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Impact of Gum Arabic Coating Pretreatment on Quality Attributes of Oven-Dried Red Raspberry (*Rubus idaeus* L.) Fruit

Veronica Morodi, Tafadzwa Kaseke and Olaniyi Amos Fawole *

Postharvest Research Laboratory, Department of Botany and Plant Biotechnology, University of Johannesburg, Auckland Park, P.O. Box 524, Johannesburg 2006, South Africa
* Correspondence: olaniyif@uj.ac.za

Abstract: The present study evaluated the effect of gum arabic (GA) edible coating pretreatment on the quality of dried red raspberries. Red raspberries were independently pretreated with varied concentrations of GA (3, 5, and 10% (*w*/*v*) by dipping for 2 min before oven-drying at 60 °C until the moisture content was below 8% (18–24 h). Raspberries dipped in distilled water were used as the control samples. Quality attributes including colour, moisture content, water activity (a_w), hardness, hygroscopicity, rehydration capacity, total soluble solids (TSS), titratable acidity (TA), pH, anthocyanin composition, ascorbic acid (AA) content, total phenolic content (TPC), antioxidant activity, peroxidase (POD), and polyphenol oxidase (PPO) enzyme activity were investigated. GA pretreatment of the raspberries improved the a_w (lower), hardness (lower), TSS, TSS/TA ratio, BrimA, AA content, and TPC, whilst it significantly ($p < 0.05$) reduced the colour properties (redness, chroma, hue angle, and total colour differences) and the total anthocyanin content when compared with the control samples. The DPPH radical scavenging activity, POD, and PPO enzymes residual activities were not significantly ($p > 0.05$) affected by GA pretreatment. Five different types of anthocyanins, including cyanidin dihexoside, cyanidin 3-*O*-galactoside, cyanidin 3-*O*-glucosyl-rutinoside, and cyanidin 3-*O*-rutinoside were identified and quantified with cyanidin dihexoside being the primary anthocyanin, varying from 951.18–1053.70 µg/g DM. GA pretreatment of raspberries between 3 and 5% could result in improved physicochemical, antioxidant properties and minimum loss of anthocyanins.

Keywords: raspberry fruit; gum arabic; pretreatment; colour; ascorbic acid; cyanidin dihexoside

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1. Introduction

Raspberry (*Rubus idaeus* L.) is an economically important commercial fruit which belongs to the Rosaceae family. The fruit is seasonal, delicate, and characterised by a sweet scent and a delicious bitter–sweet taste [1]. The global production of raspberries has shown significant growth over the past few decades, with an estimated annual production of 800,000 tonnes in 2017, translating to an approximately 64% increase in production from 2010 [2]. According to FAO [2], Russia, Poland, and United States of America are the top three producers of raspberries, contributing about 66% of the global production output. Meanwhile, production in South Africa is still relatively small due to high costs of production, limited harvesting, postharvest handling and storage knowledge of the fruit [3]. The significant increase in the raspberries' global production has been driven by the rising demand attributed to its 'superfruit' status due to the presence of many health-benefitting compounds: vitamin C and E, folic acid, flavanols, phenolic acids, ellagitannins, and anthocynanins [4,5]. Raspberries may reduce skin aging, and prevent chronic diseases such as cancer, diabetes, arthritis, and obesity [6]; as a result, the fruit has been widely studied by the cosmetic, agricultural, pharmaceutical, and food industries [7].

Despite its acclaimed health benefits and growing popularity, the fruit is highly perishable due to its high moisture content (83–86%), making it susceptible to microbial growth

and decay, thus limiting its shelf-life to only about 2–5 days [8]. Physiological disorders such as softening due to rapid ripening and senescence, moisture loss, mechanical damage, and pathogens (*Botrytis cinerea* and *Rhizopus* sp.) are the major factors contributing to postharvest losses in raspberries [9]. These challenges have considerably affected the marketing and storability of raspberries, especially in South Africa. Currently, the raspberry industry relies on cold storage to extend the shelf life of raspberries; nonetheless, the approach only extends the shelf-life to a few days. Alternatively, raspberries can be processed into products such as juices, jams, jellies, wines and dried fruits. Dried raspberries can be consumed as snacks or used in the formulation of other foods such as cereal mixes or bakery and dairy products, increasing raspberries' economic value [5].

A method of interest specific to the current study, hot-air drying, has been used to preserve the quality of fruits and vegetables by decreasing the moisture content and water activity, minimising microbial spoilage, enhancing the storage stability, and product diversification. Furthermore, hot-air drying is versatile, cost-effective and practical [10]. The main drawback of hot-air drying of fruits and vegetables is that it negatively impacts the physical, nutritional, and sensory properties (colour, texture, flavour) of the dried products compared to low temperature drying methods such as freeze-drying and vacuum drying [11]. To prevent or minimise these quality losses, the pretreatment of the fruits or vegetables before drying has been recommended.

Among these pretreatment methods, edible coatings have drawn the interest of many researchers due to their ability to retain the colour and functional compounds and minimise the shrinkage of the dried product [12]. The edible coatings preserve the quality of the dried products by forming a thin layer on the surface of the product, providing a selective barrier against moisture, carbon dioxide, and oxygen [13]. Furthermore, the edible coatings can also carry antioxidants, which enhance the edible coatings' protective effect by preventing the loss of bioactive compounds during the drying process. Due to its non-thermal nature, edible coating pretreatment can be a potential substitute for thermal pretreatments such as blanching, microwave, and infrared, which may present challenges such as poor rehydration, structural collapse, quality degradation, nutrient losses and high energy consumption [14]. Among various edible coatings, gum arabic (GA), a derivative of the sap from the Acacia tree, is abundant, inexpensive, biodegradable and non-toxic. Evidence from the literature has shown that GA alone or combined with other biopolymers has been successfully applied on tomato slices and grapefruit slices to preserve the dried products' quality [10,15].

Despite the evidence in the literature on the application of edible coatings as a pretreatment method for drying fruits and vegetables, the current study is the first to evaluate the effect of GA edible coating pretreatment on the quality functional attributes of dried red raspberries. The findings of this research might assist in the development of a practical, inexpensive, and sustainable protocol for drying red raspberries that raspberry farmers and processors can adopt.

2. Materials and Methods

2.1. Plant Material and Chemicals

Fresh red raspberries (*Rubus idaeus* L.) 'Microprop' were procured at commercial maturity from The Field Berry Farm (De Deur) Johannesburg, Gauteng Province, South Africa (26.3585° S, 27.9199° E). The fruit were transported to the Postharvest Research Laboratory, University of Johannesburg, and stored at 5 ± 0.1 °C (95% relative humidity), and used within 72 h. Gum arabic was bought from Sigma Aldrich, South Africa. Chemicals including methanol (absolute), metaphosphoric acid, sodium carbonate, sodium acetate, hydrochloric acid, ferric chloride, ascorbic acid, 2,6-dichlorophenolindophenol dye, 2,4,6-tri(2-pyridyl)-s-triazine (TPTZ), Folin-Ciocalteu's phenol reagent, 2,2-diphenyl-1-picrylhydrazyl (DPPH), gallic acid, cyanidin-3-glucoside, and 6-hydroxy-2,5,7,8-tetramethyl-chroman-2-carboxylic acid (Trolox), leucine encaphalin were purchased from Sigma-Aldrich, South Africa and were of analytical grade.

2.2. Gum Arabic Coating Preparation

Gum Arabic (GA) coating solutions were prepared at three concentrations, namely 3%, 5%, and 10% (*w*/*v*) in distilled water at 50 ± 0.1 °C on a hot plate with continuous stirring using a magnetic stirrer. The samples were cooled to room temperature (25–27 °C) before use.

2.3. Pretreatment and Drying

Raspberries (1 kg) were independently dipped in each of the prepared GA coating solutions for 2 min, after which they were air-dried using a fan (Goldair, Johannesburg, South Africa) for 2 h Raspberries dipped in distilled water for 2 min were used as the control samples. The control and GA-pretreated raspberries were dried in a hot-air oven (EcoTherm Economy, Labotec, Johannesburg, South Africa) at 60 ± 0.1 °C until the moisture content was below 8% (18–24 h). The dried raspberries were then packed in clear polyethylene zip lock bags, tightly sealed and stored in a sealed cardboard box in darkness at 25 ± 0.5 °C. All experiments were carried out in triplicate.

2.4. Determination of Physicochemical Properties

2.4.1. Moisture Content, Water Activity, and Colour

Moisture content was measured using a moisture analyzer (KERN DBS 60-3, Germany) at 105 °C. The water activity (a_w) was determined using a Novasina electronic dew point water activity meter CH 8853 (Lachen, Switzerland). Before and after drying, the colour properties, including L* (lightness/darkness), a* (redness/greenness), and b* (yellowness/blueness), were measured using a calibrated Chromometer (CR-10 plus, Konica Minolta, Osaka, Japan). The Chroma (C*), hue angle (h°) and total colour difference (ΔE) were calculated using Equations (1)–(3).

$$h^\circ = \tan^{-1} b^*/a^* \quad (1)$$

$$C* = \sqrt{a^{*2} + b^{*2}} \quad (2)$$

$$\Delta E = \sqrt{(L_0{}^* - L^*)^2 + (a_0{}^* - a^*)^2 + (b_0{}^* - b^*)^2} \quad (3)$$

where $L_0{}^*$, $a_0{}^*$, and $b_0{}^*$ represent the colour parameters measured on fresh raspberries.

