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Research Article

Retention of bilberry anthocyanins in wood hemicelluloses coated microcapsules under the impact of humidity and packaging conditions

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Abstract

Anthocyanins are attractive natural colorants with recognized health-promoting properties, but their susceptibility to moisture and oxygen limits their shelf-life in powdered formulations. This study investigated the storage stability of spray-dried bilberry anthocyanins microencapsulated with galactoglucomannans (GGM) and glucuronoxylans (GX), two emerging wood hemicelluloses, compared with gum Arabic (GA). The effects of relative humidity (RH; 8 and 33%) and packaging atmosphere (vacuum and sealed conditions) on anthocyanin degradation, color stability, antioxidant retention, and powder physicochemical properties were evaluated over 24 weeks of storage at ambient temperature. Anthocyanin degradation followed first-order kinetics under all storage conditions. Storage at 8% RH provided the highest stability, limiting anthocyanin loss to 17–22% and extending half-life ($t_{1/2}$) values to 62–69 weeks, whereas 33% RH storage resulted in anthocyanin loss of 48–57% and $t_{1/2}$ of 23–30 weeks. Despite slight differences in degradation kinetics, GGM, GX, and GA provided comparable protection against anthocyanin degradation during storage. Vacuum packaging increased $t_{1/2}$ values by 5–10 weeks compared with sealed packaging. Anthocyanin degradation was strongly correlated with polymeric color formation and non-enzymatic browning ($R^2 > 0.905$), indicating progressive pigment transformation during storage. Chromatographic analyses showed that cyanidin-based anthocyanins were more susceptible to degradation under high RH conditions than malvidin derivatives. Increased RH also promoted particle agglomeration, structural collapse, and surface cracking, facilitating oxygen diffusion and accelerating anthocyanin loss. These findings identify moisture control as the primary determinant of anthocyanin stability and highlight wood hemicelluloses as sustainable protective matrices for the development of shelf-stable natural functional powders.

Keywords

Spray-dried microcapsules, natural colorant, storage stability, degradation kinetics, browning index

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1. Introduction

Increasing consumer demand for clean-label products and regulatory restrictions on synthetic additives have promoted the use of natural colorants in food systems. Among naturally occurring pigments, anthocyanins have attracted considerable attention because they provide vibrant red, purple, and blue colors while also exhibiting antioxidant, anti-inflammatory, cardioprotective, and neuroprotective activities (Chen et al., 2023; Zafra-Stone et al., 2007). Anthocyanins are widely distributed in fruits and vegetables, with bilberry (*Vaccinium myrtillus* L.) recognized as one of the richest natural sources of anthocyanins (Chu et al., 2011; Patra et al., 2026). Bilberry anthocyanins comprise a complex mixture of cyanidin, delphinidin, malvidin, peonidin, and petunidin glycosides that contribute to both the characteristic color and functional properties of the fruit (Chu et al., 2011; Lätti et al., 2011).

Despite their desirable bioactivity and coloring capacity, anthocyanins are inherently unstable and readily undergo degradation during processing and storage. Their stability is influenced by several environmental factors, including temperature, oxygen, light, pH, moisture content, and water activity (Chen et al., 2023; Kırca et al., 2007). Anthocyanin degradation during processing and storage involves oxidation, hydration, cleavage, and polymerization reactions that lead to pigment loss, browning development, color deterioration, and decreased antioxidant capacity, thereby reducing the quality and functionality of anthocyanin-rich products (Cai et al., 2022; Chen et al., 2023). To improve anthocyanin stability and facilitate their application in food systems, microencapsulation has emerged as one of the most effective protective strategies. Among available microencapsulation technologies, spray drying is particularly attractive due to its scalability, cost-effectiveness, and ability to produce stable powders suitable for incorporation into food formulations (Ijod et al., 2024). The effectiveness of spray drying largely depends on the properties of the protective materials, which governs encapsulation efficiency, moisture uptake, oxygen diffusion, particle morphology, and the retention of encapsulated compounds (Díaz-Montes, 2023; Halahlah et al., 2023a). Biopolymers such as starches, maltodextrins, gum Arabic (GA), gelatin, casein, and whey proteins are among the most widely used protective materials in spray-dried microencapsulation due to their favorable technological properties and protective effects on encapsulated compounds (Mahdavi et al., 2014). However, the growing demand for sustainable and value-added ingredients has prompted the exploration of novel biopolymers derived from renewable resources, including forest-based polysaccharides.

Our previous studies demonstrated that galactoglucomannans (GGM) and glucuronoxylans (GX), recovered from forest-industry side streams through pressurized hot water extraction (Kilpeläinen et al., 2014), are promising protective materials for spray drying of bilberry anthocyanins (Halahlah et al., 2023c; Halahlah et al., 2025). They achieved encapsulation efficiency of up to 81–86%, comparable to GA. Moreover, powders prepared with GGM and GX exhibited greater phenolic retention and approximately two-fold higher antioxidant activity than powders produced with GA. Their favorable performance is attributed to their high water solubility, thermal stability, low viscosity, and residual lignin content, which is rich in phenolic compounds and confers emulsifying properties that enhance anthocyanin stabilization (Castro-López et al., 2021; Halahlah et al., 2023b; Mikkonen, 2020). However, the practical application of encapsulated anthocyanins depends not only on initial encapsulation efficiency but also on their ability to maintain stability throughout storage. Relative humidity (RH) and oxygen availability are among the most important factors affecting the stability of spray-dried powders. Moisture uptake increases water activity and molecular mobility, promotes particle agglomeration and structural collapse, and accelerates oxidative and non-enzymatic degradation reactions. Oxygen exposure can enhance anthocyanin oxidation, leading to pigment loss and browning development. Changes in powder microstructure during storage may further influence oxygen diffusion pathways and consequently alter degradation rates (Baeza and Chirife, 2021; Otto et al., 2024; Xue et al., 2024). Despite the recognized importance of these factors, limited information is available regarding how storage humidity and packaging conditions affect anthocyanin stability in powders encapsulated with wood hemicelluloses.

In addition, anthocyanins do not degrade uniformly during storage. Individual anthocyanin species exhibit different susceptibilities to degradation depending on their chemical structures, particularly the degree of hydroxylation, methoxylation, and glycosylation of the anthocyanidin backbone (Chen et al., 2023). Although the storage stability of encapsulated anthocyanins has been extensively investigated, most studies have relied on measurements of total anthocyanin content (Ben-Othman et al., 2024; Weber et al., 2017). Information on the storage behavior and relative stability of individual anthocyanin compounds within wood hemicelluloses-based encapsulated powders remains comparatively limited. Such information is essential for understanding degradation mechanisms and designing more effective protective matrices.

This study investigated the effects of RH and packaging atmosphere on the storage stability of spray-dried bilberry anthocyanins microencapsulated with GGM and GX in comparison with GA. Anthocyanin degradation kinetics, color stability, polymeric color formation, browning development, antioxidant retention, individual anthocyanin profiles, water activity, particle size distribution, and powder morphology were evaluated during 24 weeks of storage. By combining physicochemical, chromatographic, and microstructural analyses, this work provides new insights into the mechanisms governing anthocyanin degradation in spray-dried powders and evaluates the potential of wood hemicelluloses as sustainable alternatives to conventional protective materials for functional powder applications.

2. Materials and methods

2.1. Materials and chemicals

GGM powders were obtained from Boreal Bioproducts (Turku, Finland), GX from the Natural Resources Institute Finland (Luke), and GA (Acacia trees) from KitchenLab (Stockholm, Sweden). Commercial bilberry juice (Marjex®, Helsinki, Finland), purchased from a local supermarket (Helsinki, Finland), has a soluble solids content of approximately 10% (w/w), as determined using a digital Atago PAL-1 refractometer (Bellevue, USA).

Chemicals and reagents, used for the determination of total phenolic content, total anthocyanins, and antioxidant activity, were purchased from Merck (Darmstadt, Germany). They included Folin–Ciocalteu reagent, 2,2-diphenyl-1-picrylhydrazyl (DPPH), 2,4,6-tripyridyl-s-triazine (TPTZ), gallic acid, methanol, glacial acetic acid, acetone, sodium carbonate, iron(III) chloride hexahydrate, potassium chloride, sodium acetate, and ethanol. For the quantification of anthocyanins by UHPLC-PDA analysis, UHPLC-grade cyanidin-3-glucoside (Merck, Darmstadt, Germany) was used as the external standard. Milli-Q water was used in all experiments.

2.2. Preparation of spray-dried bilberry anthocyanin microcapsules

The spray-dried anthocyanin powders were prepared as previously described (Halahlah et al., 2025). Briefly, aqueous dispersions (10%, w/w) of GGM, GX, and GA were prepared by dispersing the respective powders in water and stirring at room temperature for approximately 15 h at 600 rpm. Bilberry juice was then added to the dispersions at a weight ratio of 1:1 (w/w) and stirred for an additional 30 min before spray drying. The feed dispersions were spray-dried using a Büchi B-290 mini spray dryer (Büchi Labortechnik AG, Switzerland) equipped with a 0.7 mm nozzle. The inlet/outlet temperatures were 179/80 °C for GGM and GA and 169/69 °C for GX. These spray drying conditions were selected based on our previous study (Halahlah et al., 2025), in which the process was optimized separately for each protective material. The selected conditions provided the highest anthocyanin encapsulation efficiency while maintaining favorable powder properties, particularly low moisture content and water activity, which are critical for long-term storage stability. The aspiration rate and compressed air pressure were maintained at 80% (32 $m^3\ h^{-1}$) and 5–8 bar, respectively. After drying, the resulting powders were collected, and immediately subjected to storage studies.

