Effect of potassium alum addition on the quality of potato starch noodles

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Abstract The objective of this study was to investigate the effects and mechanism of potassium alum (PA) on potato starch noodles to provide a theoretical basis for future searches for new alternatives for PA that are not harmful to the body. The amount of amylose leakage significantly decreased with the PA addition. SEM showed that the honeycomb structure of potato starch noodles with 0.1% PA was diminished. The number of internal pores and average area decreased from 92 and 580.09 units to 72 and 652.58 units, respectively. NMR indicated that the water holding capacity of potato starch noodles containing PA was greater than that of the control. XRD showed that the crystal structure of potato starch noodles was destroyed and that the crystallinity of potato starch noodles with PA increased from 16.65 to 19.64%. G' and G'' increased, but $\tan\delta$ decreased. FT-IR showed that PA caused the O–H stretching vibration in the absorption peak to move in the high-field direction and that the ionic bond was dominant in the PA-potato starch interaction system.

Keywords Potato starch noodles · Potassium alum · Quality · Mechanism

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Introduction

Starch noodles, produced from starch purified from various plant sources, are a major category of Asian noodles. The art of starch noodle making is believed to have originated in China and spread throughout East Asia (Tan et al. 2009; Ge et al. 2014). Potato starch has many unique properties, such as high whiteness, high transparency and viscosity after pasting, and high phosphorus content, and has been widely used as a raw or auxiliary material in the processing of food, especially of noodles (Noda et al. 2006; Kaur et al. 2007; Sandhu et al. 2010). Potato starch noodles, which are mainly comprised of potato starch, are a traditional Chinese food and very popular because of their good taste and smooth and slippery texture (Kim et al. 1996). However, the quality of potato starch noodles made from pure potato starch does not meet consumer demands (Wang et al. 2010). PA ($KAl(SO_4)_2 \cdot 12H_2O$), a naturally occurring sulfate mineral, is always used to improve the textural properties of potato starch noodles and reduce cooking loss. Called “ming fan” in China, PA is a common additive in cereal flour and starch-based products, such as fried bread sticks and noodles (Sasaki et al. 1999; Li et al. 2011), and its chemical name is potassium aluminum sulfate dodecahydrate. However, many studies have indicated that PA introduces aluminum ions, which are harmful to people's health and can lead to Alzheimer's disease, indigestion and other diseases (Buchta et al. 2003; Gomez et al. 2008; Choy et al. 2016).

Therefore, it is imperative to search for new alternatives to PA that are not harmful to human health and could improve the quality of products. Many researchers have investigated the application of PA and found suitable substitutes. Sandhu et al. (2010) added equal proportions of rice and potato starch to produce noodles of

acceptable quality. Muhammad et al. (1999) found that up to a 17% substitution with phosphorylated tapioca starch improved the quality characteristics and decreased the cooking loss of potato starch noodles. Kaur et al. (2005) added glycerol monostearate (GMS) to potato starch noodles, which substantially changed the physicochemical, thermal, rheological, and textural noodle properties. Nevertheless, the quality of potato starch noodles has been improved to only a limited extent with these methods, and these starches modified by means of chemical treatments or chemical additives cannot replace PA completely. Furthermore, these chemicals are unpopular with consumers because of their associated health hazards, and none meet the requirements for green food (Kasemsuwan et al. 1998; Prabhasankar et al. 2007).

Although a substantial amount of quality improvement work has been done, the mechanism by which PA improves the quality of potato starch noodles has rarely been reported (Li et al. 2011). The objective of this study was to determine the effect of PA on the water holding capacity, structure and rheological properties of potato starch noodles. The results of this study are also expected to provide a preliminary explanation for how PA improves the quality of potato starch noodles and contribute to replacing PA with healthy alternatives in the future.

Materials and methods

Materials

Potato starch was procured from Borui Starch Co., Ltd., Dingxi, China. PA was purchased from Shanghai Chemical Reagent Co., Ltd., China. All chemicals used in this study were of analytical grade.

