

Original article

Effect of popping water content and amylose/amylopectin ratio on the physical properties of expanded starch products with different shear histories

Daniel Beech,¹ John Beech,² Joanne Gould^{1*} & Sandra Hill³

1 Division of Food, Nutrition and Dietetics, University of Nottingham, Sutton Bonington, Leicestershire, UK

2 Real World Business Solutions Ltd, Melton Mowbray, Leicestershire, UK

3 Biopolymer Solutions Ltd, Sutton Bonington, Leicestershire, UK

(Received 21 July 2022; Accepted in revised form 15 September 2022)

Summary Aerated starch products are a staple of the food industry, with particular relevance in the snack market. Water plays a crucial role in the formation of such products due to its utility both as a blowing agent and as a starch plasticiser. Amylose/amylopectin ratio and shear are traditionally also important factors in starch expansion. In this study, waxy, normal and high amylose maize starch variants were expanded using a rice-cake style popping head at water contents between 0% and 24%. This range of water contents was achieved by drying the material at 105 °C and then rehydrating by suspension over water at 50 °C until the desired water content was reached prior to popping. Sample types were further subdivided into low shear (native) and high shear (extruded) processing prior to popping. Processing history, amylose content and water content all influenced the water interaction properties of the popped products. However, density was largely governed by water content with little apparent influence of other factors. An optimum water content range to produce low-density (and therefore high desirability in industry) products was identified in the region of ~15–21% water. Samples popped below this range exhibited suboptimal expansion whilst those popped at higher water contents experienced violent blowout.

Introduction

Expanded, starch-based products are prevalent throughout the food industry both for human consumption and for use as animal feed. A significant factor in the desirability of such products in the human snack food market is the incorporation of air bubbles into the starch structure, which gives rise to a light, crispy texture. Water plays a crucial role in the creation of such products, acting both as a plasticiser and as a blowing agent (Sjöqvist & Gatenholm, 2007; Zubair *et al.*, 2020; Mitchell & Hill, 2021).

If all the water became steam in the processing and could be retained by the matrix, even at 1% water, there should be sufficient gas for entrapment to create products of the low bulk densities ($\sim 100 \text{ kg m}^{-3}$) often desired by the snack industry (Campbell & Mougeot, 1999). It would therefore seem that it is the waters' role in manipulation of the starchy matrix that would be key to the importance of the relationship between water level and expansion.

It could be expected that melted starchy matrices would behave very differently if starches with different amylose to amylopectin ratios were compared. Also, changing starch structure, by subjecting it to the temperatures and shear within a thermomechanical extruder, would be expected to alter the bubble wall structure as to affect a difference in expansion. However, findings in the paper by Beech *et al.* (2022) indicated that after being expanded in a popping head, at a water level of 12%, the bulk densities of native starch and powders created from starches that had been previously thermomechanically extruded were not greatly different. The variances, in terms of expansion, between waxy, normal and high amylose maize starches were also not obvious, except that native waxy maize starch, unlike the high amylose and normal maize starch, retained much of its crystalline structure when directly popped.

In addition to the starches, the only other ingredient manipulated in the work of Beech *et al.* (2022) was the water levels. As water molecules are such an integral part of the starch behaviour, it was considered that

*Correspondent: E-mail: joanne.gould@nottingham.ac.uk

doi:10.1111/ijfs.16092

© 2022 The Authors. *International Journal of Food Science & Technology*

published by John Wiley & Sons Ltd on behalf of Institute of Food, Science and Technology (IFSTTF).

This is an open access article under the terms of the [Creative Commons Attribution](https://creativecommons.org/licenses/by/4.0/) License, which permits use, distribution and reproduction in any medium, provided the original work is properly cited.

the starch/water interactions may have been such that the variances due to the starches were not obvious.

Water molecules are located in different areas within native starch granules. The crystal forms for the amylopectin in the maize starches are categorised as A type and eight water molecules are associated with each monoclinic crystal (Wang *et al.*, 1998; Bertoft, 2017). If the crystals can be melted, then this portion of water, along with other plasticisers, is available to increase the mobility of the starch chains and to lower the viscosity of the system. Positioning of the water in the granule is relevant for the melting of the crystallites within the native starch. Starch is considered to lose its crystal structure in different co-operative stages. Firstly, the water molecules need to mobilise the 'spacers' between the crystalline regions and that aids in the uncoiling of the helices (Waigh *et al.*, 1998; Vermeylen *et al.*, 2006). Waigh *et al.* (2000a, 2000b) suggested that at low water contents (<20%), there would be insufficient mobility of the spacer regions between the amylopectin helices of native starch and the crystals would have to melt directly and therefore would require higher thermal activation energy.

Water contents of starch after the initial loss of native order may be relevant to starch order. Recrystallisation during holding, when the starch is above the glass transition, may occur and affect performance (Ottenhof & Farhat, 2004). This may be relevant to the holding conditions of post-extruded samples used in this work where their moisture content was at ~26% during extrusion and are slowly dried down to ~11–14% for storage. The water content of the starches as they are processed and held will therefore be of relevance to starch structure, and it could be expected that different starch types could behave differently.

The extrusion process itself will have a marked impact on the starch structures and as the starch could be fragmented it could be expected that the low molecular weight elements would contribute to a reduction in the glass transition temperature (T_g) (Trommsdorff & Tomka, 1995; Sjöqvist & Gatenholm, 2007; Mitchell & Hill, 2021).

The mobility of the system is imperative for steam expansion. To entrap the steam, the matrix needs to be able to undergo biaxial expansion, and therefore, suitable alignment of starch molecules and their mobility are vital. The glass transition temperatures for starches at 12% water (wwb) are in the region of 100 °C, but the temperature for mobility increases to 150 °C if the available water for plasticisation drops to 5% (Van der Sman & Meinders, 2011). The loss of water, and hence, reversion of the plasticised rubbery matrix to a glass, is also very important for the matrix as it expands. During the expansion, phase vapour is

trapped inside the bubble. If the sample cools whilst the matrix is still mobile then, as the steam turns back to liquid and the pressure required to hold the expansion is lost, the bubble will decrease in size. Thus water/steam levels during expansion are dynamic and critical for creation of expanded networks.

A further complication on reconciling the water levels in the starch with its performance is the hysteresis in the a_w and water content. Samples may have the same water level, but can show differences in the a_w , which is indicative of the location of the water within the system (Aviara, 2020). This could be an important feature for the starches used in the current study as the water contents used were below and above that normally occurs for the starches when stored in ambient conditions.

