

# Thyme and basil essential oils included in edible coatings as a natural preserving method of oilseed kernels

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## Abstract

**BACKGROUND:** Sunflower seeds are susceptible to developing rancidity and off-flavours through lipid oxidation. Edible coatings and essential oils have proven antioxidant properties in different food products. The purpose of this study was to evaluate the combined effect of using an edible coating and thyme and basil essential oils to preserve the chemical and sensory quality parameters of roasted sunflower seeds during storage.

**RESULTS:** 50% DPPH inhibitory concentration (IC<sub>50</sub>) values of 0.278 and 0.0997 µg mL<sup>-1</sup> were observed for thyme and basil, respectively. On storage day 40, peroxide values were 80.68, 70.28, 68.43, 49.31 and 33.87 mEq O<sub>2</sub> kg<sup>-1</sup> in roasted sunflower seeds (RS), roasted sunflower seeds coated with carboxymethyl cellulose (CMC) (RS-CMC), roasted sunflower seeds coated with CMC added with basil (RS-CMC-A), thyme (RS-CMC-T) and butylated hydroxytoluene (RS-CMC-BHT), respectively. RS-CMC-T and RS-CMC-BHT presented the lowest peroxide values, conjugated dienes and *p*-anisidine values during storage. RS-CMC-BHT, RS-CMC-T, and RS-CMC-A showed the lowest oxidized and cardboard flavour intensity ratings. On storage day 40, roasted sunflower flavour intensity ratings were higher in RS-CMC-T and RS-CMC-A.

**CONCLUSIONS:** Thyme and basil essential oils added to the CMC coating improved the sensory stability of this product during storage, but only thyme essential oil increased their chemical stability.

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**Keywords:** thyme; basil; oxidation; antioxidant; stability

## INTRODUCTION

Non-oleic sunflower seeds contain between 800 and 850 g kg<sup>-1</sup> polyunsaturated fatty acids, with 600–650 g kg<sup>-1</sup> linoleic acid. Besides acyl glycerides, the lipid composition of sunflower oil shows several different compounds like sterol and stanol esters, tocopherols, carotenoids and phospholipids, among others, where the tocopherols act as natural antioxidants that contribute to preserve the oil stability.  $\alpha$ -Tocopherol makes an oil resistant to photo-oxidation, while  $\gamma$ -tocopherol provides stability against oxidation. Sunflower oil is rich in  $\alpha$ -tocopherol but poor in  $\gamma$ -tocopherol.<sup>1</sup>

Oxidation reactions occur in sunflower lipids during storage. Aliphatic aldehydes, ketones and alcohols are products of secondary lipid oxidation reactions that are related to rancidity and off-flavours, and also those molecules are related to negative sensory attributes such as oxidized, cardboard and painty flavours. The intensity ratings of these negative attributes increase during storage, as has been observed in numerous food products as a consequence of the deterioration process.<sup>2–4</sup> One way to prevent or reduce lipid oxidation reactions in a food product is to use edible coatings<sup>3–5</sup> or to add antioxidants.<sup>3,6–9</sup>

Edible coatings have an important function in preserving quality and safety in fresh and processed foods during transportation and storage. These coatings are a type of biodegradable active packaging material that can extend shelf-life and improve sensory, safety and functional properties. The biodegradability of edible coatings

based on biopolymers is a characteristic of these compounds that can help the environment by decreasing the pollution effect. For all these advantages, edible coatings have been adopted for the food industry in many products.<sup>10</sup>

Many essential oils are natural antioxidants considered safe because they are obtained from aromatic plants used for culinary ingredients.<sup>7,8,11,12</sup> However, the use of essential oils as antioxidants in food remains limited because of their strong flavour, which makes some products unacceptable for consumers. Several authors have reported changes in the organoleptic properties of food when these oils are used.<sup>7,8</sup> To minimize the amount of essential oil, one option could be its inclusion in edible coatings as vehicles for this compound.<sup>13</sup>

Edible films prepared with carboxymethyl cellulose (CMC), methyl cellulose (MC) or whey protein isolated (WPI) have been demonstrated to delay the oxidation process in roasted peanuts, retarding the increase in lipid oxidation indicators and in the

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intensity ratings of oxidized and cardboard flavours.<sup>4</sup> CMC is the coating that has shown to be more effective against oxidation reactions than MC and WPI.

Roasted sunflower seeds have high susceptibility to rancidity during storage owing to their fatty acid composition. However, there are not many storage studies concerning the shelf-life of this kind of food product. The use of edible coatings to prevent or decrease deterioration reactions has not been studied in sunflower seeds; even the antioxidant effects of essential oil have never been evaluated in this food product. On the other hand, thyme and basil essential oils included in CMC edible coating have never been researched as a procedure for food preservation.

Considering the high susceptibility of sunflower kernels to lipid oxidation, the addition of edible coatings as protective barriers and essential oils as natural antioxidant agents could be an alternative to prolong the shelf-life of this food product. The purpose of this work was to evaluate the combined effect of essential oils (thyme and basil) and edible coating to preserve the chemical and sensory quality in sunflower seeds during storage.

## EXPERIMENTAL

### Materials

Sound and mature seeds of sunflower (*Helianthus annuus*) (2011 crop) were provided by Argensun SA, Victoria, Buenos Aires, Argentina. Damaged sunflower kernels were manually removed before processing.

