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Impact of Bacterial Cellulose Nanocrystals-Gelatin/Cinnamon Essential Oil Emulsion Coatings on the Quality Attributes of '*Red Delicious*' Apples

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Abstract: This study aimed to assess the effectiveness of bacterial cellulose nanocrystals (BCNCs)-gelatin (GelA)/cinnamon essential oil (CEO) emulsion coatings containing various CEO concentrations (1200, 1800, and 2400 µL/L) in retarding ripening and senescence of '*Red Delicious*' apples during cold storage (60 days at 4 °C). Coatings decreased the weight loss (WL) (~3.6%), as compared to uncoated fruit (~4.8%). A direct relationship between CEO concentration and respiration rate/ethylene production was also disclosed. Flesh firmness was higher for coated samples, with better results detected especially when the highest amount of CEO was applied (36.48 N for the 2400 µL/L delivered dose vs. 32.60 N for the 1200 µL/L one). These findings were corroborated by additional tests on the surface color, total acidity, soluble solids content, pH, ascorbic acid, and activities of polyphenol oxidase (PPO) and peroxidase (POD). This study demonstrated the capability of BCNCs-GelA/CEO systems to dramatically enhance the storability and quality of apples during refrigerated storage, thus avoiding undesired losses and increasing the economic performance of fresh fruit industries.

Keywords: chemical analyses; cold storage; ethylene; firmness; fruit color; respiration rate; shelf life

1. Introduction

Apples (*Malus domestica* cv. Borkh) contain many nutritive and useful compounds, such as vitamins, organic acids, flavonoids, and polyphenols, and are considered one of the most consumed fruits all over the world. Nevertheless, one of the main issues associated with their commercialization is the rapid aging that occurs during postharvest storage due to respiration behavior and ethylene (C_2H_4) production, which renders apples highly perishable as with most climacteric fruit, thus causing an abatement of their shelf life [1,2].

In an attempt to thwart such limitation, multiple efforts have been devoted to the search for effective techniques and/or substances applied to fruits [1,3,4]. Within this frame, edible or biodegradable coatings as physical barriers applied on the surface of fruit have been successfully exploited to improve the surface appearance of apples (especially visual gloss) while controlling the transfer of water vapor, oxygen (O_2), and carbon dioxide (CO_2),

thus slowing down the physiological ripening by actively regulating the respiration rate. Moreover, active coatings have also been used to protect the products against microbial spoilage [5–14].

To this end, several studies have been recently carried out to test a range of coating formulations, with the latter being successfully demonstrated to boost the postharvest shelf life and quality of apples. For example, starch-gellan coatings with thyme essential oil were deposited on '*Golden Delicious*' apples [15]; wheat straw arabinoxylan and β-glucan stearic acid ester coatings were evaluated on '*Rich Red*' apples [16], and '*Royal Delicious*' apples [17]; corn-starch-based coatings with and without calcium were tested on '*Cashew*' apples [18]; polyvinyl alcohol coating incorporating carvacrol was tested on '*Golden Delicious*' apples [14]; fenugreek and flaxseed coatings with corn oil were also evaluated on apples [19]. Other coatings used on apples have been chitosan with banana peel extract [20]; starch-based coating on '*Gropps pink*' apples [21]; chitosan coating combined with ethanolic extract of licorice [22]; tragacanth gum coating containing aloe vera on '*Red/Golden Delicious*' apples [23]; nanochitosan coating on '*Golab Kohanz*' apples [4]; and chitosan coating mixed with olive leaf and pomace extracts [24]. In general, all the above studies highlighted the importance of incorporating the active molecules within a main polymer network or a more sophisticated encapsulating system, with the main goals of controlling the release over time and preserving the stability of the active compound (e.g., from oxygen, light, etc.).

In our previous studies, emulsion coatings based on cinnamon essential oil (CEO) embedded in a bacterial cellulose nanocrystals (BCNCs) cage and stabilized by fish gelatin (GelA) were developed and their film-forming properties were deeply investigated [25,26]. Interestingly, coatings incorporating CNCs for fruit during the postharvest process are comparatively new. As a matter of fact, some recent investigations have discussed the potential application of CNC-based coatings on fruit crops, namely CNC Pickering emulsion incorporated with chitosan deposited on '*Bartlett*' pears [27]; CNC-based pectin incorporating lemongrass essential oil on strawberries [28]; CNC-oleic acid Pickering emulsion incorporated with chitosan on '*Bartlett*' pears [29]; CNC-reinforced chitosan on '*D'Anjou*' and '*Bartlett*' pears [30]; CNC-reinforced gelatin on strawberries [31]; chitosan-nanocellulose composite on strawberries [32]; and acerola puree and alginate reinforced with cellulose whiskers on acerola fruit [33]. In general, based on the outcomes emerging from the above studies, CNCs can be advantageously used for the production of edible coatings to prolong the postharvest shelf life and storability of fruit. Unfortunately, the principal mechanisms of CNCs as key compounds in food coatings, especially for different kinds of fruit, are still unclear and, hence, need to be better understood [34].

Therefore, this study was designed to evaluate the effectiveness of BCNCs-GelA/CEO coatings on the overall quality of apples of the '*Red Delicious*' variety, properly chosen because of its large availability on the market. Toward this goal, several physical (weight loss and surface color), physiological (CO_2 and C_2H_4 production rates), mechanical (flesh firmness), and biochemical (total acidity, total soluble solids, vitamin C, pH, and enzyme activities) attributes were monitored during 60 days of cold storage.

2. Materials and Methods

2.1. Raw Materials and Chemicals

Apple fruits belonging to the '*Red Delicious*' variety were obtained in their commercial maturity stage from an orchard in Meshgin city (Ardabil province, northwest of Iran).

All the chemicals and reagents used in this study for the BCNCs-GelA/CEO nanoemulsion preparation were the same as those reported in our previous works [25,26]. The generation of BC and nanocrystals thereof was performed according to the static fermentation protocol and the sulfuric acid hydrolysis, respectively [35]. For chemical analyses, monosodium phosphate (NaH_2PO_4), disodium phosphate (Na_2HPO_4), polyvinylpolypyrrolidone (PVPP), Triton X-100, catechol, *p*-phenylenediamine, hydrogen peroxide (H_2O_2), and potassium iodate (KIO_3) were obtained from Merck (Darmstadt, Germany).

