Colour changes of extrudates from wheat semolina with added cocoa bean shells

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Abstract. Response surface methodology was employed to evaluate the influence of cocoa bean shells content (5 % and 10 %), moisture content (14 % and 20 %) and temperature of the matrix (160 °C and 180 °C) on the colour characteristics of extruded products from wheat semolina with added cocoa bean shells. The extrusion cooking was performed using a Brabender 20 DN single-screw extruder. A NS800 spectrophotometer was used to determine the lightness (L), redness (a), yellowness (b) and total colour difference (ΔE) of extrudates. The extrusion process resulted in a noticeable darkening of the samples relative to the non-extruded mixtures. The average values of lightness varied from 60.68 ± 0.81 to 69.04 ± 0.81, redness from 7.11 ± 0.10 to 8.20 ± 0.25, yellowness changed from 15.29 ± 0.19 to 16.92 ± 0.48 and for the total colour difference from 2.26 ± 0.17 to 4.08 ± 0.10, respectively. The moisture content exerted the greatest influence on the total colour difference, followed by the temperature of the matrix, whereas the cocoa bean shells content had no significant effect.

1 Introduction

The cocoa bean, widely utilized in chocolate production, consists of two primary components: the nib and the shell (Fig. 1). While cocoa nibs serve as the main raw material for chocolate manufacturing, the cocoa bean shell constituting approximately 15 % of the whole bean is typically discarded as a by-product [1]. The cocoa bean shells contain a range of bioactive and nutritional components including vitamins, antioxidants, phenolic compounds, lipids, carbohydrates, and dietary fiber [2]. The high antioxidant activity and the presence of valuable substances make them a promising ingredient for a wide range of food applications [3, 4].

Fig. 1. Cocoa fruit components [3].

In recent times, the growing accumulation of waste and by-products from the agricultural and food sectors has posed serious environmental and economic concerns, primarily as a result of pollution [5, 6]. Cocoa bean shells, often considered as waste, represent a significant economic and environmental opportunity for renewable resource utilization [7]. The valorization of waste and by-products contributes to reducing their negative environmental effects and supports the formulation of food products with improved sensory quality, physicochemical stability, and nutritional value [8]. The rising consumer interest in healthy and sustainable foods is creating new opportunities for products enriched with cocoa bean shells [2].

Extrusion cooking is a widely utilized and efficient food processing method, classified as a high-temperature, short-time (HTST) technique, for producing a diverse range of food products [9, 10]. It combines multiple unit operations including mixing, heating, shearing, and forming into a single, continuous process, enabling the transformation of raw materials into structured foods with desirable texture and extended shelf life [11, 12]. The growing consumer awareness of the importance of nutritious and functional foods is prompting manufacturers to enhance their products with various health-promoting components. In this context, the integration of extrusion technology with the addition of cocoa shells offers a promising approach for the development of functional food products [13].

Colour accompanied by flavor and texture is a crucial attribute of food quality, significantly influencing the consumer perception and acceptance [14]. The customers primarily associates the quality of food products with their appearance and color; therefore, color is a key indicator in the evaluation of extruded products [15]. Despite being a high-temperature, short-time processing method, extrusion inevitably leads to changes in the color of the resulting products. These

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color changes in extrudates can be partially controlled through the formulation of the raw material blends and by adjusting processing parameters such as moisture content, temperature, screw speed, and others [16, 17]

The present study was conducted to evaluate the influence of cocoa bean shells content, moisture content and temperature of the matrix on the colour characteristics of extruded products.

2 Materials and methods

2.1 Materials

Cocoa bean shells. The experimental investigations were carried out using cocoa bean shells, as illustrated in Fig. 1. These shells were a by-product of chocolate manufacturing and were generously supplied by 'Gaillot Chocolate', Plovdiv. The average chemical composition of the cocoa bean shells was as follows: moisture content – 10 %, proteins – 14 %, carbohydrates – 45 %, fats – 3 %, and theobromine – 1.2 %.

