

The use of starch matrix-banana fiber composites for biodegradable maxillofacial bone plates

Lamis R. Darwish

Department of Biomedical Engineering,
Faculty of Engineering, Helwan University
Cairo, Egypt.

Mohamed Tarek El-Wakad

Department of Biomedical Engineering,
Faculty of Engineering, Helwan University
Cairo, Egypt.

Mahmoud Farag

Department of Mechanical Engineering
The American University in Cairo
Cairo, Egypt

Mohamed Emara

Department of manufacturing Engineering,
CIC- Canadian International
Cairo, Egypt

Received: July 25, 2019. Revised: August 16, 2021. Accepted: September 23, 2021. Published: November 27, 2021.

Abstract— Starch based green composites have been studied as potential materials to be used in several biomedical applications. This paper explores utilizing starch based composites reinforced with pseudostem banana fibers in fabricating biodegradable maxillofacial bone plates. Corn starch plasticized by 30 wt.% glycerin and 20 wt.% distilled water was used as a matrix. The produced thermoplastic starch (TPS) matrix is reinforced with pseudostem banana fibers at different weight fractions using hot pressing at 5 MPa and 160°C for 30 minutes. Our experimental results showed that increasing the banana fibers weight fraction progressively improved the mechanical properties reaching a maximum at 50 wt.% fibers. The improvement in the mechanical properties of starch/banana fibers composite was attributable to the strong interaction between fibers and the starch matrix, as evidenced by a series of scanning electron micrographs of the fracture surface. Furthermore, experiments investigating thermal properties and water uptake also showed that the best results are achieved at the 50 wt.% banana fibers. The experimental results show that the starch matrix-banana fiber composites satisfy the maxillofacial bone fixation requirements.

Keywords- Biodegradable composites; Thermoplastic cornstarch; Banana fibers; maxillofacial bone plates.

Metallic implants made of titanium alloys are widely used for internal fixation of bone fractures of the human skeleton. These implants are regarded as the gold standard for the majority of the fracture fixation treatments due to their outstanding mechanical properties [1-3]. However, metallic implants have several significant drawbacks such as stress shielding effect which leads to bone resorption, implant loosening and, consequently, the need for a second operation to remove the implants especially in pediatrics. This second operation has several risks such as infection, removal problems of jammed implants, implants migration and associated extra health care costs [2-4]. Consequently, in order to eliminate stress shielding it has been investigated for many years to use a material that would degrade and gradually lose its strength at the same rate of bone healing.

Starch based green biomaterials have recently been proposed as having a great potential for several applications in the biomedical field as they are totally biodegradable, biocompatible, and inexpensive [5-7]. These materials were

shown to have a great potential to be used in the bone related therapy applications [8-10], ranging from bone tissue engineering scaffolds [11], to bone cements [12].

Starch is the major polysaccharide constituent of photosynthetic tissues and of many storage organs in plants [13]. Worldwide corn represents the major commercial source of starch. It is a semicrystalline polymer composed of a mixture of amylose, a linear polysaccharide and amylopectin, a highly branched polysaccharide [4]. However, before being thermally processable as for thermoplastic polymers, starch must be converted to thermoplastic starch (TPS) by the addition of specific plasticizers combined with the application of heat and shear forces [14]. A plasticizer is required for preparing TPS because the plasticizer can reduce brittleness and avoid the cracking of starch based materials during handling and storage [15]. Various plasticizers have been used for TPS such as formamide, ethanolamine, sorbitol, sugars, and glycerol [16].

However, TPS has disadvantages when compared to synthetic biodegradable polymers, such as it has a strong ability to absorb water, and has poor mechanical and thermal properties [15]. The reinforcement of the TPS matrix with natural lignocellulosic fibers seems to be the logical alternative in order to increase their mechanical performance and to preserve the green character of the final material [17].

Consequently, this work proposes (investigates) using glycerol plasticized starch matrix reinforced with pseudostem banana fibers (BFs) at different weight fractions in the fabrication of maxillofacial bone plates. Pseudostem banana fiber is a hard fiber obtained from the leaf sheaths. It is considered to be the strongest commercially available hemicelluloses fiber due to its high cellulose content [18-20]. Extensive experiments have been conducted to investigate the resulting plates.