2.3. Storage stability design

The experimental design for the storage stability study is shown in Figure 1. Three types of spray-dried bilberry anthocyanin microcapsules, prepared using GGM, GX, and GA as protective materials, were evaluated under two storage conditions: RH and packaging.

In the RH experiment, 5 g of powder was spread in Petri dishes (50 × 10 mm) and stored at 8% and 33% RH in airtight chambers maintained using saturated KOH and $Mg(NO_3)_2$ solutions, respectively. These RH levels were selected based on our previous storage study of GGM- and GX-based microcapsules, in which a wider range of RH conditions (11−75% RH) was investigated (unpublished data). In that study, the microcapsules exhibited rapid structural changes and deterioration at RH levels above approximately 55%, with substantial changes occurring within 1–3 weeks. Therefore, RH levels below this threshold were used in the present study to enable a 24-week storage evaluation while maintaining the physical integrity of the microcapsules. In the packaging experiment, 5 g of microcapsules was either sealed in polypropylene bags (SB) or vacuum-sealed in the same type of bags (VB). The bags (10 × 12 cm) were prepared from packaging films consisting of biaxially oriented polypropylene (20 µm) and anti-fog polypropylene (25 µm), which were supplied by FreshServant Oy (Seinäjoki, Finland) and used for modified atmosphere packaging. According to the supplier's specifications, the film had oxygen and carbon dioxide permeabilities of less than 848 and 3389 $cm^3\ m^{-2}\ 24\ h^{-1}$, respectively. The oxygen permeability is relevant to anthocyanin stability because oxygen exposure can promote oxidative degradation of anthocyanins during storage. Approximately 50 g of each microcapsule formulation was stored in zipper bags (20 × 30 cm) under ambient laboratory conditions and served as the control (CB).

All samples were stored at room temperature and protected from light for 24 weeks. At predetermined intervals, one replicate from each treatment was withdrawn for analysis. For the control samples, a small amount of powder was collected at each predetermined sampling interval, after which the bags were immediately resealed and returned to storage. This procedure was intended to simulate repeated handling during practical use and to account for the potential exposure to oxygen and moisture associated with opening and resealing the package.

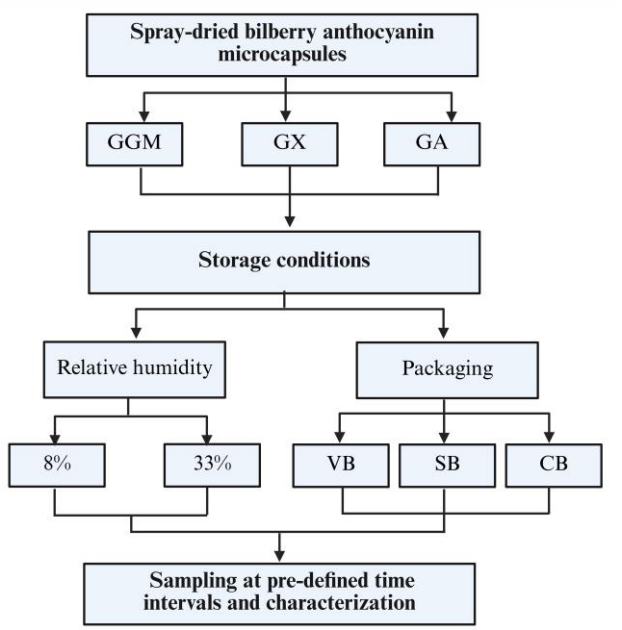

Figure 1. Experimental design for the storage stability study of GGM, GX, and GA bilberry anthocyanin microcapsules. The study comprised two relative humidity conditions (8% and 33%) and three packaging conditions (sealed bags, SB; vacuum-sealed bags, VB; and control bags, CB), evaluated separately during 24 weeks of storage at room temperature.

2.4. Analysis of anthocyanin stability and bioactive properties

2.4.1. Extraction of bioactive compounds

Bioactive compounds were extracted as previously described (Halahlah et al., 2023c). Briefly, powders were treated with a water–ethanol–acetic acid solvent mixture (pH 3) under stirring and ultrasonication for 1 h to disrupt the protective matrix and recover anthocyanins. The resulting extracts were used for determination of total anthocyanin content (TAC), polymeric color, browning index, color density, individual anthocyanin profiles, total phenolic content (TPC), and antioxidant activity.

2.4.2. Total anthocyanins

TAC was determined using the pH differential method (Lee et al., 2005). An aliquot of the extract was diluted in pH 1.0 (0.025 M KCl) and pH 4.5 (0.4 M CH_3COONa) buffers, and absorbance was measured at 510 and 700 nm using a UV–Vis spectrophotometer (UV-1800, Shimadzu, Kyoto, Japan). TAC was expressed as cyanidin-3-glucoside equivalents per gram of microcapsules.

2.4.3. Polymeric color, browning index, and color density

Polymeric color, browning index, and color density were determined using the metabisulfite bleaching method (Giusti and Wrolstad, 2001). Briefly, 1.0 mL of the extract was mixed with either 0.25 mL of 1.0 M $K_2S_2O_5$ or citrate–phosphate buffer for determination of polymeric color and color density/browning index, respectively. After equilibration for 20 min, absorbance was measured using a UV–Vis spectrophotometer (UV-1800, Shimadzu, Kyoto, Japan). Color density, polymeric color, polymeric color index, and browning index were calculated using Eqs. (1)–(4) (Jiang et al., 2019b; Wrolstad et al., 2005). In these equations, A represents the absorbance measured at the specified wavelength and DF is the dilution factor. For Eqs. (1) and (4), absorbance values were obtained from samples mixed with citrate–phosphate buffer, whereas for Eq. (2), absorbance values were obtained from samples mixed with $K_2S_2O_5$.

$$\text{Color density} = [(A_{420nm} - A_{700nm}) + (A_{520nm} - A_{700nm})] \text{ x DF} \quad \text{(Eq. 1)}$$

$$\text{Polymeric color} = [(A_{420nm} - A_{700nm}) + (A_{520nm} - A_{700nm})] \text{ x DF} \quad \text{(Eq. 2)}$$

$$\text{Polymeric color index} = \left(\frac{\text{Polymeric color}}{\text{Color density}}\right) \text{ x } 100 \quad \text{(Eq. 3)}$$

$$\text{Browning index} = (A_{420nm}/A_{520nm}) \quad \text{(Eq. 4)}$$

2.4.4. Degradation kinetics of anthocyanins

The degradation kinetics of anthocyanins were calculated using first-order reaction rate constants (K) from Eq. 5 and half-life ($t_{1/2}$) from Eq. 6, as follows:

$$\ln(C_t / C_0) = -K * t \quad \text{(Eq. 5)}$$

$$t_{1/2} = -\frac{\ln 0.5}{K} \quad \text{(Eq. 6)}$$

Where C_0 is the initial TAC in the microcapsules (day zero), C_t is the TAC at a certain time t (week) at room temperature, K is the first-order degradation kinetic constant (per week), and $t_{1/2}$ is the half-life (week) which is defined as the time required to achieve an anthocyanin degradation of 50% compared to initial concentration (Mercali et al., 2013).

2.4.5. Individual anthocyanins by liquid chromatography

Individual anthocyanins were identified by ultrahigh performance liquid chromatography/mass spectrometry (UHPLC-QTOF-MS) using a Waters HSS T3 C18 column (150×2.1 mm, 1.8 µm; Waters, Milford, MA, USA). 1 mL of the extract was filtered through a 0.20 µm PTFE syringe filter into a UHPLC vial and injected into the chromatographic system. Compound identification was based on mass spectra, retention times, UV–Vis spectra of the standard compounds, and published literature data. Quantification was performed using a reversed-phase UHPLC-PDA method previously developed (Kylli et al., 2010; Obón et al., 2011) and validated by our laboratory. All individual anthocyanins were grouped into one group and quantified as cyanidin-3-glucoside equivalent.

2.4.6. Total phenolic content and antioxidant activity

Total phenolic content (TPC) was determined by the Folin-Ciocalteu method (Grigorakis et al., 2020). Briefly, 0.02 mL of extract was mixed with Folin–Ciocalteu reagent (0.05 mL), Milli-Q water (0.78 mL), and 20% sodium carbonate (0.15 mL), incubated in the dark for 60 min, and the absorbance was measured at 750 nm using a UV–Vis spectrophotometer (UV-1800, Shimadzu, Kyoto, Japan). Results were quantified against a gallic acid calibration curve (0.05–0.5 mg mL^{-1}, $R^2 = 0.990$) and expressed as mg gallic acid equivalents (GAE) g^{-1} powder.

Antioxidant activity was determined by DPPH radical scavenging and ferric reducing antioxidant power (FRAP) assays according to Arnous et al. (2001). For the DPPH assay, 0.025 mL of extract was mixed with 0.975 mL of DPPH solution, incubated in the dark for 60 min, and analyzed at 515 nm. For the FRAP assay, 0.05 mL of extract was mixed with 0.95 mL of FRAP reagent, incubated at 37 °C for 30 min, and analyzed at 593 nm. Absorbance was measured using a UV–Vis spectrophotometer (UV-1800, Shimadzu, Kyoto, Japan). Results were quantified using Trolox calibration curves (200–2000 µmol, $R^2 = 0.991$ for DPPH; 100–2000 µmol, $R^2 = 0.990$ for FRAP) and expressed as µmol Trolox equivalents (TE) g^{-1} powder.

