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Investigation of the effects of lights, temperatures and packaging materials on the virgin rapeseed oil flavors during storage

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ABSTRACT

How external factors affect virgin rapeseed oil (VRO) flavor was explored from the perspective of key flavor-related compounds (KFCs). Light (sunlight, ultraviolet light, and dark), temperature (ambient temperature and 60 °C), and packaging materials (glass and plastics) were selected. Induction periods (IPs) of samples in packaging materials and surface hydrophilicity of the materials were measured. The results showed that aldehydes increased from 3.654 to 10.183 to 82.762–262.580 and 10.147–50.076 mg/kg under 60 °C and ultraviolet light, respectively, in six VROs. At 60 °C, alkenals were only detected at about 60–100 meq/kg, while nitriles, pyrazines, and isothiocyanates could not be detected at the end. Compared with those in polyethylene terephthalate (203.5–452.0 min), and polypropylene (207.0–458.5 min) plastic bottles, VROs samples in glass bottles showed longer IPs (226.0–486.0 min) representing better oxidation stability as well as flavor preservation during storage (p < 0.05). Glass was determined enhanced hydrophilicity with contact angle of 44.53° compared with polyethylene terephthalate (59.80°), and polypropylene (77.10°) plastic that promoted combination with polar KFCs and other compounds, which was helpful for increasing surface tension at oil/air interface with restricted oxygen diffusion to reduce oxidation rate, and preventing volatilization of KFCs.

1. Introduction

Virgin rapeseed oils (VROs) are manufactured by a simple pressing process from roasted rapeseeds and then subjected to sedimentation/filtration (Mao, Zhao, Huyan, Liu, & Yu, 2019; Jing, Guo, Wang, Zhang, & Yu, 2019). Owing to the high proportion of key flavor-related compounds (KFCs) well preserved in VRO production, such as nitriles, isothiocyanates, pyrazines, and aldehydes (Wang, Zhang, Chen, Jing & Zhang, 2020), VROs are well recognized by consumers for their unique flavors (Zhang, Lu, Yang, Li, & Wang, 2011). However, these flavors are susceptible to deterioration or loss because of changes in KFCs during storage, so packaging materials are used to prolong the shelf life of these oils (Pristouri, Badeka & Kontominas, 2010). There is thus a need to study the changes in KFCs during storage and the performance of

different packaging materials in preserving VRO flavors.

External factors, such as oxygen, light, and temperature, induce pronounced changes in the flavors of edible oils during storage. Studies have reported that flavor-related compounds of olive oils were reduced by 94% after 12 months of storage, and the positive sensory attributes of fruitiness, greenness, bitterness, and pungency were also found to exhibit massive losses (Njoroge, Ukeda and Sawamura, 1996; Malheiro, Casal, Rodrigues, Renard and Alberto, 2018). Goicoechea and Guillén (2014) studied the headspace composition of corn oil samples stored at ambient temperature for different periods of time, with aldehydes being shown to be the most abundant group of secondary volatile oxidation compounds. The relative contents of hexanal, heptanal, pentanal, octanal, nonanal, and decanal in rapeseed oil increased with oxidation levels (Xu, Yu, Li, Chen, & Wang, 2017). Oil oxidation causes the development

Abbreviations: VRO, virgin rapeseed oil; KFCs, key flavor-related compounds; IPs, induction periods; PVs, peroxide values; PET, polyethylene terephthalate; PP, polypropylene; HS-SPME/GC-MS, headspace solid-phase microextraction/gas chromatography-mass spectrometry; Sy 28, Shanyou 28; Qy 10, Qinyou 10; Fy 7, Fengyou 7; Ly 6, Longyou 6; Cy 47, Chuanyou 47; My 3, Menyuan 3; ROAVs, relative odor activity values; OTs, odor thresholds.

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of an undesirable rancid smell because of high levels of low-molecular-weight oxidation products, such as aldehydes, ketones, hydrocarbons, alcohols, and esters (Wang, Jiang, & Hammond, 2005; Choe & Min, 2006; Mahesar, Bendini, Cerretani, Matteo, & Sherazi, 2010). Most studies have primarily focused on changes in oxidation products, so information regarding changes in other KFCs (such as nitriles, pyrazines, and isothiocyanates) in VROs under different temperature and light conditions during storage is limited.

Packaged foods have a prolonged shelf life, which is normally ascribed to packaging barriers that protect products from oxygen, heat, and light (Hu, Huyan, Ding, Dong, & Yu, 2020; Kanavouras, Hernandez-Münoz, Coutelieres, & Selke, 2004). Different packaging materials, such as plastic, ceramic, and glass, were shown to be associated with significant differences in shelf life among edible oils on the basis of quality parameters (Silvagni, Franco, Bagno, & Rastrelli, 2012; Mishra & Sharma, 2011; Ramezani, 2004). This is related to the hydrophilic surfaces of the materials because packaging materials with enhanced hydrophilicity are likely to adsorb hydroperoxides and other polar compounds better, to reduce the spread of polar chemicals at air/oil interfaces and increase the surface tension of air/oil interfaces with decreased oxygen diffusion (Hu et al., 2020; Huyan, Ding, Mao, Wu, & Yu, 2019). Thus, hydrophilic packaging materials can retard oil oxidation. Flavor-related compounds, including nitriles, isothiocyanates, and pyrazines, in VROs with polar functional groups probably combine with hydrophilic packaging materials and reduce their volatilization. This could account for the differences in the ability of different packaging materials to preserve flavor.

