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The Influence of Carbon Fiber Content on the Tensile, Flexural, and Thermal Properties of the Sisal/PMMA Composites

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Abstract: The polymethyl-methacrylate (PMMA)-based composites have been developed for denture-based materials. Sisal/PMMA and sisal/carbon/PMMA composites are a good choice for denture-based materials. Therefore, the different properties are presently studied to explore the effect of carbon fiber content on the tensile, flexural, and thermal properties of untreated sisal (US)/treated carbon (TC)/PMMA composites. The composites were made by hand lay-up and cold press with 20% fiber loading and different US/TC ratios of 2:1, 1:1, and 1:2. The carbon fiber was treated by soaking it in a solution of 68% HNO₃ for various durations from 24 to 96 hours. The scanning electron microscopy (SEM) results revealed an excellent bonding strength between the fibers and the matrix and a high degree of fiber dispersion in the matrix. These phenomena led to the composites with a 1:2 US/TC ratio reaching the maximum tensile strength (57.70 ± 6.15 MPa) and flexural strength (108 ± 6.61 MPa). The mechanical properties of the composites improved by adding carbon fiber to the sisal/PMMA composite. The thermogravimetric (TGA) analysis indicated that the thermal stability of the US/TC/PMMA composites increases by the addition of carbon fiber.

Keywords: Carbon, Composite, TGA, Mechanical properties, PMMA, Sisal

1. Introduction

Natural fiber composites have recently attracted worldwide research interest, especially composite for biomedical applications due to their lightweight, high corrosion-resistance, low cost, and functional environmental aspects. Sisal fiber (*Agave Sisalana*) is a natural fiber used as the reinforcement material for biomedical composites because of its biocompatibility, biodegradable, antimicrobial, and anti-inflammation¹⁻³. Polymethyl-methacrylate (PMMA) based composites have been developed for biomedical devices such as bone plate and screw, bone cement, and dental applications⁴. In addition, PMMA reinforced with various filler types (natural fibers, synthetic fibers, particles, fiber-particle hybrid) for denture-based materials have been reviewed⁵.

The sisal/PMMA composites with different fiber contents from 2.5% to 10% have been studied for dental applications, resulting in the highest flexural strength of

about 57 MPa for 10% alkali-treated fiber loading and 52 MPa for the untreated fiber content⁶. With a similar fiber content, the flexural modulus resulted in the highest value of around 2.7 MPa for alkali-treated sisal and 2.5 MPa for the untreated sisal. Besides, kenaf (2 wt.%) reinforced PMMA composite has been studied for denture, which resulted in the flexural strength of 82.5 MPa⁷.

Carbon fiber, known for its superior properties, is combined with sisal fiber to produce hybrid composites to improve the properties of the corresponding composite. Carbon fiber is compatible with human tissue and biologically appropriate. A study has investigated the composites by combining sisal and carbon fibers to reinforce the polyester for characterizing the tensile and bending properties affected by different ratios of sisal fiber to carbon fiber⁸. The findings revealed a significant improvement in tensile and bending properties, remarkably higher than the previous results⁶. The tensile strength and tensile modulus were 122.11 MPa and 2.98

GPa. Meanwhile, the flexural strength and flexural modulus were 176.53 MPa and 13.47 GPa. It suggests that the inclusion of carbon fiber helps enhance the mechanical properties of the composites.

PMMA based composites reinforced with different concentrations of barium sulfate (BaSO_4) granules have been developed for biomedical applications, especially for preparing spacer devices in prosthetic infection⁹). The addition of BaSO_4 granules is intended to achieve high porosity, making antibiotics easier to absorb and deliver. The effects of the different particle fillers on the mechanical properties of PMMA-based composites have been studied. The addition of 5-20% hydroxyapatite (HA) microparticles increased the flexural modulus of PMMA¹⁰). The introduction of silica (SiO_2) particles improved the bending properties but decreased the impact strength¹¹). These properties changed with the volume fraction of SiO_2 particles. The smaller the particle size, the higher the mechanical properties due to the excellent strengthening mechanism. Small particles are considered to have a higher degree of dispersion in the matrix than larger particles, which inhibit the sliding of the matrix chains. Therefore, the stress transfer from the matrix to the fine particles is better than coarse particles. The insertion of 3% TiO_2 nanoparticles exhibited an optimum impact strength¹²). Recently, nanosized natural particles of pomegranate peels (PPP) and ajwa dates seed powder (SPDA) with PMMA fillers have been investigated. The results demonstrated significantly higher flexural properties of the composite than the ceramics nanoparticles mentioned above¹²). At a fraction of 1.6% pomegranate peels, flexural strength and flexural modulus attained the highest values of 114 MPa and 5 GPa, respectively¹³).

