ORIGINAL RESEARCH

Effects of Spray-Drying and Freeze-Drying on Bioactive and Volatile Compounds of Smoke Powder Food Flavouring

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Abstract

Transforming liquid smoke to powder form can provide convenience for use and storage. Liquid smoke was prepared by fast pyrolysis technology using a fluidised bed and converted to smoke powder by spray-drying or freeze-drying processes. Both drying processes effectively retained the bioactive compounds in the powder encapsulates with retention efficiencies up to 80%. The bioactive capacities were approximately two times higher than liquid smoke. Spray-drying did not induce thermal damage to the bioactive compounds, and the dominant compounds were retained in the powders. Gas chromatography–mass spectrometry and principal component analysis indicated that the chemical composition was not significantly changed after both drying methods, but small molecular carbonyls, furans and phenols were partially lost. The spray-dried particles had a spherical shape, while freeze-dried particles had irregular shapes because of different powder preparation methods. The particle size of spray-dried powders was in the range of 6.3 to 6.9 μm, while the value for freeze-dried powders was decreased from 580.4 to 134.7 μm by increasing the maltodextrin concentration. The freeze-dried powders performed better in terms of flowability and cohesiveness because of their relatively high density and large particle size. This study revealed that both encapsulation methods could efficiently prepare smoke powder. Spray-drying process would be suitable for large-scale production, while freeze-drying could be used to optimize the encapsulation efficiency of bioactive compounds.

Keywords Bioactive · Smoke flavouring · Principal component analysis · Spray-drying · Freeze-drying · Encapsulation

Introduction

Liquid smoke is a smoke flavouring food ingredient that can increase the food shelf life (Dimakopoulou-Papazoglou & Katsanidis, 2017). Compared with traditional food smoking, food treatment with liquid smoke is fast, safe and environmental-friendly (Arvanitoyannis & Kotsanopoulos, 2012; Šimko, 2005). Liquid smoke is usually prepared by smouldering, a slow process by burning wood with limited oxygen. Alternatively, fast pyrolysis can rapidly and efficiently convert wood into liquid smoke at an inert atmosphere (Xin et al., 2020). Fast-pyrolysis prepared liquid smoke showed a high antioxidant capacity and an equivalent safety level to commercialised liquid smoke (Xin et al., 2021). Liquid smoke is primarily composed of organic acids, carbonyls, phenols, furans and furfurals (Xin et al., 2020). A complex mixture of these bioactive compounds contributes to the organoleptic, colouring, antimicrobial and antioxidant effects on treated food (Maga, 2018). These bioactive compounds are sensitive to light, high temperatures and oxygen (Taruna Syah et al., 2016). Spray-drying or freeze-drying can encapsulate the bioactive compounds from liquid smoke to increase their stability and avoid undesirable interactions with other compounds. In addition, a solid product is more convenient for use and storage when compared to the original liquid of the product.

Spray-drying can encapsulate plant-based bioactive compounds, including liquid smoke (Belščak-Cvitanović et al., 2015). Ariestya et al. (2016) spray-dried coconut shell liquid smoke and applied the encapsulates to preserve tilapia fish meat. The results showed that the smoke powder inhibited microbial growth and maintained the quality of the fish meat throughout nine-day cold storage. Taruna Syah et al. (2016) investigated the impacts of spray-drying process conditions on the produced smoke microcapsules. The increased encapsulant

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concentration and spray-drying inlet temperature improved the microcapsules' product yield, phenol retention and solubility. Saloko et al. (2014) prepared smoke microcapsules derived from coconut shells for application on tuna fish. These smoke microcapsules had high values of total phenolic content, total acid content and radical scavenging activity. The smoke microcapsules were sufficient to maintain the freshness of the tuna fish meat for 48 h at room temperature.

Freeze-drying is a common method to encapsulate heat-sensitive bioactive compounds (Celli et al., 2015). To the best of our knowledge, there are no data about the freeze-drying of liquid smoke. Freeze-drying had been used to encapsulate red wine (Sanchez et al., 2013), sumac extract (Caliskan & Dirim, 2016) and acerola plum (Rezende et al., 2018). A previous study showed freeze-dried powders had better flowability, wettability and solubility, while spray-dried powders presented less moisture content and water activity values (Rezende et al., 2018).

To the best of our knowledge, there is limited information about the freeze-drying of liquid smoke. It is unclear whether spray-drying or freeze-drying better encapsulates the bioactive compounds or produces better smoke powder products. Spray-drying is an efficient process that is simple, scalable and low cost. However, the high processing temperatures can quickly damage active ingredients. Freeze-drying is particularly suitable for drying heat-sensitive materials (Desai & Jin Park, 2005). Compared with spray-drying, it often results in greater product uniformity and better stability. In this study, liquid smoke produced by the fast pyrolysis process was encapsulated by spray-drying and freeze-drying. The process yield, bioactive properties, chemical composition and physical properties were investigated. The effects of drying processes on encapsulation performance could be of significant interest to food ingredient research and the industry.

