

Article

Encapsulation of Bilberry Extract with Maltodextrin and Gum Arabic by Freeze-Drying: Formulation, Characterisation, and Storage Stability

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Abstract: Anthocyanins are polyphenolic plant pigments associated with antioxidant and health-promoting properties. However, their application in the food industry is limited due to their poor stability. The purpose of this study was to encapsulate anthocyanin-rich bilberry (*Vaccinium myrtillus* L.) extract by freeze-drying and to investigate the effects of different wall materials and extract contents on the physicochemical and bioactive properties of the obtained encapsulates. Ethanolic bilberry extract was encapsulated with the use of maltodextrin (16.5–19.5 DE) (MD), gum Arabic (GA), and their combination in a 1:1 *w/w* ratio (MIX). Bilberry solids to wall material ratios were examined at 20:80, 30:70, and 40:60. All encapsulates showed an attractive red colour and low water activity values ($a_w \leq 0.3$) that indicated a low risk of microbial spoilage. In general, the biggest losses of total phenolic compounds and anthocyanins during three-week storage in the dark and at room temperature (20 ± 2 °C) were detected in the case of encapsulates with a higher content of bilberry extract (MIX30 and MIX40, and GA30 and GA40, respectively). The use of maltodextrin provided the best protection to bilberry anthocyanins during forced storage. Overall, the obtained encapsulates show suitable potential for the development of food products with added nutritional benefits.

Keywords: encapsulation; bilberry; polyphenols; anthocyanins; freeze-drying; maltodextrin; gum Arabic; food colourants



Citation: Todorović, A.; Šturm, L.; Salević-Jelić, A.; Lević, S.; Osojnik Črnivec, I.G.; Prisljan, I.; Skrt, M.; Bjeković, A.; Poklar Ulrih, N.; Nedović, V. Encapsulation of Bilberry Extract with Maltodextrin and Gum Arabic by Freeze-Drying: Formulation, Characterisation, and Storage Stability. *Processes* **2022**, *10*, 1991. <https://doi.org/10.3390/pr10101991>

Academic Editor: Roberto Pisano

Received: 15 August 2022

Accepted: 28 September 2022

Published: 2 October 2022

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1. Introduction

Over the past decades, there has been a growing demand for food products that not only fulfil basic nutritional demands but also provide functional benefits to consumers. Food that, apart from providing necessary nutrients, prevents or reduces the consequences of nutrition-related diseases and increases the physical and mental well-being of consumers is commonly known as functional food [1].

Polyphenols are secondary plant metabolites associated with many health-promoting benefits, mainly because of their antioxidant, anti-inflammatory, and antimicrobial properties [2]. In addition to their biological activities, anthocyanins, a class of polyphenols, are natural pigments responsible for the red, blue, and purple colour of various fruits, vegetables, and flowers [3]. When consumed at normal dietary intake levels, anthocyanins have been shown to have no negative effects on consumers' health [4]. This makes them a promising alternative to artificial food dyes, which have been linked to allergic reactions and neurobehavioural effects in children [5,6].

European blueberry (*Vaccinium myrtillus* L.), also known as bilberry, is a deciduous shrub from the Ericaceae family. It is native to northern and central Europe, but is also found in some parts of North America and Asia [7]. Bilberries are rich in anthocyanins,

flavonoids, and phenolic acids, as well as fibres, minerals, organic acids, and vitamin C [8]. Due to their high polyphenolic content, regular consumption of bilberries and bilberry products has been associated with the prevention of cardiovascular and neurodegenerative diseases, type 2 diabetes, cancer, inflammation processes, etc. [9–13].

However, polyphenols, especially anthocyanins, are also sensitive to many environmental factors (high temperature, light, oxygen, pH, enzymes, etc.), resulting in reduced bioavailability and colour during food processing, storage, and consumption [14].

Encapsulation is a technology based on applying a physical barrier around the bioactive component to offer protection from undesirable environmental conditions [15]. Hence, it can be applied to improve the stability and bioavailability of polyphenolic compounds [16]. Techniques employed for the encapsulation of bioactives include spray drying, freeze-drying (lyophilisation), extrusion methods, emulsification, and coacervation [16–19]. Encapsulated polyphenol extracts can be applied as active components in functional foods and, in the case of anthocyanin encapsulates, as food colourants. Successful incorporation of polyphenol encapsulates into various food products, including jams [20], milk products [21,22], and confectionery [23], has been reported.

Freeze-drying is a dehydration process in which a frozen solvent is removed through sublimation [24]. It is usually followed by grinding, and as a result, highly stable powders with suitable reconstitution properties are obtained [25]. The procedure is simple, flexible, and easily scalable, but its main advantage is the absence of high temperatures, which, unlike spray drying, makes lyophilisation especially suitable for heat-sensitive compounds such as polyphenols [24]. Due to low water content, freeze-dried encapsulates are also easier to incorporate into foods compared to capsules obtained through extrusion or coacervation. Apart from polyphenols, freeze-drying has been applied for the encapsulation of carotenoids [26], flavours [27], essential oils [28], etc.

Maltodextrin and gum Arabic are some of the most common materials for encapsulation due to their excellent barrier properties, high water solubility, and neutral taste, smell, and colour. Maltodextrins are partial starch hydrolysis products with dextrose-equivalent values below 20. Their ability to reduce the hygroscopicity and stickiness of dried powders makes them a great option for encapsulating fruit juices that tend to become adhesive upon drying due to their high sugar and organic acid content [29,30]. Gum Arabic is a dried exudate of the acacia tree and a popular wall material thanks to its emulsifying properties and low viscosity [31]. Its chemical composition is complex and represents a mixture of arabinogalactan oligosaccharides, polysaccharides, and glycoproteins [32].

To the best of our knowledge, encapsulation of bilberry extract using freeze-drying and maltodextrin and gum Arabic has not been reported to this date. For that reason, the purpose of this paper was to develop and characterise such encapsulates.

2. Materials and Methods

2.1. Materials

Bilberries (*Vaccinium myrtillus* L.) were provided by a producer from the Raška district (Serbia). After harvest, the berries were frozen and stored at $-18\text{ }^{\circ}\text{C}$ until further use.

Ethanol (96% *v/v*) was bought from Reahem d.o.o. (Novi Sad, Serbia). Maltodextrin (16.5–19.5 DE) and gum Arabic were purchased from Sigma-Aldrich Chemie GmbH (Steinheim, Germany).

Folin–Ciocalteu reagent, sodium carbonate, gallic acid monohydrate, sodium acetate, 2-azino-bis-3-ethylbenzothiazoline-6-sulphonic acid (ABTS reagent), and manganese (IV) oxide were also purchased from Sigma-Aldrich Chemie GmbH (Steinheim, Germany). Potassium chloride was bought from Kemika (Zagreb, Croatia). All the used chemicals were of analytical grade.

