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# Analysis of the effect of recent reformulation strategies on the crystallization behaviour of cocoa butter and the structural properties of chocolate

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ABSTRACT

Chocolate is a complex soft material characterized by solid particles (cocoa powder, milk solid particles and sugar crystals) dispersed in a crystallized fat matrix mostly composed of cocoa butter (CB). Important chocolate properties such as snap, and visual appearance are strongly dependent on the internal molecular arrangement (polymorph), size and shape, as well as the spatial distribution of CB crystals within the chocolate mix.

In recent years confectionary companies have put increasing effort in developing novel chocolate recipes to improve the nutritional profile of chocolate products (e.g., by reducing the amount of high saturated fat and sugar content) and to counteract the increasing price of cocoa butter as well as sustainability issues related to some chocolate ingredients. Different reformulation strategies can dramatically affect the crystallization thermodynamic and kinetic behaviour of cocoa butter; therefore, affecting the structural and sensorial properties of chocolate.

In this review we analyse how different reformulation strategies affect the crystallization behaviour of cocoa butter and, hence, the structural and sensorial properties of chocolate. In particular, this work discusses the effect of: (1) CB replacement with emulsions, hydrogels, oleogels and oleofoams; (2) CB dilution with limonene or cocoa butter equivalents; (3) replacement or reduction of the amount of sugar and milk in chocolate. We found that there is certainly potential for successful novel alternative chocolate products with controlled crystalline properties; however, further research is still needed to ensure sensory acceptance and reasonable shelf-life of these novel products.

## 1. Introduction

Chocolate is a popular confectionary product throughout the world and its supply is increasing, with sales in the UK in 2020 estimated to be worth £6.7 billion, a £200 million increase from 2019 (Euromonitor, 2019). Despite its popularity, there are some negative health, economic and environmental issues associated to some of the ingredients that make up this product. The main ingredients of chocolate are cocoa mass, cocoa butter (CB), sugar and, for milk chocolate, milk fat. CB is the main contributor to the high fat content of chocolate; between 30% and 40% of the weight of chocolate is fat (Do et al., 2007) and consuming high amounts of this nutritional group is known to contribute to the overconsumption of calories (National Health Service, 2019). Additionally, CB is particularly high in saturated fat. Table 1 shows the typical values of saturated fat content of dark chocolate products marketed in several countries. The British National Health System, NHS (National Health Service, 2019), recommends cutting back on foods that are high in saturated fat, due to its association with increased blood cholesterol, leading to a higher risk of coronary heart disease.

Together with cocoa butter, another chocolate ingredient that can contribute to health problems is sugar, whose excess intake has associations with dental problems and overconsumption of calories (SACN, 2015).

There are also economic reasons to reformulate chocolate products. Due to the recent increase in demand for these items, the cost of cocoa powder and CB has become higher: in 2019 alone, the price of CB increased by 4% in the USA and 5% in Europe (International Cocoa Organ, 2019). Hence, reducing or replacing CB in chocolate is essential to develop products with improved manufacturing costs and more convenient selling prices.

Environmental concerns are instead mostly related to the low sustainability of milk fat used in milk chocolate. The increasing consumer demand for vegan, dairy-free products is another incentive for the reduction or elimination of this ingredient from chocolate formulations

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**Table 1**
Amounts of saturated fatty acids in dark chocolate marketed in different countries.

| Country | Amount of Saturated Fatty Acids (g/100 g) | Reference |
|---|---|---|
| Australia | 18.2 | Food Standards Australia New Zealand (2019) |
| Denmark | 18.2 | National Food Institute (2019) |
| France | 22.5 | ANSES (2020) |
| New Zealand | 17.4 | New Zealand Institute (2019) |
| UK | 16.8 | Public Health England. Co (2019) |
| USA | 18.5 | U.S. Department Of Agriculture (2019) |

(Veganuary., 2019). All these considerations highlight the need for alternative confectionary products that tackle these problems, whilst being able to provide a high-quality product that can compete with traditional chocolate.

Changing the traditional chocolate formulation to create a new product can lead to manufacturing challenges, including those related to changes in the kinetics and thermodynamic of CB and fats crystallization from the chocolate melt. Controlling this unit operation is critical as the crystal structure of CB dictates the sensory properties and shelf life of chocolate (Dimick et al., 1999). CB can crystallize in six polymorphs with form V ($\beta_2$) being the ideal structure, as it contributes to the glossy appearance of chocolate, its melting in the mouth and the snap when broken (Beckett, 2008). Because of the complex physical and chemical interactions between CB and other chocolate ingredients, even small changes in product formulation can lead to significant modification to the crystallization behaviour of cocoa butter and hence the final properties of chocolate. For this reason, this review will focus on the effects that recent chocolate reformulation strategies were found to have on the crystallization of the fat phase in chocolate.

Different strategies have been considered to tackle the problems created by the traditional chocolate formulation. Reducing the content of CB in chocolate can reduce the total amount of saturated fatty acids and provide a health and economic benefit. Nevertheless, simply reducing CB content increases the hardness of the chocolate (Ashrafie et al., 2014) and this should be counteracted by the addition of other ingredients to dilute CB and maintain suitable mechanical properties (Do et al., 2008). CB can also be partially replaced with mixtures of similar triglycerides (cocoa butter alternatives) or even different edible soft matter systems including emulsions, hydrogels, oleogels and oleofoams, as well polysaccharides and proteins. (Ashrafie et al., 2014; Rezende et al., 2015; Heymans et al., 2017; Francis and Chidambaram, 2019; Li and Liu, 2019; Li and Liu, 2019; Jin et al., 2019a).

