Jurnal Teknologi

Full Paper

EVALUATION ON TENSILE AND MORPHOLOGICAL PROPERTIES OF CHITOSAN IN PBAT BIO-COMPOSITE

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Article history
Received
24 July 2024
Received in revised form
16 January 2025
Accepted
26 January 2025
Published Online
22 August 2025

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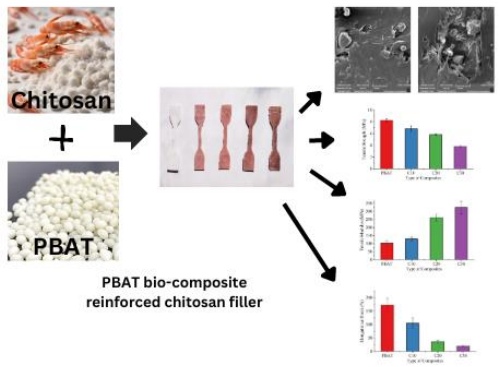
Graphical abstract



Abstract

This study explores the processibility, tensile properties and morphological analysis of biodegradable composite based on Poly (butylene adipate-co-terephthalate) (PBAT) as polymer matrix and chitosan as filler at different concentration, 0, 10, 20 and 30 wt.%. The composite sample with 30 wt.% (338 MPa) chitosan led to a higher increase of the tensile modulus of PBAT compared to the 10 wt.% (135 MPa) and 20 wt.% (275 MPa). Chitosan filler caused a drastic reduction of the tensile strength, 8.4 MPa to 3.8 MPa as the chitosan content increases from 0 wt.% to 30 wt.%. In addition, the morphological analysis suggests that the chitosan was evenly distributed in the PBAT matrix, even though it was agglomerated within the matrix. Furthermore, SEM analysis revealed a weak adherence between the matrix and the filler. The results demonstrate that the current study confirms the idea that chitosan can be effectively used as filler material to enhance the environmentally friendly qualities of polymers.

Keywords: Green polymer, bio-composite, biodegradable, chitosan

Abstrak

Kajian ini mengkaji pemprosesan, sifat tegangan dan analisis morfologi bio-komposit berdasarkan Poli (butelena adipat-ko-tereftalat) (PBAT) sebagai matriks polimer dan kitosan sebagai pengisi dengan kandungan peratusan yang berbeza; 0, 10, 20 dan 30 wt.%. Sampel menggunakan 30% (338 MPa) menunjukkan peningkatan modul tegangan yang lebih tinggi berbanding dengan 10 wt.% (135 MPa) dan 20 wt.% (275 MPa). Pengisi kitosan menyebabkan penurunan drastik dalam kekuatan tegangan, iaitu 8.4 MPa kepada 3.8 MPa apabila kandungan chitosan meningkat daripada 0 wt.% kepada 30 wt.%. Di samping itu, kajian morfologi menunjukkan bahawa kitosan bergumpal dalam matriks, walaupun disebarkan secara serata di dalam matriks PBAT. Tambahan pula, melalui analisa SEM, ikatan diantara kitosan dan PBAT adalah lemah antara matriks dan pengisi. Hasil kajian menunjukkan bahawa kitosan boleh digunakan secara berkesan sebagai bahan pengisi untuk meningkatkan kualiti mesra alam polimer.