The optimum water content to achieve starch expansion likely varies depending on the processing method and starch type but is typically quoted in literature as being in the region of 9–14% (Boischot *et al.*, 2003; Kraus *et al.*, 2013; Van der Sman & Bows, 2017; Panak Balentić *et al.*, 2018). In the current study, starches with different amylose and amylopectin ratios were studied at a range of moisture contents. Native starches and starches that had been previously extruded as pellets and then ground back to powders were compared. The method used for expansion was by popping using a specially created device similar to a rice-cake popping machine. In this way, the starches should reflect their starch structure when subjected to heat, but minimal additional shear. To gain some assessment of the state of the starch after their expansion, their water solubility and water absorbance was assessed (Devi *et al.*, 2009; Liu *et al.*, 2017; Ye *et al.*, 2018). The current study was undertaken to establish whether the samples handled water differently and that for maximum expansion the various samples would require nominally distinct water contents. If this was the case, then some understanding of the relationship between the samples and their expansion behaviour may be able to be elucidated.

Materials and methods

Materials

Three maize starches that represent different amylose: amylopectin levels were used for the work. The samples were popped without prior processing (native) or from extruded pellets (P).

Native maize starches of waxy (Amioca, Ingredion UK Ltd, Manchester, UK) normal (Meritena, Tereos UK & Ireland Ltd, Normanton, UK) and high amylose (Hylon VII, Ingredion UK Ltd, Manchester, UK) varieties were purchased from Univar UK Ltd (Bradford, UK).

Extrusion

All extrusion works were carried out using a Thermo Prism TSE MC co-rotating twin-screw extruder (Thermo Fisher Scientific Inc.) with a barrel length of 960 mm and bore diameter of 24 mm, fitted with a 4.1 mm diameter pinhole die. Starch powders were fed into the extruder at a constant rate of 8 kg h⁻¹ via gravimetric feeder, and RO water was pumped in at a rate of 1.536 kg h⁻¹, resulting in a total feed water content of 26% (wet basis), which included water associated with the native starch powders. Screw configuration and temperature profile, as well as actual recorded temperatures in heating zones which exceeded the set temperature, and specific mechanical energy (SME) are shown in Fig. 1. Screw speed was 300 r.p.m. The die temperature was set at 60 °C to produce unexpanded starch pellets. The extruder screws had identical configurations, consisting mainly of forward conveying elements with small regions of shear mixing elements to increase SME.

Following extrusion, samples were dried at 105 °C for 2 h and left out on trays to equilibrate overnight prior to long-term storage in plastic containers.

Water content adjustment

Uncooked starch powders and extruded pellets were milled using a coffee grinder (De’Longhi Appliances Srl, Treviso, Italy) if necessary and passed through a 106 µm sieve. All samples were then dried at 105 °C until constant weight. These dried samples were then suspended over water in foil trays within a closed container kept at 50 °C until they had rehydrated to the desired water content, as measured by weight increase. Samples considered to be 0% water were not rehydrated and were instead popped immediately upon drying to constant weight.

Popping

The sieved, hydrated powder samples were finally expanded or ‘popped’ using a heat press (custom build, University of Nottingham) to create final products similar to miniature rice cakes. Samples (1.8 g) were weighed into an aluminium pie tray which had been moulded to fit the dimensions of the heat press base platen recess (diameter 45 mm, depth 10 mm). The heat press head and base platens were both set to 230 °C, and all samples were pressed at this temperature for 4 s at a gauge pressure of 4 bar.

Analyses

Product density

Product density was calculated using rapeseed displacement (AACC, 2000).

Water absorption and solubility indices

Popped samples were ground, sieved (106 µm) and dried at 105 °C. Powdered sample (0.3 g) was mixed with 10 mL of RO water and left to hydrate and sediment overnight. The resulting suspension was centrifuged at 2045 g for 10 min, and the supernatant was evaporated at 105 °C. Water absorption index (WAI) was calculated as the weight of the pellet per gram of sample, and water solubility index (WSI) was calculated as the weight of the dry solids in the supernatant as a percentage of the initial sample weight. This method is a modified version of that described by Anderson *et al.* (1970).

Differential scanning calorimetry (DSC)

Approximately 8 mg samples were mixed with RO water in a ratio of 1:3 sample to water and hermetically sealed in stainless steel DSC pans. Samples were

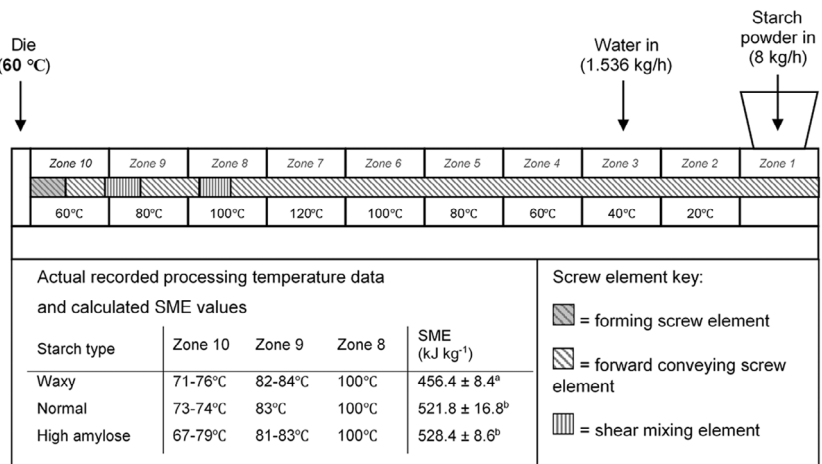


Figure 1 Screw configurations and set temperature profiles used to create unexpanded starch pellets. Actual temperatures recorded in zones 10, 9 and 8 are given as well as the calculated SME obtained from two replicates ± standard deviation. Subscripts denote samples significantly different one from another at the 5% level.

scanned from 0 to 150 °C at a rate of 10 °C min⁻¹ using a DSC 3+ (Mettler-Toledo Ltd, Leicester, UK).

Polarised light microscopy

Comminuted samples were diluted approximately ten-fold in RO water for microscopic analysis under polarised light. A Nikon Eclipse Ci microscope was used to generate 20× magnified, polarised images which were captured using NIS-Elements software (Nikon UK Limited, Surbiton, UK).

Statistical analysis

Statistical analyses were carried out using Genstat 21st Edition software. Two- or three-way ANOVA, as appropriate, followed by the Tukey test was used to identify significant effects of factors, interactions between factors and relationships between data groups.