2.2. Extraction

Extraction of bilberry polyphenols was performed according to Može et al. [33], with some modifications. A total of 250 g of frozen bilberries were homogenised for 2 min

in 750 mL of ice-cold ethanol (70% *v/v*). The homogenates were extracted for 3 h at 100 rpm using a GFL 3005 orbital shaker (GFL, Burgwedel, Germany) in the dark at room temperature. After that, the extract was filtered through cotton gauze, followed by filtration through a medium-course filter paper with an 8–12 μm pore diameter (Munktell, Falun, Sweden). Extracts were collected in sealed plastic bottles and stored in the freezer. The sediments from the gauze were reextracted in 500 mL of ice-cold 70% *v/v* ethanol for 2 h, in the dark, at room temperature. Homogenisation and filtration were performed in the same manner as previously described. The obtained extracts were mixed and stored in a freezer until evaporation, which was performed on the Devarot rotary vacuum evaporator (Elektromedicina, Ljubljana, Slovenia) under reduced pressure at 40 °C. The evaporated extract was reconstituted with ~5 mL of distilled water per 60 mL of non-evaporated extract. After that, the evaporated extract was freeze-dried using a BETA 2-8 LD Plus freeze-dryer (Martin Christ Gefriertrocknungsanlagen GmbH, Osterode am Harz, Germany) for around 27 h, at –40 °C and a pressure of 0.12 mbar. The freeze-dried extract was protected from light and water vapour and stored in the refrigerator until further use.

2.3. Encapsulation

The wall materials used for encapsulation in this study were maltodextrin (16.5–19.5 DE) (MD), gum Arabic (GA), and the combination of these two in a 1:1 (*w/w*) ratio (MIX). Three bilberry solids to wall material ratios were used (20:80, 30:70, and 40:60), and as a result, nine different formulations were obtained: MD20, MD30, MD40, GA20, GA30, GA40, MIX20, MIX30, and MIX40 (the numbers in the designations represent the mass fraction of bilberry solids in the obtained encapsulates). Wall material solutions were prepared by dissolving 1 g of wall material (MD, GA, MIX) in 10 mL of distilled water. The solutions were left to hydrate overnight on a magnetic stirrer at room temperature at 300 rpm.

Freeze-dried bilberry extract (1.5 g) was dissolved in 15 mL of distilled water and sonicated in the Bandelin Sonorex TK52 ultrasonic water bath (Bandelin Electronic GmbH & Co. KG, Berlin, Germany) for 30 s at room temperature. After that, the required amounts of dissolved extract were slowly added to the prepared wall material solutions on the RT 5 magnetic stirrer (IKA, Staufen im Breisgau, Germany) and stirred for 5 min at 300 rpm. After stirring, the solutions were quantitatively transferred into centrifuge tubes and diluted with distilled water to a volume of 30 mL. Then, the prepared solutions were homogenised using a Micra D-9 homogeniser (Micra GmbH, Heitersheim, Germany) at 11,000 rpm for 30 s. The homogenised solutions were frozen in liquid nitrogen and freeze-dried using an Alpha 1-2 LD Plus freeze-dryer (Martin Christ Gefriertrocknungsanlagen GmbH, Osterode am Harz, Germany). Freeze-drying was performed for 48 h at –50 °C and a pressure of 0.38 mbar to 0.27 mbar. The obtained freeze-dried materials were ground into powders using mortar and pestle and stored in hermetically sealed flasks at room temperature (20–22 °C) in the dark until further use.

2.4. Colour

The colour of the encapsulates was measured using a Chroma Meter CR-400 (Konica Minolta, Tokyo, Japan) and reported in CIE Lab parameters (L^* , a^* , and b^* values), where L^* was used to denote lightness, a^* —redness (+) and greenness (–), and b^* —yellowness (+) and blueness (–). Chroma (C^*) (colour intensity) and hue (h°) (colour shade) values were calculated from a^* and b^* using the following equations:

$$C^* = \sqrt{a^{*2} + b^{*2}}, \quad (1)$$

$$h^\circ = \arctan\left(\frac{b^*}{a^*}\right) \quad (2)$$

Prior to measurement, the powders were placed into a crucible and gently compacted. Measurements were performed in triplicates at different locations of the sample.

2.5. Water Activity

Water activity (a_w) of the encapsulates was determined using an Aqualab TDL water activity metre (Meter Group Inc., Pullman, WA, USA). Measurements were performed in triplicates.

2.6. Morphological Analysis

The morphological characteristics of the encapsulates were investigated by Scanning Electron Microscopy (SEM) using an FEI Quanta 250 (FEI Company, Hillsboro, OR, USA) scanning electron microscope equipped with an Everhart-Thornley detector. Before imaging, the samples were mounted on sample holders using double-sided conductive carbon tape and coated with gold.

2.7. Fourier Transform Infrared Spectroscopy (FTIR)

FTIR analysis was performed using a Frontier spectrophotometer (PerkinElmer, Waltham, MA, USA) equipped with a diamond prism-based attenuated total reflectance GladiATR module (Pike Technologies, Fitchburg, WI, USA). A small quantity of the samples was placed on the diamond surface and clamped with a consistent low but sufficient pressure. Infrared spectra were obtained against an air blank with 16 scans per spectrum and a spectral resolution of 16 cm^{-1} in the spectral interval $4000\text{--}450\text{ cm}^{-1}$.

2.8. Total Phenolic Content

The total phenolic content (TPC) of the obtained encapsulates was determined by the Folin–Ciocalteu method according to Terpinč et al. [34], with some modifications. Briefly, dissolved encapsulates (0.20 mL) were mixed in a vortex with 0.55 mL of distilled water and 0.125 mL of freshly prepared Folin–Ciocalteu reagent (1:2 diluted) and left in the dark for 7 min. After that, 0.125 mL of Na_2CO_3 was added to the mixture, followed by mixing in a vortex and 30 min incubation. After incubation, centrifugation was performed for 10 min at $8200\times g$. The absorbance was measured in triplicates at 765 nm on an 8453 Hewlett Packard UV-Visible spectrophotometer (Hewlett Packard, Böblingen, Germany). Distilled water was used as a blank. Total phenolic contents were determined from the gallic acid calibration curve ($R^2 = 0.9993$), and the results were expressed in mg of gallic acid equivalents (GAE) per gram of encapsulate.

2.9. Total Monomeric Anthocyanins Content

The total monomeric anthocyanins content (TAC) of the encapsulates was determined by the pH differential method according to Lee et al. [35]. Distilled water was used as a blank. Measurements were performed in duplicates, and the results were calculated using the molecular weight and molar extinction coefficient of cyanidin-3-glucoside (449.2 g/mol ; $26,900\text{ L}\times\text{mol}^{-1}\times\text{cm}^{-1}$) and expressed in mg of cyanidin-3-glucoside equivalents (CGE) per gram of encapsulate.

