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The Effect of Packing Material on Storage Stability of Sunflower Oil

Grujić Slavica[1,1], Aleksić Vojislav[2], Vukić Milan[2], Petrović Zoran[2]
[1] Faculty of Technology, University of Banja Luka, Banja Luka, Bosnia and Herzegovina
[2] Faculty of Technology Zvornik, University of East Sarajevo, Bosnia and Herzegovina

Abstract: Edible plant oils, including sunflower oil, with high content of unsaturated fatty acids, are subject to hydrolytic and oxidative deterioration during storage. This study investigates the impact of packing material, clear uncoloured glass bottles and clear uncoloured polyethylene terephthalate (PET) bottles, in which the refined sunflower oil was packaged, and impact of storage conditions, on the speed of oil deterioration. Oil was filled in the bottles, hermetically sealed and kept in a dry place, at room temperature, and illuminated by artificial light. Results of the oxidative stability of sunflower oil assessment during six month storing in controlled conditions, showed that photo-oxidation adversely has an affect on product quality characteristics and coses forming of primary oxidation products. Peroxide number in oil, immediately after bottling was 0,230 mmol O_2/kg oil, while after 6 months of storage value reached 7,800 mmol O_2/kg oil (in glass bottle), and 8,600 mmol O_2/kg oil (in PET bottle). Anisidine number in oil immediately after bottling was 5,5 and after 6 months of storage in a glass bottle it was 39,6 and 57,40 when oil was packaged in PET bottle. Averages of total oxidation values in sunflower oil packed in glass and PET packaging increased from 5,5 (immediately after bottling) to 55,20 in oil packaged in glass bottle, and 57,40 in oil packaged in PET packaging. The other results obtained in the paper confirmed the need for protecting sunflower oil against oxidative spoilage during storage, providing packaging in colored packages and storing on dark, dry place.

Keywords: sunflower oil, oxidative stability, packing material

Introduction

Fats and oils play an important role in the formation of aroma, flavour and texture of food products and impact on food nutritive value. Fats and oils are often added to food products as ingredient during processing. Final food products may contain pure oil or fat, or may be composite complexes of fats and proteins, carbohydrates, water and other ingredients. Oil is available on the market packed in different packages, with very attractive design, made of clear, transparent packaging materials. Packaging should be attractive for purchasers or consumers, but it also should protect fats or oils from the point of processing and packaging, during storage and to the moment of consumption as foodstuff or its ingredient. Glass and PET are suitable for food contact materials for edible oil packaging, and provide adequate protection during oil shelf life. Monitoring changes in fats or oils during products storage is an important factor with influence on developing and manufacturing of high quality products. These changes depend on the type of lipids, its origin and content, or products composition.

As it is well known, ingredients incorporated into food product as a complex system, change over time and can obtain new physical and chemical properties. The oxidation of fats is one of the most common chemical changes that has a significant impact on flavour, aroma, nutritional quality and texture of products (Grujić, 1994; Shahidi and Zhong, 2005; Craven and Lencki, 2011; Yettella and Castrodale, 2011). Compounds formed during the oxidation of the lipids are responsible for rancid flavours and aromas forming. Considering the fact that the oxidation of fat is not the only cause of spoilage of food products, the hydrolysis of lipids should be also mentioned as a frequent provoker of lipids deterioration. In practice, two types of food spoilage most frequently occur: hydrolytic rancidity and oxidative rancidity. Hydrolytic rancidity

[1] Corresponding author e-mail: grujicslavica@yahoo.com

is the result of the formation of free fatty acids and soaps. Reactions of this type develop in reaction of triglycerides and water in the presence of a catalyst or enzyme lipase. Oxidative rancidity is the result of a wide range of reactions that occur under the influence of oxygen. The process of lipid oxidation takes place in three phases: an initiation or induction phase, a propagation phase, and a termination phase.

Oxidation of unsaturated fatty acids is the main reaction responsible for the degradation of lipids (Muik, Lendl, Molina-Dıaz, and Ayora-Canada, 2005; Morales, Marmesat, Dobarganes, Marquez-Ruiz and Velasco, 2011). Indeed, the oxidation level of oil and fat is an important quality characteristic for food industry. Under mild conditions, molecular oxygen reacts with the double bonds following a free radical mechanism, and so-called auto-oxidation reaction takes place. The oxidation of fats primarily means deterioration of their quality and safety for consumption, and as a result economic loss can be expected. In addition, oxigen species that have carcinogenic effects are created during the oxidation, and they can lead to distortion of the cardiovascular system and can decrease the safety of oil for consumption (Pezzuto and Park, 2002).

