

# **Property Enhancement of Starch Based Bio-composite Film Using Reinforcements & Additives: A Comprehensive Review**

**Sabiqune Nahar Niti**

Department of Materials Science and Engineering  
Rajshahi University of Engineering & Technology  
Rajshahi-6204, Bangladesh  
[2013030@student.ruet.ac.bd](mailto:2013030@student.ruet.ac.bd)

**Maisha Chowdhury Karabi**

Department of Materials Science and Engineering  
Rajshahi University of Engineering & Technology  
Rajshahi-6204, Bangladesh  
[2013012@student.ruet.ac.bd](mailto:2013012@student.ruet.ac.bd)

**Shaira Anowara**

Department of Materials Science and Engineering  
Rajshahi University of Engineering & Technology  
Rajshahi-6204, Bangladesh  
[2013052@student.ruet.ac.bd](mailto:2013052@student.ruet.ac.bd)

**Maliha Rahman**

Department of Materials Science and Engineering  
Rajshahi University of Engineering & Technology  
Rajshahi-6204, Bangladesh  
[maliha@mse.ruet.ac.bd](mailto:maliha@mse.ruet.ac.bd)

## **Abstract**

To solve the plastic pollution problem, starch-based bio-composite films can play a vital role in reducing the use of conventional plastic packages. This work provides a comprehensive review on the reinforcements used for the performance enhancement of the conventional starch-based films. To achieve this objective, firstly fillers, additives or strategies for enhancement in mechanical, thermal, barrier, and biodegradation properties are reviewed critically. Later, different fillers and additives which are being used already are compared to find the best option. It is observed that starch-based films are promising for sustainable packaging. However, reinforcements can enhance the properties to make it more suitable for packaging applications.

## **Keywords**

Starch, Reinforcements, Biodegradable, Elongation, WVP (Water Vapor Permeability)

## **1. Introduction**

Every year polyethylene is produced over 140 million tons which leads to massive accumulation of non-recyclable waste in landfills and oceans. A serious threat to ecosystems, wildlife and human health by the entry of microplastics

into the food chain is being posed as plastics reckon for 60-95% of marine debris (Cheng et al., 2021)(Barkoula et al., 2008)(Cozier, 2014) . A non-renewable resource, petroleum is used for the derivation of most synthetic polymers which are not biodegradable and persist in nature for hundreds of years(Xie et al., 2013). A shifting reflection toward sustainable packaging, there is a demand for biodegradable plastics is rising with bio-composites predicted to account for 20% of total plastic.

Starch being emerged as one of the most promising candidates is used with renewable agricultural resources from where bioplastics derivation is done which are gaining global attentions. Sources like- corn, potato, cassava etc. are abundantly available for the production of starch, a natural polysaccharide (Röper and Koch, 1990)(Fonseca-García et al., 2021). For sustainable packaging applications starch being the most suitable as it is inexpensive, biodegradable and has good film-forming ability (Fonseca-García et al., 2021)(Lörcks, 1998). Like traditional plastic films, there also has been a transparency nature in starch-based films (Lörcks, 1998). However, brittleness, poor thermal stability, and high-water absorption are some inherent drawbacks of neat starch films due to their hydrophilic nature (Bodirlau et al., 2013)(Hazrol et al., 2022).

Significant research progress has been made to address the drawbacks of starch-based films and get the balance between mechanical strength, barrier performance, and biodegradability. Investigation of different fillers, plasticizers, and processing methods from many studies, yet a review with a systematic comparison over the types and effects of the reinforcements on film properties can be a useful tool to initiate widespread application of starch as ecofriendly packaging material. Therefore, the impact on performance from the contemporary advances in starch-based bio-composite films by focusing on reinforcement strategies and processing methods is an outline which is the goal of the review.

