Cumhuriyet Science Journal
e-ISSN: 2587-246X
ISSN: 2587-2680
Cumhuriyet Sci. J., 42(2) (2021) 299-309
http://dx.doi.org/10.17776/csj.837862

Determination of the chemical structure of diet biscuits with modern enstrumental techniques

İdil KARACA AÇARI [1, *]

[1]*Bioengineering Department, Faculty of Engineering and Natural Sciences, Malatya Turgut Ozal University, 44210, Malatya / TURKEY*

Abstract

There are too many deficiencies in the studies in terms of diet biscuit in the literature. This study aims to fill this gap in the literature. The % moisture content, structural content verification, thermal stability, degradation properties and % ash content of diet biscuit samples, structural characterizations, surface micromorphology and detailed structural analysis were determined with fast, precise, new instrumental techniques. Thermal degradation of the lemon-fiber diet biscuit sample started at 250 °C in four stages and completed at 585 °C. There is three-stage thermal degradation in wholemeal diet biscuit samples, starting at 229 °C and completed at 580 °C. There are 46.06% C, 1.60% N, 0.12% S and 6.68% H in lemon-fiber diet biscuits, and 45.51% C, 2.39% N, 0.15% S and 7.05% H in whole-wheat diet biscuits. Ca, K, Mg, Na, Zn element contents in diet biscuit samples were determined by AAS technique. In the diet biscuit samples, K was determined as the element with the highest amount and the element with the least amount was Zn. As a result, the work is comprehensive and original. It is believed that the study will fill an important gap in the literature and will be a useful resource for researchers.

Article info

History:
Received:08.12.2020
Accepted:02.05.2021

Keywords:
Enstrumental techniques,
Diet biscuit,
Structural characterization,
Thermal analysis,
Morphologies analysis.

1. Introduction

Bakery products are the most popular processed foods in the world [1]. Biscuits, included in the bakery products class, represent the most popular snack category in all population groups around the world. The reason why biscuits are made from simple, inexpensive, easily available raw materials, very acceptable taste, availability in different flavors and long shelf life are the reasons that make them the most consumed bakery products all over the world [2, 3]. The name biscuit originated in France for the first time, as a result of baking a type of bread twice in order to provide better protection [4]. A popular food eaten by both children and adults, biscuits typically contain ingredients (fat and sugar) that make them unhealthy. The traditional method of making biscuit dough is to use semi-solid fat at room temperature [5]. Briefly, biscuits are high-energy, easily digestible foods based mostly on wheat flour, oil and sugar [6, 7]. The high calorie content of the biscuits disrupts the balanced diet of the consumer. Therefore, recently, in line with the attempts to improve nutritional profiles, fat, sugars and energy levels have been reduced [8]. Therefore, dietary forms have been developed and presented to consumers. The diet (light) product consists of low-fat, artificial sweeteners and fiber-added products used instead of sugar. Dietary product consumption habit started in the 1960s [9]. Dietary product usage is very common nowadays. Today, health problems related to nutrition such as obesity, diabetes, hypertension, hypercholesterolemia and gastrointestinal problems are increasing. This increase brought the awareness that people should pay more attention to their diet. In the studies, the reasons for individuals to prefer dietary products were investigated. These are slimming, feeling healthier, eating a balanced and healthy diet, low-calorie foods are healthier than other foods, low-calorie foods prevent weight gain, and low-calorie foods taste good [10]. With the increasing use of dietary products, various diet products have started to be produced. Diet biscuits are widely consumed among diet products. The chemical structure contents of the dietary biscuit samples (lemon-fiber and wholemeal) selected within the scope of the study were determined by instrumental techniques. Structural characterizations of dietary biscuit samples were checked with Fourier transform infrared spectroscopy

*Corresponding author. *e-mail address: idil.karaca@ozal.edu.tr*

(FTIR), atomic absorption spectrometry (AAS), elemental analysis techniques, surface morphology and structures with scanning electron microscope (SEM) / energy dispersive X-ray spectroscopy (EDX) techniques, and also thermal properties with differential thermal analysis (DTA) and thermogravimetric analysis (TGA) techniques.

2. Materials and Methods

2.1. Materials

In this study, lemon-fiber and wholemeal diet biscuit samples of brand A were used. In the diet biscuit samples selected for this stud were determined respectively general analytical properties, structural characterizations, surface morphology and structure determinations and thermal properties by appropriate techniques.

2.1. Preparation of samples

Biscuits are highly hygroscopic and quickly absorb moisture when exposed to the atmosphere [11, 12]. For this reason, the preparation of the sample was carried out very quickly by grinding in a porcelain mortar. The powdered diet biscuit samples were stored in a dry airtight glass jar until analysis.

2.2. Determination of % moisture content

Moisture content in diet biscuit samples was determined by gravimetric method based on drying process in Ecocell branded oven at 105 °C [13,14]. The weighing values of the samples were taken at certain time intervals. The weighing process was continued until the difference between the two weightings was 0.5% [15]. The % moisture content in diet biscuit samples was calculated according to the formula below.

$\% \text{ Moisture} = (W_1-W_2)/W_1-Wx100$

Based on the formula given, the sample weight before drying is shown as W_1, the dried sample W_2 and the tared sample container W which is dried and brought to constant weight. Experiments were performed in triplicate.

2.3. Thermal properties and % ash content

Thermal analysis includes methods based on the measurement of the change in physical properties as a function of temperature by heating the samples in a controlled manner. Analyzes were performed with Shimadzu system 50 model thermogravimetric analysis (TGA), Shimadzu system 50 model differential thermal analysis (DTA) devices. Ash content of diet biscuit samples was determined by thermogravimetric analysis, which is one of the thermal analysis methods, instead of traditional dry burning ash determination method [13].

2.4. Structural characterization

Chemical structure characterization was determined by Fourier transform infrared spectroscopy (FTIR) analysis, elemental analysis and atomic absorption spectrometry (AAS) techniques. LECO brand CHNS-932 model device was used in elemental analysis and Perkin Elmer AAnalyst AAS device was used for determination of mineral content. The change in the chemical bond structures of the dietary biscuit samples was investigated by Thermo Matson 1100 brand Fourier transform infrared spectroscopy (FTIR) analysis using the attenuated total reflection (ATR) technique in the 4000-400 cm-1 wavelength range.