The previous study has compared the properties between the alkali-treated sisal/carbon fiber/PMMA hybrid composites and sisal/PMMA by adding maleic-anhydride-grafted polypropylene (MAPP) as the coupling agent. It suggested that hybridization by carbon fiber is more effective in improving the mechanical properties of the composite than by the addition of MAPP¹⁴). According to the study of PMMA-based biomedical composites, the development of the sisal/carbon/PMMA composite materials is of considerable interest for an alternative denture-based material because it is still scarce. Due to its biocompatibility, carbon has been commonly used in medicine, bioengineering, and biomaterials¹⁵).

In the current study, alkali-treated sisal (TS) and untreated sisal (US) fibers were combined with treated carbon (TC) fibers to reinforce the PMMA matrix. The tensile strength of TS/TC/PMMA composite results is used to optimize the modified surfaces of carbon fibers and the treatment condition. The integration of US and optimum TC fibers in various ratios is applied to reinforce the PMMA matrix. This study investigated the effects of carbon fiber content on the composite's tensile,

flexural bending, and thermal properties and discussed the changes in the properties from the relationship between the variations of carbon fiber content and the fracture surface morphology. Thermogravimetric analysis (TGA) is conducted to confirm the thermal degradation of US/TC/PMMA composites. The obtained results are compared to the previous studies. For the denture-based material application, the highest flexural strength achieved by the present composite is used as a basis of the simulation method to obtain the compressive strength.

2. Materials and methods

2.1 Materials

Sisal fiber obtained from Balittas-Malang, Indonesia, and Toray 1700sc 120000-50C carbon fiber with a tensile strength of 4.9 GPa and tensile modulus of 250 GPa supplied by Hobbyrover, China, were used as the reinforcement materials. PMMA (ISO 1567 Type II Class I), purchased from Dental Jaya, Indonesia, was used as a matrix material. NaOH and Nitric acid (HNO_3) were utilized to surface modification of sisal and carbon fibers.

2.2 Fiber treatment and composite fabrication

There were three kinds of fibers used: treated carbon (TC) fiber, untreated sisal (US) fiber, and alkali-treated sisal (TS) fiber. The carbon fibers were immersed in a 68% HNO_3 solution at room temperature for 24, 48, 72, and 96 hours, then dried in an oven at 80°C for 6 hours and designated as TC. US fibers were immersed in water for 24 hours after being washed in flushing water. These fibers were then dried at 80°C for 30 minutes before being chopped into a length of approximately 6 mm. US fibers were partially treated in an alkali solution (6% NaOH) for 4 hours before being neutralized with 1% acetic acid (CH_3COOH) called TS. Both TS and US fibers were combined with TC to strengthen the PMMA matrix and designated TS/TC/PMMA and US/TC/PMMA.

First, the TS/TC/PMMA composites with the 20 vol.% fibers, a sisal/carbon ratio of 1:1, and TC at each exposure duration to HNO_3 of 24, 48, 72, and 96 hours were made by hand lay-up in the cold press mold at 2.1 MPa for roughly 60 minutes and then subjected to a tensile test. Second, US/CT/PMMA composites were fabricated with the 20 vol.% fibers and the sisal/carbon ratios of 2:1, 1:1, 1:2 using the same process as TS/TC/PMMA composites. TC used was obtained from an optimum value of the TS/TC/PMMA composite's tensile strength. In this case, the tensile and bending tests were conducted on the US/CT/PMMA composites.