I. MATERIALS AND METHODS

A. Materials

Regular native cornstarch (28% amylose) with 11% moisture and reagent grade glycerin (99.7% purity) were used as received. Banana pseudostem was received from a local

supplier. The pseudostem was chopped manually into 30 mm length pieces. The raw banana fiber was obtained by mechanical separation of the fiber from these chopped pieces. The extracted banana fiber was treated with 0.5% NaOH solution at 90 °C for 30 min, and then washed with running tap water until the PH value reached 7. The alkali treated fiber was then oven dried.

B. Sample preparation

Corn starch was plastisized by 30 wt.% glycerin and 20 wt.% distilled water. More details about the plasticization technique can be found elsewhere [21]. The resulting thermoplastic starch matrix was emulsified with distilled water then reinforced with alkali treated banana fibers at 40%, 50% and 60% weight fractions using hot pressing at 5 MPa and 160 °C for 30 minutes.

C. Morphological characterization

Morphological characteristics of the tensile fracture surface of TPS/banana fiber composites were examined using ZEISS Scanning Electron Microscope (SEM). Images were captured in a liquid nitrogen cooled atmosphere.

D. Mechanical tests

Composite samples (specimen) were cut from the compression molded sheets considering the dimensions ratio mentioned in ASTM D5083-10. Tensile and three point flexural tests were performed using a universal testing machine (Instron 3382) with a cross head speed of 5 mm/min. Tests were performed at room temperature. Ten specimens were tested for each fiber weight fraction.

E. Water absorption measurements

Composite samples were oven dried at 100 °C for 4 hours. These samples were weighted immediately after their removal from the oven. Then they were stored for 7 days in desiccators set up at 100% RH using distilled water. The increase in weight was measured on daily basis. The water absorption can be calculated using the following equation:

$$\text{Water absorption (\%)} = [(w_f - w_i) / w_i] * 100 \quad (1)$$

where w_f is the final sample weight and w_i is the initial sample weight. All measurements were performed in duplicate.

F. Thermal properties

Thermogravimetric analysis (TGA) was performed using a TA Q500 TGA analyzer (TA Instruments, New Castle, DE, USA). The samples were heated from room temperature to 700 °C at a heating rate of 20 °C/min using synthetic air flow of 60 mL/min. The derivative of TGA curves (DTG) was obtained using TA analysis software.

II. RESULTS AND DISCUSSION

A. Morphological characteristics

Fig. 1 shows the scanning electron micrographs of the tensile fracture surfaces of TPS/BF composites at 40%, 50%, 60% BF wt.%. The good interfacial adhesion between TPS matrix and treated BF can be clearly seen in fig. 1 (a-d). This good adhesion was evident by the fracture of the BF at the surface, the decreased number of fibers pull outs and holes, and the absence of gap between the BF and the TPS matrix. This good adhesion is mainly due to the similarity in polarity between the BF and the TPS.

On the contrary, increasing the BF wt.% up to 60% caused deterioration in the wettability of the BFs with emulsified TPS matrix. This deterioration is due to the agglomeration of the BFs. This was evident by the increased number of pulled out fibers and holes (fig. 1 (e)), also, the surface cleanliness of the pulled out fibers (fig. 1 (f)).

B. Mechanical properties

The ultimate tensile strength (UTS) and young's modulus (YM) for the TPS/BF composites with different BF content are shown in fig. 2. Both of the UTS and YM increase linearly with increasing the BF content up to 50%. The UTS and YM were 30 MPa and 4.6 GPa, respectively, in the samples with 50% BF. The only study in the literature used pseudostem banana fibers as reinforcement for glycerol plasticized cornstarch was by Guimarães et al. [22]. The maximum banana fibers wt.% reported in this study was 35% and it gave 3.56 MPa and 74.35 MPa for UTS and YM, respectively.

Besides the flexural strength and modulus for the same fiber content patterns followed the same behavior of UTS and YM. The flexural strength and modulus are shown in fig. 3. The maximum flexural strength and modulus were 80 MPa and 7.5 GPa, respectively, in the samples with 50% BF.