Results and discussion

Sample hydration prior to popping

Rehydration of powdered samples generally had little effect on their observed physical properties, although some clumping occurred as water content increased. These clumps were manually dispersed before the samples were weighed out for popping. An exception to this was when waxy maize starch samples, having been extruded to pellets, ground, dried and hydrated to water contents greater than 15%. In these samples, the powder coalesced into a solid, rubbery block, which could not be returned to powder form in a way which preserved water content. Consequently, no extruded waxy starch samples in the water content range of 18–24% could be popped or analysed. Commercial pre-gelatinised starch (PGS), which can be produced by extruding waxy maize starch, is known to form a hard-homogenous gel when dispersed in water (Ayed *et al.*, 2020). Whilst the water contents used in this study are insufficient for this to occur, PGS has been found to preferentially adsorb water at particle surfaces before deeper penetration into the particles takes place. As such, these surface interactions may allow the formation of a continuous solid even with incomplete hydration of the ‘powder’ as a whole.

Experiments in native and pregelatinised wheat starch have found that T_g of both materials fell below room temperature at moisture contents above 22%, and therefore in the rubbery state (Zelezak & Hosney, 1987). T_g of maize starches has been found to increase with amylose content (Liu *et al.*, 2010) in part due to the relative mobility of amylopectin branch points and the ability of linear amylose chains to form partially crystalline interactions with each other, increasing the relative energy required to separate

amylose entanglements during gelatinisation compared with those of amylopectin. This is supported by observations in the present study that the waxy starch experienced more clumping following the hydration step than did the amylose-containing starches. It therefore seems likely that the formation of a rubbery block by the extruded waxy maize starch at high water contents is caused by surface hydration of the starch particles reducing the T_g below room temperature. Within the water content ranges investigated in the present study, the samples with higher amylose contents maintain their T_g above room temperature, and therefore, these samples are in the glassy state.

Sample creation

As can be seen in Fig. 2a, samples popped at water contents of 0–6% generally lack structural integrity and readily disintegrate. The products are also flat with minimal, patchy expansion. Starting from 9% water, the samples are more cohesive, forming discs which remain intact when handled. Expansion is also more uniform and greater in magnitude in these samples. Crumbling is still observed at the edges of some samples popped at water contents below 12%, particularly in the high amylose starches. As the popping water content reaches 18% and above, the disc shape of the samples begins to become distorted as a result of the polymer melt escaping from the heat press platen. This edge effect could reflect the low viscosity of the sample allowing sample to seep between the side and heat press head, or that the gap between the sides and the head allows steam to escape, removing a portion of the starchy material as it does so, or it could be a combination of these factors. This phenomenon of sample blowout becomes particularly apparent at 24% water, where excess expansion can be seen in all sample types, and in the samples produced from native waxy and high amylose starches, there is evidence of bubble wall collapse resulting in shrivelled, fractured structures.

As the popping process involves the application of both pressure and temperature the means by which fused, homogenous samples, particularly those at low water contents, were produced is not clear. Whilst the platen temperatures of 230 °C should be more than sufficient to cause melting in both amylose and amylopectin (Delcour & Hosney, 2010), the short cooking time of four seconds is unlikely to allow all of this heat to transfer to the sample. As such, the formation of disc-like products may have been the result of particle jamming rather than a true fusion brought about by melting. To determine the role of jamming in product formation, native powders of the three starch types were pressed in the popping head (4 bar gauge pressure) at their unmodified water content (~12%) for

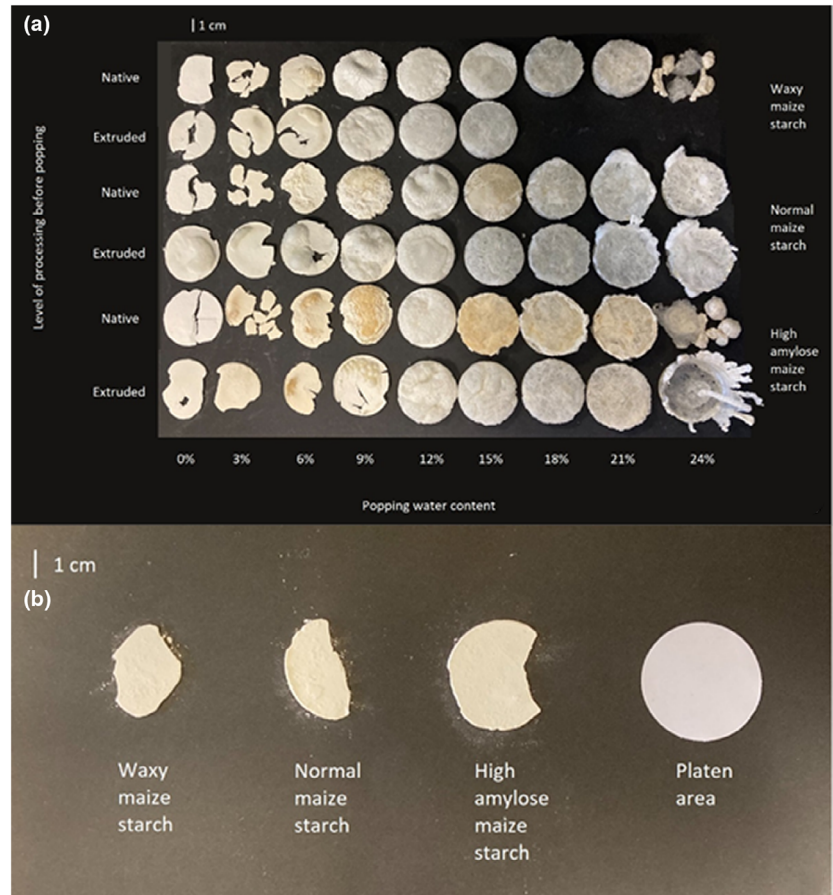


Figure 2 Representative examples of (a) the popped products made for each starch type, processing level and popping water content and (b) samples made by pressing native starches in the absence of heating.

one minute with the heaters turned off. Representative samples obtained are shown in Fig. 2b. These samples initially took the form of continuous disc shapes moulded to the shape of the platen recess. However, they were very fragile and readily snapped and crumbled when moved or handled. This behaviour is shared with the heated samples of water content $\sim 0\text{--}6\%$, which suggests that the primary factor in the shaping of these samples is particle jamming rather than melting. The cold-pressed samples appear to better resist crumbling as amylose content increases, but this was not quantified. The relationship between the cold-pressed samples and amylose could be due to the heterogeneous shapes of high amylose starch granules and higher packing density (Cai *et al.*, 2014).

