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Green revolution: Chemical modifications of starch-based materials for sustainable plastic synthesis

Stephanie Yen Nee Kew *

Faculty of Engineering and Life Sciences, Universiti Selangor, 45500, Bestari Jaya, Selangor, Malaysia.

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Abstract

The pursuit of sustainable alternatives has become even more significant given the pressing global crisis associated with conventional petroleum-based plastics. Among the most promising polysaccharides, starch-based synthesized plastics have garnered a lot of attention owing to their abundance, cost-efficiency, and ability to offer a viable solution that complies with the tenets of the Green Revolution. However, their intrinsic shortcomings, such as low thermal stability and tensile strength, have underscored the need for the discovery of various modification strategies. In light of this, the present review offers insights into several types of chemical modifications, their effects on plastic properties, and the overall effectiveness of starch-based plastics as ecologically sound polymers. Through a systematic analysis of relevant literature, findings reveal that the properties of starch-based polymers are distinctly impacted by each modification method, showing varying efficacy. A feasible technique for achieving a high water absorption index in plastics may be enzymatic hydrolysis, but acidic modification could be better suited for improved moisture content and solubility. In terms of tensile strength, alkaline modification appears favorable; nonetheless, determining the optimal concentration is essential for attaining the highest tensile strength. Therefore, understanding the influences of these chemical modifications is vital to optimizing the development of greener plastics in materials science. Emerging approaches to modification were also laid out to revolutionize plastic properties by exploring new enzymes or alternative alkaline substances, fine-tuning process parameters, and innovating additive combinations, all of which are necessary for advancing the utility and practicality of modified plastics in diverse applications.

Keywords: Molecular Modification; Polysaccharide; Sustainable; Polymer; Fabrication

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1. Introduction

In recent years, innovative strategies have been developed in the endeavor to search for sustainable substitutes for traditional petroleum-based plastics, such as the consumption of natural waste. This is attributed to the rising awareness of plastic pollution and the rapidly increasing waste build-up that conventional plastics pose. In 2016, the aggregate manufacturing of plastic products was estimated to be 3.35×10^8 tons, exhibiting approximately 4% of the yearly inflation rate [1]. If the post-use generated plastic waste is not properly disposed of, it may likely exert several adverse effects on the environment [2]. Hence, the adoption of available raw materials like banana peels has emerged as a sustainable approach to minimize the utilization of resources and environmental discharges.

It is renowned that starch-based materials are among the most prominent crops in the world at large such as banana, tapioca, wheat, corn, and potatoes. Due to its abundant storage on Earth [3], low cost [4], and high amount of cellulose content to be processed into green plastics, these starch-based materials could be a potential material for future use in plastic synthesis. Figure 1(a) and (b) illustrate the synthesized plastics using starch-based materials, specifically banana peels. Generally, the rich fibrous content of starch-based materials and their polymeric composition can potentially

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[*] Corresponding author: Stephanie Yen Nee Kew

contribute to the overall longevity and durability of the synthesized plastic. The prospect of using starch-based materials to fabricate plastic is in line with the global movements of the Green Revolution in the efforts to alleviate climate change and lessen environmental destruction.

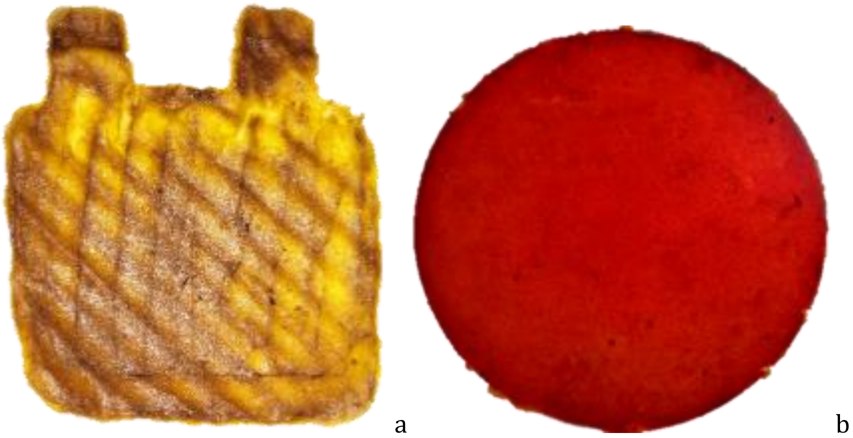

Figure 1 Green plastics using banana peels as starch-based materials in (a) molded plastic form and (b) film form.

