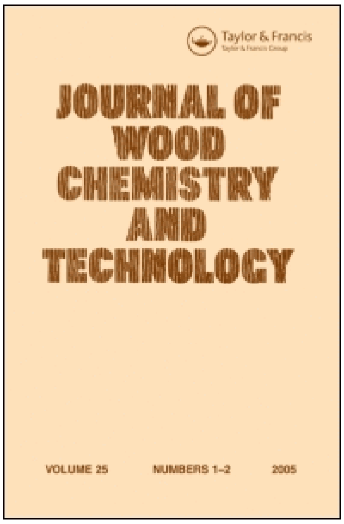

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Ultra-Violet Radiation-Cured Biofiber Composites from Kenaf: The Effect of Montmorillonite on the Flexural and Impact Properties

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Abstract: Biofiber composites, cured by ultra-violet (UV) radiation were produced using kenaf fibers as the reinforcing agent and unsaturated polyester as the matrix. This research work focused on the effects of the incorporation of kenaf fiber, montmorillonite (MMT), and cetyl trimethyl ammonium bromide-modified MMT (CTAB-MMT) in the unsaturated polyester composite. Overall, the incorporation of kenaf fibers in the form of mat had improved the flexural and impact properties of the composites. Addition of MMT into the kenaf fiber-polyester system showed an improvement up to 1% MMT after which it decreased. The increase was attributed to better stress transfer mechanism in the matrix. However, further increase in the MMT loading had resulted in the decrease in the properties, which was believed to be due to agglomeration. Modification of MMT with CTAB had produced composites with higher flexural and impact properties as compared to those without modification. This was attributed to a combination of effective dispersion of MMT in the matrix, availability of effective high aspect ratio MMT, and enhanced compatibility between CTAB-MMT with the matrix.

Keywords: Biofiber, composites, kenaf, lignocellulosic, ultraviolet radiation

INTRODUCTION

The use of natural fiber reinforcement in synthetic polymer composites has gained importance recently. Natural fiber is renewable and can be used in fiber-reinforced composites to replace glass and other non-renewable fibers. These fibers are cheaper and less abrasive to tools. Kenaf has gained research interest to be used as a lignocellulosic filler when compared to other lignocellulosic fillers. One of the advantages is the absence of silica content, which

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is critical in reducing abrasiveness to the processing equipment.[1] The density of kenaf, 0.1–0.2g/cm^3,[2] is much lower than glass fiber (2.55 g/cm^3),[3] carbon fiber (1.77 g/cm^3)^3, oil-palm empty fruit bunch (0.7–1.55 g/cm^3),[4] and hemp (1.48 g/cm^3)[5] This low density gives kenaf core an advantage to be used in applications such as automotive interior panelling to reduce fuel consumption.[6] Market price for lignocellulosic filler is much cheaper than conventional fillers such as kevlar, glass fiber, and carbon fiber.[7] However, incorporation of lignocellulosic filler has been shown to adversely affect the strength of the composites.[8−12] This situation may outweigh the importance of economy posed by lignocellulosic composites. From various studies in nano composites, nano filler addition into polymer matrix increases the strength as well as the modulus of the composites.[13−17] With its high surface area and aspect ratio, the nano filler is expected to enhance the properties of lignocellulosic–polymer composites.

The thermoset-fiber-reinforced polymer matrix composites are currently produced from unsaturated polyester resins by thermally induced initiators. Examples are polyester pre-pregs, sheet molding compounds, bulk molding compounds, and so on.[18] The production of these composites involves use of high temperatures and requires expensive molds and hydraulic presses. Room-temperature curing of such systems takes a long time. UV-curable systems are used in surface coatings and have distinct advantages such as being solvent free, environmentally friendly, and energy efficient.[19] The entire curing process occurs at room temperature and hence does not require expensive high-temperature equipment for the curing.[20] In view of the aforementioned advantages, attempts have been made to produce kenaf–unsaturated polyester composites filled with nanofiller using UV technology for curing.