2.10. Antioxidant Capacity

The antioxidant capacity (AOC) of the encapsulates was measured using the ABTS assay according to Abramovič et al. [36], with some modifications. The radical cation of ABTS ($\text{ABTS}^{\bullet+}$) was produced by dissolving 25.1 mg of ABTS reagent in 10 mL of mQ water and adding 50 mg of MnO_2 . The solution was mixed in a vortex and left overnight (16–18 h) in the dark. After that, the solution was centrifuged for 5 min at 3000 rpm and filtered through a $0.2\text{ }\mu\text{m}$ syringe filter. The samples were prepared by dissolving them in distilled water. Dissolved samples ($30\text{ }\mu\text{L}$) were mixed with $50\text{ }\mu\text{L}$ of $\text{ABTS}^{\bullet+}$ solution and $920\text{ }\mu\text{L}$ of mQ water. The mixture was mixed in a vortex, and after 4 min, the absorbance was read at 734 nm. Measurements were performed in triplicates, and distilled water was used as a blank. The total antioxidant capacity of the encapsulates was read from the gallic acid calibration curve ($R^2 = 0.9793$), and the results were expressed in mg of gallic acid equivalents (GAE) per gram of encapsulate.

2.11. Thermal Properties

Thermal properties of the encapsulates were investigated by differential scanning calorimetry (DSC) using Discovery DSC 2500 (TA Instruments, New Castle, DE, USA) and TRIOS data analysis software (TA Instruments, New Castle, DE, USA) for all measurements. The samples (~4 mg) were placed in hermetically sealed aluminium pans, with an empty pan run as a reference. The experiments were carried out under a nitrogen atmosphere with a flow rate of 50 mL/min. Each sample was heated at a rate of 5 °C/min from 0 to 100 °C.

2.12. Storage Stability

Total phenolic content, total monomeric anthocyanins content, and antioxidant capacity of the encapsulates were determined after the encapsulation, as well as during the following three weeks. Measurements were performed weekly to investigate potential changes in the encapsulates. The samples were stored in centrifuge tubes at room temperature (20 ± 2 °C), protected from light and humidity.

2.13. Forced Storage Test

A forced storage test was performed to investigate the effects of higher temperatures on the monomeric anthocyanins content and the antioxidant capacity of the encapsulates. The samples were stored in microcentrifuge tubes for five days in a WB-30 water bath (Kambič d.o.o., Semič, Slovenia) at different temperatures (40, 50, and 60 °C) in the dark. After the water bath, the samples were diluted in distilled water and sonicated in the water bath for 10 min. Measurements were performed in duplicates, as previously described.

2.14. Statistical Analysis

Statistical analysis was performed using the SPSS Statistics 26 software (IBM, Armonk, NY, USA). Analysis of variance (ANOVA) and Tukey's test for pair comparison were carried out using a significance level of 95% confidence ($p \leq 0.05$). The results are presented as means \pm standard deviation.

3. Results

3.1. Colour

The obtained encapsulates and their chroma (C^*) values are shown in Figure 1, while all the CIELab colour coordinates are given in Table 1. As expected, all encapsulates exhibited low h° values, closest to the angle for red colour (0°). The L^* values indicate that the higher extract content resulted in a darker colour of the encapsulates. The same conclusion was reached by Estupiñan-Amaya et al. [24] regarding freeze-dried Andean blueberry (*Vaccinium meridionale* Sw.) encapsulates prepared with variable proportions of maltodextrin (20–50%). The C^* values of GA and MIX powders were also influenced by extract content, resulting in significantly ($p \leq 0.05$) less intensely saturated GA20 and MIX20 encapsulates (Figure 1). However, the most intensely saturated were the maltodextrin encapsulates, regardless of the extract concentration. This is in line with the findings reported by Kalušević et al. [37] for spray-dried soybean coat extract encapsulated with maltodextrin, gum Arabic, and skimmed milk.

3.2. Water Activity

Water activity (a_w) values of the bilberry extract encapsulates are also shown in Table 1. A low water activity (≤ 0.3) indicates a low risk of microbial spoilage and (non) enzymatic reactions, implying a long shelf life [38]. According to a study conducted by Baeza et al. [39], low water activity also shows a positive impact on anthocyanins' stability. Lowering a_w resulted in a lower degradation rate of anthocyanins in freeze-dried encapsulated elderberry pulp during 90 days of storage at 38 °C. Low water activity results in low molecular mobility and, therefore, a lower rate of physicochemical degradation reactions [40].

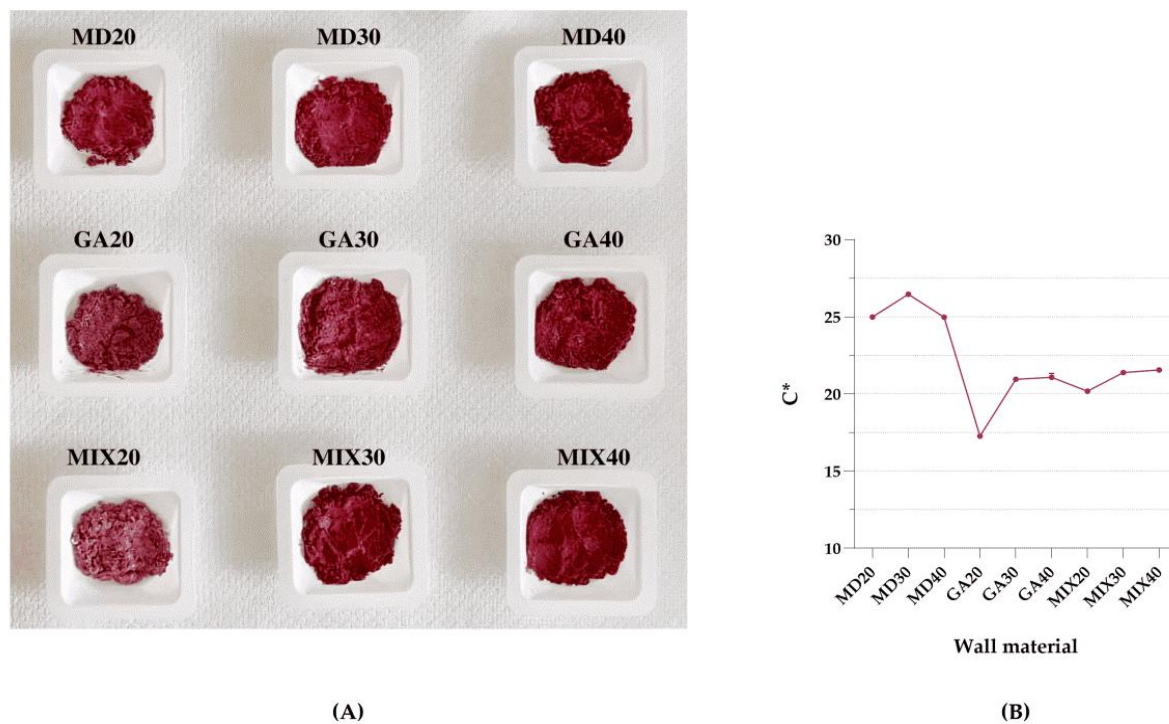


Figure 1. (A) Photographs and (B) chroma (C^*) values of the obtained encapsulates.

