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Original research paper

COMPARISON OF DARK AND MILK COCOA TOPPINGS PRODUCED BY A FIVE-ROLL MILL AND A BALL MILL

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Abstract: Chocolate toppings are confectionery products comprised of non-fat dark cocoa solids and sugar particles dispersed in cocoa butter, as a continual matrix. On the other hand, dark cocoa toppings contain cocoa powder and sugar dispersed in vegetable fat that is cheaper than cocoa butter, but with adequate characteristics. Also, with the addition of a 7% non-fat milk fraction, this product is regarded as a milk cocoa topping. This kind of product, dark and milk cocoa toppings, does not demand long-term conching or tempering. This study aimed to determine and compare the impact of the production process on rheology, particle size distribution and content of moisture, fats, sucrose and lactose. Based on the rheology properties, it was found that the values of the Casson and linear viscosity of the samples produced using a five-roll mill and conching were lower compared to those produced using a ball mill. Regarding particle size distribution, the results showed that the volume-weighted mean parameter D (4,3) was lower for the milk and dark cocoa confectionery topping produced in the ball mill. Additionally, the values of the yield stress in samples produced in the ball mill were 2-fold higher for the milk cocoa topping and 4.5-fold higher for the dark cocoa topping compared with the values of the samples produced using the five-roll mill and conching. The near-infrared spectroscopy (NIRS) analysis did not show any significant difference among the samples. It can be concluded that the difference between the samples produced in the ball mill and the five-roll mill and conching exists and that the ball mill can be used for the production of high-quality cocoa toppings thus providing a shorter time of production.

Key words: *cocoa toppings, five roll mill, ball mill, rheology, particle size distribution*

INTRODUCTION

Chocolate and products similar to chocolate are popular confectionery products recognized and consumed worldwide. In 2020, the market for this type of product was estimated to be worth £6.7 billion in the UK, which is a £200 million increase compared to the report from

2019 (Euromonitor, 2019). Chocolate toppings are a mixture of dry powder ingredients such as sugar and cacao with cocoa butter, stabilizers and aromas (Rulebook, 2019a). Because of the increase in demand for this kind of product and the price of cocoa butter which repre-

sents a main fat phase in the chocolate topping, it was necessary to use cheaper vegetable fats, with specific properties that would minimally influence the quality of the produced toppings by decreasing it (Jahurul et al, 2013; Naik & Kumar, 2014). The alternative fats to cocoa butter were palm, palm kernel, and coconut oil, because of their fatty acid composition and properties similar to cocoa butter (Talbot, 2015). However, products similar to chocolate, like cocoa toppings were accepted by the consumers which opened the market for this type of products. The main difference between chocolate and cocoa toppings is in the selected fat phase and the content of cocoa solids (Lonchamp & Hartel, 2004). Dark cocoa toppings are a mixture of powdered sugar and cacao combined with cheaper vegetable fat, with a minimum of 4% fat-free cocoa particle dry matter and at least 25% of the total fat. Additionally, the second type of cocoa topping is milk cocoa topping which according to the Rulebook of the Republic of Serbia has to contain at least 4% fat-free cocoa solids, at least 25% total fat, and at least 7% milk non-fat solids (Rulebook, 2019b).

Cocoa toppings are mainly produced using a five-roll mill for grinding the mass and conching to develop a pleasant aroma and flavour. The optimal size of the ground particles can be achieved using the five-roll mill which consists of five smooth, steel rolls. The obtained results of the particle size values will depend on the selected values of the speed of the rollers and the thickness of the mass layer between the rollers.

The optimal particle size distribution is between 15 – 30 µm. The conching represents a process in which cocoa mass is mixed and homogenized for a long period to develop aromatic compounds and optimize the rheological properties of products (Beckett, 1999; Pajin, 2014, Lončarević et al., 2019). Since the process that includes using a five-roll mill and conching is long-lasting, it was necessary to find another process that would require less time for the production of cocoa toppings.

