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Optimization of dark chocolate formulation with roasted coriander cake as cocoa substitute

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ABSTRACT

Since worldwide demand for cocoa is predicted to exceed supply in the upcoming years, this study aimed to find a substitute for cocoa powder in dark chocolate formulation. In the first phase, Coriander cake, a by-product of extracting oil from coriander seeds, was roasted at 165 °C respectively for 2, 4, 6, 8, and 10 min, and physicochemical characteristics (pH, aw, and color parameters), compositional properties, and total phenolic content were examined and compared to cocoa powder. Coriander cake roasted for 6 min was almost equal to cocoa powder and qualified for use in dark chocolate formulation. In the second phase, roasted coriander cake (RCC) was used as a partial replacement for sugar and cocoa powder. The physicochemical and rheological characteristics of RCC chocolates were evaluated. Appropriate mathematical models defined by using best-fitting analysis described results. According to the results, the fitted models illustrated a desirable coefficient of determination (\geq 90 %). The optimization of the variables indicated that using RCC 6.08 g, Sugar 14.68 g, and Cocoa 12.23 g produced the optimized RCC chocolate with the highest desirability while maintaining the quality characteristics without undesirable changes. The samples' flow behavior and physicochemical properties showed that RCC could be used as a cocoa substitute.

1. Introduction

Chocolate is a cocoa-based emulsion and is one of the most popular confectionery products in the world. The main types of chocolate are dark, milk, and white, which differ in cocoa solids, milk fat, and cocoa butter (Barišić et al., 2021). The demand for chocolate is a highly competitive market, in which variables such as food safety, efficiency, affordability, taste, and high quality play a significant role in consumer demand. The increasing number of issues related to cocoa production has worsened the global market, which requires many cocoa beans (Del Prete & Samoggia, 2020). Cocoa, the main ingredient in chocolate and similar products, is obtained from the seeds of the fruit of the Theobroma cacao tree, which is native to the South American region (Gomes dos Santos & Fontes, 2020). Cocoa powder is known for its attractive taste and aromatic properties. Conversely, cocoa contains compounds such as theobromine and caffeine, which have the potential to cause adverse health effects (Loullis & Pinakoulaki, 2018). Furthermore, the continuous increase in prices coupled with constraints on cocoa resources due to increasing global demand has encouraged certain researchers to investigate alternative sources of cocoa (Akdeniz et al., 2021). Furthermore, this type of research is of great importance because the innovation of new products, where compounds act as substitutes for cocoa in the manufacture of chocolate, aligns with consumer preferences and promotes a more sustainable use of natural resources. Chocolate manufacturers are looking for alternative, natural ingredients that can lead to lower-calorie, more affordable, and healthier chocolate products, including those made with plant-based oils instead of cocoa butter(Toker, Sagdic, et al., 2016). Nuts, fruits, and vegetables serve as sources of polyphenolic compounds(Miller et al., 2006). Numerous studies have demonstrated that many unfamiliar plant tissues are not only edible and enhance food flavor, but also possess high biological activity and offer significant health benefits (Jurikova et al., 2012; Milião et al., 2022) Integrating these components or their by-products into new formulations is essential for efficient waste management, cost reduction, and improving of products containing bioactive compounds(Beres et al., 2019). Waste and by products from plants, including