2.4.2. Hardness, Hygroscopicity, and Rehydration Capacity

The hardness of the dried raspberries was measured using the texture analyzer (Agrosta Calib 2018, Normandy, France) with a 35 mm diameter compression probe. The maximum force (N) at the first compression was recorded as hardness. Three replicates were used in all experiments. Hygroscopicity was measured following the method described by Nthimole et al. [8]. Briefly, about 2 g of the dried raspberries was placed inside a desiccator containing a solution of saturated sodium chloride (75% RH). The results expressed as a percentage (%), were calculated as the ratio of the weight of samples after 7 days to the initial weight of the dried raspberry samples. For rehydration capacity, the samples (1 g) were submerged in 20 mL distilled water at 25 °C, and the weight of the samples was measured at a 10 min interval for 1 h. The initial and final weights of the samples were used to calculate the rehydration capacity.

2.4.3. Total Soluble Solids, pH, and Titratable Acidity

In three replicates, 1 g of dried raspberries (ground) were mixed with 10 mL distilled water, vortexed (30 s), and sonicated (Sonic Clean 705, Labotec, Johannesburg, South Africa) at a frequency, power, and temperature of 50 Hz, 600 W and 25 °C, respectively, for 15 min. The samples were then centrifuged (Biofuge, Thermo Fischer Scientific, Horsham, Sussex, United Kingdom) at 8385× *g* and 4 °C for 5 min to obtain a supernatant that was used in the analysis of the total soluble solids (TSS), pH, and titratable acidity (TA). The TSS was

measured using a digital hand refractometer (PT-32, ATAGO, Tokyo, Japan). The pH was measured using a pH meter (Insmark LS128 model, Mumbai, India). To measure titratable acidity, the supernatant (2 mL) was diluted in 90 mL distilled water and titrated against 0.1 M sodium hydroxide to a final pH of 8.2 using an automated pH Titrator (Orion Star T910, Thermo Fischer Scientific, Horsham, Sussex, United Kingdom). The results were reported as mg/100 mL of citric acid. The TSS and TA were used to calculate the TSS/TA ratio. The BrimA was calculated using Equation (4) below.

$$\text{BrimA} = \text{TSS} - k \times \text{TA} \quad (4)$$

where k is the tongue's sensitivity index. A value of 2 was used to prevent a negative BrimA index.

2.5. Microstructure Analysis

The GA-pretreated and dried raspberry samples were examined using a scanning electron microscope (SEM) (Tescan Vega 3, Brno, Czech Republic) to determine the effect of pretreatment on the morphology of the dried samples. The samples were placed on an adhesive tape and gold coated using a sputter-coating attachment from Balzers (Emscope SC-500). The coated samples were observed under SEM at 40× and 500× magnification.

2.6. Bioactive Phytochemicals and Antioxidant Activity Determination

2.6.1. Ascorbic Acid, and Total Phenolic Content

The modified method described by Adetoro et al. [16] was used to determine the ascorbic acid content of the dried raspberries (ground) using 1% metaphosphoric acid (MPA) in a ratio of 0.1:10. The mixture was vortexed, sonicated, and centrifuged (Thermo-Fischer Scientific, Sussex, United Kingdom) at 8385× g and 4 °C for 5 min. The extracts (0.5 mL) were mixed with 0.025% of 2,6-dichlorophenolindophenol dye before the samples were incubated in a dark place at room temperature (24–25 °C) for 10 min. The absorbance was measured at 515 nm using a UV-visible spectrophotometer (SP-UV 300, Cape Town, South Africa) using 1% MPA as the blank. Ascorbic acid (AA) standard curve (0–120 µg/mL; R^2 = 0.9952) was used, and the results were presented as mg AA per gram of dried raspberries (mg AAE/g DM).

The dried raspberry's total phenolic content (TPC) was determined using the Folin–Ciocalteau method according to Nthimole et al. [8]. Briefly, 1 g of ground dried raspberries were added to 10 mL of 50% cold methanol, vortexed for 30 s, and then centrifuged (Thermo-Fischer Scientific, United Kingdom) at 8385× g, 4 °C for 5 min. The extracts (50 µL) were added to 450 µL of 50% methanol, followed by 500 µL of the Folin–Ciocalteu reagent and 2.5 mL of 2% sodium carbonate. The samples were incubated in darkness at room temperature for 40 min. The sample absorbances were then measured at 725 nm using a spectrophotometer (SP-UV 300, Cape Town, South Africa). A gallic acid standard curve (0–10 ug/mL; R^2 = 0.9916) was used, and the results were presented as mg gallic acid equivalent per gram of dried raspberries (mg GAE/g DM).

2.6.2. Liquid Chromatography-Mass Spectrometry Analysis for Individual Anthocyanins

The extracts for liquid chromatography-mass spectrometry analysis (LC-MS) were prepared by mixing 1 g of the dried raspberries with 10 mL of methanol/1% formic acid solution, after which the samples were vortexed for 1 min, and ultrasonicated (Separation Scientific, Cape Town, South Africa) for 1 h. An aliquot of 2 mL of the samples was centrifuged at 16,435× g for 5 min, and the supernatant obtained was used for LC–MS/MS analysis [17].

The profiling of individual anthocyanins was performed according to the method by Alberts et al. [18]. Briefly, a Waters Acquity UPLC system (Waters Corporation, Milford, MA, USA) consisting of a binary pump, vacuum degasser, autosampler, column oven and Micromass Xevo tandem quadrupole mass spectrometric detector (Manchester, UK) equipped with ESI probe was used. Reversed phase separation was performed using

an Acquity BEH C18 column (2.1 mm × 100 mm, 1.7 m particle size) at a temperature of 50 °C. Mobile phase A consisted of 7.5% formic acid in water, and mobile phase B consisted of acetonitrile. The gradient started with 1% mobile phase B isocratically for 0.5 min followed by a linear increase to 15% at 15 min, 23% at 20 min and 28% at 25 min. Column clean-up at 100% was performed using mobile phase B (for 1 min), followed by re-equilibration for 4 min and the total run-time was 30 min. The flow-rate was 0.1 mL/min throughout, and an injection volume of 10 µL was used. Peak identification was done using the spectral characteristics of analytes, retention times, and cyanidin-3-*O*-glucoside as a reference compound.

The confirmation of molecular formulas of the fragment ions was done using a Waters Synapt G2 quadrupole-time-of-flight, and high-resolution LC–MS/MS analysis was carried out using a mass spectrometer. The LC method and source conditions were identical to those described above for LC–MS/MS analysis, except that collision energy of 60 V was used. Leucine encaphalin (m/z = 556.2771) was used as reference (lock) mass and the instrument was calibrated with sodium formate. The resolution of the instrument was 20,000–24,000 in the mass range, and mass accuracy was better than 2 ppm. The MassLynx 4.1 software (Central Analytical Facilities (CAF), Stellenbosch, South Africa) was used to acquire and process the data. The primary and subtle differences and similarities between the samples were revealed using the metabolomics approach. Four metabolites were annotated and quantified.

2.6.3. Quantification of the Individual Anthocyanins

Standard solutions (5–250 mg/L) of cyanidin 3-*O*-galactoside in 90:10 water: methanol containing 0.5% formic acid were prepared by serial dilutions. The other compounds were quantified using the calibration curves and the cyanidin 3-*O*-galactoside, which was selected based on the principle of structure-related target analyte/standard (functional group and/or chemical structure). The concentration of the individual anthocyanin compounds was reported in µg/g DM. The total anthocyanin content was calculated by summing the concentration of all the individual anthocyanin compounds.

2.6.4. Antioxidant Capacity

The DPPH radical scavenging activity of the dried raspberries was determined following the method described by Fawole and Opara [19]. Under dim light, 15 µL of the dried raspberries' extracts were added to 735 µL of 100% methanol and 0.1 mM DPPH solution (750 µL) and the samples were incubated in the dark for 30 min at room temperature. The absorbances were measured at 517 nm under dim light using a UV-visible spectrophotometer (SP-UV 300, ThermoFischer Scientific, Shanghai, China). A standard curve consisting of AA (0–2 mM; R^2 = 0.9993) was used and the results presented as an AA (mM) equivalent per g of dried raspberries (mM AAE/g DM).

The ferric-reducing antioxidant power (FRAP) was determined according to Benzie and Strain [20]. The FRAP working solution was prepared from 300 mM sodium acetate buffer, pH 3.6; 10 mM of 2,4,6-tri[2-pyridyl]-s-triazine and 20 mM Ferric chloride at 37 °C in a ratio of 10:1:1. Methanolic extracts (150 µL) of the dried raspberries were added to 2850 µL of the FRAP working solution in a cuvette, and the samples incubated for 30 min and in the dark before the absorbances were measured at 593 nm under dim light using a UV-visible spectrophotometer (SP-UV 300, ThermoFischer Scientific, Shanghai, China). Trolox (0–1 mM; R^2 = 0.9999) was used as the standard curve, and results were presented as mM Trolox equivalent per gram of the dried raspberries (mM TE/g DM).

