



Effect of Cross-Linking with Ginger and Garlic on the Properties of Cassava Starch

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Abstract

Cross-linking is a type of chemical modification to improve granule stability with new covalent bonds, thus providing desired functional properties. The present study was undertaken to modify cassava starch with active components of ginger and garlic and to determine the properties of modified cassava starch. The cassava starch was modified with 5% ginger (A), 10% ginger (B), 5% garlic (C) and 10% garlic (D). The degree of cross-linking was determined from the viscosity values and chemical and functional properties of all cross-linked starches were analyzed by standard methods. Results of the study showed that higher concentrations of ginger and garlic increased the degree of cross-linking. Native cassava starch had higher protein content ($0.6 \pm 0.09\%$) than cross-linked cassava starches (0.11 ± 0.02 - $0.57 \pm 0.01\%$), amylose content was higher in cross-linked starch (22.7 ± 0.52 - $27.2 \pm 0.19\%$) than native starch ($23.2 \pm 1.20\%$). Water and oil absorption capacities were not significantly different between native ($0.96 \pm 0.01 \text{ g g}^{-1}$) and cross-linked starches (0.87 ± 0.03 - $0.93 \pm 0.04 \text{ g g}^{-1}$). Cross-linked starch had higher swelling power (14.5 ± 0.45 - $14.8 \pm 0.36 \text{ g g}^{-1}$) and solubility (8.1 ± 0.42 - $9.0 \pm 1.36\%$) when compared to those of native starch ($11.1 \pm 0.84 \text{ g g}^{-1}$ and $7.1 \pm 0.62\%$, respectively). Lower viscous properties were observed for cross-linked (1975.00 - 2565.00 cP) starch than that of native starch (3733.66 cP). Cassava starch could be modified with ginger and garlic and distinctive properties were exhibited by the cross-linked starches.

Key words: Cassava starch, cross-linking, garlic, ginger, pasting properties

Introduction

Native starches, irrespective of their source, are undesirable for many applications (Singh et al., 2007) because of their inability to withstand processing conditions such as extreme temperature, diverse pH, high shear rate and freeze-thaw variation. In order to improve the desirable functional properties, native starches are often modified. Modified starches have wide applications as binders, fillers, emulsion stabilizers, consistency modifiers and adhesives (Daramola and Osanyinlusi, 2006). Starch modification can be broadly grouped into four classes viz., physical, chemical, enzymatic and biological modifications. Among these modification methods, chemical means is the most frequently used process (Daramola and Osanyinlusi,

2006). Chemical modification of starch involves the treatment of native starch with specific chemical reagents. This definition includes acetylated, oxidized, lintnerized, pyrodextrinized, hydroxypropylated, cross-linked starches etc. (Kaur et al., 2004). Chemical modification of cassava and other plant starches with improved qualities and applications have been reported by several workers (Ahmed et al., 2005; Jyothi et al., 2005).

The active components of ginger root such as gingerol, dehydroginger-dione and shogaol (Kikuzaki and Nakatani, 1993) can be used as natural modifying agents for cassava starch. The modified starches are characterized by stable viscosity, high swelling power, which influences texture, a determining factor in consumer acceptance of some foods and improved solubility that depicts enhanced

digestibility (Daramola and Osanyinlusi, 2006). Cross-linking treatment is intended to add chemical bonds at random locations in a granule and it stabilises the granule and strengthens the relatively tender starch (Acquarone and Rao, 2003). The most common type of chemical modification technique is cross-linking, the derivatization of starch by using a bi- or poly-functional chemical reagent that is able to react with two or more different hydroxyl groups on the same or different starch polymers (Wurzburg, 1986; Thomas and Atwell, 1999). It is well known that cross-linked starches are less sensitive to processing conditions (high temperature, extended cooking time, low pH and high shear during mixing, milling, homogenization and pumping) than native starches (Whistler and BeMiller, 1997). Cross-linking reinforces the already present hydrogen bonds in the granules with new covalent bonds. As a result, cross-linked starch is more resistant to acid, heat and shearing than native starch (Hoseney, 1994; Mirmoghtadaei, et al., 2009).