The ball mill is a one-part equipment used for the grinding and mixing of the cocoa mass, typically vertical or horizontal cylinders, equipped with a rotating shaft with arms centrally placed. About 60-80% of the available volume is filled with grinding media in the form of balls that can be produced out of steel,

ceramics, or some other material allowed for use in the food industry. The filling material was weighed and then placed in the grinding chamber to form a suspension and subsequently ground by compression and shear. Additionally, a ball mill must contain a system for recirculation of the mass to achieve the optimal values for the particle size. Cocoa mass passes through a layer of steel balls that rotate along a rotating shaft with arms (Alampresse, Datei & Semeraro, 2007; Pajin, 2014). A temperature control system (made up of a water jacket equipped with temperature sensors and thermostats controlled by an electric board) allows the initial melting of solid fats and ensures that the product does not suffer thermal damage, such as a burned aroma or milk derivative decay, and it performs the substitutive action of traditional conching (Ziegler & Hogg, 1999).

To date, no scientific literature sources have so far published any results that involve testing the influence of different milling technologies on the quality of produced cocoa toppings, only for fat fillings (Lončarević et al., 2017).

This study aims to examine and compare the obtained results of produced cocoa toppings, dark and milk, using different milling technologies regarding the particle size distribution, rheological properties, content of moisture, fat, sucrose, and lactose. Additionally, this study aimed to determine the impact on the quality of the produced samples and the possibility of replacing two-part equipment, the five-roll mill and conching with the one-part equipment, the ball mill.

MATERIALS AND METHODS

Material

Dark cocoa topping (DCT) consisted of sugar powder (54.47%), hydrogenated stearin from palm kernel as a fat phase (26.83%), cocoa powder (16.77%), lecithin and polyglycerol polyricinoleate (0.6%) and sorbitan tristearate (0.98%).

On the other hand, milk cocoa topping (MCT) consisted of sugar powder (50.89%), hydrogenated stearin from palm kernel (28.73%), milk powder (13.98%), vanillin (0.1%), lecithin and polyglycerol polyricinoleate (1.05%) and sorbitan tristearate (0.98%).

Ingredients were kindly provided by the Barry Callebaut industry.

Preparation of dark and milk cocoa topping

Laboratory samples, dark and milk cocoa toppings, were produced at the Barry Callebaut industry (Novi Sad, Serbia) under industrial conditions, using the five-roll mill and conching.

At first, dry conching lasted one hour at the temperature of 65 °C, and the next step was the wet conching which lasted for another 30 minutes at 55 °C.

The complete process lasted for 2.5 hours. Additionally, the analysed samples, dark and milk cocoa toppings were produced using a laboratory ball mill at a temperature of 60 °C for 1.5 hours with the maximum speed of the mixer which is an integral part of a ball mill. Ingredients were the same for both batches.

Determination of particle size distribution of dark and milk cocoa toppings

Particle size distribution (PSD) of produced toppings was determined using a Mastersizer 2000, laser diffraction particle size analyser (Malvern Instruments, England). Analysed samples were dispersed in sunflower oil, and the analysis was conducted using a Hydro 2000 μ P unit.

Gathered results were processed via Mastersizer 2000 software and shown as the volume-based PSD and described by PSD parameters: volume mean diameter $D[4,3]$ and parameters $d(0,1)$, $d(0,5)$, $d(0,9)$ that represent the particle sizes where 10, 50 or 90% of the total particle volume include particles that are smaller than that size.

Determination of rheological properties of dark and milk cocoa toppings

The equipment used to determine the rheological properties of cocoa toppings was a Rheo Stress 600 (Haake, Germany) rheometer, equipped with coaxial cylinders Z20DIN at 40 ± 1 °. First, the shear rate was increased from 0 s^{-1} to 60 s^{-1} in three minutes and kept constant for one minute at 60 s^{-1} . Then, the shear rate decreased from 60 s^{-1} to 0 s^{-1} in three minutes.

Determination of fat, moisture, sucrose and lactose contents using Near-Infrared Spectroscopy (NIRS)

The content of fat, moisture, sucrose and lactose was determined with a MPA II multi-

purpose analyser (Bruker, USA). The results are shown as a percentage values.

