Starch-based materials encapsulating food ingredients: Recent advances in fabrication methods and applications

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Abstract

Encapsulation systems have gained significant interest in designing innovative foods, as they allow for the protection and delivery of food ingredients that have health benefits but are unstable during processing, storage and in the upper gastrointestinal tract. Starch is widely available, cheap, biodegradable, edible, and easy to be modified, thus highly suitable for the development of encapsulants. Much efforts have been made to fabricate various types of porous starch and starch particles using different techniques (e.g. enzymatic hydrolysis, aggregation, emulsification, electrohydrodynamic process, supercritical fluid process, and post-processing drying). Such starch-based systems can load, protect, and deliver various food ingredients (e.g. fatty acids, phenolic compounds, carotenoids, flavors, essential oils, irons, vitamins, probiotics, bacteriocins, co-enzymes and caffeine), exhibiting great potentials in developing foods with tailored flavor, nutrition, sensory properties, and shelf-life. This review surveys recent advances in different aspects of starch-based encapsulation systems including their forms, manufacturing techniques, and applications in foods.

Keywords: Starch-based encapsulation systems; Food ingredients delivery; Starch modification; Food fortification; Functional food

Abbreviations

CD	Conjugated dienes
CFU	Colony-forming units
GIT	Gastrointestinal tract
HMT	Heat-moisture treatment
MCT	Medium-chain triglycerides
O/W	Oil-in-water
OSA	Octenyl succinic anhydride
POV	Peroxide value
RDS	Rapidly digestible starch
RS	Resistant starch
SC-CO_2	Supercritical carbon dioxide
SDS	Slowly digestible starch
SFEE	Supercritical fluid extraction of emulsions
SGF	Simulated gastric fluid
SIF	Simulated intestinal fluid

1 Introduction

The encapsulation of food compounds or substances is particularly important in the design and production of food products with improved quality features, as most bioactive food ingredients are easy to deteriorate and lose activity when being exposed to processing environments and harsh conditions such as oxygen, light, UV, and acidic or alkaline conditions. Encapsulation can also enable the controlled release and target delivery of associated ingredients (Drosou, Krokida, & Biliaderis, 2017; Fang & Bhandari, 2010). To realize encapsulation, various fabrication methods such as spray-drying (No & Shin, 2019; Sharif et al., 2017a), coacervation (Zhao et al., 2019), and extrusion (Chen et al., 2020; Poletto et al., 2019) have been used.

Starch, as a biopolymer resource in nature, can be used as an important encapsulation material due to its low cost, high availability, and diverse functionality (e.g. water retention and tailorable viscosity) (Hoyos-Leyva, Bello-Pérez, Alvarez-Ramirez, & Garcia, 2018d). More importantly, starch itself is an important food ingredient; by physicochemical modification, starch can possess good film-forming and emulsification properties, and suitable digestion resistance (No & Shin, 2019; Subpuch, Huang, & Suwannaporn, 2016), which are desirable properties for encapsulation. Some modified starches such as Capsul® (an OSA-modified starch) could form low-viscosity suspensions and avoid the agglomeration and film-formation before spray-drying (Santiago et al., 2016). Also, starch can form complexes with small molecules, which makes it highly suitable for developing encapsulation materials for food and wider applications (Ades, Kesselman, Ungar, & Shimoni, 2012; Ashwar, Gani, Gani, Shah, & Masoodi, 2018; Chen et al., 2017; Shao, Zhang, Niu, & Jin, 2018). In

102 particular, V-type inclusion complexes can be formed by amylose with hydrophobic functional
103 components such as fatty acids (Cui et al., 2021; Di Marco, Ixtaina, & Tomás, 2020; Fanta, Kenar, &
104 Felker, 2015), fatty acid esters of vitamins (Dries, Knaepen, Goderis, & Delcour, 2017b), phenolic
105 compounds (Wang, Chen, & Liu, 2020a), and volatile aroma compound (Shi, Hopfer, Ziegler, &
106 Kong, 2019), since the inner helical cavity of amylose is hydrophobic (Shi et al., 2019). Resistant
107 starch (RS) can resist the digestion and hydrolysis of all kinds of enzymes in the stomach and the
108 duodenum and can be degraded by colonic microbiota (Martínez-Ortiz et al., 2017; Muhammad,
109 Ramzan, Huo, Tian, & Bian, 2017).

110 In recent years, great efforts have been made to fabricate starch into porous starch, microgels,
111 molecular aggregates, starch granule aggregates, and other types of particles using different
112 techniques. These starch-based systems are capable of encapsulating, protecting, and delivering a
113 wide range of food components, and thus display great potentials in the development of innovative
114 food products with improved flavor, nutrition and sensory properties as well as extended shelf-life.
115 While some reviews (Garcia, Garcia, & Faraco, 2020; Hoyos-Leyva et al., 2018d; Qi & Tester, 2019;
116 Rodrigues & Emeje, 2012; Rostamabadi, Falsafi, & Jafari, 2019; Zhu, 2017) have summarized the
117 advances in starch-based encapsulation systems, they have their specific focuses (e.g. the application
118 prospects of certain encapsulation systems, different starch systems for encapsulation, or medical
119 applications). Boostani and Jafari (Boostani & Jafari, 2021) reviewed the fundamental concepts of
120 the controlled release of encapsulated food ingredients.

121 Complementary to these previous reviews, this article surveys the recent studies mainly in the
122 past ten years on starch-based encapsulation systems with a particular focus on three aspects related

to food applications: a) the types of starch-based encapsulation systems, b) their fabrication methods, and c) their recent applications for the encapsulation of food substances.

2 Types and fabrication methods of starch-based encapsulation systems

In the past decade, various starch-based encapsulation systems have been exploited. Below, basic aspects of starch modification facilitating encapsulation are presented, and then, different starch-based encapsulation systems and their fabrication methods are discussed briefly.

2.1 Basic aspects of starch modification

To fabricate starch-based encapsulants, various modification methods (physical, chemical and enzymatic modifications) have been used to improve the functional characteristics (e.g. water solubility, hydrophobicity, amphiphilicity, emulsifiability, digestion resistance, film-forming ability, thermal stability, and adsorption capacity) of native starches. Physical modification methods involve the use of moisture, heat, pressure, irradiation, and pressure. Chemical modification methods introduce functional groups onto starch molecules via chemical reactions (e.g. crosslinking, acetylation, esterification, oxidation, and acid hydrolysis) without altering the morphology or size distribution of starch granules (Alcázar-Alay & Meireles, 2015; Obadi & Xu, 2021). Enzymatic modification mainly involves the use of hydrolyzing enzymes to modify starch molecular or granule structures.

141 The physical modification of starch usually can improve the water solubility of starch and can
142 reduce the granule size of starch (Alcázar-Alay & Meireles, 2015). Maize starch treated with high
143 pressure showed a smaller particle size and could be used as an effective Pickering emulsion
144 stabilizer (Villamonte, Jury, & de Lamballerie, 2016). Furthermore, heat-moisture treatment (HMT)
145 is in favor of the formation of the ordered crystalline structure, which increases the RS content
146 (Noor, Shah, Gani, Gani, & Masoodi, 2018).
147 Cationization and hydroxypropylation could reduce the gelatinization temperature of starch by
148 disrupting double helices within the amorphous regions of starch granules (Singh, Kaur, &
149 McCarthy, 2007). Crosslinked starches showed increased gelatinization temperatures as crosslinking
150 reduces the mobility of amorphous chains within starch granules (Singh et al., 2007). Acetylation can
151 improve, for example, the water solubility and emulsion ability of starch by incorporating the
152 lipophilic alkenyl groups from, for example, OSA onto hydrophilic starch chains, which enhances the
153 amphiphilicity of starch (Jain, Winuprasith, & Suphantharika, 2019; Li, Fu, Luo, & Huang, 2013; No
154 & Shin, 2019; Spada, Noreña, Marczak, & Tessaro, 2012). Succinylation can weaken the internal
155 hydrogen bonding in starch granules (Arshad, Ali, & Hasnain, 2018). Moreover, the RS content of
156 starch can be improved after phosphorylation (Ashwar et al., 2018).
157 Enzymatic modification of starch can increase the amylose content in starch, which is beneficial
158 to the formation of V-type inclusion complexes and improve the thermal stability of these complexes
159 (Liu et al., 2019; Reddy, Lee, Lim, & Park, 2019; Wang et al., 2020a). Besides, enzymatic
160 modification can also result in hollows or pores on the granule (Dura, Błaszczak, & Rosell, 2014;
161 Zhang et al., 2012). The pores on the surface or in the interior of the starch granule give starch

suitable adsorption capacity and porous starch prepared by enzymatic hydrolysis could be used as an encapsulant (Lei et al., 2018).

2.2 Types

The types of starch-based encapsulation systems can be classified into the following categories: porous starch, microgels, molecular aggregates, starch granule aggregates, and other types of particles. These different forms of starch-based encapsulation systems and their fabrication methods are summarized in **Table 1** and discussed in detail below. Porous starch is a novel modified starch that has abundant tunable micro-sized pores extending from the surface to the center of the starch granule (Xie, Li, Chen, & Zhang, 2019). Porous starch has attracted considerable attention due to its high specific surface area, large pore volume, excellent adsorption performance, enhanced active sites, and good mechanical stability (Fang, Fu, Liu, & Cu, 2020). Due to its absorbability, biocompatibility, nontoxicity, and biodegradability, porous starch is a promising encapsulant for liquid bioactive substances (Lei et al., 2018).

The special “sponge-like” structure of porous starch greatly increases the specific surface area and thus can improve its performance as an encapsulant (Belingheri, Ferrillo, & Vittadini, 2015a; Lei et al., 2018). The porosity has a significant influence on the capacity, surface area, release kinetics, and efficiency of porous starch. Based on the pore size, porous starch could be classified into microporous (≤ 2nm), mesoporous (2–50 nm), macroporous (50–200 nm), and gigaporous materials (≥200 nm) (Oliyaei, Moosavi-Nasab, Tamaddon, & Fazaeli, 2020).

The advantages of using porous starch as a carrier to encapsulate flavors include not only high flavor-loading capacity but also has good retention of volatile molecules over time (Belingheri, Giussani, Rodriguez-Estrada, Ferrillo, & Vittadini, 2015b). It is very important to select a suitable solvent to dissolve liquid flavor, and the solvent that has similar polarity to flavor can maintain a higher flavor content over time than other solvents (Belingheri et al., 2015a). Belingheri et al. (Belingheri et al., 2015a) compared the liquid tomato flavor retention ability among three solvents including medium-chain triglycerides (MCT), propylene glycol, and triacetin. Their results showed that the products using MCT and triacetin as solvents for plating had similar flavor retention ability with the products produced by spray-drying (Belingheri et al., 2015a).