2.3 Mechanical and thermal characterization

The tensile test was performed according to the ASTM D638-01 using a universal testing machine (UTM, Zwick

Roell Z020) with a maximum load of 0.1 MPa and a crosshead speed of 5 mm/min. A three-point bending test following the ASTM D790-02 with a maximum load of 0.1 MPa and a 1 mm/min crosshead speed. Each case needs at least five composite specimens. Tensile test was done on the TS/TC/PMMA composites, while tensile, bending, and thermal tests on the US/TC/PMMA composites. A simulation approach was used to assess the compressive strength of the US/TC/PMMA composite, starting with the creation of geometry in Autodesk Inventor 2021 software and then inserting it into the ANSYS R18 program for compression testing simulation. The compression test geometry is designed following ASTM D 695.

The thermal test by thermogravimetric analysis (TGA, TA SDTQ600) was performed on the samples (US fiber and US/TC/PMMA composites) weighing 13-15 mg at a maximum temperature of 300°C and heating rate of 20°C/min. The weight loss of each sample was recorded and analyzed.

The scanning electron microscopy (SEM, TESCAN VEGA3 LMU, HITACHI SU-3500) was employed to characterize the surface morphology of carbon fibers and the tensile fracture of US/TC/PMMA composites. The bending specimens tested were examined with a digital optical microscope from the crack areas on the tension side.

2.4 Statistical analysis

The comparison of the mean bending strength was performed by Analysis of Variance (ANOVA) using Minitab-14 software. Multiple comparison testing was conducted with the Tukey method on comparisons between the data group without Carbon and three data group with carbon obtained p -Value < 0.05. It means that the addition of carbon does affect the bending strength and tensile strength.

3. Results and discussion

3.1 Surface morphology of carbon fibers

Figure 1 displays the SEM images of the untreated and treated carbon fibers of various exposure times to HNO₃. Surface treatment improves the surface roughness making good binding at the carbon fiber/matrix interface. The magnified images Fig.1b to Fig.1e demonstrate a slight change in surface roughness (as marked by white triangles): i.e., nearly identical to the previous HNO₃-treated carbon fibers¹⁶. Other studies^{17,18} demonstrated evidence of striation-like structure on the surfaces of HNO₃-treated carbon fibers, which is quite different from this finding. The difference in treatment temperatures might cause discrepancies in those surface structures, although the HNO₃ concentration and treatment time are in the same range. In this study, the HNO₃-treated carbon fibers with different surface roughness combined with alkali-treated sisal (TS) fibers

strengthen the PMMA composites. Further, the tensile strength of these TS/TC/PMMA composites is optimized and discussed.

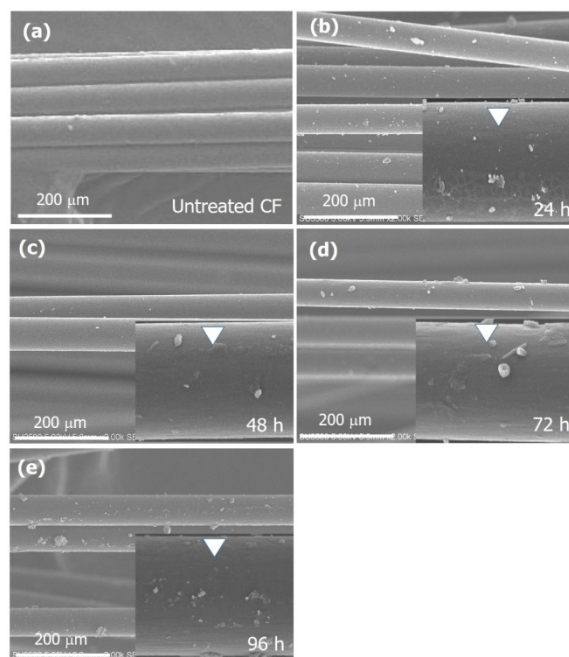


Fig. 1: SEM images of untreated carbon fibers (a) and treated carbon fibers for 24 h (b), 48 h (c), 72 h (d), and 96 h (e) of various exposure to HNO₃.