Statistical analysis

All experiments were rerun three times except for the sensory analysis which was conducted eight times. The gathered results were processed using a statistical method ANOVA and the means were compared by one-factor ANOVA with simultaneous comparisons by Duncan's test at a significance level of 0.05 using the Statistica 13.0 software.

RESULTS AND DISCUSSION

Rheological properties

The influence of different milling technologies on the rheological properties of produced cocoa toppings, dark and milk cocoa toppings is shown in Fig. 1 and Fig. 2. The measurement data showed the thixotropic flow of the samples. As shown in Fig. 1 different milling technology affected the values of yield stress, which relates to the energy required to initiate topping flow and is important in keeping solid particles in suspension and the coating of solid surfaces (Lončarević et al, 2021). The value for the dark cocoa topping (DCT) produced in a ball mill was 4.5-fold higher compared to the DCT sample produced using a five-roll mill and conching. Additionally, the sample of milk cocoa topping (MCT), produced with the five-roll mill and conching had the value of yield stress 2-fold lower (2900 mPa) as opposed to the values of MCT produced in a ball mill (5900 mPa).

Further, the results of the analysis of Casson and linear viscosity showed a significant difference ($p < 0.05$) between the samples (Fig. 2). In Fig. 2 (on the left), the samples DCT and MCT produced in a ball mill had significantly ($p < 0.05$) higher values of Casson viscosity, 3020 and 2020 mPa·s, compared to the DCT and MCT produced with different milling technology (2360 and 1520 mPa, respectively).

The same trend was present regarding the values of linear viscosity in the examined samples (Fig. 2, on the right). The increase in the viscosity, same Casson and linear, can be explained by the presence of the smaller particles in the samples that are produced using a ball mill, since the smaller particles have greater surfaces which, as a consequence, require increased amounts of fat to cover them

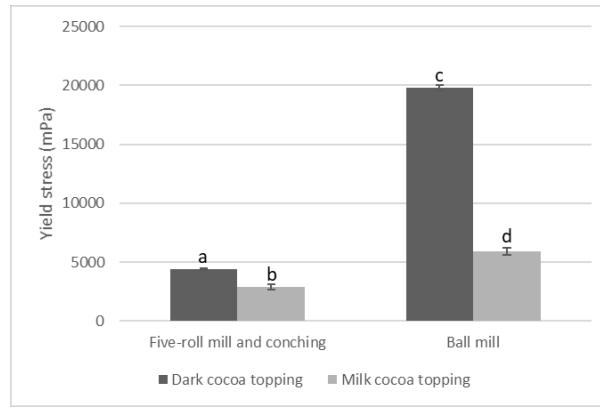


Figure 1. Yield stress in DCT and MCT samples produced using a five-roll mill and conching and a ball mill

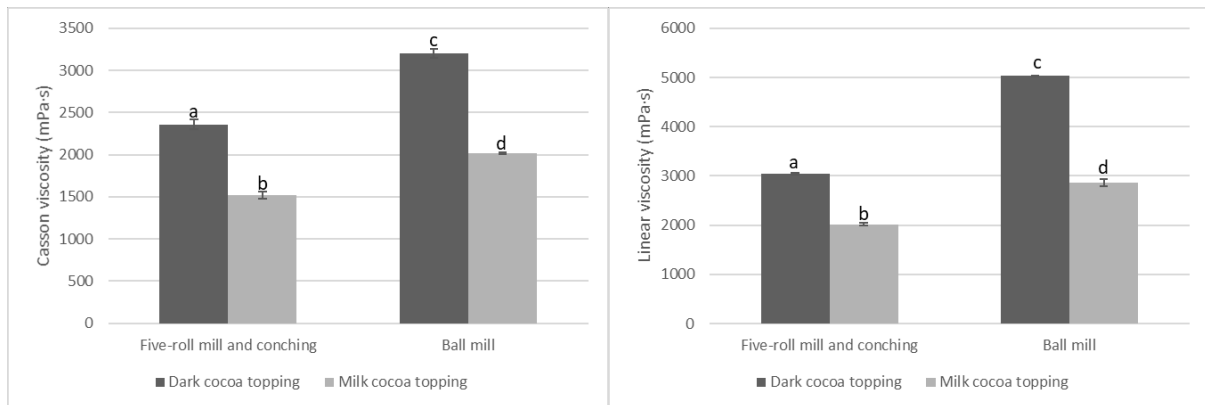


Figure 2. Casson (left) and linear (right) viscosity in DCT and MCT samples produced using a five-roll mill and conching and a ball mill

