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Encapsulation of functional lipophilic components in surfactant-based colloidal delivery systems: Vitamin E, vitamin D, and lemon oil

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ABSTRACT

The fabrication and stability of surfactant-based colloidal delivery systems (microemulsions and emulsions) suitable for encapsulation of lipophilic active agents (vitamins and flavours) was investigated. An emulsion titration method was used to study the influence of surfactant type (Tween 20, 60 and 80) and oil type (Vitamin E, vitamin D_3 and lemon oil) on the incorporation of lipophilic components into surfactant micelles. Oil-in-water emulsions were formed and then different amounts were titrated into surfactant micelle solutions. The influence of surfactant-to-oil ratio (SOR) and oil type on the formation of colloidal dispersions was examined using dynamic light scattering and turbidity measurements. SOR, oil type, and surfactant type had a pronounced influence on the nature of the colloidal dispersions formed. Microemulsions could not be formed using vitamin D or E in 1% Tween solutions, due to the relatively large size of the lipophilic molecules relative to the hydrophobic interior of the surfactant micelles. On the other hand, microemulsions could be formed from lemon oil at relatively high SORs. There was not a major impact of non-ionic surfactant type (Tween 20, 60 or 80) on the formation and properties of the colloidal dispersions. However, Tween 20 micelles did appear to be able to solubilise less lemon oil than Tween 60 or 80 micelles, presumably due to their smaller dimensions. This study provides useful information for the rational design of food grade colloidal delivery systems for encapsulating flavour oils, oil-soluble vitamins, and other functional lipids for application in foods and beverages.

1. Introduction

There are a number of commercial applications where lipophilic active components (such as bioactive lipids, flavours, antimicrobials, antioxidants, nutraceuticals, and drugs) need to be incorporated into aqueous media so that they are suitable for oral consumption, e.g., foods, health care products, and pharmaceuticals (Given, 2009; McClements, Decker, Park, & Weiss, 2009; Sonneville-Aubrun, Simonnet, & L'Alloret, 2004; Velikov & Pelan, 2008). One of the most convenient means of achieving this aim is to encapsulate the lipophilic active components within colloidal-based delivery systems, such as microemulsions, nanoemulsions, or emulsions (Acosta, 2009; McClements, 2011; Sagalowicz & Leser, 2010; Velikov & Pelan, 2008). This type of delivery system consists of small lipid-based particles suspended within an aqueous medium (McClements, 2011). In this manuscript, we focus on colloidal delivery systems suitable for utilisation within food and beverage products. The rational design of edible colloidal delivery systems for specific applications depends on a number of different factors (McClements et al., 2009). First, the delivery system must be fabricated entirely from food grade ingredients using economically viable processing operations. Second, the delivery system must remain physically and chemically stable over the range of environmental conditions (e.g., temperature fluctuations, light, oxygen, and mechanical forces) that food and beverage products are usually exposed to during their manufacture, storage, and utilisation. Third, the delivery system should not adversely influence the physicochemical or sensory properties of the food or beverage product that it is incorporated into, e.g., appearance, texture, or flavour. Fourth, the delivery system should be capable of releasing the active component in the appropriate place within the human body after consumption, e.g., the oral cavity for flavour molecules, or the gastrointestinal tract for vitamins and nutraceuticals. It is therefore highly challenging to develop effective delivery systems for utilisation within commercial applications. There appears to be no single strategy that is suitable for encapsulation of every kind of lipophilic component. Instead, colloidal delivery systems must be carefully designed for each particular application.

One of the biggest challenges is to identify the most appropriate colloidal delivery system for each particular type of lipophilic component that needs to be encapsulated (Given, 2009; McClements, 2011; Velikov & Pelan, 2008). In this study, we therefore examined the suitability of various surfactant-based delivery systems

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(microemulsions and emulsions) for encapsulation of three different functional lipophilic components: vitamin E acetate; vitamin D_3; and lemon oil (Fig. 1). Vitamin E is a fat-soluble vitamin that exists in eight different forms, with the α-tocopherol form being the most active in humans (Traber & Packer, 1995). Vitamin E is considered to be one of the most potent lipid-soluble antioxidants *in vivo* (Hoppe & Krennrich, 2000). It functions as the major radical scavenging antioxidant in lipoproteins and efficiently interrupts the chain propagation of lipid oxidation, thus protecting polyunsaturated fatty acids and low density lipoproteins from oxidation. For this reason, there is increasing interest in fortifying many foods with vitamin E (Sagalowicz & Leser, 2010). Some studies suggest that the bioavailability of vitamin E may be increased when it is delivered in colloidal form rather than in bulk form (Feng, Wang, Zhang, Wang, & Liu, 2009). Vitamin D commonly comes in two different forms: vitamin D_2 (ergocalciferol) and vitamin D_3 (cholecalciferol). Vitamin D_2 found in plants, is the product of ultraviolet B (UVB) (290–315 mm) irradiation of ergosterol, and can be consumed as a supplement or in fortified foods (Holick, 2007; Lee, O'Keefe, Bell, Hensrud, & Holick, 2008). Vitamin D_3 a product of UVB irradiation of 7-dehydrocholesterol, and is synthesized in the human epidermis or consumed in the form of oily fish, fortified foods, or supplements. A number of colloidal delivery systems have previously been utilised to encapsulate oil-soluble vitamins (such as vitamins A, D and E), including, nanoemulsions (Chen & Wagner, 2004; Shukat & Relkin, 2011), emulsions (Sanchez-Paz et al., 2008), solid lipid nanoparticles (Dingler, Blum, Niehus, Muller, & Gohla, 1999; Ma et al., 2007), microemulsions (Chiu & Yang, 1992), and filled hydrogel particles (de Britto, de Moura, Aouada, Mattoso, & Assis, 2012).