3.2 Tensile and flexural properties

The TS/TC/PMMA composites (Fig.2) reveal that the composite's tensile strength and tensile modulus with TC exposed longer than 48 hours (to HNO₃) gradually decrease. Therefore, exposure time to HNO₃ for 48 hours was selected for carbon fiber treatment and further characterization, during which TC was combined with the US to strengthen PMMA.

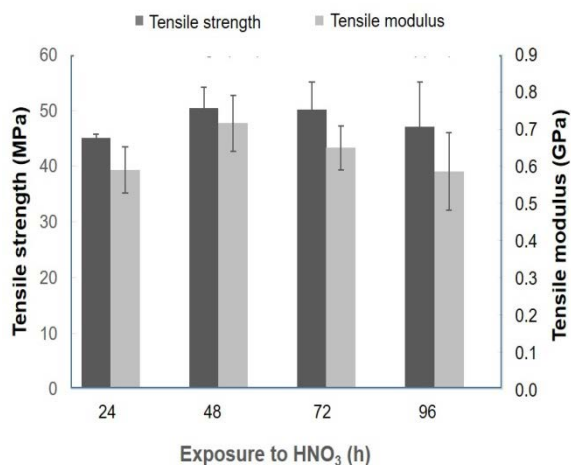


Fig. 2: Tensile properties of TS/TC/PMMA composites with different TC.

Data (mean ± SD) for tensile and flexural strengths in

Table 1 are obtained from statistical software (TestXpert II) integrated with the mechanical testing machine (Zwick/Roell). Analysis of variance with one data group of specimens without carbon (US/PMMA) and three data groups of specimens with carbon (US/TC (2:1)/PMMA, US/TC (1:1)/PMMA, and US/TC (1:2)/PMMA) resulted in p -Value = 0.000. When p -Value < 0.05, H_0 is rejected, meaning that data groups have significant differences. The Tukey method on comparisons between the data group of US/PMMA and three data groups of US/TC (2:1)/PMMA, US/TC (1:1)/PMMA, and US/TC (1:2)/PMMA obtained p -Value < 0.05. It means that the addition of carbon does affect both the tensile and the bending strengths. However, the Tukey method on comparison among the three data groups of US/TC (2:1)/PMMA, US/TC (1:1)/PMMA, and US/TC (1:2)/PMMA showed p -Value > 0.05, indicating that no significant difference between them. These statistical analysis results summarize that the addition of carbon fiber into the sisal/PMMA composite has provided a significant improvement in tensile and flexural properties, but no significant difference due to US/TC ratio.

Table 1. Mean and standard deviation (SD) of tensile and flexural strengths values for the tested specimens

Specimens	Tensile strength (MPa)	Flexural strength (MPa)
	Mean ± SD	Mean ± SD
US/PMMA	38.60 ± 2.69 ^a	63.20 ± 4.88
US/TC (2:1)/PMMA	53.10 ± 6.04 ^b	94.60 ± 8.49
US/TC (1:1)/PMMA	56.00 ± 4.73 ^b	100.00 ± 21.8
US/TC (1:2)/PMMA	57.70 ± 6.15 ^b	108.00 ± 6.61

SD: denoted same letters indicate no statistically significant differences specimens (p -Value>0.05).

Figure 3 demonstrates the tensile strength of US/TC/PMMA hybrid composites with different US/TC ratios. This finding has verified that the addition of TC at all ratios significantly improves the tensile properties of the hybrid composites from 27.30 – 33.10%.