Further addition of BF causes deterioration in the mechanical properties of the composite due to the agglomeration of the BF. This agglomeration decreases the wettability of the fiber with the emulsified TPS matrix, thus decreases the attachment between the fibers and the matrix. These results are in agreement with the SEM fracture surface investigation.

C. Water absorption

Water absorption is an important factor for the application of the TPS composites in the biomedical field. The results of water absorption for the TPS/BF composites with different BF wt.% are shown in fig. 4 . It can be clearly noticed from the figures that increasing the BF wt.% decreases the water absorption of the TPS/BF composites. These results are ascribed to the fact that starch is more hydrophilic than cellulose, thus the presence of fibers reduce the hydrophilicity of the starch based composites. The enhancement in the water absorption property of TPS/BF composites and its cause conforms to the results documented in previous studies [22-25].

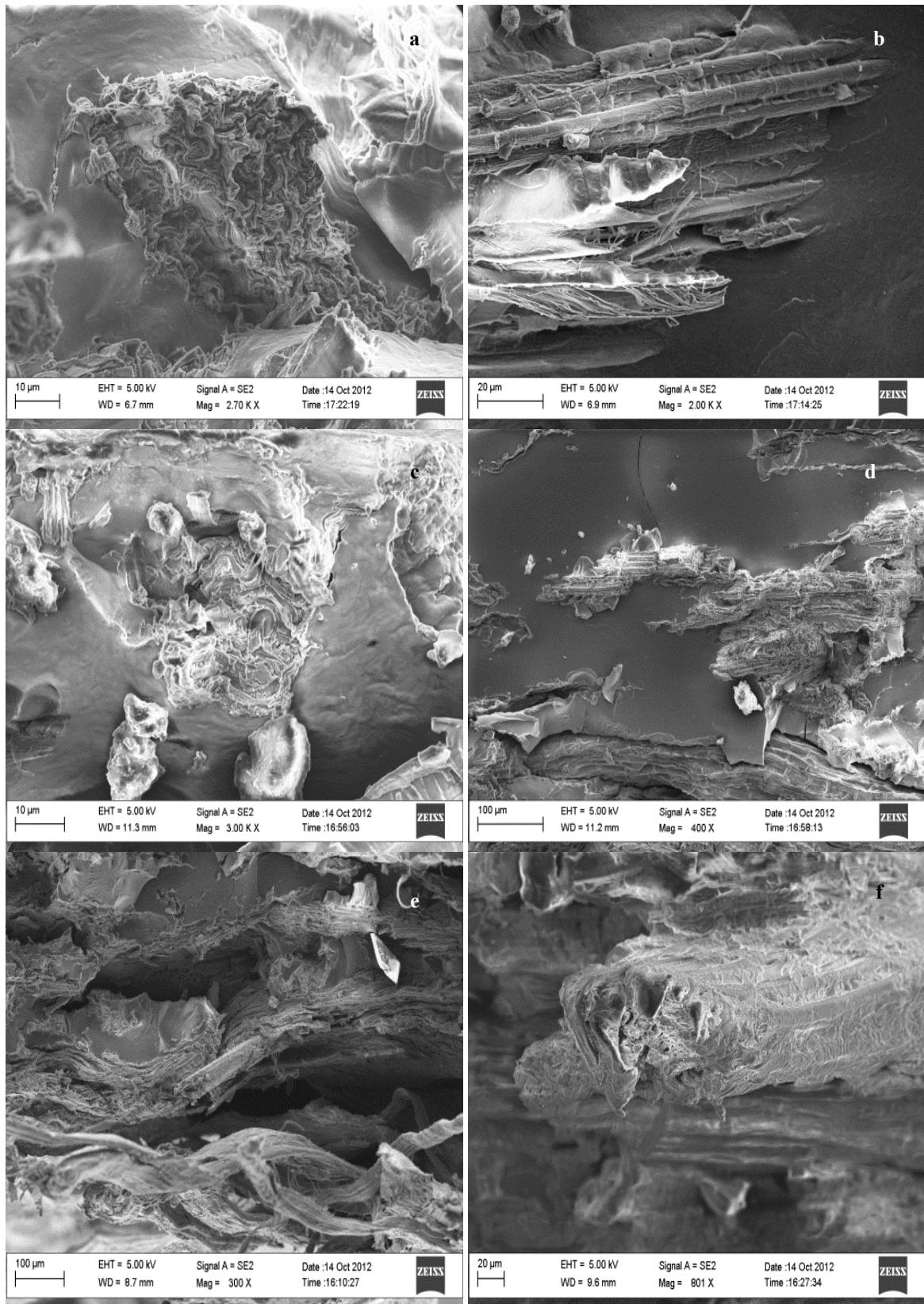


Fig.1. SEM micrographs of the tensile fracture surfaces of the (a,b) 40% BF, (c,d) 50% BF, and (e,f) 60% BF composite

