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# Preparation, structure-property relationships and applications of different emulsion gels: bulk emulsion gels, emulsion gel particles, and fluid emulsion gels

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**Abstract**

*Background:* In recent years, there has been increasing interest in emulsion gels, due to their better stability during storage and potential for prolonged intestinal drug release compared to emulsions. There are three kinds of emulsion gels, classified according to their morphological properties: bulk emulsion gels, emulsion gel particles and liquid emulsion gels.

*Scope and approach:* This paper provides a comprehensive review of the mechanisms and procedures of different methods for preparing different emulsion gels and relationships between structures and properties of emulsion gels. The applications of emulsion gels in the food industry are finally discussed.

*Key findings and conclusions:* Different emulsion gels result from different preparation methods, and have various structure-property relationships and applications. Many methods can be used to prepare bulk emulsion gels, involving different matrix materials, processing techniques, and purposes. This can result in different structures of gel matrixes and emulsion droplets and interactions between them, which can influence the structures of bulk emulsion gels and then their mechanical and release properties. On the other hand, extrusion and impinging aerosol methods are two methods for preparing emulsion gel particles, while liquid emulsion gels can be prepared by Pickering emulsions and disrupted gel systems. Rheological, syneresis and swelling properties are critical for gel particle suspensions, while flow behaviour and release properties are important to liquid emulsion gels. In addition, fat replacements and delivery systems are main applications of emulsion gels in the food industry. However, current research has mainly focused on bulk emulsion gels, so further studies on emulsion gel particles and liquid emulsion gels are required.

**Keywords:** emulsion gel; preparation; interaction; structure; property; fat replacer; delivery.

## 1. Introduction

Emulsion gels, also known as emulgels or gelled emulsions (Balakrishnan, Nguyen, Schmitt, Nicolai, & Chassenieux, 2017), are complex colloidal materials in which both emulsion droplets and gels exist (Dickinson, 2012). According to the state of emulsion droplets in gels, structures of emulsion gels can be divided into two categories: emulsion droplet-filled gels and emulsion droplet-aggregated gels (Fig. 1). In emulsion droplet-filled gels, the continuous phase (e.g., protein- and polysaccharide-based gels) forms a continuous gel matrix which can be defined as the support of emulsion gels, and emulsion droplets are embedded in this gel matrix. In emulsion droplet-aggregated gels, emulsion droplets aggregate together and form a network structure, such that the gel matrix is disrupted by the aggregated emulsion droplets. In most cases, the structural state of emulsion droplets is a combination of these two different structures (i.e., partially droplets filled in the gel matrix and partially aggregated droplets), probably owing to the inhomogeneous distribution of emulsion droplets. Moreover, according to the interactions between emulsion droplets and the gel matrix, emulsion droplets can be divided into active and inactive fillers (Fig. 2). Active fillers are mechanically connected to the gel network through emulsifiers by noncovalent and/or covalent bonds, especially when emulsifiers are natural molecules (e.g., proteins, egg lecithin, and soy lecithin); in contrast, inactive fillers have little chemical or physical affinity with the molecules of gel matrix, especially when low molecular weight (LMW) emulsifiers or no emulsifiers are used and matrix materials have weak emulsifying properties (Van Vliet, 1988).

Preparing emulsion gels normally includes two steps: first preparing emulsions and then turning emulsions into gels. During the last decade, emulsion gels have received growing interest, due to their advantages compared to emulsions, such as higher stability during storage, owing to decreased oil movement and oxygen diffusion within the systems (Cofrades, Bou, Flaiz, Garcimartin, Benedi, Mateos, et al., 2017; Corstens, Berton-Carabin, Elichiry-Ortiz, Hol, Troost, Masclee, et al., 2017; Lim, Kim, Choi, & Moon, 2015; Ma, Wan, & Yang, 2017; Sato, Moraes, & Cunha, 2014), controlled and prolonged gastric and/or intestinal drug release because of the protection by the gel matrix (Corstens, et al., 2017; Guo, Bellissimo, & Rousseau, 2017), and practical applications, including overcoming the textural problems caused by lipid particles in food products and mimicking the effect of fat on hardness and water-holding capacity of meat products (Alejandre, Poyato, Ansorena, & Astiasaran, 2016; Brito-Oliveira, Bispo, Moraes, Campanella, & Pinho, 2017).

Bulk emulsion gels, emulsion gel particles and liquid emulsion gels are three kinds of emulsion gels (Fig. 3), which exhibit their own particular properties, due to their different morphological properties. The size and shape of bulk emulsion gels are determined by the emulsion volume and emulsion containers of different shapes and sizes used during the emulsion gel preparation, and bulk emulsion gels can also be broke into smaller pieces with different sizes and shapes. Therefore, mechanical properties (including viscoelastic and textural properties) of bulk emulsion gels are important. Emulsion gel particles are normally spherical with sizes (diameters) from nano to macro (Ching, Bansal, & Bhandari, 2017). Thus, mechanical properties are also important for the macrogel particles. However, emulsion gel particles can be dispersed in aqueous media, and gel particles allow swelling or de-swelling as a function of environmental conditions, allowing turning of their size and/or physicochemical properties (Torres, Tena, Murray, & Sarkar, 2017). There are two types of fluid emulsion gels: gel-like Pickering emulsions and disrupted emulsion gel systems. Fluid emulsion gels do not have solid shapes, but they have higher viscoelasticity than conventional emulsions. Gel-like Pickering emulsions are similar to bulk emulsion gels, exhibiting a solid state (Zou, Guo, Yin, Wang, & Yang, 2015), while disrupted emulsion gels normally exhibit fluid characteristics (Soukoulis, Cambier, Hoffmann, & Bohn, 2016). This paper provides an overview of the current knowledge of preparation methods, structure-property relationships, and applications of different emulsion gel systems.

## 2. Preparation of different emulsion gels

### 2.1. Bulk emulsion gels

As shown in Table 1, proteins (e.g., myofibrillar protein, whey protein, soy protein, gelatin, bovine serum albumin, sodium caseinate, and casein), polysaccharides (e.g., carrageenan, gellan gum, agar, alginate, and inulin), and LMW compounds (e.g., sapoin glycyrrhizic acid and the mixture of *β*-sitosterol and *γ*-oryzanol) are normally used as matrix materials in bulk emulsion gels. According to the gelation process, preparation methods for bulk emulsion gels include heat-set and cold-set (including one-step cold-set, cold-set after heat treatment, enzyme treatment, acidification treatment, addition of ions, and self-assembly) gelation methods. Choosing an appropriate method depends on the matrix materials (i.e., proteins, polysaccharides or LMW compounds) and applications of prepared emulsion gels such as mimicking food processing (i.e., heating process of meat), protecting encapsulated nutrients, controlled release of encapsulated nutrients, or obtaining better mechanical properties. For heat-set, one-step cold-set, cold-set after heating, and self-assembled gelation methods, the concentration of proteins, polysaccharides and LMW compounds in the water phase should be higher than the critical gelation concentration to guarantee the gelation. However, for gelation methods based on enzyme treatment, acidification treatment, or addition of ions, the concentration of matrix molecules can be below the critical gelation concentration, especially to avoid gelation during the pre-heating process (Ye & Taylor, 2009).

#### *2.1.1. Protein-based bulk emulsion gels*

Several methods have been studied for preparing protein-based bulk emulsion gels: heat treatment, cold-set after pre-heating, acidification, addition of ions, and enzyme treatment, depending on the gelation properties of proteins (Farjami & Madadlou, 2019).

Heat treatment can denature proteins, and denatured protein molecules can aggregate and form three-dimensional structures through chemical forces (i.e., disulfide bonds, electrostatic interactions, hydrophobic interactions, hydrogen bonds, and ionic bonds) under appropriate conditions (e.g., protein concentration, pH, and ionic strength) (Tolano-Villaverde, Torres-Arreola, Ocaño-Higuera, & Marquez-Rios, 2015). Proteins (e.g., myofibrillar protein (MP), whey protein isolate (WPI), and soy protein isolate (SPI)), which undergo heat-induced gelation, can be used as matrix materials to prepare heat-induced bulk emulsion gels. However, recent studies have focused on MP- and WPI-based bulk emulsion gels (Guo, Ye, Lad, Dalgleish, & Singh, 2013; Wang, Zhang, Chen, Xu, Zhou, Li, et al., 2018). Studying heat-induced MP-based bulk emulsion gels is important to develop high quality processed meat products such as sausages and surimi, because the interactions between MPs and fat globules or oil droplets play an important role in textual properties and stability of meat products. In addition, heat treatment is the most common method for producing WPI-based bulk emulsion gels in order to investigate the interactions between emulsifiers and the WPI-based gel matrix (Chen, Dickinson, Langton, & Hermansson, 2000).

One-step cold-set or cold-set after heat treatment is normally used for preparing gelatin-based emulsion gels. The gelation mechanism of gelatin is that, when the gelatin solution is cooled below 30ºC, a self-assembly process of gelatin occurs and helices are created (Gómez-Guillén, Giménez, López-Caballero, & Montero, 2011). Heat treatment (above 40ºC) is normally used to increase the solubility of gelatin before cold-set treatment. However, for cold-soluble gelatin, the thermal process is not necessary (Pintado, Ruiz-Capillas, Jimenez-Colmenero, Carmona, & Herrero, 2015). In addition, ethanol has been used to denature proteins and produce cold-set whey protein emulsion gels (Xi, Liu, McClements, & Zou, 2019).

The mechanism of acid-induced protein gelation is that the acidification, usually carried out by adding glucono-δ-lactone (GDL), decreases the pH and neutralize the surface charges of protein aggregates and a gel network then forms by hydrophobic interactions and Van der Waals forces (Ringgenberg, Alexander, & Corredig, 2013). Before acidification, heat treatment is normally used to denature proteins and form protein aggregates. In such cases, two different processes can be used to produce acid-induced protein-based emulsion gels: using pre-heated-induced protein aggregates to form emulsions (Lu, Mao, Zheng, Chen, & Gao, 2020) or heating native protein-stabilized emulsion to form protein aggregates (Ye & Taylor, 2009) before acidification. However, heating native protein-stabilized emulsions may lead to droplet aggregation, which limits the application of emulsion gels for encapsulation of heat-sensitive compounds (Mao, Roos, & Miao, 2014).

Addition of ions (normally $Ca^{2+}$ in the form of $CaCl_2$) can promote soluble protein aggregates to form a gel network by ionic crosslinks (Wang, Luo, Liu, Adhikari, & Chen, 2019). It has been reported that structures of $CaCl_2$-induced SPI emulsion gels were mainly composed of particulate protein-coated and  were different from filamentous gel networks formed by MTGase and GDL (Tang, Chen, & Foegeding, 2011). In addition, the concentration of $Ca^{2+}$ can affect the structures of protein-based emulsion gels; Sok Line, Remondetto, & Subirade (2005) found that low calcium concentrations (e.g.,11.7 mM $Ca^{2+}$) induced emulsion gels with a fine-stranded structure, while high calcium concentrations (e.g., 40 mM or 68 mM $Ca^{2+}$) led to random aggregates. Therefore, $CaCl_2$ at a concentration of 8–20 mM is normally used to produce $Ca^{2+}$-induced emulsion gels (Liang, Leung Sok Line, Remondetto, & Subirade, 2010; Tang, Chen, & Foegeding, 2011; Ye & Taylor, 2009).

