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PII: S0144-8617(96)00024-0

Carbohydrate Polymers 30 (1996) 275–286

0144-8617/96/$15.00 + .00

High moisture twin-screw extrusion of sago starch: 1. Influence on granule morphology and structure

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(Received 20 November 1994; revised version received 13 November 1995; accepted 15 November 1995)

Effects of barrel temperature (81–149°C) and screw speed (315–486rpm) on extrusion processing of sago starch in a co-rotating twin-screw extruder under a high moisture system (34–47%) were investigated using response surface methodology. Structural changes were characterised by measuring water solubility index (WSI), water absorption index (WAI), degree of gelatinisation (DG), dextrose equivalent (DE) and high performance size-exclusion chromatography (HPSEC) profiles of the extrudates. Thermomechanical processing of sago starch in the twin-screw extruder at the high moisture (34–47%) system led to shear-induced limited degradation and starch phase transitions (a composite melting gelatinisation process). Strong positive correlations between WAI, WSI and DG showed that gelatinisation was the fundamental mechanism in this high moisture system rather than dextrinisation. Processing-induced solubility increased at the expense of water absorption. Low WSI (4.5–18.1%) is ascribed to the presence of structures of either granular *crystallite* remnants or rearrangement of bonds during extrusion. Copyright © 1996 Published by Elsevier Science Ltd

INTRODUCTION

Development of extrusion cooking technology requires an understanding of how process variables and their interactions affect the thermomechanical transformation of a feed material (Vainionpää, 1991). Early studies on extrusion processing of starch are limited to investigating the effects of variables, such as temperature and feed moisture content, on product properties (Anderson *et al.*, 1969a, 1970; Mercier & Feillet, 1975). Such empirical studies have favoured the use of Response Surface Methodology (RSM). This technique, first introduced by Box & Wilson (1951), provides a useful approach for relating raw material and processing parameters with product attributes (Olkku *et al.*, 1983). Starch extrusion systems so-investigated include: wheat-based material (Olkku & Vainionpää, 1980; Meuser *et al.*, 1982; Paton & Spratt, 1984; Linko *et al.*, 1985; Vainionpää & Malkki, 1987 and Vainionpää *et al.*, 1984, 1989); corn grits or corn starch (El-Dash *et al.*, 1983; Owusu-Ansah *et al.*, 1983, 1984; Artz *et al.*, 1990); corn, rice and potato flours with whey protein concentrate (Kim & Maga, 1987); soya (Frazier *et al.*, 1983); cassava (Grossmann *et al.*, 1988); cowpea meal (Kennedy *et al.*, 1986) and sorghum (Phillips & Falcone, 1988). More detailed understanding of extrusion systems could allow response surface analysis to be used for estimating product characteristics given specific operational variables and system parameters, or to determine the required extrusion conditions given specific product characteristics.

Research has led to a growing awareness of the importance of variables (screw speed, feed rate and die geometry) which control the mechanical history and residence time of material in the extruder (van Zuilichem *et al.*, 1975; Owusu-Ansah *et al.*, 1983; Meuser *et al.*, 1984). Structural characterisation of extrudes derived from cereal starches have indicated that these materials have undergone macromolecular degradation (Colonna & Mercier, 1983), reflected as changes in melt rheology (Vergnes & Villemaire, 1987) and product functional properties such as water solubility (Fitton, 1986), water absorption and dispersion viscosity (Doublier *et al.*, 1986).

This study is a continuation of work previously

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reported (Govindasamy *et al.*, 1996a) on extrusion of sago starch in a Brabender single-screw extruder. In this investigation, twin-screw extrusion was used to pretreat sago starch for subsequent saccharification, focusing on employment of this technology as an integral part of a commercial processing operation. In this and a subsequent paper (Govindasamy *et al.*, 1996b) we report on the influence of extruding sago starch at high feed moisture contents, similar to those of the starch after centrifugation. Based on a multifactorial experimental design, extrusion conditions were identified and suitable trials carried out. Subsequently, the influence of extrusion processing parameters on saccharification were established. This paper reports on the structural changes induced during extrusion processing of sago starch as characterised by water solubility index (WSI), water absorption index (WAI), degree of gelatinisation (DG), dextrose equivalent (DE) and molecular weight profiles of the extrudates using high performance size-exclusion chromatography (HPSEC).

MATERIALS AND METHODS

Materials

Sago starch obtained from a commercial producer, PPES Sago Industries (Mukah) Sdn Bhd (Sarawak, Malaysia) was used as the feed material for all the experiments. Moisture content of the raw material was determined by drying to a constant weight in a convection oven at 120°C for 2h and found to be about 13% (wet weight basis; wb.).

Extruder

Experiments were conducted with a co-rotating intermeshing CLEXTRAL BC21 twin-screw extruder (Clextral Co., Firminy, France). The diameter of the screws was 25mm. The barrel, 400mm long, with a length to diameter ratio (L/D) of 16:1 was divided into four modules. The first module (100mm) was fitted with a feeding port. The temperature of the barrel was controlled by four iron/constantan thermocouples located inside the metal of the four barrel modules and cooled by water circulation. Cooling was provided by water from a closed-loop recirculator. All three zones were heated to set points by conduction heating element systems. The total configured screw length was 400mm.

The extruder was driven by an AC drive of 5.5kW (Model IP 55) and was operable over the range of 0 to 2000rpm. Starch was introduced into the extruder using a volumetric twin-screw feeder (Model T-20, K-Tron-Soder AG, Niederlenz, Switzerland). Frequent calibration resulted in a coefficient of variation of the starch feed rate close to 4%. A variable stroke dosing pump (Model DKM K20, CLEXTRAL, Clextral Co., Firminy, France) was used to add water to the feed solids. Water was introduced into the first module of the extruder.

Instrumentation and data collection

Four thermocouples (J type) were used to monitor barrel temperature along the length of the extruder in the four zones. A temperature probe J type (SAGANA 316L, France) and a pressure transducer (PT 415, Dynisco) were inserted into the die plate to measure product temperature and pressure at the die. Barrel temperature was controlled in four separate zones by Eurotherm 808 Digital Temperature controllers, which were microprocessor controlled (Eurotherm Ltd, Dardilly, France).