## 2. Pure Starch Film

Comparing with the reinforced composites pure starch films have disclosed their limited strength and stability by some experiments. The tensile strength of a thermoplastic starch (TPS) film was reported to be 7.12-7.33 MPa, with 53-68% elongation at break (Piyada et al., 2013). However, these values differ largely depending on the test conditions. The values of water vapor permeability (WVP) were generally high starting from  $4.64 \times 10^{-7} \text{ g}\cdot\text{m}^{-1}\cdot\text{h}^{-1}\cdot\text{Pa}^{-1}$  to  $9.61 \times 10^{-7} \text{ g}\cdot\text{m}^{-1}\cdot\text{h}^{-1}\cdot\text{Pa}^{-1}$  in terms of barrier behaviour but depending on formulation and casting method there were some reports which noted lower values such as  $2.91 \times 10^{-11} \text{ g}\cdot\text{m}^{-1}\cdot\text{s}^{-1}\cdot\text{Pa}^{-1}$  and  $1.4\text{--}1.2 \times 10^{-10} \text{ g}\cdot\text{s}^{-1}\cdot\text{m}^{-1}\cdot\text{Pa}^{-1}$ . A major drawback had been found in moisture absorption from water uptake with 48% at 100% RH and up to 70% at 98% RH after 72 hours. A rapid breakdown with glycerol-plasticized TPS losing 70% of weight in 22 days and control films fully degrading within 30 days in soil composting was confirmed from the studies of biodegradation (Barkoula et al., 2008).

## 3. Property Enhancement of Starch Film

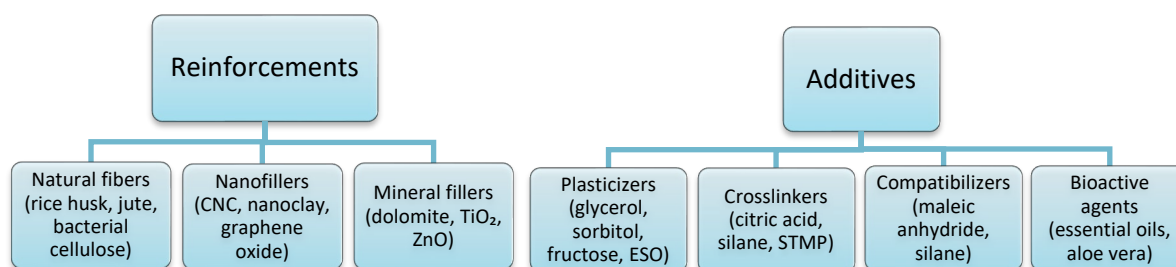


Figure 1. Types of Reinforcements and additives (Lomelí-Ramírez et al., 2023) (Silveira et al., 2020)

When starch is combined with natural fibres, nanoparticles, or other biobased reinforcements, significant improvements in properties have been reported. Types of these materials are illustrated in Figure 1.

By using these reinforcements, enhancement of mechanical strength, thermal stability, and barrier properties as well as maintenance of biodegradability and renewability are done (Bodirlau et al., 2013). Reduction of brittleness and improvement of flexibility are achieved by the plasticizers such as glycerol, sorbitol, and vegetable oils (Mallick et al., 2020). Hence for an equilibrium environmental sustainability along functional performance the development of starch-based bio-composite films provide a pathway to packaging design materials.

Table 1. Enhancement of Tensile Strength (TS), Elongation (EB), Young's Modulus (YM)

