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Wood hemicelluloses as sustainable wall materials to protect bioactive compounds during spray drying of bilberries

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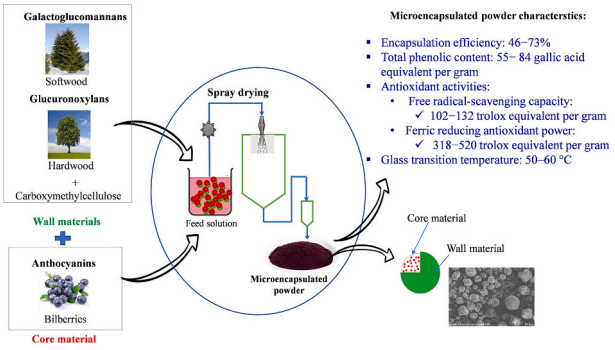
HIGHLIGHTS

- Studied wood hemicelluloses included galactoglucomannan and glucuronoxylan.
- Wood hemicelluloses are effective wall materials for spray drying of bilberries.
- Encapsulation efficiency of galactoglucomannan and glucuronoxylan was 71–73%.
- Adding carboxymethylcellulose reduced hemicelluloses' encapsulation efficiency.
- Hemicelluloses added phenolic and antioxidant activities to their bilberry powders.

GRAPHICAL ABSTRACT



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ABSTRACT

The most widely-used wall materials for spray-dried microencapsulation have limitations in cost-effectiveness, health benefits and sustainability. Wood hemicelluloses, by-products of the forestry industry, including galactoglucomannans and glucuronoxylans have the potential to be utilized as innovative wall materials. This study investigated the applicability of galactoglucomannan and glucuronoxylan and their mixtures with carboxymethylcellulose as wall materials for microencapsulation of bilberry juice, in comparison to gum arabic. The results indicated that galactoglucomannan and glucuronoxylan have a relatively high anthocyanin encapsulation efficiency (71–73%), which was similar to that of gum arabic (76%). The addition of carboxymethylcellulose reduced the encapsulation efficiency of wood hemicelluloses to 46–54%. Microencapsulated powders prepared with wood hemicelluloses were considerably higher in total phenolic content and antioxidant activities than those prepared with gum arabic, and mixtures of wood hemicelluloses with carboxymethylcellulose. The results indicate that wood hemicelluloses are efficient wall materials for spray-dried microencapsulation of bioactive compounds.

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Nomenclature

GGM	Galactoglucomannan
GX	Glucuronoxylan
GA	Gum arabic
CMC	Carboxymethylcellulose
GGM + CMC	Galactoglucomannan and carboxymethylcellulose
GX + CMC	Glucuronoxylan and carboxymethylcellulose
FD-J	Freeze-dried bilberry juice
WM	Wall material
TAC	Total anthocyanin content
TPC	Total phenolic content
EE	Encapsulation efficiency
SAC	Surface anthocyanin content
ITAC	Initial total anthocyanin content
GAE	Gallic acid equivalent
TE	Trolox equivalent
DPPH	Free radical-scavenging capacity
FRAP	Ferric reducing antioxidant power
SEM	Scanning electron microscopy
FTIR	Fourier transform infrared spectroscopy
XRD	X-ray diffraction
DSC	Differential scanning calorimeter
T_g	Glass transition temperature
T_m	Melting temperature

1. Introduction

Berries are high in natural polyphenol components, which act as powerful antioxidants and have a variety of pharmacological activities that can benefit human health [1]. Bioactive compounds contained within berries have been found to exhibit cardioprotective effects, anti-platelet aggregation properties, inhibition of the growth of cancer cells and oxidation of low-density lipoproteins [2]. Berries and berry products are important sources of polyphenols for the Nordic population [3]. Among the regularly consumed berries, bilberries (*Vaccinium myrtillus*, wild European blueberries) have the highest levels of polyphenols, particularly anthocyanins [4]. Anthocyanins are responsible for the characteristics and color, and greatly contribute to the health benefits of bilberries [5]. A number of studies have indicated that regular consumption of bilberries can reduce the risk of cardiovascular diseases and cancer [6].

However, these beneficial bioactive compounds, including anthocyanins, are unstable due to their sensitivity to high temperature, light, pH, oxygen and processing conditions, which can lead to the loss of their functional and antioxidant properties [7]. Additionally, due to the short bilberry fruiting season, the high operational costs of current preservation and processing activities (e.g., freezing and freeze drying) and the susceptibility of bilberries to microbial spoilage in the fresh form, leads them to currently be commercially unavailable to global consumers [8]. To combat this, microencapsulation of the bioactive compounds in bilberries with a suitable technique is an essential approach to protect them, to develop specifically stable components, additives and supplements, and to control the targeted release [9]. Although many techniques have been developed to encapsulate bioactive compounds, spray drying is the most often used technology in pharmaceutical and food industries due to its speed, cost-effectiveness, flexibility, high throughput, low energy consumption and widely accessible equipment [10]. Recent developments to the traditional spray dryer including the use of an ultrasonic nozzle and electrostatic collector which provides a wider range of applications and process flexibility. Furthermore, the use of a high-resolution camera to measure the particle shape and sphericity during spray drying was introduced in a recent study [11]. Spray drying is a commonly used technique to dry many bioactive compounds such as polyphenols, carotenoids, and essential oils, vitamins, polyunsaturated fatty acids, and many other heat-sensitive materials, because the drying time is extremely short (a few seconds), during which little degradation of bioactive compounds in drying materials occurs [12].

The high encapsulation efficiency (>70%) of bioactive compounds in berries by spray drying has been well reported [13–15]. However, in these studies, conventional wall materials (WMs) such as gum arabic (GA), guar gum, starches, maltodextrins, and proteins have been used. The utilization of conventional WMs has numerous limitations, including the lack of additional nutritional value in the final products. Most of them also display issues relating to their emulsifying properties, viability or sustainability. Maltodextrins, for example, lack strong emulsifying properties representing a limiting characteristic in the spray drying process [16]. Although GA has good emulsifying properties and retention of volatile compounds [17], its high expense limits its use. Meanwhile, some plant-based and animal protein-based WMs have shortcomings in terms of sustainability. For example, current starch production methods rely on conventional land-based agriculture. Environmental concerns associated with the continuous growth of agricultural land cast doubt on the sustainability of future terrestrial agriculture in providing stable, safe and secure food sources [18]. Similarly, dairy proteins are associated with methane emissions from livestock management [19], and microcapsules produced with such proteins are unsuitable for vegans and lactose-intolerant consumers. As a result, seeking new green types of cost-effective and sustainable WMs is necessary to meet the increased need of customers for “clean” and healthier food products and/or ingredients.

Biomass valorization into value-added products is a highly appealing waste and by-product management approach with several advantages such as carbon neutrality, renewability, and sustainability by not interfering with food and feed resources as was recently demonstrated [20]. Woody biomass is the most abundant organic source on the earth, producing around 5.6×10^{10} megagrams of carbon annually in the biosphere. Even though the majority of wood mass including hemicelluloses can be recovered through forest industry processes and valorized into bio-based products, only 4.8% of the biomass produced each year is utilized by humans, including as food, pulp, paper, energy, furniture, construction materials clothing and chemicals [21]. The major worldwide concerns, such as global warming, climate change and food security, all points to the necessity of using various biomass sources, ranging from agricultural and forest (woody) biomass to food wastes to ensure sustainable development. However, such development should be well engineered, considering the sustainability of the various aspects of the developing process. There are different tools for assessment the sustainability of forest industry such as techno-economic analysis, life cycle assessment, energy analysis, exergy analysis, and the combination of these techniques which have been discussed in depth in a recent work [22]. When woody biomass managed conservatively and used economically, forests can be viewed as a major renewable and sustainable resource [22,23].

