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Recent Advances in Thermoplastic Starch Research: A Review

Ochei Emeka Stephen.[1*], Apeh Solomon Enebi[2], Mrs Fajugbagbe Atinuke[3]

[1*] Nigerian Building and Road Research Institute.

[2] Nigerian building and road research institute.

[3] Nigerian building and road research institute.

*** Correspondence:** Ochei Emeka Stephen

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ABSTRACT: As a sustainable substitute for traditional plastics made from petroleum, thermoplastic starch (TPS), a biodegradable and renewable polymer made from natural starch sources, has attracted a lot of interest. A detailed review of current developments in TPS research is provided in this paper, with an emphasis on material development, techniques for processing, property improvement, and applications. Technological advancements in plasticization techniques, such as the application of new plasticisers and blending agents, have enhanced the mechanical strength, water resistance, and thermal stability of TPS. TPS's potential has been further enhanced by nanocomposites and hybrid materials, which allow for customised qualities for particular industrial uses like packaging, agricultural, and biomedical devices. Furthermore, long-standing issues with TPS's moisture sensitivity and retro-gradation have been resolved by current efforts in interaction and chemical modification. This study focuses on the biodegradability and environmental effects of TPS-based materials, emphasising their function in circular economy initiatives. In order to inform the creation of next-generation biodegradable materials, this study summarises the most recent research on TPS and describes current trends and future directions in the field.

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Introduction

Finding sustainable substitutes for traditional synthetic polymers has become more urgent due to the environmental concerns about plastic pollution and the depletion of fossil fuel resources. Since thermoplastic starch (TPS) is inexpensive, renewable, and biodegradable, it has become a viable option among biopolymers (Diyana *et al*., 2021).

Derived from natural starch sources such as corn, potato, and cassava, TPS is produced by destructuring native starch granules in the presence of plasticizers under heat and shear, resulting in a processable material with thermoplastic behaviour (Guarás *et al*., 2021). Despite its advantages for the environment, TPS's intrinsic drawbacks—such as poor mechanical qualities, high water sensitivity, and ageing from retro-gradation—have historically prevented its widespread use (; Khan *et al*., 2017). Overcoming these obstacles has been the focus of substantial scientific efforts in recent years. Developments in the use of new plasticisers, polymer mixing, and chemical changes have improved TPS's performance and adaptability, boosting its viability for uses in disposable goods, packaging films, and agricultural mulch (Rahardiyan *et al*.,2023, Pérez *et al*., 2010;).

According to Zhang *et al*. (2014), the incorporation of nanoparticles and the production of TPS-based nanocomposites have provided new avenues for enhancing structural and barrier qualities. These developments have ushered in a new era of biodegradable polymer research, together with eco-friendly processing methods and a better comprehension of the physicochemical behaviour of TPS. This review seeks to give a thorough summary of the most recent developments in thermoplastic starch research, with an emphasis on the production of new materials, ways to prepare them, improving existing properties, and developing applications. It also looks into the issues that still need to be resolved and identifies potential avenues for further study and practical use.

Preparation and Properties of Thermoplastic Starch

When native starch undergoes thermomechanical treatment, its granular structure is broken down in the presence of plasticisers to produce thermoplastic starch (TPS), a

biodegradable substance. In order to produce TPS, starch is usually heated and mixed with plasticisers such sorbitol, glycerol, water, or other polyols at high temperatures. This breaks down the hydrogen bonds that hold starch molecules together and produces a molten, processable form (Ma *et al.*, 2021). With temperatures varying from 90°C to 180°C depending on the plasticiser and starch supply employed, this transformation is frequently carried out using extruders, mixers, or internal batch mixers (Parulekar *et al*., 2020). The process ability and ultimate characteristics of TPS are significantly influenced by the kind and concentration of plasticiser used. According to Kargarzadeh *et al.* (2019), glycerol is the most popular because of its high compatibility and efficiency in reducing the glass transition temperature (Tg) of starch. Higher glycerol content, however, typically leads to greater water sensitivity and decreased mechanical strength. It has been demonstrated that adding glycerol to other plasticisers, like citric acid or urea, enhances the TPS films' water resistance and mechanical integrity (Zhang *et al*., 2020). Although partial recrystallisation may happen over time, compromising TPS's stability and mechanical performance, the majority of its structural components are amorphous regions as a result of the starch's gelatinisation and plasticization (Avérous & Halley, 2009). The amylose-to-amylopectin ratio, processing settings, and the starch plant source all have a major impact on the shape of TPS. When compared to low-amylose starch, high-amylose starch often yields TPS with higher tensile strength and thermal stability (Jiang *et al*., 2022).

