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Interaction of packaging materials and vegetable oils: oil stability

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Abstract

The effects of different plastic films (polyethyleneterephthalate, polyvinylchloride, polypropylene and polystyrene) on the stability of olive, sunflower and palm oils were studied at 24 and 37°C during 60 days of storage. The changes in peroxide value (PV) and thiobarbituric acid value (TBA) were significantly higher ($p \leq 0.05$) in the plastic bottles than in glass. Our study indicates that the plastic permeability has played a major role in oil stability. However, both butylated hydroxyanisole (BHA) and butylated hydroxytoluene (BHT) were found to leach out from plastics films into vegetable oils during storage. The rate of oxidation was not reduced by antioxidant migration from plastic films to oils. Natural antioxidant (vitamin E) retarded the oxidation rate, and this was dependent on its concentration in oils examined. The results showed that the ranking of stability of oil samples is PVC≥PET > PP≥PS. Further, the stability was dependent on the type of oil. Palm oil exibited high stability properties while the highest oxidation rate was observed in sunflower oil. In addition, increasing storage temperature accelerated the oxidation and limited the stability of vegetable oils.

1. Introduction

Oils are an important part of the human diet. More than 90% of the world oil production from vegetable, animal and marine sources is used as food or as an ingredient in food products (Formo et al., 1979). Sunflower oil is used widely for deep frying. Palm oil is a major tropical product of great economic importance to a large number of developing countries and of considerable versatility within the edible oil industry (Clegg, 1973). It is used extensively in the manufacture of cooking fats, high quality confectionery fats and in filled milk type products. Olive oil, one of the world's most important and ancient oils, is the most widely used oil in the countries bordering on the Mediterranean Sea. It is used almost entirely for edible purposes as a cooking and salad oil (Formo et al., 1979). A large variety of packaged oils and fats is available in the retail trade. Glass, metals, and different kinds of plastic films are used for packaging of vegetable oils. Storage stability and shelf-life for fats and oils are now receiving attention among nutritionists, food processors, government regulators and consumers (Kaya et al., 1993). The quality and shelf-life of the packaged food are mainly determined by the barrier properties of the package against moisture, oxygen and the interaction of food constituents with the packaging materials (Sharma et al., 1990). Hence, the major function of packaging is to minimize reactions that affect the stability of the contained products (Karel & Heidelburgh, 1975; Gilbert & Mannheim, 1982). When certain reactions occur spontaneously without external agents, packaging does not affect stability. In most cases, however, the environmentally omnipresent gaseous reactants, water vapour and oxygen, can seriously restrict stability under normal food storage and distribution conditions (Gilbert, 1985).

One of the most important reactions leading to quality loss is rancidity of the food products. Rancidity is the development of an off-flavour by oxidation and hydrolysis which makes the food unacceptable (Labuza, 1971; Frankel, 1982; Paquette et al., 1985; Robards et al., 1988). Though most of the plastic films were found to be almost inert towards food constituents, a small amount of monomeric and oligomeric constituents or additives used in their manufacture, to provide stability, plasticity and other desirable functional characteristics, are known to migrate into foods. It was shown that migration can be particularly extensive through direct contact with fatty food surfaces and at high temperature (Crompton, 1979; Shepherd, 1982).

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Additives, such as plasticizers, antioxidants, antistatic agents and lubricants, are compounded into the basic polymer before being molded into the respective plastic materials (Jayaraman & Vasundhara, 1976).

Maximum limits of leached-out substances of various plastic materials used for food packaging applications were agreed; their rates of migration influenced the quality and stability of foods. One of the additives, fat-soluble phenolic antioxidants present in the compounded polymer, will migrate at an appreciable rate into the stored fat. As representatives of these, butylated hydroxyanisole (BHA) and butylated hydroxytoluene (BHT) have been under study (Mahadeviah, 1975; Crosby, 1981). Many studies have followed the loss of the antioxidants from packaging materials to fat-releasing foodstuffs (Niebergall & Hartmann, 1983; Freytag et al., 1984; Baner et al., 1992). An extension of the shelf-life of the products was reported by Miltz et al. (1988).

Vitamin E is a fat-soluble vitamin; it is found at high concentrations in vegetable oils. Tocopherols, especially the α-isomer, have a great influence on shelf-life, preserving oils from rancidity by interrupting the chain reactions involved in the formation of hydroperoxides (Shahidi & Wanasundara, 1992).

Kiritsakis (1984) studied the oxidative stability of olive oil stored in glass and polyethylene (PE) plastic bottles. He concluded that glass bottles provide better protection from oxidation than polyethylene plastic bottles do. Sharma et al. (1990) studied the effect of plastic film contact, including PE, PP and BHA and BHT incorporated in polyethylene, on the storage stability of refined sunflower oil and groundnut oil at 37°C. These authors concluded that changes in peroxide value and thiobarabituric acid were significantly less in the presence of plastic films than in control samples. Both BHA and BHT were found to leach out from plastic films into vegetable oils during storage. Nkpa et al. (1990, 1992) have shown that crude palm oil, packaged in clear plastic bottles, sealed polyethylene film and clear glass bottles, recorded higher total oxidation values than oils packed in either lacquered metal or amber and green glass bottles. Lacquered metal cans gave the greatest protection against oxidation. Kaya et al. (1993) studied the effect of permeability, and transparency of the packages (PET and glass bottles) on the shelf-life of sunflower and olive oils. The determinations were based on the oxidative stability of oils by measuring their peroxide values. The storage stability of oil increased in the following order with respect to packaging materials: PET < clear glass < coloured glass. Sirokhman (1983) controlled the oils quality by determination of peroxide, thiobarabituric acid values and acid number. Satue et al. (1995) reported that the extent of oxidation in oils was frequently evaluated by measuring peroxide value (PV). This index is related to the hydroperoxides, the primary oxidation products, which are unstable and readily decompose to form mainly mixtures of volatile aldehyde compounds. Because these compounds are directly responsible for rancid flavour (Frankel, 1982), they are considered important markers of oxidative rancidity.