Many methods of analysis have been developed to access the extent of oxidative deterioration, which are related to the measurement of the concentration of primary or secondary oxidation products or of both. The processes of lipid oxidation and deterioration progress in oils of plant and animal origin and in products containing them, can be identified and followed up with other tests, such as: Active Oxygen Test (AOM), Oxidative Stability Index (OSI), Iodine Number, Hexanal value, Headspace Profile, Free Fatty Acids (FFA), and Thiobarbituric acid (TBA) test (Shadi and Zhong, 2005) and with Fourier Transform Raman Spectroscopy, in recent times (Muik et al., 2005). Gas chromatography is used for the determination of volatile compounds formed during the lipid oxidation.

With intention to protect edible vegetable oil from the sunlight impact, it is bottled and stored in different packages, manufactured from material which can absolutely, partly or in no way transmit sunlight and artificial light (light-yellow, amber, green, transparent coloured or uncoloured glass and PET (polyethylene terephthalate) bottles, containers, or in metal cans inside coated with a protective lacquer (Nkpa et al, 1990; Caponio, Bilancia, Pasqualone, Sikorska and Gomes, 2005). During oil storage, it is necessary to conduct evaluation of the hydrolytic stability of the oil and its oxidative changes, periodically. Nkpa et al (1990) analysed oil changes during the storage period of 100 days, measuring values of free fatty acids, peroxide number and value of anisidine value. Abramovich and Abram (2005) investigated the sustainability of oil made from the seeds of native plants *Camelia sativa*, using a peroxide value of oil and anisidine value as indicators of changes and degrees of oil spoilage. Results published by Manzocco, Panozzo and Calligaris (2011) indicated that shelf life estimation of photosensitive foods under actual or accelerated conditions cannot be correctly achieved if the effect of light and temperature are not taken into account. Most of the studies focused on changes which take place in the oil in a shorter period of time (up to 6 months), although most manufacturers consider that a bottled oil is sustainable up to 12 months (Caponio et al., 2005), and so indicate it on the labels.

Light, oxygen, humidity and temperature are some of the external factors that adversely affect the composition and quality characteristics of fats and oils during and after processing. Light is the cause and initiator of the reactions that lead to the spoilage of fats and oils. Although fat does not absorb visible light spectrum, the oxidation can be induced by light which was absorbed by the oil impurities (for example, the pigment chlorophyll). It is generally accepted interpretation that auto-oxidation of fat includes formation of free radicals.

Contact of fat and oxygen may occur in different ways. Certain amount of oxygen can be retained or trapped in the oil during the production, filling and packaging. Oxygen may be kept in headspace of bottles, cans or vessels walls. Oxygen causes the formation of hydroperoxides, compounds that are associated with oil rancidity. Heating can affect the stability, safety and acceptability oil for consumption. Oil protection from the elevated temperature influence is rarely provided. Oil containers are usually made from materials with low insulating ability. Warm ambient temperatures encourage the onset of oxidation in a silent way. It is thus very important to evaluate the effects of photosensitized oxidation of lipids on quality characteristics of oils and fats. Auto-oxidation proceeds through a free radical chain reaction through the attack on the double bond at room temperatures. Peroxide is the main product that gives rise to objectionable flavour in food products.

Edible oil in Europe is usually packed in a transparent or coloured glass packaging or plastic packaging, and less often in lacquered metal packaging. There are often situations in which the vegetable oil is exposed to direct sunlight and artificial light in the system of distribution and retail operation. On the other hand, in household, oil is usually kept in closed dark place or room. A numerous researches were conducted on examination of oil quality during the production, processing and transport, but a few published papers are related to the examination of used packaging and packaging material impact to the oil quality during storage.

The aim of this paper was to investigate possibility for reducing the speed and degree of oil deterioration occurring as a result of hydrolytic and oxidative changes during storage. Influence of packaging materials (clear uncoloured glass bottle and PET bottle) in which the refined sunflower oil was packaged, also the impact of storage conditions on the speed of oil deterioration should be checked during the research.

Materials and Methods

Samples

Fresh refined sunflower oil (water content 0,11%), without the addition of any antioxidants, was supplied from local factory, sampled according to the BAS EN ISO 5555 method and filled in the bottles of one litre capacity.