Biodegradability, renewability, and affordability are just a few of the favourable qualities that make TPS a viable substitute for petroleum-based plastics, particularly in packaging applications. However, its low mechanical qualities, high water sensitivity, and retrogradation limit its performance. In order to get over these restrictions, TPS is frequently mixed with different polymers or undergoes chemical modification to improve its performance characteristics (Chen *et al*., 2020). To strengthen TPS matrices and enhance their barrier qualities, thermal stability, and biodegradability, current research also focusses on the addition of nanofillers and crosslinking agents (Fabra *et al*., 2018).

Reinforcement of Thermoplastic Starch

Many studies have been conducted on reinforcement techniques as a result of thermoplastic starch's (TPS) associated drawbacks, which include low mechanical strength, poor moisture resistance, and limited thermal stability. In the process of integrating different fibres and fillers to TPS, its performance can be greatly enhanced, opening up new uses in the biomedical, agricultural, and packaging industries.

Natural Fiber Reinforcement

Since the hydroxyl groups of starch and lignocellulosic fibres have strong interfacial interactions, adding natural fibres to TPS, such as cellulose, hemp, sisal, or jute, has been demonstrated to improve tensile strength, Young's modulus, and thermal stability (Muller *et al.*, 2017).

For instance, cellulose nanocrystals (CNCs) have drawn a lot of interest because of their high aspect ratio and crystallinity, which give the TPS matrix rigidity and resistance qualities (Nazrin *et al.*, 2021). Aspect ratio, filler content, dispersion, and interfacial adhesion are some of the variables that affect the degree of reinforcement.

Nanomaterials and Hybrid Fillers

Through the production of nanocomposites, nanomaterials including metal oxide nanoparticles, graphene oxide, and montmorillonite (MMT) clay have been used to reinforce TPS. Along with improving TPS films' mechanical and thermal qualities, these materials also enhance their ability to resist and antibacterial qualities (Ma *et al.*, 2020). MMT, in particular, creates a tortuous path for gas molecules, significantly reducing water vapor and oxygen permeability (Liu *et al.,* 2019). Although graphene-based nanofillers provide better reinforcing effects, they frequently need to have their surfaces modified to improve their compatibility with the hydrophilic TPS matrix.

There has also been research into hybrid reinforcing techniques, which mix two or more filler categories. Combining CNCs and MMT, for example, can have synergistic effects where MMT improves the resistance performance and CNCs

provide stiffness (Zhao *et al.,* 2018). One important consideration is the dispersion of nanofillers; inadequate dispersion can result in the accumulation and stress concentration sites, which can impair performance.

Compatibilization and Surface Modification

Several compatibilizers and surface treatments are applied to enhance filler-matrix compatibility. Citric acid, silanes, and maleic anhydride have been utilised to alter the starch backbone or filler surface in order to facilitate hydrogen or covalent relationship between phases (Ibrahim *et al.*, 2021). These changes improve the mechanical characteristics by reducing phase separation and improving the stress transfer between the matrix and filler.

Biodegradability and Environmental Performance

The effect of reinforcement techniques on the biodegradability of TPS is also assessed. Certain fillers, like graphene or specific metal oxides, may inhibit microbial activity, while others, like cellulose and MMT, are biodegradable and even speed up breakdown (Khalil *et al*., 2018). Therefore, it is important to carefully assess how reinforced TPS components would affect the environment, especially when applied to products that can only be used once.

In general, reinforcing is still a key component of TPS development, and multifunctional composites that combine enhanced mechanics, resistance qualities, and biodegradability without sacrificing sustainability are becoming more and more popular.

Blends and Composites of Thermoplastic Starch

In order to address the shortcomings of thermoplastic starch (TPS), including its low mechanical strength, poor water resistance, and high tendency to retrograde, researchers have looked into combining TPS with other synthetic and biodegradable polymers. One of the best ways to enhance TPS's mechanical, thermal, and barrier qualities while maintaining its sustainability and biodegradability is to blend and produce composites.

Blending with Biodegradable Polymers

Performance enhancements have been shown in blends of TPS with biodegradable polymers, including poly (lactic acid) (PLA), polycaprolactone (PCL), and poly (butylene adipate-co-terephthalate) (PBAT). While PLA's natural brittleness can be problematic, PLA/TPS blends, for example, show superior tensile strength and decreased water sensitivity when compared to plain TPS (Wang *et al*., 2020). According to Tang *et al.* (2012), compatibilizers such epoxy-functionalized polymers or maleic anhydride are frequently added to TPS in order to enhance interfacial adhesion with hydrophobic polymers like PLA. PCL's versatility and strong TPS compatibility make it another commonly used companion in TPS blends. It has been demonstrated that PCL and TPS blends exhibit enhanced moisture resistance and elongation at break, rendering them suitable for application in both packaging and agricultural (Yew *et al.,* 2015). Likewise, materials that combine TPS and PBAT balance biodegradability, strength, and flexibility (Müller *et al.,* 2017).

