Effects of microwave and heat-moisture treatments on color characteristics, particle size and water distribution of potato starch

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Abstract

Keywords:

Potato starch
Microwave
Heat-moisture
treatment
Color
Particle size

Article history:

Received
27.09.2021
Received in revised
form 17.10.2021
Accepted
30.12.2021

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DOI:

10.24263/2310-
1008-2021-9-2-4

Introduction. The purpose of this study is to assess the effect of microwave and heat-moisture treatments on color characteristics, particle size and water distribution of potato starch.

Materials and methods. Native potato starch was modified by microwave and heat-moisture treatments. Such treatments of potato starch were done: single microwave treatment (MW); heat-moisture treatment assisted by microwave pre-treatment (MW-HMT), and heat-moisture treatment assisted by microwave post-treatment (HMT-MW).

Results and discussion. In general, HMT caused a slight increase of lightness (L^* values), while single MW caused a slight decrease of lightness, indicating that the color of all the HMT treated samples became brighter and the color of the single MW treated sample (MWS) became darker. The color of MW-HMT samples became more reddish, while MWS, HMT and HMT-MW samples became more greenish. Although there were significant differences of the difference of color (ΔE), it could conclude that all treatments did not markedly change potato starch color for ΔE was always below 5. Median diameter (D_{50}), particle diameter of volume ($D(4,3)$) and particle diameter of surface ($D(3,2)$) of all treated starch were higher compared with native starch (NS), while the value of specific surface area was significantly decreased by MW and HMT, indicating that these treatments can cause expansion, partial gelatinization and agglomeration of starch granules, resulting in large particle size of starch granules. MW and HMT treatments of potato starch caused the relaxation time T_{21} shifted toward faster relaxation times compared with native starch. Although the water in all starch samples was the main water which at least accounted for 90%, three peaks were observed in relaxation time T_2 of MW treated starch (MWS, MW-HMT and HMT-MW), and two peaks were observed in relaxation time T_2 of NS and HMT samples, which indicated the MW treated starch had three different state water, while NS and single HMT treated starch only had two different state water. Furthermore, MW and HMT treatments could change the water distribution and improve the interaction between starch and water.

Conclusions. Although MW and HMT treatments did not markedly change potato starch color, these treatments can cause expansion, partial gelatinization and agglomeration of starch granules, resulting in large particle size of starch granules. MW and HMT treatments could change the water distribution and improve the interaction between starch and water.

Introduction

Various modification methods have been used to enhance starch properties, including physical (Wang et al., 2018; Colussi et al., 2020), chemical (Ariyantoro et al., 2018) and enzymatic or combined ways (Li et al., 2019). Physical modifications are widely used for starch modification not only because of safety and simplicity, but also because of sustainability and environmental friendliness (Chandrasekaran et al., 2013; Kaur et al., 2012). Microwave treatment is widely used in the food industry at the working frequency of 915 MHz to 2450 MHz in most part of the world (Hoz et al., 2013). The microwave frequency, system composition, density, temperature and other factors influence the dielectric properties which are crucial indexes to evaluate the ability of food to absorb and transform microwave electromagnetic energy to thermal energy. The dielectric properties of starch-based materials are distinct due to their discrepant compositions, moisture content, metal ion concentration, porosity, and other parameters of a starch-based system (Tao et al., 2020). Recent researches show that microwave treatment can lead to changes in granule morphology, molecular chain structure and crystalline structure, which consequently affects its functional properties, such as solubility (Singh et al., 2012), swelling capacity (Deka and Sit, 2016), gelatinization (Oyeyinka et al., 2021), retrogradation (Chen et al., 2021) and digestion ability (Zeng et al., 2016). For example, microwave treatment decreased the swelling power, transparency and thermal properties of millet starch, destroyed the original appearance of the starch granules and formed smaller and lamellar gel blocks (Li et al., 2019). Heat-moisture treatment (HMT) is another commonly used physical treatment that involves the treatment of starch granules at low moisture content (<35%) for a certain time period (15 min-16 h) at relatively high temperatures (90 °C -120 °C) above glass transition temperature but below the gelatinisation temperature (Li et al., 2020; Chatpapamon et al., 2019). Previous researches show that HMT promotes changes in X-ray diffraction (XRD) patterns, granule morphology, gelatinization properties, swelling power and enzyme digestibility (Chen et al., 2017; Zhou et al., 2020). Moreover, HMT improves the liquefaction of amylose and quality of recrystallization due to molecular chain arrangement inside the starch granule (Jiranuntakul et al., 2011).