Screw configuration

The screws consisted of different modules of various lengths (25 and 50mm) and decreasing flight pitch (33.5, 25.0, 16.7mm) from feeder to die. Bilobal mixing cams or kneading discs of pitch 6.2mm were also used. The screw elements could be slipped onto and assembled on the splined shafts to obtain different configurations. The screw configuration that was used is shown in Fig. 1. The first two sections of the extruder were used only to receive and mix the sago starch feed and water to the required moisture content, and to convey this mixture to the final two working (or compression) zones. Each preceding bilobal cam is placed one turn anti clockwise on the splined shaft with respect to the previous cam.

Die assembly

The central feed die plate assembly consisted of a die with four 2mm diameter orifices fitted to a central feed die plate by two bolted clamps. The die plate was composed of a 10mm diameter cylindrical channel.

Extrusion conditions: multifactorial experimental design

In order to assess the effects of operating parameters on extrudate physicochemical properties, a central composite rotatable response surface experimental design was used (Mullen & Ennis, 1979). The design was based on five levels of three variables (Table 1). The three process variables selected were; *M*, total feed moisture (% wb); *T*, barrel temperatures in zones 3 and 4 (°C) and *S*, screw speed (rpm). All other parameters were kept constant including feed-rate (5.25kg/h), and temperature of the zones 1 and 2 (40 and 80°C, respectively).

Operating ranges and five standardised levels were selected, by experimentation, for each variable

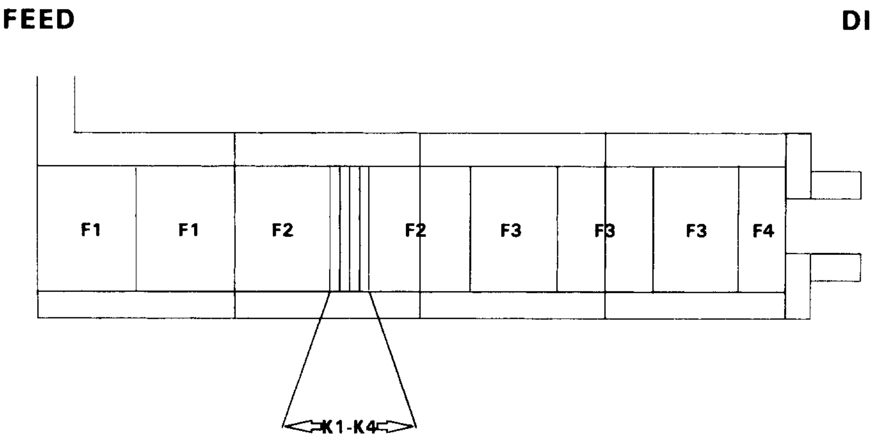

Fig. 1. Schematic diagram of the screw configuration used in extrusion processing of sago starch. F: forward screw element; F1 (pitch = 33.5mm, length = 50mm), F2 (pitch = 25.0mm, length = 50mm), F3 (pitch = 16.7mm, length = 50mm) and F4 (pitch = 16.7mm, length = 25mm); K: kneading disks; circular die with four orifices, ϕ = 2mm each.

Table 1. Levels of independent variables

		Levels				
Variable	Code	$-\alpha$	-1	0	$+1$	$+\alpha$
(1) Feed moisture (%)	M	34.3	37.0	41.0	45.0	47.4
(2) Barrel temperature (°C)	T	81	95	115	135	149
(3) Screw speed (rpm)	S	315	350	400	450	486

(Table 1). Among all possible combinations, 20 extrusion conditions were carried out in a random order (eight factorial points, six axial points and six replicates of the central point to form a central composite design with $\alpha = 1.682$) to allow the fitting of a second order model (Table 2). Coded values ranged from -1.682 to $+1.682$ across the response surface tested. The second order polynomials were computed with a statistical software (Statgraphics version 6.0, Mannugistics, Inc., Rockville, USA). Response surfaces were generated by showing the effects of two process variables, while the third was maintained constant, for each response.

Steady state operation

Before any intentional change was imposed on the extrusion system, initial steady state conditions were established and maintained for 10min. The system was considered to have attained steady state when measured process variables (torque, die pressure and mass temperature) became stable. Initial operating conditions were:

(1) Screw speed	400rpm
(2) Barrel temperatures	zone 1 = 40°C
	zone 2 = 80°C
	zones 3 and 4 = 115°C
(3) Feed-rate	5.25kg/h
(4) Feed moisture	37% (wb)

Added moisture (kg/h) was calculated using the method quoted in Lu *et al.* (1992).

Upon changing the operating environment, steady state was again established, data and samples collected for 15min.

Table 2. Values of independent variables of each experiment in dimensionless coded (m, t, s) and physical (M, T, S) units for the 2^3–2nd order factorial central composite experimental design

m	t	s	M (%)	T (°C)	S (rpm)
-1	-1	-1	37.0	95	350
-1	-1	$+1$	37.0	95	450
-1	$+1$	-1	37.0	135	350
-1	$+1$	$+1$	37.0	135	450
$+1$	-1	-1	45.0	95	350
$+1$	-1	$+1$	45.0	95	450
$+1$	$+1$	-1	45.0	135	350
$+1$	$+1$	$+1$	45.0	135	450
$+\alpha$	0	0	47.4	115	400
$-\alpha$	0	0	34.3	115	400
0	$+\alpha$	0	41.0	149	400
0	$-\alpha$	0	41.0	81	400
0	0	$+\alpha$	41.0	115	484
0	0	$-\alpha$	41.0	115	315
0	0	0	41.0	115	400
0	0	0	41.0	115	400
0	0	0	41.0	115	400
0	0	0	41.0	115	400
0	0	0	41.0	115	400
0	0	0	41.0	115	400

Drying and grinding of samples

Extrudates were dried in a convection oven at 40°C to constant weight and sealed in plastic bags prior to cooling. Extrudates were ground in a laboratory cross beater mill, type SK1 (F. Kurt Retsh Gmbh & KG, Germany) fitted with 0.5mm screen. All dry ground samples were stored refrigerated at 4°C in tightly screwed plastic bottles.