This study analyzed the changes of KFCs (aldehydes, nitriles, pyrazines, isothiocyanates) under different light and temperature conditions at different peroxide values (PVs). Glass, polyethylene terephthalate (PET), and polypropylene (PP) plastic bottles were selected as packaging materials, and their performance at preserving KFCs during storage was investigated. The induction period values (IPs) of samples in the three packaging materials were determined through Oxitest analysis. The surface hydrophilicity of the packaging materials was characterized through Young's contact angle measurements.

2. Materials and methods

2.1. Materials and chemicals

Six varieties of rapeseed (*Brassica napus*), namely, Shanyou 28 (Sy 28) and Qinyou 10 (Qy 10) from Hanzhong County in Shanxi Province; Fengyou 7 (Fy 7) from Dali County in Shanxi Province; and Longyou 6 (Ly 6), Chuanyou 47 (Cy 47), and Menyuan 3 (My 3) from Gansu (Jinchang), Sichuan (Mianyang), and Qinghai (Menyuan) Provinces, respectively, were collected from the chief rapeseed-producing areas in China. The moisture contents of all rapeseeds were 6–7 g/100g. Cylindrical glass, PET, and PP plastic bottles, about 11.3 cm in height and 8.5 cm in width, were also purchased from Chinese markets.

2.2. Sample preparation

2.2.1. Preparation of VROs

Each rapeseed type (Sy 28, Qy 10, Fy 7, Ly 6, Cy 47, and My 3; 1000 \pm 0.5 g) was roasted in a roasting machine (Jiangsu Maisi Machinery Equipment Co., Ltd., Jiangsu, China) at 120 °C for different times (Table A1\). The corresponding roasting conditions of each sample were optimized in a preliminary experiment to obtain the VRO products with higher sensory scores. The temperatures of the rapeseeds after roasting were measured using a handheld digital temperature probe (Guangdong Uni-T Technology Co., Ltd., Guangdong, China). Then, the seeds were pressed using an oil pressing machine (Westinghouse, Inc., USA). The obtained products were then degummed using saturated salt water at ambient temperature to obtain VROs.

2.2.2. Determination of KFC changes of samples under different light conditions

Samples of VROs weighing 30.0 g were placed in a closed glass bottle at ambient temperature with exposure to sunlight (40 W, exposure height of 15 cm) or ultraviolet light (40 W, exposure height of 15 cm), or being placed in the dark. Based on our preliminary experiments, no significant changes in KFCs at low PVs (less than 20 meq/kg) were observed (p>0.05, data not shown). Thus, the range of PVs was extended to about 20–100 meq/kg to monitor the changes in KFCs in VROs. PVs of samples reached 100 meq/kg in 45 days under sunlight, 25 days under ultraviolet light and 180 days under dark with no addition of the chemicals. The PVs of samples were determined according to the AOCS Cd 8b-90 method. Samples (1.0 g) were collected in a headspace bottle for GC-MS analysis.

2.2.3. Determination of KFC changes of samples at different temperatures Samples of VROs weighing 30.0 g were placed in a closed glass bottle packed with silver paper to avoid light at ambient temperature or 60 $^{\circ}$ C. Based on our preliminary experiments, no significant changes in KFCs at low PVs (less than 20 meq/kg) were observed (p>0.05, data not shown). Thus, the range of PVs was extended to about 20–100 meq/kg to monitor the change in KFCs in VROs. PVs of samples reached 100 meq/kg in 15 days at 60 $^{\circ}$ C and 45 days at ambient temperature with no addition of the chemicals. The PVs of samples were determined according to the AOCS Cd 8b-90 method. Samples (1.0 g) were collected and added to a headspace bottle for GC-MS analysis.

2.2.4. Determination of flavor-related changes of samples in different packaging materials

My 3, Fy 7, and Sy 28 obtained higher sensory scores among the six VROs as recorded by a sensory panel in a preliminary experiment, so they were selected for further analysis. Samples of VROs per 100.0 g were placed in closed glass, PET, and PP plastic bottles at ambient temperature for simulation of the daily storage of VROs. The samples were collected every 30 days six times. Samples weighing 1.0 g and 15.0 g were used for GC-MS analysis and sensory analysis, respectively.

