

Cite this: *Sustainable Food Technol.*,  
2025, 3, 96Received 3rd July 2024  
Accepted 4th November 2024

DOI: 10.1039/d4fb00201f

rsc.li/susfoodtech

# Innovations and stability challenges in food emulsions

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When two immiscible liquids are mixed, they naturally stay in separate phases. This is because these liquids, due to their molecular properties, cannot spontaneously blend into a uniform mixture. Over the years, research has been focused on achieving long term stability in emulsions and significant progress has been made. But in the food industry, emphasis on sustainability has led to increased interest in methods that can achieve emulsion stability through green practices. This includes use of biopolymers and biodegradable materials, innovations to reduce food waste and food conservation. Emulsions have also been used in many innovative applications such as coatings, films, 3D printing inks, encapsulation systems and fat replacers. This review aims to briefly introduce different types of emulsions, their physical instabilities, recent innovations and how they align with sustainability and regulatory requirements.

## Sustainability spotlight

Food emulsions are thermodynamically unstable systems, thus in order to manufacture them, kinetic stability provision is necessary. For that, the employment of substances such as surfactants is required. However, in a scenario of population growth and global warming, it is crucial to provide the referred stability integrated in a sustainable and conscious approach, which has the potential to increase food demand and reduce negative environmental impact. The role of emulsions in such a context is not restricted to its constituents synthesis and production, though. Innovations concerning these colloidal dispersions might impact human health positively, and are also suitable biodegradable alternatives to synthetic polymers, and toxic conservants. Emulsions are all that, plus compatible with the future, considering the additive manufacturing. This work is aligned with the following UN sustainable development goals: Zero Hunger (SDG 2), Good Health and Well-being (SDG 3), Industry, Innovation, and Infrastructure (SDG 9), Responsible Consumption and Production (SDG 12), Climate Action (SDG 13), Life Below Water (SDG 14) and Life on Land (SDG 15).

## 1 Introduction

Emulsions are fundamental in the food industry, as they have the ability to provide essential properties for a wide range of products, which are going to be further discussed in this review article. The importance of emulsions in food is shown by a very interesting fact: the term “emulsion” is deeply rooted in human dietary history. Indeed, the very existence of the word “emulsion” is tied to the milk consumption of other animals by mankind. This is curiously justified as the etymology of “emulsion” traces back to the Latin “*emulsionem*,” which means “to milk”.<sup>1</sup> This emphasizes the crucial role that emulsions have played in the human knowledge of food and its components.

Besides the nutritional benefits, milk, which is the first food consumed by humans, also offers a rich sensory experience.<sup>2,3</sup> This sensory appeal comes from the dispersion of oil in water while proteins play a key role in stabilizing this mixture. This results in a product with pleasing texture, taste and aroma. This principle applies to many emulsified products, both dairy such

as ice cream<sup>4</sup> and non-dairy. However, production of such systems poses challenges from a physicochemical point of view. Even when emulsions are formed, they can phase separate over time. Therefore, stability is a critical aspect of emulsions. Without stability, these systems become less viable and cannot do their intended purpose.<sup>5</sup>

Achieving stability in emulsions opens up the door not only to the production of food itself but also the use of them as intermediates with new applications in the food industry. Additionally, in the current social scenario where environmental and economic challenges are present, to ensure the stability of emulsions while adhering these specifications becomes each day more important. Therefore, it is crucial to promote sustainability practices in emulsion stability, which requires great efforts.

Thus, this review article aims to serve as a comprehensive compendium that highlights the key innovations of emulsions within the food industry, and to further examine their alignment with sustainability principles. Through an assessment of these innovations, it is possible to better understand how they contribute to sustainable practices in food production. To achieve this, however it is essential to integrate existing theoretical knowledge about the various types of emulsions and

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their inherent tendencies toward instability. This foundational understanding will provide context for the discussion of recent advancements and their implications for sustainability in food applications.

## 1.1 Instabilities

**1.1.1 Thermodynamic instability.** If analyzed the final result of mixing two immiscible liquids, it is concluded that the fate of such an experiment is the disassociation of the liquids and the consequent formation of two distinct phases. Emulsions, widely used in the food industry, however, have their existence supported precisely by antagonizing the previously introduced principle: it is described as an immiscible liquid colloiddally dispersed in another, forming a continuous phase and a dispersed one.<sup>6</sup>

From a physicochemical point of view, this tendency of non-dispersion between immiscible liquids, as long as they are in thermodynamic equilibrium, is translated as the existence of an extremely low degree of entropy ( $\Delta S$ ). Thus, considering the Gibbs free energy of formation, given in eqn (1), the product of entropy and temperature is disregarded. Therefore, the Gibbs free energy variation is always positive, since it only considers the product of the interfacial tension ( $\gamma$ ) by the increase in the interfacial area ( $\Delta A$ ), as observed in eqn (2). Such context, supported by the second law of thermodynamics, leads to the conclusion that the formation of emulsions is, in fact, non-spontaneous.<sup>7,8</sup>

$$\Delta G_{\text{formation}} = \gamma \Delta A - T \Delta S_{\text{config}} \quad (1)$$

$$\Delta G_{\text{formation}} = \gamma \Delta A \quad (2)$$

As a result, emulsions are assumed to be thermodynamically unstable systems. Consequently, for the formulation of such systems, it is necessary to postpone their eventual destabilization, which, in other words, culminates in making such dispersions kinetically stable.<sup>5</sup> The main physical instabilities commonly observed are classified into: flocculation, coalescence, Ostwald ripening, phase inversion and gravitational instabilities (creaming and sedimentation).<sup>9</sup>

**1.1.2 Physical instabilities.** Characterized by the irreversible increase of the dispersed phase droplets size, it is mutual the fate of both instabilities, coalescence and Ostwald ripening. They differ, however, in the mechanism behind this phenomenon. In coalescence, there is a rupture of the continuous phase film that separates the droplets, while in Ostwald ripening there is a diffusion of droplets with smaller diameters to larger ones, because of the Laplace pressure.<sup>10</sup> It is worth mentioning that food is often comprehended by complex structures. Thus, emulsion instabilities, even if irreversible, should not be totally depreciated when it comes to general food aspects. As exposed by Costa *et al.*,<sup>11</sup> optimal partial coalescence can reduce recrystallization in ice creams, consequently enhancing their ideal texture.

When it is observed the reversible condition of droplets gaining a higher conformational degree, leading to the formation of flakes, there is flocculation, which can be a precursor to

coalescence, Ostwald ripening, and gravitational instabilities. Gravitational instabilities, conducted by Stokes' law (eqn (3)), are defined by the reversible aggregation of droplets in the upper or lower fraction of the continuous phase, characterizing them as, respectively, creaming and sedimentation. It is interesting to note that Stokes' law describes the terminal velocity ( $u$ ) of a spherical body (in this case, referring to the dispersed phase droplet) moving in a Newtonian fluid (continuous phase). Therefore, by decreasing  $u$  when changing the observed variables, the rate at which the gravitational instabilities manifest is reduced.<sup>12-16</sup>

$$u = \frac{(\rho_2 - \rho_1)gd^2}{18\mu} \quad (3)$$

where:  $u$  = terminal velocity;  $g$  = acceleration by gravity;  $d$  = droplet radius;  $\rho_2$  = aqueous phase density;  $\rho_1$  = oily phase density;  $\mu$  = aqueous phase density.

Finally, considering the interchangeability of the initial disposition of the aqueous and oil phases, there is phase inversion,<sup>9</sup> often used in the emulsion manufacturing process.<sup>17</sup> Among the aforementioned instabilities, those that lead to an irreversible increase in droplet size are responsible for complete phase separation.<sup>18</sup>

**1.1.3 Types of emulsions and their susceptibility to instability.** Emulsions are generally categorized according to specific parameters, such as the arrangement and number of aqueous and oil phases, the droplets size, the droplets concentration and the type of stabilizer used to reduce the interfacial tension between the liquids. These different types of emulsions, in their individuality, confront instabilities in a unique way.<sup>19</sup>

Admitting the dispersed phase of an emulsion composed by oil and the continuous phase by water, these are called oil-in-water (O/W) emulsions (Fig. 1a), and the other way around, water-in-oil (W/O) emulsions (Fig. 1b). Despite such generality, there are also multiple emulsions, classified as water-in-oil-in-water (W/O/W) and oil-in-water-in-oil (O/W/O).<sup>20</sup> Multiple emulsions, especially W/O/W (Fig. 1c), are of great interest to the food industry. But given their complexity, maintaining the stability of these systems is more challenging when compared to conventional emulsions.<sup>21</sup>

Under the condition of droplet size greater than 1  $\mu\text{m}$ , the emulsions are considered macroemulsions; if it is between 20 nm and 500 nm, they display greater stability, less tendency to creaming, sedimentation and aggregation, and are categorized as nanoemulsions, hence exhibit nanotechnological applicability; and if it is between 10 nm and 50 nm, they are microemulsions.<sup>13,22,23</sup> Microemulsions, however, behave like colloidal solutions. Consequently, they are thermodynamically stable and kinetically unstable and are not considered real emulsions.<sup>24,25</sup>

High internal phase emulsions (HIPes) are those in which the concentration of the dispersed phase ( $\phi$ ) exceeds 74% of the continuous phase (Fig. 1d). As a result, they expose unique rheological characteristics, such as a considerable high viscosity. Thus, they are able to confront creaming and sedimentation with more effectiveness.<sup>26</sup>



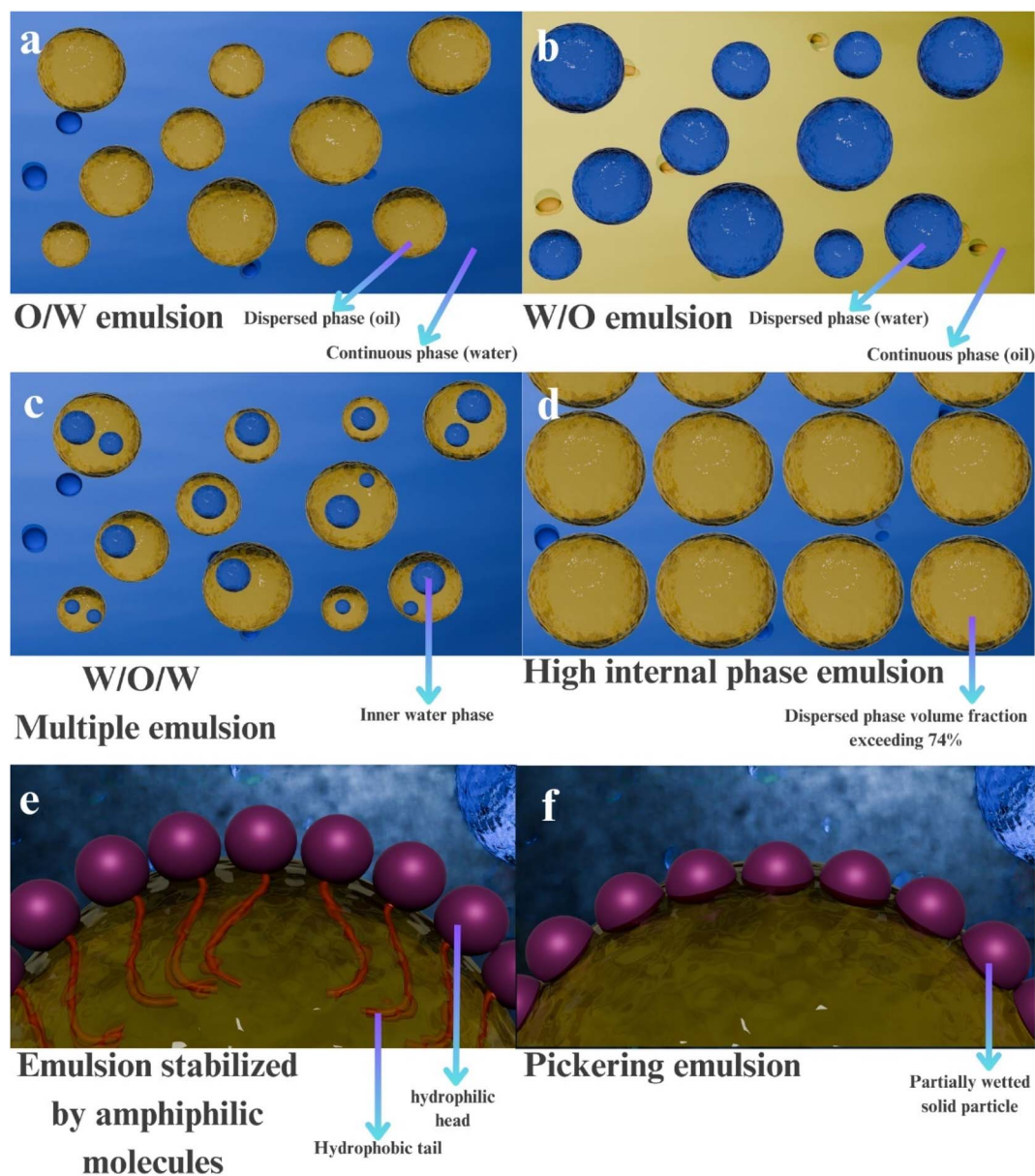


Fig. 1 Structural comparison of the main emulsion types ((a): oil in water emulsion; (b): water in oil emulsion; (c): water in oil in water emulsion; (d): high internal phase emulsion; (e): emulsion stabilized by amphiphilic molecules; (f): pickering emulsion). Created with Blender.

