Journal of Food Engineering 240 (2019) 56–64

Contents lists available at ScienceDirect

Journal of Food Engineering

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

# Effect of mono- and diglycerides on physical properties and stability of a protein-stabilised oil-in-water emulsion

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ARTICLE INFO

*Keywords:*
Mono- and diglycerides
Milk protein
Droplet size
Creaming
Stability
Emulsions

ABSTRACT

Mono- and diglycerides (MDGs) are emulsifiers used to modify physical properties and creaming stability in protein-stabilised emulsions. This study aims to understand the effect of different MDGs and sodium stearate on oil droplet size and creaming stability of emulsions. Model emulsions with eight MDG compositions and a control (protein only) were prepared by microfluidisation. Emulsion droplets and creaming stability were characterised by droplet size, zeta potential, viscosity and creaming index during aging (28 days at 25 °C). Emulsions containing 0.2% MDGs produced 15–30% smaller oil droplets and 17–27% lower polydispersity indices compared to the control. Sodium stearate (6% of MDGs) increased zeta potential by 12.6–17.3 mV in emulsions containing saturated MDGs and 1.8–5.0 mV in unsaturated MDGs. Unsaturated MDGs showed better creaming stability than the control after 28 days of aging with no improvement observed for saturated MDGs. Unsaturated MDGs are promising emulsifiers to improve creaming stability of protein-stabilised emulsions.

## 1. Introduction

Oil-in-water emulsions are the basis of many food products such as mayonnaise, whipped cream, salad dressings, sauces, soup, milk, infant formulae and beverages (McClements, 2015). Emulsions are important in food systems to deliver nutrients, taste and flavour (Hu et al., 2017). Oil-in-water emulsions can contain fat levels as high as 70–80% in mayonnaise and as low as 0.2% fat in lemon juice. Homogenised whole milk contains about 3.3% milk fat and some flavoured milk-based beverages have as high as 10% fat. Fat plays an important role in modulating the sensorial experiences in food such as flavour, mouthfeel and creaminess.

An oil-in-water emulsion consists of fine oil globules dispersed in the aqueous phase in the presence of at least an emulsifier. In the milk and beverage industry, homogenisation is used to overcome the energy barrier of forming an emulsion (Van Der Meeren et al., 2005) or to reduce the size of fat globules into smaller one (Bylund, 2015). Emulsifiers are essential to initiate formation, provide stability and produce desirable physicochemical properties of the emulsions. Emulsifiers adsorb to the oil-water interface created during the homogenisation process and lower the interfacial tension, which allows the formation of stable oil globules.

An emulsifier is a type of surface-active agent that can alter the surface tension of a fluid (Hernandez Sanchez et al., 2015; Krog and Vang Sparsø, 2003). Emulsifiers contain both the hydrophilic and hydrophobic functional groups in the same molecule. Milk protein and low-molecular weight emulsifiers such as mono- and diglycerides are common emulsifying agents in milk-based beverages. Milk is a naturally occurring emulsion, where casein and whey proteins have surface-active structures that facilitate emulsion formations in dairy products (Adjonu et al., 2014; Genot et al., 2003; McClements, 2004).

Commercial mono- and diglycerides are generally mixtures and denote the partially hydrolysed structures of triglycerides (Krog and Vang Sparsø, 2003). Monoglycerides have one hydrophobic fatty acid esterified to the hydrophilic glycerol molecule while diglycerides have two fatty acids. Commercial mono- and diglycerides have a wide range of compositions in term of monoglyceride content, fatty acid types and presence of a co-emulsifier, such as sodium stearate. Molecular distillation is the typical manufacturing process to increase the monoglyceride content in the emulsifier. The types of fatty acid influences the melting behaviour of the mono- and diglycerides, where mono- and diglycerides composed of saturated fatty acids usually have higher melting points than the unsaturated version of the same carbon number. Mono- and diglycerides are oil-soluble molecules and have very low solubility in water, making dispersibility in water a challenging task in the industry.

In order to improve dispersion of mono- and diglycerides in water, co-emulsifiers are added to mono- and diglycerides and have been

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https://doi.org/10.1016/j.jfoodeng.2018.07.016
Received 23 December 2017; Received in revised form 29 June 2018; Accepted 12 July 2018

marketed as a 'self-emulsified' form for many years (Brokaw and Lyman, 1958). Effective co-emulsifiers for mono- and diglycerides are polar types that are soluble in water, such as sodium stearate, which improves dispersibility of mono- and diglycerides in water (Krog and Vang Sparsø, 2003). Sodium stearate has both hydrophilic and hydrophobic components, which allows it to function as an emulsifier. Brokaw and Lyman (1958) discussed the effect of co-emulsifiers on gel preparation with mono- and diglycerides, however, there is no literature to explain the effect of sodium stearate on emulsion properties and stability.

Most commercial products such as milks, ice-creams and spreads contain at least an emulsifier to stabilise the emulsion system. Complete separation of oil and water is rare, but the differences in density between oil globules and the water phase often lead to formation of a ring at the top of the emulsion. This phenomenon is known as creaming (Taherian et al., 2006). Creaming is defined as the upward movement of oil globules in the emulsion due to their lower density compared to the water phase (McClements, 2015). Creaming stability is the resistance of the emulsion to cream in the emulsion. The cream layer can be easily dispersed back into the emulsion by shaking. Creaming is not a problem for products in a carton or can, however, creaming in clear packaging may give consumers the perception of inferior quality.

