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Towards new food emulsions: designing the interface and beyond

Claire Berton-Carabin and Karin Schroën

Emulsions are ubiquitous in foods, and decades of research work have led to advanced, although often empirical, control over the formulation and functionality of those systems. However, the conventional strategies to make food emulsions have to be revisited, due to the trends in the food sector area that have emerged in recent years. This includes a strong focus on naturalness, health and sustainability, which promotes the use of plant-derived ingredients, ideally obtained from mild processing, and thus, by essence, far from pure and well-characterized. Adapting to this change of mind while ensuring the physicochemical stability of emulsions is a challenge, and requires that researchers invest effort into deep characterization of the emulsions' microstructure and dynamics, for which tools to characterize multiple scales are, more than ever, an essential need.

Address
Wageningen University, Laboratory of Food Process Engineering, 6700 AA Wageningen, The Netherlands

Corresponding author:
Berton-Carabin, Claire (claire.carabin-berton@wur.nl)

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Introduction

Many food products contain two or more immiscible phases, often oil and water, which exist as one phase dispersed in the other as colloidal droplets. Such systems are water-in-oil (W/O) emulsions represented, for instance, by butter and margarine, or oil-in-water (O/W) emulsions, as found in a broad range of beverages, milk, infant formulas and other dairy-based products, mayonnaise, dressings and sauces (Figure 1) [1••]. Decades of fundamental and applied work have built advanced state-of-the-art knowledge on the factors that affect the properties and stability of emulsion systems. However, in the current context of high consumer demand for healthy and clean-label foods, and of transition to more sustainable ingredients, some of the established paradigms and formulation rules need to be reconsidered, which is a challenge for both the academic and private sectors. We thus aim at identifying directions that seem promising, and even essential, for the development of the next generations of food emulsions.

Main properties of food emulsions

Emulsifiers

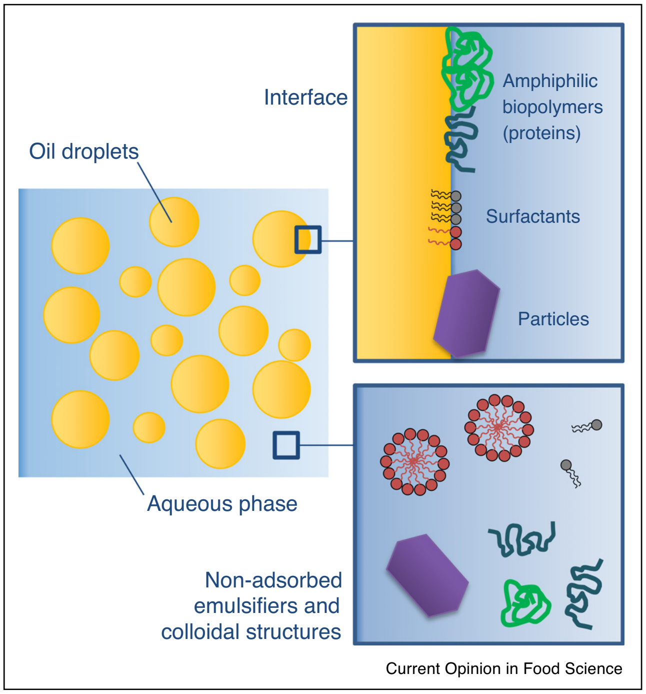
Because of the molecular incompatibility between oil and water, emulsions are thermodynamically unstable, and strive for minimizing the interfacial area between oil and water, which involves the physical destabilization of the system and ultimate complete phase separation. It is possible to retard this process to time scales that are substantially larger than typical storage times, which can be achieved by using emulsifiers. Emulsifiers are surface-active molecules that consist of hydrophilic and hydrophobic parts, which makes them able to adsorb at the oil-water interface [2,3•] (Figure 1). Two main categories of food emulsifiers exist: low molecular weight emulsifiers (LMWEs), which are represented by, for example, lecithins, polysorbates, or monoglycerides and diglycerides [4]; and amphiphilic biopolymers, of which the main example is proteins (e.g. whey proteins, caseins) [5].

In the past decade, interest has been rising in the use of more sustainable food ingredients such as plant proteins, of which the functional properties are not that thoroughly characterized yet. Furthermore, a third category of food emulsifiers, colloidal particles, has become popular. Such particles, when having affinity for both oil and water, can anchor at the interface, forming a strong mechanical interfacial barrier in so-called Pickering emulsions. Interfacial layers containing Pickering particles are much thicker than conventional emulsifier-based ones; the particles need a certain size, and wettability, to rather irreversibly nest in the interface and give emulsions high physical stability. Although the proof of concept and development of Pickering emulsions had historically been done using inorganic particles and non-food oils, many biobased particles have now been identified as useful in that respect [6••,7].