The current study was undertaken to evaluate the effect of different factors including (1) type of packaging material (glass, polyethyleneterephthalate (PET), polyvinylchloride (PVC), polypropylene (PP), and polystyrene (PS)), (2) oxygen permeability, (3) storage time and temperature and (4) antioxidants, either in the oils (Vitamin E) or in the plastics (BHA and BHT), on the stability and quality of market vegetable oils (olive oil, sunflower oil, and palm oil).

2. Materials and methods

2.1. Materials

2.1.1. Oils

Commercially refined sunflower, virgin olive and refined palm oils were obtained from a local company. Oils were free of synthetic antioxidants and preservatives.

2.1.2. Chemicals

Butylated hydroxyanisole (BHA) and butylated hydroxytoluene (BHT) were from Across Chemical N.V. (Belgium).

Vitamin E (α-tocopherol), butylalcohol and 2-thiobarbituric acid were from Sigma Co (St. Louis, MO).

All solvents were HPLC grade were obtained from Across Chemical.

2.1.3. Packs

Transparent glass bottles (1.5 l and 8 dm^2) with screw cap were used. Polethyleneterephthalate (PET), polyvinylchloride (PVC), polypropylene (PP), and polystyrene (PS) bottles (1.5 l and 8 dm^2) were obtained from SOLVAY Brussels (ELTEX B 4020). Thickness, permeability to oxygen (from the manufacture) and antioxidants content (BHA and BHT) of each package types are given in Table 1.

2.2. Methods

2.2.1. Preparation

Oils were transferred to clean and dry glass jars and plastic packages (PET, PVC, PP and PS) in an anaerobic cabinet (Forma Scientific model 1024: 84%–8%–8%–N_2–CO_2–H_2 atmosphere). Headspace oxygen was negligible and the same since the containers were filled just below the seal line and under anaerobic conditions. There was the same net content of the samples. Enough containers for each oil sample were subjected

Table 1
Plastic materials properties

Plastic type	Thickness (μm)	Permeability ($cm^3 m^{-2} 24 h^{-1}$)	Antioxidants % (w/w)	
			BHA	BHT
PET	46	8–10	0.017	0.035
PVC	44	8–15	0.180	0.180
PP	44	150	0.070	0.105
PS	45	415	0.060	0.087

PET = Polyethylene terephthalate.
PVC = Polyvinylchloride.
PP = Polypropylene.
PS = Polystyrene.
BHA = Butylated hydroxyanisole.
BHT = Butylated hydroxytoluene.

to each storage condition so that no container, once removed from storage and used for analyses, had to be reused.

2.2.2. Storage conditions

Oil samples in glass and plastic bottles were stored in the dark at 24 and 37°C for 60 days. The experiments were run in duplicate and analyses were done in triplicate.

2.2.3. Analysis

Oils were sampled after 20 and 60 days of storage. The oil samples were analyzed for acid value as mg KOH g^{-1} oil, peroxide value (PV) as milli-equivalents of peroxides per 1000 g of sample, iodine values, Kries test, and thiobarbituric acid (TBA) value as described by Kirk & Sawyer (1991).

2.2.4. Antioxidant (BHA and BHT) analysis in films and oil

The levels of BHA and BHT in the tested films were determined by extraction followed by high pressure liquid chromatography (HPLC) analysis. For the antioxidant extraction, 5 g of the plastic film were cut into small pieces and extracted with 150 ml of acetonitrile in a Soxhlet extraction apparatus for 12 h. The extracts were then filtered and diluted with the solvent to a constant volume of 200 ml (Miltz et al., 1988).

The concentration of antioxidants migrated from film to oil was determined by the method of Niebergall & Hartmann (1983). Ten grams of oil was dissolved in 10 ml chloroform and this sample was directly injected into a Gilson high performance liquid chromatograph (Model 805), mobile phase, acetonitrile-H_2O (87:13 v/v); the column (250×4.6 mm) was packed with Lichrosorb RP-18-10 μ; U.V. absorbance detector, 225 nm; flow rate, 1.25 ml min^{-1}.

Concentration of antioxidants was determined from the plotting of serial concentrations of standard BHA and BHT against their areas of peaks.

2.2.5. Vitamin E analysis

Ten grams of oil was saponified with aqueous KOH (50% w/w) contained in methanolic ascorbic acid (0.5% w/v) and BHT for 20 min with a reflux condenser under a stream of nitrogen. After cooling, the residue was extracted twice with diethyl ether after the addition of butyl alcohol as an internal standard. The diethyl ether extract was washed with distilled water five times and filtered through Na_2SO_4. After evaporation, the sample was derivatized then injected into 9 GC (Carlo Erba Model 4130), with a flame ionization detector; the column was 25 m×0.25 m fused silica OV-1. In the same manner, a sample of α-tocopherol and butyl alcohol was used as a standard (Slover et al., 1983).

2.2.6. Statistical analysis

Every treatment was performed twice. Triplicate analyses were taken from the test samples at each specific time interval. Mean values and standard deviation were calculated at each time interval and analyzed by SPSS version 6 (SPSS Inc.444 N. Michigan Avenue, Chicago, Illinois, 60611) for analysis of variance (one way, two way and three way) and F-ratio (at level 0.05).

