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Thermal, flammability, and antimicrobial properties of arrowroot (*Maranta arundinacea*) fiber reinforced arrowroot starch biopolymer composites for food packaging applications

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ABSTRACT

Using the solution casting method, a novel biodegradable thermoplastic arrowroot (*Maranta arundinacea*) starch (TPAS) films containing arrowroot fiber (AF) at different concentrations (0, 2, 4, 6, 8, and 10 wt%) were developed and characterized in terms of thermal, antibacterial activity, water vapor permeability (WVP), biodegradability, and light transmittance properties. The TPAS/AF-10 biocomposite film revealed a higher degradation temperature (313.02 °C) than other biocomposite films, indicating better thermal stability. Furthermore, increasing AF concentration led to a significant ($p < 0.05$) reduction in the linear burning rate and WVP of the biocomposite films from 248.9 to 115.2 mm/min and 8.18×10^{-10} ×g. $s^{-1}.m^{-1}$. Pa^{-1} to 5.20×10^{-10} ×g. $s^{-1}.m^{-1}$. Pa^{-1}, respectively. The addition of fibers in the surface structure had a significant impact on remarkable drop in opacity (91.1 to 74.1%). In addition, the incorporation of AF and control film showed an insignificant effect against three pathogenic bacteria, including *Staphylococcus aureus* (ATCC 43300), *Escherichia coli* (ATCC 25922), and *Bacillus subtilis* (B29). The soil burial findings demonstrated that the weight loss of TPAS/AF biocomposite films was significantly higher than TPAS film. Overall, the reinforcement of arrowroot fiber with TPAS film improved the properties of biocomposites for environmentally friendly food packaging applications.

1. Introduction

Over the past decades, global plastics production has been continuously growing. The production of these materials is now expected to surpass 350 million tons [1]. Packaging applications account for approximately 39.7% of total plastic demand and are regarded as the world's leading market for plastics [1,2]. However, the majority of the polymeric materials used in this field are derived from non-renewable resources, raising concerns about environmental pollution problems.

Therefore, natural polymers are a suitable option in the packaging market to diminish the use of non-biodegradable and non-renewable sources. Hence, due to its low cost, abundance, as well as thermoplastic characteristics, starch has been deemed one of the most potential alternatives for forthcoming materials [3,4]. Furthermore, several studies have found that starch from a number of plant sources, including wheat, corn, potato, and rice, have promising film-forming abilities [5–9]. Insight into the various methods involving the development of starch into a biopolymer has advanced significantly in recent years [10,11]. Starch is now a competitive alternative to petroleum-based polymeric materials in applications where long-term reliability and high mechanical strength are not required.

Over the last few decades, natural cellulosic fibers have played an essential role in a wide range of uses owing to their intrinsic sustainable as well as environmental benefits. They are being studied as green and

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sustainable substitutes for conventional synthetic fibers for a variety of applications [12]. Composite materials, including natural fibers, have several benefits compared to conventional materials, including flexibility, higher stiffness, ease of processing, recyclability, as well as an environmentally friendly nature.

Because of their widespread availability and renewable nature, thermoplastic materials derived from starch are one of the most potential substitutes for developing bio-based materials [13–15]. Over the last few years, the addition of natural fibers in biocomposite films as filler materials has been found to lower costs while improving overall performances [16]. There are numerous vegetable fibers that could be used to reinforce films, including sugar cane, cassava, corn, and wheat straw.

Arrowroot (*Maranta arundinacea*) tuber belongs to the *Marantaceae* family and is the source of a significant amount of starch, fiber, and carbohydrates [17]. Arrowroot starch is a locally produced native starch in Indonesia and Malaysia derived from arrowroot tubers with unique properties such as digestibility and gel-forming ability, as well as the highest amylose content (40.86%) [18], competing with corn starch (28–33%), cassava starch (16–19%), wheat starch (30–32%), and potato (18–20%), which are all required for film production. Therefore, the Indonesian government has prioritized the development and cultivation of arrowroot, which has the potential to replace wheat flour in Indonesia [19]. Previous studies have documented that the film-forming properties of starch depend on the amylose content; strong and stiff films are made by linking linear chains by hydrogen bonding. Hence, the high amylose content of arrowroot starch develops stronger films than other starch origins.

In previous research, arrowroot starch thermoplastic films were produced via a solution casting method utilizing extracted starch from rhizomes using a low cost, simple, as well as optimized technique [20]. However, control TPS has some drawbacks, such as highly hydrophilic and possesses poor mechanical properties. Therefore, in order to overcome this problem, reinforcement of TPS with natural fiber is a viable approach to increase the properties of this biopolymer while maintaining its biodegradability characteristic. During the arrowroot starch extraction process, a considerable amount of waste arrowroot fibers is obtained. Therefore, the residue after starch extraction could be used in food packaging applications. A variety of fiber reinforcements have lately been utilized in the context of biopolymers, such as potato peel waste [21], *Dioscorea hispida* [22], corn husk [23], cassava bagasse [24], and sugar palm [25]. According to the findings of arrowroot bagasse fiber [26], the fiber has better chemical composition and physical properties compared to other biomaterials, as shown in Table 1. Thus, this research proposes a use for this residue as a reinforcing agent, along with a chemical and morphological characterizations to analyse its impact on reinforced materials.

However, to the best of our knowledge, studies dealing with arrowroot starch reinforced arrowroot fiber biocomposite films have not been performed. In this research, TPAS/AF biocomposites were developed by solution casting method, and the objective of this work was to investigate the influence of AF incorporation at various fiber loadings (0, 2, 4, 6, 8, and 10%) on the thermal, water barrier, antibacterial, and optical properties of produced films. The findings suggested that the films made from plasticized arrowroot starch and incorporating AF could be useful for food packaging.

2. Materials and methods

2.1. Materials

Arrowroot tubers were purchased from Norient Jaya Sdn. Bhd., Kuala Lumpur, Malaysia. Arrowroot bagasse fibers and starch were extracted from tubers following the process outlined by Tarique et al. [26]. Arrowroot fiber was washed, dehydrated, pulverized, and screened through mesh sieve (<300 μm). Arrowroot fibers contain 45.97% cellulose, 30.18% hemicellulose, 2.78% lignin, 4.29% ash, and a density of 1.11 g/cm^3 [26]. The glycerol (with 99% purity) was supplied by Evergreen Engineering & Resources Sdn. Bhd., Selangor, Malaysia.