According to these results, the tensile strength of TS/TC/PMMA with TC at 48 hours exposure time to HNO₃ (Fig.2) is lower than that of US/TC/PMMA at US/TC (1:1) (Fig.3), although using a similar sisal/carbon ratio of 1:1. The former had higher tensile strength than the latter due to the higher surface roughness of TS fibers than US fibers, resulting in better interfacial bonding between the TS fibers and PMMA matrix. However, as sisal fiber was alkali-treated, the exposed surface area of cellulose increased¹⁹⁾ because the non-cellulosic components, such as hemicellulose and lignin, dissolved in alkali solution. As a result, the number of hydroxyl groups on the surface of TS fiber increased, making it more hydrophilic than US fiber²⁰⁾. Consequently, TS fibers tended to agglomerate similar to

TC fibers. Therefore, when TS and TC were combined to reinforce the PMMA matrix, they hardly dispersed within the matrix, leading to uneven distribution of the interfacial bonding between the fibers and the matrix. The current study performed the mechanical (tensile and flexural) characterization and thermal properties of US/TC/PMMA composites.

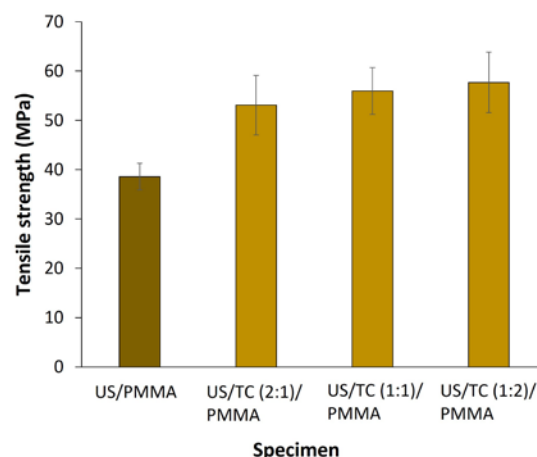


Fig. 3: Tensile strength of US/TC/PMMA hybrid composites with various US/TC ratios.

Figure 4 displays the flexural strength of US/TC/PMMA hybrid composites. The flexural strength increases with the carbon fiber content, following the same pattern as the tensile properties (Fig.3). All composite specimens did not fail, but cracks formed after the bending test (Fig.5). Changes in the mechanical properties are discussed from the morphological perspective in Fig.6 and 7.

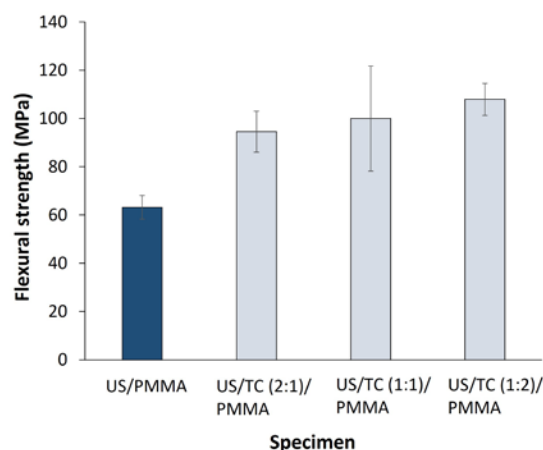


Fig. 4: Flexural properties of US/TC/PMMA hybrid composites with various US/TC ratios.

This study resulted in substantially flexural strength of 63.20 ± 4.88 MPa for US/PMMA at 20% fiber content (Fig.4) higher than a previous study⁶⁾ using 10% fiber

content. The low sisal fiber content of 10% resulted in an inadequate mix with the PMMA matrix. It usually leads to lower stiffness²¹). Moreover, a study investigating sisal/carbon/polyester hybrid composites⁸) resulted in a similar pattern to the current findings; the flexural properties were significantly higher than the tensile properties for both untreated and treated sisal fibers and different sisal/carbon ratios. The matrix material used in this research is different from the previous ones. The use of raw sisal fiber and the sisal/carbon ratio of 1:1 was utilized to compare the tensile strength, which was higher (56.00 ± 4.73 MPa) than in the previous study (38.3 MPa). Inversely, the flexural strength of this study (100.00 ± 21.8 MPa) was lower than the earlier work (131.48 MPa). Apart from the difference in the use of the matrix material, the fiber length is also different. This study used 6 mm fiber length, 4 mm longer than the previous work. It contradicts an earlier finding²²), stating that the mechanical properties of carbon-reinforced composites improved as carbon fiber length increased from 2 mm to 4 mm, then decreased until fiber length reached 6 mm. The very short fiber contributed to weak interfacial interaction with the matrix, and the fibers did not show the significant reinforcing effect²³). On the other hand, random-oriented long fibers made the fiber dispersion worse, resulting in a failure mechanism due to a lack of fibers in some matrix areas²²). Usually, carbon fiber tends to clump together in this case, making it difficult to obtain a high degree of dispersion¹⁵).