References	Additive / Reinforcement	Effects	Remarks
(Teodoro et al., 2015)	Acetylated cassava starch nanoparticles (0.5→ wt%)	TS ↑ (47.05→69.50 MPa); EB ↓ (329→307%) YM ↑ (315.78→828.68 MPa);	FTIR confirmed acetylation; lower crystallinity favorable; excessive loading caused agglomeration
(Gao et al., 2020)	Soybean oil	TS ↑ (5.21 MPa); EB ↑ (36.5%)	Slight barrier ↓
(Liu et al., 2010)	Bamboo cellulose crystals (1–20 wt%)	TS ↑ (2.5→12.8 MPa); EB ↓; YM ↑ (20.4→210.3 MPa); optimum ~8 wt%	Optimum at ~8 wt%; higher loading reduced properties due to poor adhesion
(Piyada et al., 2013)	Rice starch nanocrystals (≤20 wt%)	TS ↑ (7.12→16.43 MPa)	↑ Interfacial bonding; aggregation at >20 wt%
(López et al., 2015)	Talc particles (5 wt%)	TS ↑ (1.19→2.34 MPa); YM ↑ (22.7→38.2 MPa); EB ↓ (62.1→59.0%)	↑ Toughness; ↑ Tear resistance
(Sadegh-Hassani and Mohamma di Nafchi, 2014)	Halloysite nanotubes (5 wt%)	TS ↑ (7.33→9.82 MPa); YM ↑ (188→376 MPa); EB ↓ (68→44%)	Improved adhesion
(Salaberria et al., 2014)	Chitin nanocrystals & nanofibers (5–20 wt%) in TPS	Nanocrystals: TS ↑ (7.1→10.8 MPa); YM ↑ (222→390 MPa); Nanofibers: TS ↑ (9.9→15.0 MPa); EB ↓; YM ↑ (330→520 MPa)	Strong filler–matrix adhesion; network-like structure at 15 wt%
(Silveira et al., 2020)	Tea tree oil (TTO)	TS ↑ (3.03 MPa); EB ↑ (25.1%)	Antimicrobial property
(Mallick et al., 2020)	Rice husk fiber (RH)	TS ↑ (30.7→40.4 MPa); EB ↑ (150→300%)	Barrier property ↓
(Hazrati et al., 2021)	Dioscorea hispida fiber (DHF)	TS ↑ (optimum 6%); YM ↑; EB ↓	Eco-friendly filler
(Soykeabk aew et al., 2012)	Bacterial cellulose (BC)	TS ↑ (58 MPa, 20×); YM ↑ (106×)	High strength & transparent
(Sanyang et al., 2016)	PLA (SPS50-PLA50)	TS ↑ (76%)	Poor adhesion, needs compatibilizer
(Gürler et al., 2021)	PLA + PPS-3APTMS	TS ↑ (10.9 MPa); EB ↑ (21.9%)	Strong bilayer system
(Osman et al., 2021)	Dolomite (sonicated)	TS ↑ (36.7% at 5% loading)	Sonication improves dispersion

(Wu et al., 2019)	Citric acid (10–15%)	TS ↑ (~29%); EB ↑ (~25–31%)	Optimal at 15% plasticizer
(Gürler et al., 2021)	3-APTMS	TS ↑ (41.3%)	Hydrophobicity ↑, biodegradability ↓
(Fonseca-García et al., 2021)	Chitosan + Pluronic F127	TS ↑ (6.49 MPa at 1% then ↓); EB ↑	Best balance at 3% filler
(Mallick et al., 2020)	PVA + RHF + Plasticizers	TS ↑ (25.9 MPa at 10% Fructose); EB ↑ (~38%)	Fructose → best balance
(Tarique et al., 2021)	Glycerol (15–45%)	TS ↓ (9.3 → 1.9 MPa); EB ↑ (47–57%)	Flexibility ↑, strength ↓
(Alvarez-Ramirez et al., 2019)	CaCl <sub>2</sub>	EB ↑ (90 → 160%); TS ↓ (-8%)	Improves elasticity

Table 2. Enhancement of Barrier properties (Water Vapor Permeability (WVP), Solubility, Water Uptake.

