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Walnut Paste: A Healthy Alternative for Nutella Consumers

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Abstract: Nutella is the best-selling chocolate and hazelnut spread in the world. It is known that the main ingredients of Nutella are sugar (55%) palm oil (23%), hazelnuts (14%), cocoa solids and skimmed milk (8%). Currently, worldwide, there is a tendency to make healthier food choices, and the sugar and palm oil from Nutella's formulation are ingredients blamed for causing certain health disorders (obesity, cardiovascular diseases and diabetes). In this order, the purpose of the research was to develop the technology for obtaining walnut paste, since walnuts have proven prophylactic properties, with a low sugar content and without palm oil. The quality of the elaborated walnut paste was determined and monitored during the shelf life in terms of oxidative stability (Acidity index (FFA) and Peroxide value (PV)), total phenols content, antioxidant activity and rheology. The obtained results demonstrated that during 4 months of storage the acidity index of the walnut paste did not register significant changes, reaching maximum values of 0.17±0,01 (Oleic g/100 g), while the peroxide values evolved up to 2.22±0,03 meq/kg oil. The slow evolution of the oxidative parameters can be due to the phenolic compounds in walnut paste that recorded a value of 47.2±0,36 mg GAE/g with an antioxidant activity (DPPH free radical scavenging) of 70±1,02 %. Concerning sensory characteristics, the walnut paste samples were positively appreciated in comparison with Nutella and no significant quality difference was observed after storage for 4 months.

Keywords: Walnut paste, Oxidative stability, Phenolic compounds, Antioxidants

Introduction

The walnut (Juglans Regia L.) fruit is considered one of the most consistent foods, and walnut culture is specified as a strategic direction for human nutrition and included by FAO and WHO in the list of priority plants (Gandev, 2007). Because of the high fat content, walnut kernels were not considered a healthy food until recently. This perception has changed a lot lately, because it has been found that they have a healthy polyunsaturated fatty acid profile, are rich in proteins, vitamins and minerals. Many authors report that walnut kernel contains a large amount of lipids (> 50% of the weight), 11% proteins, 5% carbohydrates and is very caloric (approx. 525 kcal/100 g). Walnut lipids have a high content of unsaturated fatty acids (up to 90%), including polyunsaturated fatty acids (PUFA) (up to 78% of the total fatty acid content), which play an essential role for the proper functioning of the human body. They also contain appreciable amounts of dietary fiber, vitamins (E, B3, B5, B6) and mineral elements (K, P, Mg) (Chatrabnous et al., 2018; B. Liu et al., 2020; Martínez et al., 2010).

The importance of walnut culture is determined by its multi-functional utility that includes food, medicine, dyes, adhesives, cosmetics, oils, furniture and sculpture (Guasch-Ferré et al., 2018; L. Liu & Dai, 2021; Rusu et al., 2020). The interest in walnuts is also determined by the nutritional value, which derives from their unique

composition, with certain nutrients and phytochemicals responsible for multiple beneficial effects of consuming walnuts and derived products (Ni et al., 2022; Ros et al., 2018). The objective of combining walnut kernel with other raw materials is to diversify and improve the nutritional and organoleptic qualities of traditional food products, but also to obtain so-called functional foods. Thus, by using certain strategies in the reformulation of food product matrix, food with a specific composition (eg: reduction of animal fat and sodium content, fortification with various bioactive substances, etc.) and acceptable physico-chemical and organoleptic properties is obtained (Otunola & Martirosyan, 2021). Due to the beneficial effects of walnut consumption on human health demonstrated by numerous researches, there has been increased interest in the development of new food products based on walnuts, such as walnut milk, various fillings for pastry products, walnut flour. Some researchers have tried to produce meat products containing walnut kernel (Ayo et al., 2005; Cofrades et al., 2004). Some studies related to the production of drinks and emulsions using walnut kernels have also been carried out (Gharibzahedi et al., 2012; Ouyang et al., 2022).

The walnut sector is a traditional branch for Republic of Moldova, being favored by the moderate climate, the fertile soils, the possibilities of cultivating the most valuable varieties from the world selection (Zimny, 2012). As in other regions of the world, the market leader of the nut spread industry in Republic of Moldova is the well-known *Nutella* spread. Today, Ferrero's product is a global success. There are studies that mention that on one hand a jar of Nutella is sold worldwide every 2.5 seconds and on the other hand there are health and environmental damage concerns due to the use of palm oil in Nutella spread manufacturing (Cova & D'Antone, 2016; Silva, 2016). Taking into account the fact that walnuts are a local product for the Republic of Moldova and that there are no Moldovan products similar to the "Nutella" spread on the market, the prime objective of this research was to develop a walnut paste with cocoa and chocolate addition that is similar to Nutella using locally grown walnuts.