2.7. Determination of Activities of Enzymes Implicated in Browning Reaction

2.7.1. Enzyme Extraction

Dried raspberries were ground into a fine powder and mixed with 0.1 M sodium phosphate buffer, pH 7; 0.05 M/L EDTA and 60 g/L polyvinyl polypyrrolidone (PVPP) in a 1:10 ratio. The mixture was vortexed for 1 min, and then ultrasonicated (Sonic Clean,

Labotec, Johannesburg, South Africa) for 15 min (50 Hz, 600 W) at a temperature of 0 °C. The extracts were centrifuged at 8385× *g*, 4 °C for 5 min and then kept at 4 °C for 2 h. The supernatants were used as the crude enzyme extracts for further analyses.

2.7.2. Polyphenol Oxidase

Polyphenol oxidase activity was determined using a modified method by Meighani et al. [21]. Crude enzyme extracts (200 µL) were added to 50 µL of 0.06 M catechol and 2.8 mL of sodium phosphate buffer (pH 7) at 25 °C. The absorbances were measured at 398 nm, 1 min intervals for 3 min using a UV-visible spectrophotometer (SP-UV 300, ThermoFischer Scientific, Shanghai, China). Sodium phosphate buffer and catechol were used as the blank solutions. A change in absorbance of 0.01 per min referred to one unit of enzyme activity per millilitre per minute (U/min/mL).

2.7.3. Peroxidase

Peroxidase activity was determined using a modified method by Shah et al. [22], in which guaiacol and hydrogen peroxide were the substrates. Crude enzyme extracts (200 µL) were added to 2.2 mL of 0.3% guaiacol (guaiacol dissolved in sodium phosphate buffer at pH 7) at 30 °C and then 0.6 mL of 0.3% hydrogen peroxide. The absorbances were recorded at 470 nm, 1 min interval for 3 min using a UV-visible spectrophotometer (SP-UV 300, ThermoFischer Scientific, Shanghai, China). A change in absorbance of 0.01 per min referred to one unit of enzyme activity per millilitre per minute (U/min/mL).

2.8. Statistical Analysis

The data were presented as mean values ± standard deviation and analysed using one-way analysis of variance (ANOVA) and STATISTICA software (STATISTICA 14.0, TIBCO, Tulsa, OK, United States). Duncan's multiple test range was used to separate the means ($p < 0.05$). The correlations between quality attributes were determined using SPSS for windows. To determine the relationships between the observations and variables, principal component analysis was performed using XLSTAT software Version 2012.4.01 (Addinsoft, New York, NY, USA). Graphical illustrations were prepared using Microsoft Excel (Microsoft cooperation v13.0, Washington, DC, USA).

3. Results and Discussion

3.1. Colour Attributes, Moisture Content, and Water Activity

The consumability of dried fruit products may be determined by physical attributes such as colour. The colour parameters a*, h°, C*, and ΔE of the control and GA-pretreated raspberries are shown in Table 1. GA pretreatment of the raspberry samples significantly ($p < 0.05$) reduced the redness (a*) by 27–35%. The redness results corroborate the C*, which also significantly decreased with the increase in GA concentration. Therefore, the lower C* in the GA-pretreated samples suggests lower colour intensity and masking of the raspberries' colour by the coating. Contrary to our findings, Islam et al. [23] reported increased redness, h°, and C* in papaya slices coated with potato starch before drying. Furthermore, Adiamo et al. [10] observed improved colour on tomato slices after GA coating pretreatment and sun-drying. Coating pumpkin slices with cassava, corn, and potato starch prior to microwave-drying enhanced the dried product's colour [24]. In the case of h°, no significant differences ($p > 0.05$) were observed among the raspberry samples. ΔE, a homogenised representative of the three colour indices (L*, a* and b*), is a more precise indicator of the colour changes in the raspberry samples during drying. Overall, smaller ΔE indicates colour properties closer to the fresh raspberry samples. The ΔE significantly varied among the samples, with the highest and lowest ΔE being exhibited by the control (15.20) and 3% GA-pretreated samples (8.11), respectively. The results indicate that pretreatment of the raspberries with 3% GA resulted in minimum loss of colour. Further increasing the GA concentration from 3 to 5, and 10% increased the ΔE (1.5–2.0-fold higher); however, this did not significantly vary from the control. The dif-

ferences in ΔE among the samples were visible, as illustrated by the pictures in Table 1. Likewise, a 20–30% decrease in ΔE was observed in carboxymethylcellulose pretreated and dried wolfberry fruit [24]. The literature has suggested the possibility of interactions between the natural colourants and the biopolymer coating material, which may enhance their stability [15]. Nevertheless, the findings of this study suggest no possible interactions between the anthocyanins (raspberry colour pigments) and the GA polymer.

Table 1. Colour attributes, moisture content, and water activity of the control, GA-pretreated and oven-dried (60 °C for 18–24 h) raspberries.

				
Pretreatment	**Control**	**3% GA**	**5% GA**	**10% GA**
a*	35.84 ± 0.99 ^{a}	26.65 ± 2.42 ^{b}	19.40 ± 0.75 ^{c}	17.38 ± 0.58 ^{c}
h°	11.88 ± 0.53 ^{a}	11.42 ± 0.76 ^{a}	10.21 ± 1.76 ^{a}	11.44 ± 1.61 ^{a}
C*	36.63 ±0.96 ^{a}	27.20 ±2.47 ^{b}	19.76 ±0.82 ^{c}	17.76 ±0.56 ^{c}
ΔE	15.20 ± 2.45 ^{a}	8.11 ± 0.95 ^{b}	13.96 ± 1.09 ^{a}	12.06 ± 1.08 ^{ab}
MC	4.60 ± 0.13 ^{b}	5.02 ± 0.44 ^{ab}	5.64 ± 0.36 ^{ab}	5.83 ± 0.37 ^{a}
a_w	0.31 ± 0.006 ^{b}	0.27 ±0.003 ^{c}	0.28 ±0.006 ^{c}	0.34 ±0.002 ^{a}

Values represent the mean ± SE (n = 3). Values in a row with different superscripts indicate a significant difference ($p < 0.05$) according to Duncan's multiple range test. GA = gum arabic; MC = moisture content; a* = redness; h° = hue angle; C* = chroma; ΔE = total colour difference; a_w = water activity. Control samples were dipped in distilled water for 2 min.

Moisture content (MC) is one of the important quality indices of dried foods as it determines the dried product's stability and susceptibility to microbial growth during storage. The MC of the control and GA-pretreated raspberries ranged between 4.60–5.83. Raspberries pretreated with 3 and 5% GA did not significantly ($p > 0.05$) vary in MC when compared with the control samples; however, a 1.3-fold increase in MC was observed when the GA concentration was increased from 5 to 10% (Table 1). Our results concur with Islam et al. [23], who observed a higher MC in papaya slices coated with the highest concentration of potato starch (3%). The higher MC could be ascribed to the thicker coating formed around the raspberries, which reduced the moisture migration from the cells during hot-air drying. Water activity (a_w) is a more precise way of measuring the stability of the dried food product to microbiological and biochemical activity. In addition, a_w affects the textural properties of dried foods. Unlike MC, the 3 and 5% GA-pretreated samples exhibited significantly lower a_w (0.27 and 0.28, respectively) compared to the control samples (0.34%). Nonetheless, samples pretreated with 10% GA showed a higher aw (0.34), which correlated with the MC results. The a_w of the dried raspberries was less than 0.6 (maximum recommended a_w for dried food products), suggesting that the a_w could prevent biochemical activities and limit microbial growth during storage, thereby prolonging the dried raspberries' shelf life [25]. Our results agreed with the findings from potato starch, gelatin-gum arabic coated and dried pumpkin, and grapefruit slices, respectively [15,26].

3.2. Hardness, Hygroscopicity, and Rehydration Capacity

Hardness, which measures the samples' resistance to compression at a specified deformation rate, is a valuable indicator of dried foods' texture. The effect of GA pretreatment on the hardness of the dried raspberries is illustrated in Figure 1a. The hardness of the GA-pretreated raspberries increased with the increase in GA concentration, with 10% GA-pretreated raspberries being three times harder than the control samples, which appeared crumbled and flaky. The ability of the coating to maintain the shape and reinforce the structural integrity could be a reason. On one hand, the increased hardness could sug-

gest enhanced firmness in the GA-pretreated raspberries, a characteristic that could be desirable to consumers. The hardness results from the present study are similar to those reported from dried pumpkin slices pretreated with cassava starch, papaya slices coated with calcium lactate, and pomegranate arils coated with sucrose [27–29]. Evidence from the literature has revealed a negative correlation between the MC and hardness of dried food products [30]; however, this kind of relationship could not be established in the current study (Table 1). In this sense, the relationship between the MC and hardness results in the present study should be interpreted with caution.

