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From theoretical aspects to practical food Pickering emulsions: Formation, stabilization, and complexities linked to the use of colloidal food particles

Karin Schroën [a,*], Xuefeng Shen [b], Fathinah Islami Hasyyati [a], Siddharth Deshpande [b], Jasper van der Gucht [b]

[a] *Wageningen University, Department of Agrotechnology & Food Sciences, Laboratory of Food Process Engineering, Bornse Weilanden 9, 6708 WG Wageningen, the Netherlands.*

[b] *Wageningen University, Department of Agrotechnology & Food Sciences, Laboratory of Physical Chemistry and Soft Matter, Stippeneng 4, 6708 WE Wageningen, the Netherlands*

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ABSTRACT

We noticed that in literature, the term **Pickering emulsion** (PE) is used as soon as ingredients contain particles, and in this review, we ask ourselves if that is done rightfully so. The basic behavior taking place in particle-stabilized emulsions leads to the conclusion that the **desorption energy** of particles is generally high making particles highly suited to physically stabilize emulsions. Exceptions are particles with extreme contact angles or systems with very low interfacial tension.

Particles used in food and biobased applications are soft, can **deform** when adsorbed, and most probably have molecules extending into both phases thus increasing desorption energy. Besides, **surface-active components** will be present either in the ingredients or generated by the emulsification process used, which will reduce the energy of desorption, either by reduced interfacial tension, or changes in the contact angle. In this paper, we describe the relative relevance of these aspects, and how to distinguish them in practice.

Practical food emulsions may derive part of their stability from the presence of particles, but most likely have **mixed interfaces**, and are thus not PEs. Especially when small particles are used to stabilize (sub)micrometer droplets, emulsions may become unstable upon receiving a heat treatment. Stability can be enhanced by connecting the particles or creating network that spans the product, albeit this goes beyond classical Pickering stabilization. Through the architecture of PEs, **special functionalities** can be created, such as reduction of lipid oxidation, and controlled release features.

1. Introduction

Conventional emulsifiers have dominated emulsion science due to their ease of use, and relatively low cost [1]. However, their future industrial use is under threat, because of changes in legislation and consumer preferences. Emulsions stabilized with e.g., low molecular weight surfactants can destabilize upon temperature changes as occur during production and shelf-life, while so-called Pickering particles have the potential to mitigate these issues (see theoretical aspects). As pointed out in our review, when working with practical food emulsion formulations, the situation may be less ideal than the theory predicts.

The current avalanche of emulsion papers that claim to revolve around Pickering stabilization is impressive [2]. The interested reader is referred to recent papers focusing on the particles used: inorganic particles [3-6], and of biological origin [7-18], as well as purpose prepared particles [12,19-22] and effects related to particle shape and morphology [20,23-25]. Fundamental aspects are reported in [23,24,26,27], including their stability and rheology [28-30] and combined effects of particles and surfactants [31]. Also specific application areas were reviewed such as: foods [13-15,32-36], pharma [37-40], chemistry [41], water purification [42] biobased films [43], controlled release [44-49], high internal phase emulsions [28,50]), gelled PEs [51-53], water-in-oil emulsions [54], and multiple emulsions [34,41].

Despite the information available, PEs are not necessarily termed as such based on solid arguments. The question that we are trying to answer here, is if these emulsions are truly stabilized by particles, or, whether other effects play a role (e.g., the presence of surface-active components and their dynamic behavior during emulsion formation

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* Corresponding author.
E-mail address: karin.schroen@wur.nl (K. Schroën).

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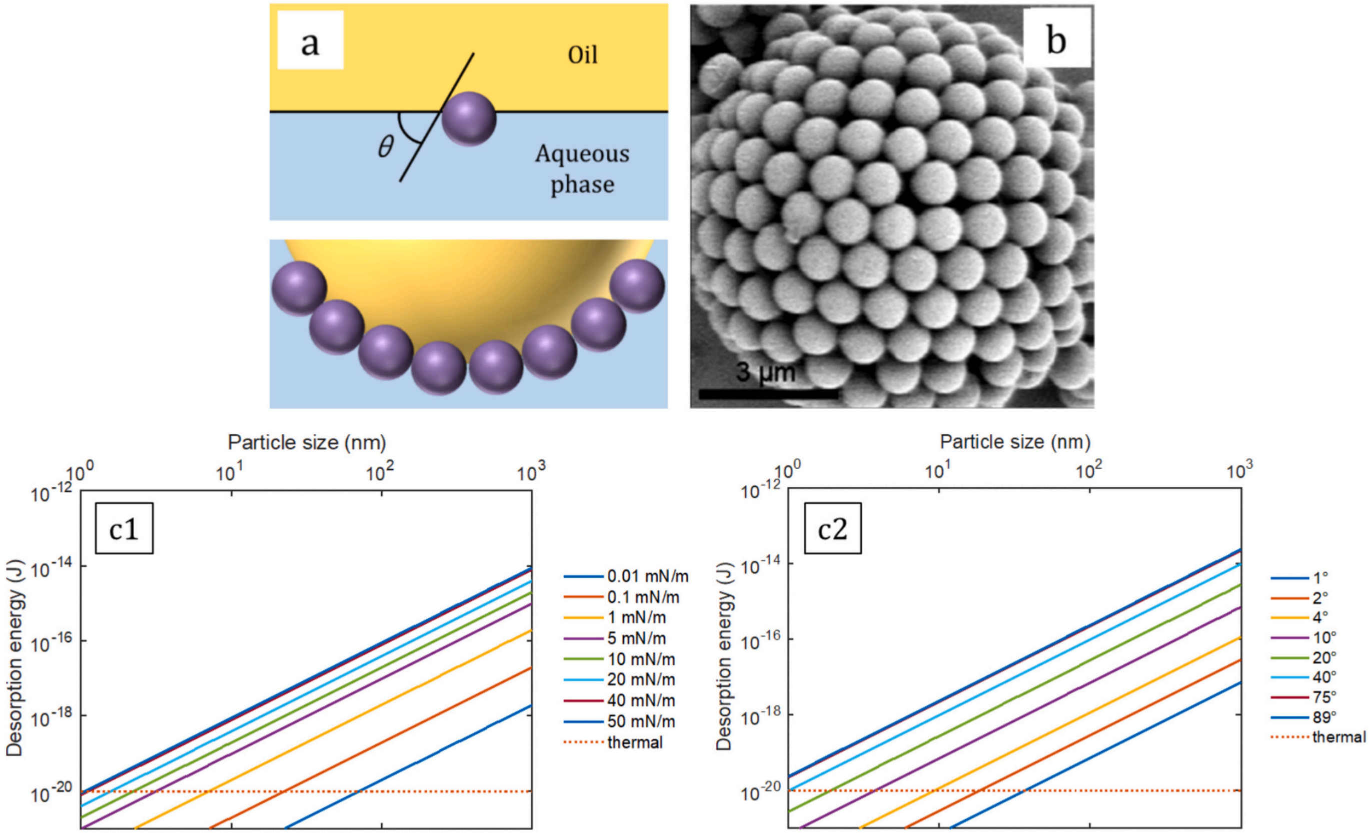

Fig. 1. (a) Position of a small spherical particle at a planar oil-water interface for a contact angle less than 90°, and corresponding positioning of particles at a curved interface of an O/W emulsion, figure adapted from [68], with permission. (b) Polystyrene particles assembled on an oil droplet in water originally from [71]. Reprinted from [32] with permission. (c) Desorption energy as function of particle size for different (c1) interfacial tension at 30° contact angle, and (c2) different contact angle at interfacial tension 30 mN/m. The dotted line represents the thermal energy at 25 °C (on the order of k_BT).

and storage) [55,56]. Besides, food particles may be deformable [57], and capable of forming interfacial or bulk networks [52,58-62] that render additional stability at the droplet level, or at the level of the product, respectively [13,22,53,63,64]. A gallant effort using deep learning techniques to make sense of this was recently published [65].

In the current review, we first describe basic Pickering stabilization, and under which conditions stable emulsions can be obtained. Next, we consider surface-active components and distinguish effects that occur at the liquid/liquid interface (interface adsorption), and solid/liquid interface (adsorption at the particle), which are aspects that are generally disregarded in reviews, especially within the food realm. In food applications, particles are expected to be deformable, and capable of adjusting their contact angle by partial (de-)swelling, or partly merging with other particles thus forming an interconnected interfacial layer, and these effects are described next. We translate these findings to emulsion formulations as would be used in practice and take the production conditions into account. Depending on the field of application, hardly any other components may be present (e.g., pharma formulations), or very many (e.g., food, personal care applications), and this will affect the opportunities to make stable emulsions greatly, as would be their further application, e.g., in dry form (see outlook section) [50].

