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FULL PAPER

Coloration Technology

Microencapsulation of Caucasian blueberries (*Vaccinium arctostaphylos* L.) anthocyanins: Colour stability in varying storage conditions

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[Correction added on 05 December 2024, after first online publication: Serpil Girgin's ORCID has been added in this version.]

Abstract

The aim of this study was to investigate the extraction of anthocyanins from the fruits of Caucasian blueberry (*Vaccinium arctostaphylos* L.) and examine the colour stability of anthocyanins encapsulated through spray drying in model systems. The trials were conducted to determine the optimal conditions for anthocyanin extraction using an ultrasonic water bath at temperatures of 25, 50 and 75°C. The ideal anthocyanin extraction occurred at 25°C using 100% acetone in an ultrasonic bath for 15 min at 35 kHz. Anthocyanin concentrations were evaluated using the pH differential method. The extracted anthocyanins were encapsulated through spray drying with maltodextrin as the encapsulating material. The encapsulation efficiency was 80.4 ± 0.20%, and the anthocyanin retention rate was 83.0 ± 0.76%. The coloured capsules were added to model systems with pH values of 4.50, 5.00, 5.50 and 6.00. The colour values (L^* 50.16 ± 0.05, a^* 4.08 ± 0.04, b^* −9.35 ± 0.01, H° 293.57 ± 0.15, C^* 10.20 ± 0.03) were measured during varying storage conditions. The results showed, that colour loss began on the fifth day and this was accelerated with higher temperatures and pH values. It was found that encapsulation significantly preserved the stability of the blue colour and the loss of the blue colour followed first-order reaction kinetics. The study showed that the blue colour of microcapsules was best preserved at pH 5.00 at 25°C. In this case, the measured total colour change (ΔE) was 5.86, the activation energy (E_a) was 37.64 kJ/mol, and the half-life ($t_{1/2}$) was determined to be 19.08 days.

1 | INTRODUCTION

The *Vaccinium* genus, belonging to the Ericaceae family, harbours approximately 450 species worldwide.[1] They are prevalent across various regions in North America, Europe, and Asia.[2] The Black Sea Region of Türkiye is a significant origin for Caucasian blueberry (*Vaccinium arctostaphylos*), bilberry (*Vaccinium myrtillus*), lingonberry (*Vaccinium vitis-idaea*), and bog bilberry (*Vaccinium uliginosum*).[3] The polyphenols, particularly the high natural anthocyanin content, found in *Vaccinium* berries (blueberry, bilberry) and, to some extent, in the leaves, make them an important source of food colourants and pharmaceutical ingredients.[4] Anthocyanins are

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one of the main components of blueberries and are responsible for their red, blue, purple, and black colours.[5] Additionally, the anthocyanin composition in many fruits is distinctive, and anthocyanin profiles are used as a fingerprint to identify plants and detect fruit juice adulteration.[6] The purplish-red colour of *Vaccinium uliginosum* berries results from the red pigment of malvidin 3-glucoside.[6] *Vaccinium myrtillus* berries typically change colour from green to blue by late July and reach full ripeness with a dark blue or black-blue colour.[7,8] The ripening period for *Vaccinium vitis-idaea* L. is in September, and the fruits are carmine-coloured and globular when mature.[9] The most abundant organic acid in lingonberries is citric acid, followed by malic acid and ascorbic acid. Thus, these fruits have a sour taste.[10] *Vaccinium arctostaphylos* fruits are round, flat, or oval in shape, and their flavour can range from sour to sweet.[11]

Anthocyanin biosynthesis in blueberry (*Vaccinium* spp.) fruits begins in the early stages of ripening.[12] The accumulation of anthocyanins continues throughout the ripening process, with the colour gradually darkening as the fruit matures. Blueberry fruits are green and vary in size during the initial 'swelling' stage of fruit development before accumulating a red pigment at the onset of ripening.[13] As the fruits ripen, the amount of anthocyanins increases, causing the colour to shift to blue. Eventually, when the fruits are fully mature, they turn dark blue or nearly black. The increase in anthocyanin content signifies a decrease in the proportion of total phenolic compounds in plants or fruits and an increase in the proportion of anthocyanins during ripening. However, the extent of this increase or decrease can vary depending on the species and variety of the blueberry.[2,12,13]

The Caucasian blueberry is a deciduous shrub with dark blue-purple[14] or purple-black fruits.[15] Known locally as 'Likarpa' in Türkiye, *Vaccinium arctostaphylos* L. is also a part of the natural vegetation in Bulgaria, the Caucasus, Georgia, and northern Iran.[14] These fruits are highly rich in valuable vitamins, antimicrobial and antitumoral compounds, as well as antioxidants like anthocyanin pigments and proanthocyanidins, which are crucial in preventing bacterial growth.[16,17] Anthocyanins are plant pigments that are formed by binding sugars to the anthocyanidin molecule.[18] The binding of sugars such as arabinose, glucose, xylose, galactose, and rhamnose to anthocyanidins is called glycosylation. The number and position of sugar molecules binding to anthocyanidin molecules, along with the type and number of aliphatic or aromatic acids associated with them, lead to the formation of various anthocyanins.[19] The anthocyanidins found in Caucasian blueberry fruits are delphinidin, cyanidin, petunidin, peonidin, and malvidin. These are bound to galactose, glucose, arabinose, and xylose monosaccharides, and in addition, delphinidin and cyanidin are also bound to rhamnose.[20] Karcheva-Bahchevanska et al[21] identified nine anthocyanins in *Vaccinium arctostaphylos* fruits. The most characteristic ones are cyanidin-3-*O*-xyloside, delphinidin-3-*O*-xyloside, malvidin-3-*O*-xyloside, and petunidin-3-*O*-xyloside. Jamshidi et al[22] reported that the level of delphinidin 3-*O*-glucoside in the extract of dried Caucasian blueberry fruits is higher than the levels of petunidin 3-*O*-glucoside and malvidin 3-*O*-glucoside. Lätti et al[20] have identified and confirmed by tandem mass spectrometry (MS/MS) detection a high variety of anthocyanins in *Vaccinium arctostaphylos* fruits from Türkiye, consisting of 19 anthocyanin peaks in a high-performance liquid chromatography diode array dection (HPLC-DAD) profile. Anthocyanins undergo modifications such as hydroxylation, methylation, glycosylation, and acylation. Therefore, the colours they provide are versatile, and their stability is variable. For example, methylation of the B-ring reduces sensitivity to oxidation and stabilises anthocyanins.[23] Blueness increases with an increased number of hydroxyl groups, while redness increases with enhanced methylation in the B-ring.[23] Glycosylation and acylation are other factors affecting the stability of anthocyanins.[19]