Materials and Methods

Materials

Kanuka woodchips were obtained from the East Cape of New Zealand and used for liquid smoke production. Kanuka (*Kunzea ericoides*) is a native tea tree species in New Zealand and is commonly used for food smoking (Essien et al., 2019). Maltodextrin was used as the encapsulation carrier, purchased from Bulk Powders New Zealand. Chemicals and standards used in this study were purchased from Sigma-Aldrich (New Zealand), including gallic acid, catechin hydrate, Folin-Ciocalteu's phenol reagent, DPPH (1, 1-diphenyl-2-picrylhydrazyl), Trolox (6-hydroxy-2,-5,-7,-8-tetramethylchroman-2-carboxylic acid), aluminium chloride, sodium carbonate, sodium nitrite and GC-grade dichloromethane.

Liquid Smoke Production and Encapsulation

Kanuka woodchips were fast-pyrolysed at 450 °C in a fluidised-bed reactor to prepare liquid smoke. The details of the reactor are illustrated in a previous study (Xin et al., 2021). The process produced liquid smoke with a yield of 30 wt% and valuable by-products, biochar and pyrolysis oil, with yields of 24 and 18 wt%. The prepared liquid smoke was an aqueous phase liquid with a brown to red colour. Maltodextrin was diluted in the fast pyrolysis liquid smoke to prepare the feed solutions for spray-drying and freeze-drying. Three feed solutions were prepared at ratios of 6:1, 5:1 and 4:1 (liquid smoke/maltodextrin, w/w).

Spray-drying of each feed solution was achieved in a Büchi B-290 spray dryer (Maia et al., 2019). Spray-drying process was maintained at an air inlet temperature of 150 °C, aspirator rate of 100%, atomising gas flowrate of 11.1 L/min (standard temperature and pressure) and feeding pump rate of 10%. The outlet temperature was in the range of 60–80 °C. Spray-dried products from the cyclone container and drying chamber were collected and weighed. The three powder products were labelled as SPD-6/1, SPD-5/1 and SPD-4/1, respectively, corresponding to the three feed solution ratios of 6:1, 5:1 and 4:1.

Freeze-drying of each feed solution was achieved using a Christ Alpha 1–2 LDplus freeze dryer (Condurache et al., 2019). The three solutions were frozen at -18 °C for 48 h before placing in the freeze-dryer chamber. The process conditions were a condenser temperature of -58.8 °C, a pressure of 6.11 mbar, a vacuum of 0.42 mbar and a drying time of 48 h. After freeze-drying, the glass-like samples were ground using a mortar and pestle following a previous study (Rezende et al., 2018). The three powder products labelled as FZD-6/1, FZD-5/1 and FZD-4/1, respectively, corresponding to the three feed solution ratios of 6:1, 5:1 and 4:1.

Six smoke powder samples were obtained from spray and freeze-drying and stored in a desiccator at around 20 °C until the analysis in the following 4 weeks. Spray-dried powders had a yellow colour like butter, while freeze-dried ones had a brown colour like chocolate.

The product yield is calculated according to Eq. (1):

$$Y_{product} = \frac{M_{powder}}{M_{TSS}} \times 100 \tag{1}$$

where $Y_{product}$ is the yield of powder product (wt.%), M_{powder} is the mass of obtained powder product and M_{TSS} is the mass of total soluble solid in the feed solution.

Determination of Bioactive Activities

The smoke powder products and kanuka liquid smoke were dissolved and diluted in distilled water to 10 mg/mL. These solutions were analysed for total phenolic content (TPC), total flavonoid content (TFC), ferric reducing antioxidant power assay (FRAP) and DPPH scavenging activity (DPPH) using an ultraviolet–visible (UV) microplate reader (EnSpire 2300, PerkinElmer).

Total phenolic content (TPC) was determined by the Folin-Ciocalteu assay by following a previous study (Munir et al., 2018). The solutions (0.025 mL) and gallic acid standards were mixed with 0.125 mL of tenfold freshly diluted Folin-Ciocalteu reagent in 96 well plates. Then, 0.125 mL of 7.5% sodium carbonate was added to each well. The plates were incubated in the dark for 60 min at room temperature. The absorbance values were measured at 765 nm using the UV microplate reader. The results were expressed as milligrams of gallic acid equivalent per gram of smoke powder or liquid smoke (mg GAE/g).