Analytical *methods*

Reducing sugar analysis

Reducing sugar was assayed according to a modified Park–Johnson method (Hizukuri *et al.*, 1981) using dextrose as the standard.

Water solubility and water absorption indices

Water solubility index (WSI) and water absorption index (WAI) were determined as described by Anderson *et al.* (1969a).

Degree of gelatinisation

Degree of gelatinised material was determined using a modified method (Govindasamy, *et al.*, 1996a) originally outlined by Wootton *et al.* (1971).

Sample preparation for high performance size-exclusion chromatography (HPSEC)

Samples were prepared and analysed by HPSEC using a modified method of Jackson *et al.* (1990) as outlined by Govindasamy, *et al.*, 1996a).

Dextrose equivalent determination

DE of the extrudates were assessed by measuring both the total dry weight and reducing sugar contents of the suspension. DE is defined as total reducing sugars expressed as dextrose and calculated as a percentage of the total dry weight.

RESULTS AND DISCUSSION

In extrusion processing of food materials, the physicochemical changes that occur are affected not only by thermal energy also by mechanical energy and possibly by pressure in the extruder (Wang *et al.*, 1991). Shear forces and thermal energy input result in starch gelatinisation in the extruder as well as substantial molecular degradation (Davidson, 1991). The product characteristics depend mostly on molecular transformations that have occurred in the extruder, ranging from compacting or sintering to complete disruption and degradation of the granular structure.

The effects of extrusion processing variables on molecular degradation could be explained using the equation developed by Rauwendaal (1986) that describes the volumetric rate of entropy generation ($\dot{S}_{\text{gen}}$) in non-compressible fluid flow:

$$\dot{S}_{\text{gen}} = \frac{1}{T}[\nabla k.\nabla T + \tau:\nabla v] \tag{1}$$

where k is the thermal conductivity, v is fluid velocity, T is the absolute temperature, and τ is the shear tensor.

∇v represents the velocity gradient applied to the material which is related to the shear rate. The symbol ':' indicates a tensorial product between the stress tensor and the velocity gradient tensor.

The first term on the right hand side ($\nabla k.\nabla T$) represents the heat supplied to the product by conduction, that is through the heating elements, which divided by the temperature gives the entropy increase of the product due to the external heating. The second term ($\tau:\nabla v$) represents the heat generated by friction, which divided by the temperature gives the entropy increase due to the friction. In the entropy generated by friction term, two effects are distinguishable; the effect due to the (a) product velocity or screw speed (v) and (b) shear stress (τ). At a constant moisture content as the screw speed increases the heat generated by friction increases. On the other hand for the same screw speed, decreasing the moisture content would result in the product viscosity increasing and so does the shear stress. For a non-Newtonian fluid, the shear stress is proportional to the apparent viscosity of the product (μ_{app}).

$$\tau = \mu_{\text{app}}\dot{\gamma} \tag{2}$$

As the shear rate ($\dot{\gamma}$) is related to the screw speed(s),

$$\dot{\gamma} \propto S \tag{3}$$

the shear stress is also proportional to the screw speed according to

$$\tau = \mu_{\text{app}}S. \tag{4}$$

The effect of screw speed is more important at a low moisture content when the product viscosity is high. At high moisture content, the product viscosity is relatively lowered and probably the effect of the screw speed on shear stress becomes less important. Thus, screw speed contributes predominantly to the entropy generated by friction which indicates the extent of degradation of the extrudate. On the other hand, at low moisture content the molecular degradation (or generated entropy) is affected by both screw speed and the shear stress. Thus, by substituting equations

[2] and [3], this results in the generated entropy due to friction (τ:∇v) is simply proportional to $\mu_{app}S^2$.

Macromolecular fractions

Extrusion of sago starch under low moisture, high temperature conditions results in rapid fracturing of granules and formation of stable fragments (Lim, 1993). In contrast, comparison of the extrudates produced under the milder conditions of this study (lower temperature and high moisture) with native solubilised starch reveals changes in macromolecular profiles but not to the extent as previously reported by Lim (1993). As shown in Fig. 2a–b, sago starch granules were fused into a coarse mass after extrusion cooking exemplifying gelatinisation. In addition, large amounts of flattened and sheared granules were observed.

Relative peak areas of the HPSEC chromatograms for each of the starch fractions of native starch (Fig. 3a) were calculated to be 70% for amylopectin (Ap), 27% for amylose (Am). A small peak eluting between the two major peaks, comprising of 3% of the total starch content, is believed to be an intermediate fraction. Structure of the intermediate material (Im) has been described as amylopectin with a lower degree of branching (Chang Rupp & Schwartz, 1988). Apparent molecular weight of amylopectin was estimated to be greater than 10^8 and that of amylose was about 8×10^5D as compared against dextran standards.