Classically, emulsions are stabilized by amphiphilic molecules that can adsorb at the liquids interfaces through their dual affinity driven by polarity (Fig. 1e). However, there are also the named Pickering emulsions, in which solid particles are responsible for reducing the interfacial tension of liquids (Fig. 1f). For this, the wettability concept is considered.<sup>27</sup> As the adsorption of such particles is irreversible, the steric stability conferred to those systems is considerably greater than observed in other emulsion types, which usually provides Pickering emulsions a longer shelf life.<sup>28</sup>

It is important to point out that the categorization of an emulsion, many times, is multifaceted. That is, a single system might share more of one classification parameter. Some examples are: Pickering nanoemulsions;<sup>29</sup> multiple Pickering emulsions;<sup>30</sup> Pickering HIPES;<sup>31</sup> O/W/O HIPES and W/O/W HIPES.<sup>32</sup>

## 2 Materials and methods

This literature review was conducted in two phases. In order to infer about the applicability of emulsions in the food industry and their use as a necessarily innovative method, it was assumed that patents are a reliable reflection of the given objective. Thus, a patent analysis was carried out in the Espacenet patent bank to identify the innovations. Then, to discuss the topics found through the patents, scientific articles were used.

### 2.1 Patent searching mechanism

Given the existence of different types of emulsions, four different search systematizations were submitted in the parent bank, as expressed in eqn (4)–(7). The feature “nftxt” was employed, and the chosen keywords were truncated with quotation marks and



articulated using the Booleans operators “AND” and “OR”, whenever necessary. The objective of each search was to find innovations of food-grade nanoemulsions, Pickering emulsions, HIPes and multiple emulsions, as they represent the main emulsion types, accordingly to Bai *et al.*<sup>19</sup> Due to their lower stability, no searching for macroemulsions was led, and because from a thermodynamical point of view, microemulsions are not real emulsions, they were discarded. The arrangement of the aqueous and oil phase was also not taken into account as a main parameter. After each search, the returned patents were ordered by the relevance feature, and then, in order of classification, the first twenty of each search were pre-selected.

$$\text{nftxt} = \text{“food-grade” AND nftxt} = \text{“nanoemulsion”} \quad (4)$$

$$\text{nftxt} = \text{“food-grade” AND nftxt} = \text{“Pickering” AND nftxt} = \text{“emulsion”} \quad (5)$$

$$\text{nftxt} = \text{“food-grade” AND nftxt} = \text{“high internal phase emulsion”} \quad (6)$$

$$\text{nftxt} = \text{“food-grade” AND (nftxt} = \text{“double emulsion” OR nftxt} = \text{“multiple emulsion”)} \quad (7)$$

With the sample space obtained, comprised of the eighty patents, it was observed that innovations related to food emulsions could be classified into five different innovative categories: “encapsulation”, “contemporaneity in the stability of emulsions”, “3D printing”, “edible coatings and films” and “fat mimetics”. These results are consistent with the novel applications in foods reported by Bai *et al.*,<sup>19</sup> except for the category concerning coatings and films.

It is indispensable to declare that the incidence of the aforementioned innovations is often concomitantly existent in the analysed patents. Therefore, for the purpose of categorizing the patents into a singular classification, the most narrowly defined one, and consequently, the less inclusive category, was always considered.

## 2.2 Eligibility criteria

The patent database was set up to retrieve patents exclusively in English and published within the last five years. Consequently, all eighty initially obtained patents already met both criteria. In regard to the eligibility criteria employed for the screening process, only emulsion innovations developed for food applications were considered. Patents concerning microemulsions were excluded, and in case of duplicate patents, only the first obtained was considered.

After a careful evaluation of the patents contemplating the criteria parameters aforementioned, 84% or 67 patents were included, while 16% or 13 were removed.

# 3 Results and discussion

## 3.1 Selected patents and their innovative classification

The highest incidence of innovation among the patents is represented by encapsulation, present in approximately 37.31% of

the obtained sample space. This is followed by Contemporaneity in the stability of emulsions, accounting for 35.82%, 3D printing with 11.94%, and both edible coatings and films and fat mimetics, each with 7.46%. Through Tables 1–5, the categorization of patents into their respective classes is evident. Within these tables, examples of the main stabilizers utilized, including emulsifiers, thickening agents, and polymers, is provided. It is crucial to note that not all stabilizers are included in the tables, particularly due to the wide range of ingredients occasionally used in patents. However, considering that the formulations may contain natural or synthetic components, random examples from each class are supplied. Furthermore, the tables explicate the primary applications of these emulsion advancements, delineating their utility in the food industry.

## 3.2 Encapsulation

The primitive function of food is to provide the necessary nutrients and energy for the proper human organism functioning. However, functional foods transcend this goal, because as they are prepared with different bioactive constituents, they culminate in the provision of extra benefits to human health.<sup>100–102</sup>

Considering the historical past regarding the functional food market, it is observed that from 2013 to 2017, there was a boost in its global revenue, which went from \$168 billion<sup>101</sup> to \$300 billion,<sup>103</sup> approximately. The given scenario demonstrates the potential of this sector, and consequently, the remarkable relevance of the production and development of this type of food for the industry, and emulsions are of enormous usefulness for functional foods because of encapsulation viability.

Although the encapsulation techniques are often implicitly present in other procedures, such as in the production of coatings and films,<sup>104</sup> in the development of fat mimetics<sup>105</sup> and others, here are discussed in publications with encapsulation taken as their central role, with the corresponding patents delineated in Table 1.

Several bioactive substances can be encapsulated in emulsions, such as: vitamins, carotenoids, flavonoids and probiotics.<sup>106</sup> However, the incorporation of these materials is not necessarily easy. The problem raised by Wackerbarth *et al.* (2009), regarding the encapsulation of carotenoids to the oil phase of an emulsion, well establishes this situation. The typical usage of organic solvents in this process, that is associated with toxicity, led the authors to develop an alternative method using bovine serum albumin (BSA). The BSA was able to form a complex with the carotenoid, which was found in the aqueous phase. Then, it was possible to proceed to emulsification and obtain an O/W emulsion, excluding the use of the said solvents for that.<sup>107</sup> This publication reinforces the importance of innovation also in the manufacture of emulsions.

An advantageous applicability of encapsulation in emulsions is the potential increase of the substances bioaccessibility/bioavailability, which make food by them structured as ideal delivery systems, as shown by patents.<sup>34,41–43</sup> It was reported by Wang *et al.* (2022), through *in vitro* studies following the INFOGEST protocol, that the bioaccessibility of





Table 1 Patents concerning encapsulation and their applicability

Patent	Stabilizer(s)	Application	References
CN112602875A	Tween 80, sodium dodecyl sulfate, lecithin, soy protein isolate, span 80	Development of a food-grade delivery system of water insoluble photosensitizers	33
CN108576778A	Sodium caseinate, octenyl succinic anhydride modified starch, tween 20	Improvement of bioavailability and solubility of lutein in functional foods	34
CN114431467A	Soybean protein isolate	Improvement of bioavailability, stability and solubility of carotenoid in functional foods	35
CN111643452A	Lecithin, tween 80	Improvement of bioavailability, stability and solubility of oxidized resveratrol in foods	36
CN111345385A	Zein, casein, chitosan	Flavor and scent-controlled release of natural substances in tea	37
AU2021106927A4	Tween 20, tween 80	Antibacterial/antimicrobial activity formulation for foods	38
CN107691948A	Tween 80, monoolein, diglyceride, monoolein	Aroma improvement of plant-based beverages	39
CN115868591A	Sucrose ester, xanthan gum, guar gum, gum Arabic	Improvement of stability and sensory aspects of camellia oil in a healthy beverage	40
CN108634169A	Sodium caseinate	Improvement of bioavailability and stability of lutein in functional foods	41
CN113812615A	Sodium caseinate, ovalbumin, whey protein	Improvement of bioavailability and stability of astaxanthin in foods	42
CN115006347A	Polyethylene glycol glycerol ricinoleate, polyoxyethylene castor oil	Improvement of bioavailability, biodegradability and biocompatibility of alpha-linolenic acid in medicines for pets. However, the invention has explicit potential for the food industry	43
CN112869119A	Casein, chitosan	Improvement of $\beta$ -carotene stability for food, focusing on the use of non-toxic and green substances	44
CN111481503A	Nano calcium carbonate	Controlled release of vitamin D3 and calcium supplementation	45
CN113974132A	SLP	Improvement of soybean oil stability	46
CN114680332A	Polyglycerol ricinoleate	Improvement of bioavailability, stability and sensory aspects of bitter substances in foods	47
CN113951511A	Modified soybean lipophilic protein	Improvement of bioavailability and stability of lycopene	48
CN107898873A	Tween 80, polyglycerol fatty acid ester, sucrose fatty acid ester, enzymatic soybean phospholipid, acacia, sodium starch octenyl succinate	Controlled release of polyphenols and stability improvement	49
CN109464395A	Polyoxyethylene monolaurate, polyoxyethylene lauryl ether, tween 20, tween 80	Development of a delivery system (no specific substance stated) suitable for food products	50
WO2021077380A1	Oxidized dextrin-currucumin/chitosan hydrochloride composite nanoparticles	Improvement of antioxidant activity and stability of encapsulated substances in functional foods	51
CN112826009A	Polyglycerol ricinoleate, polysorbate, sucrose fatty acid ester, sorbitol	Improvement of stability, solubility and sensory aspects of pigments for food coloring	52
WO2023055301A2	Octenyl succinic anhydride modified starch	Environmentally friendly encapsulation method of water-soluble substances	53
CN109078175A	Span 80, sodium lauryl sulfate	Food supplement of iron with higher stability and better sensory aspects	54
EP3925451A1	Milk proteins, polyglycerol polyricinoleate	Controlled release of bioactive substances, and improvement of stability and sensory aspects in nutritional foods	55
WO2021021026A1	Span 80, tween 20	Improvement of proteins solubility and stability in foods and beverages	56
WO2022140849A1	Gum Arabic, polysorbates, sugar esters, <i>Quillaja saponin</i>	Improvement of cannabis sensory aspects in food matrices	57

Table 2 Patents concerning contemporaneity in the stability of emulsions and their applicability