Storage conditions

Bottles (0,25-0,40 mm thickness of PET bottle wall and 2,4-3,2 mm thickness of glass bottle wall) were filled with refined sunflower oil to the top, hermetically sealed and free space between the cap and product was minimal. Immediately after bottling (in a clear uncoloured glass bottles and in clear uncoloured PET bottles) and sealing, samples were kept in controlled conditions, on a dry and cold place (φ=55%, t=20^{o}C), illuminated by artificial lighting (110 lx). Samples of sunflower oil were divided into two groups depending on the type of packaging in which they were stored: (a) clear uncoloured glass bottles and (b) clear uncoloured PET bottles.

Analysis

Before bottling, refined sunflower oil was sampled from bulk (tank) according to the BAS EN ISO 5555 method. Bottled samples of sunflower oil were stored in controlled conditions for 6 months. Samples were transferred to the Laboratory before analysis in cardboard boxes. Immediately after bottling and then every two months, an analysis of oils was carried out determining: free fatty acids - acid value (BAS EN ISO 660), peroxide value (BAS EN ISO 3960), iodine value (BAS EN ISO 3961), saponification value (BAS EN ISO 3657) and anasidine value (BAS EN ISO 6885).

The total oxidation value (Totax) was calculated from the relationship [*1*], where abbreviations indicate: Totax - total oxidation value; PV - peroxide value; AV - anasidine value (Abramovich and Abram, 2005):

Totax = 2 (PV) + AV [*1*]

The sample and ambient temperature were measured by thermometer (HANNA“ HI 98810). Intensity of illumination was measured using a portable luminometer (3M™ Clean-Trace™ NG Luminometer). Results are presented as average values of four individual measurements.

Results and Discussion

Results of average values for the quality parameters determined in sunflower oil, packed in different types of packaging materials and stored for 6 months under controlled conditions are shown in the following tables 1-6.

The initial values in examined refined sunflower oil, immediately after bottling were: 0,11% water, peroxide value 0,230 mmol O_2/kg oil, acid value 0,060 mg KOH / g oil, iodine value 131 mg J_2/100 g and saponification number 186 mg KOH / g oil, anisidine value 5,5 and total oxidation value 5,96. The results of these parameter measurement showed that the sunflower oil samples had appropriate quality and were in accordance with current regulations on the quality of edible oils (Anonymous, 2011). The water content in examined samples during the six month storage was stable, without changes, since the package was hermetically sealed.

Deterioration of lipids in polyunsaturated oils, and sunflower oil is one of those oils, results in oxidation and changes of peroxide value. Table 1. shows that sunflower oil during storage in controlled conditions, on a dry and cold place (φ=55%, t=20°C), illuminated by artificial lighting, had an increase of the average peroxide values from 0,230 mmol O_2/kg oil measured immediately after bottling, to 7,800 mmol O_2/kg oil after six months storage in glass bottles, and to the highest value of 8,600 mmol O_2/kg oil after six months storage in PET bottles.

Table 1. Average peroxide values (mmol O_2/kg) sunflower oil stored in glass and PET bottles

Packaging type	Storage conditions	Peroxide value (mmol O_2/kg oil)			
		Storage period (months)			
		0	2	4	6
clear uncolored glass bottle	T=20°C, φ=55%, artificial lighting	0,230	0,940	3,780	7,800
clear uncolored PET bottle	T=20°C, φ=55%, artificial lighting	0,230	0,800	4,180	8,600

It is important to point out that the relatively high peroxide values were measured in sunflower oil stored in PET bottles after four month (4,180 mmol O_2/kg oil) and after six month oil storing (8,600 mmol O_2/kg oil), as demonstrated in Table 1. Obtained peroxide values are higher than maximal acceptable value of 7 mmol O_2/kg oil, as allowed by appropriate regulation (Anonymous, 2011). It could be established that the artificial light accelerates the oxidative processes, resulting in oil spoilage, what is in agreement with the results obtained by other authors (Nkpa et al., 1990; Nkpa et al., 1992; Abramovich and Adam, 2005; Ozkan, Simsek and Kuleasan, 2007; Raza et al., 2009).

In our research, we found that PET bottles provide less oil protection from destructive effect of the light during storage, in comparison with glass bottle protection, and that findings confirmed the results of the other researchers (Manzocco et al., 2011). Amber or green coloured glass bottles provide better protection for edible oils, but Nkapa et al (1990) could not prove it, based on peroxide value measuring. Abramovich and Adam (2005) also indicated that the peroxide number is not used for examination of the early stages of fat oxidation and its accuracy is questionable, but it is used in a numerous tests during examination of oil sustainability. They used the monitoring of changes in peroxide value in oil kept under different conditions, as good and helpful indicator for comparing the effect of different storage conditions on rate of oxidative development process in oil.

