FUNCTIONAL CLEAN-LABEL STARCH: SUSTAINABLE PRODUCTION TECHNOLOGIES AND FOOD APPLICATIONS

Abstract

Starch, the major source of carbohydrate and energy in our daily food has been modified by chemical additives for decades in the food industry particularly due to high demand of starch-based food products. But recently there is a boost to the "Cleanlabel" tag among the health-conscious consumer community. The term clean label starch invariably means starch with least processing, additive free and understandable ingredient list. This movement has inclined the research community towards finding native and underutilized sources of starch. But the major problem is that these native sources are not industrially suitable for food manufacturing and has certain limitations. In this scenario, physical and enzymatic modification of starch plays an important role. Starch modified by above two methods are simply considered as "starch" without any Enumbers.This study also focuses on the use of starch in the food sector, including the microencapsulation of active substances such as nutraceuticals, vitamins, tastes, minerals, and even fertilisers for controlled release and targeted delivery. It is also utilised to make biodegradable and nano-composite films for packaging, as well as starch-protein-lipid complexes for decreased starch digestibility. It is worth noting that starches may be utilised in small amounts alongside protein to create plant-based alternatives to meat. The biocompatibility, biodegradability, low cost, and broad availability of starch make them ideal for all these uses.

Keywords: Functional clean-label starch; starch sources; physical and enzymatic modifications; functional properties; food applications; consumer acceptability

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I. INTRODUCTION

Today's human lifestyle has drastically changed compared to previous generations. With the advent of technology, most people lead sedentary lifestyles, spending hours in front of screens for work or leisure. This shift has significant implications for our health and wellbeing, making it crucial to adapt a balanced diet. According to an ICMR report, an average adult from urban India consumes 1943 Kcal/day, 289 g carbohydrates, 51.6 g fat and 55.4 g protein. In rural India, an average adult consumes 368 g of carbohydrates, 36 g of fat and 69 g of protein per day [1]. But as per WHO recommendation, 45% to 65% of total calories should be Carbohydrates, 10% to 35% of total calories should be protein, and rest 20% to 35% of total calories should be fat [2]. Among many forms of carbohydrate, we are familiar with in our daily life, Starch is the most important and versatile one. It consists of two polymers-amylose and amylopectin. Amylose is a linear polysaccharide composed of Dglucose units joined by the α -1.4-glycosidic linkages in a helical structure and consist of 10%–30% of natural starch. The characteristic blue-violet colour that appears when starch is treated with iodine is due to the formation of the amylose-iodine complex. Amylopectin is a branched-chain polysaccharide composed of D-glucose units linked primarily by α -1,4glycosidic bonds but with occasional α -1,6-glycosidic bonds and consist 70%–90% of natural starch[3].Commercially starch is available as a white powder and is used extensively in the food business as a viscosity/freeze-thaw agent, emulsifying stabiliser in addition to its nutritional benefits. It has a variety of physical qualities dependent on its composition (for example, swelling, gelatinization, and gelation).The thermal and shear instability of native starch, as well as its propensity to retrograde after chilling or freezing, limits its use in industrial applications and lowers the quality of food products. Chemical modification is done mostly to counteract the limitations, but it lowers the nutritional benefit. As an alternative, clean label starch has emerged as a popular choice. Now clean label food is a consumer driven movement that calls for a return to true food by demanding food containing natural, familiar, and simple ingredients that are easy to recognize, understand and free from synthetic chemicals. Clean label starch refers to a type of starch that undergoes minimal processing to retain its functional properties and is free from chemical modifications, artificial additives, chemicals, and preservatives. Clean label can be achieved by using nonconventional and underutilized starch resources, Physical and enzymatic modification, Nano encapsulation, Nano-sized starch, starch-protein and starch-lipid complexes etc [4]. Nonconventional and underutilized starch resources include various fruit seed, grains, normally overlooked and discarded plant parts (banana peel, cassava stem), rhizomes, bamboo shoots, bulbs etc. Exploring non-conventional and underutilized starch resources in clean label starch offer opportunities for diversification, sustainability, and innovation in the clean label starch industry, promotes biodiversity, and supports sustainable agricultural practices. Physical and enzymatic modifications offer clean label alternatives to chemically modified starches, as they utilize natural processes and enzymes. These modifications can improve the functionality, stability, texture, and digestibility of starch, thereby enhancing its performance in various food applications. The use of nano-sized starch in clean label starch formulations can provide functional advantages, such as improved texture, reduced staling, enhanced mouthfeel, and increased bioavailability. In nano encapsulation, proteins, polysaccharides or lipids form a matrix around the target substance (flavours, nutrients, bioactive compounds) providing stability, improved solubility, controlled release, and protection against environmental factors. Consumer acceptability and market suitability are to be considered

closely before introduction of clean label starch prepared through any of the abovementioned approaches.

II. NON-CONVENTIONAL AND UNDERUTILIZED STARCH SOURCES: IDENTIFICATION, SELECTION, EXTRACTION, AND CHARACTERIZATION

The major starch-producing crops are maize, potato, cassava, and wheat and these starches dominate the current markets. So, the underutilized, unconventional, and minor starches refer to those starches apart from maize, potato, cassava root, and wheat [5]. Along with identification and selection of such sources, finding economically feasible extraction process and improving starch yield are also very crucial for popularizing them.

Table1: Non-conventional and underutilized starch sources

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Among fruit seeds, Jackfruit *(Artocarpus heterophyllus Lam.)* is a promising cheap source of starch due to low cost and widespread cultivation and high content of starch in its seeds (60–80%, dry matter basis). "King of Fruits" Mango *(Mangifera indica)* is another such viable option due to abovementioned reasons and 58–80% starch content in kernel [26]. Avocado, Litchi (also rich in polyphenol) seeds and banana pulp are generally waste in fruit processing industry but they can be used as potential sources of starch also.

Millets are another example of underutilized starch source. Apart from ease of cultivation, these starch can be used as fat replacer in ice creams and to form capsules [27], [28].

Pseudocereals namely quinoa, amaranth [29], buckwheat sago palm are now emerging as unconventional starch sources. Among them, amaranth starch is extracted from perisperm of grains and sago starch yield is being improved using pre-extraction treatments.