Total flavonoid content (TFC) was determined using quercetin as the standard by an aluminium chloride method (Essien et al., 2020). The solutions (0.025 mL) and quercetin standards were mixed with 0.1 mL of distilled water and 0.01 mL of 5% sodium nitrite in 96 well plates. Then, 0.015 mL of 10% $AlCl_3$ and 0.050 mL of 1 mol/L NaOH were added to the well plates. After adding 0.05 mL of distilled water, the plates were incubated in the dark for 60 min at room temperature. The absorbance values were measured against a reagent blank at 510 nm. The results were expressed as milligrams of quercetin equivalent per gram of smoke powder or liquid smoke (mg QE/g).

Ferric reducing antioxidant power assay (FRAP) was conducted following a previous report (Kheirkhah et al., 2019). FRAP reagent was prepared by mixing the 2,3,5-triphenyltetrazolium chloride (TPTZ) solution (10 mmol/L TPTZ in 40 mmol/L HCl), 20 mmol/L $FeCl_3$ and 300 mmol/L sodium acetate buffer (pH 3.6) at a ratio of 1:1:10. Smoke samples (0.001 mL) and Trolox standards were mixed with 0.2 mL of FRAP reagent in 96 well plates. The absorbance values were measured at 593 nm against a reagent blank after incubation for 60 min at room temperature. The results were expressed as milligrams of Trolox equivalent per gram of smoke powder or liquid smoke (mg TE/g).

A previous study was followed to determine DPPH scavenging capacity (Essien et al., 2020). The DPPH (1,1-diphenyl-2-picrylhydrazyl) reagent was firstly dissolved in ethanol at a concentration of 40 mg/L. Then, 0.2 mL of DPPH solution was mixed with 0.01 mL of each sample, Trolox standards and blank in 96 well plates. The absorbance values were measured at 517 nm after incubation for 60 min at room temperature. The DPPH scavenging capacity was expressed as milligrams of Trolox equivalent per gram of smoke powder or liquid smoke (mg TE/g).

The retention efficiency of each bioactive value is calculated according to Eq. (2):

$$RE = \frac{V_{Powder} \times Y_{Product}}{V_{LS}/P_{TSS}} \times 100 \qquad (2)$$

where RE is retention efficiency (%), V_{Powder} is the bioactive value (TPC, TFC, FRAP or DPPH) of powder products, $Y_{Product}$ is the yield of powder products, V_{LS} is the bioactive value of pure liquid smoke and P_{TSS} is the mass percentage of total soluble solid in the feed solution.

GC–MS and Principal Component Analysis

Gas chromatography-mass spectrometry (GC–MS) analysis was conducted to identify the volatile compounds. The smoke powder products and pure liquid smoke were dissolved and diluted in distilled water to a concentration of 250 mg/mL. Then the solutions were mixed with dichloromethane at a volume ratio of 1/2 (solution/dichloromethane), and the mixtures were agitated at 200 rpm for 6 h. The dichloromethane extracts were then filtered by 0.2 μm syringe filters. Each dichloromethane extract was prepared in triplicate for GC–MS analysis. This preparation method followed a previous study with minor modifications (María D. Guillén & Ibargoitia, 1998).

The GC–MS instrument (Shimadzu QP-5000) was equipped with a DB-5HT column (30 m×0.25 mm×0.1 μm). Dichloromethane extract (1 μL) was injected at an injection temperature of 280 °C. The oven temperature was increased from 50 to 250 °C by 20 °C/min and then was held for 5 min. Mass spectra were operated in electron ionisation mode at 70 eV, and the mass range was 50–300 amu for acquisition. Volatile compounds were identified by comparing the mass spectra with those in the library NIST and by comparing the data results with previous studies (Petzold et al., 2014; Taruna Syah et al., 2016; Xin et al., 2021). Their relative abundances were expressed as the peak area percentages of the total ionisation chromatogram (TIC).

Principal component analysis (PCA) as a useful chemometric tool can reduce the experimental data dimension for ease of interpretation. It can transform a large set of interrelated variables into principal components. This study used principal component analysis (PCA) to understand the difference between liquid smoke and produced powders in terms of their chemical composition. The smoke flavouring samples were set as the observations, and peak area percentages of identified compounds by GC–MS were set as the variations. A MATLAB toolbox was used with MATLAB R2020a software to

conduct PCA (Ballabio, 2015). The toolbox gives visualising results, including plots of scores and loadings and numerical values of eigenvalues.

Determination of Physical Properties

The morphology of the smoke powders was determined using a scanning electron microscope (Hitachi SU-70 Schottky field SEM). Smoke powder samples were sprinkled on the double-sided conductive carbon tabs and glued onto the SEM mounts. The samples were coated with platinum for 100 s at room temperature.

Particle sizes of the smoke powders and pure maltodextrin were measured by a particle size analyser (Malvern Mastersizer 2000, Malvern Instruments Ltd) with a dry dispersion module (Scirocco 2000). The weighted-average volume diameter was expressed as diameter $D_{[4,3]}$, assuming spherical particles with the same volume as the actual particles. Particle size distribution (PSD) is calculated according to Eq. (3):

$$PSD = \frac{d(0.9) - d(0.1)}{d(0.5)} \qquad (3)$$

where d(0.9), d(0.5) and d(0.1) are respectively particle diameters at 90, 50, and 10% of the cumulative size distribution curve from Mastersizer analysis results (Tupuna et al., 2018).

