

# Modification and Application of Starch Acquired by Industrial Waste

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**Abstract:** Fruits are a staple of many diets and are consumed in large quantities. Additionally, the world population is expanding and dietary changes have increased demand for this product, which has led to an increase in waste. In addition, a large amount of waste is produced during the processing of fruit, including juice processing, pulp extraction, jams, and frozen pulp. Skin, pulp, seed debris, and pomace are examples of wastes that are rich in substances with added value, such as starch. One of the most prevalent biodegradable polymers in nature, starch finds extensive application in the food and beverage, bioplastic, paper, textile, and biofuel industries. Because of its low toxicity, facile synthesis, relative abundance, and biodegradability, starch has drawn a lot of interest. Starch is made up of amylose and amylopectin. However, because of its increased water absorptivity and poor thermo-mechanical qualities, natural starch cannot be employed directly. As a result, native starch requires modification prior to usage. The main methods for modifying starch are enzymatic, chemical, and physical. Chemical alteration techniques are among the most widely used in enterprises. The chemical starch modification approaches as well as the physical, enzymatic that have been developed in the last few years are covered in detail in this study. Furthermore, the present-day uses of modified starch have been examined in the domains of superabsorbent, packaging, adhesives, medicines, and widely used in food industries.

**Keywords:** Amylopectin, Amylose, Enzymatic modification, Modification, Starch.

## I. INTRODUCTION

Rising worldwide population has resulted in increased food processing demand, as well as a rise in food processing waste. According to a recent assessment by the Food and Agriculture Organisation (FAO), around 1.3 billion tonnes of food are

wasted each year. [32] Food waste is generated by food companies and post-harvest agro-processing, which constitutes a substantial environmental danger both before and after market. These wastes are often disposed of in landfills or used to make compost. However, developing value-added items from these food wastes and turning them into economically viable goods would not only boost waste recovery but would also result in higher profits [1].

The Food and Agriculture Organisation (FAO) defines waste as “food losses of quality and quantity by the process of the supply chain taking place at production, post-harvest, and processing stages”. Food waste is a factor in the projected increase in global population to over 9.69 billion people by 2050 [33]. As a result, providing an appropriate supply of food to a rapidly rising population presents a global challenge. As a result, in recent years, there has been a growing interest in study into food waste reduction and valorisation. Furthermore, raising awareness about food waste is regarded as a political responsibility around the world. It is estimated that the manufacturing and processing of fruits and vegetables will contribute 14.8% of all food waste produced; additionally, in communities, fruit and vegetables make up almost 50% of all food waste. These wastes should not be disposed of; doing so could have negative effects on the environment as well as the food and farming sectors. In this context, studies to transform such waste into high-value products have been suggested [2].

The search for new starches that do not compete with human populations’ food supplies and the growing emphasis on starches generated from wastes, particularly fruit waste, are the two factors contributing to the industry’s increased demand for starch. Fruit starches have a variety of morphologies, structures, and characteristics, which influence their physical properties and, as a result, utility. This enables us to supply the dynamic food as well as non-food industries with starches that have special properties [2].

Amylum, another name for starch, is a carbohydrate that exists in plants. All around the world, it is a significant source of nourishment for humans. It serves as both a necessary dietary item and a biomaterial that is used globally for many purposes. Grain and cereals, rhizomes, seeds, tubers, and roots are among the major sources of starch for living things. Because different plant species have varied glucose chain lengths and amylose/amylopectin ratios, starches originating from diverse plant sources have distinct compositions and structures. Similar variations can be found in the fat and protein composition of storage tissues. It consists of highly stretched amylopectin chains around roughly 65-75% and straight amylose chains, which are 24-36% in proportion. The high concentration of acidified phosphate groups in vegetable starches results in increased viscoelastic power and consistency [3].