Microbial transglutaminase (MTGase) can be used to promote cross-links between protein molecules and improve the properties of protein-based emulsion gels (Gaspar & de Goes-Favoni, 2015). Compared to other methods, enzyme treatment is a safe method to produce protein-based emulsion gels with high quality under mild process conditions (35–37ºC) and without producing any side-products (Tang, Luo, Liu, & Chen, 2013). It was found that the gel strength of MTGase-induced SPI-based emulsion gels was much higher than that of GDL- or $CaCl_2$-induced emulsion gels (Tang, Chen, & Foegeding, 2011). Two points should be highlighted when enzyme treatment is used to prepare protein-based emulsion gels. Firstly, the order of adding enzyme into emulsions may influence the properties of emulsion gels. Tang, Yang, Liu, & Chen (2013) found that adding enzyme prior to emulsification required less enzyme, but induced emulsion gels with higher stiffness compared to adding enzyme after emulsification. Secondly, although the formation of protein aggregates is not necessary for producing enzyme-induced gels, unfolding the compact structures of globular proteins (e.g., SPI, WPI, and MP) can provide more target glutamine and lysine residues for the MTGase treatment. For example, pre-incubation of SPI and egg white protein (Alavi, Emam-Djomeh, Salami, & Mohammadian, 2020; Tang, Yang, Liu, & Chen, 2013), pre-oxidation treatment of MP (Wang, Xiong, & Sato, 2017), and breaking down disulfide bonds in bovine serum albumin (Kang, Kim, Shin, Woo, & Moon, 2003) can improve gelation by MTGase. However, it has been found that heated SPI-stabilized emulsions after emulsification could not form gels following enzymatic treatment (Tang, Chen, & Foegeding, 2011).

### *2.1.2. Polysaccharide-based bulk emulsion gels*

Several methods have been studied for preparing polysaccharide-based bulk emulsion gels, such as heat-set, cold-set after pre-heating, addition of ions, and self-assembly (crystallisation), depending on the gelation properties of polysaccharides.

Curdlan is a water-soluble $\beta$-(1,3)-glucan extracted from *Alcaligenes* faecalis, and curdlan-based emulsion gels can be obtained after heating emulsions, while cold-set after pre-heating is normally used to prepare carrageenan-, agar-, and gellan gum-based emulsion gels (Jiang, et al., 2019). The gelation mechanism involves forming double helices and cross-linking helical domains to create a three-dimensional structure during cooling (Nishinari & Takahashi, 2003). These are all cold-set and thermo-reversible gels. For producing cold-set emulsion gels, polysaccharides should be dissolved at a high temperature (normally more than 70ºC), and/or emulsions should be prepared at a medium temperature (normally between 45ºC and 70ºC), after which emulsion gels are formed at a low temperature (normally less than 25ºC).

The addition of ions is normally used to produce alginate-based emulsion gels. Alginate, a linear unbranched natural polysaccharide, is derived from brown seaweed extracts (*Phaeophyceae*) (King, 1983). Sodium alginate has the ability to form 'egg-box' shaped gels when the sodium ions are replaced by divalent cations (mostly calcium in the food industry) (Ching, Bansal, & Bhandari, 2017). Two different methods can be used to prepare alginate-based emulsion gels. Pintado, Ruiz-Capillas, Jimenez-Colmenero, Carmona, & Herrero (2015) added $CaSO_4$ into an alginate-based emulsion to produce an alginate-based emulsion gel directly. Sato, Moraes, & Cunha (2014) used a different method to produce emulsion gels, in which CaEDTA was added to the alginate-based emulsion first, after which the acid was then introduced to liberate calcium ions.

Inulin is an oligosaccharide which includes 2 to 60 fructose molecules connected by β-(2→1) glycoside bonds (Glibowski & Pikus, 2011). Inulin with a crystal structure can disperse in an aqueous environment and form a suspension in which most of the crystals do not change their structures, except some of smallest crystals dissolving in water. Amorphous inulin can change its structure to crystallite in water (Glibowski & Pikus, 2011). Then, small crystallites can aggregate to form larger clusters, which ultimately interact to form a gel (Bot, Erle, Vreeker, & Agterof, 2004). Paradiso, Giarnetti, Summo, Pasqualone, Minervini, & Caponio (2015) compared three different homogenization technologies (i.e., mechanical, ultrasonic and cold ultrasonic homogenization) to prepare inulin-based emulsion gels, and found that ultrasonic homogenization is a suitable method to prepare emulsion gels with better textural properties compared to the other two homogenization technologies.

*2.1.3. Self-assembly of low molecular weight compound-based bulk emulsion gels*

Many LMW organic compounds, such as glycyrrhizic acid and a combination of β-sitosterol and γ-oryzanol, can be used as oil-structuring agents, due to their self-assembly, to replace solid fats and provide required sensory and flavor properties in food products (Pernetti, van Malssen, Flöter, & Bot, 2007; Wan, Sun, Ma, Yang, Guo, & Yin, 2017). These organic compounds, when in a water or oil phase, can form soft solid-like structured gels, which are known as oleogels or organogels (Co & Marangoni, 2012), and they can be also used to produce emulsion gels.

Saponin glycyrrhizic acid (GA) is a monodesmosidic saponin which is comprised of a hydrophobic triterpenoid aglycon moiety (18 β-glycyrrhetinic acid) attached to a hydrophilic diglucuronic unit. GA molecules have both gelation and emulsifying properties, owing to their self-assembly ability and amphiphilic structures. GA cannot structure vegetable oil directly because of its low solubility in oil. However, GA molecules can self-assemble into long nanofibrils in water, and nanofibrils not only absorb at the oil-water interface but also further assemble and entangle to create a supramolecular hydrogel in water phase. Wan, Sun, Ma, Yang, Guo, & Yin (2017) investigated GA-based O/W emulsion gels and found that, for more polar oils, GA fibrils had a higher affinity to the oil-water interface, leading to the formation of a lot of fine multilayer emulsion droplets with smaller droplet size. Ma, Wan, &

Yang (2017) used GA to produce GA-based water-in-oil-in-water ($W_1/O/W_2$) emulsion gels; a $W_1/O$ emulsion was prepared first, before being mixed with GA solution at 80ºC, and GA-based $W_1/O/W_2$ emulsion gels were formed at room temperature by the self-assembly of GA.

The combination of β-sitosterol and γ-oryzanol can self-assemble in an oil phase to form a helical ribbon, and then these tubules can aggregate and form networks, which are known as oleogels or organogels. Thus, the combination of β-sitosterol and γ-oryzanol can be used to prepare gelled W/O emulsions. However, the oil phase should be prepared at high temperature (∼100ºC) to dissolve β-sitosterol and γ-oryzanol, and W/O emulsions should also be prepared at 90ºC to prevent the gelation of oil phase during emulsification. It has been reported that, when a mixture of β-sitosterol and γ-oryzanol was used to prepare W/O emulsion gels, the presence of water weakened the tubules and reduced the firmness of gelled emulsions, due to the hydration of β-sitosterol and the transition of crystals from anhydrous and hemihydrate into monohydrate forms (Bot, den Adel, Regkos, Sawalha, Venema, & Flöter, 2011). On the other hand, it was found that reducing the water activity and using oils with low polarity could promote the formation of tubular microstructures of oryzanol and sitosterol in emulsions (Sawalha, den Adel, Venema, Bot, Floter, & van der Linden, 2012).

## 2.2. Emulsion gel particles

Gel particles or gel beads can be divided into three categories according to their size: macrogel particles (> 1 mm), microgel particles (0.2–1000 μm), and nanogel particles (< 0.2 μm) (Ching, Bansal, & Bhandari, 2017). In the food area, studies have mainly focused on macrogel and microgel particles, and alginate was the matrix material most frequently used to produce gel particles. Ching, Bansal, & Bhandari (2017) reviewed current technologies for producing alginate hydrogel particles (e.g., simple dripping, jet back up extrusion, spinning disk, atomization, impinging aerosol method, emulsification technique, microfluidics, and templating method), but studies on producing emulsion gel particles have rarely been reported. As shown in Table 2, methods used to prepare emulsion gel particles include simple dripping, electrostatic extrusion, and the impinging aerosol method.

Lević, Pajić Lijaković, Đorđević, Rac, Rakić, et al. (2015) used electrostatic extrusion technique to prepare alginate-based emulsion beads with diameters in the range from 960 to 1650 μm, in which a syringe pump and electrostatic immobilization unit (at a voltage of 6.5 kV) were used to extrude an alginate-based emulsion through a needle (22 gauge) into a collecting solution (0.015 g/ml of $CaCl_2$ solution). The reason for using an electrostatic immobilization unit is that electrostatic forces can disrupt the liquid filament at the tip of the needle and create a charged stream of small droplets. However, bigger beads were formed with the diameters in the range from 2100 to 2350 μm without applying voltage, i.e., extrusion by syringe or simple dripping, which is thus a simple method to produce emulsion gel particles, but this method usually leads to large particle sizes. Ching, Bansal, & Bhandari (2016) developed a spray aerosol method to prepare alginate-based emulsion microgel particles with the size of 36.2 to 57.8 μm. A fine aerosol mist of alginate-based emulsion and an aerosol mist of 0.5 M calcium chloride solutions are created at the top and bottom of the chamber, respectively, using an air atomizing nozzle. Two mists combine in the chamber, and emulsion gel particles form in the chamber and are collected at the base of chamber. This is an effective and continuous method to produce emulsion gel particles with small size, but this method needs a special spray aerosol system.

## 2.3. Fluid emulsion gels

Apart from bulk emulsion gels and emulsion gel particles, fluid emulsion gels are the third type of emulsion gels. Fluid emulsion gels are different from bulk gels and gel particles with solid shapes, but they have higher viscoelastic properties than conventional emulsions. Fluid emulsion gels mainly include two types according to their preparation methods: gel-like emulsions and disrupted emulsion gel systems (Table 3).

### 2.3.1. Gel-like emulsions

Pickering emulsions are a kind of emulsions which are stabilized by amphiphilic solid particles, and can be divided into three categories: polysaccharide particle-, protein particle- and mixture-stabilized Pickering emulsions. Pickering emulsions are considered as better delivery systems than conventional emulsions, owing to their enhanced storage stability against oxidation and coalescence and lower susceptibility to lipolysis. Pickering emulsions can turn into gel-like emulsions under appropriate conditions (e.g., proper solid particle type, solid particle concentration, oil phase concentration, pH, and ionic strength). It has been reported that gel-like emulsions could be formed with 6 wt% preheated soy globulins at high glycinin contents (> 75%) with soy oil at a oil volume fraction (φ) of 0.3, and that G′ and G″ values of gel-like emulsions increased as the increase of glycinin contents (from 75% to 100%), while neither unheated soy globulins nor preheated soy globulins with low glycinin contents could form gel-like emulsions (Luo, Liu, & Tang, 2013). This was probably because the formation of a gel-like network was largely attributed to hydrophobic interactions between denatured glycine molecules absorbed at the interface of oil droplets. However, Xu, Liu, & Tang (2019) found that, with increasing oil fractions (φ = 0.1 to 0.88), a 0.5 wt% soy *β*-conglycinin-stabilized Pickering emulsion could turn into a gel-like emulsion at an oil fraction of 0.7. It was also found that, with increasing wheat gluten level (emulsifier in oil-in-glycerol emulsions, 0.25–1.0 wt%), gel-like emulsions could be formed at high wheat gluten contents (>= 0.5 wt%) (Liu, Chen, Guo, Yin, & Yang, 2016). Shao & Tang (2016) found that, with increasing oil fraction (0.2 to 0.6), pea protein-based Pickering emulsions changed from liquid to a gel-like state, while Zou, Guo, Yin, Wang, & Yang (2015) found that zein/tannic acid complex-stabilized Pickering emulsion gels with high oil volume fraction (φ > 0.5) could be successfully produced. Therefore, the oil phase and emulsifier contents should be high enough to assure that solid particles absorbed at the surface of neighboring oil droplets can connect and/or react with each other (Wouters & Delcour, 2019).