2.5. Surface morphology-structure determination

Micromorphological properties of diet biscuit samples were determined with Leo EVO 40 model scanning electron microscope (SEM) at different magnifications. Structural determination was performed with "Quantax" brand energy dispersive X-ray spectroscopy (EDX) device with Bruker 125 eV detector.

2.6. Statistical analysis

Statistical analysis for % moisture and % ash of the samples was performed using GraphPad Prism. Results are presented as mean ± SD, unless otherwise stated, and statistical significance was accepted as $p < 0.05$.

3. Results and Discussion

3.1. % Moisture content of diet biscuit samples

Moisture contents which are the general analytical properties of dietary biscuit samples are given in figures 1. When we look at figure 1 for moisture content, we see that this value is 2.85 ± 0.06 for lemon-fiber diet biscuit, and 2.77 ± 0.09 for wholemeal diet biscuit. There is no statistically significant difference between the% moisture values of the diet biscuit samples.

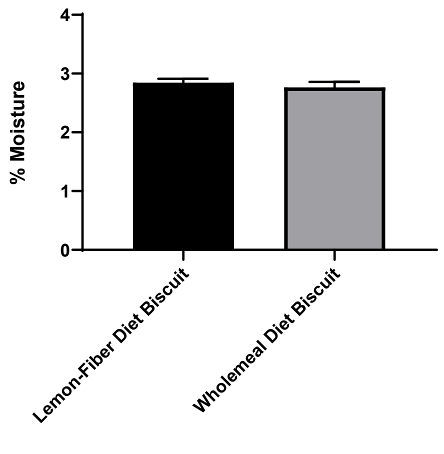

Figure 1. % Moisture Content in Diet Biscuit Samples.

There is no study similar to our study for dietary biscuit types in the literature. For this reason % moisture contents were discussed according to the existing biscuit studies in the literature not by comparing them with diet biscuit varieties. Nakov et al. determined the moisture in the biscuits containing different amounts of inulin between 2.5-2.94% [7]. Moisture ranges are compatible with our moisture ranges. Adeola et al. determined the moisture ratio between 6.30-7.90% in biscuits made of banana, pigeon pea and sweet potato flour at different rates [16]. In this study the moisture content is quite high compared to the diet biscuits we used in the study. Low moisture content of diet biscuits is known. Low moisture content in biscuits extends their shelf life. Ullah et al. made biscuits by adding different amounts of alfalfa seed flour to refined wheat flour. They observed that the amount of the moisture content in biscuits changed from 3.57% to 3.26%, respectively [17]. Their moisture content is higher than the diet biscuits we used in the study. This is an expected situation. Kumar et al. made biscuits containing seaweed in different proportions. They gave moisture ratios of these biscuits by comparing them with normal biscuits. They determined the moisture as 4.19 ± 0.07% in the normal biscuit sample. They determined the moisture content of the biscuit samples containing 1%, 5% and 10% seaweed as 4.08 ± 0.12%, 3.97 ± 0.1%, 4.03 ± 0.16, respectively [18]. In this study, we see once again that the moisture content of normal biscuits is higher than that of diet biscuits. Agu et al. made biscuits containing different proportions of wheat flour and boiled and unripe banana flour. They determined the moisture content of these biscuit samples as 1.84 ± 0.17%, 2.01 ± 0.11% and 2.55 ± 0.28% [19]. When compared to the diet biscuits we use, we see that the moisture content is low. As a result, biscuits have different moisture values due to the different additives they contain. As a result, we can say that diet biscuits contain lower moisture than other biscuits. Because diet biscuits contain fiber and bran. The water was retained by the fiber and bran [20].

3.2. Determination of thermal properties and % ash content of diet biscuit samples

Thermal properties of dietary biscuit samples were determined by TGA and DTA analyzes. Thermal analysis of dietary biscuit samples is used to verify the structural content of samples, to determine their thermal stability and degradation properties, as well as to offer an important and sensitive alternative to the determination of classical moisture and ash content by gravimetric method. The results of the TGA analysis performed within the scope of the study are given in figure 2. In these analyzes, wholemeal diet biscuits and lemon-fiber diet biscuits gave slightly different thermograms. 3 different mass loss can be seen in the TGA thermogram of a wholemeal diet biscuit sample given in figure 2 (A). The first mass loss seen between 65 °C and 120°C is due to the moisture in the biscuit structure [21]. This mass loss resulting from the removal of structural moisture is approximately 2.05%. The second mass loss is seen between 180 °C and 400 °C and is caused by the degradation of the carbohydrate structures and starch in the biscuit structure [21]. This mass loss was determined to be approximately 59.27%. The final mass loss is due to thermal decomposition of difficult degraded structures such as bran structure. It is approximately between 400 °C and 500 °C and is around 30.39% [22]. According to TGA analysis results, the amount of ash at 600 °C was determined to be 7.47±0.04% for a wholemeal diet biscuit. There are four different mass losses in the lemon-fiber diet biscuit structure. The first mass loss occurs between 75 °C and 120 °C, about 1.65% and is due to the removal of structural moisture. The second mass loss is due to the degradation of aromatic compounds added to the diet biscuit structure. It is seen between 140 °C and 240 °C and is approximately 10%. The third mass loss, approximately 55.42%, is due to the degradation of the capillary components, carbohydrate and starch, and is between 240-400 °C. The final mass loss between 400 °C and 580 °C is due to the thermal decomposition of hard-graded components and is approximately 25.22%. The amount of ash at 600 °C for lemon-fiber diet biscuit is 6.51±0.03%. There is a statistically significant difference between lemon-fiber biscuit ash ratio and wholemeal biscuit ash ratio ($p < 0.05$). The wholemeal biscuit ash ratio is due to the excessive bran content it contains. The bran biscuit ash ratio is due to the excessive bran content it contains. Ash contents were discussed according to the existing biscuit studies in the literature not by comparing them with diet biscuit varieties. Majzoobi et al. determined the ash content in the biscuit samples containing palm syrup and palm liquid sugar in the range of 5.30 ± 0.51 - 25.3 ± 0.3% and 0.60 ± 0.03 - 27.1 ± 0.2%, respectively [23]. Increasing excess of ash content compared to the ash content of diet biscuit samples is due to the additives of palm syrup and palm liquid sugar in their structure. Adeola et al. determined the ash ratio in the range of 0.9-2.40% in biscuits made of banana, pigeon pea and sweet potato flour at different rates [16]. Ullah et al. made biscuits by adding different amounts of alfalfa seed flour to refined wheat flour. They observed that the amount of ash in biscuits changed from 1.37% to 1.92%, respectively [17]. Ash content is in accordance with our results, and their moisture content is higher than the diet biscuits we used in the study. Kumar et al. made biscuits containing seaweed in different proportions. They gave moisture and ash ratios of these biscuits by comparing them with normal biscuits. They determined the amount of ash in normal biscuits as 1.28 ± 0.01%. In other biscuit samples, they determined the amount of ash as 1.37±0.02%, 1.68±0.00% and 2.23±0.03% respectively [18]. The amount of ash is less than the wholemeal diet biscuit we used in the study. Agu et al. made biscuits containing different proportions of wheat flour and boiled and unripe banana flour [19]. They determined the ash amount as 2.94 ± 0.16%, 3.16 ± 0.03%, 3.68 ± 0.18%, respectively. As a result, we can say that the fiber and bran content of the diet biscuit samples is higher and therefore the ash content is higher.