Potassium iodide (KI), starch, hydrochloric acid (HCl), sodium hydroxide (NaOH), and phenolphthalein were purchased from Mojallali (Dr. Mojallali Industrial Chemical Complex Co., Tehran, Iran).

2.2. Coating Preparation

The obtainment of BCNCs-GelA/CEO emulsions involved a three-step procedure that began with the preparation of BCNCs water suspensions at a loading of 0.4% *w*/*w* [35]. The latter was then added with different volumes of CEO (600, 900, and 1200 µL) at pH 5 to finally yield a series of CEO concentrations, that is, 0.8, 1.2, and 1.6% (*v*/*w*). Ultimately, the CEO/BCNCs system (75 g) was added dropwise into the fish gelatin solution according to the protocol reported in our previous work [25]. The final BCNCs-GelA/CEO emulsions included BCNCs and gelatin at 0.06% *w*/*w* and 3% *w*/*w* concentration, respectively, while the final concentrations of CEO were 0.12, 0.18, and 0.24% (*v*/*w*) (or 1200, 1800, and 2400 µL/L).

2.3. Sample Preparation

Apple fruits were harvested by hand and selected based on uniformity in maturity, color, and size. Apples were immediately dipped into the specific coating solution (0, 0.12, 0.18, and 0.24% *v*/*w* of CEO) for 15 s at 23 °C so that 4 different batches of 23 apples each were finally obtained, namely C0 (coated samples, but without CEO), C1200 (coating with 0.12% *v*/*w* of CEO), C1800 (coating with 0.18% *v*/*w* of CEO), and C2400 (coating with 0.24% *v*/*w* of CEO). Then, the coated samples were dried at ambient conditions for 12 h. Uncoated apples were also used as a control. Apples were then put into a plastic basket with apple trays, packed in traditional packing papers, and kept under controlled conditions (4 ± 0.5 °C, 80–85% RH) for up to 60 days. Throughout the whole storage window, a 20-day time span was chosen for sampling operations, upon which several characterization techniques were performed, as extensively described in the following subsections.

2.4. Sample Characterization

2.4.1. Weight Loss (WL)

The WL was measured using an analytical balance (GF-600, A&D Weighing, Tokyo, Japan). The results are presented as percentage WL [36] and were achieved by using the following formula (Equation (1)):

$$\text{WL } (\%) = (W_i - W_t)/W_i \times 100 \quad (1)$$

where W_i and W_t represent the initial (t = 0 days) and time-dependent weight of fruit, respectively. Final WL values are from at least five replicates.

2.4.2. Respiration Rate

The CO_2 production rate of fruit was determined according to the method previously described by Maftoonazad and Ramaswamy [7], but with slight modifications. Upon sampling, fruits were kept in ambient conditions before experiments to enable them reaching the equilibrium temperature. For each batch, three apples with known mass and volume were placed in a sealed glass jar (2 L in volume). A CO_2/O_2 gas detector device (OXYBABY 6, WITT, Witten, Germany), equipped with a needle sensor, was inserted into the jar every 30 min over the course of 4 h to assess the evolution of CO_2 concentration (%). Regardless of the sampling time (0, 20, 40, and 60 days), the respiration rate (R_{CO_2} in µmol/kg·h) was calculated as reported in Equation (2) [7,37–40]:

$$R_{CO_2} = (y_{CO_2} - y_{i,CO_2})/(t - t_i) \times V_f/W \quad (2)$$

where y_{i,CO_2} is the initial gas concentration in the jar (% *v*/*v*), y_{CO_2} is the final gas concentration (% *v*/*v*), t_i is the initial time and t is the final time (4 h), W is the total apple mass inside the jar (kg), and V_f is the free volume in the jar (mL). Each measurement set was composed of at least three replicates.

2.4.3. C_2H_4 Production Rate

Three apples with known mass and volume, collected from each investigated batch, were put into a 2 L airtight glass jar. After 24 h, the C_2H_4 production was quantified by sampling 1 mL of gas from the jar headspace using a glass syringe. The sampled gas was then injected into a gas chromatograph (Agilent 7890A, Santa Clara, CA, USA). To compensate for the extracted air, 1 mL of environmental air was injected into the jar after sampling. GC adjustments were as follows: injector and oven temperature: 90 °C; detector temperature: 180 °C; carrier gas (helium) flow rate: 7 mL/min. The following equation (Equation (3)) was then used to calculate the C_2H_4 production rate ($R_{C_2H_4}$, in μmol/kg·h) [40]:

$$R_{C_2H_4} = (y_{C_2H_4} - y_{i,C_2H_4})/(t - t_i) \times V_f/W \quad (3)$$

where y_{i,C_2H_4} is the initial gas concentration in the jar, $y_{C_2H_4}$ is the gas concentration at the final time (24 h), ti is the initial time and t is the final time (24 h), W is the apple mass (kg), and V_f is the free volume in the jar (mL). The final results arose from three replicates.

2.4.4. Flesh Firmness

Apples' tissue firmness was measured via penetration tests using an Instron Universal Testing Machine (STM-20) equipped with a 100 N load cell. First, the apples were peeled, and then a stainless steel spherical tipped rod (11 mm in diameter) was used to penetrate the apple pulp up to 9 mm deep at a constant driving probe speed of 20 mm/min. During the experiment, the apple specimens were firmly fixed on a steel plate and the rod moved downwards. The maximum force in the force-deformation plot was considered the flesh firmness [41]. For each batch, final flesh firmness values were from six replicates.