*2.3.2. Disrupted gel systems*

Fluid emulsion gels can also be prepared by breaking down bulk emulsion gels (Leon, Medina, Park, & Aguilera, 2018). Soukoulis, Cambier, Hoffmann, & Bohn (2016) investigated so-called sheared oil-in-gel (o/g) emulsions prepared by stirring an alginate-based emulsion gel system at 1000 rpm for 6 h during the gelation process. Torres, Tena, Murray, & Sarkar (2017) developed a method to produce starch-based gel emulsions by homogenizing the bulk emulsion gels. This is a simple method to produce fluid emulsion gels with small dispersed gel particles (5–50 μm in diameter), but the gel matrix-covered structure may be destroyed, leading to separation of the gel matrix and oil droplets during homogenization, which may influence the stability of oil droplets and/or encapsulated nutrients during storage.

## 3. Structure-property relationships of different emulsion gels

3.1. Bulk emulsion gels

Some properties of bulk emulsion gels are emphasized in the food industry, such as mechanical properties (e.g., rheological, and textural perception), and release properties (including stability during storage and targeted-release in digestion). Many factors (e.g., structures of the gel matrix, structures of emulsion droplets, and interactions between the gel matrix and droplets) can influence the structures of bulk emulsion gels and then their mechanical and release properties.

Common food emulsions include single emulsions (O/W and W/O emulsions) and multiple emulsions ($W_1/O/W_2$ and $O_1/W/O_2$ emulsions). After turning emulsions into bulk emulsion gels, their structures usually do not change. Thus, the structures of emulsion gels also include single structures (i.e., O/W and W/O) and multiple structures (i.e., $W_1/O/W_2$ and $O_1/W/O_2$). The matrix materials of O/W and $W_1/O/W_2$ emulsion gels are protein-, polysaccharide-, or organic compound-based hydrogels, while the matrix materials of W/O and $O_1/W/O_2$ emulsion gels are organic compound-based oleogels (also known as organogels or structured oil). Moreover, properties of O/W and $W_1/O/W_2$ emulsion gels and W/O and $O_1/W/O_2$ emulsion gels differ, because the properties of emulsion gels mainly depend on the properties of matrix materials (i.e., protein-, polysaccharide-, or organic compound-based gels), although the properties of emulsion droplets and the interactions between the gel matrix and droplets also influence the properties of emulsion gels. However, O/W emulsion gels have been studied more widely than W/O, $W_1/O/W_2$ and $O_1/W/O_2$ emulsion gels, so the following discussions in this review will focus on O/W emulsion gels unless other structures are emphasized.

#### *3.1.1. The structure-mechanical property relationships of bulk emulsion gels*

Mechanical properties of bulk emulsion gels are closely associated with other properties (e.g., storage stability, oral perception, and controlled release) and their applications. The most common mechanical properties of bulk emulsion gels are dynamic modulus (i.e., storage and loss modulus), Young's modulus, fracture strength (i.e., strain and stress), yield strength, and hardness. There are many ways or tools to measure mechanical properties of bulk emulsion gels, such as rheometry, dynamic mechanical analysis (DMA), and textural analysis (Anseth, Bowman, & Brannon-Peppas, 1996).

##### *3.1.1.1. Matrix structures*

For protein-based bulk emulsion gels, use of different proteins and methods can lead to different protein matrix structures and mechanical properties, owing to different gelation mechanisms and resultant different molecular forces between protein molecules in the gel matrix. Globular proteins (e.g., SPI, WPI, and MP) and non-globular proteins (e.g., gelatin, casein, and sodium caseinate) have been widely used as matrix materials in producing bulk emulsion gels.

The heat-set gelation method has been most used to prepare globular protein-based emulsion gels, but globular protein-based emulsion gels can be also prepared through acidification treatment, addition of ions, enzyme treatment, and MDA modification. For heat-induced emulsion gels, noncovalent cross-links (i.e., electrostatic interactions, hydrophobic interactions, and hydrogen bonds) and intermolecular disulfide bonds are the main forces between globular protein molecules (Wu, Xiong, & Chen, 2011). The main linking forces in the glucono-δ-lactone (GDL)-induced emulsion gels are hydrophobic interactions and Van der Waals forces, while salt-bridges are the main linking forces in salt-induced emulsion gels, and TGase-induced emulsion gels involve more covalent cross-links (i.e., ε-(γ-glutamyl)-lysine (G-L) cross-links). Therefore, different preparation methods may lead to different mechanical properties of globular protein-based emulsion gels (Liang, et al., 2020; Wang, Xiong, & Sato, 2017; Ye & Taylor, 2009); for example, it was found that $CaSO_4$-induced SPI-based emulsion gels were stiffer with higher rigidity than MTGase-induced gels which performed better elasticity (Wang, Luo, Liu, Zeng, Adhikari, He, et al., 2018).

Gelatin can form gels under one-step cold treatment or cold treatment after pre-heating as descried in section 2.1.1.2. Cold-set gelatin gels, a kind of elastic polymer gel, are formed with flexible and random-coil protein chains. Therefore, gelatin-based emulsion gels are similar to gels with active-fillers (bound droplets) in which stress concentration phenomena play a larger role compared to friction phenomena (Sala, van Vliet, Cohen Stuart, Aken, &

van de Velde, 2009). Other non-globular protein-based emulsion gels are normally prepared with enzyme treatment and acidification treatment. For example, although the main linking forces in acid-induced casein gels are also noncovalent cross-links, the firmness of acid-induced sodium caseinate gels was lower than that of acid-induced WPC gels, probably due to their differences in gelation mechanism (Kiokias & Bot, 2005).

Overall, contributions to the connectivity of a three-dimensional protein network arise from four different kinds of molecular forces: covalent bonds, electrostatic interactions, hydrogen bonding and hydrophobic interactions. The presence of covalent bonds leads to permanent 'chemical' cross-links within the network, whereas the other three types of weaker 'physical' forces contribute to a complex set of more temperature-dependent interactions (Chen & Dickinson, 1999b). In addition, process parameters (e.g., temperature, protein content, ionic strength, pH, the presence of other components, ultrasound pretreatment, and high-pressure homogenization) also can influence the structures and mechanical properties of protein-based bulk emulsion gels (Bi, et al., 2020; Chen & Dickinson, 2000; Cheng, et al., 2019).

Firstly, temperature can influence the degree of denaturation of proteins, and thus affect the stability of protein-stabilized emulsions and mechanical properties of emulsion gels. Generally, a high degree of denaturation of proteins results in low stability of protein-stabilized emulsions but better mechanical properties of emulsion gels (Kiokias & Bot, 2005; Ye & Taylor, 2009). Chen & Dickinson (2000) also found that gelation temperature could influence the rate of gelation and the dynamic modulus of acid-induced sodium caseinate-based emulsion gels by changing the strength of physical bonding rather than the network structures.

Secondly, the influence of protein content on the mechanical properties of emulsion gels depends on the state of emulsion droplets. The mechanical properties of droplet-filled gels and inactive droplet-aggregated gels mainly depend on the gel strength of gel matrix structures, while interactions between the gel matrix (i.e., protein and polysaccharide) and lipid droplets contribute more to the active droplet-aggregated gels (Pintado, Ruiz-Capillas, Jimenez-Colmenero, Carmona, & Herrero, 2015). Therefore, increasing protein content can increase the gel strength of both kinds of emulsion gels but for different reasons (i.e., increased gel strength of protein matrix for droplet-filled gels and inactive droplet-aggregated gels, but strengthened interactions between the gel matrix and droplets and increased gel strength of protein matrix for active droplet-aggregated gels). For example, it has been reported that increasing the concentration of sodium caseinate can decrease the gelation time ($T_{gel}$) of sodium caseinate/sunflower oil emulsion-based gels (Montes de Oca-Ávalos, Huck-Iriart, Candal, & Herrera, 2016).

Thirdly, ionic strength and pH can influence intermolecular repulsion and gel structures in emulsion gels. For example, at low ionic strength (< 50 mM NaCl) and pH values (below 4 or above 6) far away from pI of whey proteins, a fine-stranded network consisting of whey protein strains with a length of ~50 nm and a diameter of ~10 nm is formed; at high ionic strength (> 150 mM NaCl) and pH values near the pI, the strains with weak intermolecular repulsion can accumulate and form a particulate network structure (Chen, Dickinson, Langton, & Hermansson, 2000; Guo, Bellissimo, & Rousseau, 2017; Langton & Hermansson, 1992). However, both fine-stranded and particulate gels exhibit high gel strength (Guo, Bellissimo, & Rousseau, 2017; Tang, Chen, & Foegeding, 2011). It was found that fine-stranded whey protein gels prepared at low ionic strength (10 or 25 mM NaCl) were rubbery and soft, but that particulate whey protein gels prepared at high ionic strength (100 or 200 mM NaCl) were hard and brittle (Guo, Ye, Lad, Dalgleish, & Singh, 2013).

Fourthly, the presence of other components (e.g., sucrose, glucose, hydroxytyrosol, rosmarinic acid, genipin, sodium pyrophosphate, insoluble dietary fiber, and EGCG) also can influence the structures and mechanical properties of emulsion gels (Chen, Ren, Zhang, Qu, Hu, & Yan, 2019; Feng, Chen, Lei, Wang, Xu, Zhou, et al., 2017; Freire, Bou, Cofrades, & Jimenez-Colmenero, 2017; Montes de Oca-Ávalos, Huck-Iriart, Candal, & Herrera, 2016; Wang, et al., 2018; Wang, Jiang, & Xiong, 2019; Zhuang, et al., 2019). Generally, if components can strengthen protein-protein interactions and/or reduce droplet size, they can increase gel strength of emulsion gels. However, if these components can interact with protein molecules and disturb the interactions between protein molecules, they can weaken the gel strength of emulsion gels, and these effects are normally dose-dependent. Overall, preparation methods can affect linking forces between protein molecules, and protein type and processing parameters can influence the network structures of the gel matrix, both of which can affect the mechanical properties of emulsion gels.

