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Incorporation of sweetener and residue of overripe banana in chocolate cookies: Study on physical, textural, and microstructural characteristics

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Abstract

Due to the rising prevalence of type 2 diabetes mellitus, the demand for dietary fibre-enriched and low sugar bakery products is increasing rapidly. The dark brown appearance of overripe banana has been found to make them one of the most wasted products. However, overripe banana contains plentiful dietary fibre and natural sweetener which could potentially be utilised as a new ingredient in baked goods. Therefore, the present work’s objective was to assess how overripe banana sweetener (OBS) and its residue (OBR) affected the physical, textural, and microstructural characteristics of chocolate cookies. The results showed that the incorporation of both OBS and OBR influenced the starch granule integrity by minimising the size, and restricting starch gelatinisation. These mitigated starch digestion, and improved glycaemic response. Overripe banana could have great potential to be utilised as a functional ingredient to develop highly nutritious cookies without compromising its physical properties.

Introduction

Chronic disease incidences are rapidly increasing, and regarded as a critical health issue globally. Diabetes mellitus (DM), one of the chronic illnesses, is listed as a global public health problem, and estimated to have caused 1.6 million deaths globally in 2016 (WHO, 2016). Based on reports, Malaysia has the greatest prevalence of DM among all the ASEAN member states (Chan, 2015). The latest National Health and Morbidity Survey (NHMS, 2019) reported that 3.9 million (18.3%) Malaysian adults are suffering from DM, which was higher than the 3.5 million reported in 2015. The increasing trend of DM has attracted people’s attention towards diabetes-related functional food to improve their blood glucose control.

Banana (*Musa* sp.) is a popular fruit traded internationally and consumed globally (Castelo-Branco *et al.*, 2017). By having significant positive impacts on health, such as reducing the risk of high blood pressure and stroke, promoting regular bowel movements, maintaining blood sugar levels, and aiding in weight reduction, banana has played a significant role in human nutrition (Sidhu and Zafar, 2018). Nevertheless, banana is highly perishable, which subjects it to rapid deterioration after harvest (Karim *et al.*, 2018). According to Symmank *et al.* (2018), the key factor lowering customers' purchase intention is the features of overripe banana, such as the dark brown colour and the squishy consistency of the pulp. Consequently, banana is categorised as an ingredient that is often wasted since many consumers relate the ripeness of a banana with the yellow colour. Despite this, overripe banana could provide a great source of dietary fibre (DF), natural sweetener, vitamins, and minerals (Kumar *et al.*, 2012). According to Yap *et al*. (2017), an overripe banana's sugar content increased dramatically from 1.26 to 12.28% from an unripe banana. Chaipai *et al*. (2018) found that the DF concentration in the banana pulp did not vary with maturity, despite the fact that the bulk of the starch converts to sugar when the banana ripens. At the moment, overripe bananas are used to create films for applications in food packaging, such as biodegradable and sustainable starch laminates by

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Alanís-López *et al*. (2011), and purée-processed films by Martelli *et al*. (2013). Nevertheless, the initiative to explore the practicality of banana in food has not been exhausted, and the benefit of overripe banana is not fully maximised. Therefore, exploitation of overripe bananas can pave a new way in developing value-added food products with an approach focused on recycling agricultural wastes.

Currently, bakery products that included functional ingredients have gained in popularity due to their ability to provide health benefits beyond basic nutritional functions. Banana pulp powder has been used as a DF source in noodles and wheat-based foods, according to earlier studies (Ritthiruangdej *et al*., 2011; Loong and Wong, 2018). There is, however, little research on the possible effects of overripe bananas in baked goods like cookies. Hence, it is critical to explore the potential of overripe bananas to be used in the food industry.