2. Theoretical aspects of pickering emulsions

2.1. *Classical pickering stabilization*

For effective emulsion stabilization, the particles need to be partly wetted by both oil and water as shown in the pioneering work of Ramsden [66] and Pickering [67]. For oil in water (O/W) emulsions, particles that are preferentially wetted by water are used, and for W/O emulsions particles are preferentially wetted by oil. After being brought into the interface the particles assume a contact angle θ relative to the two liquid phases (see Fig. 1), and acquire a high desorption energy (ΔE, J) (Eq. (1)) [68,69,70]:

$$\Delta E = \pi r_p^2 \sigma (1 - |cos\theta|)^2 \tag{1}$$

Where r_p the particle radius (m) and σ the interfacial tension between oil and water (N/m). The same equation can also be used for particle-stabilized foams [12]. It is good to mention that the desorption energy depends on the shape of the particles, with anisotropic particles having much greater desorption energy e.g., [23,24], and thus higher potential for stable emulsion preparation. For now, we keep our discussion limited to spherical particles, and in the outlook section we elaborate on particle morphology.

In Fig. 1c, we systematically varied the particle size and investigated the effect of (a), interfacial tension, and (b), contact angle on desorption energy. The dotted line corresponds to a typical thermal energy of particles at room temperature. If this energy is higher than the desorption energy, particles may be released from the interface, as is the case for very small particles, and particles with low contact angles. The interfacial tension of the liquid interface only becomes relevant at low values.

Particles need to overcome an energy barrier that is of the same order of magnitude as the desorption energy, before being able to nest at the interface [62,72]. Once this barrier is overcome, the particles will remain practically irreversibly attached to the interface, since the desorption energy greatly exceeds the thermal energy of the particles at room temperature (this may become relevant during heat treatment, which may also contribute to additional stability if e.g., gelation takes place [52]). Conventional emulsifiers diffuse and spontaneously adsorb

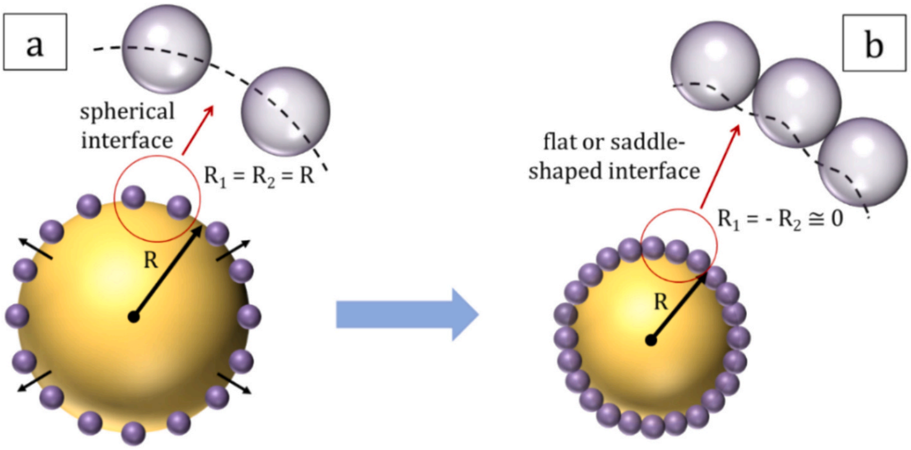

Fig. 2. Schematic presentation of the Ostwald ripening arrest by solid particles: (a) droplet surface not completely covered by adsorbed particles (b) completely covered with a dense adsorption layer of particles. Figure adapted from [84], with permission.

to interfaces, and to some extent desorb, leading to emulsion instabilities over time.

The interfacial layer formed by particles has a thickness generally similar to their size and is much thicker than emulsifier layers. When assuming that all particles used are in the interface, the amount of particles needed to stabilize droplets of size d_d can be calculated using Eq. (2) [27]:

$$d_d = \frac{4C\,\rho_p\,d_p V_d}{m_p} \tag{2}$$

Where C is the degree of coverage, ρ_p the particle density (kg/m^3), d_p the particle diameter (m), V_d the volume of dispersed phase (m^3), and m_p the mass of particles (kg).

Since particles add to the overall droplet density, theoretically, this can be used to density-match both phases, en create emulsions free of gravitational separation (naturally buoyant droplets). This is only possible for monodisperse droplets (e.g., made by microfluidics [73-75]), and may be less relevant for classical emulsification technologies, which lead to a wide range of droplet sizes [1,76,77].

The sections above relate to particles, and in practice other components would be present, as discussed in the section on the effect of surface-active components.

2.2. *Destabilization of pickering emulsions*

2.2.1. *Ostwald ripening*

The physical destabilization of emulsions may proceed through two distinct mechanisms, known as Ostwald ripening (Fig. 2) and coalescence (see next section) [78]. Ostwald ripening occurs because of Laplace pressure differences between droplets of different size [79]. The interfacial tension at the microscopic level in particle-stabilized interfaces is relatively high in the absence of amphiphilic stabilizers, and even for a perfect hexagonal particle packing, approximately 10 % of the interface area still experiences direct contact between the two fluids, [80], thus allowing Ostwald ripening to take place [81]. Small droplets possess higher Laplace pressure than larger droplets and tend to dissolve faster. The rate at which this happens depends on the solubility in the continuous phase [82,83].

The Ostwald ripening process halts when the interface between neighboring particles adopts a saddle-shaped configuration, with curvature radii $R_1 = -R_2$ (as illustrated in Fig. 2) becoming equal in magnitude but opposite in sign. Consequently, Ostwald ripening is 'arrested' in such emulsions because the Laplace pressure in the droplets is nearly zero ($P_{LP} = \sigma/(1/R_1 + 1/R_2) \approx 0$) [80,84]. When the ratio of particle size to droplet size exceeds 1:10, specific faceted shapes can also reduce the mean curvature to zero [85]. Complete particle coverage is not needed for stability; instead, a percolated network of particles with sufficient yield stress is adequate [86]. Please note that the presence of the particles does not prevent component diffusion; rather, it reduces the available space for diffusion [87]. In literature, this is often interpreted as a barrier effect, while it would be better to interpret this as part of the interface being fully permeable, while another part is not/hardly. Whether soft particles can prevent Ostwald ripening is still under debate [88,89], see also the application section.

Depending on the solubility of the droplet constituents in the continuous phase Ostwald ripening will be a faster or slower process [83,90] that may not even be relevant relative to the lifetime of an emulsion when made with vegetable oil [90,91]. Ostwald ripening has been reported to be suppressed or completely prevented in particle-stabilized emulsions or foams (for which Ostwald ripening is more relevant given the high diffusivity of gas) [92], even over years [93]. Although the droplet size may change because of ripening effects, it eventually reaches a stable state [88,91,94-98], due to the formation of a dense particle layer on the interface. In the presence of surface-active components, Ostwald ripening will be reduced because of Laplace pressure reduction, but particle displacement from the interface may occur inducing instability (see application section).

2.2.2. *Droplet coalescence*

Droplet coalescence spans many length and time scales [99,100], and that is affected e.g., by temperature. Work on the bulk level [101] and on smaller length scales using microfluidic techniques [102-104] also at elevated temperatures [105], and enhanced gravity [106,107] has been reported. For surfactant-stabilized emulsions, coalescence three stages have been identified: (i) droplet approach and formation of liquid bridges between interfaces, (ii) growth of the liquid bridges, and (iii) droplet reshaping during merging. The coalescence of particle-coated droplets remains poorly understood due to the particle shell that introduces fundamentally different mechanisms (limited and arrested coalescence [49,108,109]).

Limited coalescence was first described in the 1920s [110], and in the 1950s Wiley gave a theoretical explanation [111]. Limited coalescence occurs when insufficient particles are present to prevent coalescence, thus increasing particle coverage and eventually halting droplet coalescence. This theory has been applied [111-117] and extended [118-120] to predict the droplet size of PEs at equilibrium.

Arrested (a.k.a. partial) coalescence, happens when merging droplets are incapable of forming a spherical droplet. Two opposing forces are at work: the Laplace driving force, which aims to minimize the total interfacial area, and an elastic resistance that opposes droplet deformation. The elastic resistance can arise from an internal network or interfacial particle jamming. Most studies related arrested coalescence to dispersed phases that contain solids, such as fat crystals, which induce a viscoelastic response [108], but also particle jamming at the fluid/fluid interface in soft materials has been reported to provide mechanical resistance against the relaxation of bubbles or droplets into a spherical shape [24,121-125].

2.3. Effect of surface-active components on interaction energy

We discuss the individual effects of interfacial tension and contact angle on desorption energy, fully knowing that interfacial tension (γ_{ow}) and contact angle (θ) cannot be varied independently. The interfacial tension is one of the components that determine the contact angle, together with the interfacial energies of the particle (s) with both liquids γ_{sw} and γ_{so} as defined by Young's law: $\cos\theta = (\gamma_{sw}\text{-}\gamma_{so})/\gamma_{ow}$. We describe combined effects in the practical application section.

2.3.1. Interfacial tension

For interfacial tensions as they occur in (food) practice (5-40 mN/m), the desorption energy of particles exceeds their thermal energy, unless they are very small (<10 nm), or have extreme contact angles (close to 0 or 180°). The hard particles used in literature are generally not that small, but soft particles of this size have been suggested, including proteins. For these particles, other effects play a role as described in 'Special Effects of Particles at the Interface'. At very low interfacial tension (0.01-1 mN/m), as would occur in micro-emulsions (not to confuse with micrometer-sized droplets) or in aqueous two-phase systems, the energy of desorption is low, and particle stabilization would not be expected to be effective. When exposed to high temperatures, e.g., 120 °C the thermal energy would be ~35 % higher than shown in Fig. 1c, thus narrowing the gap between both energies, possibly leading to one exceeding the other.