2.2.5. Oxitest analysis of samples in different packaging materials

Similarly, My 3, Fy 7, and Sy 28 (each 50.0 g) were placed in closed glass, PET, and PP plastic bottles in the dark at ambient temperature for 30 days. The PVs of the samples were monitored during storage and controlled within 15 meq/kg for successful analysis. Samples of 10.0 g were used for Oxitest analysis for the calculation of IPs.

2.3. HS-SPME/GC-MS analysis

For HS-SPME/GC-MS analysis, a modified version of the method reported by Wang et al. (2020) was used. A total of 1.0 g of each sample was placed in a 20 mL vial sealed with an aluminum crimp cap equipped with a needle-pierceable polytetrafluoroethylene/silicone septum. Divinylbenzene/carboxene/polydimethylsiloxane (DVB/CAR/PDMS, 50 μ m/30 μ m coating, 1 cm length) fiber (Supelco Inc., Bellefonte, PA, USA) was used for headspace sampling. Volatile compounds were analyzed on a GC-MS apparatus (QP2010; Shimadzu, Kyoto, Japan) operating in electron ionization mode (EI, 70 eV). Ion source and GC-MS transfer line temperatures were set at 230 °C and 250 °C, respectively. The DB-17MS column (60 m \times 0.25 mm \times 0.25 μ m) was programmed from 40 °C (held for 3 min) to 120 °C at 4 °C/min $^{-1}$ and to 240 °C at 6 °C/min $^{-1}$ (held for 9 min). Helium was used as a carrier gas (1 mL/min $^{-1}$). Compounds with >85% similarity to the NIST library were identified.

Both 2-octanol (0.53 mg/mL, 7 μ L) and o-dichlorobenzene (1.00 mg/mL, 20 μ L) were used as internal standards to quantify the KFC levels in VROs. In a preliminary experiment, there was no significant difference in the quantitative results of the two internal standards. Thus, the levels of different compounds were obtained as mean values from the

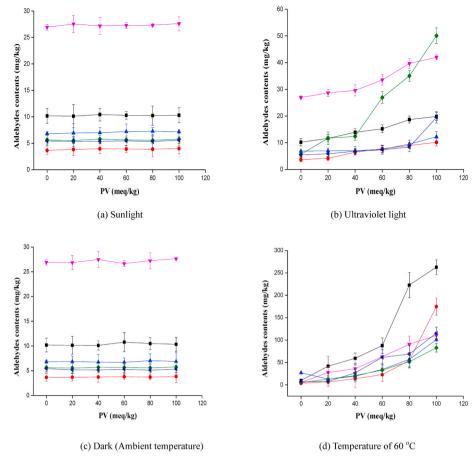


Fig. 1. Effects of different conditions on aldehydes changes (-■-Fy 7 - - Ly 6 - Sy 28 - Wy 3 - Qy 10 - Cy 47).

quantitative results of 2-octanol and o-dichlorobenzene.

2.4. Sensory analysis

Ten trained panelists (five females and five males, aged 25–35 years) from Northwest A&F University were employed to evaluate all of the flavors in the VROs. Different oil samples were coded with three digital codes randomly. Evaluation was carried out at ambient temperature one at a time, with a 5 min interval between samples. Each panelist conducted the evaluation in triplicate with a 30 min interval, and mean values were calculated. The sensory panel smelled the different VROs and rated the intensities of five flavor attributes (i.e., fruity flavor, oil fragrance, nutty flavor, pungent flavor, and rancid smell) with a five-point scale, where 0 indicated that the descriptor was not perceived and 1–10 referred to intensity from the lowest to the highest.

2.5. Contact angle measurements of water and oil droplets on packaging materials

The method for measuring the contact angle of water and oil droplets on packaging materials was based on that described by Huyan et al. (2019). Young's contact angle was measured with a JY–PHB contact angle instrument (Jinhe, Chengde, China). A drop of liquid was deposited on a surface by manually placing a tangent to the drop at its base through the goniometer sessile drop technique. Water was directly deposited on polymer surfaces and angles were calculated after holding the drop in place for 10 s. Contact angles for oils on material surfaces

were immediately calculated when drops were deposited on the surfaces. Contamination, which can affect the accuracy of the contact angle measurements, was prevented by cleaning the solid surfaces, needles, and chamber with acetone before and after every measurement.

2.6. Oxitest analysis

Samples were analyzed on an oxidation test reactor (VELP OXITEST, Rome, Italy). Samples weighing 10.0 g were used for the analysis. Heating temperature and test pressure were set at 110 $^{\circ}\text{C}$ and 6 bar, respectively. When the variation curves of pressure over time changed suddenly (the pressure was <1.5 bar), the apparatus terminated the determination process and then calculated the IPs of samples automatically.

2.7. Statistical analysis

All analyses in this study were performed in triplicate and results are expressed as mean values. The data were subjected to analysis of variance for the determination of significant differences between samples using Minitab 20.0 using the Tukey method. p < 0.05 was considered significant.

