



## Review

## Food-grade particles for emulsion stabilization



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## ABSTRACT

The use of solid, food-grade particles as emulsion stabilizers is highly promising in the field of food science and technology. Food-grade particles (e.g. fat and wax crystals, protein particles, and protein-polysaccharide complexes) can form alternatives to inorganic particles (e.g. silica, alumina or clays), which are incompatible and mostly unserviceable when considering food and nutraceutical applications. This review provides a concise insight into the concept of Pickering stabilization with the emphasis on the available food-grade particles that have been researched in this field. The innovative food applications of particle-stabilized emulsions are also discussed.

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## 1. Introduction

In the last few years, micro- and nano-particles of biological origins have been extensively explored as colloid stabilizers (i.e. stabilization of liquid–liquid and/or liquid–vapor interfaces) for application in bio-related fields, such as foods and nutraceuticals. Interestingly, this concept has been applied for centuries in food products, without being scientifically researched. For instance, in mayonnaise (a concentrated oil-in-water emulsion which has been used for ages), the fine mustard particles contribute to physical and colloidal stability of the emulsion by preventing the coalescence of oil droplets (Binks, 2007).

As its name implies, particle stabilized emulsions use solid particles to obtain stable emulsions. This concept was first introduced as Pickering stabilization in the early 1900s (Pickering, 1907; Ramsden, 1903). The mechanism of stabilization is quite different from conventional emulsion stabilization, in which the oil-water interface is generally stabilized using low molecular weight components with amphiphilic characteristics. These low molecular weight emulsifiers decrease the interfacial tension and introduce

stability with steric hindrance and/or electrostatic repulsion between the droplets (Rayner et al., 2014). Solid particles can also adsorb on the oil-water interface and create a mechanical barrier that protects the emulsion droplets against destabilization (Dickinson, 2012). However, unlike low molecular weight emulsifiers, the adsorption of particles is considered to be irreversible. Therefore, Pickering emulsions show an outstanding stabilization against coalescence and Ostwald ripening in comparison to emulsions stabilized with low molecular weight emulsifiers (Kargar, Fayazmanesh, Alavi, Spyropoulos, & Norton, 2012; Timgren, Rayner, Sjöö, & Dejmek, 2011).

Pickering emulsion droplets are stabilized by particles which feature a partial dual wettability by both the oil and water phase. This property leads to the accumulation of the particles at the oil-water interface. Aforementioned particles can stabilize both oil-in-water (O/W) and water-in-oil (W/O) emulsions. The type of emulsion that is preferably stabilized by a particle can be deduced from the contact angle at the oil–particle–water interface (Fig. 1). The liquid phase that wets the solid more than the other liquid, will likely be the continuous phase. Particles with a contact angle smaller than 90° will form an O/W emulsion, while particles with a contact angle greater than 90° usually form W/O emulsions (Finkle, Draper, & Hildebrand, 1923).

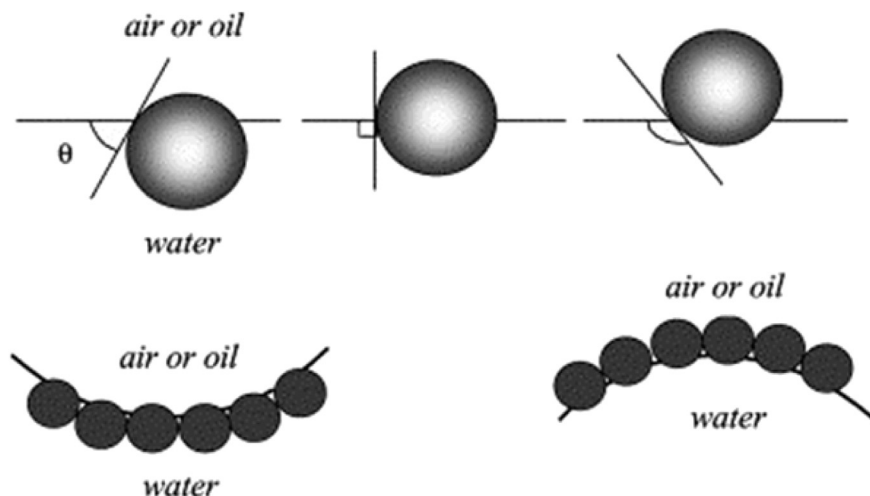
Particle wettability and surface activity of insoluble particles may be further altered by adsorption of suitable surfactants or by chemical modification (Binks, Rodrigues, & Frith, 2007). However,