2.2. Development of TPAS/AF biocomposite films

The conventional solution casting method had been used to develop biopolymer composites. To make a film-forming solution, the AS was dispersed in distilled water at a concentration of 5% *w*/w. Based on previous research [20], 1.5 g (*w*/w, starch basis) glycerol was added into the solution. According to the formulation shown in Table 2, arrowroot bagasse fibers were incorporated into the solution as a reinforcing agent at concentrations ranging from 0 to 10% by weight. These mixtures had been heated for 15 min in a thermostatic bath (Daihan Scientific, Singapore) at 80 ± 3 °C with gentle and continuous stirring. To remove air bubbles developed during solution making, the biopolymer composite-forming solution were cooled and placed in a vacuum desiccator before casting. Thereafter, 50 g of each solution was cast in a 120 mm diameter Petri dish that functioned as a surface of the casting, resulting in a good surface finish. For dehydration, the cast plates were positioned in an airflow oven set to 45 °C for 24 h. Subsequently, dehydrated composite films were conditioned at 25 °C for 24 h before being separated from plates. Finally, the biocomposite specimens were held for a week at 25 ± 3 °C and 52% relative humidity before being characterized. Fig. 1 displays the graphic images of the developed TPAS and TPAS/AF biocomposite films.

2.3. Characterization of TPAS/AF biocomposite films

2.3.1. Thermogravimetric analysis (TGA)

A TA Instrument Model Mettler-Toledo AG, (Switzerland) Model TGA-6 thermogravimetric was used to test the thermal stability of the TPAS and TPAS/AF composite films. Thermal analysis of materials provides some fundamental information about a material's thermal

Table 2
Various compositions of TPAS and TPAS/AF composites.

Sample	Glycerol (g)	Starch (g)	Fiber loading (%)
TPAS	1.5	5	0
TPAS/AF-2	1.5	5	2
TPAS/AF-4	1.5	5	4
TPAS/AF-6	1.5	5	6
TPAS/AF-8	1.5	5	8
TPAS/AF-10	1.5	5	10

Table 1
Chemical and physical properties of arrowroot fibers and other biomaterials [26].

Biomaterial	Cellulose (%)	Hemicell-ulose (%)	Lignin (%)	Ash (%)	Density (g/cm^3)	Moisture content (%)	Water holding capacity (%)
Arrowroot bagasse fiber	45.97	30.18	2.78	4.29	1.11	7.99	131.45
Arrowroot husk fiber	37.35	26.85	8.89	8.79	1.23	5.51	58.01
Dioscorea hispida	5.63	4.36	2.79	1.28	1.47	9.15	–
Cassava	10.04	29.26	3.12	3.36	1.45	14.92	258.05
Corn husk	45.7	35.8	4.03	0.36	1.49	7.81	78.8
Corn hull	15.30	40.4	2.87	0.88	1.32	8.59	37.4
Cornstalk	10.8	60.3	1.98	1.97	1.42	11.1	93.7

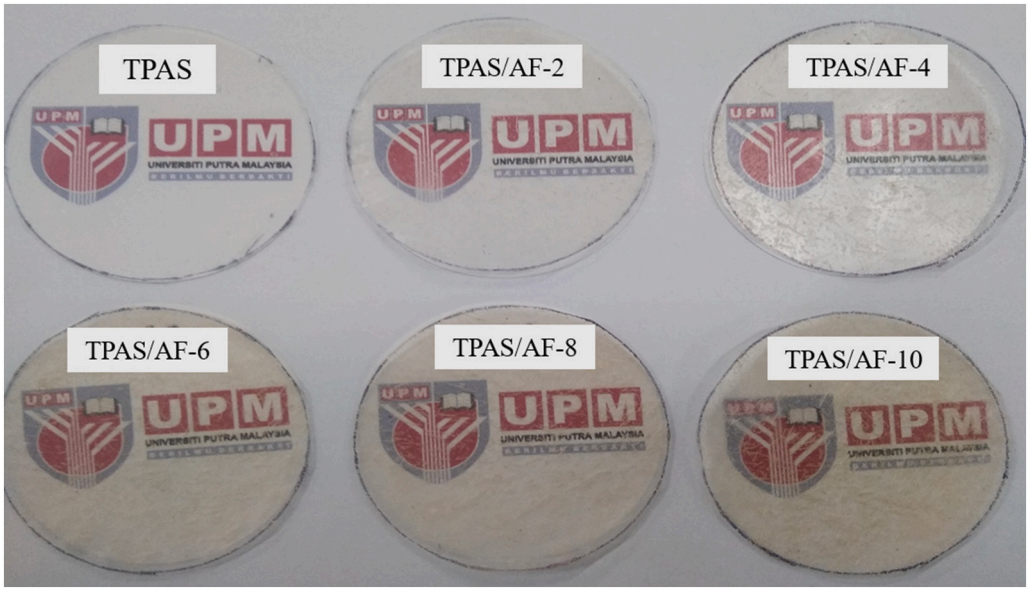

Fig. 1. Visuals of TPAS and TPAS/AF biocomposite films with different fiber loadings.

stability. Experiments were performed using a 50 mL/min nitrogen gas flow at a temperature range of 25–450 °C with 10 °C /min constant heating rate and an alumina crucible with a pinhole. TGA is a technique used to characterize the decomposition and thermal stability of materials under different conditions. TGA analysis primarily examines changes in thermal stability in terms of percentage weight loss as a function of temperature. The specimens, weighing between 10 and 15 mg, were placed in ceramic crucibles, and tested in a nitrogen atmosphere.

2.3.2. Antimicrobial activity

Antimicrobial activity test of composites was tested against *E. coli* (ATCC 25922), *B. subtilis* (B29), and *S. aureus* (ATCC 43300) agar diffusion method. The composite film samples were cut into 6 mm diameter using paper puncture under sterile conditions. After that, biocomposite samples were immersed in 0.5 mL McFarland broth standard agar containing 1 × 108 CFU/mL. The specimens were cultured in various microorganisms, then covered with a combination of bacterial suspension on the plate's surfaces. The test plates were incubated at 30 °C for 24 h. The diameter of contact area of films with agar surface was measured. For each formulation, the tests were conducted in triplicate.