Alternative ingredients to replace sucrose are mostly polysaccharides and sugar alcohols, which have less calories and equal sweetening power to sucrose. Plant based milks such as peanut, cowpea milk have instead been considered to improve the sustainability of milk chocolate products (Aidoo et al., 2012). This review will look at these strategies for creating new alternative chocolate related products with a focus on the effect on chocolate crystallization behaviour and final crystal structure stability.

## 2. Strategies for novel fat reduced chocolate products

Reducing the amount of CB would be the easiest solution for the reduction in saturated fatty acids in chocolate. However, this solution would also increase the viscosity of liquid chocolate and the hardness of solid chocolate, affecting both manufacturability and product quality (Aidoo et al., 2014). Hence, CB is often only partially replaced by different compounds, attempting to reduce the fat content while maintaining the properties needed for an acceptable product. Such changes can affect the crystalline structure of CB as illustrated in the next paragraphs.

### 2.1. Addition of limonene in reduced CB chocolate

Limonene is a cyclic monoterpene that has been considered for use in reduced CB chocolate products since 1999 (Beckett, 1999). The addition of limonene changes the kinetics of CB crystallization as it dilutes the CB, causing fewer CB crystals to form and disrupting the crystal packing. (Do et al., 2008). Such disruption lowers the chemical potential of the CB crystal network, depressing the melting point of the fat mixture (Ray et al., 2012). The lower melting point induced by limonene counteracts the increase in viscosity and hardness caused by the reduction of CB. However, the higher the amount of limonene that is added to chocolate, the more likely and quickly it is to bloom (Juhaida et al., 2019). Fat bloom is due to CB melting and recrystallizing as a more stable polymorph (form VI) at the surface of chocolate products (Juhaida et al., 2019). The appearance of fat bloom reduces the quality of the chocolate so it should be avoided, not accelerated. The dilution of CB with limonene to decrease chocolate viscosity was also found to lead to a quicker polymorph conversion of form V to form VI (Do et al., 2008; Juhaida et al., 2019). Polarised light microscopy of CB and limonene mixtures showed an increased crystallization rate in the early stages of isothermal crystallization compared to pure CB (Ray et al., 2012). A mixture of limonene and CB were also found to crystallise faster at 17 °C than 20 °C due to the higher concentration of α crystals at 17 °C for a quick α-mediated polymorphic transformation to β' (Rigolle et al., 2016).

The amount of limonene in an alternative chocolate product would have to be controlled to keep the conversion of CB crystals from form V to form VI to a minimum, while providing a substantial CB reduction and the desired viscosity. Limonene has also been shown to accelerate the polymorphic conversion of form V to form VI also in some cocoa butter equivalents (Miyasaki et al., 2016). The aforementioned studies do not include any sensory analysis; hence, a better mouthfeel related to decreased solid chocolate hardness has not been confirmed yet. Also, the limonene may impart a lemon flavour to the chocolate and sensory analysis would confirm if it is at an acceptable level for consumers. Currently, limonene holds potential to be used in a novel alternative chocolate product but more investigation looking into the effect of this compound in CB and mixtures of CB and other CB replacing fats on sensory perception is required.

### 2.2. Replacing CB with emulsions, hydrogels and oleogels

The use of emulsions and gels have been considered to create different reduced fat foods and the same strategy could be used to reduce the caloric content of chocolate (Norton et al., 2013; Nguyen et al., 2017). A low-cost solution to reduce CB content is to replace it with water. A water-in-CB emulsion (see Fig. 1) with 40% water (all water droplets below 5 μm diameter size) has been suggested to be comparable with normal milk chocolate (Prosapio and Norton, 2019). The use of margarine processing equipment to incorporate water in molten CB could be easily incorporated into chocolate processing plants, and it has been shown to create form V CB crystals (Sullo et al., 2014). Previous work on water-in-CB emulsions showed that shells of sintered CB crystal formed at the oil/water interface, stabilizing the emulsion (Norton and Fryer, 2012). Different emulsifiers were also added to further stabilize water-in-CB emulsions. Carrageenan was tested but increasing its concentration past 1.5% w/w led to an increase in unstable CB polymorphs formation, particularly form II (Sullo et al., 2014). The emulsifier soybean lecithin caused flocculation of the water droplets, leading to an increase in chocolate viscosity. This is not desired as decreasing the amount of fat itself, increases chocolate viscosity (Sullo et al., 2014). The emulsifier polyglycerol polyricinoleate (PGPR) was found to provide the best rheological properties in liquid chocolate (Prosapio and Norton, 2019). This shows how the use of additives is important when using water as a fat replacer and how it is essential to find one which does not cause unstable CB crystallization.