Wheat semolina. Wheat semolina with an initial moisture content of 13 % was collected from the Dimitar Pilev Mill Complex in Konush village. It was mixed with cocoa bean shells in set amounts, and water was added to reach the target moisture levels shown in Table 1.

Table 1. The experimental design with natural and coded values.

№	Natural values			Coded values		
	Cocoa bean shells content, %	Moisture content, %	Temperature of the matrix, ºC	X_1	X_2	X_3
1	5	14	180	-1	-1	+1
2	5	20	160	-1	+1	-1
3	5	20	180	-1	+1	+1
4	10	20	160	+1	+1	-1
5	5	14	160	-1	-1	-1
6	10	20	180	+1	+1	+1
7	10	14	180	+1	-1	+1
8	10	14	160	+1	-1	-1

2.2 Extrusion processing

A Brabender 20 DN single-screw laboratory extruder [18] was employed to conduct the extrusion process, using a range of processing settings as detailed in Table 1. The following conditions were kept constant throughout the experiment: nozzle diameter – 3 mm; screw compression ratio – 3:1; extruder screw speed – 200 min^{-1}; feeding screw speed – 30 min^{-1}; and temperatures in the first and second extruder zones 140 °C and 150 °C, respectively.

2.3 Analysis methods

2.3.1 Statistical processing

A full factorial experimental design (N = 2^3) was employed during extrusion processing. The explanatory variables were cocoa bean shell content (X_1), moisture content (X_2) and temperature of the matrix (X_3). The experimental design, including both the natural and coded values of these three factors is detailed in Table 1. The variation levels of the factors were selected based on preliminary studies and literature data [19]. Each experimental condition had three replicates.

A model employing a linear regression with interaction terms, using standardized or scaled independent variables, was utilized to represent the relationships between the factors:

$$y = b_0 + \sum_{i=1}^{n} b_i X_i + \sum_{i=1}^{n} \sum_{j=1}^{n} b_{ij} X_i X_j \quad (1)$$

where: b_o, b_i и b_{ij} were a free coefficient, coefficient of linear effect and coefficient of interaction, respectively.

Fisher's F-statistic was used to assess the adequacy of the models. All statistical analyses were conducted using the software 'Statgraphics XVII Centurion' (trial version).

2.3.2 Assessment of colour characteristics

The extrudates were ground using a laboratory grinder to a particle size of 200 µm. Their colour was then measured instrumentally with an NS800 spectrophotometer (3nh, China), following the CIE Lab color space system. In this system: *L* represents lightness (*L* = 0 for black, *L* = 100 for white), +*a* indicates red, -*a* indicates green, +*b* indicates yellow, and -*b* indicates blue. The total colour difference (Δ*E*) was estimated using the formula given below:

$$\Delta E = \sqrt{(L - L_0)^2 + (a - a_0)^2 + (b - b_0)^2} \quad (2)$$

where: *L*, *a* и *b* were the values of the extruded samples and L_o, a_o и b_o were the values of the non-extruded mixtures (Table 2).

Table 2. Colour parameters of non-extruded mixtures.

Cocoa bean shells content (%)	L_o	a_o	b_o
5	69.12	5.73	14.50
10	65.13	6.62	14.72

3 Results and discussion

The mean values and standard deviations for colour characteristics – lightness (*L*), redness (*a*), yellowness (*b*) and total colour difference (Δ*E)* depending on the three tested factors – cocoa bean shells content (X_1), moisture content (X_2) and temperature of the matrix (X_3) are shown in Table 3.

Table 3. Colour changes of extrudates.