Water content alone may be insufficient to predict the popping behaviour of starches and their subsequent functional properties. Also, important is the manner in which the water content was reached. Starchy materials show hysteresis in their water sorption isotherms. When dried and rehydrated, starches show a_w values greater than in untreated material for the same water content (York, 1981; Delcour &

Hoseney, 2010; Aviara, 2020). The hysteresis may occur because the limited absorbed water associates in a different manner with the pre-dried material. When, on drying, water molecules vacate the pores between the starch, the starch molecules associate and this association is not broken by the reintroduction of the condensing water vapour, and therefore a higher content of free water (Al-Muhtaseb *et al.*, 2004).

Native starch samples were popped at 12% water, either directly from untreated powder or after having been dried and rehydrated as described in the materials and methods section. The properties of these samples are provided in Table 1. Hydration method and starch type were found to significantly affect WAI, WSI and density ($P < 0.05$). There was also significant interaction between these factors for WAI and WSI ($P < 0.05$), but not for density. Waxy starch, which had been dried and rehydrated, had higher WAI and lower WSI than the untreated equivalent, suggesting that less overall starch conversion occurred when popping the rehydrated sample. This aligns with the concept that the rehydrated sample, although having the same water content, has water molecules in less

Table 1 Functional properties of samples popped at 12% water, either directly from native starch powder or after having been dried and rehydrated

Hydration status	Starch type	Water Absorption Index – WAI (g g ⁻¹)	Water Solubility Index – WSI (%)	Density (kg m ⁻³)
Native	Waxy	3.68 ± 0.29 ^a	52.89 ± 3.53 ^d	302.57 ± 98.77 ^b
	Normal	4.70 ± 0.16 ^c	17.78 ± 0.19 ^b	147.96 ± 23.25 ^a
	High amylose	3.92 ± 0.10 ^{ab}	8.78 ± 0.77 ^a	227.00 ± 75.39 ^{ab}
Dried and rehydrated	Waxy	7.54 ± 0.12 ^d	33.67 ± 1.76 ^c	201.29 ± 30.23 ^{ab}
	Normal	4.48 ± 0.34 ^{bc}	9.44 ± 1.17 ^a	142.25 ± 36.64 ^a
	High amylose	3.91 ± 0.37 ^{ab}	8.67 ± 5.51 ^a	143.75 ± 16.56 ^a

Average values (at least three replicates) ± standard deviation. Samples within the same column labelled with different letters are statistically different to one another to a significance level of $P < 0.05$.

intimate association with the non-dried starch molecules. For the normal starch, WSI was reduced when rehydrated compared with untreated, whilst WAI was not affected which may also suggest a slight reduction in starch conversion. No significant differences in WAI and WSI were found between rehydrated and untreated high amylose starch samples, but it is known that for the high amylose starches the hysteresis in the isotherms is small and is considered to reflect that amylose will be more strongly internally associated than amylopectin than the water fraction (Al-Muhtaseb *et al.*, 2004).

Density of the popped cakes was not significantly affected by rehydration in any of the starches, although collectively density was lower for samples popped from rehydrated starches than from untreated starches. Whilst untreated waxy starch had significantly higher density than normal starch, the rehydrated waxy starch density was not significantly different to any of the other samples. It seems that the hysteresis phenomenon in the rehydrated starches leads to an overall reduction in starch conversion despite increasing a_w . This may be due to a reduction in the ability of water to penetrate deep into the granule and disrupt amylopectin coils. This is further supported by the observation that the effect of hysteresis seems to reduce as amylose level increases and, by extension, amylopectin level decreases.

Starch structural changes

DSC analysis of the samples revealed a number of trends in the crystallinity remaining in samples after different processing regimes (Fig. 3). Firstly, at popping water contents at and below 12% (native starch moisture), there was a large amount of crystallinity remaining after popping in the samples that were not extruded, except for the high amylose variant whose endotherm broadens and flattens considerably between 0% and 12% water. This is reduced considerably in the samples which were extruded before popping. At 15% water and above, the original endotherms have

been lost. The exception to this is the waxy starch popped from native, which retains a small amount (~7%) of its original crystalline structure at 15% water but ultimately does lose all crystallinity by 24% water. Whilst the original crystalline order is lost almost universally at popping water contents of 15% and above, some samples see the emergence of new/shifted endotherms. This is particularly apparent in the non-extruded high amylose samples in which, despite initially losing crystallinity, the endotherm re-emerges as the water content increases further, albeit shifted downward in temperature.

These observations are largely supported by observation of the samples *via* polarised light microscopy, the micrographs from which are presented in Fig. 4. The prevalence of intact starch granules generally reduces with increasing water content, as well as being far fewer in extruded samples compared with native samples at all water contents. Interestingly, there is evidence of film forming in the micrographs associated with the samples popped from extruded high amylose maize starch, most notably at water contents of ~12% and above.

In terms of starch conversion, the waxy and normal maize starches behave much as would be expected. There are clear signs of starch conversion in the DSC endotherms and micrographs of both starches, increasing with popping water content and occurring to a much greater extent in the extruded pellets. WAI and WSI (Fig. 5) also provide evidence of this. The waxy samples popped from native starch initially increase in both WAI and WSI as the starch structure opens up. The WSI continues to rise, whilst the WAI falls as water content continues to increase as the amylopectin is further degraded. In comparison, extruded waxy pellets start at a much higher WSI, which continues to rise whilst WAI falls with increasing popping water content because the processes of fragmentation and dextrinisation have already begun during the extrusion step. Similarly, the native normal samples showed an increase in both WAI and WSI as water content increased. The WAI did not begin to decrease as it did