2.3.3. Water vapor permeability (WVP)

In a desiccator, the samples were conditioned according to the set of working conditions of 53 ± 1% relative humidity and 23 ± 2 °C. The WVP test was performed in agreement with ASTM E96–95 [27], with minor changes described by Sanyang et al. [28]. The testing was performed three times. At first, 20 g of silica gel was filled into the cup (dia: 30 mm). The film specimens were taken into circular forms and mounted to the mouth of circular cups. The cup with film sample was weighed and recorded before being placed in a constant relative humidity (75% RH, 25 °C). The cups were weighted on a constant time interval till the equilibrium value was achieved. At last, the test cup with the increased weight was weighed and used to calculate the WVP using Eq. (1).

$$\mathrm{WVP} = \frac{\Delta \mathrm{m} \times \mathrm{d}}{\mathrm{A} \times \mathrm{t} \times \mathrm{P}} \tag{1}$$

where m (g), d (mm), A (m^2), t (s), and P (Pa) represent the increased weight of the test cup, film thickness, exposed area of the film, transmission time interval, and partial pressure of water vapor over the sample, respectively.

2.3.4. Biodegradability of TPAS/AF biocomposite films

The weight losses of TPAS and TPAS/AF biocomposite specimens buried in compost soil in constrained humidity conditions were calculated to determine the biodegradability and the degradation rate of biocomposite films in soil. The physicochemical properties of soil were as follows: pH: 6.52; carbon: 1.14%; nitrogen: 0.07%; phosphorous: 96.6 g/g; and potassium: 45.93 g/g. Each film specimen was buried 10 cm beneath the soil surface in triplicate, with a size of 20 mm × 20 mm as shown in Fig. 5. The specimens were buried at 2, 4, 6, 8, and, 10 days intervals, with each specimen being dug out from the soil simultaneously. After that, the specimen was gently cleaned with distilled water to remove impurities and dried in an airflow oven at 65 °C to achieve a consistent weight. Eq. (2) was used to calculate the weight loss of the test specimens.

$$\text{Weight loss } (\%) = \left(\frac{W_i - W_f}{W_i}\right) \times 100 \tag{2}$$

where (W_i) and (W_f) are the pre-burial weight and post-burial weight of the specimens, respectively.

2.3.5. Flammability testing of TPAS/AF biocomposite films

Flammability testing was performed on all specimens using a horizontal burning testing in accordance with ASTM D635 [29] with minor modifications. Specimens with the dimension of (125 mm × 10 mm × 0.2 mm) were prepared, and two lines were taken as reference marks at 25 mm and 100 mm from one end of the specimen. After which, on one end of the sample, a fire was started with natural gas. The total length burned as well as the time it took for the flame to spread to 25 mm and 100 mm reference points were recorded. Eq. (3) was used to calculate the linear burning rate of specimens.

$$\mathrm{V} = 60\,\mathrm{L/t} \tag{3}$$

where V, L, and t represent the linear burning rate (mm/min), burnt length, as well as time (minute), respectively.

2.3.6. Light transmittance analysis of TPAS/AF biocomposite films

Film light transmittance (T_r) was measured using a UV–visible spectrophotometer Model (UV-1800, Shimadzu, Japan) in the range of 200–800 nm wavelength, with an accuracy of 0.1 nm. The light

transmittance of biocomposite films was calculated at 800 nm. A blank glass plate has been used as a reference.

2.4. Statistical analysis

An analysis of variance (ANOVA) on the experimentally obtained data was carried out using SPSS software. Duncan's test was used to compare means at a 5% significance level ($p \leq 0.05$).

3. Results and discussion

3.1. Thermogravimetric analysis (TGA)

Fig. 2 shows the thermal stability degradation temperatures of TPAS and TPAS/AF biopolymer composite films at various fiber loadings. The TGA curve was obtained by plotting the weight reduction (mg) of composite film samples against temperature (°C), while the DTG curve was obtained by plotting the derivative of weight reduction (mg) as a temperature-dependent (°C). Podshivalov et al. [30] found similar findings in potato starch reinforced with gelatin (0–50 wt%) and plasticized with glycerol, and Sanyang et al. [31] also observed similar results in sugar palm starch-based films.

The initial degradation process occurred below 100 °C due to the elimination of water molecules from the biocomposite film samples. Besides, weight loss at this phase could also be associated with dehydration or vaporization of low molecular weight chemicals and loosely bound water in biocomposites. This happened with all other film specimens, along with the control TPAS film. It also can be observed that the onset decomposition temperature of biocomposite films varied between 35 and 36 °C, occurred at a higher temperature than the TPAS control film, which started at 26.45 °C; this indicated good interaction between the matrix and fibers. Further heating resulted in a strong peak in the DTG curve of TPAS film at 314 °C, corresponding to nearly 88% weight reduction, which was likely attributable to saccharide ring degradation in TPAS biopolymer film. At a temperature lower than 100 °C, the TPAS biopolymer lose more mass compared to TPAS/AF biocomposite films. These behaviors could be linked to the TPAS biopolymer containing more moisture compared to TPAS/AF biocomposites [22,32].

The second thermal degradation of samples occurred between 160 and 280 °C and was linked with the volatilization of glycerol along with chemisorbed molecules of water. This temperature range of glycerol degradation of TPAS and TPAS/AF biocomposite films supported the findings of Podshivalov et al. [30] and Ilyas et al. [32] on potato and sugar palm starch, respectively. Similarly, Zhong and Li [33] observed

