(3): 115–120.

[4] Sapiai, N., Jumahat, A. and Hakim, R. N. 2014. Tensile and Compressive Properties of Hybrid Carbon Fiber / Kenaf Polymer Composite. *Advances in Environmental Biology*. 8(8): 2655–2661.

[5] Manap, N., Jumahat, A. and Ludin, A. 2014. Tensile and compressive properties of glass reinforcement in kenaf reinforced epoxy composite. *Advances in Environmental Biology*. 8(8): 2673–2681.

[6] Bakar, N. H., Hyie, K. M., Ramlan, A. S., Hassan, M. K. and Jumahat, A. 2013. Mechanical Properties of Kevlar Reinforcement in Kenaf Composites. *Appl. Mech. Mater.* 465: 847–851.

[7] Joshi, S., Drzal, L., Mohanty, A. and Arora, S. Mar. 2004. Are natural fiber composites environmentally superior to glass fiber reinforced composites?. *Compos. Part A Appl. Sci. Manuf.* 35(3): 371–376.

[8] Herrera-Franco P. and Valadez-González, A. Mar. 2004. Mechanical properties of continuous natural fibre-reinforced polymer composites. *Compos. Part A Appl. Sci. Manuf.* 35(3): 339–345.

[9] Abe K. and Ozaki, Y. Dec. 1998. Comparison of useful terrestrial and aquatic plant species for removal of nitrogen and phosphorus from domestic wastewater. *Soil Sci. Plant Nutr.* 44(4): 599–607.

[10] Amaducci, S., Amaducci, M. T., Benati, R. and Venturi, G. Mar. 2000. Crop yield and quality parameters of four annual fibre crops (hemp, kenaf, maize and sorghum) in the North of Italy. *Ind. Crops Prod.* 11(2–3): 179–186.

[11] Nishino, T., Hirao, K., Kotera, M., Nakamae, K.and Inagaki, H. Jul. 2003. Kenaf reinforced biodegradable composite. *Compos. Sci. Technol.* 63(9): 1281–1286.