### Essential oil extraction

The studied species were *Thymus vulgaris* (thyme) and *Ocimum basilicum* (basil). Plant material was provided by Facultad de Ciencias Agropecuarias, Universidad Nacional de Córdoba, Argentina. For harvesting, the plant material was cut 5 cm above the soil surface in August 2010. Plant samples were air dried at room temperature to preserve the phenolic compounds.<sup>14</sup>

Essential oils were extracted by hydrodistillation for 2 h in a Clevenger-type apparatus. The resulting essential oils were dried over anhydrous sodium sulfate and kept in dark-glass flasks in a freezer at  $-18^{\circ}\text{C}$  until use.<sup>15</sup> The essential oils were analysed by gas-liquid chromatography and mass spectrometry (GC-MS).

### Essential oil chemical analysis

A PerkinElmer Clarus 600 GC-MS instrument (Shelton, CT, USA) coupled with an ion trap mass detector was used for determining the chemical composition of the essential oils. A capillary column DB-5 (30 m, 0.25 mm i.d. and 0.25 mm coating thickness) was used for separation of the essential oil components. Helium was used as carrier gas ( $0.9\text{ mL min}^{-1}$ ). The temperature programme in the gas chromatograph oven was  $60^{\circ}\text{C}$  for 5 min,  $5^{\circ}\text{C min}^{-1}$  rate and  $250^{\circ}\text{C}$  final temperature. The ionization process was performed by electron impact at 70 eV. Mass spectral data were acquired in the scan mode in the  $m/z$  range 35–450. The identification of compounds was made by comparing retention times and mass spectra using NIST libraries. The structure identification of the main components was confirmed by co-injection of authentic standards (Sigma, St Louis, MO, USA). Quantification of the peak was made using the mass result reported by the mass detector. The results were expressed as a percentage (w/w).

### Free radical scavenging activity (test DPPH-FRSA) and total phenolic content

The method used by Chen and Ho<sup>16</sup> was followed. The essential oils were added in  $0.05\text{ mmol L}^{-1}$  2,2-diphenyl-1-picrylhydrazyl (DPPH) methanolic solution, and their final concentrations were 5.77, 2.77, 1.39, and  $0.69\text{ }\mu\text{g mL}^{-1}$ . Absorbance of the samples was measured after 30 min on a spectrophotometer (PerkinElmer, Lambda 1A, UV-visible spectrophotometer) at 517 nm. The percent inhibition of the DPPH radical was calculated according to the following equation:

$$\% \text{ DPPH inhibition} = 1 - \left[ \frac{A - A_b}{A_o} \right] \times 100$$

where  $A$  is the absorbance of DPPH solution with the essential oils,  $A_b$  is the absorbance of 60% methanol with the essential oil and  $A_o$  is the absorbance of DPPH solution.

The 50% inhibitory concentration ( $\text{IC}_{50}$ ) was calculated from the curve obtained by plotting inhibition percentage versus final essential oil concentration.<sup>17</sup>

### Edible coating preparation

First, sunflower seeds were dry roasted at  $150^{\circ}\text{C}$  in an oven (model 600, Memert, Schwabach, Germany) for 20 min until reaching a medium roast degree measured as colour Hunter lightness (L)  $50 \pm 1.0$  using a Minolta colorimeter.<sup>4,18</sup> CMC (Parafarm<sup>®</sup> Droguería Saporiti SACIFIA, Buenos Aires, Argentina) was used to prepare the coating solution. CMC was dissolved in sterile deionized water at  $5\text{ g kg}^{-1}$  w/v. Glycerol ( $19\text{ g kg}^{-1}$ ) was used as plasticizer. Roasted sunflower seeds were then coated with  $20\text{ g kg}^{-1}$  CMC solution using a stainless steel coating pan that was kept rotated at 28 rpm for 5 min during the coating process.<sup>19</sup>

Treatments: (a) RS = roasted sunflower seeds without additives as the control sample; (b) RS-CMC = roasted sunflower seeds coated with CMC; (c) RS-CMC-T = roasted sunflower seeds coated with CMC added with thyme essential oil ( $0.1\text{ g essential oil } 100\text{ g}^{-1}$  of product); (d) RS-CMC-A = roasted sunflower seeds coated with CMC added with basil essential oil ( $0.1\text{ g essential oil } 100\text{ g}^{-1}$  of product); and (e) RS-CMC-BHT = roasted sunflower seeds coated with CMC with butylated hydroxytoluene (BHT) added ( $0.02\text{ g BHT } 100\text{ g}^{-1}$  of product). The essential oils and BHT were incorporated into the CMC edible coating solution before coating the seeds.

### Storage

Samples (100 g) were packaged under normal atmosphere in plastic bags (size  $27 \times 28\text{ cm}$ ; Ziploc, Johnson & Son, Buenos Aires, Argentina) and stored at room temperature ( $23^{\circ}\text{C}$ ). Samples were removed from storage at day 0, 10, 20, 30, and 40 for chemical and sensory analyses.

### Chemical analysis

The oil was extracted from roasted sunflower seeds by a cold pressing process using a 20-ton press (HE-DU, Hermes I; Dupraz SRL, Córdoba, Argentina).

Lipid oxidation indicators were analysed in sunflower oil obtained from stored samples:

- Peroxide value (PV) was evaluated following the AOAC method.<sup>20</sup> The result was expressed as milliequivalents of active oxygen per kilogram of oil.