№	*L*	*a*	*b*	Δ*E*
1	69.04 ± 0.81*	7.11 ± 0.10*	15.48 ± 0.88*	3.37 ± 0.12*
2	68.00 ± 0.57	7.57 ± 0.39	16.71 ± 0.28	3.43 ± 0.23
3	66.75 ± 1.14	7.53 ± 0.36	15.92 ± 0.67	2.40 ± 0.21
4	62.08 ± 0.20	7.37 ± 0.31	15.29 ± 0.19	2.28 ± 0.19
5	66.87 ± 0.79	7.59 ± 0.34	16.52 ± 0.49	2.83 ± 0.22
6	60.68 ± 0.81	8.20 ± 0.25	16.22 ± 0.62	4.08 ± 0.10
7	65.36 ± 1.34	7.89 ± 0.68	16.18 ±0.74	2.26 ± 0.17
8	63.51 ± 0.92	8.13 ± 0.12	16.92 ± 0.48	2.73 ± 0.48

*Standard deviation based on three-fold repeatability

The colour attribute values of the extrudates range from 60.68 to 69.04 for lightness (L), from 7.11 to 8.20 for redness (a), from 15.29 to 16.92 for yellowness (b), and from 2.26 to 4.08 for the total colour difference (ΔE). The extruded samples displayed a darker colour compared to the non-extruded blends, as evidenced by their lower lightness (L) values (Table 3) relative to those of the non-extruded mixtures (L_0) (Table 2). Similar results were obtained by [20] according to which the addition of pumpkin flour at 10 % and 20 % concentrations in the preparation of rice flour extrudates resulted in their darker colour compared to the controls due to the Mayard reactions taking place.

The follow adequate models at confidence interval 95 % with significant coefficients were obtained:

$$L = 65.287 - 2.379X_1 - 0.908X_2 - 0.619X_1X_2 - 0.833X_2X_3 \quad (3)$$

$$R^2 = 93.52\ \% \quad F = 0.11 < F_c = 3.2$$

$$a = 7.674 + 0.225X_1 + 0.1896X_2X_3 \quad (4)$$

$$R^2 = 41.48\ \% \quad F = 0.45 < F_c = 2.9$$

$$\Delta E = 2.924 + 0.125X_2 + 0.105X_3 + 0.216X_1X_2 + 0.226X_1X_3 + 0.48X_1X_2X_3 \quad (5)$$

$$R^2 = 87.07\ \% \quad F = 1.02 < F_c = 3.6$$

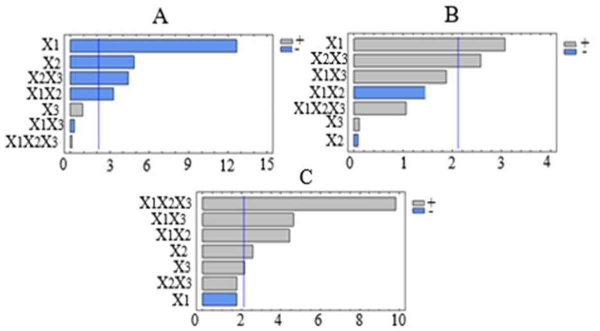

Fig. 2. Pareto charts: A – lightness (L); B – redness (a); C – the total colour difference (ΔE).

The standardized Pareto charts show the influence of cocoa bean shells content (X_1), moisture content (X_2) and temperature of the matrix (X_3) on the lightness (Fig. 2 A), redness (Fig. 2 B) and total colour difference (Fig. 2 C) of extrudates. According to the lightness (Fig. 2 A) the factors cocoa bean shells content (X_1), moisture content (X_2) as well as the factor interactions (X_2X_3) and (X_1X_2) are significant. The biggest negative effect has the factor cocoa bean shells content (X_1), followed by the moisture content (X_2). This tendency is verified by the obtained equation coefficients – 2.379 for (X_1) and 0.908 for (X_2). As for the redness (Fig. 2 B) the data indicate that only factor (X_1) and factor interactions (X_2X_3) are significant. The both examined factors have a positive effect on the redness. With regard to the total colour difference (Fig. 2 C) it is clearly that both - the single factors and the interactions between factors are significant, with the exception of factor (X_1) and (X_2X_3). Among the single factors, the greatest positive influence is exerted by factor (X_2), followed by factor (X_3) within the factor interactions, the most significant is ($X_1X_2X_3$), then (X_1X_3) and (X_1X_2). These dependencies are confirmed by the obtained coefficients in the equation 5.