3. Results and discussion

3.1. Packaging materials and oil stability

Because edible oils are subject to oxidative rancidity, packaging in plastics poses some problems. Interaction between oxygen permeate and unsaturated fatty acid glycerides is the major cause of quality deterioration in vegetable oils during storage. Table 1 presents the thickness and the permeability to O_2 of tested plastic types. At constant thickness of plastics it is noticed that the permeability is dependent on the plastic types. Oxygen is a major influence of food shelf-life, because it contributes to the oxidation of lipids and is essential for the growth of aerobic food spoilage microorganisms and insect pests (Maloba et al., 1996). The effect of plastic film properties (e.g. thickness and permeability) on the quality and stability of olive, sunflower, and palm oils in terms of changes in acid value, PV, and TBA values, are given in Tables 2–4, respectively.

3.1.1. Olive oil

At 24° and 37°C, the olive oil packed in glass bottles had not increased significantly ($p \leq 0.05$) in peroxide value (PV) after 20 days, while the increase was significant ($p \leq 0.05$) in the oils in PET, PVC, PP, and PS bottles. It reached three times higher in the oil in PP and PS bottles (Table 2). The PV did not differ significantly ($p \leq 0.05$) between the oil stored in PET, PP, and PS at 24° and 37°C after 20 days but it did for PVC. However, after 60 days, all the plastics altered significantly

Table 2
Effects of different kinds of plastic bottle on the storage stability of virgin olive oil at 24 and 37°C after 20 and 60 days of storage

Tests	Storage temp. (°C)	Storage period (days)	Package type				
			Glass	PET[1]	PVC	PP	PS
Acid value		0	$_{v}0.16^{a}$	$_{v}0.16^{a}$	$_{v}0.16^{a}$	$_{v}0.16^{a}$	$_{v}0.16^{a}$
(mg KOH g^{-1})	24	20	$_{v}0.16^{a}$	$_{w}0.22^{b}$	$_{v}0.17^{b}$	$_{x}0.29^{b}$	$_{x}0.29^{b}$
		60	$_{v}0.18^{b}$	$_{w}0.23^{b}$	$_{w}0.23^{c}$	$_{x}0.35^{c}$	$_{x}0.45^{c}$
	37	20	$_{v}0.17^{b}$	$_{w}0.24^{b}$	$_{w}0.23^{c}$	$_{x}0.34^{c}$	$_{y}0.29^{b}$
		60	$_{v}0.19^{c}$	$_{w}0.28^{c}$	$_{w}0.27^{d}$	$_{x}0.44^{d}$	$_{y}0.49^{d}$
Iodine value		0	$_{v}90.24^{a}$	$_{v}90.24^{a}$	$_{v}90.24^{a}$	$_{v}90.24^{a}$	$_{v}90.24^{a}$
	24	20	$_{v}87.67^{b}$	$_{v}87.55^{a}$	$_{v}88.56^{a}$	$_{w}79.34^{b}$	$_{x}70.52^{b}$
		60	$_{v}84.22^{c}$	$_{vw}82.98^{b}$	$_{w}79.31^{b}$	$_{x}69.87^{c}$	$_{x}66.67^{c}$
	37	20	$_{v}86.00^{c}$	$_{w}82.09^{b}$	$_{v}86.24^{c}$	$_{x}76.74^{b}$	$_{y}68.31^{c}$
		60	$_{v}82.00^{d}$	$_{w}75.31^{c}$	$_{w}72.71^{d}$	$_{x}60.43^{d}$	$_{x}61.43^{d}$
Peroxide value		0	$_{v}5.81^{a}$	$_{v}5.81^{a}$	$_{v}5.81^{a}$	$_{v}5.81^{a}$	
(meq kg^{-1})	24	20	$_{v}6.97^{b}$	$_{w}8.00^{b}$	$_{v}7.43^{b}$	$_{x}15.36^{b}$	$_{y}14.50^{b}$
		60	$_{v}7.29^{b}$	$_{w}10.07^{c}$	$_{w}9.36^{c}$	$_{x}22.00^{c}$	$_{y}23.36^{c}$
	37	20	$_{v}7.16^{b}$	$_{w}9.28^{b}$	$_{v}8.81^{d}$	$_{x}15.69^{b}$	$_{y}14.86^{b}$
		60	$_{v}8.64^{c}$	$_{w}12.57^{d}$	$_{w}11.69^{e}$	$_{x}23.78^{d}$	$_{x}25.02^{d}$
TBA value		0	$_{v}0.05^{a}$	$_{v}0.05^{a}$	$_{v}0.05^{a}$	$_{v}0.05^{a}$	$_{v}0.05^{a}$
(mg malonaldehyde kg^{-1} oil)	24	20	$_{v}0.06^{ab}$	$_{v}0.06^{a}$	$_{w}0.16^{b}$	$_{x}0.19^{b}$	
		60	$_{v}0.07^{bc}$	$_{w}0.10^{b}$	$_{v}0.08^{b}$	$_{x}0.30^{c}$	$_{x}0.32^{c}$
	37	20	$_{v}0.07^{cd}$	$_{v}0.08^{c}$	$_{v}0.07^{b}$	$_{w}0.21^{d}$	$_{w}0.22^{b}$
		60	$_{v}0.08^{d}$	$_{w}0.12^{d}$	$_{w}0.12^{c}$	$_{x}0.33^{e}$	$_{y}0.43^{d}$
Vitamin E		0	$_{v}9.69^{a}$	$_{v}9.69^{a}$	$_{v}9.69^{a}$	$_{v}9.69^{a}$	$_{v}9.69^{a}$
(mg/100 ml oil)	24	20	$_{v}8.19^{a}$	$_{w}6.54^{b}$	$_{w}7.50^{b}$	$_{x}3.61^{b}$	$_{y}4.71^{b}$
		60	$_{v}6.06^{b}$	$_{w}5.05^{c}$	$_{w}5.34^{c}$	$_{x}3.59^{b}$	$_{y}2.46^{c}$
	37	20	$_{v}7.07^{c}$	$_{w}6.00^{b}$	$_{w}6.26^{d}$	$_{x}3.09^{c}$	$_{y}4.31^{b}$
		60	$_{v}6.95^{b}$	$_{w}5.01^{c}$	$_{w}5.93^{c}$	$_{x}2.01^{d}$	$_{x}2.42^{c}$
Antioxidants		0	nd[2]	0^{a}	0^{a}	0^{a}	0^{a}
BHA (ppm)	24	20	nd	2.01^{b}	12.21^{b}	7.30^{b}	5.54^{b}
		60	nd	3.51^{c}	40.05^{c}	21.24^{c}	15.15^{c}
	37	20	nd	2.53^{b}	14.58^{d}	8.20^{d}	7.05^{d}
		60	nd	4.61^{d}	45.24^{e}	23.71^{e}	17.03^{e}
BHT (ppm)		0	nd	0^{a}	0^{a}	0^{a}	0^{a}
	24	20	nd	2.10^{b}	15.01^{b}	8.25^{b}	6.14^{b}
		60	nd	4.11^{c}	44.25^{c}	23.05^{c}	18.52^{c}
	37	20	nd	3.03^{d}	17.26^{d}	9.63^{d}	8.27^{d}
		60	nd	5.13^{e}	47.4^{e}	25.72^{e}	19.65^{e}

