

## **Effect of High Temperature Curative and Moulding Temperatures on Tensile and Thermal Properties of Engineering Thermoplastic Abs/Modified Kenaf Fibre (Kf) Composites**

Saliu, Hafsat R.<sup>1\*</sup>; Ishiaku, U.S.<sup>1</sup>; Yakubu, M.K.<sup>1</sup>; Kolawole, E.G.<sup>1</sup>; Adefila, S.S.<sup>2</sup>; Abu Bakar, M.B.<sup>3</sup> & Moh'd Ishak, Z.A.<sup>3</sup>

*Department of Textile Science and Technology, Ahmadu Bello University, Zaria, Nigeria.*

*Department of Chemical Engineering, Ahmadu Bello University, Zaria, Nigeria.*

*School of Materials and Mineral Resources Engineering, Engineering Campus, Universiti Sains Malaysia, 14300 Nibong Tebal, Penang, Malaysia.*

---

**Abstract:** *This work is aimed at incorporating natural bast fibre, kenaf into engineering thermoplastic ABS at its high melting temperature of 230°C, without degradation of the fibre. The idea was carried out by pre-coating the fibre with 40% thermoset which was later cured at high temperature in Haake internal mixer where compounding took place with ABS. Poly (acrylonitrile-butadiene-styrene) (ABS) is a known popular engineering thermoplastic for its unique properties which include its excellent mechanical response, good processing characteristics, chemical resistance and fine surface appearance. ABS composites reinforced with modified kenaf fibres have resulted into increased tensile properties up to the optimum level of the curative loading as it improved the thermal stabilities of the fibres in the composites. The moulding temperatures have also affected the tensile properties significantly.*

**Keywords:** *ABS; Thermoset; Kenaf fibre; Curative; Moulding temperature.*

---

### **I. Introduction**

Poly (acrylonitrile-butadiene-styrene) (ABS) is a thermoplastic consisting of a styrene monomer with the addition of an acrylonitrile monomer and a butadiene monomer. Chemically, this thermoplastic family of plastics is called "terpolymers", in that they involve the combination of three different monomers to form a single material that draws from the properties of all three. It is the butadiene component (a derivative of natural rubber) that is largely responsible for the strength and impact resistance of the plastic. ABS is a popular engineering thermoplastic because of its unique properties, including excellent mechanical response, good processing characteristics, chemical resistance and fine surface appearance. It consists of styrene-acrylonitrile (SAN) continuous phase partially grafted to a dispersed polybutadiene (PB) phase of elastomeric nature. The commercial ABS is produced in the main by emulsion copolymerization of styrene-acrylonitrile in the presence of rubber, and the final product is usually constructed with the physical mixture of styrene-acrylonitrile copolymer as well as the graft copolymer of styrene-acrylonitrile into the rubber. ABS copolymer is an engineering plastic used widely in industry owing to its good mechanical and processing properties. ABS is usually filled with rigid inorganic fillers, such as calcium carbonate (CaCO<sub>3</sub>), talcum powder, kaolin and glass beads, in order to enhance its strength and stiffness and reduce the production cost. ABS is widely used in the automotive industry, telecommunications, business machines and consumer market, mainly because its property-price profile is intermediate between the lower priced commodity thermoplastics and the more expensive high performance engineering plastics. It is also used with the appropriate reinforcements that may be incorporated to the plastic matrix in order to enhance functional properties. One of the main drawbacks of ABS is its lower thermal stability and inherent flammability (Karahaliou & Tarantili, 2009).

Natural fibre-reinforced composites have attracted substantial interest as a potential structural material. The attractive features of natural fibres have been their lower density, greater deformability, less abrasive and lower cost. The properties of the fibres are determined by the physical, mechanical and chemical properties of the morphological constituents and their interfaces. Natural fibre consists of highly crystalline cellulose fibrils spirally wound in a matrix of amorphous hemicellulose, lignin and, eventually, pectin. Lignin and pectin act mainly as bonding agents. The characteristic values of the structural parameters vary from one fibre to another (Bos and Donald 1999).

Kenaf plant is an annual plant that can be harvested 2-3 times a year. It can grow to reach 3-4 meters within 4-5 months and has three layers - bast, core, and pith. Kenaf bast represents one third of the plant. Core and pith represent the rest. Kenaf fibres provide high stiffness and strength values. They also have higher aspect ratios making them suitable to be used as reinforcement in polymer composites (Sanadi et al. 1995). Kenaf bast fibre has been reported to have superior mechanical properties than the other parts of the plant. Its superiority in

flexural strength and excellent tensile strength has made it a good candidate for many applications (Aji et al. 2009).