Bamboo is another underutilized source of starch and has promising application in food and paper industry. Legumes namely lentil, kidney beans, lablab, pigeon pea, chickpea, black bean etc also has significant content of starch but starch isolation is difficult because of the insoluble flocculent protein and fibre.

Rhizomes like lotus, ginger and turmeric are recently being used to extract starch. Ginger starch can be used as a satisfactory texture agent for foods manufactured at high temperature, or a stabilizing agent for sterilized products such as infant foods and UHT products [23].

Most commonly cassava root is used for starch extraction. But recent studies have shown that cassava stem, which is normally discarded as a waste product, can also be used to obtain starch. Lily bulbs and bulbils, which are underground and aboveground starch storage organs, are also being used as starch source these days.

III.PRODUCTION TECHNOLOGY &THEIR CHARACTERISATION AND FUNCTIONAL PROPERTIES

Native starches are unsuitable for use in the food industry due to thermal and shear instability, insolubility in water and organic solvents, tendency towards retrogradation, poor cold storage stability, high viscosity at low concentrations, and poor resistance to acid and extreme temperatures[30], [31]. To obtain "Clean Label Starch," native starch is modified by physical, enzymatic, and starch blending methods. Starch modified by these methods is considered an ingredient rather than an additive and is accompanied without any Enumbers[32].

Figure 1: Flowchart of different industrial methods to modify native starch

1. Physical Modification: Physical modification results in the packing of molecules in starch granules, resulting in damage/rearrangement of molecules. The functional properties and characteristics of starch, such as digestibility, thermal stability, and gels and hot pastes, change[33], [34]. Native starches are subjected to several mechanical actions such as shear pressure, combination of temperature and moisture, and irradiation to alter the size of starch granules and interactions between amylose and amylopectin in the crystalline and amorphous regions[30], [33]. Although these changes are not as significant and persistent as those brought about by chemical modification methods, physical treatments can create starches with characteristics that are fairly similar to those achieved by chemical modification. Physically, starch can be thermally and non-thermally modified. Thermal treatments include pre-gelatinization, hydrothermal treatment (heatmoisture treatment and annealing), and granular cold-water swelling. Non-thermal treatments include ultrasonication, high hydrostatic pressure (HHP) treatment, pulsed electric field (PEF) treatment, use of cold plasma, freezing, thawing, freeze drying, and irradiation. Physical modifications are very suitable for large-scale industrial production because of their high efficiency, lack of chemical reagent by-products, low cost, and ease of operation[30], [33], [34].

Thermal Methods

 Pregelatinization: Pregelatinization is the most widely used physical process for starch modification, which involves total gelatinization followed by drying[32], [33]. Pre-gelatinized starch can be produced using three methods: drum and roll drying, extrusion drying, and spray drying[34]. These procedures cause the starch granules to swell irreversibly and dissolve the hydrogen bonds between starch molecules in such a way that minimal molecular rearrangement occurs. These starches instantly swell and produce a viscous slurry when placed in water. Therefore, no further application of heat is necessary, making them suitable for manufacturing heat-sensitive foods. Pre-gelatinization characteristic features include disrupted granular structure, absence of optical birefringence, and fragmentation [32]–[34].

During drum and roll drying, the starch slurry is coated as a thin layer on the outside surface of the drum as the interior of the drum dryer is heated. A knife connected to the dryer instantly dried the starch slurry in the form of a film/flake after one revolution. The scraped dried film is then processed into flakes or dust. Wiriyawattana et al. (2018) found that compared with the control group, each sample of pre-gelatinized rice berry flour exhibited significantly higher water absorption indices and swelling capabilities. The peak, trough, and final viscosities, pasting temperature, and setback value of the rice berry flour also decreased following drum drying. This is a typical technique used in the food manufacturing industry for items such as porridge and gruel[32].

Extrusion drying is a high-temperature short-time (HTST) technique with various applications. Starch was compressed by an extruder and subjected to mechanical shear force, high temperature, and generally low humidity during the process[32], [33]. The extrudate was dried and crushed to different sizes using the following procedure: According to Colonna et al. (1984) and Doublier et al. (1984), depolymerization may occur during the production of pre-gelatinized starch, and the molecular weight of wheat starch amylose and amylopectin is reduced during the extrusion process. These modifications can be attributed to the significant shear force created in the extruder. Koa et al. (2017) evaluated the impacts of extrusion on barley and observed that extrudates had greater transverse expansion, faster starch digestion, and rapidly digestible starch than nonextrudates[34].

Spray drying has emerged as the most commonly used microencapsulation technology for components because of its excellent solubility, excellent drying and emulsifying properties, non-hygroscopic behaviour, bland taste, inertness, and economic efficiency. Izidoro et al. (2011) investigated the effect of ultrasonication on green banana starch. In the case of spray drying and ultrasonic treatment of starch, the amount of resistant starch was markedly lower, probably due to less starch crystallisation. The researchers showed that spray drying enhanced the swelling ability, solubility, and capacity to absorb starch[32], [33].