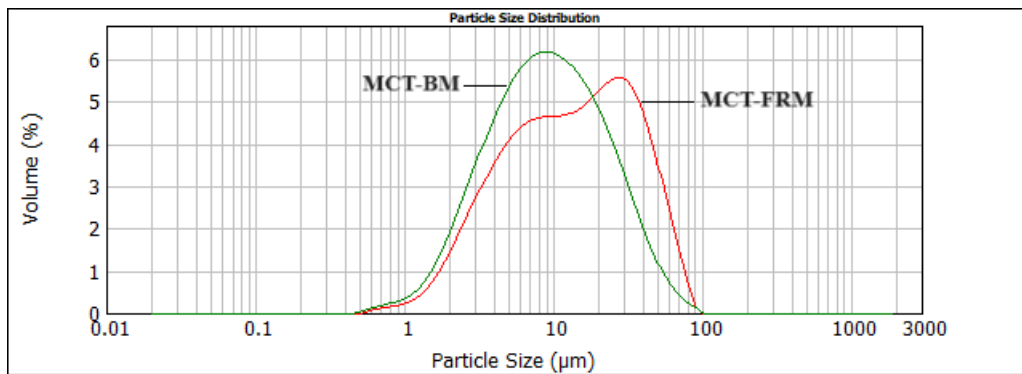


Figure 3. Particle size distribution in MCT sample produced with a ball mill (MCT-BM) and five-roll mill and conching (MCT-FRM)

(Lončarević et al., 2018).

Influence of the milling technologies on the particle size distribution

The obtained results of the influence of the different milling technologies on the particle size distribution in produced samples are presented in Figs. 3 and 4. First, the analysis was conducted on MCT samples, and the presented

curves are characteristic for samples refined by a five-roll mill and a ball mill (Lončarević et al., 2019).

As shown, the milk cocoa topping produced in a ball mill (MCT-BM) had smaller values (13.082 µm) for the mean volume-weighted parameter compared to the MCT produced with a five-roll mill and conching process (MCT-FRM).

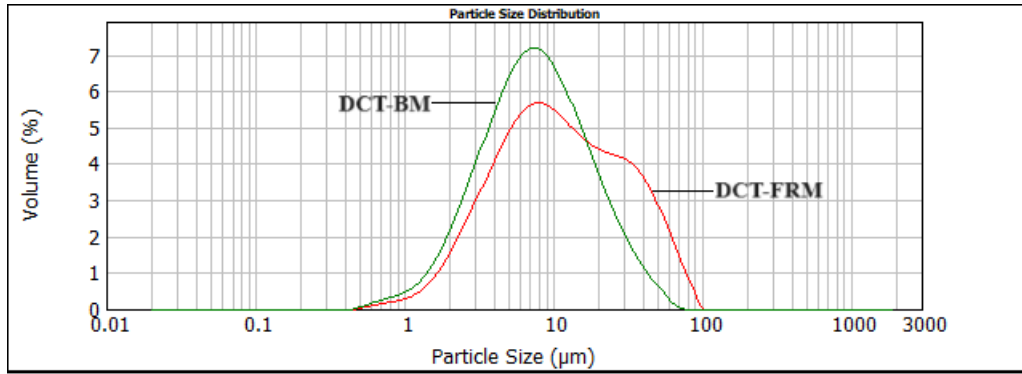


Figure 4. Particle size distribution in DCT sample produced with a ball mill (DCT-BM) and five-roll mill and conching (DCT-FRM)

Table 1.