Selecting microporous starch to encapsulate oil via a simple plating procedure can avoid the oxidation of the oil caused by the heating step, and this step, which exists in spray-drying, usually can induce the initial oxidation of the encapsulated oil (Belingheri et al., 2015a; Belingheri et al., 2015b). Moreover, porous starch can also be used to carry plant oils (Belingheri et al., 2015b; Lei et al., 2018) and probiotics (Li, Thuy Ho, Turner, & Dhital, 2016a). Lei et al. (Lei et al., 2018) used purple sweet potato to prepare porous starch granules with a high loading ratio by enzyme treatment. Olive oil was encapsulated in microspheres by impregnating. In comparison with free olive oil, the encapsulated olive oil showed significantly enhanced oxidative stability (Lei et al., 2018).

Glenn et al. (Glenn et al., 2010) fabricated porous starch microspheres based on high-amylose maize starch via atomization and air classification and the mean particle size was divided into two groups, 5 μm and 100 μm. Small-size particles could both adsorb essential oil and maintain a dispersible powder state; In contrast, larger particles tended to gather together, which may be caused

by multiple spheres colliding with each other in a molten or partially molten state (Fang et al., 2020). The open structure of porous starch microspheres can facilitate the absorption of the encapsulated compounds but provide little resistance to evaporation (Glenn et al., 2010). To overcome this drawback, some researchers proposed adding a coating material on porous starch to form a composite (Benavent-Gil, Rodrigo, & Rosell, 2018; Li et al., 2016a). Gelatinized starch is frequently used as a coating material, and this additional shell can provide a better barrier and further protection against harsh environmental conditions (Benavent-Gil et al., 2018; Li et al., 2016a).

Microgels describe small particles whose size typically range from 100 nm to 1000 μm and has a three-dimensional network consisting of crosslinked biopolymer molecules that traps a considerable amount of solvent (usually water) (McClements, 2017). Thus, microgels are usually hydrogels. This type of particle is also sometimes referred to as nanogel, hydrogel bead, biopolymer particle, or microsphere (McClements, 2017). Biopolymer microgels are typically prepared using a two-step process involving particle formation and particle gelation (McClements, 2017). The internal structure of microgels can be homogeneous or heterogeneous, and the most common types of heterogeneous microgel have either a core-shell or dispersion structure (McClements, 2017). The particle gelation for starch-based microgel usually can be categorized into ionic gelation and cold-set gelation. Ionic gelation refers to mixing starch with polysaccharides (e.g. sodium alginate and pectin) containing negatively charged carboxyl groups to form hydrogels by electrostatic interaction between carboxyl groups and calcium ions (Fangmeier, Lehn, Maciel, & Volken de Souza, 2019; Poletto et al., 2019). Starch-based microgels could also be formed by cross-linking the oxidized starch or carboxymethyl starch with trisodium metaphosphate (Li et al., 2020a; Zhang et al., 2015; Zhang et al., 2017). Zhang

et al. (Zhang et al., 2017) reported a procedure of assembling chitosan and carboxymethyl starch on the surface layer of a microgel to prepare double-layer microgel complexes which could be used for intestinal-targeted drug delivery. These negatively charged microgels prepared by cross-linking reaction were mainly reported on loading drugs due to their excellent muco-adhesive and pH-responsive properties (Li et al., 2020a), and there are few reports on using such microgels to embed food components. Various materials (e.g. emulsified liquids, probiotic cells, and bioactive molecules) can be encapsulated in microgel particles (Dafe, Etemadi, Dilmaghani, & Mahdavinia, 2017; Mun, Kim, Shin, & McClements, 2015b; Torres, Tena, Murray, & Sarkar, 2017).

Emulsification is an important method to create microgels from colloids. It should be noted that hydrophobic bioactives need to be dissolved in lipid droplets or other hydrophobic vehicles before encapsulation. For example, Torres et al. (Torres et al., 2017) presented a systematic study on the formation of emulsion microgel particles (**Figure 1**). By mixing an O/W emulsion stabilized by OSA-modified waxy maize starch (a commercial product) with a native wheat gel and subjecting the mixture to refrigeration and high-pressure homogenization, emulsion microgel particles were obtained, which can be used for the delivery of lipophilic molecules. However, the encapsulation efficiency and stability of this type of particles still need further verification (Torres et al., 2017).

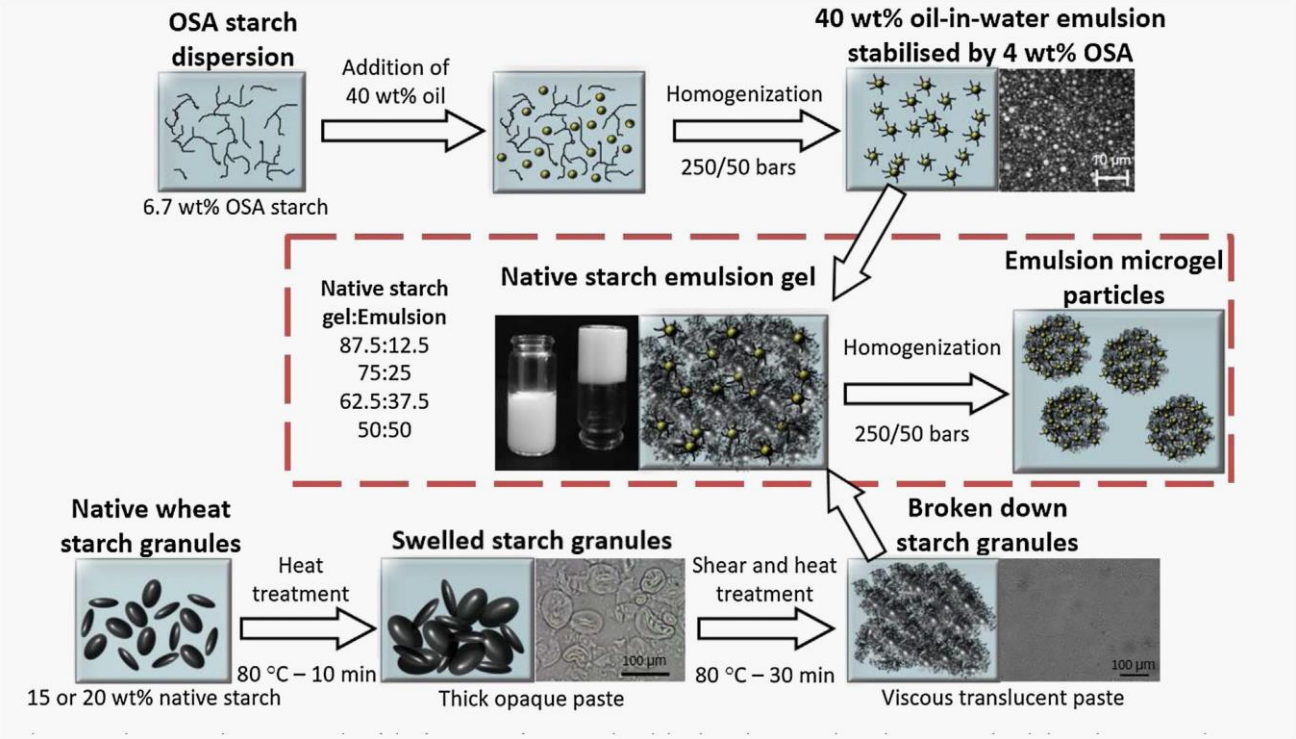


Figure 1 Schematic diagram and micrographs images of emulsion particles. Adapted from Ref. (Torres et al., 2017) with permission from Elsevier, Copyright 2017.

Aggregated particles can be categorized into those formed by molecular aggregation and those formed by starch granule aggregation. Molecular aggregation occurs in aqueous solutions based on starch alone or starch combined with other compounds. Examples of molecular aggregates are self-assembled aggregates, coacervates, and V-type starch inclusion complexes. For starch granule aggregation, some starches (e.g. taro starch and rice starch) can form spherical starch aggregates by spray drying their starch suspensions in the presence of bonding agents (e.g. carboxymethyl cellulose and gelatin) (Beirão-da-Costa, Duarte, Moldão-Martins, & Beirão-da-Costa, 2011; Hoyos-Leyva, Bello-Pérez, Agama-Acevedo, & Alvarez-Ramirez, 2018a; Hoyos-Leyva, Bello-Perez, Agama-Acevedo, Alvarez-Ramirez, & Jaramillo-Echeverry, 2019).

Apart from the types of particles mentioned above, particles can also be formed by just mixing the encapsulant and encapsulated material in different ways (in a solution or by extrusion). This latter group of particles is named as normal particles in this review.

The fabrication methods of starch-based encapsulation systems are discussed in detail **section 2.3** below.

2.3 Fabrication methods

Microcapsules can be prepared using different methods depending on the physical and chemical properties of the encapsulants and the encapsulated materials, the product application purposes, and the size, morphology, and release mechanism of the encapsulated materials. **Table 2** summarizes the synthesis methods of porous starches that are widely used at present and will be discussed in detail in **section 2.3.1**. **Table 3** lists some examples of encapsulation types based on different fabrication methods, along with the particle sizes and the advantages and disadvantages of the encapsulation systems, and these are discussed in detail from **section 2.3.2** to **section 2.3.8**.

2.3.1 Preparation of porous starch

Porous starch has been reported to be synthesized by enzymatic treatments (Benavent-Gil & Rosell, 2017; Benavent-Gil et al., 2018; Dura et al., 2014; Lei et al., 2018; Li et al., 2016a; Yang et al., 2019; Zhang et al., 2012), solvent exchange (Oliyaei, Moosavi-Nasab, Tamaddon, & Fazaeli, 2019; Oliyaei et al., 2020), microwaving (Majzoobi, Hedayati, & Farahnaky, 2015), combinations of physical methods and enzyme treatment (Majzoobi et al., 2015; Xie et al., 2019), a combination of gelatinization and atomization (Glenn et al., 2010), and a sacrifice template approach (Fang et al.,

2020). Among these fabrication methods, enzymatic treatment has drawn significant attention due to the high catalytic capability, mild reaction conditions, and substrate specificity (Dura et al., 2014). The widely used enzymes are α-amylase and glucoamylase. Maize starch granules naturally have pores, cavities, and channels, which make this type of starch more suitable to be modified into porous starch via enzymatic treatment compared to other starch cultivars (Li et al., 2016a).

The source of enzyme, enzymes/starch ratio, reaction time, temperature and pH can influence the pore size, pore frequency, and adsorption capacity of porous starch (Benavent-Gil et al., 2018; Lei et al., 2018; Li et al., 2016a; Yang et al., 2019). Lei et al. (Lei et al., 2018) reported that the adsorption capacity increased first and then decreased with an increase in the ratio of enzymes (α-amylase and glucoamylase)/purple sweet potato starch (<0.6%), reaction time (<12 h), temperature (<45 °C), and pH (<5). In this regard, the number, depth and diameter of pores increased with an increase in the reaction time, temperature, or pH. The highest adsorption capacity (~43.84%) was achieved with an enzymes/starch ratio of 0.6%, a reaction time of 12 h, a temperature of 45 °C, and a pH value of 5. Further increase in the enzymes/starch ratio, reaction time, temperature, and pH led to the breakage of the porous structure of starch granules into smaller fragments, thereby reducing the adsorption capacity (Lei et al., 2018).