- **Hydrothermal Treatment:** Hydrothermal treatment alters the physicochemical and functional characteristics of starch without disrupting the structure of the granules. In general, two methods are used: 1) heat-moisture treatment and 2) annealing This occurs when starch polymers change from an amorphous to semicrystalline state. Hydrothermal treatment takes place under the starch gelatinization temperature, preserving the granular structure and keeping the starch in a movable rubbery condition throughout modification[32], [33].
- **Heat Moisture Treatment:** HMT is performed by heating starch at temperatures (80– 140 \degree C, usually over 94 \degree C) above its glass transition temperature for a defined amount of time (1–24 h) with restricted moisture levels (10–40% w/w, commonly 35%)[32], [34]. When different kinds of starch were subjected to HMT treatment, the gelatinization temperature, peak temperature, and conclusion temperature all tended to increase while the gelatinization enthalpy typically decreased[34]. When HMT treatment is carried out at a higher temperature, the granule size increases, the viscosity properties diminish, and the enthalpy reduces. During treatment, the solubility and swelling power decrease as the water content and temperature increase, resulting in greater crystallinity (crystalline perfection), higher gel density and increased amylose cross-linking. Pasting temperature and time increased significantly at 25% and 30% moisture content, whereas peak viscosity and breakdown appeared to decrease in various experiments on the viscosity properties of HMT starch. Depending on the type of starch, HMT has different effects on the setback or final viscosity. According to reports, depending on the type of starch used, the qualities, including the stability of paste gels, may change. HMT is used to make resistant starch (RS) and slowly digestible starch (SDS) with high thermal stability. According to Cham and Suwannaporn (2010), HMT is better suited for the manufacturing of dried and semi-dried rice noodles because it requires stronger tensile strength and gel hardness. HMT starches may also be used in the preparation of bread, noodles and pasta[32], [34].
- **Annealing:** ANN is carried out in a surplus of water (more than 60%) at a temperature higher than the glass transition temperature but lower than the starch gelatinization temperature (usually $5-15$ °C lower)[35]. Annealing different forms of starch under different moisture content, temperature, and duration conditions results in uneven trends in several functional qualities. It has been done for as little as 0.5 h and as much as 192 h[34]. According to studies, ANN causes higher granule stability, decreased birefringence, increased porosity, crystalline perfection, an increase in the number of starch chain interactions, and reduction in amylose leaching[36]. Increased gelatinization temperature and decreased temperature range of phase transition with increasing annealing temperature are well known changes in thermal characteristics[34]. Chung et al. (2000) also observed that ANN induces rearrangement of starch molecules, resulting in decreased swelling power and solubility as well as increased gel strength[37]. Annealing increases the pasting temperature and thermal stability while decreasing the peak and final viscosity[34]. Simultaneously, Jacobs et al.'s (1996) study showed a deviation from this pattern, noting that ANN increases the peak

and final viscosities of rice and wheat starches[4]. Similar to HMT, ANN is also utilized to produce RS and SDS with high thermal stability[4]. ANN treatment is useful when rice noodles need to have a softer texture[32].

- **Granular Cold-Water Swelling:** Granular cold-water-swelling (GCWS) starch, also known as instant starch, is a form of pre-gelatinized starch that can be prepared in four ways. The first method involves heating amylose-containing starch in an alcoholic solution. The heated starch suspension was spray-dried in the second approach. This method has been used for a long time, and reports by Pitchon et al. (1981) have confirmed variations in the properties of starch according to their properties. The starch was treated in the third way using an aqueous alkaline alcohol solution at room temperature. The fourth approach is instantaneous controlled pressure drop (DIC)[34]. These methods modify the elemental structure of the D- glucopyranosyl unit, resulting only in physical modification. GCWS demonstrated stronger light transmittance and solubility than PG while having lower crystallinity and swelling power in buckwheat starch[34]. It can be used in conjunction with other non-thermal physical therapy methods, such as ultrasonic treatment or high-pressure treatment.
- **Non-thermal Methods:** Non-thermal physical modification methods are the most effective to overcome the limitations of thermal methods. As a result of the use of high temperatures in thermal methods, there is a loss of nutrients, flavours, vitamins & minerals, colour, texture, etc. In this context, non-thermal methods are useful for preventing food losses[33].
	- **Ultrasonication:** Ultrasonication refers to the application of sound waves with a frequency higher than the human hearing threshold (>20 kHz) to starch granules suspended in solution or gelatinized starch[32], [33]. Ultrasound induces high temperatures, significant shear stress, and free radicals that alter the functionalities and structural alterations of starch in a starch–water system. The magnitude of these alterations was determined by the ultrasonic frequency, duration, temperature, moisture content of the entire system, and starch type[38]. Since starch molecules cannot absorb ultrasound's sonic energy, they must be converted into a chemically useful form to generate cavitation, which in turn causes shear pressures that rupture polymer chains, thereby damaging starch granules and causing the rapid breakdown of microbubbles[32], [38]. The amount of degradation was found to increase with longer treatment time and was more strongly influenced by starch with larger granules (such as potato starch). In addition, more severe granule fractures were observed around the Maltese cross[32]. According to previous studies, in the presence of oxygen, less damage was done to the starch surface, whereas in the presence of air, the starch surface was more rugged. There was nothing damaged in either vacuum or CO2. The pits were deep and enormous, and the surface was smoothed with hydrogen[32]. Due to the damage to the starch granule surface caused by sonication, water solubility and swelling power are increased, whereas crystallinity, peak and final viscosity, pasting properties, and the corresponding level of polymerization are decreased[32], [34], [38]. However, Cham et al. (2010) and Carmona-Garc et al. (2016) concluded that peak viscosities increased following ultrasonication. After

ultrasonication for 15 min, the rheological characteristics of G′ and G′′ increased and decreased after 30 min[33]. The retrogradation enthalpy of rice starch was decreased because of the high ultrasonic power and high intensity, which successfully raised the onset temperature^[4]. Ultrasound-treated starch has many applications such as emulsifier, edible film, enzymatic encapsulation and delivery system, V-type inclusion complex formation, and bioethanol formation[4].

 High-Pressure Treatment: High hydrostatic pressure processing (HHP) is a technique used to alter starch in an aqueous solution over a relatively short period (under 30 minutes) using high pressure (generally between 100 and 600 MPa)[39]. Under high-pressure treatment settings, starch properties vary significantly depending on the treatment pressure, pressurization temperature, and pressurization duration[32]. HHP encourages water molecules to penetrate starch granules, weakening the double helices in the crystalline area and destroying the internal crystalline structure of starch[30]. A number of starches, such as maize, waxy corn, rice and wheat, partially gelatinise at a constant temperature and pressure of more than 300 MPa, but gelatinise at 600 MPa[38]. Since potato starch is more resistant to pressure than other starches, it requires a pressure of more than 600 MPa to fully gelatinise All types of starch can be gelatinised using HPP at temperatures below zero, provided that the pressure is sufficiently high. In addition to pressure, a lower starch gelatinisation pressure is necessary. Baszczak et al. (2005) [4]found that after potato starch was subjected to a 600 MPa highpressure treatment for 2–3 min, the outside of the granules appeared to be more resistant to change and exhibited a very compact, condensed layer, whereas the interior of the granules, which had a relatively coarse structure, clearly showed destroyed and gel-like structures. The starch became gelatinized under pressure, as evidenced by the decrease in birefringence and the formation of a gel-like structure. The degree of gelatinisation increased significantly as the duration of the pressurisation treatment was increased up to 60 min, after which a plateau was established. The consistency index, melting enthalpy, and differential scanning calorimetry peak temperature increased with increasing pressure and pressurization duration[4]. Guo et al. (2015) discovered that high-pressure (100– 500 MPa) treated lotus seed starch had significantly greater pasting temperature, peak viscosity, trough viscosity, and final viscosity compared with native starch. However, for breakdown and setback, the results were the opposite: According to several previous studies on the impact of high-pressure processing on the rheological properties of different starches, including sorghum, rice, and mung bean, the moduli of most starch gels dramatically increased as the treatment pressure level increased[4]. High-pressure homogenised starch also shows reduced T0, Tp and gelatinisation enthalpies (H). In many food companies and bakeries, high-pressure processing offers enormous potential for energy savings. The resultant creamy texture paste can substitute oil in low-fat foods (such as mayonnaise, confectionery food items, sweets, and dairy products) without heat treatment when starch pastes with a concentration of more than 15% are processed using this method. It also produces RS, which is effective in the control of colorectal cancer and diabetes[4].