In terms of polysaccharide-based emulsion gels, polysaccharide type, preparation methods, and processing parameters can influence the structures of the polysaccharide-based gel matrix. Cold-set gellan gun-, agar-, and κ-carrageenan-based emulsion gels are a kind of polymer gels with strand-based structures (Kim, Gohtani, Matsuno, & Yamano, 1999; Wang, Neves, Kobayashi, Uemura, & Nakajima, 2013). They normally show a predominantly elastic behavior, which resemble gelatin-based emulsion gels but differ from WPI-based emulsion gels with particulate structures (Sala, van Vliet, Cohen Stuart, Aken, & van de Velde, 2009). The network structures of alginate gels are in the shape of ‘egg-box’, in which sodium ion is replaced by a divalent cation, and each cation can bind with four G residues to form a three-dimensional network structure (Ching, Bansal, & Bhandari, 2017), which can be affected by freeze-thawing treatment (Li, Gong, Hou, Yang, & Guo, 2020). Inulin gels are formed by connection of microcrystals, and their rheological properties resemble that of fat crystal-based networks in oil (Nourbehesht, Shekarchizadeh, & Soltanizadeh, 2018). However, there are no studies on comparing mechanical properties of emulsion gels formed by different kinds of polysaccharides.

In addition, the influence of polysaccharide content on the mechanical properties of emulsion gels depends on emulsifier type and gel structures. Most natural polysaccharides, except gum Arabic and some kinds of pectin, have weak emulsifying abilities compared to proteins and synthetic emulsifiers (Charoen, Jangchud, Jangchud, Harnsilawat, Naivikul, & McClements, 2011). Hence, the interactions between the gel matrix and emulsion droplets in polysaccharide-based emulsion gels with/without synthetic emulsifiers are normally weak, and increasing polysaccharide content can increase their gel strength, mainly due to the decreased void spaces and increased gel strength of the gel matrix (Kim, Gohtani, Matsuno, & Yamano, 1999). However, when proteins are used as emulsifiers, increasing polysaccharide content can increase the gel strength of emulsion, mainly due to increased interactions between polysaccharide molecules and droplets and/or the gel strength of polysaccharide gels. Although studies on the effects of ionic strength and pH on the mechanical properties of polysaccharide-based emulsion gels have rarely been reported, Ozturk, Argin, Ozilgen, & McClements (2015) found that ionic strength and pH did not have significant influences on the stability of a gum Arabic-stabilized emulsion, which was different from a WPI-stabilized emulsion because of their different emulsification mechanisms (i.e., electrostatic repulsion for WPI and steric repulsion for gum Arabic). Therefore, it is proposed that the influence of ionic strength and pH on the structure and mechanical properties of polysaccharide-based emulsion gels differs from that on protein-based emulsion gels.

For LMW organic compound-based emulsion gels, saponin glycyrrhizic acid (GA) and the combination of β-sitosterol and γ-oryzanol have been investigated to prepare emulsion gels by self-assembly. GA, β-sitosterol and γ-oryzanol have physical properties, so they have been used to prepare different types of emulsion gels. GA can dissolve in water, and GA molecules can self-assemble to form long nanofibrils and gels in water phase, and so can be used to prepare emulsion gels with O/W or $W_1/O/W_2$ structures. The combination of β-sitosterol and γ-oryzanol can self-assemble in an oil phase to form a helical ribbon, then these tubules can aggregate and form a network, and so can be used to prepare gelled W/O emulsions. Processing parameters (e.g., organic compound content and solvent type) also can influence the structure and mechanical properties of organic compound-based emulsion gels. Ma, Wan, & Yang (2017) found that an emulsion stabilized by GA at a low concentration (0.5 wt%) could not form a gel, but self-standing emulsion gels could be formed and the viscoelastic modulus also significantly increased with increasing GA concentration (1–4 wt%). It was also found that no tubules were formed but only sitosterol and oryzanol crystals were present in emulsion gels at 16% total sterol concentration, while there were tubules next to the crystals at 32% total sterol concentration (Bot, den Adel, Regkos, Sawalha, Venema, & Flöter, 2011).

In addition, the polarity of solvents (i.e., oil in W/O emulsions) can influence the water activity of W/O emulsions and structures of the oil phase. It has been reported that more water molecules bind to the β-sitosterol molecules and formed monohydrate crystals in higher polarity oils (e.g., eugenol and castor oil), which hindered the formation of tubules and resulted in weaker emulsion gels compared to less polar oils (e.g., decane and limonene) (Sawalha, den Adel, Venema, Bot, Floter, & van der Linden, 2012). However, studies on comparing structures and mechanical properties of emulsion gels prepared with different kinds of organic compounds have rarely been reported. Over all, many factors (e.g., gel matrix type, preparation method, and process parameters) can affect the gel structures of bulk emulsion gels and thus their mechanical properties.

*3.1.1.2. Structures of emulsion droplets*

The structure of emulsion droplets can influence the mechanical properties of bulk emulsion gels as well. Structures of emulsion droplets are normally influenced by oil phase (e.g., oil type, oil content, and droplet size), and emulsifier type (e.g., low molecular weight emulsifiers or proteins). In the food industry, emulsifiers mainly include two categories: low molecular synthetics (e.g., Span 80, Tween 80, and monoglycerides) and natural molecules (e.g., proteins, egg lecithin, and soy lecithin) (Chen, Mao, Hou, Yuan, & Gao, 2020). Emulsifiers can not only decrease the interfacial tension and thereby increase the stability of emulsions but also affect the interactions between droplets and the gel matrix leading to active or inactive fillers (Van Vliet, 1988). Therefore, the effect of emulsion droplets on the mechanical properties of emulsion gels depends on not only emulsion droplets (i.e., oil type, oil content, and droplet size) but also the interactions between droplets and the gel matrix (Farjami & Madadlou, 2019).

The effect of active fillers on the rheological properties of emulsion gels mainly depends on the stiffness of the oil droplets and the droplet volume fraction (Sala, van Vliet, Cohen Stuart, Aken, & van de Velde, 2009). The Kerner model can explain the effect of active fillers on the mechanical properties of emulsion droplet-filled gels (Kerner, 1956):

$$\frac{G'_{gel}}{G'_{matrix}} = \frac{15(1-v_m)(M-1)\phi_f}{(8-10v_m)M+7-5v_m-(8-10v_m)(M-1)\phi_f} + 1 \quad (1)$$

where $M = \frac{G'_{filler}}{G'_{matrix}}$, and $G'_{gel}$, $G'_{filler}$, and $G'_{matrix}$ are the shear modulus of the overall gel, the filler droplets and the gel matrix, respectively, $\phi_f$ is the actual droplet volume fraction, and $v_m$ is the Poisson's ratio of the gel matrix. In addition, the Kerner model modified by Lewis and Nielsen can be used to explain the effect of active fillers on the mechanical properties of emulsion droplet-aggregated gels (Lewis & Nielsen, 1970):

$$\frac{G'_{gel}}{G'_{matrix}} = \frac{15(1-v_m)(M-1)\psi\phi_f}{(8-10v_m)M+7-5v_m-(8-10v_m)(M-1)\psi\phi_f} + 1 \quad (2)$$

where $\psi\phi_f$ is the effective volume fraction of fillers, which takes into account the crowding effect of fillers and can be expressed as follows (Lewis & Nielsen, 1970):

$$\psi\phi_f = \left[1 + \left(\frac{1-\phi_{max}}{\phi_{max}^2}\right)\phi_f\right]\phi_f \quad (3)$$

where $\phi_{max}$ is the maximum volume fraction of the fillers. According to Eq. (2), increasing the shear modulus and the effective volume fraction ($\psi\phi_f$) or actual volume fraction ($\phi_f$) of fillers can increase the mechanical properties of emulsion gels, which has been supported by many studies (Gwartney, Larick, & Foegeding, 2004; Li, Kong, Zhang, & Hua, 2012; Oliver, Scholten, & van Aken, 2015; Oliver, Wieck, & Scholten, 2016; Tang, Yang, Liu, & Chen, 2013). However, the Kerner model and the modified Kerner model are used under the assumption that M or $G'_{matrix}$ do not change with changes in other factors (e.g., $\phi_f$ and $G'_{filler}$) (Chen & Dickinson, 1998a; Oliver, Berndsen, van Aken, & Scholten, 2015), especially at oil volume fractions (φ) below 0.2 and protein (i.e., gel matrix) contents above 6 wt% (Guo, Bellissimo, & Rousseau, 2017). However, the shear modulus of filler droplets ($G'_{filler} = 4\gamma$ / d, where γ is surface tension and d is the average diameter of the oil droplets ) is influenced by oil type, oil content, droplet size, emulsifier type, emulsifier content, and process parameters (Farjami & Madadlou, 2019; Sala, van Vliet, Cohen Stuart, Aken, & van de Velde, 2009; Van Vliet, 1988). The shear modulus of the gel matrix ($G'_{matrix}$) is influenced by droplet size, oil content, gel matrix type, preparation method, and process parameters (Sato, Moraes, & Cunha, 2014). Therefore, when taking those factors (e.g., droplet size, process parameters, and high oil content), which can affect the mechanical properties of both filler droplets and the gel matrix, into account, the Kerner model and the modified Kerner model cannot be applied. For instance, it has been reported that increasing the size of olive oil droplets in a gelatin-based emulsion gel led to a weaker gel strength, probably due to the increase in interfacial area, a higher amount of gelatin adsorbed to the interface, and a lower quantity of protein available in the continuous phase (Sato, Moraes, & Cunha, 2014); however, it was found that increasing the size distribution of dispersed vegetable fat in a WPI-based emulsion gel led to an increase in firmness, probably because of a larger number of contacts between droplets (Kiokias & Bot, 2006). Oliver, Wieck, & Scholten (2016) found that increasing the casein content (from 4% to 9%) could decrease the relative Young's modulus of emulsion gels at high oil volume fractions ($\phi_f > 0.15$), probably owing to the higher inhomogeneity of casein-based gel matrix and increased effective volume fraction of droplets at lower casein concentration; this indicated that the effective volume fraction ($\psi\phi_f$) plays a more important role than $G'_{matrix}$ in affecting the mechanical properties of emulsion gels with high matrix inhomogeneity and at high oil volume fractions.



The effect of inactive fillers on the rheological properties of emulsion gels depends on the properties and concentrations of LMW emulsifiers, droplet size, and oil content, although there have been few studies on modelling the effect of inactive fillers on the rheological properties of emulsion gels. Chen & Dickinson (1999a) investigated the effect of LMW emulsifiers on the viscoelastic properties of heat-set whey protein-based emulsion gels, and found that the elastic modulus of heat-set whey protein-based emulsion gels decreased after adding a low level of diglycerol monolaurate (DGML, the surfactant/protein molar ratio (R) = 4) and diglycerol monooleate (DGMO, R = 4–32), while high levels of emulsifiers (R = 32 for DGML, and R = 64 for DGMO) could recover the storage and loss modulus of emulsion gels, probably due to depletion flocculation of the emulsion prior to heat-treatment. However, it has been reported that Tween 20 (R = 0.25–8) always decreased the mechanical properties of emulsion gels, and a high addition level (R = 8) could even break down the network structure of proteins and lead to a liquid-like emulsion (Chen & Dickinson, 1998b). It has been found that increasing oil content decreased fracture stress and stress intensity factor of agar gels and $\kappa$-carra-geenan-locust bean gum gels (Koç, Drake, Vinyard, Essick, van de

Velde, & Foegeding, 2019). It has also been found that increasing solid lipid content could increase the gel strength of emulsion gels at an emulsifier content of 4 g/100 g, but decreased the gel strength at an emulsifier content of 2 g/100 g (Geremias-Andrade, Souki, Moraes, & Pinho, 2017).