Physicochemical stability and related issues

Emulsions are, in essence, unstable systems. They may destabilize physically through different phenomena, the main ones being [1••]: flocculation, when two or more droplets stick to each other while retaining their individual integrity, forming flocs; coalescence, when droplets merge after rupture of the interfacial film, leading to

Figure 1



Schematic representation of an oil-in-water (O/W) emulsion and its different phases. Such systems comprise molecules and colloidal structures with affinity for both oil and water, which partition between the different available phases. Drawing is not to scale.

larger droplets; gravitational separation between the continuous phase and the droplets (e.g. creaming for O/W emulsions), which is due to the density difference between oil and water; and phase inversion, that is, transition to a W/O emulsion from an O/W emulsion, or *vice versa*. Although some of these phenomena are sometimes used purposely to induce-specific structures (e.g., partial coalescence of the fat droplets in ice cream, or droplet flocculation in cream cheese), they are generally unwanted, as they lead to defects in the texture and appearance of the products. The droplet size is an important property of emulsions and may vary by orders of magnitude depending on the application, from around 0.1–0.2 μm to around 100 μm. This directly impacts the total interfacial area, and thus the required amount of emulsifier to cover it, and can be notably of importance in emulsions combining small droplet sizes and aggregated emulsifiers, where adjacent droplets may share adsorbed aggregates [8].

Another major concern regarding the stability of food emulsions is their chemical stability when they contain labile components. These mainly include lipophilic components, for instance polyunsaturated fatty acids (PUFAs), which are well recognized for their health benefits [9]; however, are also highly sensitive to oxidation. This leads to the degradation of the product's nutritional quality, and generates off-flavors. It is well known that lipid oxidation in O/W emulsions depends on the structure and composition of the system, through intricate and interrelated pathways [10,11]. A prominent development in this field over the past decade has been the conceptualisation of the so-called cut-off effect [12,13•,14], according to which antioxidants that have been lipophilized by grafting of an alkyl chain are more efficient to counteract lipid oxidation in emulsions than their unmodified hydrophilic counterparts, which is attributed to their preferred location at the oil–water interface, where oxidation is supposed to be initiated. Beyond a certain alkyl chain length, a decrease in the antioxidant activity is observed, which is attributed to the aggregation and/or intra-droplet location of these molecules. From this, it has be concluded that positioning antioxidants at the right place in those multiphase matrices seems to be a key strategy for controlling lipid oxidation.

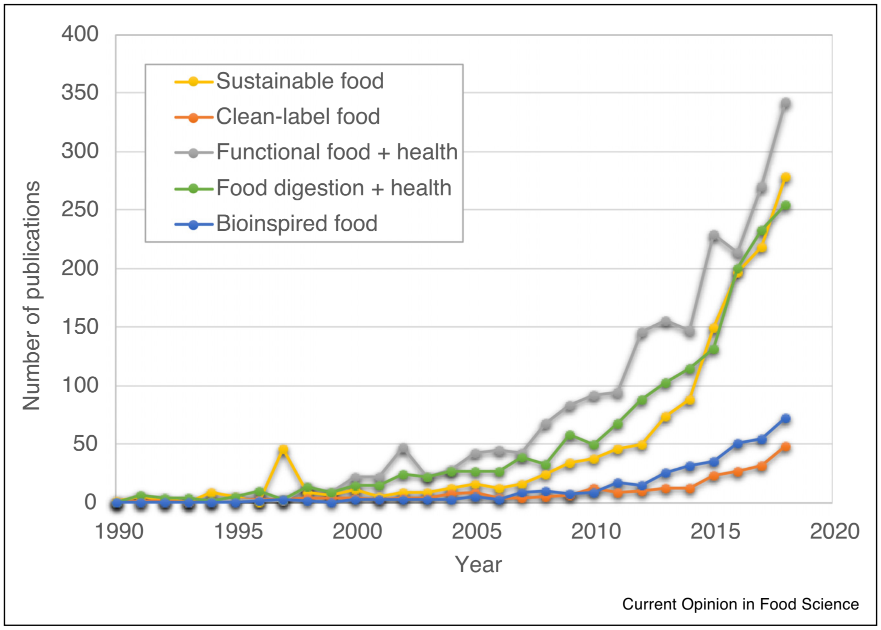
Trends and challenges in the design of food emulsions