Particle size parameters of milk (MCT) and dark (DCT) cocoa topping produced using a ball mill (BM) and five-roll mill and conching (FRM)

Cocoa toppings	Particle size parameters (µm)			
	d(0.1)	d(0.5)	d(0.9)	D[4,3]
MCT-BM	2.739±0.057 ^a	9.074 ±0.134 ^a	29.057±0.377 ^a	13.082±0.156 ^a
MCT-FRM	3.222 ±0.017 ^b	13.550±0.077 ^b	44.502±0.182 ^b	19.384±0.074 ^b
DCT-BM	2.540±0.012 ^c	7.581±0.032 ^c	22.720±0.227 ^c	10.608±0.144 ^c
DCT-FRM	3.058±0.075 ^d	10.756±0.297 ^d	42.672±0.842 ^d	17.486±0.609 ^d

Values represent the average of triplicates ± SD. Means with different letters in superscript in columns are significantly different between cocoa topping samples ($p < 0.05$)

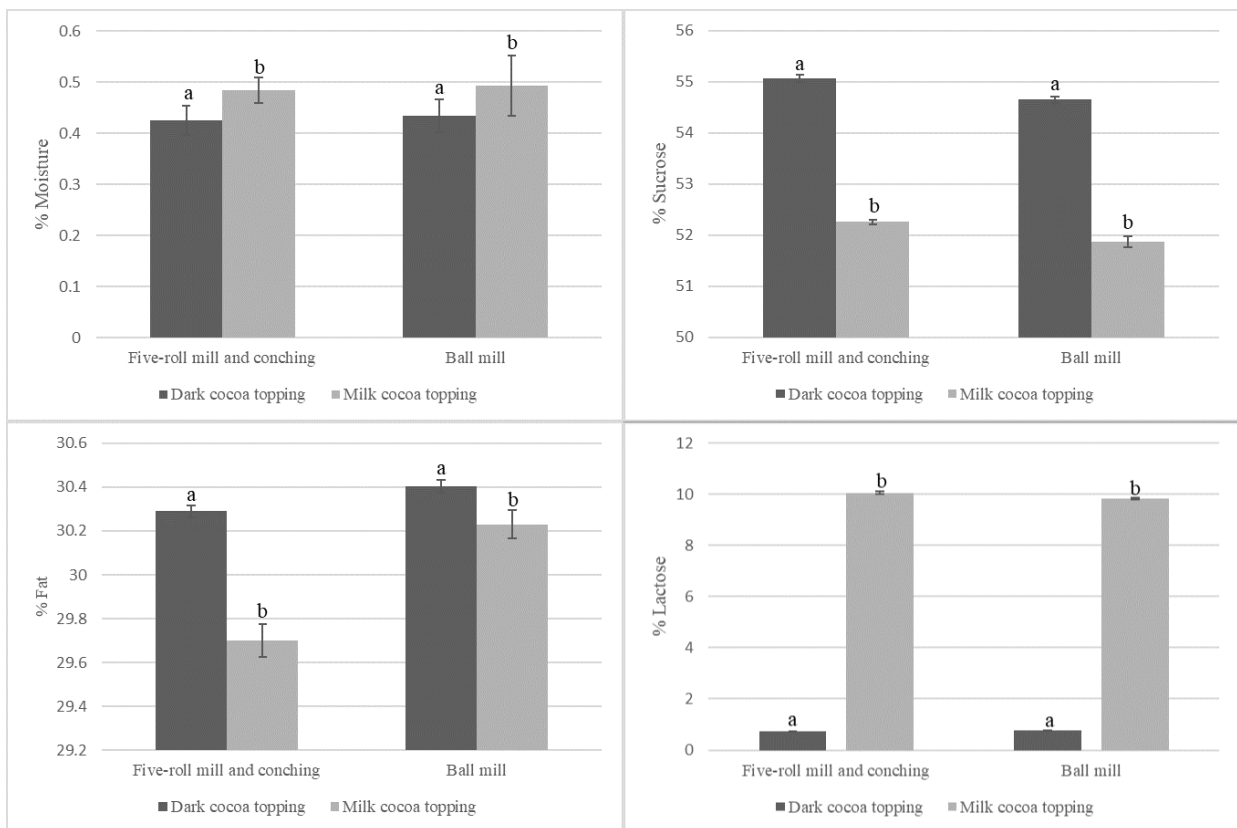


Figure 5. Results of NIR spectroscopy analysis of moisture, fat, sucrose and lactose content in produced dark and milk cocoa toppings

