LWT - Food Science and Technology 84 (2017) 617–625

Contents lists available at ScienceDirect

LWT - Food Science and Technology

journal homepage: www.elsevier.com/locate/lwt

Preparation characteristics of monodisperse oil-in-water emulsions by microchannel emulsification using different essential oils

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ARTICLE INFO

Article history:
Received 8 February 2017
Received in revised form
19 April 2017
Accepted 7 June 2017
Available online 14 June 2017

Keywords:
Functional foods
Essential oils
Microchannel emulsification
O/W emulsion
Emulsifiers

ABSTRACT

In this study, MC emulsification was used for obtaining monodispersed essential oil-in-water (O/W) emulsions. The essential oils used were lemongrass oil (LGO), lemon oil (LO), and (R)-(+)-Limonene (LMN). Sodium dodecyl sulphate (SDS), Tween 20, and bovine serum albumin (BSA) were used as hydrophilic emulsifiers. Spontaneous diffusion of Tween 20 aqueous solution into LGO caused generation of the oil droplets containing smaller aqueous ones. The use of BSA resulted in wetting of the channel when formulating LGO droplets, unlike SDS and Tween 20. Monodispersed essential O/W emulsions with an average droplet diameter (d_{avg}) of 23–30 μm and a coefficient of variation (CV) of <5% were observed regardless of the essential oil and emulsifier types. LGO was also diluted with different triglyceride oils at a mixing ratio of 25–75 g/100 g. The droplet generation behaviour changed slightly after each dilution. The advantage of using an oil mixture is that less aqueous phase is diffused into the dispersed phase and that more stable emulsification is obtained. The resultant O/W emulsions were monodisperse with a d_{avg} of 20–30 μm and CV of <5%.

1. Introduction

Essential oils are natural products that exhibit multiple functions such as attractive scent and medicinal properties. They are extracted from natural sources like flowers and herbs by steam distillation and expression. The extracted liquid with a yellowish colour is soluble in lipids and organic solvents and consists of complex volatile compounds which are characterized by strong odour. Plant essential oils are rich in terpenes, terpenoids, phenol derivatives, and aliphatic components (Bakkali, Averbeck, Averbeck, & Waomar, 2008; Sidibe, Chalchat, Garry, Lacombe, & Harama, 2001) and have numerous functional attributes. Citronellol is abundant in citrus oils. Plant essential oils have been widely used in the food flavours, perfumes, and aromatherapy. Essential oils have great importance owing to their biological functions, especially in human health. Nowadays, around 300 essential oils are commercially available for applications in various industries including pharmaceuticals, agronomics, foods, sanitary, cosmetics and perfumes. Essential oils possess antibactericidal, antivirucidal, antifungicidal, antiparasitical, and antinsecticidal activities as well as prooxidant effects.

Lemongrass is a common plant in South and Southeast Asian countries including Thailand. The essential oil extracted from lemongrass has a yellowish colour and mixed fragrance of citrus, grass, and lemon. Key compounds of lemongrass essential oil for medicinal properties include acyclic monoterpene, myrcene, citronellal, geranyl acetate, nerol, neral, geraniol, and high amount of citral (Tovar, Pinto, Wolf-Maciel, Batistella, & Maciel, 2011). Lemongrass essential oil has been used for treating spasmodic affections of the bowels, gastric irritability, and cholera. Lemon oil had been largely used in food, beverage and cosmetic industries. Limonene was the main compound in lemon oil. Investigation on the formulation of essential oil-in-water (O/W) emulsion which had different compositions including pure limonene compound would be useful for industries. Molecular structure and partition coefficient (log P) of citral and limonene was presented in Table 1.

Essential oil-in-water (O/W) emulsions as colloid-base delivery systems have been prepared by both top-down and bottom-up

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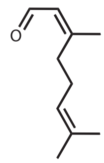
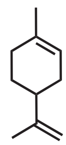
http://dx.doi.org/10.1016/j.lwt.2017.06.014
0023-6438/

Table 1
Molecular structure and partition coefficient of citral and limonene.

	Citral	Limonene
Molecular structure		
Partition coefficient (log P)	3.5	4.4

approaches (Engl, Backov, & Panizza, 2008). Essential O/W emulsions are prepared by the top-down approach using homogeniser(s), and essential O/W microemulsions are prepared by the bottom-up approach by spontaneous emulsification. Recent studies have reported that high-pressure homogenisation is capable of preparing essential O/W emulsions with droplet sizes smaller than 0.1 μm for food and beverage applications (Rao & McClements, 2011; Salvia-Trujillo, Rojas-Grau, Soliva-Fortuny, & Martin-Belloso, 2013). During each cycle of high-pressure homogenisation, a coarse essential O/W emulsion is introduced into an intensive energy field to prepare a submicron emulsion. This high-energy process results in considerable temperature elevation of the emulsion samples, which may cause degradation of heat-sensitive compounds and flavour loss. Although essential O/W submicron emulsions can be obtained using rotor-stator homogenisation, the resultant emulsions are polydisperse and their properties cannot be easily controlled.