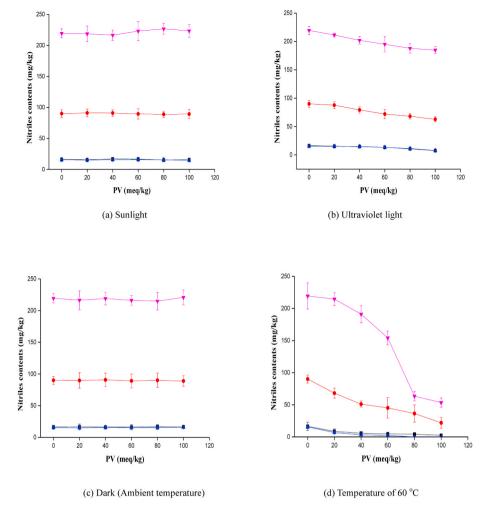


Fig. 2. Effects of different conditions on nitriles changes (→ Sy 28 → My 3 → Qy 10 → Cy 47).

3. Results and discussion

3.1. KFC changes under different light and temperature conditions

The KFCs, including aldehydes, nitriles, pyrazines, and isothiocyanates, in VROs make dominant contributions to the flavor profile with relative odor activity values (ROAVs) \geq 1.0 (Wang et al., 2020; Xu et al., 2017). A total of 23 KFCs (12 aldehydes, 6 nitriles, 3 pyrazines, and 2 isothiocyanates) were measured in the VROs (Tables A2–A7). Furthermore, contents of reducing sugar and thioglycoside may also be different, leading to different contents of their products, namely pyrazines, and nitriles, isothiocyanates, in fresh VROs during roasting.

3.1.1. Effects of different light conditions on KFC changes

Light, especially ultraviolet light, can promote the oxidation of fatty acids to produce large amounts of secondary oxidation products or decompose the flavor-related compounds. The effects of different light conditions on the changes of KFCs in VROs were analyzed as shown in Figs. 1–4.

As shown in Figs. 1–4, there were no significant changes in the four KFCs with increasing PVs under sunlight and dark conditions (p>0.05). Under ultraviolet light, aldehyde levels were significantly increased from 3.654 to 10.183 mg/kg to 10.147–50.076 mg/kg in the six VROs (p<0.05). Because the contents of endogenous antioxidants may vary with the cultivars of the rapeseeds, and therefore caused the differences in the

generation rate of aldehydes in VROs. Although aldehydes account for the oil fragrance and fruity flavor of rapeseed oils, they produce a strong rancid smell at high concentrations (Morales, Rios, & Aparicio, 1997; Giri, Osako, & Ohshima, 2010). Due to the different contents of reducing sugar and thioglycoside in six rapeseeds, their products, namely, nitriles, pyrazines, and isothiocyanates, showed differences in the contents in fresh VROs during roasting (Figs. 2–4). Their levels still showed no significant differences in the range of 0–100 meq/kg under ultraviolet light. Thus, the energy from ultraviolet light was mainly used for the auto-oxidation of fatty acids in oils, whereas ultraviolet light as well as sunlight may not be sufficiently powerful for the further decomposition of nitriles, pyrazines, and isothiocyanates. Consequently, VROs should be kept in the dark to prevent the auto-oxidation of fatty acids.

3.1.2. Effects of different storage temperatures on KFC changes

High temperatures can also promote the oxidation of fatty acids and the decomposition of flavor-related compounds. The levels of aldehydes, nitriles, pyrazines, and isothiocyanates are shown in Tables A8–A13.

As shown in Figs. 1–4, all four KFCs showed no significant changes at ambient temperature (p>0.05). At 60 °C, the levels of aldehydes increased sharply from 3.654 to 10.183 to 82.762–262.580 mg/kg in the range of 0–100 meq/kg (p<0.05), especially at about 60–100 meq/kg (Fig. 1). As shown in Tables A8–A13, pentanal, hexanal, and heptanal from linoleic acid were the main groups of aldehydes increasing in abundance at 60 °C. Notably, the groups of 2-alkenals (mainly *trans-2-*

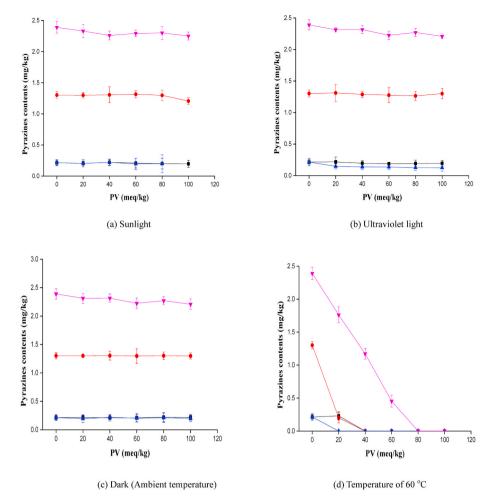


Fig. 3. Effects of different conditions on pyrazines changes (-■- Sy 28 -●- My 3 -▲- Qy 10 -▼- Cy 47).

pentenal and *trans*-2-heptenal) and 2,4-alkadienals (mainly *trans*, *trans*-2,4-heptadienal, and *trans*, *trans*-2,4-decadienal) were also identified in six VROs at 60 $^{\circ}$ C at 60–100 meq/kg, representing the intensification of oxidation-related rancidity, while they were not found at ambient temperature.