Patent	Stabilizer(s)	Application	References
CN113100417A	Pollen particles	Development of food-grade solid particles (for Pickering emulsions) based on pollen, which are eco-friendly and exhibit high biocompatibility and degradability	58
CN110403908A	Crystalline fat particles of hydrogenated vegetable oil, tween 20, tween 40 or tween 60	Low-cost food-grade and stable solid particles (for Pickering emulsions) based on vegetable oil. The patent claims low toxicity and eco-friendly characteristics	59
CN112741299A	Wax modified starch nanocrystals	Flavor controlled release system for foods, based on food-grade temperature-sensitive particles	60
CN114041607A	Bacterial cellulose, deamidated soybean protein isolate	Development of a safe nutritious food stabilized with a biotechnological emulsifier (bacterial cellulose)	61
CN115444124A	Beeswax, sodium caseinate, tween 40, tween 80	Low-cost food-grade lipid Pickering particles. The patent claims low toxicity and eco-friendly characteristics	62
WO2022073439A1	Tween 80-modified nano-selenium	Development of food-grade solid particles (for Pickering emulsions) based on selenium which are biologically active	63
CN111772171A	Soy protein isolate, lactoferrin nanoparticles	Development of food-grade stable solid particles (for Pickering emulsions) based on soy protein isolate and lactoferrin	64
CN109452447A	Soy protein isolate, anthocyanin, glycinin	Development of food-grade stable solid particles (for Pickering emulsions) with good emulsifying properties, based on soy protein isolate and anthocyanin	65
WO2021104144A1	Phytosterol particles	Development of food-grade solid particles (for Pickering emulsions) with good emulsifying properties, based on phytosterol, which is a green resource	66
CN114009741A	Citric acid stearate, lipid particles, sodium caseinate, guar gum, xanthan gum	Development of green and non-toxic food-grade solid particles (for Pickering emulsions) with good emulsifying properties, based on citric acid stearate	67
CN108851019A	Bacterial cellulose particles, sodium alginate, iota-carrageenan, low methoxy pectin, polyglycerol ricinoleate, span 20	Development of food-grade delivery system stabilized by solid particles (for Pickering emulsions) with good emulsifying properties, based on bacterial cellulose	68
CN113368049A	Edible fat, sodium caseinate, phospholipids	Development of food-grade fat crystal nanoparticles (for Pickering emulsions) with good emulsifying properties	69
CN109793836A	Black tea extract, nano-selenium	Development of food-grade solid particles (for Pickering emulsions) with good emulsifying properties and biological activity	70
CN113208120A	Nano-citrus peel dietary fiber	Development of food-grade solid particles (for Pickering emulsions) from natural biological waste (citrus peel) with good emulsifying properties	71
CN113197300A	Soy protein isolate solid particles	Development of food-grade solid particles (for Pickering emulsions). The patent claims eco-friendly characteristics and green technology usage	72
WO2021008172A1	Crystalline starch ( $\nu$ -type)	Green preparation method of food-grade crystalline particles (for Pickering emulsions). The patent claims eco-friendly characteristics	73
CN113367323A	Lignin/silicon dioxide nanoparticle	Green preparation method of food-grade solid particles (for Pickering emulsions) based on hydrolysis of lignin, silicon and chitosan. The patent claims eco-friendly characteristics	74
CN111820398A	Soy protein isolate nanoparticles	Development of food-grade solid particles (for Pickering emulsions) based on soy protein isolate, with good emulsifying properties	75
CN111808301A	Zein particles, chitin nanofibers	Development of food-grade solid particles (for Pickering emulsions) based on zein particles and chitin nanofibers, with good emulsifying properties	76
CN112515169A	Carrageenan, chondroitin sulfate, whey protein, bovine serum protein, casein, egg white protein	Development of food-grade emulsions stabilized by natural substances: Protein and polysaccharide. The method does not use synthetic surfactants, and the patent claims a concern with materials safety	77





Table 2 (Contd.)

Patent	Stabilizer(s)	Application	References
CN114259467A	Modified egg white protein nanoparticles, polyglycerol ricinoleate, gelatin	Development of food-grade emulsion stabilized by natural substances and with a lower concentration of synthetic emulsifiers	78
CN110368322A	Whey protein, pectin, soybean lecithin, span 80, polyglycerol polyricinoleate	Development of food-grade emulsion stabilized by whey protein, pectin. The patent claims eco-friendly characteristics	79
CN112120212A	Hyaluronic acid-based modified gliadin nanoparticles	Development of food-grade solid particles (for Pickering emulsions) with good emulsifying and stabilizing properties	80
CN111317142A	Glycerin, tannic acid, sodium alginate, polyglycerol ricinoleate, biosurfactant triterpene saponins	Development of food-grade emulsion stabilized by natural substances	81

docosahexaenoic acid in the small intestine was higher when introduced to the food matrix (omelette) encapsulated by a Pickering emulsion.<sup>108</sup> Similarly, He *et al.* (2023) conducted an *in vitro* study to evaluate the bioaccessibility of unencapsulated and encapsulated capsaicin, both in an O/W emulsion and in W/O/W. According to the results obtained, the previous statement is echoed, since the parameter in question increased considerably with encapsulation. Moreover, in this specific case, the values obtained for the W/O/W emulsions were even higher than those for O/W.<sup>109</sup>

Yet on the paper of He *et al.* (2023), by conducting an *in vivo* study in mice, it was observed from the analysis of the histological sections of the stomachs of these animals, that the gastric mucosa did not suffer changes when capsaicin was administered through multiple emulsions. Damage, however, was found when the substance was merely dissolved in corn oil and administered. The witnessed reduction of gastrointestinal irritation through encapsulation demonstrates that the technique addressed might be used to alter sensory characteristics, mainly through multiple emulsions.<sup>109</sup>

Despite their contributions to bioavailability knowledge, *in vitro* studies have limitations that need to be acknowledged and therefore may not translate to practical applications. Static *in vitro* digestion methods like INFOGEST simplify the digestion process – a very complex process – and do not simulate physiological interactions accurately. They maintain constant conditions like pH which do not reflect the natural human body variations, especially during gastric digestion. Additionally, in this case the enzyme activity is fixed across different phases, and the substrate type and concentration are not evaluated. Static models also do not take into account anatomical variations and their singularities, so precision is compromised. These factors might limit the usefulness of *in vitro* studies when it comes to making conclusions about digestion and bioaccessibility in real life.<sup>110</sup> To better illustrate this topic, it is viable to consider the study by Salvia-Trujillo *et al.* (2017). The formulation in being discussed, an O/W emulsion developed for the encapsulation of vitamin D<sub>2</sub>, was tested both *in vitro* and *in vivo*. When evaluating the relationship between droplet size and the bioaccessibility of the cited vitamin, it was noted that smaller droplets had better results in the *in vitro* tests, while larger droplets were more effective in the *in vivo* studies,<sup>111</sup> which precisely demonstrates the limitation of this type of test.

However, it is crucial to say that the results of *in vitro* tests can be extremely valuable, since there are also positive correlations between the results of these tests and real-world scenarios. The literature indicates, however, that more studies are needed on the outcome of this type of test and how it relates to *in vivo* tests.<sup>112</sup>

Thus, it can be inferred that, with the advancement of research, it will be possible to better analyze how this scenario is linked to practical applications when taking into account effectiveness. In terms of cost and time, *in vitro* tests are advantageous over real applications, such as *in vitro* studies in human and animal cells.<sup>112</sup> Therefore, they display greater potential for scalability, especially for preliminary studies.<sup>113</sup>

Among the organoleptic properties that can be changed with the use of encapsulation in multiple emulsions, in the context

Table 3 Patents concerning 3D printing and their applicability

Patent	Stabilizer(s)	Application	References
CN110692800A	Cod protein	Development of a food-grade 3D printing ink stabilized by protein	82
CN114847469A	Sea bass protein microgel particles	Development of a “clean label” food-grade 3D printing ink stabilized by protein, which might be used as a nutrient delivery system	83
CN115245203A	Casein, soybean protein, whey protein, pea protein, peanut protein	Development of a food-grade 3D printing ink stabilized by protein	84
CN115606788A	Burdock cellulose nanocrystal-chitosan	Development of a food-grade emulsion suitable for potential use as a 3D printing ink	85
CN113040369A	Cod protein	Development of a food-grade 3D printing ink stabilized by protein, with freeze–thaw stability	86
CN115746082A	Cod protein	Development of a food-grade emulsion suitable for potential use as a 3D printing ink, with freeze–thaw stability	87
CN114573832A	Chitosan hydrochloride	Development of a food-grade emulsion suitable for potential use as a 3D printing ink, stabilized by a non-toxic, biodegradable substance with good biocompatibility	88
CN115336760A	Functional proteins (lactoferrin), hydrophilic polyphenols (epigallocatechin gallate), anionic polysaccharide ( $\kappa$ -carrageenan, low-methyl ester pectin, sodium alginate)	Development of a food-grade emulsion suitable for potential use as a 3D printing ink	89

Table 4 Patents concerning edible coatings and films

Patent	Stabilizer(s)	Application	References
WO2019039947A1	Polyoxyethylated sorbitan, polyoxyethinlated sorbitan monostearate, glycerol, alginate acid	Development of an edible coating to enhance stability and organoleptic properties of fresh or minimally processed products, such as fruits, fresh cereals, juice, and vegetables. The patent claims eco-friendly characteristics	90
WO2020010173A1	Pullulan, glycerol, xanthan gum, locust bean gum, plasticizer	Development of an edible film with antimicrobial activity, which acts as a lipid carrier of the active substance (essential oils, such as cinnamaldehyde, eugenol and/or thymol) to enhance stability of fruits and vegetables	91
CN113575868A	Sucrose fatty acid ester, pectin	Development of an edible coating to enhance organoleptic properties of turtle meat	92
CN114176116A	Whey protein isolate, OSA starch, silicon dioxide	Development of an edible film/coating to enhance stability of duck necks	93
CN110810091A	Tween 80, sodium alginate, polyglycerol ricinoleate	Development of an edible coating to enhance stability and freshness of fruits and vegetables	94

of food, it is intuitive to think about palatability, which in fact is feasible.<sup>47</sup> It is possible, for example, to reduce bitterness of some peptides,<sup>114</sup> or even increase the perception of sweetness<sup>115</sup> or saltiness,<sup>116</sup> maintaining exactly the same amount of sucrose and sodium chloride. Flavor control is not restricted to multiple emulsions though,<sup>47</sup> and can also be achieved through other types of emulsions, such as O/W, W/O and HIPEs.<sup>117</sup> As demonstrated by some patents, in addition to palatability, through encapsulation it is also possible to control the odor of bioactive compounds.<sup>37,54</sup>

Another function that might be obtained through encapsulation is reduction of the lipid oxidation rate.<sup>40</sup> Wang *et al.* (2022) validated this using wheat gluten nanoparticles modified with rice bran as solid particles to form a Pickering emulsion. These colloidal particles possess polyphenols in their structure, and due to this fact, greater oxidation stability to the soybean oil used was shown.<sup>118</sup> However, emulsifiers are not necessarily the only responsible for this behavior. Kumar and Kumar (2020), for example, demonstrated, in a multiple emulsion (W/O/W), that it is possible to encapsulate a substance with antioxidant activity



Table 5 Patents concerning fat mimetics

Patent	Stabilizer(s)	Application	References
CN115633785A	Lipid Pickering fat globules, sodium caseinate, guar gum, xanthan gum	Fat level reduction in whipped cream. The patent claims environmental protection and safety	95
CN115646232A	Sodium caseinate, whey protein isolate, whey protein concentrate, soybean protein isolate, collagen, gelatin	Substituting both partially hydrogenated vegetable oil and saturated fatty acids with the specified proteins	96
CN115812840A	Almond protein isolate	Preparation of low-fat food (specially mayonnaise) and substitution of animal fat. The patent states that the protein used is green, safe, easy to obtain, and low-cost	97
US2022015382A1	Sodium caseinate, proteins, hydrocolloids, amphiphilic polymers	Reducing fat levels in food without adverse effects on organoleptic properties	98
CN112314714A	Solid fat particles	Reducing fat levels in food without compromising organoleptic properties, while also improving lipid digestibility and shelf life	99

to increase the stability of the oil phase. In this case, the berry extract of *Murraya koenigii* was encapsulated in the aqueous phase that is internal to the oil droplets, and then the mentioned effect was observed. The study also showed that when comparing the oxidation results for the encapsulated and non-encapsulated extract, although the antioxidant activity is exhibited in both cases, the activity was prolonged in the first condition.<sup>119</sup>

Considering the possibilities introduced, it is often possible to obtain them concomitantly. Odor and taste are usually associated, as noted in a previously addressed patent.<sup>37</sup> But it is also possible to increase the stability of the encapsulate, while improving the parameters of solubility, palatability and odor.<sup>54</sup>

From Table 1, it is clear that many patents use biopolymers and natural substances as stabilizers for encapsulation. Specifically, as shown in Fig. 2, among the analyzed formulations, 7 are classified as natural only, representing 28% of the total. Following this, 2 formulations are categorized as natural modified only, making up 8% of the total. In contrast, 6 formulations are identified as synthetic only, comprising 24% of the overall count. Notably, there are 10 mixed formulations, which account

for 40%, that represents blends of natural substances with synthetic and/or natural modified components. However, there is a significant part of patents that rely on synthetic emulsifiers like tween 20, tween 80 and span 80 to accomplish this job. The dependence on these elements creates worries about probable effects on environment and health,<sup>120,121</sup> and this highlights the importance of finding more sustainable options. Even though biopolymers and natural ingredients are widely used in patents for encapsulation, there is still not enough focus on sustainability or green characteristics in most of them. This brings attention to a significant requirement for more focus on eco-friendly methods in encapsulation technologies.