The process of converting starch-based materials into decomposable polymers involves chemical modifications. For the extraction of cellulose and other constituents from banana peels, main chemical treatments such as alkaline acid and enzymatic hydrolysis can be employed [5]. These treatments can render the starch-based materials to be more favorable for plastic synthesis by altering their structure. In general, the alkaline solutions such as sodium hydroxide (NaOH) used are responsible for disintegrating the complex polysaccharides, namely lignin and hemicellulose to break open the peel's structure for easier extraction of cellulose. Subsequently, the rest of the constituents are further broken down by acid treatment, which is frequently sulfuric acid (H_2SO_4). In this context, the concentrated acid can help increase the purity of the cellulose product and generate enhanced stability of the nanofiber aqueous suspension [6]. Meanwhile, enzymatic hydrolysis involves cellulose molecules being selectively targeted and broken down into monomers.

Within the scope of achieving optimized and improved quality in the synthesis of green polymers from starch-based materials, an understanding of the chemical modifications is required. This is because although starch-based plastics have potentially become an appealing solution, their inherent drawbacks, such as low tensile strength and thermal stability, limit their broad application and use. Additionally, a gap exists in the extant literature that examines the many chemical processes utilized to modify starch-based materials for optimal green plastic synthesis despite its widespread application, particularly in packaging [7]-[9]. Therefore, this systematic review provides a thorough overview of the various chemical modifications adopted in synthesizing sustainable starch-based plastics to achieve the desired results, highlighting the different chemical treatment reaction mechanisms, their effects on plastic properties, and their effectiveness in plastic synthesis. Overall, the present review aims to promote the development of green materials and facilitate the shift towards a more environmentally friendly plastics sector.

2. Material and Methods

The present systematic review was conducted to identify the findings of relevant articles on various chemical modifications in the synthesis of plastic utilizing starch-based materials. This review generally adopts an exhaustive literature search strategy, rigorous inclusion and exclusion criteria, and extensive data extraction methodologies (Figure 2).

2.1. Literature Search Strategy

The data collection process began with a search strategy across multiple databases, which include Google Scholar, PubMed, Scopus, and Web of Science, to ensure a comprehensive coverage of the literature. During the search, several relevant keywords, such as chemical modifications, banana peels, plastic, synthesis, and its variations were used. Grey literature sources were also screened to search for additional articles that met the objectives of this systematic review. As a result, 300 records were yielded.

2.2. Study Selection Process

This stage involved a screening process where the titles and abstracts of searched studies were screened according to the predefined criteria, including inclusion and exclusion criteria. The importance of applying these predetermined criteria is to establish full-text review eligibility. Articles that have relevant data, such as those reporting on the chemical modification method employed for synthesizing plastic using banana peels, are included. In addition, to ensure that the systematic review reflects the recent state of study in this field, the inclusion of articles must be within the past 10 years in terms of the publication date. This eventually narrowed the selection down to a total of 25 articles. The exclusion criteria, on the other hand, encompassed articles that were out of scope, not written in English, had no full-text availability, exhibited insufficient details, did not explicitly report information related to chemical modifications, or lacked clarity in the reporting of relevant outcomes, thus excluding 274 articles. Every remaining article was carefully reviewed for data relevancy, retaining the sum of 25 entries. In the final dataset, after eliminating duplicate studies, only 6 records were subjected to in-depth analysis.