Bulk density and tapped density were determined by following a reported method (Caliskan & Dirim, 2016). Briefly, 20 g of each powder sample or maltodextrin was gently loaded into a 100 mL graduated cylinder. The sample weight was divided by the measured volume to obtain bulk density (ρ_{bulk}, g/mL). The tapped density (ρ_{tapped}, g/mL) was obtained after tapping the cylinder 120 times and then measuring the sample volume.

Carr index (CI) and Hausner ratio (HR) were evaluated for the flowability and cohesiveness of the six powder samples and maltodextrin (Caliskan & Dirim, 2016). The CI and HR values are calculated according to Eqs. (4) and (5):

$$CI = \frac{\rho_{tapped} - \rho_{bulk}}{\rho_{tapped}} \times 100 \qquad (4)$$

$$HR = \frac{\rho_{tapped}}{\rho_{bulk}} \qquad (5)$$

Water content of the powder samples and maltodextrin was measured by weight difference before and after placing the sample in a drying oven at 105 °C until no more weight loss.

Statistical Analysis

All of the drying processes and analyses in this study were conducted in triplicate, and the results were presented as mean values ± standard deviation ($n = 3$). Analysis of variance (ANOVA) was performed, and the difference between means was analysed using Duncan's test. Statistical significance was considered at $p < 0.05$. All statistical analysis was performed using SPSS 9.05 (Chicago, USA).

Results and Discussion

Product Yield

At the mass ratios of 6:1, 5:1 and 4:1 (liquid smoke/maltodextrin, w/w), three solutions were spray-dried and freeze-dried separately. In spray-drying, the product in the drying chamber, cyclone and cyclone container was weighed and collected. The solid formed in freeze-drying chamber was weighed and collected as freeze-drying product. Product yields in these two drying processes are shown in Fig. 1. Freeze-drying with the highest maltodextrin ratio (FZD-4/1) showed the highest yield of 96.9 ± 0.8 wt%. Spray-drying with the lowest maltodextrin ratio (SPD-6/1) showed the lowest yield of 73.3 ± 0.5 wt%. The comparison of the two drying methods with the same solutions showed that freeze-drying could lead to a higher yield. This trend was also reported in the studies of spray and freeze-drying of strawberry flavour (Pellicer et al., 2019), black glutinous rice (Laokuldilok & Kanha, 2015) and orange powder (Barbosa et al., 2015). The loss of volatile compounds occurred in spray-drying due to a
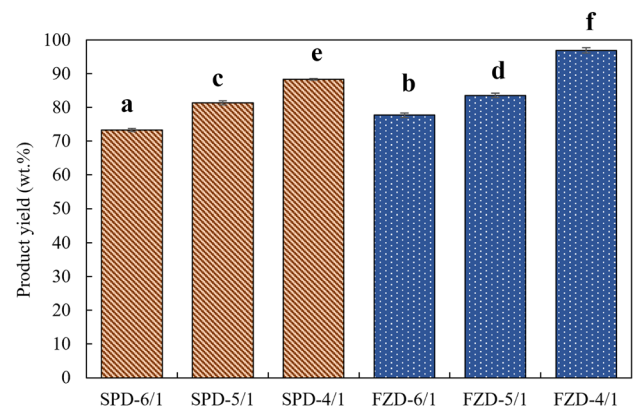
Fig. 1 Product yields in spray and freeze-drying of liquid smoke and maltodextrin solutions. Error bars correspond to standard deviations of the mean ($n = 3$); the columns with different letters indicate significant difference at $p < 0.05$

Table 1 TPC, TFC, FRAP and DPPH values of liquid smoke and smoke powders

	SPD-6/1	SPD-5/1	SPD-4/1	FZD-6/1	FZD-5/1	FZD-4/1	Liquid smoke
TPC, mg GAE/g	82.9[e]	76.1[d]	73.1[c]	86.4[f]	75.7[d]	65.4[b]	40.1[a]
± std	0.4	0.7	1.3	0.9	1.0	0.2	0.6
TFC, mg QE/g	100.6[f]	95.2[d]	86.3[c]	101.2[f]	88.0[c]	77.0[b]	38.5[a]
± std	4.3	2.5	1.4	1.9	2.5	0.7	0.5
FRAP, mg TE/g	91.7[d]	86.4[b,c]	82.7[b]	89.7[c,d]	85.4[b,c]	81.5[b]	46.9[a]
± std	1.2	1.5	2.9	1.9	2.3	2.1	2.8
DPPH, mg TE/g	39.2[d,e]	45.3[e]	41.3[d,e]	36.8[c,d]	32.5[c]	23.5[b]	13.6[a]
± std	4.7	2.3	1.0	2.4	2.0	3.6	0.7

Results in this table are expressed as mean ± standard deviation. Mean values with the same superscript letters within a row are not significantly different at $p < 0.05$

high temperature. Meanwhile, the fine particles not captured by the spray drier cyclone also contributed to the mass loss.