EXPERIMENTAL

Materials

Unsaturated polyester resin P9728 was purchased from Euro Chemo Pharma Sdn. Bhd., Penang, Malaysia. Kenaf bast fiber was obtained from Kenaf Fibre Industries, Kelantan, Malaysia. Sodium montmorillonite with cationic exchange capacity (CEC) of 145 meq/100 g was purchased from Nanocor (USA). Photoinitiator Irgacure 1800 was supplied by Ciba Specialty (Singapore) Pte. Ltd.

Preparation of Composites

Kenaf bast fibers were cut into 5 mm length. The fibers were then immersed in water in a deckle box with a dimension of 20 cm × 20 cm. Then, the water was drained off and a fiber mat was formed.[21] The fiber mat was dried in an oven at 105°C overnight. The mats were then pressed using hot press Gotech Testing Machine Inc. model GT-7014-100, at 100°C, 500 kg/cm^2 for 1 h, to obtain mat with thickness of approximately, 1 mm. Three different loadings (1%, 3%, and 5% based on the overall weight of the composite) of cetyl trimethyl ammonium bromide-modified MMT (CTAB-MMT) and montmorillonite (MMT) were mixed separately with unsaturated polyester resin using a three roll mill mixer for 1 h. Three percent of photoinitiator Irgacure 1800 was added to the mixture. The mixture was then poured onto a kenaf bast mat. The impregnation of the mixture into the mat was carried out by using a hand roller. The impregnated mat was sandwiched between two glass panels. Finally, the mats were passed through 1st UV machine, model M20-1-Tr-SLC, for twenty passes at a conveyor speed of 10 m/min, to ensure complete cure of the sample. The machine consisted of a medium pressure mercury arc lamp with the UV radiation wavenumber of 180–450 nm.

Modification of MMT with CTAB

First, 150 g of MMT was dispersed in 1 L distilled water with vigorous stirring to form a uniformly suspended solution. An excessive amount of cetyl-trimethyl ammonium bromide (CTAB) was diluted into a mixture of distilled water and ethanol with a ratio of 1:1 (vol./vol.). Then, CTAB aqueous solution was added to the MMT suspended solution and stirred continuously at 80°C for 12 h. The obtained CTAB-MMT was washed repeatedly with distilled water to eliminate excess CTAB until no AgBr precipitate was detected through titration with 0.1 N $AgNO_3$ solution.

A small sample of CTAB-MMT was ground to fine flour and mixed with KBR to form KBR disc for Fourier transform infrared analysis (FTIR). The analysis was carried out using Nicolet Avatar 360, operating at 4 cm^{-1} resolution. Each run was comprised of 200 scans.

X-ray diffraction analysis was carried out using a Shimadzu X-RD 600 X-Ray Diffractometer, equipped with a CuKα radiation ($\lambda = 1.542$ Å).

Testing

The samples were cut according to the types of testing—flexural and impact tests. Flexural test was carried out on samples with dimension of 10 cm $\times$ 1.5 cm $\times$ 0.2 cm. The test was conducted according to ASTM D790, that is, a three-point bending method using Universal Testing Machine model STM-10 at a cross-head speed of 1.0 mm/min. The Izod impact test was carried out on

