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Current Research Addressing Physical Modification of Starch from Various Botanical Sources

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Abstract

Physical modification is simple, cheap and safe because it requires no chemicals or biological agents when compared to other methods of starch modification. It is more connected to the emerging concept of 'green chemistry' for environmentally friendly applications. Physical modification of starch can improve water solubility and reduce particle size. The methods involve the treatment of starch granules under different temperature/moisture combinations, pressure, shear and irradiation. Physical modification also includes mechanical attrition to change the particle size of starch granules. Physical modification techniques are generally given preference as they do not involve any chemical treatment that can be harmful for human use. The broad classification of starch physical modification into those that are thermal and others that is non-thermal. The thermal processes involve the ones in which the starch granule structures are destroyed (all pre-gelatinization processes) and the ones in which the granules are preserved (hydrothermal processes: annealing and heat-moisture treatment). In disparity, non-thermal processes are the application of high pressure, sound, pulse electric field and irradiation to alter the physicochemical and functional properties of natives for better applications in the food and non-food industries. Modification of starch is an ever evolving industry with numerous possibilities to generate novel starches which includes new functional and value added properties as demanded by the industry. This review aims to summarize the latest developments and recent knowledge regarding physically modified starches.

Keywords: Pre-gelatinization, Hydrothermal treatment, Annealing, Heat-moisture treatment, Non-thermal modification

Introduction

Starch is undeniably the most important polysaccharide in the human diet. It is only second to cellulose in terms of abundance of organic compounds in the biosphere [1]. The attractiveness of starch usage in the food and non-food industries could be ascribed to its cheapness, abundance, biodegradability and non-toxic nature. Starches are easily obtained from various botanical sources, e.g., cereal, legume, root and tuber and green fruit [2]. The need for native starch (NS) modification is due to the inherent deficiencies in its properties. Native starches (NSs) are insoluble in water, easily retrograde with associated syneresis and most significantly gels and pastes produced by NSs are unstable at high temperature, pH and mechanical stress. Due to these inherent NS inadequacies, there is need for modification to better the functional and physicochemical properties for suitable industrial applications. Modification of starches can be broadly divided into four-physical, chemical, biotechnological and enzymatic or their combinations properly called dual modification [2-6]. Amongst them, physical methods are more acceptable since they are general chemical-free and hence considered safer for human consumption [7]. Physical modification of starch is more connected to the emerging concept of 'green technology' or 'sustainable technology' for environmentally friendly applications [8]. Physical modification could be generally classified into thermal and non-thermal modification. The thermal modification consists of pre-gelatinization and the hydrothermal processes-annealing (ANN) and heat moisture treatment (HMT). In pre-gelatinization, the granular structure of starch is totally destroyed as a result of heating, there is de-polymerization and fragmentation and so the molecular integrity of the starch is not preserved. In disparity, ANN and HMT involve heating starch in water at a temperature below the gelatinization temperature (GT) and above the glass transition temperature (Tg). Consequentially, the granular structure of starch is preserved.

The physical non-thermal processes involve methods dealing with the preservation

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of food as a result of their impact on microbial organisms that cause fermentation. These are processes that use pressure, ultrasound (US), pulsed electric field (PEF) and radiation to manipulate the physicochemical and functional properties of starches. Generally, the purpose of starch modification is to better its properties for various applications. The aim of this overview is to discuss the recent trend in the physical modification of various starches. Furthermore, unit operations, physicochemical properties and applications of the various physical modification methods will also be discussed.

Physical Modification of Starch

The physicochemical and functional properties of native starches must be altered and modified in order to meet the demand for industrial applications. Most starch granules are inert, insoluble at ambient temperature, highly resistant to enzymatic hydrolysis and unstable under various temperatures, shears, pH and therefore lack specific functional properties [2]. To make the NSs convenient for industrial applications, these starches are often modified to acquire desired properties such as solubility, heat tolerance, texture and adhesion [9-11]. Broadly, physical modification of various starches can be classified into thermal and non-thermal processes. Another classification of physical modification is based on whether starch granular structures are destroyed or preserved. The thermal processes are pre-gelatinization and hydrothermal processes (ANN and HMT). In the former the starch molecular integrity is smashed and destroyed, but they are preserved in the latter (ANN and HMT). Non-thermal physical modification of starches involves the utilization of high hydrostatic pressure (HHP) [12,13], ultrasound (US) [14-16], pulsed electric fields (PEF) treatment [17,18] and microwave treatment [19] to alter the physicochemical and functional properties of starches in order to achieve desired products. There are many merits associated with physical modification, it is simple, cheap and does not involve the introduction of chemicals or biological agents into the modified starch [2,20]. Rapid development is taking place in the field of physical modification as a result of the above stated quality linked to it. Recent trend in physical modification are as follows: corona electrical discharges [21], PEF treatment [18], micronization in vacuum ball mill [22], mechanical activation with stirring ball mill [23], instantaneous controlled pressure drop (DIC) process [24], multiple deep freezing and thawing [25], osmotic pressure treatment [26], thermally inhibited treatment (dry heating) [27], superheated starch [28] and iterated syneresis [29]. These new physical methods of starch modification are summarized in Table 1.