Delphinidin is particularly effective in the antioxidant activity of anthocyanins. The antioxidant properties of delphinidin are attributed to the presence of three hydroxyl groups in the B-ring. Pelargonidin, which has one hydroxyl group in the B-ring, is a less effective antioxidant than delphinidin. However, replacing one hydroxyl group with a methoxy or adding a glucoside can reduce antioxidant activity.[19] They possess the ability to absorb oxygen radicals due to their high antioxidant capacity.[24,25] Their content of anthocyanins provides a protective effect against chronic diseases such as cancer, cardiovascular diseases, and diabetes.[26–28] In recent years, natural pigments have been used in meat products for colour preservation, extending shelf life, antioxidant effect, and antimicrobial activity.[29,30] Szpicer et al[31] reported that adding microcapsules of anthocyanins obtained from *Aronia melanocarpa* to a meatball mixture consisting of gluten-free and soy-free analogues at a rate of 2.74% increased the antioxidant potential of the meatballs. A similar study by Su et al[30] observed that microcapsules prepared using raspberry juice and maltodextrin (2:3, w/v) increased the redness in meatballs prepared with 2% or 3% addition, preventing lipid oxidation and colour deterioration, thereby extending the shelf life of the minced meat. As carriers of functional compounds for food products,[32] microcapsules play an important role in the protection and stabilisation of natural food colourants.[33] Additionally, they help preserve flavour and

nutritional value, extending the shelf life of products and offering healthier options to consumers.[33,34]

Anthocyanins, responsible for red, blue, and purple hues in many fruits, are structurally polyphenolic and water-soluble.[35] Despite their potential as food colourants in the industry, they are highly sensitive to light, oxygen, and exposure.[36,37] Therefore, encapsulation techniques serve as an appropriate method to preserve the properties of anthocyanins and enable industrial applications.[38] Spray drying can be defined as microencapsulation within a biopolymer matrix, facilitating rapid encapsulation of the bioactive compound by swiftly removing moisture from the product.[39,40] This process enhances the product's resistance to oxygen, moisture, light, and other factors. Due to its low cost, ease of application, and feasibility for industrial-scale implementation, this method is widely accepted as a prevalent approach for microencapsulation.[39,41,42] Maltodextrin, often employed as a wall material for microencapsulation, is preferred due to its high solubility, low viscosity, low sugar content, and ability to produce colourless solutions. Furthermore, arabic gum functions as an effective polysaccharide in stabilising emulsions and may particularly suit spray drying processes.[43–45]

In this study, anthocyanins were extracted from Caucasian blueberries for the first time comparing different solvents (acetone, ethanol, methanol, distilled water). Microparticles were obtained through spray drying where maltodextrin encapsulated the anthocyanin-rich Caucasian blueberries extract. Stability assessments were conducted at various storage periods and model systems. The gelatine-based model systems, with pH values set at 4.50, 5.00, 5.50 and 6.00, were supplemented with the same amount (15 mL) of blue microcapsules and stored at 25, 35 and 45°C. Colour measurements were taken at regular intervals during storage (twice a day until day 7, and once a day until day 15). Differences in colour values (L^*, a^*, b^*, H°, C^*) were monitored according to changes in temperature and pH.

This study will contribute to the literature by examining the colour loss kinetics of microcapsules obtained from Caucasian blueberries in Türkiye in detail. It is expected to provide guidance on the potential uses and storage processes of Caucasian blueberries microcapsules. This study could make a valuable contribution to the development of microencapsulated products in the food industry.

2 | MATERIALS AND METHODS

2.1 | Materials

The fruits of *Vaccinium arctostaphylos* L. were collected from Kuskunlu Village (40°57′05″N, 38°54′20″E) in Tirebolu district of Giresun province, Türkiye, at an elevation of approximately 220 m above sea level. These samples were stored at −18°C until laboratory analysis. Table 1 shows some physicochemical characteristics of the berries of Caucasian blueberries.

TABLE 1 Physicochemical characteristics of Caucasian blueberries.

Characteristics	Mean values (± standard deviation)
pH	2.80 ± 0.07
Dry matter (g/100 g)	26.21 ± 1.23
Moisture (g/100 g)	73.79 ± 1.21
Ash (g/100 g)	0.32 ± 0.05
Anthocyanin	80.56 ± 1.75 mg/100 g fresh fruit (3.00 ± 0.01 mg/g dry weight)

Note: The experimental analysis was performed in triplicate, and the standard deviation is given.

2.2 | Anthocyanins extraction

The extraction of anthocyanins from Caucasian blueberries was conducted using a modified solid–liquid extraction method, based on the procedure outlined by Liu et al.[43] In brief, initially, Caucasian blueberries preserved at −18°C were thawed at 4°C. Subsequently, 1.3 kg of the sample was placed into a heating container. The sample was mixed in 1:2 ratio with acidified water (0.1% hydrochloric acid) and boiled for 45 min. The resulting solution was then filtered through Whatman No. 1 filter paper and allowed to cool at 4°C in amber bottles until the subsequent processing step.

2.3 | Evaluation of the ability of several solvents to extract anthocyanins from Caucasian blueberries

The Caucasian blueberries, preserved frozen at −18°C, were thawed at 4°C. The sample was adjusted to a solvent ratio of 1:10. Thus, 1 g of Caucasian blueberries was weighed, and different concentrations (100%, 75%, 50%, 25%) of various solvents (acetone, ethanol, methanol, and pure water) were added with a total volume of 10 mL. The mixture was vortexed (Vortex Dragonlab MX-F, China) for 1 min and incubated in an ultrasonic bath (Bandelin-sonorex RK 102 H, Germany) at 35 kHz under different temperature (25, 50, 75°C) for 15 min. Subsequently, the extracts were filtered through Whatman

No.1 filter paper and diluted with buffer solutions (pH 1.00 and 4.50). The absorbance values of the samples were measured using a spectrophotometer (510 and 700 nm).[46] The monomeric anthocyanin content was calculated according to Equation (1).

2.4 | Anthocyanin chelation with iron

A 4.10^{-3} mol/L iron stock solution was prepared by dissolving iron(III) chloride hexahydrate ($FeCl_3 \cdot 6H_2O$) in a sodium acetate buffer with a pH of 4.50. Next, 32 mL of the anthocyanin solution and 48 mL of the iron stock solution, adjusted to pH 3.30 with acetic acid, were taken. A mixture of anthocyanin and iron ions (molar concentration 1:3) was prepared. The initial pink colour of the mixture was turned into blue by adjusting the pH to 5.00.[45]

2.5 | Blue microcapsule production

The maltodextrin to be used as a coating material in a 4:1 ratio was adjusted to a dry matter content of 7% in the emulsion, followed by the addition of a certain amount of water and mixing at 15,000 × *g* for 2 min using a homogeniser (Heidolh Homogeniser SilentCrusher M, Germany). According to the method by Uyan,[47] the slow addition of ferric anthocyanin chelate was carried out, maintaining the stirring process at 15,000 × *g* for 5 min. The production of blue microcapsules was achieved using a spray dryer (Labplant Spray Dryer SD-06, UK) under the following drying conditions: 180°C inlet air temperature, 10 feed pump speed, and 50 m^3/h aspirator speed.

2.6 | Total anthocyanin content

The quantification of total anthocyanin was done using the pH differential assay.[48] Monomeric anthocyanin content was determined based on the following equation.

$$A = (A_{510} - A_{700})\mathrm{pH}_{1.0} - (A_{510} - A_{700})\mathrm{pH}_{4.5} \quad (1)$$

$$\text{Anthocyanins} = \frac{A \times 1000 \times \text{MW} \times \text{DF}}{\mathcal{E} \times L} \quad (2)$$

The amount of anthocyanins was calculated using the molecular weight (MW = 449.2 g/mol) and molar absorption coefficient ($\mathcal{E}$ = 26,900 1/M/cm) of cyanidin-3-glucoside and expressed as milligrams of cyanidin-3-glucoside equivalents per 100 g fresh weight. DF is the dilution factor and *L* the path length (1 cm).