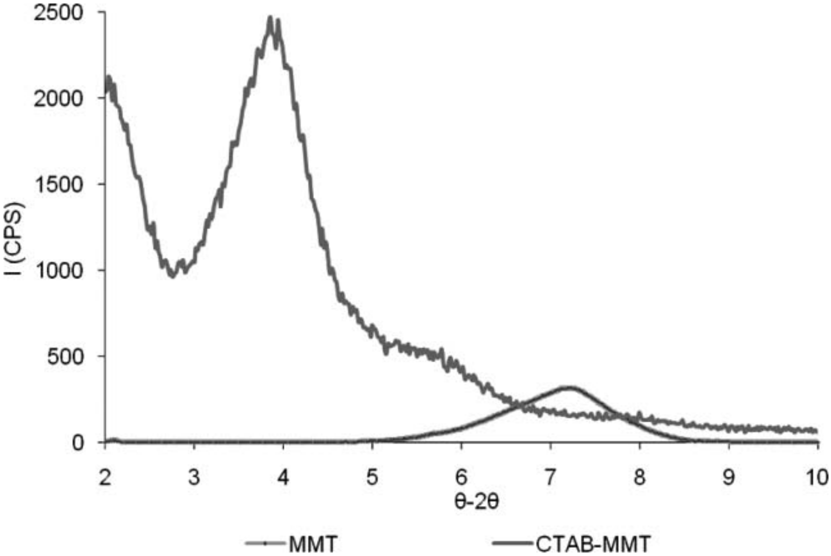

Figure 1. XRD analysis of MMT and CTAB-MMT.

unnotched samples with dimension of 6.5 cm × 1.2 cm × 0.2 cm using an Impact Pendulum Tester (Zwick) model 5101 according to ASTM D252.

RESULTS AND DISCUSSION

Analysis of XRD for MMT and CTAB-modified MMT (CTAB-MMT) is shown in Figure 1. For MMT, the peak is detected at 7.08°, corresponding to basal spacing of 12.48 nm. For CTAB-MMT, the peak is at 3.89°, corresponding to basal spacing of 22.69 nm. Thus, it is obvious that as the result of CTAB modification, the basal spacing increases 10.21 nm. The increase in basal spacing corresponds to greater separation of MMT platelet, which indicates intercalation.[22]

Figure 2 shows the infrared spectra of MMT and CTAB-MMT. The peaks shown for MMT are 460, 520, and 1031 cm^{-1}, corresponding to Si-O-Si deformation, Si-O-Al, and Si-O stretching (REF), respectively. The new peaks detected for CTAB-MMT, 2852 and 2924 cm^{-1}, are assigned to stretching vibrations for $–CH_2$ and $–CH_3$ groups.[23] This indicates the presence of CTAB, in the form of long alkyl chain on the MMT.

The effect of kenaf fiber and MMT loading on flexural strength of the kenaf–unsaturated polyester composites are depicted in Figure 3. The results show that the flexural strength increases as the kenaf fiber loading is increased. All samples display significantly higher strength than those without kenaf fiber.

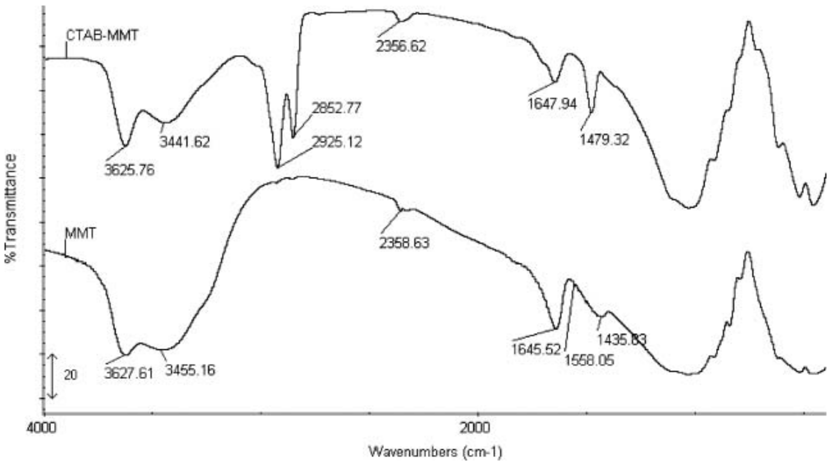

Figure 2. FTIR spectra of MMT and CTAB-MMT.