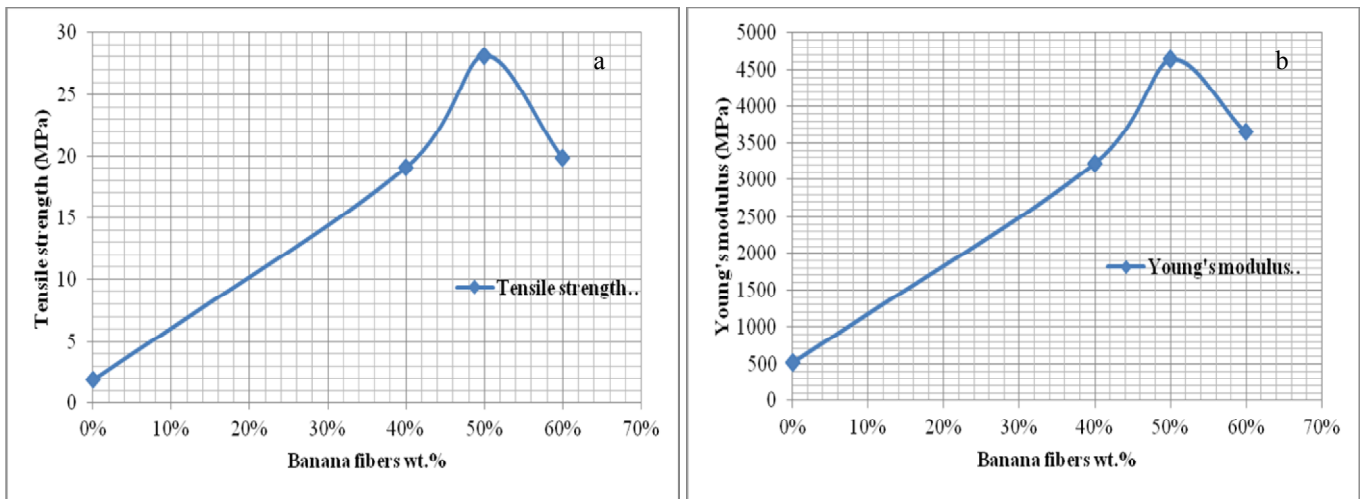


Fig. 2. Tensile properties of the TPS/BFs composites at different BFs wt.% (a) tensile strength (MPa) and (b) Young's modulus (MPa)