2.7 | Blue microcapsules physical properties

2.7.1 | Powder yield

Powder productivity was calculated by dividing the dry matter of the powder sample obtained using the spray dryer by the dry matter of the initially provided sample.[49]

2.7.2 | Water activity

The water activity of the capsules was measured at 25°C using a water activity meter (Novasina AG, Lachen, Switzerland).

2.7.3 | Moisture

The moisture content was determined according to method AOAC (1925).[50] Samples of 1.5 g were weighed into drying dishes which were previously brought to constant weight and tared. These samples were dried in an oven (Samheung APin, Korea) at 105°C until reaching a constant weight. After the drying process, the samples were transferred to a desiccator, cooled to room temperature, and weighed. The dry matter content was calculated as grams of dry matter per 100 g using Equation (3).

$$M\,(\%) = [(G2 - G)/(G1 - G)] \times 100 \quad (3)$$

where, *M* represents the percentage of dry matter in the sample, *G* is the tare weight of the weighing dish, *G*1 is the total weight of the sample and the tare weight of the weighing dish, and *G*2 is the total weight of the sample and the tare weight of the weighing dish after drying.

2.7.4 | Solubility

For the solubility analysis, 1 g of microcapsules was weighed into a beaker and mixed with 100 mL of water using a homogeniser for 5 min. at 15,000 × *g* to ensure dissolution. The solution was transferred to centrifuge tubes and centrifuged at 3000 × *g* for 5 min. Then, 25 mL was taken from the supernatant and placed into a previously weighed dry matter container. The sample was dried for 5 h. in an oven at 105°C. Solubility was calculated using the differences in weight.[51]

2.7.5 | Colour analysis

The colour parameters *L** (lightness), *a** (redness/greenness), and *b** (yellowness/blueness)[47] of the

microcapsules (3 g of capsules) were measured using a colorimeter (Konica Minolta, Chromameter CR-400 Series, Osaka, Japan), employing a white standard plate. The colorimeter was calibrated with black and white tiles prior to use. Samples were added to clear plastic dishes (1 cm depth and 3.5 cm diameter) and placed against a white tile. The colour measurements were taken using the D65 illuminant and a 2° observation angle, with diffuse illumination. According to the equations, chroma (C^*) (Equation (4)), hue angle (H°) (Equation (5)), colour differences (ΔE^*) (Equation (6)), Δ_a^*, Δ_b^*, Δ_L^* (Equation (7)) were calculated.

$$C^* = \sqrt{(a^*)^2 + (b^*)^2} \tag{4}$$

$$H^\circ = \arctan\frac{b^*}{a^*} \tag{5}$$

$$\Delta E^* = \sqrt{(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2} \tag{6}$$

$$\Delta_a^* = a^*_{\text{standard}} - a^*_{\text{sample}}; \Delta_b^* = b^*_{\text{standard}} - b^*_{\text{sample}}; \Delta_L^* = L^*_{\text{standard}} - L^*_{\text{sample}} \tag{7}$$

2.7.6 | Encapsulation efficiency

The retention rate of anthocyanins, as per the study by Tonon et al,[52] was calculated by dividing the amount of anthocyanins in the dry product after encapsulation by the amount of anthocyanins in the feed mixture supplied to the spray dryer, utilising the following equation.[53]

$$\text{Encapsulation efficiency}(\%) = \frac{\text{Amount of anthocyanins in the final product}(\text{g}/100\text{g})}{\text{Amount of anthocyanins in the feed mixture}(\text{g}/100\text{g})} \times 100 \tag{8}$$

2.7.7 | Bulk density and Carr Index value

The bulk density and Carr Index were determined according to the methods outlined by Mol[48] and Turchiuli et al.[54] Approximately 15 mL of the powdered product, ensuring no air voids, was placed into a container (25 mL measuring cylinder), and the volume (ρ_{bulk}) was recorded. Subsequently, after manually tapping the container 180 times to achieve uniform distribution, the volume for tapped density (ρ_{tapped}) was measured. The Carr Index value used to evaluate the flowability characteristics of the microcapsules was calculated using the following equation.

$$\text{Carr Index} = \left(\rho_{\text{tapped}} - \rho_{\text{bulk}}\right)/\rho_{\text{tapped}} \tag{9}$$

2.8 | Model system preparation

Approximately 2.5 g of sodium benzoate was dissolved in 276 mL of water, followed by the addition of 1150 g of sucrose. The mixture was heated up to 100°C. Subsequently, 50 g of gelatine, soaked in cold water for 5 min and drained, was added to the sucrose mixture, ensuring dissolution without boiling. Model systems were divided into portions of 300 g each, adjusting their pH values to 4.50, 5.00, 5.50, and 6.00. Next, 30 g of microcapsules were dissolved in 60 mL of distilled water, and 15 mL from the solution with a measured pH value of 5.60 was added to each pH-adjusted model system. The coloured model systems were divided into 15 g portions in Petri dishes with a diameter of 6 cm.[48]

2.9 | Storage phase

Differently coloured model systems at different pH values were stored in triplicate at 25, 35, and 45°C in an oven (Samheung APin, Korea). The L^*, a^* and b^* colour values of the model systems were measured using a colorimeter (Konica Minolta Chromameter CR-400 Series), and ΔE, H° and C^* values were calculated. Based on colour loss, the model systems at 45°C were stored for 5 days, those at 35°C and under ultraviolet (UV) for 7 days, and those at 25°C for 15 days. Colour measurements were taken every 12 h for the samples after the seventh day at 25°C. After the seventh day, colour measurements for the samples at 25°C were taken every 24 h.[48]

2.10 | Calculation of kinetic parameters related to blue colour (b^*) change

2.10.1 | Kinetic coefficients calculation

The degree and kinetic coefficients of the degradation reaction of blueness value due to heat treatment were determined by testing zero-order, first-order and second-order reaction kinetics, selecting the model that provided the highest correlation. Following the identification of the suitable model, kinetic coefficients for the change in the b^* value were calculated. Zero-order, first-order and

second-order reaction kinetic models were formulated according to the equations[55]:

$$\text{zero}-\text{order reaction kinetics} -\frac{d[C]}{dt}=k \quad (10)$$

$$\text{first}-\text{order reaction kinetics} -\frac{d[C]}{dt}=k[C] \quad (11)$$

$$\text{second}-\text{order reaction kinetics} -\frac{d[C]}{dt}=k[C]^2 \quad (12)$$

where C is the b^* value at time t, k the reaction rate constant, and t time (in days).