In general, it is assumed that the interfacial tension of the interface is not affected by the presence of particles (e.g., inorganic silica nanoparticles [126]) unless present at high concentrations and given sufficient time to adsorb [127]. Still, in many papers dealing with PEs (we refrain from using references), effects on interfacial tension are reported under diffusion-controlled conditions e.g., occur during automated droplet tensiometry. Since particles would need to be brought into the interface using a force that greatly exceeds any force generated by diffusion, these effects can safely be ascribed to the presence of surface-active components that are able to diffuse. In emulsions produced with these ingredients, most probably a dual stabilization process is at work, leading to mixed interfaces, which is relevant when working with less defined ingredients [128,129] as elaborated on in the application section. Alternatively, when the particles are added to a pendant droplet, the density difference of the particles with the oil will lead to a change in interface shape, which has been misinterpreted as an interfacial tension effect as demonstrated using (non-surface-active) glass beads and whey protein [130]. Monitoring the Bond number as is standardly available in droplet volume tensiometers would prevent this type of error.

2.3.2. Contact angle

From Fig. 1 c2, it is clear that particles with a contact angle of 90° have the highest desorption energy, and are preferred from a stability point of view [131], and the geometry plays a relevant role in this as well e.g., [23,24,25], as discussed in greater detail in the outlook section. However, measuring the contact angles of particles is difficult due to their size. Often, a small tablet is made, on which the contact angle is measured (e.g., [132]), and this may give an impression of the contact angle, but the roughness of the tablet (and possibly porosity as well) will influence the measured value leading to misinterpretations [133]. In the practical application section, a number of methods are discussed.

The contact angle is influenced by component adsorption, which may (considerably) influence the energy of desorption of the particle. As is immediately clear from Fig. 1 c2, and Eq. (1), any contact angle close to 0 (or 180) degrees leads to the thermal energy exceeding that of desorption, and thus emulsion destabilization. Surface modification with octenyl succinic anhydride (OSA) is popular given its GRAS status [134-134], and as summarized by Olawoye et al. [138] but cannot prevent adsorption induced wettability changes.

The question that would need to be answered is to which extent contact angles are affected by surface-active components, or swelling/deswelling of either the particle or any deposited layer [22,139,140]. Besides, from Young's law ($\cos\theta = (\gamma_{sw}\text{-}\ \gamma_{so})/\gamma_{ow}$), it is also clear that a change in interfacial tension will influence the contact angle, and therefore influence the desorption energy, even without adsorption to the particle. In the section dedicated to the application of PEs in food, we will try to shed further light on this.

2.4. Special effects of particles at the interface

2.4.1. Interactions

When particles are present at the interface, they show interactions amongst each other (this section), and deformation (next section). In the application section, we discuss (interconnected) layers at the interface, and network formation in the bulk. Upon particle adsorption, three types of inter-particle interactions may arise [141]. These may be direct interactions between particles, such as van der Waals, electrostatic, and hydrophobic interactions, that depend on particle characteristics [142]. The second type relates to the confining influence exerted by the interface (e.g., capillary interactions) [143]. The third interaction is induced by external fields [144,145], which is very powerful for tuning colloidal assembly at fluid interfaces [144-147], but not that relevant for application in food.

For particles adsorbed at a fluid/fluid interface, the van der Waals forces can be expressed by an effective Hamaker constant: $A_{eff} = A_{p1} + f^2(3\text{-}2f)(A_{p1}\text{-}A_{p2})$, with A_{P1} and A_{P2} the particle Hamaker constants in fluid 1 and 2, respectively, and the depth of submerging in fluid 2 ($f = (1 - \cos\theta)/2$ with θ the contact angle) [148]. Typical values are 0.1-1 k_BT, which indicates that these interparticle interactions are small [149]. Charged particles undergo mutual electrostatic interactions, and at the oil/water interface dipoles are generated due to surface charge density differences on the oil and water sides of the particle, possibly leading to dipole-driven repulsion [150]. At high surface charge (ξ-potential > ±30 mV), mutual repulsion may reduce the maximum adsorbed amount [151,152], at low surface charge, (< 30 mV) particles may aggregate and form a network in the continuous phase [64]. Hydrophobic interactions reduce unfavorable interactions between hydrophobic surfaces and water [153]. When hydrophobic patches are present, particles could manifest end-to-end attraction (e.g., Janus-particles) [154]. Adsorbing polymers to the particle is a common way to prevent aggregation caused by the mentioned attractive forces, because they exert a steric repulsion [155], which is also relevant when using complex ingredients. If adsorption is not sufficiently permanent, this may lead to the reduction of interfacial tension or bridging of particles (both at the interface and in the bulk).

The Bond number: $Bo = \Delta\rho g r^2/\gamma > 1$, with g the gravitational constant, $\Delta\rho$ the density difference, and γ the interfacial tension can be used to determine if particles with radius r may deform the interface leading to capillary forces. For the small particles used in food, this criterion is generally not met, although surface roughness, chemical heterogeneity [156,133,72,157,158,159], anisotropic shape [160,161,162,163,164,165,166] and other factors may cause pinned contact lines, and induce interface deformation at low Bond number, which is expected to be relevant for food emulsions. Depending on the curvature of fluid/fluid interfaces within an emulsion, particles move along gradient lines, adhering to the principle of minimizing energy to form structures [167,168,146,169,143]. Both in theory and practice [170,171] deformation of an anisotropically curved interface was shown to result in anisotropic capillary interactions amongst isotropic particles (e.g., cubic arrangements of spherical particles) [172]. Particles with different

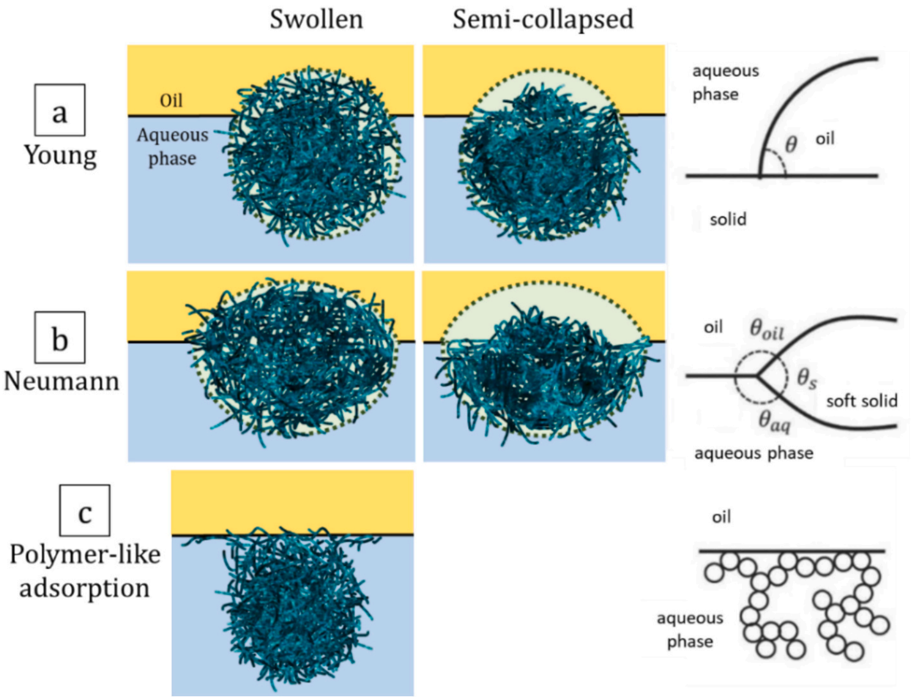

Fig. 3. Schematics of adsorption scenarios for an isolated microgel particle. Young (a) and Neumann (b). The left-side images correspond to a swollen state in both aqueous and oil phases whereas the right-side images show a collapsed state in the oil phase and a swollen state in the water phase. The light green is the microgel size in the swollen state. An alternative to these scenarios is a polymer-like adsorption (c). Figure adapted from [57], with permission. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

shapes (disks, spheres, and cylinders), experience capillary forces and migrate along the curvature gradient to sites of high deviatoric curvature [160,173,143,166]. Capillary forces between anisotropic particles are millions of times greater than the thermal energy ($\sim k_BT$) which promotes particle aggregation at the interface, thus increasing filling efficiency and fortifying the emulsion stability [64]. Further, colloidal particles have been shown to nucleate into crystalline patches on a liquid droplet surface [173] rather than forming large crystals [174,175,176,177], as this morphology minimizes the elastic energy induced by the curvature [174,175,176,177].

2.4.2. Deformation

Flexible particles, e.g., microgels, can adsorb and partly (de)swell at liquid interfaces [178,179,180]. The adsorption energy of core-shell gel particles was found similar to that of solid particles of the same size ($\sim 10^6\ k_BT$). Interestingly, these particles adsorb spontaneously, so similar to polymers (and food components), and rather different from solid particles (as discussed in the process section) [57]. Because of the high desorption energy, the particles form dense packings [57], and are significantly compressed compared to the bulk solution.

