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Abstract

The aim of the study is to produce gluten-free cookies characterized by high quality, high nutritional value, and good physical properties using chestnut and buckwheat flour as flour substitutes. Five different formulations of cookie samples were made using different amounts of two types of flour. The approximate analysis, amount of phenol, antioxidant levels and physical characteristics of the cookie samples were estimated. Results showed that the protein in the flour of buckwheat has a statistically significant higher quantity (12.22%) than the flour of chestnut (6.51%); these results were consistent with the approximate analyzes of cookies. Cookies samples made with a mixture of flours 25% CNF + 75% BWF and 50% CNF + 50% BWF contained higher levels of protein than cookies samples made with only CNF. The percentage of total dietary fiber in the samples of cookies made of 100% CNF, 75% CNF + 25% BWF and 50% CNF + 50% BWF were significantly higher than the other samples. Total phenolic contents were almost similar in all cookies samples. The antioxidant activity was significantly higher in cookies made of 100% CNF and 75% CNF + 25% BWF at (160.4 mg/100 mg) and (154.3 mg/100 mg), respectively. Results of physical properties showed that the spread ratio was significantly lower in cookies samples made from CNF compared to BWF. The hardness indicator was significantly increased with increasing amounts of buckwheat flour. The study concluded that mixing the two types of flours produced gluten-free cookies characterized by high product quality and good properties.

Keywords: chestnut flour, buckwheat flour, gluten-free cookies, alternative flours.

Introduction

Celiac disease is an autoimmune disease in which the immune system, by mistake, attacks healthy body tissues. In celiac disease, an error occurs in the immune system, as it considers the compounds found in gluten a danger for the human body and attacks it. This leads to damaging the intestinal wall, which hinders the body's ability to utilize nutrients (**Jnawali et al., 2016**). Glutens are present in any food containing grains, such as pasta, cake, breakfast cereals, and many types of bread. If a celiac patient eats food that contains gluten, it will lead to the appearance of symptoms in the digestive system, like diarrhea, stomach aches, flatulence and farting, indigestion and constipation. There is no recovery from this disease, however, following a diet free from gluten help in controlling symptoms and protecting from complications of this disease for long periods of time (**Peter et al., 2007**).

The development of products free of gluten is a big challenge in the food industry, particularly in the baked goods industry, as gluten has a major manufacturing role in forming the compositional structures for those products (**Capriles et al., 2015**). Characteristics of foods free of gluten must be studied and focused on, as traditional gluten-free flours contain high amounts of carbohydrate and small amounts of protein and antioxidants. Therefore, ingredients rich in dietary fiber should be used in gluten free food products for nutritional quality improvement (**Dicairano et al., 2018**).

Chestnut flour is produced by grinding dry chestnuts. This flour contains high content of starches (50% - 60%), sucrose (20% - 32%), and high-quality proteins, most of which are essential amino acids (5% - 8%), dietary fibers (4% - 10%) and a small amount (2% - 4%) of fat (**Littardi et al., 2020**). Chestnuts are good sources of vitamins, minerals (**Borges et al., 2008**), and antioxidants, it has been found to be related to decreasing the risk of cardiovascular diseases or stroke (**Ribeiro et al., 2007**).

Buckwheat (*Fagopyrum esculentum*) comes from the pseudo-cereal family. Buckwheat is gluten-free, which makes it a good alternative to wheat for people with gluten sensitivity or some digestive problems triggered by wheat products. It is considered one of the grains that has a high nutritional value, especially since it contains high percentages of dietary fibers, antioxidants and various minerals, which makes it a recommended food to fight many diseases, such as obesity, high blood pressure and diabetes (Li *et al.*, 2022).

Further development and innovation is required to increase the diversity of gluten-free dietary products to make them better tasting and more nutritionally valuable. This could be achieved by mixing natural ingredients that are rich in nutrients and bioactive contents (Capriles *et al.*, 2016).

The aim of the study was to produce gluten-free cookies characterized by high quality, high nutritional value and good physical properties using chestnut and buckwheat flour as flour substitutes.

Materials and methods

Materials

Buckwheat flour and chestnut flour are provided by the iHerb store website. The other ingredients (margarine, sugar and sodium bicarbonate) were bought from supermarkets. The chemical materials used in the study were bought from the El-Gomhoria Company in Cairo, Egypt.

Methods

Cookies production

Five different formulations of cookie dough were made using the following ingredients (g /100 g of cookie dough): flour (43.0), sugar (30.0), margarine (20.0), water (6.0), and sodium bicarbonate (1.0). The components of the five cookie samples were added in the same proportions unless water, was added to adjust the moisture of the flour.