Starch modification involves the alteration of chemical, physical and functional properties of native starches. This alteration has the main objective of producing more satisfactory products for specific food applications. The starches most commonly modified for commercial uses are those from normal maize, cassava, potato and waxy maize. Modified starches are used to improve viscosity, shelf stability, particulate integrity, processing parameters, texture, appearance and emulsification. Different starches have different properties and are used in the food industry for nutritional, sensory and even aesthetic purposes (Rapaille, 1995). Cross-linking at low levels, have a substantial effect on properties such as granule swelling, paste viscosity and retrogradation (Reddy and Seib, 1999). The objectives of the present study were to modify cassava starch with natural cross-linking reagents such as ginger and garlic and to determine the properties of cross-linked starches.

Materials and Methods

Cassava native starch (CNS) was isolated from cassava roots according to the method of Vasanthan (2001). Ginger and garlic were purchased from local market in Salem, Tamil Nadu. Starches were modified with two cross-linking agents viz., ginger and garlic at two different concentrations (5 and 10% based on starch dry weight),

according to the method described by Daramola and Osanyinlusi (2006) with slight modification. Ginger and garlic were cleaned and peeled. The peeled roots were crushed and mixed with starch-water suspension and stirred periodically for 30 min at room temperature. Ginger and garlic grates were sieved out and starch milk obtained was washed and the water was removed using suction pump. The starch obtained was dried using a hot air oven and ground with a mortar and pestle to obtain modified starch, namely A (cassava native starch with 5% ginger), B (cassava native starch with 10% ginger), C (cassava native starch with 5% garlic) and D (cassava native starch with 10% garlic).

Chemical analyses

The degree of cross linking of the cross-linked starches was determined from the viscosity values, according to the method of Chatakanonda et al. (2000). The peak viscosities of the starch samples were recorded using a Rapid Visco Analyzer (RVA Tech master, Perten, Instruments Japan). The starch slurry (10% by weight) was heated from 50-95°C heating rate at 12 °C min⁻¹ at 160 rpm and then held at 95°C for 2 min. Afterwards the paste was cooled to 50°C at 12°C min⁻¹ and finally kept at 50°C for 2 min. The degree of cross linking was calculated as follows:

$$\text{Degree of cross linking} = \frac{A - B}{B} \times 100$$

where, A is the peak viscosity in RV units of the native starch and B is that of the cross-linked starch.

The crude protein and amylose contents were determined by the method of AOAC (1990) and Williams et al. (1970) respectively. Measurement of water/oil absorption capacity was done by the method of Beuchat (1977). Swelling power and solubility pattern were studied by the method of Leach et al. (1959) at a temperature interval of 10°C from 40-90°C. In this method, one gram of the starch was suspended in 50 ml of distilled water and kept in a water bath for 30 min. The suspension was then removed and centrifuged for 15 min at 2200 rpm. The supernatant was allowed to evaporate at 120°C. The residue thus obtained was subjected to the measurement of the percent solubility of starch in water. The swelling power was calculated as weight of the sediment per gram of dry starch.

Pasting properties of native and modified cassava starch were measured using a Rapid Visco Analyzer (RVA Tech master, Perten, Instruments Japan) (Xiao et al., 2012). Viscosity of starch was recorded with starch suspensions (moisture content 12%, sample 3 g, water 25 ml) that underwent a controlled heating and cooling cycle under constant shear, where the sample was held at 50°C for 1 min, heated from 50-95 °C at 5 °C min⁻¹ and held at 95 °C for 2.7 min, cooled from 95 °C to 50 °C at 5°C min⁻¹ and held at 50 °C for 2 min. The speed of blending was maintained at 160 rpm. Pasting parameters such as peak viscosity, trough viscosity, breakdown, final viscosity, set back, peak time and pasting temperature were recorded. The data were statistically analyzed using single factor ANOVA in Microsoft Excel (2003). There were three replicates for each treatment.