Previous studies have reported the impacts of single microwave treatment and single heat-moisture treatment on morphological structural properties, thermal and physicochemical and digestibility of starch (Sui et al., 2015; Zhou et al., 2020). However, few studies exploring the effect of microwave treatment and heat-moisture treatment on the color characteristics, particle size and water distribution of potato starch have been reported.

Therefore, the purpose of this study is to assess the effect of microwave and heat-moisture treatments on color characteristics, particle size and water distribution of potato starch.

Materials and methods

Materials

Potato tubers locally known as Favorita cultivar were purchased from a local market in Hezhou city, China. Native starch from potato tubers was extracted with the technology of wet milling extraction as previous method of Dhritiman Deka and Nandan Sit (Deka and Sit, 2016) and the dried starch was ground and passed through 80 mesh sieve and kept in airtight plastic containers for further analysis.

Starch samples with single and dual modification

Heat- moisture treatment. Heat-moisture treatment was performed according to the method of Deng (Deng et al., 2021) with some modifications. 70 g native starch powder (NS) was weighted in 500 mL Duran laboratory bottle, and ultrapure water was added to adjust moisture content to 25%. After thorough mixing, the glass containers were sealed and equilibrated for 24 hours at 25 °C, samples were then reacted in a hot-air oven (DH411C, Yamato Scientific Co.Ltd., Japan) at 90 °C for 1.5 h, 4 h, 8 h and 12 h respectively. After heat-moisture treatment, the starch samples were dried in a drying oven at 45 °C for 24 h to make sure the moisture content less than 12 %. The dried potato starch pulverized for 45 s using a universal pulverizer, passed through an 80-mesh sieve, vacuum-packed in polyethylene bags, and stored in an airtight container for conducting further studies. The prepared starches were named HMT1.5, HMT4, HMT8 and HMT12.

Microwave treatment. Adjustment of starch moisture content (25 %) and equilibration were carried out according to the method in Section 2.2.1. Then the starch was placed flat into a petri dish with a diameter of 18 cm, covered with microwave plastic film and evenly pricked 10 holes with toothpicks. The petri dish with starch was exposed in a microwave oven (Galanz, G80F20CN2L-B8(RO), Guangdong Galanz Microwave Appliance Manufacturing Co., LTD, China) to perform the microwave treatment with 400 W power for 5 min (increasing the power level or time of heating in microwave will cause the starch granules to burn). The treated starch sample was dried and stored as the method in Section 2.2.1. The prepared starches were named MWS.

Dual modification.

(1) Heat-moisture treatment assisted by pre- treatment of microwave (MW-HMT): The MWS sample was treated with heat-moisture treatment according to the method in Section 2.2.1 to obtain MW-HMT1.5, MW-HMT4, MW-HMT8 and MW-HMT12.

(2) Heat-moisture treatment assisted by poste-treatment of microwave (HMT-MW): HMT samples were treated with microwave according to the method in Section 2.2.2 to acquire HMT1.5-MW, HMT4-MW, HMT8-MW and HMT12-MW.

The treated samples were analyzed for physicochemical, structural and digestive properties. Untreated native potato starch (NS) was used as control.

Color characteristics

Color measurements of NS and treated starch were carried out using a colorimeter (CR-400, Konica Minolta Inc., Japan.) after the calibration of the equipment with a standard-white reflection plate. L^* indicates lightness, which varies from black ($L^*=0$) to white ($L^*=100$), a^* is greenness / redness value, which varies from green (-60) to red (+60) and b^* is blueness/yellowness value, which varies from blue (-60) to yellow (+60). Color difference (ΔE) between treated starch sample and the native starch (NS) was calculated with the equation:

$$\Delta E = \sqrt{(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2} \quad (1)$$

$$\Delta L^* = L^* - L_0^* \quad (2)$$

$$\Delta a^* = a^* - a_0^* \quad (3)$$

$$\Delta b^* = b^* - b_0^* \quad (4)$$

where L^* , L_0^* is the lightness of treated starch and NS; a^* , a_0^* is the greenness/ redness value of treated starch and NS; b^* , b_0^* is the blueness/yellowness value of treated starch and NS. The smaller value of ΔE indicates the smaller color difference between treated starch and NS.