2.5. Determination of physicochemical properties of microcapsules

Water activity was measured using a water activity meter (LabMaster, Novasina, Zurich, Switzerland). Particle size and its distribution were analyzed in dry mode using a static light scattering approach (Mastersizer 3000, Malvern Instruments Ltd., Malvern, UK). Particle morphology was observed using a scanning electron microscope with field emission (FESEM, S-4800, Hitachi, Tokyo, Japan). Details of these analyses were reported in our previous study (Halahlah et al., 2023c).

2.6. Experimental design and statistical analysis

Data were analyzed by one-way analysis of variance (ANOVA) followed by Tukey's post hoc test to evaluate differences among storage conditions. Statistical significance was established at $p < 0.05$ using IBM SPSS Statistics software. All analyses were performed in triplicate, and where applicable, results are presented as mean ± standard error. For microstructural observations, one representative image was selected due to the similarity among replications.

3. Results and discussion

3.1. Effect of storage conditions on anthocyanin stability and bioactive properties

3.1.1. Degradation kinetics of anthocyanins and half-life values

As shown in Figure 2, anthocyanin degradation was strongly influenced by storage conditions. The lowest degradation rates were observed at 8% RH, whereas the highest rates were generally observed in the control samples. Under the same storage conditions, the degradation profiles of the GGM, GX, and GA microcapsules were relatively similar, despite being spray-dried at different temperatures. Anthocyanin degradation followed first-order kinetics under all storage conditions (R^2 = 0.906–0.995), in agreement with previous studies on spray-dried anthocyanin-rich powders during storage (Tonon et al., 2010; Zorić et al., 2014). The K and $t_{1/2}$ values are presented in Table 1. Lower K values and higher $t_{1/2}$ values indicate greater anthocyanin stability.

Table 1. Kinetic parameters of anthocyanin degradation (K), anthocyanin loss (%), and half-life ($t_{1/2}$) of microcapsules stored under two RH levels (8% and 33%) and those stored in three packaging conditions including sealed bags (SB), vacuum-sealed bags (VB), and control bags (CB) after 24 weeks of storage.

Microcapsules	Storage conditions	K (week $^{-1}$)	R^2	Anthocyanin loss (%)	$t_{1/2}$ (weeks)
GGM	8% RH	0.010	0.985	17.4	68.6
	33% RH	0.014	0.948	29.0	50.0
	SB	0.019	0.952	37.0	37.3
	VB	0.017	0.952	33.7	41.5
	CB	0.023	0.989	47.8	29.9
GX	8% RH	0.011	0.983	19.1	61.8
	33% RH	0.013	0.971	28.4	52.4
	SB	0.023	0.949	46.2	30.0
	VB	0.017	0.906	36.1	38.8
	CB	0.026	0.969	56.6	26.8
GA	8% RH	0.010	0.974	21.9	66.1
	33% RH	0.015	0.979	30.9	47.1
	SB	0.026	0.934	48.5	26.2
	VB	0.022	0.945	42.6	30.9
	CB	0.030	0.995	53.0	23.3

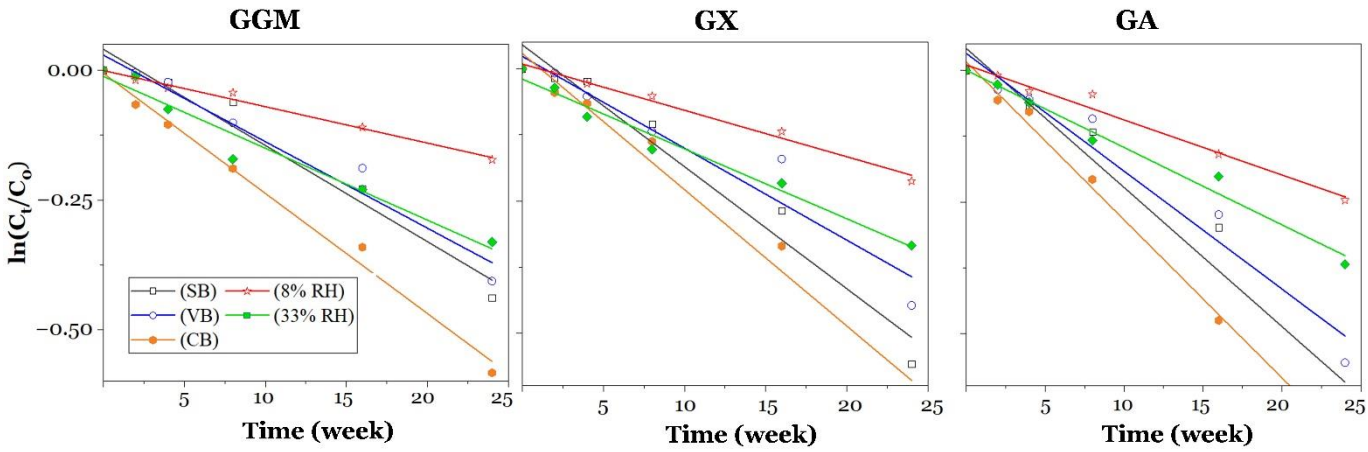

Figure 2. First-order degradation kinetics of anthocyanins in GGM, GX, and GA microcapsules stored under different conditions for 24 weeks, expressed as $\ln(C_t/C_o)$, where C_t is the anthocyanin concentration at a given storage time and C_o is the initial anthocyanin concentration. SB: sealed bags, VB: vacuum-sealed bags, CB: control bags, and RH: relative humidity.

The similar degradation behavior of GGM, GX, and GA microcapsules (Figure 2) indicates that storage conditions had a greater influence on anthocyanin stability than the protective matrix itself. Powders stored at 8% RH exhibited the lowest K values (0.010–0.011 week^{-1}), corresponding to anthocyanin losses of only 17–22%, and the highest $t_{1/2}$ values (62–69 weeks), whereas control powders showed the highest K (0.023–0.030 week^{-1}), resulting in anthocyanin losses of 48–57%, and the shortest $t_{1/2}$ values (23–30 weeks) after 24 weeks of storage. Increasing RH from 8% to 33% increased anthocyanin losses by approximately 10–15% and reduced $t_{1/2}$ by 15–20 weeks. These results confirm that moisture was the primary factor governing anthocyanin stability. Similar to most chemical reactions, anthocyanin degradation rates depend on the mobility of reactants (Fang and

Bhandari, 2011). Elevated water activity increases molecular mobility and promotes the formation of less stable hydrated carbinol pseudo-bases, thereby accelerating anthocyanin degradation. In contrast, under low humidity conditions, water remains strongly bound to polar sites of the protective matrix, limiting degradation reactions (Tonon et al., 2010). Accordingly, powders stored at 8% RH exhibited the highest anthocyanin retention due to their low water activity. In contrast, control powders showed the greatest anthocyanin loss, which may be attributed to their higher water activity during storage (details in Section 3.2.1). The anthocyanin losses observed in the control powders were comparable to those reported for spray-dried cherry anthocyanin powders encapsulated with maltodextrin, which retained only about 50% of their initial anthocyanin content after 25 weeks at 20 °C, highlighting the limited stability of anthocyanins under ambient storage conditions (Zorić et al., 2017).

The relatively low stability of bilberry anthocyanins may also be related to their structural characteristics. Unlike anthocyanins from some fruits such as grapes, bilberry anthocyanins are predominantly non-acylated. Acylation can enhance anthocyanin stability through intramolecular copigmentation, reducing hydration reactions and color loss during storage (Stintzing et al., 2002; Zozio et al., 2011). Consequently, the absence of acyl groups may increase the susceptibility of bilberry anthocyanins to moisture-induced degradation.

Oxygen exclusion improved anthocyanin stability during storage. Vacuum-sealed powders exhibited $t_{1/2}$ values 5–10 weeks longer than those of powders stored in sealed bags, suggesting that oxidative reactions contributed to anthocyanin degradation. The effect of oxygen was more pronounced in GX microcapsules than in GGM and GA microcapsules. Oxygen retained within particle pores and interparticle voids can diffuse into the microcapsule core, promoting anthocyanin oxidation. Oxygen transfer is influenced by the thickness and density of the protective matrix. The slightly greater anthocyanin stability observed in GGM microcapsules may be related to their thicker protective layer (3.50 μm) compared with that of GX microcapsules (3.10 μm) (Halahlah et al., 2024). A thicker protective layer can increase the diffusion distance for oxygen, thereby reducing its access to the encapsulated anthocyanins (Alvarado et al., 2022). In addition, increased moisture content and temperature were also suggested to facilitate the diffusion of oxygen into microcapsules thereby promoting oxidative degradation (Reineccius and Yan, 2016).

Overall, moisture control was the most effective strategy for preserving anthocyanins during storage, whereas oxygen exclusion provided a secondary protective effect. The comparable stability among GGM, GX, and GA microcapsules indicates that wood hemicelluloses can serve as effective and sustainable alternatives to conventional protective materials for long-term stabilization of anthocyanins.

