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Recent progress of starch modification assisted by ultrasonic wave

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Abstract

Starch is a kind of natural polymer, which is the main energy source for human body and important industrial raw materials. As a new non-thermal processing technology, ultrasonic has the advantages of green energy-saving, safety, high efficiency and convenient application. It plays an important role in starch modification process. In this paper, the effects of ultrasound on the multi-scale structure and physicochemical properties of starch were reviewed. The application of ultrasonic synergistic modification in starch modification was also analyzed, which is expected to provide some reference for the green modification of starch and its industrial production and application.

Keywords: ultrasonic; starch; enzymatic hydrolysis; synergistic modification.

Practical Application: Provide some reference for the green modification of starch and its industrial production.

1 Introduction

Starch is a kind of natural polymer, which has a series of advantages, such as wide source, renewable, low cost and so on, it is widely used in food, chemical industry, textile, medicine and other fields (Gilet et al., 2018). However, due to the limitation of structure, natural starch has many deficiencies, such as insoluble in cold water, easy aging, and not easy to react with other substances (Maniglia et al., 2021). Therefore, it is important to modify starch to provide starch with special good properties for food processing and industrial production (Luo et al., 2022; Moreira et al., 2022).

It is a research hotspot in the field of starch deep processing to seek new effective modification methods to meet the consistent application needs. As a new non-thermal processing technology, ultrasonic has become a hot spot in the field of food processing because of its advantages of green energy-saving, safety, high efficiency and convenient application (Araújo et al., 2022; Chew & Ali, 2021; Xu et al., 2022). Ultrasonic treatment is mainly used to modify starch by mechanical effect and cavitation effect, and affects the surface structure, crystal structure and amorphous structure of starch grains, which can degrade starch and reduce the gelatinization viscosity of starch. The degradation efficiency of amylose is more obvious, which can endow starch with some good properties. The application of ultrasonic technology in starch modification has been widely concerned, and gradually become a research hotspot in the field of starch modification.

This paper discusses the application of ultrasonic technology in starch modification, points out the key factors of the effect of ultrasonic on starch modification, and clarifies the mechanism of the effect of ultrasonic on starch modification efficiency. The aim of this study is to provide theoretical guidance for the highly efficient directional modification of starch by ultrasonic technology, and to promote the development of starch deep processing industry.

2 Effect of ultrasonic pretreatment on starch structure

Ultrasound is a physical modification method that utilizes frequencies higher than the threshold of human hearing. In the starch-water system, the physical effects (such as hole effect, mechanical effect, etc.) generated by ultrasonic treatment will produce strong shear force, high temperature and free radicals in the local area, which will change the multi-scale structure of starch (as shown in Table 1). The appearance of cracks and holes on the surface of starch particles induces changes in the morphology/ultrastructure of polymers (Yang et al., 2019a), thus changing the structure and properties of starch (Zhu, 2015).

2.1 Effect of ultrasonic pretreatment on morphology and crystal structure of starch granules

Ultrasonic treatment can induce cracks and pores in starch granules, which affect the multi-scale structure of starch and then affect its functional properties (Sujka & Jamroz, 2013). Ultrasonic treatment at T15 and T30 for 15 and 30 min on different sources of cereal starch (wheat, barley, rice and maize) resulted in the formation of depressions and pores on the surface of starch granules, but had little effect on their overall integrity (Kaur & Gill, 2019). Zuo *et al*. showed that low-power

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Table 1. Effects of ultrasonic wave on starch modification.

Starch type	Modification conditions	Main results	Reference
arrowhead tuber starch	tri-frequency power ultrasound (20/40/60 kHz)	The solubility, water-holding and oil-holding capacity are significantly increased	(Raza et al., 2021a)
Starch-lipid complex	mono: 20 kHz, 40 kHz, 60 kHz; dual: 20/40 kHz, 40/60 kHz, 20/60 kHz, and tri: 20/40/60 kHz)	Ultrasonic treatment may contribute to the formation of the complex and present a dense network structure	(Raza et al., 2021b)
rice starch	150, 300, 450 and 600 W, 20 min	Changed the morphology and physical properties of rice starch, rather than fine structure	(Yang et al., 2019c)
potato starch	60, 105 and 155 W, 30 min, 20 kHz	Ultrasonic treatment can form grooves on the surface of starch grains, and has a great effect on the cluster structure, especially on the crystalline region	(Zhu et al., 2012)
corn and cassava starch	High power ultrasound with short time	Grooves and notches appeared on the surface of starch granules with crystallinity and calorific value decreased.	(Rahaman et al., 2021)
maize starch and potato starch	500 W, 60 min, 20 kHz	The cavitation effect of ultrasonic makes the inner space of starch granules loose, the structure destroyed and the content of damaged starch increased	(Wang et al., 2022b)
plantain (large granule size) and taro (small granule size) starches	25 kHz, 80 W (20, 50 min)	Ultrasonic treatment had little effect on the particle size distribution of starch, but it had more effect on the larger particle size of starch	(Carmona-García et al., 2016)