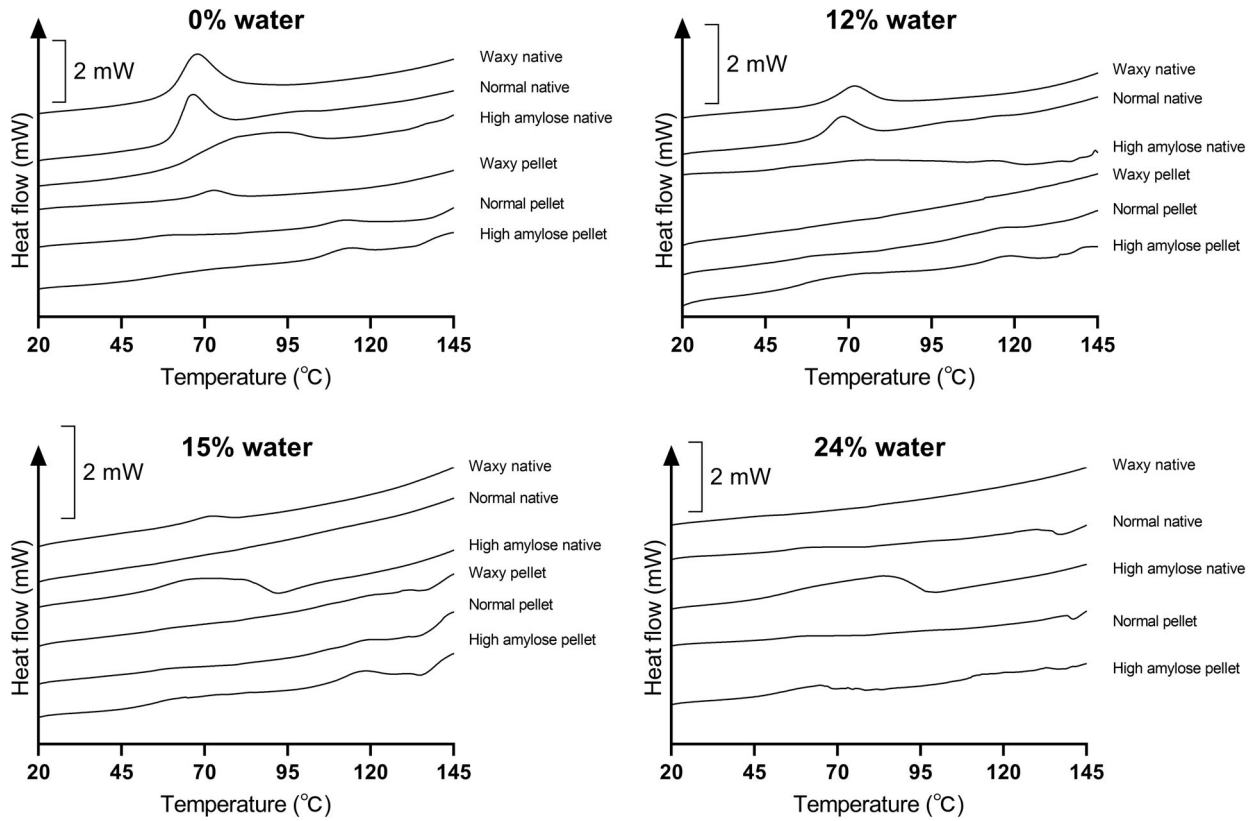


Figure 3 DSC thermograms of expanded starch samples when heated in excess water, having been popped (using native or pelleted extrudates) at different water contents.

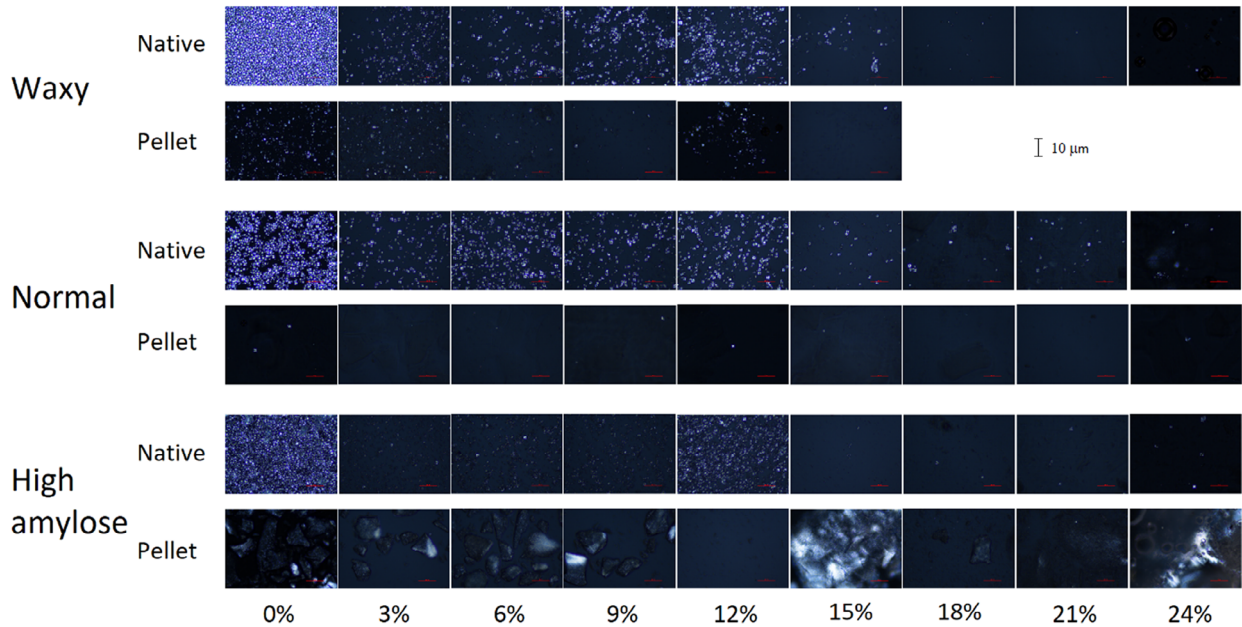


Figure 4 Polarised light micrographs of expanded starch samples popped at different water contents.