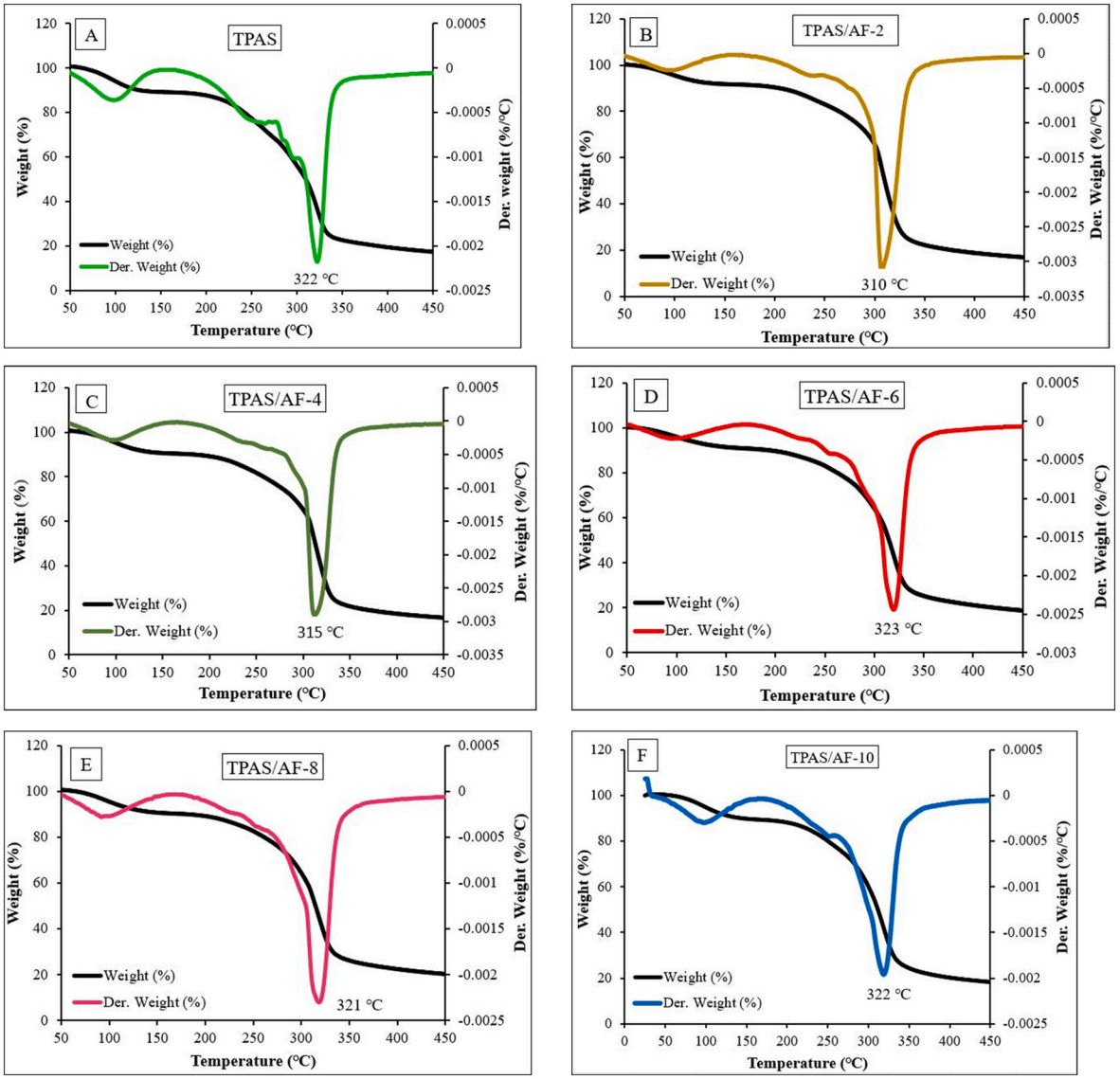

Fig. 2. TGA and DTG curves of TPAS and TPAS/AF biocomposite films with different fiber loadings.

that the thermal decomposition of kudzu starch-based biopolymers plasticized with glycerol was in the range of 150–280 °C. The maximum thermal decomposition rate was attained at a temperature over 280 °C, as noted by the major weight losses of both TPAS biopolymer and TPAS/AF biocomposites.

The start of thermal decomposition of TPAS biopolymers was found at about 300 °C, as per Fig. 2(A). This could be associated with the loss of hydroxyl groups, depolymerization, as well as decomposition of the polymeric carbon chain of starch, which is the primary constituent of biocomposites [34]. The optimum decomposition temperatures of TPAS and TPAS/AF-6 were 314 and 323 °C, respectively, indicating that TPAS/AF-6 biocomposite film was more thermally stable compared to TPAS biopolymer film. Meanwhile, the DTG peak of TPAS/AF-6 biocomposite shifted to a higher temperature indicating better thermal stability compared to other film formulations. This can be associated with the good adhesion between matrix and fibers. It was also observed that the increase in fiber loadings increased the thermal stability of biocomposite films. Previous studies also found that the reinforcement of fiber into starch biopolymer enhanced the thermal stability of film samples by strengthening husk fiber into corn starch-based film and cellulose nanofibril into chitosan/oregano essential oil [23,35].

Nevertheless, Table 3 demonstrates that an increase in arrowroot fiber concentration lowered the weight loss of biocomposites at temperatures above 300 °C, leading to an improvement in the thermal stability of biocomposites. Char is the substance that remains after all volatile substances in the material have been pyrolyzed. The amount of char residue of TPAS/AF biocomposites increased with the addition of AF, with an increase of 16.67% from 12.18% when 2 wt% of AF was added to TPAS (control AS). The increased thermal strength of TPAS/AF biocomposite films could be attributed to the existence of huge carbonates in the AF, which corresponded to the biocomposite's thermal decomposition results. These findings were consistent with previous research, which demonstrated that incorporating natural fibers within the matrix improved the thermal stability of the biopolymer matrix [36,37]. It can be summarized that adding AF to biocomposite films increased their thermal properties, in which good thermal stability is an important characteristic of food packaging materials. It could also withstand high temperatures during composite fabrication.

3.2. Antibacterial analysis of TPAS/AF biocomposite films

Abral et al. [38] conducted an experiment on poly (vinyl alcohol)/ginger nanofiber bionanocomposite films without any antimicrobial agent and found the antimicrobial effect against both gram-positive and gram-negative bacteria. These results were found because ginger has been reported to contain monoterpenoid, sesquiterpenoid, phenolic compounds, aldehydes, ketones, and alcohols, which showed a broad antimicrobial spectrum of activity against different microorganisms [39]. Similarly, the rhizomes of arrowroot contain different chemical compounds such as alkaloids, glycosides, phenolic compounds, terpenoids, flavones, and tannins, which are attributed to antibacterial and antifungal properties [40]. Following this, the positive results may have been expected without an antimicrobial agent against both gram-positive and gram-negative bacteria.