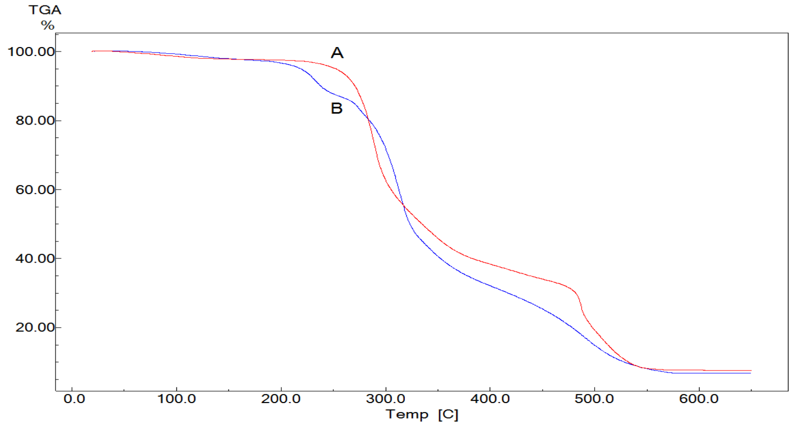

Figure 2. TGA Thermograms of Diet Biscuit Samples (wholemeal diet biscuit (A) and lemon-fiber diet biscuit (B).

In Figure 3, DTA thermograms of diet biscuit samples are given. These thermograms have been seen to be compatible with TGA thermograms. Thermal decomposition in wholemeal diet biscuit starts at approximately 229 °C and is completed at approximately 580 °C with a three-stage thermal decomposition [24]. Lemon-fiber diet biscuit structure shows a thermal degradation structure that starts at 250 °C and ends at 585 °C. This structure degrades in four stages and has a different thermogram than the wholemeal diet biscuit structure.

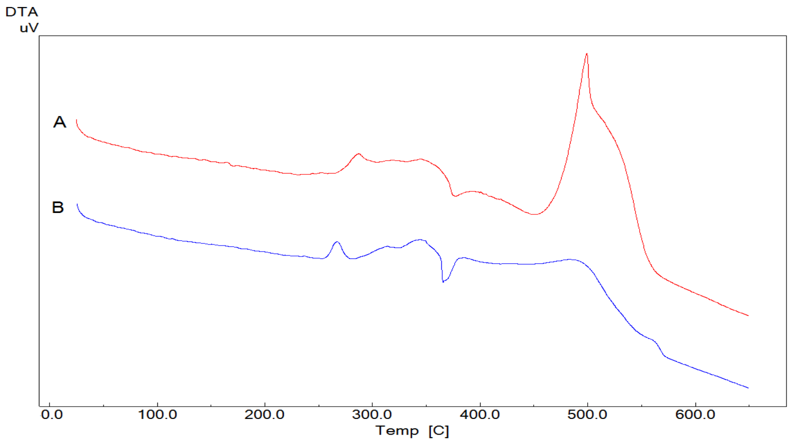

Figure 3. DTA Thermograms of Diet Biscuit Samples (wholemeal diet biscuit (A) and lemon-fiber form biscuit (B).

3.3. Structural characterization of diet biscuit samples with elemental analysis, FTIR and AAS techniques

In this part of the study, diet biscuit samples were examined structurally and information about their chemical structures was obtained. Significant element percentages in diet biscuit samples were determined with the results of elemental analysis. The results are given in table 1. According to this, it was determined that 46.06% C, 1.60% N, 0.12% S and 6.68% H in lemon-fiber diet biscuit structures. Wholemeal diet biscuit structures contain elements in the ratios of 45.51% C, 2.39% N, 0.15% S and 7.05% H. When these values are compared with the elemental analysis results of classical biscuit structures in the literature, it is seen that the N and S elemental content is high [25]. This situation is diet biscuits contain more bran and bran protein than normal biscuits. Especially N and S elements in protein structure directly contribute to the biscuit structure.

Table 1. Percentages of Elements Determined in Diet Biscuit Samples.

Sample Name	Element Percentages
Lemon-fiber diet biscuit	46.06% C, 1.60% N, 0.12% S and 6.68% H
Wholemeal diet biscuit	45.51% C, 2.39% N, 0.15% S and 7.05% H

FTIR analysis was used to illuminate the chemical structure of the diet biscuit samples and the FTIR spectra obtained are given in figure 4. FTIR spectra provide useful structural information that can help identify samples enriched with specific functional food ingredients [18].