Table 1: Recent physical modification of starch

Physical modification Reference
Pulsed electric field treatment Han et al. [18]
Corona electrical discharges Nemtanu and Minea [21]
Micronization in vacuum ball mill Che et al. [22]
Mechanical activation with stirring ball mill Huang et al. [23]
Instantaneous controlled pressure drop (DIC) process Maache-Rezzoug et al. [24]
Multiple deep freezing and thawing Szymonska et al. [25]
Osmotic pressure treatment Pukkahuta et al. [26]
Thermally inhibited treatment Chiu et al. [27]
Superheated starch Steeneken and Woortman [28]
Iterated syneresis Lewandowicz and Soral- Smietana [29]

Pre-gelatinized starch (PGS)

PGSs are starches that have undergo gelatinization and consequently are depolymerized, fragmented and the granular structure is entirely destroyed as a result of cooking [2,30,31]. The pre-gelatinization process is achieved by drum drying, spray drying and extrusion cooking. The properties associated with PGS permits instant dissolution in cold water without heating. Due to the harsh treatment (gelatinization and severe drying) used to obtain PGS, it is porous, possessed higher water absorption index (WAI) and water solubility index (WSI) than that of the NS [32]. PGS has been reported to be amorphous when studied by X-ray diffractomtery (XRD) and the irregular starch granules of the NS altered to concave spherical shape by pre-gelatinization as revealed by scanning electron microscopy (SEM). The higher WAI and WSI values of PGS when compared to the NS were ascribed to their higher macromolecular disorganization, degradation and weaker associative forces between them [2].

PGS with various degrees of gelatinization and degradation could be obtained through extrusion [8]. According to the latter author, drum-dried starch possessed much increased water absorption, swelling and solubilization than other methods of PGS production and usually accompanied by decreased apparent viscosity.

There are certain limitations associated with PGS which have reduced its applications in certain foods [4]. These include grainy texture, inconsistent and weak gels. These demerits have been surmounted by the development of granular cold water swelling starch (GCWS). The latter can exhibit cold water thickening despite keeping its granular integrity, it possess higher viscosity, more homogeneous texture with higher clarity and has more processing tolerance than PGS [33,34]. In disparity to native starch, PGS and GCWS can rapidly absorb water and increase their viscosity at ambient temperature [4]. This useful functionality have made them applicable in a range of products synthesized at low temperature containing heat-labile components (e.g., vitamins and coloring agents) and instant food [4]. The PGS and GCWS are also applied in cold desserts, instant baby foods, pie filings, gravies, soups and sauces [35-37].

Undeniably, the functional and physicochemical properties of various modified starches determine their applications in the food industry. PGSs have been solely utilized as thickener in many instantaneous products, such as baby food, instant soups and desserts [32], due to its ability to immediately form pastes when dissolve in cold water [31]. Due to the above reason, PGS is also favored in the making of thermally sensitive foods [4].

Hydrothermal modifications

ANN and HMT are the two hydrothermal treatments that modify the physicochemical properties of starch, without destroying the granular structure [38]. The similarity of both processes is that they take place at a temperature above the Tg and below the GT. So the structural molecular integrity of the starch granules is preserved in both cases because they are operating at a temperature that is below the disorder temperature-GT. They differ in the water content and temperature associated to both processes. There is a need to distinguish between ANN and HMT for purpose of clarity. ANN is the treatment of starch in excess

(> 60% w/w) or at intermediate (40-55% w/w) water content, while starch treatment below 35% (w/w) water content is appropriately called HMT [38]. The hydrothermal modification has significant effect on starch functionality. The following physicochemical properties are affected by both hydrothermal processes; granule morphology and crystallinity, double helix content, amount of amylose-lipid complexes, gelatinization and pasting, swelling power (SP) and solubility, gel properties and susceptibility to acid and enzymatic hydrolysis [38].

