Enrichment of dark chocolate with free and microencapsulated white tea and jujube extracts: Impacts on antioxidant, physicochemical, and textural properties

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Abstract

In this study, dark chocolate was enriched with white tea (WT) and jujube extracts in form of free extract and microcapsules. Each form of two extracts was added to dark chocolate in concentrations of 1, 3, 5, and 7% w/w. The microcapsules were produced using pectin/casein (10% w/w), and their particle size distribution (PSD) and Fourier transform infrared spectroscopic analysis were performed. Phenolic content, antioxidant activity, pH, moisture content, color parameters and texture of enriched dark chocolates were measured. As expected, addition of extracts caused a significant increase in total phenolic content and antioxidant properties of dark chocolates compared to the control sample (no extract added) in both free and microencapsulated forms, with a stronger effect of WT extract than jujube extract.

Keywords: dark chocolate; jujube; microcapsules; phenolic; white tea

Introduction

Chocolate is a functional product because of its high level of flavonoid compounds and positive effects on human health. Recent decades have witnessed increased consumer demand and expectations from the food market, so that they have more choices than in the past. For instance, the global chocolate market grew by about 5% through 2020 (Chen *et al.*, 2022). Accordingly, organic chocolates, polyphenol-rich cocoa chocolates, probiotic chocolates, and prebiotic chocolates can be found in the market. In fact, now, interest in functional foods has increased due to their positive health effects as well as nutritional values (Tolve *et al.*, 2018).

According to European laws, dark chocolate is a confectionery product containing at least 35% dry cocoa solids, 18% cocoa butter and 14% non-fat cocoa solids with any added sugar (Saputro *et al.*, 2017). Cocoa and cocoa products, such as dark chocolates, are rich in flavonoids, with procyanidins being the most abundant chemical compounds among other polyphenols (Kim *et al.*, 2014; Shin *et al.*, 2022). In addition to their antioxidant and anti-inflammatory properties, cocoa polyphenols have shown beneficial cardiovascular, metabolic, antiradical and anti-dermatological effects as well as eliciting effects on mood (Shin *et al.*, 2022; Sim *et al.*, 2016). The polyphenol contents of cocoa beans and their products depend on the region in which they are grown and the processes to which they are subjected to, such as temperature (Urbańska and Kowalska, 2019). One of the best methods to enhance flavor and phenolic content of chocolate is to add polyphenolic compounds derived from natural sources (Belščak-Cvitanovic *et al.*, 2012). In

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this study, Jujube and white tea (WT) extract were analyzed as sources of natural polyphenolic compounds for formulation of dark chocolates.

Jujube, *Ziziphus jujube* Miller, belongs to the Rhamnaceae family. *Ziziphus* is derived from a Hebrew word. Jujube tree, which is one of the native plants of Iran, is found in Khorasan, Golestan, Mazandaran, Fars, Isfahan, Yazd, Hamedan and Kerman provinces. However, the province of South Khorasan in Iran leads in terms of the area under cultivation and production of jujube (Shams Najafabadi *et al.*, 2017).

Jujube fruit is a rich source of nutritional compounds and vitamins, and thus known as the "king of vitamins." Besides this, it is a dietary supplement with high contents of bioactive compounds (Niu *et al.*, 2021); and because of potential antioxidant activity, it can be used as a natural antioxidant ingredient in foods and pharmaceuticals (Liu *et al.*, 2020; Rashwan *et al.*, 2020). Studies have demonstrated that jujube has anticancer, anti-inflammatory, anti-diabetes, and cardioprotective characteristics, with liver protective, antioxidant, and antiinsomnia properties. Jujube also protects digestion and stimulates the nervous and immune systems (Guo *et al.*, 2015; Rashwan *et al.*, 2020; Wang *et al.*, 2016). Jujube contains considerable amounts of carbohydrates, minerals, vitamins, fibers, amino acids, fatty acids and phenolic compounds, such as gallic, chlorogenic, and caffeic acids, which are considered the main components of human health (Bahrasemani Koohestani *et al.*, 2019). Jujube fruit is classified as a "functional food" because of health benefits that could reduce the risk of specific chronic disease as well as affects target functions beyond its basic nutritional functions (Astrini *et al.*, 2020).

Tea can be divided into three main types, such as unfermented (green and white tea), semi-fermented (Oolong tea) and fully fermented (black) tea. WT is prepared from very young tea leaves harvested once a year. The leaves are steamed and dried after harvesting to prevent oxidation and produce light and pleasant taste (Alcazar *et al.*, 2007; Liczbiński and Bukowska, 2022).

White tea contains several biologically active compounds that are believed to have a wide range of physiological properties, including stimulant, antidepressant, anti-inflammatory, antioxidant, antimicrobial, antihypertensive, anti-infective, anti-diabetic anti-mutant, and anticancer characteristics. WT decreases blood lipids, protects the nervous system, and improves immune responses (Dias *et al.*, 2013). Although one of the less-studied ingredients, WT contains the highest level of phenolic compounds among all types of tea (Liczbiński and Bukowska, 2022).

Since most phenolic and bioactive compounds in plant extracts and essential oils are biologically unstable, their stability and bioavailability can be improved using new methods, such as encapsulation. Polyphenols are sensitive to heat, light, oxygen, acidic pH and water (Delfanian and Sahari, 2020). The encapsulation technique involves creating a coating layer around the bioactive material to protect sensitive compounds and preserve them from loss during processing or storage (Ozkan *et al.*, 2019). This method is an efficient mode to of improve the delivery system of sensitive ingredients and enabling their controlled release (Laličić-Petronijević *et al.*, 2017; Reis *et al.*, 2022). Besides this, phenolic compounds when added directly to chocolate formulations impart undesirable sensory properties such as flavor, taste and color (Delfanian and Sahari, 2020). Therefore, in this study, microencapsulated forms of WT extract and jujube extract (JE) were prepared to enrich the dark chocolate formulation in comparison to addition of free form.

The results of literature review indicate that no study has been conducted on the enrichment of dark chocolate with WT and jujube extracts in microencapsulated or free forms. Therefore, this study aimed to produce functional dark chocolate containing free and microencapsulated jujube and WT extracts and evaluate its physicochemical, antioxidant and texture properties.

Materials and Methods

Extraction of extracts

White tea (*Camellia sinensis*) was purchased from Timan Company (Lahijan Welfare Tea, Iran). Aqueous extract of WT was prepared by mixing WT leaves in distilled water in 1:10 ratio at 100°C. The resulting mixture was boiled with continuous stirring for 10 min and solid particles were separated using Buchner funnel and Whatman filter paper (No. 4). The obtained extract was dried at 60°C and stored at 4°C for subsequent experiments. Jujube fruit was dried in ambient conditions, powdered in an electric mill with 40 mesh sieves, and kept at -20°C until extraction (less than 3 days). Aqueous extract of jujube was prepared by mixing jujube powder in distilled water in 1:10 ratio at 100°C. The resulting mixture was boiled for 10 min, passed through Buchner funnel and Whatman filter paper (No. 4), dried at 60°C, and refrigerated for subsequent experiments (Saral *et al.*, 2019).

