Trends in Food Science & Technology 55 (2016) 98–108

Contents lists available at ScienceDirect

Trends in Food Science & Technology

journal homepage: http://www.journals.elsevier.com/trends-in-food-science-and-technology

Review

# Emulsion microgel particles: Novel encapsulation strategy for lipophilic molecules

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ARTICLE INFO

*Article history:*
Received 18 April 2016
Received in revised form
1 June 2016
Accepted 12 July 2016
Available online 18 July 2016

*Keywords:*
Emulsion microgel particle
Filler-matrix interaction
Whey protein
Swelling
Matrix erosion

ABSTRACT

*Background:* Lipophilic molecules such as flavours, essential oils, vitamins and fatty acids are difficult to deliver in food matrices owing to their limited solubility, rapid oxidation and degradation during physiological transit. Among the technologies available to deliver lipophilic molecules, emulsion microgel particles are a relatively new class of soft solid particles of discrete size, shape, and interesting release properties.

*Scope and approach:* Relevant literature concerning the processing of emulsion gels and emulsion microgel particles has been reviewed. Factors affecting the mechanical properties of protein-stabilised emulsion gels with key emphasis on the role of "active" and "inactive fillers" are discussed. Technologies for creation of emulsion gel particles using top-down and bottom-up approaches has been covered. Special attention was dedicated to the release mechanisms from emulsion microgel particles via swelling and erosion.

*Key findings and conclusions:* Emulsion gels with "active fillers" offer the potential to create emulsion microgel particles using top-down approach. Polymer extrusion, multiple emulsion templating, fluid gels are few routes for creating emulsion microgel particles using bottom-up approaches. Although whey protein has been well researched, modified starch, plant proteins need to be investigated for design of new emulsion microgel particles that can act as surfactant and bulk gelling agents in their own right through intelligent tuning of processing conditions. If designed carefully with an end goal of "controlled delivery" in mind, responsiveness to oral temperature, gastric enzymes, intestinal pH etc, can be built into emulsion microgel particles so that they may find novel applications in food, pharmaceutical and personal care industries.



## 1. Introduction

Lipophilic active molecules, such as fat soluble vitamins, flavourings, fatty acids and essential oils pose challenges for their application in food matrices as they are water insoluble. They tend to oxidize rapidly in the presence of air, light and heat. Additionally, due to their hydrophobic nature, most of these compounds are difficult to deliver in human physiology and are generally partially absorbed by the body or their biological activity is partly or fully degraded during their transit. Thus, there is a huge need to protect these lipophilic compounds without environmental degradation and tailor their release at a physiological site, such as burst release of flavours or essential oils in mouth or protect the omega-3 fatty acids during gastric transit and release them in the intestine.

A wide range of technologies have been developed to encapsulate lipid molecules, such as emulsions, emulsion gels, liposomes, micelles, nanoparticles, etc. Each of these have their own specific advantages and disadvantages in terms of protection, delivery, cost, regulatory status, ease of use, biodegradability and biocompatibility (McClements & Li, 2010). Among these, emulsions gels are an alternative technique that allows stabilization and delivery of lipophilic compounds in food matrices. Emulsion gels are frequently produced in food products, such as, sausages, yogurt, dairy desserts, cheese, etc. (Mun, Kim, Shin, & McClements, 2015). Currently, there has been an upsurge in research efforts in the domain of emulsion gels resulting in engineering of novel soft solids, such as emulsion fluid gels and emulsion microgel particles. To understand different terminologies used in the literature, definitions of each of these classes of emulsion gels with their corresponding microstructures are included in Table 1.

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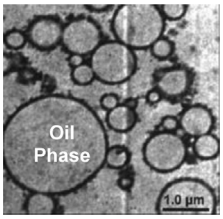
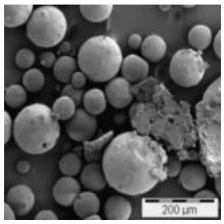
http://dx.doi.org/10.1016/j.tifs.2016.07.006
0924-2244/

**Table 1**
Definitions and microstructures (at various length scales) of different emulsion gel based strategies for delivery of lipophilic molecules. (A) Transmission electron micrograph (TEM) of emulsion gels (reproduced from Anton, Le Denmat, Beaumal, and Pilet (2001). (B) Scanning electron micrograph (SEM) of emulsion microgel particle (reproduced from Egan et al., 2013).