- *p*-Anisidine value (AV) was evaluated following the IUPAC method.<sup>21</sup> Absorbance was measured at 350 nm in a UV–visible diode array spectrophotometer (HP 8452 A, Hewlett-Packard, Palo Alto, CA, USA).
- Conjugated dienes (CD) were evaluated following the COI method.<sup>22</sup> The result was expressed as the sample extinction coefficient *E* (1%, 1 cm).

### Sensory descriptive analysis

Ten panellists (six women and four men) were trained in eight sessions of 3 h each.<sup>23</sup> For descriptive analysis, a 'hybrid' method confirmed by Spectrum TM (Sensory Spectrum, Inc., Chatham, NJ, USA) and Quantitative Descriptive Analysis (Tragon Corp., Redwood City, CA, USA) was used.<sup>3,8</sup> The attribute definitions and intensity ratings of the warm-up and references are shown in Table 1.<sup>23,24</sup> A 150 mm unstructured linear scale was used for sample evaluation. Roasted sunflower seed samples (10 g) were packaged in plastic cups with lids. The samples were identified by a three-digit number code. For testing samples, a completely randomized block design was used. Sample evaluation was performed in booths under fluorescent light at room temperature.

### Statistical analysis

For the present experiment three different lots of sunflower seeds were used. Three parallel repetitions of the coating process were carried out. The data were analysed using InfoStat software, version 2010p. Means and standard deviations were calculated. Analysis of variance and Duncan tests ( $\alpha = 0.05$ ) were used to detect significant differences between treatments. Correlations between dependent variables were estimated using Pearson coefficient. Linear equations were used for regression between storage time and dependent variables.

## RESULTS AND DISCUSSION

### Essential oil analysis

The major compounds of thyme essential oil were thymol (429.3 g kg<sup>-1</sup>), *o*-cymene (267.3 g kg<sup>-1</sup>),  $\gamma$ -terpinen (40.1 g kg<sup>-1</sup>), carvacrol (36.5 g kg<sup>-1</sup>),  $\beta$ -linalool (29.7 g kg<sup>-1</sup>), eucalyptol (23.1 g kg<sup>-1</sup>), caryophyllene (17.8 g kg<sup>-1</sup>) and caryophyllene oxide (11.6 g kg<sup>-1</sup>). Structurally, phenolic compounds comprise an aromatic ring, bearing one or more hydroxyl substituents. The potential antioxidant activity is in relation to this chemistry of phenolic compounds. Cymene has a single aromatic group and thymol has one aromatic ring and one —OH group. For that reason these two components have great potential as a natural antioxidant.<sup>25</sup> The basil essential oil consisted mainly of oxygenated monoterpene  $\beta$ -linalool (477.7 g kg<sup>-1</sup>). Others major compounds were toluene (86.9 g kg<sup>-1</sup>), cinnamic acid, methyl ester (63.4 g kg<sup>-1</sup>), eucalyptol (59.9 g kg<sup>-1</sup>),  $\tau$ -cadinol (43.4 g kg<sup>-1</sup>), estragole (35.4 g kg<sup>-1</sup>), 2-methoxy-3-(2-propenyl)-phenol (31.9 g kg<sup>-1</sup>),  $\alpha$ -bergamotene (22.1 g kg<sup>-1</sup>), germacrene D (21.7 g kg<sup>-1</sup>),  $\gamma$ -cadinene (18.4 g kg<sup>-1</sup>),  $\alpha$ -bulnesene (13.6 g kg<sup>-1</sup>) and methyl cinnamate (13.3 g kg<sup>-1</sup>). Terpenoids are a kind of terpene modified by enzymatic action that can show antioxidant activity due to the ability of hydrogen transfer, electron transfer or quenching singlet oxygen.<sup>26,27</sup> Thymol and linalool are terpenoids found in essential oils of some plants. The chemical composition of thyme and basil essential oils indicates that these two essential oils could have antioxidant activity.

### Free radical scavenging activity on DPPH and total phenolic content

The total phenolic content and DPPH IC<sub>50</sub> are presented in Table 2. Thyme and basil essential oils showed significant differences in these chemical parameters. Basil essential oil exhibited a higher value for total phenolic content but a lower IC<sub>50</sub> value than thyme essential oil. Many essential oils obtained from different aromatics plants have demonstrated antioxidant properties. Hussain *et al.*<sup>28</sup> observed that basil essential oil reduces radical DPPH, but its essential oil extracted in winter and spring crops shows greater radical scavenging activity than crops harvested in autumn and summer. Dambolena *et al.*<sup>11</sup> reported that the essential oil of *Ocimum gratissimum* had a high content of eugenol with remarkable antiradical effects in the DPPH assay. In this same research, the authors also detected that the essential oil of *O. basilicum*, which is rich in linalool, did not have antiradical activity. Viuda-Martos *et al.*<sup>12</sup> observed that thyme essential oil has the highest content of total phenols (913.17 mg GAE (gallic acid equivalents) L<sup>-1</sup>) and TBARS (thiobarbituric acid reactive substances) values (80.76), but black cumin showed the highest percent inhibition of DPPH radicals (95.89%) and the highest ferric reducing antioxidant capacity (FRAC) value (3.33 mmol L<sup>-1</sup> Trolox). The essential oils of basil, thyme and oregano, rich in linalool, thymol and carvacrol, respectively, are proposed as preservatives in foods because of their high potential antioxidant activities.<sup>29</sup>