High temperatures can promote the thermal degradation of nitriles (mainly 3-methyl-2-butenenitrile and 5-cyano-1-pentene), pyrazines (mainly 2,5-dimethyl-pyrazine), and isothiocyanates (mainly 4-isothiocyanato-1-butene) in VROs (Tables A8–A13). As shown in Figs. 2–4, continuous reductions of nitriles, pyrazines, and isothiocyanates were observed at 60 °C (p < 0.05), with these molecular groups becoming undetectable at the end of the experiment. Therefore, VROs should be protected from high temperatures.

3.1.3. Comparative analysis of KFC changes under different light and temperature conditions

Oil oxidation is the main cause of flavor deterioration, which is influenced by the energy from light or heat (Choe & Min, 2006). An increase in PV indicates the presence of primary oil oxidation, mainly due to hydroperoxide formation, which can be degraded into aldehydes, ketones, and acids, among others in secondary oil oxidation. The formation and degradation of hydroperoxides is a dynamic process during oil oxidation. They are relatively stable with low degradation rates at low temperatures (Choe & Min, 2006; Dolde & Wang, 2011a). Therefore, aldehydes exhibited no significant increases even at high PVs at ambient temperature, and sunlight and dark conditions. Hydroperoxides

exhibited low degradation rates under these conditions, which were still in the primary stage of oil oxidation.

At high temperatures, hydroperoxides are readily decomposed, as shown in Tables A8–A13. Aldehydes (mainly pentanal and hexanal) were generated rapidly at 60 $^{\circ}$ C. Abundant 2-alkenals and 2,4-alkadienals were only measured at 60 $^{\circ}$ C in the range of 60–100 meq/kg, whereas these compounds were absent under other conditions, even at the same PVs. High temperatures exerted powerful effects on hydroperoxide degradation and accelerated secondary oxidation, with the production of pentanal, hexanal, 2-alkenals, and 2,4-alkadienals at high PVs because of the high energy input compared with that under other conditions.

For nitriles, pyrazines, and isothiocyanates, different light conditions and ambient temperature had no significant effects, and continuous reductions in their levels were observed at 60 $^{\circ}$ C. The reason for this might be substantial breakdown of molecular chains at high temperatures compared with that under other conditions (Zhang 2011).

3.2. Determination of flavor-related changes in different packaging materials

Glass and plastic are usually used as packaging for edible oils (Kiritsakis, Kanavouras & Kiritsakis, 2002; Pristouri, Badeka, & Kontominas, 2010). The careful selection of food packaging materials has been shown to confer good quality on vegetable oils (Hu et al., 2020).

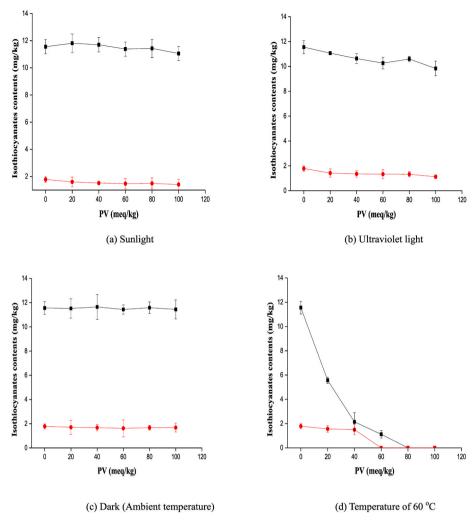


Fig. 4. Effects of different conditions on isothiocyanates changes (→ My 3 → Cy 47).

3.2.1. Changes in the KFCs of samples in the three packaging materials

The performance of glass, PET plastic, and PP plastic bottles for flavor preservation was evaluated. The changes of aldehydes, nitriles, pyrazines, and isothiocyanates of samples in the three packaging materials were also used to characterize the flavors of VROs. The trends of change of these four KFCs in the My 3 sample (used as an example for illustration) in the three packaging materials during storage are plotted in Fig. 5.

As shown in Fig. 5, changes in the KFCs of VROs showed no significant differences among the three packaging materials during 0–60 days. During 60–180 days, KFC levels varied among the different packaging materials. The levels of aldehydes of samples in the PET and PP plastic bottles, mainly hexanal, pentanal, 2-alkenals, and alkadienals, were higher than those in glass bottles, indicating more severe oxidation-related rancidity in the plastic bottles. The levels of nitriles, pyrazines, and isothiocyanates of samples in the glass bottles were higher than those in the PET and PP plastic bottles. The glass bottles showed better performance in terms of preserving the original KFCs. Changes of the four KFCs in Sy 28 and Fy 7 showed similar trends (Figs. A1–A2).