First sample: cookies made of 100% chestnut flour (100% CNF).

Second sample: cookies made of 100% buckwheat flour (100% BWF).

Third sample: cookies made of flour mixes (50% CNF and 50% BWF).

Fourth sample: cookies made of flour mixes (75% CNF and 25% BWF).

Fifth sample: cookies made of flour mixes (25% CNF and 75% BWF).

The melted margarine was mixed with sugar in a mixer for 5 minutes. Add the water and mix for 5 minutes. The flour with sodium bicarbonate were put in, and mixed well for two minutes. Keep the dough for about half an hour. The dough is cut in a circular shape with a diameter of 40 mm. After that, cooked in the electric oven at 180°C for a quarter hour. Finally, keep the cookie samples in tightly sealed containers.

Chemical analysis

Approximate analyzes

The total content of ash, protein, fat, carbohydrate and dietary fiber were determined according to **AOAC (2005)**. The amount of carbohydrates was estimated by subtracting (protein + ash + fat) from 100. Total energy was calculated using the formula of **Chaney (2006)** for transformation factors: 9 per gram of fats, 4 per gram of carbohydrates and proteins.

Analysis of total phenolic content and antioxidant activity

The total phenolic content of the extracts of cookies was determined using the Folin and Ciocalteu reagents, following the method described by **Chandra et al. (2014)**. The antioxidant activity of the extracts of cookies was measured using the DPPH assay as described by **Malencic et al. (2007)**.

Physical analysis

Diameters and thickness of samples of cookies were evaluated using a caliper device. The spread ratio was calculated as the average of the diameter was divided by the thickness. All evaluations were done in triplicate. The value of hardness was obtained according to **Mamat and Hill (2014)**. The color of the cookie samples was evaluated using color Chroma meter, results were presented as, darkness / lightness (L^* values), redness (a^* values) and yellowness (b^* values).

Statistical analysis

Results were analyzed using SPSS software, version 18. An analysis of variance (ANOVA) was used for comparison. The values considered significant differences at $P < 0.05$ (**Snedecor and Cochran, 1980**).

Results and Discussion

Approximate analyzes of chestnut and buckwheat flours

According to the results presented in Table (1), samples of flour showed significantly differences in the nutrients analysis. It was observed that the percentage of protein in the flour of buckwheat showed a significant increase (12.22%) compared with the flour of chestnut (6.51%). Total energy values were also significantly higher in buckwheat flour (330.51) compared to chestnut flour (273.62). Percentages of ash and total dietary fiber were significantly higher in chestnut flour (3.66%) and (15.01%) compared to buckwheat flour (1.21%) and (7.64%) respectively. Regarding the values of fats and carbohydrates, there were no significant differences between the two types of flours.

The results are proportional with (**Ssoronja et al., 2017**) and (**Lopes et al., 2016**) which demonstrated that chestnut flour contains proteins at 5.30% and 5.70%, respectively. The results of the study were similar with (**Torbica et al., 2012**) which indicated that protein, ash and fat of buckwheat flour were 12.3, 2.2 and 2.9%, respectively and (**Kaur et al., 2015**) showed that carbohydrate values in buckwheat flour were 69.7%.

Table (1): Approximate analyzes of Chestnut and Buckwheat flour

Approximate analyzes	Chestnut flour	Buckwheat flour
Ash (g/100 g)	3.66±0.2 ^a	1.21±0.1 ^b
Protein (g/100 g)	6.51± 0.2 ^b	12.22± 0.3 ^a
Fat (g/100 g)	1.80±0.1	1.92±0.2
Carbohydrates (g/100 g)	69.43±0.3	73.52±0.4
Total dietary fiber (g/100 g)	15.01±0.1 ^a	7.64±0.2 ^b
Total energy (kcal/100g)	273.62±0.6 ^b	330.51±0.6 ^a

Data presented as means ± SD. Values in the same row with different letters are significantly different ($p < 0.05$).

Approximate analyzes of cookies samples

The baked cookies, with varying concentrations of chestnut and buckwheat flour were also subjected to nutrient analysis, which is presented in Table (2). Cookies made of 100% BWF contained the highest percentage of proteins (7.23%), while cookies made of 100% CNF contained the lowest percentage of protein (4.83%), which is consistent with the results of the flour analysis, as the amounts of protein in buckwheat flour showed high percentages and the lower percentages were in chestnut flour. Cookies made of a mixture of flour 25% CNF + 75% BWF and 50% CNF + 50% BWF, contained higher levels of protein (6.62%) and (6.0%) respectively, than cookies made of only chestnuts, which indicates that mixing the flour was found to be effective on protein levels. These results are consistent with the results demonstrated by (Sedej *et al.*, 2011), where whole grains flour of buckwheat was used in the manufacture of free-gluten products.