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- **Pulsed Electric Field Treatment:** Pulsed electric field treatment is a promising non- thermal technology that uses high-intensity (more than 10 kV cm^{-1}) electric pulses for brief (40 µs) periods to treat starch materials in a processing chamber[4]. The starch's morphology is almost completely disrupted when the intensity approaches 50 kV cm⁻¹, resulting in the formation of a gel network from fragment clustering[4]. PEF treatment damages the outer dense protective layer of the granule. This leads to increased water absorption and swelling of the granules, resulting in particle aggregation[30]. During pasting at a high electric field intensity of more than 50 kV cm^{-1} , the setback value, peak viscosity (as seen in tapioca starch–water dispersion solutions), and breakdown viscosity decrease[4]. According to thermal analysis using differential scanning calorimetry, PEF tends to lower gelatinization temperature and ΔH . In addition, the lower gelatinization temperature raises the possibility that PEF disorganizes the crystalline region[4]. When combined with wheat, potato, and pea starches, PEF may vary the starch digestion rate, creating RS, SDS, and RDS. The amount of RDS was remarkably enhanced with electric field intensity for all starches, whereas in all electric field intensities, the amount of SDS decreased in the treated specimens. Different RS tendencies for various forms of starch were observed. When potato starch and wheat flour were treated with low-intensity PEF (2.86 and 4.29 kV cm⁻¹, respectively), the RS content increased considerably. However, when the electric field strength was enhanced $(5.71, 7.14, \text{ and } 8.57 \text{ kV cm}^{-1})$, the value did not vary significantly[4]. The RS of pea starch did not change significantly at any electrical intensity. Variables that affect the effectiveness of PEF treatments include the strength of the electric field, the length of treatment, and the temperature of the food[38]. In view of the significant benefits of low processing temperature, quicker treatment and consistent treatment intensity, PEF has been widely used in pasteurisation, enzyme deactivation, extraction, molecular modification and chemical reaction enhancement[30].
- **Plasma:** The plasma technique of starch modification is a novel approach that changes starch in various ways based on the type of plasma and the excited gas, the conditions, and the energy generation[38]. This method causes increased surface energy, increased or decreased hydrophilicity, oxidation, bond breakage or depolymerization, and crosslinking of starch[38]. During plasma treatment, two important competing processes are cross-linking and depolymerization. Crosslinking improves starch integrity and inhibits dissociation, whereas depolymerization results in more fragmented starch molecules and a lower capacity to reflect light. These responses are affected by the type of plasma used and the treatment circumstances[38].Wongsagonsup et al. (2014)[38] used jet atmospheric argon plasma treatment to nonchemically modify tapioca starch. They employed two types of starch suspension - granular starch and cooked starch, each of which was exposed to with 50 W and 100 W plasma for 5 minutes. They discovered that granular starch treated with 50 W and cooked starch treated with 100 W had reduced paste clarity when compared to untreated control samples, whereas granular starch treated with 100 W and cooked starch treated with 50 W had the reverse effect, i.e., higher paste clarity. The cross- linking process was prominent in the first example, whereas the depolymerization reaction ruled in the second. In comparison to untreated samples, the water binding capacity and

swelling power of plasma-treated corn and tapioca starches increased. The plasma treatment reduced the peak temperature (T_p) and enthalpy H of corn starches while increasing the T_p and enthalpy H of tapioca starch. When the pasting qualities of corn and tapioca starch were examined, it was discovered that after the treatment, the pasting temperature for corn and tapioca starch decreased, but the peak viscosity (PV) elevated. SEM revealed that there were no cracks or holes created on the surface of either starch granule, while corn starch granules displayed uneven surfaces and deposits following exposed plasma treatment[38]. Cold plasma treatments are believed to have no or limited effects on the physical, chemical, nutritional, and sensory properties of different products due to their nonthermal character[38].

- **Freezing, Thawing and Freeze Drying:** As a result of syneresis, amylose and amylopectin are leached upon thawing, resulting in a change in chemical properties. Freezing and thawing of potato starch resulted in particle corrosion and higher specific surface area. In addition, the water matrix formed during severe freezing compresses starch granules, which may cause amylopectin leakage from the interior to the surface. Lyophilization reduces the crystallinity in the inner structure of starch and affects the surface. This damage allows enzymes to enter starch granules and improve digestibility[34].
- **Irradiation:** Electromagnetic radiation, such as gamma rays γ and microwaves, is considered to be a physical modification of starch[39].

Cobalt-60 (60Co) and cesium-137 (137Cs) are the chief sources of gamma radiation used for starch modification. Irradiation by gamma rays generates free radicals that not only break starch chains but also produce carbonyl groups along with carboxylic acids in faba bean starch irradiated at various dosages (0–15 kGy) using a 60Co-ray source. The results showed a decrease in relative crystallinity with increase in carboxyl content, lower viscosities and better freeze-thaw stability. This may be related to the irradiation-induced shortening of starch chains[39]. These very intense and penetrating radiations may also affect the morphological characteristics of starch, depending on the dosage[30].