3.1.2. Color stability and pigment transformation

Polymeric color development and non-enzymatic browning can form colourless or brownish adducts which can lead to color loss and also can be used as an indicator for anthocyanins degradation (Jiang et al., 2019b; Muche et al., 2018). As shown in Figure 3, polymeric color increased during storage in a manner similar to the browning index. Both parameters showed a strong negative correlation ($R^2 > 0.900$) with TAC in all microcapsules, indicating that anthocyanin losses during storage were consistently accompanied by increases in polymeric color and browning. Similar relationships between anthocyanin degradation and the development of polymeric pigments and browning were reported in other anthocyanin-rich systems during storage and thermal processing (Jiang et al., 2019b; Martynenko and Chen, 2016; Sinela et al., 2017).

For polymeric color, the greatest increase was observed in the control powders (> 25%), followed by samples stored at 33% RH (11–14%) and those in sealed or vacuum-sealed conditions (13–18%), whereas powders stored at 8% RH showed the smallest increase (< 10%). Comparable increases were reported for strawberry powders, where polymeric color increased by more than 15% after 60 days of storage under ambient conditions (Ngo et al., 2007), and for black carrot anthocyanins, which showed increases exceeding 30% after more than 10 months at 30 °C (Türkyılmaz et al., 2012). Polymerization reactions may result from endogenous enzymes including residual peroxidase activity, and residual polyphenol oxidase activity, that probably remain active after spray drying (Hager et al., 2008; Kader et al., 1999). Another potential mechanism for anthocyanins polymerization could be condensation reactions of anthocyanins with other phenolic compounds such as tannins (Brownmiller et al., 2008). These polymerization and condensation reactions were suggested to increase during storage, with their rates positively influenced by moisture content and storage temperature (Muche et al., 2018; Türkyılmaz et al., 2012).

For browning index, the lowest values were observed in powders stored at 8% RH (~ 0.25), corresponding to an increase of less than 10% during storage. In contrast, the browning index increased by approximately 1.5-fold at 33% RH, 2-fold in vacuum- and sealed-packaged powders, and 2.5-fold in the control powders. Non-enzymatic browning is commonly associated with reactions involving sugars, amino compounds, and proteins under conditions of elevated moisture, temperature, or prolonged storage (Boye, 1999; Garzón and Wrolstad, 2001). The presence of sugars (e.g., from bilberry juice) and their degraded products can also accelerate the anthocyanin degradation, since this reaction rate follows the rate of conversion of sugars to furfural. Typically, furfural is a derivative from aldopentose's, as well as hydroxymethylfurfural, which is a derivative from keto-hexoses (are products resulting from the Maillard reaction) that condense together with the anthocyanins, leading to the formation of compounds with brown coloration (Tsai et al., 2004; Von Elbe and Schawartz, 1996). In addition, the degradation of anthocyanins can result in the formation of chalcone (colorless form of anthocyanins), and the chalcone is further degraded to brown products during improper storage or thermal treatment (Jiang et al., 2019a).

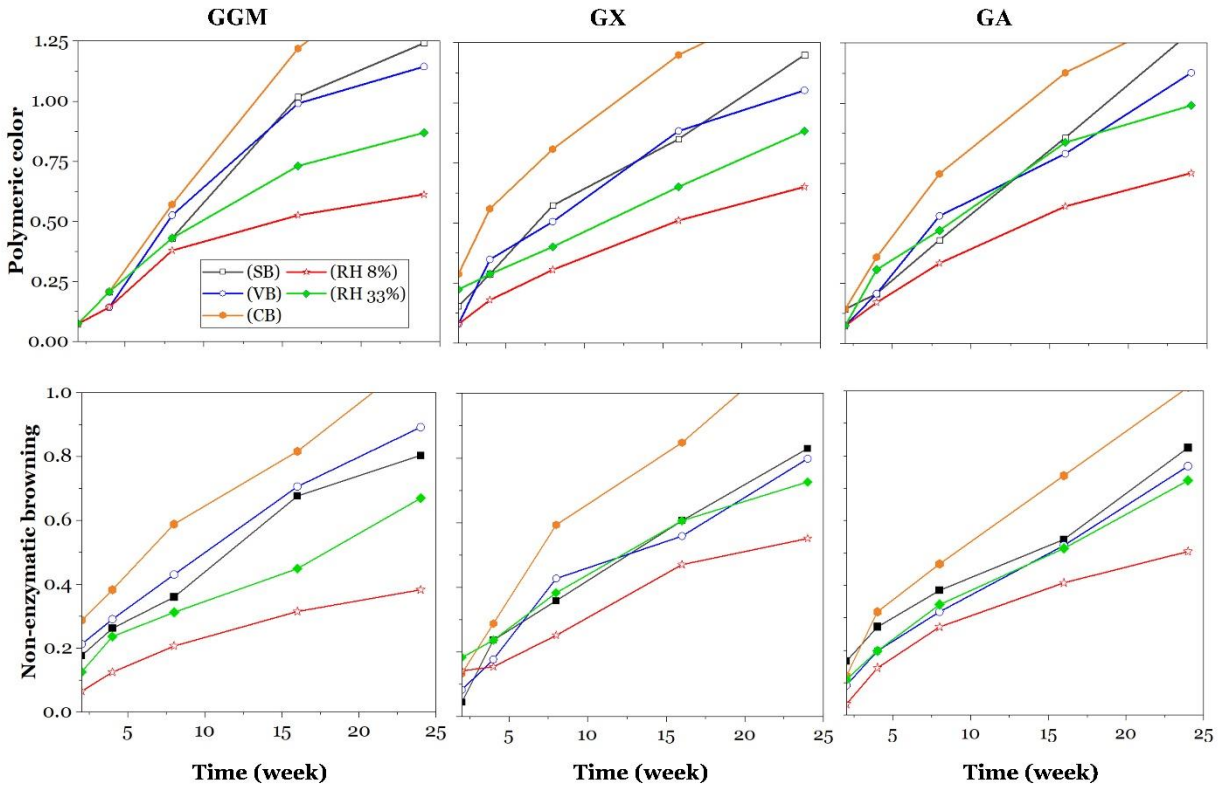

Figure 3. Polymeric color development and non-enzymatic browning index of GGM, GX, and GA microcapsules stored under different conditions for 24 weeks. SB: sealed bags, VB: vacuum-sealed bags, CB: control bags, and RH: relative humidity.

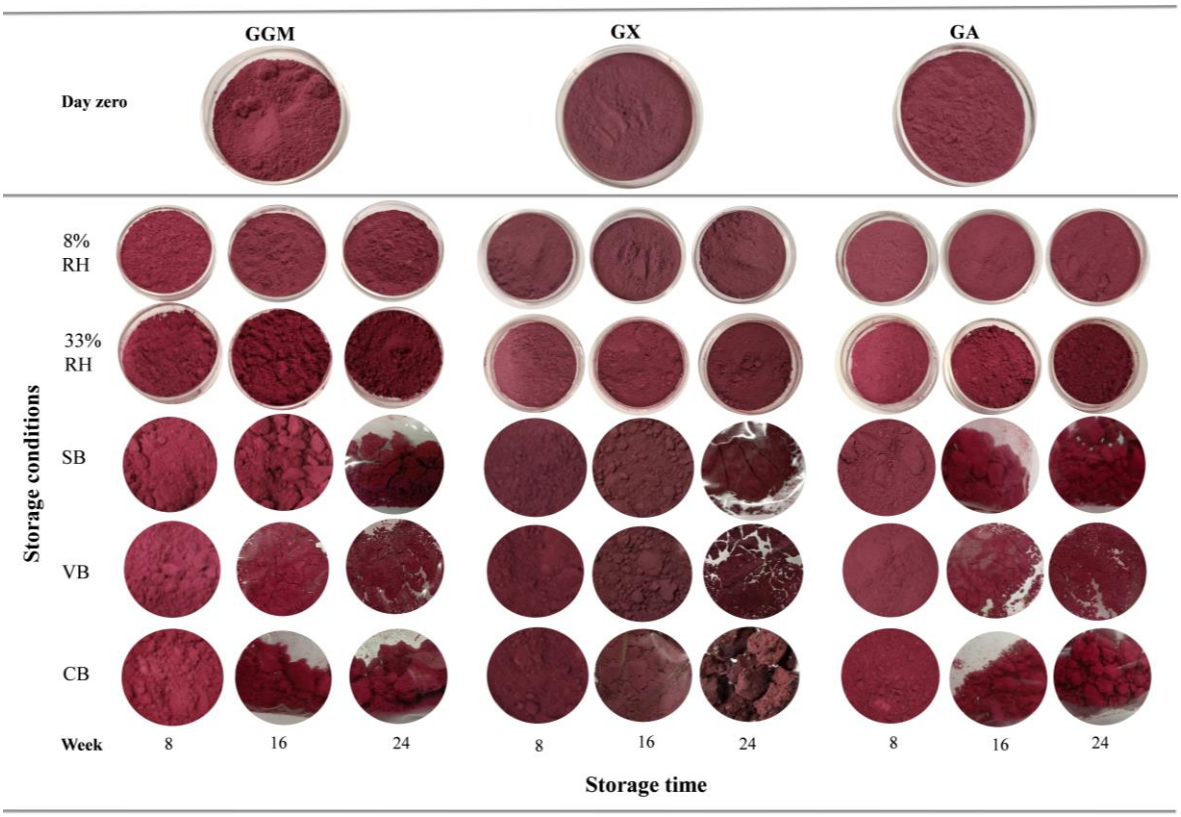

Figure 4. Images of GGM, GX, and GA microcapsules stored under different conditions for 24 weeks. SB: sealed bags; VB: vacuum-sealed bags; CB: control bags; RH: relative humidity. Images were captured using a smartphone camera and provided a qualitative representation of colour changes during storage.