While milk protein and mono- and diglycerides both adsorb to the oil-water interface, the interaction between them at the interface can influence droplet properties, flow behaviour and emulsion stability. The use of a low-molecular weight emulsifier is found to affect emulsion stability due to the capability to displace milk protein from the oil-water interface (Dickinson and Tanai, 1992; Matsumiya et al., 2010; Munk et al., 2013). However, oil-soluble emulsifiers such as mono- and diglycerides do not displace protein at low concentrations (Dickinson and Hong, 1994). Little information is available on the effect of mono- and diglyceride composition including fatty acid type, monoglyceride content and sodium stearate content, and their effect on physical properties and emulsion stability. The effect of monoglyceride content and sodium stearate content on emulsifiers' behaviour in emulsions remains a gap in literature. The objective of this study is to investigate the effect of different mono- and diglyceride composition and sodium stearate on droplet size distribution, zeta potential, viscosity and stability of an emulsion.

## 2. Materials and methods

### 2.1. Materials

Saturated mono- and diglycerides, Grindsted Mono-Di HP 40 and Dimodan HP, were donated by Danisco New Zealand while unsaturated mono- and diglycerides, Radiasurf 7148 and Radiamuls MG 2905K, were a gift from Oleon Malaysia (Table 1). Sodium caseinate and whey protein concentrate 80% were provided by Tatua Co-operative Dairy Company Ltd. (Morrinsville, New Zealand). Commercially available hydrogenated coconut oil was obtained from Davis Food Ingredients (Auckland, New Zealand). Sodium stearate was provided by Peerage Product Limited (Christchurch, New Zealand). Caster sugar was purchased from the local supermarket. Sodium azide was purchased from Sigma-Aldrich.

**Table 1**
Mono- and diglycerides composition.

| Sample | Description |
|---|---|
| Sat-40 | Saturated mono- and diglycerides with low (40% w/w) monoglyceride content |
| Sat-90 | Saturated mono- and diglycerides with high (90% w/w) monoglyceride content |
| Unsat-50 | Unsaturated mono- and diglycerides with low (50% w/w) monoglyceride content |
| Unsat-90 | Unsaturated mono- and diglycerides with high (90% w/w) monoglyceride content |

### 2.2. Preparation of model emulsions

Twelve model emulsions with different mono- and diglyceride composition (2 levels of fatty acid saturation x 2 levels of monoglyceride content x 3 levels of sodium stearate) and a control emulsion were prepared by microfluidisation. Each sample formulation was prepared in duplicates except control with four replicates. Samples were prepared across four days with a control on each day to evaluate emulsion repeatability across days. Water-phase ingredients consisted of sodium caseinate (0.8% w/w), whey protein concentrate (0.2% w/w), caster sugar (6% w/w) and sodium azide (0.02% w/w), reconstituted in Milli-Q water at 50 °C. The water-phase was continuously agitated using an Ultra-Turrax (IKA-Werke GmbH and co. KG, Stufen, Germany) at 10,000 rpm and was simultaneously heated to 75 °C. The four compositions of mono- and diglycerides (0.2% w/w) (Table 1) were added to hydrogenated coconut oil (1.1% w/w) at 65 °C to form the oil-phase. Sodium stearate was added to mono- and diglycerides at 3% or 6% w/w level as required. The oil-phase was mixed with the water-phase solution which was then homogenised for 3 min at 75 °C to form a coarse emulsion. The coarse emulsion was passed through a microfluidiser for a single time at 550 bar and 65 °C. The microfluidiser was pre-heated by circulating water at 70 °C. Each emulsion was prepared in duplicate. The control emulsion was prepared using the same method with the mono- and diglycerides not added.

### 2.3. Droplet size and polydispersity index of model emulsions

Droplet size and size-range distributions of the model emulsions were measured by dynamic light scattering using a Malvern Zetasizer Nano S (Malvern Instruments Ltd, Malvern, Worchestershire, UK). The measurements were carried out using diluted samples of 1:1000 with 6% sugar solution. The diluted sample was analysed for 60 s at 25 °C in triplicate. Duplicate measurements were carried out for each sample. The average droplet size was expressed as the intensity-weighted mean diameter (Z-average) and the width parameter was presented as the polydispersity index (Malvern Instruments Limited, 2011).

### 2.4. Zeta potential

Zeta potential of the model emulsions was determined using the Malvern Zetasizer Nano S. Samples were diluted at 1:20 with 6% sugar solution. The diluted sample was analysed at 25 °C in triplicate. Duplicate measurements were carried out for each sample.

### 2.5. Viscosity

Viscosity of the model emulsions was performed using a double gap geometry (measuring cup DG42 PO and rotor DG43 DIN53544 Ti) and 11.5 mL sample volume at 20 °C with a Rheostress 1 (Haake, Germany) rheometer. Measurements were carried out in duplicate for each sample at the shear rate range of 0–200 $s^{-1}$. The average viscosity was expressed as apparent viscosity at a shear rate of 100 $s^{-1}$. This shear rate resembles common processes such as pumping, pouring, mixing and stirring to prepare food emulsions (McClements, 2015; Shama and Sherman, 1973).

### 2.6. pH

The pH of model emulsions was tested at room temperature using a pH209 (HANNA Instruments, Woonsocket, RI) pH meter.