Results in Table (2) showed that the percentage of fat and carbohydrates didn't show any significantly differences between all samples. The percentages of total dietary fiber in the samples

of cookies made of 100% CNF, 75% CNF + 25% BWF and 50% CNF + 50% BWF were significantly higher than the other samples (13.51%), (12.81%) and (12.42%) respectively. While, the value of dietary fiber was the lowest in cookies made with 100% BWF, this is consistent with the lower percentage of dietary fiber in flour analysis of buckwheat. The highest value of total energy was in samples of cookies made by 100% BWF, while the lowest value was in samples of cookies made by 100% CNF. Regarding ash, there are no noticeable variations in different cookie samples.

Table (2): Approximate analyzes of cookies samples made from different amounts of flours

Approximate analyzes	100% CNF	100% BWF	50% CNF +50%BWF	75% CNF +25%BWF	25% CNF +75%BWF
Ash (g/100 g)	3.69±0.2	3.51±0.1	3.60±0.2	3.59±0.1	3.41±0.3
Protein (g/100 g)	4.83± 0.2 ^c	7.23± 0.3 ^a	6.0± 0.4 ^c	5.42± 0.1 ^d	6.62± 0.1 ^b
Fat (g/100 g)	17.71±0.3	16.90±0.2	16.60±0.3	17.22±0.2	16.91±0.4
Carbohydrates (g/100 g)	69.23±0.3	71.52±0.4	70.15±0.6	69.67±0.5	79.63±0.2
Total dietary fiber (g/100 g)	13.51±0.3 ^a	11.52±0.2 ^c	12.42±0.3 ^c	12.81±0.1 ^b	11.98±0.2 ^d
Total energy (kcal/100g)	397.62±1.6 ^e	409.71±1.9 ^a	403.66±1.4 ^c	400.69±1.9 ^d	406.67±1.8 ^b

CNF: Chestnut flour, BWF: Buckwheat flour. Data presented as means ± SD. Values in the same row with different letters are significantly different (p<0.05).

Total phenolic content and antioxidant activity

The amounts of total phenol in the cookie samples are summarized in Table (3). The total phenol expressed in gallic acid equivalents was almost similar in all cookies samples. The levels of phenolic content in 100% CNF and 100% BWF were (405.1 mg/100 g) and (393.1 mg/100 g) respectively.

The results agreed with (De Vasconcelos *et al.*, 2010), demonstrated that chestnut fruits contain high and varying amounts of total phenols, mainly gallic and ellagic acids (280 - 910 mg/100 g and 610 - 2560 mg/100 g). Use of gluten-free products that contain high levels of phenolic compounds is of great benefit to patients with celiac disease due to their ability to be linked with dietary fiber, making it easily absorbed of intestines, as well as considering phenols as antioxidants (Rocchetti *et al.*, 2018).

Antioxidant activity of cookies samples presented in Table (3). Antioxidants were significantly higher in cookies made of 100% CNF, 75% CNF + 25% BWF and 50% CNF + 50% BWF, the values were (160.4 mg / 100 mg), (154.3 mg / 100 mg) and (148.4 mg / 100 mg) respectively.

The results agreed with the study by (Paciulli *et al.*, 2018) which confirmed a significantly increasing in levels of antioxidant activity after cooking baked biscuits containing chestnut flour. This was attributed to the effects of millard reactions and compounds forming during heat treatment.

Table (3): Phenolic content and antioxidant activity of cookies samples made from different amounts of flours

Parameters	100%CNF	100%BWF	50% CNF +50%BWF	75% CNF +25%BWF	25% CNF +75%BWF
Total phenolics (mg/100 g)	405.1±2.8	393.1±7.3	399.2±5.4	402.07±6.3	405.7±7.1
Total antioxidant activity (mg/100mg)	160.4±4.3 ^a	136.4±2.8 ^e	148.4±3.1 ^c	154.3±5.3 ^b	141.4±4.1 ^d

CNF: Chestnut flour, BWF: Buckwheat flour. Data presented as means ± SD. Values in the same row with different letters are significantly different (p<0.05).

Physical characteristics of the cookies

The data in Table (4) showed that there are no significantly variations of physics characteristics in the diameters, thickness and masses of cookies samples. The spread ratio was significantly lower in cookies made of 100% CNF compared to 100% BWF.