Cookies are small, flat, and sweet bakery items that are very popular because they are ready to eat, handy, have a long shelf life, inexpensive, and readily available. Cookies are one of the most popular baked goods that are consumed daily by Malaysians (Kasim *et al.*, 2018). Hence, cookies could be considered to be better vehicles for fortification. However, cookies in the market not only lack in DF content, but are also high in fat and sugar contents. These attributes makes them unhealthy choices for daily consumption. Recently, Ng *et al*. (2020b) used overripe bananas to create high-DF, low-GI chocolate cookies, focusing on how the incorporation of overripe banana sweetener (OBS) and overripe banana residue (OBR) affected the nutritional content and blood glucose level. The OBS is the sugar taken from the overripe banana pulp, whereas the OBR is the leftover banana pulp after the sugar has been removed from it. The OBS was used to partially replace sugar in order to lower the amount of sugar in the chocolate cookies, while the OBR, which has greater fibre content than wheat flour, was used to partially replace wheat flour. The findings demonstrated that the incorporation of OBS and OBR had a substantial impact on the nutritional properties of the chocolate cookies, particularly their sugar content (25.12 - 17.67%), DF content (3.18 - 7.77%), and glycaemic index value (63 - 50). As a result, an overripe banana could be a source of DF and a natural sweetener that improve the chocolate cookies' nutritional value and glycaemic reaction. However, a number of variables might be involved in the variations in blood glucose response. One of the factors mentioned earlier is the food structure, which could be affected during food processing, which could, in turn, interfere with digestion or adsorption mechanisms, thus influencing the postprandial blood glucose level (Hodges *et al*., 2019; Cai *et al*., 2021). In light of this, the present work aimed to determine the influence of various incorporation levels of OBR and OBS on the primary characteristics of cookies, including textural, physical, and microstructural characteristics.

Materials and methods

Processing of raw material

A stage 4 fully matured *Musa acuminata* cv. *Berangan* banana was purchased in Kota Bharu, Kelantan, Malaysia, from a neighbourhood fruit stand. The sample was kept at room temperature (25°C), with a relative humidity of 80 to 85%, until the fruit spontaneously over-ripened (stage 5) without the application of a ripening agent. The maturity stage of a banana is assessed based on physical observation and a colour chart (Karim *et al.*, 2018). Based on a modified version of the method proposed by Albuquerque *et al*. (2005), liquid sugar was extracted from bananas. The banana pulps were mixed with water at a 1:3 ratio, homogenised, and then centrifuged at 15,000 *g* for 25 min at 4°C. Whatman No. 4 filter paper was used to separate the non-fibrous portions and the liquefied banana pulp. Both the extracted supernatant and the remaining residue were then further processed into OBS and OBR, respectively.

Preparation of OBS

The method established by Tadakittisarn *et al.* (2007) was modified slightly in order to dehydrate the extracted supernatant. By drying 50 g of banana liquid at 60°C for 16 h in a thermal dehydrator (Anywin FD770, China), the moisture content was removed. Before analysis, the concentrated syrup, known as OBS, was kept in a screw cap bottle at 4°C.

Preparation of OBR

Following the method used by Wijewardana *et al*. (2016), the residue (w/v) obtained after the filtering process was dried in a conventional oven (Memmert, Germany) at 55°C for 24 h. It was then ground into powder using an electronic blender, which was afterwards sieved to create fine powder (125 m diameter). The residue powder obtained, known as OBR, was stored in a screw cap bottle at 4°C.

Development of chocolate cookies

The chocolate cookie recipe and technique of preparation were modified slightly from Mohan *et al*. (2018). The ingredients for the chocolate cookies, including baking powder, butter, castor sugar, cocoa powder, corn flour, eggs, margarine, and wheat flour, were all obtained commercially. Ng *et al*. (2020a) deemed the incorporation of 8% overripe banana pulp powder to replace wheat flour to be the optimum formulation in terms of sensory qualities. Since OBR and overripe banana pulp powder have the same chemical makeup, a chocolate cookie with 8% OBR was used in the present work. As shown in Table 1, OBS was used to partially replace castor sugar at the ratios of 10, 15, and 20%.

A small digital scale was used to accurately weigh each component (OHAUS Scout, USA). The first stage used an electric hand mixer (Khind HM200, Malaysia) to mix the butter, castor sugar, and egg in a mixing bowl. The mixture was then continually mixed for 4 min to get a frothy cream consistency (light colour and smooth surface). The mixture was beaten again for 5 min after all the dry ingredients had been added, and it was then refrigerated for 2 h. A 5 cm-diameter mould was used to form the chilled dough into a 3 mm-thick disc. The cookies were then placed on a baking sheet, and cooked for 12 min at 170°C. The cookies were then allowed to cool at room temperature for another hour before being pulverised into powder, and kept at 4°C until further analyses.