Color which is perceptible by the human eye is the first quality parameter that consumer observes, its assessment is thus essential. Representative images of the microcapsules during 24 weeks of storage are shown in Figure 4. In the current study, the loss of color density of microcapsules was highly correlated with the loss of TAC and the increase of polymeric color/browning index of microcapsules. Typical anthocyanin degradation usually yields faded red dark products and/or pattern of change like brown color development which imply the decay of anthocyanin pigments. Among all storage conditions, microcapsules stored at 8% RH exhibited highest stability of anthocyanins and lowest polymeric color and browning index rates and hence most of their color density were preserved over 24 weeks of storage. Meanwhile, the loss of color density and the evolution of dark brick red pigments in control microcapsules occurred faster after 14 weeks of storage. The accelerated color deterioration in control powders may be attributed to exposure to ambient conditions, as anthocyanins are highly sensitive to temperature and are known to degrade even below 20 °C (Ersus and Yurdagel, 2007; Garzón and Wrolstad, 2001; Gradinaru et al., 2003; Jimenez et al., 2010). Similar color shifts from bright red to brick-red were reported for anthocyanin-rich grape products when storage temperature increased from 4 °C to 25 °C, which was also an indicative for anthocyanin degradation (Amendola et al., 2010).

3.1.3. Stability of individual anthocyanins

The freshly-prepared GGM, GX, and GA microcapsules (day 0) contained TAC values of 8.56, 7.60, and 8.84 mg g^{-1} powder, respectively. Anthocyanins of eight individual compounds were identified in bilberry juice and recovered in the microcapsules after spray drying. Those compounds, tentatively identified by LC-MS include delphinidin, cyanidin petunidin, peonidin, malvidin and their derivatives and were in comparative agreement with previous reports for bilberry anthocyanin profiles (Lätti et al., 2008; Mustafa et al., 2022). As shown in Table 2 and Figure S1 (day 0), cyanidin-based anthocyanins (compounds A3, A4, and A5) were the predominant compounds, accounting for approximately 60% of total anthocyanins, followed by delphinidin-based anthocyanins (compounds A1 and A2, ~ 20%) and malvidin-based anthocyanins (compounds A7, ~ 12%).

Consistent with the TAC degradation trends discussed in Section 3.1.1, all individual anthocyanins decreased during storage, although the extent of degradation varied among anthocyanin classes and storage conditions. For all microcapsules, storage at 8% RH resulted in the highest retention of individual anthocyanins, whereas the greatest losses were observed in the control powders. For example, cyanidin arabinoside (A5), one of the major anthocyanins, retained 84–88% of its initial concentration at 8% RH but only 45–51% in the control powders after 24 weeks. Similar trends were observed for cyanidin galactoside (A3) and cyanidin glucoside (A4). Intermediate losses were observed under 33% RH and in sealed or vacuum-sealed powders, confirming the strong impact of moisture and oxygen on the stability of individual anthocyanins. The degradation patterns of individual anthocyanins were generally similar among GGM, GX, and GA microcapsules, further supporting the conclusion that storage conditions exerted a greater influence on anthocyanin stability than the protective matrix.

The degradation behavior also differed among anthocyanin classes. Cyanidin-based anthocyanins, particularly compounds A4 and A5, were among the least stable compounds and exhibited losses exceeding 35% under 33% RH conditions. Delphinidin-based anthocyanins showed similar degradation patterns, whereas malvidin galactoside (A7) consistently exhibited greater stability under all storage conditions. For example, under control conditions, cyanidin arabinoside retained only 45–51% of its initial concentration, while malvidin galactoside retained approximately 51–53%. Previous studies have shown that cyanidin- and delphinidin-based anthocyanins are less stable during processing and storage due to the presence of hydroxyl (OH) groups on the B-ring, particularly at the 3′ position, compared with the methoxy (OCH_3) substitution found in other anthocyanins (Brooks and Celli, 2019; Xu, 2013). For example, malvidin-based anthocyanins, which contains OCH_3 groups at the 3′ and 5′ positions, exhibited greater stability than cyanidin-based anthocyanins across all microcapsules. These observations are consistent with previous studies reporting accelerated degradation of cyanidin-rich anthocyanins from *Hibiscus sabdariffa* L. flowers and grapes at increased water contents and water activity levels (Garzón and Wrolstad, 2001; Gradinaru et al., 2003; Lai et al., 2009).

Additional evidence of anthocyanin degradation was observed in the chromatograms of GGM and GX control powders after 24 weeks of storage (Figure S1), where new peaks appeared. Similar observations were reported for cyanidin-rich anthocyanins stored at temperatures of 4−37 °C (Sinela et al., 2017). As anthocyanins are glycosylated anthocyanidins, these peaks could be attributed to the hydrolysis of the 3-glycoside structure (Dias et al., 2020). Anthocyanins were also reported to decompose into phloroglucinol aldehyde and benzoic acid derivatives such as syringic acid and 4-hydroxybenzoic acid (de Villiers et al., 2011). Moreover, protocatechuic acid and gallic acid are known degradation products of cyanidin and delphinidin, respectively (Kern et al., 2007). These structural changes can directly influence the stability of anthocyanins and the functionality of these compounds. Further studies are needed to identify unknown degraded compounds and to elucidate the mechanisms involved in the formation of anthocyanin degradation derivatives.

Overall, the results indicate that low water activity and limited oxygen exposure are critical for preserving both the concentration and composition of individual anthocyanins during storage.

3.1.4. Retention of phenolics and antioxidant activities

As shown in Figure 5, all microcapsules showed a gradual decline in TPC and antioxidant activities (DPPH and FRAP) during storage, with the extent of loss strongly dependent on storage conditions. The retention of TPC and antioxidant activity followed the order: 8% RH > 33% RH, VB, SB > CB, which was consistent with the retention of TAC (Section 3.1.1). Powders stored at 8% RH retained more than 85% of their initial TPC and antioxidant activity, whereas control powders exhibited the greatest losses, with antioxidant activity decreasing to approximately 45–75% of the initial values, depending on the microcapsule type and assay. Similar reductions in TPC and antioxidant capacity during prolonged storage were reported for spray-dried anthocyanin-rich powders from açaí and blueberry stored under ambient conditions (Jiménez-Aguilar et al., 2011; Tonon et al., 2009).