### Storage study

#### Chemical analysis

Peroxide value (PV), *p*-anisidine value (AV), and conjugated diene content (CD) in roasted sunflower samples (RS, RS-CMC, RS-CMC-T, RS-CMC-A and RS-CMC-BHT) stored at 23 °C are presented in Fig. 1. Sunflower samples showed increases in PV and CD values during storage. RS had higher PV and CD and exhibited significant differences during storage after day 10 in comparison with coated roasted sunflower seeds (RS-CMC, RS-CMC-T, RS-CMC-A and RS-CMC-BHT). RS-CMC-T and RS-CMC-BHT presented the lowest PV and CD during storage. RS also exhibited higher AV and showed significant differences during storage after day 20 with respect to coated roasted sunflower samples (RS-CMC, RS-CMC-T, RS-CMC-A and RS-CMC-BHT). RS-CMC-BHT presented the lowest AV during storage, followed by RS-CMC-T. The results of the oxidation chemical indicators during storage evidence that the CMC edible coating helps to preserve the roasted sunflower seeds against lipid oxidation process but, also, the addition of these essential oils (especially thyme essential oil) as natural antioxidant improves this protection effect against oxidation reactions.

The bioactive nature of different essential oils from aromatic plants has been reported in other research studies in which the essential oil was applied directly to the food or to different edible coatings as vehicles for these natural compounds. Jouki *et al.*<sup>30</sup> investigated the quality changes of rainbow trout fillet wrapped with quince seed mucilage (QSM) film incorporated with 0–20 g kg<sup>-1</sup> (v/v) thyme or oregano essential oil, as natural preservatives, during storage for 18 days at 4 °C. These authors determined that the PV of rainbow trout fillets wrapped with QSM film (QSMF) was significantly lower than that of the unwrapped fillets (control sample), indicating that the QSM films were effective in reducing lipid oxidation. These results also indicated that QSM-based films enriched with oregano or thyme essential oils were effective in reducing PV in rainbow trout fillets stored at 4 ± 1 °C. The highest concentration of oregano or thyme essential oils (20 g kg<sup>-1</sup>) had the most obvious effect in

**Table 1.** Definitions of attributes, standard references and warm-up used for descriptive analysis, and the results of roasted sunflower samples obtained at storage day zero.

Attribute <sup>a</sup>	Reference	Reference intensity <sup>b</sup>	Warm-up intensity <sup>b, c</sup>	Sample intensity ratings <sup>b</sup>
<i>Appearance</i>				
Brown colour	Cardboard (lightness value, $L = 47 \pm 1$ )	65	45	44.73 $\pm$ 0.43
Roughness	Corn flakes <sup>d</sup>	110	18	17.73 $\pm$ 0.43
Glossiness	White bean <sup>e</sup>	35	17	17.20 $\pm$ 0.36
<i>Aromatics<sup>i</sup></i>				
Roasted sunflower	Dry roasted sunflower <sup>f</sup>	44	60	59.80 $\pm$ 0.45a (RS) <sup>i</sup> 57.60 $\pm$ 2.51b (RS-CMC) 57.00 $\pm$ 4.47b (RS-CMC-BHT) 55.60 $\pm$ 5.18c (RS-CMC-T) 55.40 $\pm$ 5.08c (RS-CMC-A)
Oxidized	Rancid sunflower	100	4	4.40 $\pm$ 0.18
Cardboard	Moist cardboard	30	9	8.83 $\pm$ 0.23
Essential oil flavour	0.1% w/w thyme essential oil in sunflower kernels	45		45.00 $\pm$ 5.77b (RS-CMC-T)
	0.1% w/w basil essential oil in sunflower kernels	51		51.00 $\pm$ 2.24b (RS-CMC-A)
<i>Tastes</i>				
Sweetness	20 g kg <sup>-1</sup> sucrose solution	20	17	16.13 $\pm$ 0.33
	50 g kg <sup>-1</sup> sucrose solution	50		
	100 g kg <sup>-1</sup> sucrose solution	100		
Saltiness	2 g kg <sup>-1</sup> NaCl solution	25	8	7.63 $\pm$ 0.51
	3.5 g kg <sup>-1</sup> NaCl solution	50		
	5 g kg <sup>-1</sup> NaCl solution	85		
Sourness	0.5 g kg <sup>-1</sup> citric acid solution	20	3	3.60 $\pm$ 0.25
	0.8 g kg <sup>-1</sup> citric acid solution	50		
	1.5 g kg <sup>-1</sup> citric acid solution	100		
Bitterness	0.5 g kg <sup>-1</sup> caffeine solution	20	15	15.97 $\pm$ 0.64
	0.8 g kg <sup>-1</sup> caffeine solution	50		
	1.5 g kg <sup>-1</sup> caffeine solution	100		
<i>Feeling factors</i>				
Astringent	8 g coffee <sup>g</sup> in 250 mL distilled water	60	30	30.07 $\pm$ 0.48
<i>Texture<sup>j</sup></i>				
Hardness	Almonds <sup>h</sup>	70	40	35.40 $\pm$ 0.89a (RS) <sup>i</sup>
				33.00 $\pm$ 2.83b (RS-CMC)
				33.75 $\pm$ 2.63b (RS-CMC-BHT)
				33.60 $\pm$ 3.58b (RS-CMC-T)
				32.75 $\pm$ 3.40b (RS-CMC-A)
Crunchiness	Corn flakes <sup>d</sup>	110	30	32.20 $\pm$ 3.03a (RS)
				28.00 $\pm$ 2.74b (RS-CMC)
				28.40 $\pm$ 3.21b (RS-CMC-BHT)
				28.00 $\pm$ 5.43b (RS-CMC-T)
				28.00 $\pm$ 2.74b (RS-CMC-A)

<sup>a</sup> Attribute definitions based on Meilgaard *et al.*<sup>23</sup>
<sup>b</sup> Mean values of all samples. Intensity ratings are based on 150 mm unstructured line scales.