Nanoencapsulation using nanoemulsions shows a dynamic balance between thermodynamic stability and sensory features. Yet, from the thermodynamics viewpoint, microencapsulation seems to provide more stability compared to nanoencapsulation.<sup>122</sup> Another possible disadvantage of emulsion nanoencapsulation might be its susceptibility degradation to environmental changes.<sup>123</sup> These are reminders of how critical it is to have precise preparation and storage conditions for maintaining the effectiveness and stability of nanoencapsulated products.

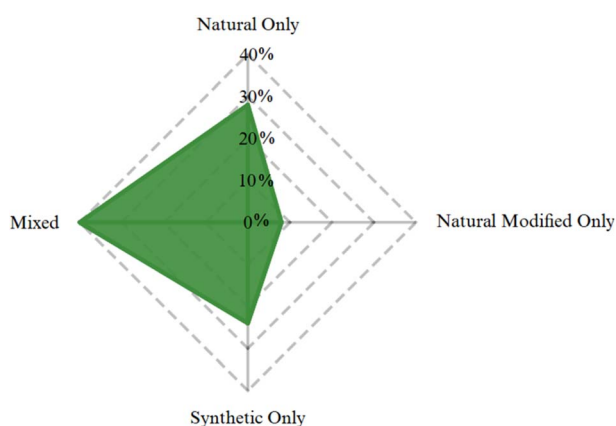


Fig. 2 Distribution of natural and synthetic substance usage in emulsion encapsulation.

### 3.3 Contemporaneity in the stability of emulsions

The multidimensional kinetic stability attributed to an emulsion is established by specific parameters, which are related to its electrostatic, steric and rheological characteristics. Therefore, the maintenance of such criteria is performed through the use of different stabilizing classes, especially emulsifiers and texture-modifying substances.<sup>8,19,124,125</sup>

Concretizing the aforementioned idealization in the context of the food industry, however, clashes with multiple variables. Among them, it is possible to mention the eventual need to meet specific types of diet; the concern of the toxicity of substances, and the minimization of the environmental impact associated to the use of sustainable resources, given the future panorama of society regarding population growth and the



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consequent increase in demand for food.<sup>126,127</sup> Therefore, in order to achieve kinetic stability, there is a need for innovations that mainly comprise both obtention of stabilizers and their respective functional use. This scenario is even better impacted if based on the ideals of circular economy.<sup>128</sup> The high incidence of this type of innovations in patents corroborates the relevance of green chemistry in the development of innovations. Among the parameters observed, it is possible to highlight the preoccupation of patents with the maintenance of the environment,<sup>58</sup> the relevance of using natural and low toxicity substances,<sup>59</sup> and even the implementation of green production methodologies.<sup>73</sup>

As previously mentioned, emulsifiers are important protagonists in providing kinetic stability to emulsions, as they are responsible for reducing the interfacial tension between immiscible liquids. However, it is important to emphasize that their application must be carried out in a quantitatively rational way. For instance, when surfactants are used excessively, it can lead to flocculation, a physical instability that is naturally undesirable.<sup>129</sup>

The term  $\rho_1$  presented in eqn (3), that refers to the density of the oil droplet in an emulsion, is equivalent to that indicated in eqn (8).<sup>130</sup> By articulating both equations, it is inferred that the abundance of surfactant impacts the emulsions also in terms of gravitational instabilities. This occurs because the rate at which these instabilities manifest themselves is postponed by reducing  $u$ . Therefore, the smaller the term  $(\rho_2 - \rho_1)$ , the smaller the value of  $u$ . Consequently, it is necessary to regulate the values of volume fraction of the emulsifier layer ( $\phi_s$ ) and density of the emulsifier ( $\rho_s$ ) so that  $\rho_1$  approaches as much as possible  $\rho_2$ . Gravitational instabilities exist whether the value of  $u$  has a positive or negative sign, this being only an indicator of the spatial arrangement of the instability: if it is positive, it indicates creaming, and if it is negative, it indicates sedimentation. This reiterates the importance of using the appropriate amount of surfactant to provide greater stability to the system.<sup>131</sup>

$$\rho_1 = \phi_s \rho_s + (1 - \phi_s) \rho_c \quad (8)$$

where:  $\rho_1$  = oily phase density;  $\phi_s$  = surfactant layer volume;  $\rho_s$  = surfactant density;  $\rho_c$  = nucleus density.

Given the importance previously addressed on the harmonization between the provision of kinetic stability to emulsions and social and environmental guidelines, an important category of amphiphilic substances synthesized by different microorganisms is partially responsible for making this bond possible: biosurfactants.<sup>132</sup> This was noticed in some patents, which used bacterial cellulose for stabilization of emulsions.<sup>61,68</sup> It is indispensable to emphasize that although according to the literature microorganisms are primarily responsible for producing these amphiphilic molecules,<sup>133</sup> they can also be synthesized by animals and plants.<sup>134</sup> Considering the data provided in Table 2, 37.88% of the stabilizers used are of plant origin, followed by 28.79% of synthetic origin. Animal-derived stabilizers make up 21.21%, while microbial sources account for 12.12% (Fig. 3). Therefore, it is possible to infer that there is a trend towards the search for stabilizers of natural origin from sources other than microorganisms, with an explicit interest in those of plant origin.

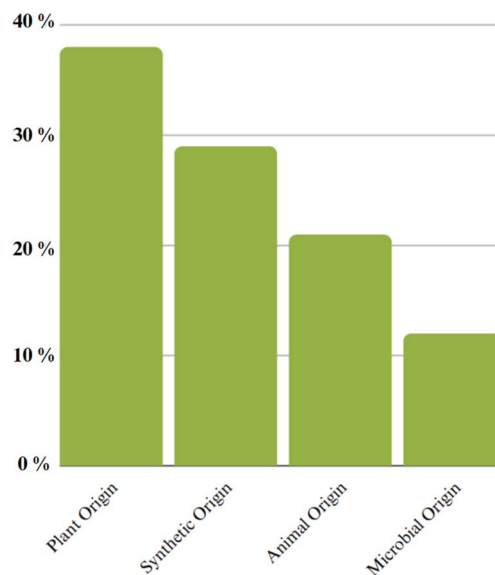


Fig. 3 Examples of biosurfactants and their corresponding sources.

Due to these diverse sources, biosurfactants can be classified based on various parameters with a greater degree of specificity, such as their molecular weight and chemical structure. For instance, microbial biosurfactants include both low molecular weight compounds, like glycolipids and lipopeptides, and high molecular weight compounds, such as polysaccharides and proteins.<sup>135–137</sup> Plant-derived biosurfactants encompass phospholipids, saponins, and protein hydrolysates.<sup>138–141</sup> While animal biosurfactants comprise a variety of substances, including proteins, lipids, and wax.<sup>142</sup> Fig. 4 provides a summary of several examples of biosurfactants, along with their origins.

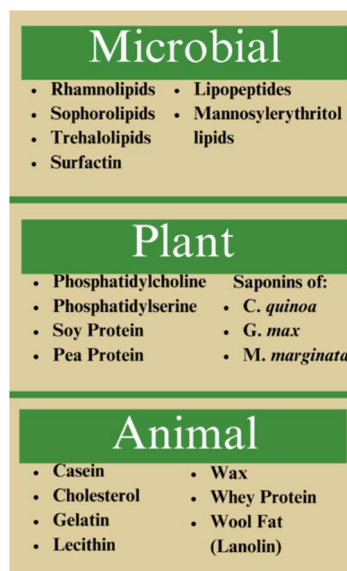


Fig. 4 Distribution of stabilizer origins in novel formulations.



Biosurfactants, in addition to potentially exhibiting good emulsifying capacity, desirable biocompatibility, biodegradability and low toxicity; in the manufacturing process they may be synthesized by microorganisms using as substrate several sustainable sources. Some examples are: low-value crude biomass, as well as sugar and fatty acids, which economically support the synthesis feasibility of these surfactants;<sup>143</sup> oily residues, which are extremely harmful to the environment. In this case, its usefulness is bilaterally worth, as it concomitantly attenuates the pollution problem and the low food availability;<sup>144</sup> and agricultural residues, which fit the same logic previously elucidated.<sup>145</sup>

However, as demonstrated in a case study, this synthesis of biopolymers by food waste faces a few challenges. One of the main issues is the pretreatment of the food waste, which needs to be done efficiently to reduce costs and production time. The transportation of perishable waste can also be expensive, so it is important to find alternatives to minimize these costs. Additionally, downstream processing can represent a significant portion of total expenses. Therefore, choosing the right technologies for this stage is crucial, as it influences both the cost and quality of the final product.<sup>146</sup>

When considering the work of Aro *et al.* (2023), it is noticed that through precision fermentation, microorganisms can also be used to produce surfactants equivalent to those obtained, originally, from animal sources. Based on this methodology,  $\beta$ -lactoglobulin, a protein present in bovine milk, which has emulsifying properties and wide application in food products, was synthesized. This accomplishment is not only beneficial to the environment, given the reduction of carbon emission related to its production,<sup>147</sup> but also to the food industry and society, as with its application, it also meets the consumer market adherent to specific diets. The presence of this specific protein acting as an emulsifier, also synthesized by precision fermentation, was reported by another author in the literature, namely used in ice creams.<sup>148</sup>

As evidenced by the innovations cataloged within Table 2, contemporary industrial practices increasingly integrate green and biocompatible emulsifiers, what reflects certain commitment to sustainable and eco-conscious manufacturing processes. The adoption of such emulsifiers offers a considerable range of advantages, ranging from environmental management to economic viability.

Therefore, to achieve this objective, a considerable number of patents primarily focused on the development of Pickering particles, which were sourced from a diverse array of natural materials including polysaccharides, fibers, novel lipid particles and proteins. These colloidal surfactants of natural origin are not only a significant alternative to synthetic emulsifiers but often also to traditional biosurfactants, primarily due to their greater effectiveness in providing stability to the system.<sup>149,150</sup>

The mechanism of Pickering emulsion formation is a key factor that explains the aforementioned increased stability. To understand this mechanism, it is important to first consider the interface between two immiscible liquids. Unlike an emulsion with classical amphiphilic surfactants, where the hydrophobic and hydrophilic portions of the surfactant organize and adsorb

due to specific affinity between these interfaces, thereby reducing the interfacial tension; in the case of Pickering emulsions, colloidal particles, which are essentially small solid spheres, partially wet in each of the two immiscible liquids with a specific contact angle ( $\theta$ ). Therefore, when  $\theta = 0^\circ$ , the colloidal particle is fully wetted by the aqueous phase, and when  $\theta = 180^\circ$ , the colloidal particle is fully wetted by the oily phase. Thus, in general, in these cases there is no emulsifying effectiveness, which highlights the importance of colloidal particles being partially wetted by both liquid phases. A  $\theta = 90^\circ$ , consequently, is considered ideal from a theoretical standpoint in terms of stability. However, it should be noted that for the range of angles mentioned above,  $\theta < 90^\circ$  applies to O/W emulsions, and  $\theta > 90^\circ$  to W/O emulsions.<sup>151</sup>

This mechanism generates considerable stability in the emulsion through steric hindrance. Moreover, once adsorbed at the interface, removing these particles is energetically costly. This parameter can be inferred through eqn (9), which returns the energy values required for this removal. As explained by Bai *et al.* (2021) and Li *et al.* (2022), generally, since the movement of the particles specifically tied to thermal energy has a significantly lower value compared to the desorption energy, removing the particles from the liquid-liquid interface becomes difficult, and therefore, their adsorption is considered irreversible.<sup>19,152</sup>

$$E = \pi r^2 \gamma_{w-o} (1 - |\cos \theta|)^2 \quad (9)$$

where:  $E$  = desorption energy;  $r$  = colloidal particle radius;  $\gamma_{o-w}$  = oil-water interfacial tension.