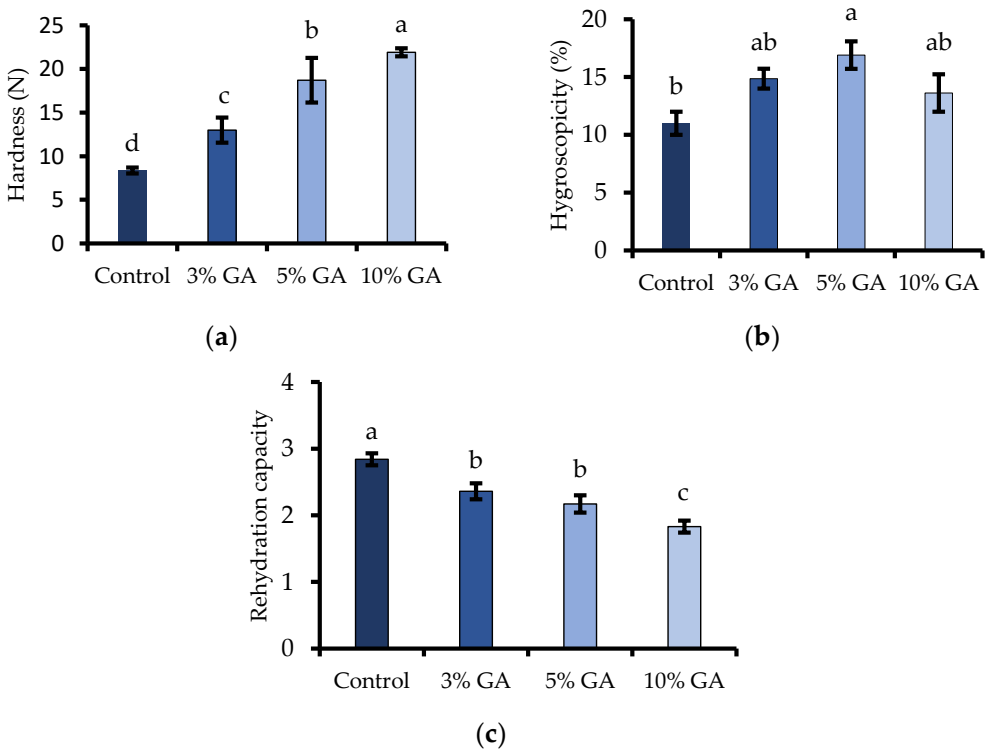


Figure 1. (**a**) Hardness, (**b**) hygroscopicity, and (**c**) rehydration capacity of the control, GA-pretreated and oven-dried (60 °C for 18–24 h) raspberries. Different letters on each bar indicate significant differences in the means ($p < 0.05$) according to Duncan's multiple range test. Vertical bars indicate the standard error of the mean. Control samples were dipped in distilled water for 2 min.

Given that hygroscopicity is vital in determining the stability of dried foods during storage, the potential of the dried raspberries to absorb the environmental moisture was evaluated. As shown in Figure 1b, the raspberries pretreated with 3% GA and 10% GA did not show significant differences in the hygroscopicity compared to the control samples. Nevertheless, the 5% GA-pretreated raspberries were significantly more hygroscopic than the control samples. The literature has reported that dried food samples exhibiting hygroscopicity values of less than 10% could be considered non-hygroscopic and those with values of 10–15% slightly hygroscopic [31]. The hygroscopicity of the samples in the current study varied from 11.00 to 16.90%. This implies that the samples are slightly hygroscopic and would, therefore, require proper packaging and storage to minimise absorption of the environmental moisture by the samples. The higher hygroscopicity of the GA-pretreated samples was expected, given the higher water-binding capacity of GA due to the high number of hydrophilic groups within the polysaccharide polymer [8]. Despite the hydrophilic properties of the GA polymer, the hygroscopicity values varied non-linearly with the increase in the GA concentration, a phenomenon that could be attributed to the presence of cracks on the coating surfaces, which facilitated the absorption of ambient moisture.

The amount and rate of water absorption during the reconstitution of dried products could affect the preparation time and sensory properties [32]. The rehydration capacity of a

dried food product could indicate the extent of structural changes which occurred during the drying process. The rehydration capacity results of the control and GA-pretreated raspberries are shown in Figure 1c. The results illustrate that the rehydration capacity gradually decreased as the GA concentration increased, with 10% GA exhibiting a 36% reduction in the rehydration capacity. In the study of Islam et al. [23], pretreating papaya slices with potato starch (1–3%) before drying significantly reduced the rehydration capacity. Apricots coated with pectin alone or combined with citric acid or ascorbic acid prior to drying showed insignificant variation in the rehydration capacity [33]. The dissimilarities in the rehydration capacity could be due to the type and concentration of the coating pretreatment, among other factors. The literature has highlighted that the rehydration process involves water absorption, swelling, and soluble compounds diffusion, which greatly depends on the cellular and structural integrity of the dried product's tissue matrix. In this regard, GA-pretreatment of the raspberries significantly reduced the porosity of the raspberry tissues, limiting the diffusion of water molecules. Meanwhile, the absence of a protective barrier on the control samples ensured that water molecules easily diffused into the raspberry tissues.

3.3. Total Soluble Solids, pH, Titratable Acidity, and BrimA

TSS is a function of all water-soluble compounds, including acids, sugars, pectins, and vitamin C [10]. The TSS in the present study varied from 5.57 °Brix (3% GA) to 6.12 °Brix (10% GA), with 5 and 10% GA-pretreated samples showing significant ($p < 0.05$) increases (7 and 9%, respectively), when compared to the control samples, indicating that increasing the GA coating concentration enhanced the TSS. The increased TSS could be ascribed to the coating layer around the raspberries, which minimised the rate of heat penetration into the raspberries and the degradation of sugars through caramelisation. However, no significant improvements were observed in the TSS when the GA concentration was increased from 5 to 10%. Osmotic dehydration (55 °C) of apricots coated with pectin alone or combined with citric acid or ascorbic acid showed that the coating pretreatments improved the soluble solids by 3% [33]. Meanwhile, the study of Adiamo et al. [10] reported no significant differences between GA-pretreated, sun-dried tomato slices and the control samples.

As shown in Table 2, no significant differences were observed between the TA of GA-pretreated raspberries and the control samples. The TA observed in the current study was generally low and varied from 0.78–0.81 mg/100 mL citric acid, indicating that GA pretreatment of the raspberries did not significantly alter the concentration of the organic acids, suggesting minimum alteration of the sensory attributes, especially taste. The sugar–acidity balance is crucial in providing the desirable bitter–sweet taste of raspberries. The TSS/TA ratio followed the same trend as the TSS in which no significant differences were observed between the control and 3% GA-pretreated samples, while the 5 and 10% GA-pretreated raspberries showed a significantly higher TSS/TA ratio when compared to the control. This is because TA was relatively lower than the TSS and showed no significant variation between the control and GA-pretreated samples. The BrimA results corroborated the TSS/TA results, given that they measured the balance between the soluble solids and the organic acids in the raspberry samples (Table 2). The effect of GA pretreatment on the pH of the dried raspberries is shown in Table 2. Pretreating the raspberries with 5% GA significantly reduced the pH, whilst pretreatment of the raspberries with 3 and 10% GA significantly increased the pH from 4.23 to 4.27 and 4.28, respectively. The low pH values (4.18–4.28) in the present study suggest that the raspberry samples could be stable during storage at appropriate conditions.

Table 2. Total soluble solids (TSS, °Brix), titratable acidity (TA, mg/100 mL citric acid), TSS/TA ratio, BrimA, and pH of the control, GA-pretreated, and oven-dried (60 °C for 18–24 h) raspberries.

Pretreatment	Control	3% GA	5% GA	10% GA
TSS	5.60 ± 0.06^{b}	5.57 ± 0.09^{b}	6.00 ± 0.15^{a}	6.12 ± 0.13^{a}
TA	0.76 ± 0.02^{a}	0.78 ± 0.02^{a}	0.81 ± 0.01^{a}	0.77 ± 0.01^{a}
TSS/TA ratio	6.80 ± 0.30^{b}	6.87 ± 0.22^{b}	7.70 ± 0.22^{a}	7.69 ± 0.13^{a}
BrimA	3.91 ± 0.03^{b}	4.00 ± 0.13^{b}	4.11 ± 0.07^{b}	4.54 ± 0.08^{a}
pH	4.23 ± 0.009^{b}	4.27 ± 0.001^{a}	4.18 ± 0.004^{c}	4.28 ± 0.014^{a}

Values represent the mean ± SE ($n = 3$). Values in a row with different superscripts indicate significant difference ($p < 0.05$) according to Duncan's multiple range test. GA—gum arabic. Control samples were dipped in distilled water for 2 min.

3.4. Microstructure Analysis

The microstructures and surface structures of the control and GA-pretreated and dried raspberry samples are shown in Figure 2. The 40× (A–D) and 500× (E–H) magnification micrographs show the microstructures and surface structures, respectively, of the control and GA-pretreated samples. At 40× it was clear that increased GA concentration protected the cells from heat-induced cell stress and structural collapse during the hot-air drying process. Control samples and 3% GA-pretreated samples showed intense cell wall rupturing, significant shrinkage, and microstructural changes. The extensive damage to the cell walls of the control and 3% GA-pretreated samples were manifested by the significant reduction in hardness of the respective samples (Figure 1a). The literature has shown that hot-air drying (60 °C) of fruits and vegetables without edible coating pretreatment caused more tissue damage and cell collapse [14,34,35]. The 5% GA-pretreated samples cells showed a round shape and were turgid, with no signs of rupturing. On the other hand, the 10% GA-pretreated raspberries cell walls were partially deformed but did not show signs of rupture. The 500× magnification micrographs revealed some cracks on the raspberries' surfaces, which could be linked to applied GA coatings. The cracks were more pronounced on the surfaces of the raspberries coated with 5 and 10% GA, which could be attributed to sample handling during the experiment. Shen et al. [15] also observed cracks on gelatin—GA composite—coated and dried grapefruit. The authors reported that the cracks did not affect the antioxidant compounds of the freeze-dried grapefruit samples, findings which are similar to the current study. The hyphae (hair-like structures) were observed on all the raspberry samples; however, they were less prominent on the 5 and 10% GA-pretreated samples, indicating that higher GA concentration provided a thicker coating around the raspberries and provided a semi-permeable protective barrier. The findings could explain the lower rehydration capacity for the GA-pretreated samples compared to the control (Figure 1c). Additionally, the intact cell walls, especially from the 5 and 10% GA-pretreated samples, could have provided a barrier against the moisture migration into the raspberry cells, hence the lower rehydration capacity (Figure 1c).