Recently, kenaf is used as a raw material to be alternative to wood in pulp and paper industries (Ashori and Raverty, 2007) and fibre reinforced plastics (FRP) to replace synthetic fibres such as glass is receiving attention (Abdullah et al. 2011), and also used as non-woven mats in the automotive industries, textiles and fibreboard (Ibrahim et al. 2011). El-Shekeil and co-workers have carried out many researches on kenaf/polyurethane composites at different point in time (2011 and 2012). Ismail et al. (2010) studied Curing Characteristics, Mechanical and Morphological Properties of Kenaf Fibre/Halloysite Nanotubes Hybrid-Filled Natural Rubber Compounds, kenaf fibre-polypropylene composites were studied by many researchers like Sanadi and Co-workers 1995, 2001; Rowell et al. 1995, 1999; Karina et al. 2007, Pan et al. 2007, Iji 2008 and Och 2008 also carried out investigation on kenaf/PLA (PolyLactic acid) and even with cotton to made composites (Wan Ahmad et al. 2007).

The inherent polar and hydrophilic nature of lignocellulosic fibres and the non-polar characteristics of most thermoplastics results in compounding difficulties leading to non-uniform dispersion of fibres within the matrix which impairs the efficiency of the composite. This is a major disadvantage of natural fibre reinforced composites. Another problem is that the processing temperature of composites is restricted to 200°C as vegetable fibres undergo degradation at higher temperatures; this restricts the choice of matrix material.

Alkali treatment can cause an increase of the fibre surface free energy. Moreover, this treatment can make the fibre surface become 'clean' due to removal of waxes, hemicellulose, pectin and part of lignin. The removal of these substances enhances the surface roughness. Therefore, the mechanical interlocking at the interface could be improved.

NaOH is one of the most common chemical treatments used to clean fibre surface and enhance its properties (El-Shekeil et al. 2012; John and Anandjiwala 2008; Mwaikambo and Ansell 2002; Sreekala et al. 1997; Geethamma et al. 1995; Bisanda and Ansell 1992).

Epoxy resins are one of the most important classes of thermosetting polymers which find their extensive use as coatings, matrix resins for composite materials and high performance adhesives. They have various desirable properties such as high tensile strength and modulus, good thermal and chemical resistance and dimensional stability. However, in many applications they have one major drawback. They are very brittle and therefore have poor thermal cycling behaviour and fairly low impact and fracture strengths.

Composite materials are attractive because they combine material properties in ways similar and beyond that found in nature. Such material often result in light weight structures having high stiffness and tailored properties for specific applications, thereby saving weight and reducing energy needs. Fibre reinforced plastic composites began with cellulose fibre in phenolic in 1908, later extending to urea and melamine, and reaching commodity status in the 1940s with glass fibre in unsaturated polymers, from guitar, tennis racquets and cars to microlight aircrafts, electronic components, and artificial joints, composites are finding use in diverse fields (Mohanty et al., 2005). Because the low interfacial properties between fibre and polymer matrix often reduce their potential as reinforcing agents due to the hydrophilic nature of natural fibres, chemical modifications are considered to optimize the interface of fibres. Jana & Prieto (2002) established in their studies on the development of natural fibre composites of high-temperature thermoplastic polymers, that thermosetting liquid epoxy resins can be a useful reactive solvent for combining wood flour particles with a high-temperature thermoplastic polymer, poly(phenylene ether) (PPE) because the composites offered a significant reduction in density and better mechanical and physical properties when compared to commercially available grades of engineering polymer blends filled with short glass fibres. Chemicals may activate hydroxyl groups or introduce new moieties that can effectively interlock with the matrix. Bledzki and Gassan (1999) reported that Sterzynski and co-workers used dimethylurea (in aqueous and methanol solutions) as a coupling agent for injection moulded flax-polypropylene composites. A dimethylurea concentration of 12 wt% leads to a 25% increase in tensile strength and a 20% increase in the Young's modulus.

Thermogravimetric analysis, TGA, of untreated and different surface modified jute fibres was performed by Mohanty et al. (2000) for the realisation of composites based on a Biopol matrix; all surface modified fabrics show the same or better thermal stability compared to untreated fabrics. In any case processing and service temperatures for vegetable fibres cannot exceed 200-220°C—limiting the availability of suitable polymeric matrix. In this work, the main objective was to obtain optimized level of the curative and moulding temperature for epoxy kenaf fibre/engineering thermoplastic ABS composites.