2.3.2 Aggregation

Self-assembly, coacervation, and the formation of V-type starch complexes can all be classified as aggregation methods here. These methods rely on the interaction of starch molecules in an aqueous solution.

Self-assembly is an aqueous solution process in which hydrophobic groups of amphiphilic polymer molecules spontaneously form aggregates through intramolecular and intermolecular associations, and this process does not require stringent reaction conditions or solvents (Yang, Han, Zheng, Dong, & Liu, 2015). In addition, by controlling the electrostatic interaction of starches (e.g. carboxymethyl starch (anionic polyelectrolyte) and spermine-modified starch (cationic polyelectrolyte)) with oppositely-charged substances, colon-targeting delivery systems can also be constructed via self-assembly (layer-by-layer self-assembly) (Zhang et al., 2020b). Using OSA-esterified waxy maize starches with different molecular masses, Xiang et al. (Xiang et al., 2020) successfully prepared spherical molecular aggregation particles loaded with naringin via self-assembly. The starch sample with a molecular mass of 8.95×10^4 Da led to higher encapsulation efficiency and higher solubility of naringin than other OSA-esterified waxy maize starches (molecular masses were 1.41×10^4 Da, 2.21×10^4 Da, 12.82×10^4 Da, and 20×10^4 Da, respectively).

Coacervation, which can be either simple coacervation or complex coacervation, is a process of phase separation and further deposition of coacervates around the core materials (Vieira da Silva, Barreira, & Oliveira, 2016). The continuous shell can be formed through the interaction between coacervate microdroplets and the surface of a water-insoluble core material, followed by a drying process to obtain a coated powder. Since no heat treatment is involved in the process of coacervation, this method is suitable for encapsulating heat-labile compounds. Zhao et al. (Zhao et al., 2019) prepared complex coacervates based on the electrostatic attraction between gelatin and OSA-modified kudzu starch (1:1, w/w) to encapsulate astaxanthin extract. The degradation rate of

astaxanthin was significantly reduced in comparison with that with gelatin used as the encapsulant. While this way of encapsulation could reduce the degradation rate of the encapsulated material, the process is complex and time-consuming, which needs to be further addressed.

The complexation method mainly refers to the synthesis of V-type inclusion complexes. This method can be classified into four categories including the classical V-amylose preparation method, thermomechanical processing, enzymatic treatment, and low-temperature infusion (Dries et al., 2017a; Obiro, Sinha Ray, & Emmambux, 2012). The low-temperature infusion method does not need to use harsh chemicals and is performed at a relatively low temperature (Dries et al., 2017a; Dries et al., 2017b). Dries et al. (Dries et al., 2017b) used potato and maize starches as raw materials to produce cold-water swelling granule starches that had the V-type crystalline structure. Using the low-temperature infusion method, the starches formed V-type inclusion complexes with ascorbyl palmitate. The encapsulated ascorbyl palmitate still maintained a high antioxidant capacity, which was about 70% that of free ascorbyl palmitate (Dries et al., 2017b). The higher the amylose content, the higher was the ability of the starch to form inclusion complexes (Wang et al., 2020a; Wang, Zhan, Jin, & Tian, 2017).

2.3.3 Emulsification

Starch microspheres, especially for sustained-release purposes, are commonly prepared by emulsification followed by a drying process (Li, Xian, Wang, Adhikari, & Chen, 2018). This method can be classified into oil-in-water (O/W) emulsification (Majeed et al., 2016), water-in-water (W/W) emulsification (Yang et al., 2020a), and multiple emulsification (Fang, Zhao, Liu, Liang, & Yang,

2019). Multiple emulsions have attracted wide interest due to their ability to entrap encapsulated materials in an inner phase in the process of the primary emulsification procedure (Marefati, Sjöö, Timgren, Dejmek, & Rayner, 2015). Spray-drying is usually combined with emulsion to design encapsulation systems with desirable features, as summarized in **Table 3**. Moreover, freeze-drying is also used by some researchers for drying emulsions (Anwar & Kunz, 2011; Bilenler, Karabulut, & Candogan, 2017; Hasani, Ojagh, & Ghorbani, 2018; Marefati et al., 2015; No & Shin, 2019; Yildiz et al., 2018). It is reported that various bioactive compounds (e.g. probiotics (Bilenler et al., 2017), pigment (No & Shin, 2019), fatty acids (Yildiz et al., 2018), polyphenols (Wang et al., 2020b), and β-carotene (Fang et al., 2019; Liang, Huang, Ma, Shoemaker, & Zhong, 2013; Sharif et al., 2017a)) have been encapsulated in emulsion and further processing allows the formation of emulsion particles or microgel particles.

The traditional emulsion preparation method usually relies on the incorporation of an emulsifier and a surface-active agent to form stable emulsion droplets (Fangmeier et al., 2019). This method is neither cost-effective nor environmentally friendly due to the addition of plant oils and synthetic surfactants (Zhu, 2019). There is a trend to use modified starch as both the emulsifier and stabilizer during the preparation of emulsions (Lin et al., 2020; Marefati et al., 2015; Sharif et al., 2017b; Yusoff & Murray, 2011), and this type of emulsions are called Pickering emulsions. It is worth noting that the emulsion was stabilized by starch granules, not starch molecules (Yusoff & Murray, 2011). The botanic source, granule size and shape, starch concentration, and emulsion time all influence the size of emulsion droplets and the stability of the emulsion (Dickinson, 2012; Ge et al., 2017; Saari, Rayner, & Wahlgren, 2019). Mechanical stirring, homogenization, and sonication can assist

emulsification by generating a more dispersed emulsion. Microfluidic emulsification is an emerging method that can achieve unprecedented control of the composition, structure, size, and mono-dispersity of emulsion droplets (Wang, Zhang, & Chu, 2014a). In addition, to avoid the broad size distribution and excessively large size of starch microspheres obtained via traditional emulsion cross-linking technique, some researchers proposed using room-temperature ionic liquids (IL) (e.g. 1-hydroxypropyl-3-methylimidazolium acetate) as the solvent of OSA-modified normal maize starch to prepare IL microemulsions (IL/O) and further synthesize starch nanoparticles using epichlorohydrin as a crosslinker via emulsion cross-linking reaction (Qi, Ji, Luo, Xiao, & Yang, 2017).

Lin et al. (Lin et al., 2020) used octenylsuccinate quinoa starch (OSQS) as a stabilizer to prepare a double-Pickering emulsion (W1/O/W2) loaded with anthocyanin (**Figure 2I**) and the inner W1/O emulsion was stabilized using polyglycerol polyricinoleate (PGPR). The droplet size of W1/O increased during 7 days of storage due to the water diffusion from the W2 phase to the W1 phase (**Figure 2II**). This novel emulsion had a less than 15% release amount of anthocyanin under simulated stomach conditions and allowed for the controlled release of anthocyanin in the simulated intestinal fluid (SIF). An emulsion is just the precursor of emulsion particles. No and Shin (No & Shin, 2019) utilized OSA-modified waxy maize starch as a stabilizer and ultrasound treatment to prepare an O/W emulsion for the encapsulation of paprika pigment, which was dried via spray-drying and freeze-drying to obtain emulsion particles. The freeze-dried emulsion particles showed better color stability than the spray-dried ones (No & Shin, 2019).

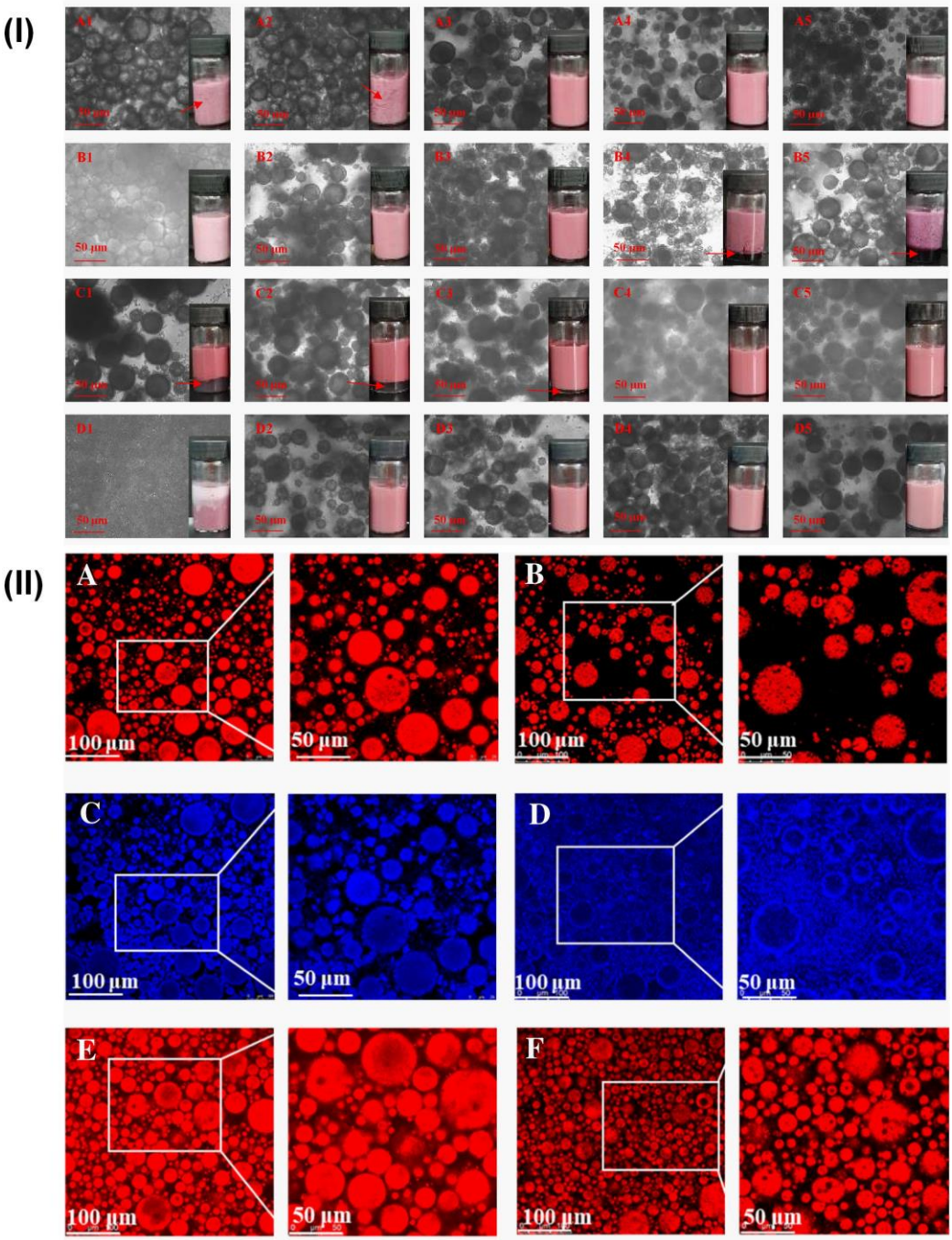


Figure 2 I) Effects of PGPR concentration (0.5%, 1.0%, 2.0%, 4.0%, and 6.0%), W1/O volume ratio (1:9, 2:8, 3:7, 4:6, and 5:5), OSQS concentration (1%, 2%, 4%, 6%, and 8%), and (W1/O)/W2 volume ratio (7:3, 6:4, 5:5, 4:6, and 3:7) on the formation of double-Pickering emulsion (stabilized by OSQS) loaded with anthocyanin (the red arrows refers to the W1 phase sinking to the bottom); II) Confocal laser scanning microscopy (CLSM) images revealing the storage stability of anthocyanin-

loaded double Pickering emulsion after storage of 7 days (the oil phase appeared red and the OSQS granules appeared blue). Adapted from Ref. (Lin et al., 2020) with permission from Elsevier, Copyright 2020.