2.10.2 | Calculation of reaction rate constant

After determining that the suitable model for the loss of blueness at the applied temperature and pH values was a first-order reaction, the rate constants (k) were determined according to the following equation. The reaction rate constant was calculated from the slope of the equation.[55]

$$\ln\frac{[C_0]}{[C]}=kt \quad (13)$$

2.10.3 | Half-life ($t_{1/2}$) calculation

The half-life ($t_{1/2}$) is defined as the time required for a 50% loss of any component. For reactions adhering to first-order kinetic models, the half-life was calculated according to the following equation.[55]

$$t_{1/2}=0.693/k \quad (14)$$

2.10.4 | Calculation of D values

The D value is defined as the time required for 90% degradation of a compound in an environment heated at a constant temperature.[55] Accordingly, the D value has been calculated using the following equation.

$$D=\ln 10/k \quad (15)$$

2.10.5 | Activation energy (E_a) calculation

Activation energy (E_a) denotes the dependency level of a reaction on the temperature degree.[56] The E_a value has been calculated using the following formula based on the Arrhenius equation.

$$\ln k=\frac{-E_a}{RT}+\ln k_0 \quad (16)$$

In the equation, E_a represents activation energy (in kJ/mol), k represents the rate constant (1/day), k_0 represents the frequency factor (1/day), R is the gas constant (8.314×10^{-3} kJ/mol/K), and T represents the temperature (in Kelvins).

$$\text{Slope}=-\frac{E_a}{RT} \quad (17)$$



FIGURE 1 Blue microcapsules obtained from Caucasian blueberries.

2.11 | Statistical analysis

Standard deviation (±SD) for all data obtained from the study was calculated using the Microsoft Excel (2010) program. Subsequently, one-way analysis of variance (ANOVA) was employed to compare the mean values. Differences at $p < 0.05$ were considered statistically significant.

3 | RESULTS AND DISCUSSION

3.1 | The properties of the microcapsules

The blue microcapsules obtained from the anthocyanin extract of Caucasian blueberries are illustrated in Figure 1. The physical properties of the blue microcapsules are provided in Table 2.

3.1.1 | Powder yield

The powder yield of the Caucasian blueberries anthocyanin extract, obtained using a spray dryer, was 80.40

TABLE 2 Physical characteristics of blue microcapsules.

Property	Quantity (± standard deviation)
Powder yield (g/100 g)	80.41 ± 0.20
Water activity	0.31 ± 0.01
Dry matter (g/100 g)	93.15 ± 0.05
Moisture (g/100 g)	6.85 ± 0.23
Colour values	L^*: 50.16 ± 0.05 a^*: 4.08 ± 0.04 b^*: −9.35 ± 0.01 H°: 293.57 ± 0.15 C^*: 10.20 ± 0.03
Ash (g/100 g)	9.29 ± 1.44
Encapsulation efficiency (%)	83.00 ± 0.76
Bulk density (g/mL)	0.34 ± 0.11
Carr Index	0.17 ± 0.05
Solubility (g/100 g)	72.83 ± 0.45

Note: L^* (black-white), a^* (red-green), b^* (yellow-blue), H° (hue angle), C* (chroma). Standard deviation of experimental analysis conducted by triplicate.

± 0.2% (g/100 g). The powder yields reported in your study are consistent with similar studies on encapsulating anthocyanins from other sources. For example: Laokuldilok and Kanha[49] encapsulated black rice anthocyanins and reported a powder yield range of 64.07% to 82.16%. Fazaeli et al[51] dried blackberries juice using a spray dryer and achieved a powder yield of 82.00%. We observed that there seems to be a direct correlation between the rise in inlet air temperature and the powder yield. This observation suggests that higher inlet air temperatures during the spray drying process could lead to higher powder yields. This could be due to increased efficiency in removing moisture from the droplets, resulting in better encapsulation of the anthocyanins. Overall, our findings suggest that spray drying is an effective method for encapsulating anthocyanin extracts, with the powder yield being influenced by process parameters such as inlet air temperature. Further optimisation of these parameters could potentially improve the powder yield and overall efficiency of the encapsulation process.

3.1.2 | Moisture and water activity

Blue microcapsules in our investigation had a moisture content of 6.85 ± 0.05% and a water activity of 0.31 ± 0.01. Laokuldilok and Kanha[49] reported that the water activity of capsules made from black rice anthocyanins varied between 0.27 and 0.48, while the moisture content ranged from 2.47% to 6.85%. *Vaccinium corymbosum* L. powder capsules had a moisture content of 3.51%, according to Atacan and Yanık.[53] The water activity of red blueberries powder capsules was found to be 0.29 by Karaca et al.[57] Our blue microcapsules fall within the same range as other products in this context, with water activity and moisture content that are comparable to those reported by Karaca et al[57] and Laokuldilok and Kanha,[49] respectively, at the higher end of the spectrum.

3.1.3 | Encapsulation efficiency

Caucasian blueberries have an anthocyanin encapsulation efficiency of 83.00 ± 0.76%. Açai fruit powder coated with maltodextrin exhibits anthocyanin encapsulation effectiveness ranging from 77.21% to 86.01%,[52] whilst grape pomace capsules demonstrate an anthocyanin retention rate ranging from 88.36% to 97.35%.[52] Previous research has shown that the ratio of the coating material (maltodextrin) to the feed temperature during encapsulation has a significant impact on the anthocyanin encapsulation efficiency.[58]

3.1.4 | Colour values

The colour values of Caucasian blueberries microcapsules were $L^* = 50.16 \pm 0.05$, $a^* = 4.08 \pm 0.04$, $b^* = -9.35 \pm 0.01$, $H^\circ = 293.57 \pm 0.15$ and $C^* = 10.20 \pm 0.03$. In another study, the colour parameters of blue colourant obtained from red cabbage were $L^* = 67.68$, $a^* = -2.15$, $b^* = -10.44$, $H^\circ = 258.36$, and $C^* = 10.65$.[48] The colour parameters of blueberries (*Vaccinium corymbosum* L.) capsules were $L^* = 82.6$, $a^* = 9.09$, and $b^* = 4.64$.[53] The properties of the acquired capsules look similar to the values reported in other investigations, suggesting that the capsules have a blue-purple colouring.

3.1.5 | Bulk density and Carr Index

Bulk density significantly affects the packing and shipping of powdered products. Dried foods with low bulk density often exhibit poor storage stability.[49] Additionally, it has been reported that an increase in bulk density is inversely related to solubility.[51] Caucasian blueberries microcapsules had a bulk density of 0.338 ± 0.11 g/mL and a Carr Index value of 16.7 ± 0.05, in that order. Good flowability is indicated by the results.[54] Fazaeli et al[51] reported that the bulk density of powder obtained from spray-dried mulberry juice ranged from 0.35 to 0.55 g/mL. Tonon et al[52] found that the bulk density of microcapsules made from açai juice ranged from 0.370 to

0.480 g/mL. The Carr Index is used to determine the flowability properties of microcapsules. A low Carr Index value is desirable for good flowability. According to the literature, microcapsules with a Carr Index value between 0.05 and 0.15 are considered to have very good flowability, those with a value between 0.15 and 0.18 have good flowability, and those with a value above 0.18 have poor flowability.[54] This information suggests that the bulk density of the Caucasian blueberries microcapsules is slightly lower than that of the spray-dried blackberries powders and at the lower end of the range for açai juice microcapsules. The Carr Index value further supports that the microcapsules have good flowability.

3.2 | Solubility property

The solubility property of the microcapsules was 72.83 ± 0.45%. This value is significantly lower than the solubility values of red grape wine microcapsules, which range from 91.84% to 97.49%.[59] However, it is comparable to the solubility characteristic of pomegranate juice capsules, which range from 68% to 92%.[60] This indicates that while the microcapsules do not dissolve as readily as red grape wine microcapsules, their solubility is within the range observed for pomegranate juice capsules, suggesting a moderate level of solubility that may still be acceptable for certain applications.