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peels, leaves, seeds, bran, kernels, pomace, and oil cake, can be sourced from numerous fruits, vegetables, cereals, oilseeds, roots, tubers, and other legumes during both the initial production and the later processing and distribution phases(Difonzo et al. Sruthi et al., 2021). Coriandrum Sativum L. (coriander) is a plant that can be annual or biennial, belonging to the Coriandrum genus within the Umbelliferae family(Lee et al., 2020). Coriander cake, a by-product of coriander seed oil extraction, is typically used as animal feed. Thus, utilizing of this byproduct in chocolate manufacturing could yield beneficial outcomes for both public health and the economy. Coriander seeds have been used in traditional medicine as a remedy for indigestion and as an ingredient in topical treatments for rheumatism(Norman, 1991) Recent research on coriander, has analyzed the composition of fatty acids, sterols, and tocopherols found in the by-product of oil extraction from its seeds(Sriti et al., 2013). Additionally, Darughe et al. (2012) examined the essential oil composition, antioxidant and antifungal attributes of coriander seeds in a butter cake recipe(Darughe et al., 2012). When coriander seed oil is extracted using cold press machinery, the oil has good quality(Sriti et al., 2009), and the residue (coriander cake) contains many beneficial compounds. Thermal treatments such as roasting seeds, nuts, fruits, and vegetables cause sensory, nutritional, and physicochemical changes, enhancing their color, flavor, and antioxidant properties due to the Maillard reaction. Numerous studies have explored the impact of various thermal methods, including roasting, on the physicochemical and color characteristics of different seeds, such as soybeans, hazelnuts, coffee beans, and corn seeds(Lemos et al., 2012; Turan et al., 2015). Roasting is commonly used in the food industry to improve flavor, bioactive compound levels, and the overall bioactivity of the food matrix (Corzo-Ríos et al., 2022; Dhull et al., 2021; Duan et al., 2025). Roasting is crucial in influencing the color of food. Color is regarded as a crucial element that consumers evaluate when assessing food items, positioning it as an important aspect of food quality that greatly affects market acceptance(Wu & Sun, 2013). The level of roasting significantly influences these markers of the end product. The primary flavor and aroma compounds in roasted foods are generated by the Maillard reaction and caramelization(Bölek, 2021). Products with a new formulation must maintain a quality comparable to common chocolate. The quality attributes influencing chocolate quality, consumer preference, and acceptance include texture, taste, and flavor. Additionally, the color of chocolate has a considerable impact on consumer decisions(Afoakwa, Paterson, & Fowler, 2008). Furthermore, the distribution of particle size in chocolate affects its flow properties, texture, and sensory perception. When particles are larger, their points of contact with one another are limited. The refining process enhances the surface area of the particles and increases the points of contact between them. Consequently, the yield stress needed to surpass the interaction of these particles increases. Moreover, larger particles contribute to a gritty texture(Beckett, 2009). Consumers are seeking low-cost, functional foods that are low in calories, sugars, and fats with high amounts of fiber. Considering its nutritional value, functional aspects, and local production potential, coriander cake, a by-product of oil extraction from coriander seeds, was used for this purpose. In the first stage, coriander cake was roasted at 165 °C for 2, 4, 6, 8, and 10 min (RCC). RCC physicochemical properties (pH, aw, and color parameters), compositional properties, and total phenolic content were investigated, and compared with cocoa powder. In the second stage, the optimized roasted coriander cake (RCC), which had the most similarity to cocoa powder in physicochemical properties, was used in dark chocolate formulation as a partial replacement for sugar and cocoa powder, and physicochemical and rheological properties of RCC chocolate was evaluated.

2. Material and methods

2.1. Preparation of roasted coriander cake (RCC)

Coriander cake was derived as a by-product from the pressing

process of coriander seed oil at a Golbaran factory in Gilan, Iran. The coriander variety (*Coriandrum sativum* L.) was cultivated in Nahavand, Iran. Coriander cake was roasted using a laboratory electric roaster (Probot PRE 1Z model, made in Germany). 100 g of the sample entered the roaster rotating chamber at 165 °C and roasted for 2, 4, 6, 8 and 10 min. After the specified time, Roasted Coriander Cake (RCC) was exposed to a two-step grinding procedure by a laboratory mill (IKA-A11, made in Germany) (first 500 μm then 200 μm) to (i) attain homogenous particle size distribution of the components used in chocolate samples (ii) do not affect the mouthfeel adversely(Akbari-Adergani et al., 2021; Alsaed & Alghzawi, 2000).

2.2. Preparation of RCC chocolate

RCC (0.0-33. 0 g/100 g) (Golbaran Gilan, Iran), sugar (0. 0-33. 0 g/ 100 g) (sugar company, Iran), Cocoa powders (0.0-33.0 g/100 g), (Altinmarka, Turkey), cocoa butter (30.48 g/100 g) (KI -kepong, Malaysia), Maltodextrin (36.02 g/100 g) (Parsipowder, Tehran, Iran), Lecithin (0.40 g/100 g) (shimistore, Tehran, Iran), and polyglycerol polyriconalate (PGPR) (0.10 g/100 g) (Palsgaard, Zierikzee, Netherlands) were used. The RCC chocolate samples were produced following the procedure by Toker, Sagdic, et al., 2016 (Toker, Zorlucan Demir, et al., 2016). The proportions of sugar, cocoa, and RCC, based on a formulation design by the design expert software, and other ingredients in fixed proportions were used to produce 2 kg of RCC chocolate samples for each formulation. In summary, sugar, cocoa, and RCC were blended using a low-speed mixer according to the proportions of each formulation. Next, the cocoa butter and lecithin, which were melted in a water bath (60 °C, 20 min), were added to the dry ingredients and mixed for 5 min at medium speed. The produced chocolate paste was subjected to refining-conching using a pilot-scale ball mill (2 h, 45 $^{\circ}\text{C},$ and 60 rpm). The melted chocolate samples were cooled to 35 °C and poured into pre-warmed polycarbonate chocolate molds (35 °C). Subsequently, they were cooled in cooling chambers (10 °C, 30 min) removed from molds, wrapped in aluminum foil, and kept at 15 °C before the analysis.

2.3. RCC analysis

2.3.1. Proximate RCC chemical analysis

The AOAC International (2023) 931.04 method were used to determine the moisture content, fat, fiber, and ash content in the RCC. Water activity of all samples was done at 25 °C using Novasina Labmaster (made in Switzerland) and a laboratory pH meter (Metrohm 827, made in Switzerland) was used for pH measurement(McCleary, 2023).