Francis and Chidambaram (Francis and Chidambaram, 2019) showed

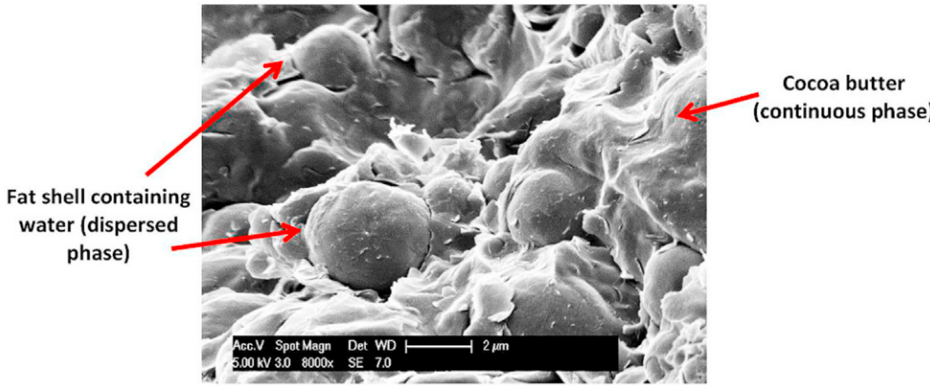


**Fig. 1.** Cryo-Scanning Electron Microscopy image of a water-in-CB emulsion. Adapted from (Prosapio and Norton, 2019).

that an acceptable alternative chocolate product could be formulated with up to 50% v/v CB substitution with a sodium alginate-pectin-citric acid hydrogel. This strategy led to the formation of the desired form V CB polymorph. This hydrogel also allowed the chocolate product to be more resistant to heat. In fact, at 50–100% v/v substitution the final product kept its original structure at temperatures up to 80 °C (Francis and Chidambaram, 2019). Heat resistance is beneficial for chocolate products in hotter climates than the UK and it avoids chocolate melting in the hands of consumers. Too high melting point can, however, have negative consequences on mouth feel and melting processes during manufacturing. Another type of hydrogel that has been considered is an agar hydrogel that can replace CB to up to 80% in volume, greater than the previous two options, while keeping chocolate quality attributes (Skelhon et al., 2013). The agar hydrogel was emulsified into the CB using shear cooling techniques (e.g., ultra-turrex equipment) (Skelhon et al., 2013). Different effects can occur with larger scale processing machinery compared to laboratory equipment; which needs to be considered when scaling up low fat chocolate production. Skelhon et al. (2013) looked at the substitution of CB with hydrogels in dark, milk, and white chocolate in contrast with other studies that tend to look at only one type of chocolate. Additionally, the hydrogel is shown to produce a chocolate with a similar visual appearance to traditional chocolate, with a substantial amount of form V CB crystals, and with no higher tendency to fat blooming than normal chocolate. (Skelhon et al., 2013). Therefore, hydrogels seem to be a promising strategy to reduce the amount of CB for an acceptable novel alternative chocolate product. However, checking the taste and sensory properties of these products is essential to confirm if hydrogels can be used, especially at high substitution levels, and still provide a high-quality product.

Oleogels have also been considered as a fat replacer for CB. Oleogels can entrap liquids and oils by forming a gel using structurants, such as ethyl cellulose, waxes and monoglycerides (Martins et al., 2018). The use of oleogels in chocolate products has been examined due to the potential to form a heat resistant material and partially replace saturated triacylglycerides with unsaturated ones (Martins et al., 2018; Marangoni, 2018). It has been shown that 100% v/v CB substitution with a monoglyceric stearate oleogel can produce a solid dark chocolate with a lower amount of solid fat and saturated fatty acid content (Li and Liu, 2019). Oleogels made from amaranth oil, also showed potential as CB replacers. For both these types of oleogels, stabilization was aided through the formation of needle-like CB crystals in a compact arrangement within the liquid phase (Li and Liu, 2019; Kamali et al., 2019). The oleogel structure also increased the oxidative value of the amaranth oil maintaining its level of essential linoleic fatty acid (Kamali et al., 2019). The intake of amaranth oil has also been suggested to protect consumers against cardiovascular disease (Martirosyan et al., 2007). However, its positive effects may be undone by the high fat and sugar levels in chocolate. These types of gels can still contain a high proportion of fat, so they are more suitable if the goal of the product is only to have a lower saturated fat content than traditional chocolate.

### 2.3. Air bubbles for healthier chocolate

Air incorporation is another strategy that has been used to reduce the calorific content of chocolate products. The addition of air bubbles does not add any nutritional value to food products, but it lowers their calorific density and significantly affects rheology and texture (Campbell and Mougeot, 1999). In confectionary applications, air bubbles can be incorporated in chocolate products by applying vacuum during tempering, followed by cooling, which entraps the bubbles in a solid matrix. Haedelt et al. (2005) reported that decreasing the vacuum pressure caused an increase in gas hold-up, as well as an increase in the mean bubble-section diameter. The setting properties of the chocolate, as modified by the addition of edible emulsifiers and non CB fats to the recipe, were found to affect the gas hold-up and bubble size. Softer chocolate allowed expansion of the air bubbles and coalescence, whereas harder formulations resulted in smaller bubble size and higher gas uptake (Haedelt et al., 2005).

Edible oil foams, also called oleofoams, have been also considered recently for the design of novel chocolate fillings with reduced calorific density and specific textures (Heymans et al., 2017) resulting in several patents related also to their use as fat replacers for cakes, pastry and biscuits (Chisholm et al., 2018; Gunes et al., 2018). Oleofoams are typically obtained by whipping a dispersion of fat crystals in a liquid oil phase. The aeration entrains the air bubbles, which are coated and stabilized by crystals through a Pickering mechanism. The fat crystals also form a network in the continuous phase, providing additional stabilization from oil drainage and bubble coalescence (Fameau and Saint-Jalmes, 2017). Common ingredients for stabilizing oleofoams for chocolate products include fatty acids and fatty alcohols (Binks et al., 2016; Fameau et al., 2015), partial glycerides (Brun et al., 2015; Goibier et al., 2019; Gunes et al., 2017; Heymans et al., 2018; Truong et al., 2019),mixtures of triacyl glycerides (Binks and Marinopoulos, 2017; Mishima et al., 2016) including CB itself (Metilli et al., 2020), and edible waxes (Saha et al., 2020). Oleofoams display high stability to drainage and coalescence, with amount of air incorporated as high as 66% v/v. The total amount and properties of the fat crystals (size, shape, and polymorphism) affect foamability, foam stability and rheological properties (Metilli et al., 2020).