Figure 1 also indicates that increasing the maltodextrin concentration led to a higher product yield in both drying processes. Maltodextrin is a common encapsulant agent for bioactive compounds. Previous studies also showed that a higher concentration of encapsulant agents improved encapsulation efficiency (Caliskan & Dirim, 2016; Rezende et al., 2018; Taruna Syah et al., 2016). However, it was suggested that the concentration of encapsulant agent should be no more than 30% in spray-drying (Ghosh, 2006), since high maltodextrin concentration also led to a dilution of bioactive compounds in the encapsulates.

Bioactive Activities

Table 1 shows the bioactive activities of pure liquid smoke and smoke powders in terms of TPC, TFC, FRAP and DPPH. Figure 2 illustrates the retention efficiency of bioactive activities via each encapsulation method. Equation (2) is used to calculate the efficiency, which eliminated the dilution effects due to the increase of the maltodextrin concentration in different solutions.

Total phenolic content (TPC) value is an important indicator of bioactive activities for liquid smoke. The phenolic compounds (produced from pyrolysis of lignin) contribute to the effects of smoke flavouring, aroma profile, antimicrobial and antioxidant capacity (Lingbeck et al., 2014). As shown in Table 1, the TPC value increased from 40.1 mg GAE/g for pure liquid smoke to the range of 65.4–86.4 mg GAE/g for smoke powders. Spray-drying and freeze-drying processes retained the phenolics to the same extent in this study. Table 1 also shows the TPC value decreased with the increase of maltodextrin concentration due to the dilution effect. Figure 2 proves that using a higher concentration of encapsulant agent increases the retention efficiency from 40 to 50%. The same effect of encapsulant agent on retention efficiency was observed in previous studies (Caliskan & Dirim, 2016; Taruna Syah et al., 2016).

Flavonoids are the largest group of naturally occurring phenolic compounds (Sulaiman & Balachandran, 2012). Total flavonoid content (TFC) of liquid smoke has rarely been reported. The results of this study indicated that TFC of a fast pyrolysis liquid smoke was approximately 38.5 mg QE/g. It was increased by more than twofold after spray-drying or freeze-drying, and both drying processes showed

Fig. 2 Retention efficiency (%) of bioactive activities in different encapsulation processes. Error bars correspond to standard deviations of the mean ($n = 3$); the columns with different letters within each cluster indicate significant difference at $p < 0.05$
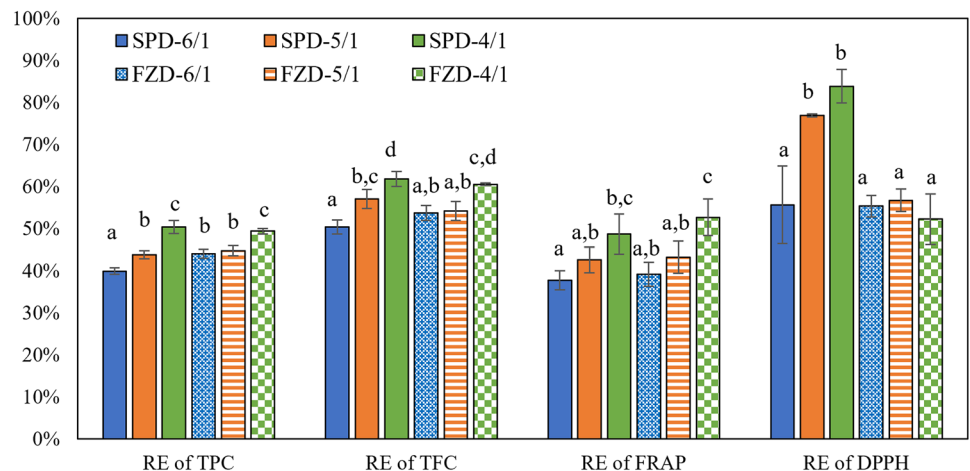
similar effects on the retention of flavonoids and retention efficiency.

Antioxidant capacity (FRAP and DPPH) of the kanuka liquid smoke produced by fast pyrolysis were much higher than those reported previously (Loo et al., 2007; Ma et al., 2013). For instance, DPPH value of commercial liquid smoke was reported in the range of 0.1 to 4.8 mg TE/g (Soldera et al., 2008). Fast pyrolysis could effectively decompose lignin polymers leading to a high proportion of phenolics in the resultant liquid smoke (Xin et al., 2021). Spray-drying or freeze-drying of the liquid smoke led to

the further increase in antioxidant capacity (approximately twofold) as shown in Table 1.