Annealing (ANN): ANN involves the heating of starch granules in excess water (76% w/w) or at intermediate water content (40% w/w) and held at a temperature below the GT and above the Tg [39-43]. Many studies by researchers have shown that annealing resulted in alterations to starch structure (increase in granular stability, starch chain interactions[within amorphous and crystalline domains of the granule], perfection of starch crystallites, formation of double helices and compartmentalization of amylopectin-amylopectin (AP-AP), amylose-amylopectin (AM-AP) and AM-AM helices) and properties (elevation of starch GTs, narrowing of GT range, decrease in swelling factor and AML and increase in hot and cold paste viscosities) [43]. It has been documented that ANN of lentil, smooth pea and wrinkled pea starches decrease granules swelling and amylose leaching (AML), and increase GTs, thermal stability and susceptibility towards digestion by alpha-amylase [44,45]. These authors ascribed the alterations to increase in crystalline perfection and increased interaction between AM-AM and AM-AP chains.

Most annealed starches are from potato, rice, wheat, maize, sago, pea and cassava. ANN of fermented and unfermented starches decreased SP and solubility, peak, setback and setback viscosities of pasting [8]. It also brings about an increased in peak temperature, transition temperatures and enthalpy of gelatinization with narrower temperature ranges. The polymorph of annealed fermented and unfermented cassava starch altered from C_A- to A-type [46,47]. Finally in this sub-section a summary of annealed cereal and tuber starches with their associated gelatinization parameters are given in Table 2.

Annealed starches could be utilized in the canned and frozen food industries [52] because they possessed improve thermal stability and decrease in extent of setback, respectively [53, 54]. The properties of annealed starches make it suitable for producing desirable properties in noodles. The physical properties of annealed starches such as decrease in granular swelling, AML and the increase in heat and shear stability [52] has also been used to improve resistant starch levels while preserving granule structure [55].

Heat moisture treatment (HMT): In HMT, the native starch is subjected to heat treatment in the present of limited amount of water (usually 35% w/w) at a temperature above Tg but below the GT. The physical properties of heat moisture treated starches depend on the botanical source of the starch and treatment conditions utilized [2]. HMT brings about alterations in functional properties such as decrease in starch SP, solubility, AML and peak viscosity but increase is observed in the pasting temperature of heat moisture treated starches [56,57].

Other previously documented research indicated that HMT can impact the structure and physicochemical properties of cereal, tuber and legume starches, as observes by important alterations in X-ray diffraction (XRD) pattern, crystallinity, granule swelling, amylose leaching, gelatinization parameters, viscosity, thermal stability, rheological characteristics, acid/enzyme susceptibility [58-62], retrogradation and pasting parameters. Generally, heat-

Table 2: Annealing and gelatinization parameters for cereal and tuber starches

	ANN parameters				Gelatinization parameters					
Starch source	Temp(°c)	Steps	Time (h)	W:S[a)]	S:W[b)]	T_o[c)](°c)	T_p[c)](°c)	T_c[c)](°c)	Δ*H* (J/g)	Δ*H* (J/g)
Waxy barley (native)	-	-	-	-	1:04:05	53	59	68.6	10.8	Qi et al. [48]
Waxy barley (annealed)	48	single	168	Excess	1:04:05	65.4	67.4	73.3	11	Qi et al. [48]
Normal maize (native)	-	-	-	-	1:05	56.9	67.7	76.5	17.6	Qi et al. [48]
Normal maize (annealed)	55	single	168	Excess	1:05	72.1	76.4	82.3	16.9	Qi et al. [48]
Waxy maize annealed)	55	single	168	Excess	1:05	73.3	77.3	83.1	17.3	Qi et al. [48]
Wheat (native)	-	-	-	-	1:03	58.4	63.2	69	100	Shi [41]
Wheat (annealed)	52	single	4	3;01	1:03	67	69.4	73.2	8.8	Shi [41]
Waxy rice (native)	-	-	-	-	1:03	76.2	81.2	87.1	19.2	Shi [41]
Waxy rice (annealed)	70	single	48	3:01	1:03	86.9	90	93.7	19.6	Shi [41]
Normal rice (native)	-	-	-	-	1:03	67.7	73.5	78.7	13.3	Hormdokand Noomhorm [42]
Normal rice (annealed)	55	single	24	3:01	1:03	71.1	74.7	79.1	11	Hormdokand Noomhorm [42]
Normal barley (native)	-	-	-	-	1:03	61.3	65.3	72.8	10	Waduge et al. [43]
Normal barley (annealed)	50	single	72	3:01	1:03	66.7	69.8	77	10	Waduge et al. [43]
Waxy barley (native)	-	-	-	-	1:03	59.3	64.9	81.8	13	Waduge et al. [43]
Waxy barley (annealed)	50	single	72	3:01	1:03	66.2	70.2	82.9	13.4	Waduge et al. [43]
Potato (native)	-	-	-	-	-	59.1	61.9	66.8	18.3	Vermeylen et al. [39]
Potato (annealed)	51	single	24	2.01	-	64.9	66.8	71.1	18.3	Vermeylen et al. [39]
Potato (annealed)	50	single	72	2.01	-	61.1	63.1	70.7	18	Kohyama and sasaki [40]
Potato (annealed)	40	multi	24	3.01	-	-	-	-	-	Nakaazara and Wang [50]
Cassava (native)	-	-	-	-	-	65.4	71.5	81.5	8.8	Atichokundomchai et al. [51]
Cassava (annealed)	51	single	72	3.01	-	70.7	74	81.3	9.4	Atichokundomchai et al. [51]