2.3. Data Extraction

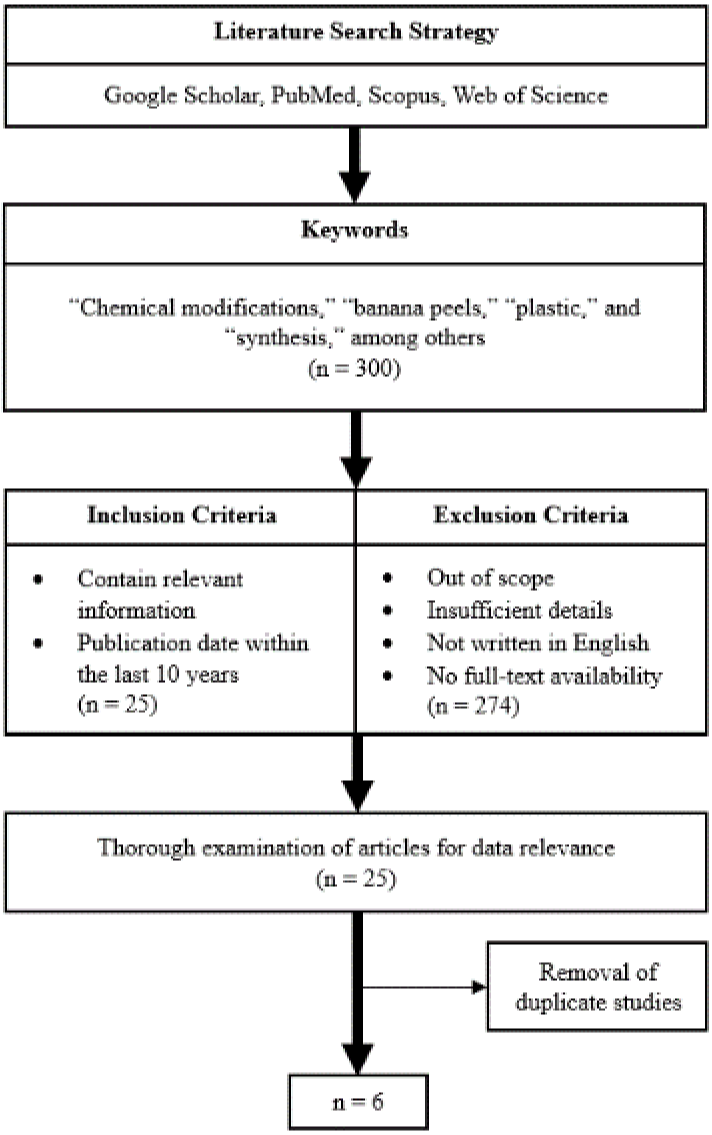

Figure 2 Data collection process

A structured technique was employed during data extraction from the included articles. For the retrieval of specific information, a standardized data extraction format was followed, to systematically gather and record information derived from each piece of literature. The details include: a) chemical modifications on banana peels, including chemicals used, chemical treatment method, and key process parameters, b) results associated with the properties of the synthesized sustainable plastics, and c) effectiveness of different chemical modifications. This meticulous data extraction procedure involves an in-depth examination of each included article, essential to ensure that all pertinent data were captured, allowing for a thorough and reliable review of the evaluation of chemical modifications of banana peels for the production of sustainable plastic. The gathered extracted data were then organized to facilitate the synthesis of findings, which is required for analysis.

3. Results and Discussion

Chemical modifications are processes that involve specific alterations to the functional groups, which ultimately lead to a substantial effect on the properties of the compound, in this case, the starch-based materials. These modifications can be achieved through a variety of techniques, namely enzymatic hydrolysis [10], acidic [11] or alkaline treatments [12]-[13], and the addition of plasticizers [14]. In this context, each of these approaches may contribute to the plastic with varying unique properties and effectiveness. Hence, understanding the impact of the various chemical treatments on the starch structure allows for the optimization of plastic formulations to meet specific uses, like packaging.

3.1. Types of Chemical Modifications

Enzymatic hydrolysis is a process that involves enzymes that facilitate the conversion of complex molecules into smaller units, specifically to break down the α-1, 4 bonds between two glucose units [15]. This is essential as simpler compounds can aid in providing the synthesized plastic with the desired qualities and attributes. Figure 3 shows the hydrolysis of starch by amylase. With relevance to this, the study of Ghizdareanu et al., [10] revealed the chemical modification of enzymatic hydrolysis using α-amylase. It is believed that α-amylase is efficient and can function well at fairly mild conditions, thereby making it more suitable for a range of industrial uses, including the synthesis of green plastics.