Chemical Composition

The volatile compounds of pure kanuka liquid smoke and smoke powders were identified by GC–MS analysis. Peak area percentages of identified compounds are presented in the Supplementary data (Table S1). The compounds were classified by their main functional groups as acids, carbonyls, esters, furans and phenols. The results showed the most abundant

Fig. 3 Scores (**a**) and loadings (**b**) plots of PC1 and PC2 for liquid smoke, spray dried powders and freeze dried powders
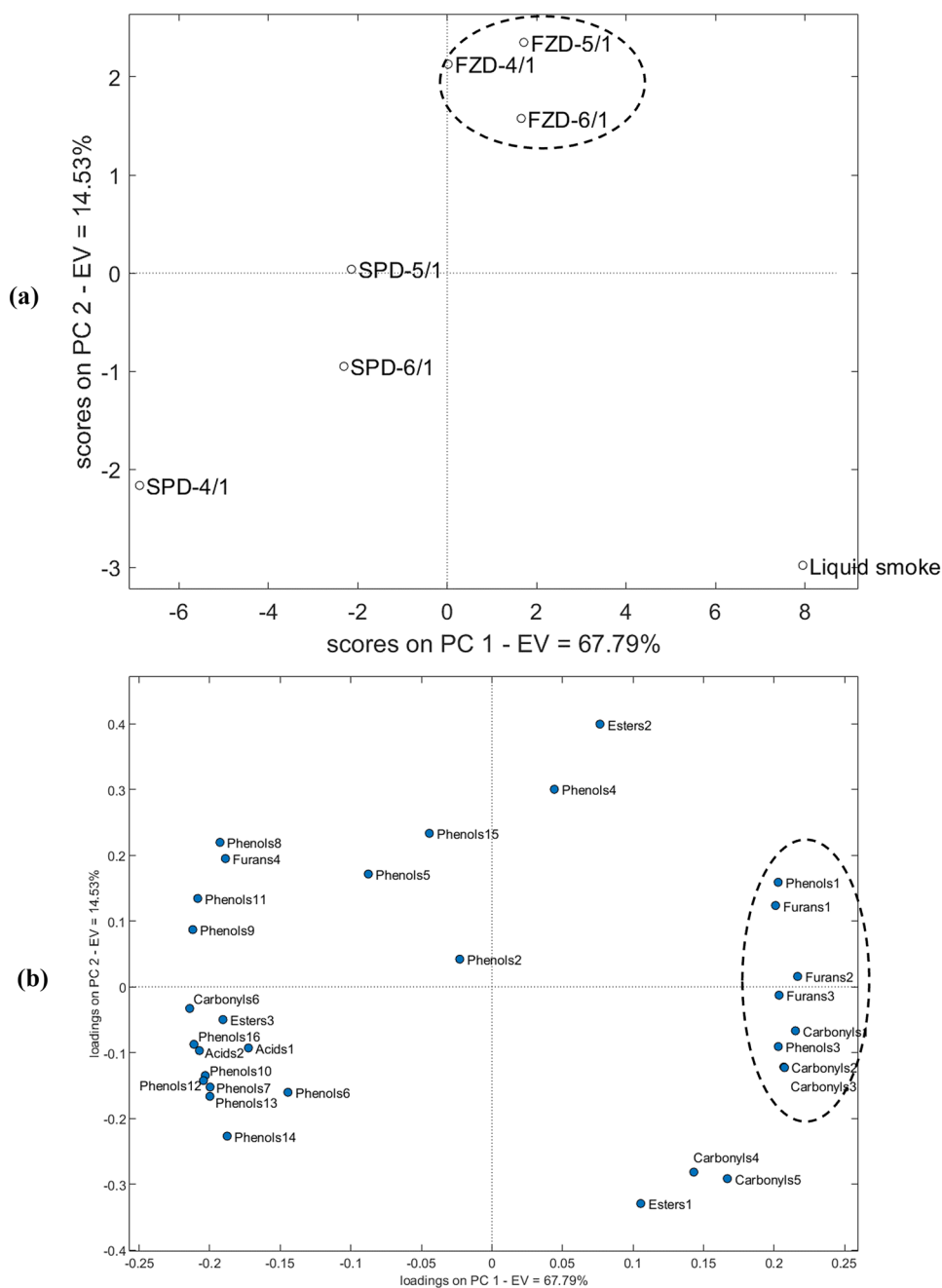
compounds in liquid smoke and smoke powders were the same, which were guaiacol, 2-hydroxy-2-cyclopenten-1-one, 2(5H)-furanone and syringol in sequence. Guaiacol, a common compound in liquid smoke, has been reported frequently in previous reports (Guillén et al., 1995; Surboyo et al., 2019). Guaiacol and its derivatives are the main smoky flavour and aroma in liquid smoke (Schranz et al., 2017). Syringol, as one of the guaiacol's derivatives, presents the most favoured aroma (Montazeri et al., 2012). 2-Hydroxy-2-cyclopenten-1-one from carbonyls group and 2(5H)-furanone from furans groups can soften the heavy smoky aroma associated with phenols and improve the overall sensory properties of the treated food (Maga, 1987).