Table 1. Water activity and CIELab colour parameters of the encapsulates.

Wall Material	a_w	L^*	a^*	b^*	h°	C^*
MD20	0.22 ± 0.00^a	51.01 ± 0.08^a	23.87 ± 0.07^a	7.41 ± 0.06^a	0.30 ± 0.00^a	24.99 ± 0.09^a
MD30	0.29 ± 0.00^b	51.25 ± 0.21^a	25.24 ± 0.05^b	7.94 ± 0.02^b	0.30 ± 0.00^{ab}	26.46 ± 0.05^b
MD40	0.23 ± 0.00^c	46.44 ± 0.14^b	23.78 ± 0.01^a	7.63 ± 0.14^c	0.31 ± 0.01^b	24.97 ± 0.03^a
GA20	0.18 ± 0.00^d	52.21 ± 0.44^a	17.11 ± 0.08^c	2.38 ± 0.05^d	0.14 ± 0.00^c	17.27 ± 0.08^c
GA30	0.28 ± 0.00^e	51.74 ± 0.37^a	20.22 ± 0.09^d	5.46 ± 0.05^e	0.26 ± 0.00^d	20.95 ± 0.10^d
GA40	0.30 ± 0.00^f	47.11 ± 1.20^b	20.11 ± 0.24^d	6.28 ± 0.11^f	0.30 ± 0.00^a	21.07 ± 0.26^{df}
MIX20	0.26 ± 0.00^g	59.61 ± 0.23^c	19.65 ± 0.09^e	4.54 ± 0.06^g	0.23 ± 0.00^e	20.17 ± 0.08^e
MIX30	0.17 ± 0.00^h	46.99 ± 0.17^b	20.31 ± 0.07^d	6.71 ± 0.01^h	0.32 ± 0.00^f	21.39 ± 0.07^{fg}
MIX40	0.17 ± 0.00^h	45.79 ± 0.83^b	20.28 ± 0.15^d	7.29 ± 0.03^a	0.35 ± 0.00^g	21.55 ± 0.15^g

Values represent the mean \pm SD of triplicate tests. Values in a column with different superscripts are significantly different ($p \leq 0.05$).

3.3. Morphological Analysis

SEM images of the freeze-dried bilberry extract are presented in Figure 2. Micrographs of the encapsulates show irregular-shaped particles with broken glass or flake-like structures. The same morphology has been reported by several authors and is typical for well-defined freeze-dried powders [24,41–43]. The average particle size reported in these studies was 2–3 μm with a wide distribution, which is also the case for our samples and likely a result of manual homogenisation.

The MD30, MD40, and MIX40 encapsulates were an exception, as they showed caking and formation of a sticky layer on the surface. A similar morphology of maltodextrin encapsulates has been reported by Ezhilarasi et al. [41] and contributed to the hygroscopic nature of fruit extract and the high dextrose equivalent value of maltodextrin. It could also be possible that the maltodextrin encapsulation capacity for the bilberry extract was lower compared to gum Arabic and that at higher extract loadings, some amount of extract was left unencapsulated in maltodextrin, thereby resulting in sticky encapsulates.

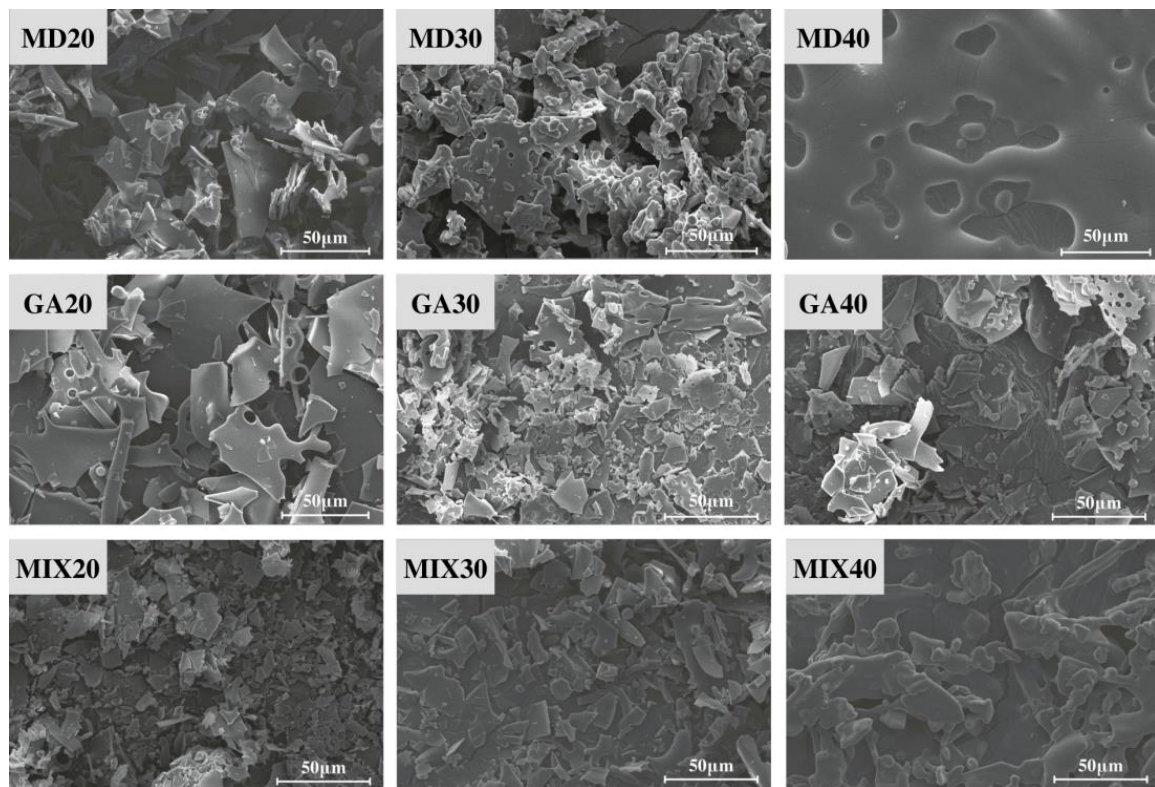


Figure 2. Micrographs of the obtained encapsulates. Specimens shown at 1000× magnification, observed at 10 kV acceleration voltage, 200 µm field width, and 9 mm working distance. Scale as indicated.