For example, micron sized, needle or platelet-like FHR-B (fully hydrogenated rapeseed rich in behenic acid) crystals enabled rapid incorporation of high amounts of air, compared to larger, aggregated crystals which required longer aeration times to reach the same air volume (Mishima et al., 2016). In some systems, such as co-crystallized monoglyceride-native phytosterol dispersed in sunflower oil, large, aggregated crystals could not be aerated at all (Truong et al., 2019). Dispersions of monoglyceride crystals of α or β polymorph in a sunflower oil phase were able to incorporate comparable amounts of air; whereas low-stability polymorphs such as sub-α did not produce any foam (Heymans et al., 2018). However, it is unclear whether the latter might have melted during the whipping process, resulting in no crystals available for stabilizing the air bubbles. Lack of control over fat polymorphism was reported to hinder air incorporation (Mishima et al., 2016) where FHR-B β' crystals were unable to incorporate any air, compared to β crystals. Finally, Binks & Marinopoulos (Binks and Marinopoulos, 2017) demonstrated that aeration of pure CB was possible if partly molten (SFC between 20 and 30%) as shown in Fig. 2. Further work by Metilli et al. (2020) determined that, in a model CB – Sunflower oil system, the air bubbles were stabilized by β (V) cocoa butter nanoplatelets; the oleofoams were stable to oil drainage up to three months of storage. However, at present there is scarce literature available regarding the acceptance of these novel materials in chocolate products by customers.

### 2.4. Proteins gels and polysaccharides as CB replacements

Protein based gels have been used in different food products to reduce their fat contents, including addition of gelatine in low fat yoghurt (Nguyen et al., 2017). Using this strategy to partially substitute CB and increase the protein content of an alternative chocolate product would appeal to active consumers looking for a product with a low fat and high protein health claim. The partial substitution of CB with collagen hydrolysate led to acceptable results with the best formulation reducing the fat and energy content by 11% and 35.8% respectively (Ashrafie et al., 2014). In this study a sensory analysis was conducted. No significant difference in acceptability was found between the control and the reduced CB chocolates, even though the rheological properties were different, and the reduced CB chocolate did not fit the standard rheological model used to test chocolate (Ashrafie et al., 2014). This shows how the rheological properties do not have to be identical to those of normal chocolate in order to have a product that is still acceptable to the consumer. However, Ashrafie et al. (2014) did not investigate the effect of collagen hydrolysate on the crystallization of CB, which could have shown why the chocolate does not have the same rheological model as normal chocolate. It is also important to consider that collagen is of animal origin and is not consumed by those following vegetarian or vegan diets.

As well as protein, polysaccharides could be used to substitute CB. β-glucan and inulin, together or separately, have been shown to be able to replace up to 50% of CB with acceptable sensory characteristics (as shown in Fig. 3) and may be able to give the final product a health claim (Rezende et al., 2015). The increase in fibre led to an increase in viscosity of liquid chocolate. There was also an increase in shear stress that may have been due to the polysaccharides disrupting the CB crystal network (Rezende et al., 2015). This may also have an effect on CB crystallization, but that was not investigated in this study and is an area for further research. Rezende et al. (2015) tested these polysaccharides on sucrose free chocolate; a formulation strategy that could provide several health claims: low in sugar, low in fat, high in fibre and beta-glucan to control blood cholesterol levels (Union, 2017).

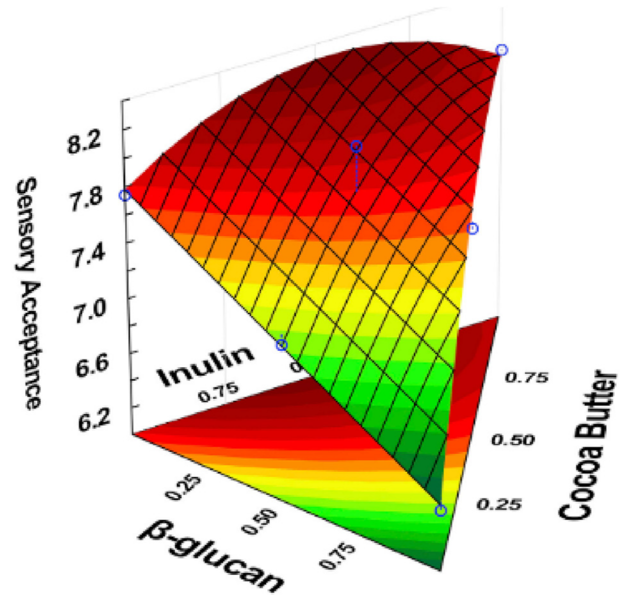


**Fig. 3.** Sensory acceptance of sucrose-free chocolates containing inulin, β-glucan, and CB at different substitution levels. 10-point hedonic scale was used (1-strongly dislike to 10-strongly like). Adapted from (Rezende et al., 2015).

## 3. Cocoa butter alternatives: economic and sustainability considerations

The use of cocoa butter alternatives (CBAs) as CB replacers aims to create alternative chocolate products with improved manufacturing costs and sustainability, depending on the origin of the replacing fat. It can also result in improved properties such as thermal resistance. For example, the use of a mango kernel fat CBA to replace CB created a chocolate with increased heat resistance for use in tropical countries (Tran et al., 2015). CBAs are usually classified based on their chemical composition, as explained in Table 2. Almost 90% of CB available on the market is made up of three triglycerides: 1-palmitoyl-2-oleoyl-3-stearoyl glycerol (POS), 1,3-distearoyl-2-oleoyl glycerol (SOS) and 1,3-dipalmitoyl-2-oleoyl glycerol (POP) (Ray et al., 2012). Chemical similarity to CB is essential for CBAs. If a CBA has a very different triglyceride composition than CB, then the crystallization behaviour and the rheological properties of the liquid chocolate might be unacceptable from a product quality point of