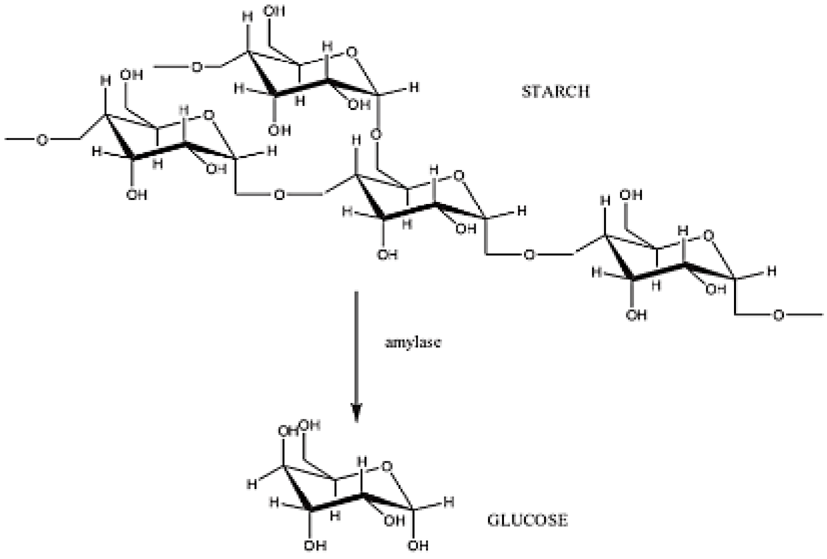

Figure 3 Enzymatic hydrolysis of starch [16].

Since starch-based plastic typically has low tensile strength and may easily deteriorate when exposed to water, modification with acid is therefore crucial [17]. In the synthesis of plastic with starch-based materials, the process of acidic modification refers to the addition of acids to alter the properties of starch. As demonstrated by Oluwasina et al., [11], dialdehyde starch and silica solutions were utilized in the acidic modification of green plastics because these acids can modify the surface topology of the plastic films. Particularly, dialdehyde starch serves as a valuable cross-linking agent (Figure 4) and helps in film-forming properties, while silica particles present in the silica solution act as reinforcing fillers and play a significant role in modifying the surface properties of the green plastics to increase the degree of compatibility with other materials.

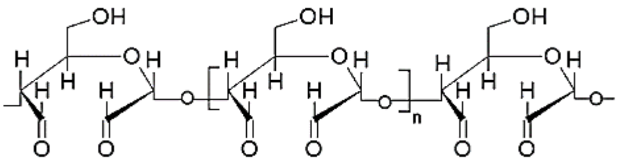

Figure 4 Structure of dialdehyde starch [18].

Alkaline treatment, on the other hand, makes use of alkaline substances that can potentially disrupt the hydrogen (-H) bonds between starch molecules and break the crystalline regions of the starch granules [19]. Hence, alkaline-treated starch-based materials possessed higher amounts of OH groups compared to untreated materials [20]. With regard to this, the finding by Narayanan et al., [12] described the utilization of sodium hydroxide (NaOH) to treat the starch-based materials which create a reinforcement effect. Furthermore, the presence of OH groups resulting from the addition of NaOH can essentially promote the chemical bridging and mechanical interlocking of polypropylene (Figure 5), thus making it a viable option in the production of green plastic products. Similarly, Qin et al., [13] also discussed the alkaline treatment using NaOH. When the starch-based materials were treated with 2% NaOH treatment, the starch molecules typically underwent an orderly rearrangement with the formation of an Eh-type crystal structure with an enlarged space of a single helix structure, which causes recrystallization to slow down. Meanwhile, during the 10% NaOH treatment, the hydroxyl groups would tend to oxidize, leading to the formation of poly-carbonyl structure and the initiation of cross-linking and degradation within the starch molecule chains.