3.4. Fourier Transform Infrared Spectroscopy (FTIR)

ATR-FTIR analysis was performed to investigate possible interactions between wall materials and bilberry extract. FTIR spectra of all samples (Figure 3) showed broad bands in the spectral region $3500\text{--}3000\text{ cm}^{-1}$, corresponding to O-H stretching of carbohydrates, carboxylic acids, and residual water [44]. Peaks at $\sim 2900\text{ cm}^{-1}$ and $\sim 2930\text{ cm}^{-1}$ were assigned to symmetric and asymmetric C-H stretching [23,45]. Peaks typical of carbohydrates were observed in pure maltodextrin (Figure 3A) at $\sim 1650\text{ cm}^{-1}$ (C=O stretching), $\sim 1150\text{ cm}^{-1}$ and $\sim 1080\text{ cm}^{-1}$ (C-O stretching), and $\sim 1015\text{ cm}^{-1}$ (angular deformations of =CH and =CH₂ bonds) [23,46]. As expected from the more complex nature of the mixture of glycoproteins and polysaccharides in the acacia harvested gum, characteristic peaks of pure gum Arabic (Figure 3B) were found at $\sim 1600\text{ cm}^{-1}$ (C=O stretching belonging to the amide I band), $\sim 1420\text{ cm}^{-1}$ (C-N stretching and N-H bending vibrations of the amide II band), and $\sim 1020\text{ cm}^{-1}$ (C-O stretching) [47,48]. The spectrum of the mixed wall materials (Figure 3C) showed peaks at $\sim 2900\text{ cm}^{-1}$, $\sim 1600\text{ cm}^{-1}$, $\sim 1420\text{ cm}^{-1}$, $\sim 1150\text{ cm}^{-1}$, and $\sim 1080\text{ cm}^{-1}$, coming from either maltodextrin or gum Arabic. Characteristic peaks of the freeze-dried bilberry extract (Figure 3A–C) at $\sim 1716\text{ cm}^{-1}$, $\sim 1606\text{ cm}^{-1}$, $\sim 1340\text{ cm}^{-1}$, and $\sim 1240\text{ cm}^{-1}$ have been previously associated with the presence of different phenolic compounds [49,50]. The intensive band at $\sim 1716\text{ cm}^{-1}$ is most probably caused by C=O vibrations, and the $\sim 1606\text{ cm}^{-1}$ peak could be due to C=C vibrations typical for aromatic compounds. The peaks at $\sim 1340\text{ cm}^{-1}$ and $\sim 1240\text{ cm}^{-1}$ have been previously assigned to C-O vibrations of phenolics [45,46].

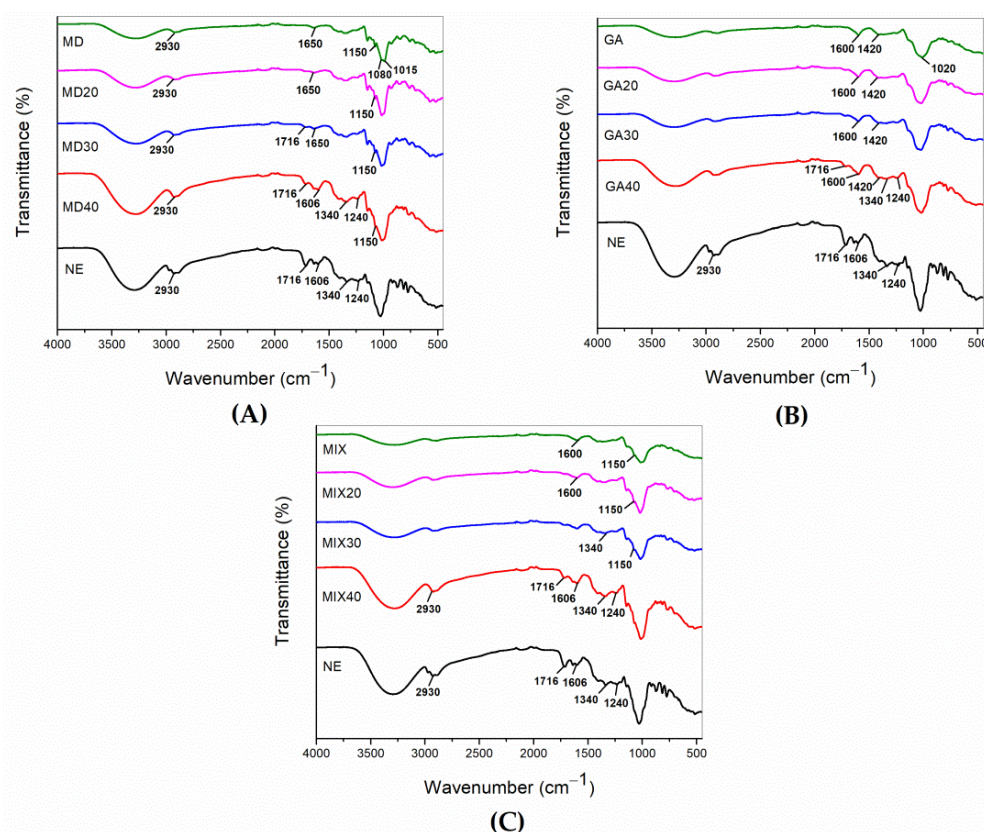


Figure 3. FTIR spectra of the obtained encapsulates, as denoted below individual lines: (A) maltodextrin (MD) encapsulates, (B) gum Arabic (GA) encapsulates, and (C) combination of maltodextrin and gum Arabic (1:1 *w/w*) (MIX) encapsulates, as well as the non-encapsulated, freeze-dried extract (NE).

The spectra of all encapsulates showed peaks related to pure wall material and/or non-encapsulated freeze-dried extract. The absence of newly formed bands and considerable peak shifting suggests that no significant interactions between the extract and wall material have occurred. This absence of covalent interactions is preferable since maintaining the structure and, therefore, the activity of bioactive compounds could contribute to the bioactive stability of encapsulates during storage and digestion [51]. In addition, by looking at the spectra recorded with different proportions of extract and wall material, it can be observed that the increase in extract content was accompanied by a gradual increase in the intensity of the bands between 1700 and 1500 cm⁻¹ and 1000 and 750 cm⁻¹. According to Estupiñan-Amaya et al. [24], a quantitative correlation of the absorption bands with the juice content of Andean blueberry encapsulates indicates that ATR-FTIR measurements could be used as a fast technique to assess the juice content in the encapsulates.

3.5. Total Phenolic Content

Changes in total phenolic content (TPC) of the encapsulates over the course of three-week storage are presented in Figure 4. TPC values after the encapsulation (Day 0) ranged from 10.00 ± 0.03 (MIX20) to 21.78 ± 0.06 mg GAE/g encapsulate (MIX40). The value reported by Casati et al. [52] was 7.69 ± 0.43 mg GAE/g encapsulate for highbush blueberry (*Vaccinium corymbosum*) pulp encapsulated by freeze-drying and with a combination of maltodextrin and gum Arabic (80:20) at a 20% ratio of encapsulating agents. Higher values in our study could be due to the fact that wild blueberries, i.e., bilberries, have higher total phenolic content than cultivated varieties such as highbush blueberries [53].

