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Impact of UV-light irradiation on sensory properties, volatile, fatty acid, and tocopherol composition of peanuts (*Arachis hypogaea L.*)

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ABSTRACT

Shelf life of peanuts is determined by their susceptibility to lipid oxidation. However, oxidation stability tests require accelerated conditions to speed up oxidation processes. This work aimed to test the use of UV radiation to accelerate the oxidation in peanuts. Shelled and fried peanuts were irradiated for 3 and 7 days, respectively with a UV lamp. The content of fatty acids, tocopherols, and volatile compounds was determined followed by colour and sensory properties of samples. Obtained results were compared with samples heated for 2 months at 70 °C in an oven. Tocopherol content showed oxidation process had just started after 3 days of UV irradiation. Moreover, aldehydes content increased 8.40 times and 11.77 times at 3 days and 7 days, respectively. Aroma, odour, sweetness scores, and a* value of peanuts were lower and permanency and rancity scores were higher after the oxidation process. Thermal treatment showed an increase in colour and b* value and a reduction in a crunchiness score highlighting temperature possible causes chemical changes which are not related to the oxidation process. Acceleration of oxidation with UV radiation can be used to study peanuts oxidation with less impact on sample colour and crunchiness when the sensorial analysis was performed.

1. Introduction

In the last five years, the production of peanuts (*Arachis hypogaea* L.) has increased by 9.7% reaching 53,638,932 tonnes in 2020. Peanuts are rich in oil (47–50 wt%) and protein (around 30 wt%). The oil fraction of peanuts is composed of unsaturated fatty acids, mainly oleic (42–52%) and linoleic acid (32–37%) (Dun et al., 2018; Juan-Polo et al., 2022). In addition, it also contains other minor compounds such as tocopherols, and volatile compounds among others (Zhang, Li, Cao, Wang, & Xue, 2020).

Different factors such as temperature, humidity, light, and the oxygen amount affect the shelf life of nuts (S. F. Mexis, Badeka, Riganakos, Karakostas, & Kontominas, 2009). Thus, storage conditions must be carefully selected, including low temperatures, i.e., from 4 °C to 15 °C; kernel moisture content around 2.5 wt%, relative humidity of about 40–60%, oxygen concentration less than 2.5%, and darkness, to slow down the oxidation reactions. Nevertheless, even at low temperatures, oxidation of peanuts proceeds in presence of oxygen (Mildner-Szkudlarz, Jeleń, Zawirska-Wojtasiak, & Wąsowicz, 2003).

Since nuts oxidation proceeds at low speed in normal storage conditions, oxidation stability tests of peanuts are time-consuming. Hence, Raisi et al. demonstrated ground almonds could remain fresh for 7 months (Raisi, Ghorbani, Sadeghi Mahoonak, Kashaninejad, & Hosseini, 2015). In ground peanuts, P.L. López et al. proved oleic acid concentration only decreased 1% at 25 °C for 21 days (López, Marchesino, Grosso, & Olmedo, 2022).

Consequently, different factors must be modified to accelerate the oxidation and reduce the assays duration. The application of high temperatures (between 40 °C and 70 °C) is the most employed approach. Recently, it was proposed that 1 day at 60 °C is equivalent to 8.79 days at 25 °C in terms of sample oxidation (López et al., 2022). However, it is important to note that, at high temperatures, oxidation products are different in comparison with the degradation process at ambient temperature. First of all, temperature roasts peanuts and they acquire a darker colour due to the b* parameter diminution (Bagheri, Kashaninejad, Ziaiifar, & Aalami, 2019). Crunchiness decreases when peanut is oxidised, but this reduction is lower when high temperature is applied (Braddock, Sims, & O'keefe, 1995). In an accelerated oxidation process,

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sweetness decreased, while bitterness increased. J.B. Mugendi et al. and E.G. Abegaz et al. demonstrated this fact in peanuts kept at 40 °C and 21 °C, respectively (Abegaz, Kerr, & Koehler, 2004; Mugendi, Sims, Gorbet, & O'Keefe, 1998). Nevertheless, K.J.H. Warner et al. pointed out there were no significant changes in sweetness when peanuts were stored at 65 °C (Warner, Dimick, Ziegler, Mumma, & Hullender, 1996). In relation to the volatile profile, compounds like 3-ethyl-1,5-octadiene or 1-octen-3-ol were only detected when temperature (60 °C) was used (Xu, Yu, Li, Chen, & Wang, 2018). Consequently, changes in the volatile compounds produced at high temperatures result in changes in the sensory properties of peanuts (López-De-Dicastillo et al., 2012).

UV radiation has been used in peanut treatments processes such as allergens reduction, enhance *trans*-resveratrol production or reduction of aflatoxin levels (Diao et al., 2015; Li et al., 2019; Pi et al., 2021). The application of UV irradiation to peanuts results in the formation of free radicals, such as lipid radicals, superoxide radicals and H_2O_2, and catalyses other oxidation processes (Shen et al., 2014). Concerning the unsaturated fatty acids present in peanuts, X. Shen et al. used a UV lamp (power 36 W) with irradiation intensity of 6.4 mW/cm^2 to evaluate the effects of UV-irradiation on them (Shen et al., 2014). A reduction of 10% and 15% in the oleic and linoleic acid content, respectively, after 40 min of UV irradiation was reported. This reduction strongly depends on the irradiation time (Shen et al., 2014).

Despite these works involving the irradiation of peanuts with UV, to the best of our knowledge, the use of UV radiation to accelerate the oxidation of peanuts and the impact of exposure to UV on the chemical composition and sensory properties of peanuts has not been deeply investigated. Thus, the main aim of this research is to investigate the UV radiation as a tool to accelerate the oxidation of peanuts and compare the chemical and sensory changes to those attained by using high temperatures as a mean for enhancing oxidation.