2.3.2. RCC color measurements

The Hunter Lab Colorimeter (Colorflex EZ model: 45/0, USA) was used to determine the color values of the sample(Sengar et al., 2022) calibrated for the CIE-L*a*b* system (L* = lightness; a* = redness; b* = yellowness).

2.3.3. RCC total phenolic content

The Folin–Ciocalteu colorimetric technique was used to quantify the total phenolic contents (TPC) in the spice seed extracts(Phuyal et al., 2020). One gram of gallic acid was dissolved in one hundred milliliters of 70 % aqueous methanol (1000 mg/L) to create a standard gallic acid solution. From the stock solution, several concentrations of gallic acid solutions (10, 25, 50, 100, 150, 200, and 250 mg/L) were made. Four milliliters of a 7.5 % Na2CO3 solution and 2.5 ml of a 10 % Folin–Ciocalteu reagent (FCR) were added to each concentration. As a result, the blue-colored mixture was thoroughly shacked and allowed to be in a dark place for half an hour. After that, a serious standard concentration absorbance was measured at 760 nm in comparison to a blank. The FCR reagent, turning them dark blue, which is then detected using a UV–visible spectrophotometer, oxidizes phenols in plant

Table 1Cocoa, Coriander cake and RCC: Physicochemical and technological characteristics.

parameter	Cocoa powder	Roasting time						
		R0 (Coriander Cake)	R2	R4	R6	R8	R10	
Moisture Content (g.100 g - 1)	2.50 ± 0.24	6.02 ± 0.05^a	$3.15\pm0.02^{\mathrm{b}}$	$2.97\pm0.02^{\rm c}$	2.45 ± 0.05^{d}	1.99 ± 0.01^{e}	$1.56\pm0.03^{\rm f}$	
Fat (g.100 g - 1)	22.81 ± 0.53	$18.01 \pm 0.24^{\rm d}$	17.97 ± 0.71^{cd}	$18.74\pm0.53^{\rm c}$	20.98 ± 0.76^{b}	22.97 ± 0.35^{a}	22.96 ± 0.41^{a}	
Fiber (g.100 g − 1)	32.21 ± 0.23	39.24 ± 0.21^{a}	27.54 ± 0.35^{b}	23.65 ± 0.17^{c}	22.46 ± 0.14^{d}	$22.12\pm0.33^{\text{d}}$	$21.01 \pm 0.10^{\rm e}$	
Ash (g.100 g - 1)	6.11 ± 0.12	4.97 ± 0.12^{c}	5.01 ± 0.07^{c}	5.05 ± 0.06^{c}	$5.11\pm0.04^{\rm c}$	$5.27\pm0.08^{\mathrm{b}}$	5.73 ± 0.11^a	
Water activity	0.18 ± 0.04	0.336 ± 0.02^{a}	$0.054 \pm 0.01^{\mathrm{b}}$	0.051 ± 0.03^{c}	$0.026 \pm 0.02^{\rm d}$	$0.024 \pm 0.03^{\mathrm{de}}$	0.022 ± 0.01^{e}	
pH	6.98 ± 0.05	6.44 ± 0.03^{a}	$6.32\pm0.05^{\mathrm{b}}$	$6.31\pm0.02^{\mathrm{b}}$	6.24 ± 0.06^{c}	6.24 ± 0.04^{c}	6.24 ± 0.07^{c}	
L*	48.56 ± 0.03	84.15 ± 0.05^a	$65.53 \pm 0.07^{\mathrm{b}}$	61.12 ± 0.58^{c}	$41.43\pm02^{\rm d}$	38.75 ± 0.23^{e}	$31.12\pm0.1^{\rm f}$	
a*	11.16 ± 0.13	$1.32\pm0.01^{\text{d}}$	7.38 ± 0.21^{c}	$9.01\pm0.11^{\rm b}$	10.54 ± 0.10^a	10.54 ± 0.16^a	10.55 ± 0.12^{a}	
b*	16.12 ± 0.06	19.84 ± 0.51^a	18.32 ± 0.14^{ab}	$17.12\pm0.22^{\mathrm{b}}$	15.21 ± 0.32^{c}	$11.35\pm0.23^{\text{d}}$	8.45 ± 0.45^e	
Total Phenolic Content (mg/100 g)	32 ± 0.18	0.96 ± 0.01^a	0.94 ± 0.02^{b}	0.94 ± 0.01^{b}	0.94 ± 0.03^{b}	0.92 ± 0.01^{c}	0.91 ± 0.01^d	

R0 = (CC), R2 = 2 min, R4 = 4 min, R6 = 6 min, R8 = 8 min and R10 = 10 min. $L^* = lightness$, $a^* = red$, $b^* = yellow$. Same letters in the same line indicate no significant difference (p < 0.05).

extracts. Every experiment was run in triplicate, and the calibration curve was plotted using the average absorbance readings at various gallic acid doses.