2.3.4 Supercritical fluid process

The supercritical fluid process is a relatively mild encapsulation process (Almeida et al., 2013). Compared with traditional encapsulation techniques, the supercritical fluid process is superior in the control of morphology, particle size, and size distribution and can overcome the degradation of thermally labile compounds (Saldaña, dos Reis Coimbra, & Cardozo-Filho, 2015; Temelli, 2018). At present, there have limited reports on the application of these techniques in food-related fields whereas they have shown great potential (Temelli, 2018). Supercritical solvent impregnation (SSI) and supercritical fluid extraction of emulsions (SFEE) are techniques commonly used for starch-based encapsulation systems at present (Aguiar, Silva, Rezende, Barbero, & Martínez, 2016; Almeida et al., 2013; Cruz, Lima Reis, Ferreira, Masson, & Corazza, 2020; Lee, Tan, Sulaiman, Smith, & Chong, 2018; Santos, Martín, Meireles, & Cocero, 2012). In this process, the widely used solvent is supercritical carbon dioxide (SC-CO_2) (Janiszewska-Turak, 2017). The low operating temperature of the supercritical fluid process and the non-toxicity and easy removal of the supercritical fluid involved make this technique suitable for encapsulating substances that are prone to oxidative degradation such as essential oils (Almeida et al., 2013) and carotenoids (Mezzomo et al., 2012).

SFEE has high encapsulation efficiency and can maintain the antioxidant activity of the encapsulated products (Cruz et al., 2020; Reis et al., 2019). Besides, the high diffusivity of SC-CO_2 in a starch matrix can achieve the deep impregnation of essential oils via SSI (Almeida et al., 2013). It is worth noting that SFEE needs to be combined with emulsification. Cruz et al. (Cruz et al., 2020) used modified waxy maize starch as an encapsulant to prepare an emulsion loaded with yacon leaf extract, which has a high concentration of phenolic antioxidants, and finally prepared microparticles via SFEE. The obtained microparticles showed high antioxidant activity than unencapsulated yacon leaf extract (Cruz et al., 2020).

2.3.5 Extrusion

According to the condition of extrusion and the type of extrudate, extrusion can be categorized into two types: hot-melt extrusion (involving screws) and injection extrusion (a screwless form). As a continuous high-temperatures screw-extrusion process, hot-melt extrusion usually requires that the encapsulants and encapsulated food ingredients be able to tolerate high temperatures and possess high flow properties (Bamidele & Emmambux, 2020).

Starch could be converted into a homogenous molten state during a hot-melt extrusion process (Chen et al., 2020). The molecular entanglement that occurred in this process provides the possibility of the encapsulation of bioactive substances (Chen et al., 2020). Injection extrusion combined with vibration technology enables the production of capsules of similar size and shape. For the formation of particles via injection extrusion, the droplets usually need to be solidified in a crosslinking solution (e.g. $CaCl_2$) to achieve ionic gelation (Dafe et al., 2017; Poletto et al., 2019).

Using injection extrusion, Dafe et al. (Dafe et al., 2017) prepared a hydrogel consisting of starch and pectin as a novel type of food-grade hydrogel particles to encapsulate *Lactobacillus plantarum*. The encapsulated probiotic cells exhibited higher viability in the simulated gastric fluid (SGF) and the SIF conditions than the non-encapsulated cells. The experimental results showed that the encapsulation efficiency increased from 72.2 to 94.8 with an increasing starch content and the hydrogel could stand against the harsh simulated gastrointestinal conditions. Regarding this, the increased starch content caused a denser hydrogel network and further resisted the diffusion of acid into the hydrogel (Dafe et al., 2017). In another study, a probiotic, *Lactobacillus acidophilus*, was encapsulated in a composite of sodium alginate and Hi-Maize® (a commercial RS) via a method of injection extrusion combined with external ionic gelation, and the composite encapsulant allowed for the viability of the probiotic at 25 °C for 120 days (Poletto et al., 2019).

Both hot-melt extrusion and injection extrusion are simple and cost-effective methods, require limited amounts of solvents, and involve mild process conditions (Obadi & Xu, 2021)(Poletto et al., 2019).

2.3.6 Fluidized bed coating

Fluidized-bed coating (spray granulation) is a one-step process to prepare coated pellets, and the most commonly used techniques are the bottom-spray fluidized-bed and top-spray fluidized-bed processes (María Chávarri, Marañón, & Villarán). The particles with good flowability and a narrow size distribution enter into the fluidized bed and are suspended due to the bottom air; then, the coating material from the bottom liquid flow is sprayed onto the particles, followed by the formation

of a coating with the evaporation of the solvent (Bachmann, Chen, Bück, & Tsotsas, 2020; Hoyos-Leyva, Chavez-Salazar, Castellanos-Galeano, Bello-Perez, & Alvarez-Ramirez, 2018e). This method can be used to encapsulate probiotics and flavor compounds (Pellicer et al., 2019; Pitigraisorn, Srichaisupakit, Wongpadungkiat, & Wongsasulak, 2017). During the process of fluidized-bed coating, the starch suspension is atomized and then coat the particles. The partially gelatinized starch granules on the microcapsules could improve the barrier ability of microcapsules against moist-heat penetration (Pitigraisorn et al., 2017). The low temperature of the drying process makes it suitable for coating or encapsulation of heat-sensitive microorganisms or lipids (Anwar & Kunz, 2011; Pitigraisorn et al., 2017). The particles that initially enter the fluidized bed can be produced using particles prepared by methods such as electrospraying (Pitigraisorn et al., 2017).

2.3.7 Other methods

In addition to the fabrication methods mentioned above, the encapsulation of food ingredients by starch can be achieved by simply mixing the encapsulant solution and encapsulated food ingredients solutions followed by ultrasonication and then freeze-drying or sprayed in chilled alcohol to achieve precipitation (Gupta, Chawla, Arora, Tomar, & Singh, 2015; Li, Shin, Lee, Chen, & Park, 2016b; Wang et al., 2018a). In some reports, this process is described as a solution mixing and solvent evaporation method (Ades et al., 2012; Gupta et al., 2015; Li et al., 2016b; Qiu et al., 2017; Zhu, Zhang, Tian, & Chu, 2018). For the solvent-evaporation method, once the encapsulant comes into contact with alcohol which was used as a dehydrating medium, it will be dehydrated and form microcapsules followed by separating the microcapsules from ethanol and evaporating residual

462 ethanol at low temperature (4–7 °C) (Gupta et al., 2015). In this way, the solvent used in this method
463 could be recycled and reused. (Gupta et al., 2015). In another study, Qiu et al. (Qiu et al., 2017)
464 prepared starch nanoparticles loaded with essential oils (menthone, cinnamon, lavender, oregano, and
465 citral) by adding hot ethanol (with essential oils dissolved) into a debranched normal waxy maize
466 starch solution followed by magnetic stirring, centrifugation and freeze-drying.

467 2.3.8 Post-processing drying methods

468 As mentioned in **section** 2.3.3 emulsification is usually combined with freeze-drying and spray-
469 drying to form particles.

470 2.3.8.1 Freeze-drying

471 Freeze-drying is a multi-stage process including freezing, sublimation, desorption, and finally,
472 storage, and the low-temperature operating environment during the drying process makes it suitable
473 for the dehydration and encapsulation of all heat-sensitive materials (Desai & Jin Park, 2005;
474 Ezhilarasi, Indrani, Jena, & Anandharamakrishnan, 2013; Laokuldilok & Kanha, 2015). This method
475 is mainly used to solidify starch-based emulsions incorporated with food ingredients and
476 biomaterials to further form emulsion powder or microparticles. These biomaterials were usually
477 heat-sensitive, such as phenolic compounds (Laokuldilok & Kanha, 2015), pigments (No & Shin,
478 2019), carotenoids (Spada et al., 2012), essential oils (Hasani et al., 2018), fatty acids (Yildiz et al.,
479 2018), and probiotics (Bilenler et al., 2017).
480 For example, Marefati et al. (Marefati et al., 2015) selected OSA-modified quinoa starch
481 granules as a stabilizer and fabricated W/O/W Pickering emulsions loaded with carmine, which is a

common food-coloring agent, through a two-step emulsification method. The oil-containing powder with high encapsulation efficiency (over 97%) and a high oil content (70 wt%) can be obtained via further freeze-drying. Some fabrication methods such as simple solution mixing (Li et al., 2016b; Spada et al., 2012) and complexation (Wang et al., 2020a) were also combined with freeze-drying to obtain the final particles.

2.3.8.2 Spray-drying

Spray-drying is the most used technology for the fabrication of starch-based encapsulation due to its low cost and easy operation. Spray-drying encapsulation involves the preparation and homogenization of dispersions, solutions, or emulsions, which are atomized in a drying chamber, and finally, the dehydration of the atomized droplets with hot air supplied to the drying chamber (Pereyra-Castro et al., 2018).

Research has shown that starch can form a “wall” around a “core” material (D-limonene), which was stable during spray-drying and can protect the enclosed ingredient for a relatively long time (Jafari, He, & Bhandari, 2007). However, a high air temperature may affect the activity of thermosensitive substances such as microorganisms (Alfaro-Galarza et al., 2020). Prepared using spray-drying, particles based on starch as the “wall” material have a larger size than the particle size of starch composite incorporated with other materials such as inulin or maltodextrin. The larger size using only starch as the encapsulant could be due to the high viscosity of the starch (Fernandes, Borges, & Botrel, 2014).