## **II. Materials and Methods**

### **Materials**

The engineering thermoplastic used as the matrix in this test is Polytac ABS which was an extrusion grade resin purchased from Chi Mei Company, NaOH and the kenaf of 3mm, all provided by Science and Engineering Research Centre (SERC), Universiti Sains Malaysia, Nibong Tebal, Malaysia.

## Methods

### Surface Treatment and Coating

Kenaf fibre of 3mm sizes was used. Firstly, the fibres were air dried in the lab at about 37°C for 2days prior to being immersed in 6% NaOH for 3hrs, washed and neutralized with 100% acetic acid, finally washed thoroughly with distilled water. Thereafter, the fibres were oven dried at 80°C for 16hrs. 40% epoxy of very low viscosity thinned with chloroform was used to coat the NaOH treated fibre, first oven dried between 30 – 40°C for 16hrs and temperature raised to 80°C for more 8hrs to dry finally.

### Thermogravimetric Analysis (TGA)

TGA was conducted from room temperature to 600°C at a heating rate of 10°C/min under nitrogen atmosphere using 10 – 15mg samples weight.

### Thermoplastic preparation

The engineering thermoplastic (ABS) was dried in a vacuum oven at 90°C for 3hrs.

### Compounding

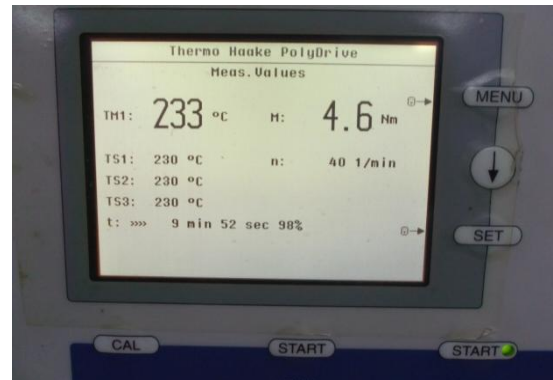
The constituent materials (ABS /Epoxy coated Kenaf fibre) after being dried were melt-mixed using a fibre loading of 12% in an internal mixer by varying the curative loading (2, 4, 6, and 8 wt.% 4,4-diaminodiphenyl sulfone, DDS). The mixing processes were performed for 10mins at temperature of 230°C using a rotator speed of 40rpm.

### Compression Moulding

The melt-mixed composites were dried at 60°C for 3h before moulded into sheets at the compression moulding machine. The composites were pre-heated for 5mins followed by compression moulding for another 5mins at 200, 210, 220 and 230°C moulding temperatures. After that, the samples were cooled through at ambient temperature for 3mins.