2.4. RCC chocolate analysis

2.4.1. RCC chocolate particle size

A particle size analyzer (Shimadzu 2101, Sald, Japan) was used to determine the particle size distribution.

2.4.2. RCC chocolate moisture content

The AOAC (1990) methods from 931.04 were applied to determine the moisture content in the RCC chocolate samples.

2.4.3. RCC chocolate rheology properties

A rheometer (Anton Paar, MCR301, Austria) was used to specify the melted chocolates' rheological properties. To melt the chocolates, they were warmed in an oven preheated to 45 °C for one hour. Before the measurement cycle, chocolate samples were subjected to pre-shear at 40 °C and 5 s $^{-1}$. As the shear rate increased from 2 s $^{-1}$ to 50 s $^{-1}$, the shear stress was calculated (ICA 2000). The Casson viscosity and Casson yield stress were derived from the data after it was fitted to the Casson model(Rasouli Pirouzian et al., 2017). Four replicate measurements' mean values were assessed.

2.4.4. RCC chocolate hardness

The Texture Analyzer (Stable Micro Systems, TA-XT-plus, UK) was used to measure the hardness using a needle geometry and a 5 g trigger force. The maximum penetrating force needed for the needle to penetrate a sample at 20 °C over a distance of 3 mm at a steady speed of 1 mm/s was recorded as hardness, measured in Nioton (N)(Homayouni et al., 2013). The average values of four replicates were evaluated.

2.4.5. RCC chocolate L*

Color characteristics were assessed on the surface of chocolate formulations at room temperature utilizing a colorimeter (Colorflex (45/0, USA). Three measurements were obtained from every chocolate sample. The color assessment involved CIE L* parameter that indicates lightness on a scale from 0 (black) to 100 (white)(Becerra et al., 2023).

2.5. Statistical analysis

To determine the best roasting time for coriander cake, analysis of variance was used with SPSS version 16 software in a completely random design. Means were analyzed using Duncan's test at a significance level of 5 %. The simplex lattice mixture design was used to examine the effect of RCC (A), Sugar (B), and Cocoa (C) on the quality properties of the chocolates. The related factors, including moisture

content, D90, Casson viscosity, Casson yield stress, hardness, and L*, were analyzed, and the fitted models were exposed to variance analysis (ANOVA) to determine significance (p < 0.05), the determination coefficient (R2), and lack of fit. Multiple response optimizations were used to determine the combination of examined variables simultaneously. The levels of ingredients were expressed as parts of the mixture, totaling (A + B + C) to 33. All estimated data were shown as mean \pm standard deviation (S·D). The calculation was done using a statistical software package (Design Expert 7.0.0 trial version; Stat Ease Inc., Minneapolis, USA).

3. Results and discussion

3.1. Characterization of RCC

As indicated in Table 1, the average levels of chemical compounds in cocoa powder for moisture content, fat, fiber, and ash were recorded as 2.50 \pm 0.24, 22.81 \pm 0.53, 32.21 \pm 0.23, and 6.11 \pm 0.12, respectively. The effects of the roasting process on the chemical composition of cocoa, coriander cake, and roasted coriander cake (RCC) are detailed in Table 1. On average, the coriander cake shows 6.02 % moisture, 18.01 % fat, 39.24 % fiber, and 4.97 % ash. With an increase in roasting time from 0 to 10 min, the moisture content declined from 6.02 % to 1.56 %. The fat content in coriander cake is 18.01 %, which is higher than the values found in many seeds and grains, positioning this product as a valuable source of fatty acids. As the roasting time increased, the fat percentage rose, potentially due to thermal degradation that results in fat being released(Oboh et al., 2010). a trend that several researchers have reported during roasting(Lee et al., 2013; Turan et al., 2015). As the roasting duration extended, the ash percentage increased from 4.97 to 5.73, which may be attributed to the reduction in moisture content. According to Table (1), among the samples, RCC roasted for 6 min is very similar to cocoa powder in terms of moisture content, fat content,

According to Table 1, the water activity of coriander cake was 0.336. After 2 min of roasting, this figure dropped to 0.054 (P < 0.05). The water activity level of cocoa powder was measured at 0.18, which is lower than that of coriander cake and higher than roasted coriander cakes. Because of its low water activity, microorganisms are unable to grow in the absence of moisture. Based on the findings (Table 1), the pH value of cocoa powder was found to be 6.98 \pm 0.05. The effect of the roasting process on pH indicates a decrease from 6.44 to 6.24 as roasting time increased, showing a downward trend. With prolonged roasting, the Maillard reaction progresses, leading to a reduction in pH. Certain by-products from the acid caramelization process, such as pyruvic acid, contribute to this decline(Alsaed & Alghzawi, 2000). The influence of the roasting process on color at various times is presented in Table 1. The L* value used as an indicator of the roasting degree in foods like coffee