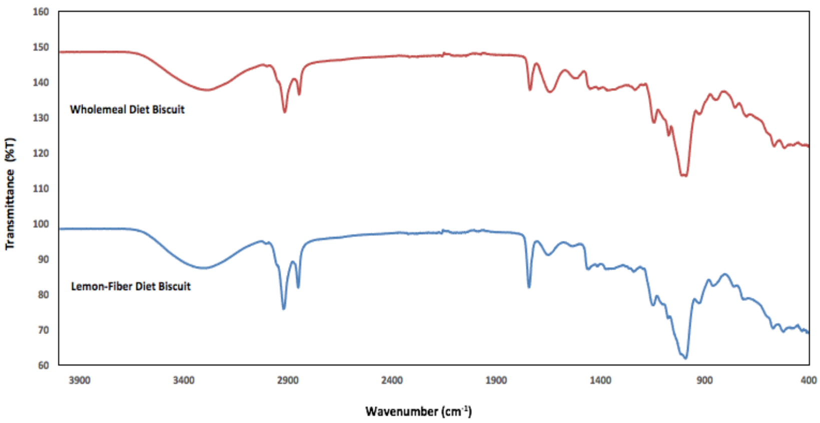

Figure 4. FTIR spectrum results of diet biscuit samples.

On the FTIR spectra of the diet biscuit samples given in figure 4, the 3 main peak structures are clearly seen. The peaks in these structures are caused by carbohydrate, protein and structural fat molecules and are clearly visible. Especially due to the carbohydrate molecules, we see peaks of free OH groups in the range of 3000-3600 cm^{-1}. These peaks are caused by both structural moisture and surface hydroxyls. In addition, a wide and severe peak in the range of about 1000-1100 cm^{-1} due to carbohydrate structures is caused by etheric stretching vibration. C-O stretching vibrations stand out as a thin peak around 1200 cm^{-1}. Due to protein structures, FTIR spectra contain amide 1 and amide 2 peaks [26, 27]. Peaks in protein structure; 1650 cm^{-1} amide 1 peak is the amide 2 peak that comes around 1548 cm^{-1}. These specifically show carbonyl stretching vibrations in protein structures. Again, due to the protein structure peptide bonds, C-N stretching vibrations were detected at 1440 cm^{-1}. At low wave numbers, especially at 825-650 cm^{-1}, aliphatic C-H vibrations, aliphatic CH stretch vibrations in the range of 2850-2950 cm^{-1} and C-C stretch vibrations at 1380 cm^{-1} are seen due to both protein structures and carbohydrate structures. A very small C = O stretching vibration is observed in diet biscuit structure especially at 1730 cm^{-1}. This peak is proof that the amount of fat in the diet biscuit structure is very low. This peak is generally seen in classical biscuits as prominent and severe. This situation is due to the fat molecules in the structure. However, it is almost non-existent in the diet biscuit structure. When FTIR spectra in Figure 3 are compared with each other, an esteric carbonyl stretch vibration around 1820 cm^{-1} is seen in the lemon fiber diet biscuit structure. This is due to the aromatic ester flavors in the biscuit structure.

Analysis of Ca, K, Mg, Na, Zn elements in dietary biscuit samples was performed by flame atomic absorption spectrometry (FAAS) technique. Samples were digested primarily by wet digestion method. According to this method, after adding 5 mL (HCl-HNO_3, 3:1 v / v) to the 0.5 g samples, 2 mL ethanol (99.8 %) was added to the mixture.The resulting mixture was then filtered through whatman 42 filter paper [28]. Calibration graph was drawn for each given element for Ca, K, Mg, Na, Zn analyzes in the samples obtained.

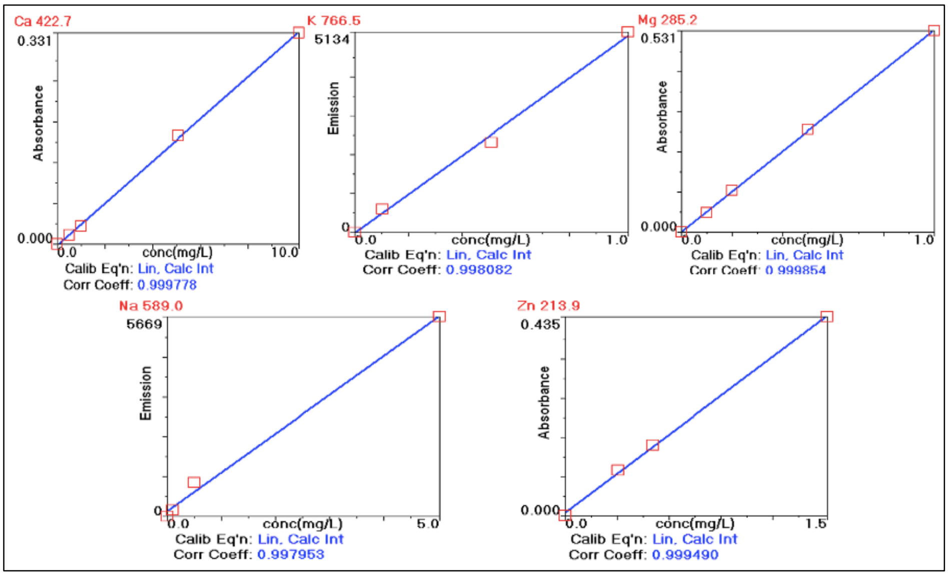

Figure 5. Calibration graph for Ca, K, Mg, Na, Zn elements

Table 2. Analytical working ranges, correlation coefficients and wavelengths of Ca, K, Mg, Na and Zn elements.

Element	Analytical working range (mg/L)	Correlation coefficient (R^2)	Wavelength (nm)
Ca	0-10.0	0.9998	422.7
K	0-1.0	0.9981	766.5
Mg	0-1.0	0.9999	285.2
Na	0-5.0	0.9980	589.0
Zn	0-1.5	0.9995	213.9

The graphics are given in figure 5. İnformation about the analytical working ranges, correlation coefficients and wavelengths of Ca, K, Mg, Na and Zn elements is given in table 2. Elemental analyzes in the prepared samples were determined in triplicate by flame atomic absorption spectroscopy. Results are given in Table 3.

3.4. Surface Morphology-Structure Analysis of Diet Biscuit Samples with SEM / EDX technique

Diet biscuit samples have a different structure than other biscuits and cookies due to the bran structure they contain and with low fat content [29, 30]. Basically, it displays more harder and rigid morphology.

Table 3. Analytical results of Ca, K, Mg, Na, Zn elements in diet biscuits samples (metal concentrations as, mg/100 g).

Sample Name	Ca	K	Mg	Na	Zn
Lemon-fiber diet biscuit	24.89±0.15	100.2±0.42	25.16±0.01	46.92±0.40	0.505±0.01
Wholemeal diet biscuit	31.01±0.32	170.3±2.97	42.22±0.01	55.73±0.11	0.813±0.01

SEM analyzes were performed to examine the micro morphological and surface structure of the apparently hard-structured diet biscuit samples and the results are given in Figure 6 and Figure 7.