However, the antibacterial activity of TPAS biopolymer and TPAS/AF biocomposites were tested using ADM against gram-positive bacteria (*B. subtilis* B29 and *S. aureus* ATCC 43300) and gram-negative bacteria (*E. coli* ATCC 25922). A TPAS film was used as a control to see if the films without filler had any antibacterial effects. The findings revealed no inhibitory zone for *S. aureus* ATCC 43300 and *E. coli* in control and TPAS/AF films against any of the tested bacteria, indicating no antimicrobial activity for these microbes. According to Nogueira et al. [41], arrowroot starch-based film with no filler has an insignificant antioxidant and anthocyanin capacity. Remarkably, no zones of inhibition were visible under the TPAS/AF biocomposites films in Fig. 3, signifying that AF could not prevent these microbes in direct contact with the film. Hajer et al. [42] noticed a similar pattern when testing the antimicrobial activity of various sodium alginate (NaAlg) and gallnut extract (GE) film-forming solutions against a representative gram-positive bacteria, *Staphylococcus aureus* (ATCC 6538), a gram-negative bacteria, *Escherichia coli* (ATCC 11775), and two pathogenic molds, *Penicillium digitatum* and *Aspergill*. However, NaAlg film-forming solutions integrated with GE at various concentrations showed no apparent inhibition on agar plates against *P. digitatum* and *A. niger,* indicating that GR has no antifungal effect against the tested pathogenic fungi. In contrast, increasing the GE concentration greatly enhanced the antimicrobial action of the composite NaAlg film-forming solutions against the tested microorganism. In fact, Gram-positive bacteria's exterior cell wall is made up of acidic polysaccharides (teichoic) and peptidoglycan, which have a lot of holes and may let bioactive compounds adhere to or penetrate the cell. Apart from the cell membrane, Gram-negative bacteria have an extra outer membrane made up of lipopolysaccharide, lipids, and proteins that may act as a barrier to bioactive substances entering the cell [43]. It can be summarized that the incorporation of an antimicrobial agent such as essential oil is required to obtain an antimicrobial film for food packaging applications. The incorporation of antimicrobial agents may improve the antimicrobial property of TPAS and TPAS/AS biocomposite films.

3.3. Water vapor permeability

In packaging applications, the hydrophobic nature of the material is the most important factor for material selection since the properties of the product could be affected whether the product is water-susceptible or not. Therefore, WVP should be as low as possible because packaging films are frequently required to prevent or at least reduce water transmission between food and surrounding [44]. In addition, decreasing the WVP of biocomposites is essential for their important possible applications. Fig. 4 shows the WVP of TPAS and TPAS/AF films. It can be observed that TPAS film had the highest WVP, which was 9.45 $\times 10^{-10} \times g.\ s^{-1}.m^{-1}.\ Pa^{-1}$. This was due to its hydrophilic character, which made them sensitive to moisture containment and surrounding humidity, which were difficult to control.

However, increasing fiber concentration from 2 to 10 wt%

Table 3
Onset temperature (T_{Onset}), temperature on maximum degradation (T_{Max}), weight loss (W_L), and mass residue for TPAS and TPAS/AF biocomposite films obtained from TGA and DTG curves.

Sample	Water evaporation			First thermal degradation			DTG Peak temperature	Mass residue
	T_{Onset} (°C)	T_{Max} (°C)	W_L (%)	T_{Onset} (°C)	T_{Max} (°C)	W_L (%)	(°C)	(%)
TPAS	26.45	100.04	12.91	165.32	297.63	75.09	314.03	12.18
TPAS/AF-2	36.33	98.15	8.74	155.06	304.15	74.88	310.22	16.67
TPAS/AF-4	35.73	98.75	10.46	163.41	309.33	73.69	315.49	16.86
TPAS/AF-6	35.05	91.93	9.69	169.07	311.01	72.07	323.15	18.72
TPAS/AF-8	35.54	96.4	10.54	166.21	311.48	70.08	321.74	20.18
TPAS/AF-10	35.55	98.55	11.08	162.82	313.02	71.13	322.73	18.27

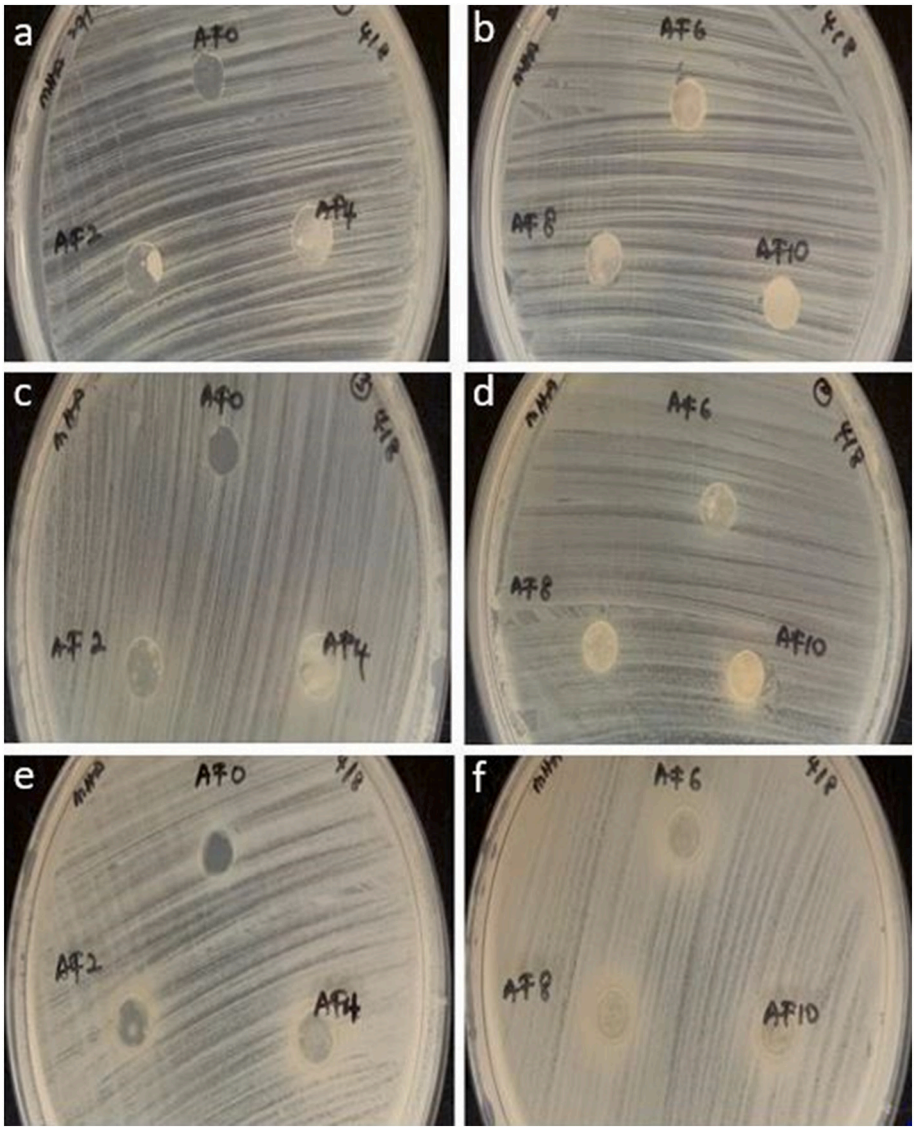

Fig. 3. Inhibitory effect of TPAS and TPAS/AF biocomposite films against *S. aureus* (a, b), *E. coli* (c, d), and *B. Subtilis* (e, f).

