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Original Research Paper

Drying behavior and locking point of single droplets containing functional oil

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A B S T R A C T

Inception of the second drying stage called locking point plays a crucial role in the microencapsulation process of functional oils by spray drying. The transition between the first and the second drying periods can directly affect encapsulation efficiency and lipid oxidation by modifying the mechanism of globules migration to the surface of droplet/particles. In this study, the locking point of a single emulsion droplet prepared by incorporating walnut oil into skim milk powder solution was determined using a droplet suspension device. The effects of drying air temperature (80–140 °C), total solid content (12–33% w/w), and oil/wall material mass fraction (0.25–1.00 w/w) were assessed on the droplet/particle drying behavior, shrinkage, and locking point. The latter was achieved by plotting the variation of drying rate against moisture content of the droplet/particle. Moreover, confocal laser scanning microscopy (CLSM) was applied in order to investigate the morphology of the produced particles. Finally, a regression function for estimating the locking point from experimental variables was developed. Overall, such research can pave the way towards increasing the encapsulation efficiency and mitigating the lipid oxidation in industrial-scale spray dryers applied for microencapsulation of functional oils.

1. Introduction

It is well-documented that long chain polyunsaturated fatty acids are essential nutrients for human growth and health. However, incorporation of these ingredients into food systems is limited owing to their extreme susceptibility to oxidation [1]. This, in turn, leads to formation of toxic hydroperoxides during processing, transportation, or storage [2]. Hence, there is an increasing interest amongst researchers in order to develop and apply conservative methodologies such as encapsulation technology to fortify foods with these nutraceuticals. Nowadays, encapsulation has appeared to be an essential technique to incorporate such invaluable sensitive ingredients into food systems. This technique can be efficiently used to protect food ingredients (i.e., flavors, essential oils, lipids, oleoresins, and colorants) against deterioration, volatile losses, and interaction with other ingredients [3,4]. The protective mechanism of this method is to envelop a solid, liquid or gaseous substance (core material) within another substance (wall material) in a sealed capsule [5].

During the past decades, various techniques such as spray drying, spray cooling/chilling, extrusion, fluidized bed coating, freeze drying, coacervation, liposome entrapment, spinning disk, inclusion complexation, centrifugal suspension separation, lyophilization, and cocrystallization have been developed and applied to encapsulate food ingredients [6,7]. Amongst them, spray drying is one of the most commonly technologies employed to encapsulate oils and flavors in a few seconds [8] because it is cheap and easy to use. However, this unique technique suffers from its lower encapsulation efficiency because of fat globules migration to the surface of droplets/particles in the initial stage of the drying process. Furthermore, higher drying air temperatures prevalent in spray drying results in higher lipid oxidation compared with the other techniques applied for the microencapsulation process. Hence, numerous

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research works have been conducted to enhance encapsulation efficiency and to reduce lipid oxidation by improving drying facilities, optimizing drying conditions, stabilizing the emulsion, modifying the emulsion preparation procedure, changing the coating material, etc. [6,9–11]. For instance, Aghbashlo et al. [6] investigated the effects of wall material composition and drying air temperature on encapsulation efficiency and peroxide value of fish oil microcapsules manufactured by spray drying. Furthermore, Carneiro et al. [9] attempted to maximize encapsulation efficiency and minimize lipid oxidation of flaxseed oil during spray drying by evaluating different wall materials at a constant drying air temperature. In another survey, Frascareli et al. [10] studied the effects of total solid content, oil concentration to total solids, and inlet drying air temperature on the encapsulation efficiency of coffee oil. Later, Goula and Adamopoulos [11] investigated the effects of core to wall material ratio, feed solids concentration, inlet air temperature, and drying air flow rate on the encapsulation efficiency of oil extracted from pomegranate seed.

However, most of those reports were made empirically based attempts to enhance the quality of the encapsulation process by carrying out a large number of drying experiments. Despite the industrial importance of functional oil encapsulation by spray drying, there are a limited number of reports on systematic investigations of the physicochemical mechanisms governing the fat globules entrapment and diffusion within an emulsion droplet. For example, Wang et al. [12] investigated the drying behavior of a single emulsion droplet containing Docosahexaenoic acid using the method called glass filament single droplet drying. They used whey protein concentrate and modified starch as wall materials at lower drying air temperatures of 70 and 90 °C. In a similar study, Han Chew et al. [13] evaluated the drying behavior and particle formation from a single high-solid emulsion droplet (based on simplified version of human milk) at an uncommon drying air temperature of 70 °C. Obviously, application of the models developed at lower drying air temperatures for simulating, designing, and optimizing of large-scale drying systems may fail due to the higher drying air temperatures prevalent in industrial spray dryers. Moreover, transition between the first and the second drying stages (locking point) as an influential parameter on the encapsulation quality has not been considered in the previous reports.