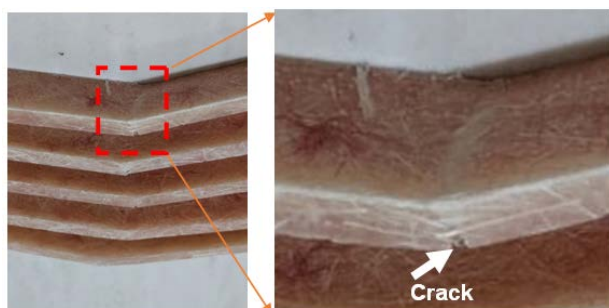


Fig.5: Photograph of bending tested specimens.

Compared to the denture-based materials made of kenaf/PMMA⁷), hibiscus sabdariffa fiber reinforced acrylic resin²⁴), and glass fiber/PMMA²⁵), the highest flexural strength (108.00 ± 6.61 MPa) of US/TC (1:2)/PMMA (Fig.4) was higher than those flexural strengths. It was comparable with nanosized-pomegranate peel's flexural strength (114 MPa) as the natural powder reinforced PMMA composite¹³). Furthermore, the maximum tensile strength (36 MPa) achieved by SiO₂ (25 μm, 12%)/PMMA¹¹) was lower than the highest tensile strength of US/TC (1:2)/PMMA (57.70 ± 6.15 MPa). However, the maximum flexural strength of the present composite (108.00 ± 6.6 MPa) was much lower than that of SiO₂/PMMA (355.19 MPa). Besides, PMMA reinforced with 1%, 3%, and 5%

ceramic nanoparticles (ZrO₂, TiO₂, and Al₂O₃)²⁶) showed the comparable flexural strength with this US/TC(1:2)/PMMA composite, especially by 3% and 5% reinforcements.

It may be due to a higher degree of particle fillers dispersion than randomly oriented fiber dispersion in the matrix. This statement suggests that the particle reinforced matrix has a better strengthening mechanism than the fibers reinforced matrix. Two critical factors influence the fiber reinforcement: fiber length and orientation, though the fiber orientation has a more substantial effect than the fiber length²⁷).

3.3 Morphological characterization

Figure 6 exhibits SEM images of morphological tensile fracture surfaces of the composite specimens. The distribution of mixed US and TC fibers appeared with reducing US/TC ratios. Fiber-pull out and debonding are observed in the US/PMMA composite (Fig.6a), although their existence is considerably low, resulting in the lowest mechanical properties among the specimens. Since the composite manufacturing did not operate in a vacuum, water molecules are trapped during the process, resulting in micro-voids in all composite specimens. The hybridized US and TC fibers also demonstrate good interfacial bonding between the fibers and the matrix. Besides, Fig. 6a, Fig.6b, and Fig.6c illustrate the disparity in the sisal/carbon ratio, as well as the actual proportions of 1:2, 1:1, and 2:1, respectively. The tensile

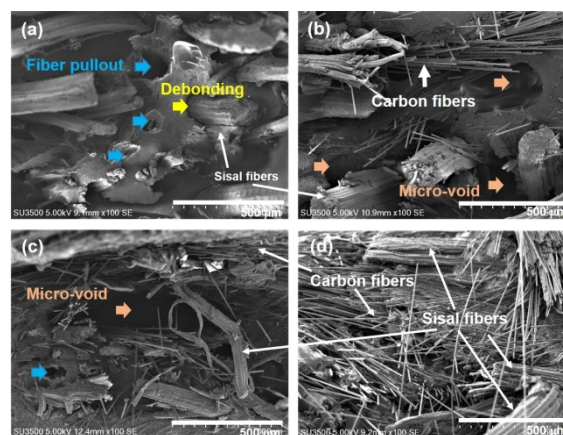


Fig.6: SEM images of the tensile fracture surface of US/TC/PMMA composites without TC (a) and with US/TC ratios of 2:1 (b), 1:1 (c), and 1:2 (d).

and bending strength increased with carbon fiber content, consistent with the previous findings⁸). Carbon fibers did not seem uniformly dispersed as individual fibers and tended to form clusters. It is one of the problems with manufacturing composites using carbon fibers.