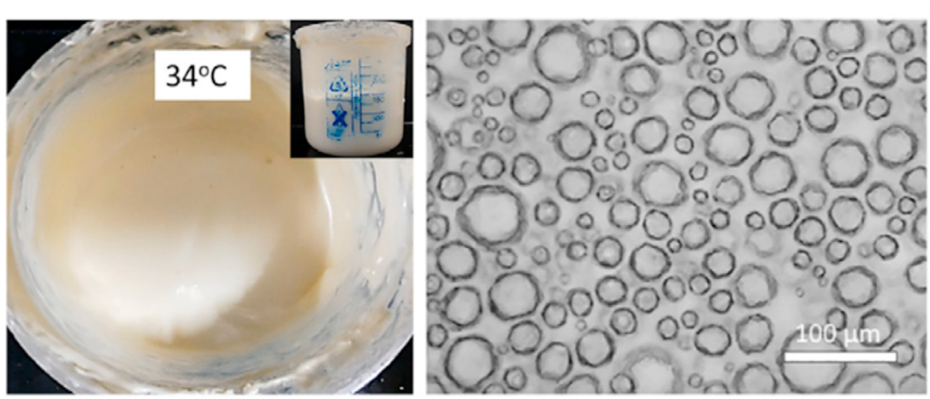


**Fig. 2.** Cocoa butter-based oleofoam obtained by whipping cocoa butter with a hand mixer. Macroscopic appearance (left) and optical microscopy image (right), showing textured air bubbles stabilized by fat crystals (adapted from (Binks and Marinopoulos, 2017) with permission from Elsevier).

**Table 2**
Definitions of CBAs, adapted from (Lipp and Anklam, 1998).

| Term | Definition |
|---|---|
| Cocoa butter alternative (CBA) | Generic term for any fat used fully or partially instead of CB in chocolate to provide similar functions. |
| Cocoa butter equivalent (CBE) | A plant-based fat, which does not contain lauric acid, used in part or wholly instead of CB and displaying similar characteristics. When mixed with CB, it does not affect the functionality of CB in chocolate. |
| Cocoa butter substitutes (CBS) | Plant fats that contain lauric acid and other fatty acids compositions not found in CB. This makes them unable to mix with CB so can only display acceptable properties with full CB substitution. |

view.

Any alternative fat chosen and its total amount in the final product must be considered carefully as it can cause detrimental changes to CB crystallization or lead to the product being classified as a 'compound' rather than 'chocolate' (Union, 2000). The origin of CBAs is also important to consider as it affects chocolate sustainability. In the next paragraphs the use of CBAs as chocolate ingredients is discussed, particularly their effect on the crystallization behaviour of chocolate.

### 3.1. Design of optimal CBEs for chocolate manufacturing

Fats of different origin can be mixed to obtain CBEs with a similar fatty acid composition to CB. Many CBEs are also obtained by chemically manipulating natural fats using enzymatic interesterification. This technique was developed in the 1980s and is used to modify the composition of CBEs to form more of the triacylglycerides (TAGs) that are normally found in chocolate. For example, a 1,3 regiospecific enzyme can be used with palm mid-fraction triglycerides and stearic acid to create triglycerides with a higher proportion of steric acid in the 1,3 positions, as is common in CB (Undurraga et al., 2001). This process needs less energy and causes less isomerism than other similar unit operations such as fractionation (Undurraga et al., 2001).

It is beneficial to use CBEs with a similar fatty acid composition to CB, as they give comparable chocolate properties such as similar melting points. Ghazani & Marangoni (Ghazani and Marangoni, 2019a) studied the phase behaviour of mixture of POS, SOS and POP and showed how slight differences in their ratio can change the melting point and crystalline structure of the mixture. Large differences between the triacylglycerides composition of CBE and CB would cause large changes in the crystallization process. Therefore, the higher the similarity between the CBE and CB itself, the higher the proportion of CBE that can be used in a chocolate related product without significant changes in the crystallization behaviour of chocolate (Ghazani and Marangoni, 2018; Jia et al., 2019). Ghazani & Marangoni (Ghazani and Marangoni, 2018) studied the crystallization behaviour of mixtures of enzymatic interesterified shea stearin and palm mid-fraction and investigated their use as CBEs. A 60:40 w/w mixture produced a CBE with a similar TAG composition and crystal structure to CB, as shown in Fig. 4 A and B (Ghazani and Marangoni, 2018). However, when mixing CB with this CBE in a ratio 85:15 w/w, the final crystal size was smaller compared to pure CB as β′ polymorph crystals formed instead of the mixture of β and β' crystals that was obtained in pure CB (Ghazani and Marangoni, 2018). It was suggested that this was due to the mixture having less trisaturated TAGs than pure CB (Ghazani and Marangoni, 2018). Partial substitution can be beneficial as some CB may help in keeping a close resemblance to normal chocolate. However, fully substituting CB may allow better control of fat crystallization during chocolate processing.

Illipe butter has also been used with palm mid fraction (to provide POP) to obtain a CBE through enzymatic interesterification (Bahari and Akoh, 2018a). When interesterified correctly, this CBE can have a very similar triacylglycerides composition and give similar crystal morphology to CB. Additionally, this CBE has the capability to crystallise in the β polymorph; therefore, products containing this ingredient are expected to have close properties to normal chocolate (Bahari and Akoh, 2018b). This was shown in white and dark chocolates samples (Bahari and Akoh, 2018b). This CBE also generated products with similar rheological properties to CB chocolate. The same study also suggested that fat