Microwave treatment induces fast dipole reorientation of polar molecules in the starch system, resulting in the formation of structural changes[30]. The first step of microwave modification is the disintegration of the amorphous area (internal chain), while the second stage is the destruction of the crystalline region (external chain)[30]. Yang et al. (2017) discovered that the relative crystallinity of waxy maize starch reduced by 10.47%, 6.42%, and 2.91% after 5 min, 10 min, and 20 min of microwave irradiation, respectively, implying that microwave irradiation could cause significant damage to the crystalline areas, the disappearance of the double helices, and degradation of starch granules. In addition, the moisture content increased (from 30% to 50%), but the relative crystallinity of millet starch decreased considerably (from 23.3% to 3.2%)[30]. As a result, a higher moisture content may boost the microwave effect on starch by increasing the heating rate (due to its high dielectric characteristics) and increasing the hydration in the amorphous region. As reported by Li et al., 2019[30], when treated with microwave at 700 W for 60 s, starch with a moisture content of 30% retained its shape but a few cracks and cavities could be encountered on the surface, starch granules with 40% water aggregated together to produce larger starch clusters, and granules with 45% moisture content completely deformed and developed big gel blocks.

2. Enzymatic Modification: Recently, enzymatic treatment has been introduced as another ―clean-label‖ approach to improve starch functionalities. In addition, enzymatic changes can be used in combination with other methods, such as physical treatments, to achieve the desired functionality of the modified starches[39]. Other advantages of enzymatically modified (EM) starches include better purity, consistency of high-quality products, lower cost and absence of undesirable products[31]. Enzyme-modified starches undergo debranching, phosphate substitution, and disproportionation, which alter starch chain length and branch point creation. These starches are characterised by a lower paste viscosity, different rheological characteristics and improved elastic properties and digestibility. When starches are exposed to enzymatic activity, holes and pits emerge in the starch granules, resulting in a porous structure that allows water molecules to enter the starch granules to facilitate gelatinization, liquid absorption, or delay the release of enclosed fluid[31]. Starch-modifying enzymes include (1) α -glucanotransferases (α GTases) which comprises branching enzyme (BE, EC 2.4.1.18), cyclodextrin glycosyltransferase (CGTase, EC 2.4.1.19), and $4-\alpha$ -glucanotransferase (4α GTase, EC 2.4.1.25), (2) maltogenic amylase (MA, glucan 1,4-α-maltohydrolase, EC3.2.1. 133), (3) α-amylase (endo-acting enzyme), (4) β-amylase (exo-splitting enzyme), (5) amylo-sucrase (AS, E.C. 2.4.1.4) and (6) descending enzymes: pullulanase (EC 3.2.1.41) and isoamylase (EC 3.2.1.68)[31], [32], [39]. Starting with 4aGTase (amylomaltase) or D-enzyme (disproportionation enzyme), transglycosylation on starch substrates is catalyzed in several steps: disproportionation-cyclization coupling and hydrolysis. By intermolecular and intramolecular transglycosylation, this enzyme can produce modified aggregates of amylopectin and cycloamylose (cyclic α -1, 4-glucan derivatives) or large-ring cyclodextrins. CGTase may also be used to create a thermoreversible starch gel[32]. CGTase may produce cyclic oligosaccharides known as cyclodextrin (6, 7, and 8 glucose residues) capable of forming inclusion complexes with appropriate guest molecules[32]. In vivo synthesis of α -1,6-glucosidic bonds in glycogen and starch by the branching enzyme Similar to the other two, it breaks down the α -1,4-glycosidic linkage. The only difference is that it shifts the cleaved α -glucan chain to the free 6-hydroxyl group in the acceptor glucan chain, resulting in the formation of an a-1,6-glycosidic bond[32]. The products of α-GTase-treated starch include cyclodextrin, thermoreversible starch gel, highly branched cyclic dextrin, slowly digestible starch (SDS), and resistant starch (RS)[32]. Maltogenic amylase induces the hydrolysis of 1,4-glucosidic linkages in starch, releasing maltose as the primary product from the chain's non-reducing ends. MA-treated starch provides improved cold storage endurance, slower retrogradation, and decreased viscosity. MA is widely used in the food industry as an antistaling agent (maintaining moisture and softness) in bread as well as other baked products to prolong their storage period[31]. α-amylase breaks α-1,4 bonds of amylose and amylopectin at random, whereas β-amylase sequentially breaks α -1,4 bonds in amylose and amylopectin and produces monomeric or oligomeric compounds. Pullulanase and other debranched enzymes break α-1,6 glycosidic bonds[40]. Amylo-sucrase transfers a D-glucopyranosyl

moiety from sucrose to the non-reducing end of an acceptor molecule, such as maltose, glycogen, maltodextrin, or sugar polysaccharide, resulting in an amylose-like polysaccharide with only α -(1 4)-glucosidic linkages, making it a powerful glucosylation approach for the production of novel amylopolysaccharides[31].

3. Starch Blending: Blending distinct native or physically modified starches improves the functional properties of native starches. By combining different starch components, additive and non-additive behaviour can be created for all starch qualities. When additive effects are observed, the properties of the blend can be predicted using starch components. In the case of non-additive effects, forecasts deviate from reality, resulting in interaction. Several parameters, such as amylose content, amylose leaching, starch and water concentration relative to granule size, and swelling power, influence the quality of starch blends. Liu and Lelievre (1992) discovered that when the overall starch concentration was low, the blend's thermal parameters were the sum of its component starches[41]. However, due to the struggle for water between the starches in the mixture, non-additive behaviour occurs at high starch concentrations. Waterschoot et al. (2015) discovered that each starch gelatinizes independently in the presence of adequate water; however, in the absence of sufficient water, the starch with a lower gelatinisation temperature gelatinizes first and leaves less water for the gelatinisation of the other starch. As a result, the other starch will gelatinise at high temperatures and change its properties. In relation to the relative granule size, when there is a larger variation in the granule size between the starch components in the mixture, the non-additive impact becomes more obvious. According to previous reports, when restricted-swelling starches were combined in a ratio comparable to that of easily swollen starches, the swelling power was dramatically reduced and the gelatinisation temperature increased[42]. [43]. Blending techniques are mainly used for the preparation of noodles, cakes and wheat replacements. A 1:1 combination of potato starch and rice starch yielded high-quality noodles in terms of cooking duration, transparency, and texture[44]. Starch blending techniques are particularly useful for the preparation of gluten-free foods.