Over last two decades, emulsification techniques using microfabricated structures have been developed to controllably prepare monodispersed emulsions (Vladisavljević, Kobayashi, & Nakajima, 2012). Microcapillary device was developed to precisely fabricate double emulsions which droplet size and number of internal droplets could be controlled (Utada et al., 2005). Recently, it increased the emulsion production rate by three-dimensional splitting design (Chen, Gao, Zhang, & Zhao, 2016). Microchannel (MC) emulsification is a promising technique that enables generation of uniform-sized droplets via MC arrays (Kawakatsu, Kikuchi, & Nakajima, 1997; Shinji; Sugiura, Nakajima, & Seki, 2002). Development of parallel MC arrays on the plate provided the advantage on the scale up production of monodispersed droplets (Engl et al., 2008). Droplet generation via MC arrays requires considerably low input energy and no external shear force, which is driven by spontaneous transformation of a dispersed phase that is passed through uniformly sized MCs (Kobayashi, Vladisavljević, Uemura, & Nakajima, 2011; S.; Sugiura, Nakajima, Iwamoto, & Seki, 2001). Droplet generation behaviour can be microscopically observed by firmly attaching MC arrays to an optically flat transparent plate (Kawakatsu, Komori, Nakajima, Kikuchi, & Yonemoto, 1999). Monodispersed emulsions prepared by MC emulsification possess better stability, and more precise analysis of their physicochemical properties is possible (McClements, 2004; Saito, Yin, Kobayashi, & Nakajima, 2006). Previous MC emulsification studies also indicated that the resultant monodispersed microcapsules and multilayer-coated emulsions have the potential to improve droplet stability and control the release rate of functional substances (Iwata, Neves, Watanabe, Sato, & Ichikawa, 2014; Nakagawa, Iwamoto, Nakajima, Shono, & Satoh, 2004).

An important factor for successful MC emulsification is the use of MC arrays with controlled surface properties. For instance, hydrophilic MC emulsification plates are preferred for preparing O/W emulsions, because this can prevent wetting of the dispersed phase to the plate surface. To date, the use of MC emulsification has successfully produced monodispersed vegetable O/W emulsions containing soybean oil, Medium-chain triglyceride (MCT) oil (Kobayashi et al., 2001), and palm oil droplets (Neves, Ribeiro, Fujiu, Kobayashi, & Nakajima, 2008). These monodispersed vegetable O/W emulsions have been stabilized by food-grade surfactants and proteins (Kobayashi & Nakajima, 2002; Saito, Yin, Kobayashi, & Nakajima, 2005). Recently, stability of monodispersed clove oil droplets prepared by MC emulsification was characterized (Purwanti, Neves, Uemura, Nakajima, & Kobayashi, 2015). To the best of our knowledge, there has been no previous MC emulsification study on the preparation of lemongrass O/W emulsions. Therefore, the aim of this study was to prepare monodispersed O/W emulsions by MC emulsification using lemongrass oil and a few other essential oils. We investigated the effects of the types of essential oils and emulsifiers as well as the mixing ratio between the essential oil and vegetable oil on the droplet generation characteristics.

2. Experiment

2.1. Chemicals

Lemongrass oil (LGO) was purchased from Naturas Psychos (Tokyo, Japan). Lemon oil (LO) was purchased from Spectrum chemical Manufacturing Co. (Gardena, USA). (R)-(+)-Limonene (LMN), refined soybean oil (RSO), polyoxyethylene sorbitan monolaurate (Tween 20), sodium dodecyl sulphate (SDS) and bovine serum albumin (BSA) were purchased from Wako Pure Chemical Industries, Ltd. (Osaka, Japan). Medium-chain triglyceride oil (MCT, Sunsoft MCT-8) was obtained by Taiyo Kagaku Co., Ltd. (Yokkaichi, Japan). Milli-Q water was used to prepare all the solutions. All the chemicals were used without any further treatments.

2.2. Measurements of density, viscosity, and interfacial tension

The liquids used for measuring density, viscosity, and interfacial tension were essential oils, essential oil mixtures, and aqueous solutions containing an emulsifier. The liquid density was measured at 25 °C with a density meter (DA-130N, Kyoto Electronics Manufacturing Co., Ltd., Kyoto, Japan). The liquid viscosity was measured with a glass capillary viscometer (SO, Shibata Scientific Technology Ltd., Tokyo, Japan) that was soaked in a water bath at 25 °C. 10 mL of the liquid was pipetted into a capillary viscometer and was subsequently left for 10 min. The viscosity was measured by the time that the meniscus took to move between the upper and lower marks labelled on the capillary. This measurement was repeated triplicate for each liquid sample, and the liquid viscosity was obtained by the following equation:

$$\eta = \rho C t \qquad (1)$$

where η is the dynamic viscosity of liquid (mPa s), ρ is the liquid density (g/cm^3), C is a constant of the capillary tube (cSt/s), and t is time (s). The viscosities of the essential oils and essential oil mixtures were measured by a capillary tube with a constant of 0.104. A capillary tube with a constant of 0.00388 was used for measuring the viscosity of the aqueous solutions. The static interfacial tension between the essential oil and aqueous solutions was obtained by the pendant drop method using an interfacial tensiometer (PD-W, Kyowa Interfacial Science Co. Ltd., Saitama, Japan) at 25 °C.

2.3. Microchannel emulsification

We used an MC emulsification setup that consists of a module equipped with an MC array plate, apparatuses to supply two liquid