For that purpose, peanut samples were irradiated by using UV radiation and results were compared with a conventional oxidation treatment by using high temperature. Chemical components, including fatty acid composition, tocopherols profile and volatile compounds, have been monitored after 3 and 7 days of UV irradiation exposure. In addition, a correlation between changes in chemical composition and sensory properties because of the different degradation processes has also been carried out.

2. Materials and methods

2.1. Reagents

Methanol (HPLC grade), isopropanol (HPLC grade) and n-hexane (99%, GC grade) were purchased from Panreac (Barcelona, Spain). Sodium methoxide 25 wt % solution in methanol, sulphuric acid (98%), tocopherol homologues (α-tocopherol, β-tocopherol, γ-tocopherol and δ-tocopherol) standards and a certified reference mixture of 37 fatty acid methyl esters (FAMEs) in dichloromethane (F.A.M.E. Mix. C4–C24) were acquired from Sigma–Aldrich Inc. (St. Louis, MO, USA).

2.2. Samples

The commercially packaged peanut kernels (shelled and fried with salt) were obtained from a Spanish supermarket and were kept in a cool place (storage temperature of 4 ± 1 °C) under modified atmosphere (N_2) until their analysis. Samples were immediately prepared before analysis in order to protect them against oxidation. Peanuts were ground in a domestic electric grinder (Moulinex, Barcelona, Spain) before the analysis, except for sensory analysis, since panelists ate entire peanuts.

2.3. Oxidation treatments

In this study, the oxidation of the peanut samples by using UV-light and the oxidation by temperature were compared. Concerning the oxidative treatment by using temperature, 1 kg of peanuts were placed into 39 cm × 26 cm x 7 cm open container in an oven (Selecta, Barcelona, Spain) at 70 °C for 2 months (Beltrán Sanahuja, Maestre Pérez, GranéTeruel, & Martín Carratalá, 2009). This time was selected to ensure that oxidation was in advanced state while the peanuts remained edible. Regarding the UV irradiation treatment, peanuts (1 kg) were placed into an open container too to expose into a light box for photography containing a UV tube (wavelength of 254 nm) for their irradiation. Samples were irradiated at 22 cm with a 230V (50Hz) UV lamp (Inecsa, Barcelona, Spain) for 3 and 7 days, respectively. A longer UV treatment rendered the peanuts inedible.

2.4. Volatile compounds analysis

2.4.1. Box-Behnken experimental design (BBD)

Volatile compounds were determined by headspace-solid-phase microextraction (HS-SPME) followed by gas chromatography (CG) coupled to mass spectrometry (MS). An optimization of extraction time, equilibration time and temperature using a response surface methodology (RSM) was proposed. To investigate the influence of the extraction time (min) (A); the equilibration time (min) (B) and the extraction temperature (°C) (C) at three levels on the selected response, a Box-Benhken Design was used. As it is shown in Table 1, a set of 16 experiments with 4 centre points were carried out in a randomised order. The sum of the areas of hexanal, heptanal, octanal and nonanal was used as the selected response. All of these volatile compounds have been reported as main aldehydes generated in the oxidation process in different nuts (Costa De Camargo et al., 2016; Mildner-Szkudlarz et al., 2003).

The significance of the effects was checked by using the analysis of variance (ANOVA). Multiple regression analysis was performed to illustrate the effect of factors on the response in a second-degree polynomial equation (Eq. (1)), where Y is the concentration, β_0 is the model constant; β_i represents the linear coefficients; β_{ii} the quadratic coefficients; β_{ij} the interaction coefficients and X_i the different factors (Sahu et al., 2018).

$$Y = \beta_0 + \sum_{i=1}^{4} \beta_i X_i + \sum_{i=1}^{4} \beta_{ii} X_{ii}^2 + \sum_{i=1}^{3} \sum_{j=i+1}^{4} \beta_{ij} X_i X_j \quad \text{(Eq. 1)}$$

The adequacy of the model was determined by evaluating the lack of fit, the coefficient of determination (R^2) and F-test obtained from the analysis of variance (ANOVA).

2.4.2. Headspace solid phase microextraction (HS-SPME) and GC-MS conditions

The volatile compounds were determined by headspace-solid-phase microextraction (HS-SPME) followed by GC-MS analysis by using an Agilent 6890N GC System (Palo Alto, CA, USA) coupled to a triple quadrupole mass spectrometry (Agilent 5973N, Palo Alto, CA, USA) equipment. The automated sample preparation procedure was performed with a Gerstel MultiPurpose autosampler (TDS-2, Gerstel GmbH, Mülheim an der Ruhr, Germany).

Samples (0.5 g of ground peanuts) were heated at 70 °C for 30 min. Afterwards, the SPME fiber (DVB/CAR/PDMS SPME, 50/30 m, StableFlex, 1 cm long) fixed on the mechanical arm was exposed to headspace of sample vial for extraction of the compounds at 70 °C, over 15 min. After exposition, the fiber was immediately desorbed for 3 min at 250 °C into the GC injector port in splitless mode. A DB-624 column (30 m × 250 μm x 1.4 μm; Agilent, California, USA) was used which was programmed from 50 °C to 250 °C (hold 12 min) at 10 °C min^{-1}. Helium was used as carrier gas (1 mL min^{-1}). MS data were recorded between 30 and 550 m/z with an electron energy of 70 eV. The temperature values of the ion source and the transfer line were 230 and 150 °C, respectively. Identification was performed by using the NIST mass spectral library. All samples were analysed in triplicate.

Table 1
Measured and predicted absolute sum of areas of hexanal, heptanal, octanal and nonanal and desirability values in Box–Behnken design (BBD).