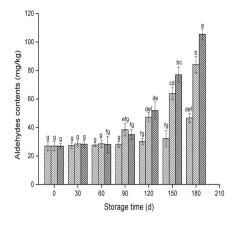
3.2.2. Changes in the sensory scores of samples in the three packaging materials

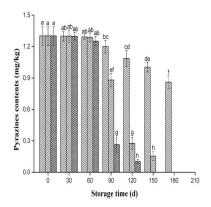
Among the four KFCs, aldehydes account for a pleasant oil fragrance and fruity aroma. However, excessive aldehydes can cause an undesirable rancid smell. Nitriles and isothiocyanates are responsible for the unique pungent aroma of VROs (Gracka, Jeleń, Majcher, Siger, &

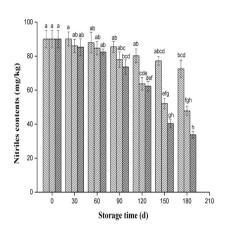
Kaczmarek, 2016), while pyrazines contribute to the nutty aroma (Wang et al., 2020). Thus, five flavor attributes of My 3, Fy 7 and Sy 28 (fruity flavor, oil fragrance, nutty flavor, pungent flavor, and rancid smell) were assessed through sensory descriptive analysis. Sensory scores of Cy 47, Ly 6 and Qy 10 (data were not shown) were very similar to that of My 3, Fy 7 and Sy 28, respectively. Three radar plots were plotted to provide graphical representations of the flavor profiles in My 3 (used as an example for illustration) in glass, PET, and PP plastic bottles during storage (Fig. A3).

Figure A3 shows that My 3 had a remarkable pungent flavor, which was mainly caused by isothiocyanates and nitriles. During storage, this pungent flavor of My 3 packaged with PET and PP plastic bottles gradually decreased in intensity and almost disappeared at the later stage, eventually being replaced by a rancid smell. However, My 3 in glass bottles still had a strong pungent flavor during storage, while the rancid smell was minimal or not perceived. The flavor-related tendencies in Fy 7 (Ly 6) and Sy 28 (Qy 10) were similar to that in My 3 (Figs. A4–A5).

The results of both sensory analysis of the flavor profiles and GC-MS analysis of KFC changes in VROs placed in the three packaging materials indicated that the glass bottles outperformed the PET and PP plastic bottles in terms of preserving the VRO flavors. Because edible oils are generally known to make direct contact with the packaging materials, the surfaces of packaging materials can influence chemical changes in oils, as reported by Hu et al. (2020). Therefore, the hydrophilicity of the surfaces of the packaging materials was further analyzed.







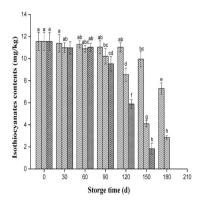


Fig. 5. Effects of different packaging materials on KFC changes in My 3 (Glass PET PP).

Table 1 Induction period values of samples in the three packaging materials. ^a

Packaging Materials	Induction period values (min)		
	Му 3	Sy 28	Fy 7
Glass PET plastic PP plastic	$\begin{array}{c} 226.0 \pm 4.24 \ ^{a} \\ 207.0 \pm 4.24 \ b \\ 203.5 \pm 3.54 \ b \end{array}$	$\begin{array}{c} 310.5 \pm 2.12 \ ^{a} \\ 296.5 \pm 0.71 \ b \\ 278.5 \pm 3.54 \ c \end{array}$	$486.0 \pm 9.90 \stackrel{a}{-} \\ 458.5 \pm 2.12 \ b \\ 452.0 \pm 4.24 \ b$

^a Values followed by different letters (a, b, c) in the same column are significantly different (p < 0.05).

3.3. IPs of samples in the three packaging materials

Oxidation constitutes a major factor behind deterioration of the flavor of edible oils (Xu, Fei, Li, Yu, & Liu, 2015). High IPs of the oil samples represented better oxidation stability (Dolde & Wang, 2011b), which were determined in glass, PET, and PP plastic bottles by Oxitest analysis (Table 1).

As shown in Table 1, IPs of My 3 (226 min), Sy 28 (310 min), and Fy 7 (486 min) in the glass bottles were significantly higher than My 3 (207 and 203 min), Sy 28 (296 and 278 min), and Fy 7 (458 and 452 min) in the PET and PP plastic bottles, respectively. Overall, the oil samples in glass bottles had better performance in terms of oxidation stability than those in the plastic bottles.