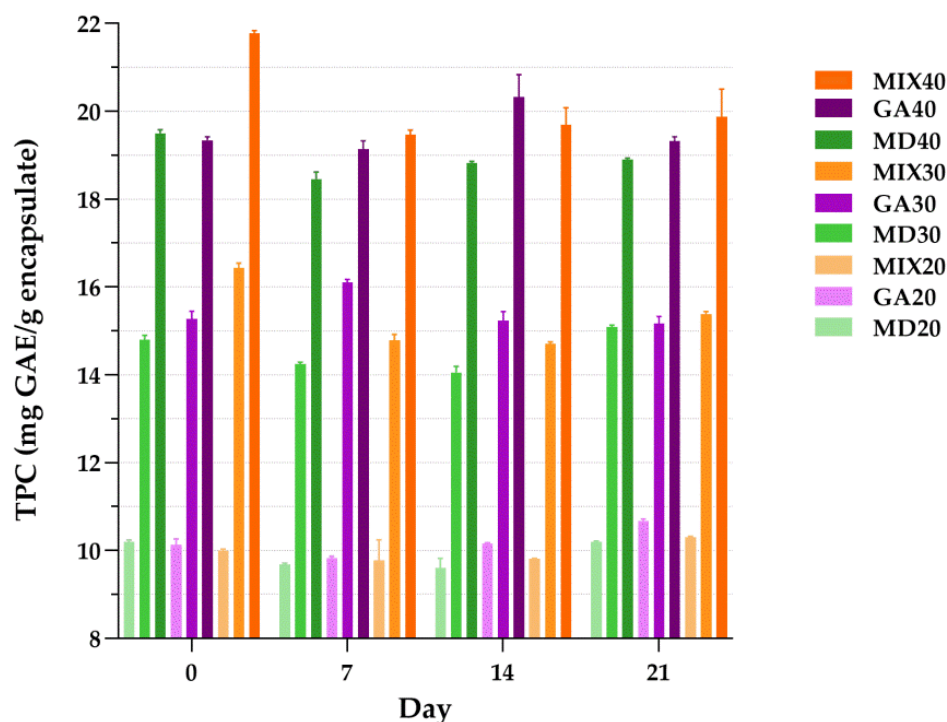


Figure 4. Total phenolic content of the encapsulates during three-week storage at room temperature ($20 \pm 2 \text{ }^\circ\text{C}$) in the dark.

Total phenolic contents of MD20, GA30, and GA40 encapsulates after three-week storage were not significantly ($p > 0.05$) different; the TPC values of MD40, MIX30, and MIX40 were lower compared to Day 0, while in the case of MD30, GA20, and MIX20 encapsulates, TPC seems to have increased ($p \leq 0.05$). Smaller fluctuations in TPC of fruit juices during storage have also been observed by several authors and attributed to the formation of compounds that could possibly react with Folin–Ciocalteu’s reagent [54,55].

3.6. Total Monomeric Anthocyanins Content

Figure 5 shows the total monomeric anthocyanins content (TAC) of the encapsulates over the course of three-week storage. Initial TAC values ranged between 6.87 ± 0.08 (GA20) and 16.10 ± 0.32 mg CGE/g encapsulate (GA40). The TAC of highbush blueberry encapsulates in the previously mentioned study by Casati et al. [52] was 0.74 ± 0.05 mg CGE/g encapsulate. Anthocyanins are present in both the peel and pulp of bilberries, while in blueberries, they are present mainly in the peel. Therefore, the anthocyanin content in bilberries is significantly higher than in blueberries, which could be the reason for the higher values obtained in our study [56].

After some smaller fluctuations, the TAC of most encapsulates was not significantly changed ($p > 0.05$) after three weeks of storage. GA30 and GA40 encapsulates occurred as an exception, where a TAC decrease was observed ($p \leq 0.05$). A decrease in anthocyanin content of the encapsulates during storage has been reported by several authors [39,57,58].

3.7. Antioxidant Capacity

Antioxidant capacity (AOC) of the encapsulates after the encapsulation and during three-week storage is presented in Figure 6. Initial AOC values ranged from 6.10 ± 0.17 (MD20) to 10.43 ± 0.29 mg GAE/g encapsulate (GA40).

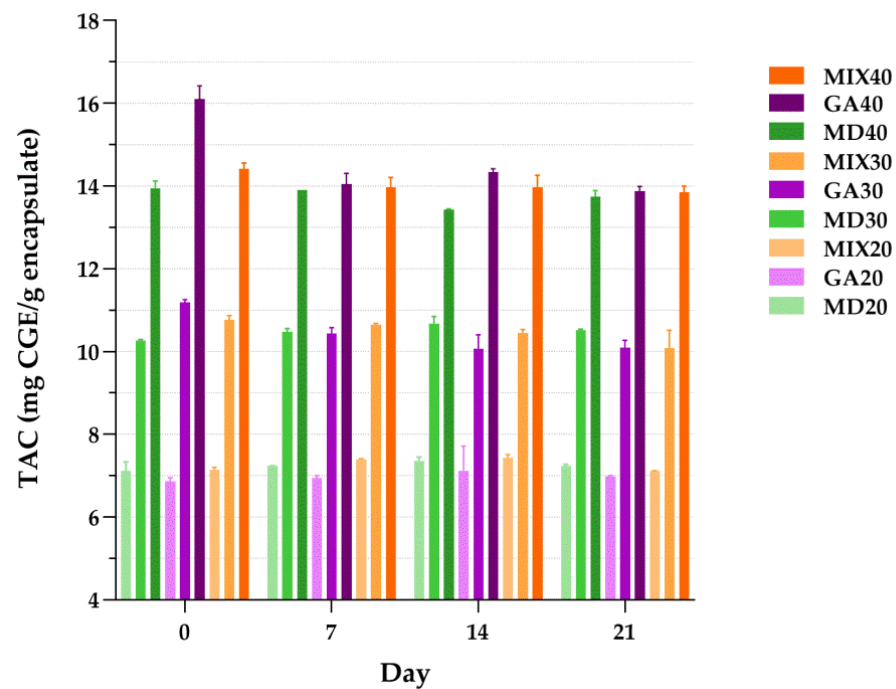


Figure 5. Total monomeric anthocyanins content of the encapsulates during three-week storage at room temperature (20 ± 2 °C) in the dark.