Run	Variables			Response		Desirability	
	A	B	C	Observed	Predicted	Observed	Predicted
1	30.0	15.0	30.0	9.5	10.0	0.63	0.66
2	17.5	0.0	30.0	6.0	5.4	0.39	0.35
3	17.5	30.0	30.0	6.4	6.2	0.42	0.40
4	5.0	15.0	30.0	2.7	3.1	0.16	0.19
5	17.5	15.0	52.5	10.8	12.2	0.72	0.81
6	17.5	15.0	52.5	11.7	12.2	0.78	0.81
7	5.0	30.0	52.5	7.4	7.2	0.48	0.47
8	5.0	0.0	52.5	3.7	3.9	0.23	0.24
9	17.5	15.0	52.5	13.9	12.2	0.93	0.81
10	17.5	15.0	52.5	12.5	12.2	0.83	0.81
11	30.0	30.0	52.5	14.1	13.9	0.94	0.93
12	30.0	0.0	52.5	12.7	12.9	0.85	0.86
13	30.0	15.0	75.0	14.9	14.6	1.00	0.97
14	17.5	30.0	75.0	10.6	11.2	0.70	0.74
15	17.5	0.0	75.0	7.5	7.7	0.49	0.51
16	5.0	15.0	75.0	6.3	5.8	0.41	0.38

2.5. Fatty acids and tocopherols analysis

Fatty acids and tocopherols analysis were carried out following the procedure as described previously (Juan-Polo et al., 2022). Instrumental conditions used for fatty acid (GC-MS) and tocopherol (HPLC) determination have been described elsewhere (Juan-Polo et al., 2022).

2.6. Instrumental colour analysis

The colour of grounded peanuts was measured by using a spectrophotometer Konica CM-3600d (Konica Minolta Sensing Europe, Valencia, Spain) by using the CIELAB colour notation system (International Commission on Illumination). The L*, a*, and b* represent three dimensions of a measured colour which gives specific colour values of the material.

The measured coordinates were used to calculate the total colour difference (ΔE) with respect to the reference peanut, as given by Equation (2). All samples were analysed in triplicate.

$$\Delta E = \sqrt{\left(L^*_{REF} - L^*\right)^2 + \left(a^*_{REF} - a^*\right)^2 + \left(b^*_{REF} - b^*\right)^2} \quad (2)$$

2.7. Sensory evaluation

Sensory analysis was performed by using a 16-member panel, 9 males and 7 females, with ages between 19 and 25 years, recruited from the university community. All of them were given a previous learning course of 60 h about sensory analysis and evaluation.

A general descriptive sensory analysis was carried out (Asociación Española de Normalización., 2017, 2018). Seven sensory attributes, directly dependent on lipid oxidation, were studied: aroma peanut, colour intensity, crunchy, peanut odour, permanency, rancity and sweetness. Firstly, the attributes were generated and defined by the panel, which were trained to recognize all of them. Concerning the "rancid" attribute, the judges were provided with oxidised and fresh peanuts to distinguish rancid and fried-peanut attributes. In a second stage, oxidised peanut samples were presented to the panellists for practice rating sessions. Panellists were tested for performance during training for precision and accuracy (agreement with accepted sensory values for fresh and oxidised peanuts).

Five peanuts per treatment were served at room temperature in plastic containers with lids labelled with 3-digit random codes. Panellists were asked to taste a control, low and high oxidation levels using UV and samples oxidised for two months in an oven. Quantitative descriptive analysis was conducted using a 10-point unipolar category numerical scale to evaluate the intensity of the attributes, where 1 was no detection, and 10 was the maximum intensity (The Spanish Association for Standardisation, 2006). The overall acceptability of each sample was measured asking if the sample was edible. During the training sessions, panellists were given reference samples with anchor points for each attribute. Distilled water and unsalted bread were provided to rinse mouths between samples. Evaluations were conducted in partitioned booths and a 10-min break was taken between samples.

2.8. Statistical analysis

Statgraphics Centurion XVI software (version 7.0.0, Stat-Ease, Inc., Minneapolis, MN, USA) was used to maximise the response obtained from the fitted BBD model. The satisfactoriness of the fitted model was evaluated using the lack of fit value, the coefficient of determination (R^2) and adjusted R^2 obtained from the analysis of variance (ANOVA). Statistical significance of model parameters was determined at $\alpha = 0.05$. Data were subjected to independent samples *t*-test and ANOVA by using SPSS software, ver. 15.0 (IBM, Chicago, IL, USA). A Tukey test was assessed at a $p \leq 0.05$ significance level to obtain differences between values.

3. Results and discussion

3.1. Tocopherol and fatty acid composition in raw and oxidised peanuts

Tocopherols are natural antioxidants that prevent lipid oxidation by interrupting the initiation or propagation step of lipid oxidation reactions (Martín, Asensio, Nepote, & Grosso, 2018; Zhang et al., 2020). Consequently, the degradation process in peanuts starts with the oxidation of tocopherols. Then, the oxidation of fatty acids becomes significant after an induction period during which antioxidants are destroyed (Canavar, 2015). Fatty acids and tocopherol contents of the samples analysed are shown in Fig. 1.

As other authors have reported, the main tocopherols in peanut samples are α- and γ-tocopherol (Juan-Polo et al., 2022; Kamal-Eldin, 2006; Martín et al., 2018; Siddeeg & Xia, 2015). Tocopherol content depends on geographical origins as well as the sample processing method (Juhaimi et al., 2018). However, γ-tocopherol is usually the major isomer with contents ranging between 1 and 6 mg per 100 g of peanut (Juan-Polo et al., 2022). In the present study, all tocopherol isomers vary significantly when the two accelerated oxidation methodologies were used. It can be highlighted that the irradiation of peanuts using UV light for 3 days led to a reduction of 4% in α-tocopherol, 39% in β-tocopherol and 31% in γ-tocopherol contents whereas no changes in the δ-tocopherol were registered. Increasing the duration of UV treatment resulted in a 27% decrease in the level of the δ-tocopherol. Oxidative effect of UV irradiation has been also demonstrated in olive oil