Table 1. Substitution levels of OBS and OBR in chocolate cookies.

Ingredient	**Amount (g)**				
	Control (0% OBR and OBS)	**8% OBR + 0% OBS**	**8% OBR + 10% OBS**	**8% OBR + 15% OBS**	**8% OBR + 20% OBS**
OBS	0	0	4.1	6.2	8.2
Castor sugar	41	41	36.9	34.8	32.8
OBR	0	6.1	6.1	6.1	6.1
Wheat flour	76	69.9	69.9	69.9	69.9

OBS: overripe banana sweetener; and OBR: overripe banana residue.

Physical properties

The physical properties of the cookies were examined using a 15 cm scale. After an hour of baking, the mean value (g) was determined by weighing ten cookies from each formulation using an analytical balance (Mettler-Toledo Dragon 204, Switzerland). By aligning ten cookies edge to edge, their diameter (D) was measured. Ten pieces of cookies were stacked to determine the cookies' thickness (T). The diameter-to-thickness ratio was used to measure the spread ratio (D/T).

Texture profile analysis

The crispiness and firmness of the cookies were assessed using the three-point break technique (Gains, 1991). The Texture Analyser TA.XTplus (Stable Micro Systems, Surrey, UK) was equipped with the Texture Exponent Software package, HDP/90 heavy-duty platform, and HDP/3PB 3-point bend rig that were required for the test. The cookie was supported by two adjustable rig base supports which were separated 20 mm apart, and maintained throughout the test for comparison purposes. Next, the cookie sample was put centrally on the base plate of the heavy-duty platform.

Pre-test speed was set to 1.0 mm/s, post-test speed was set to 10.0 mm/s, test speed was set to 3.0 mm/s, compression distance was set to 3 mm, the distance between probe and cookie was set to 10 mm, trigger force was set to 50 g, and the (return to start) option was selected. After obtaining the trigger force, the force was steadily increased until the cookie was snapped in two. Firmness refers to the greatest or peak force that was needed to fracture the cookie. The mean distance compacted prior to the breaking value was considered as "crispiness." The program then produced a curve and the values of interest.

Microstructure characteristics

A FEI ESEM Quanta 450 FEG scanning electron microscope was used to study the cookie's morphological properties (FELMI-ZFE, Austria). Samples were cut into small pieces, and placed on aluminium stubs with carbon conductive doublesided adhesive tape to improve conductivity followed by gold sputtering coating in a sputter coater device (Baltex SCD005, Hi-Tech Germany). All the samples were viewed at 5 kV at an electron beam-accelerating voltage with magnifications of 500× and 1,000×.

Statistical analysis

Based on the significance threshold of $p < 0.05$, the study findings of physical characteristics and texture patterns were compared using a One-way analysis of variance (ANOVA) and Tukey's *post hoc* test to identify the significant mean differences of the samples. SPSS Version 24.0 was used for all statistical analyses (SPSS Inc., Chicago, Illinois). For all evaluations, information was gathered from three batches of OBS, OBR, and chocolate cookies. The results were then presented as mean ± standard deviation (SD).

Results and discussion

Physical properties

Table 2 shows the physical properties and texture profile of chocolate cookies incorporated with OBR and OBS. Physical characteristics showed a modest reduction in weight (6.40 to 6.34 g), diameter (50.43 to 50.28 mm), and spread ratio (16.81 to 16.76) when 8% OBR was incorporated, but no appreciable ($p > 0.05$) variations from the control group of chocolate cookies. However, there were no differences in thickness between the control and the 8% OBR-chocolate cookies (3 mm). This agreed with Ajila *et al.* (2008) and Nassar *et al.* (2008) for cookies incorporated with mango and citrus peel powder, respectively. The OBR's high pectin content, which has a potent capacity to bind water and produce extra hydrogen bonds, has previously been shown by Ng *et al*. (2020b) to result in higher water absorption (Rosell *et al*., 2001). As a result, pectin's competition with gluten for moisture or its interactions with gluten prevents gluten from being hydrated (Quiles *et al*., 2018). Therefore, the diluting of the gluten proteins, which prevented the proteins from forming a continuous and strong network, was most likely the cause of the decrease in the diameter, weight, and spread ratio of the cookies when OBR was incorporated. This agreed with Zaker *et al*. (2016) who proved that cookies incorporated with high fibre ingredients did not show a significant reduction in physical properties when the incorporation levels ranged from 0 - 10%. However, significant results were obtained when the incorporation levels exceeded 10%.