Particle size distribution

A laser diffraction particle size analyzer ((BT-2001, Baxter Instruments Co. LTD, China) equipped with a dry dispersion unit was used to determine the particle size distribution of all the starch samples according to the manufacturer's instructions. The starch particle size parameters included particle diameter of volume (D(4,3)), particle diameter of surface (D(3,2)), specific surface area (S.S.A.), D50 represents the corresponding particle size which is smaller than 50% of the sample particles.

LF-NMR spin– spin relaxation (T_2) measurements

Low-field nuclear magnetic resonance (LF-NMR) spin-spin relaxation measurements were carried out using a Niumag Benchtop Pulsed NMR Analyzer (NMI120X, Niumag Electric Corp., Shanghai, China) to determine the water distribution of native and treated starch. A saturated NaCl_2 solution was used to equilibrate the water content of the samples for two weeks at 25 °C until the water activity of the samples ($a_w = 0.724$) was constant and consistent according to the method of Fan (Fan et al., 2013). Approximately 1 g of equilibrated starch was placed into the NMR tube with diameter of 15 mm to measure T_2 using the Carr-Purcell-Meiboom-Gill (CPMG) sequence. The measurement temperature was 32 ± 0.1 °C and the proton resonance frequency was 18.0 MHz. Typical pulse parameters were as follows: the time of 90° plus (P_1) was 9.5 μs and the time of 180° plus (P_2) was 19.04 μs , the waiting time (TW) between subsequent scans was 3500 ms, data from 5000 echoes were acquired as 4 scan repetitions, and each measurement was performed at least 3 times.

Statistical analysis

All the experiments were conducted in triplicate unless otherwise stated. The Statistical analysis was performed on Data Processing System (version 7.05) and charts were done in Origin Pro 8. Data were analyzed using ANOVA with Duncan's multiple range test, and the values were considered significantly different when $p \leq 0.05$.

Results and discussion

Color characteristics

The color values of the native potato starch (as control) and treated starch were showed in Table 1. In general, HMT caused a slight increase of lightness (L^* values), while single MW treatment caused a slight, although significant, decrease of lightness, indicating that the color of all the HMT treated samples became brighter and the color of the single MW treated sample (MWS) became darker. The a^* values of treated samples varied significantly in different ways depending on the treated ways. Although there was no significant difference of a^* values in starch samples with the same treated method, the color of MW- HMT samples became more reddish, while MWS, HMT and HMT-MW samples became more greenish. As can be seen from Table 1, all the treated samples became more yellowish, as significant increase of b^* values were obtained for treated samples. Although there were significant differences of the difference of color (ΔE), it could conclude that all treatments did not markedly change potato starch color from the analysis of ΔE , given that ΔE was always below 5, indicating that color differences was no visible differentiated of all the treated starch samples (Solaesa et al., 2021; García-Viguera and Zafrilla, 2001).

Table 1

Color characteristics of native and treated potato starch samples

Samples	L*	a*	b*	ΔE
NS	99.38±0.05 ^{cd}	4.80±0.01 ^{bcd}	-2.38±0.01 ⁱ	-
MWS	98.71±0.02 ^e	4.73±0.03 ^g	-2.21±0.03 ^h	0.70±0.02 ^c
MW-HMT1.5	99.31±0.24 ^d	4.81±0.03 ^{abc}	-2.02±0.06 ^f	0.42±0.05 ^f
MW-HMT4	99.46±0.20 ^{bcd}	4.84±0.02 ^a	-1.94±0.05 ^e	0.48±0.03 ^c
MW-HMT8	99.51±0.20 ^{abcd}	4.83±0.01 ^{ab}	-1.71±0.03 ^{bc}	0.70±0.02 ^c
MW-HMT12	99.54±0.06 ^{abcd}	4.80±0.03 ^{abcd}	-1.60±0.01 ^a	0.80±0.02 ^a
HMT1.5	99.38±0.07 ^{cd}	4.76±0.03 ^{defg}	-2.15±0.02 ^g	0.24±0.02 ^g
HMT4	99.59±0.02 ^{abc}	4.77±0.02 ^{cdef}	-2.03±0.02 ^f	0.41±0.01 ^f
HMT8	99.49±0.12 ^{abcd}	4.78±0.02 ^{cde}	-1.85±0.01 ^d	0.55±0.03 ^d
HMT12	99.66±0.08 ^{ab}	4.75±0.04 ^{efg}	-1.68±0.04 ^b	0.76±0.03 ^{ab}
HMT1.5-MW	99.70±0.05 ^a	4.74±0.01 ^{fg}	-2.14±0.01 ^g	0.41±0.03 ^f
HMT4-MW	99.50±0.05 ^{abcd}	4.75±0.01 ^{efg}	-1.85±0.01 ^d	0.55±0.02 ^d
HMT8-MW	99.68±0.13 ^{ab}	4.77±0.01 ^{defg}	-1.89±0.01 ^d	0.58±0.06 ^d
HMT12-MW	99.70±0.11 ^a	4.77±0.03 ^{def}	-1.74±0.02 ^c	0.72±0.05 ^{bc}