Production of microcapsules

To prepare microcapsules, the polymers of pectin (Sigma Aldrich, Germany) and casein (Merck, Germany) were

suspended in distilled water (10% w/v) under constant mechanical vibration (on a magnetic hot plate; FALC, Italy). Sodium hydroxide (4 M) was added to adjust pH to 8.0 ± 0.1. After complete suspension, the polymer and extract were added in 1:1 ratio. The microcapsules were obtained by gradually lowering of pH from 8 ± 0.1 to 3 ± 0.1 with 1-M citric acid and applying an additional constant vibration for 30 min. The samples were then freeze-dried (Model DW1.0-110; Heto-Holten A/S, Denmark) (Baracat *et al.*, 2012).

Preparation of chocolate samples

In order to prepare chocolate, sugar was first crushed in a domestic grinder and passed through a 20–38-µm laboratory sieve. For 100 g of dark chocolate, 20 g of cocoa butter (Farmand Co., Iran) was first melted in an oven at 60°C; then 26 g of cocoa powder (Farmand Co., Iran) and sugar were added to the melted cocoa butter (up to 100 g of formulation). The resulting mixture was placed in a blender at 50 rpm for 4 h and then in a paraffin bath at 65°C with mild agitation for 24 h for conching process. Temperature of the prepared sample were reduced to 45°C, and maintained for 30 min. Then the sample was exposed to 28°C for 10 min, finally raising temperature to 32°C in order to vaporize IV crystals (Chen *et al.*, 2022). The prepared sample was immediately transferred to a plastic mold, stored at 15°C for 30 min, and finally packed in an aluminum foil under refrigerated conditions. To produce enriched chocolate, WT and jujube extracts were added to the chocolate formulation at four levels (1, 3, 5 and 7%) in free and microencapsulated forms during the mixing stage of formulation.

Measurements

Particle size distribution of microcapsules

Particle size was measured using a dynamic light scattering (DLS) device (FRITSCH ANALYSETTE 22, Nano Tec, Germany). Approximately 5 g of sample was suspended in 50 mL of hexane at ambient temperature (25°C). The sample was dispersed ultrasonically for 2 min to ensure its separate suspension. The resultant obtained from the device was used to calculate and report parameters of the largest (D90) and the smallest (D10) particle sizes on a micrometer scale (Sim *et al.*, 2016).

Fourier transform infrared spectroscopy (FTIR) test of microcapsules

The sample was measured with an FTIR device (Spectroma 2 Perkin-Elmer, USA) in a wave range of 500–4000 cm^{-1} to obtain the required peaks (Lončarević *et al.*, 2018).

Sample preparation for total phenol test and antioxidant activity

Samples were prepared to determine the content of phenolic compounds and antioxidant activity according to Todorovik *et al.* (2015). Chocolate samples were refrigerated and crushed manually in a mill. To remove fat, 0.2 g of each product was extracted thrice with 10 mL of N-hexane. The defatted samples were air-dried for 24 h to evaporate the remaining solvent. The required compounds and polyphenols were defatted from cocoa and extracted twice with 5 mL of extraction solvent (70 + 29.8 + 0.2 v/v/v of acetone + distilled water + acetic acid) in an ultrasound for 30 min. The extracted mixture was centrifuged at 3,000 rpm for 10 min. After filtration, the supernatant was used to measure total phenolic content and antioxidant activity (Todorovic *et al.*, 2015).

Measurement of total phenolic content

Total phenolic content was determined by the Folin–Ciocalteu method. Extract sample (100 µL) was mixed with 100 µL of Folin–Ciocalteu reagent. Then 1,000 µL of 20% sodium carbonate and 8.8 mL of distilled water were added to the mixture, kept in dark for 30 min, and the absorbance was read using a spectrophotometer at 700 nm. Different dilutions of gallic acid (25–250 mg L^{-1}) were used as a standard, and the results were calculated as equivalent of gallic acid (mg) per gram of chocolate (Godocikova *et al.*, 2017).

Antioxidant activity

The free radical scavenging activity of the produced chocolate samples was measured by spectrophotometry based on the decolorization of 2,2-diphenyl-1-picrylhydrazyl (DPPH) purple solution as a reagent. Briefly, 1 g of the product was dissolved in 10 mL of distilled water, and then 100 µL of this solution was mixed with 4 mL of DPPH ethanol solution (25 mg/g). The samples were kept in dark at room temperature for 1 h, and their absorbance was measured at 517 nm by a spectrophotometer (Model T60 UV, USA). The inhibitory percentage of DPPH free radicals by the produced oral films was calculated by using Equation (1) (Ruiz-Navajas *et al.*, 2013).

$$I\,(\%) = \frac{A_{\text{Control}} - A_{\text{Sample}}}{A_{\text{Control}}} \times 100 \qquad (1)$$

In Equation (1), A_{control} is the control absorbance rate (DPPH methanol solution at 517 nm) and A_{sample} is the absorbance rate of chocolate sample containing free extract or microcapsules. Three replications of each sample were measured.

Color features

The color of chocolate samples was determined by measuring the color components of L^* (lightness), a^* (redness/greenness), b^* (yellowness/blueness) and ΔE (total color difference) using a Hunterlab colorimeter (Minolta

model CR-410; Tokyo, Japan) standardized with a white tile (L^* = 98.14, a^* = -0.23, and b^* = 1.89). The samples were placed on a standard white screen, and the amount of color was measured using Hunter parameters with three replications for each sample, and the results were reported as a mean of data.

Moisture content and pH

Moisture content and pH of the samples were measured by the Karl Fisher method (Association of Official Analytical Chemists [AOAC], 2005) and the AOAC (2005) method, respectively.

Textural characteristics

For this test, chocolate samples were incubated at 20°C for 2 h. The hardness of chocolate samples (5 × 40 × 25 mm) was tested using a texture analyzer (model TA PLUS Texture Analyzer, USA) equipped with a smooth-bottom mandrel (piston) with a diameter of 1.6 mL and a speed of 90 mm per min. The maximum force at a depth of 4 mm was reported as hardness. A load cell of 500 N was used in this experiment (Farzanmehr *et al.*, 2018).

Scanning Electron Microscopic (SEM) monitoring test

The microstructure and morphology of chocolate samples were examined by SEM (XL30 Philips Company, the Netherlands) with a voltage of 26 kV and a magnification of 4,000–12,000× (Gultekin-Ozguven *et al.*, 2016).