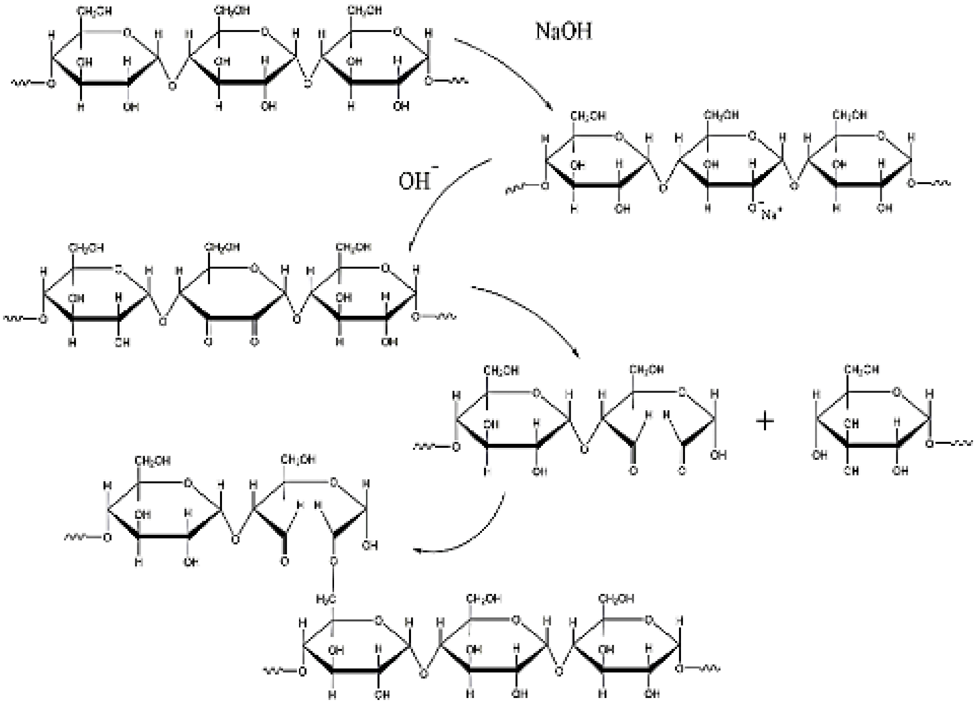

Figure 5 Alkaline treatment on starch-based material [13].

Another technique for chemical modification is employing additives, such as plasticizers. The plasticizer effect can generally lead to an increase in the molecular mobility, interstitial volume of the polymer matrix, and overall hydrophilicity of green plastics [21]. Through the mixing of plasticizers with starch-based materials for green plastic synthesis, the properties of the material can be tailored according to the specific requirements for various purposes. A recent study by Chapain et al., [14]) stated the use of different plasticizers, such as urea, glucose, glycerol, and distilled water. The hydrogen bonds formed between the starch and other components are mainly contributed by the added urea. Glucose, alike urea, encourages cross-link formation and hydrogen bonding network in the green plastic matrix. Meanwhile, distilled water facilitates the formation of the cohesive plastic structure, while glycerol aids in lowering the glass transition temperature, which is vital for easier processing of green plastic fabrication. Similarly, Wahyuningtiyas and Suryanto, [22] also evaluated the effects of glycerol as a plasticizer but this study focused on varying concentrations from 0% to 3%, which significantly impacted the way compounds combine, disintegrate, and alter during the plastic synthesis process.

Table 1 Types of chemical modifications.