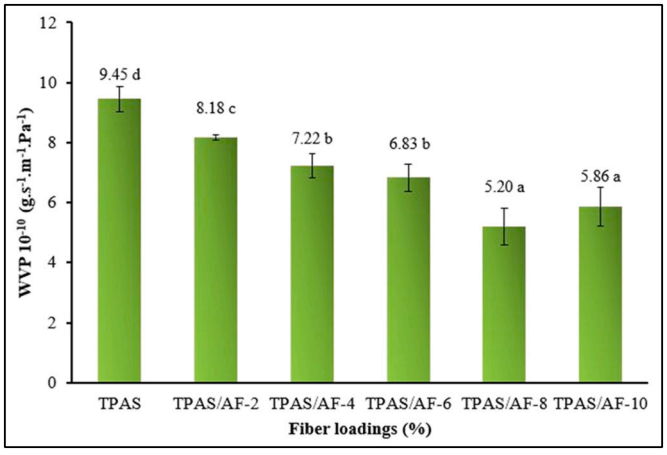

Fig. 4. Effect of fiber loadings on the WVP of the TPAS/AF biocomposite films compared with TPAS film. The letters give significant differences ($p < 0.05$) between means obtained in Duncan's test.

significantly ($p < 0.05$) decreased WVP of resulting TPAS/AF biocomposites from 8.18 to 5.20 × 10^{-10} ×g. $s^{-1}.m^{-1}$. Pa^{-1}. This reduction was observed around 30% for biocomposite film reinforced with the highest fiber loading (10 wt%) than the neat TPAS film ($p < 0.05$). Such interactions between fiber and matrix may have reduced the free volume of the polymeric matrix and resulted in the formation of a denser polymeric matrix, lowering the diffusion of water molecules through films. In line with our result, Uthaya Kumar et al. [45] found a significant reduction in WVP of seaweed-based biocomposite films when extracted neem leaves were increased from 0.5 to 7.5 wt%. According to these researchers, such behavior could be associated with the development of hydrogen bonds between (-OH) groups in neem extract as well as seaweed, which could restrict the accessibility of hydrogen groups to establish hydrogen bonds with water, thereby reducing the hydrophilic propensity of resulting biocomposites. Similarly, this finding is in agreement with the findings of Kan et al. [46], who experienced a decrease in WVP of gelatin-chitosan films when the amount of filler was increased from 2 to 4 wt%. These effects were attributed to the introduction of the extract, which formed a sinuous path that slowed the diffusion of water through film, lowering the WVP of final film samples. In agreement with our findings, Lopez et al. observed a considerable reduction in the WVP of TPS films and reinforced TPS films when *Pachyrhizus ahipa* starch extraction (PASR) (0.5% *w*/w) was added. The

addition of PASR to TPS films reduced the WVP values by 33%. Muller et al. [47] found a similar reduction in WVP in cassava starch films reinforced with fibers (0.30 g fiber/g dry starch) and 0.3 g glycerol/g dry starch. Bitencourt et al. [48] also found that gelatin-based films reinforced with higher concentrations of Curcuma ethanol extract improved their water barrier characteristics. However, based on the findings of Bitencourt et al. [48], a more reasonable explanation is the formation of cross-links as a result of interactions between the gelatin and phenolic compounds existing in the Curcuma ethanol extract, which could reduce the free volume in the polymer matrix, resulting in reduced interactions between water molecules in additivated films. It can be summarized that bioplastics with low WVP are suitable for use in food packaging.

3.4. Biodegradability of TPAS/AF biocomposite films

Fig. 6 displays the weight loss of TPAS and TPAS/AF biocomposites as a function of decomposition time after degradation testing. Soil burial test of biocomposite films was monitored for 20 days. During the earlier biodegradation stage, water diffusion induced swelling in samples, which helped the bacterial attack [23]. The TPAS film sample had lost 64% of its weight after 8 days, while the composite films had lost 66.7, 67.7, 71.7, 73.2, and 78.7% of their weight for TPAS/AF-2, TPAS/AF-4, TPAS/AF-6, TPAS/AF-8, and TPAS/AF-10, respectively, as shown in Figs. 5 and 6. For the TPAS sample and TPAS/AF-10, the average degradation rate was 8 and 9.84%/day, respectively. Throughout the analysis, the weight loss of TPAS/AF biocomposite films was significantly higher than TPAS biopolymers, as shown in Table 4.

The TPAS/AF-10 biocomposite sample took 12 days to decompose completely. On the other hand, the biocomposite samples with 2, 4, 6, and 8% fiber loadings had lost 73.5, 76.2, 83.3, 87.3, and 93.6%, respectively. The weight loss of the control film was lower compared to composite samples. González [49] ascribed this occurs due to the existence of a connection between the water content and bacterial activity of the soil, where the higher water content of the film resulted in higher bacterial activity within the film. López et al. [50] reported consistent results when leafwood fiber was used as a filler with wheat starch. Avérous et al. [51] found similar results when adding fibrous residue of *Pachyrhizus ahipa* plant into thermoplastic corn starch films. This finding was also in line with those of Hazrati et al. [22], who investigated

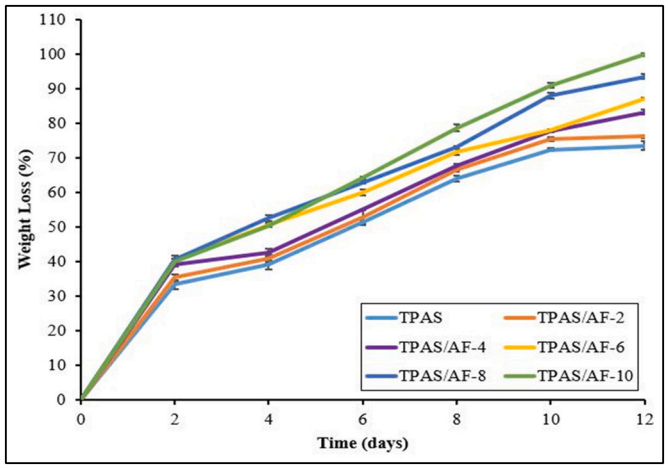

Fig. 6. Weight loss analysis of TPAS and TPAS/AF biocomposite films in biodegradability test.