References	Additive / Reinforcement	Effects on Barrier	Remarks
(López et al., 2015)	Talc particles (5 wt%)	WVP ↓ 26–54%	Created tortuous pathway for vapor/gas
(Versino et al., 2015)	Cassava bagasse fiber (1.5%)	WVP ↑ 54.13%	Makes the material more permeable to water vapor.
(Mehanny et al., 2016)	Date palm & flax fibers (≤60 wt%)	WVP ↓ Water uptake (48 → 38%)	Lower hydrophilicity of fibers reduced absorption
(Babae et al., 2015)	Kenaf cellulose nanofibers (10 wt%)	$9.61 \times 10^{-7} \rightarrow 8.26 \times 10^{-7} \text{ g/m} \cdot \text{h} \cdot \text{Pa}$	WVTR 7.47 → 6.42 g/m <sup>2</sup> h; acetylation ↑ WVP
(Sanyang et al., 2016)	PLA (SPS50-PLA50)	WVP ↓ 96.7%; WA ↓ 65.9%	Strong barrier
(Lomeli-Ramírez et al., 2023)	CNCs	WVP ↓ (20.7%)	Strong reinforcement
(Gürler et al., 2021)	PLA + PPS-3APTMS	WVP ↓ 55%; Solubility ↓; Uptake ↓ 83%	Very effective
(Gürler et al., 2021)	3-APTMS	WVP ↓ 8.7%; Solubility ↓; Uptake ↓ 51%	Hydrophobic crosslinker
(Wu et al., 2019)	Citric acid (10–15%)	WVP ↓ (19–32%); Swelling ↓ (73–86%)	Best balance at 15% plasticizer
(Fonseca-García et al., 2021)	Chitosan + F127	WVP ↓ ( $21 \times 10^{-11} \rightarrow 2.76 \times 10^{-14}$ ); Solubility ↓ (42.6 → 3.3%)	Strong barrier
(Mallick et al., 2020)	PVA + RHF + Plasticizers	WVP ↑ with concentration	Fructose → lowest WVP
(Tarique et al., 2021)	Glycerol (15–45%)	WVP ↑; Solubility ↑; Moisture ↑	Plasticizer effect
(Mallick et al., 2020)	Rice husk fiber (RH)	WVP ↑; (6.44 → 12.6 g · mm/day · m <sup>2</sup> )	Barrier reduced

### 3.1 Reasons for The Results among The Starch Based Systems

From Tables 1 and 2, improvements with specific additives have been shown evidently by the thermal and functional modifications of starch-based bio-composite films. A bilayer film was fabricated with a melting temperature of 175 °C and enhanced thermal stability by incorporating PLA with PPS-3APTMS (Gürler et al., 2021). Degradation slows down at higher concentrations when glycerol was used as a plasticizer at 15–45% (Tarique et al., 2021), while the onset ( $T_0$ ) and peak ( $T_p$ ) degradation temperatures was strongly reduced in combination with poly(vinyl alcohol) and rice husk fiber (Mallick et al., 2020). Ionic crosslinking through  $\text{CaCl}_2$  improved thermal stability (Alvarez-Ramirez et al., 2019). A strong inhibition with 77% reduction (Figure 2) of *S. aureus* and 65% reduction of *C. albicans* was shown by tea tree oil in terms of antimicrobial activity (Silveira et al., 2020), while aloe vera gel exhibited progressive antimicrobial

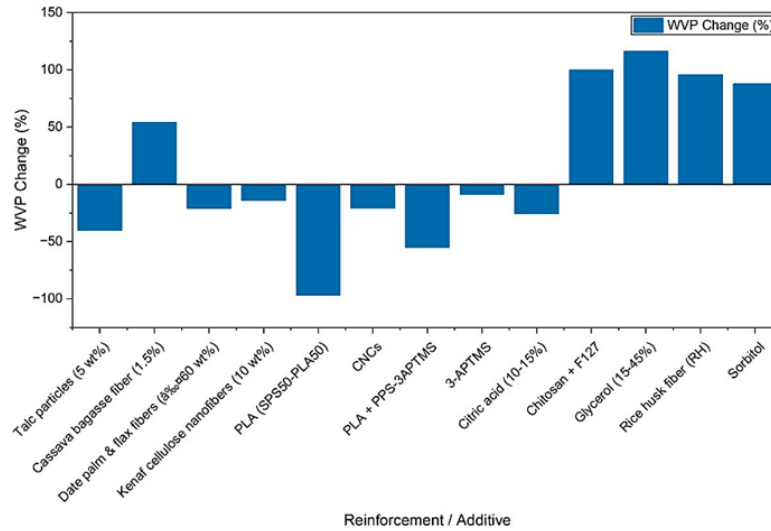


Figure 2. Tensile Strength and Elongation of Reinforced Starch Films (Table-1).