An interesting feature identified in patents that can be attributed to emulsions when using colloidal surfactants is the provision of functionality. In a specific case, a thermo-responsive emulsion was developed.<sup>60</sup> But it is also possible, for example, to create nanoparticles responsive to light incidence,<sup>153</sup> pH variations,<sup>154</sup> temperature,<sup>155</sup> and even, concomitantly, pH and temperature.<sup>156</sup>

It is noteworthy to mention that Pickering particles commonly originate from raw materials usually undergo chemical and physical modifications. Even though these modifications are usually singular to each situation, similarities arise in how Pickering fat particles are made. Normally, a chosen fat is stirred up, heated and then cooled down again, all done with water and certain surfactant present, and this way it results in a suspension of fat particles. The surfactant used for this process might come from synthetic sources such as tween 20, tween 40 or tween 60, or could use natural alternatives like sodium caseinate, according to previous studies. Differences in the parameters for heating and cooling, speed of agitation, and time duration are usual.<sup>59,69</sup>

Moreover, even though emulsifiers of biotechnology origin were reported (bacterial cellulose), they were relatively less common compared to other kinds, accounting for 12.12% (Fig. 3). However, as they were reported in the form of solid particles, they are still considered Pickering particles.<sup>61,68</sup> It is important to state that the applicability of biotechnology encounters important difficulties because methods and processes for purification are complex, which increases the



overall cost. However, the employment of agricultural residues and other lower cost substrates can lower the general production cost.<sup>157</sup>

Texture modifiers, in this context understood mainly by biopolymers, are responsible for the rheological modification related to the viscosity of emulsions. Even though biopolymers might be used as biosurfactants, by using them as texture modifiers the viscosity of the continuous phase is increased, which increases the steric hindrance of the system and reduces the Brownian movement of the droplets. The sum of these phenomena results in greater stability, not only in relation to the increase in droplet size, but also in relation to their disposition.<sup>19,158</sup>

The relationship between viscosity and stability is corroborated specially when it comes to the gravitational instabilities. According to eqn (3), it is noticed that by increasing the viscosity ( $\mu$ ), the terminal velocity ( $u$ ) of the droplets decreases, thus delaying the creaming and sedimentation rate.<sup>15,16,159</sup>

Among the various biopolymers used in food emulsions, a very important class are cellulose derivatives, which can be obtained from various sustainable sources. In the work of Costa *et al.* (2022), it was demonstrated the possibility of extracting cellulose and derivatives from textile residues, whether from mixed fabrics or composed of pure cotton. After cellulose extraction by acid hydrolysis, in the study two types of cellulose derivatives were also obtained: carboxymethylcellulose in its sodium salt form (NaCMC) and cellulose acetate (CellAcet).<sup>160</sup> Cellulose derivatives can also be obtained from various agricultural residues, such as carboxymethylcellulose extracted from corn husk,<sup>161</sup> and microcrystalline cellulose (MCC) obtained from grain leftovers, residues of Chinese herb, and also from multiple stalks, namely those present in sweet sorghum and Jerusalem artichoke.<sup>162</sup>

The role of microorganisms is also of great importance for the production of biopolymers, with xanthan gum being the most expressive in this context. Agricultural residues, such as potato crop residues (PCR), can be used in the sustainable production of this biopolymer. As pointed out by Soltaninejad *et al.* (2022), the PCR, after treatment by the organosolv process, underwent enzymatic hydrolysis and then the resulting hydrolyzate could effectively be used by the bacteria *Xanthomonas campestris* for the production of xanthan gum.<sup>163</sup>

A key benefit of the patented innovations is seen in how they reduce the amount of emulsifiers needed for effective industrial use. This decrease helps to save resources and reduce potential environmental effects associated with excessive chemical use.<sup>59</sup> Additionally, using green emulsifiers from natural substances like citrus peel (an organic waste) demonstrates ideas related to circular economy and waste valorization.<sup>71</sup>

The cost-effectiveness of some green and biocompatible emulsifiers further accentuates their appeal to industries seeking to optimize operational expenditure. By offering lower costs, low toxicity profiles and eco-friendly attributes,<sup>58,59,62,72</sup> these emulsifiers align, to a certain extent, with contemporary sustainability mandates, thereby enhancing corporate social responsibility initiatives. When green emulsifiers are used, it gradually reduces the reliance on synthetic options.<sup>59,62</sup> In some

cases, this facilitates the complete elimination of synthetic emulsifiers, promoting a shift towards more sustainable innovation methods.<sup>58,61</sup> This change could play an important role in lessening the environmental impact of industrial activities while also boosting new ideas within green chemistry as well as biotechnology.

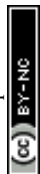
In spite of the benefits these advancements bring, an important negative aspect results from how synthetic substances, particularly emulsifiers, linger in formulations. Despite the aim for greener practices, some industrial processes might still need a reliance on artificial emulsifiers.<sup>59,62,79</sup> This highlights how complex it can be to completely switch over to green options within current industrial systems.<sup>164</sup>

Moreover, the fact that source materials can be renewed is not an assurance of sustainability in their use. Even though green emulsifiers might come from renewable sources,<sup>66</sup> it is very important to consider a broader sustainable cycle, which includes parameters such as how they are cultivated, methods used for getting resources, logistics for transportation and finally what happens at end-of-life disposal stage.<sup>165</sup>

### 3.4 3D Printing

Diverse conceptual markers are responsible for fragmenting the industrial revolutions, and among them, it stands out the existence of unique technologies linked to their historical moment. However, the unique characteristic subordinated to such technologies often has its source on the improvement of pre-existing techniques. This narrative can be seen by comparing the primitive principle of “mass customization” (MC) for Industry 4.0 and its progress toward “mass personalization” (MP) for Industry 5.0.<sup>166</sup> Both MC and MP have a market and production objective centered around the creation of customizable goods. The key difference between them is attached to the degree of product customization for the customers, which is higher in the MP. In this context, the additive manufacturing or 3D printing is of great importance, because it allows the metamorphosis of the materially intangible, formulated in computers, into the tactile form. This mechanism is crucial for both MC and MP, because 3D printing enables the synthesis of highly customizable materials with and increased flexibility, despite the unique characteristics of each process. As this allows linking digital design to actual production, scalability and customization of products to individual preferences are benefited. Therefore, 3D printing is the key to moving from mass customization to true mass personalization.<sup>167,168</sup>

It is evident that such materialization, which can be performed for food production, depends on the use of food-grade inks. Their effectiveness in the printing process is expressed specially by rheological parameters, particularly given by an adequate viscosity and the existence of shear thinning behavior. Emulsions used for 3D are fluids. That is, they do not have a specific shape, are prone to modifications under external pressure, and have the ability to flow naturally. There are two basic types of fluids: Newtonian fluids, which exhibit a linear relationship between viscosity and shear stress, and non-Newtonian fluids. Non-Newtonian fluids can display two



different behaviors: shear thinning, where viscosity decreases as shear stress increases, and shear thickening, where viscosity increases with shear stress.<sup>169</sup> This is why it is important for food-grade inks to display shear thinning behavior; because as shear stress is applied, they become less viscous and can be printed easily, as it will flow more effectively through the 3D printer nozzle tip.<sup>170</sup>

HIPES might display the desired rheological aspects aforementioned, hence they may be used as 3D printing inks.<sup>171</sup> All patents found about food-grade 3D printing inks, present in Table 3, were obtained by the methodology applied for HIPES specific search, which highlights the relevance of its applicability for this purpose. In these emulsions, with regard to their stabilization, it was observed that mainly proteins were used, but as well other substances of natural origin, such polysaccharides in association with other biopolymers.

The Pickering HIPE elaborated by Liu *et al.* (2023), which used zein, tannic acid and sodium alginate complexes to stabilize the emulsion, was effectively developed as an ink for the process discussed here. As demonstrated, it is important to obtain emulsions that exhibit shear thinning behavior, so that there is compatibility of the material with the extrusion speed, *i.e.*, the length of the extruded layer divided by the extrusion time exerted by the additive manufacturing machines. The amount of surfactant used is also an important indicator of printing quality. It was described that with an increase of colloidal particles, within the range tested, it was possible to obtain samples with higher resolution and better structure.<sup>172,173</sup> It is pertinent to observe that even though Pickering HIPES have higher viscosity than classical HIPES,<sup>174</sup> their printing effectiveness can still be assessed empirically through their rheological properties. Then, the underlying principle that allows them to support printing remains consistent from this perspective.

In the article made by Cen *et al.* (2023) it is shown that food-grade inks are not limited to HIPES. This is corroborated by the fact that among the different emulsions tested, the most stable and viable one had a  $\phi = 65\%$ , and under this particular condition, it exposed better shear thinning behavior when compared to other  $\phi$  values, which would variate at 5% in the spectrum of 50% to 70%. Considering the emulsion composition, it was not even possible to produce and test a HIPE, since at  $\phi = 70\%$ , phase separation was already reported.<sup>175</sup> Yet on food-grade inks stability, it was perceived through patents the concern in the development of compositions that can be submitted to freeze–thaw processes.<sup>87,88</sup>

Food-grade inks made of emulsions, once formed, may be applied in various circumstances of pertinent interest to the food industry. Building upon them, the development of structurally more complex products has become feasible. Tay *et al.* (2023), for example, have developed an emulsion-based salmon mimetic consisting of plant-derived proteins. The printed food structurally mimics the myomere and myosepta of salmon, utilizing red lentil protein and pea protein for the emulsions, respectively.<sup>176</sup> Similarly, in the work of An *et al.* (2023), the authors demonstrated, with the production of edible

decorations, the possibility of printing structures of considerable detail through emulsions.<sup>177</sup>

A notable feature of emulsions used in 3D printing food inks is their possible impact on sustainability efforts in the food industry. As precise deposition of food materials is made possible by this technology, it can help cut down on wasted foods and promote better use of ingredients. This aspect relates to wider attempts to reduce waste and support accountable resource management.<sup>178</sup>

The approach regarding the efficiency of food waste reduction through 3D printing is widely discussed in the scientific literature. Burke-Shyne, Gallegos, and Williams (2020), through interviews, developed a qualitative study on 3D printing exclusively related to the food sector and identified four main guiding themes; among them, sustainability, specifically linked to the reduction of food waste.<sup>179</sup> Additionally, the valuable issue of sustainability tied to food waste reduction through additive manufacturing is reported, for example, through the use of fruit and vegetable residues as raw materials for the production of inks.<sup>180</sup> It is important to note that this positive impact generated by 3D printing, however, is not only related to the origin of raw materials but also to the printing process itself and its relation to logistics and management purposes. To better discuss this, Dhir *et al.* (2023), in their quantitative work related to data collection in industries of different sizes, inferred in the significant reduction of waste generated through additive manufacturing.<sup>181</sup> According to Ramundo *et al.* (2020), this reduction is due to some intrinsic factors of 3D printing, such as accurate production scale and greater efficiency in raw material management.<sup>182</sup>

As referenced in Table 3, the information shows that emulsions used for stabilizing 3D printing food inks can be stabilized by non-toxic and biodegradable substances, which was directly mentioned in a patent.<sup>88</sup> By using this method to stabilize 3D printing inks, not only printing safety is granted, but environment friendly principles are followed. Moreover, the emulsion character of these 3D printing food inks opens up the possibility of creating nutrient delivery systems within printed food products.<sup>83</sup> Thus, this innovative approach to food production highlights the multifaceted benefits of emulsions in 3D printing.