Results and Discussion

The degree of cross-linking of the modified starches was determined from the RVA viscosity values and the results are presented in Table 1. The degree of cross-linking ranged from 30.15-46.14% and this result is in agreement with the findings of Xiao et al. (2012) who reported that the degree of cross-linking of rice starch ranged from 18.71–72.41%. It was observed that the degree of cross-linking increased with increasing concentration of natural cross-linking reagents (ginger and garlic active components). Xiao et al. (2012) reported that the degree of cross-linking increased with increasing concentration of epichlorohydrin (EPI). Higher level of cross-linking of starch retarded the swelling of granule and resulted in a lower peak viscosity. The increased cross-linking could be attributed to the bi-functional entities containing an activated double bond as well as hydroxyl/methoxy phenyl residues (Stauffer, 1990) and sulphur compounds present in active ginger and garlic components such as gingerol, dehydroginger-dione,

shorgal and allicin (Daramola and Osanyinlusi, 2006).

The crude protein content of native and cross-linked starches is presented in Table. 1. The protein content of cross-linked starches was significantly lower than that of native cassava starch (0.60%). With increasing concentration of cross-linking agents, the protein content decreased. Native starch cross-linked with garlic 5% (0.57%) had higher protein content than all other modified starches. This result is in conformity with the report of Aparicio-Saguilán, et al. (2008) who stated that the protein content of cross-linked banana starch ranged from 0.53 to 0.55% and the reagents used in the chemical modification of starch solubilized an important fraction of proteins, thus reducing their levels in the cross-linked preparation (Aparicio-Saguilán et al., 2005).

Amylose contents of native and cross-linked cassava starches are presented in Table 1. The amylose content of native starch was significantly lower than that of the cross-linked starches. With increase in the concentration of cross-linking agents, the amylose content increased. Native starch cross-linked with ginger at 10% concentration had higher amylose content (27.2%) than native starch cross-linked with garlic at 10% level (26%), native starch cross-linked with ginger at 5% (25.0%) and native starch cross-linked with garlic 5% (22.7%). The amylose contents of native and cross-linked starches ranged from 22.7-27.2%. This result is similar to the reports of Dzogbefia et al. (2008) who stated that the amylose content of native cassava starch was 24.5%. Cross-linking with ginger and garlic components might have occurred in the outer regions of the granules and was located on the amylopectin molecules. Though cross-linking preferentially occurred in amylopectin, some amylose molecules were cross-linked to amylopectin (Jane et al., 1992). These amylose molecules will be protected from acid hydrolysis. Therefore, amylose hydrolysis decreased after cross-linking resulting in higher percentage of amylose and consequently lower

Table 1. Degree of cross-linking, protein and amylose contents of native and cross-linked cassava starches

Sample	Degree of cross-linking (%)	Crude protein (%)	Amylose (%)
NCS	Nil	0.60±0.09	23.2±1.20
A	39.36±9.86	0.44±0.07	25.0±1.21
B	46.14±12.66	0.11±0.02	27.2±0.19
C	30.15±13.45	0.57±0.01	22.7±0.52
D	37.46±6.19	0.21±0.08	26.0±0.19
CD (0.05)	15.39	0.08	1.13

Mean ± standard deviation; NCS = native cassava starch; A = cassava native starch with 5% ginger; B = cassava native starch with 10% ginger; C = cassava native starch with 5% garlic; D = cassava native starch with 10% garlic

relative crystallinity compared to that of the acid-modified non cross-linked starch (Atichokudomchai and Varavinit, 2003). Wurzburg (1986) reported that during acid modification, the amount of starch amylose increases indicating that acid preferably hydrolyses amylopectin.

The water absorption capacity (WAC) and oil absorption capacity (OAC) of the native and cross-linked starches are presented in Table 2. The modified starches with higher degree of cross-linking exhibited a decrease in the WAC and OAC. Compared to WAC and OAC of native starch (1.1 g g^{-1} and 1.5 g g^{-1} , respectively), the cross-linked starches exhibited higher levels of WAC ($0.9\text{--}0.9 \text{ g g}^{-1}$) and OAC ($1.1\text{--}1.3 \text{ g g}^{-1}$) values. On the other hand, Yook et al. (1993) reported a decrease in the water uptake of rice starch upon cross linking with EPI. Kaur et al. (2004) observed a considerable lowering of WBC of cross-linked potato starches in comparison to the native counterparts. Choi and Kerr (2003) reported that cross-linking at low concentration of phosphorus oxychloride had little effect on the dynamic water properties of wheat starch.