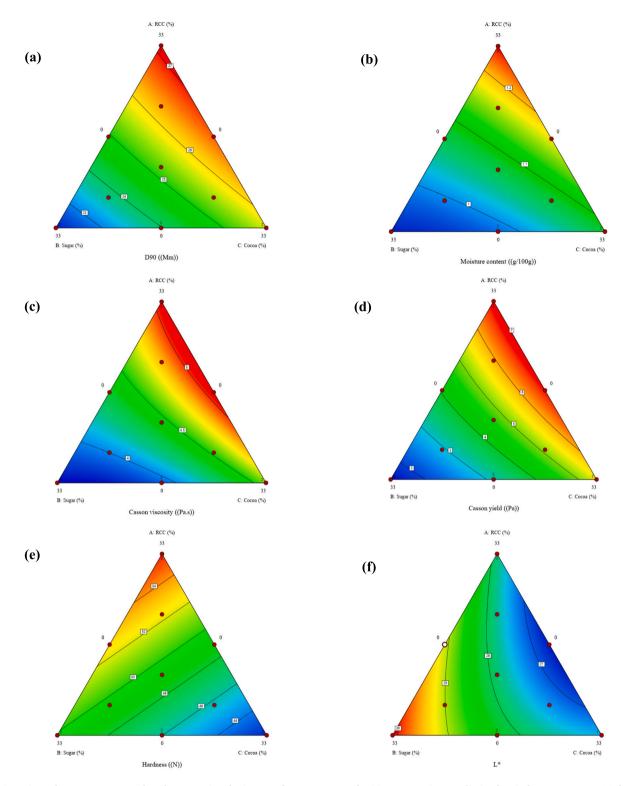


Fig. 1. Estimated response contour plots demonstrating the impact of sugar, cocoa and RCC concentration on physicochemical parameters: a D90, b Moisture, c Casson viscosity, d yield value, e hardness and f L^* .

and cocoa and functions as a temperature-time reference concerning the impact of the final roasting phase(Sacchetti et al., 2009). By increasing the roasting time, the L* parameter values decrease. A lower value of this parameter indicates that the samples' brightness decreases. These changes in color result from the Maillard browning reaction and the formation of hydroxymethyl furfural(Fallico et al., 2003). The trend in the a* value rising from 1.32 in the coriander cake to 7.38 after 2 min of

roasting indicates a shift in the color spectrum from green to red, which continues to intensify throughout the process. These color transformations observed during the thermal treatment of edible powders can be used in products that require a dark brown color with a high nutritional value. Moreover, as roasting time increases, the b* values decline, reflecting a reduction in the blue color in the samples. The L* and a* values of the samples roasted for 6, 8, and 10 min were similar to cocoa

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powder roasted at 135 °C for 30 to 40 min(Zyżelewicz et al., 2013). Phenolic compounds are significant antioxidants due to their ability to donate either a hydrogen atom or an electron, leading to the formation of stable radical intermediates, which in turn help inhibit the oxidation of various biological molecules(Cuvelier et al., 1992). Furthermore, numerous oilseeds and their by-products have been studied for their phenolic content as safe natural antioxidant sources(Wettasinghe et al., 2002). The total phenol content of coriander cake during the roasting process is presented in Table 1. The research indicated that the total phenol content consistently declined as roasting time increased. The most significant loss of total phenol occurred at 10 min of roasting, while the least loss was observed between 2 and 6 min. During roasting, the total phenol content decreased from 0.96 mg GAE/g to 0.91 mg GAE/g. similar patterns of reduction were observed during the conventional roasting of cocoa beans(Arlorio et al., 2005). The reduction in polyphenolic compounds in coriander cake is strictly correlated to the oxidation and thermal degradation of these compounds. High temperatures have been shown to adversely affect and reduce the water content and the polyphenolic content in all roasted samples during conventional roasting. The phenol content decreased between 32.63 % (cocoa beans Arriba) and 54.74 % (cocoa beans Ghana) during traditional roasting methods. A similar declining trend was noted in the roasting of pistachio beans under different roasting conditions concerning chemical composition(Bogoeva-Gaceva et al., 2007). A significant reduction in phenolic content was documented during the conventional roasting of a cocoa bean model system(Oliviero et al., 2009). Additionally, it is important to recognize that the effectiveness of polyphenol extraction is affected by various factors, such as the variety of coriander, drying methods, extraction techniques, and solvents in the extraction process. The chemical structure of coriander is influenced by different factors, including climate, variety, soil type, irrigation, and cultivation techniques. This important by-product is mainly used as animal feed, however, due to its rich nutritional value; it has potential uses in the food industry.