A very fractal but monolithic structure is seen in the lemon-fiber diet biscuit structure given in figure 6. An uninterrupted and cavitated SEM surface image is seen. Due to starch structures, spherical structures are seen in places. However, the microstructure shows irregular and small porosity.

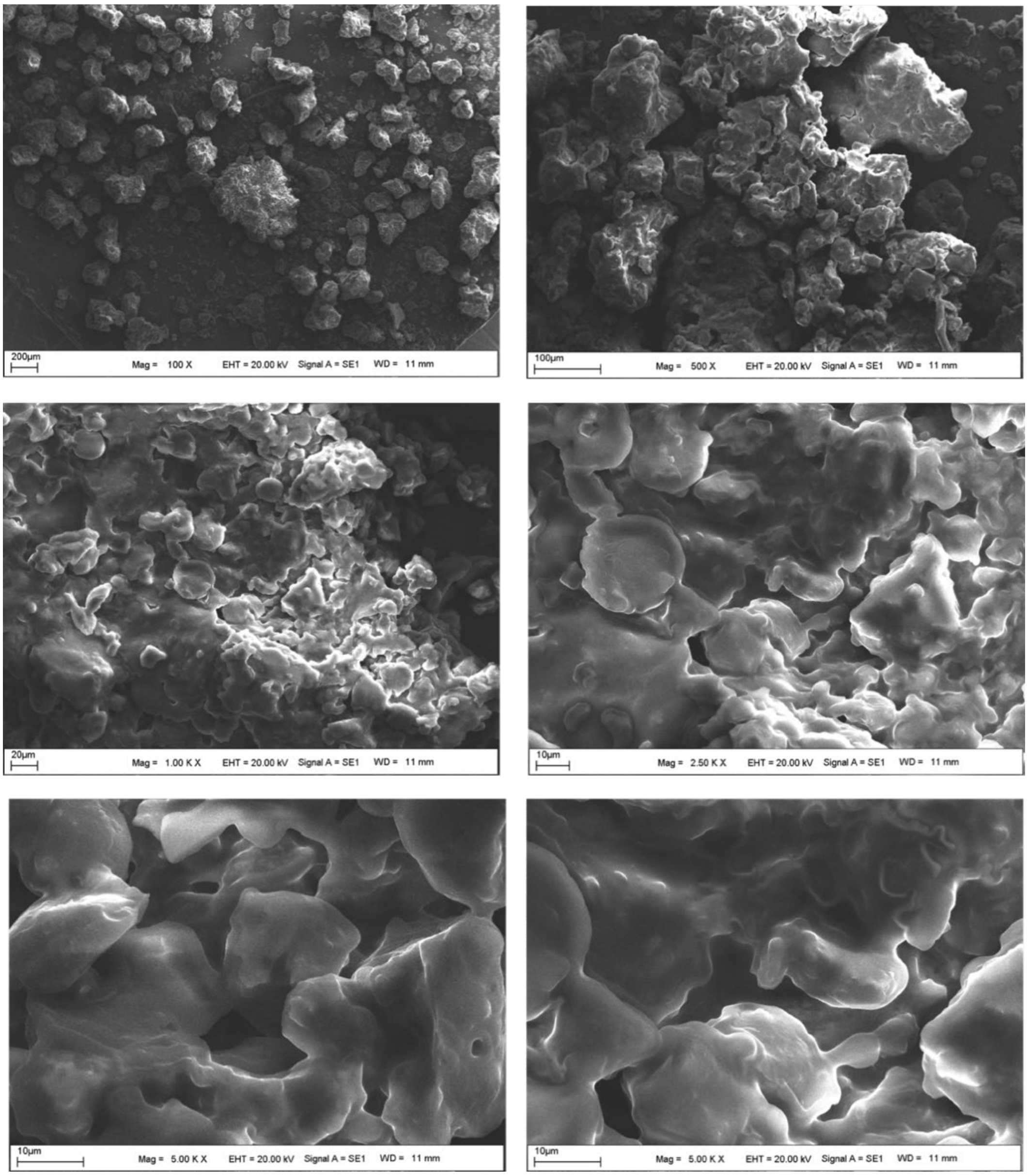

Figure 6. SEM Images of Lemon-Fiber Diet Biscuit Sample at Different Magnifications.

A high cavital structure is seen in the wholemeal biscuit structure in figure 7. In places, bran fibers are visible and starch structures are frequently encountered. In particular, the porosity was seen to be greater. Although the biscuit structure exhibits a monolithic structure, it contains fractal and granular parts in places. The evaluation of the starch spherical structures of the lemon-fiber diet and wholemeal diet biscuit samples given in Figure 6 and Figure 7 is given in detail in figure 8. In the SEM images of the lemon-fiber diet biscuit sample given in figure 8 (A) and (B) the more prominent spherical structures are clearer and distinct. However, it is seen that the starch structures in the structure of wholemeal diet biscuit in figures (C) and (D) are better adhered to the main surface and structurally fused.