3.3 | Change in anthocyanin content based on temperature and solvent percentage

Caucasian blueberries had the maximum concentration of monomeric anthocyanins when they were immersed in an ultrasonic bath with 100% acetone for 15 min at 25°C and 35 kHz. Moreover, it was shown that the quantity of monomeric anthocyanins reduced as the extraction temperature rose and the percentage composition of the solvents (acetone, ethanol and methanol) declined. This decline was ascribed to the negative association between temperature and time increased anthocyanin degradation. In a similar study by Flores et al,[61] the acetonic extract of whole blueberries was reported to yield a total monomeric anthocyanin content of 178.9 mg/L. Furthermore, the methanolic extract of whole blueberries was found to have a total monomeric anthocyanin content of 185.1 mg/L. These results highlight the variations in anthocyanin content based on the type of solvent used and contribute to the existing knowledge on anthocyanin extraction methods.

Table 3 displays the total monomeric anthocyanin contents (in mg/L) of Caucasian blueberries under tested extraction procedures.

3.4 | Effects of storage conditions on stability

3.4.1 | Effect of temperature and pH on colour parameters

Blue-purple microcapsules were used to colour each model system, which had pH values of 4.50, 5.00, 5.50 and 6.00. The systems were then held at various temperatures. Table 4 presents the model systems' original colour values prior to the application of temperature. As a result, changes associated with pH fluctuation were noted, the model systems' a^* values are positive, and the negative b^* value is constant across all models. The colour tone was determined by the H° value, which places red at 360°, blue at 270°, and green at 180°. The models' initial H° values showed a purple tint that leant toward blue.

In terms of hue, pH 4.50 and pH 5.50 are the most similar to blue, whereas the model system with a pH of 5.00 is closer to blue. The C* value is a metric for colour intensity that gets brighter as it gets further from the matte plane's centre (0). It is clear by looking at the C^* values of the models that the pH 4.50 and pH 5.00 models have more vibrant colour. Table 5 provides the values for colour parameters. The blue-coloured model systems were held at three different temperatures (25, 35, and 45°C).

The colour values differ for capsules obtained through spray drying of *Vitis labrusca* L. (fox grape, muscadine grape) and capsules obtained through spray drying with whey protein/maltodextrin ((L^*) 51.45, (a^*) 15.19, (b^*) −10.31). This discrepancy may be attributed to both the fruit variety and the wall materials used during encapsulation.[58] When examining the effect of heat treatment on colorimetric parameters, it was found that the untreated blueberries anthocyanin samples had L^*_0, a^*_0, and b^*_0 values of 32.41 ± 0.31, 4.06 ± 0.32, and 0.13 ± 0.06, respectively. During heat processing, the a^* values decreased, while the L^* values and b^* values increased.[62] This implies that temperature fluctuations significantly impact the colour values of the capsules.[63] The increase in L^* and b^* values suggests that the capsules become lighter and more yellow as the temperature increases, while the decrease in a^* values indicates a reduction in redness. These changes in colour parameters highlight the sensitivity of the microcapsules to heat, which could affect their visual appeal and applicability as food colourants in heat-processed products. The stability of these colour parameters at various temperatures should be considered when evaluating their potential industrial applications.

After looking over the data, it was discovered that all of the models' a^* values were consistently positive, giving the model systems purple hues. In every model, the b^* value produced negative results; the greatest variation

TABLE 3 Total monomeric anthocyanin content (mg/L ± standard deviation) depending on the extraction conditions of Caucasian blueberries.

Extraction solvent	Concentration of solvent (%)	Extraction temperature (°C)	Total monomeric anthocyanin content (mg/L ± standard deviation)
Acetone	100	25	199.14 ± 7.67
		50	115.78 ± 2.79
		75	—[†]
	75	25	128.95 ± 0.75
		50	105.94 ± 5.26
		75	87.12 ± 3.69
	50	25	99.43 ± 3.79
		50	82.20 ± 3.37
		75	78.87 ± 5.53
	25	25	86.40 ± 4.14
		50	65.27 ± 3.48
		75	58.76 ± 3.20
Ethanol	100	25	79.89 ± 1.74
		50	66.57 ± 1.96
		75	80.32 ± 4.14
	75	25	81.91 ± 5.83
		50	75.26 ± 1.33
		75	70.91 ± 3.92
	50	25	76.41 ± 3.45
		50	63.24 ± 5.65
		75	76.12 ± 4.03
	25	25	36.18 ± 5.36
		50	54.70 ± 3.98
		75	56.78 ± 4.02
Methanol	100	25	88.21 ± 2.71
		50	92.70 ± 4.88
		75	—[†]
	75	25	88.89 ± 5.19
		50	92.99 ± 4.40
		75	106.37 ± 2.17
	50	25	66.69 ± 4.71
		50	74.96 ± 4.28
		75	81.65 ± 3.93
	25	25	42.26 ± 4.37
		50	43.69 ± 3.66
		75	83.79 ± 4.14
Distilled water	100	25	12.30 ± 3.64
		50	17.53 ± 3.07
		75	27.21 ± 0.90

[†]Extraction process was not carried out at 75°C for acetone and methanol, as their boiling points are 56 and 64.7°C, respectively.

was noted at pH 6.00, and the lowest at pH 5.00. Figure 2 shows the total colour changes at various temperatures and pH levels till the end of storage. Up until the fifth day, the total colour changes were computed for all temperature levels. Observations indicated that the rate of colour loss in the models increased with temperature. The minimal colour change within the model systems occurred at 35°C and pH 5.00, while the most significant colour change was at 45°C and pH 4.50. Initial H° measurements revealed purple hues in all samples. As the pH increased, the coloured model systems exhibited less blueness and more purple tones. Colour degradation in the pH 6.00 model systems began between 25 and 45°C, and at 35°C in the pH 5.50 model systems. Consequently, colour changes occur more rapidly with increasing pH values. Studies have shown that anthocyanins are generally stable at pH values of 3.5 and below but tend to decompose at higher pH levels. Specifically, at pH values above 7.0, anthocyanins decompose depending on their substituent groups.[64]

More specifically, the pH affects the structure of anthocyanins, leading to the production of different colours. In an acidic environment (pH ~1.0), anthocyanins exist in their red flavylium cation form, which is highly soluble in water. As the pH increases between 2.0 and 4.0, they begin to transition into a bluish quinoidal base form. At neutral pH levels (around 5.0 to 6.0), colourless forms such as carbinol pseudobases and chalcones become dominant due to structural changes. Beyond pH 7.0, anthocyanins tend to degrade, with their stability and colour significantly affected by the surrounding environment and the specific substituent groups present. The pH-dependent colour transition and stability are crucial for understanding anthocyanin behaviour in various applications, particularly in food and beverage formulations.[64,65]

The H° value is a parameter that defines the tone of a colour and is arranged counterclockwise on a chromatic

TABLE 4 Pre-storage colour parameters (L^*, a^*, b^*, H°, C^*) of model systems coloured with microcapsules.

pH	L^* (lightness) (±SD)	a^* (red/green) (±SD)	b^* (yellow/blue) (±SD)	H° (hue) (±SD)	C^* (chroma) (±SD)
4.50	34.63 ± 0.28	3.79 ± 0.41	−3.13 ± 0.29	320.42 ± 1.82	4.91 ± 0.48
5.00	36.08 ± 0.92	3.04 ± 0.24	−3.07 ± 0.43	314.91 ± 2.07	4.32 ± 0.46
5.50	35.66 ± 1.57	2.34 ± 0.29	−1.78 ± 0.02	322.61 ± 3.43	2.94 ± 0.23
6.00	33.43 ± 0.19	2.64 ± 0.07	−1.73 ± 0.13	326.73 ± 3.16	3.16 ± 0.08

Note: SD, standard deviation.