*3.1.2. The structure-release property relationships of bulk emulsion gels*

Bulk emulsion gels, especially O/W emulsion gels, are often used for the delivery and release of oil-soluble bioactive compounds and nutrients, such as α-tocopherol (Liang, Leung Sok Line, Remondetto, & Subirade, 2010) and β-carotene (Soukoulis, Tsevdou, Andre, Cambier, Yonekura, Taoukis, et al., 2017). Compared to emulsions, emulsion gels can provide better protection for encapsulated compounds and show slower release behavior (Cofrades, et al., 2017). Many studies have focused on the matrix erosion, lipid digestion and controlled release of encapsulated compounds during digestion of emulsion gels. The digestion behaviors of protein- and polysaccharide-based emulsion gels differ in the gastrointestinal tract because of different digestion processes of proteins and polysaccharides. For protein-based emulsion gels, Liang, Leung Sok Line, Remondetto, & Subirade (2010) found that gel loss (i.e., matrix erosion owing to protein degradation) and release of α-tocopherol occurred in both simulated gastric fluid (SGF) and simulated intestinal fluid (SIF), respectively, which indicated that release of α-tocopherol was controlled mainly by matrix erosion because of protein degradation. However, under simulated gastrointestinal (GI) conditions (0.5 h SGF followed by 6 h SIF), gel loss and release of α-tocopherol only occurred in the SGF step, probably due to the formation of a viscous layer at the surface of gels. Moreover, gel rigidity of protein-based emulsions is an important factor affecting the lipid digestion in GI digestion. It has been reported that gastric digesta of a soft gel, prepared with 10 or 20 mM NaCl, mainly consisted of individual oil droplets and small gel particles (~10 mm), while gastric digesta of a hard gel, prepared with 100 or 200 mM NaCl, mainly consisted of small gel particles (~10 mm) after 240 min gastric digestion, and the remaining network structure of gel particles hindered further breakdown during intestinal digestion (Guo, Ye, Lad, Dalgleish, & Singh, 2016). It was also found that digestion of emulsion gels in the intestinal step was delayed by denser, more spatially heterogeneous protein matrixes (Guo, Bellissimo, & Rousseau, 2017). In terms of polysaccharide-based emulsion gels, although there are fewer reports about their digestion, it was found that oil droplets could be released from agar-based emulsion gels during GI digestion in both SGF and SIF steps (2.0 h SGF followed by 4–14 h SIF), while emulsifier type (glycerol monolaurate with different degrees of polymerization) affected the size distribution of released oil droplets (Wang, Neves, Kobayashi, Uemura, & Nakajima, 2013).

Bulk emulsion gels are also used for the delivery and release of volatile flavor compounds, such as ethyl butyrate, ethyl hexanoate, ethyl octanoate, propanol, diacetyl, pentanone, hexanal, and heptanone (Hou, Guo, Wang, & Yang, 2016; Mao, Roos, & Miao, 2014). The release of volatile compounds in the oral cavity is normally measured by a simulated nose breath device (Hou, Guo, Wang, & Yang, 2016) or gas chromatography (GC) headspace analysis (Mao, Roos, & Miao, 2014). The release rate of volatile compounds depends on the gel matrix structure, oil content, the nature of volatile compounds, and the interactions between flavor compounds and food ingredients (particularly oils in O/W emulsion gels) (Boland, Delahunty, & van Ruth, 2006; Guichard, 2002). It has been reported that the release rate of ethyl butyrate was significantly lower in a SPI/sugar beet pectin (SBP) complex-based emulsion gel with a compact network than SPI- or SBP-based emulsion gels, but the release rate of aroma compounds with higher hydrophobicity was not significantly influenced by the structures of emulsion gels, probably because of their high affinity for the lipid phase rather than interacting with proteins and/or polysaccharides (Hou, Guo, Wang, & Yang, 2016). Mao, Roos, & Miao (2014) also found that emulsion gels with higher storage modulus at a low oil content (20%) had lower release rates and partition coefficients of the volatiles, and that increasing oil contents (from 5% to 20%) significantly decreased the release rate of heptanone, probably owing to its highly lipophilic characteristics.

## 3.2. The structure-property relationships of emulsion gel particles

Although emulsion gel particles and bulk emulsion gels have similar structures (i.e., active fillers, inactive fillers, emulsion droplet-filled gels, and emulsion droplet-aggregated gels) and structure-property relationships, their physical characteristics and length scales differ (Ching, Bansal, & Bhandari, 2016).

Firstly, the rheological behavior of gel particles differs to that of bulk gels, because the microgel particle system is a suspension (usually gel particles in water). The rheological properties of microgel particle suspensions are influenced by three parameters: volume fraction ($\phi$), particle modulus (modulus of particles that make up the suspension) and interaction potential (Ching, Bansal, & Bhandari, 2016). The volume fraction ($\phi$) can be determined using the equation below (Ching, Bansal, & Bhandari, 2016):

$$\phi = \frac{\frac{m}{\rho}}{\frac{m}{\rho}+v} \quad (4)$$

where $\phi$ = final microgel suspension volume fraction, m = mass of microgel concentrate, ρ = density of microgel concentrate measured with a 50 mL calibrated pycnometer, and v = volume of water added to microgel concentrate. Eq. (4) was modified by the equations developed by Suzawa & Kaneda (2010), who calculated the volume fraction by the weight and density of emulsions but did not consider the weight loss (normally water loss) of gel particles during gelation. At low volume fraction, the flow behaviour is determined by the continuous phase; at higher volume fraction, softer microgels will exhibit a lower storage modulus compared to hard microgels (Adams, Frith, & Stokes, 2004). Ching, Bansal, &

Bhandari (2016) found that, at the same volume fraction, suspensions with more deformable alginate-based micorgels exhibited a lower bulk modulus. However, it is technically difficult to investigate the rheological properties of macrogel particles, although their mechanical properties could be investigated by a texture analyser. It has been reported that, with increasing oil contents in alginate-based macrogels, the elastic modulus of particles decreased, which indicates that oil droplets in alginate-based emulsion gel particles without emulsifiers were inactive fillers (Ching, Bansal, & Bhandari, 2016).

Secondly, syneresis and swelling properties are important properties of gel particles (Ching, Bansal, & Bhandari, 2017). It was found that alginate-based emulsion gel particles shrank less if they had higher oil content, and that the swelling was more pronounced for smaller particles, probably owing to the larger contact surface, but was less pronounced at increased oil contents, probably because of droplets acting as physical barriers for water transport (Lević, et al., 2015).

Thirdly, encapsulation efficiency (EE), loading capacity (LC) and encapsulation yield, important parameters in encapsulation processes of emulsion gel particles, are affected by properties and contents of matrix material, emulsifier, and oil. It has been reported that increasing alginate contents in the water phase could increase the oil EE in lupin protein isolate (LPI)-stabilized emulsion gel particles, probably due to the formation of a stronger gel matrix and better crosslinking on the external surfaces of particles (Piornos, Burgos-Díaz, Morales, Rubilar, & Acevedo, 2017). However, when the protein content was higher than the saturation concentration, or the oil content was very low, in which case excessive free protein molecules existed in the water phase, the aggregation of non-adsorbed protein molecules could lead to lower emulsion stability and lower EE (Guzey & McClements, 2006). In addition, Ruffin, Schmit, Lafitte, Dollat, & Chambin (2014) found that, compared to native WPI, using pre-heated WPI at 80ºC for 30 min as emulsifier in pectin-based emulsion gel particles slightly improved the yield and stability of encapsulated vitamin A, because of the increased viscosity of denatured WPI dispersions and the decreased particle size of emulsions.

## 3.3. The structure-property relationships of fluid emulsion gels

### *3.3.1. Gel-like emulsions*

The oil content, particle content, and surface charge of particles can affect the rheological properties of gel-like Pickering emulsions and release behavior of encapsulated compounds from such emulsions (Shao & Tang, 2016; Xu, Liu, & Tang, 2019). For the effect of oil content, Dai, Sun, Wei, Mao, & Gao (2018) found that zein/gum arabic complex-stabilized Pickering emulsion gels solidified at high oil volume fractions in emulsions ($\varphi >= 0.5$), and increasing oil volume fractions ($\varphi = 0.5$–$0.7$) increased the G′ and G″ of gel-like emulsions, probably due to more interactions between emulsion droplets (Xiao, Wang, Gonzalez, & Huang, 2016). It was also reported that a gel-like emulsion at $\varphi = 0.6$ exhibited much lower release rate of *β*-carotene but higher stability during digestion than a Pickering emulsion at $\varphi = 0.3$ (Shao & Tang, 2016). In terms of the effect of particle content, Xu, Liu, & Tang (2019) found that increasing soy *β*-conglycinin contents from 0.2 to 1.0 wt% led to a progressive decrease in droplet size, but a progressive increase in stiffness of the gel-like emulsions at $\varphi = 0.8$. Liu, Gao, McClements, Zhou, Wu, & Zou (2019) also found that increasing pre-heated WPI contents from 2.5 to 10 wt% led to a progressive increase in gel strength, hardness, WHC, and stability of the gel-like emulsions at 75 vol% oil; they also found that increasing protein contents could increase the bioaccessibility of β-carotene because of the reduced aggregation of the oil droplets and retarded degradation of β-carotene during digestion, owing to a dense WPI-based gel structure around droplets. In addition, the surface charge of (nano)particles can affect their emulsification and interfacial behavior (Larson-Smith, Jackson, & Pozzo, 2012). It has been reported that electrostatic screening by adding NaCl could improve the performance of soy glycine nanoparticles in forming gel-like emulsions and increase stiffness of the resultant gel-like emulsions, due to enhanced diffusion and adsorption of solid particles at the interface (Liu & Tang, 2016).

### *3.3.2. Disrupted gel systems*

Although there are few studies on the structure-property relationships of disrupted gel systems, Torres, Tena, Murray, & Sarkar (2017) found that increasing starch contents (from 15 to 20 wt%) and oil fractions (from 0 to 20 wt%) could improve the elastic modulus of starch-based disrupted gels stabilized by octenyl succinin anhydride (OSA) modified starch, which fitted the Kerner model. It has been reported that, compared to alginate-based emulsions and bulk emulsion gels, sheared oil-in-gel (o/g) emulsions exhibited higher bioaccessibility of encapsulated β-carotene after *in vitro* digestion, due to the lower unbound calcium content and higher colloidal stability throughout gastrointestinal passage, whereas encapsulated β-carotene in the bulk emulsion gels exhibited highest chemical stability (Soukoulis, Cambier, Hoffmann, & Bohn, 2016).

## 4. Applications of emulsion gels in the food industry

### 4.1. Use of emulsion gels as fat replacers in meat products

Emulsion gels formed by myofibrillar proteins (MPs), water and lipid not only contribute to the sensory properties (appearance and flavor) but also relate to the textural properties (water- and oil-holding, and cooking losses) of meat products (Wang, et al., 2018; Zhao, Zou, Shao, Chen, Han, & Xu, 2017). Additives, such as extracts from herbs and spices, polyphenols, and NaCl, can influence structures of emulsion gels and the properties of meat products (Wang, et al., 2018; Zhao, Zou, Shao, Chen, Han, & Xu, 2017). Wang et al. (2018) found that a low level of rosmarinic acid (RA) (12 μM/g protein) could protect thiol and ε-$NH_3$ groups in MP-based emulsion gels from oxidation, and thus improve the structure and water- and oil-holding abilities of emulsion gels; however, a high level of RA (300 μM/g protein) could induce interactions between RA and MPs, which led to aggregation of MPs and a poor emulsion gel network, while a high level of NaCl (0.6 M) could promote these interactions.