Table 2. Effect of OBS and OBR incorporation on physical properties and texture profiles of chocolate cookies.

Parameter	Control (0% OBR and OBS)	8% OBR + 0% OBS	8% OBR + 10% OBS	8% OBR + 15% OBS	8% OBR + 20% OBS
Weight (W, g)	6.40 ± 0.04^{a}	6.34 ± 0.03^{a}	6.36 ± 0.03^{a}	6.38 ± 0.02^{a}	6.42 ± 0.04^{a}
Diameter (D, mm)	50.43 ± 0.02^{a}	50.28 ± 0.02^{a}	50.35 ± 0.05^{a}	50.41 ± 0.03^{a}	50.46 ± 0.04^{a}
Thickness (T, mm)	3.00 ± 0.00^{a}	3.00 ± 0.00^{a}	2.96 ± 0.01^{a}	2.94 ± 0.01^{a}	2.90 ± 0.01^{a}
Spread ratio (D/T)	16.81 ± 0.07^{c}	16.76 ± 0.01^{c}	17.01 ± 0.02^{b}	17.15 ± 0.01^{b}	17.40 ± 0.03^{a}
Firmness (kg)	1.05 ± 0.03^{c}	1.23 ± 0.04^{a}	1.19 ± 0.05^{a}	1.17 ± 0.02^{ab}	1.14 ± 0.03^{b}
Crispiness (mm)	0.38 ± 0.04^{a}	0.35 ± 0.02^{a}	0.34 ± 0.03^{a}	0.33 ± 0.05^{a}	0.31 ± 0.02^{a}

Mean ± SD values in similar row with different lowercase superscripts differ significantly ($p < 0.05$). OBS: overripe banana sweetener; and OBR: overripe banana residue.

The weight and diameter of the chocolate cookies increased when OBS was incorporated at percentages of 10, 15, and 20%, but there was no appreciable variation ($p > 0.05$) from the 0% OBS-chocolate cookies. In comparison to chocolate cookies with 0% OBS, there was a significant ($p < 0.05$) increase in the spread ratio of cookies with increasing levels of OBS (17.01 to 17.40), but no significant difference was found for cookies with 10 and 15% OBS. Contrarily, the thickness of the chocolate cookies somewhat decreased with the incorporation of OBS (2.96 to 2.90 mm), although there were no appreciable changes ($p > 0.05$) from the thickness of the chocolate cookies without OBS. This agreed with Handa *et al*. (2012), who found that the fructooligosaccharide-sweetened cookie had an increased diameter and spread ratio, as well as a slightly decreased thickness. The crystallised sucrose does not fully dissolve during dough mixing, so the undissolved sugars will dissolve during baking, causing the cookie to spread (Handa *et al.,* 2012). Cookies incorporated with OBS were shown to be larger in spread ratio than cookies without OBS, mainly due to the higher solubility of OBS as compared to sucrose. The trend could have been due to the proportion of sucrose (50.39%), fructose (16.33%), and glucose (13.65%) in OBS (Ng *et al*., 2020b). Several studies have claimed that a sweetener with higher solubility as compared to sucrose is likely to maintain its dissolved nature longer during baking, which would facilitate the flow in dough mixing, and result in a more homogenous distribution of the dough, causing an increase in spread ratio (Handa *et al*., 2012; Rodriguez-Garcia *et al*., 2022). The spread of the cookies is influenced by the rate of dough flow, which is associated with the viscosity of the dough (Panghal *et al*., 2018). The high-water absorption of OBS could attract more water, thus causing a decrease in dough viscosity, and leading to an increase in the spread ratio of chocolate cookies. In addition, the increment of the spread ratio of the cookie prepared with liquid sugar compared to crystal sugar is associated with lateral expansion due to the dissolution of sugar increasing the solvent volume in the dough, which contributes to cookie spreading (van der Sman and Renzetti, 2019; Niroula *et al*., 2020). A higher spread ratio of cookies developed with different types of syrups in comparison to sucrose has been published by Majzoobi *et al*. (2016). Furthermore, OBS has a higher affinity for binding water in comparison to sucrose, thereby limiting the water for gluten development, and causing poor gas retention, which results in the decreased thickness of the cookies. In general, the thickness of the cookie results from gluten development which leads to expansion during baking. However, the amount and types of sugar in the formulation can also affect the thickness of the end product. This is because sugar preferentially attracts water over gluten protein, takes up more water, and inhibits gluten development, leading to a decrease in cookie thickness (Ho and Pulsawat, 2020). Kweon *et al*. (2014) claimed that while concentrated aqueous sugar solutions could function as anti-plasticisers, sugars are plasticisers of the biopolymer of wheat flour. Thus, starch gelatinisation during baking and the production of gluten during dough mixing are avoided.