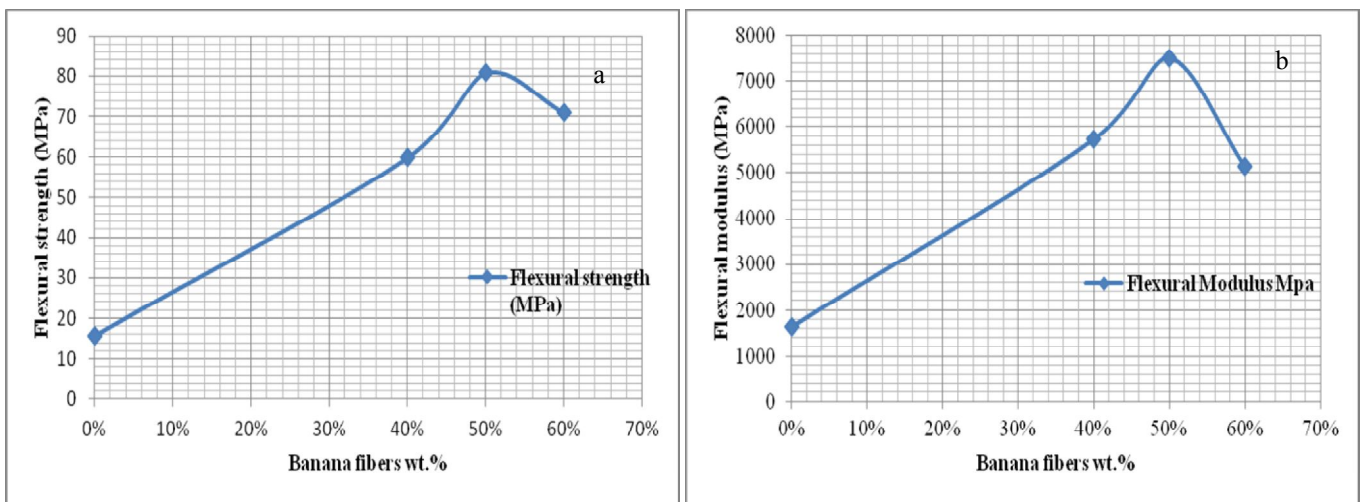


Fig. 3. Flexural properties of the TPS/BFs composites at different BFs wt.% (a) flexural strength (MPa) and (b) flexural modulus (MPa)

D. Thermogravimetric analysis

Fig. 5 shows the TGA curves and their corresponding derivative (DTG) curves of the 40%, 50% 60% BF composites. The first weight loss that occurred just above the room temperature till 100 °C is related to the evaporation of water. It appeared as a small peak around 60 °C and 100 °C in the DTG curves of 40% and 50% BFs composite. This peak disappeared in the higher BFs wt.% (60%), which indicates that less water was absorbed by the composite. Wattanakornsiri et al [26] reported the same peak disappearance in the high fibers content. The weight loss in

the temperature range of 150 °C to 200 °C is related to the evaporation of glycerin. This glycerin evaporation appeared in the DTG curves of the TPS and the TPS/BF composites as a small peak right before 200 °C. These peaks were also observed by other authors [24-27]. Peaks group (I) observed in the DTG curves of the TPS/BF composites at the temperature range of 325 °C to 335 °C are attributed to the thermal degradation of starch. The weight loss in the temperature range of 365 °C to 377 °C, which appeared in the DTG curves as peaks group (II) are related to the degradation of the treated BFs. Starch and BFs decomposition peaks were observed by other authors [24].

It can be noticed from the TGA curves and table I that the onset degradation temperature of starch increased with increasing the BFs wt.%. Prachayawarakorn et al. [28-29] reported the same observation and assigned it to the possible formation of hydrogen bond linkage between TPS and cellulosic fibers.

Furthermore, The weight loss of the 40% ,50% and 60% BFs composites at the peaks group (I) temperatures, which are related to the decomposition of starch, are 41%, 38.7% and 38.3%, respectively. It can be noticed that the weight loss decreased by increasing the BFs content.

Thus, The decrease in the weight loss as well as the increase in the onset degradation temperature of starch indicate that the thermal stability of the TPS/BFs composites improves with increasing the BFs wt.%. Similar observations were also reported by other research groups [24-27]. They attributed this improvement in the thermal stability of TPS by increasing the fibers content to the higher thermal stability of cellulosic fibers compared to starch and the good compatibility of both polysaccharides.