Table 2. Water and oil absorption capacities of the native and cross linked starches

Sample	Water absorption capacity (g g^{-1})	Oil absorption capacity (g g^{-1})
NCS	0.96 ± 0.01	1.45 ± 0.05
A	0.92 ± 0.06	1.16 ± 0.03
B	0.87 ± 0.03	1.09 ± 0.10
C	0.93 ± 0.04	1.27 ± 0.20
D	0.88 ± 0.02	1.23 ± 0.16
CD (0.05)	0.08	0.05

Mean \pm standard deviation; NCS = native cassava starch; A = cassava native starch with 5% ginger; B = cassava native starch with 10% ginger; C = cassava native starch with 5% garlic; D = cassava native starch with 10% garlic

Table 3. Swelling power of native and modified cassava starches

Sample	Temperature ($^{\circ}\text{C}$)					
	40	50	60	70	80	90
NCS	2.2 ± 0.19	3.0 ± 0.19	3.6 ± 0.14	5.1 ± 0.26	11.2 ± 20.84	15.4 ± 2.88
A	3.1 ± 0.74	4.6 ± 0.81	5.6 ± 0.61	10.7 ± 0.10	14.5 ± 0.45	11.0 ± 0.20
B	3.5 ± 0.02	5.4 ± 0.67	6.2 ± 0.61	11.4 ± 0.50	14.7 ± 0.46	11.0 ± 0.20
C	3.5 ± 0.63	4.6 ± 0.64	5.8 ± 0.73	11.0 ± 0.07	14.9 ± 0.36	11.2 ± 0.50
D	3.5 ± 0.03	5.5 ± 0.57	6.3 ± 0.07	11.8 ± 0.30	14.8 ± 0.45	11.0 ± 0.19
CD (0.05)	0.63	0.86	0.72	0.41	0.76	1.86

Mean \pm standard deviation; NCS = native cassava starch; A = cassava native starch with 5% ginger; B = cassava native starch with 10% ginger; C = cassava native starch with 5% garlic; D = cassava native starch with 10% garlic

The swelling powers of native and cross-linked starches are given in Table 3. The results are consistent with the earlier reports (Liu et al. 1999; Hirsch and Kokini, 2002). According to Kartha and Srivastava (1985) with increase in the degree of cross-linking, the granular structure of starch becomes more compact and swelling and solubility are reduced. Swelling power of native starch gradually increased at higher temperature ($40\text{--}90^{\circ}\text{C}$), but that of cross-linked starch increased up to 80°C and then decreased at 90°C . These results are in conformity with that of Carmona-Garcia et al. (2009) who reported that the swelling power of cross-linked banana starch increased with increasing temperature, but at 90°C , it decreased. This could be due to the bi-functional entities containing an activated double bond as well as hydroxyl/methoxy phenyl residues present in active ginger components such as gingerol, dehydroginger-dione and shogaol (Kikuzaki and Nakatani, 1993).

The solubility of native and cross-linked starches in water is given in Table 4. The solubility of the native starch was $1.1\text{--}8.5\%$ and those of the modified starches in the four sets of experiments were $1.9\text{--}7.1$, $2.8\text{--}6.4$, $2.2\text{--}7.1$ and $2.7\text{--}6.9\%$ respectively. The cross-linked starches showed lower solubility in comparison to the native starch. The solubility decreased with an increase in reagent concentration, which could be attributed to the increase in the degree of cross-linking and is also consistent with the swelling power observed. Due to the strengthening of the starch granules through cross-links, less disintegration took place during gelatinization and lowering of solubility. Cross-linking reduces the solubility which leach out of the swollen granules on cooking and the extent of solubilization is affected by the degree of cross-linking; the fewer the cross-links the higher would be the solubility (Wurzburg, 1986). A lower solubility of the cross-linked starches was reported earlier by

Table 4. Solubility (%) of native and modified cassava starches

Sample	Temperature (°C)					
	40	50	60	70	80	90
NCS	1.1±0.19	1.9±0.17	2.3±0.33	6.3±0.29	7.1±0.45	8.5±0.44
A	1.9±0.47	2.6±0.17	3.6±0.77	6.7±1.32	8.8±1.55	7.1±0.62
B	2.8±0.42	3.3±0.12	4.1±0.63	7.7±1.23	9.0±1.36	6.4±0.72
C	2.2±0.73	3.0±0.10	4.1±0.31	7.0±0.33	8.7±0.58	7.1±0.27
D	2.7±0.58	3.5±0.50	4.2±0.14	7.9±0.58	8.1±0.42	6.9±0.83
CD (0.05)	0.75	0.37	0.70	1.23	1.48	0.86