Kata kunci: Kelestarian polimer, bio-komposit, biodegradasi, kitosan

1.0 INTRODUCTION

The popularization of polymer products usage was increasing year by year where the polymer or plastic really benefited in human daily life. The environment has been seriously impacted by the significant increase in polymer particles brought on by unsustainable polymer. This has resulted in harm to the ecological system and poses serious hazards to human health. Due to the significant damage it inflicts, this has become a prominent worldwide issue [1-3]. Therefore, to coup with this threat, there are numerous new polymers that have been researched and reported from renewable resources where it able to fulfill the demands of biodegradable and biocompatibility polymer materials. Moreover, the biodegradable polymer is very suitable to be utilized for industrial applications, packaging, and structural applications [4]. Polylactic acid (PLA), polyhydroxyalkanoates (PHA) and polycaprolactone (PCL) are examples of the biodegradable polymer [5] that are commonly used for biodegradable matrices for bio-composites. Polybutylene adipate co-terephthalate (PBAT) is one of the polymers that is synthetic and fully degradable polymer from a fossil resource which derived from petroleum-based products [6]. PBAT also contains special properties such are biodegradable due to aliphatic unit, have an excellent mechanical property where it is more flexible compared to biodegradable composites such are PLA and polybutylene-co-succinate (PBS). Due to its properties, PBAT has a potential to be a substitute for high-density polyester (HDPE) where it commonly suitable for packaging applications [7], and much films for agricultural applications [8]. However, PBAT has a low modulus [9] and rigidity which hinders its practicality implementation for packaging. There are many attempts of research and report to enhance the mechanical properties of these materials and at the same time maintaining their excellent properties and biodegradability. Combination of PLA with others biodegradable polymer such as PBAT has resulted that the matrix is able to produce a biodegradable polymer which exhibits with an enhanced modulus and rigidity [10, 11]. Next, the issue is enhancement of modulus of the polymer composites by reinforced it with natural fibers. Natural fibers are prone to be more famous due to their mechanical properties, lightweight, nontoxic properties and effectively low in cost [12, 13].

Chitosan is derived from chitin that is composed from glucosamine and N-acetyl glucosamine [14]. Moreover, chitosan is one of the natural biopolymers that have a modifiable structure and abundant functional group make it that chitosan able to be process in many sizes and shapes [15]. Chitosan can be procured from the exoskeleton of animals such are crabs, eggshell, shells, and cells wall of fungi [14, 16, 17]. In addition, chitosan has an antibacterial behavior, decent mechanical, thermal properties, biocompatibility, easy process ability and low viscosity [16, 18]. Filla *et al*., [19] proved that the reinforcements of chitosan with polymer does not affect the polymer properties negatively which results that chitosan is efficient to be utilized as bio-filler. In addition, Zhao *et al*., [20] stated that the reinforcement of chitosan with the polyvinyl alcohol (PVA) able to improve the performance of the chitosan film via formation of intermolecular of the chitosan with the polymer. Therefore, chitosan is suitable for utilized in various fields including water treatment, food processing, biomedical, and agricultural [15].

In the pursuit of expanding the use of chitosan in polymer applications and managing the massive quantity of residues produced, it is vital to comprehend how chitosan interacts with compatibilized matrices like PBAT. Moreover, based on the available information, there is a scarcity of research papers that have addressed the use of chitosan-derived fillers in PBAT. By employing chitosan as a reinforcing agent, the composite material produced in the present study is both durable and environmentally benign. In addition, the matrix utilized was the completely biodegradable polymer PBAT. As a result, the components that were employed were biodegradable and favorable to the environment. PBAT composites were produced by incorporating chitosan additives at different concentrations (0%, 10%, 20%, and 30%).

2.0 EXPERIMENTAL WORK

2.1 Raw materials preparation

The polymer PBAT (ecoflex F mix C1200) was acquired from BASF SE, located in Ludwigshafen, Germany. Table 1 provides a comprehensive overview of the physical and mechanical properties of the PBAT polymer according to the supplier. The Chitosan 38906, with a molecular weight of 345,500 g mol-1 and a degree of deacetylation of 84.5%, was acquired from Chemiz (M) Sdn Bhd in Selangor, Malaysia.