**Figure1a:** The epoxy treated kenaf fibre and ABS thermoplastic.



**Figure 1b:** The compounding meter of Haake PolyDrive.



**Figure 1c:** The Compression Moulding Machine



**Figure 2:** Universal (Instron 3366) Testing Machine

**Tensile testing**

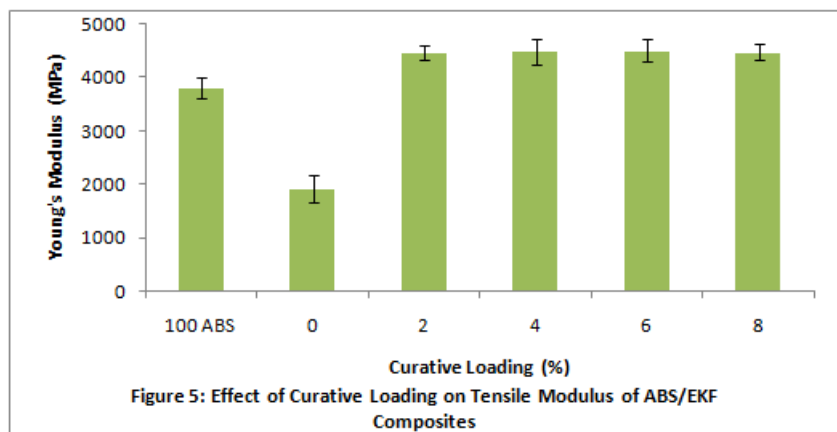
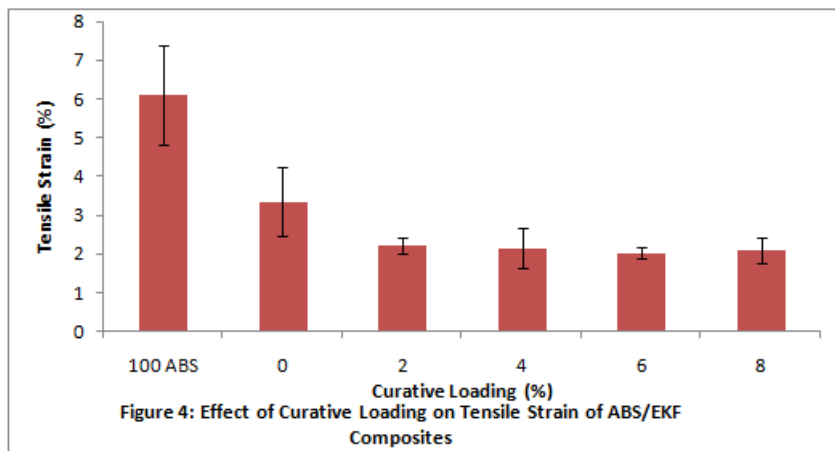
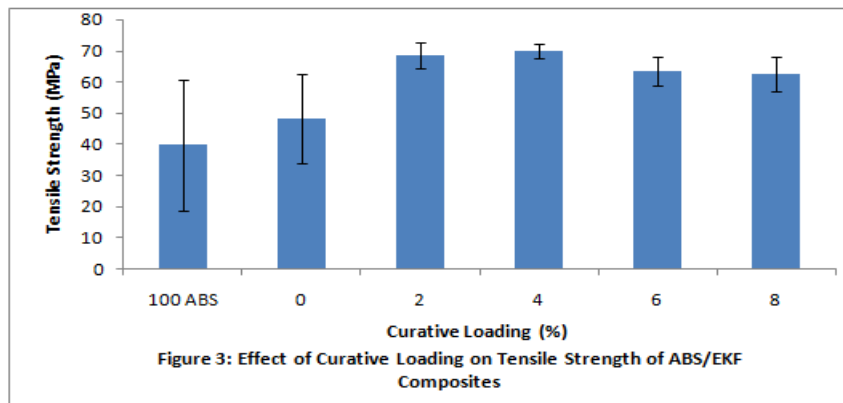
Tensile test was conducted using the universal testing at  $23 \pm 2^\circ\text{C}$  and  $50 \pm 5\%$  relative humidity in accordance to ASTM D882 - 97 with a guage length of 100mm and cross-head speed of 1mm/min. The specimens of 130 x 13 x 1mm were prepared by cutting with a band saw. Five measurements were carried out on each sample.

**Scanning Electron Microscopy (SEM)**

Fractured from the un-notched Izod impact test samples of composites were observed under a field-emission scanning microscope. Prior to observation, the samples were sputter-coated with a thin layer of gold to avoid electrical charging during examination.