Potato starch noodle preparation

Fifteen grams of potato starch (with a moisture content of 16.01%) with 0.1 g of PA was dispersed in 10.0 mL of distilled water at 50 °C. Then, 45 mL of boiling water was added and stirred to a paste, and another 85.0 g of potato starch was then added to the paste to make the starch dough (with a moisture content of 35%). The starch dough was covered with plastic wrap and allowed to rest for 5 min at room temperature (26–30 °C). Then, the starch dough was extruded into a strip shape using a Hydraulic Pressure Straight Starch Noodle Machine (XingTai DingBang Machinery Manufacturing Co., Ltd, HeBei, China). The starch noodles were boiled for 3–5 s and then soaked in a cold-water bath for 60 min. The starch noodles were drained and placed in cool ventilated air, with a final moisture content of $53.70 \pm 0.23\%$, and used as the

experimental group. Potato starch noodles without the PA addition were used as the control group.

Low-field nuclear magnetic resonance analysis

The water distribution and migration in potato starch noodles were observed with an MRI system (Shanghai Niumag Electronics Technology Co., Ltd., Shanghai, China). Approximately 0.5 g of cooked potato starch noodles wrapped in preservative film was placed in an NMR tube 18 mm in diameter. Transverse relaxation (T_2) was measured using the Carr–Purcell–Meiboom–Gill (CPMG) pulse sequence. The typical pulse parameters were as follows: spectrometer frequency (SF) = 22 MHz, number of sampling points (TD) = 104,492, echo number (NE) = 5000, waiting time of repeat sampling (T_w) = 4000 ms, accumulative frequency (NS) = 8, and echo time (T_E) = 0.205 ms. The test sample height was no more than 20 mm each time (Yu et al. 2018). The signal was collected 3 times and used to improve the signal stability and for data repair. Potato starch noodles without the PA addition were used as the control group.

Scanning electron microscopy (SEM)

Noodle sheets were cut into pieces at a length of 3.0 cm, quickly frozen at -30 °C for approximately 2 h and then vacuum freeze-dried (FD-100 s, China) at a vacuum degree of 1 Pa for 24 h. After freeze drying, dry starch noodles were cross split with a sharp blade; the cross sections were mounted on specimen holders with colloidal graphite, sputter-coated with gold and observed at magnifications of $200\times$ and $500\times$ with a scanning electron microscope (QUANTA FEG, USA). The sizes and numbers of pores in the experimental and control groups were compared after dehydration. The scanning electron microscope images were processed using MATLAB 7.0 image analysis software (MathWorks, USA), and the number and average area of the pores were obtained according to the method provided by Yue (2012).

Determination of amylose leakage

The amount of amylose leakage from starch was determined by the method provided by Zhu et al. (2011). Potato starch and PA were mixed at a concentration of 2% (w/w) (the quality ratio of potato starch and PA was 10:0, 9:1, 8:2, 7:3, or 6:4), and the solution was then gelatinized for 20 min in a boiling water bath. After cooling at 25 °C, the solution was centrifuged at 5000 r/min for 20 min, and the supernatant was diluted with distilled water to 100 mL. Then, 1 mL of the solution was aliquoted and mixed with 1 mL of CH_3COOH (1 mol/L) and 2 mL of iodine solution

(0.01 mol/L) in a 100 mL volumetric flask and brought to a final volume of 100 mL with distilled water. Then, 1 mL of acetic acid (1 mol/L) and 2 mL of iodine solution (0.01 mol/L) were mixed, and distilled water was used to bring the volume to 100 mL as the blank control. All samples were kept away from light at 25 °C for 20 min, after which the absorbance at 620 nm was determined using a UV-2000 UV–Vis spectrophotometer (UNICO, NJ, USA). Potato amylose was used as a standard sample, and the standard curve was $y = 0.30214x + 0.00446$ ($R^2 = 0.98483$). The analysis was replicated at least 3 times.