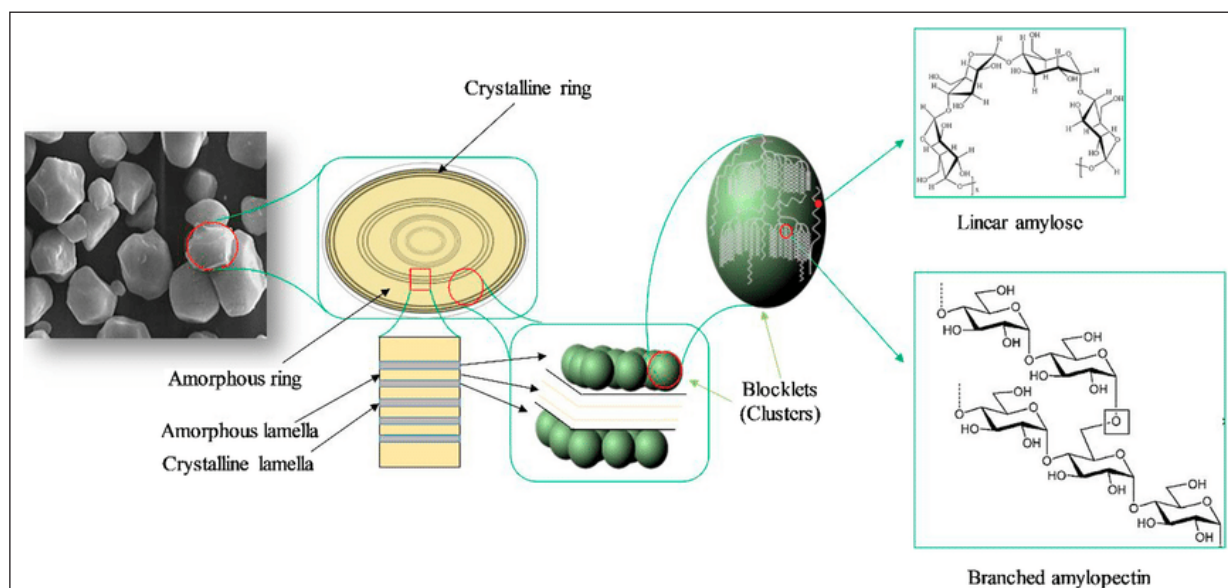
The starch is often used in its unprocessed form in industry and is rarely consumed in its entirety due to its sensitivity to pH, and fluctuations in temperature. A significant number of phospholipids are present in oat starches, while tubers were discovered to be abundant in phosphorus due to their esterified structure. To the best of our knowledge, nevertheless, no prior research has examined the value-adding of fruit waste for the recovery of starch. As a result, this review offers substitutes for starch recovery that can be used to increase the value of fruit wastes. This strategy might benefit the environment and the economy. The primary fruit wastes that are considered viable sources of starch are outlined in this review, along with information on its isolation, content, structures, qualities, and uses [2].

## II. STRUCTURE OF STARCH

Considering the variety of starch granule shapes, it is remarkable to find that the internal structural components of the plants are distributed equally, ignoring the organs of the plants, such as the roots, endosperm, stems, and other sections. Under an optical microscope, a “Maltese cross” with arms extending from the hilum may be seen; this is thought to be the catalyst for granule development. This pattern of birefringence suggests an elevated level of order since it indicates that the particles, or at least a significant fraction of the molecules, which are arranged radially inside the granules [34].

### *Amylopectin and Amylose*

Amylose is the small, straight or slightly branched part of starch. Even though bifurcation amylose molecules often have shorter chains than linear amylose molecules, the molecules are frequently larger. Amylose’s molecular size differs throughout starches as well. The main, highly branched component of starch is amylopectin. Compared to amylose, amylopectin is substantially larger. The weight of the typical molecular weight (Mw) is estimated by physicochemical techniques such light scattering, viscometry, and ultracentrifugation to be between 107 and 108 Da, but the amount of mean values (Mn) are only 105 to 106 Da. Consequently, the index of polydispersity (Mw/Mn) is extremely high. Three size-fractions with estimated average DPn-ranges of 800-2200, 4300-8300 and 12400-25500 are discovered in amylopectin from various sources, with the large molecules typically predominating [3].