Texture profile

Crispiness and firmness are textural characteristics of significant interest in the assessment of baked goods due to their association with freshness (Pereira *et al.,* 2013). Table 2 shows the effect of OBR and OBS incorporation on the texture profile of chocolate cookies. The change in crispiness, which varied from 0.35 to 0.38 mm, was not statistically significant ($p > 0.05$). When grape skin and seed flours were included in the formulation of cookies, comparable trends in the findings were seen (Kuchtová *et al*., 2018). OBR was also proven to increase the hardness of the chocolate cookies, going from 1.05 to 1.23 kg. In comparison to the control chocolate cookies, it was considerably ($p < 0.05$) higher at 8% more for the OBR-chocolate cookies. This agreed with Varastegani *et al*. (2015), who noted that the hardness of the cookie was enhanced by the addition of papaya pulp flour as a DF source. The large decrease in moisture content (2.58 to 2.44%) due to the increase in DF content (3.18 to 7.77%) in the cookie when the OBR was added may be the cause of the chocolate cookie's increased firmness (Ng *et al*., 2020b). The matrix became harder by reacting with the gelatinised starch. As a result, it became stiffer, rendering it less available for dough expansion during baking. The DF component has a high capacity for water absorption. As a result, cookies had more compact structure and greater degrees of hardness.

Both firmness and crispiness of the chocolate cookies were found to decrease with increasing levels of OBS. The firmness of chocolate cookies decreased from 1.19 to 1.14 kg, significantly at 20% OBS in comparison with the 0% OBS-chocolate cookies. However, no significant ($p > 0.05$) differences between 0 and 10% OBS-chocolate cookies were observed, as well as the 15 and 20% OBS-chocolate cookies. Despite a decrease in crispiness from 0.34 to 0.31 in chocolate cookies, no significant differences ($p > 0.05$) were found amongst the formulations that had different OBS levels. The results specified a lower snapping characteristic, contributing to a softer cookie texture which agreed with Ayyappan *et al.* (2016), who used oxylooligosaccharides to replace sugar (5 - 15%) in cookies, which demonstrated a decrease in both hardness and fracturability as compared to control cookies. A similar result for the effect of sucrose replacers has also been reported by Majzoobi *et al*. (2016), who used date syrup in cookie. As sucrose was replaced by OBS, lesser force was needed to break the cookies which can be attributed to their higher spread ratio. Furthermore, cookie firming involves the recrystallisation of sucrose, leading to a firmer and drier texture (Handa *et al.*, 2012). This agreed with the use of high fructose corn syrup (Zargaraan *et al.*, 2016) and raffinose (Belcourt and Labuza, 2007) which effectively prevented the recrystallisation of sucrose. When sucrose is replaced with sweeteners with higher solubility, their anti-plasticising effect inhibits the development of gluten and starch gelatinisation, causing a decrease in firmness and crispiness of the cookie (Ho and Pulsawat, 2020). In the present work, OBS was highly soluble, which explains why it did not recrystallise as it bounded more water, thus leading to softer cookies.

Microstructure characteristics

Figure 1 shows the microstructure characteristics of four different formulations chocolate cookies incorporated with OBR and OBS using SEM with visual magnifications of 500× and 1,000×.