- data not reported; a) water:starch; b) starch:water; c) Onset (To), midpoint (Tp) conclusion (Tc) temperatures of gelatinization; ΔH Enthalpy of gelatinization.

moisture treated starches tended to bring about higher GT, lower paste viscosity, a decrease in granular swelling and an increase in thermal stability [10,63]. The most important reported effect of HMT was the shift in crystalline structure from B- to A-type for potato starch [39] and yam starch [64] and a transition from C-type to A-type for sweet potato starch [65]. However, some starches were resistant to changes in crystallinty due to HMT. Such examples were normal corn starch [44] and rice starch [59].

In another study on breadfruit starch, after its subjection to HMT, there was no apparent change in starch granule morphology, but decreased was observed in the molecular weight and an increased in the AM content of the modified starch [66]. The HMT-modified breadfruit starch was more thermally stable than the native starch. The increased enzyme resistance of the physically modified starch was ascribed to the rearrangement of molecular chains, more compact granule structure [66] and more perfect crystalline structure.

The baking quality and freeze-thaw stability were improved when heat-moisture treated potato starch is used to replace maize starch [67,68]. The excellent freeze-thaw stability and organoleptic properties of heat-moisture treated cassava starch is utilized in pie-filling [69]. At the industrial level, heat-moisture treated starches also find applications in the preparation of infant foods [24]. HMT promotes retrogradation and formation of RS3 (Singh et al., 2005). RS3 is type III resistant starch that is formed through a retrogradation mechanism due to processing in this case and can also be produced by intentional modification [70].

Non-thermal physical modification of starches

In a world that is asking for environmental sustainability and food security, innovation is also a key for the sustained growth of the food industry [71]. Such innovative food processing technologies have advantage of physical phenomena (unit operations) like high hydrostatic pressure (HHP), ultrasound (US) and pulsed electric field (PEF) treatment [72]. Some of the merits of these emerging technologies include decreased energy utilization, less consumption of water and extended shelf life of processed food thereby enhancing global food security [71, 72]. These technologies cause biological, chemical and physical modifications leading to alterations in sensory, textural and nutritional properties [73]. Furthermore, these non-thermal technologies (HHP, PEF and US, etc.) also retain freshness, nutritional value and sensory characteristics of food items without any significant thermal degradation [71].

Non-thermal physical modification is an alternative to the traditional heating processes [2, 30, 31]. The high-energy traditional thermal treatments usually diminish cooking flavors and cause loss of vitamins and essential nutrients in the desired product. The concept of non-thermal treatment was born to minimize the demerits inherent in traditional heating. Compared to traditional thermal processes, the non-thermal processes kills most pathogenic or spoilage micro-organisms and inactivate enzymes, but minimize the loss of color, taste, texture, nutrients and heat labile functional components of food [2].

Some of the non-thermal processes are conducted at high hydrostatic pressure (HHP) [12, 13], using ultrasound effect [15, 74], pulsed electric field treatment [18], and microwave treatment of starches [19] from different botanical sources. It was reported that various non-thermal treatments impact the physicochemical properties of starch differently.