effects, reaching a maximum at 40% loading against *E. coli*, *Klebsiella*, and *Bacillus* (Mallick et al., 2020). Improvement of bioactivity was done by using citric acid at 10–15% concentration, yielding antimicrobial enhancements of 5% against *E. coli* and 11% against *S. aureus*, along with improved barrier properties (Wu et al., 2019). A dense yet flexible films was produced along other functional effects including- ionic conductivity improvement with  $\text{CaCl}_2$ , increasing from  $3 \times 10^{-5}$  to  $7 \times 10^{-5}$  S/cm while reducing crystallinity (Alvarez-Ramirez et al., 2019), and higher crystallinity and film thickness with D-allulose (Ployetchara and Gohtani, 2018).

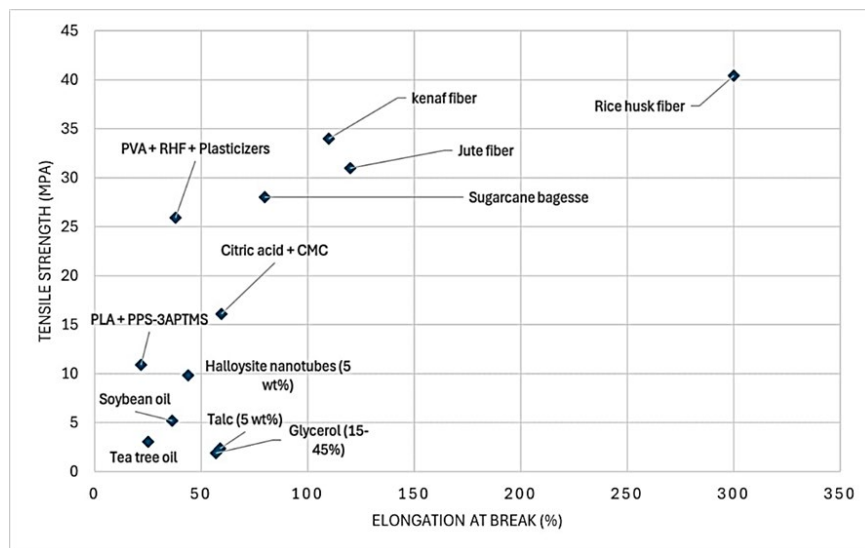


Figure 3. Barrier Property of Reinforced Starch Films (Table-2).

Figure 2 and 3 show the effect of different reinforcements and additives on the mechanical and barrier performance of starch-based films. As seen in Figure 2, most nanofillers and natural fibers enhance tensile strength, although their influence on elongation varies plasticizers and rice husk fiber improve ductility, while rigid fillers like talc or halloysite reduce flexibility despite strengthening. In Figure 3, Water vapor permeability reduces when certain additives like-talc, PLA and citric acid creates a tortuous diffusion paths or crosslinking networks while rice husk fiber tend to increase permeability due to hydrophilic nature. During the design of the starch-based bio-composite films for packaging applications there should be a balance between strength, flexibility, and barrier properties which are found from the highlighted results.

#### 4. Future Outlook

With green surface modifications and advanced processing techniques, filler dispersion and interfacial compatibility can be addressed evidently in future research. It remains a central challenge in achievement of an optimal balance between strength, barrier resistance, and biodegradability. Utilization of abundant agricultural residues such as rice husk or jute fiber and developing low-energy fabrication methods can be considered in future assessments for having scalability and cost-effectiveness. Ultimately, ensuring that starch-based biocomposites are not only effective in the laboratory but also viable and sustainable for industrial packaging applications, prominence should be placed on long-term stability, and life-cycle assessments.