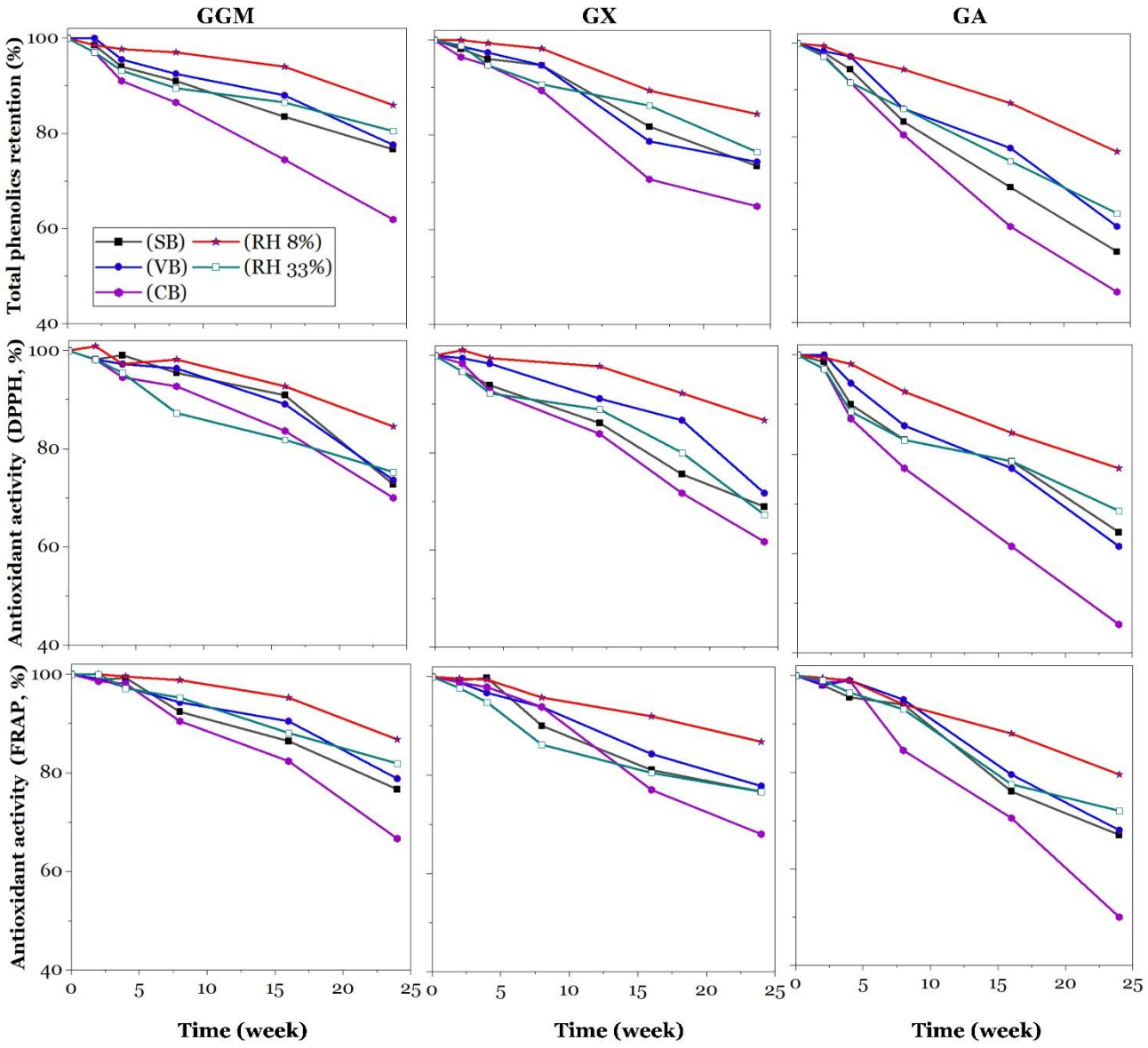

Figure 5. Retention of total phenolic content and antioxidant activities, including DPPH radical scavenging activity and ferric reducing antioxidant power (FRAP), of GGM, GX, and GA microcapsules stored under different conditions for 24 weeks. SB: sealed bags; VB: vacuum-sealed bags; CB: control bags; RH: relative humidity.

A slight increase or fluctuation in antioxidant activity was observed during the first 4–8 weeks of storage, particularly for GGM and GX microcapsules, before a gradual decline occurred. Similar fluctuations in antioxidant capacity were reported for processed black raspberry anthocyanins stored at 25 °C for 24 weeks (Hager et al., 2008). This behavior may be attributed to the hydrolysis and/or degradation of phenolic compounds from bilberry juice, leading to the formation of new compounds with antioxidant activity (Atrooz, 2008; Howard et al., 2010; Jiménez-Aguilar et al., 2011).

Table 2. Changes in individual and total anthocyanin contents of GGM, GX, and GA microcapsules after 24 weeks of storage relative to day 0. Results are expressed as mg g^{-1} dry weight.

Microcapsules	Peak	Tentative compounds	Day 0	Week 24				
				8% RH	33% RH	SB	VB	CB
GGM	A1	Delphinidin galactoside	1.02	0.87	0.72	0.61	0.62	0.51
	A2	Delphinidin arabinoside	0.58	0.48	0.41	0.35	0.37	0.29
	A3	Cyanidin galactoside	1.54	1.33	1.08	0.93	0.96	0.77
	A4	Cyanidin glucoside	1.69	1.44	1.10	1.05	1.07	0.85
	A5	Cyanidin arabinoside	1.82	1.60	1.16	1.09	1.09	0.84
	A6	Petunidin galactoside	0.57	0.49	0.40	0.34	0.36	0.29
	A7	Malvidin galactoside	0.94	0.83	0.68	0.59	0.60	0.47
	A8	Peonidin glucoside	0.39	0.33	0.28	0.24	0.25	0.21
	Total		**8.56 ± 0.55^{f}**	**7.24 ± 0.34^{d}**	**5.83 ± 0.39^{c}**	**5.20 ± 0.62^{b}**	**5.31 ± 0.34^{b}**	**4.22 ± 0.45^{a}**
GX	A1	Delphinidin galactoside	0.96	0.82	0.67	0.58	0.61	0.48
	A2	Delphinidin arabinoside	0.61	0.52	0.43	0.37	0.39	0.31
	A3	Cyanidin galactoside	1.67	1.42	1.17	1.00	1.05	0.84
	A4	Cyanidin glucoside	1.40	1.19	0.88	0.84	0.88	0.70
	A5	Cyanidin arabinoside	1.38	1.17	0.83	0.85	0.87	0.62
	A6	Petunidin galactoside	0.45	0.38	0.31	0.27	0.28	0.22
	A7	Malvidin galactoside	0.72	0.63	0.50	0.45	0.45	0.37
	A8	Peonidin glucoside	0.40	0.34	0.28	0.24	0.25	0.20
	Total		**7.60 ± 0.27^{d}**	**6.18 ± 0.48^{c}**	**5.09 ± 0.35^{b}**	**4.61 ± 0.42^{b}**	**4.79 ± 0.36^{b}**	**3.75 ± 0.43^{a}**
GA	A1	Delphinidin galactoside	1.05	0.90	0.74	0.63	0.66	0.53
	A2	Delphinidin arabinoside	0.67	0.57	0.47	0.40	0.42	0.33
	A3	Cyanidin galactoside	1.72	1.46	1.20	1.03	1.08	0.86
	A4	Cyanidin glucoside	1.62	1.38	1.02	0.97	1.02	0.81
	A5	Cyanidin arabinoside	1.85	1.54	1.17	1.11	1.11	0.93
	A6	Petunidin galactoside	0.59	0.50	0.41	0.35	0.37	0.29
	A7	Malvidin galactoside	0.91	0.77	0.66	0.57	0.57	0.46
	A8	Peonidin glucoside	0.43	0.37	0.30	0.26	0.25	0.24
	Total		**8.84 ± 0.34^{f}**	**7.32 ± 0.45^{d}**	**5.97 ± 0.53^{c}**	**5.33 ± 0.47^{b}**	**5.49 ± 0.38^{b}**	**4.44 ± 0.28^{a}**

The data of total anthocyanins are shown in means ± standard errors (n = 3) and means with different letters in the same row indicate significant differences at p < 0.05. SB: sealed bags, VB: vacuumed bags, CB: control bags, RH: relative humidity.

After 24 weeks, GA microcapsules showed approximately 15–20% lower retention of TPC and antioxidant activity than GGM and GX microcapsules, particularly under sealed, vacuum-sealed, and control storage conditions. This can be explained by the fact that GGM and GX themselves naturally contain proportions of lignin compounds which add additional phenolics to their microcapsules (Halahlah et al., 2023c). Anthocyanins are generally more susceptible to degradation than other phenolic compounds because of their highly reactive flavylium cation structure, which is prone to nucleophilic attack in the presence of moisture (Enaru et al., 2021; Sadilova et al., 2007). Therefore, the additional phenolic constituents in GGM and GX microcapsules may have partially compensated for anthocyanin losses, contributing to the higher retention of antioxidant activity during storage.

3.2. Effect of storage conditions on physicochemical properties

3.2.1. Water activity

Water activity is an important indicator of storage stability, as values below 0.30 generally inhibit microbial growth and slow chemical deterioration (Beuchat et al., 2013; Lavelli and Vantaggi, 2009). From Table 3, except for powders stored at 8% RH, which showed a slight decrease in water activity, all other samples exhibited increasing water activity over time. The greatest increase was observed in the control powders, where water activity rose from approximately 0.20 to 0.44 after 24 weeks, indicating substantial moisture uptake from the surrounding environment. Similar increases were reported for spray-dried strawberry anthocyanin powders formulated with maltodextrin during long-term storage (Castaño-Peláez et al., 2022). Powders stored at 33% RH reached a stable water activity of approximately 0.34 after 8 weeks, reflecting equilibrium with the surrounding environment, whereas vacuum-sealed powders showed a smaller increase, likely due to moisture transfer through the packaging. These results demonstrate the hygroscopic nature of the microcapsules and their ability to absorb moisture during storage.

Table 3. Changes in water activity of microcapsules stored under two RH levels (8% and 33%) and those stored in three packaging conditions including sealed bags (SB), vacuum-sealed bags (VB), and control bags (CB) over 24 weeks.

Storage conditions	Time	Microcapsules		
		GGM	GX	GA
	Day 0	0.22 ± 0.01^{b}	0.21 ± 0.01^{b}	0.21 ± 0.02^{b}
8% RH	8	0.2 ± 0.01^{b}	0.18 ± 0.01^{b}	0.19 ± 0.01^{b}
	16	0.18 ± 0.01^{a}	0.16 ± 0.02^{a}	0.18 ± 0.01^{a}
	24	0.16 ± 0.00^{a}	0.15 ± 0.01^{a}	0.16 ± 0.01^{a}
33% RH	8	0.30 ± 0.01^{d}	0.30 ± 0.30^{d}	0.29 ± 0.01^{cd}
	16	0.31 ± 0.02^{d}	0.33 ± 0.01^{d}	0.32 ± 0.00^{d}
	24	0.32 ± 0.01^{d}	0.34 ± 0.03^{d}	0.33 ± 0.02^{d}
SB	8	0.26 ± 0.01^{c}	0.26 ± 0.02^{c}	0.25 ± 0.03^{c}
	16	0.31 ± 0.02^{d}	0.33 ± 0.02^{d}	0.28 ± 0.01^{cd}
	24	0.36 ± 0.01	0.36 ± 0.01	0.34 ± 0.02
VB	8	0.24 ± 0.01^{bc}	0.25 ± 0.01^{bc}	0.24 ± 0.02^{bc}
	16	0.27 ± 0.01^{c}	0.29 ± 0.03^{cd}	0.27 ± 0.02^{c}
	24	0.30 ± 0.02^{d}	0.32 ± 0.00^{d}	0.29 ± 0.01^{cd}
CB	8	0.28 ± 0.01^{cd}	0.28 ± 0.01^{cd}	0.28 ± 0.02^{cd}
	16	0.33 ± 0.02^{d}	0.36 ± 0.02^{d}	0.34 ± 0.02^{d}
	24	0.41 ± 0.01^{e}	0.44 ± 0.03^{e}	0.40 ± 0.02^{e}

The data are shown in means ± standard errors (n = 3), and means with different letters in the same column indicate significant differences at $p < 0.05$.