ultrasound treatment (2.5 and 4.1 W) reduced the particle size of waxy rice starch, whereas there was no significant change in starch granule surface and starch molecular weight (Zuo et al., 2009). However, the high-power ultrasonic treatment destroyed the granule morphology of kiwifruit starch, formed pores and cracks on its surface, and reduced the starch particle size and short-range molecular order (Wang et al., 2022a).

Under the ultrasonic pretreatment system, the amorphous area of starch particles is more easily destroyed by ultrasonic than the crystalline area, and cracks and pores are formed on the surface of starch particles, which improves the uniformity of starch particles, the increase of ultrasonic power can make starch granule smaller, granule deformation and surface roughness (Aijun et al., 2011). When corn starch was treated by ultrasound, Han *et al*. found that the amorphous region was degraded first, and then the crystalline region was degraded, resulting in the continuous decrease of relative crystallinity (Han et al., 2019). The proportion of B1, B2 and B3 in waxy corn starch after ultrasonic treatment was lower, while the proportion of chain A was higher. The distribution of chain length, double helix, single helix and amorphous form of pre-treated starch, especially α-1, 4-glycosidic bond and α-1, 6-glycosidic bond had great influence (Yang et al., 2019b). Falsafi *et al*. reported that ultrasonic irradiation using a horn sonicator or an ultrasonic bath reduced the crystallinity of oat starch, while the crystal structure of A-Type remained unchanged (Falsafi et al., 2019).

The efficiency of ultrasonic treatment is affected by many factors, such as ultrasonic treatment power and frequency, treatment time and temperature, and the properties of starch suspension, i. e. starch concentration and source (Falsafi et al., 2019; Sujka, 2017). The starch granules of potato, wheat, corn and rice were suspended in water or ethanol and treated by ultrasonic (170 W, 30 min), especially in potatoes and wheat starches (Sujka & Jamroz, 2013). In recent years, it has been found that starch treated by dual-frequency ultrasound has more obvious damage than starch treated by single-frequency ultrasound (Hu et al., 2015).

2.2 Effect of ultrasonic pretreatment on molecular weight of starch

Molecular weight is one of the important basic parameters for studying the molecular structure of starch, which directly affects the physicochemical properties of starch. When Sonication in starch, the molecular structure and rheological properties of starch were significantly affected. Ultrasonic irradiation is a novel method for degrading polymer compounds, particularly in view of the reduced molecular weight, which is achieved by splitting the most susceptible chemical bonds without causing any change in chemical property (Gogate & Prajapat, 2015). The reduction of molecular weight due to ultrasound-induced chain breakage is attributed to the cavitation of local high pressure, high temperature and shear stress, which leads to starch chain breakage, the resulting starch dextrin has a smaller molecular size and narrower molecular weight distribution (Zhu, 2015). The degree of decrease in molecular weight decreases with time. The molecular weight of ultrasonically degraded starch has a certain limit. When the molecular weight of the starch sample is close to the limit, the ultrasonic effect is weakened.

The higher the ultrasonic intensity, the faster the rate of molecular weight change. The higher the temperature of starch paste, the higher the movement ability of the molecule, the molecular resistance of potato starch paste was reduced, and the change of molecular weight of potato starch paste in ultrasonic field was restrained (Li et al., 2017a). After ultrasonic treatment, the relative molecular weight (Mn, Mw, Mp) of potato starch paste decreased to a certain extent, and increased with the extension of the treatment time (Li et al., 2017b). In summary, ultrasonic degradation of starch, resulting in starch surface holes, molecular weight distribution tends to reduce, viscosity and other properties of change, to enhance the subsequent process of saccharifying enzyme degradation of starch (Haiming et al., 2011).