Some encapsulation types such as starch spherical aggregates (Hoyos-Leyva et al., 2018a; Hoyos-Leyva et al., 2019), microcapsules (Hong et al., 2019; Santiago et al., 2016), and V-type

503 starch inclusion complexes (Marinopoulou et al., 2019) have been widely used to encapsulate
504 various compounds via this drying method, such as vitamins (Hoyos-Leyva et al., 2018e; Subpuch et
505 al., 2016), probiotics (Alfaro-Galarza et al., 2020; Avila-Reyes, Garcia-Suarez, Jiménez, San Martín-
506 Gonzalez, & Bello-Perez, 2014), fatty acids (Marinopoulou et al., 2019; Tangsrianugul,
507 Suphantharika, & McClements, 2015), anthocyanins (Santana, Cano-Higuita, de Oliveira, & Telis,
508 2016; Santiago et al., 2016), and carotenoids (Liang et al., 2013).

509 ### 2.3.8.3 Electrospraying

510 Microcapsules with similar morphological characteristics could be obtained by electrospraying,
511 which does not require heating and the use of organic solvents, and can be used to encapsulate heat-
512 sensitive nutrients without toxicity issues (Pérez-Masiá et al., 2015; Pitigraisorn et al., 2017). In the
513 process of electrospraying, electrically charged jets from a viscoelastic polymer solution are
514 produced under a high-voltage electric field and further form particles by evaporation of the solvent
515 in the regions of lower potential (Drosou et al., 2017). Similarly as the spray-drying process as
516 mentioned in **section 2.3.8.2**, encapsulation could be achieved by spraying the entire solution that is
517 formed by dissolving, dispersing, or emulsifying the encapsulated substance in an aqueous solution
518 of the encapsulant (Drosou et al., 2017).

519 A stable electrospraying process could be obtained when the electrostatic forces inside the
520 droplet could overcome the surface tension of the starch solution, otherwise Taylor cones will not be
521 formed and the solution will drip (Pérez-Masiá, Lagaron, & López-Rubio, 2014). In the process of
522 electrospraying, the addition of surfactants could improve the electrospraying of a resistant maize
523 starch (Fibersol®) solution, as the surfactants could decrease the surface tension and stabilize the

electrospraying process (Pérez-Masiá et al., 2014). In addition, the type and amount of surfactant could also influence the size and size distribution of obtained particles (Pérez-Masiá et al., 2014). To obtain particles with satisfactory physicochemical characteristics, some research tends to combine two or more fabrication methods, such as electrospraying combined with fluidized-bed coating (Pitigraisorn et al., 2017). For example, Pitigraisorn et al. (Pitigraisorn et al., 2017) prepared moist-heat-resistant multilayered microcapsules. The core was sodium alginate encapsulating *Lactobacillus acidophilus*, which was coated with stearic acid and egg albumen as the first inner layer by electrospraying and then with cassava starch as the second outer layer by fluidized-bed coating. This method could lead to high encapsulation efficiency (93%) (Pitigraisorn et al., 2017).

3 Starch-based encapsulation systems for food applications

Starch-based encapsulation systems have been widely applied in food applications. The existing and potential applications of such systems developed in recent years are listed in **Table S1** in the **Supplementary Material** and are discussed in detail below.

3.1 Encapsulation of fatty acids

It is known that omega fatty acid-rich oils such as plant and fish oils are chemically unstable and susceptible to oxidation and deterioration. To reduce fatty acid oxidation and the unpleasant flavor of oxidation products, a commonly used method is encapsulation using starch or starch composites (Lei et al., 2018; Serfert, Drusch, & Schwarz, 2010; Yildiz et al., 2018). The encapsulation efficiency of

543 particles and the oxidative stability of the encapsulated oil are important parameters to evaluate the
544 encapsulation effectiveness (Belingheri et al., 2015b; Lei et al., 2018; Wang et al., 2020c; Yildiz et
545 al., 2018). Encapsulation efficiency describes the amount of oil that is encapsulated in particles
546 (Anwar & Kunz, 2011). The particle morphology can also provide some indication of the
547 encapsulation effectiveness for fatty acids. Specifically, particles with low air permeability and a
548 good protective effect often have a particle surface with no fissures or cracks (Wang et al., 2020c).
549 OSA-modified starch has an excellent oil-load capability for spray-drying due to its good film-
550 forming property and emulsifiability (Arshad et al., 2018; Wang et al., 2020c; Yang et al., 2020b).
551 However, the hydrophobic groups of OSA-modified starch may decrease the hydrophilicity and
552 water-dispersibility of the resulting particles (Wang et al., 2020c). This shortcoming can be overcome
553 by combining it with other polysaccharides or water-soluble hydrocolloids (Arshad et al., 2018;
554 Wang et al., 2020c). Wang et al. (Wang et al., 2020c) prepared microgel particles loaded with algal
555 oil with a size range of 223.70–644.46 nm via emulsification and spray-drying, and the encapsulant
556 was mainly based on OSA-modified starch (Capsul®) but combined with other polysaccharides such
557 as inulin, maltodextrin, and chitosan. The oxidation stabilities of free and encapsulated algal oils
558 were measured using the Rancimat accelerated oxidation method. The microgel particles using OSA-
559 modified starch, chitosan, and inulin (OSA/CS/IN) had the highest induction time which was three
560 times that of free algal oil (0.67 h) (Wang et al., 2020c). While low water-dispersibility of microgel
561 particles resulted when only OSA-modified starch was used as an encapsulant, a combination of
562 OSA-modified starch and inulin or maltodextrin could improve the water-dispersibility of the
563 microgel particles suitable to encapsulate ingredients in food or beverage products (Wang et al.,

564 2020c). In addition, microgel particles based on OSA/CS/IN showed a smoother surface than those
565 based on OSA-modified starch (OSA) or a combination of OSA-modified starch, maltodextrin, and
566 inulin (OSA/MD/IN) as the encapsulant. The difference in the surface smoothness could be
567 attributed to the high molecular flexibility of the OSA/CS/IN mixture and the formation of
568 electrostatic forces between OSA-modified starch and inulin (Wang et al., 2020c).
569 Conjugated linoleic acid, whose main absorption site is the small intestine, has numerous
570 physiological activities. However, conjugated linoleic acid has poor water solubility and high oxygen
571 sensitivity, making it tend to deteriorate in the upper part of the digestive tract such as the stomach
572 and difficult to reach the small intestine in its active form (Yang et al., 2020b). Based on
573 emulsification, Yang et al. (Yang et al., 2020b) prepared an encapsulant comprised of OSA-modified
574 waxy maize starch (a commercial product from Cargill) and xanthan gum, which showed effective
575 protection of conjugated linoleic acid and enabled the targeted delivery of this acid to the small
576 intestine (Yang et al., 2020b). He et al. (He et al., 2016). reported microgel particles also based on
577 OSA-modified waxy maize starch (a commercial product from Fonovo Food Ingredients Co., Ltd)
578 and xanthan gum but prepared by the combination of emulsification and spray-drying, which could
579 effectively avoid the direct contact of conjugated linoleic acid with non-absorptive sites. However,
580 spray-dried microgel particles using native sorghum starch or OSA-modified sorghum starch had
581 poor powder flowing property and some of the microgel particles had wrinkles on the surface
582 (Arshad et al., 2018), which may decrease the oxidative stability of the encapsulated fatty acid during
583 processing and storage. Microgel particles with wrinkled surfaces can capture oil droplets more

584 easily than those with smoother surfaces, which means a lower encapsulation efficiency and the
585 accelerated oxidation of fatty acid (Wang et al., 2020c).

586 The fabrication methods for particles can also influence the protective effect of OSA-modified
587 starch against lipid oxidation. Anwar et al. (Anwar & Kunz, 2011) evaluated four biopolymers
588 (soluble soybean polysaccharides (SSP), maltodextrin, hydroxypropyl β-cyclodextrin, and OSA-
589 modified starch (Hi-Cap® 100)) in combination as encapsulants for fish oil. For all combinations of
590 encapsulants, the encapsulation efficiency of particles prepared by spray granulation (SG) was higher
591 (>96%) than those obtained by spray-drying (SD) (68.03–89.97%) or freeze-drying (FD) (<50%). A
592 mixture of OSA-modified starch and SSP (65:10, w/w) as an encapsulant was the most effective at
593 preventing lipid oxidation as OSA-modified starch have both hydrophobic and hydrophilic groups
594 and can act as both a stabilizer and a surfactant to protect the fish oil from oxidation and improve the
595 emulsion stability (Anwar & Kunz, 2011). After storage at 21 °C for 5 weeks, the POV and propanal
596 content of the microgel particles based on OSA-modified starch/SSP prepared by SG were about 10
597 meq/kg oil and about 10 μmol/kg oil respectively and were lower than those values of the microgel
598 particles based on the same encapsulant prepared by SD or FD. In this regard, the higher processing
599 temperature of SD (180 °C) than SG (70 °C) could induce the formation of primary oxidation
600 products and further cause rapid degradation of the primary oxidation products (Anwar & Kunz,
601 2011). In addition, the irregular, flake-like and porous structure of products produced by FD may
602 accelerate the oxidation of fish oil. Thus, the microgel particles via SG could be used as a multi-
603 protection system for the encapsulation of fish oil (Anwar & Kunz, 2011).

Belingheri et al. (Belingheri et al., 2015b) obtained two types of particles by plating sunflower oil on porous starch (StarrierR®) or microgel particles based on gum Arabic and maltodextrin prepared by spray-drying. The peroxide value (POV) and conjugated dienes (CD) of the encapsulated oils were measured after exposure to heat and light to evaluate their oxidative stability. As compared with the maximum POV (about 18 meq O_2/kg) of bulk oil obtained before the decrease in POV (indicating possibly secondary oxidation) under the light exposure level (300–600, Klux) at any temperature (25–40 °C), the highest POVs of the oils encapsulated in the porous particles and microgel particles were 29.24 meq O_2/kg and 29.71 meq O_2/kg, respectively and did not show possibly secondary oxidation under the highest temperature and light exposure level (40 °C, 600 klux) (Belingheri et al., 2015b). Concerning the similar POVs of the porous particles and microgel particles, the "open'' structure of the porous starch matrix could be highly accessible to light and further promote the oxidation of the encapsulated oil; the oil on the surface of the microgel particles may suffer more rapid lipid oxidation (Belingheri et al., 2015b). The CD level of sunflower oil encapsulated in the porous starch was 2.52 under the highest temperature and light exposure level (40 °C, 600 klux) whereas this index of the oil encapsulated in the microgel particles was observed up to 2.59 at heating temperature (36 °C) and light exposure (600 klux), indicating plating on porous starch for the encapsulation of sensitive oil has a better effect than using spray-drying (Belingheri et al., 2015b). The simple plating procedure avoided the oxidation of the oil caused by the heating step, and this step which exists in spray-drying usually could induce the initial oxidation of the encapsulated oil (Belingheri et al., 2015b).