The surface morphology of the fractured bending specimens could not be characterized comprehensively in the current work because the composites specimens did not fail. Thus, the composites were fractured

manually on the cracks to observe the fiber dispersion in the matrix, as displayed in the optical micrographs (Fig.7). In all composite specimens, the degree of fiber dispersion in the matrix is considerably homogenous. The fibers are primarily unbroken, suggesting that the fibers can withstand the bending load well. The morphology of tensile and bending fracture surfaces shown in Fig.6 and Fig.7, respectively, indicates a good relationship between these morphologies and the mechanical test result.

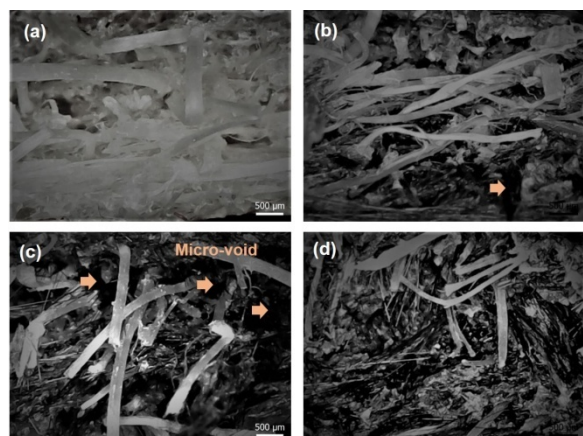


Fig. 7: Optical micrographs of the bending failure surface of US/TC/PMMA composites without TC (a) and with US/TC ratios of 2:1 (b), 1:1 (c), and 1:2 (d).

3.4 Thermal analysis

Figure 8 displays TGA curves of US fiber and the US/TC/PMMA composites obtained from 30°C to 300°C. The US fiber curve illustrates one stage of degradation, having a transition temperature from 73.27°C to 212.37°C (Table 2). The initial weight loss below 100°C occurred due to the evaporation of water molecules from the sisal fiber or the fiber losses moisture²⁸⁻³⁰. Further degradation to the final transition temperature at 286.25°C was caused by the decomposition of cellulose and hemicellulose with a weight loss of 11.09%^{31, 32}. The current untreated sisal TGA curve pattern is similar to that stated in the previous outcome³¹.

TGA curves of the US/TC/PMMA composites are different from the US fiber, with the maximum peaks (T_m) of the composites lower than T_m of the US fiber (Table 2). Hybridization of the US and TC fibers embedded in the PMMA reduced weight loss by around 20%. The composite containing the highest volume fraction of carbon fiber has produced the lowest weight loss (7.45%) (Table 2). PMMA as the matrix material appeared to protect the sisal fiber from degradation in these composites. The interaction between US and TC fibers and PMMA inhibited the degradation process, leading to the composite's more robust thermal property than the US fiber. Based on the TGA thermograms in the previous studies^{28, 30, 33}, the thermal stability of the composite reinforced with the alkali-treated fiber was

higher than that of the hybrid composite reinforced with raw fiber and carbon fiber. The non-cellulosic constituents contained in the natural fibers caused thermal degradation.