For comparison purposes, we also studied the encapsulation of an oil-soluble flavour (lemon oil) that has quite different molecular characteristics to the oil-soluble vitamins. Lemon oil is one of the most common flavour oils used to prepare soft drinks (Edris & Abd El-Galeel, 2010; Given, 2009; McClements, 2005). The composition of lemon oil depends on its biological origin, as well as the various processing steps used to isolate, purify and refine it. Commercial lemon oils contain a wide variety of lipophilic components with different molecular characteristics. The most important flavour constituent is citral, but other constituents are also important, such as limonene, myrcene, octanal, and gamma-terpinene (Schieberle & Grosch, 1988). Various types of colloidal delivery systems have also been developed to encapsulate flavour oils, such as microemulsions, nanoemulsions, and emulsions (Rao & McClements, 2011a, 2011b, 2012b; Yang, Tian, Ho, & Huang, 2011).

In this study, we investigated the influence of oil type and concentration on the formation and stability of colloidal dispersions prepared using three different food-grade non-ionic surfactants (Tween 20, 60 and 80). In particular, we wanted to understand the influence of different lipophilic components on the design and fabrication of colloidal delivery systems suitable for application within the food and beverage industry.

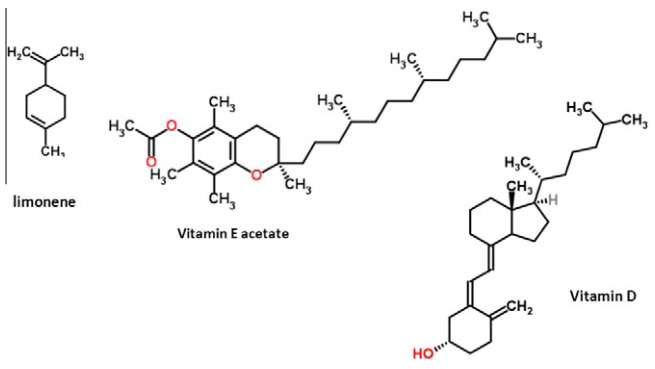

Fig. 1. Molecular structures of different lipophilic components used in this study: vitamin D_3; vitamin E acetate; limonene (the major constituent of lemon oil). The structures were obtained from ChemSpider.

2. Material and methods

2.1. Materials

Vitamin D_3 (BASF, Ludwigshafen, Germany) in medium chain triglycerides and acetate vitamin E were kindly donated by PepsiCo (Valhalla, NY). Lemon oil reference (4×, SC020207) with ≈60% limonene, <18% citral + citral acetate, and <15% gamma-terpinene was donated by International Flavours and Fragrances (Union Beach, NJ). Non-ionic surfactants (Tween 20, Tween 80) were purchased from Sigma–Aldrich Co. (St. Louis, MO). Tween 60, citric acid and sodium benzoate were provided by PepsiCo (Valhalla, NY). Double distilled water was used in the preparation of all solutions and colloidal dispersions.

2.2. Characterisation of the lipophilic compounds

The physicochemical properties of the three different oil phases were characterised to better understand their influence on the formation and stability of colloidal dispersions. The density of the three compounds was measured using a digital density metre (DE50, Mettler-Toledo Laboratories, UK). The oil–water interfacial tension of the different oil phases was measured using a drop shape analysis method (DSA 100 Tensiometer, Kruss, Germany). The viscosity of vitamin E (which was highly viscous) was measured using a dynamic shear rheometer (Model KVX2000, Kinexus Rotational Rheometer, Malvern Instruments, Malvern, UK). The viscosity of lemon oil and vitamin D_3 (which had relatively low viscosities) was measured using a capillary viscometer. The refractive index of the oils was measured using a handheld refractometer (Fisher Scientific). All measurements were performed at least in triplicate. The data from these measurements is compiled in Table 1.

2.3. Emulsion preparation

Initially, an aqueous surfactant solution was prepared by dispersing 1% (w/w) of the selected surfactant in an acidic buffer solution (0.8% citric acid, 0.08% sodium benzoate, pH 2.6). A 10% w/w oil-in-water stock emulsion was then formed by blending 10% w/w oil phase with 90% w/w aqueous phase using a high-speed blender for 1 minute, and then passing the resulting coarse emulsion through a high pressure homogenizer five times at 9000 psi (Microfluidics 110L, Microfluidics Corp., Newton, MA, USA) to reduce the particle size. After preparation, these stock emulsions were stored at 20 °C prior to utilisation.

2.4. Emulsion titration method

The ability of the different oils to form microemulsions or emulsions under a particular set of experimental conditions was determined using an emulsion titration experiment (Fig. 2). Aliquots of stock emulsion (10% w/w oil, 1% w/w surfactant) were titrated into aqueous surfactant solutions (1% w/w surfactant) to form a series of samples containing increasing amounts of oil phase, i.e., decreasing surfactant-to-oil ratios. The samples were then stored overnight at ambient temperature and their particle size, turbidity, and appearance were recorded. These experiments were performed with a number of different non-ionic surfactants: Tween 20, Tween 60 and Tween 80.