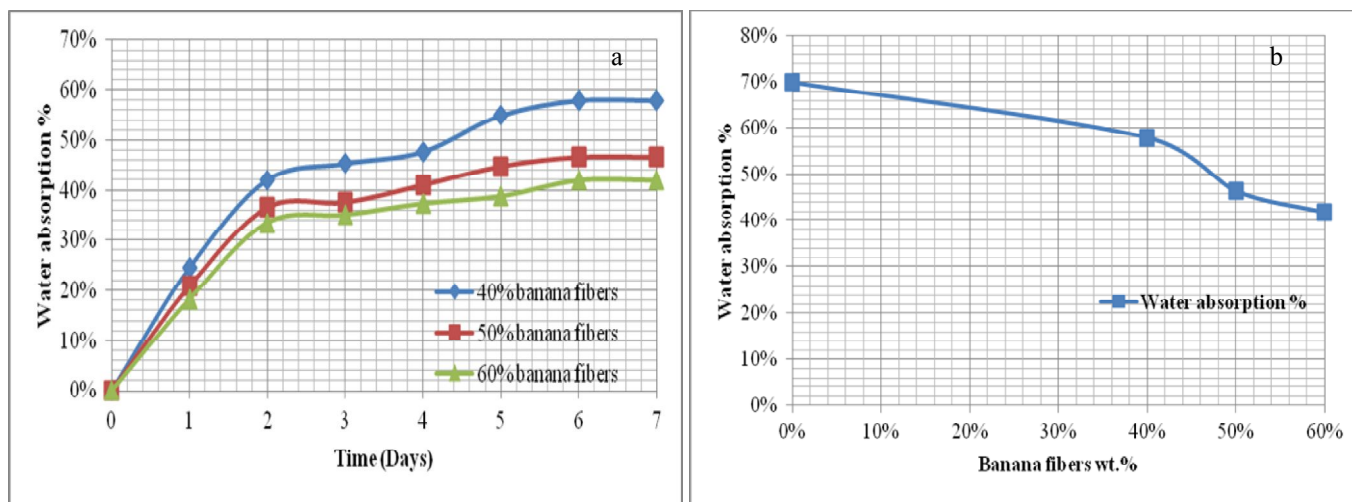


Fig. 4. Effect of BFs wt.% on (a) water absorption of the composites over a period of 7 days and (b) water absorption saturation level of the composites after 7 days

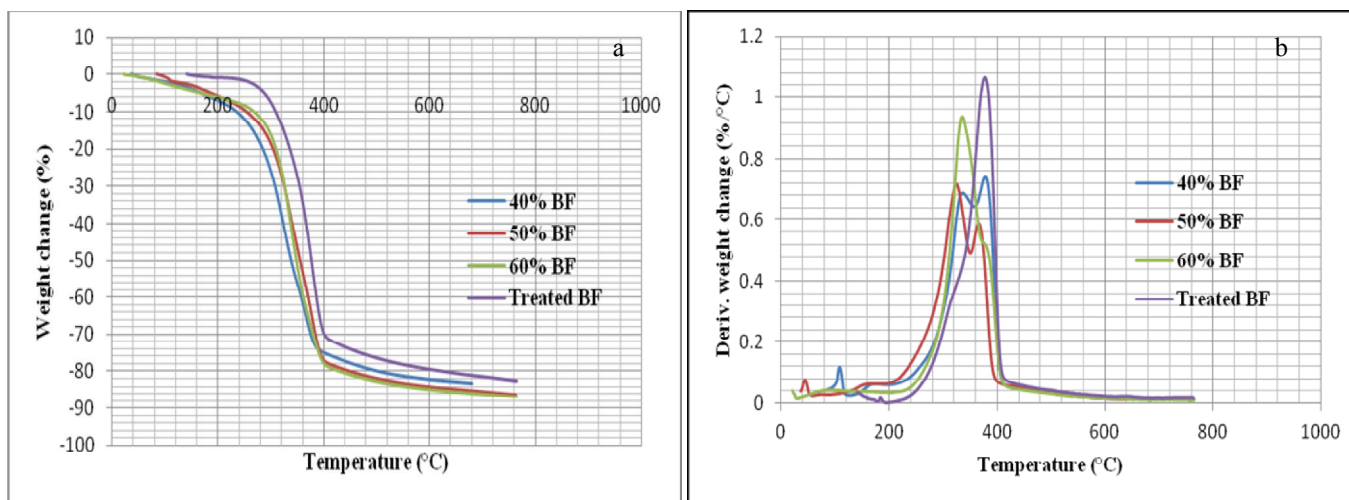


Fig. 5. (a) TGA and (b) DTG of TPS/BFs composites with different BFs wt.%

III. CONCLUSION

This paper addressed the developing of green cornstarch composites for maxillofacial bone plates' fabrication. Glycerol plasticized starch was reinforced with pseudostem banana fibers at different weight fractions. Extensive experiments had been conducted in order to investigate the morphological, thermal and mechanical properties for the proposed composite. The optimum mechanical properties were obtained at 50 wt.% BFs. Furthermore, Incorporating BFs into the TPS matrix improved the thermal properties of the composite. Thus the mechanical and thermal properties of this composite nominate it to be used in the fabrication of maxillofacial bone plates. However the water resistance property was the major drawback of the fabricated composite. Future work may explore blending biodegradable synthetic polymer with the TPS matrix in order to overcome the water resistance drawback.