It is well-established that the inception of the second drying stage called locking point has a direct effect on particle shrinkage and morphology [14–16] as well as on the encapsulation quality during drying of a droplet containing functional ingredients [10,17]. Hence, an optimal locking point can not only profoundly improve the encapsulation efficiency but also mitigate the lipid oxidation by controlling the fat globules leakage to the surface of droplets/particles. However, to the best of our knowledge, there is no information up to now on the mechanistic investigation of single emulsion droplet drying in order to comprehensively scrutinize parameters affecting the locking point. Therefore, the main objective of this study was to evaluate the effects of drying air temperature, total solid content, and oil/wall material mass fraction on drying behavior, crust formation history, shrinkage, and structure formation of a single droplet prepared by incorporating walnut oil (core material) into skim milk powder solution (wall material) using a droplet suspension apparatus. Such insights can then be employed to simulate, design, and optimize industrial-scale spray drying systems being used for encapsulation of functional ingredients in order to maintain their characteristics.

2. Materials and preparations

Skim milk powder with 1% fat was provided by J.M. Gabler Saliter GmbH, Germany. The chemical composition of skim milk powder is given in Table 1. Walnut oil was generously provided by Sanabio GmbH, Germany. Table 2 presents the fatty acid profile of walnut oil used in this study.

Table 1
Composition of skim milk powder used for experiments.

Component	Mass fraction (%)
Fat	1.0
Protein	35.5
Carbohydrate	51.7
Ash	8.5

The wall material solution was prepared by reconstituting skim milk powder in deionized water at 40 °C. The solution was then cooled and left overnight to enhance the hydration of coating material. Afterwards, oil-in-water emulsion was prepared by gradually adding a given amount of walnut oil to the solution. The emulsion was homogenized for 20 min using an ultrasonic probe with a diameter of 3 mm, power of 70 W, and frequency of 20 kHz (Bandelin, Sonopuls HD2070, Germany). In this study, total solid content (12–33% w/w) and oil/wall material mass fraction (0.25–1.00 w/w) were considered as experimental variables during preparation of emulsions used for drying experiments (Table 3).

The size of oil droplets in the prepared emulsions was measured by a Zetasizer (Malvern Instruments Ltd., UK). It should be noted that the size of oil droplets has a profound effect on the quality of encapsulation.

3. Single droplet drying device

A schematic illustration of the droplet suspension apparatus used throughout this study for drying of a single liquid droplet is depicted in Fig. 1. The advantage of this system over other equipment reported in literature [14] is its capability to dry a single droplet at higher drying temperatures (up to 200 °C). The main unit was a steel cylinder (chamber) with an internal diameter of 24 mm. The chamber was covered with an electrical resistance and an insulator to heat the drying medium and to prevent heat losses to the ambient, respectively. Two thermocouples (Type K, Conatex GmbH, Germany) were set at the air outlet and inside the chamber. Humidity of inlet and outlet air was measured by a dew point mirror (Optidew Vision-Michell Instruments) and an infrared spectrometer (NGA 2000, Fisher-Rosemount), respectively. To insert a droplet onto the tip of a thin polyamide wire, a syringe with a long needle was utilized. An electrical heater was used to heat inlet air at a known humidity ratio. The heated air was then pumped into the drying chamber from the bottom. A mixing unit was used to adjust the inlet air humidity and mass flow rate. To achieve desired drying conditions, saturated and dry air streams with volumetric flow rates in the range of 1–500 ml/min were mixed. The mass flow controllers (F-201CV) were supplied from Bronkhorst Maettig GmbH, Germany. To track and analyze the outer structural changes and droplets/particles size during the drying process, an image recording system including a high speed camera (MC-1009AP/MD, Horn Imaging GmbH, Germany),

Table 2
Fatty acid profile of walnut oil used for experiments.

Fatty acid	Mass fraction (%)
Palmetic acid (16:0)	6.0–8.0
Palmitoleic acid (16:1)	<0.1
Stearic acid (18:0)	1.0–3.0
Oleic acid (18:1)	14.0–21.0
Linoleic acid (18:2)	54.0–65.0
Gamma-linoleic acid (18:3)	10.0–16.0
Eicosenoic acid (20:1)	<0.2