3 Effect of ultrasonic pretreatment on physicochemical properties of starch

The change of multi-scale structure of ultrasonic pretreatment starch will inevitably lead to the change of swelling, gelatinization, rheology and other physical and chemical properties (Zhu, 2015). Ultrasonic technology can create and improve the functionality and stability of starch-based products, and improve the swelling and solubility of starch from different grains. Moreover, rapid digestible starch (RDS) and resistant starch (RS) significantly increase with time, after ultrasonic treatment, the in vitro digestibility and the content of resistant starch increased, which was related to the changes of starch particle size, crystallinity, physicochemical properties and starch structure (Kaur & Gill, 2019). Starch granules of potato, wheat, maize and rice were suspended in water or ethanol and treated with ultrasound (20 kHZ, 170 W, 30 min), the starch treated by ultrasonic has higher liposuction and water absorption capacity, lower viscosity and higher solubility and expansion. In addition, ultrasonic treatment of potato starch shows a high degree of pasty clarity, which is a desirable property in many food applications. The results also showed that water was better than ethanol as the medium for ultrasonic treatment of starch (Sujka & Jamroz, 2013). Similarly, Sujka *et al.* found that ultrasonic treatment of potato, wheat, corn and rice starch in water and ethanol causes changes in particle porosity, and that ultrasonic treatment in water-based reaction systems, all starches showed significant changes in specific surface area, but only potato starches showed significant increases in specific surface area in the ethanol system (Sujka, 2017).

The solubility, swelling degree, peak viscosity, final viscosity and retrogradation value of wheat starch increased with the increase of ultrasonic treatment time because of the destruction of the aggregation structure of starch, however, the gel strength of starch paste decreased with the decrease of disintegration value (Ying, 2019). Monroy *et al.* ultrasonic-treated tapioca starch changed at the molecular level, reflecting the stability of starch paste under freezing conditions (Monroy et al., 2018). The peak viscosity and gel properties of corn starch treated by ultrasonic wave with different frequencies (20 kHz, 25 kHz and 20 kHz + 25 kHz) were significantly decreased, and its thermal stability and retrogradation were enhanced, while its cold stability was unchanged, the peak viscosity decreased with the increase of ultrasonic frequency (Hu et al., 2015). Ultrasonic treatment reduced the peak viscosity of potato, Xiaomi and waxy corn starches, and the lower viscosity may be attributed to physical damage to the starch granules, increasing the water permeability of hydration (Han et al., 2019; Yang et al., 2019b). Mechanical oscillation and cavitation during ultrasonic treatment can lead to long chain breakage and decrease the interaction force between starch particles, thus resulting in the decrease of viscosity (Wang et al., 2022c). In addition, ultrasonic-assisted enzymatic treatment is more effective in improving the solubility of starch, mainly because it degrades the structure of loose starch granules and makes it easier for water to enter the reticular structure. Enzyme treatment and ultrasonic treatment can effectively reduce the molecular weight of starch, thus improving the solubility of starch in water.

4 Enzymatic hydrolysis of starch enhanced by ultrasonic pretreatment

Ultrasound can accelerate enzyme reactions by acting on different targets. It can modify enzymes and substrate macromolecules, which helps to improve enzyme activity and product yield, the synergistic effect of ultrasound and enzymes has been widely reported to improve the catalytic rate (Wang et al., 2018). For many years, ultrasound has been used as a method to inactivate enzymes, and some work has shown that under mild conditions, ultrasound does not inactivate all enzymes, but has a positive effect on enzyme activity, can be used to speed up the enzyme reaction (Nguyen & Le, 2013). Ultrasonic pretreatment of starch shortened the time of liquefaction, significantly increased the dextrose equivalent (DE) value of saccharification process, and increased the rate constant of amylase hydrolysis (Li et al., 2018). Therefore, the suitable ultrasonic pretreatment conditions can create better conditions for enzymatic hydrolysis of corn starch. An evaluation of the kinetics of enzyme kinetics and starch degradation showed an increase in reaction rate and enzyme-substrate affinity. According to the thermodynamic results, ultrasonic enzymatic hydrolysis requires less energy than enzymatic hydrolysis (Wang et al., 2017). Ultrasonic irradiation (0.5 W/cm^2, 40 kHz) did not affect the optimum temperature and pH value of the enzyme, and improved the thermal stability of the enzyme, and increased the activity of alliinase by about 47.1% (Wang et al., 2011). Wang *et al.* studied the effect of ultrasonic treatment on the activity of glucoamylase. The activity of glucoamylase increased by 21.07% and the value of Vmax and Km increased after ultrasonic treatment at 60 °C (420 W, 10 min), Ea, ΔG, ΔH decreased. The results showed that the content of tryptophan and tyrosine on the surface of saccharifying enzyme increased by 17.8% and 12.41%, respectively (Meng et al., 2018). The results showed that ultrasonic treatment could change the activity of glucoamylase, mainly by changing the conformation of glucoamylase.