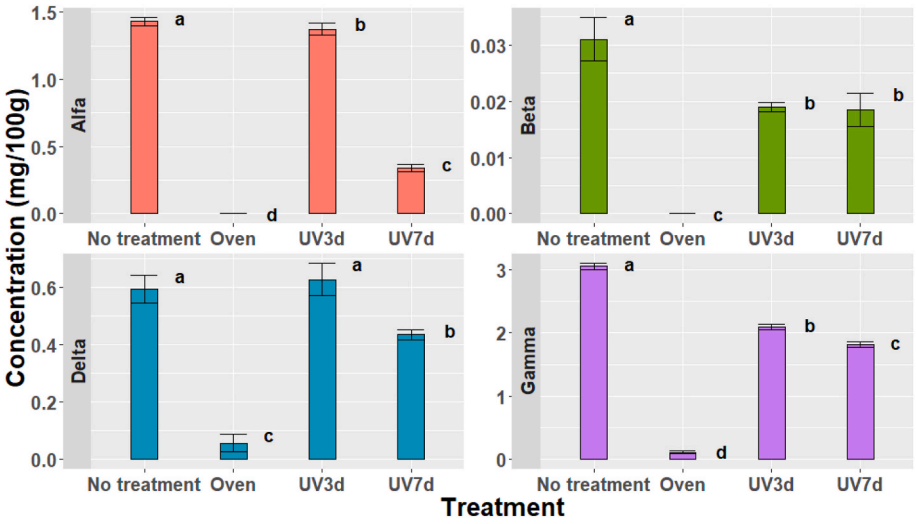

Fig. 1. Tocopherol concentration (mg 100 g^{-1} peanut) as a function of accelerated oxidation conditions. Results expressed as the mean ± standard deviation of three replicates (α = 0.05). Different letters within the same graph indicate statistically significant different values ($p < 0.05$).

(Poulli, Mousdis, & Georgiou, 2009).

Concerning peanut samples oxidised by temperature, the obtained results showed that the compounds α- and β-tocopherol were not detected in any sample after the treatment and the content of γ- and δ-tocopherol was significantly lower. Losses of 91% in the total tocopherol content were obtained after samples being 2 months in an oven. These results showed that the high-temperature treatment produced higher oxidation of the sample, however, it should be noted that with the 7-day UV treatment there was already a 49% drop in the overall level of tocopherols, indicating that the oxidation process was accelerated when UV radiation was applied. In both treatments the degradation rate of tocopherols was: α-tocopherol > γ-tocopherol≈ β-tocopherol > δ-tocopherol. Similar results were reported by Franklin et al. (Franklin et al., 2017).

Six main fatty acids were identified in peanut samples (Table 2): Palmitic acid (C16:0), stearic acid (C18:0), oleic acid (C18:1), linoleic acid (C18:2), eicosenoic acid (20:1), and behenic acid (22:0). As was stated previously (Canavar, 2015), oleic acid was the main fatty acid in peanuts, with an abundance of 35 wt % approximately. The differences in fatty acid composition obtained among unoxidised and oxidised peanuts during the UV treatment were not significant. Öner Canavar demonstrated that when α-tocopherol and γ-tocopherol had decreased 51% and 46%, respectively, after 2 months at room temperature, fatty acid content had not decreased yet (Canavar, 2015). After the oxidation treatment at 70 °C for 2 months, the content of unsaturated fatty acids experienced a significant decrease of 12%, 16% and 36% for C18:1; C20:1 and C18:2, respectively, as corresponding with a more advanced oxidation status. The oxidation was more effective for C18:2. This was in accordance with previous studies carried out in nut samples oxidised by temperature that state linoleic acid reacts with oxygen 10 times faster than oleic acid does (Beltrán Sanahuja et al., 2009; Coultate, 2007).

Taking all that into account, chemical composition confirmed that the peanut samples which were in the oven were more oxidised than those which were under UV light. Although, it is remarkable the reduction in tocopherols in only 7 days of UV light exposure.

3.2. *Optimization of HS-SPME procedure for volatile compounds determination in peanut samples*

The optimization of HS-SPME extraction procedure of volatile compounds in peanuts was carried out to obtain the optimal levels of the three studied independent variables: A, extraction time (min); B, equilibration time (min); C, extraction temperature (°C), their quadratic effects and their interactions, leading to the highest headspace areas of hexanal, heptanal, octanal and nonanal.

The pareto chart estimates the statistical significance of the factors, and interactions between them, that had the greatest effect on the

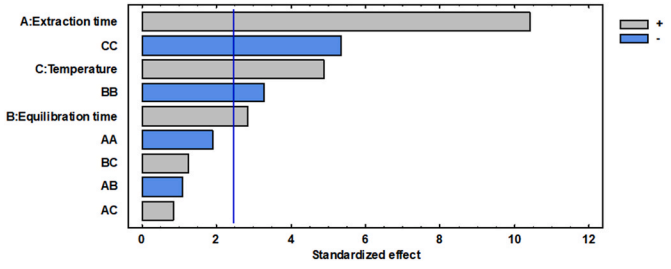

Fig. 2. Pareto chart of factors and interactions obtained from BBD for the response.

Table 2
Fatty acids (g 100 g^{-1} peanut) concentration as a function of storage time for accelerated oxidation conditions by UV light and thermal treatment.