2.4.5. Color Measurement

Apples' color was evaluated using a uniformly inside-lighted chamber. For each batch of samples, five apples were kept separately in the same conditions of cold storage. Two pictures (3025 × 4032 pixels, focal length of 4.15 mm, RGB space color) of both sides of each sample were taken every 20 days during the 60-day storage period. Fiji image processing software (the latest version of ImageJ v.2.2.0/1.53c) was used for image processing. At first, the images were converted into the CIELab color space and color channels (L*, a*, b*) were obtained separately. Then, to unveil the effect of the coating on color changes over time, L*, a*, and b* values were determined and the total color difference (ΔE) was calculated as follows (Equation (4)):

$$\Delta E = \text{sqrt}\ (\Delta L^{*2} + \Delta a^{*2} + \Delta b^{*2}) \quad (4)$$

where ΔL*, Δa*, and Δb* are the differences between L*, a*, and b* of samples every 20 days of storage time in comparison to the first day of storage [7]. Ten images of both sides of five samples were captured and taken as replicates.

2.4.6. Total Soluble Solids (TSS), Total Acidity (TA), pH, and Vitamin C

As far as TSS measurements are concerned, a laboratory blender was employed to extract juice from fruits. The latter was then monitored in terms of Brix index (%) through a scaled refractometer (RHB-32/ATC, Brix 0–32%, 0.2% accuracy; Hong Kong, China). Three replicates were used for each batch.

TA was determined by titration using NaOH 0.1 M in the presence of phenolphthalein as an indicator solution to reach a pH of ~8 and a light pink color. For this purpose, the fruit extract was filtered using a cheesecloth; 5 mL was separated, poured into an Erlenmeyer flask, and diluted with distilled water to reach 200 mL. The titration proceeded until the color of the diluted extract turned to light pink. Finally, total titratable acidity was expressed as mg of malic acid equivalent per mL of juice (mg/mL) according to the following formula (Equation (5)) [42]:

$$\text{Total acidity} = (N \times V_1 \times Eq,wt)/V_2 \quad (5)$$

where N is the normality of used NaOH (mol_{Eq}/mL), V_1 is the consumed volume of NaOH (mL), whereas Eq,wt is the equivalent weight of dominant acid (mg/mol_{Eq}), and V_2 is the volume of utilized fruit juice (mL). The equivalent weight of malic acid is 67 mg/mol_{Eq}.

A pH meter (Metrohm-827, Zofingen, Switzerland) was used to measure the pH of the fruit extract every 20 days [7].

Ascorbic acid (vitamin C) was quantified by titration using a starch solution (5% *w*/*w*) as an indicator. Briefly, 100 g of apple tissue was homogenized by blender, and the obtained extract was filtered using a cheesecloth to obtain the apple juice. The latter was diluted right away using distilled water to reach 100 mL volume, 20 mL of which was finally poured into an Erlenmeyer flask. Afterward, 150 mL distilled water was added to the flask, together with 5 mL potassium iodide 0.006 M and 5 mL HC1 1 M solutions. Titration was carried out using potassium iodite 0.002 M until a blackish or dark blue color was obtained. According to the following redox reaction (Equation (6)):

$$2IO_3^- + 10I^- + 12H^+ \rightarrow 6I_2 + 6H_2O \quad (6)$$

The moles of potassium iodite being known, the ascorbic acid moles in the fruit juice were calculated. Based on the above reaction, each ion of iodite (IO_3^-) gives 3 ions of iodide (I_2) that react with the ascorbic acid in the fruit juice. With the concentration (C, mol/L) and volume (V, mL) of KIO_3 used in the titration known, the number of moles (n) was calculated as follows (Equation (7)):

$$n = C \times V \quad (7)$$

Overall, by knowing the stoichiometry of the following reaction (Equation (8)):

$$\text{Ascorbic acid} + I_2 \rightarrow 2I^- + \text{dehydroascorbic acid} \quad (8)$$

Considering the molecular weight of vitamin C (176.12 g/mol), the amount of ascorbic acid was assessed and subsequently expressed as mg/100 mg (or %) of fruit juice.

Irrespective of the measured parameter (TSS, TA, pH, and vitamin C), five replicates were performed for each batch of samples.

2.4.7. Polyphenol Oxidase (PPO) and Peroxidase (POD) Activity

For the analysis, the enzyme extract was prepared from the fruit tissues using a sodium phosphate buffer 0.4 M, which originated from a solution of disodium hydrogen phosphate (Na_2HPO_4) (pH = 6.5) and a solution of sodium di-hydrogen phosphate (NaH_2PO_4) (pH = 6.5). Then, the enzyme extractive solution was produced, adding 4% (*w*/*v*) of polyvinylpyrrolidone (PVP) and 1% Triton X-100 (*v*/*v*) to the sodium phosphate buffer 0.2 M (pH = 6.5). Finally, fruit tissues were homogenized, and 10 g of the extract was mixed with 20 g of the enzyme extractive solution, followed by centrifugation (LISA, Château-Gontier, France) for 10 min at 4 °C and 7500 rcf. The supernatant was separated carefully to evaluate enzymatic activities. Three replicates were considered for each batch.

To assess the PPO activity, 75 µL of supernatant was mixed with 3 mL sodium phosphate buffer 0.05 M (pH = 6.5) containing catechol 0.07 M. For control purposes, the same method was adopted, but distilled water was used instead of the enzyme extractive solution. The absorbance of the solution was measured using a spectrophotometer (NanoDrop One C, Thermo Scientific, Waltham, MA, USA) at 420 nm and 25 °C for 10 min in kinetic mode. Enzymatic activity was determined based on absorbance changes measured every minute per g of sample [43,44].

To assess the POD activity, 500 µL of supernatant was mixed with 1 mL sodium phosphate buffer 0.05 M (pH = 6.5). To start the reaction, 1 mL sodium phosphate buffer 0.05 M (pH = 6.5) containing 1% *w*/*v* P-phenylenediamine and 500 µL hydrogen peroxide 1.5% *v*/*v* was added. Then, the absorbance of the prepared solution was measured at 485 nm and 25 °C for 10 min in kinetic mode. The enzyme activity was determined based on the absorbance changes measured every minute per g of sample [43,44].