This clearly shows that the fibers, which are in the form of the mat, are able to reinforce the composite. This also indicates some degree of compatibility between the fiber and unsaturated polyester matrix. This may come either in the form of primary bonding, ester bonding, or secondary bonding, hydrogen bonding. The former is possible through the reaction of OH groups from kenaf with COOH of maleic anhydride residue of polyester and the latter through the interaction of either hydrogen or oxygen from either of the components. As for the effect of MMT incorporation, composites exhibit the highest flexural strength at 1% MMT, after which it decreases. Thus, the result indicates that the incorporation of MMT has significantly improved the stress transfer mechanism

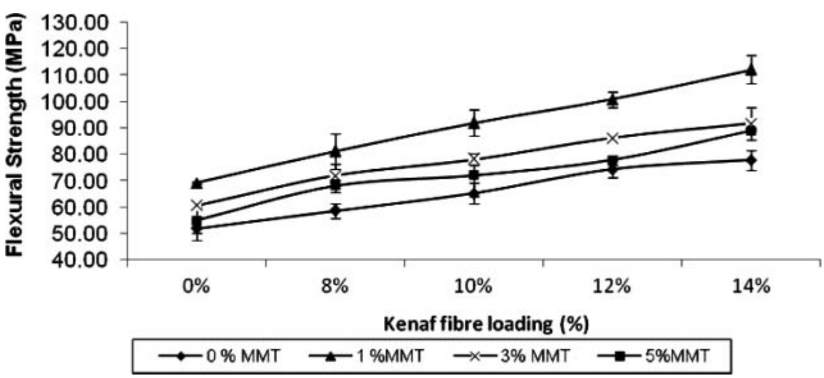

Figure 3. The effect of kenaf and MMT content on the flexural strength of kenaf–unsaturated polyester composite.

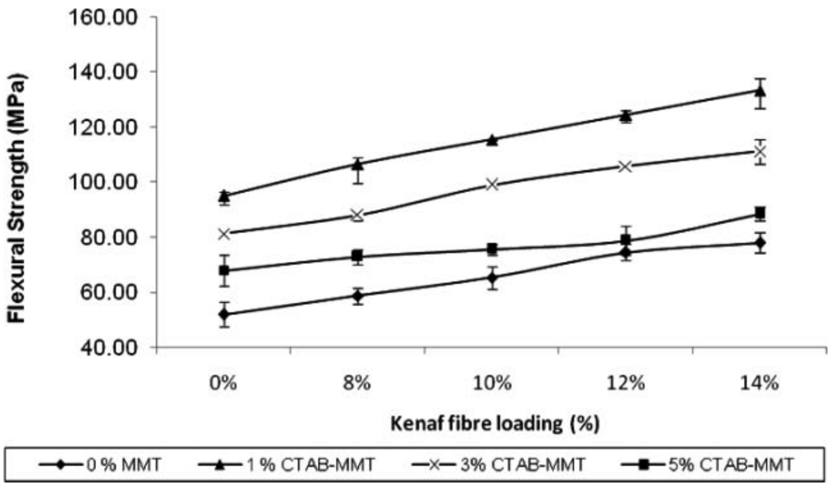

Figure 4. The effect of kenaf and CTAB-MMT content on the flexural strength of kenaf–unsaturated polyester composite.

of the samples. The MMT loading of 1% is sufficient to produce a significant increase in flexural strength as compared to those without MMT. However, further increase in the amount of MMT decreases the strength. This may be due to the agglomeration of MMT as observed by several previous studies.[24–26]

Figure 4 shows the effect of CTAB modification of MMT on flexural strength of the kenaf–unsaturated polyester composites. Overall, the trend is