Mean values in the same column not sharing a superscript to the right are significantly different. Mean values in the same row not sharing a subscript to the left are significantly different.

[1] See Table 1 for further details.

[2] Not determined.

($p \leq 0.05$) in PV. A sharp increase in thiobarbituric acid (TBA) value in olive oil after 20 days of storage was observed and it was also pronounced in PP and PS bottles. Relatively, the acid and iodine values were altered at the end of storage period, and were affected by packaging materials. The three-way analysis of variance showed that the three variables (package type, temperature and time) interacted to significantly effect ($p \leq 0.05$) all the tested parameters (acid, iodine, peroxide, and TBA values) (Table 2). The interaction between time and temperature (two way analysis) was affected significantly ($p \leq 0.05$) in the tested parameters. After 60 days, the ranking of oxidative stability in olive oil was glass > PVC = PET > PP > PS at both temperatures. Jimenez & Qujano (1973) studied the organoleptic tests of two virgin olive oils stored in glass, tin, polyethylene and PVC bottles. PVC gave significantly better flavour; however, the greatest flavour deterioration occurred in polyethylene containers which gave significantly poorer results than other packaging materials. Gutierrez et al. (1988) have already shown that olive oil packed in permeable PE and PP should be sold within four weeks; however, air-impermeable PVC bottles are able to secure the oil quality for 3 months.

3.1.2. Sunflower oil

The type of packaging material affected the degree of stability of sunflower oil during the storage period. With regard to the changes in tested parameters, the PVC and PET bottles were the best packaging containers compared to glass control, followed by PP and PS (Table 3).

Table 3
Effects of different kinds of plastic bottle on the storage stability of sunflower oil at 24 and 37°C after 20 and 60 days of storage