Using porous starch from purple sweet potato obtained via enzymatic modification, Lei et al. (Lei et al., 2018) successfully prepared olive oil–loaded porous particles by plating, and the oxidation stabilities of free and encapsulated olive oils were determined by measuring the POV. The POV of olive oil encapsulated in porous particles showed a very slow increase compared with free olive oil during storage for 4 days at 60 °C and the encapsulation could delay the decrease in oxygen pressure, demonstrating that this porous starch-based encapsulation system can improve the oxidation stability of the oil (Lei et al., 2018). An optimal loading ratio (33.22%) could be obtained with a suitable mass ratio of olive oil to porous starch (3:1), embedding temperature (40 °C), and embedding time (>50 min) (Lei et al., 2018).

Apart from OSA-modified starch and porous starch, hydroxypropylated starch was also used as an encapsulant (Yildiz et al., 2018). Yildiz et al. (Yildiz et al., 2018) used four encapsulants (pea protein isolate, pea protein isolate–hydroxypropylated starch composite, Tween 20, and sodium dodecyl sulfate) to prepare microgel particles loaded with canola oil (rich in omega-3 fatty acid) using emulsification followed by freeze-drying. It was found that the particles based on pea protein isolate–hydroxypropylated starch composite (1:1, w/w) as an encapsulant had the lowest POV and the highest release value for omega-3 fatty acid in the SGF (37.9%) or in the SGF and SIF (91.3%) compared with those based on other three encapsulants. Thus, this composite encapsulant can improve the antioxidant activity of omega-3 fatty acid and be applied in healthy food products (Yildiz et al., 2018).

3.2 Encapsulation of antioxidants

3.2.1 Phenolic compounds

As a kind of natural antioxidants with potential health benefits to humans, the effectiveness of phenolic compounds often depends on their bioactivity, stability, and bioavailability (Fang & Bhandari, 2010). However, phenolic compounds are vulnerable to heat, oxidants, and light and thus can be easily deteriorated and lose their original functions (Fang & Bhandari, 2010; Mehran, Masoum, & Memarzadeh, 2020; Palupi & Praptiningsih, 2016; Santiago et al., 2016). Some phenolic compounds (phenolics and tocopherols) are easy to degrade when exposed to solution conditions (Gomes et al., 2019). Moreover, phenolic compounds that are orally taken usually have low bioavailability (the fraction of ingested ingredients that enters the circulatory system and is accessible for storage and biological activities (Flores & Kong, 2017)) due to the high sensitivity to the gastrointestinal tract (GIT) environments of the human body (Annunziata et al., 2020). These issues may be addressed by encapsulation.

Vitaglione et al. (Vitaglione et al., 2013) studied the bioavailability in the human body of cocoa polyphenols (flavonols and phenolic acids) from cocoa-nut cream, encapsulated or not. Cocoa polyphenols–loaded molecular aggregation particles based on high-amylose maize starch were obtained via a complexation method followed by spray-drying. Three groups were set up in the experiment: cocoa-nut cream containing 20% (w/w) cocoa (CC), cocoa-nut cream containing 1.5% (w/w) free cocoa polyphenol extract (FPC), and cocoa-nut cream containing 1.5% (w/w) encapsulated cocoa polyphenol extract (EPC). The content of catechin, for example, for CC, FPC,

and EPC in serum was 0 nmol, 22.1 nmol, and 1.59 nmol, respectively. Regarding the higher content of catechin for FPC than EPC, it was hypothesized that the encapsulant that contained a higher amount of dietary fiber might slow the stomach emptying rate and further blunting the absorption of catechin; the content of catechin in serum after CC consumption indicated that the absorption of catechin was dose-dependent compared to FPC (Vitaglione et al., 2013). The total flavanol contents in the fecal sample for EPC and FPC were 151 nmol and 28 nmol, respectively, which indicates the encapsulation of flavanols could achieve the delivery to the gut (Vitaglione et al., 2013). In addition, the sensory score for the bitterness of cocoa-nut cream that was incorporated with cocoa polyphenols (1.5%, w/w) in the encapsulated form (6.0) was higher than the score for the cream with the free cocoa polyphenols (4.8), suggesting encapsulation could effectively mask the unwanted bitterness of cocoa polyphenols, this could be attributed to the fact that high-amylose maize starch was not easy to be accessed by saliva enzymes (Vitaglione et al., 2013).

Gomes et al. (Gomes et al., 2019) used a combination of OSA-modified starch (Capsul®) and inulin (1:1, 2:1, and 1:2, w/w) to encapsulate Brazil nut residue (which is rich in phenolic substances), for which normal particles (5.59–8.04 μm) were prepared by spray-drying. OSA-modified starch could reduce wrinkles by expanding the particles before spray-drying, which is conducive to the formation of particles with a smooth and uniform surface, as a smooth outer surface is fundamental features to ensure the protection of active ingredients (Gomes et al., 2019). After 120 days of storage at 27 °C, these three types of particles exhibited a high retention level of total phenolic compounds (69.5–71.9%) suggesting effective maintenance of the total phenolic

683 compounds and their antioxidant capacity (Gomes et al., 2019). These particles containing phenolic
684 substances can be used for functional foods.
685 Tea polyphenols are mainly absorbed in the small intestinal (Fang & Bhandari, 2010). Tea
686 polyphenols usually have low oral bioavailability due to the degradation caused by gastric acid,
687 enzymes, and microorganisms before reaching the small intestine (Fang & Bhandari, 2010). To solve
688 the problem of the low intestinal transport efficiency for tea polyphenols especially catechin, Shao et
689 al. (Shao et al., 2018) used Pickering emulsion stabilized by taro starch granules to encapsulate tea
690 polyphenols, when taro starch was dissolved in excess water and heated to 50 °C; the starch
691 gelatinized and tightly combined with tea polyphenols to form a layer of molten starch barrier,
692 thereby improving the retention rate (67%) of the tea polyphenols (Shao et al., 2018). The
693 amphiphilicity of taro starch and its nano-scale granule size resulting from milling (467.93 nm)
694 allowed it to be adsorbed at the oil-water interface to stabilize the Pickering emulsion (Shao et al.,
695 2018).
696 Hong et al. (Hong et al., 2019) used maize starch with different relative debranching degrees
697 (16.95%, 34.03%, 38.52%, and 44.96%) and mixed it with xanthan gum (100:2.5, w/w) as a
698 composite encapsulant to carry tea polyphenols and all the obtained micron-scale normal particles
699 (A40, B40, C40, and D40) prepared by spray-drying presented an encapsulation efficiency of over
700 80%. In addition, in simulated gastrointestinal fluids, the release of tea polyphenols encapsulated in
701 particles with 34% debranching degree was about 30% after 2 h and about 80% after 4 h but the
702 other three kinds of particles exceeded 40% and reached 100% at the same time points (Hong et al.,
703 2019). An increased amylose content and decreases in the relative molecular mass and molecular size

704 of the debranched maize starch might contribute to the digestion resistance of the encapsulant (Hong
705 et al., 2019). The encapsulant based on maize starch with a relative debranching degree of 34% had a
706 continuous, dense, porous, and crosslinked gel network structure and thus could achieve the slow
707 release of tea polyphenols; the *in vitro* release results also confirmed better performance of this
708 encapsulant in improving the bioavailability (Hong et al., 2019).
709 Mehran et al. (Mehran et al., 2020) used maltodextrin and an acetylated maize starch (Pregeflo®
710 CH 20) in combination (1: 0, 1: 0.25, 1: 0.5, and 1:1, w/w) to encapsulate Iranian borage extract
711 (IBE) (which contains anthocyanins) by spray-drying and the *in vitro* release of anthocyanins in the
712 SGF and the SIF was measured. Acetylated maize starch and maltodextrin, due to their good film-
713 forming property, could encapsulate heat-sensitive anthocyanins during a spray-drying process
714 (Mehran et al., 2020). Acetyl groups imparted starch with emulsifiability and increased its water
715 resistance, and the interaction between the flavylium cation of anthocyanins and the acetyl groups of
716 acetylated starch prevented anthocyanins from changing into other unstable forms (Mehran et al.,
717 2020). The encapsulation efficiency for the composite encapsulants of all four ratios were all above
718 93%. The 1:1 (w/w) encapsulant had the highest encapsulation efficiency (97%) and the stability of
719 anthocyanins encapsulated in this encapsulant was improved by 48% compared with unencapsulated
720 anthocyanins (Mehran et al., 2020). Besides, acetylated starch is stable under an acidic medium (pH
721 2.0) but under high pH conditions (pH 6.5), the acetyl group in the modified starch begins to
722 dissociate, and a repulsive force generated between starch chains eventually causes the prepared
723 particles to rupture (Mehran et al., 2020). The release content of anthocyanins from crude IBE and
724 from the encapsulated IBE in the SGF was 93.2% and 45.2% respectively after 120 min. This means

725 the 1:1 (w/w) encapsulant could partly protect anthocyanin in the SGF and the encapsulated IBE
726 could achieve stable and sustained release of anthocyanins in the SIF, suitable for target delivery to
727 the intestine (Mehran et al., 2020). However, in the case of choosing acetylated starch as the “wall”
728 film material, it is necessary to consider the wrinkled and convex-concave surface of the particles
729 caused by an increasing amount of acetylated starch (Mehran et al., 2020), as a smooth outer surface
730 is favorable for the protection of active ingredients (Gomes et al., 2019).
731 The ginkgo leaf contains abundant bioactive ingredients that are beneficial to human health,
732 such as polyphenols and quercetin; however, the low oral bioavailability of ginkgo biloba extract
733 (GBE) limited their application in food products (Wang et al., 2018a). Wang et al. (Wang et al.,
734 2018a) reported a method to prepare normal particles containing GBE, and in this method, ethanol
735 solution containing GBE was added into gelatinized damaling starch (26.7% amylose). As compared
736 with the rapid release of free GBE in the SGF and the SIF, which reached the maximum at 1.5 h and
737 2 h, respectively, the release profile of the encapsulated GBE (EGBE) in the SGF and the SIF could
738 be divided into three stages: initial rapid release (1 h and 2 h, respectively), immediate release (1–6 h
739 and 2–7 h, respectively), and sustained release (after 6 h and after 7 h, respectively) (Wang et al.,
740 2018a). In addition, the release of EGBE in the SGF was faster than in the SIF, especially in the
741 initial rapid release stage. The first stage of faster release in the SGF could be due to the protonated
742 state of EGBE and the sustained release of EGBE could be attributed to the gradual thinning
743 encapsulant with increasing digestion time.
744 Li et al. (Li et al., 2020a) used an oxidized potato starch microgel to encapsulate micelle-like
745 nanoparticles formed by lysozyme and quercetin via self-assembly. This method combined the

advantages of oxidized starch microgels in resisting complex gastrointestinal conditions and of protein micelles in encapsulating hydrophobic bioactive substances. The lysozyme as a protein carried hydrophobic quercetin and improved the solubility of quercetin in water (Li et al., 2020a). The release contents of quercetin only encapsulated in micelle-like nanoparticles in the SGF (incubating for 2 h) and the SIF (incubating for 4 h following incubating in SGF) were 8.17% and 30%, respectively, whereas those of quercetin encapsulated by the oxidized potato starch microgel were negligible in the SGF (incubating for 2 h) and 7.9% in the SIF (incubating for 4 h following incubating in SGF) after 6 h. Besides, the results of *in vivo* test (**Figure 3**) showed that the microgel particles could adhere to the intestine and achieve the target release of quercetin, due to the carboxyl-abundant of oxidized starch (Li et al., 2020a).