However, while health concerns around some meat products containing high fat content (over 27%) have increased in recent years, reducing fat content usually negatively influences consumer acceptance and textural properties of final products (Oliver, Scholten, & van Aken, 2015). In order to avoid undesirable textural changes and improve the nutritional value of meat products (e.g., sausages and patties), promising methods have been studied, such as replacing fat with unsaturated oil (Oliver, Scholten, & van Aken, 2015) or structured oil (e.g., olive, linseed, fish, perilla, and sunflower seed oil encapsulated in emulsion gels formed with SPI, WPI, sodium caseinate, carrageenan, gelatin, alginate, chia flour, oat bran, or inulin) (Alejandre, Poyato, Ansorena, & Astiasaran, 2016; de Souza Paglarini, de Figueiredo Furtado, Biachi, Vidal, Martini, Forte, et al., 2018; de Souza Paglarinia, Martinib, & Pollonio, 2019; Freire, Cofrades, Perez-Jimenez, Gomez-Estaca, Jimenez-Colmenero, & Bou, 2018; Freire, Cofrades, Serrano-Casas, Pintado, Jimenez, & Jimenez-Colmenero, 2017; Glisic, et al., 2019; Pintado, Herrero, Jimenez-Colmenero, Pasqualin Cavalheiro, & Ruiz-Capillas, 2018; Poyato, Astiasarán, Barriuso, & Ansorena, 2015; Serdaroglu, Nacak, & Karabiyikoglu, 2017). However, these methods may lead to undesirable sensory quality changes (e.g., color parameters and sensory acceptability) (Serdaroğlu & Öztürk, 2017). Oliver, Scholten, & van Aken (2015) found that physical properties of fat or oil and structural properties of the gel matrix could influence the rheological properties of fat-filled emulsion gels or oil-filled emulsion gels. Hence, the properties of fat in meat products should be considered, and the gelling agent and oil should be chosen carefully when emulsion gels are used as a fat replacer

(Freire, Cofrades, Serrano-Casas, Pintado, Jimenez, & Jimenez-Colmenero, 2017). It has been reported that combining emulsion gels and animal fat could be a good method to produce healthier meat products with acceptable sensory properties (de Souza Paglarini, et al., 2019). In addition, emulsion gels help to control sodium availability and perception by changing sodium mobility and binding behavior, and can thus allow reduction of the salt content in meat products (Okada & Lee, 2017). However, most studies have focused on bulk emulsion gels and their uses in solid foods, and so more studies on emulsion gel particles and their uses in liquid foods are needed.

## 4.2. Emulsion gels used as delivery systems to encapsulate and release food nutriments

Absorption of encapsulated lipophilic food nutrients (e.g., β-carotene, curcumin, n-3 fatty acid, vitamin A, and α-tocopherol) in emulsion gels include several steps: release from the gel matrix as the result of mechanical, chemical and enzymatic processes throughout the oral processing and gastrointestinal passage, incorporation in the co-digested lipid droplets, interaction with endogenous lipid surface active compounds (mainly bile salts and phospholipids) promoting the formation of mixed micelles, and eventual transportation of the mixed micelles to the small intestinal epithelium (Soukoulis, Cambier, Hoffmann, & Bohn, 2016; Yonekura & Nagao, 2007). Polysaccharides (e.g., alginate, κ-carrageenan, and starch) and proteins (e.g., gelatin and WPI) are normally used as gelation materials in producing emulsion gels encapsulating lipophilic food nutrients, but their digestion behaviors differ. Protein-based emulsion gels are mainly disrupted in gastric digestion as the result of enzymatic hydrolysis by pepsin, and the remaining protein-based network structures can hinder further breakdown during intestinal digestion (Guo, Ye, Lad, Dalgleish, & Singh, 2016; Liang, Leung Sok Line, Remondetto, & Subirade, 2010). On the other hand, polysaccharide-based (especially alginate-based) emulsion gels are less sensitive to gastric fluid than protein-based emulsion gels, and may protect the encapsulated nutriments from harsh gastric environment, and the remaining gel structures can be further disrupted during intestinal digestion (Wang, Neves, Kobayashi, Uemura, & Nakajima, 2013; Xu, et al., 2019). However, emulsion gels normally give low effective bioavailability of encapsulated lipophilic compounds, due to insufficient digestion of the gel matrix and resulting unreleased and undigested lipid phase (Liang, Leung Sok Line, Remondetto, & Subirade, 2010; Zhang, et al., 2016). Therefore, it is important to choose appropriate materials for different nutrients, which can protect encapsulated nutrients and control their release, and also do not inhibit release in the targeted gastrointestinal tract (Zhang, et al., 2016). Although emulsion gels may not improve the final bioaccessibility of encapsulated food nutrients, they can improve emulsion structures and stability of nutrients during storage, and exhibit slow release effects in the gastrointestinal passage compared to emulsions (Brito-Oliveira, Bispo, Moraes, Campanella, & Pinho, 2017; Ma, Wan, & Yang, 2017; Soukoulis, Cambier, Hoffmann, & Bohn, 2016; Zhang, et al., 2016).

## 5. Conclusions

Various preparation methods of emulsion gels are available for different matrix materials (e.g., heat treatment, enzyme treatment, acidification treatment, and addition of ions for protein-based emulsion gels, cold-set and addition of ions for polysaccharide-based emulsion gels, and self-assembly for LMW compound-based emulsion gels), purposes (e.g., cold treatment for protecting encapsulated nutrients and better mechanical properties), and emulsion gel types (e.g., internal gelation for bulk emulsion gels, external gelation for emulsion gel particles, self-assembly for gel-like Pickering emulsions, and mechanical stir for disrupted emulsion gels). Due to differences in the morphological properties among different emulsion gels, different physical properties are emphasized, such as the importance of mechanical and release properties for bulk emulsion gels, syneresis and swelling properties for emulsion gel particles, rheological properties for microgel particle suspensions, and flow behaviour and release property for fluid emulsion gels. In terms of bulk emulsion gels, many factors (e.g., structures of gel matrix and emulsion droplets and interactions between them) can influence their structures and thus mechanical and release properties. Structures of the gel matrix in bulk emulsion gels are affected by matrix material, preparation method, and process parameters, while structures of emulsion droplets are affected by oil type, oil content, droplet size, and emulsifier type. In terms of emulsion gel particles, oil content and particle size can influence their syneresis and swelling properties. The rheological properties of microgel particle suspensions are influenced by volume fraction, particle modulus, and interaction potential. In terms of gel-like Pickering emulsions, their rheological and release properties also are influenced by many factors (e.g., oil content, particle content, and surface charge of particles). Finally, two main applications of emulsion gels in the food industry are as fat replacers in meat products and delivery systems for food nutrients.

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**Table 1**

Selected examples of materials and methods used to prepare bulk emulsion gels.

| Materials | Methods | Matrix | Emulsifier/oil category and content | Structure | References |
|---|---|---|---|---|---|
| Protein | Heat treatment | Myofibrillar protein (MP) | MP/soybean oil (φ = 0.25), peanut oil (5%) or lard and peanut oil (0–15%) | O/W | (Feng, et al., 2017; Wang, et al., 2018; Wu, Xiong, & Chen, 2011; Wu, Xiong, Chen, Tang, & Zhou, 2009) |
| | | Myofibrillar protein | SPI/canola oil (10%) | O/W | (Jiang & Xiong, 2013) |
| | | Chicken protein isolate (CPI) | CPI/pork backfat (20%) | O/W | (Zhao, Zou, Shao, Chen, Han, & Xu, 2017) |
| | | Whey protein isolate (WPI) | WPI, glycerol monopalmitate, Tween 20, DGML, DGMO, or lecithin/canola oil (20%), soybean oil (20%), sunflower oil (φ = 0.05–0.25) or triolein (φ = 0.3) | O/W | (Chen & Dickinson, 1998a; Chen & Dickinson, 1999a; Chen & Dickinson, 1999b; Chen, Dickinson, Langton, & Hermansson, 2000; Guo, Bellissimo, & Rousseau, 2017; Guo, Ye, Lad, Dalgleish, & Singh, 2013; Gwartney, Larick, & Foegeding, 2004) |
| | One-step cold-set | Cold-soluble gelatin | No emulsifier/olive oil (52%) | O/W | (Pintado, Ruiz-Capillas, Jimenez-Colmenero, Carmona, & Herrero, 2015) |
| | Cold-set after heat treatment | Gelatin | No emulsifier/sunflower oil (φ = 0.3) | O/W | (Sato, Moraes, & Cunha, 2014) |

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|---|---|---|---|---|
| | Gelatin | WPI/sunflower oil or fat (50% in emulsions) or medium-chain triglycerides (40% in emulsions) | O/W emulsion filled | (Oliver, Berndsen, van Aken, & Scholten, 2015; Oliver, Scholten, & van Aken, 2015; Sala, van Vliet, Cohen Stuart, Aken, & van de Velde, 2009) |
| Enzyme treatment (TGase) | Myofibrillar protein | SPI or MP/canola oil (10% or 25%) | O/W | (Jiang & Xiong, 2013; Wang, Xiong, & Sato, 2017) |
| | Soy protein isolate (SPI) | SPI/soy oil (φ = 0.2–0.6) | O/W | (Tang, Luo, Liu, & Chen, 2013; Tang, Yang, Liu, & Chen, 2013; Tang, Chen, & Foegeding, 2011) |
| | Bovine serum albumin | Bovine serum albumin/*n*-tetradecane (φ = 0.45) | O/W | (Kang, Kim, Shin, Woo, & Moon, 2003) |
| | Sodium caseinate | Sodium caseinate/olive oil (52%) or sunflower oil (45%) | O/W | (Lim, Kim, Choi, & Moon, 2015; Pintado, Ruiz-Capillas, Jimenez-Colmenero, Carmona, & Herrero, 2015) |
| | Sodium caseinate | PGPR/perilla oil (80% in $W_1$/O) | $W_1/O/W_2$ | (Freire, Bou, Cofrades, & Jimenez-Colmenero, 2017) |
| | Gelatin | Sodium caseinate/perilla oil (80% in $W_1$/O) | $W_1/O/W_2$ | (Flaiz, Freire, Cofrades, Mateos, Weiss, Jimenez-Colmenero, et al., 2016) |
| Acidification treatment (GDL/citric acid) | Soy protein | Soy protein/soy oil (40% or φ = 0.2–0.3) | O/W | (Fang Li & Hua, 2013;Li, Kong, Zhang, & Hua, 2012; Tang, Chen, & Foegeding, 2011) |