Additionally, the value of parameter $d(0,9)$ for MCT-BM was 29.097 μm , whereas for MCT-FRM, this value was 44.052 μm . The parameter $d(0,9)$ implies that 90% of the total particle volume is smaller than the value of this parameter. Also, a significant difference ($p < 0.05$) was present among the parameters $d(0,1)$ and $d(0,5)$. Refer to Table 1 for the previously mentioned data.

Furthermore, the same analysis was conducted for the dark cocoa toppings produced using two milling technologies, and measurement data are presented in Fig. 4 and Table 1. The results showed a significant difference in the values of examined parameters. The DCT-BM sample had lower values for parameters $D[4,3]$ and $d(0,9)$ when compared to the DTC-FRM sample (Table 1). and rheology analysis data are in agreement. The optimal interval for particle size distribution is between 15 – 30 μm . According to the collected data, the produced samples using a five-roll mill and conching had values between this interval. The toppings produced in a ball mill had particle size distribution lower than optimal. Since the MCT-BM and DCT-BM samples had lower values for the parameter $D[4,3]$, the increase in Casson and linear viscosities was expected in these samples, and the particle size distribution

NIR spectroscopy

Near-infrared spectroscopy (NIR spectroscopy) is a non-destructive assessment technique which uses infrared electromagnetic wavelengths to gather information about the sample composition (Johnson, Walsh, Naiker & Ameer, 2023). In confectionery products, NIR is generally used for quantitative measurements of key parameters such as fat, sugar, protein, and moisture content (da Costa Filho, 2009; Amorim, Duarte, de Oliveira, de la Fuente & Gómez-Cortés, 2020). The NIR spectroscopy analysis data are presented in Fig. 5. The chemical composition of toppings produced by various milling processes is consistent, except for moisture content and the levels of volatile compounds.

CONCLUSIONS

The study investigated the effects of different milling technologies on the quality of produced samples of dark and milk cocoa toppings. Toppings were produced using a ball mill and a five-roll mill with conching.

The results showed that:

- Samples produced in a ball mill had an increase in linear and Casson viscosity, leading to a 4.5-fold increase in yield stress of DCT-BM, and a 2-fold increase for MCT-BM sample. Additionally, the Casson viscosity for DCT-BM increased 1.28 fold and for the MCT-BM sample, the increase was 1.33 fold. The same trend was present regarding the values of linear viscosity.
- Particle size distribution analysis showed a significant ($p < 0.05$) decrease in mean volume-weighted parameter $D[4,3]$ in samples produced using a ball mill compared to the samples produced with a five-roll refiner and conching. Since the presence of smaller particles influences the viscosity by increasing it, these results coincide with rheology analysis data.

In summary, different milling technology slightly influenced the quality of produced samples. The viscosity of the samples produced in a ball mill was higher than the compared sample, but not to the point where this increase will be regarded as negative during the covering of the product.

Presumably, this increase can be lowered by shortening the production time, thus impacting the particle size distribution and refining the particles to the optimal range. Overall, the quality of the produced samples is intact and it can be concluded that the ball mill can be used for the production of this type of confectionery product, simultaneously saving time and energy, compared to the different milling process that includes five-roll mill and conching.

AUTHOR CONTRIBUTIONS

Conceptualization, L.I.; Methodology, Z.D., P.J. and N.I.; Investigation, formal analysis, validation, S.M and P.T.; writing-original draft preparation, S.M.; Writing-review and editing, L.I. and S.M.; Supervision, P.B.

DATA AVAILABILITY STATEMENT

Data contained within the article.