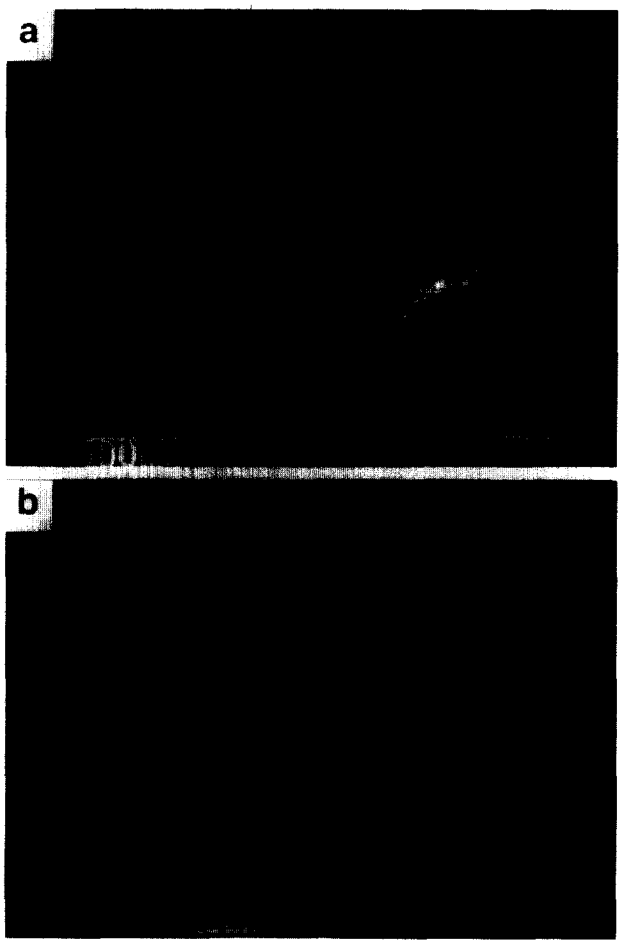

Fig. 2. Scanning electron micrographs of sago starch extrudates derived from twin-screw extrusion at 41% feed moisture, 115°C barrel temperature and 486rpm showing a coarse mass (C) with flattened and sheared granules (F) at (a) lower and (b) higher magnifications.

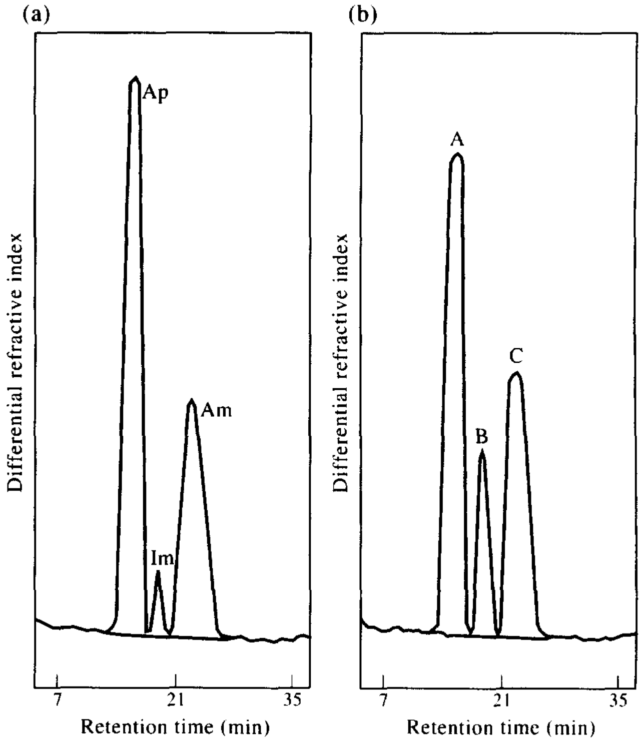

Fig. 3. HPSEC chromatogram of 0.5% (w/v), 1N NaOH solubilised native and extruded sago starch. (a) Native starch: Amylopectin (Ap), Intermediate (Im) and Amylose (Am); (b) extruded at 47% moisture content, 135°C barrel temperature and 350rpm screw speed: Peaks A, B and C corresponding to materials of apparent molecular weight smaller than Amylopectin, Intermediate material and Amylose respectively.

Three populations of molecules could be detected in solubilised extrudates (Fig. 3b); peak 'A' corresponding to an apparent molecular weight smaller than amylopectin, peak 'B' corresponding to the intermediate material and peak 'C' corresponding to amylose (MW = 10^5). Elution profiles of the high moisture extruded starch, with the exception of a slight shift in molecular weight of amylopectin and an increase in intermediate material were generally similar to those of native starch (Fig. 3a). Amylose was not affected by the extrusion process. This phenomenon is also evident during low moisture extrusion (Lim, 1993) and coextrusion with a thermostable α-amylase in the single-screw extruder (Govindasamy, *et al.*, 1996a). This also agrees with Craig & Stark (1984) who suggested that physical damage of wheat starch resulted in increased solubility of amylopectin compared to amylose, unlike the heating of starch in water that liberated amylose preferentially. There was little change in residence time of peaks 'A', 'B' and 'C' as compared to the chromatographic profile of native starch. It is suggestive that peak 'A' may represent the slightly degraded amylopectin and peak 'C' encompassing the native amylose and the degraded products with molecular size in the range of that of native amylose. The increase in relative area of peak 'B' (could be due to some degraded products).

An index for degree of degradation (DGR) of starch in the extrudates was arbitrarily assigned to a decrease in percentage of material present in the range of molecular weights corresponding to amylopectin. Degree of degradation was in the range of 0–10%. Limited degradation, as indicated by increasing amounts of intermediate material at the expense of amylopectin, is typical of shear-induced degradation where small molecular fragments are unlikely to form but the more shear sensitive macromolecular polymers are broken (Davidson, 1991). Variations in starch degradation with processing conditions under high moisture extrusion system was insignificant as DGR could not be described by a regression model. These results suggest that the changes in the processing conditions of high moisture extrusion (barrel temperature and screw speed) were too mild to cause significant starch degradation.

Dextrose Equivalent (data not shown) for all extrudates were less than 1. This substantiates the interpretation of HPSEC analysis in that there was little formation of small molecular weight compounds. Reinikainen *et al.* (1986) reported similar DE values (below 2) for products following thermomechanical extrusion processing of wheat starch and mono- and oligosaccharides were undetected. The DEs of the sago extrudates were affected significantly by all three variables; feed moisture content, barrel temperature and screw speed (Table 3).

Dextrose Equivalent increases with decreasing moisture content and increasing barrel temperature (Fig. 4). Higher mass temperature and mechanical energy generated at both these conditions would have enhanced starch macromolecular degradation.