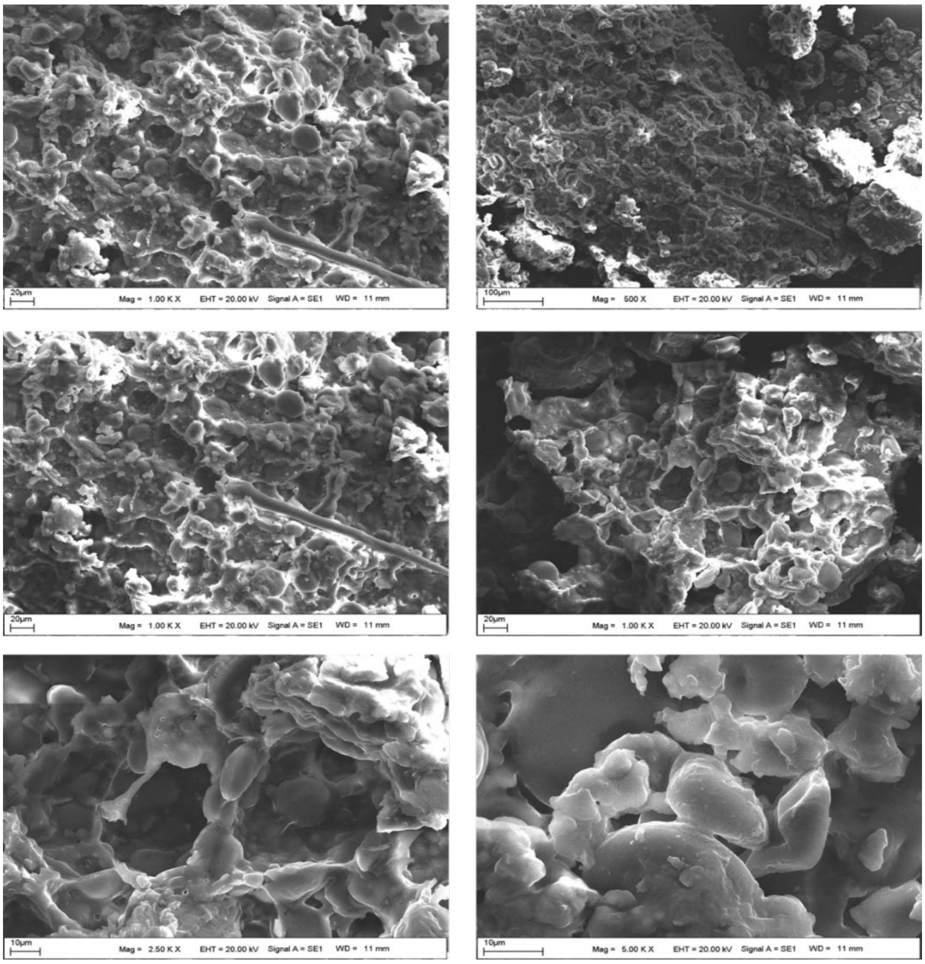

Figure 7. SEM Images of Wholemeal Diet Biscuit Sample at Different Magnifications.

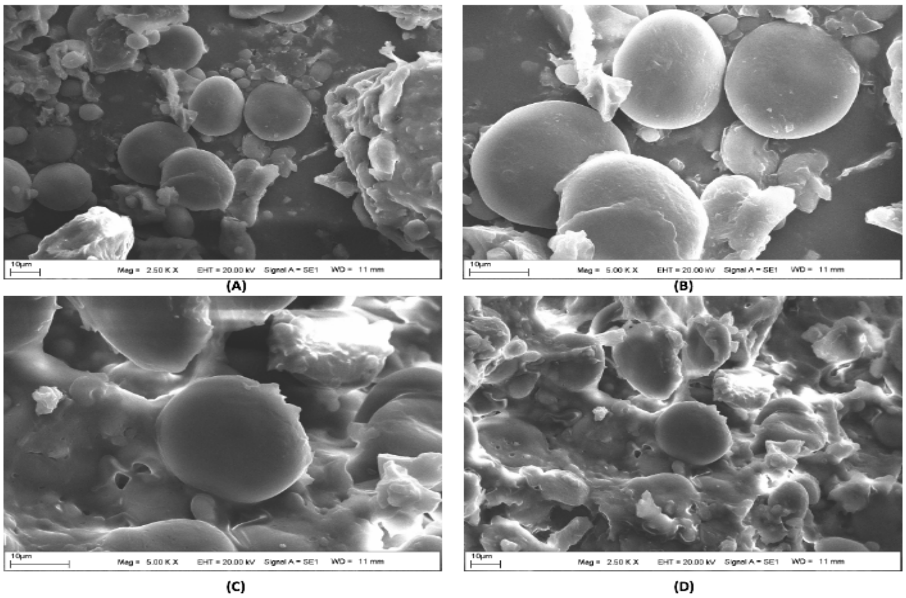

Figure 8. SEM Images of Starch Structures in Diet Biscuit Samples at Different Magnifications.

EDX spectra of diet biscuit samples were taken for a detailed surface analysis. The results of these analyzes are given comparatively in figure 9. When we look at the surface analysis results of this diet biscuit, although the peak intensities are different, we see the surface peaks of Ca, K, Mg, Na elements as well as P and Cl elements.

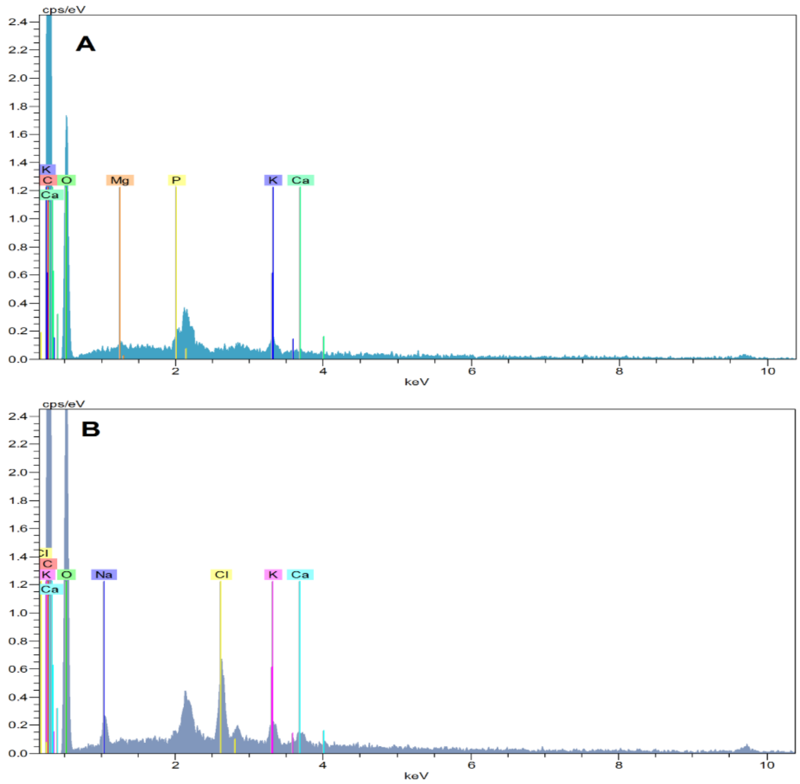

Figure 9. EDX Spectra for Lemon-Fiber diet biscuit (A) and wholemeal diet biscuit (B) samples