The technological advancement expressed through the patents present in Table 3 is considerably significant regarding the use of 3D printing in the food industry. The reduction in the use of synthetic surfactants, for instance, is grounded in the use of cod protein, which acts as a natural stabilizer, culminating in the development of a technology that is not only safe but also characterized by sustainability and nutritional richness.<sup>82</sup> The prominence of proteins in the technological advancement permeating 3D printing in the food sector is also observed in other patents, such as those employing sea bass proteins and other animal and plant sources. The sea bass protein emulsion, for example, exhibits enhanced stability and the ability to protect bioactive compounds through an innovative combination of thermal and enzymatic crosslinking, establishing a new paradigm for the development of high-performance food materials. Furthermore, it can be observed that the developed



emulsions possess characteristics that are not only ideal during extrusion but also self-supporting, thus enabling prolonged storage without compromising the structural integrity of the printed food.<sup>83,84</sup>

The commitment to nutritional quality is based on technological advancements applied not only to the use of proteins themselves but also to other substances. The combination of burdock cellulose with chitosan, for instance, not only proposes an effective method for delaying lipid digestion but also promotes the creation of healthier food formulations.<sup>85</sup>

Printing food with 3D technology also has its own drawbacks and limitations. One of these is the slow speed of the printing process. In comparison to traditional methods, 3D printing uses a technique known as layer-by-layer deposition, which can be time-consuming. This limitation in speed might prove problematic in certain scenarios, especially within industrial environments where fast operation and high productivity are crucial factors. However, the characteristic of 3D printing food requiring significant time can also be seen as an advantage. This aspect creates a special market where people find pleasure in observing their food being printed through 3D technology. The fascinating show of seeing complex designs of food come to life one layer at a time can add value to the whole eating experience, turning it into entertainment. This type of technology, however, seems to not be appropriate for particular groups of consumers, especially older people, individuals with dysphagia, and those with disabilities.<sup>183</sup>

Although additive manufacturing is associated with an overall reduction in carbon footprint, both in relation to the printing process itself and transportation issues,<sup>184</sup> and the viability of alternative production methods, such as cultivated meat printing, which can reduce the total carbon footprint by up to 96% when compared to traditional methods,<sup>185</sup> studies on the environmental impact of 3D printing have been developed. Particularly the high energy consumption required by the printing process, mainly due to the time it takes, is linked to controversies, with potential implications for air quality, which may negatively affect climate change and soil quality.<sup>182</sup>

Another concern that needs more attention is the study on nutritional value of food made by 3D printing. Parameters like bioavailability and digestibility, especially for inks based on emulsion, need more focus.<sup>186,187</sup> But there are encouraging findings in the literature, such as a significant improvement in the bioavailability of  $\beta$ -carotene from High Internal Phase Emulsions (HIPEs) used in 3D printing inks, that is especially helpful for elderly individuals.<sup>188</sup> Although these results are promising, more research is needed to better comprehend and improve the nutritional components of 3D printed food.

It is essential to balance the increase in bioavailability of emulsions with the specific needs of the target audience for which they are formulated. This requires considering factors such as diets and distinct groups of people, which enhances the personalized nature of 3D printing. Thus, formulations aimed at meeting the nutritional requirements of athletes, the elderly, and children show great potential, as they allow for adjustments to both the nutritional profile and sensory properties of foods, tailoring them to the particular demands of these groups.<sup>189</sup>

Additionally, a significant obstacle to the widespread usage of 3D printing is its high cost. Usually, traditional methods of 3D printing need costly tools and components that are not feasible for numerous applications because they are too expensive. But there are ongoing attempts to create cheaper options. For instance, Demircan and Özçelik (2023) have created a laboratory scale 3D food printing machine that works using the syringe-pump mechanism. This development considerably lowered costs without compromising on the quality of printing.<sup>190</sup> Therefore, these advancements in cost-effective printing technologies hold promise for expanding the accessibility of 3D printed food products to a broader market.

### 3.5 Edible coatings and films

Although, so far, the panorama of the topic that covers the stability of emulsions has been specifically described, in the food industry these intrinsically unstable systems from a thermodynamic point of view can be used, interestingly, as protagonists in terms of granting stability to different food products. At first glance, this scenario may appear as paradoxical, however, it corroborates the potential effectiveness of the kinetic stability provided to emulsions.<sup>191,192</sup> It is important to highlight what it means to provide kinetic stability to an emulsion. As mentioned earlier, emulsions are inherently unstable. In other words, at some point, they will inevitably separate into phases. Therefore, promoting kinetic stability means the same as decreasing the rate at which phase separation occurs, consequently postponing this inevitable event.<sup>122</sup>

The development of edible coatings and films has emerged as a promising solution when it comes to food conservation, as evidenced by patents cataloged in Table 4. In order to increase the shelf life of numerous foods, such as meat products,<sup>93,193</sup> fruits,<sup>90,91,94,194</sup> and vegetables,<sup>90,91,94,195</sup> it is plausible to use coatings and films produced from emulsions. In general, they demonstrate functional properties, providing extra protection to what is being involved. As demonstrated by a patent, coatings and films can also be used for other purposes, such as to enhance the taste of certain foods. For a particular case, the validation of the developed coating usage included a taste assessment through experimental methods. Mangos coated the formulation underwent sensory evaluation by 15 trained judges. Coated mangoes achieved a perfect taste score of 5.0, attributed to enhanced fruity aroma, while control had a score of 4.8. At 14 days, coated mangoes retained a high score of 3.9, while control samples decomposed. These results highlight effectiveness of coatings in preserving taste and extending shelf life, validating their application in food packaging.<sup>90</sup> The discrimination between coatings and films is essentially based on the method of application and formation of such systems, and the formulation of both can be even equivalent, as described by Wigati *et al.* (2023).<sup>196</sup>

**3.5.1 Emulsion-based edible coatings.** When emulsions are used as coating systems, they are applied directly on the surface of the food that is desired to envelop, through different methods, such as immersion (Fig. 5a) and spraying (Fig. 5b).<sup>197</sup>



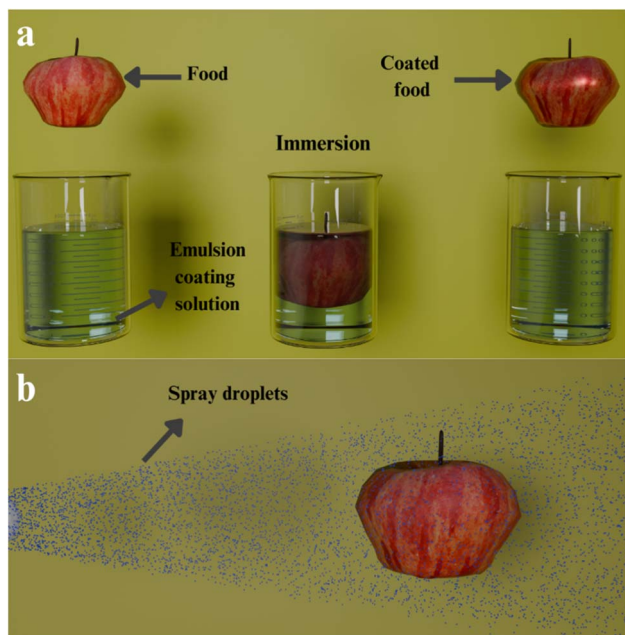


Fig. 5 Illustrative demonstration of immersion and spraying techniques in food coating, using apples as examples (a): immersion technique; (b): spraying technique). Created with Blender.

Afterwards, the drying process is employed, enabling the layer formation. According to Trinh *et al.* (2022),<sup>198</sup> for a single formulation, the drying temperature may take place at different degrees for distinct foods, such as, in this case, at 70 °C for bananas and strawberries, and at room temperature for apples.

Considering the various substances that may be used to produce coatings, essential oils (OE) deserve to be highlighted, especially due to their antimicrobial and antioxidant activities.<sup>199</sup> The results obtained by Kazemeini *et al.* (2021), regarding the effectiveness of *Trachyspermum ammi* essential oil (TAEO) in coatings for the protection of turkey fillets against the bacterium *Listeria monocytogenes*, corroborate the superiority of nanoemulsions for this function. It was observed that, especially on day twelve, the count of *Listeria monocytogenes* was lower in the coatings formulated by nanoemulsions than in those by emulsions of larger droplet size and with referent

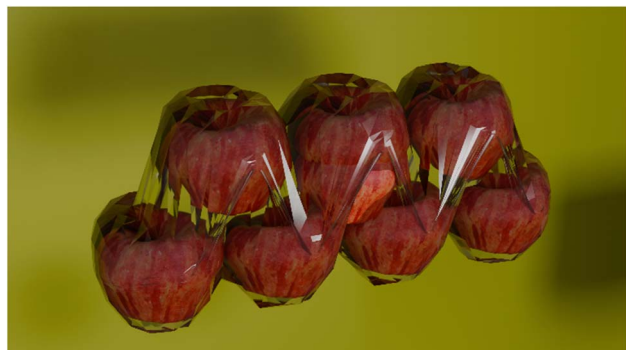


Fig. 6 Illustrative demonstration of food (apples) wrapped by emulsion-based films. Created with Blender.

composition, that is, both for alginate 3% containing 0.5% TAEO, and for alginate 3% containing 1% TAEO.<sup>200</sup> Considering specifically the patents in Table 4, it shows clearly that essential oils have been used. This involves cinnamaldehyde, eugenol, thymol,<sup>91</sup> tea tree essential oil as well as cinnamon essential oil.<sup>93</sup> It is interesting to see how a combination of cinnamon essential oil and salicylic acid was used in one case; both substances serve an antimicrobial purpose.<sup>94</sup> Moreover, a mix of various chemically natural antioxidants was also shown.<sup>90</sup> The presence of this variety of substances makes it clear that encapsulated materials in coatings and films are not only limited to essential oils though, and highlights the possible use of different materials for similar purposes.

**3.5.2 Emulsion-based films.** On the other hand, the film formation process is usually independent of the food desired to be wrapped (Fig. 6). With this technique, it is feasible to obtain materials with good biodegradability that may be considered better alternatives to the synthetic polymers traditionally used, such as plastics.<sup>196,201</sup> The formation of films may arise by different methods, the main ones being solvent casting and extrusion.<sup>197</sup>

By consulting the literature, it is observed that the incorporation of Pickering emulsions in the production of films has been widely used. The work of Xu *et al.* (2023) precisely exemplifies this, in which the emulsion, formulated with oregano essential oil and stabilized by TEMPO-oxidized chitin nanocrystals, was introduced to a glucomannan matrix. Due to the essential oil, it was possible to obtain an active film with antioxidant properties, justified by the presence of phenols and terpenes, and also antimicrobial activity. This last effect was tested and approved specifically against *E. coli* and *S. aureus*, both pathogenic bacteria important in the context of food.<sup>202</sup> Similarly, it was by Fasihi *et al.* (2023) demonstrated the use of a Pickering emulsion in films based on carboxymethylcellulose (CMC)/polyvinyl alcohol (PVOH), with the employment of ginger essential oil (GEO) to make the packaging structure bioactive and protect, specifically, breads. Antioxidant and antimicrobial activities were also observed in the film.<sup>203</sup> It is reiterated, as observed in both cases, the importance of using substances with good biodegradability and low toxicity.

The applicability of films in the food industry, regarding food degradation, is not exclusively restricted to the addressed bioactive functionality. As expressed in the study of Ran *et al.* (2022), emulsions can be used for the development of intelligent films. As for the mentioned work, although there is the undesirable presence of an important value of water vapor permeability, it was possible to develop a film that indicates, colorimetrically, the presence of spoilage in pork meat, upon change in pH.<sup>204</sup>

**3.5.3 Further implications of edible coatings and films.** It is possible to observe that the patents presented in Table 4 related to food preservation have their innovative character grounded and distributed across four different objectives: extending shelf life; reducing food waste; ensuring safety; and reducing environmental impact. Although the primary target of the patents is generally the increase in shelf life of the food involved in films and coatings, the other aforementioned intentions hold significant value. They not only further promote



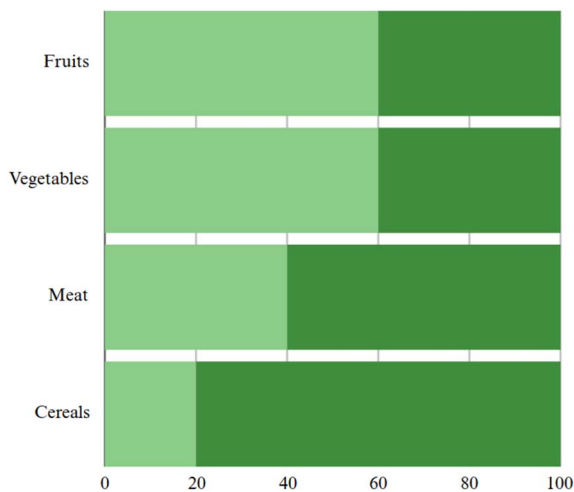


Fig. 7 Incidence of food types in patent formulations for edible coatings and emulsion-based films for food preservation.

the innovative character of the patents but also strengthen their connection to sustainable practices. The encapsulation of antioxidant substances obtained specifically from food waste of fruits, vegetables, and cereals, for example, exemplifies this scenario, as there is a considerable interplay between reducing food waste, lowering environmental impact, and extending the shelf life of food.<sup>90</sup> Additionally, some patents indicated in their scope the importance of using substances properly regulated by oversight and regulatory bodies, ensuring that the innovations not only fulfill their functions but also comply with food safety standards.<sup>91</sup> Substances of low toxicity, which do not harm the environment and are economically viable, were also highlighted, reinforcing not only the practical applicability of the technologies but also signaling a commitment to sustainability and social responsibility, which are essential aspects in the search for innovative solutions in the food sector.<sup>93</sup> Still regarding patents aimed at the development of coatings and films for food preservation, it was observed, as illustrated in Fig. 7, that fruits, vegetables, meats, and cereals were the primary foods targeted for conservation. Among these innovations, most formulations are intended for application to fruits and vegetables (60% each), followed by meat (40%) and, to a lesser extent, cereals (20%). It is important to note that some patents apply to more than one type of product, resulting in overlap across the analyzed categories.