#### 5. Conclusion

After reviewing several studies on the development of starch-based bio-composite films using different reinforcements and additives, the following conclusions can be drawn:

1. Starch-based bio-composite films are promising sustainable alternatives to conventional plastics due to their biodegradability, renewability, and low cost.
2. Pure starch films face inherent downsides such as brittleness, poor thermal stability, and high-water absorption, which limit their direct packaging applications.
3. Reinforcements using natural fibres (rice husk, jute, bacterial cellulose), nanofillers (CNC, nano clay, graphene oxide), and mineral fillers (talc, TiO<sub>2</sub>, ZnO) significantly enhance mechanical strength, thermal resistance, and barrier properties.
4. Plasticizers like glycerol, sorbitol, and epoxidized soybean oil improve flexibility and reduce brittleness, though excessive loading lowers tensile strength.
5. Crosslinkers such as- silanes, calcium ions and citric acid are there from which citric acid (10–15%) improve both mechanical and barrier properties, while also adding antimicrobial benefits, making them highly effective additives.
6. Bilayer and compatibilized systems, especially starch–PLA composites, provide superior adhesion, barrier performance and biodegradability compared to single-component films.

7. Overloading of nanoparticles or fibres leads to agglomeration and property deterioration, highlighting the need for optimized formulation.
8. Reinforced starch-based films combine environmental sustainability with enhanced performance, making them strong candidates for replacing petroleum-based plastics in eco-friendly packaging applications.

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## **Biographies**

**Sabiqune Nahar Niti** is an undergraduate student at Rajshahi University of Engineering and Technology (RUET), currently focusing on sustainable packaging through starch-based materials. She is dedicated to growing up as a researcher and is actively involved in various projects aimed at finding eco-friendly solutions. She is passionate about expanding her knowledge in the research field and aspires to contribute to the development of more sustainable, environmentally friendly materials. With a strong desire to make a positive impact, she is eager to enhance both her academic and practical understanding of the field.

**Maisha Chowdhury Karabi** is an undergraduate student at Rajshahi University of Engineering and Technology (RUET), currently working on starch-based packaging. She has a strong ambition to become more proficient in the research world and is actively involved in various research projects based on sustainability, advanced materials and green source-based energy sectors etc. She is dedicated to advancing her knowledge and skills, aiming to make meaningful contributions to the field of sustainable materials and packaging. With her passion for research, she strives to have a positive impact on the future.

**Shaira Anowara** is an undergraduate student at Rajshahi University of Engineering and Technology (RUET), aspiring to build expertise in sustainable material development. Her academic interests are centered around the use of biodegradable and eco-friendly resources, with a strong focus on innovative packaging solutions. She is actively engaged in research-oriented activities and projects that aim to replace conventional plastics with greener alternatives. With a growing passion for scientific exploration, she strives to expand both her technical skills and research abilities. Shaira hopes to contribute to the advancement of environmentally responsible materials, driven by her motivation to create meaningful, long-lasting impact in the sustainability sector.

**Maliha Rahman** is an Assistant Professor in the Department of Materials Science and Engineering at Rajshahi University of Engineering & Technology (RUET). With a strong research background in biopolymers, natural fiber-reinforced composites, biomaterials, and advanced nanocomposites, she actively contributes to the development of sustainable materials for industrial and biomedical applications. Her work focuses on eco-friendly polymer composites, additive manufacturing (3D printing) using bio-based materials, and structural performance optimization of biomedical devices and implants. She has published research on natural fiber-reinforced polypropylene composites, renewable bio-based thermoplastic polyurethane for FDM 3D printing, and material design for mechanical and biomedical systems. Her research contributions have earned significant citations, reflecting the impact of her work in materials science and sustainable engineering. She continues to advance research that integrates renewable resources with modern manufacturing, aiming to support environmentally responsible innovations in both industrial and medical sectors.