Sample Treatment	Exposure time	Fatty acids (g $100g^{-1}$ peanut)					
		C16:0	C18:0	C18:1	C18:2	C20:1	C22:0
UV	0 days	2.347 ± 0.018 [a]	0.812 ± 0.008 [a]	35.4 ± 0.8 [a]	5.06 ± 0.07 [a]	0.218 ± 0.007 [a]	0.649 ± 0.015 [a]
	3 days	2.23 ± 0.04 [a]	0.761 ± 0.014 [a]	35.6 ± 0.3 [a]	4.67 ± 0.06 [a]	0.201 ± 0.006 [a]	0.602 ± 0.015 [a]
	7 days	2.38 ± 0.13 [a]	0.8 ± 0.05 [a]	36 ± 2 [a]	4.2 ± 0.3 [a]	0.22 ± 0.018 [a]	0.65 ± 0.04 [a]
Oven	0 days	2.347 ± 0.018 [a]	0.812 ± 0.008 [a]	**35.4 ± 0.8** [a]	**5.06 ± 0.07** [a]	**0.218 ± 0.007** [a]	0.649 ± 0.015 [a]
	2 months	2.24 ± 0.17 [a]	0.77 ± 0.05 [a]	**31 ± 2** [b]	**3.23 ± 0.16** [b]	**0.183 ± 0.014** [b]	0.63 ± 0.05 [a]

* Results expressed as the mean ± standard deviation of three replicates (α = 0.05).
** Different superscripts for each compound within the same column and storage treatment indicate statistically significant different values ($p < 0.05$).

response (Fig. 2). The extraction time (A) was the most significant factor that affected the extraction of volatiles, followed by the quadratic term $(C)^2$, extraction temperature (C), the quadratic term $(B)^2$ and, finally, the equilibration time (B). Regarding the significant and positive effect of the extraction time, in reported works, it has been confirmed that longer times led to greater retention of the substances until fiber saturation (García, Martínez, Landete, Moya, & Sanahuja, 2021; Trujillo-Rodríguez et al., 2014). However, extraction time depends on the compound. In general, for small compounds, such as hexanal, the signal decreased after 10–20 min exposure of the fibre to the headspace (Ezquerro, Pons, & Tena, 2003; Pastorelli, Valzacchi, Rodriguez, & Simoneau, 2006). Increasing the extraction temperature has been reported to be a good way of improving the extraction recovery (García, Pérez, Butsko, Moya, & Sanahuja, 2020), but high temperatures are also associated with the unwanted generation of artefacts such as CC interactions, which have been shown to have negative effects (Clarke, Mannion, O'Sullivan, Kerry, & Kilcawley, 2019). In hazelnuts, Pastorelli et al. tested different temperatures (50–80 °C) and found at 60–70 °C the maximum peak area of hexanal was obtained (Pastorelli et al., 2006). Finally, the effect of equilibration time is positive. The use of equilibration times equal to 30 min is widely employed in studies analysing volatile compounds in peanuts (Baker et al., 2003; Costa De Camargo et al., 2016; Xu et al., 2018).

The final second-order polynomial equation is detailed in Equation (3), where A, B and C represent extraction time, equilibration time, and extraction temperature, respectively:

$$\text{Sum of Areas} = -1.56 \times 10^8 + 5.00 \times 10^6 * A + 2.53 \times 10^6 * B + 6.12 \times 10^6 * C - 6.43 \times 10^4 * A^2 - 3.08 \times 10^4 * AB + 1.61 \times 10^4 * AC - 7.72 \times 10^4 * B^2 + 1.98 \times 10^4 * BC - 5.60 \times 10^4 * C^2 \quad (3)$$

Analysis of variance (ANOVA) was performed to monitor the effect of the studied variables in the response and to evaluate the reliability of the fitted model. A high value was obtained for the coefficient of determination (R^2) of 96.89, with adjusted R^2 values quite close to R^2 (92.22), confirming the accuracy of the fitted model. A value of 0.8135 ($p > 0.05$) was obtained for the 'Lack of Fit' test suggesting that the model was reliable in predicting the response. Therefore, the model explained the response adequately. The optimal extraction conditions obtained in the present study were 30 min of equilibration time, and 15 min of extraction time at 70 °C of temperature. Similar conditions have been employed previously in the extraction of volatile compounds in peanuts (Baker et al., 2003).

After the optimization of the volatile extraction parameters, the optimal conditions were employed in the extraction of volatile compounds in peanut samples. Identified compounds (Table 3) were grouped under five headings: aldehydes, acids, alkenes, furans and pyrazines.

Aldehydes (hexanal, heptanal, octanal and nonanal) were the most abundant group of volatiles associated with rancid nuts (López et al., 2022; Mildner-Szkudlarz et al., 2003; Stamatios F. Mexis & Kontominas, 2009). The registered signal of aldehydes increased with increasing oxidation either by irradiation with UV or by heating. Initially (day 0), the signal of hexanal, typically employed as an indicator of lipid oxidation (Grosso & Resurreccion, 2002; S. Y. Lee, Trezza, Guinard, & Krochta, 2002), was higher than the heptanal, octanal and nonanal signals. As oxidation proceed, nonanal was the main oxidation aldehyde produced in both treatments.

Nonanal originates from the autoxidation of oleic acid (Costa De Camargo et al., 2016). Oleic acid was the major fatty acid in peanut samples (around 35%), so nonanal represented 43% and 54% of total aldehydes after 7 days of UV-light and 2 months in the oven, respectively. Wang et al. also identified nonanal as the major aldehyde in roasted peanuts after 8 weeks stored at 21 °C (Wang, Adhikari, & Hung, 2017). Nevertheless, Warner et al. proved that hexanal was the main oxidation product in roasted peanuts after 68 days stored in an air convection oven at 65 °C (Warner et al., 1996). It should be stressed that

Table 3
Volatile compounds 10^8 (Area/peanut mass (g)) identified in peanut samples under accelerated oxidative treatment of UV light and thermal treatment (70 °C).