Sustainable ingredients

A leading trend in food design is the current transition to more sustainable foods and ingredients (Figure 2), which, for emulsions, mostly comes down to the origin of the proteins used as emulsifiers. Although dairy proteins are known for their high functionality, in the light of the current protein transition it is desirable to consider plant-derived alternatives [15,16]. Substantial work has already been done to characterize the interfacial and emulsifying properties of various plant-derived protein ingredients, for a large part, focusing on soy proteins [17,18]. Yet, the use of soy proteins has been subjected to debate, due to, for example, allergenicity and genetic modification issues, especially in Europe. Therefore, in countries where soybean production is scarce [19], efforts are currently made at considering other plant protein sources, of which some have been proved useful as food emulsifiers, such as pea [20,21,22•], lupin [23,24], or lentil proteins [25–27].

The use of plant proteins as emulsifiers has been extensively reviewed lately, for instance by Sharif *et al.* [28], Lam *et al.* [29•] and Burger and Zhang [30]; despite the great interest in their use, some drawbacks still need to be overcome. First, a true sustainability superiority over animal-derived proteins can only be achieved if the separation process applied to obtain the protein ingredient is not too energy-consuming and water-consuming (this is often not the case for high purity isolates, which are generally subjected to a thermo-aggregation step at low pH, leading to protein aggregation and poor solubility). Thus, mildly refined plant protein fractions should be preferred, which implies that the non-protein ingredients present in these fractions (e.g. polysaccharides) should be

Figure 2



Number of publications in the past two decades with the terms 'sustainable food', 'clean-label food', 'functional food + health', 'food digestion + health', or 'bioinspired food' as title and/or topic (*Web of Science*, March 2019).

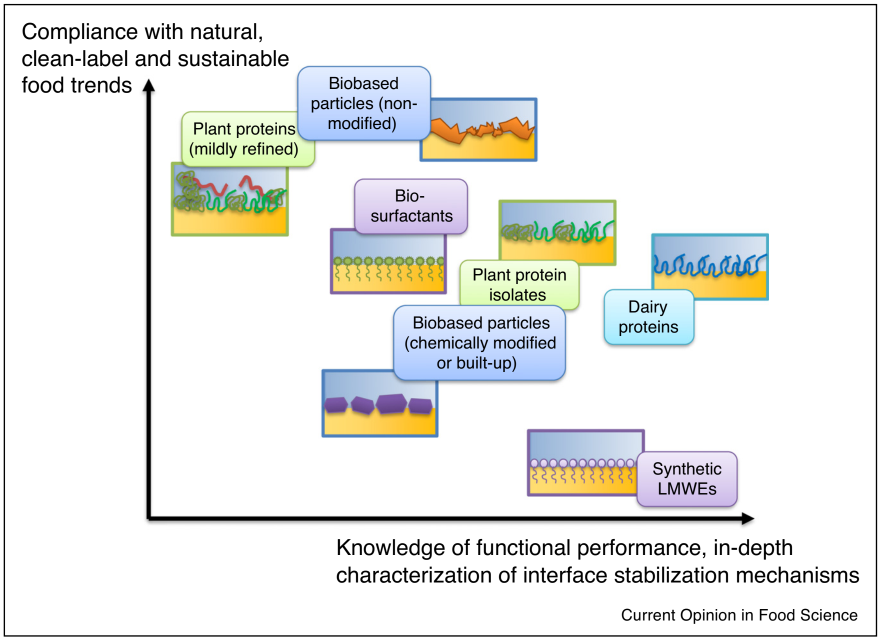
an integrate part of the emulsion design strategy [31]. Second, many plant protein ingredients show limited solubility in aqueous media at neutral or acidic pH, which may limit their ability to rapidly adsorb at fluid interfaces [30,32]; besides, they exhibit relatively large surface loads compared to, for example, dairy proteins [3•], possibly requiring the use of higher emulsifier concentrations. As previously mentioned, the interfacial behavior of plant proteins is not comprehensively understood yet, and that leads us to conclude that there is still a considerable gap to bridge. The knowledge that needs to be generated should be directed towards systematic understanding of interfacial and emulsifying properties of plant proteins in relation to their molecular and supramolecular structures (Figure 3), and linking this to emulsion stability, which is a first step towards actual food applications; in a later stage, also sensory properties, digestibility, nutritional value and so on, need to be considered.