Table 1 Properties of PBAT used in this study

Properties	PBAT
Density (g/cm^3)	1.25-1.27
Melt flow index (g/10 min @ 190°C)	2.7 – 4.9
Melting Temperature (T_m, °C)	110 - 120
Tensile yield strength (MPa)	36
Tensile modulus (GPa)	-
Tensile elongation (%)	560

2.2 PBAT/Chitosan Composite Fabrication

PBAT/Chitosan composites were prepared by melt mixing PBAT with varying concentrations of chitosan (0%, 10%, 20%, and 30% w/w) using a Rheomixer batch mixer (HAAKE™ Rheomix Lab Mixers, Germany) at a temperature of 120 °C and a rotational speed of 60 rpm. Following the mixing process, the composites were pelletized using a pelletizing machine (Crusher model:1506 s, MIKASA Sangyo Co., Ltd, Japan). The pellets were subsequently subjected to hot pressing at a temperature of 120 °C for a duration of 7 minutes, utilizing a square plate of 150 mm x 150 mm x 20 mm. Next, the composites underwent a post-curing process for 24hours at room temperature before being cut following the regular testing procedure.

2.3 Tensile and Morphological Analysis

To determine the tensile strength, the composite specimens were cut to the dimensions of 115 mm × 61 mm × 3 mm, following the ASTM D638 Type IV standard. Prior to commencing the testing procedure, in accordance with ASTM rules, all samples were subjected to conditioning at a temperature of 22 °C and a relative humidity of 50%. The tests were conducted using a Universal Testing Machine (5 kN Blue Hill INSTRON Universal Testing Machine, INSTRON Corp., United States). During the test, the specimens underwent stretching at a velocity of 20 mm/min, while the gap between the fixtures was adjusted to 25 mm. Each group of samples underwent five tests, and the average value was calculated based on the valid values.
The surface morphology of the films was examined using a scanning electron microscope (SEM) (EM-30AX COXEM, Daejeon, Korea). The voltage used for acceleration remained constant at 10 kV throughout the scanning procedure.

3.0 RESULTS AND DISCUSSION

3.1 Tensile Properties

The impact of varying chitosan concentrations in PBAT on the tensile characteristics of the composite material is illustrated in Figure 1-3. As the chitosan content of the PBAT composite increases gradually, its tensile strength and elongation at break exhibit a declining trend, as illustrated in Figures 1 and 2. The tensile strength of the PBAT composite decreases from 8.4 MPa to 3.8 MPa as the chitosan content increases from 0% to 30%. The test results indicate that the tensile properties of chitosan filled PBAT composites are considerably diminished in comparison to those of pure PBAT.

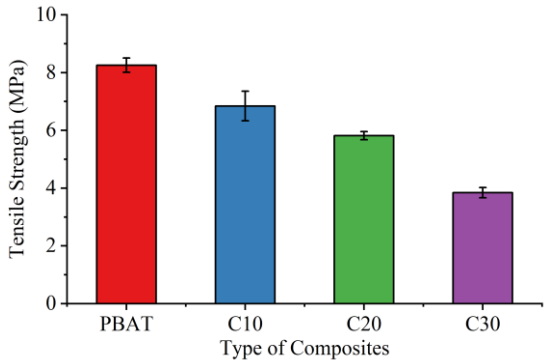


Figure 1 Tensile strength of PBAT/chitosan blends as a function of different amount of chitosan content

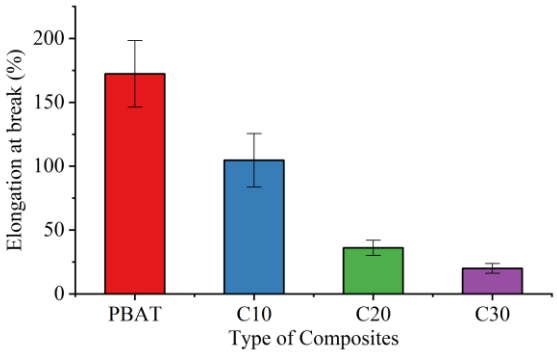


Figure 2 Elongation at break of PBAT/chitosan blends as a function of different amount of chitosan content

The use of inorganic filler-filled polymers often results in a significant deterioration of the tensile properties of the composite as the content of inorganic filler particles, such as chitosan, increases. The specific surface area of the inorganic infill will influence the quantity of molecular chains that can be adsorbed, thereby increasing the material's propensity to agglomerate. Increasing the quantity of chitosan resulted in a decrease in the tensile properties of the PBAT composites.