The influence of cocoa bean shell content (X_1), moisture content (X_2) and temperature of the matrix (X_3) on lightness (L) is shown in Fig. 3. The lowest value of lightness (60.68) was recorded for extrudates containing 10 % cocoa bean shells, 20 % moisture content and 180 °C temperature of the matrix while the highest value of (69.04) was observed at 5 % cocoa bean shells, 14 % moisture content and the same temparature of the matrix, respectively. The darkening of colour with increasing cocoa bean shells content could be explained by the melanins they contain, which are susceptible to oxidation and this leads to colour change [21].

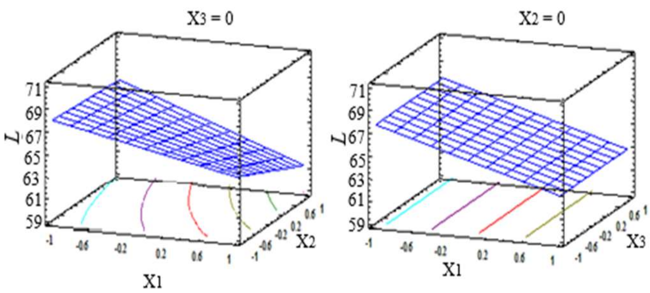

Fig. 3. Response surface of the lightness (L), depending on the cocoa bean shells content (X_1), moisture content (X_2) and temperature of the matrix (X_3).

The increase of moisture content lead to a decrease of the lightness (L) of extrudates which is more pronounced at a higher level of the factor temperature of the matrix (Fig. 4).

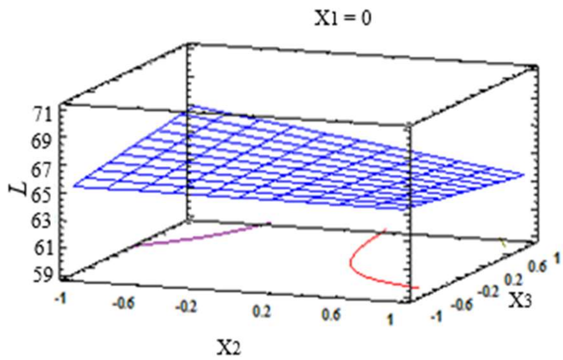

Fig. 4. Response surface of the lightness (L), depending on the moisture content (X_2) and temperature of the matrix (X_3).

The obtained results are consistent with those reported by [22]. The authors extruded products from three types of legumes with varying moisture content ranging from 20 % to 33 %, using a laboratory-scale Wenger X-5 extruder at temperatures of 110 ºC, 121 ºC, 132 ºC and 150 ºC, with a screw speed of 700 min^{-1} and a nozzle diameter of 3.5 mm. They established that the colour of the obtained extrudates depends on the initial moisture content of the mixture and the values of the lightness decrease with the increase of moisture content.

The influence of the three examined factors on the redness (a) of extrudates is illustrated in Fig. 5 and Fig. 6. As shown in Fig. 5, the redness values increase with higher cocoa bean shells content, while changes in the other two parameters have no significant influence on redness. The observed increase in redness may be attributed to the presence of tannins and caffeine in the cocoa bean shells, as well as to Maillard reactions occurring between individual constituents involved in the extrusion process [20, 23].

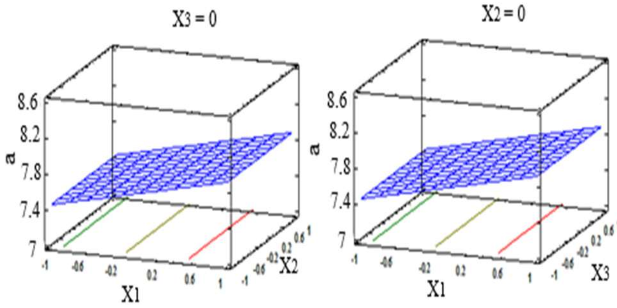

Fig. 5. Response surface of the redness (*a*), depending on the cocoa bean shells content (X_1), moisture content (X_2) and temperature of the matrix (X_3).