Statistical analysis

The results were evaluated with a completely randomized statistical design using a factorial method. Data were statistically analyzed using the SAS statistical software, version 9.1. Mean values of treatments were compared using Duncan's multiple range test at an alpha probability level of 0.05 ($P < 0.05$) using the same software. Processing variables, including the type of extract (free and microencapsulated) and concentration levels (1, 3, 5, and 7%), were performed in three replications.

Results and Discussion

Particle size distribution of microcapsules

Particle size and distribution of microcapsules play an important role in the physical properties of colloidal systems, such as stability during storage, turbidity and rheological properties, and are effective in attributes, such as bioavailability and organoleptic and sensory properties, of the contained foods (Lončarević *et al.*, 2018). In this study, the microencapsulation efficiencies of WT and jujube extracts were 84.1% and 74.5%, respectively. The results of particle size distribution analysis by DSL showed that the average diameter of microencapsulated particles containing WT extract (D50 = 21.84 ± 0.55 μm) was smaller than that of jujube extract (D50 = 53.5 ± 0.65 μm) (Table 1). This could be due to different chemical composition of two extracts. The particle diameter of microcapsules depends on the material properties, concentration, viscosity of microencapsulation material, and the dryer operating conditions (Emadzadeh *et al.*, 2021; Junior *et al.*, 2018). Besides this, researchers suggested that increasing the ratio of pectin in mixture increased the particle size of emulsion because of the presence of nonabsorbable pectin on the surface of oil particles, resulting in aggregation and increased particle size (Azizanbari *et al.*, 2013; Emadzadeh *et al.*, 2021).

Table 1. Particle size parameters of microencapsulated white tea extract (MW) and microencapsulated jujube extract (MJ).

MJ	MW	Particle size parameters (μm)
3.11±0.05	2.3±0.08	D_{10}
53.5±0.65	21.84±0.55	D_{50}
261.17±1.4	157.23±1.87	D_{90}

Tolve *et al.* (2018) evaluated the chemical and sensory properties of dark chocolates enriched with different concentrations of microencapsulated phytosterols, and established that diameter D90 values of all chocolate samples were in the range of 19–25 μm; this was a good result because a desired dark chocolate requires particles with a maximum diameter size of 35 μm. Deou *et al.* (2022) reported that particle size reduction in a defined domain in chocolate formulation provided stability against sedimentation and migration phenomena during its shelf life. In addition, a smaller particle size in chocolate improves sensory properties. The particle size distribution is significant because it can be optimized without general changes in chocolate formulation to achieve the desired rheological properties (Tolve *et al.*, 2018).

FTIR analysis of microcapsules

FTIR spectroscopy is a widely used method to analyze and identify polymers and some of their additives. Each substance has a specific infrared spectrum and is specific to that molecule—the same as a fingerprint. Almost all covalently bonded organic or inorganic compounds absorb different frequencies of electromagnetic radiation from the infrared region (Choo *et al.*, 2016). In this study, FTIR spectroscopy was used to detect and confirm the formation of microcapsules and to create an interaction between extracted polyphenolic compounds and capsule coating. The FTIR test results of free and microencapsulated WT and jujube extracts are depicted in Figure 1.

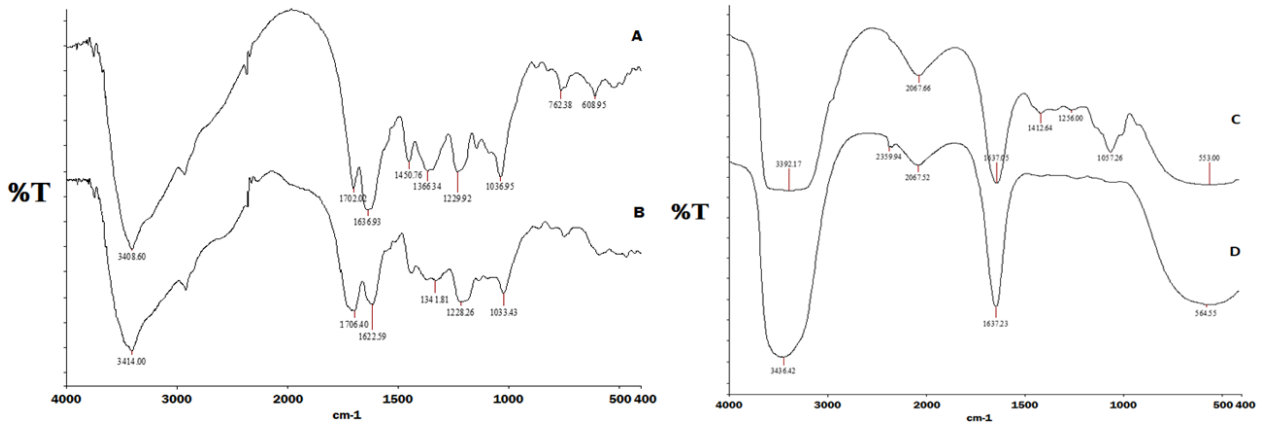

Figure 1. FTIR patterns of (A) white tea extract, (B) microencapsulated extract of white tea (MW), (C) jujube extract and (D) microencapsulated jujube extract (MJ).

In Figure 1(a), WT extract has absorption peaks in different areas, so three strong peaks are observed in the range of 3408.6, 1639.93, and 1036.95 cm^{-1}, which belong to C–H and C–O groups. The peaks observed at 3408.6 and 1639.93 cm^{-1} corresponded to hydrogen-bonded alcohols, phenols and C=O in aldehyde, ketone, carboxylic acid, and ether, respectively. The peak observed at 1036.95 cm^{-1} related to C=C group in alkene. In Figure 1(b), characteristic peaks of the microencapsulated extract of WT included three strong peaks at a wavelength of 1706.4 cm^{-1} corresponding to the C=O group in ether, carboxylic acid, aldehyde and ketone. Wave number 1341.81 cm^{-1} belonged to the C-H group in alkanes, and wave number 1228.26 cm^{-1} belonged to the C-O group in alcohol, ether and carboxylic acid. The absorption peak of 1622.59 cm^{-1} belonged to the C=C group in alkene, and peak at 3414 cm^{-1} was variable and somewhat wide in alcohol with hydrogen bonding and phenols (Pradini *et al.*, 2018).