Furthermore, the reduced strength can be attributed to the absence of shear yielding during the mix stretching process. The results indicate that the inclusion of chitosan in the blends causes the chitosan to be dispersed with PBAT, resulting in increased steric hindrance. This limits the tensile performance of PBAT [21]. In addition, Dammak *et al.*, [7] shows the similar results as the elongation at break (%) of its composite PBAT with maleic anhydride (MA).

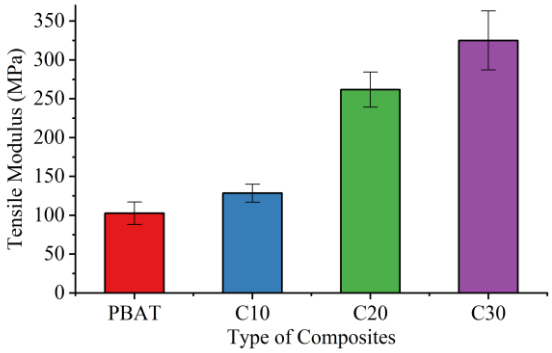
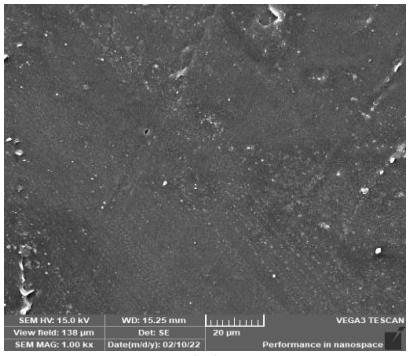


Figure 3 Tensile modulus of PBAT/chitosan blends as a function of different amount of chitosan content

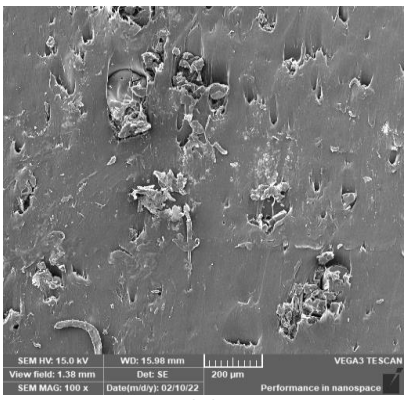
Figure 3 summarizes the tensile modulus of PBAT composites. The bar chart highlights that the addition of chitosan filler caused a linear increase of the tensile modulus upon increasing the filler concentration. More in detail, the tensile modulus of PBAT composite increase from 109 MPa up to 135 MPa, 275 MPa and 338 MPa. The different tensile modulus increase observed is probably due to the higher reinforcing action of chitosan to the PBAT matrix. Previous study from Li *et al.*, [22] indicates similar findings regarding the increment of tensile modulus of PBAT blends with thermoplastic starch modified with lignin content increasing. Another similar finding found on Botta *et al.*, [23] which indicated that the tensile modulus of PBAT/microcrystalline cellulose MCC increasing due to the increment of MCC content in the blend.

3.2 Morphological Analysis

Figures 4 and 5 display the impact of chitosan on the microscopic morphology of the PBAT composite. Figure 4 displays the surface of the composite material, while Figure 5 exhibits the fractured surface resulting from the tensile strength test.



(a)



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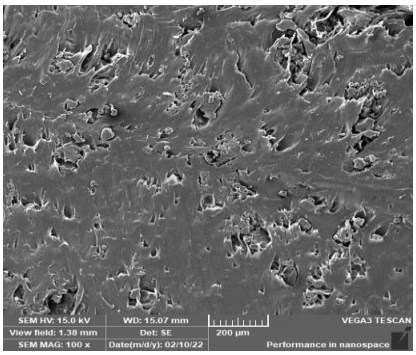
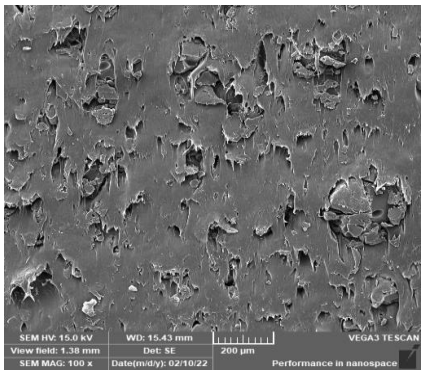