Retention time		UV			Oven	
	Compounds	0 days	3 days	7 days	0 days	2 months
Aldehydes						
9.051	Hexanal	0.536 ± 0.016 ^{a}	3.3 ± 0.2 ^{b}	4.7 ± 0.2 ^{c}	0.536 ± 0.016 ^{a}	2.31 ± 0.05 ^{b}
11.152	Heptanal	0.101 ± 0.004 ^{a}	0.51 ± 0.04 ^{b}	0.71 ± 0.02 ^{c}	0.101 ± 0.004 ^{a}	1.06 ± 0.03 ^{b}
13.066	Octanal	0.21 ± 0.02 ^{a}	1.676 ± 0.12 ^{b}	2.18 ± 0.07 ^{c}	0.21 ± 0.02 ^{a}	3.98 ± 0.09 ^{b}
14.820	**Nonanal**	0.35 ± 0.02 ^{a}	4.3 ± 0.3 ^{b}	6.1 ± 0.2 ^{c}	0.35 ± 0.02 ^{a}	8.6 ± 0.3 ^{b}
16.430	Decanal	nd	0.219 ± 0.018 ^{a}	0.403 ± 0.013 ^{b}	nd	nd
Acids						
13.357	Hexanoic acid	0.24 ± 0.03 ^{a}	4.5 ± 0.4 ^{b}	9.4 ± 0.3 ^{c}	0.24 ± 0.03 ^{a}	12.8 ± 0.3 ^{c}
16.353	**Octanoic acid**	0.086 ± 0.003 ^{a}	0.33 ± 0.03 ^{ab}	1.00 ± 0.03 ^{b}	0.086 ± 0.003 ^{a}	15.9 ± 0.6 ^{c}
17.777	**Nonanoic acid**	0.1 ± 0.02 ^{a}	0.262 ± 0.019 ^{a}	0.81 ± 0.08 ^{a}	0.1 ± 0.02 ^{a}	13.54 ± 0.12 ^{c}
Alquenes						
14.318	(E)-2-Octenal	nd	2.406 ± 0.17 ^{a}	3.21 ± 0.1 ^{b}	nd	3.00 ± 0.08 ^{d}
15.990	(E)-2-Nonenal	nd	0.58 ± 0.05 ^{a}	0.88 ± 0.03 ^{b}	nd	1.92 ± 0.07 ^{d}
17.541	2-Decenal, (E)-	nd	0.65 ± 0.03 ^{a}	1.4 ± 0.06 ^{b}	nd	nd
13.650	2,4-Heptadienal, (E,E)-	0.033 ± 0.004 ^{a}	0.09 ± 0.006 ^{b}	0.102 ± 0.008 ^{c}	0.033 ± 0.004 ^{a}	nd
18.482	**2,4-Decadienal, (E,E)-**	nd	0.246 ± 0.018 ^{a}	0.401 ± 0.009 ^{b}	nd	0.407 ± 0.012 ^{b}
Furanes						
12.437	2-Pentyl-furan	0.085 ± 0.006 ^{a}	0.32 ± 0.03 ^{b}	0.429 ± 0.011 ^{c}	0.085 ± 0.006 ^{a}	0.81 ± 0.03 ^{e}
18.330	5-Butyldihydro-2(3H)-furanone	nd	nd	0.184 ± 0.006 ^{a}	nd	1.16 ± 0.09 ^{c}
19.778	Dihydro-5-pentyl-2(3H)-furanone	nd	nd	0.134 ± 0.008 ^{a}	nd	nd
Pyrazines						
11.260	**2,5-Dimethyl pyrazine**	0.729 ± 0.039 ^{a}	0.404 ± 0.27 ^{b}	0.393 ± 0.021 ^{b}	0.729 ± 0.039 ^{a}	nd
11.471	2,3-Dimethyl-pyrazine	0.0334 ± 0.0003 ^{a}	nd	nd	0.0334 ± 0.0003 ^{a}	nd
12.876	2-Ethyl-6-methyl-pyrazine	0.0769 ± 0.003 ^{a}	nd	nd	0.0769 ± 0.003 ^{a}	nd
14.220	**3-Ethyl-2,5-dimethyl-pyrazine**	0.155 ± 0.008 ^{a}	0.186 ± 0.011 ^{b}	0.19 ± 0.009 ^{c}	0.155 ± 0.008 ^{a}	nd
15.491	3,5-Diethyl-2-methyl-pyrazine	0.033 ± 0.003 ^{a}	nd	nd	0.033 ± 0.003 ^{a}	nd

* Compounds were identified based on NIST library.
** Results expressed as mean ± standard deviation of four replicates ($\alpha = 0.05$).
*** Different superscripts for each compound within the same column and storage treatment indicate statistically significant different values ($p < 0.05$).

hexanal is formed through the autoxidation of linoleic acid, which is very susceptible to oxidation and its content plays a decisive role in the shelf-life of peanuts (Wang et al., 2017; Yin et al., 2022). Hence, differences in the level of hexanal produced during oxidation could be attributed to differences in the initial fatty acid composition of the sample (López et al., 2022). Fried nuts, which linoleic acid content is higher than in roasted samples, promote hexanal *versus* nonanal production (García, Sanahuja, et al., 2021).

It is interesting to note that after 7 days of UV light irradiation, the second most abundant aldehyde was hexanal (33%) while octanal was the second most abundant aldehyde in the high-temperature treatment. Octanal, is another oxidation product of oleic acid (Yin et al., 2022). This indicates different oxidation results when comparing the two treatments. High temperatures induced the formation of oleic acid-derived aldehydes while UV radiation produced a mixture of oleic and linoleic acids derived aldehydes. It could be an indication that UV and heat oxidation proceeded through different oxidation mechanisms (Van Dyck, 2010). The different mixture of aldehydes can induce changes in the sensory evaluation of oxidised samples.

In addition, further oxidation of those aliphatic aldehydes generated their corresponding organic acids (Morales, Rios, & Aparicio, 1997). Hexanoic acid has been detected as the major acid in different oxidation studies (J. Lee, Xiao, Zhang, Ebeler, & Mitchell, 2014), which is in agreement with the results of the UV treatment. However, when high temperature was used the production of hexanoic, octanoic and nonanoic acids was similar, indicating a different oxidation mechanism.