3.4. Surface hydrophilicity of packaging material

Young's contact angles were used to characterize the hydrophilicity

Table 2Contact angle measurement of water and oil droplets on polymer surfaces. ^a

Packaging materials	Contact angles (°)	
	Water	Vegetable oils
Glass	$44.53\pm4.45~^{a}$	39.15 ± 1.25 ^a
PET plastic	$59.80 \pm 9.00^{\ b}$	39.50 ± 2.40 ^a
PP plastic	77.10 \pm 2.70 $^{\mathrm{c}}$	55.20 ± 3.50 b

^a Values followed by different letters (a, b, c) in the same column are significantly different (p < 0.05).

of material surfaces and investigate the shape of oil or water droplets held for 10 s on the material surfaces (Fig. A6). The contact angles of My 3, Sy 28, and Fy 7 on each surface were measured. No significant differences between them were observed (p < 0.05). Average contact angles of VROs and water droplets are presented in Table 2.

As shown in Table 2, water droplets on glass (44.53°) had smaller contact angles than those on PET (59.80°) and PP (77.10°) plastics (p < 0.05). This suggested that glass was more hydrophilic than PET and PP plastics. The order of hydrophilicity (polar) was as follows: glass > PET plastic > PP plastic. Moreover, the contact angles of VRO drops on glass and PET plastic were similar to each other, and lower than the contact angle on PP plastic.

Based on the above results, the oxidation stability and flavors of samples in glass bottles were superior to that of samples in PET and PP plastic bottles. As Huyan et al. (2019) concluded, complex packaging materials with high surface hydrophilicity can retard oil oxidation. The surface of glass with better hydrophilicity than PET and PP plastics is

likely to attract more compounds, including hydroperoxides and compounds with polar functional groups. As a result, few polar compounds were positioned at air/oil interfaces, resulting in increased surface tension and decreased oxygen diffusion, which in turn retarded the oil oxidation (Huyan et al., 2019). Therefore, glass bottles are preferable for extending the shelf life.

For nitriles, pyrazines, and isothiocyanates, a hypothetical situation involving VROs not being packaged at all could be introduced for discussion. These KFCs with polar functional groups were likely to integrate with the air in order to separate from low-polar oil phases, resulting in the loss of VRO flavors from the perspective of the polarity of media. When more hydrophilic packaging materials were present, these KFCs tended to combine with the materials to decrease volatilization. Consequently, the glass bottles showed better performance in preserving the flavor of VROs than the PET and PP plastic bottles.

4. Conclusion

In this study, aldehydes, nitriles, pyrazines, and isothiocyanates were used as KFCs of VROs to characterize the whole flavor of these oils. The changes of VROs under different light and temperature conditions were measured in the range of 0-100 meg/kg. Glass, PET, and PP plastic bottles were selected to investigate their performance at preserving KFCs during storage. IPs of samples in the three packaging materials were determined to evaluate their oxidation stability through Oxitest analysis. The surface hydrophilicity of the materials was also characterized by measuring Young's contact angle. The results revealed that the aldehyde levels of the VROs dramatically increased at 60 °C and under ultraviolet light. Among them, alkenals were only detected in the range of 60-100 meq/kg at 60 °C, and nitriles, pyrazines, and isothiocyanates were not detected at the end of the experiment at 60 °C. High energy associated with a high temperature accelerated oxidation as well as fracture of the molecular chains. The IPs of samples in glass bottles with enhanced hydrophilicity were higher than those of the samples in PET and PP plastic bottles, indicating superior oxidation stability. The surface of glass was determined to have higher hydrophilicity, which is likely to attract more compounds, including hydroperoxides and compounds with polar functional groups. In this way, the surface tension at the oil/air interface was increased, which restricted oxygen diffusion, and KFCs were attracted by the glass surface, reducing their volatilization to the air. Thus, it is preferable to preserve VROs in glass bottles.

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Ethical approval

This article does not contain any studies with human or animal subjects.

Declaration of competing interest

Mengzhu Wang, Mengmeng Wang, Zongyao Huyan, Qi Li, Keqing Hu, Jinwei Li, Xiuzhu Yu declare that they have no conflict of interest.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.lwt.2022.113089.