Hemicelluloses are cell wall polysaccharides that account for 20–30% of wood dry mass [24], but they are currently considered as low-value by-products of the forest industry. During cellulose refining, hemicelluloses typically end up in waste pulping liquor and/or are burned for energy [23]. Wood hemicelluloses, including galactoglucomannans (GGM) from sprucewoods and glucuronoxylans (GX) from birchwoods, have been successfully extracted using a highly safe and environmentally friendly method known as pressurized hot water extraction [25]. After safety evaluation according to food legislation, the extracted hemicelluloses could be incorporated in many food products

[26,27]. GGM and GX have been demonstrated to have excellent emulsifying properties [28], low viscosity in aqueous solutions at high concentrations [29] and high thermal stability [30]. In addition, in an earlier study (unpublished data), feed solutions of GGM and GX with bilberry juice at concentrations of up to 10–15% (*w*/w) prepared by simply stirring at 600 rpm for 30 min were physically stable for a week and had a low viscosity, which is suitable for spray drying. It was also observed that carboxymethylcelluloses (CMC) have a high affinity towards GGM and GX. The high affinity between WMs could lead to crosslinking and the formation of a complete film between them [31], and therefore the building of a protective layer around the core material [31,32]. Therefore, the presence of CMC in feed solutions is expected to improve the protection of bioactive compounds during spray-dried microencapsulation of GGM and GX. These reported properties of GGM and GX, along with their abundant, sustainable and low-cost sources, indicate that they could be excellent WMs for spray-dried microencapsulation of bioactive compounds, and viable alternatives to currently used wall materials. However, such added-value applications of wood hemicelluloses, especially for spray drying of bilberries in the production of functional powders have so far not been exploited, and their ability to protect bioactive compounds during spray drying is still unknown.

The aim of this study was to investigate the applicability of GGM and GX alone, and in combination with CMC, as WMs for spray-dried microencapsulation of bilberry juice to protect phenolic and anthocyanin compounds and their antioxidant activities during spray drying, in comparison to GA. The physiochemical properties of microencapsulated powders, including morphology, molecular structure, thermal characteristics, particle size and solubility, were also determined.

2. Materials and methods

2.1. Materials and reagents

Extracts of GGM and GX were recovered from wood saw meal by semi-pilot-scale pressurized hot water extraction, as described by Kilpeläinen [33], and provided by Natural Resources Institute Finland (Luke). The extracts were then spray-dried using a pilot-scale spray dryer (Mobile-minor, Niro Atomizer Co., Ltd., Copenhagen, Denmark) at inlet and outlet air temperatures of 180 and 70 °C, respectively, to obtain dried powders (GGM and GX). Information about the chemical composition of GGM and GX has been reported by Mikkonen [34]. GA from Acacia trees was purchased from KitchenLab (Stockholm, Sweden). Na-CMC (Texturecel™ CRT 30 PA) was kindly provided by the DuPont Company (Delaware, USA). A commercial bilberry juice produced by Marjex® (Helsinki, Finland) was purchased from a local supermarket (Helsinki, Finland). According to the manufacturer, the bilberry juice contains; 10.2% carbohydrates, 0.8% protein and 0.6% fat. The soluble solid content of the bilberry juice was determined by using a digital refractometer (Atago PAL1, Bellevue, Washington, USA), and the result was around 10% (*w*/w). 2,2-Diphenyl-1-picryl-hydrazyl (DPPH) and 2,4,6-tripyridyl-*s*-triazine (TPTZ) were obtained from Sigma-Aldrich (St Louis, USA), iron (III) chloride hexahydrate, potassium chloride, sodium acetate and sodium carbonate from Merck (Darmstadt, Germany), and gallic acid and Folin-Ciocalteu reagent from Sigma-Aldrich (Buchs, Switzerland). Ethanol (≥99.5%) was purchased from the Altia company (Helsinki, Finland) and methanol was obtained from Sigma-Aldrich (Helsinki, Finland). All chemicals and reagents were of analytical grade. Milli-Q water was used as a solvent for all experiments.

2.2. Preparation of feed solutions for spray-dried microencapsulation

Feed solutions were prepared in two consecutive steps: (1) preparing the wall material solutions, and (2) mixing the wall material solutions with bilberry juice as discussed below.

2.2.1. Preparation of wall material solutions

Aqueous solutions (10% *w*/w) of GGM, GX and GA were prepared by magnetic stirring (600 rpm) overnight (~15 h) at room temperature (22 °C). CMC solution (7% *w*/w) was dissolved in warm water at about 45–50 °C and stirred overnight (~15 h) until complete dissolution was achieved. Dissolving WMs directly into bilberry juice was avoided to prevent introducing oxygen into the solutions while stirring, which could lead to the oxidation of anthocyanins present in the bilberry juice. Furthermore, any involvement of heating in the preparation of CMC solutions could cause degradation of bioactive compounds if bilberry juice was used as a solvent to dissolve CMC. The 10% (*w*/w) concentration of hemicellulose solutions was chosen based on preliminary trials, which indicated that a high viscosity of hemicellulose solutions was noticeable at higher concentrations (e.g., >15%, *w*/w) causing difficulties in spray drying.

2.2.2. Preparation of feed solutions consisting of wall material and bilberry juice

Feed solutions of GGM, GX, GA, GGM + CMC and GX + CMC with bilberry juice were prepared with solid ratios as indicated in Table 1. When a single WM was used (GGM, GX and GA), the solid ratio between WM and juice was 1:1, while in mixtures of WMs (GGM or GX with CMC), the solid ratio between GGM (GX), CMC and juice was 1:0.7:1. The final solid concentrations of the feed solution were about 10% (*w*/w). The WM solutions and bilberry juice were mixed under stirring at 600 rpm for 30 min before spray drying. All feed solutions were stirred with a magnetic stirrer in closed Duran glass bottles to avoid introducing oxygen, which can induce the degradation of phenolic compounds and anthocyanins in bilberry juice [35].

2.2.3. Microencapsulation by spray drying

The spray drying process was performed using a mini-spray dryer (B-290, Buchi Labortechnik AG, Flawil, Switzerland) coupled with a 0.70-mm spraying nozzle. The conditions used were as follows: an aspiration rate of 80%, an inlet air temperature of 150 ± 2 °C and outlet air temperature of 70 ± 2 °C, a feed flow rate of 10–15 mL/min and compressed air pressure of 5–8 bar. The powders collected from the product collection vessel were stored in an air-tight container and kept in a dark and dry place.

2.3. Microencapsulated powder characterization

2.3.1. Extraction of bioactive compounds

The wall structure of microencapsulated powders was deconstructed as follows: 100 mg of microencapsulated powders was added to 10 mL of an ethanol, acetic acid and water mixture (45:5:50 by volume). This solvent combination was found to be better than water for the extraction of anthocyanins and was adapted from da Rosa [36] and García-Gurrola [37]. The dispersion was agitated using a magnetic stirrer at 600 rpm for 15 min and then ultrasonicated for 30 min in an ultrasonic bath (Branson 3210, Branson, Danbury, USA) with a frequency of 47 kHz. The dispersion was centrifuged (SL 8R centrifuge, Thermo Scientific,

Table 1
Sample codes, sample names and solid ratios between wall materials and bilberry juice.

Sample codes	Sample names	Wall material to juice solid ratio (w/w)
GGM: J	Galactoglucomannan: bilberry juice	1:1
GX: J	Glucuronoxylan: bilberry juice	1:1
GA: J	Gum arabic: bilberry juice	1:1
GGM + CMC: J	Galactoglucomannan and carboxymethyl cellulose: bilberry juice	1:0.7:1
GX + CMC: J	Glucuronoxylan and carboxymethyl cellulose: bilberry juice	1:0.7:1
FD-J	Freeze-dried bilberry juice	–

Osterode, Germany) at 10,000 rpm for 10 min and the supernatant was then collected. The same procedure was repeated twice for the sedimented solids, and the collected supernatants were combined. The extracts were then used to determine the total anthocyanin content, total phenolic content (TPC) and antioxidant activities in microencapsulated powders, as described in the next sections. All the experiments were performed in triplicate.

2.3.2. Extraction of surface anthocyanins

One hundred milligrams of microencapsulated powder was dispersed in 5 mL of acetone [38]. The dispersions were stirred by magnetic stirring (500 rpm) at room temperature for 1 min. They were then centrifuged (SL 8R centrifuge, Thermo Scientific, Osterode, Germany) at 10,000 rpm for 10 min and the supernatant was collected for quantification of the surface anthocyanin content of microencapsulated powders.

2.3.3. Determination of the encapsulation efficiency of anthocyanins

The anthocyanin content in the feed solutions and the extracts obtained in sections 2.3.1 and 2.3.2 was determined with the same method as reported by Lee [39] with a slight modification. For each solution, an aliquot was diluted with two buffer solutions of pH 1.0 (potassium chloride, 0.025 M) and pH 4.5 (sodium acetate, 0.4 M). The absorbance of each dilution was measured at 510 nm (A_{510}) and 700 nm (A_{700}) against a distilled water control using a UV–Vis spectrophotometer (UV-1800, Shimadzu Europe GmbH, Duisburg, Germany). The anthocyanin content in each solution was calculated as milligrams of cyanidin-3-glucoside equivalents per gram of total solid in the solution using eq. (1). Cyanidin 3-O-glucoside is the most abundant anthocyanin in bilberry fruits [40].

$$Anthocyanin\ content = \frac{A \times M_W \times D_F \times V}{\varepsilon \times L \times m} \quad (1)$$

where $A = [(A_{510} - A_{700})pH_{1.0}] - [(A_{510} - A_{700})pH_{4.5}]$, M_W is the molecular weight of cyanidin-3-glucoside (449.2 g/mol), D_F is the dilution factor, V is the sample volume (mL), ε is the molar absorptivity of cyanidin-3-glucoside (26,900 L/mol.cm) L is the path length (1 cm), and m is the sample weight (g).