X-ray analysis

Potato starch noodles were milled into powder in a mortar prior. Then, the powder was sieved through 120 mesh screens. The crystal structure of starch in the noodles was determined according to the method of Waduge et al. (2006) with an X-ray diffractometer (X'Pert PRO, Holland). The test parameters were as follows: voltage, 40 kV; current, 40 mA; sweep range, 5° to 50°; and data acquisition step width, 0.02°.

Infrared spectrum analysis

Potato starch noodle powder was obtained via the X-ray analysis method. The powder (2 mg) was mixed with previously dried spectrograde KBr powder (100 mg). Upon exhaustive grinding, the mixture was pressed into a tablet. The spectrum of the tablet was recorded by a Fourier transform infrared spectroscopy (FT-IR) instrument (TENSOR II, BRUKER, Germany) at 27 °C. The spectra were obtained after 32 scans and ranged from 4000 to 400 cm^{-1} with a resolution of 4 cm^{-1} (Wang, et al. 2018).

Rheological properties of potato starch dough

The starch doughs were prepared as described above. The rheological properties of starch dough were determined using a DHR-2 dynamic rheometer (TA Instruments, USA) fitted with parallel plate geometry (40 mm diameter, 2.0 mm gap). The dough (approximately 1.0 g) was placed in the rheometer measuring system, and the edges were covered with paraffin oil to prevent drying. The doughs were rested between the plates for 5 min before testing to relax the residual stresses (Petrofsky and Hoseney 1995). Doughs were sheared at a 1% strain through a frequency sweep from 0.1 to 10 Hz at a constant temperature of 25 °C (Wang et al. 2018). The results are expressed in terms of storage modulus (G'), loss modulus (G'') and $\tan\delta$ (G''/G').

Statistical analysis

SPSS statistical software (Version 16.0, SPSS Inc., Chicago, IL, USA) was used to certify significant differences by one-way analysis of variance (ANOVA) and T test, and differences were considered significant at $p < 0.05$. The figures were plotted using Origin 8.5.1 software (Microcal Software, Northampton, MA, USA).

Results and discussion

Effect of PA on moisture migration in potato starch noodles

Moisture distribution and moisture migration have direct impacts on the rheology and stability of food. MRI is a powerful tool for characterizing water mobility and distribution in foods during processing and storage (Marcone et al. 2013; Syamaladevi et al. 2016). The atlas can directly reflect the different forms of water in samples, with the transverse relaxation time (T_2) as the abscissa and the value of the NMR signal as the ordinate. Smaller T_2 values are correlated with the water and nonaqueous components in the sample being closer and with smaller degrees of freedom (Bertram et al. 2002; Mortensen et al. 2005; Rondeau-Mouro et al. 2015).

Water and starch importantly affect the mobility of water molecules (Doona and Baik 2007). The effects of the PA addition on water inversion in potato starch noodles are shown in Table 1, which reveals 5 peaks at T_{20} (0.01–0.2 ms), T_{21} (0.66–2.66 ms), T_{22} (4.64–14.17 ms), T_{23} (16.30–132.19 ms), and T_{24} (151.99–533.67 ms) in the experimental group. The control group had only 4 peaks at T_{21} (0.38–1.00 ms), T_{22} (3.05–4.64 ms), T_{23} (12.33–114.98 ms), and T_{24} (132.19–533.67 ms).

T_{20} , T_{21} and T_{22} represented water tightly bound to starch with minimum degrees of freedom. Compared to that in the control, in which the peak area of the tightly bound water in potato starch noodles accounted for approximately 12% of the total area, the PA addition resulted in an increased area of up to 15%, which indicates that PA can increase the water holding capacity in potato starch noodles. T_{23} represented weakly bound water, with a liquidity between that of tightly bound water and free water. The signal to amplitude ratio of the peak area corresponding to the experimental group was approximately 54%, while that of the control group was approximately 55%. These results indicated that the main peak of the experimental and control groups was T_{23} , and the moisture content of potato starch noodles was mainly in the form of weakly bound water. T_{24} represents free water, which has the greatest mobility. The peak area of the experimental

Table 1 The peak area percentage and relaxation time of potato starch noodles with PA (experimental group) and without PA (control group)