IV. MICROENCAPSULATION PROPERTIES OF STARCH

Microencapsulation is a versatile technology that involves the encapsulation of active substances within a protective matrix, allowing controlled release and targeted delivery. Over the years, starch has emerged as a promising material for microencapsulation due to its biocompatibility, biodegradability, low cost, and widespread availability. Starch, a polysaccharide composed of glucose units, exhibits unique physicochemical properties that make it an ideal candidate for microencapsulation. Starch can form a gel-like matrix when heated with water, enabling easy entrapment of active compounds within its network structure. Furthermore, the presence of hydroxyl groups on starch molecules facilitates chemical modifications and enhances the stability and functionality of the microencapsulated products[45]. Various techniques have been employed to encapsulate active ingredients using starch as the encapsulating material. The most commonly used techniques include spray drying, coacervation, and emulsion methods. Spray drying involves atomizing a solution or suspension containing the active ingredient and starch, followed by rapid drying to obtain microcapsules. Coacervation utilizes phase separation between the core material and the starch matrix, forming a coacervate phase that encapsulates the active compound. Emulsion methods involve the emulsification of a starch solution containing the active substance, followed by solvent evaporation or gelation to produce microcapsules [46].

The versatile properties of starch-based microencapsulation have found numerous applications in different industries. In the food industry, microencapsulation of flavors, vitamins, and nutraceuticals using starch provides enhanced stability, controlled release, and targeted delivery, thereby improving the quality and shelf life of food products. Flavor encapsulation is crucial for improving the stability and shelf life of volatile compounds in food products. Starch-based microencapsulation has been utilized to protect and preserve flavors, allowing their controlled release during consumption. For instance, microencapsulation of citrus oils using starch-based systems has been shown to enhance the stability and release of the flavors in various applications, such as beverages, confectionery, and bakery products [47].The encapsulated flavors remain protected from degradation, evaporation, or interaction with other food ingredients until they are released, resulting in enhanced sensory experiences for consumers.

Vitamins are essential micronutrients that are prone to degradation due to environmental factors, such as heat, light, and oxygen exposure. Microencapsulation using starch offers a solution for improving the stability and bioavailability of vitamins in food and dietary supplements. Starch-based microcapsules have been used to encapsulate vitamins, such as vitamin C, vitamin E, and B-group vitamins, providing protection against degradation and ensuring their controlled release during digestion [48]. These encapsulated vitamins can be incorporated into functional foods, beverages, and dietary supplements, offering improved nutritional value and prolonged shelf life.

In the pharmaceutical field, starch microcapsules have been utilized for the controlled release of drugs, protecting them from degradation and enabling sustained drug release, reducing dosing frequency and improving patient compliance [49]. Nutraceuticals, including bioactive compounds and herbal extracts, possess various health-promoting properties. However, their incorporation into food products is challenging due to issues related to stability, solubility, and taste. Starch-based microencapsulation has been employed to overcome these challenges and enhance the delivery of nutraceuticals. For example, curcumin, a bioactive compound found in turmeric, has been successfully encapsulated within starch microcapsules. This encapsulation approach improves the stability, solubility, and bioavailability of curcumin, enabling its incorporation into functional foods and dietary supplements. Similarly, polyphenols from plant extracts, such as green tea or grape seed extract, can be effectively encapsulated using starch-based systems, providing protection against degradation and allowing for their controlled release in the body. Furthermore, the agricultural sector has also benefited from starch microencapsulation, with controlled release formulations of fertilizers and pesticides offering improved efficiency, reduced environmental impact, and enhanced nutrient utilization [50].

However, despite its many advantages, starch-based microencapsulation also presents some limitations. One major challenge is the susceptibility of starch to moisture, which can lead to premature release of encapsulated materials. This issue can be mitigated by incorporating hydrophobic coatings or blending starch with other polymers to improve its moisture resistance [51]. Furthermore, starch-based microcapsules may suffer from limited encapsulation efficiency and poor mechanical strength, particularly when exposed to harsh processing conditions. Ongoing research focuses on developing novel approaches to optimize

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the encapsulation process and improve the performance of starch microcapsules [52]. The field of starch microencapsulation is constantly evolving, with ongoing research aimed at addressing the existing limitations and exploring new possibilities. One exciting avenue for future development lies in the incorporation of functional additives into starch-based microcapsules. By introducing bioactive compounds, such as antioxidants or antimicrobial agents, into the encapsulation matrix, the resulting microcapsules can offer additional health benefits and extended product shelf life [53]. Furthermore, the combination of starch with other natural polymers or nanoparticles holds great potential for enhancing the encapsulation properties, stability, and release kinetics of microcapsules [54]. Another area of active research is the development of stimuli-responsive starch microcapsules. By modifying the surface properties or incorporating specific components, such as pH-sensitive polymers or temperature-responsive materials, the release of encapsulated substances can be triggered or modulated in response to external stimuli. This approach allows for precise control over the release profile, enabling targeted delivery and customized release patterns.

Moreover, advances in nanotechnology have paved the way for the development of nanoscale starch particles for microencapsulation purposes. Nanosized starch particles exhibit enhanced surface area, improved dispersibility, and increased reactivity, resulting in improved encapsulation efficiency and stability. Furthermore, nanoscale starch particles can be employed in novel encapsulation techniques, such as electrostatic assembly or layer-bylayer deposition, providing precise control over the encapsulation process [55]. The microencapsulation properties of starch have revolutionized various sectors, offering improved product stability, controlled release, and enhanced performance. As research and development in this field progress, the potential applications of starch-based microencapsulation are boundless, promising a bright future for this versatile technology.

V. STARCH FOR BIODEGRADABLE FILMS, NANOPARTICLE BASED COMPOSITE FILMS, AND THEIR FOOD APPLICATIONS

Packaging materials are essential for food hygiene and quality, and petroleum-based polymers and materials account for a large part of the commercial packaging worldwide. These materials have strong processing properties, such as water and vapor barriers, transparency and low cost, but they may have serious environmental impacts, such as carbon dioxide emissions and accumulation over time, as a result of recycling problems with synthetic packaging[56]. Consumer concerns about the disposal of plastic in the environment have increased industrial interest in biodegradable films. Starch is widely used in the production of biodegradable and edible films due to its excellent film-forming characteristics, neutral organoleptic properties, low cost and availability. Biodegradable films are dissimilated into the environment by breaking the polymer chains into smaller components of monomers or dimers[56].