3.2.2. Particle size

Particle size is an important factor affecting powder stability because it influences porosity, oxygen transfer, chemical oxidation, and the release behavior of encapsulated compounds (Castaño-Peláez et al., 2022). Changes in particle size distribution during storage are presented in Figure 6, while the corresponding D[3,2] and D[4,3] values are summarized in Table S1.

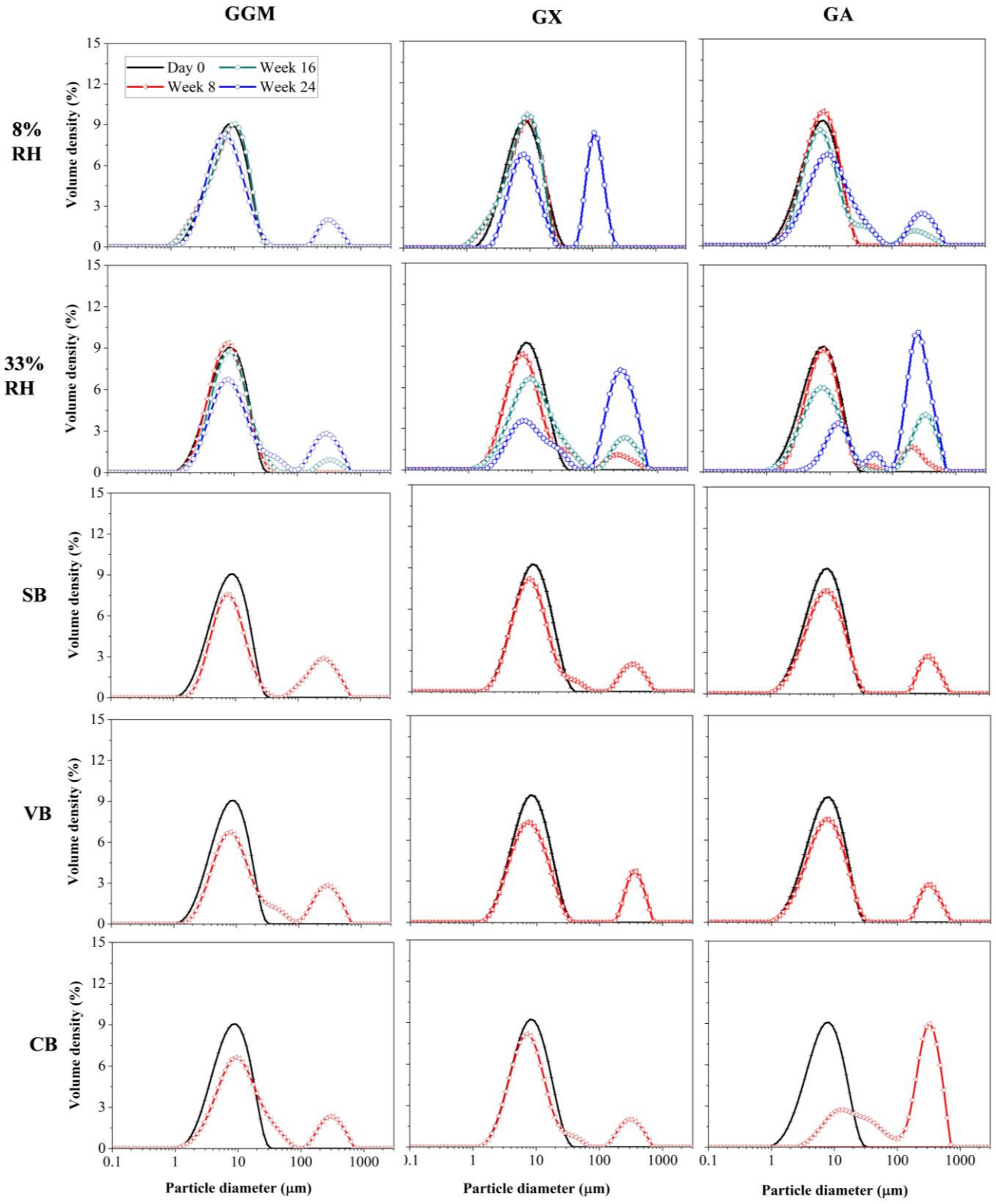

Figure 6. Particle size distributions of microcapsules stored at 8% and 33% RH and in three packaging conditions: sealed bags (SB), vacuum-sealed bags (VB), and control bags (CB), measured at day 0 and after 8, 16, and 24 weeks of storage.

Freshly prepared microcapsules exhibited a narrow unimodal particle size distribution with the main population centered at approximately 5–10 µm. During storage, particle size distributions gradually shifted toward larger particle sizes, indicating particle agglomeration. Powders stored in sealed bags (SB), vacuum-sealed bags (VB), and control bags (CB) developed bimodal particle size distributions by week 8, with a primary peak at 5–10 µm and a secondary peak at approximately 100–300 µm. Similar trends were observed for microcapsules stored at 33% RH after 16 weeks and for those stored at 8% RH after 24 weeks of storage. It was observed that particle agglomeration in microcapsules stored in bags increased significantly after weeks 16 and 24, with many agglomerates exceeding 1000 µm. Consequently, particle size measurements were not further reported for these samples because extensive agglomeration no longer accurately represented the particle population. The GGM, GX, and GA microcapsules exhibited comparable levels of agglomeration, indicating that storage conditions played a more significant role than the protective matrix.

The strongest agglomeration was observed in control powders, which also showed the largest increase in water activity (Section 3.2.1). Increased moisture promotes particle stickiness and enhances interparticle interactions, resulting in the formation of larger aggregates (Bhandari and Ho, 2020). In vacuum- and sealed-bag samples, particle compression during packaging, particular under vacuum sealing, may have further increased particle–particle contact and strengthened van der Waals interactions, thereby contributing to agglomerate formation (Palzer, 2009; Palzer, 2011).

Although the formation of larger particles could theoretically reduce oxygen diffusion because of a lower surface-area-to-volume ratio and a longer diffusion path to the particle core (Amidon et al., 2009; Linke et al., 2020), this effect was insufficient to compensate for the negative impact of moisture uptake. This is evident from the control powders, which exhibited the greatest degree of agglomeration but also the highest anthocyanin degradation rates (Section 3.1.1). Therefore, the observed agglomeration was primarily associated with increased hygroscopicity and moisture uptake rather than improved protection against oxidation.

3.2.3. Microcapsules morphology

The morphology of GGM, GX, and GA microcapsules stored at 8% RH, 33% RH, and in control bags is shown in Figure 7, as these conditions exhibited the highest, intermediate, and lowest anthocyanin retention, respectively.

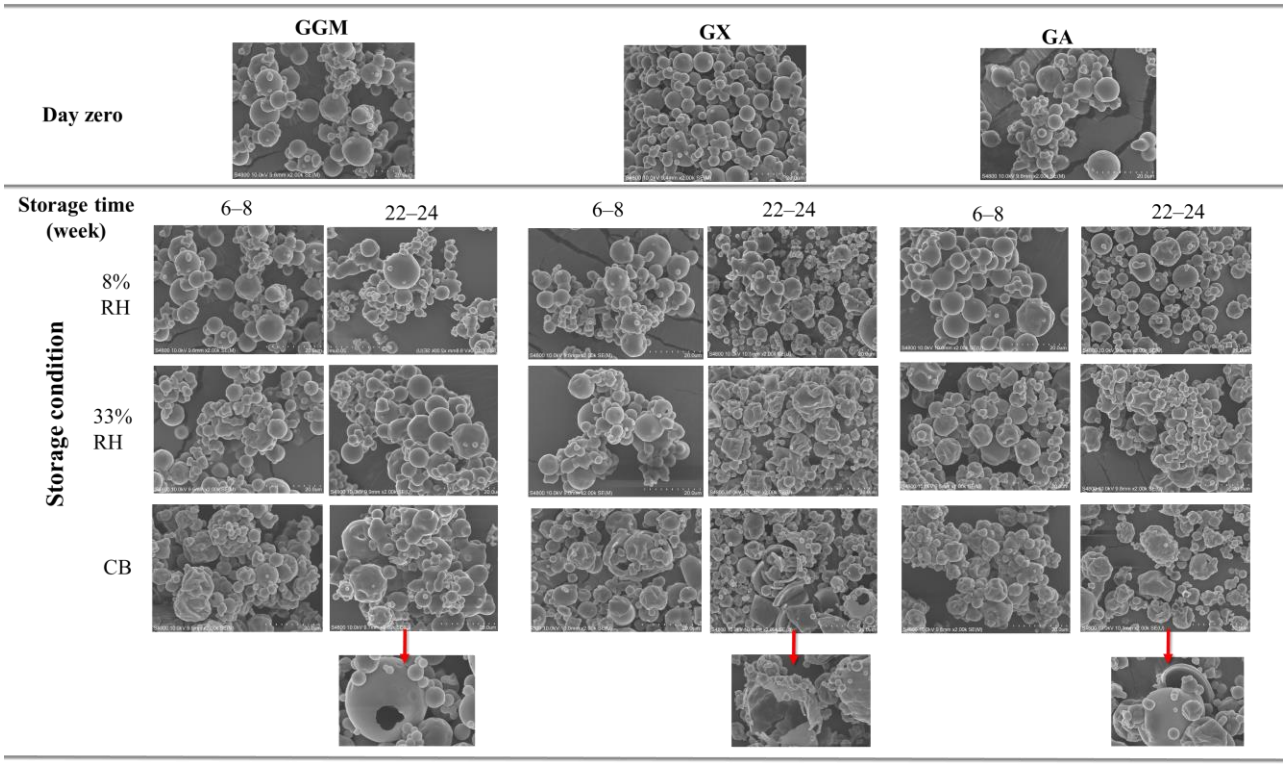

Figure 7. Morphology of microcapsules stored at 8% RH (best anthocyanins retention), 33% RH (second best anthocyanins retention), and CB (lowest anthocyanins retention). CB: control bags, RH: relative humidity. The scale bar of all samples was 20 µm.