References	Types of chemical modifications	Chemicals used	Process parameters	Reaction mechanisms
Ghizdareanu et al., [10]	Enzymatic hydrolysis	α-amylase	Enzyme to substrate ratio is 357 U/g; corn starch to water ratio is 1:2.8; incubation temperature is 48°C	Starch molecules are broken down into monomers, smaller units, which helps strengthen the intermolecular bonds between molecules and increase chain flexibility
Oluwasina et al., [11]	Acidic modification	Dialdehyde starch; silica solution	Different concentrations from 60 to 100% for both dialdehyde starch and silica solutions	Interaction between dialdehyde starch solution and starch matrix
Narayanan et al., [12]	Alkaline modification	Sodium hydroxide (NaOH)	Double steps compounding method; injection molding process (210°C, 30 sec of residence time); various concentrations at 0, 10, 20, 30, and 40 wt%	NaOH introduced reinforcement effects that enhance the matrix-filler interaction via chemical bridging and mechanical interlocking
Qin et al., [13]	Alkaline modification	Sodium hydroxide (NaOH)	Melt extrusion; various concentrations of NaOH ranged from 2% to 10%	2% NaOH treatment: formation of Eh-type crystal structure and a slowed down recrystallization; 10% NaOH treatment: oxidization of hydroxyl groups led to poly-carbonyl structure as well as cross-linking and degradation
Chapain et al., [14]	Additive modification	Plasticizers, such as glycerol, distilled water, glucose, urea	Functional group analysis conducted at ambient temperature	Formation of hydrogen bonding between N-H urea and starch
Wahyuningtiyas and Suryanto, [22]	Additive modification	Plasticizers, such as glycerol	Various glycerol concentrations at 0, 2, 2.5, and 3%	Combination, decomposition, and transformation of compounds

3.2. Effects of Chemical Modifications on Plastic Properties

Different chemical modifications have been analyzed to evaluate their effects on the green plastic properties. Enzymatic hydrolysis, as studied by Ghizdareanu et al., [10] indicated green plastics with higher transparency, increased solidity, and compacity due to enhanced intermolecular bonding in the polymer matrix. This hydrolysis method can also improve the hydrophobicity of green plastics because there is an increase in the formation of new hydrophobic bonds after the molecular structure changes. Moreover, the capacity of the plastics to dissipate energy under stress can potentially be enhanced due to the resilient structure formed within the synthesized plastic material. This in turn led to better mechanical properties. The use of plasticizers as another strategy of chemical modification, as found by Chapain et al., [14] can produce plastics with higher tensile strength and water absorption ability. In this relation, the enhanced tensile strength is contributed to by the increased mobility of the polymer chains, enabling the plastics to effectively withstand stress and deformation without breaking. In principle, molecules from plasticizers tend to intercalate between the chains of polymer, which causes the volume within the matrix to expand. This volume expansion in turn creates more spaces, allowing water molecules to penetrate easily, thus greater water absorption. Conversely, the finding of Wahyuningtiyas and Suryanto, [22] emphasized the effects of a range of glycerol concentrations, observing that the resultant plastics had a higher moisture content, better decomposition, and longer lifespans. These outcomes generally indicate that concentration is an important factor in defining plastic properties.

Meanwhile, Oluwasina et al., [11] determined that acidic treatment can improve the tensile strength, while simultaneously increasing their film solubility and moisture content, rendering them to become more susceptible to degradation in diverse environments. This higher capacity for moisture absorption is likely due to the increased porous structure of the polymer matrix caused by the presence of silica particles, while dialdehyde starch plays a major role in improving tensile strength by forming additional chemical bonds in the matrix. Furthermore, Narayanan et al., [12] observed that the tensile strength of green plastics can be increased by alkaline treatment. The increased tensile strength is highly likely to be influenced by changes in the plastic’s internal structure and surface properties due to the added alkaline, thereby creating a stronger bond between its polymeric matrix and any reinforcing fibers. The finished product would be a more durable and resilient green plastic, as the material is capable of tolerating greater tensile stresses without breaking. Nevertheless, Qin et al., [13] pointed out that while alkaline modification may have imparted on the tensile strength properties, increasing NaOH concentrations can decrease the tensile strength and, at the same time, increase elongation at break. The reduced tensile strength is attributed to the excessive cross-linking and degradation, which eventually weaken the structural integrity of the plastic.

Table 2 Effects of various chemical modifications on plastic properties.