2.4.8. Sensory Analysis

Sensory evaluation was performed according to the method reported by Meilgaard, Civille, and Carr [45] based on the taste, odor, firmness, color, and overall acceptability of fruits, previously stored for 60 days. Trained panelists evaluated the quality of both coated and uncoated apples (0, 1200, 1800, and 2400 µL/L of CEO) using the hedonic test with a 9-point scale (9 = like extremely, 5 = neither like nor dislike, 1 = dislike extremely) for each quality attribute.

2.5. Statistical Analysis

All collected results were treated in the form of a completely randomized design (CRD) and expressed as means ± standard deviations. One-way analysis of variance was used to check for differences among samples. The significance level (p) was fixed at 0.01 and used for comparison of means using Minitab 18 statistical software (Coventry, UK).

3. Results

3.1. Weight Loss

WL% was observed for both uncoated and coated fruits throughout the 60-day cold storage period. As displayed in Figure 1a, a more pronounced WL% was observed for the uncoated samples ($p < 0.01$) over the coated ones after 60 days, irrespective of the CEO loaded in the coating formulation. Peculiarly, C1200 and C1800 presented almost the same WL (1.33% and 1.35%, respectively) after the first 20 days of storage, whereas they differed slightly at the very last sampling time (3.52% and 3.66%, respectively), notwithstanding the absence of statistical differences ($p > 0.01$). Moreover, time affected the increase of WL for all the treatments significantly ($p < 0.01$). Amongst treatments, C2400 was the most effective in minimizing the incipient WL increase, plausibly due to its capability to act as a semipermeable membrane that slowed down the moisture transfer across the surface of the fruit.

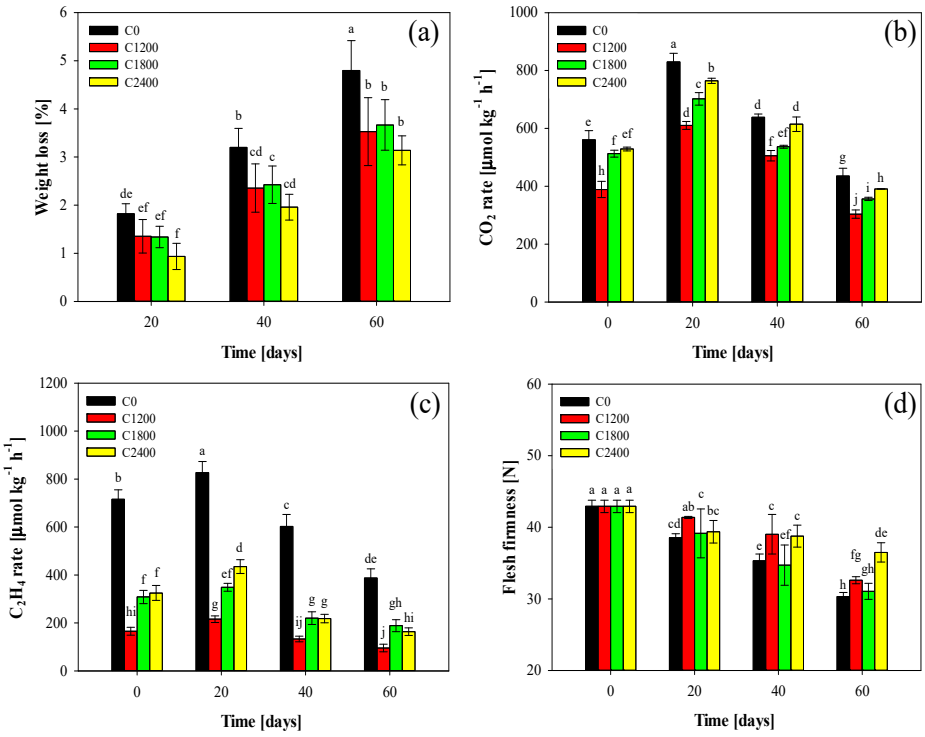

Figure 1. Values of (**a**) weight loss, (**b**) respiration rate, (**c**) ethylene production rate, and (**d**) flesh firmness of untreated (control) and coated (CEO concentration = 1200–2400 µL/L) apple fruits during 60 days of cold storage (4 °C). Different letters above the bars denote significant ($p < 0.01$) differences among mean values.

In full agreement with our previous findings [26], the protective effect against water losses was eventually emphasized at the highest applied concentration of essential oil, owing to the larger presence of hydrophobic CEO droplets which might have consistently inhibited evaporative phenomena. These results are consistent with previously reported studies in which biopolymer coatings were deposited on apples [4,16,19,21,22,24] while being in contrast with the results achieved by Sapper et al. [14,15], who reported on the coating ineffectiveness to prevent or reduce weight loss from apples. The authors explained this result according to two main reasons: (i) the very hydrophilic nature of the applied polymers, which increased the water vapor permeability, and (ii) the low amount of solids that adhered to the surface of the fruit, which yielded final coatings that were too thin. Overall, it can be stated that the coating employed in this work is capable of improving the shelf life of apple fruit by curtailing evapotranspiration and water loss.

3.2. Respiration Rate and c_2h_4 Production

As displayed in Figure 1b, different ($p < 0.01$) respiration rate values were observed among treatments within the different time intervals. As expected, the respiration rate increased for all the treatments after 20 days of storage, indicating that ripening continued after harvesting due to physiological processes. A different scenario was observed after 40 and 60 days of storage, that is, a sudden decrease in the respiration rate was observed for both uncoated and coated fruits. A decidedly similar trend was attained for C_2H_4 production rates at the same time intervals seen for CO_2 production (Figure 1c). According to the ripening pattern of climacteric fruit (e.g., apples), the respiration rate displays a sudden upsurge from the preclimacteric minimum to the climacteric peak (Figure S1 of the Supplementary Material). During this sharp rise, a series of biochemical changes is triggered by the autocatalytic production of C_2H_4, which dictates the transition from ripening to senescence. Concurrently, an increase in the respiration rate is involved, which finally leads to the ripening of the fruit [46]. For these reasons, it can be concluded that the ripening process of apple fruit continued for up to approximately 20 days of storage (abrupt increase of CO_2 and C_2H_4 up to the respiratory climacteric point), after which a rapid decrease of both CO_2 and C_2H_4 was generated.