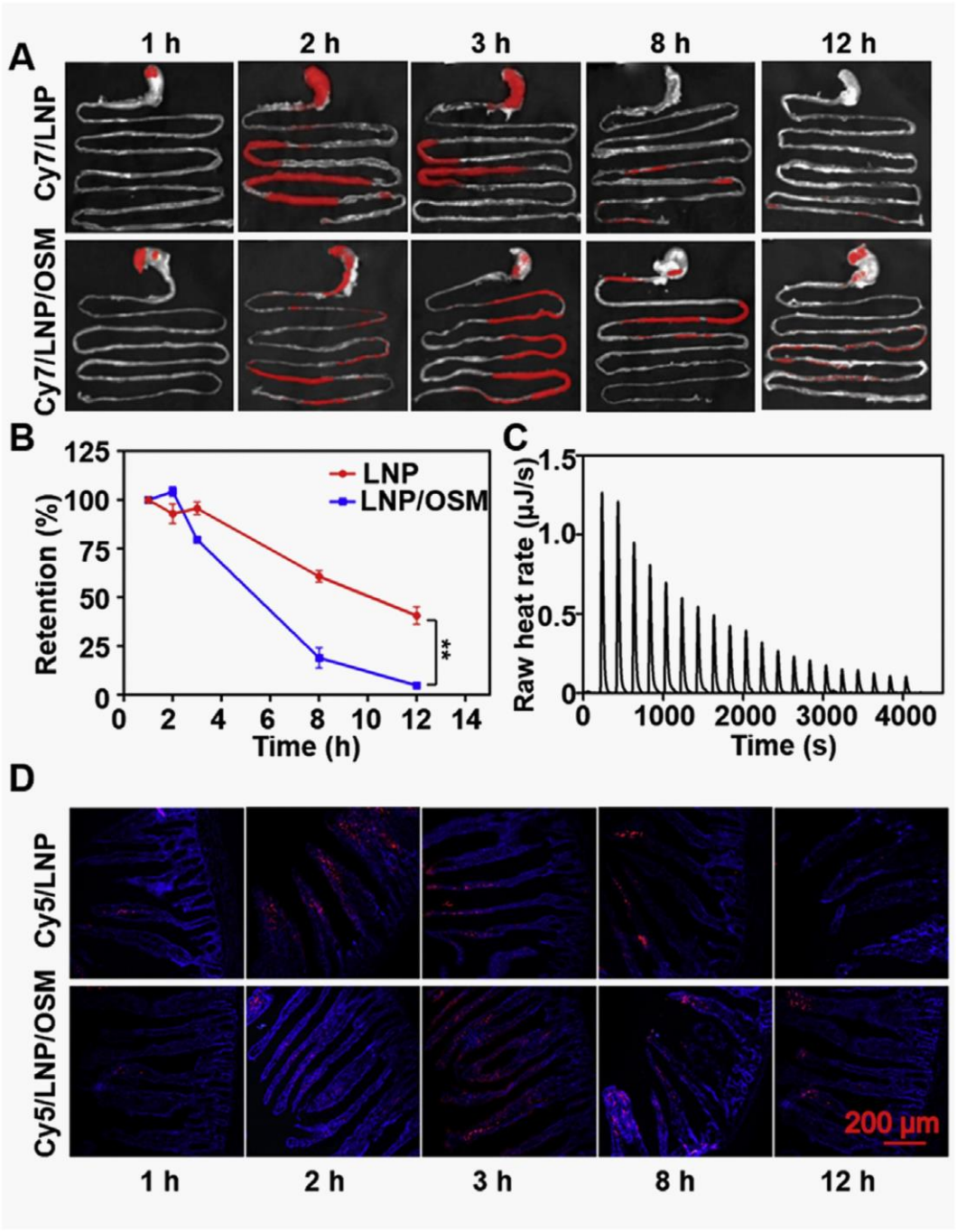


Figure 3 A) Fluorescent bioimaging images of the distribution of micelle-like nanoparticles and microgel particles loaded with quercetin in the stomach and the whole intestine of the rat; B) Retention of micelle-like nanoparticles and microgel particles loaded with quercetin in the intestine of rat after oral administration of nanoparticles and microgel particles; C) The mucoadhesive behavior of oxidized potato starch microgel investigated by isothermal titration calorimetry; D) The

763 distribution and absorption of micelle-like nanoparticles and microgel particles by intestinal villi.
764 Reprinted from Ref. (Li et al., 2020a) with permission from Elsevier, Copyright 2020.
765
766 Zanoni et al. (Zanoni, Primiterra, Angeli, & Zoccatelli, 2020) used spray-drying to encapsulate
767 polyphenols (mainly anthocyanins) extracted from red cabbage and red chicory with an OSA-
768 modified starch Capsul® as an encapsulant. The formed normal particles (diameter: 1–30 μm) had
769 high encapsulation efficiency for polyphenols extracted from red cabbage (79%) and red chicory
770 (88%). The difference in encapsulation efficiency of OSA-modified starch for the two types of
771 anthocyanins could be due to their different ways and degree of interaction with OSA-modified
772 starch (Zanoni et al., 2020). Besides, due to the amphiphilic nature of grafted functional groups,
773 OSA-modified starch exhibited excellent film-formation property during spray-drying and improved
774 the stability of anthocyanins (Zanoni et al., 2020). The encapsulated anthocyanins and the
775 unencapsulated counterpart had almost identical color at different pH values, which indicates that the
776 encapsulation process almost did not affect the colors of anthocyanins in the visible range. Regarding
777 the different colors at pH 6 or 7 of the red cabbage extract (un-encapsulated or encapsulated), the
778 starch could have affected the optical properties of anthocyanins by interacting with their structures
779 (Zanoni et al., 2020). In addition, encapsulation improved the retention of antioxidant stability for
780 polyphenols from red cabbage (20–30%) and red chicory (44–55%), as compared with the un-
781 encapsulated polyphenols (Zanoni et al., 2020).
782 As a polyphenol, curcumin is known for its antioxidant and antimicrobial activities, whereas its
783 low water solubility and easy degradation have limited its application (Acevedo-Guevara, Nieto-

784 Suaza, Sanchez, Pinzon, & Villa, 2018; Li et al., 2016b). By loading curcumin into normal particles
785 based on soluble starch prepared via a simple solution mixing method, the solubility of curcumin in
786 water could be increased by 715 fold and the antioxidant ability was also retained (Li et al., 2016b).
787 In another study, Acevedo-Guevara et al. (Acevedo-Guevara et al., 2018) prepared curcumin-loaded
788 normal particles based on native banana starch (NBS) or acetylated banana starch modified by acetic
789 anhydride (ABS) prepared using the same method. The obtained particles allowed for the controlled
790 release of curcumin in the SGF and the SIF, and the release amount of curcumin in ABS was 15%,
791 lower than that in NBS, after 120 min in the SGF whereas two kinds of particles showed similar
792 release profiles in the SIF. Regarding the lower release in the SGF achieved by particles based on
793 acetylated banana, Acevedo-Guevara et al. (Acevedo-Guevara et al., 2018) indicated that, compared
794 with NBS, ABS, due to its more hydrogen-bond-accepting sites, had greater hydrogen-bonding
795 interaction with curcumin molecules (hydrogen-bond-donor) (Acevedo-Guevara et al., 2018).

796 ### 3.2.2 Carotenoids

797 Souza et al. (Souza et al., 2018) used an OSA-modified starch (Capsul®), whey protein isolate,
798 maltodextrin, and their mixtures to prepare normal particles loaded with lycopene-rich tomato
799 concentrate by spray-drying. Among the various encapsulant formulations, the lycopene
800 encapsulated in an all-starch material showed the highest antioxidant capacity (27.24 ± 11.28, μmol
801 Trolox/g) (Souza et al., 2018). OSA-modified starch has enhanced amphiphilicity and emulsifiability
802 (Souza et al., 2018). Thus, compared with other encapsulants (whey protein isolate, maltodextrin,
803 and their mixtures), the OSA-modified starch provided better protection to the lipophilic compound

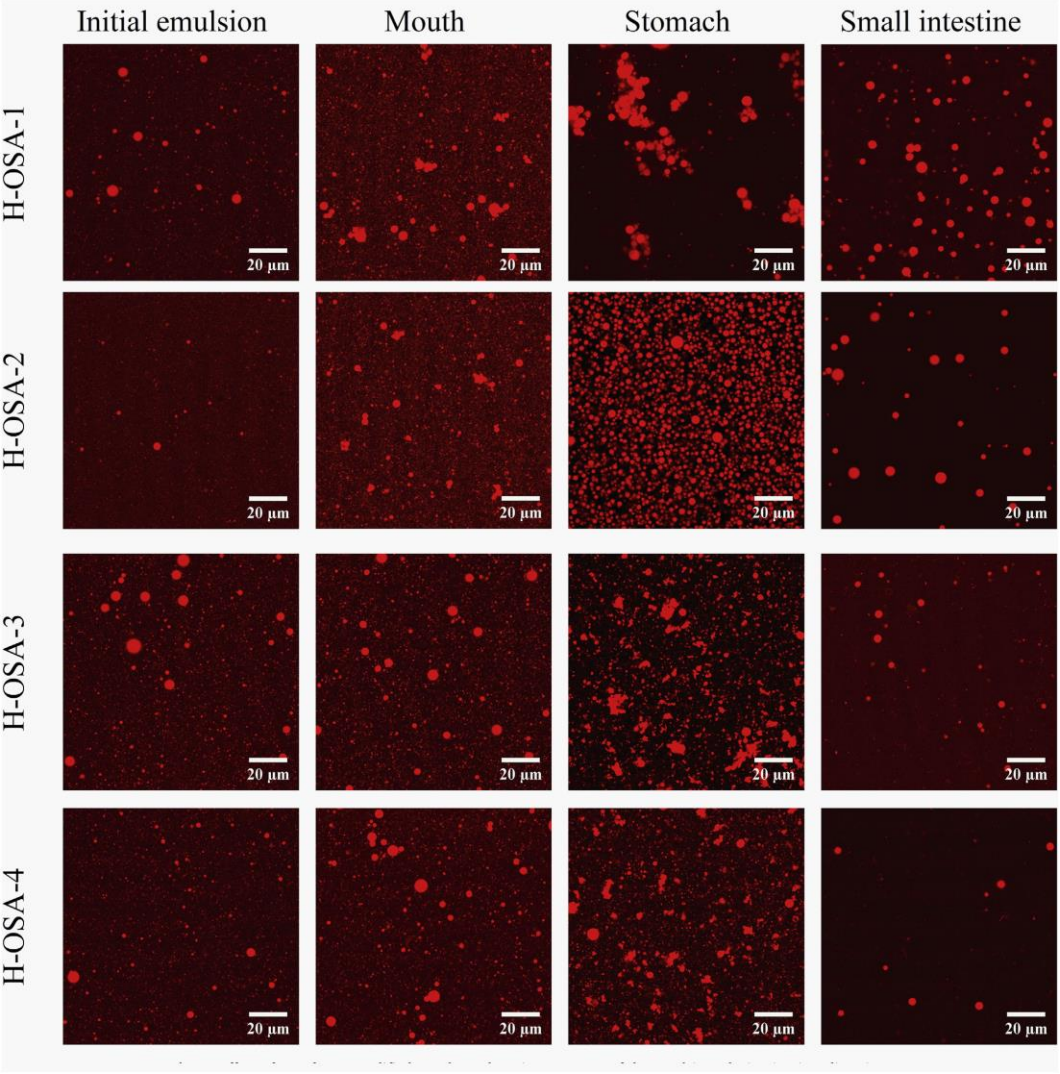
804 lycopene in normal particles during spray-drying and storage, thereby reducing the degradation of
805 lycopene (Souza et al., 2018).