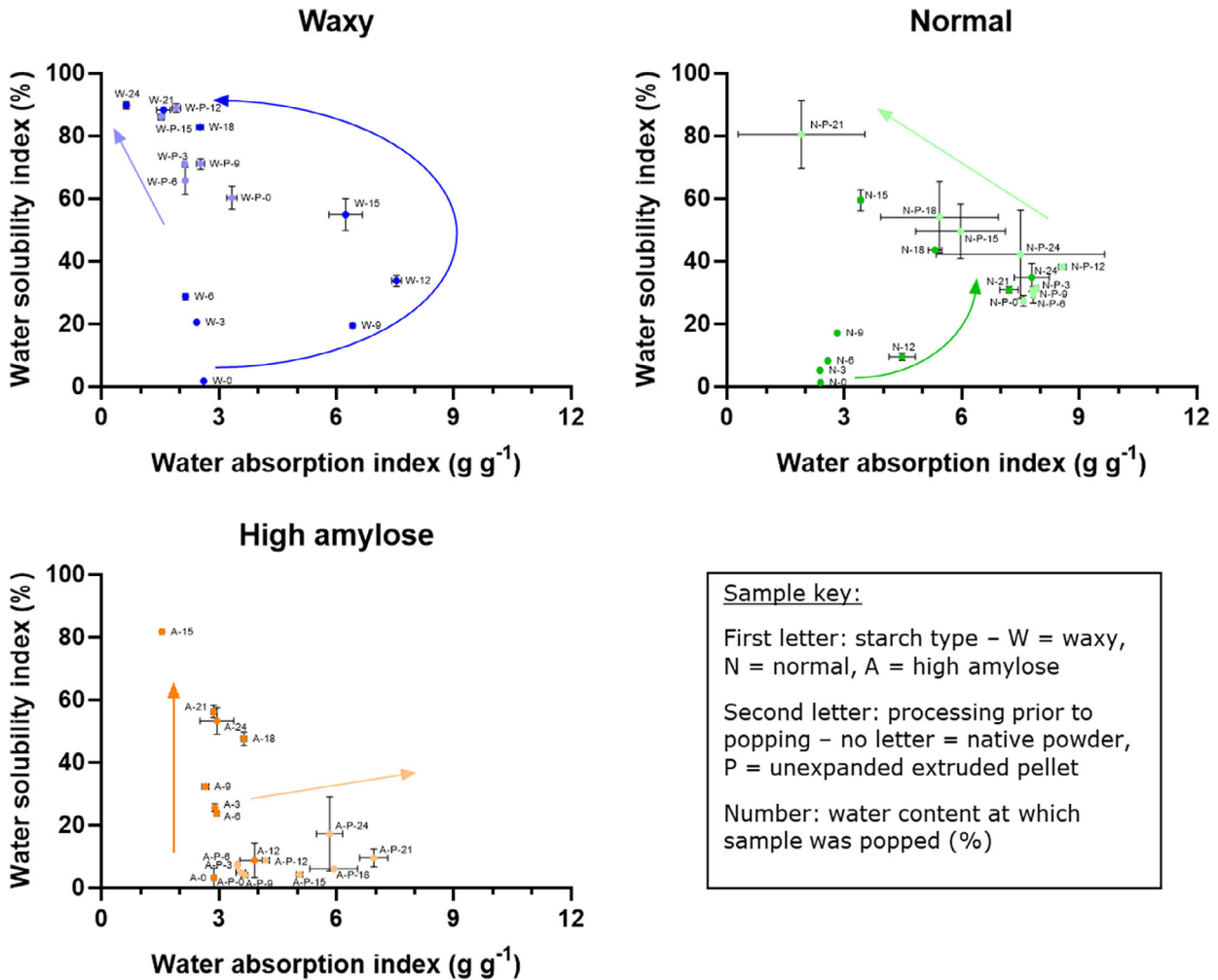


Figure 5 Water absorption and solubility indices of the popped starch samples. Arrows indicate approximate direction of increasing water content prior to popping for each sample type based on amylose/amylopectin ratio and shear history.

in the waxy samples presumably because the amylose in normal maize starch is more resistant to physical modification and degradation as well as reinforcing the amylopectin structures and promoting retrogradation. The extruded normal pellets initially have high WAI and WSI compared with their native counterparts, with WAI decreasing and WSI increasing as water content rises. It seems likely that in this case, the combination of both extrusion and popping, and in particular the high shear of extrusion, are sufficient to degrade and dextrinise the amylose-containing starch. The high amylose starch, on the contrary, responds to the processing very differently to the other starches. Increasing the water content at popping generally increased WSI in the native high amylose starch, but had little influence on WAI. However, for high amylose starch which had been previously extruded, increasing the popping water content led to increased

WAI as well as a minor increase in WSI, although much less so than was observed in the native samples.

The water content of the popped products, after storage at room temperature, was assessed by drying and is presented in Fig. 6. The general trend observed in all starch types and processing histories was a positive correlation between popping water content and product water content. The range of product water contents is much smaller than that of the popping moisture contents (1.8–9.2% vs 0–24%), with all samples popped at 6% water and above losing water, and all samples popped at 0% water gaining water in the time after popping.

Product expansion/density

Taking 12% water prior to popping as a midpoint, reducing the water content leads to a product with

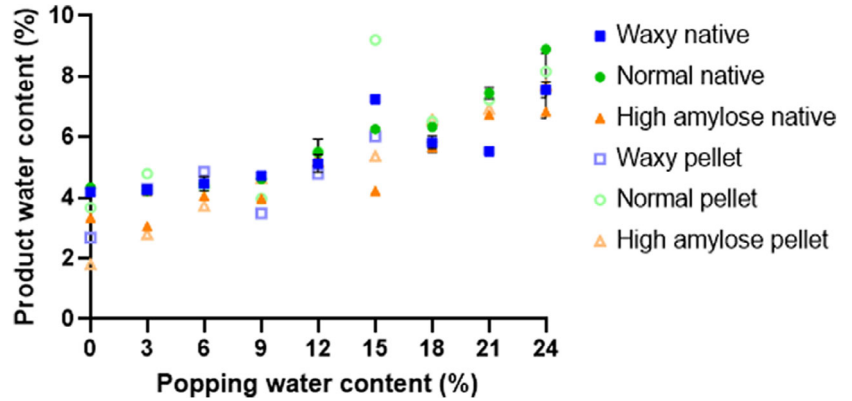


Figure 6 Water contents of expanded starch samples after popping and having been allowed to equilibrate at room temperature vs the water contents at which they were popped.

higher density, generally increasing with each 3% step down regardless of starch type or shear history (Fig. 7). This effect is particularly pronounced in the extruded high amylose samples. Whilst density is also affected by water content in the >12% samples, the differences are far smaller. This occurs despite evidence that the extent of starch conversion continues to rise in these samples, suggesting that the dominant factor affecting product density is the amount of blowing agent available to drive expansion. Minimum density is achieved in the approximate region of 15–21% water before popping. Based on these observations, it seems that sufficient blowing agent is present for maximum expansion to occur from 15% water and that raising the water content above this value does not confer any additional benefit. Starting from water contents of approximately 15%, it was observed that sample material began to escape from the heated platen during the popping process. This phenomenon was also observed and termed ‘blowout’ by Orts *et al.* (2000) at water contents greater than 18.5% when popping mixtures of wheat starch and whole grain brown rice at 210 °C for 10 s. They reported that blowout did not affect density measurements but presented a practical limitation in that

substantial loss of material would not be tolerated in industrial processing. In the present study, it has been observed that the blowout phenomenon also leads to irregularly shaped products, which presents an additional problem as this lack of uniformity is also unlikely to be tolerable in industry. Furthermore, whilst Orts *et al.* (2000) popped at water contents up to 22%, the present study found that at 24% moisture, the blowout became so severe in some samples (native waxy and high amylose and arguably extruded high amylose) that a portion of the sample material was cooked on the outside of the heated platen. This portion of the sample lost water throughout the cooking time and ultimately collapsed leading to drastically different density and texture. Based upon the prior considerations, the optimum water content range for starch expansion when popping under the experimental conditions is deemed to occur in the region of 15–21%, with lack of blowing agent being the limiting factor below this range and blowout/bubble collapse becoming dominant above. It is also in the range of 15–21% popping water that most samples had industry-desirable densities of 100 kg m^{-3} (Campbell & Mougeot, 1999), with 18% being the only water content at which all samples reached this