As far as the effect arising from the coating deposition on the apple samples is concerned, uncoated fruit exhibited higher gas production rates than those associated with the coated ones, thus highlighting the crucial role of coatings in controlling the permeability of CO_2 and C_2H_4. Interestingly, a direct relationship between CEO concentration (from 1200 to 2400 μL/L) and respiration rate/C_2H_4 production was observed, confirming our previous findings on the permeability properties of BCNCs-GelA/CEO films, which showed that increasing CEO concentration within the formulation prompted the CO_2 transmission rate, especially for high relative humidity values (50%) [26]. This can be explained by retrieving the plasticizing effect of CEO, which might have increased the free volume of the main biopolymer phase (gelatin) of the coating network, being responsible for the enhanced mass transfer phenomena. The plasticizing effect of CEO has been compared to that of other common plasticizers, such as glycerol, sorbitol, and even water, though with some differences [26]. In other words, CEO increased the intermolecular free volume within the main gelatin network, thus hindering the hydrogen bonding pattern of native gelatin, due to both steric reasons (small molecule with low molecular weight) and partial chemical incompatibility with the main biopolymer phase. These results are corroborated by the findings of Sapper et al. [15], who reported the same trend in respiration rates when comparing coated and uncoated fruit. In particular, they observed an increase in the CO_2 production rate when carvacrol essential oil was added to the main polymer phase of polyvinyl alcohol. The same authors detected the same effect on the CO_2 reduction when starch-gellan coatings were used [14]. However, when lecithin-encapsulated essential oil was added to the starch-gellan coatings, no significant differences between coated and uncoated samples were observed, again owing to the plasticizing effect of essential oil, as reported before. Deng et al. [47] showed that both C_2H_4 and CO_2 production was significantly reduced

compared to uncoated fruit when using cellulose nanofibers emulsion coatings on bananas, thus delaying the ripening and senescence of fruit over the storage time. In a more recent work, Deng et al. [29] investigated the postharvest storage of pear fruit covered by CNC Pickering emulsion chitosan coatings. All the coated samples exhibited lower C_2H_4 production than control (uncoated) fruit, with the best performance belonging to the coating formulations including CNCs. The authors explained this behavior in terms of superior hydrophobicity of coatings containing CNCs.

3.3. Flesh Firmness

According to the results depicted in Figure 1d, firmness decreased over time for both uncoated and coated samples. In particular, flesh softening can be initially due to the enzymatic activity in the tissue cell walls of the fruit, which is also controlled by C_2H_4 production [48]. Fruit cell walls generally consist of cellulose microfibrils embedded in a hemicellulose matrix with a pectin network serving as the anchoring agent between adjacent cell walls [49]. Cell-wall modifications that occur during maturing and ripening of fruit tissues include decreased cell-cell adhesion, reduced apoplastic pH, depolymerization and solubilization of pectins and hemicelluloses, and a general increase in cell-wall flexibility and porosity [50]. A key enzyme involved in cell-wall degradation and associated with textural changes during apple ripening is polygalacturonase (PG) [51]. Accordingly, the lower tissue softening for the coated samples compared to the uncoated ones could be explained in terms of the curbed C_2H_4 production, in agreement with what was obtained by Sahraei Khosh Gardesh et al. [4] and Zhang et al. [20]. After 40 days of refrigerated storage, the incremental softening was likely due to C_2H_4 accumulation and senescence. It is noteworthy that there was a significant difference ($p < 0.01$) between batches during the storage period, which confirms the influence of CEO concentration and time on the softening of fruit tissues. Control samples exhibited lower firmness values throughout the time of analysis compared to the other treatments. This may be related to the higher WL, respiration rates, and C_2H_4 production, which led concurrently to more intense tissue softening among the samples by speeding up the aging process [14,15]. At the same time, a direct inhibition of PG mediated by CEO can be hypothesized, as recently demonstrated by López-Gómez and co-workers [52]. Amongst treatments, C1200 had the highest flesh firmness force during the first 40 days of storage. This result (retarded tissue softening for C1200 compared to C2400 and C1800) is not surprising, considering the superior performance of the C1200 treatment as far as respiration rate and C_2H_4 production are concerned (Figure 1b,c). At the same time, despite a lower respiration rate and lower C_2H_4 production than C2400, the C1800 treated samples exhibited lower firmness values which, in turn, were comparable to those from uncoated samples ($p > 0.01$). This trend was probably due to the slightly higher WL of C1800 than C2400. The latter showed a moderate firmness decline, except for a sharp decrease in the first 20 days, which can potentially be linked to the respiratory climacteric of apples. Finally, a possible difference among treatments could be due to the CEO concentration and its plasticizing effect.

3.4. Color Measurements

During growth or preharvest and postharvest storage, the color of the fruit varies due to the synthesis of carotenoids and anthocyanins, as well as the degradation of chlorophyll [53]. The graph of Figure 2a reveals that L* values of coated fruits were appreciably higher than those belonging to the control samples over sampling time. In this regard, two hypotheses might be inferred to explain this observation. First, since the L* variations are associated with PPO activity that modifies the tissue surface, coatings could have possibly restricted the enzyme action by reducing the respiration process (Figure 1b), as concluded by Zambrano-Zaragoza et al. [54] and Gardesh et al. [4]. Second, applying the coatings altered the optical properties of the apple's surface owing to the ability of the BCNC-based gelatin polymeric system to reflect light. As can be seen, L* values increased for all the treatments for up to 40 days, whereas a reduction occurred during the last observation

window (40–60 days). This reduction could likely be attributed to the onset of degradation of apple peel pigments after 40 days of storage. Overall, no significant differences were observed for L* values ($p > 0.01$) among coated samples in terms of lightness, irrespective of the investigated time spot.