Table 4
Biodegradability analysis of TPAS and TPAS/AF biocomposite film.

Sample /Day	TPAS	TPAS/ AF-2	TPAS/ AF-4	TPAS/ AF-6	TPAS/ AF-8	TPAS/ AF-10
2	33.3 ± 1.3^{a}	35.3 ± 1.0^{a}	39.2 ± 0.6^{a}	40.2 ± 0.5^{a}	40.6 ± 1.1^{a}	40.0 ± 0.8^{a}
4	39.0 ± 1.2^{b}	40.9 ± 1.2^{b}	42.5 ± 1.2^{b}	50.6 ± 0.6^{b}	52.6 ± 0.9^{b}	50.4 ± 0.5^{b}
6	51.4 ± 0.4^{c}	52.3 ± 2.2^{c}	55.1 ± 0.1^{c}	60.0 ± 1.0^{c}	62.9 ± 0.4^{c}	64.2 ± 0.3^{c}
8	64.0 ± 0.9^{d}	66.7 ± 0.7^{d}	67.7 ± 0.7^{d}	71.7 ± 0.9^{d}	73.2 ± 0.2^{d}	78.7 ± 0.9^{d}
10	72.4 ± 0.3^{e}	75.5 ± 0.5^{e}	77.8 ± 0.5^{e}	78.1 ± 0.1^{e}	87.9 ± 0.8^{e}	90.9 ± 0.7^{e}
12	73.5 ± 1.3^{e}	76.2 ± 0.5^{e}	83.3 ± 0.7^{f}	87.3 ± 0.3^{f}	93.6 ± 0.7^{f}	100 ± 0.5^{f}

*For the same column, different letter indicates significant difference ($p < 0.05$).

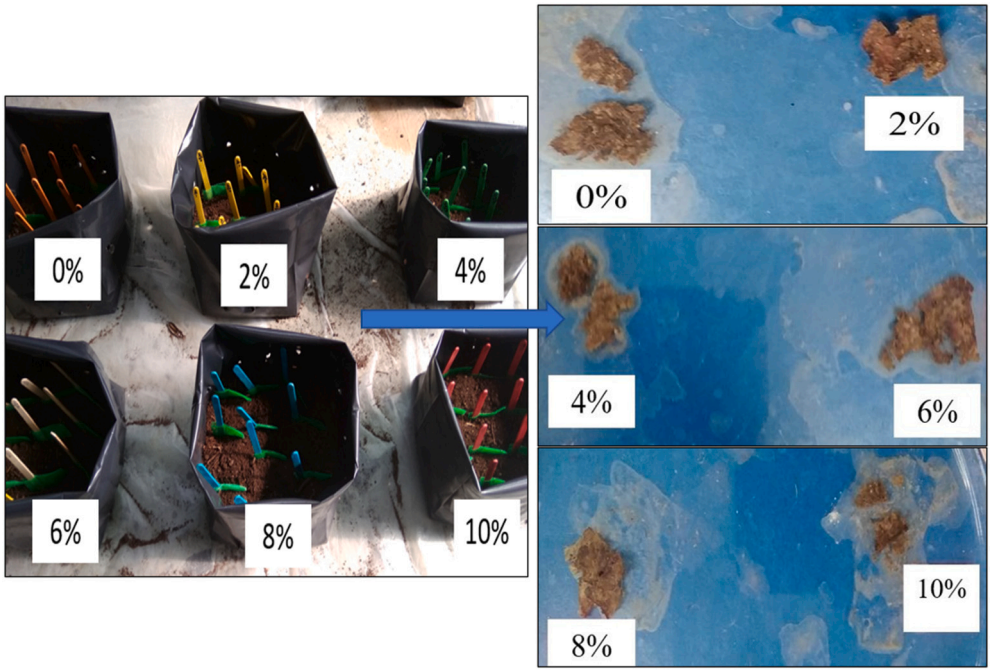

Fig. 5. Soil burial setup (left) and photographs of biodegraded films (at 8 days) of TPAS and TPAS/AF biocomposite films (right).

Dioscorea hispida/Dioscorea hispida fiber biocomposite films and found that biocomposites with higher fiber concentrations were more degradable than control film. Besides that, the results indicated that the biocomposite film with higher AF loading exhibited greater potential biodegradation, making it more vulnerable to bacterial attack. However, based on the results of related studies, a more plausible explanation is that in the presence of an aqueous medium, this microbe, in the form of bacteria or fungi, affects the biocomposite films [52,53]. As soil bacteria consume the surrounding starch, the biocomposite films lose the integrity of the structure, diminishing the interfacial strength between the starch matrix and the TPAS/AF biocomposites [54]. Indeed, the findings revealed that the TPAS/AF biocomposite films will not have an environmental impact, implying that the TPAS/AF biocomposite films are entirely biodegradable.

3.5. Flammability analysis of TPAS/AF biocomposite films

Plastics have evolved into one of the highly usable materials of humankind. Plastics can quickly burn if subjected to enough heat in the presence of oxygen owing to their chemical composition. Due to the high burning rate of plastics, a lot of effort has been spent on studying and mitigating the flammability of these products, such as the use of flame retardant compounds to inhibit or reduce burning. This experiment was used to identify and evaluate the burning properties of bioplastics. The linear burning rate in Fig. 7 shows the effect of different fiber loadings on the flammability of TPAS biopolymer and TPAS/AF biocomposite films. Fig. 7 illustrates the average burning rate for each sample graph, which was significantly ($p < 0.05$) decreasing with increasing fiber loadings (0, 2, 4, 6, 8, and 10%) in specimens. It can be observed in Fig. 7 that biocomposite films with 8% and 6% fiber loading possessed better flammability characteristics because the lowest average burning rates were 104.4 and 106.7 mm/min, while the average burning rates for fiber concentrations of 2 and 4% were 152.7 and 113.7 mm/min, respectively. This occurrence can be associated with the presence of lignin in fibers that are predicted to arise in a char formulation that helps to prevent flammability [55].