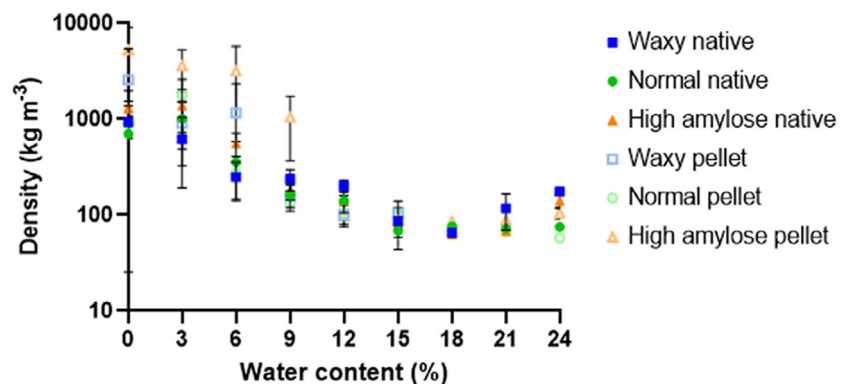


Figure 7 Densities of expanded starch products popped at different water contents as measured by rapeseed displacement.

benchmark regardless of starch type or processing history. This optimum range is higher than is generally reported in literature, where values quoted typically fall in the region of ~9–14% (Boischoit *et al.*, 2003; Kraus *et al.*, 2013; Van der Sman & Bows, 2017; Panak Balentić *et al.*, 2018). One possible explanation for this is that the popping head creates a closed system in which starch melting and superheating of sample water can occur. This could theoretically allow for greater quantities of steam to flash off at once when the pressure is released and therefore exert a greater evaporative force on the starch bubble wall. Compared with more conventional popping methods such as microwaving, which due to being an open system loses water as steam continually throughout the cooking process, more benefit may be derived from increasing water content in such a closed system.

Conclusion

During starch expansion *via* popping head, water plays a crucial and complex series of roles, acting not only as a blowing agent but also as a plasticiser and in promoting starch conversion and loss of starch macromolecular order. Whilst physical jamming of particles can account for a proportion of the formation of continuous, cohesive products, jamming alone gives rise to delicate and brittle shapes both with and without the application of high temperature. Therefore, it is clear that to achieve a truly fused product it is insufficient to apply only heat and/or pressure and that water is required to plasticise and convey mobility to the starch. The response of starch to popping is affected by its hydration history. Native starches that are pre-dried and then hydrated showed reduced levels of starch conversion as compared to native starch granules at the same moisture content. This is likely due to the removal of and inability to replace water bound deep within starch granules and which provide mobility to the amorphous portions of the granules, thus allowing loss of order to occur. This phenomenon is reduced as the amylose/amylopectin ratio increases. Furthermore, the hydration mechanics of starchy materials were influenced by both starch type and shear history, most apparent in this study from the formation of a rubbery mass from extruded waxy maize starch at water contents of 18+% despite incomplete hydration. An optimum water content at which to carry out the popping process, in order to achieve the most aerated/lowest density products, was identified in the region ~15–21%. This range was consistent regardless of starch type and shear history. Amylose/amylopectin ratio and shear history greatly affected the water interactions of products created and the measures of crystallinity within them. Despite this, as

previously reported (Beech *et al.*, 2022), the ultimate densities of the products were largely unaffected by these factors, particularly when popping within the optimum water content region. As such, water content during popping appears to be the most important factor determining sample aeration/density. Further study would be of merit to follow the molecular changes of the starches as they are processed by the different expansion techniques. This would allow for a deeper understanding of the macromolecular assembly, microstructural arrangements and changes to the glass transition temperatures found in such samples and how these contribute to the physical properties observed.

Acknowledgments

This work was supported by the EPSRC [grant number EP/R512321/1]; Real World Business Solutions Ltd and Biopolymer Solutions Ltd. The authors would like to thank Val Street, Steven Johnson and Darrell Cobon of University of Nottingham for their assistance with extruder operation.

Author contributions

Daniel Beech: Data curation (lead); investigation (lead); methodology (equal); visualization (equal); writing – original draft (lead); writing – review and editing (equal). **John Beech:** Conceptualization (equal); methodology (equal); supervision (equal). **Joanne Gould:** Funding acquisition (equal); project administration (lead); resources (lead); supervision (equal). **Sandra Hill:** Conceptualization (equal); funding acquisition (equal); methodology (equal); supervision (equal); visualization (equal); writing – review and editing (equal).

Conflict of interest

The authors declare no conflict of interest.

Ethical approval

Ethics approval was not required for this research.

Peer review

The peer review history for this article is available at <https://publons.com/publon/10.1111/ijfs.16092>.

Data availability statement

All data supporting this study are openly available from the University of Nottingham data repository (Beech, 2022) at <http://doi.org/10.17639/nott.7226>.