Quadratic term for screw speed had a significant effect on DE (Table 3). As the entropy generated due to friction is proportional to μS^2 at lower hydration levels, increases in the quadratic term for screw speed would increase the entropy generated and consequently the macromolecular degradation. Furthermore, as expected the influence of the screw speed was markedly dependent on feed moisture content (Fig. 5). At high moisture contents, increase of specific mechanical energy with increasing screw speeds was presumably responsible for

Table 3. Best selected prediction equations for all dependent variables

Dependent variables	Independent variables	Coefficient	R^2† (adjusted)	*p*-value
DE	constant	0.051	0.80	0.0001
	M***	−0.027		
	T**	0.023		
	M^2***	0.036		
	T^2**	0.024		
	S^2*	0.015		
	M*S*	−0.039		
WSI	constant	5.764	0.94	0.0000
	M***	−2.119		
	S**	1.516		
	M^2*	1.079		
	T^2***	2.297		
	S^2***	4.054		
WAI	constant	2.784	0.87	0.0000
	M***	−0.374		
	M^2**	0.280		
	T^2*	0.484		
	S^2***	1.369		
	M*T**	0.508		
	T*S***	−0.565		
Degree of gelatinisation	constant	51.481	0.76	0.0000
	T**	8.387		
	M^2***	17.541		
	S^2*	6.039		

***: $p < 0.001$.
**: $p < 0.01$.
*: $p < 0.05$.
†: R^2 values were adjusted for the degree of freedom.

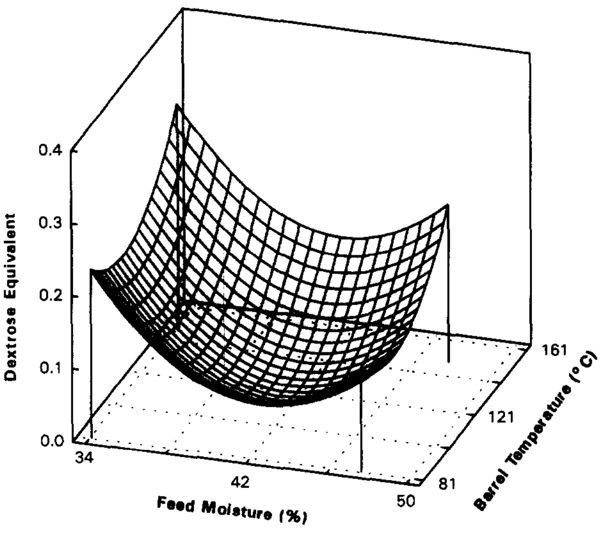

Fig. 4. Influence of feed moisture and barrel temperature on dextrose equivalent of sago extrudates at screw speed 400rpm.

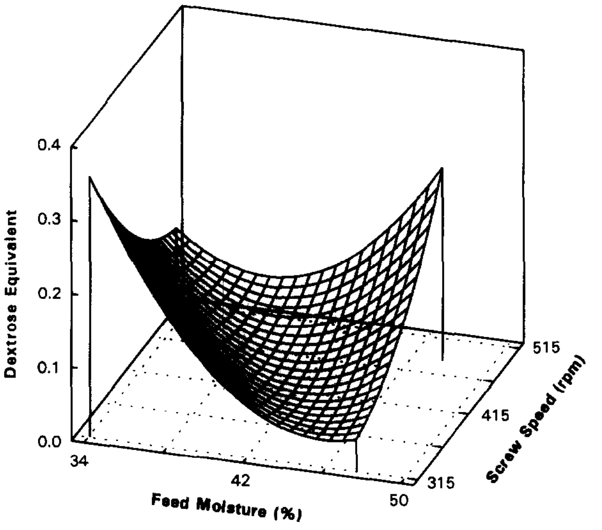

Fig. 5. Influence of feed moisture and screw speed on dextrose equivalent of sago extrudates at barrel temperature 115°C.

relatively more molecular degradation, manifested as higher DE values (Fig. 5).

Structural analysis of the extruded starch suggests that limited degradation of macromolecular components had occurred and it is therefore possible that many of the changes in the functional properties were caused by gelatinisation, resulting in various extents of granular structure relaxation. The latter is more likely to be the major mechanism occurring during high moisture extrusion rather than dextrinisation as in the low moisture processing of sago starch in a single-screw extruder (Lim, 1993).

Granule architecture

Changes in microstructure are shown in parallel studies of water solubility and absorption. Water solubility is often used as an indicator of degradation of molecular components (Kirby *et al.*, 1988). Following extrusion, water solubility of sago starch was increased (4.5–18.1%) compared to the native material which had negligible water solubility (less than 0.2%).

Absence of maltodextrins suggests that extrusion, under the conditions reported, partially solubilises sago without major breakdown of macromolecules. Increased solubility can occur as a consequence of changes to the macromolecular structure or as an independent mechanism leading to increased mobility of starch components resulting in leaching of carbohydrate material from entangled molecules (Colonna & Mercier, 1983). In addition, Jackson *et al.* (1990) had demonstrated that at least part of the increased solubility observed by other workers was due to an extrusion-induced increase in molecular solubility and dispersibility of amylose and amylopectin.

Volume of swollen gelled particles which maintain their integrity in aqueous dispersion is indicated by WAI (Mason & Hoseney, 1986). This index was found to correlate with dispersion viscosities measured at ambient temperature (Mason & Hoseney, 1986). Native sago starch with a WAI of about 1.60g/g dry matter is not unusual in respect to other starches. Water absorption capabilities of sago starch was increased on extrusion (2.73–7.07g/g). In contrast, sago starch extrudates produced under both low and high moisture systems, in a Brabender single-screw extruder, had lower WAIs. At both moisture levels (15–24% and 21–38%) in the single screw, WAIs (1.94–4.77g/g, at lower moisture and 2.89–5.24g/g, at higher moisture) were lower compared to other starch extrudates (Lim, 1993). In contrast, extrudates produced using the twin-screw extruder were comparable with products formed from other starches (Colonna *et al.*, 1989). Limited WAI and poor water solubility could well be related to inaccessibility of some compact structure to water (Meuser *et al.*, 1987). Such structures being ascribed to remnants of granular crystallites or rearrangement of bonds during extrusion.