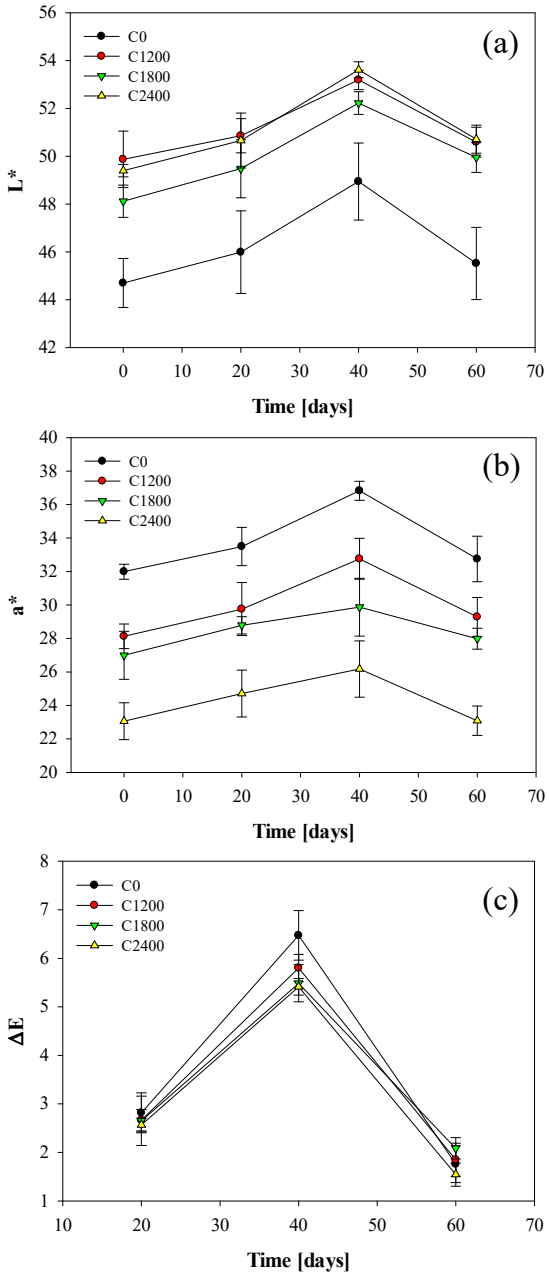

Figure 2. Values of (**a**) L*, (**b**) a*, and (**c**) ΔE of untreated (control) and coated (CEO concentration = 1200–2400 μL/L) apple fruits during 60 days of cold storage (4 °C). Different letters above the bars denote significant ($p < 0.01$) differences between mean values.

As depicted in Figure 2b, a* values changed over time similarly to L* values, though the lowest values pertained to the coated samples, indicating that a decrease in redness occurred in the presence of the coating, especially for the highest concentrations of CEO adopted. Because an increase in CEO has been shown to lead to a proportional increase in haze and a decrease in transparency of films [26], it can be plausibly concluded that coatings influenced the redness of apples by diminishing their surface color. Nevertheless, a* values increased during the first 40 days (Figure 2b), which is due to the chlorophyll

breakdown reactions leading to its reduction [21,55] and fruit ripening. At the same time, red pigments increased (through anthocyanin synthesis) [28] until full pigment development was completed after approximately 40 days.

After this time, pigments started to degrade up to 60 days, causing a decrease in a* values for all the treatments. As regarding the total color difference value (ΔE), there were no significant differences ($p > 0.01$) between uncoated and coated samples, even though high variations over time were observed for each treatment up to 60 days of storage. Only after 40 days did control samples show higher color differences compared to other treatments as a result of full ripening. This finding might be caused by the counteracting effect exerted by L* and a* parameters.

3.5. TSS, TA, pH, and Vitamin C

Figure 3 shows TSS, TA, pH, and vitamin C values. In particular, the TSS of all the treatments underwent a significant ($p < 0.01$) increase throughout the whole monitoring time (Figure 3a) as compared to control samples analyzed at the beginning of the storage phase. It must also be noticed that the TSS values of coated samples were lower than those of uncoated ones, whatever the selected sampling time. It is well-known that the respiration phenomenon strictly relies on the presence of a substrate. In many tissues, this substrate acts as storage of carbohydrates, such as starch. These complex molecules are broken down into simple sugars that enter the respiratory pathway to provide energy for plant sustenance. When these substrates are utilized and completely oxidized, different amounts of oxygen are consumed concerning the amount of CO_2 evolved [56]. By knowing that starch hydrolysis starts during fruit ripening and storage [57–59], we can indisputably assert that the lower detected TSS values of coated apples rather than uncoated samples can be ascribed to the slower respiration process, being also corroborated by the previously shown results of Figure 1b, which demonstrate the lower total sugar content in apples' tissue.

However, within a group (i.e., within the same temporal step), TSS decreased when the CEO concentration in the emulsion coating increased. According to Tanda-Palmu and Grosso [60], this trend can be explained by considering that the higher the WL value, the higher the TSS. Moreover, following our previous findings related to the WL of different treatments (Figure 1a), the higher the CEO concentration, the lower the TSS values (for the same reason, the highest TSS value was obtained for the control samples). In addition to the above explanation, the direct relationship between TSS and WL could be linked to the metabolic reactions involved during the fruit ripening, that is, the intense conversion of starch into sugar and the increase in sweetness of fruit during the storage time [28,61]. Overall, these results indicated that the application of coatings can protect the fruit during storage against the external atmosphere, namely by controlling gas exchange and thus maintaining the TSS of the coated fruit for a longer time than the uncoated fruit.

As shown in Figure 3b, TA values decreased up to the end of storage time for all the treatments. At each time of analysis, the coated samples had higher TA compared to the control, plausibly owing to the lower respiration rate compared to the uncoated fruit. Accordingly, organic acids (here represented by malic acid) are used quickly during respiration reactions [23,32,47,62]. Therefore, TA levels were inversely related to the coating treatment, with the lowest value for the uncoated sample and the highest value for the C2400 treatment, in line with the respiratory behavior depicted in Figure 1b. The progressive decrease of TA over time is supported by the trend of TSS during the 60-day temporal window, as well as by the pH value evolution (Figure 3c). These observations are in line with other studies in which different coatings were used on apples [23,62], strawberries [28,32], pears [29,30], and bananas [47]. As a general trend, the authors reported that TSS values were higher for control samples than for coated fruit, and the increase of TSS during storage was related to the hydrolysis of starch into sugar, WL, and the solubilization of cell-wall pectins, polyuronides, and hemicelluloses. The same authors also reported a reduction tendency for TA during storage, with higher TA values for the coated samples in comparison with control fruit, as observed in this work.