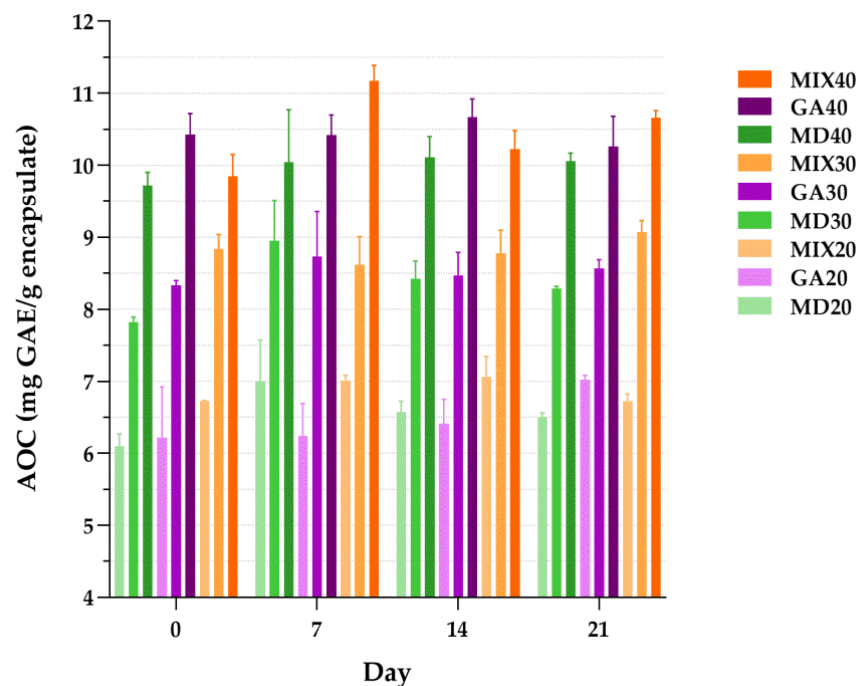


Figure 6. Antioxidant capacity of the encapsulates during three-week storage at room temperature (20 ± 2 °C) in the dark.

At the end of the three-week storage period, AOC values of GA20, GA40, MIX20, and MIX30 encapsulates did not significantly differ ($p > 0.05$) from the initial values, while in the case of GA30, MIX40, and all MD encapsulates, an increase in AOC was observed ($p \leq 0.05$). The increase in antioxidant capacity could be attributed to newly formed polyphenol polymers and phenolic degradation products of anthocyanins [59,60]. According to Pinelo et al. [60], polyphenols show a tendency to undergo polymerisation and form oligomers with larger areas available for charge delocalisation. This occurs only to

a certain extent, after which the increased molecular complexity reduces the availability of hydroxyl groups that react with free radicals, resulting in a decreased antioxidant capacity.

3.8. Thermal Properties

A differential scanning calorimetry analysis was performed to examine possible interactions between bilberry extract and wall materials. The obtained thermograms are shown in Figure 7. The analysis was performed in a temperature range from 0 to 100 °C in order to better distinguish the effects of extract-wall material interactions on non-water-related thermal behaviour within the temperature window that encapsulates would be exposed to during preparation [43]. Pasteurisation, a technique used to preserve various foods, including milk and fruit products, is carried out within this temperature range. Therefore, the potential application of encapsulates as food colourants in such products was investigated.

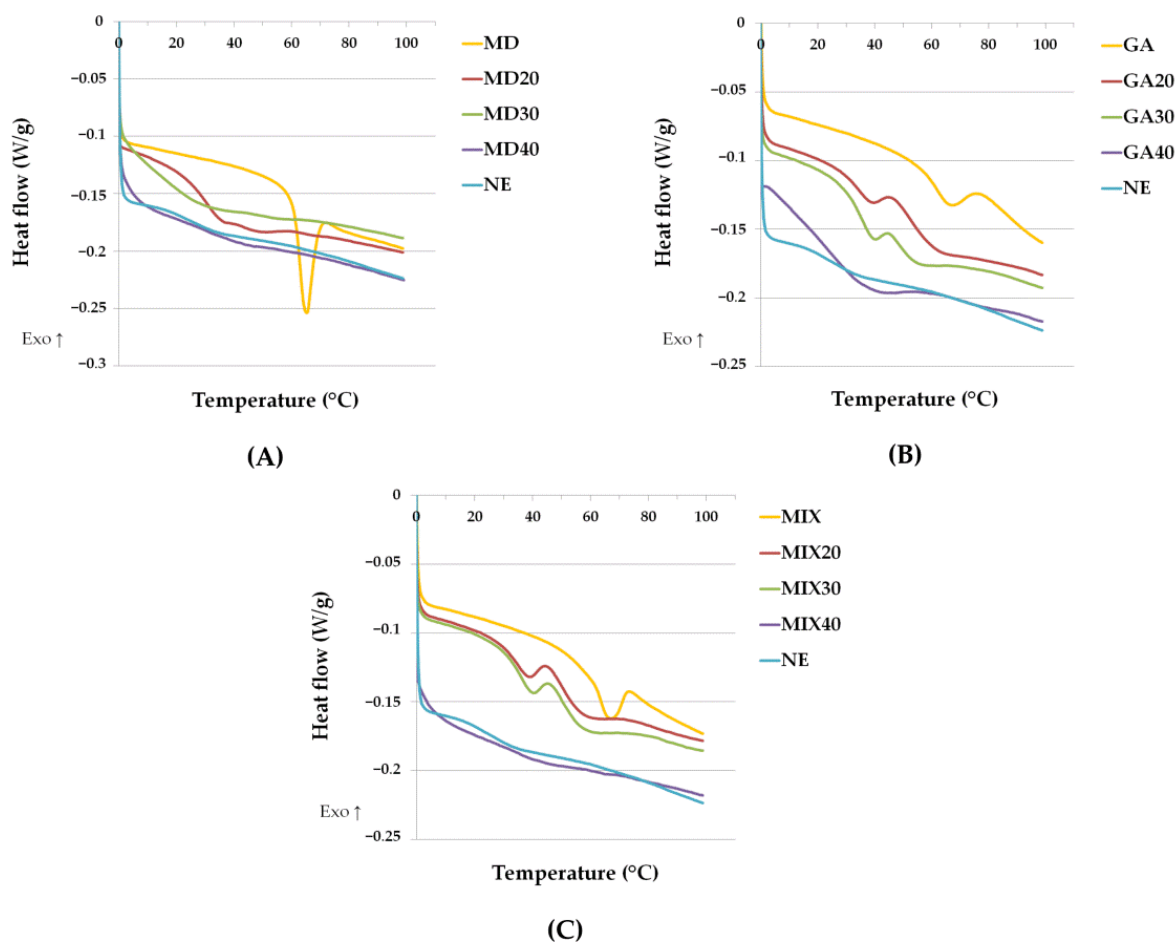


Figure 7. Thermograms of the pure wall materials and obtained encapsulates: (A) maltodextrin (MD) and MD encapsulates, (B) gum Arabic (GA) and GA encapsulates, and (C) combination of maltodextrin and gum Arabic (1:1 *w/w*) (MIX) and MIX encapsulates, as well as the non-encapsulated, freeze-dried extract (NE).

All pure wall materials' thermograms (Figure 7A–C) showed an endothermic peak between 50 and 70 °C. This peak corresponds to a glass transition with enthalpic relaxation and has been observed in several other studies [51,61]. The thermogram of the non-encapsulated freeze-dried extract did not show any prominent peaks.