Blending with Synthetic Polymers

Although completely biodegradable mixtures are the best, TPS has also been mixed with traditional synthetic polymers like polyethylene (PE) and polypropylene (PP) to reduce costs and achieve partial degradability. However, because of the polarity mismatch, these mixes frequently have poor miscibility. Reactive extrusion using functionalised polyolefins is one compatibilization technique used to enhance phase dispersion and interfacial contact (Gunawardene *et al*., 2021, Formela *et al*.,2018, Maubane *et al*., 2018). Although these materials are more eco-friendly than pure plastics, their limited biodegradability limits their use in sustainable applications.

TPS-Based Composites

TPS-based composite materials are produced by adding organic or inorganic fillers to the polymer matrix, such as metal oxides, lignin, nanoclays, or natural fibres. These composites provide improved the ability to resist qualities, thermal stability, and mechanical performance. The aromatic character of lignin, for example, has been demonstrated to enhance hydrophobicity and improve UV resistance in TPS-lignin composites (Zhang *et al.,* 2019). Moreover, TPS-based bionanocomposites

containing montmorillonite or halloysite nanotubes show a markedly higher modulus and a much lower water vapour permeability (Jiang *et al*., 2020).

Morphology and Compatibility

The ultimate characteristics of TPS composites and blends are largely determined by component interfacial compatibility, dispersion, and shape. Inadequate mechanical properties arise from phase separation caused by poor interfacial adhesion. Compatibility is thus still a major concern in TPS blending investigations. Many techniques have been investigated to improve compatibility and stability, including reactive blending, in situ compatibilizer synthesis, and coupling agent use (Martinez *et al*., 2022, Palai *et al*., 2019, Ma *et al*., 2016).

All things considered, TPS composites and blends offer a flexible foundation for customising qualities to particular uses while preserving environmental advantages. The functional capacities of these materials are being further expanded by developments in nanotechnology and compatibilizer design.

Surface Modification and Cross-Linking

Thermoplastic starch's (TPS) hydrophilic nature and retro-gradation susceptibility limit its mechanical strength, water resistance, and durability, which in turn affect its performance. Chemical cross-linking and surface modification have become essential methods for overcoming these constraints and enhancing the interfacial stability, environmental resilience, and general usefulness of TPS-based materials.

Surface Modification

Reducing moisture absorption and improving TPS's compatibility with hydrophobic polymers are the main goals of surface modification approaches. Physical techniques like UV irradiation, corona discharge, and plasma treatment have been used to change the surface energy and improve the adhesion characteristics of TPS films (Fazeli *et al.,* 2019). These surface treatments add functional groups (such as carbonyl, hydroxyl, and carboxyl) that improve covalent or hydrogen bonding interactions with other coatings or polymers.

Conversely, chemical surface modification entails coating or grafting TPS with functional or hydrophobic compounds. Alkyl ketene dimer (AKD), fatty acids, and silanes have been utilised to make TPS surfaces more hydrophobic, which greatly lowers water sensitivity and enhances dimensional stability (Wang *et al.,* 2020). In composite applications, silane coupling agents, for instance, can improve interfacial adhesion by forming covalent connections with the hydroxyl groups on starch chains.

Chemical Cross-Linking

Cross-linking reduces molecular mobility and increases mechanical strength, water resistance, and thermal stability by creating covalent or ionic connections between starch chains, improving TPS's integrity and resilience. Citric acid, epichlorohydrin, sodium trimetaphosphate (STMP), and glutaraldehyde are examples of common cross-linking agents (Martínez-Villadiego *et al*., 2024). Citric acid is a bio-based cross-linker that has been extensively researched since it is non-toxic and may esterify hydroxyl groups when heated, creating a more rigid, three-dimensional network (Liu *et al*., 2018). It's important to properly regulate the amount of cross-linking. Excessive cross-linking might cause reduced processability and brittleness. The shelf life of TPS-based materials can be extended and solubility and retrogradation can be considerably decreased with optimal cross-linking (García-Guzmán *et al*., 2022).