Jujube extract also had several absorption peaks in different regions (Figure 1[c]). The absorption band of 3392.17 cm^{-1} represented the stretching vibrations of OH group in phenolic compounds; the absorption band of 2067.66 cm^{-1} belonged to alkaloids with C-F structure; the absorption band of 1637.05 cm^{-1} related to the NH2 functional group of amides; wavelength of 1412.64 cm^{-1} was characteristic of C-H in alkanes; and wavelength range of 1057.26–1256 cm^{-1} characterized the C-O group of ethers, alcohols and carboxylic acids. In Figure 1(d), the peaks of microencapsulated jujube extract (MJ) included the wavelength of 3436.42 cm^{-1} belonging to the OH group of phenolic compounds of extract; the wavelength of 2067.52 cm^{-1} represented the CF group of alkaloids; and the wavelength of 1637.23 cm^{-1} related to the NH2 structure of amides. Peak at a frequency of 2359 cm^{-1} was unexpected and probably was due to presence of CO_2 gas in microcapsules (Choo *et al.*, 2016). According to the above-stated data and comparison of absorption peaks of the patterns related to free and microencapsulated extracts, it could be concluded that microcapsule formation changed or eliminated absorption at some areas compared to free extract, indicating interaction between microcapsule wall constituents and compounds of WT and jujube extracts. SEM results also confirmed these interactions.

Total phenol content (TPC)

As secondary metabolites, phenolic compounds are an important group of plant compounds formed in response to environmental stresses. Owing to their hydroxyl groups, these compounds can counteract free radicals and act as electron or hydrogen donors (Lončarević *et al.*, 2019). These compounds also show antioxidant activity by inhibiting the decomposition of hydroperoxides to free radicals (Godocikova *et al.*, 2017). The results showed that chocolate samples containing free and microencapsulated extracts of WT generally contained more phenolic compounds than samples containing jujube extract and the control sample, with a statistically significant difference ($P < 0.05$). By free form addition of WT and jujube extracts, results suggested a higher polyphenolic concentration for WT rather than jujube extract. However, in encapsulated form, another factor that could be responsible was the water absorption rate of coatings. As reported by other studies, the rate of release of phenolics increased with increase in water absorption rate of coatings (Delfanian and Sahari, 2020). Considering the higher moisture content of MJ versus microencapsulated white tea extract (MW) (as seen in Table 2), polyphenolics release of MW was more sustained and delayed. It was also observed that the effect of encapsulation was higher on increasing phenolic compounds in dark chocolate than free extract; so dark chocolate with microencapsulated extract contained more phenolic content than the sample containing free extract for all treatments

Table 2. Texture, moisture content, and pH of dark chocolate enriched with free extracts of jujube and white tea as well as microencapsulated extracts of jujube and white tea in different concentrations.

	Moisture content (%)				pH			
	1%	3%	5%	7%	1%	3%	5%	7%
Sample								
CH	0.067±0.006^{e*}	–	–	–	7.58±0.08^{a}	–	–	–
CH-WE	0.211±0.05^{d}	0.235±0.03^{d}	0.243±0.05^{d}	0.254±0.08^{c,d}	7.41±0.05^{a,bc}	7.38±0.03^{b,c}	7.33±0.01^{b,cd}	7.18±0.04^{d}
CH-MW	0.283±0.01^{c,d}	0.356±0.04^{b,c}	0.418±0.02^{b}	0.437±0.05^{b}	7.46±0.06^{a,bc}	7.49±0.07^{a,b}	7.41±0.02^{a,bc}	7.5±0.08^{a,b}
CH-JE	0.204±0.04^{d}	0.239±0.02^{d}	0.231±0.01^{d}	0.267±0.03^{c,d}	7.44±0.05^{a,bc}	7.35±0.12^{b,c,d}	7.28±0.07^{c,d}	7.33±0.06^{b,cd}
CH-MJ	0.812±0.01^{a}	0.837±0.03^{a}	0.844±0.01^{a}	0.883±0.02^{a}	6.12±0.08^{e,f}	6.24±0.01^{e}	6.15±0.04^{e,f}	6.02±0.11^{f}
Stiffness (N/m)								
CH	64104.1±580^{a}	–	–	–	–	–	–	–
CH-WE	49387.5±423^{d}	47553.4±311^{e}	5892.2±842^{e,f}	44968.8±690^{f,g}	–	–	–	–
CH-MW	35586.6±188^{h}	31456.3±322^{i}	25879.5±745^{k}	25048.1±422^{k}	–	–	–	–
CH-JE	54289.3±749^{b}	51756.9±254^{c}	0882.6±852^{c,d}	43658.6±984^{g}	–	–	–	–
CH-MJ	28434.8±356^{j}	25474.3±241^{k}	4892.5±175^{k}	20658.6±398^{L}	–	–	–	–

CH: dark chocolate-control sample; CH-WE: dark chocolate-white tea extract; CH-MW: dark chocolate-microencapsulated white tea extract; CH-JE: dark chocolate-jujube extract; CH-MJ: dark chocolate-microencapsulated jujube extract.
*Superscripted alphabets demonstrating statistical significant differences.

(Figure 2). This was due to the increased stability of phenolic compounds in microencapsulated state relative to free extract during the thermal process of producing dark chocolate (Delfanian and Sahari, 2020; Jiang *et al.*, 2020). In addition, the amount of phenolic compounds in dark chocolate also depended on extract concentration, signifying that the amount of phenolic compounds showed a significant upward trend with increasing concentration of free or microencapsulated extract.

According to the results, measured values for control (CH), WT 7%, JE 7%, MW 7% and MJ 7% were 33.2 ± 3.57 mg/g (gallic acid), 98.45 ± 2.5 mg/g, 72.46 ± 1.3 mg/g, 112.32 ± 2.8 mg/g and 88.23 ± 2.4 mg/g respectively. It suggested that encapsulation improved the stability of phenolic compounds in dark chocolate formulation with WT and jujube extracts by 14.1% and 21.7%, respectively (Figure 2). Results of other studies have confirmed the role of pectin-based encapsulation in protection and