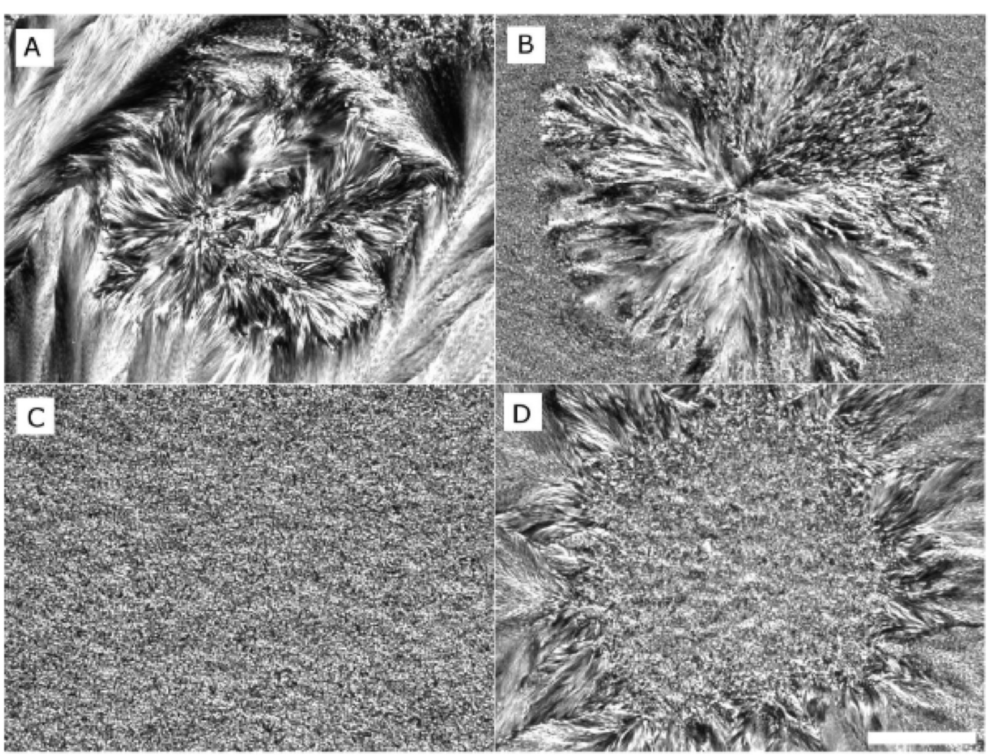


**Fig. 4.** Polarised light micrographs of CB (A), shea stearin and palm mid-fraction CBE, (B), CB and CBE 85:15 w/w mixture (C), and shea stearin and palm mid-fraction in a 60:40 w/w non-interesterified mixture (D). Bar represents 20 μm. Adapted from (Ghazani and Marangoni, 2018).

blooming in dark chocolate could be delayed with the addition of a 0.5% w/w sugar ester to the CBE. However, illipe butter comes from the nut of the *Shorea Stenoptera* tree, which is native to certain areas of Indonesia and Malaysia. This plant has a decreasing population with a classification of near threatened from the ICUN Red List (Randi et al., 1256). If the use of illipe butter increases, the population of the *Shorea Stenoptera* tree might decrease even further, making illipe butter an unsuitable choice for chocolate.

Palm fat can also be made into a CBS that is comparable to CB. The palm mid-fraction, refined bleached deodorized palm kernel oil and palm stearin are all formed through fractionation of palm fat and have different melting profiles (Biswas et al., 2016). Mixing these different palm fat variations can form a CBS with better properties than using just one of them, especially if they are combined to give compatible physicochemical behaviour to CB (Biswas et al., 2016). It was found that a 14.9/59.6/25.5 (%w/w) blend of palm mid-fraction, refined bleached deodorized palm kernel oil and palm stearin gave crystals with similar morphology to CB (Biswas et al., 2017a). However, other properties such as polymorphism and melting profile were different to CB, which could hinder the acceptability of this CBS in alternative chocolate products. A further study used enzymatic interesterification to control the melting range of this palm based CBS and tested the new material by blending it with CB (Biswas et al., 2017b). A 5 g CBS/100 g blend gave no significant difference in the sensory testing to regular chocolate (Biswas et al., 2017b). A higher amount of substitution would be preferable for companies who are looking to lower the amount of CB in their products. However, the sensory analysis was significantly different for a 20 g CBS/100 g blend and it would likely not be accepted by consumers (Biswas et al., 2017b). Both the 5 and 20 g CBS/100 g blend mixtures were found to have similar melting profiles and polymorphism to CB, showing that interesterification and then blending of this CBS with CB can change the crystallization behaviour of the final triglyceride mixture (Biswas et al., 2018). These mixtures presented oleic and palmitic fatty acids, especially as 2-oleoyl-1,3-dipalmitoylglycerol (POP), in similar amounts to CB, which could explain their compatibility with CB (Biswas et al., 2018). Palm fat has high saturated fatty acids, which could be reduced by using hybrid palm fat stearin, which has less saturated fatty acids than normal palm stearin (Ruedas et al., 2020), this was not tested in a chocolate formulation so it may not provide the solid fat content needed due to its higher unsaturated fatty acid content.

### 3.2. Increasing the sustainability of CBAs

While one can formulate CBEs and CBSs that can be successfully included in chocolate, there may still be some drawbacks, including the poor sustainability of certain compounds, for example palm oil (Azhar et al., 2017). A way to tackle this is by using high oleic sunflower oil or high oleic-high steric sunflower oil. Sunflowers can be grown in more temperate climates and can provide a more sustainable alternative to fat extracted from tropical plants. Bootello et al. (2012) used high oleic-high stearic sunflower oil with palm mid-fraction to produce a CBE that is more compatible with CB than just sunflower oil on its own. Without the palm mid-fraction, this CBE would not contain significant amounts of -oleoyl-1-palmitoyl-3-stearoylglycerol (POS) and POP, which would negatively affect the crystallization behaviour (Bootello et al., 2012). In fact, POS is important in CB crystallization as its stability and content were found to dictate CB polymorphism stability (Ghazani and Marangoni, 2019b). Kadivar et al. (2014) showed using only interesterified high oleic sunflower oil with CB that the amount of form V CB crystals decreased with increasing the content of the CBE because of the slower crystal growth rate in mixtures compared to pure CB. High oleic-high stearic sunflower oil has also been shown to provide more compatible physiochemical properties with CB than high oleic sunflower oil (Kadivar et al., 2016). An earlier study looked at olive pomace oil and using enzymatic interesterification to produce a CBA without any palm fats (Ciftci et al., 2009). However, increasing the concentration of olive pomace oil in the produced CBA also decreased the content of the major CB fatty acids, which led to a decrease in the oxidative stability of the blend and a reduced shelf life. These examples show how challenging it is to form a sustainable CBA with all the desired properties.