Table 2. The average acid number value (mg KOH / g) of sunflower oil stored in glass and PET bottles

Packaging type	Storage conditions	Acid number (mg KOH / g oil)			
		Storage period (months)			
		0	2	4	6
clear uncolored glass bottle	T=20^{O}C, φ=55%, artificial lighting	0,060	0,100	0,225	0,360
clear uncolored PET bottle	T=20^{O}C, φ=55%, artificial lighting	0,060	0,101	0,220	0,320

Table 2. represents the average acid values measured in sunflower oil samples, stored in transparent uncoloured glass and PET bottles. There was an increase in acid values of oil samples regardless of the type of packaging material used for packaging, glass bottles (0,360 mg KOH / g oil) and PVC bottles (0,320 mg KOH / g oil). Based on changes in acid value and peroxide value Caponio and co-authors (2005) found that olive oil can maintain its qualiy more than 12 months, if it is stored in a dark and cold place after packaging in bottles. A comprehensive study was carried out to investigate the extent of changes in sunflower oil under auto-oxidation and photo-oxidation conditions (Raza, Adnan, Qureshi, Asim, Najaf and William, 2009). The free-fatty acid (FFA) contents of the auto- and photo- oxidized samples increased from 0,056 to 0,290% and 0,056 to 0,369%, respectively.

Decomposition of triglycerides can occur under the enzyme lipase influence, in the presence of water (Freja, Mozzon and Lercker, 1999). However, in our examination, the measured values of water content in sunflower oil samples (0,11%) were relatively low and within the permitted values (Anonymous, 2011). The packaging in which the oil was stored was hermetically sealed, so it did not allow penetration of water inside the packaging. That is why is reasonable to exclude water as promoter of hydrolysis of triglycerides. It is possible that small amount of water pass through the walls of plastic containers. The effect of that water impact is negligible in our study, as can be confirmed by comparing the results obtained by measuring the acid number in oil packed in glass and PVC bottles (Table 2).

The increase in content of free fatty acids should be caused by endemic species of microorganisms, particularly thermophilic lipolytic fungi that can get into the oil in various stages of processing and transport within the plant. Several authors, as stated by Nkpa et al (1990), verified the presence of these bacteria in palm oil. Changes in free fatty acid values in analysed sunflower oil samples were statistically significant, showing the phenomenon of lipid oxidation (Tian, Dasgupta, Purnendu, and Shermer, 2000; Anwar, Bhanger and Kazi, 2007).

Table 3. The average iodine number values (mg J_2/100 g) of sunflower oil stored in glass and PET bottles

Packaging type	Storage conditions	Iodine number (mg J_2/100 g oil)			
		Storage period (months)			
		0	2	4	6
clear uncolored glass bottle	T=20^{O}C, φ=55%, artificial lighting	131	130	128	125
clear uncolored PET bottle	T=20^{O}C, φ=55%, artificial lighting	131	131	126	123

Decrease in iodine value is an indicator of lipid oxidation (Naz, Sheikh, Saddiqi and Sayeed, 2004). After four months of sunflower oil storage, we find a drop in iodine value (Table 3), which may be the result of light impact and creation of radicals on unsaturated hydrocarbon chain, which links appear connecting the complex compounds (dienes and polymers). As shown in Table 3, faster changes of iodine number occur in sunflower oil packed in transparent PET packaging. There was no change in saponifcation value and acid value during oil storage, what can be attributed to the lack of water in oil, as one of the factors that influences the process of splitting of triglycerides and saponification. Similar results were published by Raza and co-authors (2009), for auto- and photo- oxidized sunflower oil samples the iodine values were found to decrease from 139,0-127,5 and 139,0-123,3.

It can be observed that the packaging of sunflower oil in transparent glass and PET packaging does not affect the deterioration of oil caused by triglycerides hydrolysis. On the other side, increasing the saponification value (Table 4) can be attributed to the light impact, showing that the transparent packaging does not prevent progress of the light through the the walls of the packaging and that edible oil packing and storing in transparent glass and PET bottles is not the best solution. It is necessary to protect the oil from penetration of light by packaging it in coloured bottles.