Tests	Storage temp. (°C)	Storage period (days)	Package type				
			Glass	PET[1]	PVC	PP	PS
Acid value		0	$_{v}1.78^{a}$	$_{v}1.78^{a}$	$_{v}1.78^{a}$	$_{v}1.78^{a}$	$_{v}1.78^{a}$
(mg KOH g^{-1})	24	20	$_{v}1.80^{a}$	$_{w}2.05^{b}$	$_{w}2.01^{b}$	$_{x}2.33^{b}$	$_{x}2.38^{b}$
		60	$_{v}1.89^{b}$	$_{w}2.19^{c}$	$_{w}2.14^{b}$	$_{x}2.59^{c}$	$_{y}2.66^{c}$
	37	20	$_{v}1.83_{a}$	$_{w}2.10^{bc}$	$_{w}2.10^{b}$	$_{x}2.49^{d}$	$_{x}2.46^{d}$
		60	$_{v}1.92^{c}$	$_{w}2.21^{d}$	$_{w}2.25^{c}$	$_{x}2.79^{e}$	$_{x}2.80^{e}$
Iodine value		0	$_{v}138.02^{a}$	$_{v}138.02^{a}$	$_{v}138.02^{a}$	$_{v}138.02^{a}$	$_{v}138.02^{a}$
	24	20	$_{v}137.31^{a}$	$_{v}138.00^{a}$	$_{v}137.51^{a}$	$_{w}123.68^{b}$	$_{x}115.97^{b}$
		60	$_{v}136.25^{ab}$	$_{w}133.4^{b}$	$_{w}135.23^{ab}$	$_{x}117.76^{c}$	$_{y}111.78^{c}$
	37	20	$_{v}135.01^{bc}$	$_{v}132.91^{b}$	$_{v}133.31^{bc}$	$_{w}118.54^{c}$	$_{x}112.31^{c}$
		60	$_{v}134.99^{c}$	$_{w}129.77^{c}$	$_{v}131.23^{c}$	$_{x}115.55^{c}$	$_{y}109.24^{c}$
Peroxide value		0	$_{v}14.88^{a}$	$_{v}14.88^{a}$	$_{v}14.88^{a}$	$_{v}14.88^{a}$	$_{v}14.88^{a}$
(meq kg^{-1})	24	20	$_{v}15.80^{a}$	$_{v}16.00^{b}$	$_{v}15.35^{b}$	$_{w}35.00^{b}$	$_{x}30.24^{b}$
		60	$_{v}17.51^{b}$	$_{w}25.95^{c}$	$_{w}22.62^{c}$	$_{x}60.95^{c}$	$_{x}61.34^{c}$
	37	20	$_{v}17.05^{b}$	$_{v}17.57^{b}$	$_{v}18.35^{b}$	$_{w}37.13^{b}$	$_{x}32.13^{b}$
		60	$_{v}18.75^{c}$	$_{w}27.28^{d}$	$_{w}24.06^{d}$	$_{x}75.84^{d}$	$_{x}75.84^{d}$
TBA value		0	$_{v}0.15^{a}$	$_{v}0.15^{a}$	$_{v}0.15^{a}$	$_{v}0.15^{a}$	$_{v}0.15^{a}$
(mg malonaldehyde kg^{-1} oil)	24	20	$_{v}0.16^{a}$	$_{w}0.18^{b}$	$_{x}0.47^{b}$	$_{y}0.51^{b}$	
		60	$_{v}0.19^{b}$	$_{w}0.26^{c}$	$_{w}0.29^{c}$	$_{x}0.86^{c}$	$_{x}0.89^{c}$
	37	20	$_{v}0.18^{c}$	$_{w}0.21^{c}$	$_{w}0.21^{d}$	$_{x}0.55^{d}$	$_{x}0.58^{b}$
		60	$_{v}0.21^{d}$	$_{w}0.32^{d}$	$_{w}0.33^{e}$	$_{x}0.89^{c}$	$_{x}0.98^{d}$
Vitamin E		0	$_{v}50.69^{a}$	$_{v}50.69^{a}$	$_{v}50.69^{a}$	$_{v}50.69^{a}$	$_{v}50.69^{a}$
(mg 100 ml^{-1} oil)	24	20	$_{v}48.12^{a}$	$_{v}47.89^{b}$	$_{v}46.66^{b}$	$_{w}35.11^{b}$	$_{w}32.12^{b}$
		60	$_{v}45.56^{b}$	$_{v}46.78^{b}$	$_{w}36.05^{c}$	$_{x}28.12^{c}$	$_{x}23.53^{c}$
	37	20	$_{v}46.92^{b}$	$_{w}43.71^{c}$	$_{w}43.12^{d}$	$_{x}30.82^{d}$	$_{w}29.26^{d}$
		60	$_{v}42.49^{c}$	$_{w}36.25^{d}$	$_{w}35.46^{c}$	$_{x}25.66^{e}$	$_{w}22.16^{e}$
Antioxidants		0	nd[2]	0^{a}	0^{a}	0^{a}	0^{a}
BHA (ppm)	24	20	nd	2.51^{b}	13.52^{b}	6.02^{b}	5.52^{b}
		60	nd	4.63^{c}	41.63^{c}	21.54^{c}	15.62^{c}
	37	20	nd	3.02^{d}	15.14^{d}	8.63^{d}	7.89^{d}
		60	nd	5.10^{e}	46.26^{e}	24.74^{e}	17.85^{e}
BHT (ppm)		0	nd	0^{a}	0^{a}	0^{a}	0^{a}
	24	20	nd	3.23^{b}	14.75^{b}	9.31^{b}	5.65^{b}
		60	nd	4.78^{c}	42.02^{c}	22.53^{c}	17.69^{c}
	37	20	nd	4.10^{d}	18.53^{d}	10.02^{d}	8.74^{d}
		60	nd	5.62^{e}	48.67^{e}	25.12^{e}	21.02^{e}

Mean values in the same column not sharing a superscript to the right are significantly different. Mean values in the same row not sharing a subscript to the left are significantly different.

[1] See Table 1 for further details.

[2] Not determined.

The 3-way analysis of variance revealed that the interaction between the three variables had a significant effect ($p \leq 0.05$) on acid, iodine, peroxide and TBA values. In the 2-way analysis of variance between temperature and time, there is a significant effect on all the tested parameters. The results confirmed that the oxidative stability of sunflower oil is less than olive oil. Permanyer (1986) showed that, as an index of oxidation of vegetable oils, all the parameters together gave an indication of the stability of the various oils. He reported that the oxidative changes depended on the nature and initial condition of the oil but, overall, virgin olive oil was more stable than refined sunflower oil. In agreement with the present study, Wilhelm et al. (1988) showed, in their study of oxidative stability of sunflower oil, olive oil, and butter oil which were used for potato frying, that sunflower showed the most rapid oxidation, followed by olive oil. This is not surprising in view of their fatty acid composition. Sunflower oil contains a relatively high amount of linoleic acid, 75%, while olive oil contains only 6%. Kaya et al. (1993) investigated the effect of packaging material and its oxygen permeability on the shelf-life of sunflower and olive oils. They concluded that the storage stability of oils was greater in glass than in PET, which reflects the role of permeable oxygen.