Source: Yu *et al.*, 2021 [35]

Fig. 1: A Schematic Representation of Starch Granules

### III. STARCH ISOLATION

With only slight adjustments, starch was extracted through extraction from several sources. 100 cc of distilled water or an alkaline solution was mixed with 5 grammes of the sample, allowed to soak for 6 or 8 hours at room temperature, and stirred continuously. The mixture was strain through a stainless steel sieve with a mesh size of 75, and any leftover material was rinsed three times using deionized water. After mixing

the filtrates, they precipitated at 4 °C for the entire night. After discarding the supernatant, distilled water was used to purify the crude starch. This process was carried out three times, and the starch cake was oven-dried for twenty-four hours at 40 °C. A pestle and mortar were used to grind the starch. Before being used, the starches were stored at room temperature in a plastic bag. Optimal conditions with a 6-hour soaking time were determined by considering the yield [4].



Fig. 2: Process of Isolation of Starch

### IV. MODIFICATION OF STARCH

While starch is often used by industry in its natural state, it is rarely consumed in its complete form. Because they are sensitive to variations in temperature, pH, and shear stresses, the majority of native starches have a limited range of direct applications. Native starches exhibit a great propensity for retrogradation and breakdown. In addition, some starch granules lack functional

qualities because they are extraordinarily resistant to enzymatic breakdown, insoluble in water at room temperature, and inert. In order to achieve specific properties like solubility, smoothness, binding, and resilience to the extremely high temperatures used in industrial processes, native starches are commonly modified. Various techniques have been developed to modify starch and change its characteristics to make it suitable for use in the future. Modification of starch involved physical methods chemical methods and enzymatic methods [5].

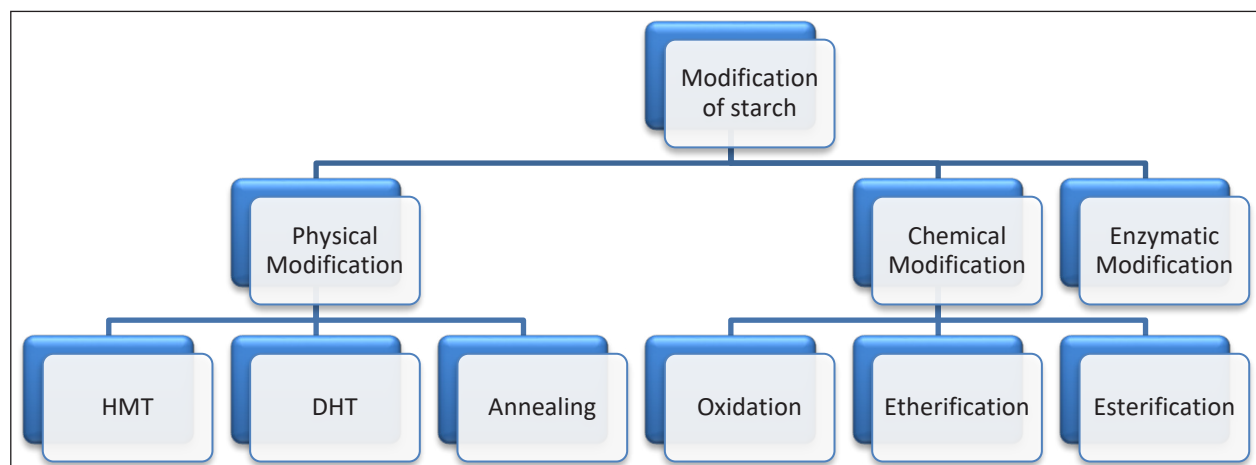


Fig. 3: Representation of Modification of Starch

#### A. Physical Methods

Physical treatments that do not alter the D-glucopyranosyl groups of the starch polymer molecules are referred to as physical modifications of starches. Physical treatments typically only alter the total structures of starch granules and the packing arrangements of the starch polymer molecules inside them, but these modifications can have a substantial effect on the characteristics of the starch itself, as well as the pastes and gels it forms and even its digestibility. The food sector is interested

in physical alterations because they eliminate the requirement for chemical reagents, which eliminates the necessity for the starch product to be labelled as modified. Thermal treatments are the most common physical therapy utilised today [6].

##### i. Heat Moisture Treatment (HMT)

Hydrothermal techniques, like HMT, involve heating starch granules above the temperature at which gelatinization occurs (i.e., at a moisture content that prevents starch from gelatinizing)

and then subjecting them to a particular duration that preserves granular integrity. High temperature (84 to 140 °C); low moisture content (10 to 35%); and exposure period (1 to 16 hours) are the conditions utilised for this starch modification. The process variables—moisture content, the temperature, and heating time—as well as the starch properties—botanical source, structure, amylose/amylopectin ratio, and organization—determine how effective the treatment is [7].