3.2. Characterization of RCC chocolate

3.2.1. D₉₀ value of RCC chocolate

The D90 particle size of chocolate has a significant influence on the texture, flow, melting point, color, and water activity characteristics (Konar, 2013). The measured particle sizes varied between 22.15 and 27.16 μ m (Fig. 1a). The particle size of the RCC chocolate samples was less than 35 μ m, which is the appropriate range for chocolate(Achaw & Danso-Boateng, 2021). The regression equation for the D90 parameter is expressed in eq. (1). A presents RCC, B presents Sugar and C presents

$$\label{eq:D90} \text{D90} = 27.14 \text{ A} + 22.24 \text{B} + 25.71 \text{C} + 0.992 \text{AB} + 0.977 \text{ AC} - 0.007 \text{ BCE} \tag{1}$$

The highest D90 value was at the maximum RCC level, and the lowest value was at the maximum sugar content. The results showed that with increasing RCC content in the formulation, the particle size of RCC chocolate increased (Fig. 1). The impact of RCC on particle size might result in other quality parameters. For instance, increasing particle size and particle size distribution can negatively affect sensory characteristics and cause a sandy texture. This problem reduces the flavor of chocolate. Puleo et al. (2020) also showed similar results by adding 8 % hazelnut paste in the formulation of cocoa creams(Puleo et al., 2020). Research showed that chocolate pastes with particles larger than 35 μm were felt as coarse or gritty, leading to a reduced overall acceptance. Additionally, Breen et al. (2019) examined the sensory attributes of dark chocolates produced with particle sizes (D90) varying from 19 μm to 33 $\mu m(Breen\ et\ al.,\ 2019)$. Consumers were able to detect differences in sensory characteristics even when the particle size of the chocolate changed by just about 5 µm. However, this problem can be solved by increasing and refining modifications during conching. For instance,

when using RCC in a ball mill, lengthening the processing time and decreasing the particle size may help reduce identified risks. Additionally, RCC can be used after its initial grinding to particle sizes smaller than 100 µm in advanced mills. Miele et al. (2019) investigated how milling affects particle size reduction of hazelnuts and cocoa, and their results showed that increasing milling time significantly reduced the particle size (D90) of the products(Miele et al., 2019). Moreover, Cavella et al. (2020) reported similar findings on the effect of ball milling on a mixture of white chocolate and hazelnuts, which resulted in a reduction in particle size and improved stability(Cavella et al., 2020).

3.2.2. Water activity of RCC chocolate

Moisture content ranged from 0.94 g/100 g to 1.26 g/100 g, which was in acceptable range (0.5–1.5 g/100 g)(Afoakwa, 2010). Higher levels of RCC significantly increased the moisture content in chocolate (Fig. 1b). The control chocolate showed an average moisture content of 0.995 g/100 g. The regression equation for moisture amount response was reported in Eq. (2).

Moisture content =
$$1.25 \text{ A} + 0.947 \text{B} + 1.08 \text{C} - 0.13 \text{AB} + 0.029 \text{ AC} - 0.116 \text{ BCE}$$
 (2)

As can be seen in Fig. 1b, an increase in RCC leads to a rise in moisture content, which may be associated with the fatty characteristics of RCC. A higher amount of RCC results in an increased fat content, thereby enhancing the hydrophobic behavior of chocolate. Since fat is unable to interact with water molecules, a greater number of water molecules remain in the chocolate, leading to an elevation in moisture content. Additionally, various factors may influence the moisture content of chocolate products, including types of chocolate(Rossini et al., 2011), processing methods(Carvalho et al., 2018)and conching temperature(Konar, 2013).

3.2.3. Rheological measurements of RCC chocolate

3.2.3.1. Casson plastic viscosity. The results showed that replacing sugar and cocoa with RCC did not significantly influence the fitting of the mathematical model, with the Casson model being the preferred choice for defining the rheological characteristics of chocolate. All chocolate samples showed shear-thinning behavior, confirming their classification as non-Newtonian fluids. The incorporation of RCC into the chocolate matrix resulted in an increase in Casson viscosity (Fig. 1c). The Casson viscosity values for the chocolate samples ranged from 3.77 to 5.07 Pa.s, with samples containing 100 % RCC demonstrating the highest viscosity, averaging 5.06 Pa.s. The viscosity of the chocolate samples was related to their D90 and moisture content. Additionally, smaller particles have more surface area, meaning more fat coats the solid particles, which can reduce viscosity(Afoakwa, Paterson, & Fowler, 2008). This may explain why the control samples showed lower viscosity (2.35 Pa. s), indicating a lower D90 (21.53 μm). The regression equation representing the response of Casson viscosity is provided in Eq. (3).