3.1.3. Palm oil

A similar trend of change was found in palm oil. Oxidation was high in PS and PP bottles compared to PET and PVC bottles (Table 4). In the 3-way analysis of variance, it is apparant that the interaction between the

Table 4
Effects of different kinds of plastic bottle on the storage stability of refined palm oil at 24 and 37°C after 20 and 60 days of storage

Tests	Storage temp. (°C)	Storage period (days)	Package type				
			Glass	PET[1]	PVC	PP	PS
Acid value		0	$_{v}6.45^{a}$	$_{v}6.45^{a}$	$_{v}6.45^{a}$	$_{v}6.45^{a}$	$_{v}6.45^{a}$
(mg KOH g^{-1})	24	20	$_{v}6.64^{a}$	$_{v}6.26^{a}$	$_{v}6.50^{a}$	$_{w}8.38^{b}$	$_{w}8.16^{b}$
		60	$_{v}7.50^{b}$	$_{v}7.64^{b}$	$_{v}7.58^{b}$	$_{w}10.42^{c}$	$_{x}9.67^{c}$
	37	20	$_{v}6.96^{a}$	$_{w}6.57^{a}$	$_{w}6.57^{a}$	$_{x}8.50^{b}$	$_{x}8.44^{b}$
		60	$_{v}7.99^{c}$	$_{w}8.48^{c}$	$_{w}8.62^{c}$	$_{x}12.40^{d}$	$_{y}11.65^{d}$
Iodine value		0	$_{v}55.25^{a}$	$_{v}55.25^{a}$	$_{v}55.25^{a}$	$_{v}55.25^{a}$	$_{v}55.25^{a}$
	24	20	$_{v}52.38^{b}$	$_{w}46.28^{b}$	$_{w}49.94^{b}$	$_{x}39.69^{b}$	$_{y}43.03^{b}$
		60	$_{v}45.51^{c}$	$_{w}37.69^{c}$	$_{w}37.36^{c}$	$_{x}22.86^{c}$	$_{x}21.34^{c}$
	37	20	$_{v}50.48^{b}$	$_{w}43.83^{d}$	$_{w}48.36^{b}$	$_{x}38.27^{b}$	$_{x}40.83^{b}$
		60	$_{v}47.12^{c}$	$_{w}35.58^{c}$	$_{w}37.02^{c}$	$_{x}22.61^{c}$	$_{x}20.49^{c}$
Peroxide value		0	$_{v}0.56^{a}$	$_{v}0.56^{a}$	$_{v}0.36^{a}$	$_{v}0.56^{a}$	$_{v}0.56^{a}$
(meq kg^{-1})	24	20	$_{v}0.59^{a}$	$_{w}0.76^{b}$	$_{w}0.76^{b}$	$_{x}1.51^{b}$	$_{x}1.82^{b}$
		60	$_{v}0.77^{b}$	$_{w}1.72^{c}$	$_{w}1.96^{c}$	$_{x}3.27^{c}$	$_{x}3.02^{c}$
	37	20	$_{v}0.64^{c}$	$_{w}1.41^{d}$	$_{x}1.97^{c}$	$_{x}1.99^{b}$	$_{y}2.68^{c}$
		60	$_{v}1.41^{d}$	$_{w}2.02^{e}$	$_{w}2.18^{d}$	$_{x}3.74^{c}$	$_{y}4.71^{d}$
TBA value		0	$_{v}0.02^{a}$	$_{v}0.02^{a}$	$_{v}0.02^{a}$	$_{v}0.02^{a}$	$_{v}0.02^{a}$
(mg malonaldehyde kg^{-1} oil)	24	20	$_{v}0.04^{a}$	$_{v}0.05^{b}$	$_{v}0.04^{a}$	$_{w}0.21^{b}$	$_{x}0.31^{b}$
		60	$_{v}0.08^{b}$	$_{w}0.11^{c}$	$_{w}0.12^{b}$	$_{x}0.38^{c}$	$_{x}0.41^{c}$
	37	20	$_{v}0.07^{b}$	$_{v}0.06^{b}$	$_{v}0.09^{c}$	$_{w}0.22^{b}$	$_{x}0.31^{b}$
		60	$_{v}0.09^{c}$	$_{w}0.16^{d}$	$_{w}0.19^{d}$	$_{x}0.52^{d}$	$_{x}0.58^{d}$
Vitamin E (mg 100 ml^{-1} oil)		0	$_{v}25.93^{a}$	$_{v}25.93^{a}$	$_{v}25.93^{a}$	$_{v}25.93^{a}$	$_{v}25.93^{a}$
	24	20	$_{v}24.40^{a}$	$_{w}22.58^{b}$	$_{w}23.15^{b}$	$_{x}12.94^{b}$	$_{w}15.02^{b}$
		60	$_{v}22.18^{b}$	$_{w}20.39^{c}$	$_{v}21.98^{c}$	$_{x}9.06^{c}$	$_{x}9.29^{c}$
	37	20	$_{v}22.70^{bc}$	$_{w}21.93^{b}$	$_{v}22.14^{b}$	$_{x}9.94^{c}$	$_{y}11.67^{d}$
		60	$_{v}21.13^{c}$	$_{w}19.65^{d}$	$_{v}20.21^{d}$	$_{x}7.39^{d}$	$_{x}6.86^{c}$
Antioxidants		0	nd[2]	0^{a}	0^{a}	0^{a}	0^{a}
BHA (ppm)	24	20	nd	0.53^{b}	8.65^{b}	4.15^{b}	4.56^{b}
		60	nd	0.74^{c}	30.02^{c}	8.56^{c}	10.85^{c}
	37	20	nd	0.85^{d}	10.74^{d}	5.86^{d}	6.02^{d}
		60	nd	1.21^{e}	41.02^{e}	15.64^{e}	15.75^{e}
BHT (ppm)		0	nd	0^{a}	0^{a}	0^{a}	0^{a}
	24	20	nd	0.72^{b}	9.85^{b}	6.02^{b}	3.62^{b}
		60	nd	0.96^{c}	27.12^{c}	16.36^{c}	12.69^{c}
	37	20	nd	1.13^{d}	11.02^{d}	8.95^{d}	7.41^{d}
		60	nd	1.68^{e}	43.01^{e}	20.54^{e}	16.02^{e}

Mean values in the same column not sharing a superscript to the right are significantly different. Mean values in the same row not sharing a subscript to the left are significantly different.