| Nomenclature and microstructure | Description | References |
|---|---|---|
| A) Emulsion gel   | "Emulsion gels", also named as "emulsion hydrogel", "emulgel", "emulsion-filled gel" are defined as soft solids where emulsified lipid droplets are entrapped in a gel matrix. Generally, the emulsified lipid droplets are referred to as "fillers" and the gelled aqueous phase is referred to as the "matrix". They are formed either by suitable application of temperature, pH, ionic strength to the emulsion made with high concentration of biopolymer (especially protein in case of protein-based emulsion gel) or by addition of a gelling agent to the continuous phase forming physical cross-links between emulsion droplets. It has the advantages of both hydrogels (*i.e.* thermodynamic stability) and emulsions (i.e., delivery of lipid soluble molecules). | (Briuglia, Urquhart, & Lamprou, 2014; Dickinson, 2012; Oliver, Scholten, et al., 2015; Sarkar et al., 2015; Satapathy et al., 2015) |
| B) Emulsion microgel particle   | "Emulsion microgel particles", "emulsion filled hydrogel particles", "emulsion gel beads" or "fluid emulsion gel" are a new class of particles formed by encapsulating several emulsion droplets into a soft gel-based shell either using a top-down or a bottom-up approach. Fluid emulsion gels are a specific case of emulsion microgel particles as they are formed by applying shear to the continuous phase whilst gelling the emulsion droplets. | (Beaulieu et al., 2002; Ching et al., 2016; Dickinson, 2015; Egan et al., 2013; Garrec & Norton, 2012; Moakes et al., 2015b; Sung et al., 2015) |

Emulsion microgel particles are a relatively new class of soft solids, particularly in food research. Emulsion microgel particles have similar polymer chemistry to emulsion gels though their physical arrangement and scale is different. Both emulsion gels and emulsion microgel particles have oil and gel phases but microgels are much smaller discrete particles with well-defined spherical shape (Thorne, Vine, & Snowden, 2011). In emulsion gels, the emulsion droplets are stabilised by emulsifiers and heterogeneously distributed in a continuous gel matrix whereas in emulsion microgel particles, emulsion droplets are stabilised by an emulsifier and gelling agent, creating a soft solid shell around several emulsion droplets which are then incorporated into a continuous gel matrix. Therefore, in emulsion gels before gelation of the matrix, emulsion droplets are rather mobile due to Brownian motion and can be unstable due to faster flocculation, coalescence and creaming. Meanwhile, in emulsion microgel particles, several emulsion droplets are entrapped into a soft solid shell providing better control of droplet size, mobility and mechanical properties (Mun, Kim, & McClements, 2015; Ruffin, Schmit, Lafitte, Dollat, & Chambin, 2014; Zhang, Zhang, Decker, & McClements, 2015; Zhang, Zhang, Tong, Decker, & McClements, 2015). Additionally, microgel particles have been demonstrated to protect against oxidation lipophilic compound such as polyunsaturated fatty acids (Augustin & Sanguansri, 2012; Beaulieu, Savoie, Paquin, & Subirade, 2002; Chung, Degner, Decker, & McClements, 2013; Mao & Miao, 2015; Matalanis, Jones, & McClements, 2011; Velikov & Pelan, 2008).

The microgel particle encapsulation method has been described as "smart" because the size, physicochemical properties of these particles are tuneable and allow the microgel to swell or de-swell, as well as degrade in response to specific temperature, pH, ionic strength, enzymatic conditions (Ballauff & Lu, 2007; Kawaguchi, 2014; Shewan & Stokes, 2013; Wei, Li, & Ngai, 2016). Hence, emulsion microgel particles can be effective for site-dependent release of lipophilic bioactives (Ching, Bansal, & Bhandari, 2016). For instance, incorporation of filled hydrogel particles in low fat dairy products have been found to retain the sensory attributed of the dairy product by controlling the release of lipophilic aroma and mimicking fat droplet functionality (Chung et al., 2013; Joye & McClements, 2014; Malone & Appelqvist, 2003; Malone, Appelqvist, & Norton, 2003; Oliver, Berndsen, van Aken, & Scholten, 2015; Oliver, Scholten, & van Aken, 2015; Pizzoni, Compagnone, Di Natale, D'Alessandro, & Pittia, 2015; Zhang, Zhang, Chen, Tong, & McClements, 2015). Hydrogel particles encapsulating hydrophilic compounds have been well studied and reviewed by Joye and McClements (2014) and McClements (2015) as well as protein-based microgels has been investigated by Dickinson (2015). Nevertheless to our knowledge, no review on emulsion microgel particles encapsulating lipophilic compounds is available. Hence, this review aims to detail the formation of emulsion microgel particles and their application for controlled release of lipophilic compounds.

We begin by covering the basic processing steps of emulsion gels since this sets the scene for the top-down approach of making emulsion microgel particles from the parent emulsion gel. In the second section, we discuss the role of oil droplet, "filler" or gel "matrix", and interactions that govern the mechanical properties of emulsion gels. We have focussed mainly on whey protein (from bovine milk) and also covered the few available publications on modified starch-based systems, since both these biopolymers have