Coatings and films that are based on emulsion present an environmentally friendly substitute to usual plastic packaging. This tackles the worry about environmental damage caused by conventional plastics, which have low biodegradability.<sup>205</sup> A significant detail shown in Table 4 is the fact that coatings and films mainly use biopolymers like alginic acid,<sup>90</sup> pullulan,<sup>91</sup> modified starch, whey protein isolate,<sup>93</sup> and sodium alginate,<sup>94</sup> materials naturally more degradable compared to polymers made from petroleum-based resources. Thus, switching to biopolymers helps lower the environmental impact associated with plastic packaging, reducing the accumulation of non-degradable waste in landfills and natural systems.<sup>205</sup>

The use of edible coatings and films display specific advantages. They may help with energy saving in comparison to usual keeping methods like freezing or controlled atmosphere storage. This decrease in energy usage is aligned with sustainability values as it promotes resource effectiveness and reduces environmental effects linked to high-energy preservation techniques.<sup>90</sup>

Additionally, edible coatings usually have few calories and do not greatly change the nutritional quality of food matrices, making them more attractive for use as a solution in food packaging. This feature keeps the natural nutritional advantages of packaged foods intact, encouraging healthier choices for consumption by people who buy these products. The importance of edible coatings and films goes beyond just thinking about nutrition; it also relates to their part in decreasing loss or waste of food. By extending the shelf life of perishable food products, these coatings contribute to the reduction of food waste at various stages of the supply chain, from production to consumption.<sup>206</sup>

In some situations, the use of coatings and films formulas can make it possible to create end-products without adding organic chemical additives. This method improves their environmental characteristics, as the decrease in chemical components reduce the risk linked with synthetic substances and promotes safer packaging solutions that are environmentally friendly.<sup>90</sup> Therefore, new types of edible coating and films, which focus on natural and renewable elements, provide a way to diminish the reliance on chemicals in making packages, and this contributes towards an industry that is cleaner as well as more environmentally accountable.

Specifically, in one methodology outlined within a patent, the use of an edible coating helps to entirely remove synthetic emulsifiers, particularly polysorbate. As the main methodology relies on polysorbate usage for achieving emulsification, an alternate approach is suggested in the same publication which implies potential for attaining a formulation without the mentioned synthetic surfactant. This progress diminishes environmental consequences while also enhancing the trust of consumers regarding safety, which encourages more acceptance of environmentally-friendly packaging solutions within this field. An interesting method employed in this patent is microfluidization, a cutting-edge technology that utilizes controlled fluid dynamics to manage and create uniform emulsions at the nanoscale level.<sup>90,207</sup>

The drawbacks of bioplastics, in general, include less mechanical strength than normal plastics, which can cause them to be easily physically damaged. Often, they also show higher water vapor permeability, possibly affecting packed items' quality by letting in moisture.<sup>208</sup> There is also a concern about unwanted leakage and interaction of elements from films into food products, which could affect how consumers accept the film because of possible alterations in parameters like taste, smell or general quality.<sup>209</sup> Another issue that is important to be considered is that the transition from traditional petroleum-based plastics to biodegradable films faces further economic and technical challenges. Despite a projected growth in the biodegradable plastics market of 21.7% annually, cost remains



a significant barrier, with biodegradable options like polylactic acid (PLA) priced at \$1.9–2.0 per kg compared to \$1.2–1.3 per kg for conventional plastics. Consequently, market adoption is still slow.<sup>210</sup>

Similarly, coatings applied to food surfaces could also bring some disadvantages. They might change the taste, smell and other sensory attributes of the food.<sup>211</sup> Furthermore, when consumers eat the food that has a coating, they might also consume this layer and possibly have allergic reactions. To exemplify this, the proteins found in Whey Protein Isolate (WPI) are  $\alpha$ -lactalbumin ( $\alpha$ -LA) and  $\beta$ -lactoglobulin ( $\beta$ -LG). Although WPI has many advantages,  $\alpha$ -LA and  $\beta$ -LG can cause allergies in certain individuals.<sup>212</sup> A patent introduces an edible coating made from WPI, demonstrating its widespread use in food-related innovations. Notably, this edible coating, developed for protecting duck necks,<sup>93</sup> raises significant concerns for individuals with allergies. Even if someone can tolerate eating duck meat, the addition of WPI in the coating makes it not suitable for those allergic to  $\alpha$ -LA and  $\beta$ -LG. This limitation could restrict access to and usage of the product among certain consumer groups with allergies.

### 3.6 Fat mimetics

When consulting in the literature the relationship between the consumption of different types of fats and their impact on human health, it is perceived that the topic is endowed with plurality. In the review work prepared by Kim *et al.* (2021), a statistically significant relationship was observed between the exacerbated consumption of saturated fats, *trans* fatty acid, and health problems. In this instance, the consumption of mono-unsaturated and polyunsaturated fats was considered healthier alternatives.<sup>213</sup> Corroborating the complexity of this topic, in the cohort study of Otto *et al.* (2012), it was presented that the association between saturated fats consumption and cardiovascular diseases depends considerably on the origin of such constituents. While the consumption of saturated fats of meat origin showed a considerable tendency to the manifestation of cardiovascular diseases, the consumption of saturated fats of dairy origin showed an inverse tendency to this risk, under specific conditions.<sup>214</sup>

Various studies also point to the relationships between the consumption of *trans* fats and health problems. Among these issues, there is a notable increase in low-density lipoprotein (LDL) and a decrease in high-density lipoprotein (HDL), which can lead to impaired vascular function, increased systemic inflammation, and a higher risk of coronary heart disease, as well as suggestions of a link to the development of atherosclerosis.<sup>215</sup>

However, some authors present in their studies even greater divergence between the mentioned association. Harcombe *et al.* (2016), for example, after completing their meta-analytical work, found no evidence to validate the relationship between fat intake and congenital heart disease or all-cause mortality.<sup>216</sup>

Considering the hypothesis of the negative relationship between excessive fat consumption and health conditions, it is natural to think about the consequent need to regulate the

nutritional value of foods through this parameter. Fats, on the other hand, play an important role in organoleptic and conformational characteristics of food.<sup>217</sup> Therefore, in order to reconcile this dichotomous situation, fat substitutes might be used, including in the form of emulsions, as noticed in patents present in Table 5. With the changing market demands and search of consumers for healthier and more nutritious foods, the replacement of fats has been widely targeted in recent years. As a result, it is projected that this market will reach \$2.79 billion by 2025,<sup>218</sup> which justifies the relevance of developing new products and scientific research in this area, and consequently, reinforces the importance of emulsions for this purpose.

The terminology that surrounds the classification of fat substitutes is dimensionally considerable to understand the role of emulsions in this context. O'Connor and O'Brien (2016) classify fat replacers into fat substitutes and fat mimetics. The mentioned systematization, which will be adopted for the elaboration of the topic, states that fat substitutes are those substances that, in their singularity, are capable of replacing the fat of interest; while fat mimetics are those more complex substances consisting of carbohydrates and/or proteins,<sup>219</sup> subdivision that might accommodate emulsions.<sup>220</sup> It is noteworthy, however, that there are divergences among authors regarding the terminology used. Some authors refer to such emulsions as fat substitutes.<sup>221,222</sup>

With the objective to produce yogurt with lower fat content, Li *et al.* (2022) used an emulsion gel as a fat mimetic, composed by vegetable oil in the oil phase, and microparticles of whey protein gel, used as an emulsifier. It was possible to reduce the fat content of yogurt from 3.14 g/100 g to 1.50 g/100 g, when compared to the traditional one, made with whole milk powder.<sup>223</sup>

The relevance of emulsions as fat mimetics is also given by the usefulness of multiple emulsions,<sup>98,99</sup> congruence observed in the publication of Eisinaite *et al.* (2017). It was shown that the W/O/W disposition implies in the existence of, necessarily, an aqueous phase (in this case represented by beet juice) dispersedly present within the oil molecule (sunflower oil), which is accommodated in an outermost aqueous phase (whey protein isolate 0.5%). Therefore, when compared to an O/W emulsion, the number of calories present per oil droplet is decreased. At the same time, the possibility of replacing animal fat with vegetable fat in meat-based foods has been exhibited.<sup>224</sup>

In agreement with what was discussed by Kim *et al.*,<sup>213</sup> the study of Silva *et al.* (2018) admitted that the consumption of monounsaturated and polyunsaturated fats is beneficial when compared to saturated fatty acids. Thus, it had its elaboration based on the objective of developing a fat substitute, using a mixture of olive oil, linseed and fish oils for that. However, as these fats molecules are prone to oxidation, the proposal of the authors was the elaboration of a multiple emulsion, in which an antioxidant, namely quercetin, was nanoencapsulated in the external aqueous phase, and another antioxidant was encapsulated in the internal aqueous phase, namely garlic oil. Thus, a multiple emulsion with considerable stability against lipid oxidation was obtained, displaying potential to be applied as



a fat mimetic.<sup>225</sup> It was observed that in patents, although in lower apparent incidence, Pickering emulsions<sup>95</sup> and HIPES<sup>96</sup> could also be used for such purposes.

Considering the aforementioned content, in addition to their role as fat mimetics, emulsions are also widely utilized as carrier systems. For example, the multiple emulsion mentioned of Silva *et al.* (2018) not only replaces traditional fats but also carries antioxidants such as quercetin and garlic oil. This dual functionality is particularly important in the fat replacer context, as these emulsions not only mimic fats but also enable the incorporation and protection of bioactive ingredients.<sup>225</sup> Thus, this dual functionality emphasizes the significance of emulsions as carrier systems in food formulations, as they are able to provide both the sensory attributes associated with fats and the ability to deliver and stabilize functional ingredients, potentially enhancing the overall nutritional profile and stability of the product.

The usage of gel emulsions as fat mimetics also has great potential in emulsified meat products. Li *et al.* (2022) effectively replaced pork fat in sausages through this method. The presence of gel emulsion in sausages, not only reduced fat content, but was also associated with an improvement of the mechanical properties of the sausages, including increase of viscosity at defined shear stress, decrease of loss by cooking, and lower rate of lipid oxidation.<sup>226</sup>

As shown in Table 5, the utilization of biopolymers and Pickering fat particles in these emulsions highlights their environmental safeguards and safety features. Occasionally, these materials are safe to use, easy to get hold of and inexpensive. This makes them even more appealing as an alternative choice for traditional fat sources.<sup>97</sup>

Another advantage of emulsions as fat mimetics is that it becomes possible to improve the digestion of lipids, which leads to better absorption and use of nutrients. Furthermore, the existence of emulsifiers and stabilizers in these formulas can prolong the shelf life of food items. This aids in enhancing safety standards as well as overall quality.<sup>99</sup>

A notable disadvantage of fat mimetics is that fat is considerably important for the body to absorb certain vitamins such as A, D, E and K.<sup>227</sup> Therefore, the reduction or replacement of fat in food formulations by emulsions could potentially lead to decreased absorption of these essential nutrients, thereby negatively impacting the overall nutritional profile of the food product.

### 3.7 Sustainability implications of food emulsion innovations

The Brundtland Report emerged in 1987 under the guidance of the World Commission on Environment and Development, led by Gro Harlem Brundtland. When this report first appeared, it signaled a new era in worldwide discussions as it introduced the idea of sustainable development. This important document set up a way to combine economic progress with protecting nature and achieving fairness among people, changing how communities handle issues related to development. As per the Brundtland Report, sustainable development is “development

that meets the needs of the present without compromising the ability of future generations to meet their own needs”.<sup>228</sup>

With the understanding of how crucial sustainable development is, the United Nations took charge in worldwide actions by setting up Millennium Development Goals (MDGs) first and then changing to 17 Sustainable Development Goals (SDGs). MDGs were put forward in 2000, and the SDGs were introduced in 2015 to expand the plans further.<sup>229</sup>

As expressed in Tables 1 through 5, a notable part of patents specifically claims their innovation as “green”, “environmentally safe”, “clean label” or “biodegradable”. This shows an observable possibility for change towards sustainability in the emulsion sector. Nevertheless, as emphasized by Marques *et al.* (2020), the connection between green chemistry and sustainable development is nuanced. Green chemistry, even when it supports sustainability goals, is not a simple and direct match.<sup>230</sup> Therefore, it cannot be assumed that all patents adhering to green chemistry principles are inherently sustainable.