Note: all values are the mean of triplicate determinations ± SD. The means within the same column with different letters are significantly different (P<0.05).

Particle size distribution

The variation of particle size could reflect the changes in agglomeration of the starch before and after modification. The particle size distribution parameters of native and treated potato starch were showed in Table 2.

Table 2

Particle size distribution of native and treated potato starch samples

Samples	D50(μm)	D(4,3) (μm)	D(3,2) (μm)	S.S.A.(m ² /kg)
NS	34.12±0.40 ^c	36.60±0.38 ^f	22.24±0.56 ^h	99.93±2.54 ^a
MWS	34.29±0.16 ^{bc}	36.97±0.07 ^{cdef}	24.06±0.07 ^g	92.35±0.26 ^b
MW-HMT1.5	34.38±0.09 ^{bc}	37.24±0.21 ^{bcd}	29.67±0.12 ^{def}	74.87±0.32 ^{cd}
MW-HMT4	34.53±0.03 ^{abc}	37.35±0.08 ^{abcde}	30.51±0.02 ^{abc}	72.83±0.06 ^{de}
MW-HMT8	34.91±0.27 ^{ab}	37.79±0.26 ^{ab}	30.88±0.23 ^a	71.96±0.53 ^e
MW-HMT12	35.14±0.68 ^a	37.95±0.65 ^a	30.76±0.56 ^{ab}	72.24±1.31 ^e
HMT1.5	34.61±0.28 ^{abc}	37.48±0.19 ^{abcd}	30.28±0.17 ^{abcd}	73.38±0.41 ^{cde}
HMT4	34.82±0.21 ^{ab}	37.52±0.22 ^{abc}	29.51±0.23 ^{ef}	75.29±0.59 ^e
HMT8	34.73±0.49 ^{abc}	37.33±0.67 ^{abcde}	29.35±0.25 ^f	75.71±0.65 ^e
HMT12	34.64±0.23 ^{abc}	37.42±0.17 ^{abcde}	29.96±0.44 ^{cdef}	74.18±1.08 ^{cde}
HMT1.5-MW	34.36±0.13 ^{bc}	37.03±0.18 ^{cdef}	23.97±0.42 ^g	92.72±1.60 ^b
HMT4-MW	34.29±0.19 ^{bc}	37.00±0.07 ^{cdef}	29.41±0.24 ^{ef}	75.55±0.62 ^e
HMT8-MW	34.32±0.11 ^{bc}	36.82±0.09 ^{def}	30.11±0.23 ^{bcd}	73.63±0.84 ^{cde}
HMT12-MW	34.54±0.07 ^{abc}	36.75±0.09 ^{ef}	30.27±0.46 ^{abcd}	73.41±1.10 ^{cde}

Notes: all values are the mean of triplicate determinations ± SD. The means within the same column with different letters are significantly different (P<0.05).

The D50, D(4,3) and D(3,2) of all treated starch were higher than NS, while the value of S.S.A. was significantly decreased by MW and HMT, which was agreed with precious studies about HMT treated lily starch (Li et al., 2020) MW treated waxy hull-less barley starch (Chen et al., 2021). The reason for the large particle size of MW and HMT modified starch was that the internal temperature of the granules rose rapidly during the MW and HMT treatment, and the internal pressure increased, causing the starch granules to expand, resulting in partial gelatinization and agglomeration of the granules (Li et al., 2020; Chen et al., 2021).