| | | | | |
|---|---|---|---|---|
| | Whey protein isolate | WPI/sunflower oil (20%) or milk fat (20–30%) | O/W | (Mao, Roos, & Miao, 2014; Ye & Taylor, 2009) |
| | Sodium caseinate | Sodium caseinate/sunflower oil (10%), vegetable fat (30%) or n-tetradecane (φ = 0.3) | O/W | (Chen & Dickinson, 2000; Dickinson & Merino, 2002; Kiokias & Bot, 2005; Montes de Oca-Ávalos, Huck-Iriart, Candal, & Herrera, 2016) |
| | Micellar casein isolate | WPI or casein/sunflower oil or fat (50% in emulsions) or milk fat (5–25% in emulsions) | O/W emulsion filled | (Oliver, Scholten, & van Aken, 2015; Oliver, Wieck, & Scholten, 2016) |
| | Whey protein isolate | WPI, Tween 20, or lactoferrin/sunflower oil or fat (50% in emulsions) or medium-chain triglycerides (40% in emulsions) | O/W emulsion filled | (Oliver, Scholten, & van Aken, 2015; Sala, van Vliet, Cohen Stuart, Aken, & van de Velde, 2009) |
| Addition of ions | Soy protein | Soy protein/soy oil (φ = 0.2) | O/W | (Tang, Chen, & Foegeding, 2011) |
| | Whey protein isolate or β-lactoglobulin | Proteins/sunflower oil (30%) or milk fat (20–30%) | O/W | (Liang, Leung Sok Line, Remondetto, & Subirade, 2010; Sok Line, Remondetto, & Subirade, 2005; Ye & Taylor, 2009) |
| Malondialdehyde (MDA) | Myofibrillar protein | MP/soybean oil (20%) | O/W | (Zhou, Sun, & Zhao, 2015) |

| | | | | | |
|---|---|---|---|---|---|
| | modification | | | | |
| Protein/protein | Heat treatment | Micelle casein/whey protein isolate | Proteins/sunflower oil (5–15%) | O/W | (Balakrishnan, Nguyen, Schmitt, Nicolai, & Chassenieux, 2017) |
| | Enzyme treatment (Tyrosinase) | Potato protein/zein | Potato protein and zein/olive oil (φ = 0.4) | O/W | (Glusac, Davidesko-Vardi, Isaschar-Ovdat, Kukavica, & Fishman, 2018) |
| Polysaccharide | Cold-set after heat treatment | κ-Carrageenan | Polysorbate 80 or no emulsifier/sunflower oil (40%) or corn oil with monoglycerides (25–75%) | O/W or bigels | (Poyato, Astiasarán, Barriuso, & Ansorena, 2015; Zheng, Mao, Cui, Liu, & Gao, 2020) |
| | | Gellan gum | Tween 80/sunflower oil (10–30%) | O/W | (Lorenzo, Zaritzky, & Califano, 2013) |
| | | Agar | Polyglycerol esters of fatty acids/soybean oil (20% in emulsions) or corn oil (φ = 0.3 in emulsions) | O/W emulsion filled | (Kim, Gohtani, Matsuno, & Yamano, 1999; Wang, Neves, Kobayashi, Uemura, & Nakajima, 2013) |
| | | κ-Carrageenan | WPI or Tween 20/medium-chain triglycerides (40% in emulsions) | O/W emulsion filled | (Sala, van Vliet, Cohen Stuart, Aken, & van de Velde, 2009) |
| | Addition of ions | Alginate | No emulsifier/sunflower oil (φ = 0.3 or 52%) or chia oil (40%) | O/W | (Herrero, Ruiz-Capillas, Pintado, Carmona, & Jiménez-Colmenero, 2018; Pintado, Ruiz-Capillas, Jimenez-Colmenero, Carmona, & Herrero, 2015; Sato, Moraes, & Cunha, 2014) |

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|---|---|---|---|---|---|
| | Self-assembly (crystallisation) | Inulin | Soy lecithin/olive oil (21–38%) | O/W | (Paradiso, Giarnetti, Summo, Pasqualone, Minervini, & Caponio, 2015) |
| Polysaccharide/ polysaccharide | Self-assembly (compatibility) | Alginate/konjac glucomannan | Egg yolk or Tween 80/rapeseed oil (10–60% in emulsions) | O/W emulsion filled | (Yang, Gong, Lu, Li, Sun, & Guo, 2020) |
| | | Xanthan/konjac glucomannan | Tween 80/rapeseed oil (20%) | O/W emulsion filled | (Yang, Gong, Li, Li, Sun, & Guo, 2019) |
| Protein/ polysaccharide | Cold-set after heat treatment | Whey protein isolate/xanthan gum | Span 80 and Tween 60/babacu seed oil (2.8%) and tristearin (1.2%) | O/W emulsion filled | (Geremias-Andrade, Souki, Moraes, & Pinho, 2017) |
| | | Soy protein isolate/xanthan gum | Span 80 and Tween 80/tristearin (4.5%) | O/W emulsion filled | (Brito-Oliveira, Bispo, Moraes, Campanella, & Pinho, 2017) |
| | | Gelatin/Agar | WPI/sunflower oil (40% in emulsions) | O/W emulsion filled | (Devezeaux de Lavergne, Tournier, Bertrand, Salles, van de Velde, & Stieger, 2016) |
| | Enzyme treatment (TGase or laccase) | Soy protein isolate/sugar beet pectin (SBP) | Tween 20, SPI, SBP or SPI and SBP/corn oil (15%) or medium-chain triglyceride oil (10%) | O/W | (Feng, Jia, Zhu, Liu, Li, & Yin, 2019; Hou, Guo, Wang, & Yang, 2016) |
| | Acidification treatment (GDL) | Whey protein isolate/carrageenan | WPI/canola oil (50%) | O/W | (Lam & Nickerson, 2014) |
| | Heat treatment and addition of ions | Gelatin/alginate | No emulsifier/sunflower oil (φ = 0.3) | O/W | (Sato, Moraes, & Cunha, 2014) |

| | | | | | |
|---|---|---|---|---|---|
| | | Whey protein concentrate/Persian gum | No emulsifier/milk fat (1%) | O/W | (Khalesi, Emadzadeh, Kadkhodaee, & Fang, 2019) |
| | Self-assembly (electrostatic attraction) | Egg yolk protein/alginate (pH < pKa of proteins) | Egg yolk protein/rapeseed oil (30%) | O/W emulsion filled | (Yang, et al., 2020) |
| Organic compounds | Self-assembly | Sapoin glycyrrhizic acid | No emulsifier/sunflower oil, algal oil, and flaxseed oil (40%) | O/W | (Wan, Sun, Ma, Yang, Guo, & Yin, 2017) |
| | | *β*-Sitosterol/*γ*-Oryzanol | No emulsifier/sunflower oil (40–90%) | W/O | (Bot, den Adel, Regkos, Sawalha, Venema, & Flöter, 2011; Sawalha, den Adel, Venema, Bot, Floter, & van der Linden, 2012) |
| | | Sapoin glycyrrhizic acid | PGPR in $W_1$/O and no emulsifier in double emulsions/sunflower oil (70% in $W_1$/O) | $W_1/O/W_2$ | (Ma, Wan, & Yang, 2017) |

1208 **Table 2**

1209 Selected examples of materials and methods used to prepare emulsion gel particles.

| Materials | Methods | Matrix | Emulsifier/oil category and content | Structure | References |
|---|---|---|---|---|---|
| Polysaccharide | External gelation (ionic gelation) | Alginate | No emulsifier or WPI/canola oil (1–10%), safflower oil (10%), thyme essential oil (1%), or D-limonene | O/W | (Benavides, Cortes, Parada, & Franco, 2016; Ching, Bansal, & Bhandari, 2016; Corstens, et al., 2017; Lević, et al., 2015) |
| | | κ-Carrageenan or alginate | Tween 80/corn oil (10%) in emulsions | O/W emulsion filled | (Zhang, et al., 2016) |
| Protein/polysaccharide | External gelation (ionic gelation) | Pectin/WPI | WPI/oily solution of vitamin A (20%) | O/W | (Ruffin, Schmit, Lafitte, Dollat, & Chambin, 2014) |
| | | Alginate/WPI | WPI/sunflower oil (0.5–20%) | O/W | (Feng, Yue, Wusigale, Ni, & Liang, 2018) |
| | | Alginate/lupin protein | Lupin protein/linseed oil ($\varphi$ = 0.15–0.69) | O/W | (Piornos, Burgos-Díaz, Morales, Rubilar, & Acevedo, 2017) |

1210 **Table 3**

1211 Selected examples of materials and methods used to prepare fluid emulsion gels.

| Materials | Methods | Matrix | Emulsifier/oil category and content | Structure | References |
|---|---|---|---|---|---|
| Protein | Pickering emulsion/self-support | / | Soy glycinin nanoparticles/soy oil (φ = 0.1, 0.3, or 0.5) or unknown oil (φ = 0.1–0.89) | O/W | (Liu & Tang, 2016; Luo, Liu, & Tang, 2013; Xu, Liu, & Tang, 2019) |
| | | / | Pea protein isolate/soy oil (φ = 0.2–0.6) | O/W | (Shao & Tang, 2016) |
| | | / | WPI/camellia oil (φ = 0.75) | O/W | (Liu, Gao, McClements, Zhou, Wu, & Zou, 2019) |
| | | / | Casein peptides/unknown oil (61% and 77%) | O/W | (Wakita & Imura, 2018) |
| | | Sarcoplasmic protein | Sarcoplasmic protein/canola oil (50%) | O/W | (Hemung, Benjakul, & Yongsawatdigul, 2013) |
| | | / | Wheat gluten/corn oil (60%) | Oil-in-glycerol | (Liu, Chen, Guo, Yin, & Yang, 2016) |
| | Electrospinning emulsion/self-support | Gelatin | Gelatin/corn oil (φ = 0.2–0.8) | O/W | (Zhang & Zhang, 2018) |
| Polysaccharide | Pickering emulsion/self-support | Starch granule | Octenylsuccinate quinoa starch OSQS/corn oil (30–60%) | O/W | (Li, Zhang, Li, Fu, & Huang, 2020) |
| | Disrupted gel systems | Starch | OSA modified starch/sunflower oil (40%) | O/W emulsion | (Torres, Tena, Murray, & Sarkar, 2017) |

| | | | | | |
|---|---|---|---|---|---|
| | (homogenization) | | | filled | |
| | Disrupted gel systems (mechanical shearing) | Sodium alginate | Tween 80/canola oil (5%) | O/W | (Soukoulis, Cambier, Hoffmann, & Bohn, 2016) |
| Protein/polysaccharide | Pickering emulsion/self-support | / | Zein and gum arabic complex (ZGAPs)/medium-chain triglyceride oil (φ = 0.1–0.7) | O/W | (Dai, Sun, Wei, Mao, & Gao, 2018) |
| | | / | Zein and chitosan complex (ZCCPs)/algal oil (20–70%) | O/W | (Wang, Yin, Wu, Qi, Guo, & Yang, 2016) |
| | | / | Zein and tannic acid complex particles (ZTP)/corn oil (φ = 0.5) | O/W | (Zou, Guo, Yin, Wang, & Yang, 2015; Zou, Yang, & Scholten, 2018) |
| | | / | *β*-lactoglobulin and gum arabic complex/medium-chain triglyceride oil (φ = 0.3–0.7) | O/W | (Su, et al., 2020) |
| | Disrupted gel systems (mechanical shearing) | WPI/alginate | WPI/olive oil (5–25%) | O/W | (Leon, Medina, Park, & Aguilera, 2018) |

**Figure legends**

Fig. 1. Structures of two idealized models of emulsion gels: (A) emulsion droplet-filled gels, and (B) emulsion droplet-aggregated gels (Dickinson, 2012).