The degree to which these physicochemical property changes of starches occur is mostly determined by alterations in the semi-crystalline arrangement of starch granules. Hoover and Manuel (1996) provided evidence that the crystalline and amorphous components were modified to differing extents by the HMT method. The significant changes were thought to be caused by chain reactions in the amorphous area of the starch granules in different plant starches, including cereal, legumes and tuber. It is exceedingly challenging to consistently describe the properties of starches treated with HMT due to the variety of reaction conditions. Among these various reaction conditions are the amount of moisture, temperature, botanicals supply, heat origin, cooling process, and treatment duration. Among these conditions, temperature and moisture have been the HMT parameters that have been studied the most (Vermeylen *et al.*, 2006; Ambigaipalan *et al.*, 2014). However, the period during heating and the amount of moisture had a greater impact on the properties of maize starch (Zhongquan *et al.*, 2015). HMT-treated starches are used in the industrial setting to prepare newborns for feeding. In the food business, HMT processing is also applied to potato starches to improve their baking qualities and freeze-thaw resilience [8].

#### *ii. Dry Heating Treatment (DHT)*

Dry heating treatment (DHT) is a straightforward, safe physical alteration technique that doesn't produce any pollution. According to some, starches can be thermally treated to generate functionality that is comparable to that achieved by chemical crosslinking—for example, by treating them with phosphorous oxychloride—while utilising just heat. Many applications that need for a stable-viscosity paste made from starch use chemically crosslinked starches; as a result, the dry heating method would be a cost-effective, time-efficient, and chemical-use-reduction option.

The pasting thickness of the starch increased due to DHT, as found by the authors. On the other hand, treated starches displayed reduced pasting viscosity, gel strength, and hydration characteristics (water absorption ratio and swelling power) when a longer time for heating was used. DHT lowered the fast digested starch content and the projected glycemic value of the rich amylose is rice starch in the *in vitro* digestibility of starch data [9].

#### *iii. Annealing*

Another hydrothermal method called annealing involves keeping starch granules in excess water (usually more than 38% w/w) at a point of temperature that is both above and below the starch's glass transition and gelatinization temperatures. Time intervals for the annealing process have varied from minutes to days. The starch undergoes a reorganisation of its semicrystalline structure during the process, which increases the stability of the granules due to the decrease free energy concentration on the molecular structure and the connections between the starch chains. It has been found that ANN can occur at a temperature as low as 16 °C less than the gelatinization temperatures, but that the process is more effective the closer the temperature at which gelatinization takes place.

The development of starches resistive to enzyme digestion and the ease of digestion of modified starches have both been linked to the increased interactions among the chains of starch and the alterations in the granule surface. Because to the crystalline structure's rearrangement after ANN, starch exhibits increased resistance to digestion, with higher amounts of SDS and RS and a lower concentration of RDS [10].

### *B. Chemical Methods*

#### *i. Starch Oxidation*

In certain formulations, oxidised starches have been demonstrated to exhibit enhanced physicochemical characteristics such adhesivity and film formation, which support the regulated release of active ingredients in drug delivery systems. Usually, a regulated pH and temperature reaction between starch slurry and an oxidising agent produces the starch derivative. The three accessible hydroxyl groups are targeted for oxidation during the oxidative process in order to produce new starch derivatives. The oxidised starch derivative, which enhances the physicochemical properties of the original starch particles, is determined by the type of oxidative method and reagents utilized. Consequently, several modifications to the molecular structure of starch occur, which modify the co-polymer to provide the derivative the required properties and enhance its appropriateness for pharmaceutical delivery. The three accessible hydroxyl groups in starch are usually the focus of chemical oxidation, which can be accomplished via oxidative and/or substitutive mechanisms, which usually entail a sequence of events. This is demonstrated by the addition of aldehyde groups to native starch granules, which causes at least two reactions. The first step is the oxidation of the hydroxyl groups to carboxyl or carbonyl linkages. The cleaving of the -(1-4) linkages between the glucopyranose units depolarizes chains. Thus, depolarization and oxidation efficiency can be linked. Oxidation efficiency can be influenced by various

factors such as the type of reaction and chemicals utilised, as well as the molecular structure and organisation of starch [11].