Principal component analysis (PCA) scores and loadings plots are shown in Fig. 3 with the first two principal components, PC1 and PC2. PC1 described the largest possible variation (67.79%), and PC2 the second-highest variation (14.53%). Since the value of PC1 is high, the variations among the seven smoke flavouring samples can be explained by the locations on the horizontal direction in the loadings plot (Fig. 3b). It shows carbonyls 1, 2, 3 (Butanal, 3-Methyl-2-cyclopenten-1-one, 2-Hydroxy-2-cyclopenten-1-one), furans 1, 2 (2(5H)-Furanone, 3-Methyl-2(5H)-furanone), and phenols 1, 3 (Phenol, Guaiacol) are located farthest to the right. Presumably, partial loss of these compounds during freeze or spray-drying processes led the changes of chemical compositions of smoke powders.

The pure liquid smoke is located far away from the powder samples, as shown in Fig. 3a indicates its chemical composition is relatively different from smoke powders. Three freeze-dried powders are clustered, indicating that the chemical composition is relatively similar. In contrast, three spray-dried powders are scattered in the scores plot. It can be concluded that maltodextrin concentration is a more important parameter in spray-drying than freeze-drying. A lower concentration led to more losses of volatile compounds.
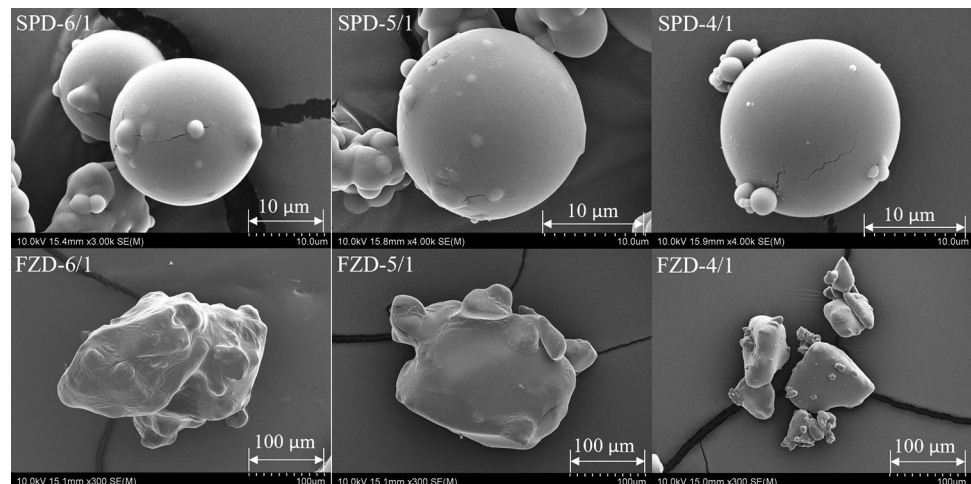
Physical Properties

SEM images of smoke powder particles are shown in Fig. 4. The particles produced by spray-drying had a spherical shape with sizes at the scale of 10 μm. The particles produced by freeze-drying had irregular shapes, and their particle sizes were relatively large at the scale of 100 μm. Because freeze-drying of liquid smoke solutions resulted in an amorphous glassy mass, it was ground using a mortar and pestle.

Table 2 shows the physical properties of the smoke powders and the carrier maltodextrin as a reference. The particle size ($D_{[4,3]}$) of spray-dried powders was in the range of 6.3 to 6.9 μm with no significant difference by using the lab-scale spray dryer. It is noted that the particle size is strongly affected by the spray-drying inlet temperature, feeding rate, feed composition or carrier type (Cortés-Rojas et al., 2015). In a previous study, a smoke powder produced by spray-drying had an average particle size of 0.013 μm (Saloko et al., 2014). That nano-encapsulate was prepared from a mixture of liquid smoke, chitosan, and maltodextrin. While in another study, the particle size of spray-dried powder was in the range of 0.5 to 12.7 μm (Taruna Syah et al., 2016). A smaller particle size would make spray-dried smoke powder perform better on food preservation by easily penetrating the treated food. The PSD value for spray-dried powders was decreased from 2.57 to 2.16 by increasing the maltodextrin concentration. The increase of maltodextrin concentration led to less homogeneous particle sizes in spray-drying, and this trend was also reported in another study of norbixin spray-drying (Tupuna et al., 2018).