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CONFLICT OF INTEREST

The authors declare no conflict of interest. The funders had no role in the design of the study; in the collection, analyses, or interpretation of data; in the writing of the manuscript, or in the decision to publish the results.

REFERENCES

- Amorim, T. L., Duarte, L. M., de Oliveira, M. A. L., de La Fuente, M. A., & Gómez-Cortés, P. (2020). Prediction of fatty acids in chocolates with an emphasis on C18: 1 trans fatty acid positional isomers using ATR-FTIR associated with multivariate calibration. *Journal of Agricultural and Food Chemistry*, 68(39), 10893-10901. <https://doi.org/10.1021/acs.jafc.0c04316>
- Beckett, S. T. (Ed.). (2011). *Industrial chocolate manufacture and use*. John Wiley & Sons.
- Alamprese, C., Datei, L., & Semeraro, Q. (2007). Optimization of processing parameters of a ball mill refiner for chocolate. *Journal of Food Engineering*, 83(4) (2007), pp. 629-636. <https://doi.org/10.1016/j.jfoodeng.2007.04.014>
- da Costa Filho, P. A. (2009). Rapid determination of sucrose in chocolate mass using near-infrared spectroscopy. *Analytica Chimica Acta*, 631(2), 206-211. <https://doi.org/10.1016/j.aca.2008.10.049>
- Euromonitor, (2019). Chocolate Confectionery in the United Kingdom [Report]. London: Euromonitor International. <https://www.euromonitor.com/chocolate-confectionery-in-the-united-kingdom/report>
- IOCCC (2000). Viscosity of cocoa and chocolate products. Analytical Method 46- 2000. Bruxelles, Belgium: International Office of Cocoa, Chocolate and Sugar Confectionery.
- Jahurul, M. H. A., Zaidul, I. S. M., Norulaini, N. A. N., Sahena, F., Jinap, S., Azmir, J., ... & Omar, A. M. (2013). Cocoa butter fats and possibilities of substitution in food products concerning cocoa varieties, alternative sources, extraction methods, composition, and characteristics. *Journal of Food Engineering*, 117(4), 467-476. <https://doi.org/10.1016/j.jfoodeng.2012.09.024>
- Johnson, J. B., Walsh, K. B., Naiker, M., & Ameer, K. (2023). The use of infrared spectroscopy for the quantification of bioactive compounds in food: A review. *Molecules*, 28(7), 3215. <https://doi.org/10.3390/molecules28073215>
- Lončarević, I., Fišteš, A., Rakić, D. Z., Pajin, B., Petrović, J., Torbica, A., & Zarić, D. B. (2017). Optimization of the ball mill processing parameters in the fat filling production. *Chemical Industry and Chemical Engineering Quarterly (CICEQ)*, 23(2), 197-206. <https://doi.org/10.2298/CICEQ151217031L>
- Lončarević, I., Pajin, B., Fišteš, A., Šaponjac, V. T., Petrović, J., Jovanović, P., ... & Zarić, D. (2018). Enrichment of white chocolate with blackberry juice encapsulate: Impact on physical properties, sensory characteristics and polyphenol content. *LWT*, 92, 458-464. <https://doi.org/10.1016/j.lwt.2018.03.002>
- Lončarević, I., Pajin, B., Petrović, J., Nikolić, I., Maravić, N., Ačkar, Đ., ... & Miličević, B. (2021). White chocolate with resistant starch: Impact on physical properties, dietary fiber content and sensory characteristics. *Molecules*, 26(19), 5908. <https://doi.org/10.3390/molecules26195908>
- Lončarević, I., Pajin, B., Tumbas Šaponjac, V., Petrović, J., Vulić, J., Fišteš, A., & Jovanović, P. (2019). Physical, sensorial and bioactive characteristics of white chocolate with encapsulated green tea extract. *Journal of the Science of Food and Agriculture*, 99(13), 5834-5841. <https://doi.org/10.1002/jsfa.9855>
- Lonchamp, P., & Hartel, R. W. (2004). Fat bloom in chocolate and compound coatings. *European Journal of Lipid Science and Technology*, 106(4), 241-274. <https://doi.org/10.1002/ejlt.200400938>
- Naik, B., & Kumar, V. (2014). Cocoa butter and its alternatives: A review. *Journal of Bioresource Engineering and Technology*, 2(1), 1-11. <https://api.semanticscholar.org/CorpusID:43231010>
- Pajin, B. (2014). *Tehnologija čokolade i kakao proizvoda*. Novi Sad, Srbija: Univerzitet u Novom Sadu, Tehnološki fakultet.
- Rulebook. (2019a). Pravilnik o kakao i čokoladnim proizvodima namenjenim za ljudsku upotrebu (2019/2024) / Rulebook on cocoa and chocolate products for human consumption/. *Službeni glasnik RS*, 24/2019, 18/2024.
- Rulebook. (2019b). Pravilnik o proizvodima sličnim čokoladi, krem proizvodima i bombonskim proizvodima (2019/2024) / Rulebook on products similar to chocolate, cream products and candy products/. *Službeni glasnik RS*, 24/2019.
- Talbot, G. (2015). Specialty oils and fats in confectionery. In G. Talbot (Ed.), *Specialty oils and fats in food and nutrition* (pp. 221-239). Woodhead Publishing. <https://doi.org/10.1016/B978-1-78242-376-8.00009-0>
- Ziegler, G. R., & Hogg, R. (1999). Particle size reduction. In S. T. Beckett (Ed.), *Industrial chocolate manufacture and use* (3rd ed.). Oxford, UK: Blackwell Science. <https://onlinelibrary.wiley.com/doi/book/10.1002/9781444301588>