Casson viscosity =
$$5.08 \text{ A} + 3.75B + 4.61C - 0.2976AB + 0.979 \text{ AC} - 0.976 \text{ BCE}$$
 (3)

3.2.3.2. Casson yield stress. The Casson yield values for the chocolate samples ranged from 1.58 Pa.s to 6.91 Pa.s. Among the different chocolate formulations, those with 100 % RCC showed the highest yield stress, with an average of 6.89 Pa.s, while formulations with 100 % sugar showed the lowest yield stress, with an average 1.6 Pa.s (Fig. 1d). As the amounts of RCC and cocoa increased and the sugar content of the formulation decreased, the yield stress increased. The size of the RCC particles influenced the rheological properties of RCC samples. Increasing RCC resulted in difficulty in the flow of larger particles, which resulted in higher viscosity and yield stress. Additionally, due to the structure of RCC, there are a greater number of active and available

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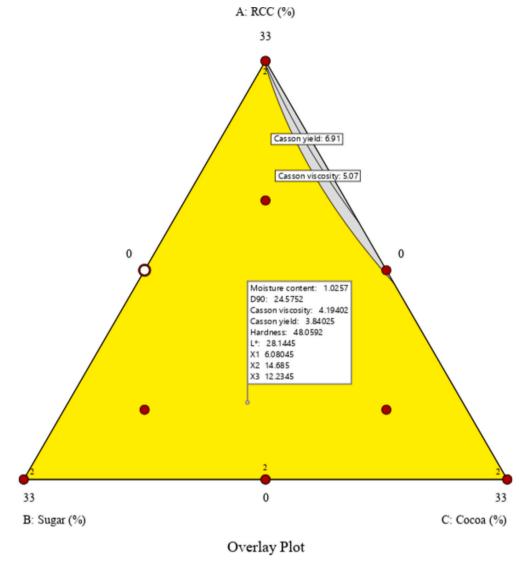


Fig. 2. Contours of estimated response surface demonstrating the point where optimum level was achieved. A = RCC; B = Sugar and C = Cocoa.

hydroxyl groups, which promote increased particle interactions and, consequently, a higher yield value. Moreover, two main factors influence the rheological properties of chocolate: 1) formulation (including particle size distribution and types and amounts of fat) and 2) processing techniques (like refining, conching, and tempering)(Toker, Zorlucan Demir, et al., 2016). Therefore, it is essential to balance formulation and processing techniques to achieve a high-quality product. The regression equation for the response of the Casson yield value is provided in Eq. (4).

Casson yield stress = 6.93~A + 1.55B + 5.46C -0.696AB +2.31~AC 1.52 BCE (4).

3.2.4. Hardness of RCC chocolate

The hardness measurements of the RCC chocolate samples varied from 41.68 N to 56.09 N (Fig. 1e). Higher levels of RCC contributed to an increase in the hardness of the chocolates. Formulations with 100 % RCC showed the greatest hardness, averaging 55.55 N, while the control samples had the lowest hardness, averaging 44.36 N. Research by Carvalho et al. (2018) showed that the addition of lyophilized grapes had a significant effect on the hardness of milk chocolate(Carvalho et al., 2018). Hardness, influenced by the types and amounts of fat, sugar, and cocoa solids in chocolate making(Barišić et al., 2021; Konar et al., 2013), is a crucial mechanical characteristic that impacts the sensory appeal of

chocolate(Hřivna et al., 2021; Machálková et al., 2015). Moreover, the variations in hardness may also be linked to the internal organization and increased intermolecular bonds in chocolate samples made with RCC. As RCC inclusion increases the fiber and carbohydrate content of the generated samples, these substances might interact with other components in chocolate, forming additional intermolecular bonds (Akdeniz et al., 2021). Similar to our results, Akdeniz et al. (2021) likewise indicated that incorporating LBF influences the hardness values of chocolates. The regression equation describing the hardness response is presented in Eq. (5).

$$Hardness = 55.53 \text{ A} + 49.84B + 42.21C$$
 (5)

3.2.5. Color of RCC chocolate

L* value of approximately 28.54 is considered acceptable for dark chocolate as reported by(Aidoo et al., 2013). The lower value of L* indicates a darker appearance. The L* values for the chocolate samples ranged from 27.5 to 30.11. Among the different chocolate formulations, those with 100 % sugar showed the highest L*, averaging 30.12, while formulations containing 100 % cocoa showed the lowest L*, averaging 27.5 (Fig. 1f). As the amounts of RCC and cocoa increased and the sugar content decreased in the formulations, the L* decreased. Typically, it has been reported that the chocolate sample with a greater concentration of

Table 2
Predicted and experimental results used in the validation of model based on optimum formulations (RCC = 6.08 g, Sugar = 14.684 g and Cocoa = 12.235 g).