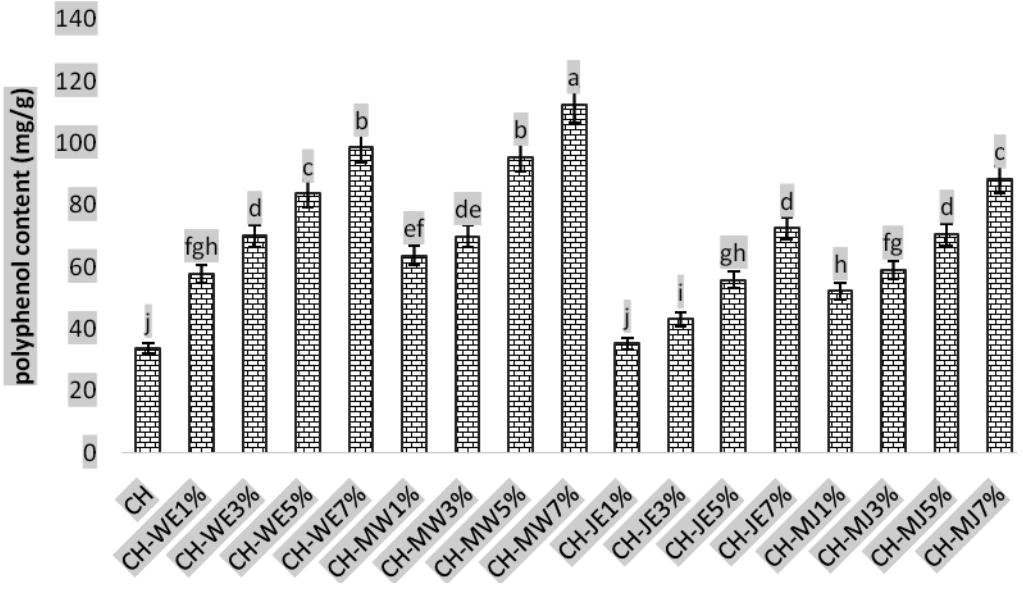

Figure 2. Effect of free and encapsulated extracts of white tea and jujube on the polyphenol content (mg/g) of dark chocolate. CH: dark chocolate-control sample; CH-WE: dark chocolate-white tea extract; CH-MW: dark chocolate-microencapsulated white tea extract; CH-JE: dark chocolate-jujube extract; CH-MJ: dark chocolate-microencapsulated jujube extract.

stability of phenolic and bioactive ingredients in different media (Delfanian and Sahari, 2020; Emadzadeh *et al.*, 2021; Jiang *et al.*, 2020).

In a similar study, Lončarević *et al.* (2019) examined the effect of green tea extract microencapsulation on the physical, sensory and functional properties of white chocolate, and reported that total phenolic content increased from 0.14 (GAE/kg) in white chocolate to 2.73 (GAE/kg) in chocolate enriched with 100 g/kg of green tea encapsulated extract. Sim *et al.* (2016) investigated the effect of polyphenols of *Garcinia mangostana* Linn plant on chocolate and found that it increased the functional properties of chocolate without changing its sensory properties. The authors reported that the addition of 3% pericarp powder increased polyphenol compounds by 13% and 50% in dark and combined chocolates, respectively. A study done by Godocikova *et al.* (2017) also revealed that total phenolic content was significantly higher in dark chocolate samples containing blackberry and service extracts than in control sample.

Antioxidant activity

The DPPH antioxidant assay is widely used to evaluate ability to inhibit or neutralize DPPH radical by antioxidants. The DPPH radical is more stable than hydroxyl and superoxide anion radicals, and this is one of its advantages (James *et al.*, 2019). Essential oils and extracts of medicinal plants, such as tea and jujube, also have a high potential to neutralize DPPH radicals because of their high levels of phenolic compounds and a high level of total antioxidant capacity (Liczbiński and Bukowska, 2022). The results showed that dark chocolate as a control had a remarkable antioxidant activity (62.45 ± 0.2), which increased significantly ($P < 0.05$) with the addition of different concentrations of free and microencapsulated extracts of WT and jujube. The highest inhibitory activity of dark chocolate was observed for the treatments containing 7% free and microencapsulated WT extracts (83.58 ± 1.00% and 85.89 ± 3.01%, respectively) (Figure 3). Accordingly, the samples containing microencapsulated extract showed higher antioxidant potential than those containing free extract because of the better preservation of phenolic compounds and their increased bioavailability. There is a direct relationship between the concentration of polyphenols and antioxidant activity (Delfanian and Sahari, 2020), because at high concentrations, the number of hydroxyl groups increases in the structure of phenolic compounds. This increases the probability of donating hydrogen to free radicals, thereby increasing the inhibitory potential of the extract (Zhang *et al.*, 2010). The present results were consistent with those of James *et al.* (2019), who reported that the free extract of green tea showed less antioxidant activity than the microencapsulated extract at storage temperature. Researchers observed a positive correlation between the total phenolic content of jujube and its antioxidant properties (Rashwan *et al.*, 2020; Zhang *et al.*, 2010), which was also observed in dark chocolates containing free and encapsulated jujube extract in the present study.

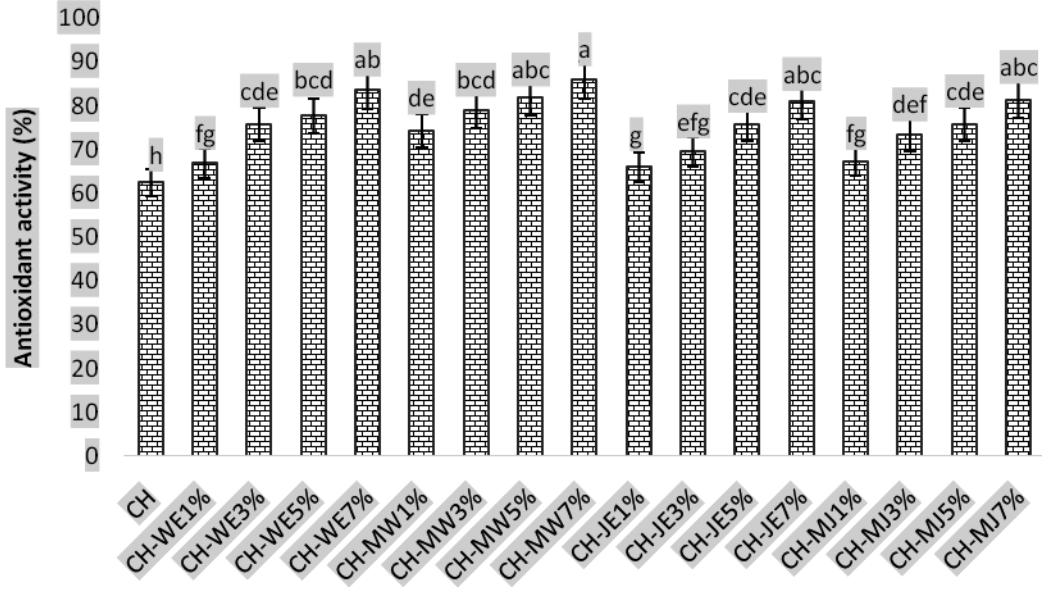

Figure 3. Effect of free and encapsulated extracts of white tea and jujube on the antioxidant properties of dark chocolate. CH: dark chocolate-control sample; CH-WE: dark chocolate-white tea extract; CH-MW: dark chocolate-microencapsulated white tea extract; CH-JE: dark chocolate-jujube extract; CH-MJ: dark chocolate-microencapsulated jujube extract.