<sup>c</sup> Medium (lightness value,  $L = 50 \pm 1$ ) roasted sunflower seeds.

<sup>d</sup> Corn flakes, Granix, Buenos Aires, Argentina.

<sup>e</sup> White bean, Grandiet, Córdoba, Argentina.

<sup>f</sup> Dry roasted sunflower, Argensun SA, Victoria, Buenos Aires, Argentina.

<sup>g</sup> Coffee, Nescafé® Clásico, Nestlé Argentina SA, Buenos Aires, Argentina.

<sup>h</sup> Almonds, Grandiet, Córdoba, Argentina.

<sup>i</sup> RS, roasted sunflower seeds without additives as the control sample; RS-CMC, roasted sunflowers coated with carboxymethyl cellulose; RS-CMC-BHT, roasted sunflowers coated with CMC added with BHT; RS-CMC-T, roasted sunflowers coated with CMC added with thyme essential oil; RS-CMC-A, roasted sunflowers coated with CMC added with basil essential oil.

<sup>j</sup> Different letters after the mean values indicate that there are significant differences ( $\alpha = 0.05$ ) between samples.

**Table 2.** Total phenolic content (expressed as mg GAE mL<sup>-1</sup>) and DPPH IC<sub>50</sub> from thyme and basil essential oils

Essential oil	Phenol content (mg mL <sup>-1</sup> ) <sup>a</sup>	DPPH IC <sub>50</sub> (µg mL) <sup>a</sup>
Thyme	5.29 ± 0.21a	0.2780a
Basil	7.46 ± 0.38b	0.0997b

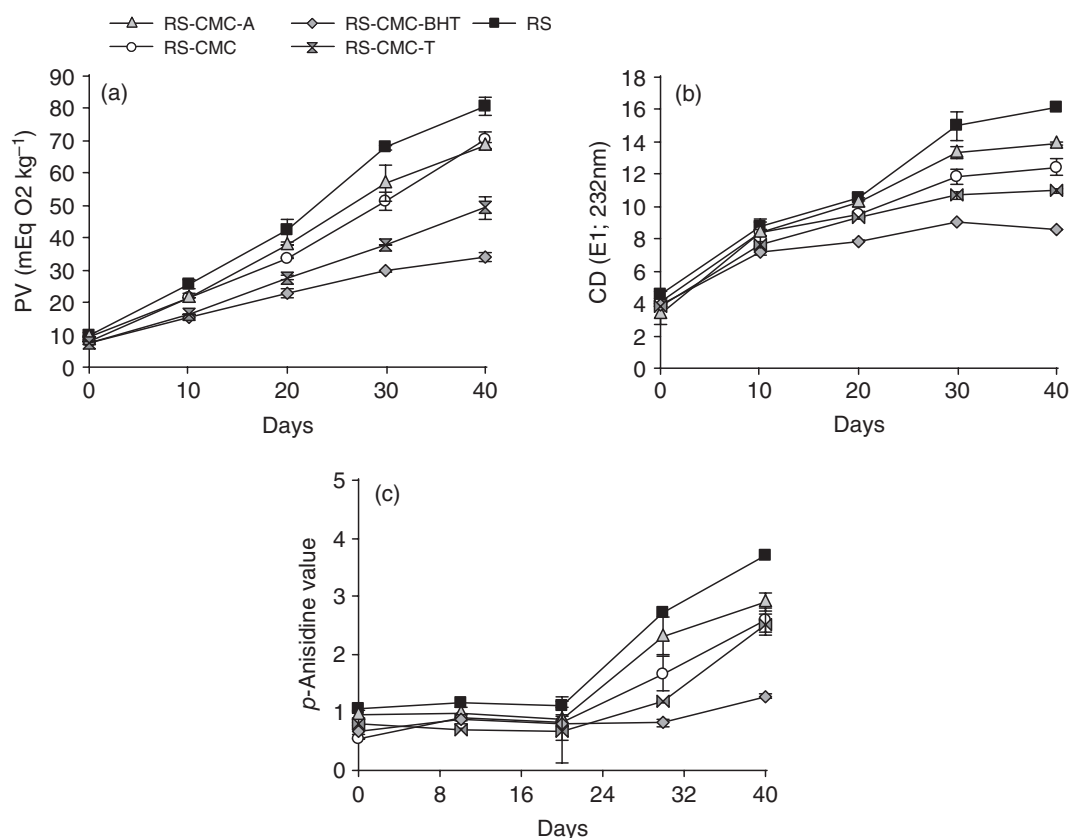
<sup>a</sup> Values with different letters within each column are significantly different (Duncan's test,  $\alpha = 0.05$ ;  $n = 3$ ).