	D90 (μm)	Moisture content (g/100 g)	caisson viscosity (Pa.s)	caisson yield stress (Pa)	Hardness (N)	L*
Predicted values Experimental values	$\begin{array}{l} 24.57 \pm 0.16^a \\ 25.20 \pm 0.27^b \end{array}$	$\begin{aligned} 1.02 &\pm 0.03^a \\ 1.04 &\pm 0.02^a \end{aligned}$	$\begin{array}{l} 4.19 \pm 0.09^a \\ 4.23 \pm 0.03^b \end{array}$	$\begin{array}{l} 3.84 \pm 0.05^a \\ 3.12 \pm 0.06^b \end{array}$	$\begin{array}{l} 48.05 \pm 0.26^{a} \\ 49.60 \pm 0.47^{b} \end{array}$	$\begin{array}{c} 28.14 \pm 0.22^{a} \\ 28.28 \pm 0.14^{b} \end{array}$

^{*} Mean of four determinations + SD. Different letters within columns indicate significant differences (p < 0.05).

sugar substitutes displayed a darker color(Aidoo et al., 2013; Shah et al., 2010). The regression equation for the response of the L* value is provided in Eq. (6).

$$L^* = 27.85 \text{ A} + 30.11 \text{B} + 27.45 \text{C} + 0.486 \text{AB} - 3.93 \text{ AC} - 2.04 \text{ BCE} \tag{6}$$

$$A = \text{RCC}; \ B = \text{Sugar and C} = \text{Cocoa}.$$

3.2.6. Optimization of the RCC chocolate formulation

After evaluating the quality parameters within the specified standard limits (D90, moisture content, caisson viscosity, yield stress, and hardness and L*), the formulation comprising 6.08 g of RCC, 14.684 g of Sugar and 12.235 g of Cocoa was identified as the highest desirability. The combination of factors that results in an overall desirability is shown in Fig. 2. According to the optimization results, the optimal ranges for D90, moisture content, caisson viscosity, caisson yield stress, hardness and L* which are very similar to the control sample, were determined to be 24.57, 1.02, 4.19, 3.84, 48.05 and 28.14 respectively (Fig. 2).

3.2.7. Model validation

The Design Expert software provided the optimal formula based on the selected parameters. The optimal sample was produced based on the optimal formulation, tested, and compared with the prediction model data. The tested values and results for the parameters are presented in Table 2. The experimental sample quality parameters are close to the values predicted by the model. These results confirm the model validation generated by the software.

Food manufacturers are increasingly looking for ways to effectively utilize unconventional plant-based raw materials, often underestimated as food ingredients. Moreover, it is important because the production of new products, the use of roasted coriander cake as a substitute for cocoa in chocolate production, meets the demands of consumers. However, the limitations of using coriander cake include the quality of the seeds, extraction methods, and subsequent processes like roasting, which affect the quality of the final product and the availability of nutrients. In addition, one of the limitations of large-scale production is that manufacturers need to ensure the amount of coriander cake as a cocoa substitute.

4. Conclusion

Coriander cake, as a by-product of coriander seed oil extraction, is highly nutritious and is commonly used for animal feed. Thus, incorporating coriander cake into chocolate formulation has significant benefits for both public health and economic development. After roasting, coriander cake was used as a cocoa substitute in the dark chocolate formulation. The first phase of this study aimed to determine the optimal roasting time to achieve a roasted coriander cake with physical and chemical properties similar to cocoa powder. Results showed that roasting coriander cake for 6 min at 165 °C, which leads to the Maillard reaction along with changes in color and aroma, makes it an appropriate substitute for cocoa powder. In the second phase, RCC was used as a substitute for sugar and cocoa powder in the RCC chocolate formulation, and 13 different experimental points were determined by mixture design. The effect of RCC on some physicochemical and rheological properties of RCC chocolate samples was investigated. The results showed that with increasing RCC content, caisson viscosity and yield stress increased. However, this slight increase did not cause a

significant change in quality. After evaluating the quality parameters within the specified standard limits (D90, moisture content, caisson viscosity, yield stress, and hardness and L*), the formulation comprising 6.08 g of RCC, 14.684 g of Sugar and 12.235 g of Cocoa was identified as the highest desirability. According to the optimization results, the optimal ranges for D90, moisture content, caisson viscosity, caisson yield stress, hardness and L* which very similar to the control sample, were determined to be 24.57, 1.02, 4.19, 3.84, 48.05 and 28.14 respectively. In conclusion, RCC can be used in chocolate as a partial replacement for cocoa powder and can enhance the functional properties of RCC chocolate and reduce product costs. This study contributes to further research on the valorization of RCC, thus helping to create strategies for developing innovative value-added chocolate formulations. Overall, the nutritional and financial benefits of CC make it a suitable alternative to cocoa. This study showed that by using some processing methods, the by-products could be valorized and help improve food security, which can be applied to other by-products.

Compliance with ethical standards

This article lacks research involving human or animal subjects.

CRediT authorship contribution statement

Javad Cheragheshahi: Conceptualization, Methodology, Investigation, Writing - review & editing. Elham Mahdian: Methodology, Investigation, Writing- review & editing. Sharareh Mohseni: Methodology, Investigation, Writing - review & editing. Ali Mohamadi Sani: Methodology, Writing - review & editing, Supervision. Zahra Nazari: Methodology, Writing - review & editing, Supervision.

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.

Data availability

The data that has been used is confidential.

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Further-reading

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