Lončarević *et al.* (2019) investigated the effect of green tea microencapsulation on white chocolate, and reported that the antioxidant capacity increased from 1.22 mmol/L Trolox in white chocolates to 16.12 mmol/L Trolox in enriched chocolates. Dean *et al.* (2016) used peanut skin phenolics microencapsulated by maltodextrin to lessen their bitterness for the fortification of milk chocolate. Results of the cited study showed that addition of 9% of encapsulated phenolics in chocolate could produce a product with an antioxidant capacity similar to that of dark chocolate and a flavor similar to milk chocolate. In another similar study, enrichment of gummy candy with betanin-liposomal nanocarriers resulted in the highest betanin stability and antioxidant activity with better sensory parameters (Amjadi *et al.*, 2018).

Texture hardness

The results of the effect of free and microencapsulated extracts of WT and jujube extracts at different concentrations on the hardness of dark chocolate samples are shown in Table 2. All treatments demonstrated less hardness than the control sample with a statistically significant difference ($P < 0.05$). Less hardness was also observed in chocolate samples containing microcapsules than those containing free extracts. This could be attributed to the hygroscopic capacity of microcapsule wall compounds (pectin and casein) in maintaining moisture, as was observed in the results of moisture content, thus reducing chocolate hardness. This was not surprising, because according to the higher moisture content of microencapsulated samples, water had a serious thickening effect on chocolates. Presence of water on the surface of sugar particles results in their sticking together and impeding the flow (Belščak-Cvitanovic *et al.* 2012). Change in the hardness of samples was concentration-dependent, intending that the hardness of chocolate samples decreased with increasing the concentration of free or microencapsulated extract. Overall, the results indicated that the samples containing microencapsulated jujube extract had less hardness than other treatments. Sample containing 7% jujube microcapsules had the lowest hardness, which showed about a 67% decrease compared to the control sample. This was attributed to the presence of simple sugars (glucose and fructose), considered as humectant substances, in jujube extract (Lončarević *et al.*, 2018).

Other studies have also reported similar findings. Verde *et al.* (2021) investigated the stability of milk chocolate with hygroscopic fibers during storage. Their results showed that after 270 days of storage, chocolates with inulin presented slightly better acceptability in relation to melting and hardness. Prosapio and Norton (2019), in developing fat-reduced chocolate by using water-in-cocoa butter emulsions, found that increasing a_w from 0.333 ± 0.165 (control chocolate) to 0.796 ± 0.006 reduced hardness of chocolate by 72%. Belščak-Cvitanovic *et al.* (2012) reported lowest hardness in dark chocolates containing freeze-dried extract of *Rubus idaeus* L. leaf extract. In another study, Rezende *et al.* (2015) reported that the addition of beta-glucan and inulin to chocolate formulation reduced its hardness because of retention of moisture, but a reverse trend was observed in larger amounts because of increased dry matter. However, all fiber-containing treatments had less hardness than other samples. Actually, there is a set of factors affecting the hardness of chocolate, and the most important ones are, the particle size distribution, composition of dark chocolate suspensions, especially a_w and fat content, and surfactants (mostly lecithin) (Afoakwa *et al.*, 2008; Deou *et al.*, 2022; Prosapio and Norton, 2019). Hardness was also affected by the production process, the properties of raw materials used (shape and surface besides size), and the length and storage conditions (Lapčíková *et al.*, 2022).

Moisture content and pH

The results showed that chocolate samples containing free and microencapsulated extracts generally contained a higher moisture content than the control sample (Table 2), with a statistically significant effect of microcapsules in increasing the moisture content of chocolate relative to free extract ($P < 0.05$). In this study, coating polymers were humectant compounds that affected the moisture content, and, therefore, the water activity of dark chocolate samples containing microcapsules. At a constant concentration, the effect of jujube extract microcapsules was higher than WT extract microcapsules in maintaining chocolate moisture, while no significant difference was observed between samples containing free extracts ($P < 0.05$). One of the reasons for this was the presence of simple sugars (glucose and fructose) in jujube extract, which are considered humectant substances (Bahrasemani Koohestani *et al.*, 2018). As observed, the moisture content had an increasing trend in all treatments with increasing concentrations, so that the highest amount (0.883 ± 0.02%) was measured in the treatment containing 7% jujube extract microcapsules (Table 2). Tolve *et al.* (2018) used whey protein isolate (WPI) for preparing phytosterol microcapsules to enrich dark chocolate with different concentrations of cocoa. Although WPI is a humectant compound, the authors reported that the microcapsules prepared with WPI did not have a significant effect on the water activity and moisture content of enriched chocolates. However, the percentage of cocoa used in the formulation significantly changed the moisture content of samples, so that the moisture content of chocolate showed an upward trend with increasing the amount of cocoa from 64% to 85%.

No definite trend was observed regarding the pH characteristics, although pH of chocolate samples generally

had a decreasing trend with the addition of free extract and microcapsules of WT and jujube; however, this decrease was not significant, except for treatments containing jujube extract microcapsules (Table 2). According to results, the control sample had the highest pH (7.58 ± 0.08) among all samples, but the pH decreased to 7.18–7.5 and 6.02–7.44, respectively, with the addition of free extract and microcapsules of WT and jujube. Maximum decrease in pH was observed for the treatments containing jujube microcapsules, especially at higher concentrations, which was in agreement with the results of Zhao *et al.* (2006). They stated that the pH of samples decreased with increasing jujube, and attributed this to the presence of pectic polysaccharides in jujube. Pectic polysaccharides are acidic and have a carboxyl group in their structures, thereby reduced the pH of samples. Both water absorption rate of coatings and pH could affect the release rate and stability of polyphenolic compounds during chocolate processing and shelf life (Delfanian and Sahari, 2020).

Color properties

Color is one of the main features for consumer's acceptance of a product. Color change in chocolates is often due to differences in their composition and process parameters during production (Tolve *et al.*, 2018). Encapsulation of phenolic compounds in nano or micro carriers appears to be an excellent tool for masking and incorporation of color into chocolates (Belščak-Cvitanović *et al.*, 2015; Lončarević *et al.* 2019). The results of comparing the mean color parameters of dark chocolate samples are shown in Table 3. The L^* index represented the lightness of sample and ranged from 0 (pure black) to 100 (pure white). It was clear that the apparent lightness of chocolate samples (L^* component) increased significantly from 20.52 ± 0.48 in the control sample to 26.08 ± 0.54 in the samples containing 3% WT microcapsules with the addition of free extract ($P < 0.05$). There was an opposite effect of jujube extract on the L^* component of chocolate samples, such that the addition of jujube extract and microcapsules reduced L^*, with the lowest level (13.88 ± 1.08) measured in the samples containing 7% jujube microcapsules. These results agreed with the results of Lončarević *et al.* (2019), who reported a change in the surface color of chocolates enriched with green tea capsules with increasing microcapsule content, so that all enriched chocolates had a somewhat lighter color than control samples during 12 months of storage. Similarly, Tolve *et al.* (2018) presented evidence that the addition of phytosterol microcapsules to dark chocolate formulation increased the L^* component, while the addition of cocoa in different concentrations decreased the level of this component.