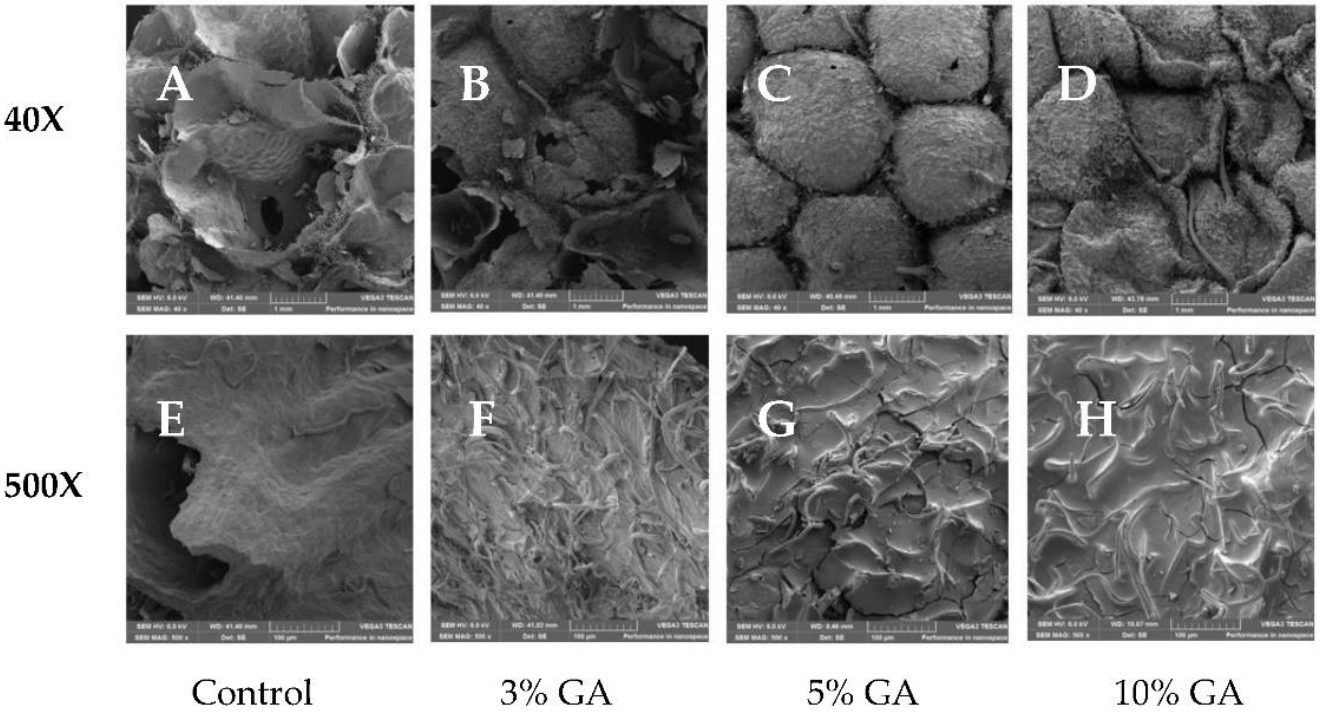


Figure 2. Scanning electron microscopy (SEM) images for the control samples, 3%, 5% and 10% GA-pretreated (**A**–**D**) (40×) and (**E**–**H**) (500×) and oven-dried (60 °C for 18–24 h) raspberries.

3.5. Ascorbic Acid, Total Phenolic Content, and Antioxidant Activity (DPPH and FRAP)

Ascorbic acid (also known as Vitamin C) is a widely known antioxidant that plays various functions in optimal health and disease prevention [27]. In the present study, the concentration of AA from the dried raspberries significantly varied from 13.61 to 14.99 mg/g DM. These findings indicate that a 100 g serving of dried raspberries far exceeds the global recommended dietary allowance (RDA) of vitamin C for healthy adult women and men [36]. Overall, increased concentration was accompanied by a significantly higher AA content, with 10% GA exhibiting a 5–6% higher concentration when compared to the 3 and 5% GA-pretreated raspberries (Figure 3a). It can be hypothesised that GA pretreatment of raspberries prior to hot-air drying minimised the degradation of the AA. Despite being significantly higher than the control samples (13.61 mg/g DM), the AA content for the 3 and 5% GA-pretreated raspberries did not significantly vary (14.29 and 14.12 mg/g DM, respectively). In the study of Islam et al. [23], AA content also significantly increased with increased concentration (1–3%) of potato starch coating on papaya slices (hot-air-dried at 50 °C). The authors attributed this phenomenon to the oxidation barrier characteristics of the starch coating, which prevented the degradation of AA during the hot-air drying process. In the same manner, the GA coating around the raspberries provided a barrier that prevented oxidation of the AA during the oven-drying process. The previous study conducted by Garcia et al. [35] on pectin-coated papaya slices also showed higher AA in coated slices than the control samples.

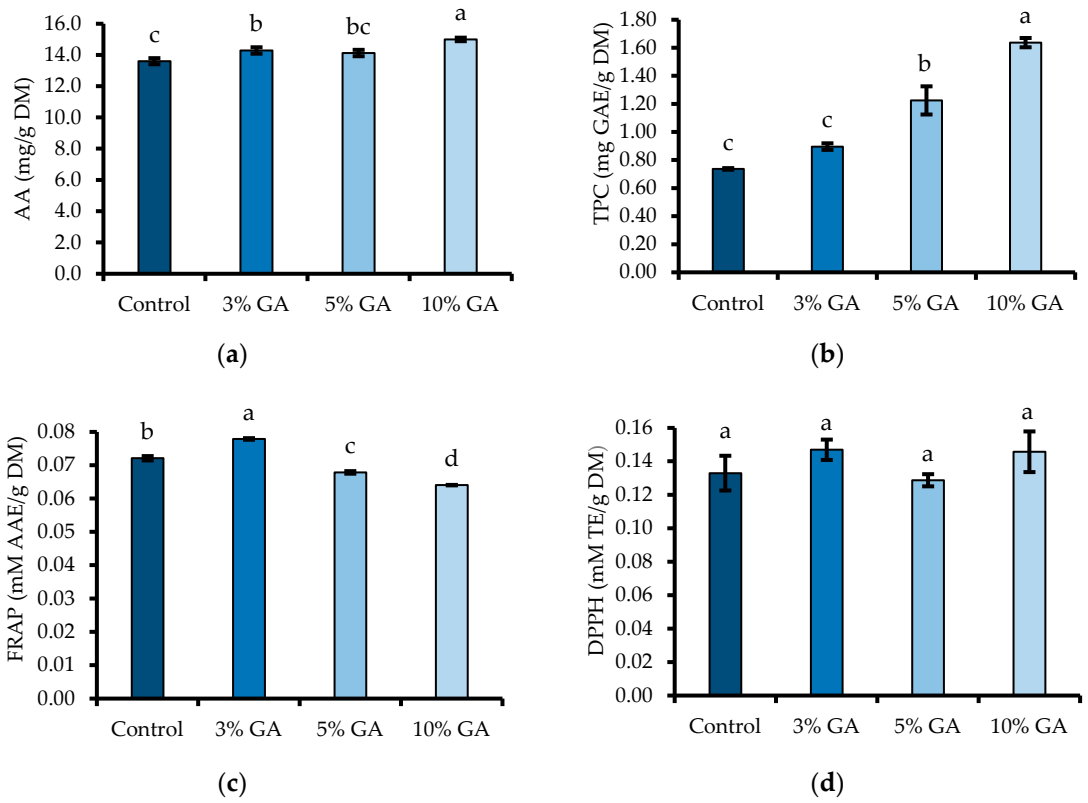


Figure 3. (**a**) Ascorbic acid (AA), (**b**) total phenolic content (TPC), (**c**) ferric-reducing antioxidant power (FRAP), and (**d**) DPPH radical scavenging capacity of the control, GA-pretreated and oven-dried (60 °C for 18–24 h) raspberries. Different letters on each bar indicate significant differences in the means ($p < 0.05$), according to Duncan's multiple range test. Vertical bars indicate the standard error of the mean. Control samples were dipped in distilled water for 2 min. GA = gum arabic; GAE = gallic acid equivalence; TE = trolox equivalence; and AAE = ascorbic acid equivalence.