From the eq. (1), depending on the solution used for analysis, the anthocyanin content can be the initial total anthocyanin content (ITAC) in feed solutions, the total anthocyanin content (TAC) in the microencapsulated powders or the total anthocyanin on the powder surface (SAC). The percentages of SAC (%) and encapsulation efficiency (EE, %) of microencapsulated powders were calculated according to eqs. 2 and 3, respectively [41,42].

$$SAC(\%) = \frac{SAC}{ITAC} x\ 100 \quad (2)$$

$$EE\ (\%) = \frac{TAC - \mathrm{SAC}}{ITAC} x\ 100 \quad (3)$$

2.3.4. Total phenolic content

The TPC of microencapsulated powders was determined by following the Folin-Ciocalteau method [43] and using the extract obtained in section 2.3.1. Briefly, 0.05 mL of Folin-Ciocalteu reagent was mixed with 0.02 mL of the extract and 0.78 mL of deionized water. After exactly 1 min, 0.15 mL of sodium carbonate (20%) was added, and the mixture was allowed to react for 60 min in the dark at ambient temperature. The total polyphenol concentration was determined by constructing a calibration curve using gallic acid as a standard with various concentrations from 0.05 to 0.5 mg/mL ($R^2 = 0.99$) and by measuring the absorbance of samples at 750 nm using a UV–Vis spectrophotometer (UV-1800, Shimadzu Europe GmbH, Duisburg, Germany). The results were expressed as milligrams of gallic acid equivalents (GAE) per gram of powder.

2.3.5. Antioxidant activities

Antioxidant activities were determined by free radical-scavenging capacity (DPPH) and ferric reducing antioxidant power (FRAP). DPPH assay involves electron and hydrogen atom transfer reactions [44]. On the other hand, FRAP reaction detects the compounds' redox potentials, based on an electron transfer mechanism. However, this methodology is not able to detect compounds that can transfer hydrogen [45].

The antioxidant capacity of microencapsulated powders, measured as the DPPH (2,2-diphenyl-1-picrylhydrazyl) radical scavenging ability, was determined by following a method described by Arnous [44] and using the extract obtained in section 2.3.1. Aliquots of 0.025 mL of the extract were placed in different test tubes and 0.975 mL of the DPPH solution in methyl alcohol was added. After shaking, the tubes were kept in the dark, and after 60 min, the absorbance was measured at 515 nm using a UV–Vis spectrophotometer (UV-1800, Shimadzu Europe GmbH, Duisburg, Germany). Methyl alcohol was used as a blank sample to calibrate the spectrophotometer. The DPPH antioxidant activity was calculated from the Trolox calibration curve against % inhibition in the range of 200–2000 μmol ($R^2 = 0.99$) and expressed as μmol Trolox equivalent/g of powder (μmol TE/g). The percentage of DPPH free radical inhibition was calculated as in eq. (4):

$$Inhibition\ (\%) = 1 - \frac{As}{Ac} x\ 100 \quad (4)$$

where As and Ac are the absorbance of sample and the control, respectively.

The reducing capacity of microencapsulated powders was estimated using the FRAP assay. This method involves the reduction of the ferric 2,4,6-tripyridyl-*s*-triazine complex (Fe^{+3}-TPTZ) to its ferrous, coloured form (Fe^{+2}-TPTZ) in the presence of antioxidants [45]. The FRAP reagent includes 2.5 mL of 10 mM TPTZ solution in 40 mM HCl, 2.5 mL of 20 mM $FeCl_3$ and 25 mL of 0.3 mM acetate buffer, pH = 3.6. Aliquots of 0.05 mL of the extract obtained in section 2.3.1 were mixed with 0.95 mL of FRAP reagent and the absorbance of the reaction mixture was measured at 593 nm after incubation at 37 °C for 30 min. FRAP was calculated based on the Trolox calibration curve in the range of 100–2000 μmol ($R^2 = 0.99$) and expressed as μmol TE/g.

2.3.6. X-ray diffraction analysis

X-ray diffraction (XRD) analysis of microencapsulated powders were obtained by using an Empyrean Alpha 1 X-ray diffractometer (Malvern Panalytical, Worcestershire, UK). The measurement was carried out with a voltage and current of 45 kV and 40 mA, respectively, by using Cu radiation (λKα1 = 1.541 Å) at 25 °C and an angular range of 2θ = 5–70°. Samples were packed into the rectangular cavity of a plastic holder and scanned at a step size and scan speed of 0.01° and 0.03°/s, respectively.

2.3.7. Scanning electron microscopy

Scanning electron microscopy (SEM) with field emission (FESEM, S-4800, Hitachi, Tokyo, Japan) was used to examine the morphology of microencapsulated powders. The powders were fixed with double carbon tape, and coated with gold/palladium at a thickness of 4 nm with two cycles (208HR, Cressington Scientific Instruments, Watford, UK). The coated samples were evaluated using a FESEM at an accelerating voltage of 10 kV, an emission current of 10 μA and a working distance of 10 mm.

2.3.8. Fourier transform infrared spectroscopy

The infrared absorbance spectra of microencapsulated powders were recorded using a PerkinElmer FTIR spectrophotometer (Spectrum One, Perkin Elmer, Wellesley, USA). The scanning frequencies ranged from 4000 to 650 cm^{-1}. The spectral resolution was 4 cm^{-1} and the number of scans was 16.

2.3.9. Solubility

The solubility of microencapsulated powder was determined following the methods of [46]. 5 g of the microencapsulated powder was added to 100 mL mQ water in closed Duran glass bottles. Agitation was performed at room temperature (22 °C) using a magnetic stirrer at 600 rpm for 30 min. Then, the dispersion was centrifuged using a temperature-controlled SL 8R centrifuge (Thermo Scientific, Osterode, Germany) at 22 °C, 10,000 rpm for 10 min. The supernatant was decantated and the remaining undissolved powders were then transferred to a pre-weighed pan and oven dried (Memmert 800, Schwabach, Germany) at 105 °C to constant weight. The increase in the mass of the moisture pan was the content of insoluble solids (W_{is}), and this was used to determine the water solubility of the microencapsulated powders. The moisture content of the powders was considered in the calculations. The total solids in the dispersion before centrifugation were calculated using the precisely measured mass of microencapsulated powders (W_{ts}). The solubility (S, %) of microencapsulated powders was calculated using the following eq. (5):

$$S\ (\%) = \frac{W_{ts} - W_{is}}{W_{ts}}\ 100 \tag{5}$$

2.3.10. Particle-size distribution

The size and distribution of particles of the microencapsulated powder were determined with a static light scattering technique in dry mode (Mastersizer 3000, Malvern Instruments Ltd., Malvern, UK) using air as the dispersion medium. Approximately 2 g of powder was loaded into a feeding tray. The dispersion air pressure was adjusted to 0.1 bar and the refractive index used was 1.53.

2.3.11. Thermal properties

The thermal properties of microencapsulated powders were determined using a differential scanning calorimeter (DSC-823e, Mettler-Toledo, Greifensee, Switzerland). Approximately 5–10 mg of sample was weighed into a 40-μL aluminium pan, sealed hermetically and scanned at a heating rate of 5 °C/min from 25 to 200 °C. The onset temperature was taken as T_g, and the sharp endothermic peaks were identified as the melting temperature.

2.3.12. Color

The color of microencapsulated powders was evaluated using a Minolta spectrophotometer (CM-2600d/2500d, Minolta, Osaka, Japan) with a CIELAB scale (L* and a* and b*). The color parameters ranged from L* = 0 (black) to L* = 100 (white), − a* (greenness) to + a* (redness), and − b* (blueness) to + b* (yellowness).

2.3.13. Moisture content and water activity

The moisture content of the microencapsulated powders was determined gravimetrically by oven (Memmert 800, Schwabach, Germany) drying at 105 °C to constant weight. Water activity at 22 °C was measured using a water activity meter (LabMaster, Novasina, Zurich, Switzerland).