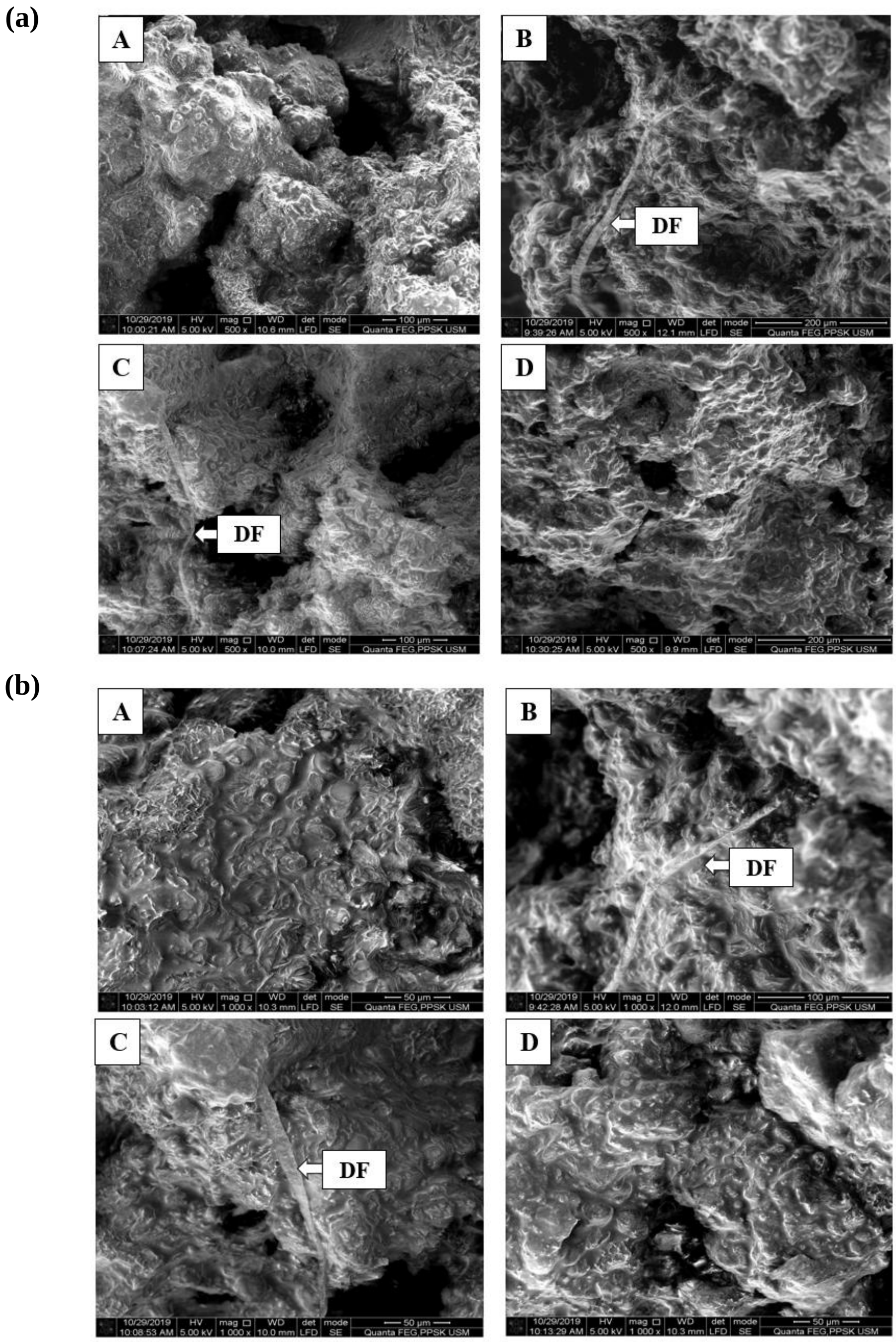

Figure 1. Scanning electron micrograph of chocolate cookies at **(a)** 500×, and **(b)** 1,000× magnifications. **(A)** control; **(B)** 8% OBR + 0% OBS; **(C)** 8% OBR + 10% OBS; and **(D)** 8% OBR + 20% OBS. OBS: overripe banana sweetener; and OBR: overripe banana residue.