### ii. Starch Esterification

The process of starch esterification is essential for changing the physicochemical characteristics and structure of starch so that it can be used as an industrial and functional material. A variety of variants of organic acids, such as anhydrides and chlorides, or inorganic acids, such as phosphate and sulphate, can be used to modify starch to produce starch esters. Because of their increased viscosity and transparency, inorganic starch esters, such as starch phosphate, are useful as adhesives, thickeners, stabilisers, and medication bulking agents [69]. It is usually preferable to use starch directly in thermoplastic [26] polymer

applications with a high degree of esterification-induced substitutes of the  $-OH$  functional group. Esterification of starch can be used to improve a material's processability, promote hydrophobicity, reduce crystallinity, boost flexibility, etc. [12].

### iii. Starch Etherification

Esterification is a modification process whereby an ether link (ROR) is formed and the hydroxyl groups are replaced by hydroxypropyl, carboxymethyl, and/or hydroxyethyl groups. While there are differences in etherification techniques, all of them usually need an alkaline catalyst to start the chemical substitutions, with sodium hydroxide being a popular choice [13].

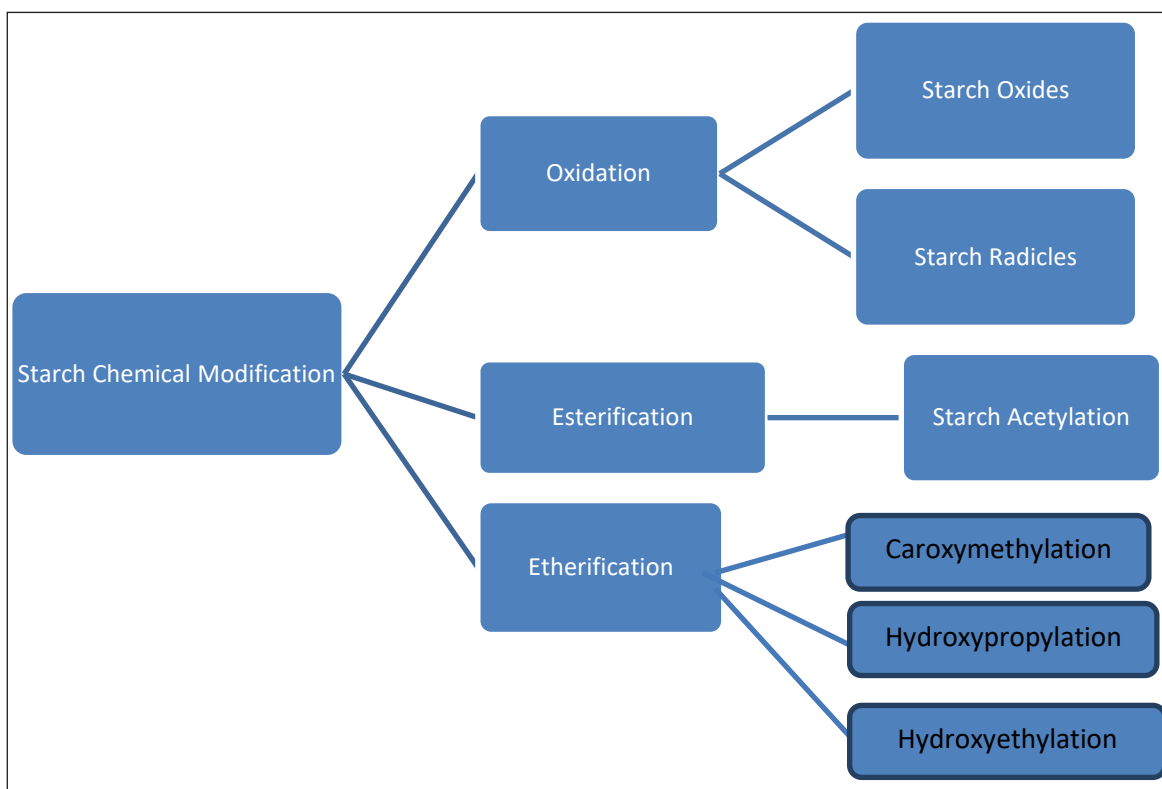


Fig. 4: Chemical Modification of Starch

## V. ENZYMIC MODIFICATIONS

Glycosyl hydrolases and transglycosylases are the two main categories of enzymes that modify starch. These include essential enzymes for functionalizing starch, like  $\alpha$ -amylase (maltogenic amylase), amylosucrase,  $\beta$ -amylase glycosyltransferase, cyclodextrin and branching enzyme (glucantransferase), and de-branching enzymes (pullulanase, isoamylase). 4-Amylomaltase  $\alpha$ -glucanotransferase Glycosyl hydrolases catalyse the hydrolytic breakdown of the glycosidic link in glycosides, generating glucose, a sugar hemiacetal, and

free aglycon. Conversely, transglycosylases catalyse the change of one glycoside into another, *Bacillus acidopullulyticus*, the fungus *Aspergillus niger*, *Bacillus stearothermophilus* B. subtilis, *Bacillus magaterium*, *N. polysaccharea*, *Deinococcus geothermalis*, *Deinococcus radiodurans*, *Rhodothermus obamensis*, *Aquifex aeolicus* and *Rhodothermus obamensi* are among the microorganisms from which these starch-modifying enzymes have been isolated. Additionally, certain enzymes have been extracted from different plant sources, including barley (malt) and ramie leaf [14].