2.4. Statistical analysis

One-way ANOVA followed by post-hoc Tukey's test was performed to differentiate the mean values of the results of different homogenization techniques. The data were tested for the normality of the distribution by analyzing the residuals. Statistically significant differences were assessed for $p < 0.05$ at a confidence level of 95% using JMP™Pro 13 (SAS Institute, Cary, USA) software. All samples were prepared in triplicates and three replicate analyses were performed for each sample, resulting in a total of nine measurements per sample and the results are expressed as mean values (± standard errors). For the characterization of microencapsulated powders, one representative replicate was selected to show in the figures due to the similarity among replications.

3. Results and discussion

3.1. Encapsulation efficiency of anthocyanins

To evaluate the loss of anthocyanins during spray drying, a comparison between the TAC in the feed solutions and in the microencapsulated powders was carried out. Around 70–80% of TAC in the feed solutions of GGM: J, GX: J and GA: J was recovered in microencapsulated powders after spray drying, while only around 50% of TAC was recovered in microencapsulated powders of GX + CMC: J and GGM + CMC: J from their feed solutions. This indicated the occurrence of anthocyanin loss during spray drying. The loss of anthocyanins could happen during the preparation of feed solutions and due to the deposition of powder on the chamber's wall. A higher stickiness and deposition of feed solution on the chamber walls was visually observed for GX + CMC: J and GGM + CMC: J feed solutions. This will increase the exposure time of anthocyanins to heat and thus increase their degradation [47]. Meanwhile, powders of FD-J exhibited better preservation of anthocyanins (>90%) due to the low drying temperature, which has also been reported in previous studies [48,49].

As indicated in Fig. 1a, the EE of GX: J, GGM: J microencapsulated powders was in the range of 71–73%, which was slightly lower than that of GA: J powders. Against expectations, the EE of GGM + CMC: J and GX + CMC: J samples was considerably lower (46–54%), indicating the negative effects of CMC addition on EE. GGM and GX had a similar EE to other reported WMs. da Rosa [36] reported that the EE for anthocyanin compounds extracted from blueberry using maltodextrin (DE20) and resistant starch (Hi-maize) by spray drying was 74–85%. Meanwhile, the EE of GA for anthocyanins from chokeberry was 79% [50], and that of inulin, starch and maltodextrin for anthocyanins from jussara fruit was 67% [51]. The low EE of the microencapsulated powders produced from GGM + CMC: J and GX + CMC: J could be caused by the high viscosity of their feed solutions. The ability of the WM to form a coating layer in spray-dried microencapsulation is related to the viscosity of its feed solution [52]. WMs inducing high viscosity of feed solutions and inappropriate solidification conditions exhibit a low EE and poor protection ability, as such WMs would be unable to flow around and coat core materials [53]. Similarly, Di Battista [54] reported that in spray-dried microencapsulation of phytosterols using gum, maltodextrin and surfactants as WMs, feed solutions with lower viscosity resulted in a higher product yield and EE. Therefore, the use of lower concentrations of CMC with either GGM or GX could improve the EE, but this requires further investigation.

As expected, there was a negative correlation between EE and SAC. As illustrated in Fig. 1a, the SAC of GA microencapsulated powders was lowest (~4%), whereas that of GX + CMC: J powders was highest (~7%). The SAC values of GX: J, GGM: J and GGM + CMC: J samples were approximately 5–6%. In summary, GGM and GX had a very high ability to retain anthocyanin compounds during spray-dried microencapsulation, indicating their efficiency as WMs.

3.2. Total phenolic content of microencapsulated powders

The TPC of microencapsulated powders prepared from various WMs alongside FD-J powder is presented in Fig. 1b. The TPC of FD-J powder was around 105 mg GAE/g, which was higher than the values reported for freeze-dried cranberry (91 mg GAE/g) and similar to freeze-dried blueberry powders (108 mg GAE/g) [55]. Regarding the microencapsulated powders, the TPC of GGM: J and GX: J samples was the highest, with values of 74 and 84 mg GAE/g, respectively. Meanwhile, the TPC of GA: J microencapsulated powders was almost half that of GGM: J and GX: J powders (40 mg GAE/g), while that of GGM + CMC: J and GX + CMC: J was around 55–60 mg GAE/g. Unlike GA and CMC, it is reported that GGM and GX naturally contain lignin-derived phenolic residues [56], which strongly contribute to the high TPC in their microencapsulated powders. The phenolic content of GGM and GX

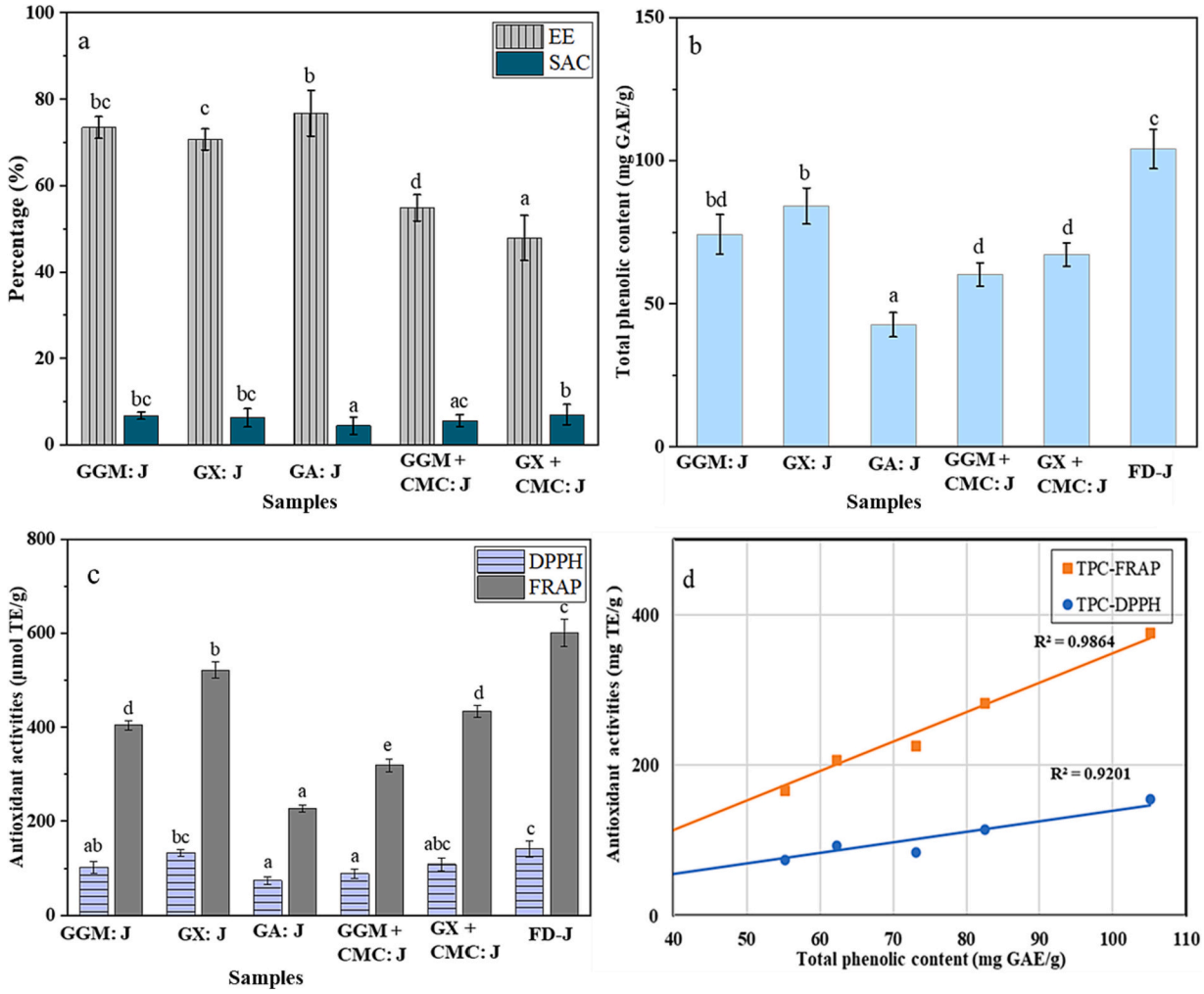

Fig. 1. (a) Encapsulation efficiency (EE) and surface anthocyanin content (SAC), (b) total phenolic content and (c) antioxidant activities, including radical scavenging activity (DPPH) and ferric reducing antioxidant power (FRAP), of microencapsulated powders prepared from GGM, GX, GA, GGM + CMC, and GX + CMC, and those of freeze-dried juice powder (FD-J), and (d) the correlation between antioxidant activities and total phenolic content. The data are presented as means ± SD (n = 3) and means in the same graph with different letters as superscripts are significantly different at $p < 0.05$. Refer to Table 1 for sample code.

powders was estimated to be around 50 and 70 mg GAE/g powder, respectively [34]. The higher phenolic content in the GX than that of GGM powder was most likely also the reason for the higher TPC in the GX microencapsulated powders as compared to GGM powders.