The results agreed with (**Demirkesen, 2016**), as the results showed a decrease in the diameter of cookies when increasing the amount of chestnut flour compared to other samples. The spread ratio is largely associated with the chemical structure of the components used in cookies and also with the mixing of sugar with fat and lower dough viscosity (**Brites et al., 2019**). In the initial stage of baking process, melted fats with sugar lower dough viscosity, leading to the dough to relax and expand (**Pareyt and Delcour, 2008**).

Regarding the hardness indicator, it significantly increased with increasing the amounts of buckwheat flour comparing with chestnut flour. The values of hardness were significantly higher in 100% BWF and 25% CNF + 75% BWF as (75.31 N) and (72.59 N) respectively, comparing with 100% CNF and 75% CNF + 25% BWF as (64.50 N) and (67.19 N) respectively.

These results agree with the results of (**Taylor et al., 2016**), a relationship between the hardness of baked goods and the flour of buckwheat was confirmed. Moreover, (**Kaur et al., 2015**) reported that adding buckwheat flour increased the value of the hardness indicator (24.6 N) for (42.30 N) of gluten-free biscuits comparing to wheat flour.

Fig. (1) shows the results of color parameters for cookies samples, as they are considered an important factor for the quality of food products and have an impact on consumer acceptance of the final products. The results showed that cookies samples made of 100% BWF had a significantly ($p < 0.05$) greater (L^* value) compared with 100% CNF.

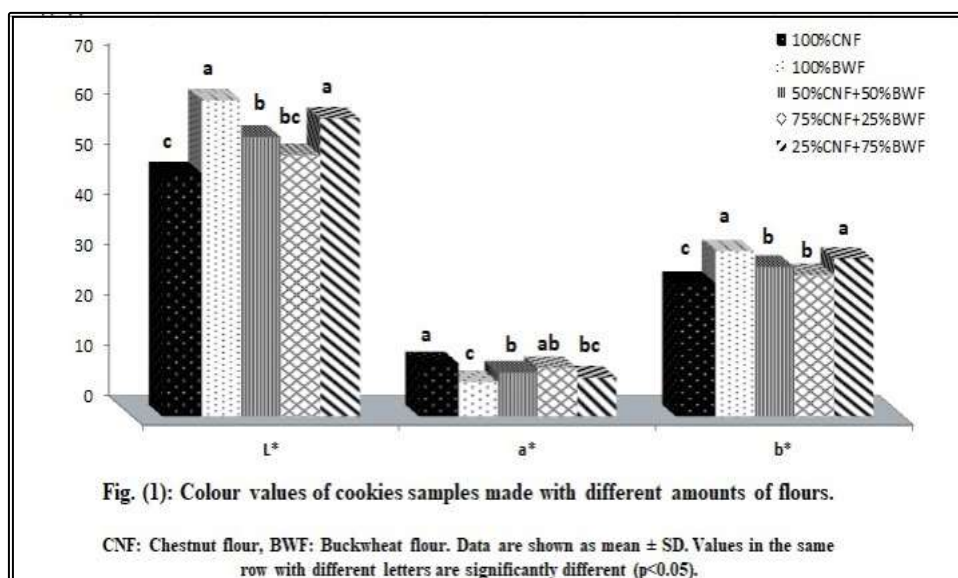
Table (4): Physical properties for cookies samples made from different amounts of flours

Physical characteristics	100% CNF	100% BWF	50%CNF + 50%BWF	75%CNF +25%BWF	25%CNF +75%BWF
Diameter (mm)	58.30±0.8	62.21±0.6	60.75±0.7	60.12±0.4	60.47±0.6
Thickness (mm)	10.90±0.5	9.50±1.0	10.2±0.6	10.54±0.8	9.84±0.9
Mass (g)	19.51±1.6	18.71±1.4	19.1±1.8	19.29±1.5	18.89±1.7
Spread ratio	5.34±0.6 ^b	6.52±0.9 ^a	5.87±0.4 ^b	5.68±0.8 ^b	6.15±0.7 ^a
Hardness (N)	64.50±3.5 ^c	75.31±4.1 ^a	69.91±2.5 ^b	67.19±2.5 ^{bc}	72.59±3.2 ^{ab}

CNF: Chestnut flour, BWF: Buckwheat flour. Data presented as means ± SD. Values in the same row with different letters are significantly different ($p < 0.05$).