Starch-based biodegradable films are made using various methods, such as solvent casting, extrusion, tape casting, compression moulding and injection moulding. Solvent casting is one of the primary methods used on a small scale to research and create new film composition. A polymer solution in a volatile solvent is applied to a flat die and sequentially deposited onto a stainless-steel belt in commercial solvent casting. Viscosity and temperature in the casting solution are crucial factors that affect the thickness. Filtration was used to remove bubbles and pollutants. The solvent was blown away using dry air, after which the

film was cooled and removed from the conveyor belt. Although it appears to be a straightforward process, some interaction processes depend on the circumstances of the process and the starch granule characteristics. Starch film production begins by heating the starch dispersion in water. In this process, amylose and amylopectin leach into the solution, creating a non-structural paste known as gelatinisation. The process can be divided into two stages, the first occurring between 60 and 70°C and the second occurring above 90°C. During the cooling stage, amylopectin and amylose have a high molecular mobility, which helps to organise the intermolecular hydrogen bond between amylose and amylopectin into a semicrystalline form, known as retrogradation. Plasticisers are often used in casting solutions in order to increase film flexibility and film formation performance. Water is most commonly used as a plasticiser in films, as it is non-toxic and food-friendly[56].

Table 2: Characteristics of biodegradable films made of starch for food packaging

Starch-based packaging films have not been widely used in the packaging sector for several reasons, such as their poor mechanical, barrier, and processing properties. In order to compensate for these shortcomings, starch films are often produced by filler elements, such as nanoparticles, in the starch matrix. The addition of starch nanoparticles to composite films alters the physicochemical, functional and mechanical characteristics of the films. Nanocomposite films have unique properties such as low solubility, low water vapor transfer rate, nontoxicity, and biodegradability, making them an attractive option for food and nonfood applications[57]. Incorporation of nanoparticles forming a dispersed phase in the film matrix restricts biopolymer chain mobility. The water vapour permeability (WVP) of composite films can be reduced by reducing biopolymer chain mobility. The thickness of the nanocomposite films increased when starch films containing different amounts of nano-starch were used. Because of its nanometric scale, nano starch reduces the WVTR of nanocomposite films, increasing the surface-to-volume ratio and disrupting the mobility of polymer chain. Due to its nano size, nano starch improves the density of nanocomposite films, which increases the resistance to water vapor. The flexibility and ductility of the nanoparticles affects the mechanical characteristics of the films. Incorporation of starch nanocrystals raises the melting temperature of films. These nanocomposites are potential future alternatives and important subjects of study, but their practical applications are limited due to their limited manufacturing and high cost. Although nanocomposite technology enhances the water barrier and mechanical characteristics of natural biopolymers, it has only found a few applications in the food industry. Optimum film composition combined with cost-effective and effective production processes may be a viable alternative for the development of novel food packaging materials[57].

VI. STARCH-PROTEIN-LIPID COMPLEXES AND ITS USES

The three macronutrients that compose most foods — lipid, proteins, and starch, are the main suppliers of energy in the average person's diet. The macronutrients in question experience alterations throughout the preparation of foods, and their interactions, which are frequently complex, determine the final quality of food items, nutritional content, and organoleptic qualities [58], [59]. In order to sustainably generate energy and produce shortchain fatty acids, the digestibility of starch should be controlled, particularly by enhancing the proportion of slowly digestible starch (SDS) or resistant starch (RS)[58]. Starch multiscale systems, such as lamellar structures, crystalline structures, morphology features and chain length distribution have been shown to regulate starch enzymatic hydration. Lipids, proteins, phenolic compounds, and protein-containing components are examples of nonstarchy substances that affect the digestibility of starch by generating starchordered structures et/or reducing activity of enzymes. Through non-hydrophilic and electrostatic interactions of starch with proteins and lipids have been resulted into increased starch ordered structures for retarding the rate of digestibility starch. For the purpose of starch

to develop into structured molecules and for its bioavailability to be modulated, proteins or lipids must interact with starch.

In recent years, substantial research has been done on modifying the digestibility of starch using ternary systems of protein, lipid, and starch. However, it hadn't been clear whether the proteins affected the creation of starch-protein-lipid aggregates^[58]. The hydrophobic tail from fatty acids is thought to react alongside starch to produce starch-fatty acids inclusion complexes, while the charge-negative carboxyl group of the fatty acid is thought to engage alongside protein to shape a ternary complex of starch-protein-fatty acid in ternary systems containing starch, lipids, and protein[58]. The development of starch-lipid inclusion complexes with the adhesion of protein to the starch interface led to a considerable reduction in starch digestibility whilst the starch-protein-fatty acid aggregates have developed[58]. Amylose-lipid complex development lowers starch sensitivity to digestive enzymes and expansion strength, delays starch retrogradation, raises gelatinization temperature, and reduces starch solubility. Via hydrophobic or electrostatic interactions between their lipophilic/hydrophobic domains, proteins may also build compounds containing lipids. Starch and protein are capable of producing combinations, given the right circumstances, that can change the physiochemical characteristics of the two - starch and protein parts.

While examining the pasting properties of sorghum flour, development of fatty acids (FAs), starch and protein ternary complexes were described[59]. With the use of differential scanning calorimetry (DSC), high performance size exclusion chromatography (HPSEC), Xray diffraction (XRD) and multi angle laser scattering (MALLS) has been useful to investigate the interactions among lipids, starch and proteins [59]. Application in the field of nutritional health products and release and delivery of non-hydrophilic drugs has been concluded as the result to the studies in regards of a self-assembled ternary complex which is able to carry sparingly soluble small molecules in the lumen of the amylose helix [59].