Materials and Methods

Materials

Walnuts from *Pescianski* variety used in the research were purchased from an orchard in the Republic of Moldova. In walnut paste formulations was also used cocoa, dark chocolate (95% cocoa) and powdered sugar that were bought from a local supermarket in Moldova.

Methods

Preparation of Walnut Paste

After walnuts shelling, the kernel was roasted in a convection oven (Rational SCC 61E, Germany) at 170 °C for 10 min. The roasted walnut kernel was then shaken on a sieve and winnowed in order to remove part of the kernel skin, which, on the one hand, is very rich in polyphenols (Jahanban-Esfahlan et al., 2019), and on the other hand gives the final product a bitter taste (M. Liu et al., 2021). The walnut kernel (83.5%), powdered sugar (10.0 %), dark chocolate (5.0 %) and cocoa (1.5 %) was then blended in a blender (KitchenAid 5KSB4026, United Kingdom) at 8000 rpm until a fine homogeneous paste was obtained. The paste was then distributed in jars, covered and kept in a dark place in order to monitor its quality during storage.

Acidity Index (FFA)

In a 250 ml volumetric flask weighed 5 g of sample with the precision of 0.01 g. Then were added 25 ml of hexane and 25 ml of ethyl alcohol. The potentiometric method consists of adding 0.1N NaOH until the pH of the solution reaches the value of 8.1(*AOCS Official Method Cd 3d-63*, 1999).

$$A(\%\ oleic\ acid) = \frac{28{,}2 \cdot N\ (NaOH) \cdot V}{m}$$

where:
V– volume of sodium hydroxide, [ml]
N – concentration of sodium hydroxide, [mol/dm^3]
m – mass of sample, [g].

Peroxide Value (PV)

In a 250 ml volumetric flask were weighed 2 g of the sample, were added 10 cm^3 of hexane, the analyzed sample was quickly dissolved, then added 15 cm^3 of 15% glacial acetic acid and 1 cm^3 of 50% potassium iodide solution, the obtained solution was mixed for 1 minute and placed in a dark place for 15 minutes. Then was added 75 cm^3 of distilled water, mixed and titrated with sodium thiosulfate solution (5,09 mM) until a pale blue colour appeared and was stable for 5 seconds. The titration was performed in the presence of 1% starch indicator solution (*AOCS Official Method*, 2003).

$$\mathrm{PV} = \frac{(\mathrm{V}_s - \mathrm{V}_{ref}) \cdot \mathrm{N} \cdot 1000}{\mathrm{m}}$$

where:
V_{ref} – volume of titrant used for blank titration, [ml]
V_s – volume of titrant used for sample titration, [ml]
N – normality of sodium thiosulfate solution.

Total Phenolic Content (TPC)

Total phenolic content was performed using a Shimadzu 1800 UV/Vis spectrophotometer, at 765 nm wavelength, using a 10 mm quartz cuvette. The results of the total phenolic content, expressed in mg GA/100g of dry weight, were obtained using the GA calibration curve (y = 0.0041x-0.1331, R^2=0.9924) (Šarolić et al., 2014).

DPPH Antioxidant Activity

The antioxidant activity of the samples was performed using a Shimadzu 1800 UV/Vis spectrophotometer and expressed as % inhibition of DPPH using the following equation (Šarolić et al., 2014):

$$AA\% = \frac{A_0 - A_t}{A_0} \cdot 100\%$$

where:
A_0 - the absorbance of the DPPH solution at the initial time of 0 s;
A_t - the absorbance of the DPPH solution after 30 min of incubation.

Rheological Properties

The viscosity of the nut paste was determined using the BROOKFIELD DVIII Ultra rotary viscometer at 25±0.2 °C.

Sensory Analysis

The sensory analysis of the walnut paste was carried out at the Department of Food and Nutrition, Technical University of Moldova. 20 panelists (aged 20 to 60 years old) rated the quality of the nut paste using a 5-point hedonic scale, from 1 - extremely dislike to 5 - extremely like (Covaliov et al., 2022). Sensory parameters such as taste, color, texture, flavor, spreadability and overall acceptability were evaluated in comparison with Nutella spread.

Statistical Analysis

All the determination were performed in triplicate with exception of sensory analysis as mentioned above. The results are presented as mean ± standard deviation (SD). Student's *t*-test was used for comparison between two means. Physicochemical properties and sensory acceptability results were analyzed with ANOVA software (2020 version); Tukey test (0.05 significance level) was applied for the comparison of mean values.