High hydrostatic pressure (HHP) is a non-thermal food processing technology that takes away some demerits of conventional thermal processing by decreasing undesirable chemical reactions which may results to undesired organoleptic properties and also impose nutritionally adverse effects [75]. High pressure involves using a uniform pressure throughout a product. In the food industry, pressures ranging from 400 to 900 MPa can be used [2]. Pivotally, HHP treatment inhibits SP of starch granules, so that their viscosity is lower than heat processed starches [76]. Additionally, starch gelatinization is obtainable at ambient temperature or below 0^0C with HHP treatment of starches from various botanical origins [2]. The molecular integrity and orderliness of potato starch treated with 600 MPa for 3 min was slightly distorted by HHP, nevertheless the crystallinity of the starch was preserved. The position of localization of amorphous and semi-crystalline growth rings within the starch granules of waxy starch is not significant when subjected to high pressure treatment (650 MPa/9 min) because disruption and complete gelatinization will obviously take place [77]. A possible application of pressurized starch could be to utilize it as a fat substitute, the starch granules might stimulate fat droplet as they are considered as micro-particles of well-defined size distribution [78].

Ultrasound (US) food processing technology use frequency in the range of 20 KHz to 10 MHz [79]. US is the sound that is above the threshold of the human ear (>18 KHz). It is produced with either piezoelectric or magnetostrictive tranducers that generate high energy vibrations. These vibrations are amplified and transferred to a sonotrode or probe, which is in direct contact with the fluid [80, 81]. Some merits as a consequent of US utilization in food processing are processing time reduction, energy efficiency and eco-friendly process [82]. Other advantages of US are reduction of processing temperature, batch or continuous process can be utilized, increased heat transfer, deactivation of enzymes and possible modification of food structure and texture [83]. The US methods have been applied to several kinds of starch (sweet potato, tapioca, potato and corn) and polysaccharides [74]. When native corn starch was subjected to HPU treatment (24 KHz), the crystalline region of the modified corn starch granules was observed to be distorted [80]. `An increase in SP, solubility and disruption of crystallinity of starch granules as studied by X-ray diffractometry were observed as a result of subjection of native granules to US treatment [84]. The best way for molecular weight reduction of polysaccharides such as starch and chitosan is to treat their aqueous solution with 360 KHz US [14]. The degradation of starch by applied US has been ascribed to OH radical formation and mechanochemical effects. High power ultrasound (HPU) is very significant in the following fields of food processing; filtration, crystallization, homogenization, extrusion, de-foaming, viscosity alteration, separation, emulsification and extraction. These unit operations are very important in the separation of gross product into its various components. Other applications of ultrasound include inactivation of enzymes and bacteria by splitting their cell membranes due to the violence of cavitation and the production of free radicals [2].

Pulsed electric field (PEF) technology is non-thermal food preservation methods which kills pathogens or spoilage micro-organisms and inactivate enzymes and minimize the loss of taste, color, texture, nutrients and heat labile functional components of foods [85]. Other merits associated to PEF are that it kills vegetative cells, no toxicity was detected and short treatment time was also observed [71]. Recent studies documented by Hans et al. [18] on PEF shows that various treatments affect the physicochemical properties of starches differently. When corn starch –water suspension were processed in PEF with electric field strength of 50 KV/cm [18], the following results were obtained. The GT and enthalpy of the modified corn starch decrease with an increase of electric field strength. Additionally, the starch lost granule shape and the crystallinity degree were decreased significantly. Meanwhile, the peak, breakdown and final viscosities of the modified corn starch were decreased with increasing electric field. The applications of PEF in the food industries resulted in food spoilage reduction, enhance food safety by increased shell life and retains freshness of food commodities [86].

Conclusion

The importance of physically modified starches cannot be overestimated. They are generally safe and do not involve the addition of chemicals or biological agents when compared to chemical or genetical modification. The classification of physically modified starches is twofold; either based on the preservation of starch granules or its destructurization. The other classification depends on thermal or non-thermal applications to alter the physicochemical properties of starch granules. The thermal division involves pre-gelatinization and hydrothermal processes. The pre-gelatinized starches (PGSs) were obtained by subjecting the native starches to harsh treatment and drying processes. Therefore, PGSs are gelatinized, depolymerized and fragmented so that they are easily soluble in water at ambient temperature. On the other hand, hydrothermal modification involves two processes-annealing (ANN) and heat moisture treatment (HMT). Both processes consist of heating native starches in water at a temperature above the grass transition temperature but below the gelatinization temperature (GT). In ANN and HMT, since the temperature of de-structurization (GT) is not exceeded, the starch granules are preserved and movement is mostly restricted to the amorphous region in the granules. In disparity, the non-thermal processes involve the utilization of high hydrostatic pressure, ultrasound and pulsed electric field treatments to bring about alterations in the physicochemical properties in starch granules.

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