(c)



(d)

Figure 4 SEM observation of PBAT/chitosan composite at different chitosan content; (a) 0 %, (b) 10 %, (c) 20 %, (d) 30%

The figures clearly demonstrate a noticeable aggregation of chitosan particles in the composite system, resulting in separate agglomerates. This suggests that the compatibility between chitosan and PBAT matrix is inadequate, and the interfacial bonding is poor. When external forces are applied to the composite, it is prone to fracturing at the interface, leading to a considerable loss in its mechanical properties. Furthermore, the analysis of Figure 4 (a-d) reveals that a decrease in the amount of chitosan results in non-uniform distribution of chitosan within the composite system, primarily due to chitosan agglomeration. As the amount of chitosan increases, the chitosan particles are

dispersed more evenly in the system, but the tendency for them to accumulate together gets more pronounced. A similar finding was found on Liu *et al*., [22] research where the 30 % content of lignin agglomerates within the PBAT matrix. Moreover, another previous study form Venkatesan *et al*., [25] recorded similar finding where the surface of PBAT become rougher as per content of AgSnO2 increase.

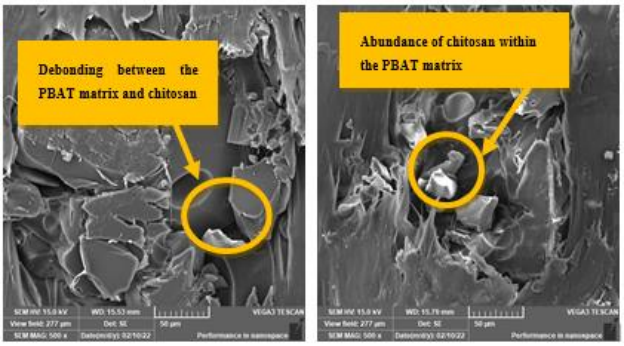


Figure 5 SEM observation of shattered surface of PBAT/chitosan composite from tensile strength test

The fractured surface of PBAT/chitosan composite was observed as shown in Figure 5. The fractured surface of the PBAT/chitosan composite indicates debonding between the PBAT matrix and chitosan, resulting in poor bonding between the two materials. Consequently, void regions were observed throughout the PBAT/chitosan matrix. The presence of a significant amount of non-bound chitosan within the PBAT matrix indicates poor interfacial adhesion in this matrix. Similar findings found on Qiu *et al*., [26] research where the PLA/PBAT poor interfacial indicate by the abundance of PBAT within the matrix. In addition, similar findings were found on Xiong *et al*., [27] where the lignin content in the PBAT matrix created a gap within the bond of the matrix which resulted in a poor bonding within the matrix.

4.0 CONCLUSION

Aiming at production of bio compostable composite to valorize industrial waste, chitosan was successfully used as filler for a PBAT via melt mixing. The tensile properties studies indicated that an increased concentration of chitosan in the matrix led to greater agglomeration of chitosan within the matrix, resulting in structural damage to the matrix. This behavior can certainly be noticed by SEM tests. Consequently, the tensile properties of the matrix, such as tensile strength and elongation at break, exhibited a steady decrease, while the tensile modulus of the matrix increased, indicating an increase in rigidity. This discovery demonstrates the potential use of this blend in future applications to enhance the production of high-quality polymer bio-composites.

Acknowledgement

This research is fully supported by GPM Grant, Universiti Teknologi MARA, 600-RMC 5/3/GPM (041/2023). The authors fully acknowledged INTROP, Universiti Putra Malaysia for the testing equipment facilities which makes this important research viable and effective.

Conflicts of Interest

The authors declare that there is no conflict of interest regarding the publication of this paper.

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