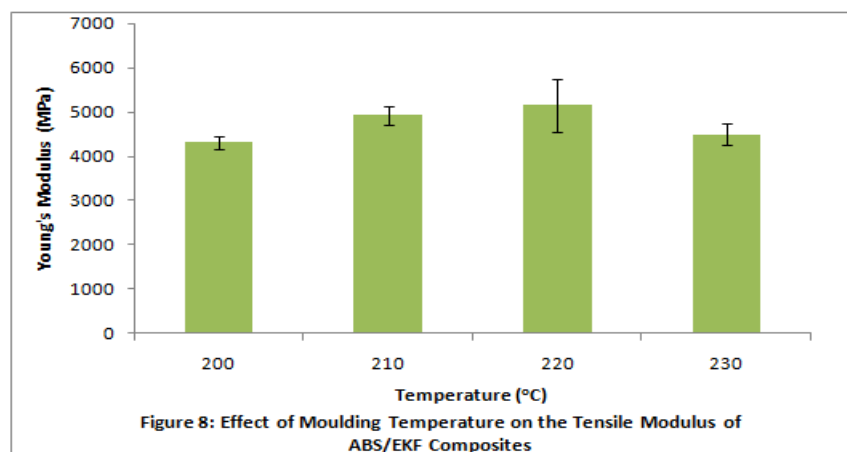
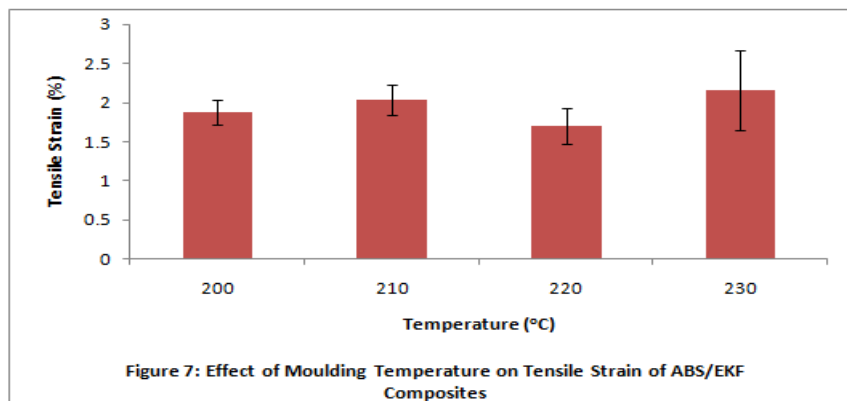
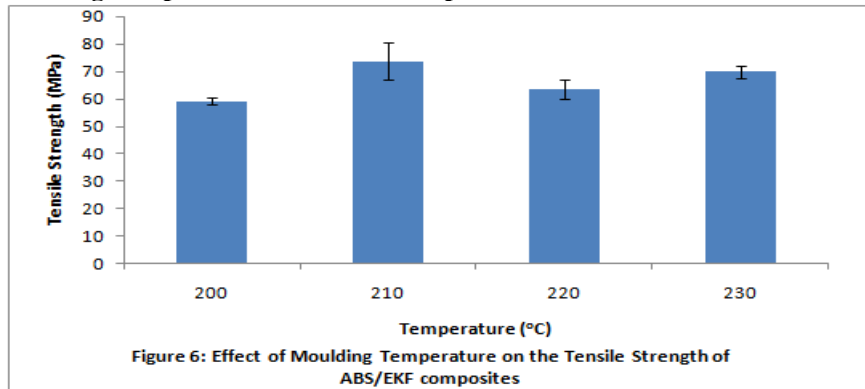
**III. Results And Discussion**

**The Effect of Curative loading on Tensile Properties**



The effect of curative loading on tensile properties of ABS/kenaf composites were depicted in Figures 3–5. From Figure 3, it was observed that tensile strength increased with increasing curative to a maximum of 4 wt.% loading signifying an optimum crosslinking of the total thermosetting resin (epoxy) used. However, with an additional 2 and 4 wt.% DDS make the strength to decrease by 9 and 11% respectively. It was clearly observed that small amount of curatives are needed to achieve greater tensile strength after cured on epoxy just as displayed in Figure 3 where the strength of 2 and 4 wt.% curative loading amounted to 68.445 and 69.85MPa respectively. This signifies increased in interfacial bonding due to increase from 2 wt.% to 4 wt.%. From Figure 4, tensile strain decreased with increasing curative up to 6 wt.%. The tensile modulus behaved in similar way like the tensile strain (Figure 5).

### The Effect of Moulding Temperatures on Tensile Properties



The effect of moulding temperatures on the tensile properties was examined and results represented in Figures 6–8. Optimum tensile strength was obtained at 210°C as it can be seen from Figure 6 which signifies no excessive degradation of the fibre and polymer at that particular temperature. However, with a further increase of temperature to 220°C, there was a 14% decrease indicating that much degradation of fibres had set in: this

being in line with the findings of Bogoeva-Gaceva et al. (2007) and El-Shekeil et al. (2011). An increase of 10% was observed when the temperature was increased to 230°C, although this is not surprising because the polymer film used to cover the composites during the moulding melted with the composite was unable to remove and therefore the strength was increased as a result of the polymer film, which might also account for the high tensile strain (Figure 7). Figure 8 showed an increase in tensile modulus from 200 to 220°C and a sudden decrease at 230°C from 5151.333 to 4490.25MPa (13% decrease).