References

- AACC (2000). *AACC Approved Methods of Analysis, 10th Ed. Method 10-05.01. Guidelines for Measurement of Volume by Rapeseed Displacement*. St. Paul, MN: AACC. The rapeseed displacement method was used in the determination of sample densities. This was necessary in order to demonstrate the extent of expansion which occurred in the different sample types. Whilst this technique is now considered somewhat archaic, the small size and fragility of the samples precluded the use of more modern techniques such as laser scanning.
- Al-Muhtaseb, A.H., McMinn, W.A.M. & Magee, T.R.A. (2004). Water sorption isotherms of starch powders: part 1: mathematical description of experimental data. *Journal of Food Engineering*, **61**, 297–307.
- Anderson, R., Conway, H. & Peplinski, A. (1970). Gelatinization of corn grits by roll cooking, extrusion cooking and steaming. *Starch-Stärke*, **22**, 130–135. Water absorption and solubility indices were determined using a modified version of the method described in this article. These measurements are used to assess the relative extent of starch conversion which has occurred in the various different samples.
- Aviara, N.A. (2020). Moisture sorption isotherms and isotherm model performance evaluation for food and agricultural products. In: *Sorption in 2020s* (edited by G. Kyzas & N. Lazaridis). Pp. 143–175. London: IntechOpen.
- Ayed, C., Bramante, F., Nwaiwu, O., MacNaughtan, W., Bakalis, S. & Foster, T.J. (2020). Water penetration into mixed and un-mixed carbohydrate powders. *Carbohydrate Polymer Technologies and Applications*, **1**, 100007. This publication was useful in describing and contextualising the unique surface water interactions which take place in pregelatinised waxy starches and ultimately lead to uneven hydration. This helped to explain why only the extruded waxy maize starch formed a rubbery block when hydrated higher water contents whilst no other samples used in the current study exhibited this behaviour.
- Beech, D. (2022). Effect of popping water content and amylose/amylopectin ratio on the physical properties of expanded starch products with different shear histories – data; University of Nottingham data repository. <https://doi.org/10.17639/nott.7226>
- Beech, D., Beech, J., Gould, J. & Hill, S. (2022). Effect of amylose/amylopectin ratio and extent of processing on the physical properties of expanded maize starches. *International Journal of Food Science & Technology*, **57**, 2298–2309. The present study is a follow up from the work reported in this paper by the same authors. Many of the ideas discussed are intended to directly build upon those presented in the previous work.
- Bertoft, E. (2017). Understanding starch structure: recent progress. *Agronomy*, **7**, 56.
- Boischot, C., Moraru, C. & Kokini, J. (2003). Factors that influence the microwave expansion of glassy amylopectin extrudates. *Cereal Chemistry*, **80**, 56–61.
- Cai, C., Zhao, L., Huang, J., Chen, Y. & Wei, C. (2014). Morphology, structure and gelatinization properties of heterogeneous starch granules from high-amylose maize. *Carbohydrate Polymers*, **102**, 606–614.
- Campbell, G.M. & Mougeot, E. (1999). Creation and characterisation of aerated food products. *Trends in Food Science & Technology*, **10**, 283–296.
- Delcour, J.A. & Hosney, R.C. (2010). *Principles of Cereal Science and Technology*, 3rd edn. St. Paul, MN: AACC International.
- Devi, A., Fibrianto, K., Torley, P. & Bhandari, B. (2009). Physical properties of cryomilled rice starch. *Journal of Cereal Science*, **49**, 278–284.
- Kraus, S., Solyom, K., Schuchmann, H.P. & Gaukel, V. (2013). Drying kinetics and expansion of non-predried extruded starch-based pellets during microwave vacuum processing. *Journal of Food Process Engineering*, **36**, 763–773.
- Liu, P., Yu, L., Wang, X., Li, D., Chen, L. & Li, X. (2010). Glass transition temperature of starches with different amylose/amylopectin ratios. *Journal of Cereal Science*, **51**, 388–391.
- Liu, Y., Chen, J., Luo, S. *et al.* (2017). Physicochemical and structural properties of pregelatinized starch prepared by improved extrusion cooking technology. *Carbohydrate Polymers*, **175**, 265–272.
- Mitchell, J.R. & Hill, S.E. (2021). Starch. In: *Handbook of Hydrocolloids* (edited by G.O. Phillips & P.A. Williams). Pp. 239–271, 3rd edn. Duxford: Woodhead Publishing.
- Orts, W., Glenn, G., Nobes, G. & Wood, D. (2000). Wheat starch effects on the textural characteristics of puffed brown rice cakes. *Cereal Chemistry*, **77**, 18–23. Orts and coworkers used a similar processing method to that used in the present study on different raw materials. They made use of what they termed a “rice cake puffing machine” to create expanded cakes from brown rice and wheat starch. Their similar observations of blowout at higher water contents support our findings and demonstrate their applicability to similar materials of different botanical origin.
- Ottenhof, M.-A. & Farhat, I.A. (2004). Starch retrogradation. *Biotechnology and Genetic Engineering Reviews*, **21**, 215–228.
- Panak Balentić, J., Babić, J., Jozinović, A. *et al.* (2018). Production of third-generation snacks. *The Croatian Journal of Food Science and Technology*, **10**, 98–105.
- Sjöqvist, M. & Gatenholm, P. (2007). Effect of water content in potato amylopectin starch on microwave foaming process. *Journal of Polymers and the Environment*, **15**, 43–50.
- Trommsdorff, U. & Tomka, I. (1995). Structure of amorphous starch. I. An atomistic model and X-ray scattering study. *Macromolecules*, **28**, 6128–6137.
- Van der Sman, R. & Bows, J. (2017). Critical factors in microwave expansion of starchy snacks. *Journal of Food Engineering*, **211**, 69–84.
- Van der Sman, R. & Meinders, M. (2011). Prediction of the state diagram of starch water mixtures using the Flory–Huggins free volume theory. *Soft Matter*, **7**, 429–442.
- Vermeylen, R., Derycke, V., Delcour, J.A., Goderis, B., Reynaers, H. & Koch, M.H. (2006). Structural transformations during gelatinization of starches in limited water: combined wide-angle X-ray scattering study. *Biomacromolecules*, **7**, 1231–1238.
- Waigh, T.A., Perry, P., Riekel, C., Gidley, M.J. & Donald, A.M. (1998). Chiral side-chain liquid-crystalline polymeric properties of starch. *Macromolecules*, **31**, 7980–7984.
- Waigh, T.A., Gidley, M.J., Komanshek, B.U. & Donald, A.M. (2000a). The phase transformations in starch during gelatinisation: a liquid crystalline approach. *Carbohydrate Research*, **328**, 165–176.
- Waigh, T.A., Kato, K.L., Donald, A.M., Gidley, M.J., Clarke, C.J. & Riekel, C. (2000b). Side-chain liquid-crystalline model for starch. *Starch-Stärke*, **52**, 450–460.
- Wang, T.L., Bogracheva, T.Y. & Hedley, C.L. (1998). Starch: as simple as a, B, C? *Journal of Experimental Botany*, **49**, 481–502.
- Ye, J., Hu, X., Luo, S. *et al.* (2018). Properties of starch after extrusion: a review. *Starch-Stärke*, **70**, 1700110.
- York, P. (1981). Analysis of moisture sorption hysteresis in hard gelatin capsules, maize starch, and maize starch: drug powder mixtures. *Journal of Pharmacy and Pharmacology*, **33**, 269–273.
- Zeleznač, K. & Hosney, R. (1987). The glass transition in starch. *Cereal Chemistry*, **64**, 121–124.
- Zubair, M., Ferrari, R., Alagha, O. *et al.* (2020). Microwave foaming of materials: an emerging field. *Polymers*, **12**, 2477.