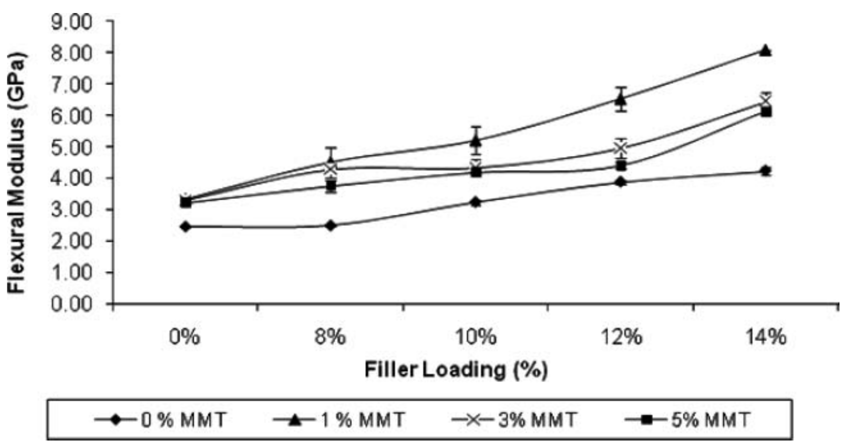

Figure 5. The effect of kenaf and MMT content on the flexural modulus of kenaf–unsaturated polyester composite.

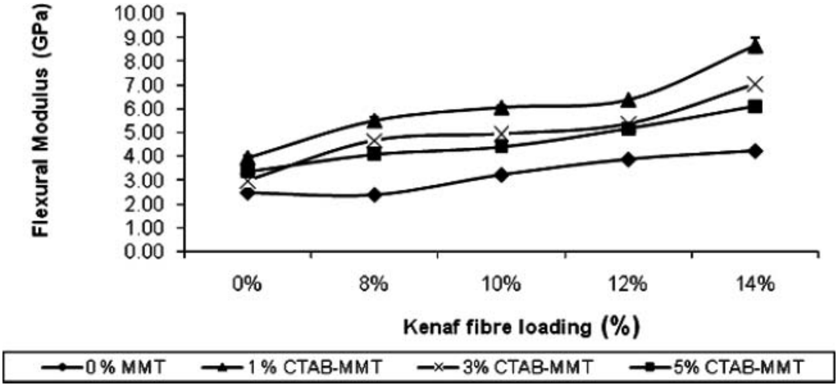

Figure 6. The effect of kenaf and CTAB-MMT content on the flexural modulus of kenaf–unsaturated polyester composite.

similar to those without CTAB modification (Figure 3). However, it can be seen that the magnitude of the strength is higher in the CTAB-MMT than in MMT composites, especially for those with 1% and 3% CTAB-MMT. This indicates that the stress transfer mechanism of the composites with CTAB-MMT is more efficient than those with MMT. This may be attributed to intercalation of MMT platelets, which leads to (i) more effective dispersion of MMT in the matrix and (ii) the availability of effective high aspect ratio MMT. Furthermore, the introduction of long polymer chain from CTAB adds hydrophobicity to the MMT, which increase the compatibility with the unsaturated polyester matrix.

The effect of kenaf fiber and MMT loading on flexural modulus of the kenaf–unsaturated polyester composites are depicted in Figure 5. In general, flexural modulus of all samples increase as the kenaf filler loading is increased. It is expected since kenaf, being a lignocellulosic material, has its own inherent stiffness that contributes to the overall stiffness of the composites. The composites with 1% MMT loading display the highest modulus, which is similar in trend for flexural strength. As shown by various studies,[27–29] incorporation of MMT imparts stiffness to the composites. Figure 6 shows that MMT modification with CTAB produces composites with higher modulus than those without (Figure 5). It is believed that the enhanced compatibility and distribution of CTAB-MMT with and in the matrix are the main factor in increasing the stiffness of the composites. With the introduction of long hydrophobic chains of CTAB on the MMT, the interaction between the former with polyester chains is expected to be increased. This, in turn, restricts the mobility of the polyester molecular chains in the matrix, resulting in an increased stiffness. The restriction in mobility is further enhanced with the ability of the polyester to flow into the intercalated spaces. The effect of kenaf