Clean-label ingredients

Consumers are nowadays more and more reluctant towards food products containing synthetic additives often referred to as their official identification number within the European Union, or E-number. This has pushed food manufacturers to try to minimize the use of such additives, and to prefer 'natural' ingredients, which is referred to as the 'clean-label' trend [33]. Although no official regulation for clean-label food exists yet, recently there has been a massive effort at investigating the potential of natural, biobased molecules or ingredients to stabilize emulsions, both physically and chemically. Already in 1993, Dickinson published a review article entitled 'Towards more natural emulsifiers' [34], yet most of the research on this topic has been published in the past five years.

Of course, many emulsifiers that have been traditionally used in food emulsions are of natural origin, such as lecithin or dairy proteins, but many others are, in fact, synthetic, such as polysorbates or sucrose esters (Figure 3). As recently reviewed [35•], alternatives to synthetic LMWEs can be compounds called bioemulsifiers or biosurfactants, such as *Quillaja* saponin [36]. Yet natural polymers are the most obvious candidates, including conventional or alternative protein sources — which can also be a way to mitigate sustainability issues, by using plant proteins. Finally, as mentioned earlier, biobased particles have recently met strong interest for stabilizing emulsions instead of conventional emulsifiers. A distinction here should be made depending on the level of processing and chemical modifications that are necessary to obtain particles with suitable functionality. Pickering particles that naturally occur and need minimal modifications, such as cocoa [37] or citrus fibers [38] are expected to be preferred over particles that need chemical modification (e.g. hydrophobization of starch granules by octenylsuccinic anhydride, OSA [39]) or that are purposely engineered from relatively pure ingredients (e.g. colloidal lipid particles [40] or protein-polysaccharides complex particles [41]).

Figure 3



Comparison of different food emulsifiers based on: (X-axis) the state-of-the-art knowledge regarding their functionality, that is, interfacial and emulsifying behavior; and (Y-axis) their potential to comply with sustainability and clean-label requirements. Obviously, the ideal emulsifier would be located in the top right corner of this graph. It should be pointed out that this graph does not include the amount of material that would be needed to stabilize an interface, that is, the surface load, which is much larger for colloidal particles compared to molecular emulsifiers.

Ensuring the oxidative stability of food emulsions without using synthetic antioxidants such as ethylenediamine tetra acetic acid (EDTA) is also a challenge, and the development of natural alternatives is of great relevance. It is well-known that proteins present in the aqueous phase of O/W emulsions can have a strong antioxidant activity [42], but whether this could fully be an industrially relevant alternative is still to be explored. Besides, it should be kept in mind that the efficiency of antioxidants in emulsions is highly matrix-dependent, and so far, effects have failed to be generalized [43••]. This is certainly due to the dynamic physical location of antioxidant molecules, due to diffusive transfer and exchange between both phases and the interface, that has often not been taken into consideration, and which could be crucial to achieve rational solutions [44••,45].

Health-promoting emulsions and controllable digestive fate

"Let food be thy medicine and medicine be thy food": here is the title of a recent review article dealing with the trends in the food sector, and notably with the increasing interest in health-promoting and functional foods [46] (Figure 2). This trend has undoubtedly influenced the design of food emulsions, and in particular their composition. Emulsions can be used as a reservoir for bioactive, health-promoting molecules such as PUFAs, vitamins, or phytochemicals [47] that are often lipophilic and can be combined directly with the dispersed oil phase. The continuous hydrophilic phase ensures compatibility with a broad range of food formulations. It is also possible to design advanced emulsion-based systems, such as double emulsions, for the encapsulation of hydrophilic components, but this is outside the scope of the present review.

Ensuring stable encapsulation of lipophilic components in emulsions has been extensively studied; in addition, the ultimate step in a food emulsion's lifespan, that is, its fate in gastrointestinal conditions, determines it efficacy as a delivery system, which has become an important field of research and an integral part of food emulsion formulation. The point of the present article is not to give a comprehensive overview of the emulsion digestion process; for that, we refer the interested reader to, for example, the reviews of Golding and Wooster [48], and more recently of Corstens *et al.* [49•], Guo *et al.* [50], and McClements [51]. When it comes to emulsion design, the composition and structure of emulsion-based systems has often been claimed to modulate their digestibility and thus control their digestive fate. Promoting the digestion and bioavailability of lipophilic bioactives (e.g. lipophilic vitamins, pigments) via rational emulsion design has been an active research area [47,52]. Independently of the targeted emulsion fate, a key question is whether the digestive model used under lab conditions is relevant to phenomena occurring in the human gastrointestinal tract [53••]. Most studies are conducted using static *in vitro* models, which are certainly useful for screening purposes, but more advanced studies including dynamic *in vitro* and *in vivo* models are essential before bringing promising systems towards applications.