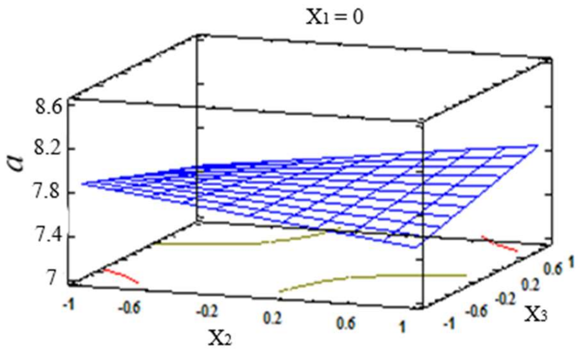

Fig. 6. Response surface of the redness (*a*), depending on the moisture content (X_2) and temperature of the matrix (X_3)

The response surface presented in Fig. 6 has a complex profile and it is of the Mini-Max'type. The highest values for redness (*a*) is observed at low level of factors moisture content and temperature of the matrix as well as at high levels of these two variables. Whereas the combination of high moisture content with low temperature, as well as low moisture content with high temperature, results in minimum redness (*a*) values.

The influence of cocoa bean shells content and moisture content at constant temperature of the matrix of 170 ºC on the total colour difference is presented in Fig. 7. It is clearly evident that the highest (Δ*E*) values are obtained at high levels of the two investigated factors while the lowest occur at 10 % cocoa bean shells content and 14 % moisture content. The lower values are an indicator of a lower reaction of the colour components in the cocoa bean shells. The moisture content significantly influences the extrusion process and the increase from 14 % to 20 % causing a more pronounced alteration in the colour of the extrudates. This is likely due to more pronounced chemical (oxidation, hydrolysis) or physical changes occurring in the cocoa components responsible for colour, as reported by Dushkova et al. (2023).

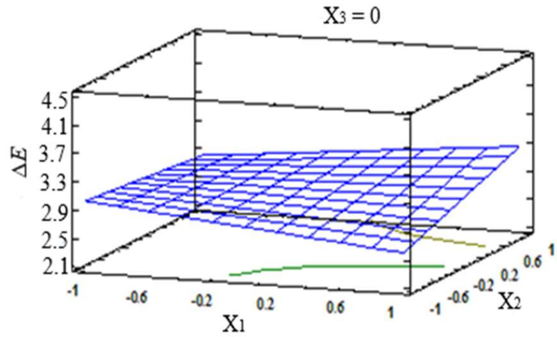

Fig. 7. Response surface of the total colour difference (Δ*E*), depending on the cocoa bean shells content (X_1) and moisture content (X_2).

The combined effect of cocoa bean shells content and temperature of the matrix at constant moisture content of 17 % on the total colour difference is shown in Fig. 8. The data indicate that the highest value of the total colour difference (Δ*E*) is obtained as the temperature of the matrix increases at the high level of factor cocoa bean shells content. Our results confirm the conclusion that extrusion temperature has a significant impact on the colour characteristics of extrudates [24]. The increase of the temperature increases the colour intensity, leading to a decrease in lightness (*L*) values and an increase in redness (*a*) and yellowness (*b*) values. At lower level of factor temperature of the matrix with the increase of the cocoa bean shells content the total colour difference (Δ*E*) values decrease which is probably due to the higher redness (*a*) values in the mixture with higher cocoa bean shells content. On the other hand the exruded products with higher redness (*a*) values are darker and exhibit lower lightness (*L*) and the total colour difference (Δ*E*) values (Table 3).