Collectively, low intensity ultrasound has positive effect on enzyme, while high intensity ultrasound has negative effect on enzyme. When enzymes and substrates are exposed to an ultrasonic field, structural changes occur and the aggregation of enzymes and substrates is destroyed, making them easier to connect. At the same time, the binding site of the enzyme can be better exposed to the substrate (Wang et al., 2018).

5 Ultrasonic coupled with other methods to synergistically modify starch

In order to improve the properties and uses of starch, double modification was introduced to optimize the function of single modified starch (Table 2). Double modification method, also known as synergistic treatment, is modified natural starch by two methods. The combination of ultrasonic and high-pressure treatment had great effects on the structure and physicochemical properties of pea starch, which could increase the content of

Table 2. Effects of ultrasonic combined treatment on structure and properties of starch.

Starch type	Modification combination	Results	Reference
mung bean starch	ultrasound combined with plasma treatment	The relative molecular weight of starch, long chain and crystallinity of amylopectin were decreased while the gelatinization temperature and gelatinization heat were increased.	(Shen et al., 2021)
Agriophyllum squarrosum starch	Ultrasound and heat-moisture treatments	The content of amylose, molecular weight, swelling potential and solubility of amylopectin decreased, while the content of resistant starch increased significantly.	(Han et al., 2021b)
potato starch	Ultrasound and freeze-thaw treatments	The composite treatment showed the coarsest structure, and the oil absorption increased from 59.62% (natural starch) to 80.2%.	(Wang et al., 2020b)
wheat starch	microwave-ultrasound	The structure of the granules showed obvious irregularity and fragmentation, and the content of resistant starch increased	(Zhang et al., 2021)
foxtail millet starch	combining physical, chemical and enzymatic methods	The swelling potential of modified starch decreased, and with the increase of modification times, the swelling potential decreased.	(Dey & Sit, 2017)
Pea starch	ultrasound and heat-moisture	The crystallinity, relative molecular weight and swelling potential of starch were decreased	(Han et al., 2021a)
Rice starch	vacuum combined ultrasound	vacuum combined with ultrasonic treatment can lead to serious shrinkage and damage of starch granules.	(Li et al., 2022)

resistant starch and reduce the blood Glycemic index of food to a certain extent, at the same time, the heat processing stability of modified starch was improved (Zhou et al., 2019). Ultrasonic treatment alone had no effect on the physical properties of corn starch, while ozone treatment alone or in combination with ultrasonic treatment was proved to be an effective starch modification technique, and the ultrasonic treatment before the ozone action will improve the subsequent ozone action (Castanha et al., 2019). The starch prepared by electrostatic field followed by ultrasound has good functional properties and can obviously improve the degree of substitution of acetylated starch (Cao & Gao, 2020). The effects of ultrasonic and microwave treatment alone and in combination on the physicochemical and functional properties of chestnut starch were studied. The surface damage of ultrasonic and microwave (UM) and microwave-ultrasonic (MU) composite modified starch was more serious, and the relative crystallinity and heat of gelatinization were lower. The UM sample showed the highest oil absorption capacity and MU sample showed the highest water absorption capacity, which provided reference starch for potential industrial applications of ultrasonic and microwave treatment of chestnut starch (Wang et al., 2020a). Dual physical modification is more effective than single physical treatment. For example, compared with single ultrasonic treatment or freeze-thaw cycle treatment, potato starch has higher oil absorption capacity after dual ultrasonic treatment and freeze-thaw cycle treatment (Wang et al., 2020b).

In recent years, some emerging processing technologies coupled with ultrasonic technology have made progress in starch processing modification. However, the most achievements are still in the laboratory or pilot stage, which requires the research and development of relevant equipment for industrial production scale. In addition, the integration of artificial intelligence and ultrasonic is helpful to boost the innovative application of ultrasonic technology in starch processing.

6 Perspectives

Ultrasonic has the characteristics of green energy-saving and high mass transfer efficiency, which can change the multi-scale results of starch and affect its physicochemical properties. It was also found that low intensity ultrasound had a positive effect on the enzymatic hydrolysis of starch, while high intensity ultrasound had a negative effect on the enzyme. In addition, in order to meet the needs of industrial starch processing diversity, ultrasonic modification with other methods is also emerging, but the coupling mechanism of different modification methods need to be further explained.

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