As previously mentioned, the presence of CMC in feed solutions generated a higher viscosity, which in turn prolonged the exposure of bioactive compounds to heat during spray drying and thus increased the loss of bioactive compounds [53,54,57]. Moreover, the addition of CMC to GGM and GX increased the solid ratio of WM to core material, which caused a diluting effect for both the core material and the proportion of GGM and GX, thus lowering the TPC in their microencapsulated powders. The results demonstrated that the use of GGM and GX as WMs in spray-dried microencapsulation of polyphenol-rich fruits makes the produced powders a superior source of phenolic compounds compared to conventional WMs (e.g., GA).

A comparison between the EE and TPC of bilberry powders microencapsulated by GGM and GX in this study, to other studies on the use of polysaccharide-based wall materials in spray-dried microencapsulation of polyphenols are reported in Table S1 in the supplementary materials.

3.3. Antioxidant activities of microencapsulated powders

As illustrated in Fig. 1c, the antioxidant activities of microencapsulated powders prepared from different WMs followed similar trends in both DPPH and FRAP assays. Due to the high TPC and TAC and the absence of WMs, the FD-J powders exhibited the highest antioxidant capacity in both DPPH and FRAP assays, which was about 140 and 600 μmol TE/g, respectively. Aaby [58] reported a lower FRAP result for freeze-dried strawberry powder, which was around 420 μmol TE/g. Regarding microencapsulated powders, GGM: J and GX: J samples had considerably higher antioxidant activities compared to GA: J and their counterparts, GGM + CMC: J and GX + CMC: J. GX: J microencapsulated powders exhibited higher antioxidant activity than GGM: J samples. For example, the DPPH of GX: J microencapsulated powders was 132 μmol TE/g, while that of the GGM: J powders was around 103 μmol TE/g. This is because of the higher phenolic content naturally present in GX than GGM powders [59,60]. GA: J exhibited antioxidant activities of DPPH and FRAP around 74 and 227 μmol TE/g, respectively. These results are in accordance with the data reported by da Silva Carvalho [61] for spray-dried microencapsulated powders of jussara berries with different maltodextrins. The authors reported that the microencapsulated powders exhibited antioxidant activities measured by DPPH and FRAP assays in the range of 60–75 and 260–300 μmol TE/g, respectively. However, these results are only approximately half of the DPPH and FRAP measurements from GGM: J and GX: J microencapsulated powders, which were in the range of 100–130 and 400–500 μmol TE/g, respectively. This indicated that the antioxidant activities can vary according to the WMs applied. In fact, unlike GA and CMC powders, GGM

and GX powders exhibited good radical scavenging activity by themselves [59,60].

As mentioned in the previous section, the addition of CMC to GGM and GX caused a diluting effect and in turn lowered the antioxidant activities. The high antioxidant activity can also result from the hydrolysis of phenolic compounds in bilberry juice and GGM/GX powders during the drying process, which can lead to the formation or release of antioxidants, meaning a higher capacity for the deactivation of free radicals [62,63]. Furthermore, it can be due to the caffeoyl group (naturally presented in GGM and GX powders), which can be acylated to anthocyanin and thus greatly contribute to the high free radical activity [64]. The total antioxidant activities were positively correlated with the TPC, and the correlations with the TPC were almost the same for both FRAP and DPPH ($R^2 > 0.90$), as illustrated in Fig. 1d. In summary, microencapsulated powders produced with GGM and GX as WMs have the added value of strong antioxidant activities when compared to GA.

3.4. Microstructure of microencapsulated powders

The X-ray diffraction patterns of spray-dried GX: J, GGM: J, GA: J, GGM: J + CMC: J and GX: J + CMC: J microencapsulated powders, and those of FD-J powders are presented in Fig. 2. A broad peak (halo) was found for all samples. This confirms that all the produced powders were mainly composed of amorphous proportions. Evaporation of water during spray drying occurs so rapidly that the solute does not have enough time to crystallize [65]. In most cases, the average time for dried particles to pass through the drying zone in spray drying equipment is approximately 5–15 s, depending on the equipment configuration [66]. Furthermore, spray drying is the most common method to obtain amorphous solids [67]. Similar XRD patterns were found for spray-dried microencapsulated powders of strawberry juice with maltodextrin, in which an amorphous structure was confirmed by the presence of broad undefined peaks in their XRD patterns [68].

3.5. Morphology of microencapsulated powders

As illustrated in Fig. 3, besides spherical particles, semi-spherical microparticles with a varied size and void were the most commonly observed morphology. Surfaces with cavities and morphological irregularities could be caused by water evaporation rates during the spray drying process [69]. The higher the temperature (faster evaporation), the smoother and more defined the surfaces [70]. Morphological analysis revealed that GX: J, GGM: J and GA: J microencapsulated powders were irregularly spherical in shape, and that GGM: J and GA: J microencapsulated powders had a smoother surface with fewer indentions, wrinkles and cracks than GX: J microencapsulated powders. A similar morphology was also observed in the SEM analysis of microencapsulated powders containing anthocyanin from barberry and blackberry with GA, maltodextrin and starch [41]. GGM + CMC: J and GX + CMC: J microencapsulated powders also had irregularly spherical-shaped particles with a dented surface. However, GGM + CMC: J microencapsulated powders exhibited a round shape with noticeable rough surfaces, while GX + CMC: J samples had smoother surfaces with less evident cracks or particle agglomerations. In SEM analysis, GX + CMC: J microencapsulated powder was similar to the spray-dried microencapsulated powders of black carrot anthocyanin with different maltodextrins [71].

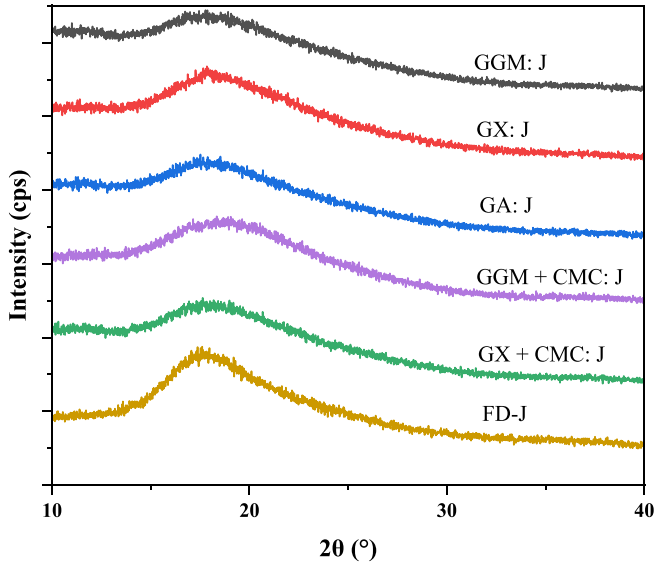

Fig. 2. XRD profiles of GGM: J, GX: J, GA: J, GGM + CMC: J and GX + CMC: J microencapsulated powders, and those of FD-J powders. Cps: counts per second. Refer to Table 1 for sample codes.

The FD-J powder had a non-spherical, irregular morphology and long semi-cylindrical particles, which could be explained by unavoidable particle agglomeration during the freeze-drying process. According to Nogueira [72], SEM analysis of freeze/spray-dried blackberry juice with starch as the WM revealed that freeze-dried powders had an irregular morphology resembling broken glass, with highly varied particle sizes. The authors also reported a larger particle size for freeze-dried powders in comparison to spray-dried powder counterparts. This characteristic was also seen in this study and was confirmed by particle size analysis (section 3.8).