A popular tropical fat that can be used to formulate CBAs is mango kernel fat, which can be obtained through supercritical fluid extraction (Jahurul et al., 2014a). It is popular because, as a waste product of mango foodstuffs, it is relatively more sustainable than other tropical fats. It also has potential health benefits due to being a free-trans-fat and an antioxidant source (Jin et al., 2019b). Chocolates using mango kernel fat have been observed to keep their shape at 32 °C, unlike CB-based products, with melting occurring at 37 °C because of a high 2-oleoyl-1, 3-distearoylglycerol (SOS) content (Jin et al., 2019a). The use of interesterification of mango kernel fat in chocolate also has the potential to decrease fat bloom as it generates a denser crystal network where plate-like crystals form instead of the classic CB spherulitic crystals (Jin et al., 2019a). Chocolate with a partial substitution of CB with mango kernel fat (80:20 w/w substitution) was found to show a lower hardness than the control chocolate (Naeem et al., 2019). This study tested fats from different varieties of mango, which showed differences between the rheological properties of each variety, with Lal Badshah having the best behaviour among the varieties (Naeem et al., 2019). This is an important point to bear in mind for all tropical fat-based CBAs as the same may be true for their varieties. Regarding crystallization, blends of mango kernel fat and palm mid-fraction showed orderly packed spherulite crystallization, like CB, with the best melting properties occurring when the mango kernel fat proportion was between 65 and 80% (Jahurul et al., 2014b). As previously mentioned, palm oil tends to be unsustainable, therefore mixing it with mango kernel fat could give a palm oil-based CBAs a better structure as well as improved sustainability. A further way to decrease the fat content of chocolate using mango kernel fat is by using it in a gelatine based emulsion gel structure with CB (Sagiri et al., 2014). However, this structure caused the fat crystals to not fully transform into a β polymorph, reducing the shelf life of the chocolate product (Sagiri et al., 2014). Furthermore, the use of gelatine to form such gel would make the chocolate product unsuitable for vegetarians and vegans.

Tea seed oil is a waste product of tea production that has also been considered as a possible CBA, when enzymatically interesterified, with the potential to reduce fat bloom in chocolate (Zarringhalami et al., 2010). Sensory analysis showed that products with CB substituted with up to 10% w/w with this CBA has acceptable sensory properties as interesterified tea seed oil does not cause any undesirable change to CB crystallization behaviour (Zarringhalami et al., 2010). This compatibility with CB may be due to high levels of oleic acid (Sarmah and Das, 2018). Tea seed oil is also high in linoleic acid, which can provide health benefits (Sarmah and Das, 2018).

Coconut products are another class of ingredients that have been considered as CB replacers in chocolate products. Coconut is cheaper and slightly more sustainable than CB in some countries, such as India. Furthermore, there are different parts of the coconut that can be used in chocolate manufacturing: coconut oil, milk, and cream (Divya et al., 2017; FAOSTAT, 2019). Substitutions of CB with either 30% coconut milk, 20% coconut cream or 10% coconut oil in chocolate were the preferred variants in sensory analysis participants. However, the rheological properties and crystallization behaviour were not tested (Divya et al., 2017). These tests would allow a closer understanding of how these coconut products affect CB crystallization and chocolate structure. Using coconut products could also lead to a different taste within the chocolate. This could allow the design of interesting novel products, but it would also need to be controlled to allow desirable chocolate properties.

Other possible sustainable plants to consider for CBAs are wild mango (with a close triacylglyceride composition to CB), Cinnamomum camphora seed oil and bambangan kernel fat (Akhter et al., 2016; Jahurul et al., 2019; Ma et al., 2019). These examples and the others discussed in this review show how research into different plant CBAs is extensive. The sustainability, price, and comparable properties to CB need to be considered for each of these novel ingredients to determine their viability. As mentioned previously, these decisions are all subject to the location of the chocolate manufacturing and consumption. For example, coconut fats may be more sustainable in India but not elsewhere. Therefore, location is important when deciding the CBA to use in a chocolate formulation.

## 4. Sugar reduction strategies

It can be hard to reduce sucrose in many food products due to its important structural role, in addition to its sweetening power. For example, in biscuits sugar gives hardness and inhibits gluten development (Gallagher et al., 2003). This ingredient also has an important role in chocolate; in fact, sucrose decreases the nucleation and growth rate of CB crystals, which affects many rheological properties of liquid chocolate (Svanberg et al., 2011). One study looking at the effect of particles on chocolate properties showed that increasing their concentration, and decreasing their size, increased the rate of crystallization kinetics on CB due to their role as heterogeneous nucleating agents (Fernandes et al., 2013). However, when considering reduced fat chocolate products, it has been shown that a blend of both small and large sugar crystals gave the best rheological properties due to their better packing abilities (Do et al., 2007).