Freshly prepared microcapsules were predominantly spherical with smooth surfaces and few dents, wrinkles, or cracks. Microcapsules stored at 8% RH largely retained their original morphology throughout storage, remaining spherical and free of visible pores or cracks, which may limit oxygen penetration and contributed to improved anthocyanin stability (de Barros Fernandes et al., 2014). In contrast, powders stored at 33% RH exhibited increased agglomeration, semi-spherical structures, and

the appearance of surface dents and cavities, likely due to moisture uptake during storage (Santana et al., 2016). The most pronounced morphological deterioration was observed in control powders, which showed collapsed surfaces, cracks, broken structures, and large agglomerates. Such structural defects can facilitate oxygen diffusion into the microcapsules, accelerating anthocyanin degradation, as previously reported for anthocyanin-rich powders during storage even at room temperature (Tonon et al., 2010). These results highlight the importance of low-humidity storage conditions for maintaining microcapsule integrity and extending shelf life.

3.3. Relationship between physicochemical changes and anthocyanin stability

The physicochemical changes observed during storage were closely linked to anthocyanin stability. Increased water activity was associated with greater anthocyanin degradation, polymeric color formation, browning development, and reductions in phenolic content and antioxidant activity, indicating that moisture was the primary factor driving quality deterioration. Moisture uptake also promoted particle agglomeration and morphological changes, including surface dents, cavities, and structural collapse, which may increase oxygen accessibility and further accelerate oxidative degradation. In contrast, powders stored at low RH maintained lower water activity, preserved microcapsule integrity, and exhibited superior retention of anthocyanins and antioxidant functionality. These findings demonstrate that anthocyanin stability is governed by the combined effects of moisture-induced molecular mobility, oxygen exposure, and structural changes within the microcapsule matrix.

4. Conclusion

This study demonstrated that anthocyanin stability was primarily governed by storage conditions rather than the protective matrix. Anthocyanin degradation in GGM, GX, and GA microcapsules followed first-order kinetics and was strongly influenced by moisture uptake and oxygen exposure during storage. Low-humidity storage (8% RH) provided the most favorable conditions, resulting in lower degradation rates, longer $t_{1/2}$ values, and greater retention of anthocyanins, phenolic compounds, antioxidant activity, and color quality. In contrast, storage under ambient conditions promoted significant moisture uptake, increased pigment degradation, and pronounced physical deterioration. Anthocyanin degradation was accompanied by increased polymeric color and browning, reduced antioxidant activity, particle agglomeration, and changes in microcapsule morphology. Among the individual anthocyanins, cyanidin- and delphinidin-based anthocyanins were more susceptible to degradation, whereas malvidin-based anthocyanins showed comparatively greater stability. Overall, the results demonstrate that wood hemicelluloses are effective and sustainable alternatives to GA for the spray-dried microencapsulation of anthocyanins, while their intrinsic phenolic compounds may provide an additional contribution to antioxidant properties during storage. From a practical perspective, bilberry anthocyanin powders should be stored under low relative humidity and with limited oxygen exposure. These conditions could be achieved through packaging systems that minimize moisture and oxygen transmission, such as vacuum packaging or packaging materials with low moisture and oxygen permeability.

A better understanding of anthocyanin degradation products and their formation pathways is needed to elucidate the mechanisms governing anthocyanin stability. Moreover, evaluating interactions between anthocyanins and hemicellulose matrices and assessing stability under realistic food processing conditions and food products will support the development of anthocyanin-enriched functional foods and nutraceutical products.

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Data Availability

The datasets generated during and/or analyzed during the current study are available from the corresponding author on reasonable request.

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Additional information

Conflict of interest

There are no conflicts of interest to declare.

Artificial Intelligence (AI) use

The authors accept full responsibility for the content of the manuscript, including the disclosure of any use of AI.
The authors used ChatGPT for language improvement.

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Author contributions

Abedalghani Halahlah: Conceptualization, Writing - original draft, Formal analysis, Investigation, Methodology
Vieno Piironen: Writing - review and editing, Supervision
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Supplementary materials

Table S1. Particle diameter (µm) of microcapsules stored under two RH levels (8% and 33%) and those stored in three packaging systems including sealed bags (SB), vacuum-sealed bags (VB), and control bags (CB) over a period of 24 weeks. D[4,3]: Volume-based diameter, D[3,2]: Surface area-based diameter.

Storage conditions	Time (week)	Microcapsules					
		GGM		GX		GA	
		D[4,3]	D[3,2]	D[4,3]	D[3,2]	D[4,3]	D[3,2]
	Day 0	8.93	6.14	9.33	6.47	8.16	5.53
SB	8	74.8	8.68	52.8	7.13	8.95	6.24
	16	-	-	-	-	-	-
	24	-	-	-	-	-	-
VB	8	75.3	9.03	81.4	7.62	61.8	6.6
	16	-	-	-	-	-	-
	24	-	-	-	-	-	-
CB	8	64.2	8.92	52.8	11.7	24	211
	16	-	-	-	-	-	-
	24	-	-	-	-	-	-
8% RH	8	9.09	5.74	8.79	5.64	8.43	6.03
	16	9.19	5.88	8.54	5.58	32.4	6.87
	24	49.1	6.82	52.8	11.7	64.2	8.92
33% RH	8	9.15	6.32	9.66	11.6	34.5	7.18
	16	29.2	7.33	63.1	12.5	102	7.97
	24	75.3	9.03	173	17.3	201	38.1

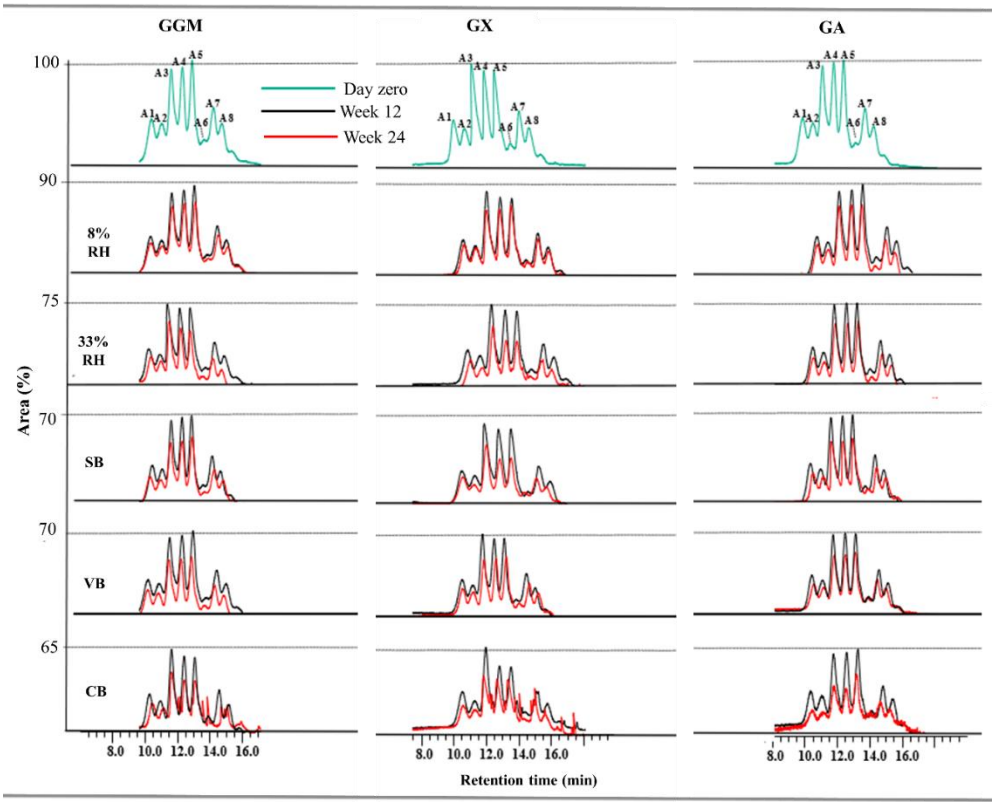

Figure S1. Chromatograms of the changes in individual (compounds A1–A8) and total anthocyanins content of microcapsules stored under two RH levels (8% and 33%) and those stored in three packaging systems including sealed bags (SB), vacuum-sealed bags (VB), and control bags (CB) over a period of 24 weeks.