The a^* index indicated the closeness of sample color to green and red, ranging from -120 (pure green) to +120 (pure red). Evaluation of the effect of treatments on the redness component (a^*) of dark chocolate samples revealed that the addition of WT and jujube free extracts had no significant effect on the amount of this parameter ($P < 0.05$). On the other hand, samples containing microcapsules had higher redness ($a^* = 1.74 \pm 0.05$) than the

Table 3. Color properties of dark chocolate enriched with free extracts of jujube and white tea as well as microencapsulated extracts of jujube and white tea in different concentrations.

	L*				a*			
	1%	3%	5%	7%	1%	3%	5%	7%
Sample								
CH	20.52±0.48^{c,d*}	–	–	–	1.74±0.05^{d,e}	–	–	–
CH-WE	20.87±0.23^{c,d}	22.06±0.25^{c}	21.78±0.18^{c}	19.65±0.38^{d,e}	1.63±0.08^{e,f}	1.68±0.1^{d,e,f}	1.43±0.02^{f}	1.55±0.02^{e,f}
CH-MW	24.35±0.31^{b}	26.08±0.54^{a}	25.67±0.25^{a,b}	25.08±0.41^{a,b}	2.82±0.1^{b}	2.97±0.07^{b}	2.09±0.15^{c,d}	1.85±0.11^{d,e}
CH-JE	20.15±0.28^{d}	19.87±0.75^{de}	19.62±0.33^{d,e}	18.59±0.42^{e}	1.59±0.11^{e,f}	1.53±0.06^{e,f}	1.68±0.08^{d,e,f}	1.87±0.12^{c,d}
CH-MJ	16.72±0.34^{f}	15.78±0.65^{fg}	15.08±0.37^{g,h}	13.88±1.08^{h}	2.21±0.18^{c}	2.78±0.14^{b}	3.47±0.33^{a}	3.12±0.27^{a,b}
	b*				ΔE			
CH	1.00±0.07^{d,e}	–	–	–	66.52±0.45^{e}	–	–	–
CH-WE	0.89±0.03^{d,e}	0.74±0.08^{e,f}	0.57±0.01^{e,f}	0.64±0.11^{e,f}	66.16±0.37^{e}	67.96±0.75^{g}	68.23±0.18^{f}	70.36±0.19^{ed}
CH-MW	3.11±0.18^{c}	3.56±0.14^{b,c}	3.92±0.21^{a,b}	4.33±0.15^{a}	64.88±0.28^{gh}	64.21±0.25^{h}	64.65±0.22^{gh}	65.28±0.22^{g}
CH-JE	1.28±0.32^{d}	1.04±0.21^{d,e}	0.72±0.15^{e,f}	0.33±0.08^{f}	69.9±0.55^{e}	70.17±0.48^{e,d}	70.41±0.42^{e,d}	71.45±0.27^{d}
CH-MJ	0.97±0.19^{d,e}	0.88±0.12^{d,e}	0.75±0.14^{e,f}	0.61±0.09^{e,f}	73.32±0.18^{c}	74.25±0.35^{b}	74.95±0.31^{b}	76.15±0.35^{a}

CH: dark chocolate-control sample; CH-WE: dark chocolate-white tea extract; CH-MW: dark chocolate-microencapsulated white tea extract; CH-JE: dark chocolate-jujube extract; CH-MJ: dark chocolate-microencapsulated jujube extract.
*Superscripted alphabets demonstrating statistical significant differences.

free form and the control, and jujube microcapsules had a significant effect on increasing the a^* index of chocolate samples (Table 3). Lončarević *et al.* (2019) reported the same results for higher a^* index in white chocolate formulations enriched with encapsulated green tea extract in different concentrations compared to the control white chocolate sample. As demonstrated, WT microcapsules with up to 3% concentration ($a^* = 2.97 \pm 0.07$) and jujube microcapsules with up to 5% concentration ($a^* = 3.47 \pm 0.33$) increased the red component of chocolate samples, but further increase had a negative effect on a^* index.

The b^* index represented the closeness of sample color to blue and yellow and ranged from -120 (pure blue) to +120 (pure yellow). Among the treatments, samples containing WT microcapsules had the highest amount ($b^* = 3.11–4.33$) of this component, which was significantly different from that of the control sample ($b^* = 1.00 \pm 0.07$) ($P < 0.05$). In jujube-containing samples, the amount of b^* component had a decreasing trend with increasing concentration from 1% to 7%, which agreed with the decrease of L^* component and increase of a* component. Shourideh *et al.* (2012) stated that humectant compounds reduced light scattering, thereby decreasing the lightness and giving a darker appearance to chocolates. Aidoo *et al.* (2014) also showed that adding polysaccharides to chocolate formulation led to increased caramelization and Millard processing, resulting in elevated color production and darker color of chocolates. In this study, the samples containing jujube extract had redder component and less apparent lightness because of the presence of polysaccharides, especially in the structure of jujube extract, and pectin and casein in the walls of jujube microcapsules. Dwijatmoko *et al.* (2016) investigated the effect of cinnamon essential oil on the sensory and color properties of dark chocolates, and reported that the addition of this essential oil at a concentration of 0.25–0.75% had no significant effect on a^* and b^* components but significantly increased the L^* component compared to the control sample. ΔE changes in CH-JE and CH-MJ altered with increase in extract concentration; this could be due to the hygroscopic properties of jujube extract and its effect on the homogeneity of chocolate as w/o suspension, as observed in Table 2. Besides this, encapsulation could reduce hydrophobic attributes and impact some properties (Reis *et al.*, 2022).

Microstructure

The SEM test was used to observe and evaluate the morphology and microstructure of emulsions and polymers. SEM images of chocolate samples (Figure 4) showed that particles of dark chocolate containing WT extract microcapsules (CH-MW 3%) had a smaller size and greater uniformity than both control sample (CH) and sample containing WT free extract (CH-WE 3%). However, a relatively different result was obtained in the jujube-containing samples, intending that the dark chocolate sample containing jujube free extract (CH-JE 3%) had a smaller size and more uniformity than the control and MJ samples (CH-MJ 3%). The microstructural examination of CH-MW 3% sample demonstrated a particle bond coated with an external fat phase, resulting in the formation of a homogeneous and uniform emulsion with a smaller dispersed phase particle size than in other samples. This uniformity in the conching phase could be facilitated because conching leads to coating of solid particles with fat, and such a homogeneous fat matrix is ideal for visual and textural properties (Sim *et al.*, 2016).