4. Conclusion

The moisture content of the diet biscuit samples used in the study was determined as 2.85 ± 0.06% in the lemon-fiber diet biscuit sample and 2.77 ± 0.09 in the diet biscuit sample with whole wheat. Accordingly, four different mass losses are observed in the lemon-fiber diet biscuit samples. First mass loss (75-120°C) from removal of structural moisture, second mass loss (140-240 °C) from decay of flavoring aromatic compounds, third mass loss (240-400 °C) from structural carbohydrate and starch degradation, fourth mass loss (400-580 °C) is due to the fiber in the structure. There are three different mass losses in the example of wholemeal diet biscuit. The first (65-120 °C) is the moisture in the structure, the second (180-400 °C) is the structural carbohydrate and starch, and the third (400-500 °C) is the degradation of the bran structures. Ash contents of diet biscuit samples were determined by TGA analysis. Ash contents were determined as 6.51 ± 0.18% in the lemon-fiber diet biscuit sample, and 7.47 ± 0.04% in wholemeal diet biscuit. Thermal degradation of the lemon-fiber diet biscuit sample started at 250 °C in four stages and was completed at 585 °C. It was observed that there was a three-stage thermal degradation in wholemeal diet biscuit samples started at 229 °C and completed at 580 °C. 46.06 % C, 1.60 % N, 0.12 % S and 6.68 % H elements were observed in the samples of lemon-fiber diet biscuit. 45.51% C, 2.39% N, 0.15% S and 7.05% H elements were obsevde in the sample of wholemeal diet biscuit. In FTIR spectra, three main peaks originating from carbohydrate, protein and fat structure are clearly observed. In the lemon-fiber diet biscuit sample, there is an esteric carbonyl stretch vibration around 1820 cm^{-1}, unlike the whole wheat diet biscuit, due to the aromatic esters that give the lemon flavor. When we look at the AAS results, we see different amounts of Ca, K, Mg, Na, Zn elements in both diet biscuit sample contents. Since a structural characterization has been carried out for the first time in dietary biscuit samples with this study, the elements determined by the AAS technique are in harmony with the biscuit samples previously made [31]. When the micromorphologies of the surface are examined with SEM, a very fractal, monolithic structure is seen in the lemon-fiber diet biscuit sample. Wholemeal diet biscuit sample exhibits a high cavital structure. Spherical starch structures are present in both biscuit samples. Surface analyzes are detailed with EDX spectra. In the EDX spectrum, we see the peaks of Ca, K, Mg, Na elements as well as P and Cl elements with different intensities on the dietary biscuit sample surfaces. We can explain both structural and superficial content differences of diet biscuit samples as follows. We know that flour containing wheat flour and wheat bran are used in the production of snacks such as biscuits etc. snacks. Contrary to expectations, flour contents are extremely complex and the composition can vary. It has different ingredient contents in bakery products (biscuits, etc.). Different ingredient contents vary greatly depending on the genetic potential of the selected wheat, growing conditions and climate. Especially the main source of mineral content in flour is soil. The amount of mineral matter obtained from the soil depends largely on the soil type, rainfall, the type and amount of fertilizer applied and environmental conditions [32]. In addition; The prominence of the P peak is due to phytic acid molecules in the structure of wholemeal [33]. Cl is caused by NaCl added to biscuit samples for aroma and preservative purposes [34]. As a result, the study showed us that the moisture, ash content, chemical structure properties, surface morphology-structure properties and thermal properties of the diet biscuit samples are different from the classical biscuit structures. The study is the first comprehensive original study done with diet biscuits. For this reason, the study will provide positive contributions to the literature in terms of originality. When we look at the literature, we see that food analysis is carried out with laborious, time-consuming traditional test methods. This study was carried out using modern estrumental techniques. The techniques used have enabled us to make interpretations both faster and with more precise results. The study has also brought a different perspective to food analysis.

Conflict of interest

The authors declare that they have no conflict of interests.

References

[1] Caleja C., Barros L., Antonio A.L., Oliveira M.B.P.P., Ferreira I.C.F.R., A comparative study between natural and synthetic antioxidant: Evaluation of their performance after incorporation into biscuits, *Food Chem.,* 216 (2017) 342-346.

[2] Chauhan A., Saxena D.C., Singh, S., Total dietary fibre and antioxidant activity of gluten free cookies made from raw and germinated amaranth (Amaranthus spp.) flow, *LWT-Food Sci Technol.,* 63(2) (2015) 939-945.

[3] Psodorov Đ., Ačanski M., Psodorov D., Vujić, Đ., Pastor K., Determination of the content of buckwheat and wheat flours in bread using GC-MS and multivariate analysis, *J. Food Nutr. Res.,* 54(2) (2015) 179-183.

[4] Vujıc´ L., Cepo D.V., Sebecıc B., Dragojevic´ I.V., Effects of pseudocereals, legumes and inulin addition on selected nutritional properties and glycemic index of whole grain wheat-based biscuits, *J. Food Nutr. Res.,* 53(2) (2014) 152-161.

[5] Boobier WJ., Baker JS., Davies B., Development of a healthy biscuit: an alternative approach to biscuit manufacture, *Nutr.,* 5(7) (2006) 1-7.

[6] Klunkin W., Savage G., Biscuits: A substitution of wheat flour with purple rice flour, *AFSE.,* 2(3) (2018) 81-97.

[7] Nakov G., Stamatovska V., Ivanova N., Damyanova S., Godjevargova T., Komlenic D.K., Physicochemical characteristics of functional biscuits and in vivo determination of glucose in blood after consumption of functional biscuits, *J. Hyg. Eng. Des.,* 22 (2018) 25-32.

[8] Kolberg DI., Prestes OD., Adaime M.B., Zanella R. A New Gas Chromatography/Mass Spectrometry (GC-MS) Method for the Multiresidue Analysis of Pesticides in Bread., *J. Braz. Chem. Soc.,* 21(6) (2010) 1065-1070.

[9] Cerci A., Cakmak Aksan A., Gümüs D., Pazarbası İ., Determination of the diet product consumption status of adults between, *Hacettepe University Faculty of Healt Sciences Journal.*, 7891 (2015) 103857.