The TPAS film showed a higher burning rate than TPAS/AF biocomposite films. This is primarily due to the glycerol plasticizer used in the production of TPAS film. Glycerol is a flammable material with a flashpoint of 176 °C [56]. This result is also related to the thermal degradation property, where TPAS/AF biocomposites possessed higher thermal stability than TPAS film, as shown in Table 3. The TPAS without filler appeared to have a significant impact on the burning rate, as evidenced by the fact that TPAS (248.9 mm/min) burned faster than TPAS/AF-10 (115.2 mm/min). In other words, these were the least flame-resistant materials that needed to be improved in terms of flammability. Because of this, some studies used glycerol phosphate instead of glycerol to improve the flame-resistant capabilities of starch-based polymer blends [55,57]. On the other hand, Younis [58] reported that petroleum-based plastic such as polypropylene demonstrated a lower burning rate (25.4 mm/min) compared to bio-based plastics. Prabhakar et al. [59] reported that the polymer matrix was dependent on the reinforcement and filler materials. Furthermore, it played no significant influence in increasing the flame resistance of the biocomposites.

3.6. Light transmission analysis of TPAS/AF biocomposite films

Starch films are finally employed in the packaging of food. Thus, the light transmission properties of films are essential for their applications [60]. Fig. 8 shows the light transmittance (*Tr*) of transparent TPAS/AF biocomposites at 800 nm wavelength. Similarly, these results provide some insight into the pattern of fiber distribution within the film matrix. For instance, the TPAS film was more transparent compared to the TPAS/AF biocomposite films, as observed in Fig. 8, indicating that fiber addition reduced light transmittance (91.1 to 74.1%). The addition of fibers on the surface structure had a significant impact on the noticeable drop in *Tr* reflects. This occurrence could be related to (a) the existence of AF reinforcement material solid particles, (b) strong interaction between both matrix and fibers, as well as (c) random distribution of the arrowroot fibers in the starch matrix [61]. The transparency values of the TPAS/AF biocomposite films were significantly ($p < 0.05$) reduced when the AF concentrations were increased from 0 to 10 wt%, which was attributable to the homogeneous distribution of the AF within the biocomposite reducing film transparency, as can be seen in Fig. 1. It was closely consistent with the findings of Pelissari et al. [61], who used banana nanocellulose to reinforce the banana starch matrix, resulting in a decrease in transparency (90 to 74%). In the meantime, Wang et al. [62] reported that adding cotton linter nanocellulose concentration within soy protein film reduced the optical transmittance of film. Mahyar et al. [63] also noticed a similar trend, reporting a decrease in the transparency of thermoplastic starch as a matrix and cellulose nanofiber (TPS/CNF) by 18% as the content of CNF increased in biocomposite. These findings have important implications because film transparency and opacity are important characteristics in several film applications, especially when the films are employed as a food covering or to improve product presentation [64].

4. Conclusions

In this research work, glycerol plasticized TPAS films with different AF loadings were successfully produced using the solution casting

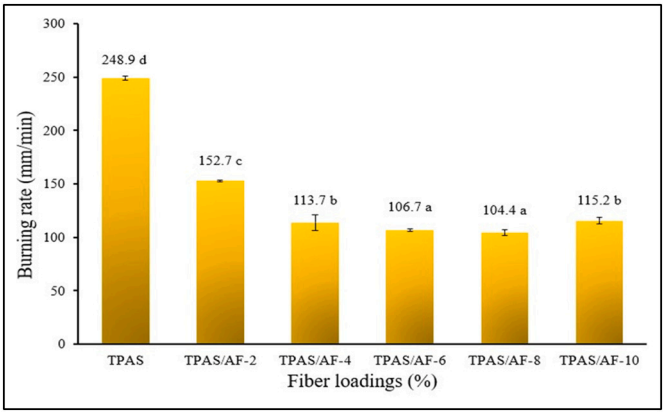

Fig. 7. The effect of various fiber loadings on burning rate of TPAS biopolymer and TPAS/AF biocomposite films. The letters give significant differences ($p < 0.05$) between means obtained in Duncan's test.

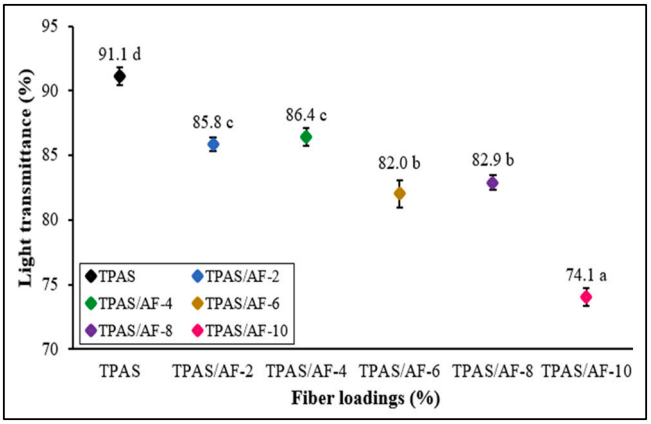

Fig. 8. Light transmittance (*Tr*) of TPAS/AF biocomposite films at 800 nm. The letters give significant differences ($p < 0.05$) between means obtained in Duncan's test.

technique and characterized. It was incredible to note that the TPAS/AF biocomposite films demonstrated a significant increase in degradation temperature from 298 to 313 °C, indicating a considerable increase in thermal stability. As the AF concentration increased, the light transmittance and linear burning rate decreased significantly. In comparison to the TPAS film, the TPAS/AF biocomposite film with 8% AF had the fastest linear burning rate and hence the shortest burnt-out time. The biodegradability results revealed that the addition of AF accelerated the biodegradation process because of the hydrophilic nature of fiber. Furthermore, the addition of AF revealed that the TPAS and TPAS/AF films had no significant action against the most prevalent pathogenic microbes, *S. aureus*, *E. coli*, and *B. subtilis*. The potential of this packaging material will be investigated further, and work will be carried out on incorporating essential oil into biocomposite films to improve antibacterial activity. Hence, this research work revealed the huge potential of TPAS/AF biocomposite films for packaging application.

CRediT authorship contribution statement

J. Tarique: Conceptualization, Methodology, Investigation, Writing – original draft, Writing – review & editing, Project administration. **S.M. Sapuan:** Supervision, Funding acquisition. **A. Khalina:** Supervision, Project administration. **R.A. Ilyas:** Supervision, Project administration. **E.S. Zainudin:** Project administration.

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