### The Effect of Curative on Thermal Properties

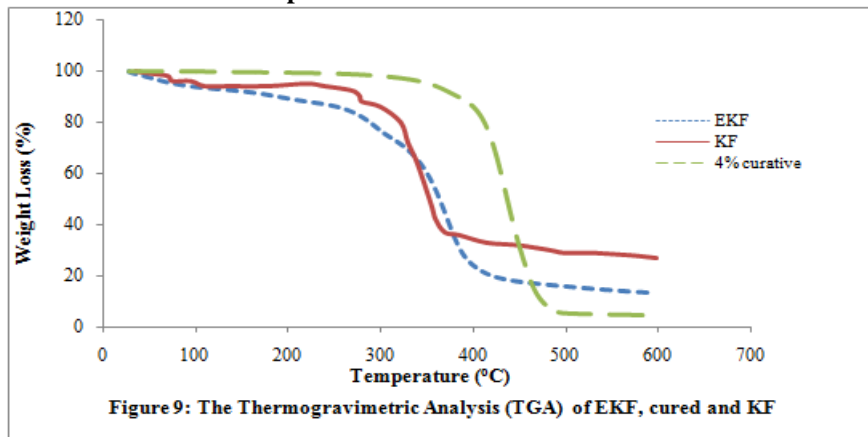


Figure 9: The Thermogravimetric Analysis (TGA) of EKF, cured and KF

It is believed that temperatures higher than 200°C would be critical for natural fibres as degradation might start affecting the mechanical properties of the fibres (Bogoeva-Gaceve et al., 2007). But the result in Figure 9 has proven otherwise as kenaf fibre has its onset degradation at 320°C and final degradation at 355°C. Furthermore, kenaf fibre coated with epoxy resin (EKF) has a final degradation of 399°C. The resin served as protective layer over the fibre preventing an early degradation and when cured at elevated temperatures during compounding made the fibres withstand the compounding temperatures without much degradation (Figure 9). The on-set degradation of cured composite started at 330°C; this temperature is called initial temperature of decomposition,  $T_i$ , having its peak degradation at 410°C and maximum degradation at 482°C referring to the temperature of maximum decomposition,  $T_m$  to give the residual ash. The 4% curative gives the best stability to thermal degradation. This might be as a result of the optimum curing stage.

### The Morphological Properties

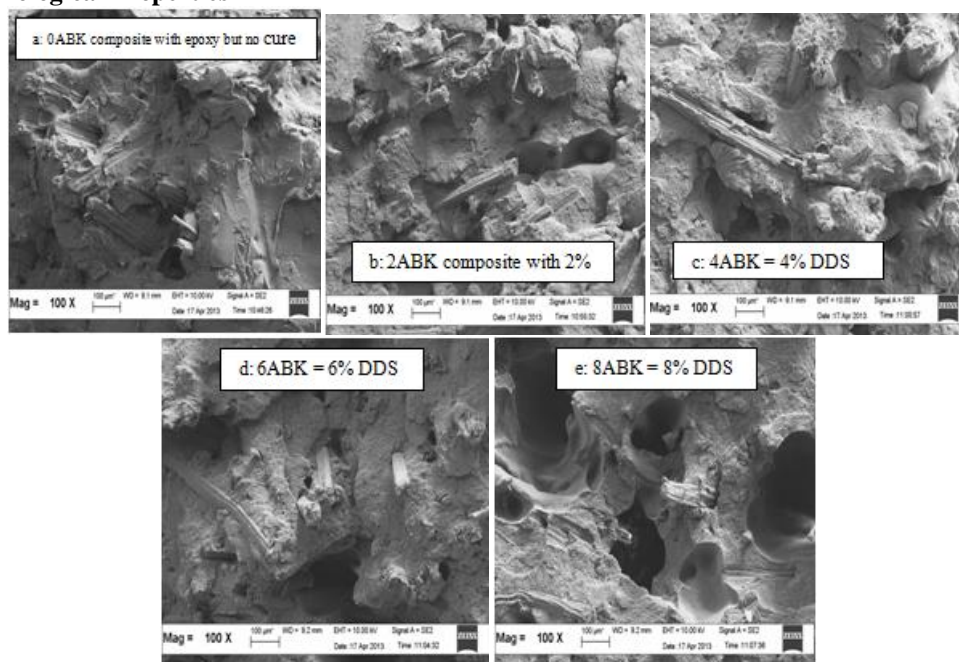


Figure 10: SEM Micrographs for the Composites (a: composite with epoxy no cure, b: with 2% DDS, c: with 4% DDS, d: with 6% DDS and e: with 8% DDS).

The impact fracture surfaces of composite samples were used for SEM to study dispersion, distribution, and state of kenaf fibres in the polymer matrices (Figures 10a to 10e). Figure 10a shows (OABK) composite with the epoxy coated fibres but no curative. There are fewer holes, no scattering of fibres, wetness of fibres and less pull outs as depicted in Figure 10b indicating poor adhesion between the fibre and the matrix ABS. These observations are similar to that of Supri & Ismail (2011) and Han et al. (2012). The observation of nodular morphology confirms the evidence of formation of epoxy coating layer on the fibres as was observed by Jana & Prieto (2002a). Consequently, Figure 10b represents the micrographs of 2 wt.% curatives (DDS) on the epoxy coated fibres composite which resulted into substantial wetting of fibre by the polymer indicating improved interactions and adhesion. Figure 10c, with 4 wt.% curative loading further showed better interactions and adhesion responsible for the best tensile strength attributes recorded. When the curative level was further increased to 6 and 8 wt.%, reduction in fibre wetting, pull outs, large holes and scattering increased as represented in Figures 10d and 10e. This can be attributed to excessive curative on epoxy fibres.