Analysis of the relationship between WSI and WAI can indicate the type of mechanism responsible for increased water solubility. Within the experimental region, changes in WSI paralleled that of WAI ($r = 0.89$) (Fig. 6), due to an increasing proportion of granules attaining a state of gelatinisation (Smith, 1991). This state is attainable under the more severe processing conditions as thermal and mechanical energy is used to disrupt long range granular order. It is presumed that dextrinisation is not a predominant mechanism in this high moisture system as solubility does not increase at the expense of water absorption capacity. Water absorption index increases exponentially with increasing WSI (Fig. 6). Similarly, Kirby *et al.* (1988) observed an elevation of WAI with increasing WSI above 32% feed

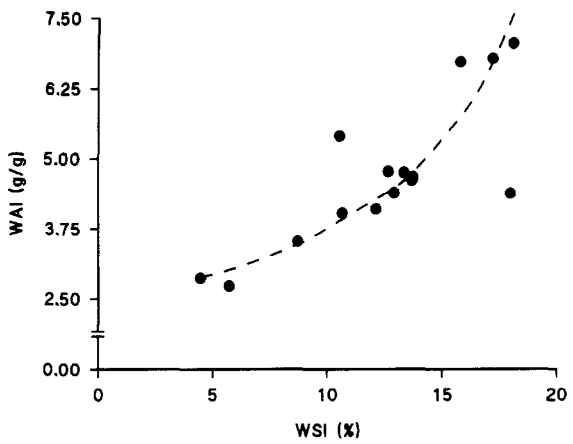

Fig. 6. Correlation between water solubility index and water absorption index (r = 0.89).

moisture and subsequent microstructure studies revealed that water absorption was related to the proportion of gelatinised material in the product.

Furthermore, significant gelatinisation (40–99%) of some of the products occurred upon extrusion processing of sago starch.

Effects of processing variables on structural modifications

Solubility of products was dependent on moisture content which has a decreasing effect (Table 3). At high moisture content (41%), there was no further increase in WSI (Fig. 7). Increases in WSI with reduction of moisture have been demonstrated for corn grits (Anderson *et al.*, 1969a, b; Conway, 1971), corn starch (Mercier & Feillet, 1975; Gomez & Aguilera, 1984) and wheat starch (Paton & Spratt, 1984).

Solubility goes through a minimum for barrel temperature at 101°C (Fig. 7) for all the moisture contents explored. At lower temperatures (below 101°C) the generated entropy ($\dot{S}_{gen}$) is mainly due to friction. On increasing the barrel temperature (above 101°C), the melt viscosity decreases so the generated entropy (indication of molecular degradation) is lowered. In addition, the contribution of heat conduction through heating elements becomes an important component at higher temperatures. Thus, elevation in the barrel temperatures (above 101°C) increases the generated entropy, suggesting higher molecular degradation manifested as improved WSI. Furthermore, the generated entropy would be greater at the lower moisture due to the higher product viscosity than the high moisture condition. Thus, the influence of barrel temperature is more pronounced at the lowest hydration level (34%).

The quadratic term for screw speed exerted a positive influence on WSI of the extrudates. However, with initial increases, there was a decrease (from 15 to 5.5%) in the WSI (Fig. 8). Increasing the screw speed above 385rpm led to an elevation in WSI. Initial increases in the screw speed would result in shorter residence time. At higher screw speed (above 385rpm), the shear rate effect predominates over the shorter residence time, raising the entropy of heat generation.

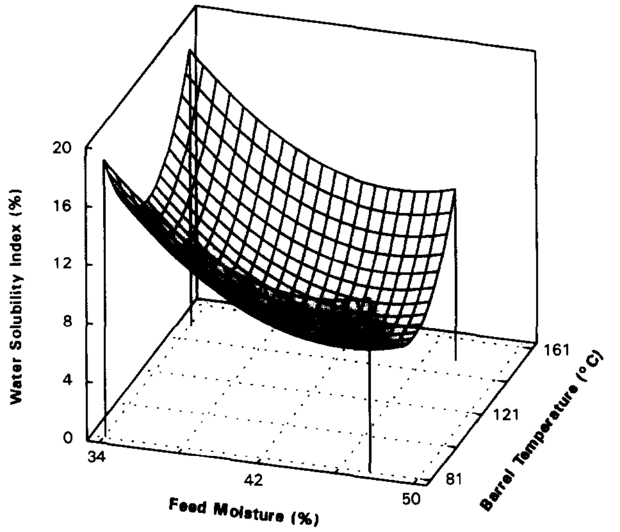

Fig. 7. Influence of feed moisture and barrel temperature on water solubility index of sago extrudates at screw speed 400rpm.

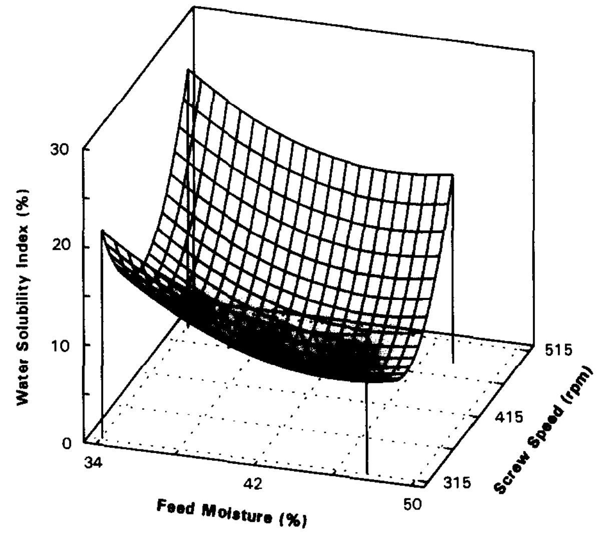

Fig. 8. Influence of feed moisture and screw speed on water solubility index of sago extrudates at barrel temperature 115°C.