It was observed that the gluten proteins of the control cookie (Figure 1a-A) produced a smooth and incessant network where the starch granules were implanted in the protein matrix, and almost entirely gelatinised. Additionally, an open structure with greater-sized starch granules was identified in the control cookie, which was linked to the entrapment and enlargement of gas bubbles throughout the baking process which potentially enlarged the surface area. This led to increased susceptibility to enzyme activities. A large number of partially gelatinised starch granules were evidently observable for cookies incorporated with 8% OBR (Figure 1a-B). The gluten matric was nearly combined, and partly disturbed by starch granules and the existence of DF, signifying a weak established gluten network that was attributed to the limited amount of water in the formulation. Furthermore, it could be observed that the structure of starch granules was disturbed by the DF component from OBR embedded in the cookies, causing irregular shape and a rough granule surface. A similar observation was reported by Indrani *et al.* (2015), who explored the impact of DF on the internal structure of bread. The addition of DF-rich OBR trapped the starch granules within the fibre matrix, leading to a more compact structure. Thus, the DF lessened the starch granule's integrity retention by interrupting the structure, minimising the size of the starch granules, and increasing the vulnerability of enzymatic hydrolysis (Wu *et al.,* 2014). DF has also demonstrated an increased water absorption capacity which especially absorbs water when there is a water deficiency, ensuring that more starch granule surfaces are guarded from water penetration, which restricts starch gelatinisation (Schuchardt *et al.,* 2016). Correspondingly, Sivam *et al.* (2013) revealed that pectin-fortified bread incorporated with 20% extra water contributed to a larger extent of starch gelatinisation and a more even microstructure than control pectin-fortified bread.

The size of starch granules decreased when OBS was incorporated into the cookies (Figures 1b-C and 1c-D), as compared to the control and OBR-cookies. The increase in the continuous phase in the cookies can also be observed as the levels of OBS increased. A similar result was reported on the effect of sugar replacers in muffins (Jingrong *et al*., 2018) and burger buns (Sahin *et al.,* 2018). It has been shown that different sugars can delay starch gelatinisation. Sugar is able to reduce the penetration rate of water by mixing with water molecules, causing the water to be less accessible for starch (Alamri *et al.,* 2016). Sun *et al*. (2014) described that the growing concentration of fructose syrup in starch promoted the inception of gelatinisation, and moved it to a higher temperature, thus producing more thermal energy required for starch granule swelling and gelatinisation. The influence of syrups on starch physicochemical properties is caused by the penetration of small sugars into the amorphous sections of starch, which prompts intricate reactions between sugars and starch components, which, in turn, contribute to the increase in the gelatinisation temperature. This will contribute to an interruption in amylose leaching, starch swelling, and hinder the start of starch gelatinisation. Mohamed *et al.* (2019) reinforced this finding as they examined the active rheological properties of date syrups in corn starch. This is attributable to the probable connection between OBS with amylose that contributes to a frailer network, and stretches phase separation.

The incorporation of OBR and OBS into the chocolate cookies was shown to disturb the structure and reduce the size of starch granules, as well as limit starch gelatinisation. All of these resulted in a reduction of the surface area of starch granules exposed to enzyme activity. Therefore, minimised starch digestion contributed to a slower release of glucose from the starch which will directly improve glycaemic responses. The findings inferred that chocolate cookies formulated with OBR and OBS were more resilient than the controlled chocolate cookie, which can be associated with the positive enhancement of glycaemic responses reported by Ng *et al.* (2020b).

Conclusion

The OBR and OBS were shown to have the ability to reduce gluten development and starch gelatinisation, causing an alteration in physical properties such as thickness and spread ratio. Meanwhile, the firmness and crispiness of the chocolate cookies decreased with increasing levels of OBS due to the higher solubility of OBS as compared to sucrose. The microstructure of the chocolate cookies was found to be affected by the incorporation of both OBS and OBR by interrupting the morphology of the starch granules, and restricting the gelatinisation of starch. This contributed to reduced starch digestion which minimised the level of postprandial blood glucose. In summary, the present work proved that the use of overripe bananas created a new and alternative approach in developing highly nutritious and diabetic-friendly chocolate cookies, without affecting the physical quality much. This would provide knowledge for further studies in developing various overripe banana-based food products that can make overripe banana a high-value food ingredient.

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