Modifications in the pasting and gelatinization capacities of a hypothetical system of starch in the presence of FAs and/or β-lactoglobulin (LG) have been observed in order to clarify the development and configurations of the starch-protein-lipid group [59]. An array of XRD, Fourier Transform infrared spectroscopy (FTIR), and Raman spectroscopy were used to characterize the multiple scale frameworks of the ternary complexes made from RVA pastes[59]. Additional studies have been done in case of proteins having isoelectric point value of less than pH 6, such as LG and whey protein isolates (WP)[58]

The development of these complexes during manufacturing can have a substantial impact on the nutritional value as well as the quality of meals made of starch. For instance, adding lipids to foods made from wheat at the extrusion process alters the final texture of the product and nutritional value, which is thought to be connected to the development of starchlipid combinations[60]. Starch-lipid-protein combinations are frequently employed as a component in applications in industry in addition to processing owing to their unique functional qualities, such as nongelingbehaviour, high viscosity, and excellent flexibility [60]. starch-lipid-protein complexes may be used as fat substitutes in order to create meals with less calories[60]. Additionally, to deal with the rising prevalence of diet-related disorders and weight gain, starch-lipid-protein complexes, being types of resistant starch can be employed as useful components in food formulations [60]. Considering the targeted delivery and regulated release of the bioactive compounds, vacant regions in the lumen of the amylose helices of starch-lipid-protein complexes could serve as potential transporters for accommodating small molecules with poor solubility in aqueous media [60]. The hydrophobic lumen of the amylose helix of the ternary starch-lipid-protein nanoparticle has been modified to contain the chemotherapeutic medication 5-fluorouracil [60]. Additional study is needed in this domain, particularly on the possible health advantages of starch-lipidprotein complexes, particularly with reference to their gut prebiotic qualities. The unique activity of starch-lipid-protein combinations offers potential for the development of novel uses, as the article mentioned above illustrates.

VII. STARCH IN DEVELOPMENT OF PLANT-BASED ALTERNATIVES AND CONSUMER ACCEPTABILITY

Starches are probably among the next most prevalent natural polymers after cellulose and lignin. Starch is mostly derived from cereals like corn and wheat in the United States. In both the food and industrial sectors, starch is used in a wide range of applications, most of which benefit from its propensity to dissolve in hot water [61]. For instance, starch pastes are used in food as a thickening and stabiliser. In many different goods, including paper, cardboard, medicine tablets, and many more, starch is frequently employed as an adhesive and binder. It also has undesirable characteristics, such as restricted solubility in cold water, a loss of viscosity and thickening power after cooking, a high tendency to retrograde, a low shear resistance, and a low thermal resistance[62]. As a result, starch is frequently mildly chemically altered to adapt its physicochemical and functional qualities for use in food[62]. For applications including food packaging, chemical containers, and mulch films during fumigation, laminated films—which include a central TPS layer that is shielded from moisture by outer polymer layers—might be an efficient barrier material.

Various research and development initiatives have been made throughout the years with the goal of creating blends and composites by combining natural polymers with synthetic polymers. Starch, a type of polysaccharide, is one of these extensively studied polymers. This was made possible by starch's low cost, biodegradability, ease of manufacture, abundance, and status as a renewable resource. Granular starch has also been used to create composite materials with polyethylene, polypropylene, polylactic acid, and many other polymers[63].

Global interest in meat-replacing goods has grown over the past few decades, and experts expect that it will continue to expand in the years to come. As a result, there are more goods on the market that resemble meat or provide customers with protein-rich substitutes. Creating the right texture, flavour, and colour for these items is one of the toughest production problems. Protein (typically derived from wheat and legumes), fat or oil, binding agents, flavours, and colouring additives make up a frequently used combination of components[64]. Starch is frequently included as a minor ingredient alongside other purified ingredients, including protein isolates, and is typically used in small amounts in commercially accessible meat substitutes and actual meat products. **Table 3** lists the ways in which starch is used in the processed meats and meat substitutes that are currently available on the market.

Table 3: Collection of meat and meat replacing products on the market that contain starch

A lot of basic research as well as commercialization have focused on thermoplastic starch (TPS). Starch can be melt processed in an extruder like other more common polymers by adding small amounts (10%-30%) of water and/or plasticizers including glycerol, sorbitol, propylene glycol, urea, and triethylene glycol. TPS-based polymers include drawbacks such as low strength, moisture sensitivity, and propensity for brittleness. The average tensile strength of TPS formulations is less than 6 MPa, which is significantly lower than that of commercial oil-based polymers such cellulose acetate (which has the highest tensile strength), maize starch, kraft paper, etc.

The market for starch-based bioplastics was valued at \$424 million in 2016 by Allied Market Research, with a CAGR of 3.7%. The European Bioplastics Association calculated that starch blends accounted for 840 million pounds, or 18.8%, of the global bioplastics capacity in 2017[65].

VIII. CONCLUSION AND FUTURE DIRECTIONS

As a result of placing a higher priority on health, well-being, and social $\&$ environmental consciousness, the health-conscious buyer market has recently turned to eating more sustainably and naturally, which has raised awareness of "clean-label food". In this context, this review provided an overview of native starches from a range of nonconventional and underutilised sources and their modification using environmentally safe physical, enzymatic, and other approaches. Physical and enzymatic modification processes can significantly enhance the characteristics of native starch by changing its physicochemical properties, functionalities and structural attributes and increasing its technical value as a clean label starch because they are free from artificial and synthetic ingredients. Furthermore, it discusses innovative uses of clean-label starch, such as the production of biodegradable and nano-composite films, starch-protein-lipid complexes, and plant-based alternatives in the food industry.

It is important to look for research possibilities to more fully comprehend and utilise uncommon and underused starches. It is crucial to emphasise the genetic variety of starch characteristics and to conduct comparisons between unconventional and industrially important starches. Such starches should be examined for modifications and uses, and a lifecycle analysis of their manufacturing should be conducted for environmentally friendly production. Relationships between an individual starch's structure, function, and utilisation are crucial for expanding applications. Moreover, Starch-based materials have a lot of potential and demand for usage in applications including fertiliser and water treatment. Therefore, it is necessary for academics and researchers to investigate this direction.

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IIP Series, Volume 3, Book 23, Part 2, Chapter 2

FUNCTIONAL CLEAN-LABEL STARCH: SUSTAINABLE PRODUCTION TECHNOLOGIES AND FOOD APPLICATIONS

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FUNCTIONAL CLEAN-LABEL STARCH: SUSTAINABLE PRODUCTION TECHNOLOGIES AND FOOD APPLICATIONS

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