References

- Choe, E., & Min, D. (2006). Mechanisms and factors for edible oil oxidation. Comprehensive Reviews in Food Science and Food Safety, 5, 169–186.
- Dolde, D., & Wang, T. (2011a). Oxidation of corn oils with spiked tocols. Journal of the American Oil Chemists' Society, 88(11), 1759–1765.
- Dolde, D., & Wang, T. (2011 b). Oxidation of crude corn oil with and without elevated tocotrienols. *Journal of the American Oil Chemists' Society*, 88(9), 1367–1372.
- Giri, A., Osako, K., & Ohshima, T. (2010). Identification and characterisation of headspace volatiles of fish miso, a Japanese fish meat based fermented paste, with special emphasis on effect of fish species and meat washing. Food Chemistry, 120, 621–631.
- Goicoechea, E., & Guillén, M. (2014). Volatile compounds generated in corn oil stored at room temperature. Presence of toxic compounds. European Journal of Lipid Science and Technology, 116, 395–406.
- Gracka, A., Jeleń, H., Majcher, M., Siger, A., & Kaczmarek, A. (2016). Flavoromics approach in monitoring changes in volatile compounds of virgin rapeseed oil caused by seed roasting. *Journal of Chromatography A*, 1428, 292–304.
- Hu, K., Huyan, Z., Ding, S., Dong, Y., & Yu, X. (2020). Investigation on food packaging polymers: Effects on vegetable oil oxidation. Food Chemistry, 315, 126299.
- Huyan, Z., Ding, S., Mao, X., Wu, C., & Yu, X. (2019). Effects of packaging materials on oxidative product formation in vegetable oils: Hydroperoxides and volatiles. Food Packaging and Shelf Life, 21, 100328.
- Jing, B., Guo, R., Wang, M., Zhang, L., & Yu, X. (2019). Monitoring flavor variations in virgin rapeseed oil caused by seed roasting. Lebensmittel-Wissenschaft und -Technologie- Food Science and Technology, 126, 109301.
- Kanavouras, A., Hernandez-Münoz, P., Coutelieres, F., & Selke, S. (2004). Oxidation derived flavor compounds as quality indicators for packaged olive oil. *Journal of the American Oil Chemists' Society*, 81, 251–257.
- Kiritsakis, A., Kanavouras, A., & Kiritsakis, K. (2002). Chemical analysis, quality control and packaging issues of olive oil. European Journal of Lipid Science and Technology, 104, 628–638.
- Mahesar, S. A., Bendini, A., Cerretani, L., Matteo, B. C., & Sherazi, S. T. H. (2010).
 Application of a spectroscopic method to estimate the olive oil oxidative status.
 European Journal of Lipid Science and Technology, 112(12), 1356–1362.
- Malheiro, R., Casal, S., Rodrigues, N., Renard Catherine, M., & Alberto, P. (2018).
 Volatile changes in cv. Verdeal Transmontana olive oil: From the drupe to the table, including storage. Food Research International, 106, 374–382.
- Mao, X., Zhao, X., Huyan, Z., Liu, T., & Yu, X. (2019). Relationship of glucosinolate thermal degradation and roasted rapeseed oil volatile odor. *Journal of Agricultural* and Food Chemistry, 67, 11187–11197.
- Mishra, R., & Sharma, H. K. (2011). Effect of packaging materials on the storage stability of physically refined rice bran oil and its blends. African Journal of Food Science, 12, 676–685.
- Morales, M., Rios, J., & Aparicio, R. (1997). Changes in the volatile composition of virgin olive oil during oxidation: Flavors and off-flavors. *Journal of Agricultural and Food Chemistry*, 45, 2666–2673.
- Njoroge, H., Ukeda, A., & Sawamura, M. (1996). Changes in the volatile composition of Yuzu (Citrus Junos tanaka) cold-pressed oil during storage. Journal of Agricultural and Food Chemistry, 44, 550–556.
- Pristouri, G., Badeka, A., & Kontominas, M. (2010). Effect of packaging material headspace, oxygen and light transmission, temperature and storage time on quality characteristics of extra virgin olive oil. Food Control, 21, 412-418.
- Ramezani, R. (2004). The effect of packaging materials and storage condition on the oxidative stability of refined sunflower oil. Food Science and Technology International Tokyo, 10, 350–354.
- Silvagni, A., Franco, L., Bagno, A., & Rastrelli, F. (2012). Thermo-induced lipid oxidation of a culinary oil: The effect of materials used in common food processing on the evolution of oxidised species. *Food Chemistry*, 133(3), 754–759.
- Wang, T., Jiang, Y. Z., & Hammond, E. G. (2005). Effect of randomization on the oxidative stability of corn oil. *Journal of the American Oil Chemists' Society*, 82(2), 111–117.
- Wang, M., Zhang, J., Chen, J., Jing, B., Zhang, L., & Yu, X. (2020). Characterization of differences in flavor in virgin rapeseed oils by using gas chromatography-mass spectrometry, electronic nose, and sensory analysis. European Journal of Lipid Science and Technology, 122, 1900205.
- Xu, L. R., Fei, T., Li, Q., Yu, X. Z., & Liu, L. (2015). Qualitative analysis of edible oil oxidation by FTIR spectroscopy using a mesh "cell. Analytical Methods, 7, 4328–4333.
- Xu, L., Yu, X., Li, M., Chen, J., & Wang, X. (2017). Monitoring oxidative stability and changes in key volatile compounds in edible oils during ambient storage through HS-SPME/GC-MS. *International Journal of Food Properties, S3*, S2926–S2938.
- Zhang, S. B., Lu, Q. Y., Yang, H. S., Li, Y., & Wang, S. (2011). Aqueous enzymatic extraction of oil and protein hydrolysates from roasted peanut seeds. *Journal of the American Oil Chemists' Society*, 88(5), 727–732.