Reactive Blending and In Situ Cross-Linking

Reactive extrusion, a recent technique, combines cross-linking and blending in a single step to produce TPS composites with enhanced characteristics without the need for extra processing steps. When multifunctional additives like polycarboxylic acids or isocyanates are used during extrusion, in situ cross-linking facilitates improved dispersion and consistent network development (Jiang *et al.,* 2021). This method has been effective in enhancing the qualities of films, particularly in mixes of TPS and polyester or TPS and lignin.

Applications and Future Outlook

For TPS to be applied to areas that need greater durability, like food packaging, mulch films, and medical scaffolds, surface modification and cross-linking

procedures are essential. To preserve TPS's positive environmental effects, more study is being done on the utilisation of bio-based and green cross-linkers. New developments in UV-induced or enzymatic cross-linking offer promising low-energy substitutes for industrial scalability.

Mechanisms of Improvement

The development of high-performance thermoplastic starch (TPS) materials hinges on understanding and exploiting the mechanisms that govern property enhancement. These mechanisms primarily involve improvements in molecular interactions, phase dispersion, crystallinity control, barrier formation, and degradation resistance. Each enhancement strategy—be it blending, reinforcement, cross-linking, or surface modification—relies on manipulating these fundamental interactions within the starch matrix.

Intermolecular Interactions and Compatibility

A critical mechanism underlying TPS improvement is the **enhancement of intermolecular interactions** between starch molecules and additives or fillers. In native TPS, hydrogen bonding dominates due to the abundance of hydroxyl groups. By introducing compatibilizers such as maleic anhydride, isocyanates, or silanes, these interactions can be tuned to promote stronger hydrogen bonding or covalent linkages between TPS and nonpolar polymers or fillers, leading to improved mechanical properties and phase stability (Jayarathna *et al*., 2022).

In polymer blends, improved compatibility reduces interfacial tension and promotes fine dispersion of phases, which is crucial for achieving uniform mechanical and thermal behavior. For instance, TPS-PLA blends within situ compatibilization demonstrate better stress transfer and reduced microphase separation (Jariyasakoolroj *et al*., 2021).

Crystallinity and Retrogradation Control

The crystalline of TPS plays a central role in determining its mechanical properties, water resistance, and biodegradability. However, high retrogradation tendency—where starch chains re-associate into crystalline regions over time—can lead to

embrittlement and shrinkage (Li *et al*., 2016). Additives such as plasticizers, nanofillers, and cross-linkers reduce chain mobility or disrupt hydrogen bonding, thus **inhibiting retrogradation** and stabilizing the amorphous phase. For example, cross-linking with citric acid forms ester bonds that hinder the realignment of starch chains, improving dimensional stability (Liu *et al*., 2018).

Filler-Matrix Interactions and Stress Transfer

The inclusion of reinforcing agents such as cellulose nanocrystals, clays, or lignin enhances the stiffness and strength of TPS through **load transfer mechanisms**. The degree of stress transfer depends on filler dispersion, aspect ratio, and interfacial adhesion. Well-dispersed fillers increase the effective surface area for interaction with the starch matrix, improving mechanical reinforcement (Son *et al*., 2024). Surface treatments on fillers (e.g., silanization) further improve interfacial bonding, which is essential for maintaining strength during moisture uptake and thermal cycling.

Barrier Formation via Tortuous Pathways

Improved **barrier properties** in TPS composites and blends often arise from the formation of tortuous diffusion pathways. Nanofillers such as montmorillonite or graphene oxide are dispersed in the matrix, obstructing the direct path of water vapor or gas molecules. This mechanism significantly lowers permeability, making TPS suitable for food and agricultural packaging (Ma et al., 2020). The extent of improvement depends on the orientation and aspect ratio of the filler particles, as well as their exfoliation in the matrix.

Hydrophobicity and Water Resistance

Mechanisms that reduce water uptake and improve hydrophobicity are vital for broadening TPS applications. Surface modification using hydrophobic coatings or the incorporation of hydrophobic polymers shifts the balance from polar to non-polar interactions, limiting moisture diffusion (Wang *et al*., 2020). Moreover, cross-linking reduces the number of free hydroxyl groups available for water interaction, further enhancing water resistance.

Degradation and Environmental Performance

While improvement mechanisms often target enhanced durability, their effect on biodegradability is also a key consideration. Reinforcements such as cellulose or lignin do not hinder microbial activity, whereas some nanomaterials like metal oxides may slow degradation. The **balance between performance enhancement and environmental impact** is therefore an essential design criterion in TPS development (Khalil *et al*., 2018).