TABLE I. Onset and peak temperatures of TPS/BFs composites

BF wt.%	T _{onset} (°C)	Temperature region		
		>350 °C		<350 °C
		Peak (°C)	Weight loss (%)	Peak (°C)
40% BF	281.25	325	41	365.9
50% BF	306.25	333.8	38.75	377
60% BF	309.375	334.9	38.33	377.5

REFERENCES

- [1] Leenslag, J. W., Pennings, A. J., Bos, R. R., Rozema, F. R., & Boering, G. "Resorbable materials of poly (L-lactide). VI. Plates and screws for internal fracture fixation". *Biomaterials*, vol. 8(1), 70-73. J. Clerk Maxwell, A Treatise on Electricity and Magnetism, 3rd ed., vol. 2. Oxford: Clarendon, 1892, pp. 68-73, 1987.
- [2] Hu, Q., Li, B., Wang, M., & Shen, J., "Preparation and characterization of biodegradable chitosan/hydroxyapatite nanocomposite rods via in situ hybridization: a potential material as internal fixation of bone fracture". *Biomaterials*, vol. 25(5), pp. 779-785, 2004.
- [3] Eglin, D., & Alini, M., "Degradable polymeric materials for osteosynthesis: tutorial". *Eur Cell Mater*, vol. 16, pp. 80-91, 2008.
- [4] Zargar Kharazi, A., Fathi, M. H., Bahmani, F., & Fanian, H. (2011). Partially resorbable composite bone plate with controlled degradation rate, desired mechanical properties and bioactivity. *Polymer Degradation and Stability*, 96(12), 2055-2063.
- [5] Alberta Araújo, M., Cunha, A. M., & Mota, M. (2004). Enzymatic degradation of starch-based thermoplastic compounds used in protheses: identification of the degradation products in solution. *Biomaterials*, 25(13), 2687-2693.
- [6] Marques, A. P., Reis, R. L., & Hunt, J. A. (2002). The biocompatibility of novel starch-based polymers and composites: in vitro studies. *Biomaterials*, 23(6), 1471-1478.
- [7] Lu, D. R., Xiao, C. M., & Xu, S. J. (2009). Starch-based completely biodegradable polymer materials. *Express Polym Lett*, 3(6), 366-75.
- [8] Azevedo, H. S., Gama, F. M., & Reis, R. L., 2003, "In vitro assessment of the enzymatic degradation of several starch based biomaterials", *Biomacromolecules*, 4(6), pp. 1703-1712.
- [9] Duarte, A. R. C., Mano, J. F., & Reis, R. L., 2010, "Enzymatic degradation of 3D scaffolds of starchpoly-(ϵ -caprolactone) prepared by supercritical fluid technology", *Polymer Degradation and Stability*, 95(10), pp. 2110-2117.
- [10] Mano, J. F., Koniarova, D., & Reis, R. L., 2003, "Thermal properties of thermoplastic starch/synthetic polymer blends with potential biomedical applicability", *Journal of Materials Science: Materials in Medicine*, 14(2), pp. 127-135.
- [11] Gonçalves, A., Costa, P., Rodrigues, M. T., Dias, I. R., Reis, R. L., & Gomes, M. E., 2011, "Effect of flow perfusion conditions in the chondrogenic differentiation of bone marrow stromal cells cultured onto starch based biodegradable scaffolds", *Acta Biomaterialia*, 7(4), pp. 1644-1652.
- [12] Boesel, L. F., & Reis, R. L., 2008, "A review on the polymer properties of Hydrophilic, partially Degradable and Bioactive acrylic Cements (HDBC)", *Progress in Polymer Science*, 33(2), pp. 180-190.
- [13] Alves, N. M., Saiz-Arroyo, C., Rodriguez-Perez, M. A., Reis, R. L., & Mano, J. F. (2007). Microhardness of starch based biomaterials in simulated physiological conditions. *Acta Biomaterialia*, 3(1), 69-76.
- [14] Wattanakornsiri, A., Pachana, K., Kaewpirom, S., Sawangwong, P., & Migliaresi, C. (2011). Green composites of thermoplastic corn starch and recycled paper cellulose fibers. *Sonklanakarin Journal of Science and Technology*, 33(4), 416.