	Peak area percentage/%			Relaxation time/ms				
	A ₁	A ₂	A ₃	T ₂₀	T ₂₁	T ₂₂	T ₂₃	T ₂₄
Experimental group	15 ± 0.07 ^a	54 ± 0.03 ^b	31 ± 0.05 ^b	0.01–0.25	0.66–2.66	4.64–14.17	16.30–132.19	151.99–533.67
Control group	12 ± 0.04 ^b	55 ± 0.08 ^a	33 ± 0.10 ^a	–	0.38–1.00	3.05–4.64	12.33–114.98	132.19–533.67

A₁, peak area percentage of bound water; A₂, peak area percentage of loosely bound water; A₃, peak area percentage of free water

Means with different letters in the same column differ significantly different at $p < 0.05$

group was approximately 31%, while that of the control group was approximately 33%. Meanwhile, the starting time of the initial peak in the experimental group was shorter than that in the control group (Table 1), which indicated that the water of the experimental group was more closely bound to the starch and other nonaqueous components. Compared to that in the control, the increase in tightly bound water with the shorter starting time of the initial peak and the decrease in free water indicated that the PA addition resulted in moisture migration from free water to tightly bound water in potato starch noodles. When PA dissolves in water, a large number of Al^{3+} ions, which have a high ionic strength and water-retaining capacity, dissociate (Sasaki et al. 1999) and hence suppress hydration of the amorphous regions of starch granules (Li et al. 2011). As a result, higher amounts of water bound to starch are mainly due to the retainment of water molecules by Al^{3+} .

Effect of PA on amylose leakage from potato starch pastes

During the gelatinization process, starch not only contains water in the interior of the starch granules but also binds to the starch molecules, causing the starch granules to swell and rupture and dissolution of the starch chain (Sandhu et al. 2010). Due to the high branching of the amylopectin molecular structure, very few amylose molecular branches exist, and thus, most of the starch that is dissolved is amylose (Nguyen et al. 1998).

The effects of the PA addition on the amylose content in potato starch pastes with different concentrations of PA are shown in Fig. 1. As the PA concentration in the system increased, the amount of amylose decreased (from 32.74% to 24.85%, 21.29%, 18.40%, 16.64%) significantly ($P < 0.05$, $r = -0.959$). On the one hand, aluminum hydroxide colloid, which has strong adsorption ability, is formed when PA dissolves in water. This colloid adsorbs on the surface of starch granules. As a result, the interaction between starch granules and water molecules is restricted (Nguyen et al. 1998), enhancing the stability and network structure of starch granules, and the pasting temperature of starch increases (Huang 2009). Additionally,

Fig. 1 The amount of amylose leakage in potato starch and PA systems mixed at different ratios. (Means with different letters are significantly different, ($p < 0.05$))

ions (Al^{3+} and SO_4^{2-}) may penetrate deeply into the granule, preferentially acting mainly on the amorphous regions, hindering the water molecules from acting on the amorphous regions (Li et al. 2011). Therefore, starch pasting becomes more difficult, and amylose leakage from starch granules is restricted. On the other hand, there may be some interaction between PA and leaching amylose or competition with starch for water (Lo and Ramsden 2000).

Effect of PA addition on the microstructure of potato starch noodles

As shown in Fig. 2a, b, d, e, the experimental and control groups showed many irregular pores, which might serve as containers for water before freeze drying. Figure 2d, e are different magnifications images of the experimental group potato starch noodles, which showed smaller and denser pores than the control group noodles (Fig. 2a, b). Figure 2c was obtained from Fig. 2a using MATLAB, and Fig. 2f was obtained from Fig. 2d. In Fig. 2c (the control group), 145 pores comprised 8.44% of the entire image, and the average area was 580.09 units. In Fig. 2f (the experimental group), there were 72 pores, which accounted for 4.71% of