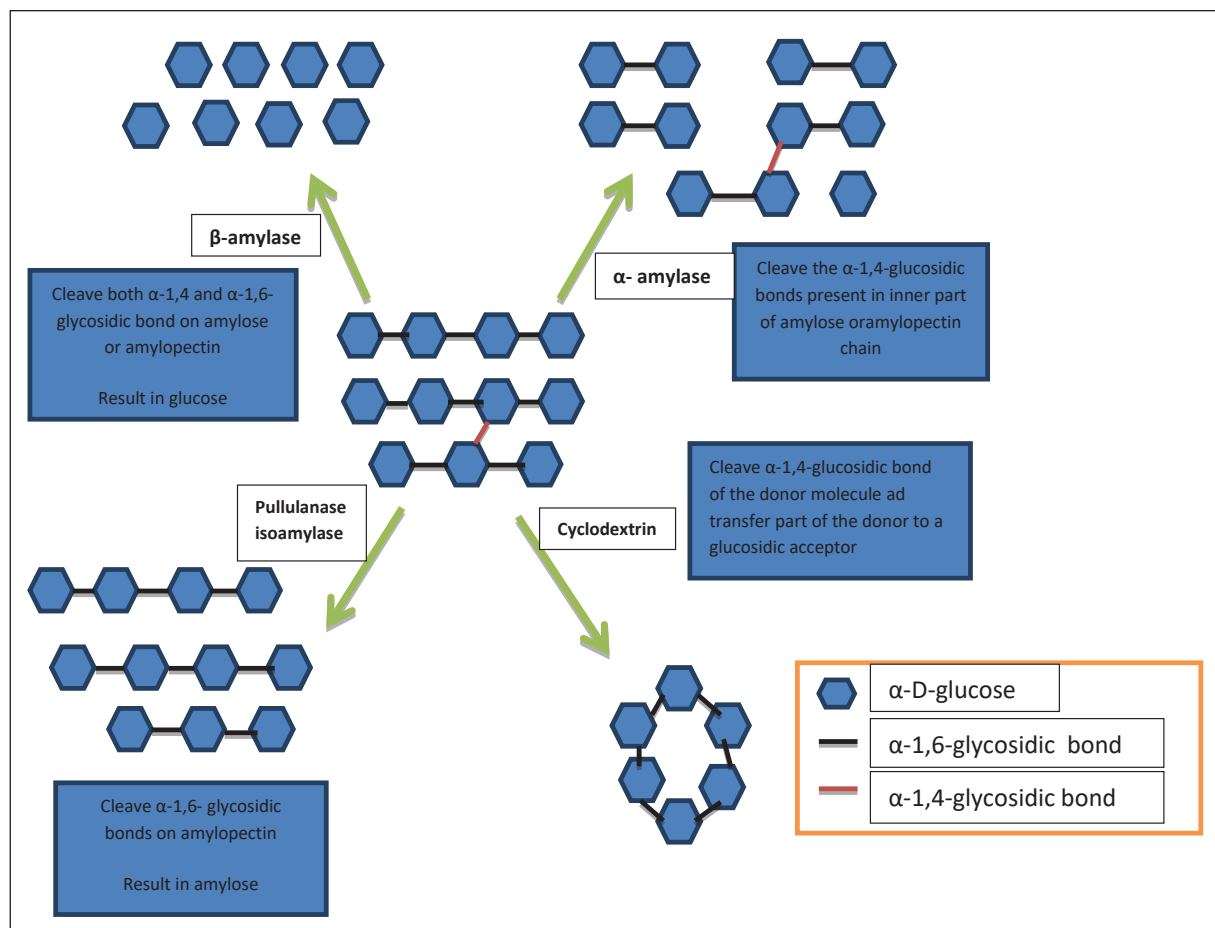


Fig. 5: Enzymatic Hydrolysis of Starch

## VI. APPLICATION OF MODIFIED STARCH

### A. Used as Self-Healing Polymeric Material

Formaldehyde-based microcapsules that are hard, cross-linked, and uniformly dispersed are commonly found inside self-healing polymers. This is carried out so that the micro-capsules can transport the dispersion around without interfering with the polymeric phase. The highly reactive (healing) fluids like epoxy compounds, GMA, and isocyanates are typically found inside the microcapsules due to the sensitive groups these fluids—which are frequently monomers—have on their chains. These liquids react fast with different functional groups on polymer chains. The material's ability to self-heal is activated when large-molecule-based micro particles move within it, clash, or even burst one or more microcapsules that carry therapeutic chemicals. Eventually, the sealant which is applied into fractures and fills gaps will both fill and cross link, depending on the cross linker type utilised in the polymerization phase and the surrounding temperature [15].

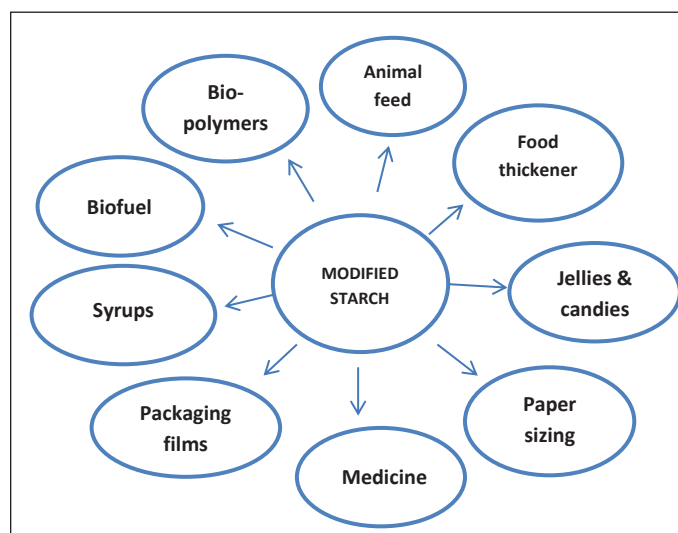


Fig. 6: Applications of Modified Starch

### B. Used in the Process of Delivery of Drugs

Many people are interested in using starch as a potential medication delivery agent and as a way to regulate the rate of dynamic drug release over prolonged periods of time because it is a biocompatible and ecologically acceptable material. To gradually regulate the release of medication into capsules, local starch—which functions as both a binding agent and a tablet preparation agent—can be modified twice. In an investigation into the effects of using several biological sources of starch in the production of pills, it was discovered that the availability of carbohydrates and the kind of modification applied to the local starch considerably impacted the rate of fragmentation, which is also linked to the rate of fluid medication factor release [15].

### C. Used as a Starch Based Film in Food Packaging

The first thing that comes to mind when thinking about using a starch-based film for food packaging is food coating film. Based on starch films, they are an excellent edible coating for foods with a greater respiration rate, such as fruits and vegetables, due to their improved oxygen-blocking capabilities. The membrane stops respiration and reduces the rate of oxidation. Because starch-based films love water, they have insufficient humidity resistance in this situation, where the coating needs to be rinsed off after application. As such, most low-cost edible starch films are flavourless, colourless, and odourless [16].