The combined effect of high desorption energy and the absence of an energy barrier to adsorption is most probably the result of dangling polymeric chains present at the surface of the particles, in combination with solvent quality effects that determine the level of collapse of the polymers inside the particle, which may be different for both phases as illustrated in Fig. 3 and modeled by [181]. This affects the contact angle as it occurs in these systems which can be different for the two phases (right side of Fig. 3, and thus the energy of desorption. It was found that softer particles exhibited greater interfacial activity than harder particles [182,183,184], with the softer particles assembled and stretched more readily, thus increasing their adsorption energy. The interfacial tension co-determines the shape of a flexible particle at the interface [179]. A larger interfacial area can be covered when the interfacial tension (γ_{ow}) is high; if the surface free energies of the particle with the liquids (γ_{op}, γ_{wp}) are large, the particle is close to a sphere. The fact that soft particles are able to adsorb at much lower energy input compared to solid particles makes them relevant for practical application (see respective section) [185,186,187,188] that is as long as these food-grade particles are able to withstand the emulsification process (see next section).

3. Process considerations

To make stable PEs, small droplets need to be timely stabilized by particles carried to the interface through the shear force applied. We briefly describe various process conditions used during emulsification (see Appendix A for an extensive overview [189,190], share relevant time scales, and consider particle integrity during emulsification.

3.1. Droplet formation

Considerable shear is used to make emulsions, and depending on the device this will be through laminar (lab- and small pilot-scale emulsification devices), or turbulent flow (full pilot-scale and really large-scale emulsification homogenizers), which makes comparison far from trivial [191]. Droplet breakup in laminar conditions can take place through plain shear flow or extensional flow. In both cases, a critical Weber number (We) can be defined for droplet breakup.

$$\mathrm{We} = \frac{\eta_c \dot{\gamma} R_d}{2\sigma} \tag{3}$$

Here, $\dot{\gamma}$ is the shear rate applied (1/s), η_c the viscosity of the continuous phase (Pa·s), R_d the droplet radius (m), and σ interfacial tension (N/m). For both situations, so-called Grace curves (relationship between the viscosity ratio and the critical capillary number for droplet break-up) are derived [192,193]. Under turbulent conditions, the flow is much more chaotic in nature and is characterized through typical eddy lengths [194,193,191]. Very recently, some of these processes were covered through a modelling approach [195]. Table A1 and A2 in the appendix give a concise overview of the emulsification methods,

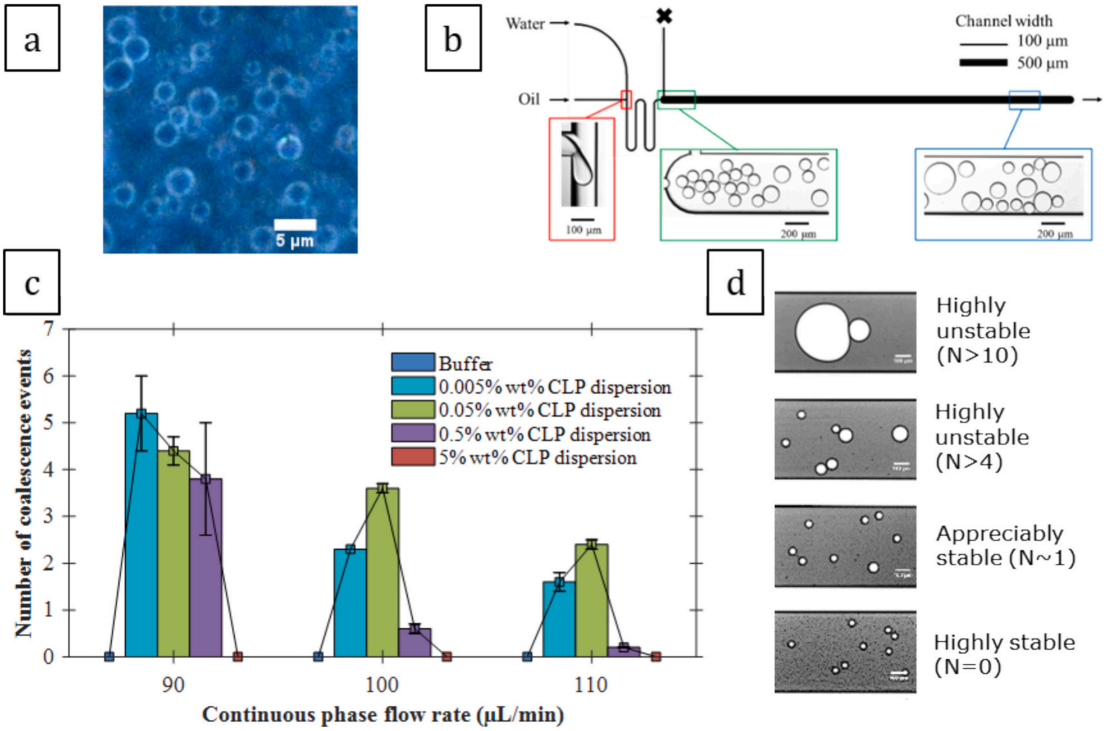

Fig. 4. (a) A polarized light image of a 1 wt% solid lipid particle, and 10 wt% sunflower oil emulsion produced by high-pressure homogenization (400 bar, 5 passes). (b) Microfluidic coalescence cell, droplet formation takes place at the T-junction (red rectangle), droplets are investigated along the coalescence channel (green rectangle to the blue rectangle). (c) Number of coalescence events as a function of continuous phase flow rate (μl/min) for sunflower oil-in-water emulsions formed with various concentrations of sodium caseinate-coated solid lipid particles in the continuous phase. The dispersed phase flow rate was 1 μl/min. No coalescence was observed for the 5 wt% dispersion. (d) selection of snapshots of the coalescence chamber. All images are reproduced with permission from [62]. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

including typical time scales.

3.2. *Force needed to bring particles to the interface*

As mentioned earlier, the size, geometry, and contact angle (including pinning) influence the desorption energy of particles, and thus the stability of emulsions. Besides that, interfacial tension (in combination with the deformability of particles) plays a role in desorption. These considerations hold for particles that are nested in the interface, but in order for *solid* particles to adhere to the interface they need to be brought there through a shear force big enough to exceed the energy barrier described in Eq. (4). As mentioned, flexible particles behave more like molecules during adsorption through dangling chains.

It was investigated how much force was needed to push particles (sodium caseinate-covered solid lipid particles), into an interface [62] using a microfluidic coalescence cell (see Fig. 4). Given its relevance for Pickering stabilization, we discuss this work in detail. The coalescence cell consists of a droplet formation part (T-junction; red rectangle) of which the continuous phase flow rate is changed to vary the force with which particles are brought to the droplet interface. Droplet interaction takes place in the coalescence channel that starts at the green rectangle and continues to the blue rectangle. At different locations, the droplet size is registered and compared to the droplet size at the time of formation, and from this, the average number of coalescence events (*N* in Fig. 4) is calculated. The snapshots shown in the right bottom part of Fig. 4 show the highly diverse outcome.

Sunflower oil droplets made in the absence of particles did not coalesce, unlike droplets formed in the presence of lipid nanoparticles that showed a coalescence dependency on the continuous phase flow rate. Coalescence was reduced to zero at high particle concentrations and continuous phase flow rates >90 μl/min (Fig. 4). At lower flow rates, particles induce coalescence and have a *de*stabilizing effect, caused by droplet bridging, with coalescence expected to continue until 'fully' covered droplets are obtained, as demonstrated for surfactant-stabilized droplets and bubbles [196–198,56]. Because of that, droplets may no longer be spherical [24,199,200,61]. To be complete, depletion attraction was excluded as a cause of coalescence [201].

The energy barrier (E_a) that needs to be overcome for particles to reach the interface has been described by an Eyring-type equation derived from the transition state theory of Kramers [202]:

$$k_{ad} = \omega_0 \cdot e^{\frac{-E_a}{RT}} \tag{4}$$

in which k_{ad} is the adsorption rate, and ω the attempt frequency that is related to particle diffusion. Under flow, the drag exerted on the particle can be estimated using Stokes law ($F = 6\pi\eta r_p v$, with η the fluid viscosity, r_p the particle radius, and v the fluid flow velocity relative to the particle [203,201]. These hydrodynamic forces affect the adsorption rate as shown in:

$$k_{ad} = \omega_0 \cdot e^{\frac{(-E_a+F\delta)}{RT}} \tag{5}$$

with δ the thickness of the interfacial region that needs to break for particle adsorption to take place. The hydrodynamic forces lower the energy barrier, leading to particle adsorption rate exponentially increasing with continuous phase flow velocity, ultimately preventing coalescence, as shown in the snapshots on the right, and the polarized light image of an emulsion prepared by high-pressure homogenization (Fig. 4).

The fact that emulsions can be destabilized by particle bridging, has been noted in the literature [204], while it has also been reported that bridging does not necessarily lead to instability [58]. For the emulsions reported in this work, it was found that when sufficiently covered, the bridged droplets could be disconnected by gentle shaking.

Table 1
Contact angle measurement techniques.