slowing the primary peroxidation process compared with others ( $P < 0.05$ ). Viuda-Martos *et al.*<sup>31</sup> studied the effect of adding orange dietary fibre (10 g kg<sup>-1</sup>), rosemary essential oil (0.2 g kg<sup>-1</sup>) or thyme essential oil (0.2 g kg<sup>-1</sup>), and the effect of storage conditions on the quality characteristics and shelf-life of mortadella, which is a bologna-type sausage. Those authors observed that the treatments resulted in lowered lipid oxidation. Orange dietary fibre and spice essential oils could find a use in the food industry to improve the shelf-life of meat products. Cardoso-Ugarte *et al.*<sup>32</sup> explored the potential antioxidant power of basil essential oil under frying conditions. Two concentrations (200 or 500 ppm) were added to palm olein (PO) to evaluate their effect on fat oxidation/degradation during repeated frying of French fries at 180 °C. Those authors detected higher oxidative stability for PO with basil essential oil at 200 ppm. Both concentrations showed lower *p*-anisidine values than PO without basil essential oil after 5 storage days. The addition of 500 ppm basil essential oil decreased total polar compounds and free fatty acid content in French

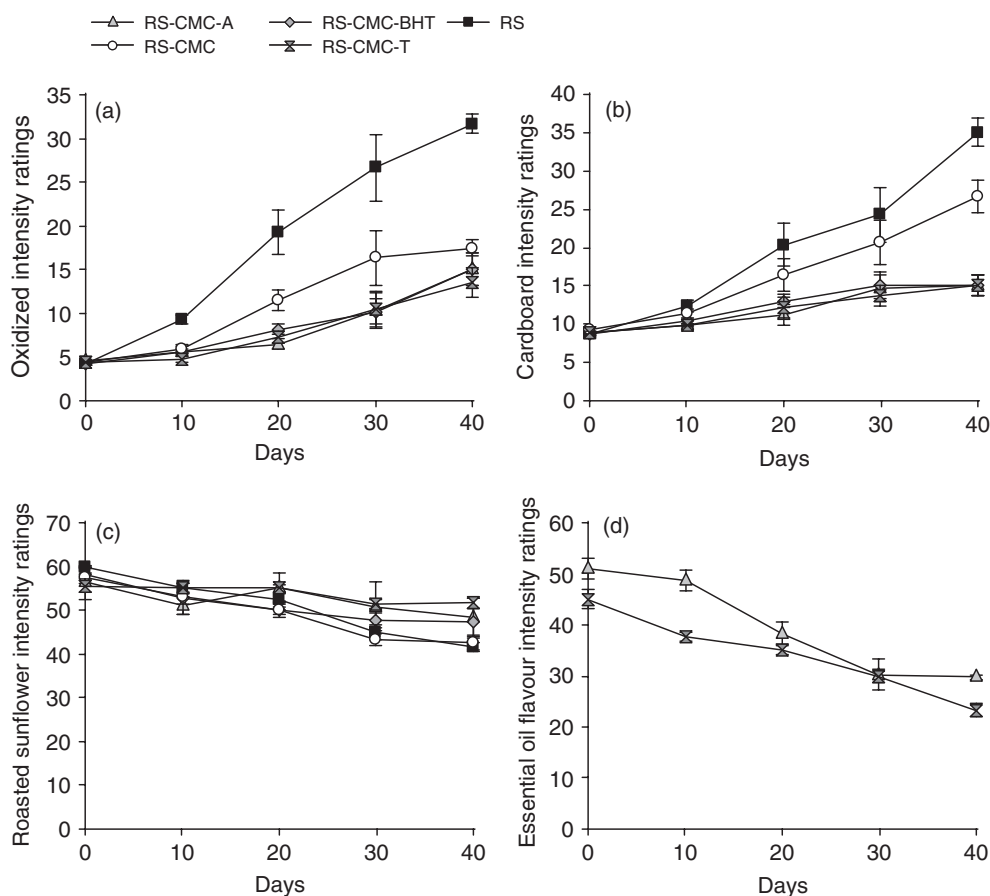
fries. Thus the addition of basil essential oil improves the performance of PO during repeated frying of French fries. Cichoski *et al.*<sup>33</sup> evaluated different concentrations of basil essential oil in relation to their antioxidant activity. Concentrations of 0.19, 0.38 and 0.75 mg g<sup>-1</sup> were tested for antioxidant activity in the internal part of Italian type salami during processing and after 30 days of storage, in terms of lipid and protein oxidation. Basil essential oil presents an antioxidant activity measured as an IC<sub>50</sub> of 12 mg mL<sup>-1</sup>. In the internal part of Italian type salami, the commercial antioxidant (control) and the formulation containing 0.75 mg g<sup>-1</sup> basil essential oil presents antioxidant activity in relation to lipids, during processing and storage. Riveros *et al.*<sup>4</sup> reported that PV, AV and CD showed less increase in coated peanuts with CMC, MC and whey protein with respect to the control sample, the samples with CMC being more stable.

#### Sensory analysis

Mean intensity ratings of sensory attributes from RS and coated roasted sunflower (RS-CMC, RS-CMC-BHT, RS-CMC-T and RS-CMC-A) obtained from descriptive analysis at day zero (fresh product) are presented in Table 1. Roasted sunflower flavour, essential oil flavour (thyme and basil), hardness and crunchiness were the attributes that showed significant differences ( $\alpha = 0.05$ ) in intensity ratings among samples. RS showed higher intensity ratings in hardness and crunchiness (35.4 ± 0.89 and 32.2 ± 3.03, respectively) in comparison with coated roasted sunflower samples. The roasted sunflower intensity rating was greater in RS (59.80 ± 0.45). The lowest rating of this attribute was observed in RS-CMC-T (55.60 ± 5.18) and RS-CMC-A (55.40 ± 5.08). This



**Figure 1.** Means and standard deviations ( $n = 3$ ) of (a) peroxide value (PV), (b) conjugated dienes (CD) and (c) *p*-anisidine value (AV) measured in roasted sunflower (RS); roasted sunflowers coated with CMC (RS-CMC); roasted sunflowers coated with CMC added with BHT (RS-CMC-BHT); roasted sunflowers coated with CMC added with thyme (RS-CMC-T) or basil (RS-CMC-A) essential oil during storage at 23 °C.