Mean ± standard deviation; NCS = native cassava starch; A = cassava native starch with 5% ginger; B = cassava native starch with 10% ginger; C = cassava native starch with 5% garlic; D = cassava native starch with 10% garlic

different workers (Liu et al., 1999; Hirsch and Kokini, 2002). According to Liu et al. (1999) who observed lowering of solubility of cross-linked waxy rice starches, cross-linking restricted swelling of granules and would also lower the solubility by increasing chain binding.

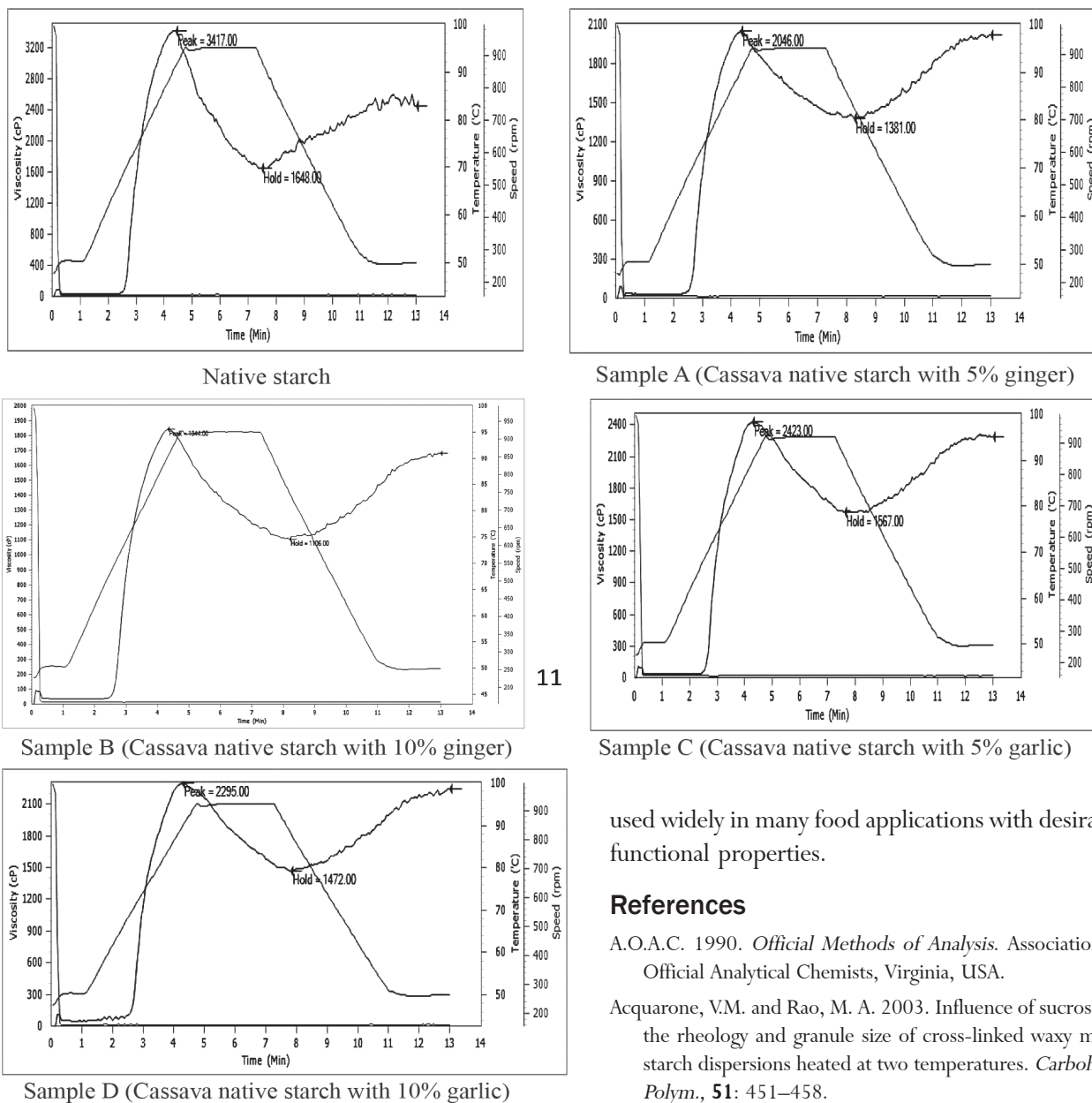
Pasting (RVA) characteristics of native and modified cassava starches are shown in Fig. 1 and Table 5. The peak viscosity of cross-linked starches (1975.0-2565.0) was lower compared to that of native starch (3733.7) and gradually it decreased with increase in the degree of cross-linking. Jyothi et al. (2006) and Xiao et al. (2012) indicated that the peak viscosity decreased at higher levels of cross-linking. Cross-linking strengthened the bonding between starch chains and increased the resistance of the granules to swelling, leading to low peak viscosity (Xiao et al., 2012). In the higher level of cross-links, there were sufficient cross-links to inhibit the swelling of starch and there was a decrease in viscosity (Kaur, 2004). Native starch had the highest trough viscosity (1747.3) than cross-linked starches (1082.0-1601.0). Trough viscosity decreased with increasing concentration of cross-linking agents. Cross-linked starches showed lower breakdown (770.0-963.3) than native (1986.3)