Technique	Principle	Pros	Cons	Reference
1. Sessile drop (including advancing and receding values)	Placing droplet on target surface	Simple Available	Sensitive to roughness and porosity leading to big differences in advancing and receiving angle Target surface preparation may affect properties	[251]
2. Tilted Plate	Droplet shape at various tilting angles	As simple as sessile drop	More sensitive to interpretation Target surface preparation may affect properties	[250]
3. Wilhelmy Plate	Force experienced by the Wilhelmy plate	Available	Target surface preparation may affect properties Sensitive to interpretation	[251]
4. Capillary rise	Height difference of liquid inside and outside capillary	As available as Wilhelmy plate Easy to interpret	Surface of interest needs to be deposited in a capillary which may affect properties	[252,253]
5. Sticking bubble	Air bubbles sticking to a target surface	Analysis in liquid Easy to interpret Insensitive to roughness	Target surface preparation may affect properties Elaborate method	[254]
6. Peel-off imprinting	Polymerization of the interfacial layer, analysis through AFM	Good impression of the contact angle in a liquid-like state	Elaborate method Limited observations	[131,255]
7. Super-resolution microscopy	Direct observation of the contact angle	Precise values for liquid/liquid High resolution Versatility Easy sample preparation	Highly specialized equipment needed Limited observations	[256,257]

3.3. *Droplet formation versus particle deposition*

3.3.1. *Time scale considerations*

Because of continued droplet break-up and coalescence during emulsification [190,83], the force applied to make small droplets exceeds the interfacial Gibbs free energy by a factor of 20 to 100, and even more due to heat dissipation [205,206,207,208,209,210]. It has been suggested that under turbulent conditions, components of which the size is similar to that of eddies would be transported fastest [193,191], and this could be beneficial for particles. In the work of Walstra, it is suggested that the higher propensity of casein at the interface relative to the much smaller whey proteins that in essence have much higher diffusivity is caused by this. For more information on the characteristics of emulsification processes, and equations see the appendix, Table A1 and A2.

3.3.2. *Particle integrity*

The massive shear forces exerted during homogenization will impact the integrity of particles, especially when these are soft particles, or possibly aggregates, as would occur in food ingredients. For bacterial cells [211], starch [6], and protein particles [212] this has been reviewed. In general, shear will not be able to destroy the molecular protein structure when dispersed in a liquid unless shear rates exceed 10^5 s^{-1} but the situation is expected to be quite different for proteins that are present at an interface e.g., [213] where they may unfold e.g., [214,215,216], and thus become susceptible to shear. For starch granules various effects have been described in relation to their structure, retrogradation, and digestive functionality [217,218,139,219 ,220,221,6], while for protein gels this is still rather limited [222,25]. It can be expected that insights on microbial cells could be used as guidelines for the disruption of flexible particles in general [211]. Last but not least, particle aggregation may be playing an important role that prevents uniform distribution of the particles across the emulsion [135,136]. In literature, suggestions were made to evaluate if a process can break up aggregates [223,224,225,226]. Especially when used as powders, rewetting of solid particles and their interaction strength (hundreds of kJ/g) can be an issue [223].

4. Interface considerations

From the previous sections, it is clear that various effects play a role in PE production. One would need to weigh the exerted shear force that leads to droplet formation within typical time scales for particle adhesion to the interface (see process considerations section). Furthermore, in the formulation of food products, not only particles will be present, but also molecular components that are expected to influence the interfacial tension, rheology, and contact angle, thanks to a plethora of effects that all influence the composition of the interface that we consider a determining factor in emulsion stability together with effects that occur in the bulk phase [128,129,227]. We begin by describing the techniques to measure interfacial effects and describe their usefulness in PE design.

4.1. *Interfacial tension and rheology measurement*

The classical interfacial tension measurement techniques are based on diffusion leading to the reduction of the interfacial tension (please see the following reviews [228,229]. These methods have shown their value, and allowed the identification of various regimes e.g., [230,228]. However, it is questionable if these methods are suited to shed light on processes happening at time scales relevant to large scale emulsification. For that, microfluidic devices may be better suited [82,231,232,233,234,235,236,56] since they allow capturing effects occurring within a sub-millisecond time range.

If particles are relatively large, as would be the case in many PEs, diffusion is slow, and the diffusive force of particles to break into the surface is not sufficient to allow them to nest there, making diffusion-based interfacial tension measurement not that relevant. When dealing with inert particles that can diffuse to the interface, no effect on interfacial tension is expected [24]. Still, in many papers, one can find a reported effect of ‘particles’ on interfacial tension. Most probably that is the result of either particle preparation methods leaving unreacted molecular surface-active species behind that even after repeated washing cannot be removed [187,237], or particle disruption by emulsification [60], or the weight of particles disturbing the shape of the droplet as discussed later.

Assuming that particles can be brought into an interface, interfacial rheology data can be gathered using the volume expansion/reduction cycle-mode of a droplet volume tensiometer that allows the identification of linear and non-linear behavior by using so-called Lissajous plots [238,239,240,241]. The elasticity of the interface is directly connected to its ability to prevent coalescence e.g., [242], and can amongst others be used to investigate mixed layers [243,244,245,20], including their interfacial displacement as recently reviewed by Hinderink [246]. For particles both at the air/water as well as oil/water interface, information has been gathered using step deformation, and the results were compared with e.g., protein-stabilized interfaces [213]. Dynamic heterogeneity was reported for many interfaces with stress relaxation following a stretched exponential decay (values of 0.4–0.6 for extension and 0.6–1.0 for compression as described by a Kohlrausch-Williams-Watts function), with the highest values corresponding to interfaces that contain particles. This indicates (slow) transfer between interface and bulk during deformation, which was confirmed through molecular dynamics simulations [213]. A microfluidic method [238,247] to investigate interfacial rheology through analysis of droplet deformation under flow while passing through a constriction [248] showed that the onset of film formation for proteins is within seconds, and much faster than previously assumed and analyzed [249]; for particles, this still needs to be tested.

4.2. *Contact angle measurement and impression thereof*

There are methods available to measure contact angles, but whether that leads to accurate values is a bit of a question, especially when seen in the light of particles that would be used in the production of food emulsions [14] that are typically non-spherical and often rough. We have put the methods in Table 1, and listed their pros and cons. For more information including protocols, please see [250,251].

Contact angle measurement is often carried out at the air/water interface, and only limitedly at the oil/water interface. For a number of methods (1-5), a target surface needs to be prepared [133,153], which is often done by compressing the particles into a tablet, or depositing them onto a carrier (film casting) and to which they may need to be attached. Typical food particles may include low molecular weight components that are released from a tablet, thus influencing the contact angle. Furthermore, methods 1-4 are sensitive to roughness, while this is not the case for method 5, that actually performs better on porous and rough surfaces. The fact that in methods 1-4, dry surfaces are used leads away from the completely wetted state that particles would have in food products. Polymerization has been used to create a peel-off imprint of the particles, which besides being time intensive is not always that straightforward to interpret [131]. Recently, an exciting new technique was proposed for particles observation at a liquid/liquid interface [257] using a super-resolution microscope in highly specialized labs. A more accessible method (albeit not trivial) is the so-called sticking bubble method that was developed for comparing the wettability of membranes (porous and rough surface) [254], or fibers [258], and shown to be instrumental in identifying surfactant-induced wettability changes in membranes [259]. Methods 6 and 7 are closest to observing the particles at the location of interest, with the super-resolution being closest and a great step forward [260]. The method is versatile and has been applied in liquid/liquid systems, including deformable particles [257], and particles with varying surface chemistry [261], and more in general, to get insights into the topology of interfaces [256] in various systems including emulsions and bubbles [260]. The fact that the method has high spatial resolution, low invasiveness, high sensitivity, simple sample preparation and so on, are all big pros, but getting it to work with the much less defined samples used for food emulsion preparation is the next challenge, especially in relation to emulating interfaces as created by emulsification devices.

4.3. *Particle, interface, and droplet impressions*

The question we ask ourselves is whether images, or more general characterization techniques, do justice to the situation as it would occur in food-related PEs and whether all aspects of the emulsions and particles also truly come to the foreground. Droplet size distributions are often measured through static and dynamic light scattering techniques, but this will not cover the full range of 'structures' present, be it particles, droplets, or even free molecules. Various fractions can be measured, but the question how to appropriately overlap the size distributions obtained, is not easy to answer [262]. This also implies that it is inherently difficult to follow the stability of PE emulsions, that is unless major instabilities such as massive creaming or oiling of occurs, but that would be far beyond the acceptability limit.

In the review of Low [109], various visualization methods for droplets, and particles have been summarized. Bright-field optical microscopy gives a quick impression of the emulsion, while phase contrast microscopy distinguishes particles and droplets, and that also holds for polarization microscopy if particles show crystallinity (see Fig. 4). For confocal laser scanning microscopy, either the inherent fluorescence of components present or that of the fluorophores attached for this purpose can be used to get an in-depth understanding of the localization of e.g., particles, including food particles [263]. The added fluorophore may change component behavior, and that needs to be taken into account consistently. Scanning and Transmission Electron Microscopy (TEM) is used to generate images of both particles and droplets e.g., [136,14], with the first technique requiring sputter coating of a dried sample which may affect the sample greatly, while TEM can be used in a (cryo-treated) liquid state to elucidate morphological differences [61]. Atomic force microscopy has been used not only to investigate droplets (and capsules) but also interfacial layers (Langmuir-Blodgett) e.g., of soft particles of which the behavior could be modeled [181].