Relaxation time (T_2) analysis

In starch-based foods, the physical properties of water can significantly affect the processing quality of starch. LF-NMR technology is the most effective technology for evaluating the distribution and state of water in the starch-water system. Relaxation time (T_2) is the time required for an excited spin-spin proton to reach dynamic equilibrium after energy exchange with adjacent protons, reflects the difference in the degrees of freedom of water. The less hydrogen protons are bound or higher degree of freedom is associated with longer transverse relaxation time (Han et al., 2009), while the more hydrogen protons are bound or lower degree of freedom corresponds to a shorter transverse relaxation time (Pitombo and Lima, 2003; Luo et al., 2020).

As shown in Figure 1-3 and Table 3, three peaks were observed in T_2 of MW treated starch (MWS, MW-HMT and HMT-MW), while two peaks were observed in T_2 of NS and HMT samples. The first peak T_{21} (0.01-1 ms) corresponded to bound water, which interacted with the surface of amylose and amylopectin chains (Tang et al., 2000).

Figure 1. Water distribution in native and single HMT treated starch by LF-NMR

Figure 2. Water distribution in native and MW-HMT treated starch by LF-NMR

Figure 3. Water distribution in native and HMT-MW treated starch by LF-NMR

Table 3

Relaxation times (T_2) and corresponding peak areas percentages of water from native and treated starch

Samples	Relaxation Time (ms)			Proportion of water in different state (%)		
	T_{21}	T_{22}	T_{23}	PT ₂₁	PT ₂₂	PT ₂₃
NS	0.62 ±0.06 ^a	5.74 ±0.57 ^b	-	90.48 ±0.98 ^c	9.52 ±0.98 ^a	-
MWS	0.59 ±0.01 ^{ab}	8.43 ±3.24 ^{ab}	30.61 ±3.02 ^a	92.56 ±0.80 ^{ab}	1.14 ±0.06 ^c	6.29 ±0.74 ^a
MW-HMT1.5	0.57 ±0.00 ^{ab}	7.73 ±2.26 ^{ab}	25.01 ±4.90 ^a	93.06 ±0.04 ^{ab}	1.58 ±0.39 ^{de}	5.36 ±0.35 ^{ab}
MW-HMT4	0.60 ±0.05 ^{ab}	8.52 ±0.70 ^{ab}	24.71 ±2.74 ^b	93.39 ±0.52 ^{ab}	3.92 ±0.88 ^c	2.69 ±0.72 ^c
MW-HMT8	0.57 ±0.00 ^{ab}	7.60 ±0.65 ^{ab}	24.66 ±0.16 ^a	93.11 ±0.75 ^{ab}	3.57 ±1.42 ^c	3.33 ±2.17 ^{bc}
MW-HMT12	0.57 ±0.00 ^{ab}	8.06 ±0.00 ^{ab}	25.76 ±1.40 ^a	93.57 ±0.30 ^{ab}	2.85 ±0.64 ^{cd}	3.57 ±0.94 ^{bc}
HMT1.5	0.57 ±0.00 ^{ab}	7.10 ±0.99 ^{ab}	-	93.32 ±0.18 ^{ab}	6.68 ±0.18 ^b	-
HMT4	0.57 ±0.00 ^{ab}	6.75 ±0.53 ^{ab}	-	92.59 ±0.26 ^{ab}	7.41 ±0.26 ^b	-
HMT8	0.57 ±0.00 ^{ab}	6.44 ±0.53 ^{ab}	-	93.31 ±1.33 ^{ab}	6.69 ±1.33 ^b	-
HMT12	0.57 ±0.00 ^{ab}	6.60 ±0.65 ^{ab}	-	92.37 ±0.94 ^b	7.63 ±0.94 ^b	-
HMT1.5-MW	0.57 ±0.00 ^{ab}	8.72 ±0.86 ^a	24.77 ±0.00 ^a	93.61 ±0.29 ^{ab}	2.32 ±0.26 ^{cde}	4.07 ±0.04 ^{abc}
HMT4-MW	0.54 ±0.05 ^b	8.72 ±0.86 ^a	28.61 ±0.18 ^a	93.97 ±0.53 ^a	2.99 ±1.13 ^{cd}	3.05 ±1.66 ^{bc}
HMT8-MW	0.54 ±0.05 ^b	7.73 ±2.26 ^{ab}	28.76 ±5.64 ^a	93.77 ±0.88 ^{ab}	2.52 ±0.13 ^{cde}	3.71 ±1.01 ^{bc}
HMT12-MW	0.54 ±0.04 ^b	8.52 ±0.70 ^{ab}	27.33 ±2.22 ^a	93.77 ±0.14 ^{ab}	3.19 ±0.40 ^{cd}	3.04 ±0.41 ^{bc}

Notes: all values are the mean of triplicate determinations ± SD. The means within the same column with different letters are significantly different ($P < 0.05$). – indicates that there is no value.