Results and Discussion

Oxidative Stability

The most common cause of the deterioration of fatty raw materials during storage is lipid peroxidation, which largely depends on the accessibility of oxygen and results in toxic oxidation products (Schwember & Bradford, 2010). The accumulation of peroxides causes the reduction of antioxidant enzymes, their antioxidant capacity and the viability of oilseeds (Bailly et al., 2002).The changes of the acidity and peroxide values of the walnut paste were monitored during storage for 4 months (Table 1). The acidity index is an important quality index for food fats and for food products with advanced fat content. According to Ghasemnezhad and Honermeier (2009), the composition and proportions of free fatty acids is one of the factors that determines susceptibility to fat degradation, because unsaturated fatty acids are much more susceptible to oxidation than saturated fatty acids (Ghasemnezhad & Honermeier, 2009).

Table 1. Physicochemical properties of walnut paste during storage

Storage time, months Parameter	0	1	2	2	4
FFA, Oleic acid g/100 g	0.10±0.01	0.10±0.01	0.13±0.01	0.12±0.01	0.17±0.01
PV, meq/kg	1.17±0.09	1.19±0.05	1.15±0.07	1.81±0.05	2.22±0.11

From the data presented for the oxidative stability indices, no legitimacy is observed in the FFA or PV values during the storage of the walnut paste. Thus, during 4 months of storage, the FFA values oscillate in the range of 0.10-0.17 g Oleic acid/100 g, and the PV values within the limits of 1.15 - 2.22 meq/kg. The oscillations recorded by these parameters can be explained by the fact that at certain stages, some compounds are involved in other reactions, passing into other forms: for example, fatty acids undergo a rearrangement, turning into conjugated dienes, which later, under the action of oxygen, turn into peroxides. Peroxides in their turn will undergo changes thus forming compounds such as aldehydes, ketones, etc., the presence of which degrades the quality of the final product (Abeyrathne et al., 2021; Subotin et al., 2021). According to Ampofo and Grilo (2022), the peroxide value of walnut oil reaches 1.80 meq O_2/kg after 4 months of storage, while FFA ranges within 0.02 – 0.03 Oleic acid g/100g (Ampofo et al., 2022). The difference in FFA content of our walnut paste and the values reported in the previously mentioned study can be linked to the low moisture contents of kernels, thus the lipase activity for FFA formation is limited. Our results are in agreement with the data obtained for the walnut paste (with no addition) accelerated storage study reported by Dordoni et al. (2019). On the other hand, Pourfarza et al. (2020) showed a different behavior of lipids in the case of hazelnut butter production, mentioning a simultaneous increasing trend for both FFA and PV during storage (Pourfarzad & Shokouhi Kisomi, 2020).

Total Phenolic Content and Antioxidant Activity

Current trends in the food field place more and more emphasis on the biologically active potential of certain ingredients due to their effect on human health (Nile & Park, 2014). Polyphenols are categorized as biologically active substances that exhibit free radical scavenging capacity. The total phenolic content and the antioxidant activity of formulated walnut paste is shown in Table 2.

Table 2. Physicochemical properties of walnut paste during storage

Storage time, months Parameter	0	1	2	2	4
TPC, mg GAE/g	47.20±1.23	44.73±0.93	41.43±0.56	37.64±0.65	32.56±0.14
DPPH, %	70.00±1.54	67.31±2.01	61.29±1.32	55.11±0.98	46.65±0.86

The data presented indicate a decrease in the total content of phenolic substances by about 31% after 4 months of storage, and the antioxidant activity, which initially reached a value of 70%, recorded a reduction of 33.35%. The tendency of the values of these parameters to decrease can be explained by the property of polyphenols to be oxidized. Bakkalbaşi et al (2012) reported a walnut phenolic content that ranged within 9.31 – 31.80 mg GAE/g (Bakkalbaşı et al., 2012). In our study the total phenolic content of walnut paste was higher, probably due to the intake of phenolic substances from cocoa, which can reach values up to 106.77 mg GAE/g (Borja Fajardo et al., 2022), and chocolate. In the same context, Dordoni et al. (2019) reported a total phenolic content of walnut paste (with no addition) of 406.9 mg GAE/100 g that showed an upward trend to 465.79 mg GAE/100 g in the 9th day of storage at 60 °C when simulating 2 years storage at 20 °C (Dordoni et al., 2019).

However, it is difficult to compare the values of the total phenolic content obtained in different researches because Folin's method requires the use of different extraction methods and solvents. Multiple researches have demonstrated the direct relationship between the total content of phenolic compounds and the antioxidant activity of vegetable raw materials (Bors & Michel, 2002; Bunea et al., 2012; Gramza et al., 2006). In our study the correlation coefficient is *r=0.99*. This is due to the fact that phenolic compounds actually act as antioxidants contributing to free radical scavenging, thus promoting healthy effects on human body (Chiva-Blanch & Visioli, 2012).