Phenolic compounds are secondary metabolites that contribute to the sensory and functional properties of plant-based foods. However, these invaluable and heat-labile compounds are affected by processes such as pretreatments and drying [10]. The effect of GA pretreatment of the raspberries on TPC is illustrated in Figure 3b, showing that the TPC was GA concentration-dependent. The TPC values ranged from 0.74–1.64 mgGAE/g DM, with the control and 10% GA-pretreated samples exhibiting the lowest and highest values, respectively. According to the results, increasing the GA concentration from 3 to 5 and 10% significantly ($p < 0.05$) increased the dried raspberries' TPC by 1.3 to 1.8-fold. These findings are quite interesting given the potential of phenolics as antioxidant, antiinflammatory, cardioprotective, cancer chemopreventive and neuroprotective compounds [12]. Drying of fruits and vegetables without pretreatment has been associated with reduced concentrations of antioxidant compounds due to the extensive exposure to light, oxygen, and heat [12]. Thus, the absence of a coating around the control samples could have exposed the phenolic compounds to oxidation and heat damage during the oven drying process. Our results agree with Islam et al. [23] and Yu et al. [25] who reported improved retention of TPC in potato starch (1–3%) and carboxymethyl cellulose (0.5–3%)-pretreated papaya slices and wolfberry fruits, respectively. Both studies established an increased TPC with an increase in coating concentration. Contrarily, osmotic dehydration of apricot cubes coated with pectin alone or pectin combined with citric acid did not increase the retention of TPC when compared to the control samples [33]. Literature has also reported that AA can scavenge and consume the oxygen molecules inhibiting the polyphenol oxidase-catalyzed reactions [33], which could also explain the significantly higher TPC and AA in the GA-pretreated samples. This synergism between AA and phenolic compounds has been

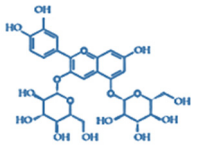
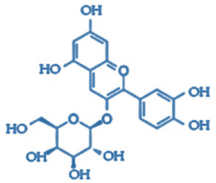
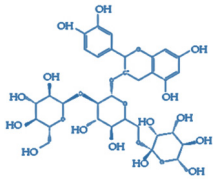
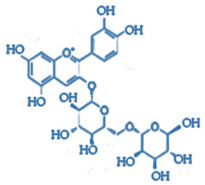
reported in the literature [37]. On the other hand, the lack of correlation between the TPC results and the antioxidant activity (FRAP and DPPH) results could suggest that heating of the GA polymer during the drying of raspberries could have resulted in the formation of compounds that absorb at the same wavelength (725 nm) as phenolic compounds.

The antioxidant activity of fruits and vegetables can be measured using various assays with varied mechanisms, including single electron transfer, hydrogen atom transfer, metal chelation, and reducing power [33]. In the current study, the antioxidant activity of the dried raspberries was measured using the DPPH radical scavenging activity (electron transfer) and ferric-reducing antioxidant power (FRAP) assays. According to Figure 3c, significantly higher FRAP was only recorded from 3% GA-pretreated raspberries, which exhibited an 8% increase in FRAP compared to the control samples. The 5 and 10% GA-pretreated raspberries showed decreased FRAP (3–6% decrease) compared with the control. On the other hand, the dried raspberries' DPPH radical scavenging activity insignificantly varied from 0.13–0.15 mM TE/g DM, despite the significant improvement in the AA and TPC. However, slightly higher DPPH radical scavenging activity was observed in the 3 and 10% GA-pretreated samples (Figure 3d). Unlike in the present study, coating pretreatment of papaya slices using potato starch before drying enhanced their DPPH inhibition (%), and the authors attributed the findings to the minimum degradation of phenolics and AA, which contributed to the higher antioxidant activity [23]. Additionally, sodium carbonate pretreatment of wolfberry fruits before drying at 40 and 50 °C considerably improved the antioxidant activity by 14–22%. Furthermore, significantly higher antioxidant activity was reported for Jerusalem artichoke pretreated with cassava modified starch and sodium caseinate coating prior to hot-air and freeze-drying, when compared with the control [38]. Meanwhile, other researchers highlighted that hot-air drying of coated fruits and vegetables could decrease the antioxidant activity due to enzymatic oxidation during the drying process, attributed to the slow movement of the water molecules from the fruit tissues.

3.6. Anthocyanin Composition

Raspberries have been reported as an important source of polyphenols, particularly anthocyanins; therefore, these valuable compounds have been targeted by researchers for breeding purposes to enhance the quality of the fruit and consumers' interests [39]. The anthocyanin profile of the dried raspberries (Figure 4 and Supplementary Materials-Figure S1), was characterised by LC-MS, and the results are presented in Table 3. Five different types of anthocyanins, including cyanidin dihexoside, cyanidin 3-*O*-galactoside, cyanidin 3-*O*-glucosyl-rutinoside, and cyanidin 3-*O*-rutinoside were identified with reference to their retention time, UV–Vis, mass spectra, and the data available in the literature. Vara et al. [39] managed to identify only two anthocyanin compounds (cyanidin-*O*-sophoroside and cyanidin-*O*-hexoside) from 'Kweli' Red Raspberries, while Szymanowska and Baraniak [40] identified three anthocyanins (cyanidin-3-*O*-sophoroside, cyanidin-3-*O*-glucoside, and cyanidin-3-*O*-rutinoside) from enzyme-treated raspberries. These findings illustrate that profiling a wide range of anthocyanins from raspberries could be challenging.

Table 3. List of anthocyanin compounds tentatively identified in the control, GA-pretreated and oven-dried (60 °C for 18–24 h) raspberries showing retention times, detected $[M-H]^-$ ion, elemental composition, MS^E fragments and UV absorbance.

No.	Experimental *m/z* $[M-H]^-$	Mass Accuracy (ppm)	Retention Time (min)	Elemental Formula	Sensitivity (ppm)	MS^E Fragments	UV (nm)	Tentative Identity	Chemical Structure	Control	3% GA	5% GA	10% GA	Ref.
1	611.1610	0.3	11.36	$C_{27}H_{31}O_{16}$	-	611.1608, 287.0554	550	Cyanidin dihexoside		✓	✓	✓	✓	[17,18]
2	449.1082	3.2	12.31	$C_{21}H_{21}O_{11}$	1.09	449.1070, 287.0558	550	Cyanidin 3-*O*-galactoside		✓	✓	✓	✓	Std *
3	757.2170	3.6	12.53	$C_{33}H_{41}O_{20}$	-	757.2189, 287.0552	550	Cyanidin 3-*O*-glucosyl-rutinoside		✓	✓	✓	✓	[17,18]
4	595.1665	2.4	13.42	$C_{27}H_{31}O_{15}$	-	595.1680, 287.0554	550	Cyanidin 3-*O*-rutinoside		✓	✓	✓	✓	[17,18]

✓ present, * Std, confirmed using a pure chemical standard, literature sources and standards were used to corroborate existing observations. GA = gum arabic. MSE fragments in bold typeface refers to the base peak (the highest peak).

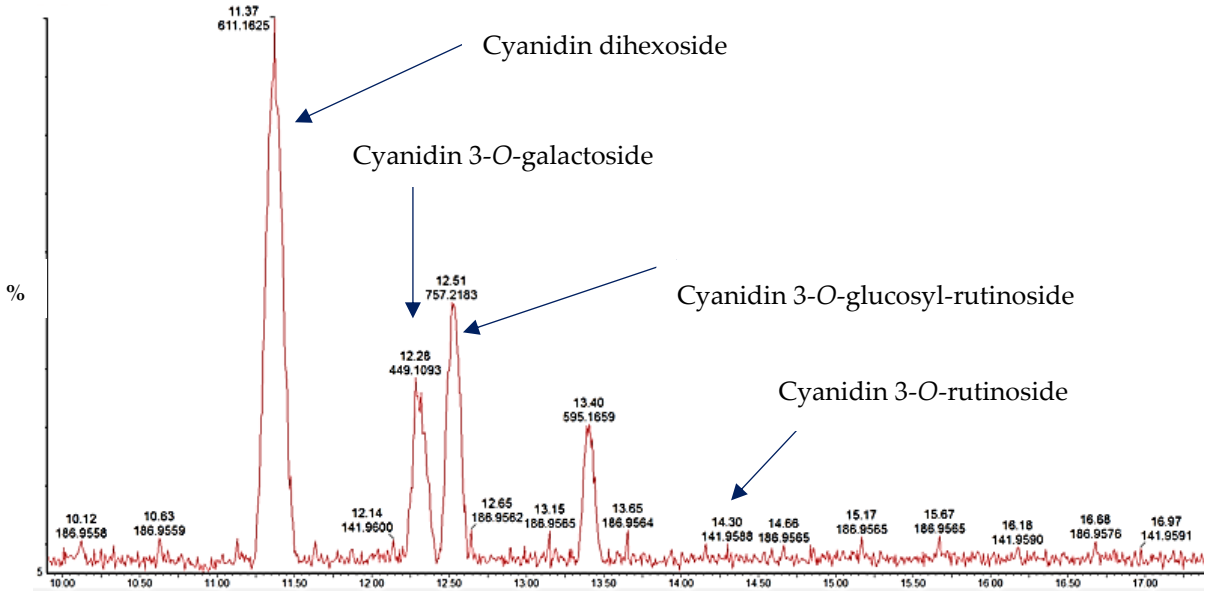


Figure 4. A typical liquid chromatography-mass spectrometry (LC-MS) chromatograph of the major anthocyanin compounds in the oven-dried (60 °C for 18–24 h) raspberries and their retention times.