It can be seen from Table 3 that the MW and HMT treatments of potato starch caused the T_{21} shifted toward faster relaxation times compared with native starch (NS), indicating that MW and HMT treatments enhanced the starch-water interaction and self-binding of starch, and consequently resulting in lower mobility of water protons. The second peak T_{22} (1-20 ms) corresponded to “semi-crystalline lamellae water” (Tang et al., 2000), which existed in the narrow space between crystals. Due to different spaces, the water in them exhibited different mobilities. Peak of NS was closer to the left than treated starch; they were followed by the single HMT treated starch (HMT1.5, HMT4, HMT8 and HMT12). The peaks T_{22} of MW treated starch were close to the right and connected with its next peak. The third peak T_{23} was located in T_2 between 20 ms and 100 ms was attributed to immobilized water, which was regarded as water inside the hexagonal channels of B-type crystal clusters and was defined as “channel water in B-type crystal” by a previous study (Chen et al., 2019). It can be observed from Figure1, the native and HMT treated starch did not have channel water in B-type crystal, while all the MW treated starch contained a certain amount of channel water in B-type crystal.

The proportion of different state water in native and treated starch were summarized in Table 3. The areas under different component represented the proportion of different state water distribution, namely PT_{21} (bound water), PT_{22} (semi-crystalline lamellae water), PT_{23} (channel water in B-type crystal), respectively. Although the water in all starch samples was the main water which at least accounted for 90%, the MW treated starch had three different state water, NS and single HMT treated starch only had two different state water. There were significant differences of PT_{21} and PT_{22} between NS and all treated starch, NS had the lowest PT_{21} but highest PT_{22} , indicating MW and HMT treatments could change the water distribution and improve the interaction between starch and water. No significant differences of PT_{21} were observed between MW treated starch and single HMT treated, but all MW treated starch had lower PT_{22} than single HMT treated starch; furthermore, all the MW treated starch contained a certain amount of channel water in B-type crystal (PT_{23}). These results indicated that MW treatment could result in a certain level of semi-crystalline lamellae water shifting to channel water in B-type crystal.

Conclusions

In this study, different treated potato starch samples were prepared with MW and HMT methods, and the color characteristics, particle size and water distribution of native and treated potato starch were evaluated by using colorimeter, laser diffraction particle size analyzer and LF-NMR. Although color differences was no visibly differentiated between NS and all treated starch, HMT treatment caused a slight increase of lightness (L^* values), while single MW treatment caused a slight decrease of lightness, indicating that the color of all the HMT treated samples (HMT, MW-HMT, HMT-MW) became brighter and the color of the single MW treated sample (MWS) became darker. Although there were significant differences of the differences of color (ΔE), it could conclude that all treatments did not markedly change potato starch color for ΔE was always below 5, indicating that color difference was no visible differentiated of all the treated starch samples. The results of particle size distribution showed that D_{50} , $D(4,3)$ and $D(3,2)$ of all treated starch were higher than NS, while the value of S.S.A. was significantly decreased by MW and HMT, indicating that MW and HMT treatments can caused expansion, partial gelatinization and agglomeration of starch granules, resulting in large particle size of starch granules. Although the water in all starch samples was the main water which at least accounted for 90%, three peaks were observed in T_2 of MW treated starch (MWS, MW-HMT and HMT-MW), two peaks were observed in T_2 of native and single HMT treated starch, which indicated the MW treated starch had three different state water, while NS and single HMT treated starch only had two different state water. There were significant differences of PT_{21} and PT_{22} between NS and all treated starch, NS had the lowest PT_{21} but highest PT_{22} , indicating MW and HMT treatments could change the water distribution and improve the interaction between starch and water.

Acknowledgements. The authors gratefully acknowledge the financial support received from the Middle aged and Young Teachers' Basic Ability Promotion Project of Guangxi (Grant No.2021KY0710) and the High-level Innovation Team and Outstanding Scholars Program of Guangxi Colleges and Universities (GJR (2018) 35). The authors are grateful to Guangxi Key Laboratory of Health Care Food Science and Technology for providing laboratory facilities and technical support during this research work.

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