3.6. FTIR spectra of microencapsulated powders

Variation in FTIR absorption bands of core and wall molecules, along with the absence and/or appearance of characteristic absorption bands, can provide supportive evidence for microencapsulation formation [73]. The characteristic bands and FTIR spectra of WMs (GGM, GX, GA, GGM + CMC and GX + CMC) and their microencapsulated powders (GGM: J, GX: J, GA: J, GGM + CMC: J and GX + CMC: J), and those of FD-J powders are reported in Table S2 and Fig. S1, respectively, in the supplementary materials. The FTIR spectra of GGM: J microencapsulated powders display characteristic absorption due to the O—H stretching vibration at 3350 cm^{-1}, shifted by 11 cm^{-1} from the value of GGM powders. The absorption band due to the asymmetric C—H stretching of $–CH_2$ groups appears at 1627 cm^{-1} for GGM: J microencapsulated powders, shifted from the value of GGM (1577 cm^{-1}). Similar changes can be seen in the FTIR spectra of GX: J and GA: J microencapsulated powders (Figs. S1b and c). These shifts could indicate possible interactions between WMs and bioactive compounds in the juice. Kalušević [74] reported that the FTIR bands of spray-dried microencapsulated powders of soybean anthocyanins with GA were shifted and/or showed bands originating from the WMs and phenolic compounds, which is evidence of microencapsulation. The bands in the FTIR spectra of GA and GA: J were consistent with previous studies on the microencapsulation of black raspberry anthocyanins with GA as the WM [75,76]. The peaks that appeared with low intensity at about 2918 cm^{-1} were characteristic of free carboxyl groups, which were negatively charged [76,77]. At first glance, the broader peak of GA: J at 3312 cm^{-1}, which was also shifted from the value of 3382 cm^{-1} for GA powders, could be evidence of the occurrence of possible interactions between the WM and bioactive compounds, according to Mansour [76].

3.7. Thermal analysis

The glass transition temperature (T_g) is an important property of spray-dried powders, as it determines the stability of the powders during long-term storage. When the storage temperature of amorphous powders is above their T_g, the powders become rubbery, increasing the molecular mobility and the rate of physicochemical changes such as collapse, caking, agglomeration, browning and oxidation. Thus, spray-

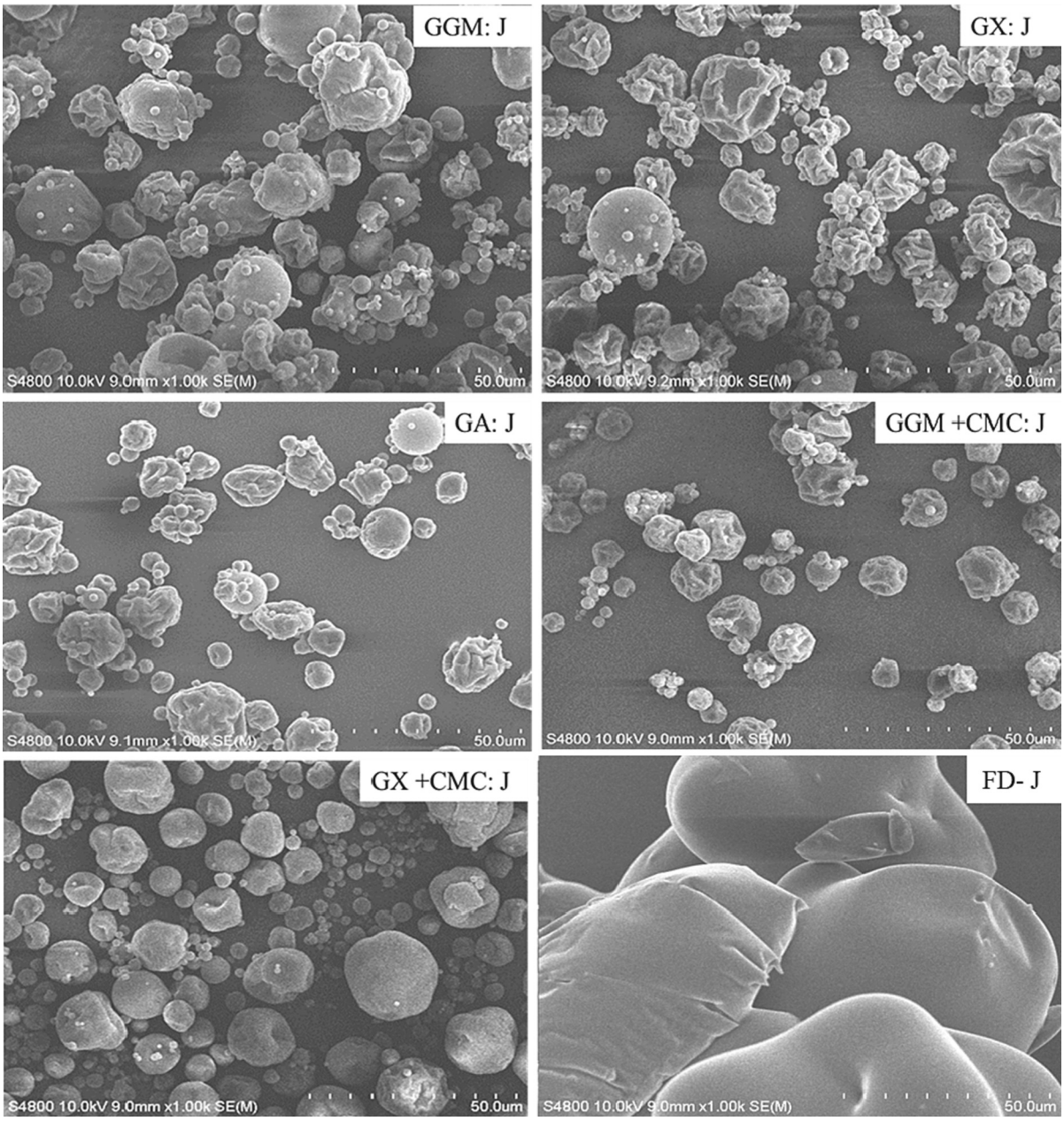

Fig. 3. SEM images of GGM: J, GX: J, GA: J, GGM + CMC: J and GX + CMC: J microencapsulated powders, and those of FD-J powders. The scale bar for all samples is 50 μm. Refer to Table 1 for sample codes.

dried powders should be stored at temperatures below their T_g [78]. The DSC curves of GGM: J and GX: J, microencapsulated powders presented in Fig. 4a reveal a major transition at about 50–60 °C. These transitions were suggested to be linked to the T_g of lignin presenting in hemicelluloses [79,80], followed by the enthalpy of relaxation of amorphous materials. A similar T_g result was also found for GA: J microencapsulated powders. As indicated in Fig. 4b, the T_g value of GGM + CMC: J and GX + CMC: J microencapsulated powders was 60–65 °C, which was slightly higher than that of GX: J, GGM: J and GA: J microencapsulated powders.

In addition, GX: J and GX + CMC: J microencapsulated powders had a tendency to form semi-crystalline regions upon heating to around 170 °C, which has also been reported for grafted birch xylans [81]. The tendency to crystallize was also observed for GGM: J and GGM + CMC: J powders but was less profound than GX: J and GX + CMC: J, which could be due to their differences in amorphous proportions. As mentioned above, crystalline proportions were not detected by XRD analysis, while they appeared in DSC upon heating. XRD analysis was performed at room temperature, while the analysis of DSC involved slowly heating the materials up to 200 °C. As the material temperature is higher than its T_g, the molecular mobility increases and the material tends to transform to its crystalline state [82].

The third change observed on the DSC curves of GX: J, GGM: J and GA: J microencapsulated powders was a sharp endothermic peak at around 170–180 °C, possibly originating from the melting of crystalline proportions. Another possible origin of the sharp endothermic peak was the decomposition of the powders, which was physically observed by opening the pan after the experiment had been completed.

3.8. Particle-size distribution

As indicated in Table 2, D[4,3] and D[3,2] values of all microencapsulated powders were 9.8–11.8 and 7.3–10.7 μm, respectively.