Fig. 2 Scanning electron micrographs (SEM) and pore graphs of potato starch noodles with PA (experimental group) and without PA (control group). **a** Control group ($\times 200$); **b** control group ($\times 500$); **c** pore graphs of control group ($\times 200$); **d** experimental group ($\times 200$); **e** experimental group ($\times 500$); **f** pore graphs of experimental group ($\times 200$)

the entire image, and the average area was 555.77 units. Compared with those in the control group, the number and average area of pores was significantly decreased in the potato starch noodle group ($p < 0.05$), and the hardness and cohesion of the potato starch noodles were significantly correlated with pore size. The small pore structure increases the abilities to resist and recover from deformation (Zhou et al. 2013). The addition of PA to potato starch noodles resulted in smaller and denser pores, mainly because a positively charged $Al(OH)_3$ colloid, which promotes formation of a tighter three-dimensional grid structure, is formed after alum ionization (Tang et al. 2018; Li et al. 2011). Therefore, these results can explain why PA can improve the elasticity, tensile strength and sensory quality of potato starch noodles.

Effect of PA addition on the crystal structure of potato starch noodles

The native potato starch structure exhibits a B-pattern, and its main peaks are at approximately 5.6° , 17° , 22° and 24° . Furthermore the diffraction peak of the native potato starch structure is high, while the peak width is narrow (Han et al. 2009). Table 2 shows that the diffraction peaks of the experimental and control groups were observed near 17.15° and 21.42° but absent at 5.6° and 24° , respectively. The

Table 2 XRD data of potato starch noodles with PA (experimental group) and without PA (control group)

	Diffraction peak/ $^\circ$		Crystallinity/%
Experimental group	17.15 ± 0^a	21.42 ± 0^a	19.64 ± 0.01^a
Control group	17.15 ± 0^a	21.42 ± 0^a	16.65 ± 0.01^b

Means with different letters in the same column differ significantly different at $p < 0.05$

results showed that the potato starch noodle crystal structure was destroyed, as the B-type crystalline structure was replaced by an amorphous state.

In potato starch noodle processing, after starch gelatinization and exertion of external mechanical force, the starch molecular structure is damaged, the amylose of the amorphous region and amylopectin of the crystal region structure are changed, the ordering and crystal degree are reduced, and starch gelatinization becomes easier (Zhu 2015). However, when PA was added, the crystallization degree of the potato starch noodles increased significantly ($p < 0.05$) from 16.65 to 19.64% (Table 2). Earlier, Singh et al. (2018) and Shevkani et al. (2017) demonstrated that a higher degree of crystallinity provides structural stability to the granules, thus making them more resistant to gelatinization. This phenomenon indicated that the addition of PA could provide structural stability to the starch granules, potentially because the aluminum hydroxide colloid, ionized from PA, adsorbed on the surface of the starch granules. This action prevented moisture from entering the particles and thus stabilized the starch granules and molecules, thereby delaying the disintegration of the starch granule crystallization region (Li et al. 2011).

FT-IR spectra analysis

Starch is a high-molecular weight polysaccharide composed of glucose units. Glycosidic bonds exist between structural units and adjacent structural units, and numerous intra- and intermolecular hydrogen bonds are formed. These hydrogen bonds play an important role in the three-dimensional structure and function of the system. Infrared spectroscopy is an effective method to analyze the molecular structure and identify the interaction of hydrogen bonds. Higher frequencies of the infrared absorption peak are correlated with lower wavenumbers and stronger hydrogen bond interactions (Pawlak and Mucha 2003).

In this study, the change in the molecular structure of potato starch noodles with the addition of PA was analyzed

using FT-IR, and the results are shown in Table 3. The potato starch noodles with PA had the same absorption peaks as the control group noodles except for the difference in displacement of the O–H peak. With the PA addition, the O–H stretching vibration absorption peak of potato starch at 3419 cm^{-1} was obviously affected and moved slightly to 3449 cm^{-1} . The decreased hydrogen bond force suggested that hydrogen bonding was not dominant in the system. Because the aluminum ion can form a strong ionic bond with –OH, the –OH bond is weakened. PA-ionized aluminum hydroxide is a type of white colloid substance that does not dissolve in water and has a strong adsorption capacity, which can cause particles to agglomerate into larger particles; this process is not reversible (Li et al. 2011). Once the positively charged $\text{Al}(\text{OH})_3$ encounters the OH^- in the starch molecule, it adsorbs together and causes the numerous particles in the system to agglomerate to form a solid colloidal system. Because aluminum ions form a strong ionic bond with –OH during this process, a more solid and stable three-dimensional lattice structure is formed, which weakens the –OH bond (Noda et al. 2018).