TABLE I: UNDERUTILISED STARCH AND ITS FUNCTIONS

<i>Starch Source</i>	<i>Functions</i>	<i>Reference</i>
Hairless Canary Seed ( <i>Phalaris Canariensis</i> L.)	As a Gel stabiliser.	[18]
Quinoa	As a Food grade quality pickering emulsions stabiliser.	[19]
Carob	bread production (gluten free).	[20]
Sweet Potato	Used in starch industry.	[21]
Chesnut ( <i>Castanea Sativa</i> Mill)	Substitute of corn-starch and lower processing temperature.	[22]
Pinhão	Used in Microencapsulation of $\beta$ -carotene.	[23]
Jackfruit seed	<ul style="list-style-type: none"> <li>• Production of Chocolate aroma.</li> <li>• Encapsulation of Rice bran oil.</li> <li>• Flour production.</li> <li>• Used in Production of reduced calorie chocolate cake.</li> </ul>	[24], [25]
Mango kernel	<ul style="list-style-type: none"> <li>• Coating purpose.</li> <li>• Edible coating for enhancing shelf-life.</li> <li>• Production of Composite film.</li> </ul>	[26], [27]
Tikhur ( <i>Curcuma Angustifolia</i> )	Mimic the properties of Tapioca Sago.	[28]
Tamarind ( <i>Tamarindus Indica</i> L.) kernel starch	For the food products requires high swelling power.	[29]
Banana	<ul style="list-style-type: none"> <li>• For the food products requires high pasting ability.</li> <li>• As a Emulsifying agent.</li> </ul>	[30]
Litchi kernel starch	Have Good gel strength and elasticity.	[31]

## VII. CONCLUSION AND FUTURE TRENDS

Fruit trash accounts for a sizable portion of the amount of food waste produced worldwide. However, its capacity to recover valuable components is limited. In this way, scientists and business have collaborated to turn fruit waste into valuable products. Utilising these wastes as an unorthodox supply of starch seems promising for this purpose. The main fruit wastes that can be utilised as a source of starch are reviewed here, along with their structural and functional traits. A thorough definition of extraction techniques is provided, along with a discussion of possible uses. The rise of the industrial sector must coexist with the valorisation of fruit pulp starch and its avenues for

large-scale processing. But the information regarding these carbohydrates is still scarce [2].

Therefore, it is essential to expand one's knowledge and look for fresh approaches to raising starch yield and improving industrial competitiveness. It is possible to change starch by chemical, physical, or enzymatic means. Though their high production costs prevent their usage in the polymer industry, enzymatic and genetic techniques find widespread application in the food and pharmaceutical sectors. Physical methods are less expensive and more environmentally friendly, but they weaken the structure of starch, which limits their commercial use. Physically modified starch has use not just in the food business but also in 3D printing and packaging. Chemical

treatments are extensively employed in the business due to their ability to modify the structural characteristics of starch, hence supplying the necessary product features [14].

Chemically modified starch and its derivatives can be made via a variety of methods, such as etherification grafts, esterification, alkaline treatment, acid hydrolysis, and dual modifications. The tensile strength, elongation at break, adhesive qualities, crystallinity, shape, and molecular weight of starch are all impacted by chemical alteration. Moreover, chemical alterations that offer strong moisture barrier, air, and foaming qualities may increase shelf life and enhance quality. Chemically modified starch has applications in the fields of packaging, biobased adhesives, biomedical sciences, agriculture, superabsorbents, and wastewater treatment. The primary disadvantages of modified starch include its poor mechanical qualities high water sensitivity, propensity to form hazardous compounds, and difficult processing. Because modified starch is gluten-free, its use as emulsifiers and stabilising agents in the food and beverage industry is anticipated to increase. The market for modified starch is expected to expand due to various causes, including the increasing need for processed foods, the increasing use of bio-based adhesives in various industrial applications, and the growing enthusiasm for starch-based products. The need for all-natural sweeteners in the beverage and food sectors is also anticipated to drive growth in the global modified starch market in the coming years, as modified starch is used in paper goods, cosmetics, and pharmaceuticals [17].

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