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**Fig. 1.** Bending behavior of oil–water interface for a contact angle less than  $90^\circ$  (left) and greater than  $90^\circ$  (right). The contact angle equal to  $90^\circ$  corresponds to the point of phase inversion. Reproduced with permission from Elsevier (Aveyard et al., 2003).

the implementation of modified particles in the food and pharmaceutical industry is quite limited because the required modifications to render them suitably surface-active are most often not permitted in foods. Notable exceptions are the inert silica particles with a hydrophobic coating of lecithin which are used as emulsifiers of triglyceride O/W emulsions (Eskandar, Simovic, & Prestidge, 2007), starch granules chemically modified with octenyl succinic anhydride (OSA) to increase their hydrophobicity (Matos, Timgren, Sjöo, Dejmek, & Rayner, 2013; Rayner, Timgren, Sjöo, & Dejmek, 2012) and other types of food-grade modified starches such as distarch phosphate and acetylated distarch phosphate (Prochaska, Kędziora, Le Thanh, & Lewandowicz, 2007). Also some hydrophobically modified cellulose fibers, such as hydroxypropyl methylcellulose phthalate (HP), can stabilize emulsions and produce superstable Pickering foams and emulsions (Wege, Kim, Paunov, Zhong, & Velev, 2008).

Particles used to stabilize food emulsions should be food-grade and therefore, the options are limited to particles of biological origin, such as fat crystals, cellulose derivatives, dairy and plant proteins, protein-polysaccharide complexes and flavonoids (Rayner et al., 2014). Pickering emulsions stabilized with food-grade particles have a wide application range. They can be used to encapsulate active ingredients or to enhance the nutritional profile, texture or structure of several processed foods (Berton-Carabin & Schroën, 2015).

This review aims to provide an insight in the Pickering stabilization concept and the preparation and characterization of the particles and the emulsions. Moreover, an overview of food-grade particles is also given with particular emphasis on innovative food applications.

## 2. Kinetic and energetic aspects of solid-particle adsorption on the interface

The kinetic aspects of solid-particle adsorption on the oil-water interface are different compared to those of low molecular weight surfactant. This is caused by the large particle size of the solid particles, compared to that of surfactant and protein molecules. The adsorption of solid particles results in stronger capillary forces between the particles trapped in the liquid films (Velikov, Durst, & Velev, 1998). Therefore, adsorbed solid particles display a high desorption energy barrier, but also show a high barrier to particle adsorption (Tcholakova, Denkov, & Lips, 2008). Consequently, the

particle adsorption shows much slower kinetics than the adsorption of low molecular weight surfactants but, once adsorbed on the interface, the adsorption of particles is considered to be irreversible due to the high desorption energy barrier. Again, this is in contrast to low molecular weight surfactants, which are in rapid and dynamic equilibrium between the oil-water interface and the bulk phase.

The energy of attachment of a particle to a fluid–fluid interface is not only related to the contact angle, but also to the tension of the oil-water interface  $\gamma_{ow}$ . Since the Pickering particles are small, gravity and buoyancy effects can be neglected. The energy ( $E$ ) required to remove a particle from the interface is given by Eq. (1).

$$E = \pi r^2 \gamma_{ow} (1 - |\cos\theta|)^2 \quad (1)$$

where  $r$  is the radius of the Pickering particle,  $\gamma_{ow}$  the surface tension of the oil-water interface and  $\theta$  the contact angle. This detachment energy is in the order of  $10^3$  kT for a 10 nm particle at angles of around  $90^\circ$ , but falls quite rapidly as the particle contact angle decreases or increases (Dickinson, 2010). When the particles are either too hydrophilic (low  $\theta$ ) or too hydrophobic (high  $\theta$ ), they remain in the aqueous or oil phase respectively, giving rise to very unstable emulsions (Binks, 2002).

## 3. Stability of Pickering emulsions

In general, Pickering emulsions provide outstanding stability against Ostwald ripening and coalescence (Kargar et al., 2012; Timgren et al., 2011). However, multiple factors will influence the overall emulsion stability. These stability-influencing properties are related to the solid particles themselves (particle wettability which is mentioned above, particle size, particle concentration and particle shape), to the oil phase (type of oil) and to the water phase (pH and electrolyte concentration) (Aveyard, Binks, & Clint, 2003; Binks, 2002).

As mentioned above, particle size is an important factor in determining the emulsion stability. The size of particles that can be used to stabilize emulsions ranges from nanometers to several micrometers. For successful stabilization, the particles should be several orders of size smaller than the emulsified droplets for the particles to be properly located around the droplets. The size of emulsion droplets is determined by the amount of particles relative to the dispersed phase. Generally, droplet size decreases with

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