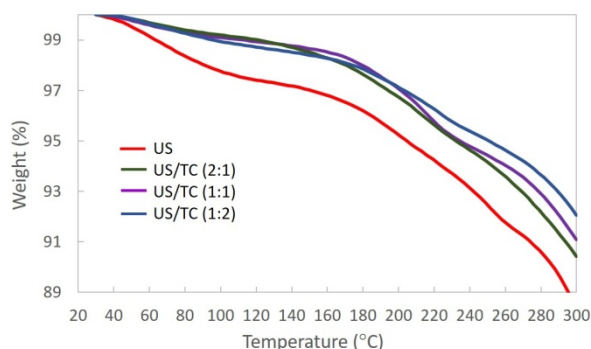


Fig. 8: TGA thermograms of US fiber and US/carbon/PMMA composites.

Table 2. Average mass loss (%) of the composites.

Sample	T_i (°C)	T_m (°C)	T_f (°C)	W_{Ti} (%)	W_{Tm} (%)	W_{Tf} (%)	Last Residue (%)
US	73,27	212,37	286,25	2,6	7,93	11,09	88,56
US/K (2:1)	106,23	198,29	267,64	1,66	4,57	8,79	90,99
US/K (1:1)	75,16	199,59	277,74	1,3	4,52	8,24	91,24
US/K (1:2)	78,76	204,45	277,54	1,47	4,59	7,45	92,41

3.5 Denture-based material application

Based on the component materials of the present composites (sisal fiber, carbon fiber, PMMA) and the flexural strength achievement, the US/TC/PMMA hybrid composites are potentially used for the denture-based material. For that application, the present result has added the compressive strength of the composite obtained by a simulation result using the method described above.

The US/TC (1:2)/PMMA hybrid composite showed the highest flexural strength selected for this simulation. The composite material is applied as a denture in maxillary lateral incisors that are vulnerable to injure³⁴. In this simulation, the force was applied by gradually increasing until reaching the composite's yield strength (101.78 MPa). The maximum force load used was 91 N. Applying the force load higher than 91 N will exceed the yield strength. The simulation results showing the stress distribution in various views (Fig. 9) indicated the maximum stress of 101.00 MPa, known as the compressive strength of US/TC (1:2)/PMMA hybrid composite.

The compressive strength of other denture-based materials evaluated experimentally resulted in various values. For examples, the compressive strength of eggshell (3%)/acrylic composite of 75.46 ± 13.9 MPa³⁵ and that of 2-5% (TiO₂-ZnO)/PMMA nanocomposites of 118.80 MPa– 85.70 MPa³⁶ can be used for comparison

with the present result, indicating the comparable values.

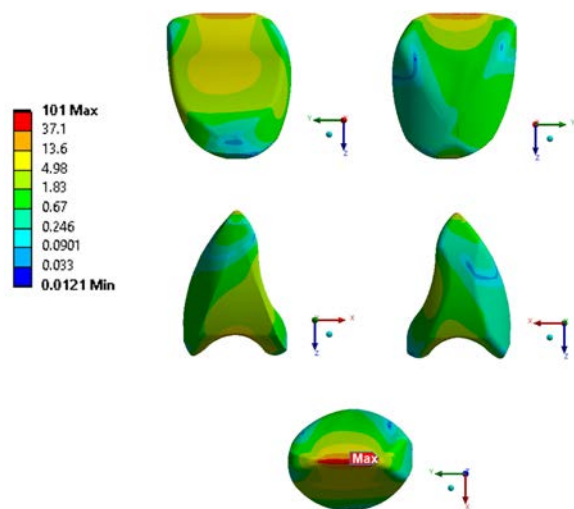


Fig. 9. The simulation results showing the stress distribution in various views.

4. Conclusions

The addition of surface-treated carbon (TC) fiber in untreated sisal (US)/PMMA composite has significantly improved tensile and flexural properties, but not due to the differences in US/TC ratios. The tensile and flexural strengths were enhanced by 33.10% and 41.48%, respectively. The thermal analysis indicated a weight loss of 11.09% in the composite without carbon fiber and decreased to 7.45% in the composite with carbon fiber.

These research findings suggested that the US/TC (1:2)/PMMA hybrid composite's flexural strength of 108.00 ± 6.61 MPa and compressive strength of 101.00 MPa are included in the range of the properties of the denture-based materials, leading to being valuable information in the development of denture materials.

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