Compound 1, presented a molecular ion $[M\text{-}H]^-$ at m/z 6111 and an MS^2 fragment at m/z 287, was identified as cyanidin dihexoside. The compound has been identified in previous studies as cyanidin-3,5-diglucoside due to the two glucose molecules attached to the cyanidin compound (Table 3) [41]. Compounds 2 and 3 presented molecular ions $[M\text{-}H]^-$, 449, and 757, respectively, and MS^2 fragments at 287 were identified as cyanidin 3-*O*-galactoside and cyanidin 3-*O*-glucosyl-rutinoside, respectively. Compound 4 presented a molecular ion $[M\text{-}H]^-$ at m/z 595 and an MS^2 fragment at m/z 287 and was identified as cyanidin 3-*O*-rutinoside. Cyanidin dihexoside was the primary anthocyanin and accounted for about 63% of the total anthocyanins identified, followed by cyanidin 3-*O*-galactoside, and cyanidin 3-*O*-glucosyl-rutinoside, which accounted for 21 and 13%, respectively, of the total anthocyanins (Figure 4). The cyanidin 3-*O*-rutinoside was identified as a minor anthocyanin (ca 3% of total), and this result concurs with Szymanowska and Baraniak [40], who observed that the compound accounted for only 5% of the total anthocyanins. The literature has reported cyanidin-3-*O*-sophoroside, cyanidin-3-*O*-glucosylrutinoside and cyanidin-3-*O*-glucoside, as the primary anthocyanins in raspberries. However, the profiling methods applied in the current study failed to identify the cyanidin-3-*O*-sophoroside. Moreover, the anthocyanin profiles of raspberries vary with cultivar, environmental conditions, agricultural practices, maturity stage, processing, and storage [39,41].

As shown in Table 4, the concentration of the individual anthocyanins in the dried raspberries was GA pretreatment dependent. The total anthocyanins varied from 1.56 to 1.65 mg/g DM. The 3 and 10% GA pretreatment of raspberries significantly reduced the total anthocyanins, whereas 5% GA pretreatment did not significantly ($p > 0.05$) change the total anthocyanins. The total anthocyanins (1.56 to 1.65 mg/g DM) from the present study were lower than those reported by Stamenkovic et al. [42] (2.50–2.87 mg/g DM) from convective and freeze-dried raspberries. Among other factors, the differences could be explained by the methods used to determine the total anthocyanins. Compared to the LC-MS method used in the present study, the pH differential method applied by Stamenkovic et al. [42] is less accurate [43], and may be associated with an overestimation of the total anthocyanin content. Sodium alginate coating (0.2, 0.4, and 0.8%) pretreatments applied on purple-fleshed sweet potato cubes before drying using a microwave-assisted spouted bed dryer showed that only 0.2% exhibited the highest anthocyanin level. The pretreatment of raspberries with 5% GA, did not significantly affect the concentration of cyanidin dihexoside, while pretreating the raspberries with 3 and 10% GA caused a 5–10%

reduction in the cyanidin dihexoside. Given the minimum influence of 5% GA on this primary anthocyanin, the concentration can be considered the optimum GA pretreatment concentration for the dried raspberries. Considering its quantity, cyanidin dihexoside can be considered a biomarker of anthocyanin-rich berry intake in humans, and this could help further elucidate the anthocyanins' health importance [44]. GA pretreatment of raspberries before drying significantly decreased (3–5%) the cyanidin 3-*O*-galactoside, while 3 and 10% GA-pretreated dried raspberries maintained the cyanidin 3-*O*-glucosyl-rutinoside at 3 and 12%, respectively (Table 4). The 5% GA pretreatment insignificantly affected the cyanidin 3-*O*-glucosyl-rutinoside. The concentration of cyanidin 3-*O*-rutinoside varied from 34.09 to 43.76 μg/g, with 5% GA insignificantly affecting the anthocyanin, and 3 and 10% GA, significantly decreasing its concentration.

Table 4. Anthocyanin concentration (μg/g DM) of the control, GA-pretreated, and oven-dried (60 °C for 18–24 h) raspberries.

Anthocyanin/Pretreatment	Control	3% GA	5% GA	10% GA
Cyanidin dihexoside	1049.07 ± 22.37 ^{a}	998.30 ± 3.39 ^{b}	1053.70 ± 15.12 ^{a}	951.18 ± 22.74 ^{b}
Cyanidin 3-*O*-galactoside	350.55 ± 3.08 ^{a}	339.09 ± 2.31 ^{b}	334.02 ± 3.58 ^{b}	336.09 ± 2.75 ^{b}
Cyanidin 3-*O*-glucosyl-rutinoside	211.78 ± 11.57 ^{c}	219.01 ± 1.65 ^{b}	209.22 ± 1.65 ^{c}	237.37 ± 2.56 ^{a}
Cyanidin 3-*O*-rutinoside	41.98 ± 0.05 ^{a}	34.09 ± 0.18 ^{b}	43.76 ± 0.53 ^{a}	37.62 ± 0.34 ^{b}
Total Anthocyanins	1650 ± 37 ^{a}	1590 ± 8 ^{b}	1640 ± 21 ^{a}	1560 ± 28 ^{b}

Values represent the mean ± SD (n = 3). Different letters $^{(a–c)}$ on each row indicate significant differences in the means ($p < 0.05$) according to Duncan's multiple range test. Vertical bars indicate the standard deviation of the mean. Control samples were dipped in distilled water for 2 min. GA = gum arabic.

3.7. Enzyme Activity (PPO and POD)

The changes in bioactive phytochemicals of plant material during processes such as pretreatment and drying could be attributed to the activity of PPO and POD enzymes. Given that thermal processing inactivates these enzymes, their residual activity is commonly used as a quality index to measure the effectiveness of thermal pretreatment and drying processes on fruits and vegetables. The effects of GA pretreatment of raspberries on POD and PPO residual activity is illustrated in Figure 5a,b. The residual activity of PPO, which catalyses the oxidation of mono- and diphenols to o-quinones [45], was significantly ($p < 0.05$) reduced (ca 39% decrease) in the 3% GA-pretreated samples (0.100 U/mL/min), whilst no significant variation was observed in the enzyme's residual activity in the control, 5 and 10% GA-pretreated raspberries (0.165, 0.145, 0.147 U/mL/min, respectively). The results suggest that even though the 5 and 10% GA pretreatment provided a thicker coating around the raspberries that provided a barrier for moisture evaporation, this did not negatively influence the residual activity of the PPO enzyme. The significantly low PPO residual activity in the 3% GA-pretreated raspberries could be linked to the considerably lower a_w in the respective samples (Table 1) and delayed oxidation during processing and storage of the packed raspberries [15]. As shown in Figure 5b, GA pretreatment of the raspberries before drying did not significantly ($p > 0.05$) affect the POD residual activity. The literature has reported that POD is one of the most thermostable enzymes responsible for performing single electron oxidation on a wide variety of compounds in the presence of hydrogen peroxide. It is important to highlight that no complete inactivation of both the POD and PPO enzymes was observed in all the pretreatments. The POD and PPO residual activities in the present study (0.1–0.112 U/mL/min) were similar to the PPO and POD residual results (0.01–0.12 U/mL/min) reported by Shen et al. [15] from pectin–GA composite coated and dried grapefruit slices. However, our results were higher than some hot-air dried 'newhall' navel oranges [46]. Factors such as the edible coating pretreatment method, drying conditions, and the nature of the plant material, among others, could be implicated in the variation of the enzymes' residual activities.

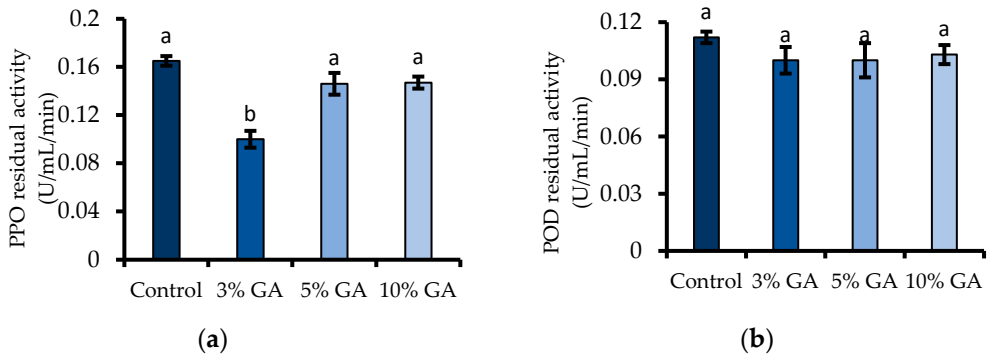
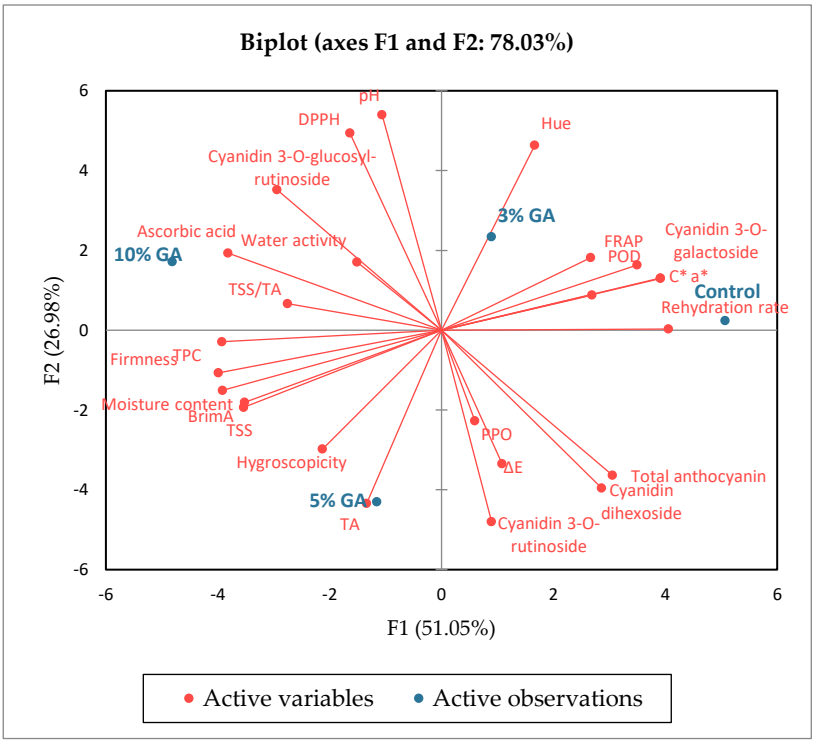