**Figure 2.** Mean and standard deviation ( $n = 3$ ) of the intensity rating of sensory attributes: (a) oxidized, (b) cardboard (c) roasted sunflower and (d) essential oil flavours evaluated in roasted sunflower (RS); roasted sunflowers coated with CMC (RS-CMC); roasted sunflowers coated with CMC added with BHT (RS-CMC-BHT); roasted sunflowers coated with CMC added with thyme (RS-CMC-T) or basil (RS-CMC-A) essential oils during storage at 23 °C.

observed result indicates that the addition of the CMC edible coating affects the intensity of texture attributes like hardness and crunchiness and the characteristic roasted flavour in the finished product. These sensory changes could affect the acceptability of this product by consumers.

The essential oil flavours (thyme and basil) were only present in RS-CMC-T and RS-CMC-A, respectively. The addition of essential oils brought about a new sensory attribute in the product, which was named essential oil flavour. This attribute is detected by the sensory panel and will probably be detected by consumers, affecting the acceptability of roasted sunflower seeds.

Changes in the intensity ratings of the sensory attributes during storage at 23 °C in RS, RS-CMC, RS-CMC-BHT, RS-CMC-T and RS-CMC-A samples are presented in Fig. 2. Roasted sunflower flavour, essential oil flavour and those attributes related to lipid oxidation, such as oxidized and cardboard flavours,<sup>24,34</sup> were the only attributes that changed their intensity during storage ( $\alpha = 0.05$ ). Oxidative rancidity is associated with the degradation of fatty acids in lipids by reactions with oxygen. Aldehydes and ketones are responsible for rancid odour and flavour and are formed as secondary lipid oxidation products as a consequence of a free radical process on unsaturated fatty acid that can cleave, releasing these volatile compounds.<sup>6</sup> Cardboard and oxidized intensity ratings increased in all samples during storage. By contrast, the intensity ratings of roasted sunflower and essential oil flavours decreased with storage time. In other studies, increases in the intensity ratings of cardboard and oxidized flavours and

a decrease in roasted flavour were also observed in different products during storage.<sup>2,3,8,24</sup>

The increases in the intensity ratings for oxidized and cardboard flavours were higher in RS than in coated roasted sunflower (RS-CMC, RS-CMC-BHT, RS-CMC-T and RS-CMC-A). Significant differences ( $\alpha = 0.05$ ) in intensities of oxidized and cardboard flavours were observed between coated roasted sunflower samples (RS-CMC, RS-CMC-BHT, RS-CMC-T and RS-CMC-A) and RS during storage. RS-CMC-BHT, RS-CMC-T and RS-CMC-A showed the lowest oxidized and cardboard intensity ratings. The addition of natural (thyme and basil essential oils) or synthetic (BHT) antioxidants into the matrix of the CMC edible coating prevented the development of these rancid flavours. The antioxidant activities of essential oils and the preserving effect of edible coatings in roasted peanuts have also been reported in other studies. Ramadan *et al.*<sup>35</sup> studied the sensory characteristics of flavoured ice cream by thyme, basil or marjoram essential oils. Those authors observed that all samples received high scores for all quality attributes in terms of sensory characteristics. The addition of essential oils had no significant effect ( $P \leq 0.05$ ) on the scores of appearance, body and texture, and also, on the characteristic of resistant to melting. The received scores of flavour were significantly ( $P \leq 0.05$ ) higher for samples flavoured with thyme and marjoram essential oils than that of basil essential oil. Erkan<sup>36</sup> investigated the quality and shelf-life of filleted hot-smoked rainbow trout in vacuum packaging (untreated) and treated with thyme oil (TO) or garlic oil (GO), after storage in vacuum-packaged (VP) conditions at 2 °C,

**Table 3.** Coefficients and adjusted  $R^2$  values from regression equations of peroxide value (PV), conjugated dienes (CD), *p*-anisidine value (AV) and sensory attributes (oxidized, cardboard, roasted peanutty, and thyme and basil flavours) in roasted sunflower samples