[10] Yıldırım Mavis Ç., Ozay Arancıoglu İ., Aldemir N., Assesment of clients'awareness and consumption frequency of diet food products who consulted to nutrition and diet counseling, *Haliç University Journal of Health Sciences.,* 1 (2018) 15-29.

[11] Guinè RPF., Barroca M.J., Pereira D., Correia P.M.R., Adsorption isotherms of Maria biscuits from different brands, *J. Food Process Eng.,* 37(3) (2014) 329-337.

[12] Diukareva G., Pak A., Gasanova A., Determination of storage conditions for new biscuits using their sorption isotherms, *Ukrainian Food Journal.,* 3(2) (2014) 249-256.

[13] Passos M.E.A. dos, Moreira C.F.F., Pacheco M.T.B., Takas I., Lopes, M.L.M, Valente-Mesquita V.L., Proximate and mineral composition of industrialized biscuits, *Food Sci. Technol.,* 33(2) (2013) 323-331.

[14] Cayuela-Sánchez Y.A., Palarea-Albaladejo J., Zira T.P., Moriana-Correro E., Compositional method for measuring the nutritional label

components of industrial pastries and biscuits based on Vis/NIR spectroscopy, *J. Food Compost Anal.,* 92 (2020) 103572.

[15] Kalendar M., Akosman C., Drying behavior of sunflower seeds in infraraed dryer and adaptation analysis to during models, *Fırat University Journal of Engineering,* 27(1) (2015) 51-56.

[16] Adeola A.A., Ohizua E.R., Physical, chemical and sensory properties of biscuits prepared from flour blends of unripe cooking banana, pigeon pea, and sweet potato, *Food Sci. Nutr.,* 6(3) (2018) 532-540.

[17] Ullah F., Ahmad S., Wahap S., Zeb A., Khattak M.K., Khan S., Kang M., Quality evaluation of biscuits supplemented with alfaalfa seed flour, *Foods.,* 5(4) (2016) 68.

[18] Kumar A., Krishnamoorthy E., Devi H.M., Uchoi D., Tejpal C.S., Ninan G., Zynudheen A.A., Influence of sea grapes (caulerpa racemosa) supplementation on physical, functional and antioxidant properties of semi-sweet biscuits, *J. Appl. Phycol.,* 30 (2018) 1393-1403.

[19] Ag H.O., Okoli, N.A., Physico-chemical, sensory, and microbiological assesments of wheat-based biscuit improved with beniseed and unripe plantain, *Food Sci. Nutr.,* 2(5) (2014) 464-469.

[20] Erinc H., Mert B., Tekin A., Different sized wheat bran fibers as fat mimetic in biscuits: its effects on dough rheology and biscuit quality, *J. Food Sci. Technol.,* 55(10) (2018) 3960-3970.

[21] Amin. T., Naik H.R., Hussain S.Z., Mir M.A., Jabeen A., In vitro digestion, physicochemical and morphological properties of low glycemic index rice flour prepared through enzymatic hydrolysis, *Int. J. Food Prop.,* 21(1) (2018) 2632-2645.

[22] Costa L.A. Des., Fonsèc A.F., Pereira, F.V., Druzıa J.I., Extraction and characterization of cellulose nanocrystals from corn stover, *Cell Chem Technol.,* 49(2) (2015) 127-133.

[23] Majzoobi M., Hansouri H., Mesbahi G.H., Farahnaky A., Golmakani M.T., Effects of sucrose substituation with date syrup and date liquid sugar on the physiochemical properties of dough and biscuits, *J. Agric. Sci. Technol.,* 18 (2018) 643-656.

[24] Díaz A., Bomben R., Dini C., Viña S.Z., García M.A., Ponzi M., Comelli N., Jerusalam artichoke tuber flour as a wheat flour substitute for biscuit elaboration, *LWT-Food Sci. Technol.,* 108 (2019) 361-369.

[25] Ahmad N., Sahrin N., Talib N., Ghani F.S.A., Characterization of energy content in food waste by using thermogravimetric analyser (TGA) and elemental analyser (CHNS-O), *J. Phys. Conf. Ser.,* 1349 (2019) 1-8.

[26] Esselink E.F.J., Aalst H.van., Maliepaar M., Duynhoven, J.P.M. van., Long-term storage effect in frozen dough by spectroscopy and microscopy, *Cereal Chem.,* 80(4) (2003) 396-403.

[27] Van Velzen E.J.J., Van Duynhove J.P.M., Pudney P., Weegels P.L., van der Maas J.H., Factor associated with dough stickiness as sensed by attenuated total reflectance ınfrared spectroscopy, *Cereal Chem.,* 80(4) (2003) 378-382.

[28] Doner G., Ege A., Evaluation of digestion procedures for the determination of iron and zinc in biscuits by flame atomic absorption spectrometry, *Anal. Chim. Acta.,* 520 (2004) 217-222.

[29] Pereira D., Correi P.M.R., Guinè, R.P.F., Analysis of the physical-chemical and sensorial properties of Maria type cookies, *Acta Chim. Slov.,* 6(2) (2013) 269-280.

[30] Tavares B.O., Sılva E-P da., Sılva V-S-N.da., Soares Jùnio M.S., Ida E.I.,Damıanı C., Stability of gluten free sweet biscuit elaborated with rice bran, broken rice and okara, *Food Sci. Technol.,* 36(2) (2016) 296-303.

[31] Barbarisi C., De Vito V., Pellicano MP., Boscaino F., Balsamo S., Laurino C., Sorrentino G., Volpe MG., Bread chemical and nutritional characteristics as influenced by food grade sea water, *Int. J. Food Prop*., 22(1) (2019) 280-289.

[32] Gül H., Determination of the effects of corn and wheat brans addition on dough and bread characteristics, Ph.D. Thesis., Department of Food Engineering Institute of Natural and Applied Sciences University of Çukurova, (2007).

[33] Ravindran V., Ravindran G., Sivalogan S., Total and phytate phosphorus contents of various foods and foodstuffs of plant origin, *Food Chem.,* 50 (1994) 133-136.

[34] Zaky AS., Pensupa N., Andrade-Eiroa Á., Tucker G.A., Du C., A new HPLC method for simultaneously measuring chloride, sugars, organic acids and alcohols in food samples, *J. Food Compos. Anal.,* 56 (2017) 25-33.