In the case of encapsulates, certain patterns related to the increase in bilberry solids can be observed, especially in the case of GA (Figure 7B) and MIX powders (Figure 7C). Thermograms of the encapsulates with lower bilberry solids content (20 and 30%) showed more similarities to the thermograms of pure wall materials, but due to the presence

of bilberry solids, the glass transition from an amorphous solid state to a rubbery state occurred at different (lower) temperatures. The reduced glass transition temperature (T_g), compared to pure maltodextrin and gum Arabic, could be a result of increased water absorption due to the hygroscopic nature of bilberry extract. Water acts as a plasticising agent, causing the T_g to decrease. Since the increase in molecular weight increases the T_g , the addition of materials such as maltodextrin or gum Arabic contributes positively to the stability of encapsulated materials [62].

The encapsulates containing 40% bilberry solids, on the other hand, resembled the non-encapsulated, freeze-dried extract more. In the case of the GA40 encapsulate, a thermally induced transition could still be observed, as opposed to MD40 and MIX40 encapsulates. This correlates with the results of morphological analysis (Figure 2), where only MD40 and MIX40 showed caking and the formation of a sticky layer on the surface.

According to Rutz et al. [63], the predominance of the wall material's characteristic thermal profile indicates a more successful encapsulation and better protection of the encapsulated component. This could suggest that, in the case of the lower extract content, the encapsulation capacity of the wall material has not been exceeded, and therefore, it could represent a better choice for designing stable encapsulate formulations.

3.9. Forced Storage Test

Figure 8 shows the total monomeric anthocyanins content (TAC) and antioxidant capacity (AOC) of the encapsulates after the forced storage test. The results show that greater losses of anthocyanins have occurred at higher temperatures (Figure 8A). The average TAC losses at 40 °C were 20%, at 50 °C—56%, and at 60 °C—89%. This behaviour is in line with the findings of Ersus and Yurdagel [58] and Jiménez-Aguilar et al. [64]. On the other hand, the average decrease in AOC at 40 °C was 8%, at 50 °C—18%, and at 60 °C—29%. A less negative impact on the AOC values could be attributed to anthocyanins' degradation products that could also exhibit antioxidant activity [59], which would also explain the slight increase in AOC that was already observed during three-week storage in Figure 6.

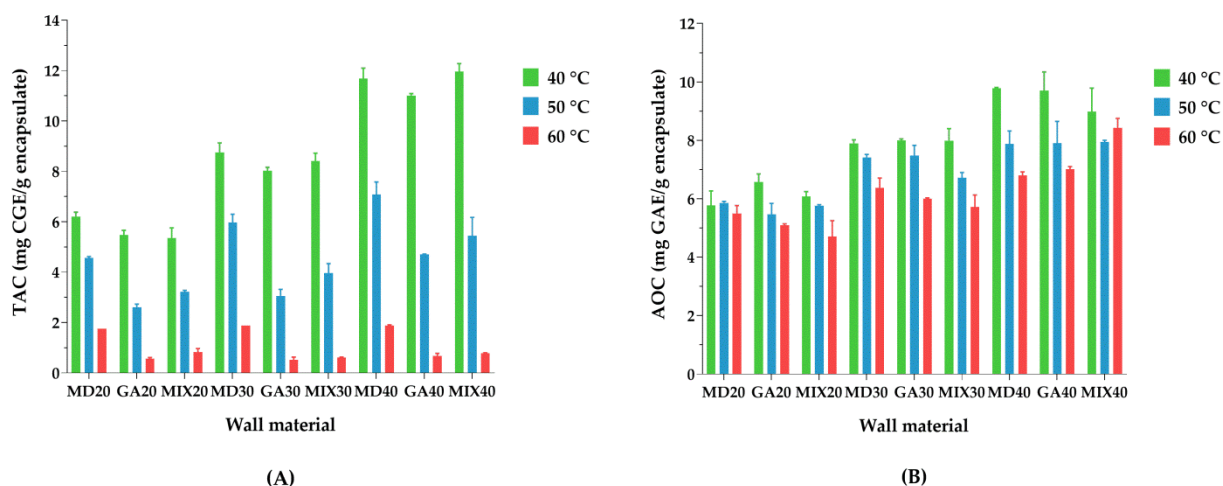


Figure 8. (A) Total monomeric anthocyanins content, and (B) antioxidant capacity of the encapsulates after forced storage test performed at different temperatures.

The use of maltodextrin led to the smallest losses of anthocyanins and antioxidant capacity during the forced storage test. In terms of extract content, the encapsulates prepared with 20% bilberry solids had the lowest losses at 50 and 60 °C. This further confirms that the higher amount of wall material truly offered better protection to the bilberry extract.

4. Conclusions

The results show that freeze-drying with the use of maltodextrin (MD), gum Arabic (GA), and the combination of these two wall materials in a 1:1 (*w/w*) ratio (MIX) is a suitable way to encapsulate bilberry (*V. myrtillus* L.) extract. The obtained encapsulates presented high phenolic and monomeric anthocyanins content after encapsulation and after three weeks of storage, as well as a preserved antioxidant capacity. The use of maltodextrin resulted in the most intensely coloured red encapsulates, regardless of the extract content, while FTIR spectra indicate successful encapsulation. The results of the SEM and DSC analysis, as well as storage tests, show that the higher wall material content resulted in a more successful encapsulation and, therefore, better protection of the bilberry extract. This suggests that there might be certain limitations regarding the encapsulation capacity at higher extract contents, especially with the use of maltodextrin.

Freeze-dried encapsulated bilberry extract shows promise for use as a functional food colouring due to its appealing colour, high polyphenol content, and low water activity. The stability and colour properties also make this an attractive prospect for the alternative protein market, where the selection of natural red colourants is scarce and whose products could also benefit from antioxidant additions. Further studies are required in order to gain a better insight into the effects of different wall materials and extract contents on the encapsulates' properties. The focus of these studies should be on the encapsulates' stability in food matrices and gastrointestinal conditions.

Author Contributions: Conceptualisation, N.P.U. and V.N.; methodology, software, validation, formal analysis, investigation, A.T., L.Š., A.S.-J., S.L., I.G.O.Č., I.P., M.S. and A.B.; resources, N.P.U. and V.N.; data curation, A.S.-J., S.L., I.G.O.Č., I.P., N.P.U. and V.N.; writing—original draft preparation, A.T.; writing—review and editing, A.S.-J., S.L., I.G.O.Č., I.P., N.P.U. and V.N.; visualisation, A.T.; supervision, N.P.U. and V.N.; project administration, N.P.U. and V.N.; funding acquisition, N.P.U. and V.N. All authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by the Ministry of Education, Science and Technological Development of the Republic of Serbia (grant number 451-03-68/2022-14/200116) and by the Slovenian Research Agency through the P4-0121 programme. Furthermore, A.T. would like to thank the Tempus Public Foundation (TPF) for their financial support.

Data Availability Statement: Data are contained within the article.

Conflicts of Interest: The authors declare no conflict of interest. The funders had no role in the design of the study, in the collection, analyses, or interpretation of data, in the writing of the manuscript, or in the decision to publish the results.

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