Variable	Sample <sup>a</sup>	Regression coefficients <sup>b</sup>		$R^2$
		$B_0$	$B_1$	
Peroxide value	RS	8.62	1.835900	0.990491
	RS-CMC	6.17	1.542100	0.991166
	RS-CMC-BHT	8.49	0.670300	0.983621
	RS-CMC-T	6.62	1.052500	0.998038
	RS-CMC-A	8.15	1.532000	0.993793
Conjugated dienes	RS	5.52	0.254200	0.899067
	RS-CMC	5.27	0.200300	0.917372
	RS-CMC-BHT	5.07	0.112400	0.758132
	RS-CMC-T	5.01	0.173900	0.882375
	RS-CMC-A	4.65	0.260100	0.929278
<i>p</i> -Anisidine	RS	0.55	0.053900	0.952316
	RS-CMC	0.67	0.041900	0.874604
	RS-CMC-BHT	0.90	0.011500	0.695613
	RS-CMC-T	0.72	0.033300	0.641377
	RS-CMC-A	0.84	0.043800	0.766420
Oxidized flavour	RS	3.75	0.723600	0.986879
	RS-CMC	3.84	0.365300	0.957533
	RS-CMC-BHT	3.39	0.261700	0.935544
	RS-CMC-T	3.33	0.239000	0.950548
	RS-CMC-A	3.30	0.255300	0.890719
Cardboard flavour	RS	7.19	0.647300	0.971046
	RS-CMC	8.01	0.442100	0.979613
	RS-CMC-BHT	8.92	0.174000	0.936757
	RS-CMC-T	8.63	0.162700	0.986443
	RS-CMC-A	8.43	0.172700	0.936869
Roasted peanutty flavour	RS	60.03	-0.462600	0.980775
	RS-CMC	57.27	-0.396100	0.957030
	RS-CMC-BHT	56.43	-0.265300	0.905892
	RS-CMC-T	56.12	-0.117000	0.816376
	RS-CMC-A	55.63	-0.165700	0.600722
Basil flavour	RS	0.00	0.000000	0.000000
	RS-CMC	0.00	0.000000	0.000000
	RS-CMC-BHT	0.00	0.000000	0.000000
	RS-CMC-T	0.00	0.000000	0.000000
	RS-CMC-A	51.74	-0.608500	0.932152
Thyme flavour	RS	0.00	0.000000	0.000000
	RS-CMC	0.00	0.000000	0.000000
	RS-CMC-BHT	0.00	0.000000	0.000000
	RS-CMC-T	44.49	-0.512500	0.980877
	RS-CMC-A	0.00	0.000000	0.000000

<sup>a</sup> RS, roasted sunflowers; RS-CMC, roasted sunflowers coated with CMC; RS-CMC-T, roasted sunflowers coated with CMC added with thyme essential oil; RS-CMC-A, roasted sunflowers coated with CMC added with basil essential oil; RS-CMC-BHT, roasted sunflowers coated with CMC added with BHT.

<sup>b</sup> Regression coefficients for the general regression equation  $Y = \beta_0 + \beta_1 X$ , where  $Y$  is the dependent variable (PV, CD, AV, oxidized, cardboard, roasted peanutty, thyme or basil flavours) and  $X$  is the independent variable (days of storage).

by measurement of microbiological, sensory and physicochemical analyses. The author observed that the acceptability scores for appearance, odour, taste and texture of untreated and treated smoked trout decreased with storage time. The limit of sensory acceptance was reached after 5 weeks for the untreated samples, after 7 weeks for thyme oil-treated samples (TO + VP) and after 6 weeks for garlic oil-treated (GO + VP) samples. Can<sup>37</sup> studied the microbiological, chemical and sensory (colour, odour, taste, flavour, texture and overall acceptance) changes occurring in rainbow trout (*Oncorhynchus mykiss*) as a function of treatment (control salted samples, salted samples with 1, 3 and 5 g kg<sup>-1</sup> thyme essential oil) and storage time. The author observed that

the sensory scores of trout samples salted with thyme decreased during storage time. However, at the end of the storage period, samples with thyme oil were acceptable by the panellists. The results of this study suggested that salting, thyme essential oil and air packing were found to be effective, easy and cheap methods of fish preservation. Riveros *et al.*<sup>4</sup> determined the sensory stability of roasted peanuts coated with CMC, MC or whey protein during storage. Those authors reported that the intensity ratings of oxidized and cardboard flavours showed lower increases in coated samples during storage, which indicated that these edible coatings had a preservative effect on the sensory properties of roasted peanuts.

The decrease in roasted sunflower intensity ratings was less in coated roasted sunflower seeds (RS-CMC, RS-CMC-BHT, RS-CMC-T and RS-CMC-A) than in RS. Roasted sunflower flavour intensity ratings obtained at the end of the storage period were higher in RS-CMC-T and RS-CMC-A. The results indicate that the use of a CMC coating added with thyme or basil essential oil improved the preservation of this product during storage.

With respect to the correlation and regression analyses, it was observed that PV, CD, AV, and oxidized, cardboard, roasted sunflower, and thyme and basil essential oil flavours were the variables that changed during storage. PV, CD, AV, and oxidized and cardboard flavours presented positive correlations (higher than 0.60). On the other hand, roasted sunflower, thyme and basil essential oil flavours with PV, CD, AV, and oxidized and cardboard flavours exhibited negative correlations. These negative correlations indicate that roasted peanutty, thyme and basil essential oil flavours decreased during storage time and, simultaneously, lipid oxidation indicators increased. The regression equations of PV, CD, AV, and oxidized, cardboard, roasted peanutty, thyme and basil essential oil flavours for RS, RS-CMC, RS-CMC-T, RS-CMC-A and RS-CMC-BHT are shown in Table 3. The dependent variables showed  $R^2 > 0.60$  in all roasted sunflower samples, indicating that these variables are good predictors of the time effect.

## CONCLUSIONS

According to the results observed in the present study, the use of carboxymethyl cellulose edible coatings improves the sensory and chemical stability of roasted sunflower seeds by preventing lipid oxidation and the development of rancid flavours. The addition of thyme or basil essential oils to the coating improves the sensory stability of roasted sunflower seeds during storage, but only thyme essential oil increased their chemical stability. Thyme essential oil could constitute an effective natural antioxidant for this product and, probably also for other foods with similar physical and chemical characteristics.

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