Table 4. The average saponification values (mg KOH / g) of sunflower oil stored in glass and PET bottles

Packaging type	Storage conditions	Saponification values (mg KOH / g oil)			
		Storage period (months)			
		0	2	4	6
clear uncolored glass bottle	T=20^{O}C, φ=55%, artificial lighting	186	186	186	187
clear uncolored PET bottle	T=20^{O}C, φ=55%, artificial lighting	186	186	187	188

The concentration of secondary oxidation products of oil formed under the influence of various packaging and storage factors of oil, can be monitored by measuring the value of oil anisidinske. Abramovich and Abram (2005) saw no increase in the anisidine value during the first months of storage of *Camelia sativa* oil. In their case, sudden and intense changes of anisidine value occurred after 10 months of oil storage in a dark cold room. The authors interpret that the 2-alkenes and 2,4-dienes as key compounds that determine anisidine value, do not develop during the early stages of storage. During their research Nkpa et al (1990; 1992), found higher values of oxidation number in the crude palm oil packaged in transparent plastic and glass bottles than in the oil packed in metal cans with a layer of lacquer on the inner side, the same as in the yellow or green coloured plastic bottles. Lacquered metal cans provide good protection against oil oxidation and are considered to be practical for larger packaging units.

Table 5. The average anisidine values in sunflower oil stored in glass and PET bottles

Packaging type	Storage conditions	Anisidine value			
		Storage period (months)			
		0	2	4	6
clear uncolored glass bottle	t=20^{O}C, φ=55%, artificial lighting	5,5	18,4	27,7	39,6
clear uncolored PET bottle	t=20^{O}C, φ=55%, artificial lighting	5,5	19,3	28,4	40,2

The results of determining anisidine value are shown in Table 5. It is noticeable that the anisidine value increased in all tested packages during the six month storing in controlled conditions, with the initial value of 5.5, immediately after bottling, to the maximum value of 39.6 in glass and 40,2 in PET bottles. These results are in agreement with peroxide number values. The anisidine values indicate the level of aldehyde, the affect of auto- and photo-oxidation reactions during oil storage.

Raza and co-authors (2009) while researching peroxide value, p-anisidine of the auto- and photo-oxidized sunflower oil samples were found to be 1,10-7,09 meq/kg and 4,55-17,3 meq/kg; 1,51-10,03 and 1,56-17,16, respectively, and confirmed the decrease in oil quality during storage.

Table 6. The average total oxidation values in sunflower oil stored in glass and PET bottles

Packaging type	Storage conditions	Total oxidation values			
		Storage period (months)			
		0	2	4	6
clear uncolored glass bottle	t=20^{O}C, φ=55%, artificial lighting	5.96	20.28	35.26	55.20
clear uncolored PET bottle	t=20^{O}C, φ=55%, artificial lighting	5.96	20.90	36.76	57.40

Deteriorative effect of storing conditions on samples shelf life are indicated in the results obtained for total oxidation values in sunflower oil stored in glass and PET bottles during the experiment realisation. Oxygen is the most critical factor affecting the quality of oil. It can access the oil by permeation of the wall of the bottle, depending of the thickness of the wall. Higher values calculated for samples stored in PET bottles can be explained with the fact that ambient temperatures affect the rate of lipid oxidation and can modify the barrier properties, diffusion and permeability of polymeric materials. Total oxidation values in sunflower oil were relative high in our research, as shown in Table 6.

Storage stability of tested oil samples decreased as time of storage increased. Our results were similar to the other published results (Ramezani, 2004; Manzocco et al., 2011). Hamilton and Rossell (1986) considered that the total oxidation value is a useful indicator for determining the beginning of a progressive spoilage of oil and it can provide good information related to the development of primary and secondary oxidation products.

Conclusions

This paper demonstrated that the refined sunflower oil bottled in a clear uncoloured glass and PET packaging is a subject to the oxidation reactions. Glass and PET bottles are suitable for packaging of edible oil. Packaging and storing oil in PET bottles, exposed to the artificial light, increases oxidative deterioration of sunflower oil, while the degree of oxidative deterioration of sunflower oil packed in glass bottles was at the edge of tolerance during the six month storing. It can be concluded that the edible sunflower oil should be stored in packaging made from coloured glass and coloured plastic or other materials of appropriate quality and storing on dark, dry place. To minimize oxidative deterioration of edible oil, it is necessary to conduct additional education of producers, distributors and workers in warehouses and retail stores, on the importance of oil protection from the light, in order to provide favourable conditions for preserving the quality during storage and product shelf life and postpone deterioration.

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