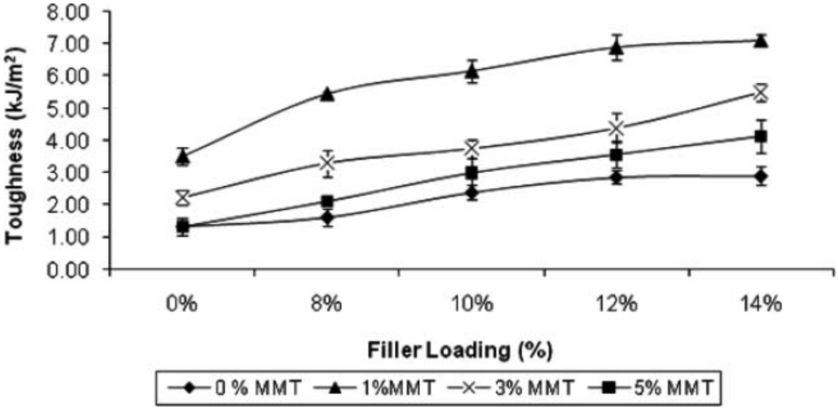

Figure 7. The effect of kenaf and MMT content on the flexural toughness of kenaf–unsaturated polyester composite.

filler loading on the CTAB-MMT composites shows the same trend as those of flexural strength, where the highest modulus is from those with 1% CTAB-MMT. The same explanation presented for the flexural strength applies to the modulus.

Toughness reflects the energy being absorbed by a sample before failure. Thus, from the toughness results (Figure 7), it is obvious that those with 1% MMT have the capability to absorb more energy before failure. This may indicate that at 1%, the stress transfer mechanism is at the highest point in the given condition, after which agglomeration may take effect. It can be seen that the composites with CTAB-MMT display higher toughness than those with MMT (Figure 8). It indicates that those with CTAB-MMT have the ability to absorb more energy before failure. This ability may be contributed by the better distribution of high aspect ratio MMT in the matrix and the introduction of long polymer chain from CTAB, which adds hydrophobicity to the MMT, which subsequently increases the compatibility with the unsaturated polyester matrix.

Figure 9 shows the effect of kenaf fiber, MMT, and CTAB modification of MMT on impact strength of the kenaf–unsaturated polyester composites. It is known that the impact strength of a composite is highly influenced by interfacial bond strength, matrix, and fiber properties.[23] The results show that the impact strength increases as the kenaf fiber loading is increased. All samples display significantly higher strength than those without kenaf fiber. This clearly shows that the fibers have the ability to reinforce the composite. This also indicates the presence of some degree of compatibility between the fiber and unsaturated polyester matrix as explained earlier. With MMT, composites exhibit the highest impact strength at 1% MMT, after which it decreases. Thus, the result indicates that the

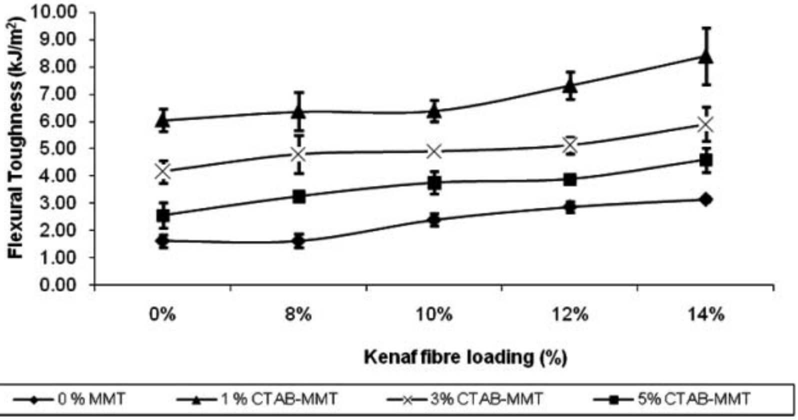

Figure 8. The effect of kenaf and CTAB-MMT content on the flexural toughness of kenaf–unsaturated polyester composite.

incorporation of MMT has significantly improved the energy absorption of the samples. The MMT loading of 1% is sufficient to produce a significant increase in impact strength as compared to those without MMT. However, further increase in the amount of MMT decreases the strength, which may be due to agglomeration of MMT as observed by several previous studies.[24–26]