TABLE 5 Changes in colour parameters (L^*, a^*, b^*) during storage (± standard deviation).

pH	Colour parameters	Temperature		
		25°C	35°C	45°C
4.50	ΔL^* (lightness)	5.03 ± 0.35	5.51 ± 0.31	8.24 ± 0.28
	Δa^* (redness)	1.75 ± 0.02	1.42 ± 0.05	0.98 ± 0.03
	Δb^* (yellowness)	−0.65 ± 0.21	−1.31 ± 0.14	−2.97 ± 0.20
5.00	ΔL^* (lightness)	5.52 ± 0.31	4.30 ± 0.26	7.50 ± 0.28
	Δa^* (redness)	1.97 ± 0.02	2.36 ± 0.02	2.31 ± 0.03
	Δb^* (yellowness)	−0.08 ± 0.11	−1.25 ± 0.12	−1.15 ± 0.14
5.50	ΔL^* (lightness)	5.00 ± 0.27	4.48 ± 0.35	6.98 ± 0.31
	Δa^* (redness)	2.12 ± 0.04	1.57 ± 0.03	1.99 ± 0.04
	Δb^* (yellowness)	−1.64 ± 0.43	3.33 ± 0.47	−1.54 ± 0.49
6.00	ΔL^* (lightness)	5.03 ± 0.32	7.23 ± 0.33	5.36 ± 0.30
	Δa^* (redness)	1.60 ± 0.01	1.36 ± 0.02	1.42 ± 0.01
	Δb^* (yellowness)	−2.14 ± 0.21	−1.83 ± 0.30	−1.64 ± 0.27

Note: The ΔL^*, Δa^* and Δb^* values were calculated considering the first day and the fifth day of storage at specified temperatures.

circle; the values on this circle are defined as 0°/360° (magenta), 90° (yellow), 180° (bluish-green), and 270° (blue).[66] Figure 3 shows the changes in H° values for model systems with varying pH levels (4.50, 5.00, 5.50 and 6.00), stored at different temperatures (25, 35 and 45°C). When the H° values of the model systems were examined, it was observed that all samples exhibited purple tones on the first day. The first signs of colour degradation were observed from the fourth day in the model system with a pH of 6.00 when stored at both 25 and 45°C, while degradation was also noted from the fourth day in the model with a pH of 5.50 when stored at 35°C. It was concluded that colour degradation occurred more rapidly at higher temperatures[48,52] and was influenced by pH. These results highlight the combined effects of temperature and pH on the stability of anthocyanins during storage.[67,68]

3.5 | Kinetic parameters for b^* value

Model systems (pH 4.50, 5.00, 5.50 and 6.00) coloured with microcapsules were subjected to heat treatment at 25, 35 and 45°C, and it was determined that thermal degradation followed first-order reaction kinetics. The first-order reaction kinetics at each temperature are illustrated in Figure 4.

Table 6 displays the kinetic parameters for the blueness-indicating b^* value. Based on these findings, it is noticed that the model system coloured with pH 6.00 maintained at 45°C experiences the fastest colour loss, whereas the model system coloured with pH 4.50 stored at 45°C exhibits the longest blue colour preservation. The system maintained at 25°C and pH 5.00 exhibits the second-longest lasting hue.

The $t_{1/2}$ for the loss of blue colour intensity was also calculated. The analyses showed that the $t_{1/2}$ values were affected by both temperature and pH. The shortest $t_{1/2}$ (5.37 days) was observed in the model systems stored at 35 and 45°C (pH 6.00). In contrast, the longest $t_{1/2}$ (19.14 days) was found in the model system (pH 4.50) stored at 45°C. These findings indicate that both temperature and pH jointly influence the $t_{1/2}$ value.

It was observed that the model system (pH 5.00) stored at 25°C had the highest D value (65.8 days), while

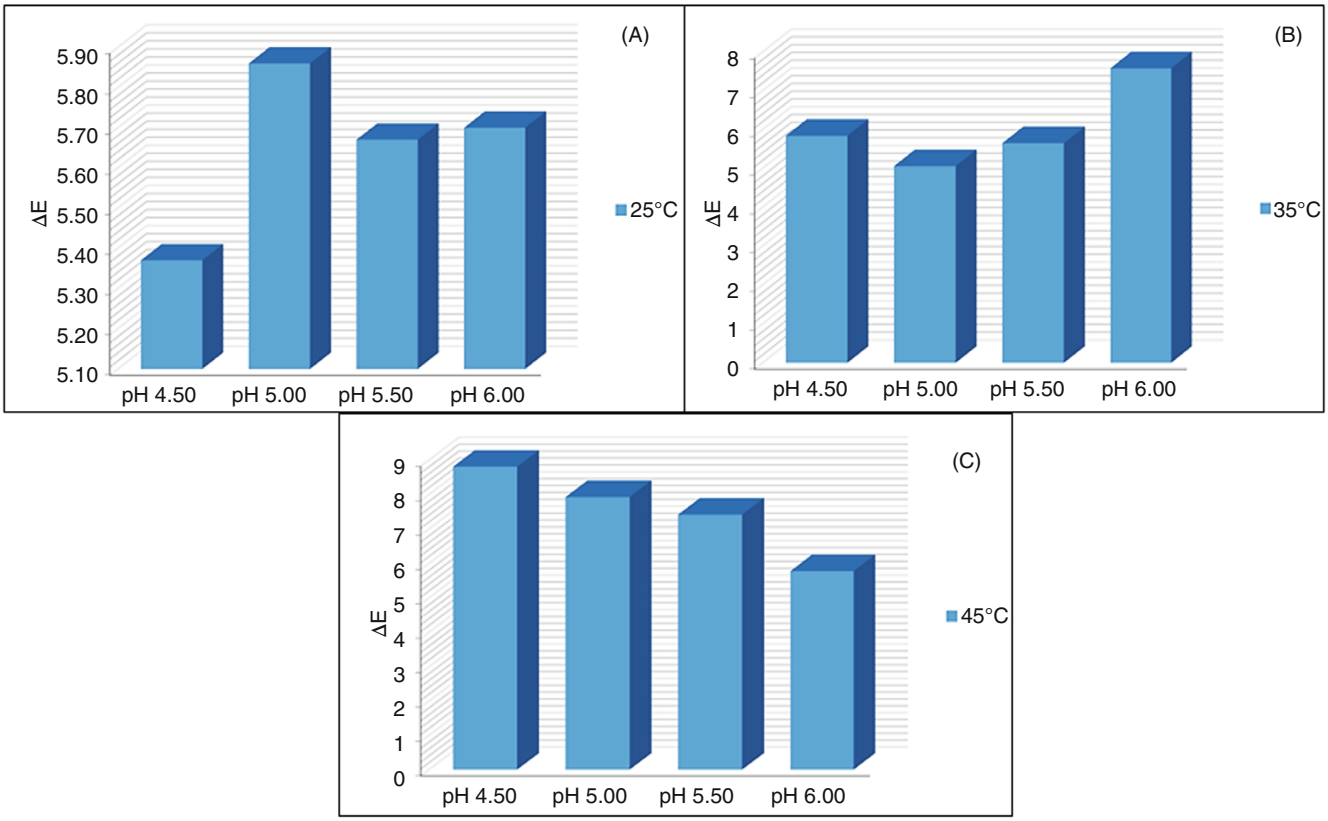


FIGURE 2 Total colour changes (ΔE) in model systems coloured with blue capsules stored at (A) 25°C, (B) 35°C and (C) 45°C.