References	**Types of chemical modifications**	**Plastic properties**
Ghizdareanu et al., [10]	Enzymatic hydrolysis	More transparent; more solid and compact; enhanced hydrophobicity; better energy dissipation properties, increased mechanical properties
Oluwasina et al., [11]	Acidic modification	Higher moisture content; greater film solubility, higher tensile strength, longer degradability
Narayanan et al., [12]	Alkaline modification	Increased tensile strength
Qin et al., [13]	Alkaline modification	When NaOH concentration increases: reduced tensile strength, increased elongation at break
Chapain et al., [14]	Additive modification	Higher water absorption, higher tensile strength
Wahyuningtiyas and Suryanto, [22]	Additive modification	Increased moisture content, improved degradation, extended shelf-life

3.3. Effectiveness of Different Chemical Modification Methods

Based on the specific process adopted, the effectiveness of various chemical modification techniques on the synthesized green plastics can vary from each other. In comparison to the control films, Ghizdareanu et al., [10] showed that the hydrolyzed films have a greater water absorption index, indicating better hydrophilicity. Additionally, the higher plastic transparency is evident from the high light transmission and contact angle. Despite its effectiveness, during the enzymatic hydrolysis process, it is advisable to incorporate modifiers, for example, cross-linking agents or surfactants to improve the properties of the produced films further. To add on, surface modifications, including chemical grafting and plasma treatment, could be effective means of modifying starch-based film characteristics for a wide range of industries by making the enzyme more accessible to the substrate. The study of Chapain et al., [14] explained that chemical modification using plasticizers, allows the plastics to effectively absorb water that lasts for 4 days, without decay, indicating the plastic is water-resistant. In addition to that, the tensile strength of 2.3 KPa signifies that approximately 2.3 kg can be supported by plastic, making it suitable for lightweight support only. To further increase the tensile strength, and reduce water absorption, it is encouraged to add reinforcing fillers, for instance, fibers, or to adjust processing parameters, in terms of pressure and temperature, to achieve thorough mixing and dispersion of the plasticizer in the starch-based polymer matrix. On the other hand, the focus of Wahyuningtiyas and Suryanto, [22] on the glycerol effect reported that increased glycerol levels can lead to higher uptake of moisture, prolonged shelf-life, and total breakdown by the 9th day. This rapid degradation indicates that the synthesized plastic is effective and useful in lowering pollution levels, as it can decompose while leaving no harmful residues behind. Indeed, the concentration of glycerol has a major impact on determining the plastic's performance; hence, optimize the plasticizer content accordingly for specific purposes. With every aspect considered, each chemical modification approach has unique benefits and may be customized to meet specific requirements when developing sustainable green plastics.

Acidic modification, as presented by Oluwasina et al., [11], revealed that compared to silica-treated films, films treated with dialdehyde solution exhibit superior compatibility and miscibility. This leads to enhanced solubility, increased moisture content, and stronger tensile strength. The greater effectiveness observed in aldehyde-treated films may result from the cross-linking of starch molecules by aldehyde groups, yielding a blend that is more compatible and homogenous. To further improve the compatibility of the additives with the starch matrix, better mixing methods or the incorporation of compatibilizers can be employed. These techniques not only increase the homogeneity of the synthesized plastic sheets but also their overall strength. In alkaline modification, Narayanan et al., [12] found a significant increase in tensile strength when the film was treated with 30 wt% NaOH. This outcome suggests that 30 wt% could be the optimal NaOH concentration for the optimal performance characteristics in the synthesized plastics. Hence, compared to polymers treated with lesser or greater concentrations, those exposed to 30 wt% NaOH are inclined to exhibit excellent mechanical qualities. However, Qin et al., [13] emphasized that tensile strength generally varied according to different NaOH concentrations. The elongation at break, on the contrary, increases with a decrease in NaOH concentration. Higher NaOH concentrations often induce significant modifications, whereas less extensive changes occur when a lower concentration is used. Principally, the ideal NaOH concentration for synthesizing starch-based polymers with desired properties differs depending on the specific application.

Table 3 Effectiveness of different chemical modification methods.