Some synthetic emulsifiers, in their original form, have been documented to have adverse effects on the environment, animal life, and microorganisms, as highlighted by Johnson *et al.* (2021). These compounds can persist in ecosystems and disrupt natural balances, posing threats to biodiversity and ecosystem health. Furthermore, the degradation products of synthetic emulsifiers may exhibit toxicity, further exacerbating environmental and safety concerns. The widespread use of these substances emphasizes the importance of exploring alternative, more sustainable options in emulsion formulations to mitigate their detrimental impacts on ecological systems, such as biosurfactants.<sup>231</sup>

However, when it comes to the mentioned terms, biosurfactants are not necessarily always superior to synthetic surfactants. Li *et al.* (2017) evaluated the biodegradability and toxicity in soil of three synthetic emulsifiers (modified heterogeneous alcohol ether, fatty alcohol methyl esters of ethoxylate and tween 80) and one biosurfactant (rhamnolipid). In this study, the more favorable values for both parameters were attributed to a synthetic emulsifier, namely fatty alcohol methyl esters of ethoxylate.<sup>232</sup>

With respect to the synthetic emulsifiers utilized within the innovations, the greatest frequency was attributed to polysorbates. Despite the problematic raised previously, the literature indicates that tween 80, a polysorbate, do not share the stated disadvantages, as biodegradability and biocompatibility are attributed to it.<sup>233</sup> However, polysorbates have also been reportedly associated with adverse effects, such as negative repercussions on gastrointestinal integrity<sup>234</sup> and alterations in the mucosal barrier of the small intestine.<sup>235</sup> However, it is very important to comprehend that the significance of biosurfactants in food emulsions and its relationship with sustainability is not solely about their biodegradability or toxicity. These materials are important components of a circular economy because they align with the main goal of reducing waste and using resources efficiently in the food industry by offering renewable options as compared to regular emulsifiers.<sup>236</sup>



SDG 2 aims to end hunger, achieve food security, improve nutrition, and promote sustainable agriculture.<sup>237</sup> Innovations in food emulsions play a crucial role in advancing SDG 2 objectives by addressing various aspects of food security and sustainability. Annually, more than 1 billion tons of agricultural waste is generated, and the prevision is that this value tends to increase as the years go by,<sup>238</sup> and at the same time, the food demand on earth increases.<sup>126,127</sup> Consequently, the utilization of biopolymers in food emulsions, particularly those derived from agricultural waste, enhances availability of food.

It is possible to infer that this practice, along with the potential utilization of other natural substances in food emulsion processes, also aligns with other SDG, such as SDG 9: Industry, Innovation, and Infrastructure,<sup>239</sup> and SDG 12: Responsible Consumption and Production.<sup>240</sup> Furthermore, the development of food-grade emulsions that function as edible coatings and films extends the shelf life and quality of different food products. By preventing spoilage and degradation, these emulsions mitigate food waste along the supply chain, thereby supporting efforts to achieve SDG 2 targets. Additionally, as emulsions contribute to the advancement of 3D printing in food, food waste is also minimized, principally because the concept of zero waste can be further supported through 3D printing.<sup>241</sup>

SDG 3 has the objective to ensure healthy lives and promote well-being for everyone, everywhere, at all ages.<sup>242</sup> Innovation in food emulsions has a big part in achieving SDG 3 goals because it provides solutions that help health and well-being. Encapsulation using emulsions can make food items with better nutrient levels by using biopolymers as stabilizers instead of artificial substances such as emulsifiers, and these include proteins,<sup>44</sup> fibers<sup>71</sup> and other beneficial substances. Also, encapsulation might help in boosting the bioavailability of bioactive substances. When active ingredients become more available for use, their intended physiological effects can be exerted more effectively which leads to greater health benefits.<sup>34</sup> In addition, the quality of functioning as a fat mimetic is crucial for emulsions, as it allows for the reduction of fat content in food products. By mimicking the texture and mouthfeel of fat, emulsions can create healthier alternatives without significantly compromising sensory qualities. This makes them valuable in the development of lower-fat foods that still deliver a satisfying eating experience.<sup>99</sup> So, using emulsions as imitations for fats in food mixtures aids also encourage healthier eating habits and matches with SDG 3.

SDG 13: Climate Action, SDG 14: Life Below Water, and SDG 15: Life on Land, demonstrate the importance of addressing environmental challenges related to climate change, marine ecosystems, and terrestrial biodiversity, respectively.<sup>243–245</sup> In the realm of food emulsion innovations, the development of edible coatings and films holds promise in reducing reliance on plastics and synthetic non-biodegradable polymers. These innovations not only enhance the shelf life of food but can also mitigate the need for energy-intensive methods,<sup>90,246</sup> which contributes to sustainability.<sup>247</sup>

It was evidenced by Cabernard *et al.* (2022) that the carbon footprint associated with plastics is really concerning, as 4.5% of the global Greenhouse Gas (GHG) emissions were linked to them.<sup>248</sup> By minimizing the use of plastics, particularly those

derived from fossil fuels, the advancement in the formulations of biodegradable edible films reduce the release of GHG,<sup>249</sup> and positively aligns to SDG 13. Every year, dolphins, fishes, turtles and other aquatic animals are negatively affected by plastic waste, which culminates not only in a diverse range of injuries for them, but in last instance, results in death.<sup>250</sup> It is estimated that annually, plastic contributes to the death of approximately 100 000 marine creatures, causing a considerable ecosystem disruption.<sup>251</sup> Therefore, edible coatings and films align with SDG 14 by mitigating the adverse impacts of plastics on aquatic ecosystems, including the pervasive issue of microplastic pollution.

In the environment, microplastics can change important parameters such as vegetation development,<sup>252</sup> soil infiltration<sup>253</sup> and microbiota.<sup>254</sup> Then it is possible to infer that by reducing plastic usage, these innovations positively impact SDG 15 by helping to protect terrestrial fauna and flora from the detrimental effects of plastic pollution.

### 3.8 Regulatory perspectives

In the food field, in order for research products to be scalable to the market, something very important is the alignment of the products developed with food regulations, which may vary depending on the country of origin, and as pointed out by Cox *et al.* (2011), different regulatory agencies may differ considerably when classifying certain emulsifiers as food additives. Approval of the use of these substances depends on several tests, such as toxicological ones.<sup>255</sup> Consequently, this might apply to the wide range of innovative colloidal particles developed and reported in patents.

Considering the Food and Drug Administration (FDA) in the United States, for example, if substances used in food are not considered “Generally Recognized as Safe” (GRAS), it is necessary to evaluate and approve them as a food additive.<sup>256</sup> Therefore, while specific substances such as alginic acid extracted from brown algae are considered intrinsically safe for direct use in food, and in this case, even as an emulsifier,<sup>257,258</sup> others, however, need to be evaluated and regulated based on evidence of their safety, which occurs, for example, with the different types of modified cellulose, which are widely used as colloidal Pickering particles.<sup>259</sup> Therefore, it can be inferred that innovations such as those classified under “contemporaneity in the stability of emulsions” will probably need to be articulated uniquely with the regulatory issues of the country in which their application at market level is desired.

This same tangential logic to substances used in food, from a regulatory point of view, also applies to other innovations, but is particularly essential in encapsulation techniques. When taking into account functional foods and/or nutraceuticals, which were considerably involved in innovations linked to encapsulation, it is important that the distinction between food and medicine is explicitly defined, as regulations regarding the substances used in encapsulating these two types of products can differ.<sup>260</sup>

With the removal of partially hydrogenated oils (PHOs), a significant source of *trans* fat, from the list of GRAS substances, and the eventual prohibition of the use of PHOs in



several instances,<sup>261</sup> the issue of fats and their relationship with dietary guidelines is corroborated. The use of fat replacers, including carbohydrate and/or protein-based fat replacers, is sustained by the FDA, and are often classified as GRAS.<sup>262</sup> However, the explicit use of emulsions, which are based on carbohydrates and/or proteins, needs to have its regulation better elucidated. Likewise, it is noteworthy that, regarding 3D printing, although regulatory issues are of paramount importance for the quality and safety of food, and the additive manufacturing market has been growing considerably in recent years, Alami *et al.* (2024) point out that there is still a lack of well-defined regulation on 3D printing in the food sector.<sup>263</sup>

According to Zhu *et al.* (2023), there are potential risks and issues that still need to be studied regarding 3D-printed foods. The lack of regulations surrounding their use, combined with concerns such as the potential release of toxins during the printing process, improper handling of waste generated, and inadequate cleaning of equipment and its relation to microbial growth, as well as the absence of evidence on how the nutritional value of food is affected by the printing process,<sup>264</sup> leads to a context where the development and implementation of well-defined regulations for 3D food printing are necessary. Other authors also discuss additional issues, such as how this could facilitate food adulteration.<sup>265</sup> Therefore, the development of laws and regulations that cover food safety in terms of the potential release of toxic substances from the materials and components of the printers; proper sanitation protocols to prevent microbiological contamination; shelf-life evaluation; possible harmful interactions between printed food matrix and further external components; as well as specific parameters regarding the impact of the printing process on the nutritional value of printed foods, is necessary. Additionally, regulations addressing traceability and authenticity of food products are important to prevent adulteration and ensure transparency throughout the production chain.

The importance of materials considered GRAS extends to the production of edible coatings and films, and the substances encapsulated in the matrix should also have their toxicological profile and safety evaluated.<sup>266</sup> This is related to something previously raised as a possible drawback of this type of innovation, which is the release of substances present in the films/coatings. In relation to this issue, specifically in the case of films, the regulation of the European Union EC 1935/2004 is highlighted, which indicates the importance of these materials, upon coming into contact with food, in general, not altering its composition, changing organoleptic characteristics, or masking the eventual decomposition of the food.<sup>267,268</sup> Some authors report that in this sphere, regulation still needs to evolve, mainly due to the potentially employed nanomaterials in this type of formulation, which at different levels, may lead to the formation of toxic substances.<sup>269</sup>

## 4 Conclusions and future outlook

This review explores the vast potential of emulsions in the food industry through a detailed analysis of patents. It was concluded that innovations in this context seek to promote the encapsulation of substances *via* emulsions, especially for the

formulation of functional foods; the use of natural compounds and the development of new stabilizers, with emphasis on biopolymers; the application of emulsions as food inks suitable for 3D printing, meeting both rheological and, potentially, nutritional requirements; the development of edible coatings and films for food preservation and sensory enhancement; in addition to the possibility of replacing potentially harmful fats by using emulsions as fat alternatives.

When considering the constant change of civilizations and the consequent adaptation of the market to meet these expectations, valuable emerging trends have been identified. The valuable potential of applying HIPES as food inks for 3D printing of food, the increasing use of natural emulsifiers and stabilizers, and the attempt to reduce or even eliminate the use of synthetic substances, corroborate and reflect the tendency of the food industry to adapt. These approaches, based mainly on principles of sustainability and green chemistry, make it clear that emulsions have a vital potential role in the evolution of food science, as they are systems capable of enabling the alignment of sustainable practices and innovation. However, it is worth noting that the use of emulsions in sustainability practices has untapped potential that could be further explored to fully meet all expected sustainable requirements. Thus, although there are signs of characteristics linked to the concept of sustainability in the patents, it is noteworthy that, in general, sustainability itself is not often addressed, so it is expected that more innovations with this focus will emerge.

Consequently, predicted progress can occur in regulatory matters, especially those related to 3D printing using food-grade inks. There might also be advancements in the development of edible coatings and films that incorporate emulsions. Such improvements are foreseen to help with food safety and lengthening shelf life, advancing sustainability goals.

## Data availability

No primary research results, software or code have been included and no new data were generated or analysed as part of this review.

## Author contributions

Felipe Kelmer Müller: project administration; conceptualization; methodology; data curation; formal analysis; investigation; writing – original draft. Fabiano Freire Costa: funding acquisition; project administration; resources; supervision; methodology, writing – review & editing.

## Conflicts of interest

The authors declare that there is no conflict of interest.

## Acknowledgements

We would like to thank the Pró-Reitoria de Pós-Graduação e Pesquisa of Federal University of Juiz de Fora, MG – Brazil for the undergraduate research scholarship.



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