Fig. 2. Schematic presentation of two kinds of fillers in emulsion gels: (A) active fillers (droplets covered by black line), and (B) inactive fillers (droplets covered by white line).

Fig. 3. Visual appearances of alginate-based (A) bulk emulsion gels, (B) emulsion gel particles, and (C) fluid emulsion gels. Preparing alginate-based emulsion gels includes two steps: first preparing emulsions with 1 wt% sodium alginate and 0.5 wt% Tween 80 in water phase and sunflower oil at 40 wt% and then turning emulsions into gels. For the preparation of bulk emulsion gel, 0.5 wt% $CaCl_2$ was added to the emulsion, and the samples were allowed to gel for 6 h in stand. For the production of emulsion gel particles, the emulsion was dropped into a 2 wt% $CaCl_2$ solution, and the samples were allowed to gel in the $CaCl_2$ solution for 6 h with mild magnetic stirring. For producing fluid emulsion gel, 0.5 wt% $CaCl_2$ was added to the emulsion, and the mixture was sheared under constant paddle stirring at 600 rpm for 6 h.

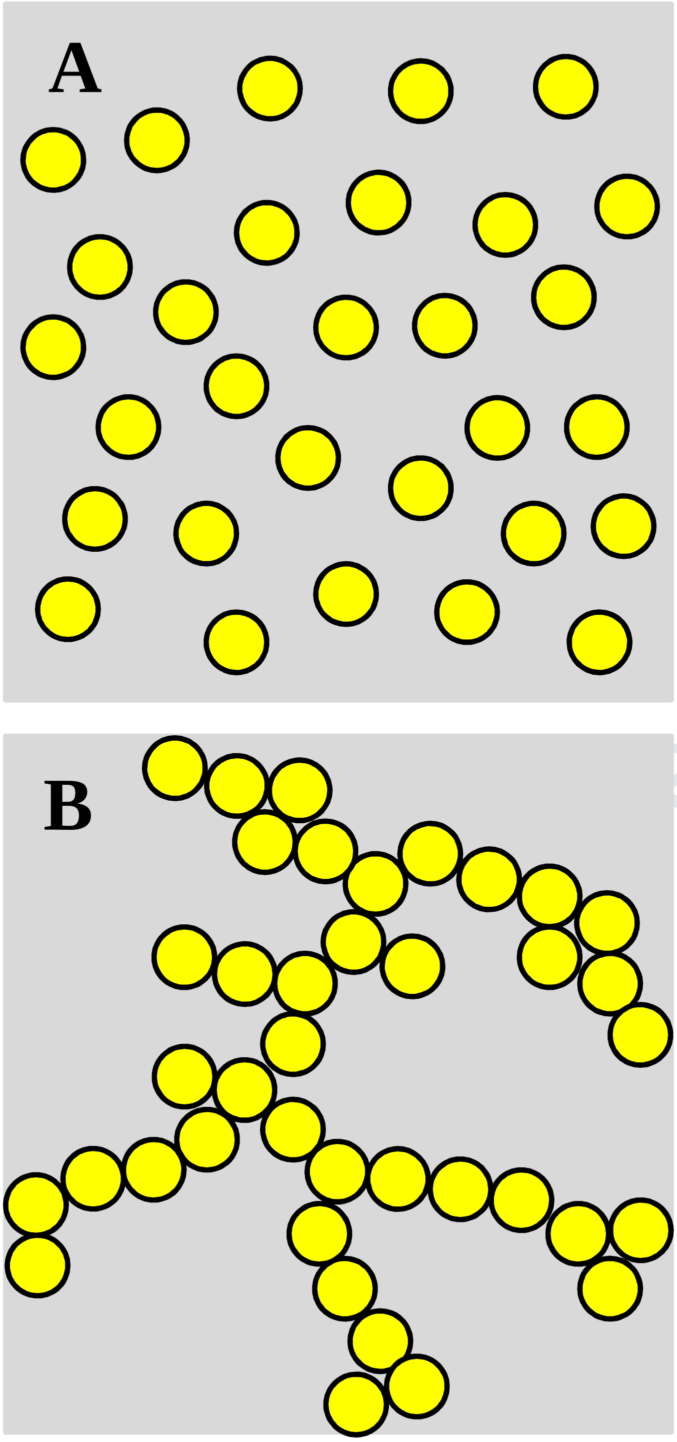


Fig. 1.

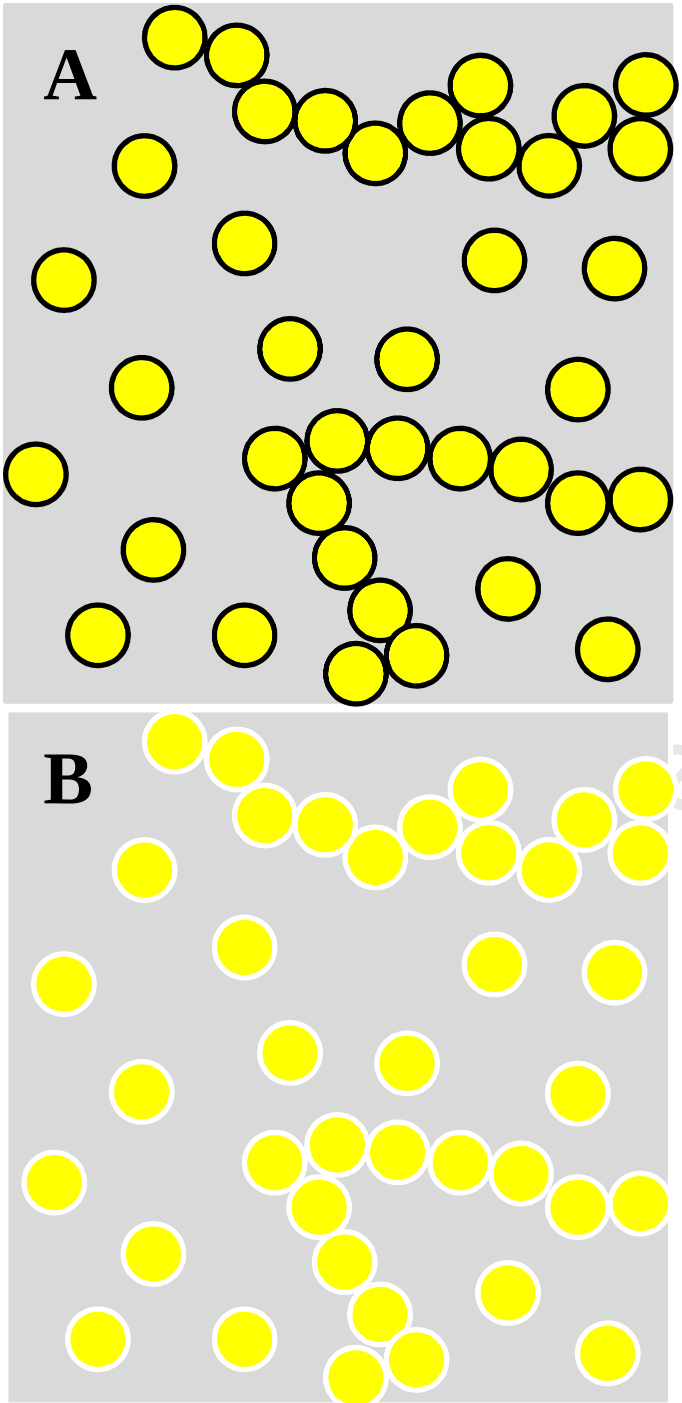


Fig. 2.

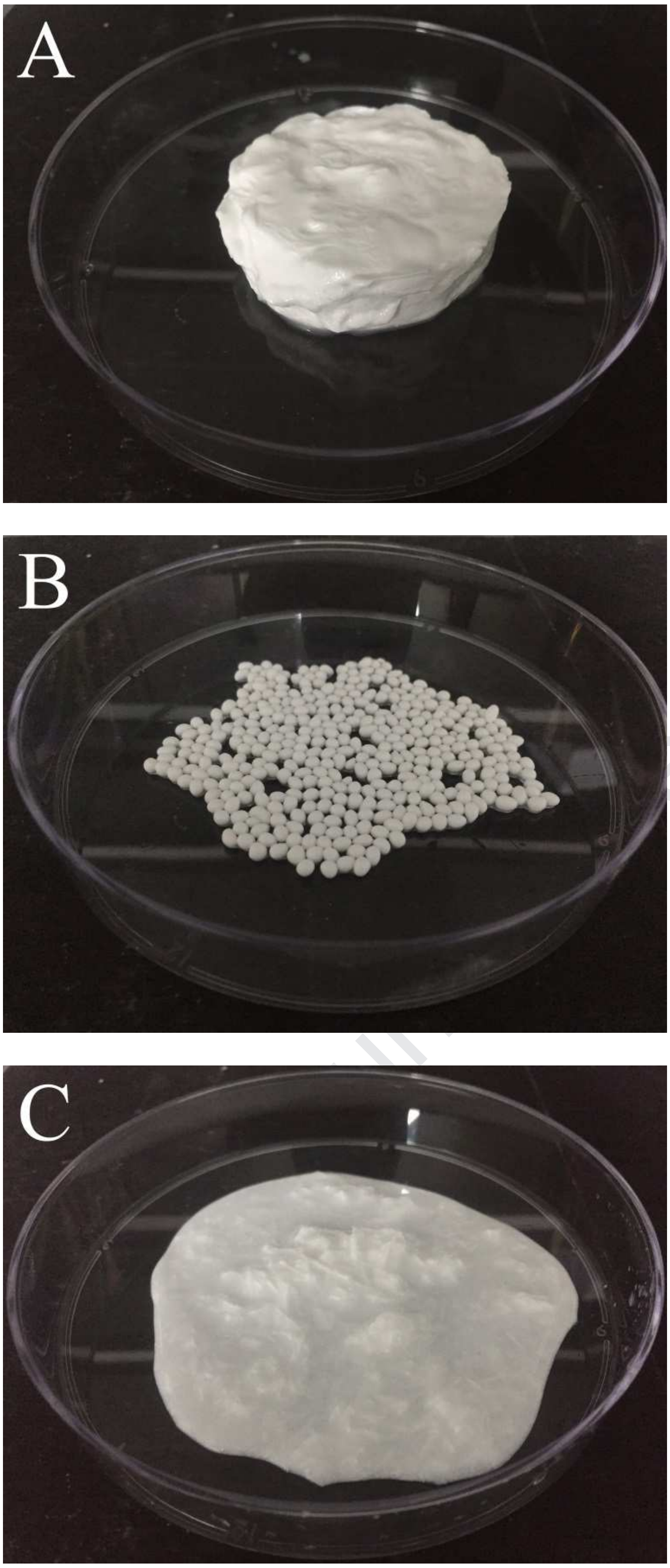


Fig. 3.

**Highlights**

- Preparation methods differ according to the emulsion gel types, gelling agents, and purposes.
- Different emulsion gels have different morphological properties and structure-property relationships.
- Structures of matrix and emulsion droplets can affect mechanical and release properties of bulk emulsion gels.
- Uses of emulsion gels as fat replacers and delivery systems were discussed.