Effect of PA addition on potato starch dough rheological properties

Potato starch dough is a mixture of starch paste and starch. Potato starch dough rheological properties are directly related to the processing characteristics of potato starch noodles. The elastic modulus (G'), viscous modulus (G'') and loss tangent ($\tan\delta$) curves with the angular frequency variation in the experimental and control groups are shown in Fig. 3. The elastic modulus represents the recoverable elastic properties of energy storage, and the viscous modulus represents the viscosity property of energy dissipation (Viturawong et al. 2008). As shown in Fig. 3, the G' and G'' values in the two groups of starch dough were increased, and G' was always greater than G'' ; thus, the loss tangent $\tan\delta$ was less than one. The pattern was very similar to the typical weak gel rheology pattern (Ptaszek

Table 3 FTIR spectra data of potato starch noodles with PA (experimental group) and without PA (control group)

Absorption peak	Peak 1 (O–H stretching vibration peak)	Peak 2 (C–H stretching vibration peak)	Peak 3 (C=O stretching vibration peak)	Peak 4 (C–H angular vibration peak)	Peak 5 (C–O stretching vibration peak)	Peak 6 (C–H bending vibration peak)
Wave number range/ (cm^{-1})	4000–3000	3300–2700	1900–1650	1500–1300	1300–1000	1000–600
Experimental group	3449 ^a	2929 ^a	1649 ^a	1372 ^a	1169 ^a	853 ^a
Control group	3419 ^b	2929 ^a	1649 ^a	1372 ^a	1169 ^a	853 ^a

Means with different letters in the same column differ significantly different at $p < 0.05$

Fig. 3 Dynamic mechanical spectra of potato starch dough with PA (experimental group) and without PA (control group), G' (elastic modulus), G'' (viscous modulus), $\tan\delta$ (loss tangent) = G''/G' , and ω (frequency)

et al. 2009), indicating that the starch dough was a type of non-Newtonian fluid with viscoelastic properties and mainly showed elastic properties. Compared with those of the control dough, the addition of PA significantly increased the G' and G'' of the starch dough, indicating that the elastic properties of starch dough were enhanced. The increase in G' and G'' after the PA addition may be due to salting-out ions, such as Al^{3+} and SO_4^{2-} , promoting the elution of amylose and formation of a lattice structure with the dissolved amylose. Higher $\tan\delta$ (G''/G') values indicate larger viscosity properties and higher elastic properties (Eidam et al. 1995). As shown in Fig. 3, the addition of PA significantly ($p < 0.05$) reduced the $\tan\delta$ values (from 0.62 to 0.51 when $\omega = 10 \text{ rad}\cdot\text{s}^{-1}$) of starch dough, indicating that the increase in G' was faster than the increase in G'' .

Conclusion

Taken together, these results demonstrate that the addition of PA strongly affects various properties of potato starch noodles to different extents. The addition of PA could improve the stability of starch molecules, inhibit starch gelatinization, prevent starch molecule leakage, increase the amount of bound water in the system, and cause potato starch noodles to have smaller and denser pores and more compact structures. The addition of PA significantly decreased the $\tan\delta$ value and made the G' value increase faster than the G'' value. Thus, when searching for new alternatives that are not harmful to our body and could improve the quality of products that replace PA in the future, the new alternatives should have the following characteristics: the ability to inhibit starch gelatinization, decrease the amount of amylose leakage, slow down the

disintegration of the starch granule crystal region, increase the G' value, and decrease the $\tan\delta$ value significantly.

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Compliance with ethical standards

Conflict of interest The authors have declared that they have no conflicts of interest.

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