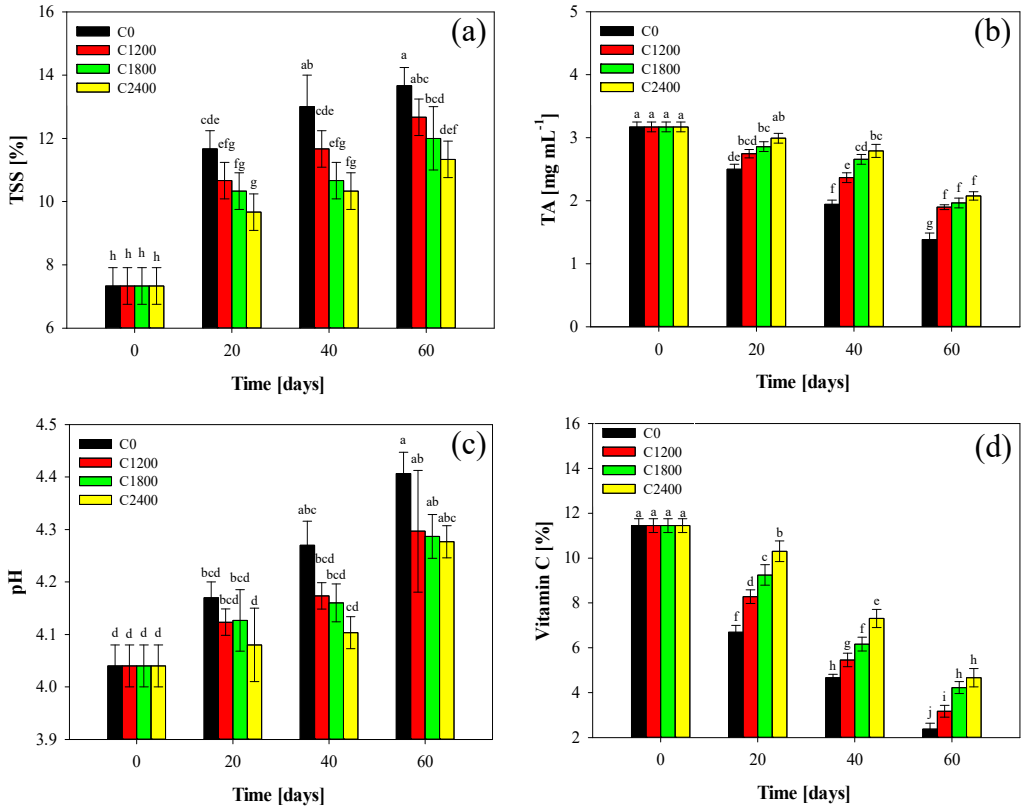

Figure 3. Values of (**a**) total soluble solids (TSS), (**b**) total acidity (TA), (**c**) pH, and (**d**) vitamin C of untreated (control) and coated (CEO concentration = 1200–2400 μL/L) apple fruits during 60 days of cold storage (4 °C). Different letters above the bars denote significant ($p < 0.01$) differences between mean values.

pH values changed significantly ($p < 0.01$) between coated and uncoated fruit and, for each treatment, as a function of time (Figure 3c). More specifically, the pH of the coated samples was lower than that of the control fruit, with the lowest pH value observed for the C2400 treatment. Similar findings were obtained by Jahanshahi et al. [23], who used a tragacanth gum coating on '*Red Delicious*' apples and reported a positive effect of the coating in maintaining pH during 120 days of cold storage.

Vitamin C is considered an important indicator for quality assessment because it is a bioactive compound due to its antioxidant properties [63]. In this work, the amount of vitamin C decreased significantly ($p < 0.01$) for all the treatments over 60 days (Figure 3d). However, its loss was reduced proportionally to CEO concentration in the coatings. At the same time, it can be noted that water loss can play a pivotal role in the decrease of ascorbic acid due to a more intense oxidation process [5,43]. Accordingly, it is plausible that the coatings reduced O_2 diffusion, decreasing the oxidation of vitamin C, which finally led to slower fruit ripening and senescence [18]. This hypothesis is further strengthened by the results of Azeredo et al. [33] obtained on acerola fruit protected with CNC-reinforced alginate coatings, in which the authors proved the effectiveness of CNC as an O_2 barrier with a reduction of the ascorbic acid oxidation rate.

3.6. PPO and POD Activity

PPO and POD activities increased during 60 days of storage for all the treatments (Figure 4a,b). PPO enzymes use O_2 to catalyze the oxidation of fruits' phenolic compounds to form quinones, which ultimately leads to the formation of brown pigments [64–66]. PODs are instead enzymes that catalyze peroxidation, oxidation-catalytic, and hydroxylation reactions. They are involved in ripening, senescence, and darkening reactions [67]. The

susceptibility of fruit to darkness, as well as PPO and POD enzyme activities, may vary according to variety and pre-/postharvest management [68].

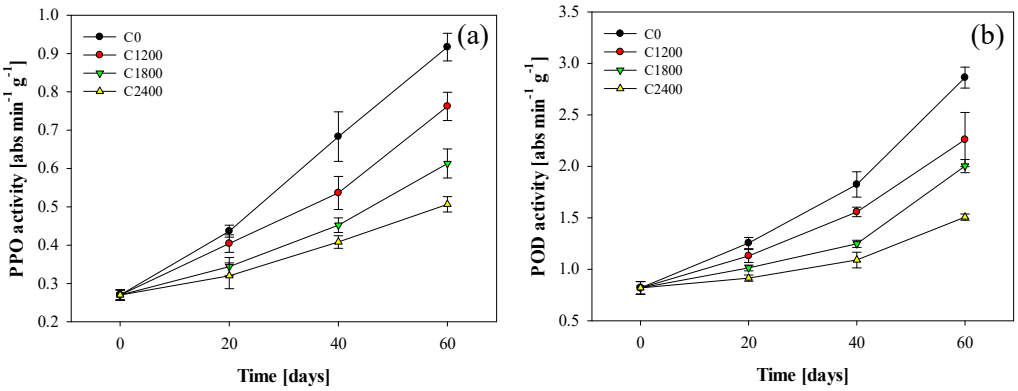

Figure 4. Values of (**a**) PPO activity, and (**b**) POD activity of untreated (control) and coated (CEO concentration = 1200–2400 μL/L) apple fruits during 60 days of cold storage (4 °C).