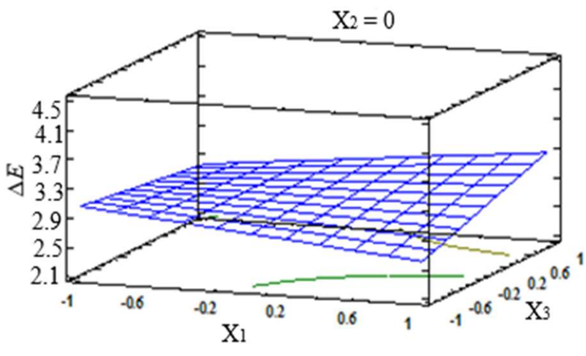

Fig. 8. Response surface of the total colour difference (Δ*E*), depending on the cocoa bean shells (X_1) and temperature of the matrix (X_3).

4 Conclusion

Reliable mathematical regression models were developed to investigate the impact of cocoa bean shell content (5 % and 10 %), moisture content (14 % and 20 %), temperature of the matrix (160 °C and 180 °C) on the colour characteristics of extruded products. The average lightness (*L*) values ranged from 60.68 ± 0.81 to 69.04 ± 0.81, redness (*a*) varied between 7.11 ± 0.10

and 8.20 ± 0.25 while yellowness (*b*) values lay between 15.29 ± 0.19 and 16.92 ± 0.48. The total colour difference (ΔE) was recorded within the interval of 2.26 ± 0.17 to 4.08 ± 0.10.