References	**Types of chemical modifications**	**Effectiveness**
Ghizdareanu et al., [10]	Enzymatic hydrolysis	Hydrolyzed corn starch films had a light transmission of 78.5 ± 0.121% per mm compared to the control film; a higher water absorption index of 2.32 ± 0.112% in hydrolyzed starch films compared to the control film (0.81 ± 0.352%); hydrolyzed starch films exhibited higher contact angle of 79.21 ± 0.171°
Oluwasina et al., [11]	Acidic modification	Better compatibility and miscibility were observed in the interaction between the starch matrix and the dialdehyde solution compared to the interaction between the starch matrix and silica solution; the dialdehyde synthesized starch films had higher moisture content (6.62-11.85%), improved solubility (4.23-7.90%) and greater tensile strength (1.63-3.06 MPa) in comparison to those synthesized with silica solution
Narayanan et al., [12]	Alkaline modification	The treated composite had higher tensile strength by approximately +45.20% with 30 wt% in comparison to the control sample
Qin et al., [13]	Alkaline modification	At 2% NaOH treatment: tensile strength was 10.03 MPa and elongation at break was 40%; at 10% NaOH treatment: increased molecular weight to 4.97×10^5 g·mol^{-1}
Chapain et al., [14]	Additive modification	Water absorption uptake reached up to 4 days without decaying; the load test indicated a high tensile strength of 2.3 KPa
Wahyuningtiyas and Suryanto, [22]	Additive modification	At higher amounts of glycerol: greater moisture absorption, longer shelf-life, and complete degradation happened on the 9th day

3.4. Future Modification Techniques

Future chemical modification methods geared towards optimizing green plastics' characteristics may expand upon and refine present chemical alterations. This can be achieved by seeking new chemical interactions and enhancing the process parameters. Enzymatic hydrolysis, which already offers advantageous plastic properties in terms of solidity, transparency, and hydrophobicity [10] can be further enhanced by exploring different enzymes, for instance, proteases, cellulases, and lipases instead of α-amylase. Fine-tuning the concentration levels of acids in incremental steps within acidic modification is known to particularly improve tensile strength, solubility, moisture content, and degradability [11], which could aid in better balancing these attributes. Moreover, integrating several acids in certain ratios may further maximize the plastic's traits for the intended use thanks to the synergistic effects yielded. While NaOH has been a common substance used in alkaline modification [12]-[13], discovering alternative alkaline chemicals, such as calcium hydroxide ($Ca(OH)_2$) or potassium hydroxide (KOH) might bring about fresh effects on reinforcement in plastic remodeling and expand the variety of modification options. Compared to NaOH, KOH may be beneficial in forming stronger bonds inside the plastic matrix for greater tensile strength. Yet, $Ca(OH)_2$ could contribute to superior resistance to environmental variables and durability since it is less soluble than NaOH. In the context of additive modification, which largely relies on glycerol [14, 22], the prospects for developing new plasticizers with higher flexibility and resistance to impact while preserving other qualities are encouraging. Likewise, lifespan and general performance can be further boosted by the synergies of mixing additives like stabilizers and fillers. Overall, the development of new additives or creative combinations of the current ones could broaden the spectrum of desirable features of green plastic.

4. Conclusion

This study highlights that the synthesis of plastics through the chemical modification of starch-based materials holds potential, as it enables the production of effective polymers that contribute to environmental sustainability. Moreover, the properties of starch-based green plastics were demonstrated to be significantly and uniquely modified by various chemical treatments, from enzymatic hydrolysis that offers enhanced transparency to acidic modifications that provide better film solubility and moisture content. Alkaline concentration, while exhibiting an interconnected interplay with NaOH, may either improve or reduce tensile strength depending on the concentration utilized and specific applications. Collectively, these approaches help develop more sustainable plastics, alleviating the adverse effects of conventional plastics. As the exploration of new modification techniques continues, a cleaner and greener world draws nearer, with plastics no longer being a cause of pollution.

Compliance with ethical standards

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Disclosure of conflict of interest

No conflict of interest to be disclosed.

Statement of informed consent

Informed consent was obtained from all individual included in the study.

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Author's short biography

Stephanie Kew Yen Nee is a young researcher with broad scientific knowledge. Her research interests span chemical elements, chemical reactions, and chemical bonding. Stephanie holds a strong appreciation for Mendeleev's Periodic Table, reflecting her deep passion for the fundamental aspects of chemistry. She is also a member of Commonwealth Chemistry, the Federation of Chemical Sciences Societies, at Cambridge, Cambridgeshire, United Kingdom. Her active participation in intervarsity symposiums, conferences, and competitions has showcased her research and expanded her professional network.