Water absorption index was influenced significantly by feed moisture content, barrel temperature and the quadratic term for screw speed. Higher WAI was obtained at lower feed moisture contents (Table 3) and the effect of moisture content was dependent on barrel temperature (Fig. 9). At increased severity of thermal input (149°C) WAI decreases whilst the converse was observed at low thermal input (81°C) with reduction in hydration levels. Since at low temperatures, the entropy generated is primarily by friction, product viscosity is an important consideration. Changes in the moisture content would then affect this product property and

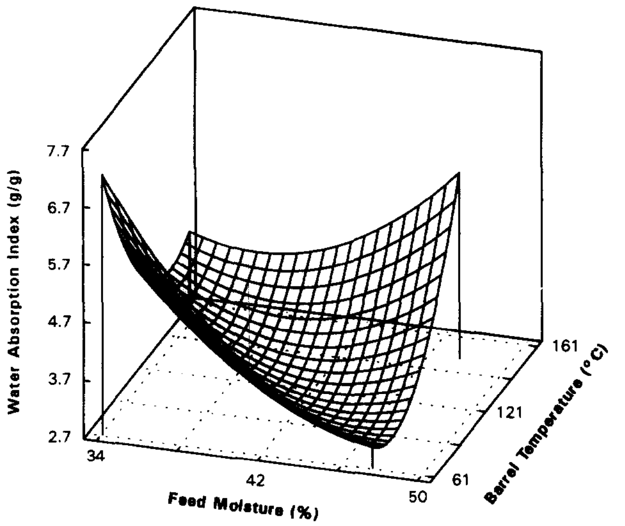

Fig. 9. Influence of feed moisture and barrel temperature on water absorption index of sago extrudates at screw speed 400rpm.

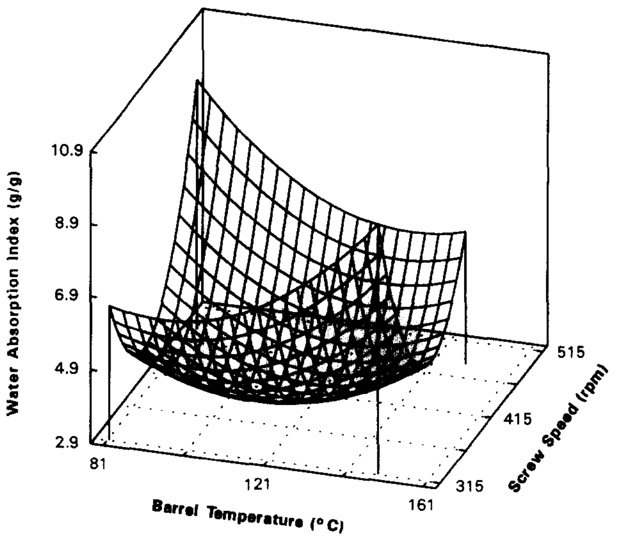

Fig. 10. Influence of barrel temperature and screw speed on water absorption index of sago extrudates at feed moisture 41%.

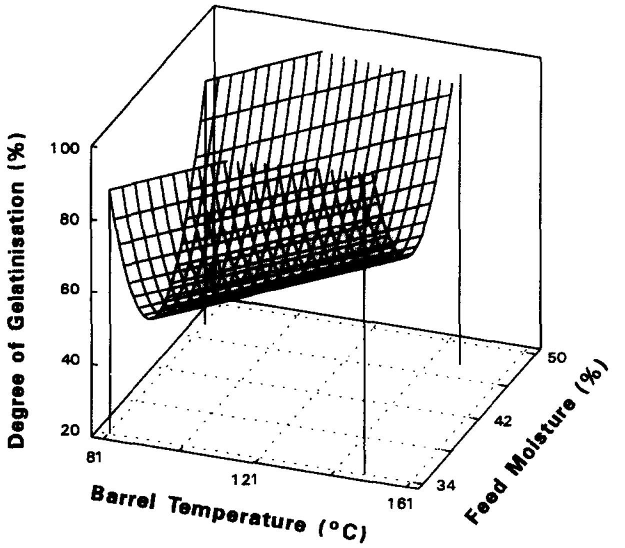

Fig. 11. Influence of barrel temperature and feed moisture on degree of gelatinisation of sago extrudates at screw speed 400rpm.

subsequently the WAI. At high temperatures, the entropy due to the heat conduction through the heating elements exerts a greater effect. Thus, changes in moisture content (or consequently the product viscosity) would have less influence on WAI.

Screw speed had the most significant effects ($p < 0.001$) on WAI (Table 3). The effect of increasing screw speed from 315 to 390rpm at lowest barrel temperature (81°C) produced a lower overall WAI of 6.7 to 4.7g/g. Further increasing the screw speed resulted in an overall increase in WAI to 7.4g/g (Fig. 10). The lower WAI of sago extrudates on increasing screw speed from 315 to 410rpm could be ascribed to shorter residence time whilst the higher WAI on further increase of screw speed from 410 to 486rpm could be related to elevation in shear rate resulting in structural modification.

The effect of barrel temperature was dependent on shear rate. At the highest screw speed (486rpm), there was a decline in WAI with elevation of the barrel temperature whilst at 315rpm, the reverse was seen (Fig. 10).

As expected increasing barrel temperature enhanced the degree of gelatinisation (DG) (Fig. 11). Feed moisture content exerted the greatest effect on gelatinisation. Increasing or decreasing the moisture content around 40% led to an elevation in the DG (Fig. 11). Processing under conditions of low moisture content of starch (below 40%) may restrict the material flow inside the extruder barrel, increase the viscosity and residence time which would increase the DG. With excess water (above 40%) acting as a lubricant, it should also be observed that at high moisture, the viscosity of the starch would be low, allowing for extensive internal mixing and uniform heating which would account for enhanced gelatinisation (Lawton *et al.*, 1972).

The quadratic term for screw speed was included in the model for DG. Increasing or decreasing the screw speed around 410rpm results in a rise in the DG (Fig. 12). The major factor in starch gelatinisation is granule swelling which depends on the strength and character of the micellar network within the granule, which in turn is dependent on the degree and kind of association. The swollen granules were increasingly susceptible to disintegration by the shear field existing in the extruder. Raising screw speed (325 to 410rpm) increases shear rate but also lowers the residence time, which reduces swelling making the granule less susceptible to shearing action. At higher screw speeds (410 to 485rpm) the shearing action presumably predominates over residence time accounting for the enhanced gelatinisation at these conditions.