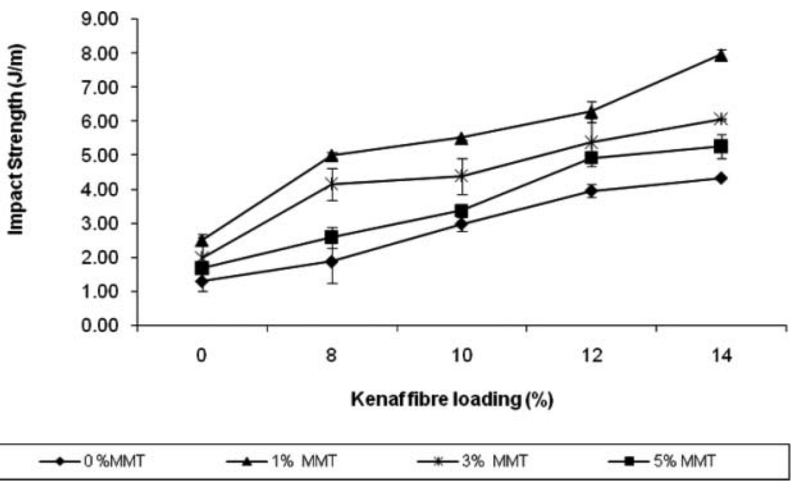

Figure 9. The effect of kenaf and MMT content on the impact strength of kenaf–unsaturated polyester composite.

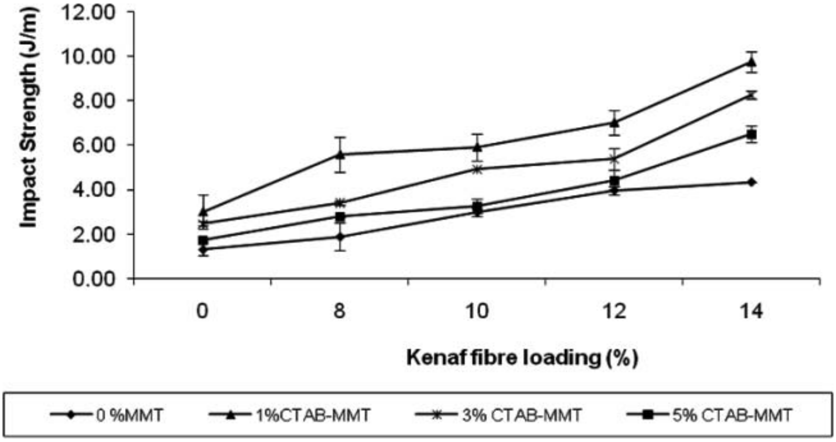

Figure 10. The effect of kenaf and CTAB-MMT content on the impact strength of kenaf–unsaturated polyester composite.

Figure 10 shows the effect of CTAB modification of MMT on impact strength of the kenaf–unsaturated polyester composites. Overall, the trend is similar to those without CTAB modification (Figure 9). However, the impact strength of those with CTAB-MMT is higher than the ones with MMA, especially for those with 1% and 3% CTAB-MMT. This indicates that the stress transfer mechanism and absorption of energy of the composites with CTAB-MMT is more efficient than those with MMT. This may be contributed by a combination of effective dispersion of MMT in the matrix, availability of effective high aspect ratio MMT, and enhanced compatibility between CTAB-MMT with the matrix.

CONCLUSIONS

The effects of the incorporation of kenaf fiber, MMT, and CTAB-MMT in the unsaturated polyester composite were investigated. Overall, it was obvious that incorporation of kenaf fibers in the form of a mat had improved the flexural and impact properties of the composites. Addition of MMT into the kenaf fiber–polyester system showed an improvement up to 1% MMT, after which it decreased. The increase was attributed to better stress transfer mechanism in the matrix. However, further increase in the MMT loading had resulted in the decrease in the properties, which was believed to be due to agglomeration. Modification of MMT with CTAB had produced composites with higher flexural and impact properties as compared to those without modification. This was attributed to a combination of effective dispersion of MMT in the matrix, availability of effective high aspect ratio MMT, and enhanced compatibility between CTAB-MMT with the matrix.

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