As seen in Figure 4, particle size uniformity in CH-MW 3% was higher than in CH-MJ 3%, which is in agreement with position sensitive detection (PSD) results. This could be due to composition of different extracts and their interactions with wall compounds. There was a visible trend of decreasing crystalline structure to a more amorphous-like (or ill-defined) crystal structure, as shown in Figure 4 for CH-MW 3% in comparison to CH-WT 3%, which it mostly raises from the surface interactions of coating materials (both casein and pectin). In an investigation on enrichment of white chocolate with blackberry juice encapsulate (maltodextrin), SEM results indicated changes in the spatial distribution of uniform crystalline network within the structure. The microstructural examination of encapsulated samples showed the interconnection of particles externally coated with the fat phase (Lončarević *et al.*, 2018). The same trend was observed by Lapčíková *et al.* (2022) in SEM results of partial substitution of skimmed milk powder with whey powder in milk chocolates. In forming more uniform particles, pectin's emulsifying and emulsion stabilizing properties could play an important role (Alba and Kontogiorgos, 2020). Datsomor *et al.* (2019) suggested that lecithin could be partially substituted by pectin extracted from Okra in milk chocolates without significant textural changes. Baracat *et al.* (2012) also concluded that wall composition and drying rate, particularly in the early stages, could affect the surface microstructure properties of microencapsulated materials. Other researchers discovered that increasing portion of pectin increased the particle size of emulsion, and attributed this to the presence of nonabsorbable pectin on the surface of oil particles, resulting in aggregation and rise in particle size (Azizanbari et al, 2013; Emadzadeh *et al.*, 2021).

Conclusion

This study mainly aimed to enrich dark chocolate with WT and jujube extract in free and microencapsulated forms. According to polyphenol content and antioxidant activity analysis, adding WT and jujube extracts resulted in a significant increase in phenolic content and dark

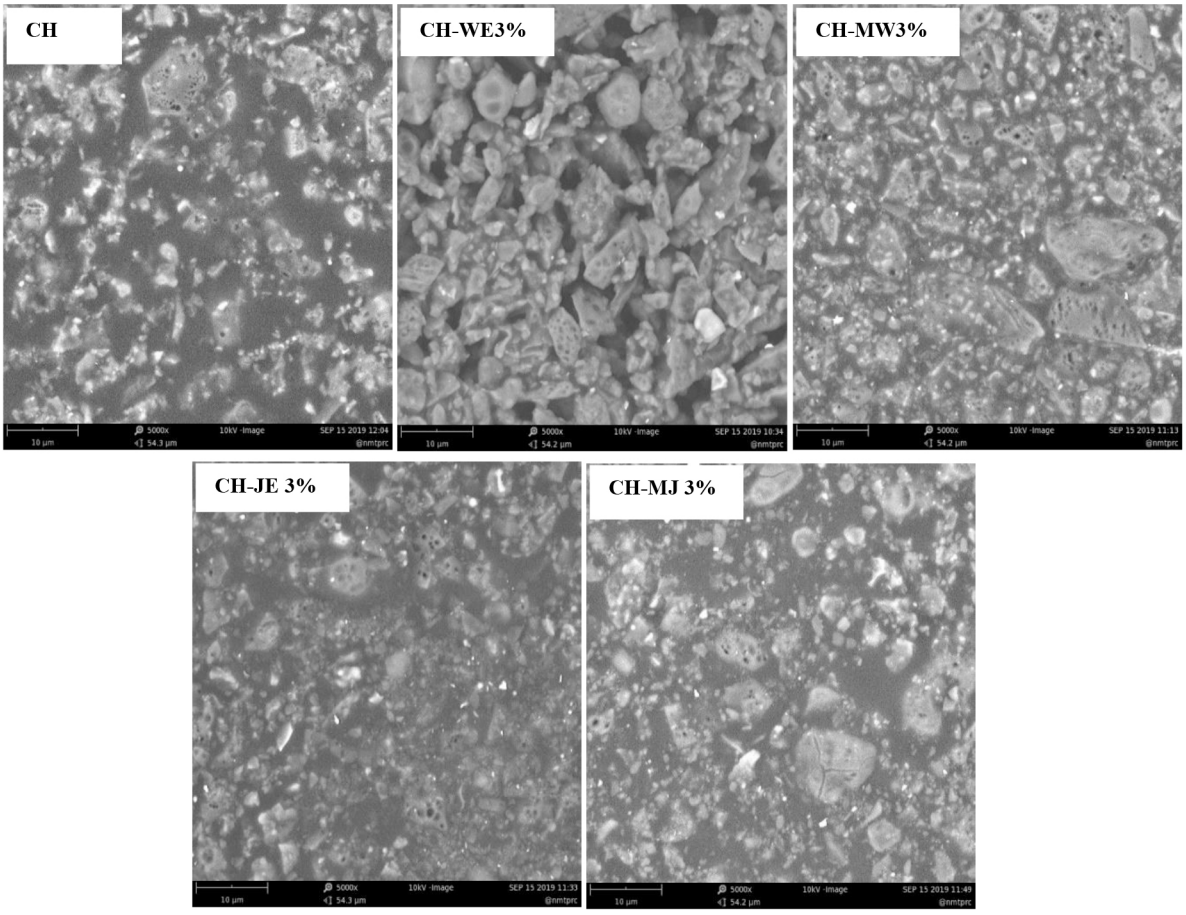

Figure 4. SEM images of control and chocolate samples enriched with WT and free jujube extracts and microcapsules. Microstructures of dark chocolate (CH) and enriched chocolates with 3% white tea extract (CH-WE3%), 3% encapsulated white tea extract (CH-MW3%), 3% jujube extract (CH-JE3%) and 3% encapsulated jujube extract (CH-MJ3%) at magnifications of ×5000.

chocolate's antioxidant properties. The effect of encapsulation on the stability of phenolic compounds of both extracts during processing of dark chocolates was obvious. TPC results demonstrated a positive and protective role of encapsulation in increasing phenolic compounds of dark chocolate formulation with WT and jujube extracts by 14.1% and 21.7%, respectively. Enrichment of chocolates by JE and MJ reduced the L^* index and gave it a darker appearance. The lowest level of L^* (13.88 ± 1.08) was measured in MJ 7% sample. Encapsulation of phenolic compounds was an excellent tool for masking and incorporation of color in chocolates. Addition of WT and jujube extracts raised the moisture content of dark chocolates, mostly in encapsulated form because of the hygroscopic properties of coating compounds (pectin and casein). This parameter, besides some emulsifying properties of pectin, reduced the hardness of dark chocolate samples. SEM images showed a visible trend of decreasing crystalline structure to a more amorphous-like structure for dark chocolates containing WT microcapsules in comparison to free-form samples. In summary, the applied method for microencapsulation of WT and jujube extracts by pectin and casein was effective in protecting the functional properties of enriched dark chocolates, and this proved to be an interesting strategy for incorporating phenolic compounds into chocolate formulations.

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