#### IV. Conclusion

The effects of high temperature curative and moulding temperatures on tensile and thermal properties of engineering thermoplastic ABS/epoxy coated Kenaf Fibre (EKF) composites were studied. From the results, it can be concluded that high temperature curative has improved the tensile strength and thermal stability of epoxy coated kenaf fibre as well as improving the wetting and reduce fibre pull out of the matrix. Also, coating natural bast fibres like kenaf with epoxy will definitely improve the thermal stability either with or without curative.

#### Reference

- [1]. Abdullah, A.H.; Khalina, A. & Ali, A. (2011). Effects of fibre volume fraction on unidirectional kenaf/epoxy composites: The transition region. *Polym. Plast. Technol. Eng.* 50 (1), 1362–1366.
- [2]. Aji, I. S.; Sapuan, S.M.; Zainuddin, E.S. & Abdan K. (2009) “Kenaf fibres as reinforcement for polymeric composites: a review”. *International Journal of Mechanical and Materials Engineering (IJMME)*, 4 (3): 239-248.
- [3]. Ashori, A. & Raverty, A. (2007). Printability of sized kenaf (*Hibiscus cannabinus*) papers. *Polym.-Plast. Technol. Eng.* 46 (7): 683–687.
- [4]. Bisanda E. and Ansell M. (1992). Properties of sisal–CNSL composites. *J Mater Sci* 27; 690–700.
- [5]. Bledzki, A.K. and Gassan, J. (1999). Composites reinforced with cellulose based fibres. *Progress in Polymer Science*, 24; 221 – 274.
- [6]. Bogoeva-Gaceva, G.; Avella, M.; Malinconico, M.; Buzarovska, A.; Grozdannov, A.; Gentile, G. & Errico, M.E. (2007). Natural Fibre Eco-composites, *Polymer Composites* 28(1), 98 – 107.
- [7]. Bos, H.L., and Donald. A.M. (1999). In situ ESEM study of the deformation of elementary flax fibres. *Journal of Material Science.* 34(13): 3029 – 3034.
- [8]. El-Shekeil, Y.A.; Sapuan, S.M.; Abdan, K. & Zainudin, E.S. (2011). “Development of a new kenaf bast fibre-reinforced thermoplastic polyurethane composite” *BioResources* 6 (4): 4662 – 4672.
- [9]. El-Shekeil Y. A.; Sapuan S. M.; Khalina A.; Zainudin E. S. & Al-Shuja’a O. M. (2012): Influence of chemical treatment on the tensile properties of kenaf fibre reinforced thermoplastic polyurethane composite. *eXPRESS Polymer Letters* Vol.6, No.12 1032–1040.
- [10]. Geethamma V.; Joseph R. & Thomas S. (1995). Short coir fibre-reinforced natural rubber composites: effects of fibre length, orientation and alkali treatment. *J Appl Polym Sci* 55; 583–94.
- [11]. Han, Seong Ok; Mehdi Karevan; I. Na Sin; Md A. Bhuiyan; Young Hun Jang; Jonathan Ghaffar & Kyriaki Kalaitzidou (2012). “Understanding the reinforcing mechanisms in kenaf fibre/PLA and kenaf fibre/PP composites: A comparative study” *International Journal of polymer Science*, 2012: 1 – 8. Doi: 10.1155/2012/679252.
- [12]. Ibraheem, S.A.; Ali, A. & Khalina, A. (2011). Development of green insulation boards from kenaf fibres and polyurethane. *Polym.-Plast. Technol. Eng.* 50 (6), 613–621.
- [13]. Iji M. (2008); Highly functional bioplastics used for durable products. In: *The Netherlands Science and Technology (Organizer and Editor); Innovative Technologies in Bio-Based Economy. Wageningen, The Netherlands, 8<sup>th</sup> April; 2008. [http://www.twanetwork.nl].*
- [14]. Ismail, H.; Norjulia, A. M. & Ahmad, Z. (2010). “Curing Characteristics, Mechanical and Morphological Properties of Kenaf Fibre/Halloysite Nanotubes Hybrid-Filled Natural Rubber Compounds” *Polymer-Plastics Tech. and Eng.* 49: 938 – 943.
- [15]. Jana, S. C. and Prieto, A. (2002). On the Development of Natural Fibre Composites of High- Temperature Thermoplastic Polymers *J Appl Polym Sci* 86, 2159 –2167.
- [16]. John, M. J. and Anandjiwala, R. D. (2008). “Recent developments in chemical modification and characterization of natural fibre-reinforced composites”. *Polym. Compos.* 29: 187–207.
- [17]. Karahaliou, E.-K.; and Tarantili, P.A. (2009). Preparation of Poly(Acrylonitrile-Butadiene- Styrene)/Montmorillonite Nanocomposites and Degradation Studies During Extrusion Reprocessing, *Journal of Applied Polymer Science*, Vol. 113, No. 4, (August 2009), pp. 2271-2281, ISSN 1097-4628.
- [18]. Karina, M.; Onggo, H. & Syampurwadi, A. (2007). Physical and Mechanical Properties of Natural Fibres filled Polypropylene Composites and its Recycle, *Journal of Bio. Sci.* 7. (2) 393-396.
- [19]. Mohanty, A.K., Misra, M. & Hinrichsen, G. (2000). Biofibres, biodegradable polymers and biocomposites: an overview, *Macromol. Mater. Eng.* 276/277, 1 – 25.
- [20]. Mwaikambo L. and Ansell M. (2002). Chemical modification of hemp, sisal, jute and kapok fibres by alkalization. *J Appl Polym Sci.* 84(12): 2222– 2234.
- [21]. Ochi S. (2008); Mechanical properties of kenaf fibres and kenaf/PLA composites. *Mech Mater*;40: (4-5) 446–452.
- [22]. Pan P.; Zhu B.; Kai W.; Serizawa S.; Iji M. & Inoue Y. (2007); Crystallization behaviour and Mechanical properties of bio-based green composites based on poly(L-lactide) and kenaf fibre. *J Appl Polym Sci*; 105:1511–20.