POREĐENJE TAMNIH I MLEČNIH KAKAO PRELIVA PROIZVEDENIH UZ UPOTREBU PETOVALJKA I KUGLIČNOG MLINA

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Sažetak: Čokoladni prelive su konditorski proizvodi koji se sastoje od nemasnih tamnih kakao delova i čestica šećera dispergovanih u kakao puteru, kao kontinualnoj fazi. S druge strane, tamni kakao prelive sadrže kakao prah i šećer dispergovan u biljnoj masti koja je jeftinija od kakao putera, ali sa adekvatnim karakteristikama. Takođe, sa dodatkom 7% nemasne frakcije mleka, ovaj proizvod se smatra mlečnim kakao prelivom. Ovakvi proizvodi (tamni i mlečni kakao preliv) ne zahtevaju dugotrajno končiranje ili temperiranje. Ovaj rad je imao za cilj da utvrdi i uporedi uticaj procesa proizvodnje na reologiju, distribuciju veličine čestica i sadržaj vlage, masti, saharoze i laktoze u kakao prelivima. Na osnovu reoloških svojstava, utvrđeno je da su vrednosti Casson i linearnog viskoziteta uzoraka proizvedenih u mlinu sa pet valjaka i uz končiranje bile niže u odnosu na one vrednosti viskoziteta uzoraka proizvedenih u kugličnom mlinu. Što se tiče distribucije veličine čestica, rezultati su pokazali da je zapreminsko-maseni srednji parametar D (4,3) bio manji za mlečni i tamni kakao preliv, proizveden u kugličnom mlinu. Pored toga, vrednosti prinosnog napona u uzorcima proizvedenim u kugličnom mlinu bile su 2 puta veće za mlečni kakao preliv i 4,5 puta više za tamni kakao preliv u poređenju sa vrednostima uzoraka proizvedenih upotrebom petovaljka i končiranja. Analiza bliske infracrvene spektroskopije (NIRS) nije pokazala značajnu razliku u sadržaju masti, saharoze i laktoze između uzoraka. Može se zaključiti da razlika između uzoraka proizvedenih u kugličnom mlinu i petovaljku postoji i da se kuglični mlin može koristiti za proizvodnju visokokvalitetnih kakao prelive, čime se obezbeđuje kraće vreme proizvodnje.

Ključne reči: kakao prelive, petovaljak, kuglični mlin, reologija, distribucija veličine čestica

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