5. Considerations for application of pickering emulsions

5.1. *General aspects*

To prevent gravitational separation, emulsion droplets need to be small, as follows immediately from Stokes' formula for the sedimentation velocity of freely dispersed droplets:

$$v = -\frac{2gr^2\,(\rho_2 - \rho_1)}{9\eta_1} \tag{6}$$

with v the sedimentation rate (m/s), g the (gravitational) acceleration (m/s^2), r the droplet radius (m), ρ_1 and ρ_2 the densities of the dispersed and continuous phase, respectively (kg/m^3), and η_1 the continuous phase viscosity (Pa·s). Depending on the effective density difference of the droplets, which is affected by adsorbed particles, droplets will either cream or sediment.

As shown in Appendix A, a high-pressure homogenizer generates droplets of typically 0.1 μm, which reduces the creaming rate greatly given its squared dependency (typically in the order of 1 mm per *year*). When particles are to be used to stabilize these droplets, they would typically need to be 1/10th of the droplet size, which makes them in the order of 10's of nanometers. These particles are most probably in the danger zone for being released from the interface (see Fig. A1 in the appendix that are revised versions for Fig. 1 c1 and Fig 2, taking 5 mN/m as interfacial tension as a realistic value in food emulsions, and an 'average' contact angle of 60°). From this figure, it is clear that for an interfacial tension of 5 mN/m, strict requirements for particle size and contact angle are needed. For non-spherical or rough particles, there may be additional effects [172,161,154] compared to the base-case described in Fig. A1, but in essence, the trends will not be that different. Even if the particles do not cover the whole interface [23,24], stable emulsions can still be achieved. It is thought that approximately 30 % uniform coverage would suffice.

The biggest difference with the scenario sketched earlier, would hold for soft particles that can swell/de-swell when present at the interface, thus changing their interaction energy. This would most probably lead to a low effective interfacial tension (at the lowest 2-5 mN/m) as would be the case for interface stabilized with proteins [264,57], and for which the scenario is given in Fig. A1.

The situation described earlier relates to a specific food product with a long shelf life, hence requiring small droplets. Alternatively, viscosity enhancement has been used to reduce sedimentation or creaming, for example by gelling or network formation [204,58,63,13,52,61,53]. The dispersibility of particles (or lack thereof) can be an indication of their propensity to form a network in the bulk phase e.g., [11,13,61]), or emulsions that obtain their physical stability through a network of particles and connected droplets [60]. Furthermore, components present at the interface can form a film [265,247,266,213], a process that can be speeded up by heat treatment [267] as well as result in

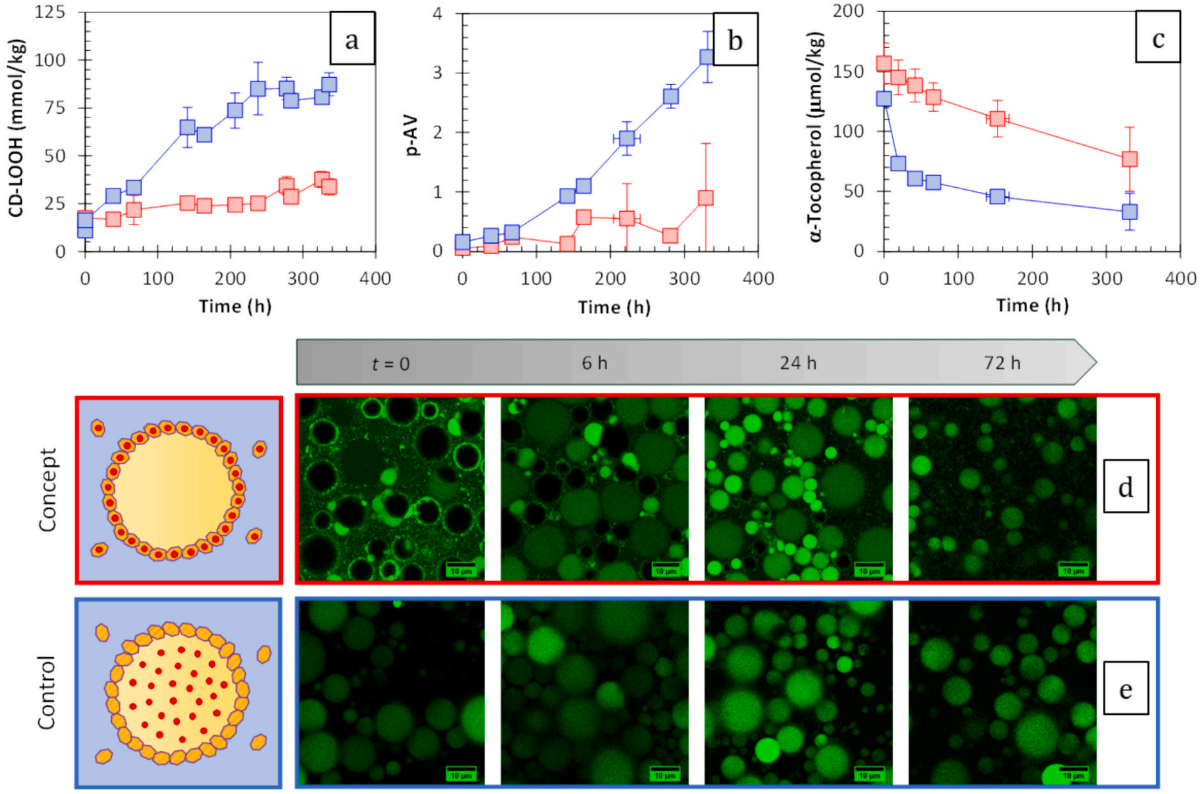

Fig. 5. Formation of (a) conjugated diene hydroperoxides (CD LOOH) and (b) aldehydes (pAV), and (c) stability of α-tocopherol during incubation of concept and control PEs (red circles and blue squares, respectively; see schematic illustrations at the bottom left) initially containing 200 µmol kg^{-1} α-tocopherol. CLSM images of the PEs with α-tocopherol initially added to the CLPs (d, concept PE) or the oil (e, control PE), taken over 0–72 h. The scale bar on all images is 10 µm. Adapted, with permission from Elsevier. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

displacement and subsequent destabilization [268,81,269,244,245]. Besides, stable emulsions have been reported through bridging mechanisms [270,271,58], for which particles need a strong continuous phase affinity, and cover a low amount of interface due to the extreme contact angle [204]. Many examples are available in the reviews mentioned in the introduction.

5.2. Choice of particles and emulsification process

5.2.1. Purpose-made versus existing particles

When deciding which particles to use for the production of PEs, two options are available: 1. starting from particles that are purpose-prepared e.g., [61] or 2. to work with particles as they happen to be available in many bio-based processes either without further treatment [128,129,8,13,14], or after receiving a surface treatment on purpose [134,135,136,137,138,272,22], or that occurs as part of the treatment given during component preparation [273], which may even lead to additional functionality (see added functionality section). Obviously, option 1, leads to better control over the particles and thus the emulsion, but option 2 is of greater practical relevance since, this would add value to streams that are currently termed by-products, or even waste [11,14,60], and thus contribute to sustainability.

Irrespective of the particles used, their structural integrity upon receiving a homogenization treatment needs to be investigated. This can be done by checking their size, which may require investigating various fractions, which is not trivial as described earlier, especially when particles of various sizes and geometries are present [274]. Alternatively, one could measure the interfacial tension of a particle dispersion after receiving a homogenization treatment. If only particles are present, the reduction in interfacial tension would be slow, or not occur, while if low molecular weight components are present because of disruption of the particles, interfacial tension would decrease much more and much faster. For soft particles, a similar reduction of interfacial tension has been reported as for molecules, albeit at a slower rate [127]. Furthermore, some papers discuss the exchange of particles, although this seems a minor effect, and if it occurs at all, it may be induced by the fact that particles are small in combination with wettability changes induced by molecular components present [275].

The insoluble part of food ingredients, as abundantly present in plant ingredients (protein, starch granules) can serve as 'particles' that stabilize emulsions, even at high oil volume fractions, albeit that oxidative stability needs to be taken into account [276] (see additional functionality section). These particles would need to remain insoluble upon receiving processing (e.g., emulsification or heat treatment), but if they do, they can be effective stabilizers [11,135,136,277].

5.2.2. Emulsification and other process requirements

In many cases, we feel that particle stabilization is part of the explanation for reported emulsion stabilities found, and that may be a small part due to the presence of molecular components that may have been part of the ingredient, or the result of loss of particle integrity. In essence, many of the food emulsions that are claimed to be PEs have mixed interfaces as investigated in depth by [186,187,188] who found that repeated rinsing of microgel dispersions still did not fully remove molecular components. Due to the focus on particles, other components in the interface are not that well documented, and it is often not realized that they are there given the focus on the visualization of particles in the interface as described in the section on particle, interface, and droplet impressions. Besides, continuous phase effects take place that can lead to enhanced stability due to increased viscosity of network formation [128,129,11,13,53]. For more information on mixed interfaces and the resulting effects, we refer to the review by McClements and Jafari [278].