[1] See Table 1 for further details.

[2] Not determined.

three variables was significant ($p \leq 0.05$) in all the tests. The parameters changed significantly as the result of the interaction between the storage time and temperatures. Previously reported effects of various packaging materials on storage stability of palm oils are that lacquered metal cans and amber glass bottles gave the greatest protection against oxidation (Nkpa et al., 1990, 1992).

The permeability to O_2 of different bottles clearly affected the oxidative degradation of vegetable oils. The ability of tested plastic bottles to permeate oxygen was shown to be a significant factor causing decreased shelf-life. Oils placed in glass bottles were changed during storage time but were less affected compared to those packed in plastic bottles. PS has a higher permeability rate (415 $cm^3 m^{-2} 24 h^{-1}$) and caused a rapid deterioration in the three types of oils. Further, PET and PVC gave a degree of stability closer to that provided by packing in glass (Tables 2–4).

3.2. Other factors that influence oil stability

3.2.1. Temperature

The criteria for the shelf-life of the oils depend on the temperature of the storage. The oxidative stability of the three types of oils was significantly different between the two storage temperatures (24° and 37°C), especially after 60 days of storage (Tables 2–4). Oil stability is usually determined under accelerated conditions (60°C and more) because ambient conditions demand an excessively long period of storage (Formo et al., 1979). Frankel (1993) found that different volatile compounds were formed according to the temperature used. These

differences in volatile formation indicate that the formation and decomposition of different hydroperoxides vary significantly with temperature of oxidation. Therefore, studies of oxidative stability at high temperatures may not be extrapolated to ambient temperature (Frankel, 1993). In addition, Satue et al. (1995) concluded that the evaluation of oxidative stability should be carried out at temperatures as low as possible, depending on the oxidative susceptibility of the oil. Maloba et al. (1996) showed that sunflower oil, which was stored in the presence of a novel oxygen-scavenging film at 37°C, became rancid more rapidly than at 23°C, owing to increased reaction rates at the higher temperature.

3.2.2. Type of oil and its initial constants

The differences in oxidative stability between oils reflect their initial constants (e.g. PV). The changes in the tested oils followed a similar pattern but the oxidative stability was considerably greater ($p \leq 0.05$) in palm oil than in olive and sunflower oils (Tables 2–4). Satue et al. (1995) reported that the oxidative stability of several commercial olive oils varied significantly with the oil having the lowest initial PV being the most stable.

The type of the oil also affected its physical and chemical properties. In general, the greater the degree of unsaturation (the higher the iodine value) the greater is the stability of the fat to oxidative rancidity.

3.2.3. Antioxidants

3.2.3.1. BHA and BHT. The initial concentration of antioxidants (BHA and BHT) in the plastic bottles used in this study is shown in Table 1. In general, the migration of antioxidants from the plastics into oils proceeded throughout the storage period at a low rate (Tables 2–4). It is observed that the migration rate is dependent on the initial concentration of antioxidants in the plastic, as well as temperature and time of storage (Tables 2–4). The interactions of temperature and storage time significantly ($p \leq 0.05$) affect the migration of antioxidants into the vegetable oils. In the 2-way analysis of variance of temperature and time, in the three type of oils, there was a significant effect on BHA and BHT migration into oils. Interestingly, it is observed that, at low levels of migrated antioxidants from plastic films into oils, there was no effect on oil stability. Even in PVC bottles, which had the highest antioxidant content and relatively higher migration of antioxidants into oils compared to other plastic types, it is not possible under experiment conditions to separate the antioxidant effect. The cereal contained in HDPE pouches impregnated with the high level of BHT had lower oxidation levels as a result of its migration into the cereal. However, Satue et al. (1995) showed that the antioxidant effectiveness of phenolic compounds in virgin olive oils can be significantly diminished in oils if their initial PV values are too high. The concentrations of leached-out antioxidants from plastic bottles into oils are reported to depend on the nature of the oil and the antioxidant type (Sharma et al., 1990). In the present study, the migration of BHA and BHT was considerably higher in sunflower oil than in olive oil and both were greater than in palm oil (Tables 2–4). It is observed that BHT migrated slightly more than BHA. Similar results were reported by Sharma et al. (1990). The kinetics of antioxidant migration were studied by Miltz et al. (1988). It was found that the loss of BHA antioxidants from HDPE followed a first order rate expression. Moreover, Figge (1972) has reported that migration of antioxidants in vegetable oils depends on the nature and thickness of films, method of fabrication, storage temperature and duration and the nature of oils and antioxidants. Kochmann et al. (1985) reported that the rate of migration is quite high from PP and PE films which swell rapidly in contact with vegetable oils and organic solvents as compared to rigid PVC films which exhibit negligible swelling. The oils having lower viscosity and lower solidification points are known to extract higher concentrations of antioxidants and other plastic additives (Sharma et al., 1990). A decrease in viscosity is known to increase the solvating action of organic solvents. (Sharma et al., 1990). Niebergall and Hartmann (1983) have reported a very rapid migration of antioxidants into vegetable oils and into ground spices having a large percentage of aromatic components.