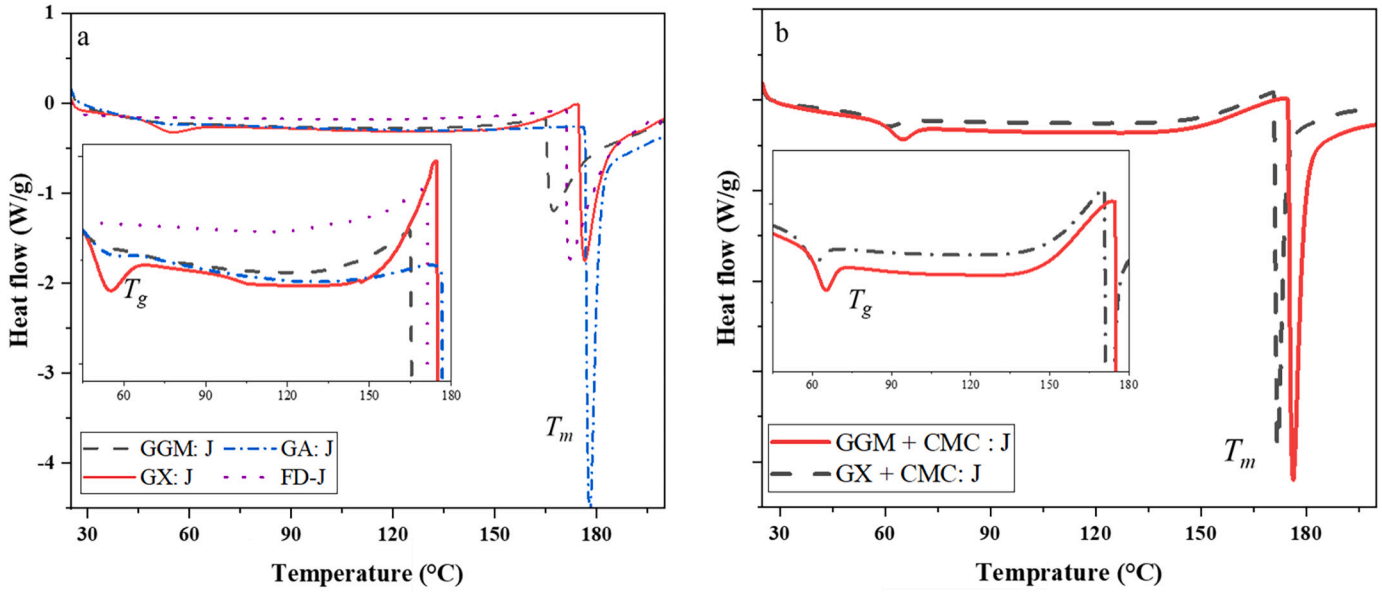

Fig. 4. DSC curves of microencapsulated powders prepared from (a) GGM: J, GX: J, GA: J and FD-J; and (b) GGM + CMC: J and GX + CMC: J. The small inset graphs represent the original graphs in the range of 50–180 °C, T_g: glass transition temperature, T_m: melting temperature of crystalline proportions and/or decomposition of the powders. Refer to Table 1 for sample codes.

Meanwhile, FD-J powders had much larger particles (D[4,3] and D[3,2] = 93 and 152 μm, respectively), implying particle agglomerations, which was confirmed by SEM analysis. This could be attributed to the nature of freeze drying and spray drying. Typically, spray drying produces a smaller particle size compared to freeze drying. This is because spray drying applies powerful atomization, whereas in freeze drying, the final particle size is dependent on the grinding procedure instead of the drying process [83]. The larger powder particle size produced by freeze drying than spray drying was also reported by Guo [84] and Pellicer [85]. The results were in line with previous similar studies such as microencapsulated powders of maltodextrins with anthocyanins obtained from banana (2–10 μm) [86] and carrot (3–20 μm) [71].

GGM + CMC: J and GX + CMC: J microencapsulated powders had shown a slightly larger particle size than that of GX: J, GGM: J and GA: J microencapsulated powders, which is possibly because of the higher viscosity of feed solutions prepared with CMC. Pang, Yusoff & Gimbun [87] reported that at the same solid concentration (10.67%), high viscosity feed solutions (whey protein isolate) induced a larger particle size of microencapsulated powders than low viscosity feed solutions (maltodextrin). As indicated in Fig. 5, the particle-size distribution of GX: J, GGM: J and GA: J microencapsulated powders exhibited similar monomodal curves, with the main peaks at around 10 μm. GGM + CMC: J and GX + CMC: J samples also had a monomodal curve, but their main peaks were at around 10–15 μm.

3.9. Solubility

As shown in Table 2, the solubility of all microencapsulated powders was very high, ranging from 91 to 99%. The lowest solubility was found for GGM: J microencapsulated powders, while the highest solubility was seen for GA: J and FD-J. This could be attributed to the fact that GA and FD-J components have superior water solubility [88]. The presence of CMC increased the solubility of GGM + CMC: J and GX + CMC: J microencapsulated powders, which is due to the full dissolution of CMC in water. It is well reported that GGM and GX powders naturally contain insoluble fractions, and GX: J has less insoluble fractions than GGM: J [60]. As compared to previous studies, the solubility of the powders in this study was similar to that of the microencapsulated powders of flexirubin and cagaita fruit extracts with GA (95–98%) [89,90], but higher than that of the microencapsulated powders of black mulberry juice with maltodextrin and GA, mulberry juice with GA, and blackberry juice with starch and GA (79–87%) [15,91]. The differences in solubility can be explained by the chemical structure of the used WMs. The occurrence of water adsorption by polysaccharides is attributed to the links between hydrogen present in water molecules and hydroxyl groups available in substrates such as polysaccharides [57].

3.10. Color

Color L* and a* and b* values, and images of GGM: J, GX: J, GA: J, GGM + CMC: J, GX + CMC: J microencapsulated powders, and those of FD-J powders are presented in Table 2 and Fig. 6, respectively. The

Table 2
Moisture content, water activity, solubility, particle size and color of microencapsulated powders prepared from GGM: J, GX: J, GA: J GGM + CMC: J and GX + CMC: J, and those of FD-J powders.

Samples	Moisture content (%)	Water activity (−)	Solubility (%)	D [4,3], μm	D [3,2], μm	a* (−)	L* (−)	b* (−)
GGM: J	6.07 ± 0.61^{a}	0.25 ± 0.03^{a}	91.4 ± 0.26^{c}	11.8 ± 0.9^{a}	7.96 ± 0.70^{a}	37.8 ± 1.81^{a}	28.7 ± 1.55^{b}	0.87 ± 0.08^{b}
GX: J	5.92 ± 0.23^{a}	0.23 ± 0.03^{a}	95.1 ± 0.20^{d}	11.03 ± 1.36^{a}	7.38 ± 0.86^{a}	29.7 ± 1.32^{d}	32.3 ± 2.43^{b}	−2.26 ± 0.44^{a}
GA: J	5.90 ± 0.20^{a}	0.24 ± 0.02^{a}	99.7 ± 0.05^{a}	9.88 ± 1.91^{a}	10.0 ± 1.97^{a}	41.2 ± 1.22^{a}	32.5 ± 1.09^{b}	−2.34 ± 1.09^{a}
GGM + CMC: J	5.39 ± 0.67^{a}	0.21 ± 0.02^{a}	95.4 ± 0.23^{bd}	10.9 ± 2.65^{a}	10.6 ± 2.66^{a}	19.4 ± 0.90^{b}	39.5 ± 2.34^{a}	−2.81 ± 0.68^{a}
GX + CMC: J	5.06 ± 0.47^{a}	0.19 ± 0.02^{a}	96.5 ± 0.21^{b}	11.1 ± 2.91^{a}	10.7 ± 2.75^{a}	18.3 ± 0.71^{b}	37.9 ± 2.54^{a}	−3.3 ± 1.02^{a}
FD-J	6.24 ± 0.44^{a}	0.21 ± 0.04^{a}	99.7 ± 0.09^{a}	108 ± 33.5^{b}	59.5 ± 15.4^{b}	12.8 ± 0.43^{c}	2.14 ± 0.09^{c}	3.30 ± 2.21^{c}

The data are presented as means ± SD (n = 3), and means in the same column with different letters as superscripts are significantly different at $p < 0.05$. Refer to Table 1 for sample code.