### *4.1. Sucrose replacers*

Different compounds have been looked at to replicate sucrose in novel chocolate products. One compound that showed potential is inulin, a polysaccharide. Rezende et al. (2015) showed that a 25% w/w inulin substitution of CB in sugar free chocolate gave a similar acceptance to normal sucrose based chocolate (Rezende et al., 2015). The amount of inulin used in chocolate must be controlled as increasing it increases liquid chocolate viscosity (Aidoo et al., 2017). However, when mixing inulin with a polydextrose this increase was not observed, and the yield stress of the reformulated product was not significantly different than normal chocolate. (Aidoo et al., 2017). Polydextrose and inulin were used as bulking agents, to replicate the rheological properties that sucrose imparts to chocolate. Whereas, intense sweeteners such as stevia provide the sweetness. At high fat content and without the use of polydextrose, 38% w/w inulin can fit the standard rheological Casson model as this ratio provides the necessary solid volume fraction (Kiumarsi et al., 2017). How inulin is used in relation to other ingredients is important for the functionality of the final product. Inulin and polydextrose mixtures generate a more packed CB crystal network leading to slower melting, which could give the chocolate a different mouthfeel (Aidoo et al., 2015). This high packing is due to inulin disrupting the crystallization of CB causing the formation of large CB crystals, and polydextrose fills in the gaps in the network (Aidoo et al., 2014, 2015). Inulin on its own has also been shown to induce the nucleation of form V CB crystals when it has the correct degree of polymerisation (Shah et al., 2010). Using inulin and polydextrose may also allow for the use of a health claim of “high in fibre” and/or “low in sugar”. However, if there is still a high fat content, the product may not be called healthy overall.

Bulk sweeteners do not need bulking agents like polydextrose and inulin. These compounds can fully replicate the structural role of sucrose as well as give sweetness. Isomalt, a mixture of glucosyl-glucitol and glucosyl-mannitol disaccharides, has been shown to have a similar effect to sucrose on the rheological properties of chocolate. (Kiumarsi et al., 2017; Bender, 2014). However, isomalt was given a low sensory acceptability for taste and texture in sensory tests; although this could be due to the addition of stevia and the use of a ball mill process to adjust the size distribution of isomalt particles (Rad et al., 2019). Another study showed that xylitol had higher sensory acceptability, but it also increased the viscosity in the liquid chocolate (Rad et al., 2019). This is because sugar alcohols such as xylitol dissolve quicker in the mouth compared to sugar, due to the higher amount of –OH groups in sugar alcohols with high water solubility in the mouth (Rad et al., 2019). The free –OH groups on the xylitol molecule also lead to more interactions with each other, and this causes a high viscosity in liquid chocolate (Rad et al., 2019). Meanwhile, other sweeteners such as maltitol and tagatose could reduce the amount of fat bloom in chocolate compared to sucrose, with tagatose shown to be the most effective at suppressing CB recrystallization in solid chocolate. (Son et al., 2018). Maltitol, isomalt and inulin all resulted in different melting properties for the chocolate compared to sucrose, which would result in different mouth feel and may affect acceptance (Kiumarsi et al., 2017). However, in this study the concentration of all three bulk sweeteners was the same as that of the sucrose in the reference sample, 38% w/w. These sweeteners all have different chemical structures and chemical-physical properties. Therefore, a different concentration may be needed to give the best CB crystallization and rheological properties to novel sugar-free chocolate products. Therefore, looking at different concentrations would give more insight into the way these sweeteners work. A mixture of isomalt and maltose in sucrose-free dark chocolate was shown to give similar properties to sucrose dark chocolate when the tempering method was changed to a CB form V seeded technique (Toker et al., 2019). Changing tempering techniques should be considered when making novel alternative chocolate products due to the effect on novel ingredients on fat crystallization.

## 5. Milk replacements with plant milk alternatives

Milk powder and anhydrous milk fat are used to make milk chocolate, and its addition affects tempering conditions as well as inhibiting fat blooming (Liang and Hartel, 2004). However, replacing milk powder could be economically beneficial and can allow the formulation of products that are lactose free and vegan. Mixture of peanut and cowpea milk has been considered as a substitute for milk powder as these two plants are common in Ghana, where cow milk is harder to find (Aidoo et al., 2012). An earlier study used sensory tests to determine the optimum amount of peanut-cowpea milk in the chocolate formulation (Aidoo et al., 2010). This milk mixture has a high milk fat content, which dilutes CB leading to slower crystal formation. This gives a softer chocolate with acceptable sensory properties (Aidoo et al., 2012). Considering both rheological properties and sensory analysis helps to understand what ranges of different rheological properties can produce an acceptable product. Plant milks may not be able to supress fat blooming like milk powder; therefore, the addition of other bloom inhibitors may be needed for this type of chocolate. Lactose free chocolate is good for lactose intolerant consumers but the lactose in the milk powder influences the chocolate properties. In particular, a change in chocolate viscosity was noticed depending on the lactose particles being amorphous or crystalline; but it is still unknown if the inclusion of lactose itself affects chocolate properties (Bolenz et al., 2014). Some studies considered the use of soy milk as a milk chocolate ingredient, but samples with complete substitution were not given a high overall sensory acceptability, since the hardness of the chocolate was increased significantly (Pandey and Singh, 2011). There was no change in CB crystallization behaviour when using up to 15%–20% of soy milk in the chocolate mass (Pajin et al., 2013; Zarić et al., 2015). Unfortunately, there is a lack of studies looking at milk replacements in milk chocolate and more research is needed in this area.

## 6. Conclusions

This review sheds some light on the effect of different potential reformulation strategies on the crystallization behaviour of chocolate. The use of soft matter such as gels, foams or emulsions shows a good potential and allows correct CB crystal growth. There are different CBAs that can be used to decrease the manufacturing cost of chocolate and keep a close resemblance to CB in terms of physiochemical properties, particularly using enzymatic interesterification. Also, there is potential for novel alternative chocolate products to provide additional health benefits if the ingredients are carefully chosen. Sucrose replacers are available, which can even add further health or functional benefits to the novel formulations. The use of plant milk in chocolate is also promising, but more research is needed to find the potential of different plant milks and their combinations.

Recent trends have shown that consumers think that taste is more important than health claims in chocolates, and premium, indulgent chocolates are currently on the rise (Euromonitor, 2019). This highlights how the formulation of novel chocolate products need to be carefully designed to ensure a high-quality product that also appeal to consumers.

## Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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