starch, which could be attributed to the strengthening of the swollen granules against breakage under conditions of high temperature and shear (Jyothi et al., 2006). Final viscosity of cross-linked starches (1678.0-2325.7) was significantly lower than native starch (2592.7). Final viscosity decreased with increasing concentration of cross-linking agents. The set back value of cross-linked starches (579.0-746.3) were lower when compared with the native starch (845.3) and it gradually decreased with the increasing degree of cross-linking. Jyothi et al. (2006) and Xiao et al. (2012) indicated that the setback values decreased at higher levels of cross-linking. The peak time of cross-linked starches (4.24-4.27 min) was higher than the native starch (4.24 min), but no significant differences were observed among the various starches. Pasting temperature of cross-linked starches increased in comparison to that of the native starch and it increased with increasing levels of cross-linking. This result is similar to the report of Jyothi et al. (2006) who stated that the pasting temperature of cross-linked cassava starches increased when compared to the native starch. Cross-linking of molecular chains makes the starch granules more ordered and consequently more energy would be required for swelling (Karthi and Srivastava,

Table 5. Pasting (RVA) characteristics of native and cross-linked cassava starches

Sample	Viscosity (cP)					Peak time (min)	Pasting temperature (°C)
	Peak	Trough	Breakdown	Final	Setback		
NCS	3733.7±533.8	1747.3±184.3	1986.3±349.9	2592.7±275.8	845.3±91.6	4.24±0.16	67.83±0.07
A	2136.0±193.7	1366.0±36.2	770.0±173.9	2075.5±77.0	709.5±74.1	4.26±0.31	67.82±0.02
B	1975.0±287.8	1082.0±33.9	876.0±256.5	1678.0±73.5	579.0±36.2	4.26±0.11	68.15±0.47
C	2565.0±239.1	1601.7±135.4	963.3±118.2	2325.7±146.1	724.0±11.0	4.24±0.04	62.38±10.2
D	2313.0±93.3	1425.0±153.8	887.3±79.3	2172.0±165.2	746.3±22.7	4.27±0.00	65.16±6.77
CD (0.05)	432.54	176.79	307.91	232.47	79.25	0.23	7.73

Mean ± standard deviation; NCS = native cassava starch; A = cassava native starch with 5% ginger; B = cassava native starch with 10% ginger; C = cassava native starch with 5% garlic; D = cassava native starch with 10% garlic



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Fig. 1. RVA patterns of native and cross-linked cassava starch

1985). With increase in the level of cross-linking, the gelatinization temperature also increased (Xiao et al., 2012).

Conclusion

The present study indicated that the active components of ginger and garlic at 5% and 10% concentrations modified the native starch of cassava with reduction in crude protein, water and oil absorption capacities. The peak viscosity of cross-linked starches was lower compared to native starch. The pasting temperature of native starch was higher than cross-linked starches. Cross-linked cassava starch could be

used widely in many food applications with desirable functional properties.

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