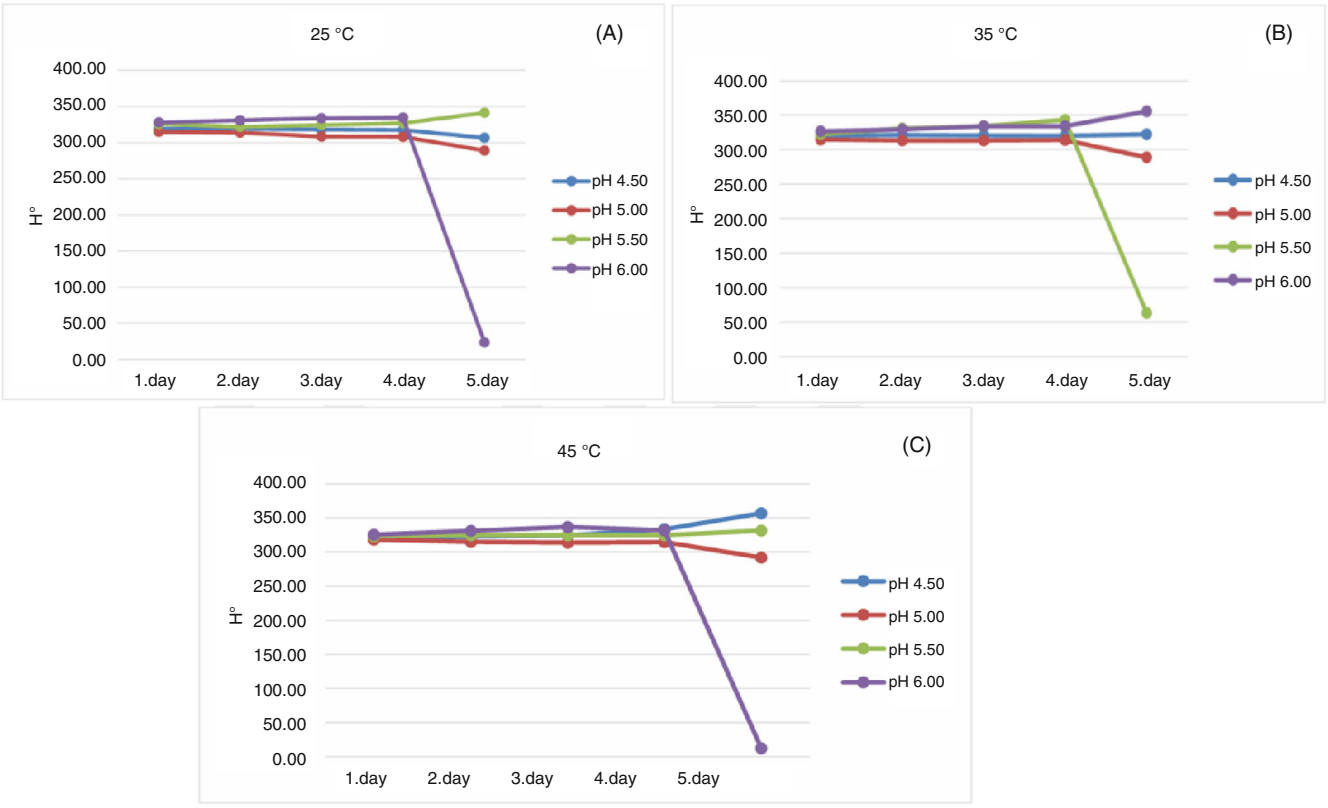


FIGURE 3 Hue angle (H°) values of model systems at different pH levels stored at (A) 25°C, (B) 35°C and (C) 45°C.

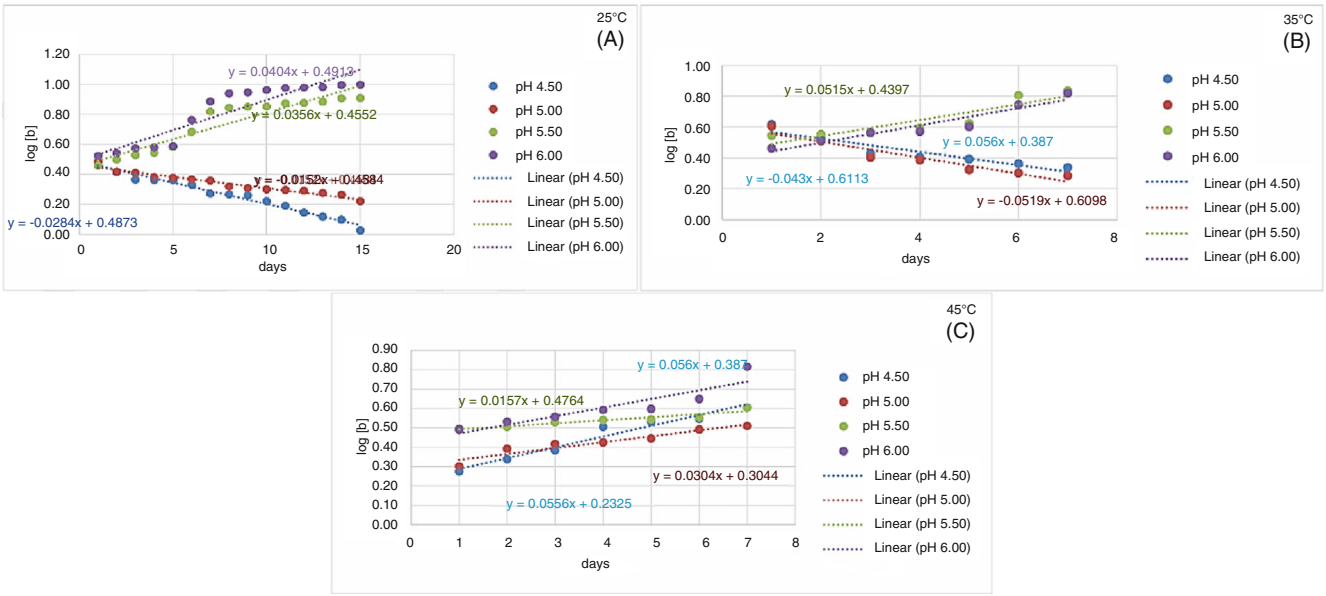


FIGURE 4 Variation of b^* values in coloured model systems at (A) 25°C, (B) 35°C and (C) 45°C depending on pH.

TABLE 6 Kinetic parameters.