Due to the different powder preparing methods, particle sizes of freeze-dried powders were much larger than the spray-dried powders. The particle size was decreased by increasing the carrier concentration, and $D_{[4,3]}$ value of FZD-4/1 (134.7 μm) was close to the maltodextrin powder

Fig. 4 SEM images of particles from spray and freeze-drying of liquid smoke

Table 2 Physicochemical properties of smoke powders and maltodextrin

	SPD-6/1	SPD-5/1	SPD-4/1	FZD-6/1	FZD-5/1	FZD-4/1	Maltodextrin
$D_{[4,3]}$, μm	6.9[a]	6.3[a]	6.4[a]	580.4[d]	500.1[d]	134.7[c]	96.1[b]
± std	0.2	0.1	0.3	29.0	7.7	2.1	0.9
PSD	2.57[g]	2.24[f]	2.16[e]	1.39[b]	1.33[a]	1.63[c]	1.99[d]
± std	0.06	0.01	0.02	0.02	0.02	0.00	0.01
Bulk density, g/mL	0.224[a]	0.290[b]	0.349[c]	0.397[d]	0.482[f]	0.513[g]	0.460[e]
± std	0.003	0.003	0.007	0.007	0.003	0.003	0.002
Tapped density, g/ml	0.275[a]	0.367[b]	0.422[c]	0.451[d]	0.552[e]	0.589[f]	0.550[e]
± std	0.002	0.005	0.009	0.004	0.005	0.004	0.005
CI, %	18.6[c]	20.9[d]	17.2[b,c]	12.1[a]	12.7[a]	13.0[a]	16.3[b]
± std	0.7	0.5	1.2	0.9	1.0	0.0	0.6
HR, %	1.23[b]	1.26[c,d]	1.21[b]	1.14[d,e]	1.15[c]	1.15[e]	1.20[a]
± std	0.01	0.01	0.02	0.01	0.01	0.00	0.01
Moisture content, wt.%	8.5[b]	10.1[c,d]	7.8[b]	10.9[d,e]	9.7[c]	12.0[e]	4.7[a]
± std	0.9	0.6	0.7	0.3	0.3	0.2	0.2

Results in this table are expressed as mean ± standard deviation. Mean values with the same superscript letters within a row are not significantly different at $p < 0.05$

(96.1 μm). A similar observation was reported by Rezende et al. (2018), and the relatively large size of freeze-dried particles could be due to its low grindability index.

As presented in Table 2, bulk and tapped density values of spray-dried powders were lower than the values of maltodextrin. In contrast, the bulk and tapped densities of freeze-dried powder were in the range of 0.397–0.513 g/mL, close to the density values of maltodextrin. With the increase of maltodextrin concentration, bulk density was increased for both dried powders. The same trend can be observed in the relation of tapped density and the carrier concentration. Similar results were reported in studies of freeze and spray-drying of sumac extract (Caliskan & Dirim, 2016), spray-drying of Ber juice (Singh et al., 2014) and spray-drying of tea extract (Nadeem et al., 2011).

Carr index (CI) and Hausner ratio (HR) were used to evaluating the flowability and cohesiveness of the smoke powders (Table 2). The powder flowability is classified by value as very good (CI < 15), good (CI = 15–20), fair (CI = 20–35), bad (CI = 35–45) and very bad (CI > 45) (Jinapong et al., 2008). As presented in Table 2, spray-dried powders can be classified as good or fair, and freeze-dried powders as very good. The powder cohesiveness based is classified as low (HR < 1.2), intermediate (HR = 1.2–1.4) and high (HR > 1.4) (Jinapong et al., 2008). The cohesiveness of spray-dried powders was found as intermediate and freeze-dried powders as low. A lower HR value means that the powder is less cohesive and freer to flow. In this study, freeze-dried powders showed better flowability and cohesiveness than the spray-dried ones.

The water content of all the smoke powders was higher than that of maltodextrin, as shown in Table 2. Since the

values were measured by oven-heating at a temperature of 105 °C, the loss of volatile chemicals could contribute to the measurement.

Conclusion

Spray-drying and freeze-drying showed good performance in encapsulating flavour and bioactive compounds of liquid smoke. Both drying processes were able to concentrate the phenolic and flavonoid compounds by approximately two times or more. Freeze-drying had a slightly higher retention efficiency than spray-drying with the same feed solution. A higher concentration of carrier agent also increased retention efficiency in both drying processes. Spray-drying did not damage bioactive compounds at high temperature conditions, while both drying processes led to a partial loss of small molecular carbonyls, furans and phenols. Compared with freeze-dried powder, spray-dried powder had a tenfold smaller particle size, which would be advantageous for food preservation. On the other hand, freeze-dried powder had more handling convenience because of higher flowability and lower cohesiveness.

Declarations

Competing Interests The authors declare no competing interests.

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