To make a true PE, solid particles need to be pushed toward the interface, for soft particles this value is considerably lower which may imply that alternative emulsification processes become relevant. Especially microfluidic techniques that allow manipulation of the droplet formation process at a very small scale have been suggested for this [73,210,4], and that also includes direct and pre-mix membrane emulsification [279,207,280]. These techniques have been recommended for their low energy input compared to homogenization [281,282,283], and may thus be less detrimental to the particles. Because of the more defined, and in some cases even uniform droplet size, these emulsions are also more stable against Ostwald ripening. For microfluidics, obtaining submicron droplets, as well as upscaling are still challenges that need to be addressed [284,285,286,74,75].

To warrant food safety, products are heated, cooled, or frozen, which may cause instability. Besides thermal energy increase (Fig. A1), this may induce (interfacial) gelation [19,63], which has been investigated in depth for starches e.g., [217] and protein [287], including continuous phase, and digestion effects, as discussed in the respective section. Furthermore, crystallization and network formation as occurs in ice cream through partial coalescence of stabilizing crystals on air bubbles was reported [288,289].

5.3. *Additional functionality of pickering emulsions*

Besides physical stability, PEs also are known for the additional functionality they may have e.g., chemical stability (lipid oxidation), or digestive stability, and how they can serve as a basis for other products.

5.3.1. *Lipid oxidation mitigation*

Lipid oxidation in emulsions is initiated at the oil-water interface [290], and implies that putting antioxidant (containing) particles at the interface yields an opportunity to enhance physical and chemical stability [273]. This was proven for emulsions stabilized with solid lipid nanoparticles. Emulsions that were identical in composition, but in which the antioxidant α-tocopherol was either delivered through the oil droplets or was incorporated in solid lipid particles (Fig. 5; [200,61]) had different oxidative behavior. Fig. 5 shows that lipid oxidation products (both primary conjugated dienes (a), and secondary aldehydes (b)) were suppressed in emulsions with the antioxidant incorporated in the particles. The drop in α-tocopherol (c) is in line with this, as are the CLSM images (Fig. 5d and e) that show an exchange of α-tocopherol starting from the interface (d, $t = 0$) to the droplet, from which it is consumed away. When added to the continuous phase, the α-tocopherol-loaded particles had no effect on oxidation, clearly indicating that particle localization was key [200].

Other examples of antioxidant-loaded Pickering particles have been published, e.g., epigallocatechin gallate (a common plant-derived antioxidant) was incorporated into mesoporous silica nanospheres, which lowered oxidation of emulsified essential oil better compared to the standard emulsifier Tween 80 [291]. Hydrophobic zein and hydrophilic phenolics were used to create composite particles that rendered relatively high oxidative stability to emulsions compared to protein-stabilized control emulsions [292,293]. These examples demonstrate the viability of Pickering particles as interfacial antioxidant reservoirs, thus providing dual (physical and oxidative) stability to emulsions.

Particles added to the continuous phase can lead to antioxidant effects, albeit through another mechanism. Maillard reaction products have been found to affect lipid oxidation in emulsions due to binding of metal and radical scavenging [294,295,296]. Starch particles [273] were claimed to form thick interfacial layers with an ability to scavenge free radicals in conjunction with regular emulsifiers [272]. It is good to mention that the differences for emulsions of comparable droplet size were actually not that big, and that the claim that thick layers would reduce oxygen diffusion is not valid given the fact that interfacial layers contain many holes. Even if perfect these layers would not be thick and dense enough to slow down gas diffusion as demonstrated extensively for edible coatings [297,298], also not when gelled, and oxygen diffusion in oil is so fast that no gradients would occur [299,300].

5.3.2. *Digestive stability*

Emulsions can be used for controlled release through the loss of interface stability under digestive conditions. For emulsions stabilized with proteins and other surface-active components, this is expected to happen early on in the digestive tract [301,302]. This may lipolysis rates through droplet coalescence (reduced surface area), and not by a direct effect of the interfacial layer, unless specific combinations of surface-active components are used [278,303]. For PEs, digestive effects have been reported [19,304,51,46,47,15,48,287,305]. For indigestible particles, the findings were very nicely summarized in the review by Sarkar [48] who concluded that the ability of Pickering particles to prevent displacement by bile salts leads to a reduced oil droplet area available for lipid hydrolysis to take place, and thus reduced formation of free fatty acids and extent of digestion. If the particles are displaced, the emulsions are no longer protected, and either are digested very rapidly or coalesce. Biodegradable particles add complexity to the digestive behavior, leading to a reduction in particle size, and most probably production of low molecular weight components of which the effect is covered in Figs. 1c and A1 (smaller particles, lower interfacial tension, and an unpredictable effect on contact angle), thus leading to lower desorption energy and emulsion stability.

When particles form a gel layer, first gel digestion by abrasion needs to take place, before lipid digestion can take place [306,307]. Furthermore, gelled layers may slow down lipase diffusion through their mesh size [287,48], which is in line with findings for gel beads containing oil droplets [308,309]. This effect was dependent on the temperature treatment given to starch particle-stabilized emulsions [267], thus also affecting the encapsulation efficiency of bioactives [219]. To be complete, droplet aggregation is expected to influence digestion by effectively enlarging the diffusive distance of digestive enzymes.

5.3.3. *Other products*

PEs are products in their own right, as well as starting points for other products such as high internal phase emulsions, both oil-in-water [28,310], and water-in-oil [54] and encapsulates e.g., [311], that can be strengthened and further functionalized by the addition of other layers that consist of spherical as well as fibrous particles [312,313]. Some of these systems have been shown to exhibit a strong pH response that can be relevant for delayed digestion [312] either of the oil or of components that are dissolved in the oil. Most of the work reported on food PEs relates to simple micrometer-sized emulsions, but also nano-emulsions have been reported [314,315], as well as double emulsions as reviewed in [310,314]. Furthermore, PEs have formed the basis for encapsulates [45,304], powders [316,317], foams [2,12] in which in some cases particles were formed at the site [318], and even more complex systems [95], that have been reported for their heat e.g., [267] as well as cold stability e.g., [319]. Besides, Pickering foams have been reported [318,95,2,12], and anti-bubbles [320] with excellent stability thanks to the previously mentioned curvature effects typical of particle-stabilized systems (see Fig. 2).

When taking inspiration from fundamental studies, it is clear that typical particles as occur in food ingredients have their benefits. They are generally not spherical and rough, and may be heterogeneous in their surface composition which may increase their desorption energy, thus making them less likely to be removed from the interface that they are stabilizing [23,20,24]. This holds even more strongly for deformable particles that are expected to be amply present in foods, or be formed upon rehydration of powders [321,322,323]. There are also a number of effects that go beyond the actual Pickering mechanism. Through their multiple charges, these particles may enhance connectivity in the interfacial layer [29,24]. Besides, particles may form a network in the bulk that adds to the stability of emulsions e.g., [61], or one that is formed upon gelling [217,52,267,53], or upon rehydration of particles

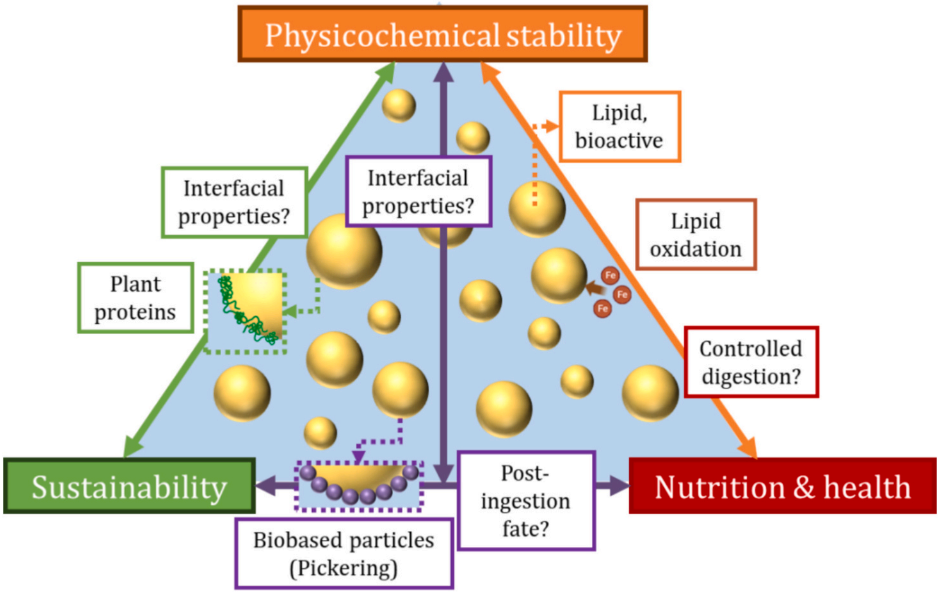

Fig. 6. Summary of various aspects related to PEs from the graphical abstract of (Berton-Carabin & Schroën, [129].

[321,322,60]), which also effectively reduces the volume fraction of free water, thus adding to viscosity of the continuous phase, and possibly network formation even at high volume fractions of the dispersed phase [54,276]. This positions emulsions made with particles well for the formation of foods and food elements with special properties as very recently reviewed in [50].