- [15] Kaewtatip, K., & Thongmee, J. (2012). Studies on the structure and properties of thermoplastic starch/luffa fiber composites. *Materials & Design*, 40, 314-318.
- [16] Hulleman, S. H., Janssen, F. H., & Feil, H. (1998). The role of water during plasticization of native starches. *Polymer*, 39(10), 2043-2048.
- [17] Girones, J., Lopez, J. P., Mutje, P., Carvalho, A. J. F., Curvelo, A. A. S., & Vilaseca, F. (2012). Natural fiber-reinforced thermoplastic starch composites obtained by melt processing. *Composites Science and Technology*, 72(7), 858-863.
- [18] Benítez, A. N., Monzón, M. D., Angulo, I., Ortega, Z., Hernández, P. M., & Marrero, M. D., 2012, "Treatment of Banana Fiber for Use in the Reinforcement of Polymeric Matrices", *Measurement*, 46(3), pp. 1065-1073.
- [19] Chaudhary, A. K., Gope, P. C., & Singh, V. K., 2011, "Studies on Fracture Performance of Bio-fiber-Silica-glass Fiber Reinforced Epoxy Hybrid Composites" In *Experimental and Applied Mechanics*, T. Proulx ed., Springer, New York, Vol. 6, pp. 363-368.
- [20] Mohanty, Amar K., Manjusri Misra, and Lawrence Thaddeus Drza, 2005, *Natural fibers, biopolymers, and biocomposites*, CRC Press, Boca Raton, Florida, pp. 90-110, Chap. 2.
- [21] Elsayed, H., Farag, M., Megahed, H., Mehanny, S., 2012, "Influence of flax fibers on properties of starch based composites", *IMECE2012-89628*, Proc. of ASME 2012 international conference of mechanical engineering, Huston, Texas, USA.
- [22] Guimarães, J. L., Wypych, F., Saul, C. K., Ramos, L. P., & Satyanarayana, K. G. (2010). Studies of the processing and characterization of corn starch and its composites with banana and sugarcane fibers from Brazil. *Carbohydrate Polymers*, 80(1), 130-138.
- [23] Curvelo, A. A. S., De Carvalho, A. J. F., & Agnelli, J. A. M. (2001). Thermoplastic starch-cellulosic fibers composites: preliminary results. *Carbohydrate Polymers*, 45(2), 183-188.
- [24] Prachayawarakorn, J., Chaiwatyothin, S., Mueangta, S., & Hanchana, A. (2012). Effect of jute and kapok fibers on properties of thermoplastic cassava starch composites. *Materials & Design*.
- [25] Prachayawarakorn, J., Sangnithidej, P., & Boonpasith, P. (2010). Properties of thermoplastic rice starch composites reinforced by cotton fiber or low-density polyethylene. *Carbohydrate Polymers*, 81(2), 425-433.
- [26] Wattanakornsiri, A., Pachana, K., Kaewpirom, S., Sawangwong, P., & Migliaresi, C. (2011). Green composites of thermoplastic corn starch and recycled paper cellulose fibers. *Sonklanakarin Journal of Science and Technology*, 33(4), 416.
- [27] Kaewtatip, K., & Thongmee, J. (2012). Studies on the structure and properties of thermoplastic starch/luffa fiber composites. *Materials & Design*, 40, 314-318.
- [28] Prachayawarakorn, J., Limsiriwong, N., Kongjindamunee, R., & Surakit, S. (2012). Effect of agar and cotton fiber on properties of thermoplastic waxy rice starch composites. *Journal of Polymers and the Environment*, 20(1), 88-95.
- [29] Prachayawarakorn, J., Ruttanabus, P., & Boonsom, P. (2011). Effect of cotton fiber contents and lengths on properties of thermoplastic starch composites prepared from rice and waxy rice starches. *Journal of Polymers and the Environment*, 19(1), 274-282.

Creative Commons Attribution License 4.0
(Attribution 4.0 International, CC BY 4.0)

This article is published under the terms of the Creative Commons Attribution License 4.0
https://creativecommons.org/licenses/by/4.0/deed.en_US