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References

1. J. Choi, N. Kim, H.Y. Choi, Y.S. Han, Effect of cocoa bean husk powder on the quality properties of pork sausages. Food Sci. Anim. Resour. **39**, 742 (2019) https://doi.org/10.5851/kosfa.2019.e62
2. F. Fetriyuna, M. Djali, A. Z. Rafi, D.A. Nurunnisa, R.C. Purwestri, Cocoa bean shells: A potential chocolate replacement in food production. Int. J. Adv. Sci. Eng. Inf. Technol. **15**, 147 (2025) https://doi.org/10.18517/ijaseit.15.1.20270
3. T.F. Soares, M.B.P.P. Oliveira, Cocoa by-products: Characterization of bioactive compounds and beneficial health effects. Molecules **27**, 1625 (2022) https://doi.org/10.3390/molecules27051625
4. A. Gil-Ramírez, S. Cañas, I.M. Cobeta, M. Rebollo-Hernanz, P. Rodríguez-Rodríguez, V. Benítez, S.M. Arribas, M.A. Martín-Cabrejas, Y. Aguilera, Uncovering cocoa shell as a safe bioactive food ingredient: Nutritional and toxicological breakthroughs. Futur. Food **10**, 100461 (2024) https://doi.org/10.1016/j.fufo.2024.100461
5. J.P. Balentić, Đ. Ačkar, S. Jokić, A. Jozinović, J. Babić, B. Miličević, D. Šubarić, N. Pavlović, Cocoa shell: A by-product with great potential for wide application. Molecules **23**, 1404 (2018) https://doi.org/10.3390/molecules23061404
6. T.A. Dendegh, B.M. Yelmi, M.J. Abdullahi, Extrusion technology: A tool for value addition to food by-products and wastes. Arch. Curr. Res. Int. **21**, 39 (2021) https://doi.org/10.9734/acri/2021/v21i330236
7. R. Campos-Vega, K.H. Nieto-Figueroa, B.D. Oomah, Cocoa (*Theobroma cacao L.*) pod husk: Renewable source of bioactive compounds. Trends Food Sci Technol. **81**, 172 (2018) https://doi.org/10.1016/j.tifs.2018.09.022
8. M.P. Raja, Dr.A. Karthiayani, Dr.P. Selvan, Dr.V. Nithyalakshmi, Development of defatted watermelon seed cake flour (*Citrullus vulgaris*) based RTE extruded snacks. Int. J. Chem. Stud. 8, 1379 (2020) https://doi.org/10.22271/chemi.2020.v8.i5s.10494
9. N. Devrajan, P. Prakash, N. Jindal, Effect of extrusion cooking on colour (L^*, a^*, b^*) of germinated buckwheat-corn based snacks. Int. J. Curr. Microbiol. App. Sci. **7**, 3413 (2018) https://www.ijcmas.com/special/7/Nalin%20Devrajan,%20et%20al.pdf
10. A.E. Lazou, Food extrusion: An advanced process for innovation and novel product development. Crit. Rev. Food Sci. Nutr. **64**, 4532 (2024) https://doi.org/10.1080/10408398.2022.2143474
11. S. Choton, N. Gupta, J.D. Bandral, N. Anjum, A. Choudary, Extrusion technology and its application in food processing: A review. Pharma Innov. **9**, 162 (2020) http://dx.doi.org/10.22271/tpi.2020.v9.i2d.4367
12. N. Yadav, D. Suvedi, A. Sharma, S. Khanal, R. Verma, D. Kumar, Z. Khan, L. Peter, Extrusion technology in food processing: Principles, innovations and applications in sustainable product development. Food Hum. **5**, 100672 (2025) https://doi.org/10.1016/j.foohum.2025.100672
13. H. Bobade, S. Sharma, Effect of extrusion on colour characteristics of honey enrichedwhole grain cereal flour extrudates. Int. J. Agric. Eng. **10**, 37 (2017) https://doi.org/10.15740/HAS/IJAE/10.1/37-42
14. Iv. Y. Bakalov, T. V. Petrova, M.M. Ruskova, K.D. Kalcheva-Karadzhova, N.D. Penov, The effect of extrusion variables on the colour of bean-based extrudates, Bulg. Chem. Commun. **48**, 407 (2016)
15. R. Valadez-Blanko, A.I.S. Virdi, S.T. Balke, L.L. Diosady, In-line colour monitoring during food extrusion: Sensitivity and correlation with product colour. Food Res. Int. **40**, 1129 (2007) https://doi.org/10.1016/j.foodres.2007.06.008
16. K. Rosentrater, K. Muthukumarappan, S. Kannadhason, Effects of ingredients and extrusion parameters on aquafeeds containing DDGS and corn starch. JAFSN 1, **44** (2009)
17. N. Poliszko, P.Ł. Kowalczewski, I. Rybicka, The effect of pumpkin flour on quality and acoustic properties of extruded corn snacks. J. Consum. Prot. Food Saf. **14**, 121 (2019) http://dx.doi.org/10.1007/s00003-019-01216-6
18. N. Toshkov, Examining the extrusion process of fish for farm species. Phd Thesis, University of Food Technologies, Faculty of Engineering (2011)
19. N. Toshkov, V. Nenov, B. Bozadjiev, N. Delchev, E. Valov, Extrusion of wheat semolina and cocoa bean shells. Food Sci. Appl. Biotechnol. **4**, 177 (2021) https://doi.org/10.30721/fsab2021.v4.i2.133
20. M. Dushkova, A. Koleva, D. Genev, A. Simitchiev, T. Petrova, M. Kakalova, Physicochemical and sensory characteristics of extrudates from rice enriched with pumpkin. Food Sci. Appl. Biotechnol. **6**, 103 (2023) https://doi.org/10.30721/fsab2023.v6.i1.224
21. A. Hagenimana, X. Ding, T. Fang, Evaluation of rice flour modified by extrusion cooking. J

Cereal Sci. **43**, 38 (2006) https://doi.org/10.1016/j.jcs.2005.09.003

22. E. Gujska, K. Khan, Feed moisture effects on functional properties, trypsin inhibitor and hemagglutinating activities of extruded bean high starch fractions, J. Food Sci. **56**, 443 (2006) http://dx.doi.org/10.1111/j.13652621.1991.tb05299.x
23. M. Kovac, D. Subaric, A. Jozinovic, J. Babic, J. Bekavac, S. Jokic, V. Barisic, D. Ackar, B. Milicevic, Application of cocoa bean shell extracts in the production of corn snack products. NFHD **10**, 69 (2021) https://hrcak.srce.hr/file/391039
24. M. Leonel, T. De Freitas, M. Mischan, Physical characteristics of extruded cassava starch. Food Sci. Technol. **66**, 486 (2009) https://doi.org/10.1590/S0103901620090004000009