- [23]. Rowell, R.M. (1995). Chemical modification of agricultural fibres for property enhanced composites. In: Olesen, Ole; Rexen, Finn; Larsen, Jorgen, eds. Research in Industrial Application of Non Food Crops, I: Plant Fibres. Proceedings of a seminar; 1995 May; Copenhagen, Denmark. Lyngby, Denmark Academy of Technical Sciences: 49-70.
- [24]. Rowell, R.M.; Sanadi, A.R.; Jacobson, R.E. & Caulfield, D.F. (1999). In: Kenaf properties, Processing and Products. Mississippi State University, Ag and Bio Engineering, Chap 32, 381-392.
- [25]. Sanadi, A.R.; Caulfield, D.F.; Jacobson, R.E. & Rowell, R.M., (1995). Renewable Agricultural Fibres as Reinforcing Fillers in Plastics: Mechanical Properties of Kenaf Fibre-Polypropylene Composites, Ind. Eng. Chem. Res., 34, 5, 1889-1896.
- [26]. Sanadi, A.R.; Hunt, J.F.; Caulfield, D. F.; Kovacsvolgyi, G. & Destree, B., (2001). High Fibre-Low Mayrix Composites: Kenaf Fibre/Polypropylene. A paper presented at The Sixth International Conference on Woodfibre-plastic Composites at The Madison Concourse Hotel, Madison, Wisconsin on May 15-16, 121-124, retrieved from <http://www.fpl.fs.fed.us/documnts/pdf/2002/sanado2apdf> on 16/08/2012.
- [27]. Sreekala M.; Kumaran M. & Thomas S. (1997). Oil palm fibres: morphology, chemical composition, surface modification and mechanical properties. J Appl Polym Sci 66:821–35.
- [28]. Supri, A.G. and Ismail, H. (2011). The Effect of Isophorone Diisocyanate-Polyhydroxyl groups modified Water Hyacinth Fibres (Eichhornia Crassiper) on properties of Low Density Polyethylene/ Acrylonitrile Butadiene Styrene (LDPE/ABS) Composites. Polymer-Plastics Tech. and Eng. 50: 113 - 120. DOI: 10.1080/03602559.2010.531428.
- [29]. Wan Ahmad, W.Y.; Salleh, J.; Yahya, M.F.; Abdul Kadir, M.I.; & Misnon, M.I. (2007). Some properties of kenaf and kenaf combined waste composites. Woodhead Publishing Limited, <http://217.218.200.220/documents/10129/21410/ECotextiles+The+way+forward+for+sustainable+development+in+textiles+12.pdf> On 17/7/2012.