Commercially, Irganox and Ionox (complex high molecular weight compounds) are employed as antioxidants for stabilization of PE and PP films. Schwope et al. (1987) have reported considerably less migration of Irganox as compared to BHT in corn oil from low density PE films. However, the beneficial effects of incorporating antioxidants in the film on the stability of potato chips and Quaker oats were reported previously (Talburt and Smith, 1959).

3.2.4. A natural antioxidant in oils: (vitamin E)

The rates of loss of vitamin E in olive, sunflower and palm oil after 20 and 60 days of storage at 24°and 37°C are presented in Tables 2–4, respectively. Like other antioxidants, the tocopherols are themselves readily oxidizable. Mild oxidation of a tocopherol opens the heterocyclic ring to form tocoquinone, which is not an antioxidant (Formo et al., 1979). In this study, the loss of vitamin E was followed by the decrease in oxidative stability. It was more pronounced in the oils packed in PP and PS. The increase in the peroxide value was in accordance with the decrease in the natural antioxidant (vitamin E). In addition to the initial loss of vitamin E as a result of oxidation, partial loss might be attributed to plastic swelling and its additives. The 3-way analysis of variance of the interacted parameters (type of oil and plastic and temperature) showed a significant effect

($p \leq 0.05$) on the concentration of vitamin E. In the present study, the presence of natural antioxidants (tocopherols) might have enhanced the stability of palm oil. However, in sunflower oil, which had a higher content, it did not. This could be explained by the difference between the two oils in the initial PV, which was clearly higher in sunflower oil (Table 3). Many contradictory results concerning the efficacy of natural and phenolic antioxidants on oil stability were reported. Loebis (1986), in his study of the effect of antioxidants on palm oil, showed that BHT was relatively ineffective, whereas α-tocopherol greatly improved the stability of palm oil. However, Satue et al. (1995) showed that α-tocopherol was not as good an antioxidant for the prevention of hydroperoxide formation as the other phenolic compounds, and at high concentrations showed a prooxidant effect.

3.2.5. Toxicological evaluation

From the toxicological point of view the accepted daily intake (ADI) of BHA and BHT is estimated to be approximately 0.1 mg kg^{-1} of body weight. To estimate the significance of the migrated levels of antioxidants in oils with regard to human health, the highest concentration of both antioxidants detected in oil was used to calculate a 'maximum potential exposure' or 'MPE' for these antioxidants. The calculation assumes that a 70 kg person would use 100 g oil per day. This means, for example, that the concentration of antioxidants (BHA + BHT) in olive oil packed in PET bottles stored at 37°C after 60 days is (0.95 100 ml^{-1} oil) 70 kg^{-1} b/w = 0.014 mg kg^{-1} day^{-1}. MPE values were compared with Acceptable Daily Intakes (ADI) for BHA and BHT. The relative ratio, calculated MPE to reference ADI, was used as a safety indicator. When the resulting ratio is below one, it indicates a 'safe' health hazard (Fig. 1). In PVC plastic bottles the ratio is more than one. It is reported that 'At levels 500 times the daily consumption of these antioxidants (50 mg kg^{-1} day^{-1}), both rodents and monkeys appeared to be free of any obvious injurious effects.' However, at larger doses (500 mg kg^{-1} day^{-1}), both BHA and BHT cause certain pathological, enzyme and lipid alterations (Formo et al., 1979).

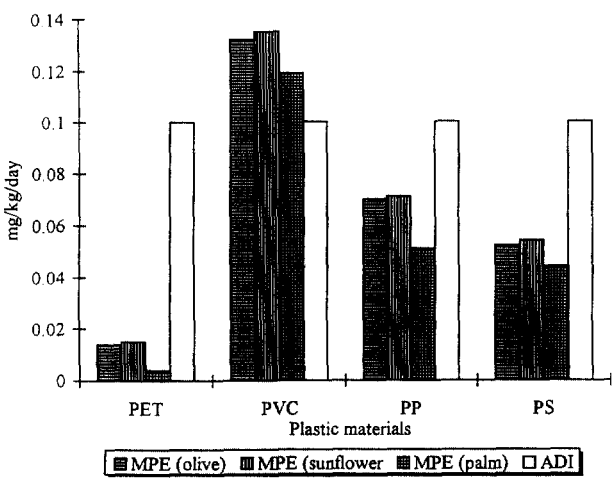

Fig. 1. Evaluation of maximum potential exposure of migrated antioxidants (BHA and BHT) observed in different kinds of oil after 60 days of storage at 37°C with different plastic types using ADI value as a health assessment reference. ADI = Acceptable Daily Intake, MPE = Maximum Potential Exposure for a 70 kg person using the worst case exposure data from the studies conducted.

4. Conclusions

It is concluded from the current study that the stability of vegetable oils is dependent on the following characteristics:

- The type of plastic film and its oxygen permeability (PS > PP > PET = PVC).
- The level of natural antioxidants in oils.
- The type of oil and its initial physical and chemical properties.
- The time and temperature of storage. It is found that there are significant differences in oil stability according to the storage time and temperature in all types of package materials.

It seems that the level of phenolic antioxidants (BHA and BHT) in the tested plastics did not play a significant role in enhancing oil stability. The synthetic antioxidants (BHA and BHT) levels caused by migrations from plastic films into oils would not pose a public health concern.

The reduction in the vegetable oil stability was faster in PS and PP bottles than PET and PVC bottles.

It is of great importance to conduct storage experiments by choosing different conditions (e.g. temperature and light) to evaluate the ability of any new polymer to extend shelf-life and overall quality. There is a need for active packaging materials that can scavenge residual oxygen from package headspaces, and any that subsequently diffuses into the package (Maloba et al., 1996).

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