Ascorbic acid as an antioxidant compound is effective in controlling the enzymatic browning caused by PPO activity [69,70]. Due to the vitamin C decrease during the storage time for coated samples (Figure 3d), it is plausible that a boost in the PPO activity occurred. This hypothesis is supported by the fact that the capability of ascorbic acid to reduce PPO activity is highly dependent on its concentration. Therefore, the inhibitory effect of ascorbic acid may be negligible at certain levels [69]. Indeed, by comparing the intensity of PPO activity among treatments, increasing the concentration of CEO in the coatings significantly reduced ($p < 0.01$) the enzyme activity, due to the antioxidant properties of this essential oil, based on its phenolic and polyphenolic compounds. In particular, cinnamaldehyde, the most representative compound of CEO, has shown a great effect against the enzymatic activity [52,70].

The obtained results agree with the investigations conducted by Eshghi et al. [43], who applied copper-loaded nanochitosan coating on fresh strawberries, and Can et al. [71], who investigated the inhibitory effect of ascorbic acid on the PPO activity of apples. Nonetheless, other studies reported contrasting results, as in the case of the investigation conducted by Othman [69], who reported an enhancement of PPO activity during eight days of storage for pineapple, mango, and papaya fruit. However, PPO activity is affected by many factors, such as temperature, pH of the medium, O_2 availability, amount and type of phenolic compounds in samples, and cultivars, which could be the reason for these contradictory results [66,69]. A similar trend to that seen for PPO was observed for POD enzyme, regardless of the considered batch, throughout the storage period, thus indicating that the coatings delayed the ripening and darkening reactions of apples (Figure 4b). However, it is obvious that POD activity demonstrated a higher rate of reaction than PPOs. Moreover, the higher the CEO concentration, as an antioxidant compound, the stronger the POD activity abatement (Figure 4b). Similar results were obtained by Othman [69] for POD enzyme activities of different fruit (pineapple, mango, and papaya) during their ripening stage.

3.7. Sensory Evaluation

The outcome of the sensory assessment of both uncoated and coated apples at the end of the cold storage period is reported in Figure 5. Based on the sensory values (hedonic scale rating), significant ($p < 0.01$) differences were observed among batches. In particular, the highest CEO concentration (2400 μL/L) allowed the preservation of the sensorial attributes at a higher level in comparison to the lowest CEO loading (1200 μL/L).

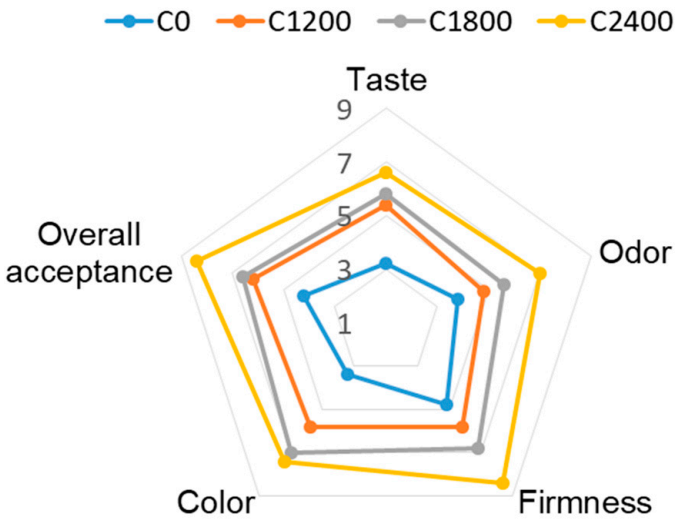

Figure 5. Radar plots showing the effect of GelA/CNCs-CEO coatings loaded with different CEO concentrations (1200–2400 μL/L) on the sensory characteristics of '*Red Delicious*' apples after 60 days of cold storage at 4 °C. Numbers indicate the averaged score attributed by panelists to each parameter upon the hedonic test.

In addition, it can be seen that coated apples, irrespective of the CEO concentration in the coating, received higher sensory scores than the control sample (uncoated apples). Thus, the deposition of active coatings had a positive effect on the overall appearance of apples, raising a positive perception among the panelists. These results might be related to both lower WL% and better equilibrium between fruit components (e.g., sugars and acids) of coated samples compared to the uncoated ones [72]. These findings are decidedly in accordance with those reported by Rashid et al. [19] and Rahimi et al. [73] for apples and peaches, respectively, where it has been proved that coatings and EO remarkably improved both sensory scores and overall acceptance of fruits during storage. Moreover, Rashid et al. [19] reported that the highest tested CEO concentration was the most effective in maintaining the sensory characteristics of apple fruits during cold storage (60 days cold storage at 5 °C), thus corroborating our results.

4. Conclusions

This investigation has shown the important effect of the BCNCs-GelA/CEO emulsion coating in improving the postharvest quality of '*Red Delicious*' apples during cold storage. In particular, the BCNCs-GelA/CEO coating retarded the fruit ripening and senescence (as demonstrated by biochemical, physical, and mechanical analyses) and reduced the evapotranspiration of fruit, thus delaying ripening and extending their shelf life.

In conclusion, due to both ease of manufacturing and efficacy, this type of coating has the potential to be commercialized by the fresh fruit industry for enhanced postharvest storability during long-term cold storage.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/coatings12060741/s1, Figure S1: Typical ripening pattern for climacteric fruits (adapted from Valente et al., 2019). Reference [74] is cited in the supplementary materials.

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