Viscosity

Rheology is the science of the flow and deformation of materials under stress and deformation. In the food industry, rheological data are needed for studying ingredient functionality in product development, determining food texture by correlating with sensory data, immediate or final product control, and process engineering calculation for equipment, etc. (Fischer & Windhab, 2011). The results of rheological properties of developed walnut paste, in terms of viscosity are presented in figure 1.

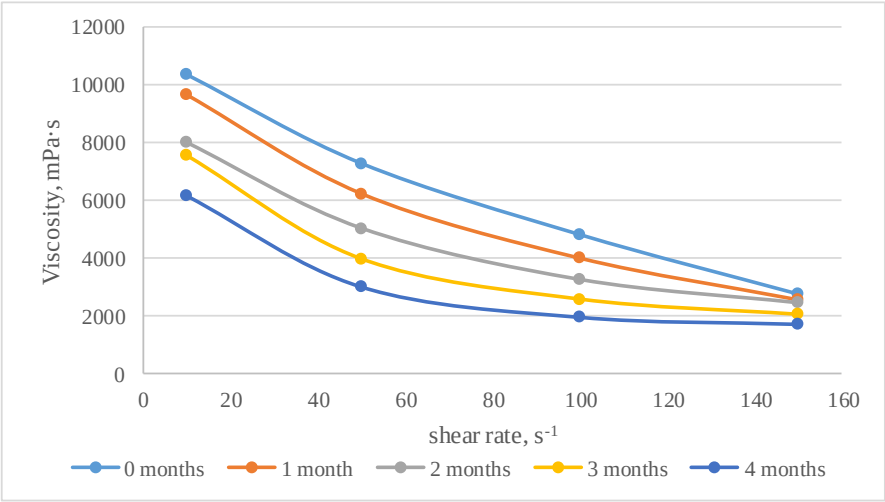

Figure 1. Evolution of apparent viscosity of walnut paste during storage

During the storage period the walnut paste exhibited non Newtonian pseudoplastic behavior, fact confirmed by the values of the apparent viscosities that showed an downward trend when the shear rate increased from 10 to 150 s^{-1} (Rehm et al., 2012). It is also worth mentioning that the reduction of apparent viscosity values during storage is due to the coalescence process of the fat droplets manifested by their fusion and the creation of an oily layer on the surface. Several researches were done in order to avoid the oil separation in nut butters or spreads. In order to avoid the oil separation in penut butter, Gills et al. (2008) tried to use palm oil and hydrogenated vegetable oils. However the results showed that the only factors that affect the quantity of separated oil are temperature and storage time (Gills & Resurreccion, 2000). Ardakani managed to reduse the oil leakage in pistachio butter using two types of emulsifiers: lecithin and mono-di glycerides (Ardakani et al., 2006). Ereifej et al. (2005) states that powdered sugar, pectin and gum arabic manage to decrease oil leakage in halva, however the differences were not significant compared to control sample (Ereifej et al., 2005).

Sensory Analysis

The results of sensory analysis of walnut paste in comparison with *Nutella* spread are summarized in Table 3.

Table 3. Physicochemical properties of walnut paste during storage

Parameter Product	Taste	Flavor	Color	Texture	Spreadability	Overall acceptability
Walnut paste	4.90±0.03	4.90±0.03	5.00±0.06	4.5±0.05	3.7±0.08	4.58±0.05
Nutella	4.50±0.04	4.70±0.05	5.00±0.00	4.8±0.08	5.0±0.00	4.8±0.03

The developed walnut paste had a mean 'overall' acceptability score of 4.58. The panelists deemed the walnut paste between "extremely like" and "neither like nor dislike". This score was lower than the score obtained by Nutella spread (4.8). The acceptability score of walnut paste was in a great measure affected by 'spreadability' compared to Nutella's spreadability. The texture of walnut paste was described as being more fluid than spreadable. The walnut paste had higher scores than the Nutella spread in terms of taste and flavour. Some panelists mentioned that the taste of Nutella is excessively sweet, while the taste of the developed walnut paste was appreciated as moderately sweet with a pleasant flavour of roasted walnuts and an "interesting" aftertaste.

Conclusion

The formulated paste had a high bioactive potential in terms of total phenolic content and antioxidant activity. The investigation of oxidative stability showed that the product is relatively stable during storage, registering slight fluctuations in the value of acidity and peroxide, however, falling within the limits recommended by the normative documents. The sensory analysis showed that the walnut paste was within the acceptance range of the hedonic scale of 4 – 5. Thus, walnuts can be successfully used in the production of a healthy alternative of *Nutella* spread. The developed product would lack an excessive amount of sugar and palm oil, at the same time it would have a good potential on the market of the Republic of Moldova.

Scientific Ethics Declaration

The authors declare that the scientific ethical and legal responsibility of this article published in EPHELS journal belongs to the authors.

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