Pyrazines are responsible for peanut aroma because of their nutty odour. They are caused by Maillard reactions between simple sugars and amino acids (López et al., 2022). In roasted peanuts, 2,5-dimethylpyrazine is the pyrazine found in highest amount (Cuicui & Lixia, 2018). It is well-known that pyrazine concentrations decreased during storage (López et al., 2022). E.G. Abegaz et al. demonstrated 2,5-dimethylpyrazine content decreased 35% after 52 weeks at 21 °C (Abegaz et al., 2004). Lipid radicals and peroxides might contribute to the degradation of these heterocyclic compounds (Bett & Boylston, 1992). Nevertheless, Warner et al. have affirmed that the amount of pyrazines (2-methyl pyrazine, 2,6-dimethyl pyrazine, and 2,3,5-trimethyl pyrazine) showed no distinct trends with storage when peanuts were stored in an air convection oven for 68 days at 65 °C (Warner et al., 1996).

In a preliminary experimental test, peanuts samples were stored at ambient temperature in an open tray and all the pyrazines, except the 2,5-dimethylpyrazine, were not detected after a month. In relation to the compound 2,5-dimethylpyrazine, its content decreased by around 32% in the same preliminary experimental test. Due to the high volatility of these compounds, our hypothesis is when peanuts are in open containers pyrazines volatilize rather quickly. Initially, samples (reference) were characterized by the presence of five pyrazines, the most important were 2,5-dimethyl pyrazine and 3-ethyl-2,5-dimethyl pyrazine. At the end of the oxidation procedures, the signal of the pyrazines decreased. After the high-temperature treatment, all five original compounds were not detected, which is related to the oxidation process and the duration of the experiment. During the UV treatment the two main pyrazines of the reference were retained while the other three were not detected. The shorter duration of this treatment limited the loss of these compounds.

Regarding the groups of alkenes and furanes, in general terms, the oxidation treatments led to an increase in the production of these compounds, although 2-decenal, 2,4-heptadienal, and dihydro-5-pentyl-2(3H)-furanone were not detected in samples kept at high temperatures.

3.3. Instrumental colour evaluation

The instrumental colour evaluation showed significant differences in a* and b* parameters (Table 4). The a* value decreased with both oxidation treatments, so samples got greener. The b* value increased after samples being oxidated. The increase in the b* value indicates that the colour of peanut kernels turned to yellow rather than blue (Bagheri et al., 2019). The temperature could oxidise polyphenols as well as increase Maillard products formation owing to the non-enzymatic browning reactions (Saklar, Katnas, & Ungan, 2001). The increase in the b* value during roasting was reported for peanuts, pistachio nuts, and sesame seeds (Bagheri et al., 2019; T. Kahyaoglu & Kaya, 2006; Talip Kahyaoglu, 2008). The influence of a* and b* values in total colour compared to fresh peanuts was shown in the ΔE value. Increasing UV-light irradiation increased the ΔE value from 0.89 ± 0.08 to 1.14 ± 0.07. Browning developed was greater after thermal treatment due to ΔE was higher (2.56 ± 0.05).

3.4. Sensory analysis

The mean values for the sensory attribute ratings from the sensory analysis are presented in Table 4. The attributes with intensity ratings that changed significantly (α = 0.05) are in bold. As can be seen, both treatments lead to significant changes in most of the sensory attributes of peanuts.

Peanut aroma and peanut odour perception scores were lower in the oxidised samples. Peanuts' aroma and odour could be related to a very studied attribute called roasted peanut flavour. After 7 days of UV light irradiation, peanut aroma and peanut odour decreased by 43% and 37%, respectively. An oxidation test in peanuts at 21 °C for 52 weeks reported a reduction of 42% in roasted peanut flavour attribute (Abegaz et al., 2004). Losses in peanut aroma and odour scores for samples after 2 months at 70 °C were slightly higher (around 65% and 68%, respectively). However, K.J.H. Warner et al. reported only a roasted peanut flavour reduction of 33% after 68 days at 65 °C (Warner et al., 1996). This fact could be explained because of the different sample origins and different oxidation levels.

Table 4
Results of sensory evaluation and instrumental colour evaluation under UV light and oven treatment at different times.

Sample Treatment	Exposure time	Sensory evaluation							Instrumental colour evaluation			
		Aroma peanut	Colour	Crunchiness	**Peanut odour**	**Permanency**	**Rancity**	**Sweetness**	L[a]	a[a]	b[a]	ΔE
UV	0 days	**6.9 ± 0.7[a]**	4.3 ± 0.2[a]	6.4 ± 0.4[a]	**7.9 ± 0.4[a]**	**4.3 ± 0.5[a]**	**2.9 ± 0.9[a]**	**3.7 ± 0.5[a]**	60.51 ± 0.1[a]	2.72 ± 0.04[a]	16.05 ± 0.04[a]	–
	3 days	**4.2 ± 0.7[b]**	4.1 ± 0.2[a]	6.1 ± 0.3[a]	**3.6 ± 0.5[b]**	**5.3 ± 0.8[ab]**	**4.3 ± 0.7[b]**	**2.8 ± 0.5[b]**	60.21 ± 0.16[a]	2.00 ± 0.09[b]	15.64 ± 0.04[b]	0.89 ± 0.08[a]
	7 days	**3.9 ± 0.6[b]**	4 ± 0.15[a]	6.0 ± 0.4[a]	**5.0 ± 0.7[c]**	**5.7 ± 0.8[b]**	**5.0 ± 0.9[b]**	**2.7 ± 0.5[b]**	60.86 ± 0.41[a]	1.77 ± 0.05[c]	15.67 ± 0.14[b]	1.14 ± 0.07[b]
Oven	0 days	**6.9 ± 0.7[a]**	**4.3 ± 0.2[a]**	**6.4 ± 0.4[a]**	**7.9 ± 0.4[a]**	**4.3 ± 0.5[a]**	**2.9 ± 0.9[a]**	**3.7 ± 0.5[a]**	60.51 ± 0.1[a]	2.72 ± 0.04[a]	16.05 ± 0.04[a]	–
	2 months	**2.4 ± 0.4[b]**	**5.2 ± 0.7[b]**	**4.2 ± 0.6[b]**	**2.5 ± 0.5[b]**	**7.5 ± 0.5[b]**	**8.0 ± 0.5[b]**	**1.8 ± 0.3[b]**	60.0 ± 0.5[a]	1.70 ± 0.17[b]	18.3 ± 0.3[b]	2.56 ± 0.05 [a]

** Different superscripts for each compound within the same column and storage treatment indicate statistically significant different values ($p < 0.05$).
[a] Results expressed as the mean ± standard deviation of three replicates (α = 0.05).