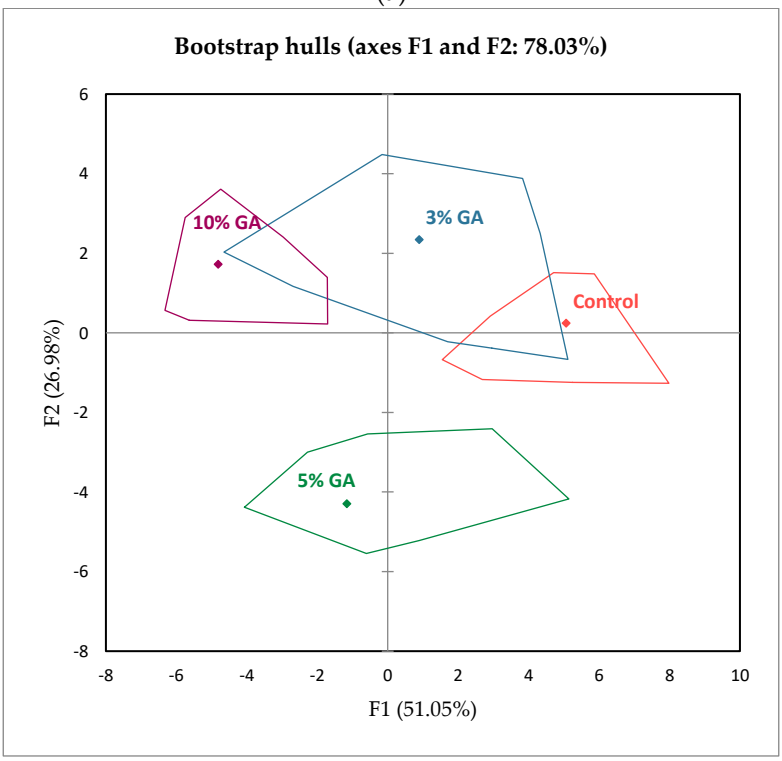
Figure 5. (**a**) Polyphenol oxidase (PPO), (**b**) peroxidase (POD) of the control, GA-pretreated, and oven-dried (60 °C for 18–24 h) raspberries. Different letters on each bar indicate significant differences in the means ($p < 0.05$), according to Duncan's multiple range test. Vertical bars indicate the standard deviation of the mean. Control samples were dipped in distilled water for 2 min. GA = gum arabic.

3.8. Princial Component Analysis

The multivariate data was subjected to principal component analysis (PCA) in order to deeply understand the effect of GA pretreatment on the oven-dried raspberries. The first two factors, F1 and F2, which had the highest eigenvalues (12.76 and 6.75, respectively), were considered the most important principal components (Supplementary Materials-Figure S2). These first two principal components (F1 and F2) accounted for 78.03% of the total variability, with F1 contributing 51.05% (primary variability along the *x*-axis) of the total variability and F2 explaining 26.98% (minor variability along the *y*-axis) of the total variability (Figure 6a). These findings demonstrate that the maximum possible variation in the quality of the oven-dried raspberries was explained by F1. The positive scores on F1 were associated with the control and 10% GA-pretreated raspberries, and these observations were correlated with FRAP, rehydration rate, POD, a*, C*, and cyanidin 3-*O*-galactoside (control) on the positive plane and cyanidin 3-*O*-glucosyl-rutinoside, AA, TSS/TA ratio, TPC, firmness, BrimA, TSS, MC and a_w (10% GA) on the negative plane (Figure 6a). The results confirmed that cyanidin 3-*O*-glucosyl-rutinoside, AA, TSS/TA ratio, TPC, firmness, BrimA, TSS, MC and a_w were significantly higher ($p < 0.05$) in the 10% GA-pretreated raspberries than the control samples. The F1 also affirmed that attributes including POD, a*, C*, and cyanidin 3-*O*-galactoside were considerably high in the control samples, and GA pretreatment significantly reduced these quality attributes. The positive scores on F2 corresponded to 3% GA-pretreated samples, which correlated with pH, DPPH radical scavenging activity, and h°, whilst the negative scores corresponded with 5% GA-pretreated raspberries and were correlated with TA, PPO, ΔE, total anthocyanin, hygroscopicity, cyanidin dihexoside, and cyanidin 3-*O*-rutinoside. However, the TSS/TA ratio, PPO, POD, ΔE and a_w were poorly represented in both F1 and F2. Overall, it can be suggested that GA pretreatment enhanced the chemical properties whilst it reduced the colour properties of the dried raspberries. These findings are supported by the agglomerative hierarchical clustering (AHC), which separated the control samples from the GA-pretreated raspberry samples (Supplementary Materials-Figure S3). The bootstrap hulls plot showed the confidence areas of the samples (Figure 6b). Despite the clear separation of the samples in the PCA biplots and AHC dendrograms, the 3% GA-pretreated samples overlapped with the control, and 10% GA-pretreated samples, indicating similarities in some of the quality attributes among the samples [47]. Nonetheless, the overlapping occurred with the quality attributes that were poorly represented in the F1 and F2 components. On the other hand, the 5% GA-pretreated samples showed no overlapping with the other samples, indicating discrete qualities separating it from the rest of the samples.



(**a**)



(**b**)

Figure 6. (**a**) Principal component analysis (PCA) biplot and (**b**) principal component analysis (PCA) bootstrap hulls showing the relationship between the quality attributes (active variables) and the pretreatments (control, 3%, 5% and 10%) (active observations) and confidence areas of oven-dried (60 °C for 18–24 h) raspberries, respectively. DPPH = 2,2-diphenyl-1-picrylhydrazyl, TSS = total soluble solids, TA = titratable acidity, TPC = total phenolic content, PPO = polyphenol oxidase, POD = peroxidase, FRAP = ferric reducing antioxidant power, C = chroma, a = redness/greenness, ΔE = total colour difference, GA = gum arabic. Control samples were dipped in distilled water for 2 min.

4. Conclusions

The effect of GA edible coating pretreatment on the quality of dried red raspberries was studied. Due to the high perishability of raspberries, the present study could provide an alternative protocol for adding value to raspberries and potentially reduce postharvest losses. The quality of the dried raspberries was dependent on the concentration of GA. GA pretreatment of the raspberries improved the a_w, hardness, TSS, TSS/TA ratio, BrimA, AA, and TPC compared to the control samples. These results are important as they may promote diversified use of raspberry fruits. Enhanced quality attributes are desired by the food industries and consumers. On the other hand, GA pretreatment reduced the colour properties (a*, C*, h°, ΔE) and the total anthocyanin content of the dried raspberries. Given these results, the application of other drying techniques on GA-pretreated raspberries, such as freeze-drying, are recommended. The DPPH radical scavenging activity, POD, and PPO enzymes residual activity did not significantly ($p > 0.05$) vary among the samples. The five different types of anthocyanins profiled and quantified (including cyanidin dihexoside, cyanidin 3-*O*-galactoside, cyanidin 3-*O*-glucosyl-rutinoside, and cyanidin 3-*O*-rutinoside) varied among the treatments. Considering that cyanidin dihexoside was identified as the primary anthocyanin but has been rarely identified in previous studies as the dominant anthocyanin in raspberries, these results may incite further studies on this compound and its potential as a biomarker in red raspberries' benefits. Based on the PCA, we can conclude that GA coating pretreatment of raspberries between 3 and 5% could result in improved physicochemical, antioxidant properties and minimum loss of the anthocyanins. However, further studies are required to evaluate the storability, shelf-life and sensory quality of the 3 and 5% GA-pretreated and dried raspberries.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/pr10081629/s1, Figure S1: Mass spectra of some of the identified anthocyanin compounds observed in the oven-dried (60 °C for 18–24 h) raspberries. Figure S2: Scree plot of variance explained by each factor of the principal component, Figure S3: Agglomerative Hierarchical Clustering (AHC) dendrograms for the control, GA-pretreated and oven-dried (60 °C for 18–24 h) raspberries.

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Abbreviations

AA	Ascorbic acid
DF	Dilution factor
DM	Dry matter
DPPH	2,2-diphenyl-1-picryl hydrazyl
FAO	Food and agriculture organisation

FRAP	Ferric reducing antioxidant power
GA	Gum Arabic
GRAS	Generally regarded as safe
LC-MS	Liquid Chromatography-Mass Spectrometry
MC	Moisture content
PCA	Principal component analysis
POD	Peroxidase
PPO	Polyphenol oxidase
SEM	Scanning electron microscope
TA	Titratable acidity
TE	Trolox equivalence
TPC	Total phenolic content
TPTZ	2,4,6-tri(2-pyridyl)-s-triazine
TSS	Total soluble solids

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