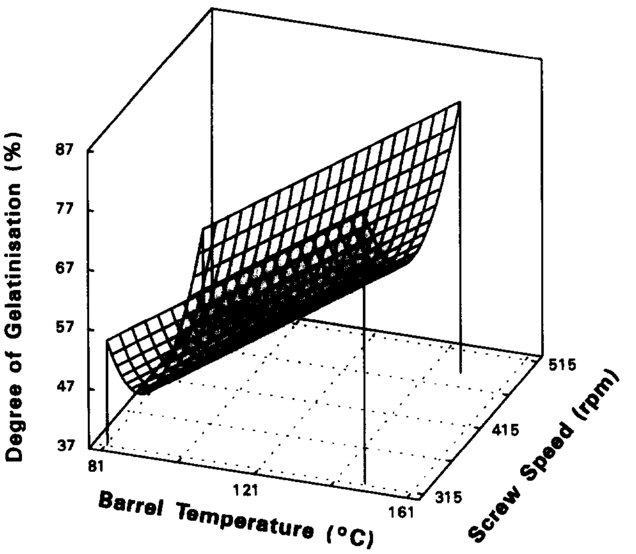

Fig. 12. Influence of screw speed and barrel temperature on degree of gelatinisation of sago extrudates at feed moisture 41%.

CONCLUSIONS

Thermomechanical processing of sago starch in the twin-screw extruder at the high moisture (34–47%) system led to both limited degradation, and starch phase transitions. Reactions in extruders, such as gelatinisation, melting, degradation as well as complexations between ingredients are influenced by the form and the intensity of energy inputs (Zheng & Wang, 1994).

Strong positive correlation between WSI and WAI emphasised that dextrinisation was not a predominant mechanism in this high moisture system (Table 4) but rather gelatinisation, which resulted in various extents of granular structure relaxation. Furthermore, DG was strongly correlated with DE ($r = 0.71$).

Limited degradation was apparent for the high moisture extruded samples since processing conditions were less severe. There was only a small amount of starch degradation. Despite the higher degree of gelatinisation, water solubility was found to be lower. Water absorption index range, on the other hand, was relatively higher and comparable to other starches. This disproved the significance of dextrinisation as the predominant mechanism in high moisture extrusion of sago starch. If dextrinisation had been an important reaction during extrusion, a much lower WAI and higher WSI would have been observed. In fact, gelatinisation was more likely to be the primary mode of starch modification in high moisture low temperature extrusion of sago starch. Additionally, the compact granular structure seemed to have become loosened to some extent facilitating water absorption.

Table 4. Correlation coefficients between dependent variables

	WSI	WAI	DG
WAI	0.8853***		
DG	0.2954	0.2239	
DE	0.5827**	0.4195	0.7118***

***: $p < 0.001$.
**: $p < 0.01$.
*: $p < 0.05$.
: not significant.

Extrusion cooking of starch leads to a degradation of amylose and amylopectin by chain splitting (Colonna & Mercier, 1983; Colonna *et al.*, 1983; Davidson *et al.*, 1984) due to effects of shear (Della Valle *et al.*, 1989; Davidson, 1991). However, in this present system amylopectin was degraded but as in the single-screw system, the amylose was apparently unaffected. Additionally mono- and oligosaccharides were undetected and the DE value remained below 1 after extrusion. Limited degradation, as indicated by increasing amounts of intermediate material at the expense of amylopectin, was typical of shear-induced degradation where small molecular fragments were unlikely to form but the more shear sensitive polymers were broken (Davidson, 1991). Extrusion, under the conditions reported, partially solubilised sago without major breakdown of the macromolecules. Sago starch granules were fused into a coarse mass after extrusion cooking, exemplifying gelatinisation.

Lower water solubilities measured for the samples reflected an increase in molecular solubility and dispersibility of amylose and amylopectin, disproving the occurrence of dextrinisation. Furthermore, solubility did not increase at the expense of water absorption capacity (Smith, 1991).

Structural analyses of the extrudates implicated the occurrence of limited shear-induced degradation of the macromolecular components and that many of the changes in the functional properties were rather caused by starch phase transitions.

The 'critical' water content for sago starch was determined to be 60% (w/w) (Fig. 13); at this moisture, there was only a single endotherm observed (Cui-Rong and Oates, submitted). As the feed moisture content used in the extruder ranged from 34–47%, it can be assumed that a composite gelatinisation melting process was occurring. At temperatures closer to or higher than phase transition temperature, the cooking process would be mainly caused by thermal energy input as the shear energy at such high temperatures was too low (due to low viscosity) to cause starch conversion (Zheng & Wang, 1994). As operating temperatures (81–149°C) used were higher than the T_p (peak temperature) in some conditions, both temperature and shear energies were influencing (Zheng & Wang, 1994) the composite melting gelatinisation process.

Physicochemical characteristics of the extrudates suggest that under these high moisture conditions,

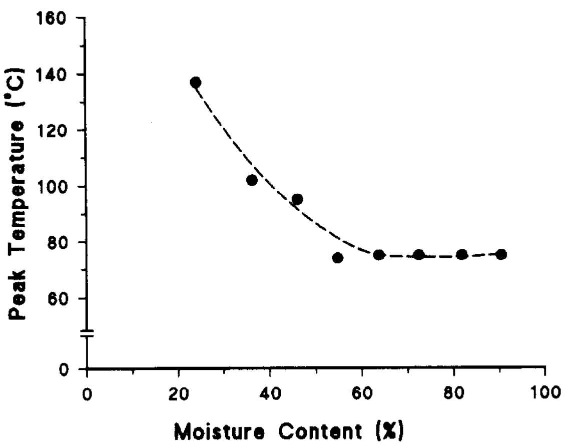

Fig. 13. Relationship between moisture content and peak temperature T_p of sago starch in differential scanning calorimetry (printed with permission from Cui-Rong and Oates).

starch macromolecular components undergo limited degradation induced by the effects of shear. It is postulated that starch granules are either split into smaller pieces or the surfaces are sheared but that, the crystalline structure is wholly or partially preserved within each fragment.

In a subsequent paper we describe relationships between structural characteristics and the thermomechanical history of the extrudate and the susceptibility of the extrudates towards saccharification.

ACKNOWLEDGEMENTS

We gratefully acknowledge the financial support made available through the National University of Singapore and Australian-ASEAN Economic Cooperation. We also wish to express our appreciation to Food Technology Department, Massey University, New Zealand for the use of the twin-screw extruder and both Brenda Looney and Byron McKillop for all their help.

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