Temperature (°C)	pH	Slope	k (1/day)	$t_{1/2}$ (day)	D Value (day)	E_a (J/mol)
25	4.50	−0.03	−0.06	10.59	35.21	69.15
	5.00	−0.02	−0.03	19.08	65.80*	37.64*
	5.50	0.04	0.08	7.60	28.08	88.16
	6.00	0.04	0.09	7.45	24.76	100.05
35	4.50	−0.04	−0.10	6.36	23.26	107.67
	5.00	−0.05	−0.12	5.82	19.35	129.95
	5.50	0.05	0.12	5.87	19.74	126.86
	6.00	0.06	0.13	5.37*	7.94*	137.94
45	4.50	0.02	0.13	19.14*	63.62	39.95*
	5.00	0.03	0.13	8.88	29.52	78.59
	5.50	0.06	0.08	5.41	17.99	143.74
	6.00	0.06	0.04	5.37*	17.88	144.77*

*Significant $p \leq 0.05$.

the system (pH 6.00) stored at 35°C had the lowest D value (7.94 days). A study on the stability of ferric anthocyanin, used as a natural blue food colouring in polysaccharide and gelatine-based model systems, reported that as the storage temperature increased, both $t_{1/2}$ and D values decreased.[45]

The Arrhenius equation is used to describe the temperature dependence of degradation rate constants. Figure 5 shows the Arrhenius plots for the coloured model systems (pH 4.50, 5.00, 5.50 and 6.00). These plots illustrate the relationship between the inverse of temperature ($1/T$) and the logarithm of the reaction rate constant (log k). The observed linearity in the Arrhenius plots indicates that the thermal degradation of the coloured model systems follow Arrhenius behaviour, allowing for the calculation of E_a values. The slope of the linear fit is directly related to the E_a value, which quantifies how sensitive the degradation reaction is to changes in temperature.

This observation supports the study's conclusions by showing that the coloured model systems deteriorate more quickly at higher temperatures. The E_a values range from 37.64 to 144.77 kJ/mol, reflecting the temperature dependence of the reaction. The model system that exhibits the highest E_a value has a pH of 6.00 at 45°C, whereas the system with the lowest E_a value has a pH of 5.00 at 25°C. The results of our work are supported by

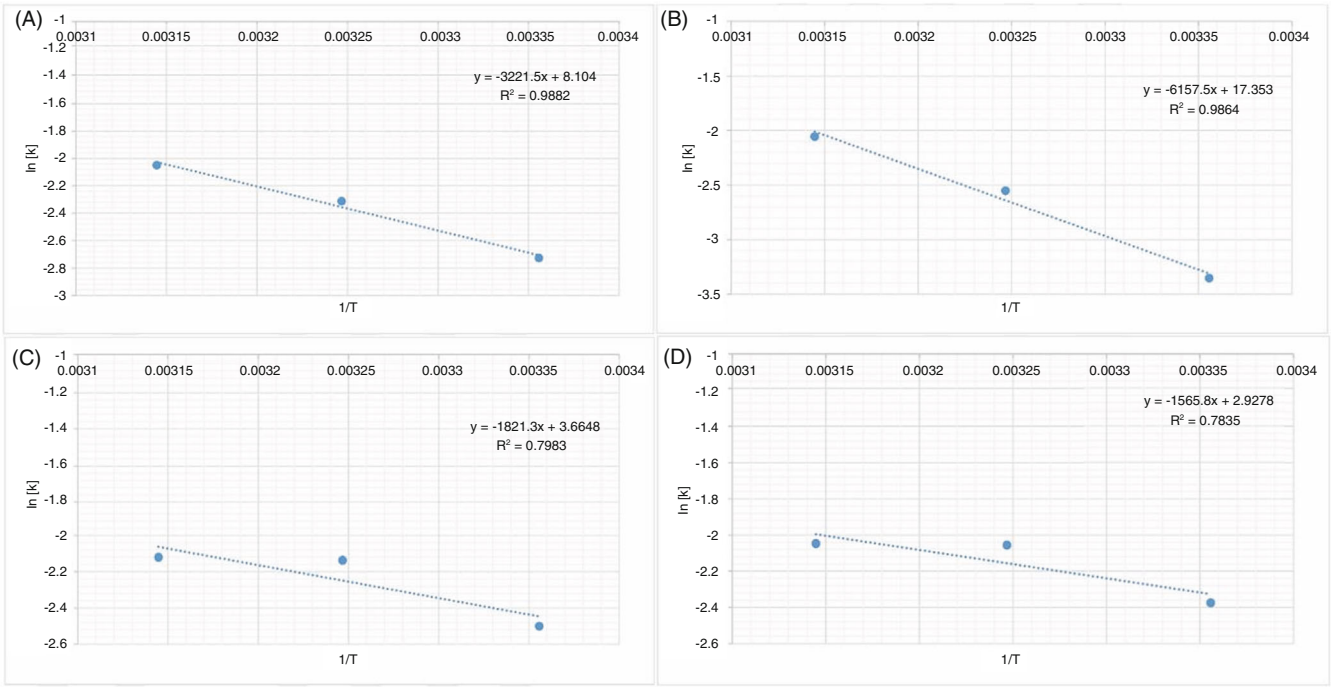


FIGURE 5 Arrhenius plots of the experimental data obtained from coloured model systems: (A) pH 4.50; (B) pH 5.00; (C) pH 5.50; (D) pH 6.00.

the observation that an increase in pH causes a discernible increase in the E_a value. This is in line with the findings of Mol,[48] who showed a similar tendency in red cabbage-derived anthocyanin capsules. In the literature, it has been reported that thermal treatments applied to fruits, vegetables, and simple food matrices result in anthocyanin losses ranging from 28% to 80%. The effect of temperature on anthocyanin degradation, particularly in relation to high E_a value, becomes especially pronounced during prolonged thermal processes, as demonstrated by kinetic and thermodynamic analyses on acerola (*Malpighia emarginata* L.).[69] Similarly, a higher E_a value was associated with greater thermal sensitivity of anthocyanins during the thermal processing of plum puree. A linear relationship effectively explained the change in both total anthocyanin content and the overall visual colour of plum puree during thermal treatment.[70] High E_a value indicates that anthocyanins in extracts are more prone to degradation when exposed to elevated temperatures.[71]

4 | CONCLUSIONS

Microcapsules obtained from Caucasian blueberries have been demonstrated to be stable and viable natural food colourants. These colourants are anticipated to be applicable in products such as mineral water, herbal teas, coffee and beer. The achieved shades of blue-purple offer significant advantages for the food industry in terms of stability and potential preference among children. Considering consumer trends and health aspects, this natural food colourant is expected to be a significant alternative to synthetic colourants, aligning with the growing demand for natural and health-conscious ingredients. The abundance of anthocyanins in Caucasian blueberries and their stability due to encapsulation are well-documented in the literature, indicating the fruit's potential for industrial production. This makes the resulting natural food colouring a valuable option for industrial applications, with the added benefit of potentially offering health advantages to consumers. As a result, the natural food colouring derived from these blueberries, with its blue-purple hues, may be highly useful in various industrial settings. Additionally, this natural food colourant may offer health benefits for end users, aligning with the increasing consumer demand for natural and health-promoting ingredients. It was discovered that the extraction of Caucasian blueberries in a variety of solvents and temperatures was most successful when done at 25°C using 100% acetone. This extraction method yielded the highest concentration of monomeric anthocyanins, highlighting the importance of optimising solvent type and temperature to maximise the extraction efficiency of valuable bioactive compounds from these blueberries.

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