It was also observed in Fig. (1) that the lightness value reduced with increasing the percentage of chestnut flour. Moreover, results showed that the (a^* value) were significantly higher in the chestnut flour cookies samples compared to the rest of the samples. While (b^* value) decreased with increasing amounts of chestnut flour in the cookies samples. The cookies made of 100% BWF had significantly ($p < 0.05$) higher (b^* value) compared with chestnut flour cookies samples.

The results of the study agreed with (Dall *et al.*, 2013) and (Ssoronja *et al.*, 2017), which showed that using chestnut flour in cookies and bread samples as a wheat substitute led to increased redness in the samples of 20 to 60% and 0 to 100%, respectively. In addition, the results agreed with (Paciulli *et al.*, 2018), as chestnut flour contains a high percentage of sugars (20%-32%) and starches (50%-60%), which are associated to the caramelization processes and reactions of maillard that occur during the baking process. These interactions cause a reduction of (L^* value) with elevation of (a^* value) in the chestnut flour cookies samples.



Conclusion

Depending on the results of the study, it can be concluded that chestnut and buckwheat flour could be combined to produce gluten-free cookies that are distinguished by their high nutritional value, high percentage of dietary fibers, high levels of phenolic content and antioxidant activity, as well as their good physical properties. Mixing chestnut flour with buckwheat flour improved the characteristics of cookies produced using only one type of flour. The study concluded that mixing the two types of flours produced gluten-free cookies characterized by high product quality and good properties.

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تقييم الخصائص الكيميائية والفيزيائية للكوكيز خالي الغلوتين المحضر بدقيق الكستناء والحنطة السوداء

سها محمد يوسف

التغذية وعلوم الأطعمة - قسم الاقتصاد المنزلي- كلية التربية النوعية- جامعة الفيوم

الملخص العربي

الهدف من هذه الدراسة هو إنتاج كوكيز خالية من الغلوتين تتصف بجودة عالية ، قيمة غذائية عالية وخصائص فيزيائية جيدة باستخدام دقيق الكستناء ودقيق الحنطة السوداء كبدائل للدقيق. تم عمل خمس تركيبات مختلفة من عينات الكوكيز باستخدام كميات مختلفة من نوعين الدقيق. تم تقييم التركيب التقريبي، قيم الفينول الكلي، مستويات مضادات الأكسدة والصفات الفيزيائية لعينات الكوكيز. أظهرت النتائج أن نسبة البروتين الموجود في دقيق الحنطة السوداء أعلى بكثير 12.22% من دقيق الكستناء 6.51% ، وكانت هذه النتائج متوافقة مع التحليلات التقريبية لعينات الكوكيز. عينات الكوكيز المصنوعة من خليط الدقيق 25% دقيق الكستناء + 75% دقيق الحنطة السوداء و 50% دقيق الكستناء + 50% دقيق الحنطة السوداء احتوت على مستويات أعلى من البروتين مقارنة بالعينات المصنوعة من دقيق الكستناء فقط. مستويات الألياف الغذائية في عينات الكوكيز المصنوعة من 100% دقيق الكستناء، 75% دقيق الكستناء + 25% دقيق الحنطة السوداء و 50% دقيق الكستناء + 50% دقيق الحنطة السوداء كانت أعلى بكثير من العينات الأخرى. قيم الفينول الكلي كانت متشابهة تقريباً في جميع عينات الكوكيز. بينما قيم نشاط مضادات الأكسدة كانت أعلى معنوياً في عينات الكوكيز المصنوعة من 100% دقيق الكستناء، 75% دقيق الكستناء + 25% دقيق الحنطة السوداء ، القيم كانت 160.4 mg/100mg ، 154.3 mg/100mg على التوالي. نتائج تقييم الصفات الفيزيائية لعينات الكوكيز أظهرت ان نسبة الانتشار كانت أقل بكثير في الكوكيز المصنوع من دقيق الكستناء مقارنة بالكوكيز المصنوع من الحنطة السوداء. كما أن هناك ميل إلى زيادة الصلابة بشكل ملحوظ مع زيادة محتوى دقيق الحنطة السوداء في عينات الكوكيز. نستخلص من الدراسة أن خلط دقيق الكستناء مع دقيق الحنطة السوداء يمكن أن ينتج كوكيز بخصائص أفضل من الكوكيز المنتج باستخدام نوع واحد فقط من الدقيق، حيث تم إنتاج كوكيز خالي من الغلوتين يتميز بجودة منتج عالية وخصائص جيدة.

الكلمات المفتاحية: دقيق الكستناء، دقيق الحنطة السوداء، الكوكيز الخالي من الجلوتين، بدائل الدقيق.