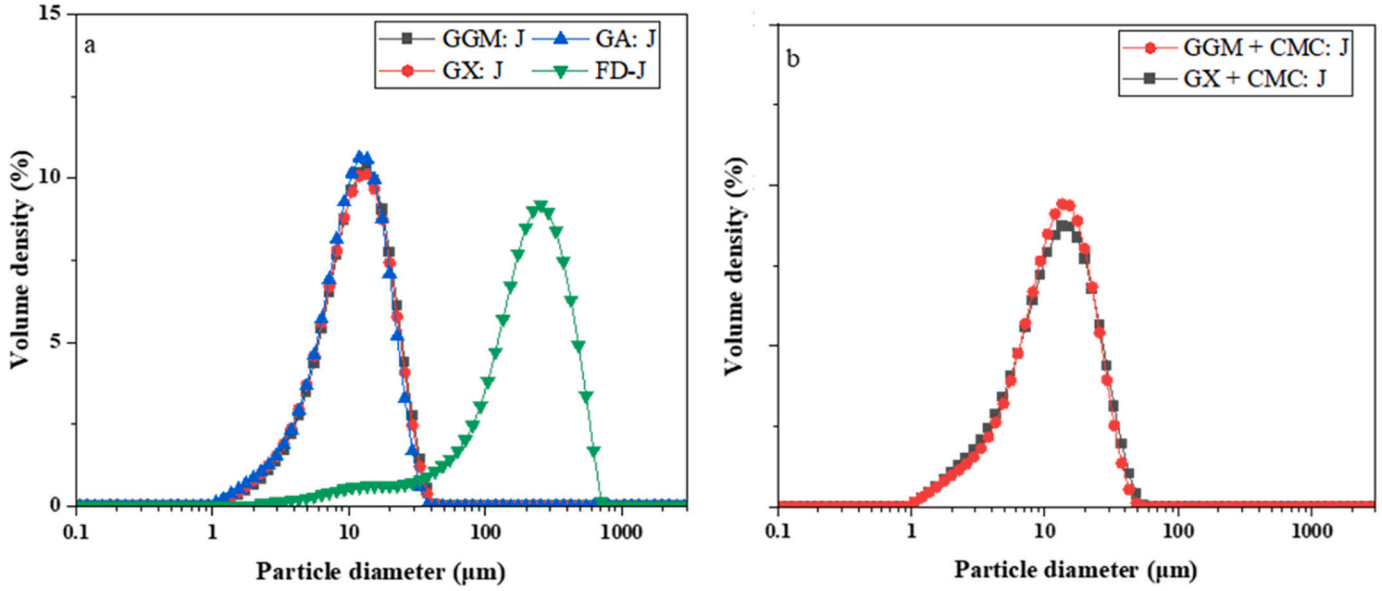

Fig. 5. Particle-size distribution of microencapsulated powders prepared from (a) GGM: J, GX: J, GA: J and FD-J, and (b) GGM + CMC: J and GX + CMC: J. Refer to Table 1 for sample codes.

results indicated that the FD-J powder had a dark color with a very low L* value, and that GGM: J, GX: J, and GA: J powders were higher in redness (+a*) than GGM + CMC: J, and GX + CMC: J powders. Microencapsulated powders with different WM formulations displayed statistically significant differences in a* values. The addition of CMC to GGM and GX increased L* and b*values but decreased the a* value of their microencapsulated powders (GGM + CMC: J, and GX + CMC: J) as compared to their counterparts without CMC (GGM: J and GX: J). This could be due to lower anthocyanin retention in microencapsulated powders of GGM + CMC: J, and GX + CMC: J and a higher ratio of WM to core material, which caused a dilution effect.

According to de Araujo [92], a* values within the range of red can confirm the presence of anthocyanins in the produced powders. In this study, the correlation coefficient between a* values and the anthocyanin content in microcapsule powders was >0.9 (data not shown). This observation was similar to that reported by de Araujo [92] and Villacrez [93], who investigated the spray-dried microencapsulation of Andes berries and pomegranate, applying different WMs (maltodextrin, GA and corn starch). They established a proportional correlation between the a* parameter values and anthocyanin content. Color is one of the important factors that attracts consumers to food. The microencapsulated powders of the bilberry fruit compounds can potentially be used as a natural red colorant in the food industry.

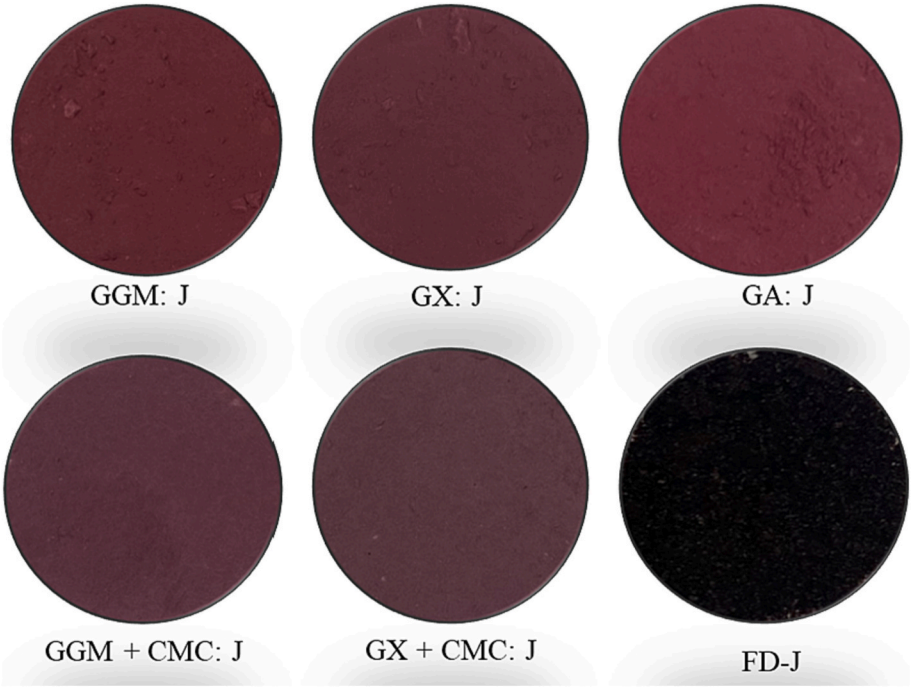

Fig. 6. Images of GGM: J, GX: J, GA: J, GGM + CMC: J, GX + CMC: J microencapsulated powders, and those of FD-J powders. Refer to Table 1 for sample codes.

3.11. Moisture content and water activity

The moisture content and water activity of microencapsulated powders prepared from different WMs are presented in Table 2. All the samples yielded water activity values below 0.3, which indicated high stability of the powders to microorganism growth, biochemical reactions and long-term storage [94]. The moisture content of all microencapsulated powders varied between 5.0 and 6.0%. The results are similar to the reported values for spray-dried cranberry juice with GA and maltodextrins (3.78–7.24%) [95,96] and mulberry with either maltodextrins or GA (1.5–3%) [91,97]. The narrow range of water activity and moisture content in the current study could be explained by the use of identical spray drying conditions (e.g., inlet and outlet temperatures and aspiration rate).

4. Conclusions and future directions

In this study, it has been demonstrated that wood-based hemicelluloses, GGM and GX, are efficient WMs for the spray-dried microencapsulation of bilberry juice. The EE values of GGM and GX were relatively high and close to that of GA. The use of CMC as a co-WM reduced the EE of GGM and GX. However, optimization of the concentration of added CMC could improve the EE, and this requires further investigation. Due to the natural presence of lignin-derived phenolic compounds in the structure of GGM and GX, their microencapsulate powders had significantly higher TPC and antioxidant activities than GA powders. The produced microencapsulated powders had an amorphous structure, irregular spherical morphology and very high water solubility.

The findings of this study provide another value-added application of wood hemicelluloses obtained from waste and by products of the forest industry. Wood hemicelluloses can replace the currently used wall materials in the production of high quality bilberry powders via a more cost effective method (i.e. spray drying) than commonly-used ones (freeze drying and freezing), thus offering an economic opportunity to bring bilberry products to global consumers. Wood hemicelluloses not only effectively protect the bioactive compounds against heating impact during spray drying, but also bring benefits of adding dietary fibres and antioxidant functionality to the final products. This study is also expected to enable the production of stable microcapsules of many other core functional compounds (such as essential oils, polyunsaturated fatty acids, bioactive peptides) and benefit microorganisms using wood hemicelluloses, which eases their utilization in many food, pharmaceutical and cosmetic products, to meet the increasing demand for natural and functional products.

This study was one of first studies to investigate spray drying of bilberry using wood hemicelluloses as wall materials, thus a fixed spray drying condition regarding solid concentrations of feed solutions and drying temperatures was explored. This could impair the EE, bioactive compounds, and functionality of produced bilberry powders. Therefore, an optimization of these parameters is required for further studies to maximize the encapsulation efficiency and ability to protect bioactive compounds. In addition, due to the amorphous structure and high hygroscopicity of bilberry powders, further studies on their storage stability as well as bioavailability is also needed to provide insight of their long-term storage and applications. Wood hemicelluloses with the presence of lignin-derived phenolic compounds might affect the flavor of the berry juice powders, thus the sensory evaluation of bilberry powders is also considered in further studies.

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CRediT authorship contribution statement

Abedalghani Halahlah: Conceptualization, Methodology, Investigation, Formal analysis, Writing – original draft. **Heikki Räikkönen:** Methodology, Investigation, Formal analysis. **Vieno Piironen:** Supervision, Writing – review & editing. **Fabio Valoppi:** Writing – review & editing. **Kirsi S. Mikkonen:** Conceptualization, Supervision, Writing – review & editing. **Thao M. Ho:** Conceptualization, Methodology, Supervision, Funding acquisition, Writing – review & editing.

Declaration of Competing Interest

There are no conflicts of interest to declare.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.powtec.2022.118148.

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