6. Conclusion/outlook

Although often claimed to lead to Pickering emulsions (PE), emulsion formulations that contain (solid) particles, mostly do not lead to emulsions that derive their stability (solely) from particles at the interface, a prerequisite for the term PE. In most practical 'Pickering' food emulsions, mixed interfaces will be present, either as a result of the composition of the ingredients, or of the emulsification technique used that may reduce particles to smaller entities (e.g., during homogenization). This leads to a reduction in interfacial tension, and possibly a change in contact angle, that both reduce the desorption energy of the particles, and thus emulsion stability. Soft particles show behavior between that of polymers and solid particles, with dangling chains readily adsorbing, and particle deformation leading to relatively high desorption energy, and high emulsion stability. As long as particles remain at the interface in sufficient amounts, droplets will be physically stable against coalescence, and Ostwald ripening even when an oil with appreciable solubility in the water phase is used. These emulsions may not be stable against gravitational separation, for which one may consider network formation of the particles in the bulk phase including particle-covered droplets as part thereof, thus making use of aggregation behavior to enhance stability.

The use of particles in food emulsion products can generate additional functionality, e.g., oxidative stability, and enhanced controlled digestion and release of components compared to regular emulsions. Besides, the use of particles is a versatile option to create food products with increased shelf-life, can contribute to health, as well as sustainable food products [129] (see Fig. 6).

CRediT authorship contribution statement

Karin Schroën: Writing – review & editing, Writing – original draft, Supervision, Investigation, Funding acquisition, Conceptualization. **Xuefeng Shen:** Writing – original draft, Validation, Methodology, Conceptualization. **Fathinah Islami Hasyyati:** Writing – original draft, Visualization, Validation, Methodology, Conceptualization. **Siddharth Deshpande:** Writing – review & editing, Supervision, Methodology, Conceptualization. **Jasper van der Gucht:** Writing – review & editing, Supervision, Funding acquisition, Conceptualization.

Declaration of competing interest

The authors declare no conflict of interest.

Data availability

Data will be made available on request.

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Appendix A. Appendix

Table A1
Some features of common methods and machines to produce emulsions.

Homogenizer Types	Flow Regime	(Un)-Bounded Flow	Energy Density ($J\cdot m^{-3}$)	Relative Energy Efficiency	Droplet sizes Achieved	Viscosity	Typical Volumes
Stirred tanks	TI, TV, LV	U	Low–High	Low	2 μm and larger	low to medium	batches up to several m^3
Colloid mill	LV (TV)	B	Low–High 10^3 to 10^8	Inter-mediate	1 to 5 μm	medium to high	4 to 20,000 $l\cdot h^{-1}$
Tooth disc disperser (e.g. Ultraturrax)	TV	B	Low–High 10^3 to 10^8	Inter-mediate	1–10 μm	low to medium	batches cm^3 up to several m^3
High-pressure homogenizer	TI, TV, (CI), LV	U	Medium–High 10^6 to 10^8	High	0.1 μm	low to medium	100 to 20,000 $l\cdot h^{-1}$
Ultrasonic probe	CI	U	Medium–High 10^6 to 10^8	Low	0.1 μm	low to medium	batches <100 cm^3
Ultrasonic jet	CI	U	Medium–High 10^6 to 10^8	High	1 μm	low to medium	1 to 500,000 $l\cdot h^{-1}$
Micro-fluidization	TI, TV	B/U	Medium–High 10^6 to 10^8	High	<0.1 μm	low to medium	up to 12,000 $l\cdot h^{-1}$
Membrane and Micro-channel	Injection STB	B	Low 10^3	Exceptionally high	0.3 μm—often larger	low to medium	batch or semi-continuous 10's $l\cdot h^{-1}$

Table from Rayner [189,190]; reprinted with permission from CRC Press. The flow regimes are: LV = laminar–viscous, TV = turbulent–viscous, TI = turbulent–inertial, CI = cavitation–inertial, STB = spontaneous transformation based. Low viscosity = like water, medium viscosity = like cream, high viscosity = like honey.

Table A2
Equations for estimating stresses exerted on droplets, expected mean particle diameters, adsorption times of surfactants, droplet deformation times, and collision times for emulsion droplets under laminar and turbulent flow conditions.

Flow Regime	Laminar—Viscous Shear or Elongational (LV)	Turbulent–Viscous Shear Forces (TV)	Turbulent–Inertial Forces (TI)
Re—flow	<1000	>approx. 2000	>approx. 2000
Re-droplet	<1	<1	>1 *
Mean Diameter ($d \approx$) #	$\frac{2\gamma We_{cr}}{\eta_c G}$	$\frac{\gamma}{\sqrt{\varepsilon \eta_c}}$	$\left(\frac{\gamma^3}{\varepsilon^2 \rho_c}\right)^{1/5}$
External stress acting on droplets (σ)	$\eta_c G$	$\sqrt{\varepsilon \eta_c}$	$\sqrt[3]{\varepsilon^2 d^2 \rho_c}$
Droplet deformation time scale (τ_{DEF})	$\frac{\eta_D}{\eta_c G}$	$\frac{\eta_D}{\sqrt{\varepsilon \eta_c}}$	$\frac{\eta_D}{\sqrt[3]{\varepsilon^2 d^2 \rho_c}}$
Duration of disruptive stresses (τ_{DIS})	$\frac{1}{G}$	$\sqrt{\frac{\eta_c}{\varepsilon}}$	$\frac{1}{2}\left(\frac{\gamma^2 \rho_c}{\varepsilon^3}\right)^{1/5}$
Surfactant adsorption time scale (τ_{ADS})	$\frac{6\pi\Gamma}{d\, m_c G}$	$\frac{6\pi\Gamma}{d\, m_c}\sqrt{\frac{\eta_c}{\varepsilon}}$	$\frac{\Gamma}{m_c}\sqrt[3]{\frac{\rho_c}{d\,\varepsilon}}$
Droplet collision time scale (τ_{COL})	$\frac{\pi}{8 G \phi}$	–	$\frac{1}{15\phi}\sqrt[3]{\frac{d^2 \rho_c}{\varepsilon}}$

Adapted from: ([1]; [193]; [191]). Symbols: We = Weber number (see text); Re = Reynolds number (see text); Re-droplet = Reynolds number but with the droplet size as characteristic dimension; Γ = surface excess of surfactant ($mol\ m^{-2}$); ε = power density ($J\cdot s^{-1}\cdot m^{-3}$); d = droplet diameter (m); γ = interfacial tension ($J\cdot m^{-2}$); η = viscosity (Pa·s); G = velocity gradient (s^{-1}); m_c = surfactant concentration in the continuous phase ($mol\cdot m^{-3}$); τ = characteristic time (s); σ = stress (Pa); ρ = density ($kg\cdot m^{-3}$). Subscripts: d = dispersed phase; c = continuous phase; cr = critical value for droplet break-up; DEF = deformation; DIS = disruptive, ADS = adsorption; COL = collision. Notes: * For $d > \eta_c^2/(\gamma\, \rho_c)$. # Only if $\eta_d \gg \eta_c$. Table reprinted with permission from CRC Press [189].

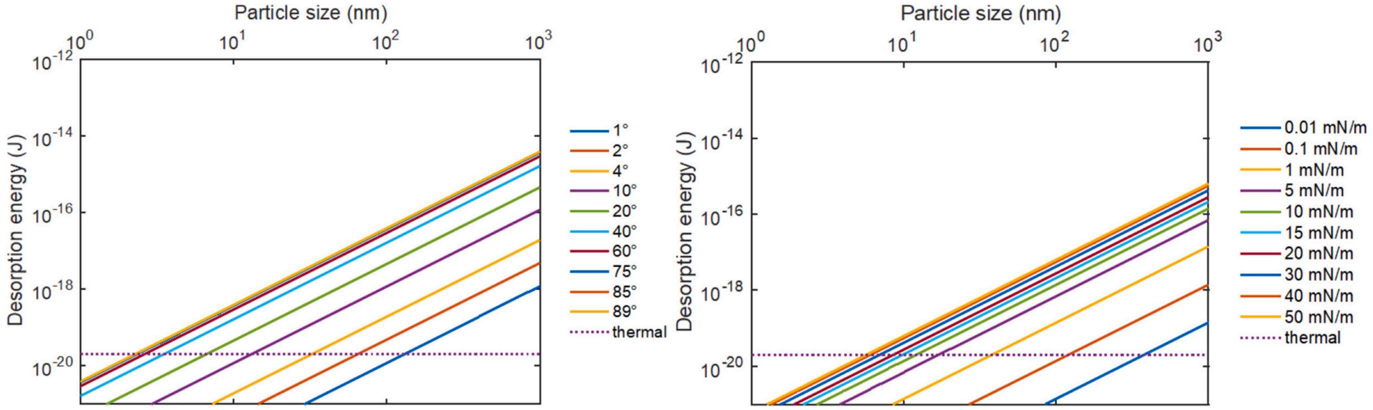

Fig. A1. Revisited version of Fig. 2, with 5 mN/m as interfacial tension in the left graph, and a contact angle of 60 °for the right graph. The thermal desorption energy has been taken as during a heat treatment (120 °C).

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