Alternatively, delaying lipid digestion has also been a hot topic, because slow and more distal hydrolysis and absorption of triglycerides are associated with enhanced satiety [49•]. This is, however, difficult to achieve as the human gastrointestinal has evolved over millions of years to optimally digest food materials and absorb their nutritional components. Some attempts have been made at manipulating the physical state of the lipid phase [54,55], or the composition and structure of the oil droplet surface [56–59], leading to mixed, sometimes contradictory, and highly matrix-dependent results. A more robust approach to delay lipolysis seems to lie in the entrapment of emulsion droplets within a hydrogel matrix, for example protein gels [60,61] or alginate beads [62•,63], which mostly revolves around the design of the gels beads and not so much around that of the emulsion and its interface.

Bioinspired emulsions

Emulsion droplets naturally exist in living organisms, in the form of, for example, oil bodies for plants, and lipoproteins or milk fat globules for mammals. For example, the oil–water interface in these emulsions has a complex composition and structure: oil bodies are covered by a monolayer of phospholipids and embedded proteins, mostly from the oleosin family, and milk fat globules are covered by a trilayer of phospholipids with embedded membrane proteins and cholesterol [64]. These systems have optimal physical and chemical stability, which is explained by their formation mechanisms, and show targeted responsiveness towards their physiological roles. For example, the natural milk fat globule membrane present in breast milk is recognized for its health benefits in infants [65••]. This has been a source of inspiration for researchers in the food science area [66•] (Figure 2). In the past few years, a number of articles have reported strategies for mimicking the milk fat globule membrane in O/W emulsions, such as incorporating bovine milk phospholipids [67] or milk fat globule membrane fragments [68], or by adsorbing an external phospholipid layer onto pre-formed droplets using electrostatic deposition [69]. The applicability of such strategies to industrial emulsion formulation depends, however, on the complexity and scaling-up possibilities of the involved interfacial engineering, which can be a real challenge.

Towards multiscale analysis and design

Achieving a high level of control over emulsion structure is only possible when the structure itself can be accurately characterized at various scales, going from molecular composition of both phases and the interface, to interface functionality, and ultimately food product stability. Food emulsions are complex systems from a composition and morphology point of view, and the various molecules present may partition among the available phases. For instance, the effect of the continuous phase fraction of emulsifiers on the physicochemical stability of emulsions is often overlooked, even though these non-adsorbed emulsifiers can play an important role as they may induce depletion flocculation [1••], enhance compositional ripening [70] and more generally, influence transport phenomena, and often have an antioxidant role [11]. Excess emulsifiers may represent a very large fraction of the total used (Figure 1), and the interfacial composition may differ substantially from the composition of the emulsifier ingredients used. Thus, the interfacial composition should be systematically characterized, also as function of time to chart exchange processes.

This can be done in different ways, by non-invasive, invasive, or destructive methods [3•]. The structure of the corresponding interfacial layer is of importance, and may be studied *in situ* by, for example, microscopy techniques, or using two-dimensional model interfaces [3•]. A drawback of most of these methods is their static character, which to some extent can be mitigated by using expanding/compressing interfaces as is possible in a Langmuir trough, and in droplet volume tensiometry, and from which data related to interface rheology can be extracted [71••]. Still there is an essential drawback; the interfacial layers are formed by passive spreading or diffusion, whereas the conditions encountered in conventional emulsification devices, such as high pressure homogenizers, are highly dynamic and involve active mass transport. An interesting approach to take these aspects into account is the use of microfluidic methods to study the formation and short-term coalescence stability of emulsion droplets, as these devices operate at very short time-scales, and involve convective transport [72•,73–75].

To wrap up, combining different techniques and scales seems essential to unravel the interfacial microstructure in food emulsions, particularly when ingredients of high compositional complexity are used, such as plant protein and biobased particulate materials. Although the optimal link between industrial conditions and those that can be achieved under lab conditions is still a difficult one to establish, more and more tools become available to do exactly that.

Conclusions

In the past few years, research in the field of food emulsions has encountered pivotal influences that open perspectives, but also identify many challenges. Some leading trends may be conflicting, such as increasing the levels of PUFAs while removing synthetic antioxidants, or striving for stable emulsion products while using less refined and well-characterized ingredients. Despite the exponential number of scientific articles published in the field of food emulsions, there is a large gap between scientific research and industrial applications. Taking the complexity of relevant components and of the structures formed as an integral part of the studies, and using a multiscale, multidisciplinary approach to deal with this complexity is expected to be key for future developments.

Conflict of interest

The authors have no conflict of interest to declare.

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- •• of outstanding interest

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