Challenges and Future Directions

Despite significant progress in the development and application of thermoplastic starch (TPS), several challenges continue to hinder its widespread commercial adoption. These limitations, along with opportunities for future research, are critical considerations in realizing TPS's potential as a sustainable alternative to petroleum-based plastics.

Challenges

1. **Moisture Sensitivity and Poor Water Resistance**

   One of the most significant drawbacks of TPS is its inherent hydrophilicity, which leads to high water absorption, reduced mechanical properties under humid conditions, and dimensional instability (Khan *et al*., 2017). While chemical modifications and blending with hydrophobic materials have been used to mitigate this, fully overcoming water sensitivity without compromising biodegradability remains a challenge.

2. **Mechanical Brittleness and Aging**

   TPS tends to exhibit low tensile strength and poor elongation at break, particularly during long-term storage due to retrogradation—the recrystallization of amylose and amylopectin chains (Si *et al*., 2024). This results in brittleness, shrinkage, and reduced usability, especially in flexible applications such as films and bags.

3. **Compatibility Issues in Blends and Composites**

   Blending TPS with other polymers often leads to **phase separation** due to differences in polarity and thermal properties. While compatibilizers can enhance phase interactions, they add complexity and cost, and their effect can be difficult to control (Moghaddam *et al.*, 2018). This limits the uniformity and reproducibility of TPS-based materials at an industrial scale.

4. **Thermal Instability During Processing**

   Starch degradation begins at relatively low processing temperatures (~180°C), limiting the choice of co-processing polymers and reinforcing agents (Zhang *et al.*, 2014). This narrows the range of composite formulations that can be achieved without thermal decomposition or discoloration.

5. **Inconsistent Biodegradability**

   Although TPS is inherently biodegradable, the rate and completeness of degradation vary widely depending on its formulation, cross-linking degree, and environmental conditions (Yao *et al.*, 2024). Furthermore, the inclusion of synthetic, non-biodegradable polymers in TPS blends can undermine environmental performance.

Future Directions

1. **Green and Functional Additives**

   Future research should focus on the use of **bio-based plasticizers**, **natural compatibilizers**, and **enzymatically derived cross-linkers** to enhance TPS performance while maintaining sustainability. These green additives can reduce environmental impact and align with circular economy principles (Khalil et al., 2018).

2. **Nanotechnology Integration**

   The incorporation of **nanomaterials**, such as cellulose nanocrystals, chitin nanofibers, and graphene oxide, offers substantial opportunities to improve

mechanical, thermal, and barrier properties through high surface area interactions (Jiang et al., 2021). Controlled dispersion and alignment of nanofillers, however, remain key challenges to be addressed.

3. **Reactive Extrusion and In Situ Functionalization**

   Advancements in **reactive extrusion technologies** can simplify processing by enabling simultaneous blending, cross-linking, and functionalization. This approach can streamline production while improving compatibility and property control (Ma et al., 2020).

4. **Lifecycle and End-of-Life Assessment**

   Future TPS research should include comprehensive life cycle assessments (LCAs) to quantify environmental impacts across production, usage, and disposal. Studies on compostability, marine degradation, and microplastic formation will provide essential data for policymakers and industries (Syranidou *et al.*, 2020).

5. **Application-Specific Optimization**

   Rather than seeking a one-size-fits-all solution, future TPS developments should focus on tailored formulations for specific applications—e.g., packaging, agriculture, or biomedical uses. This includes integrating intelligent packaging features (antimicrobial agents, oxygen scavengers) or creating TPS-based hydrogels for drug delivery.

Conclusion

Recent advances in thermoplastic starch (TPS) research have significantly expanded the potential of starch-based materials as sustainable alternatives to conventional petroleum-based plastics. Innovations in plasticization techniques, starch modification, blending with biopolymers, and the incorporation of nano-reinforcements have led to notable improvements in the mechanical, thermal, and barrier properties of TPS. Furthermore, increasing understanding of starch–plasticizer interactions and processing conditions has facilitated the development of

TPS formulations tailored for diverse applications ranging from packaging to biomedical fields.

Despite these advancements, challenges remain in ensuring the long-term stability, moisture sensitivity, and scalability of TPS materials for industrial applications. Continued research is needed to optimize formulations, develop cost-effective processing methods, and enhance the biodegradability profile without compromising performance. Integrating multidisciplinary approaches, including materials science, polymer chemistry, and environmental engineering, will be essential in driving TPS technology forward. As the global push for sustainable materials intensifies, thermoplastic starch stands out as a promising and versatile biopolymer poised to play a critical role in the transition toward a circular and bio-based economy.

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