It is important to note that scores of these attributes were lower for the samples treated at high temperatures than for the UV. This could be related to the duration of the treatment as well as the different volatile profiles produced. Reductions in roasted peanut aroma and odour scores correlated highly with pyrazines (specifically, 2,5-dimethylpyrazine) (Baker et al., 2003). It has been suggested that the loss of roasted peanut flavour in peanuts during storage is a result of the masking of pyrazines by off-flavour compounds derived from lipid oxidation (Abegaz et al., 2004; Mugendi et al., 1998). For example, hexanal and 2-octenal produce beany flavour while hexanoic acid possesses penetrating goaty, sweaty, and cheesy aromas (Franklin et al., 2018; Warner et al., 1996). The aroma of octanoic acid is described as waxy, rancid, and oily (Franklin et al., 2017).

As it has been explained before, aldehydes and organic acids content increased with oxidation level, like rancity attribute. Initially, rancity attribute of studied samples was evaluated with a 2.9 score and it increased to 5 and 8 after 7 days of UV irradiation and 2 months of oven treatment, respectively. The obtained results are in agreement with previously cited studies, in which rancity score increased from 0.5 to 5.6 (Warner et al., 1996). It is interesting to notice rancity score of samples after 7 days of UV light irradiation show that there can be significant flavour fade in peanuts in the absence of appreciable oxidation (without changes in fatty acid content) (Mugendi et al., 1998).

Initially, rancity attribute of studied samples was evaluated with a 2.9 score and it was increasing to 5 and 8 after 7 days of UV light and 2 months of oven treatment, respectively. The obtained results are in agreement with previously cited studies, in which rancity score increased from 0.5 to 5.6 (Warner et al., 1996). It is interesting to notice that rancity scores of samples after UV irradiation indicated that the oxidation process was going on, although there were no changes in the fatty acid profile of the samples yet. Permanency scores followed a similar trend that rancity.

As regards the sweetness scores, they decreased during the treatments as was reported in other studies where the authors highlighted that as the oxidation process progresses, the rancity increases while sweetness decreases (Abegaz et al., 2004; S. Y. Lee et al., 2002; Mugendi et al., 1998).

Crunchiness was just a bit affected by temperatures lower than 60 °C (S. Y. Lee et al., 2002). However, at 70 °C the value of this parameter decreased 34% in 2 months. UV light did not affect crunchiness parameter. Similar results were found for colour because browning and caramelization reactions take place during oxidation (Bagheri et al., 2019). Thus, only thermal treatment had a significant impact on colour attribute determined by panellists, although instrumental colour measurements detected changes due to the UV treatment as well.

It is remarkable samples at day 0 were identified as edibles by all panellists. By contrast, all panellists agreed that, after 2 months in the oven, peanuts were not edibles. Samples oxidised by UV for 3 days and 7 days were edible for 52% and 30% of panellists, respectively. The decrease in overall acceptance and flavour in all samples is probably related to the increase in oxidised flavour intensity and the decrease in roasted peanut flavour intensity (Grosso & Resurreccion, 2002). N.R. Grosso et al. determined that the overall acceptance in roasted peanuts after being stored at 40 °C was shown around 5 (neither like nor dislike) on the hedonic scale after 66 days of exposure (Grosso & Resurreccion, 2002).

4. Conclusions

In this study, the use of UV irradiation as an accelerated oxidation procedure for peanuts has been investigated and compared with conventional accelerated methodology (thermal oxidation). The irradiation of the samples during 3 and 7 days reduced the levels of tocopherols, especially after 7 days, while did not affect the levels of the main fatty acids present in the peanuts. The volatile profile also confirmed the oxidation of the samples treated with UV due to the pyrazine's losses and the production of hexanal, nonanal and decanal. Changes in colour instrumental determinations were also detected.

It is interesting to note that in the case of volatiles, the profile of compounds determined was different in each oxidation treatment, which is indicative that the oxidation mechanisms may be different. This can be a consequence of the shorter duration of the UV treatment and the fact that it is carried out at lower temperatures.

The panellists determined that the UV treatment changed the sensory profile of the samples in the variables peanut aroma, peanut odour, rancidity, permanency, and sweetness. However, the high-temperature treatment also changed the perception of all colour and crunchiness, which are more influenced by the temperature.

In conclusion, UV irradiation is proposed as a peanut's and other food products accelerated oxidation methodology. The main advantage of this methodology is that the time needed to develop the studies can be significantly reduced.

In future work it will be necessary to better optimize the irradiation conditions so that the UV oxidation process proceeds through mechanisms more similar to those that take place when high temperatures are used. In this way, accelerated UV oxidation could replace the use of high temperatures for food oxidative stability studies.

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CRediT authorship contribution statement

Adriana Juan-Polo: Conceptualization, Methodology, Formal analysis, Data curation, Writing – original draft, Writing – review & editing. **Ana Beltrán Sanahuja:** Conceptualization, Methodology, Formal analysis, Data curation, Writing – original draft, Funding acquisition, Writing – review & editing, Supervision. **María Monedero Prieto:** Funding acquisition, Writing – review & editing, All authors have read and agreed to the published version of the manuscript. **Carmen Sánchez Reig:** Funding acquisition, Writing – review & editing. **Arantzazu Valdés García:** Conceptualization, Methodology, Data curation, Writing – review & editing, Supervision. **Salvador E. Maestre Pérez:** Conceptualization, Methodology, Formal analysis, Data curation, Funding acquisition, Writing – review & editing, Supervision.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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