806 To improve the processing and utilization of astaxanthin, Zhao et al. (Zhao et al., 2019) prepared
807 a kind of molecular aggregation particles using OSA-modified kudzu starch and gelatin as a complex
808 encapsulant (1:1, w/w) to encapsulate heat-labile astaxanthin via a complex coacervation method.
809 The negative charge of kudzu starch due to the presence of surface proteins, lipids, and phosphates or
810 the negatively charged carboxylic acid carried by OSA could promote the electrostatic interaction
811 between kudzu starch and gelatin (Zhao et al., 2019). The results of stability analysis showed that the
812 retention rate of astaxanthin encapsulated in molecular aggregation particles was 82% whereas the
813 retention rate of astaxanthin encapsulated in gelatin was 71% after storage at 25 °C for 10 days.
814 Thus, encapsulating astaxanthin in molecular aggregation particles can delay the degradation of
815 astaxanthin and further improve its bioavailability (Zhao et al., 2019).

816 In a study (Rocha, Fávaro-Trindade, & Grosso, 2012), lycopene-loaded microgel particles using
817 modified starch (Capsul®) as an encapsulant via spray-drying were added into a cake and the color of
818 the cake was measured using a colorimeter. The external surfaces of the obtained particles had no
819 fissures, cracks or interruptions and could better protect lycopene. The cake with encapsulated
820 lycopene had a homogenous color distribution and a stronger color than the cake without lycopene
821 (Rocha et al., 2012).

822 In addition, it was found that a higher degree of substitution (DS) of OSA-modified waxy maize
823 starch might reduce the flocculation and coalescence of the OSA-modified waxy maize starch-
824 stabilized-emulsion loaded with β-carotene during *in vitro* digestion (**Figure 4**) (Lin, Liang, Zhong,

825 Ye, & Singh, 2018). Specifically, OSA-modified waxy maize starch with higher DS values had more
826 carboxyl groups and thus may contribute to forming a more rigid and compact surface, thereby
827 resisting the flocculation and coalescence of emulsion droplets (Lin et al., 2018).
828



829
830 **Figure 4** Confocal laser scanning microscopy (CLSM) images showing the microstructure of OSA-
831 modified waxy maize starch-stabilized emulsions loaded with β-carotene during *in vitro* digestion
832 (H-OSA-1, H-OSA-2, H-OSA-3, H-OSA-4 represent OSA-modified hydrolyzed waxy maize
833 starches with DS = 0.0158, 0.0249, 0.0340, and 0.0416, respectively). Reprinted from Ref. (Lin et
834 al., 2018) with permission from Elsevier, Copyright 2018.

Color is also an important quality indicator of foods that determine their acceptance by consumers (Azeredo, 2009; Chranioti, Nikoloudaki, & Tzia, 2015). As an alternative to chemical colorants, natural pigments could have both beneficial functional properties and are safe for humans (Yamashita et al., 2017). Chranioti et al. (Chranioti et al., 2015) successfully prepared normal particles loaded with saffron extract (which are rich in crocin) and beetroot extract (which are rich in betacyanins and betaxanthins) via freeze-drying. Five different encapsulant combinations including gum Arabic, maltodextrin, gum Arabic–modified starch, modified starch–chitosan, and modified starch–maltodextrin–chitosan were used, where the modified starch was an OSA-modified starch (Clear-gum® CO-01). The obtained normal particles were added into a chewing gum model system and the color stability of chewing gum were measured using a colorimeter. The chewing gum incorporated with coloring extracts encapsulated in gum Arabic–modified starch (1:1, w/w) had the greatest value of b* (saffron, 36.20 and 38.76) at different storage temperature (25 °C and 40 °C) (Chranioti et al., 2015).

Apart from the particle forms mentioned above, starch-based hydrogels could also provide good encapsulation for lipophilic nutraceuticals (Lin, Liang, Ye, Singh, & Zhong, 2017; Mun, Kim, & McClements, 2015a; Mun et al., 2015b). Mun et al. (Mun et al., 2015a) designed three delivery systems (corn oil-in-water (phosphate buffer dispersed with whey protein isolate) emulsion, rice starch hydrogel, and filled rice starch hydrogel) incorporated with β-carotene (**Figure 5A**) to study the bioaccessibility of β-carotene. The filled starch hydrogel matrix could protect lipid droplets (corn oil) from extensive aggregation and maintained the semi-solid structure of the hydrogel under

simulated oral and gastric conditions, but the structure of the filled rice starch hydrogel disintegrated in the simulated small intestine due to the amylase activity from the pancreatin or to dilution and shearing (**Figure 5B**). Besides, the bioaccessibility of β-carotene in the filled starch hydrogel (as high as over 50%) was higher than that in the emulsion (1–23%, depending on lipid concentration) or in the starch hydrogel (≈ 1%) (Mun et al., 2015a). Mun et al. (Mun et al., 2015b) further used a starch-based hydrogel (mung bean or rice starch) filled with lipid droplets (corn oil) stabilized by whey protein isolate or Tween 20 to encapsulate β-carotene, and the bioaccessibility of β-carotene in the digesta after being digested in the SGF and the SIF was evaluated. β-Carotene directly incorporated into the hydrogel had very low bioaccessibility (<1%) whereas that incorporated into lipid droplets (corn oil) stabilized by whey protein isolate or Tween 20 containing 4 wt% lipid (corn oil) had higher bioaccessibility (14% and 54%, respectively) (Mun et al., 2015b). The starch hydrogel could improve the bioaccessibility of β-carotene by preventing the lipid droplets from aggregation, and the lipid digestion rate could be decreased significantly by adding the lipid droplets into the mung bean starch hydrogel due to the high amylose and protein contents of mung bean starch (Mun et al., 2015b). The higher amylose content of mung bean starch than rice starch was conducive to the formation of a stronger gel, and the tightly-packed structures stabilized by hydrogen bonding during retrogradation could slow down the digestion of starch by amylase, thereby inhibiting the release of lipid droplets (incorporated with β-carotene) at the initial digestion stage into the intestinal fluids and could improve the bioaccessibility of β-carotene (Mun et al., 2015b).

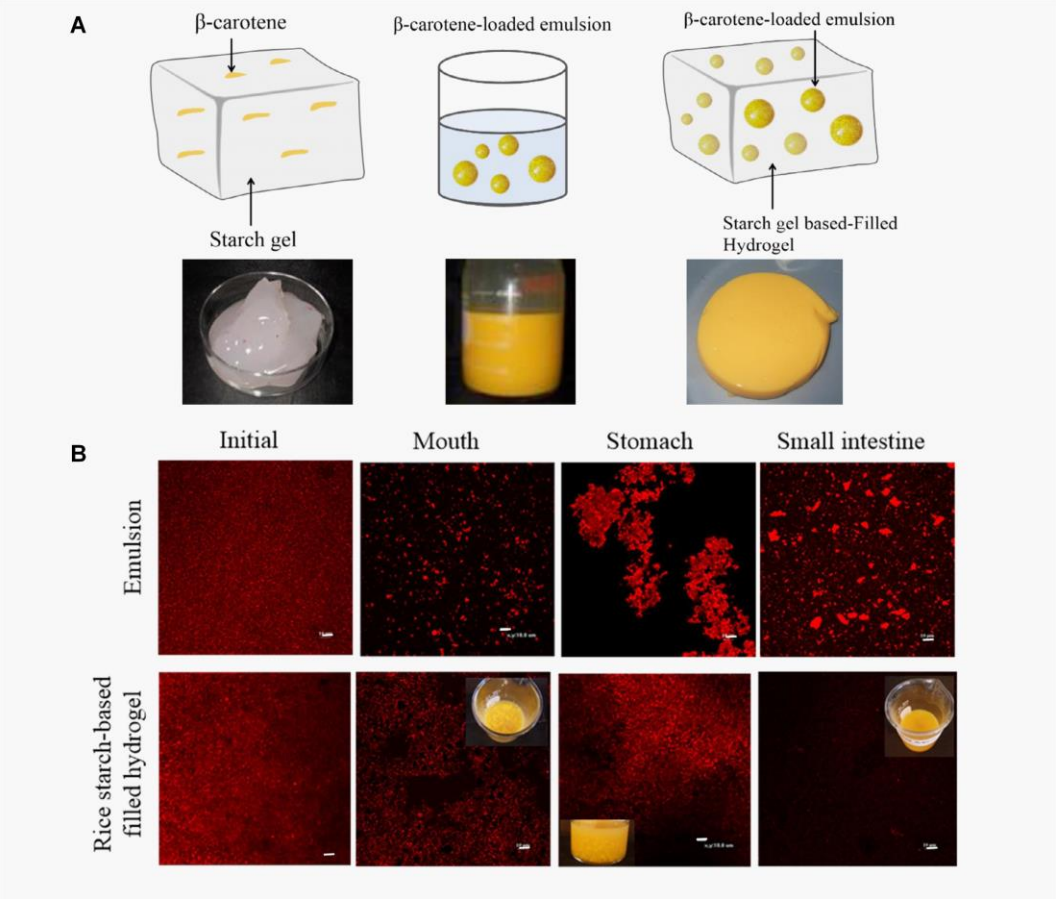


Figure 5 A) Three delivery system (rice starch hydrogel, emulsion, and filled rice starch hydrogel) designed to control the bioaccessibility of β-carotene; B) The microstructures of the emulsion and the filled rice starch hydrogel in simulated gastrointestinal conditions (scale bars = 10 μm). Adapted from Ref. (Mun et al., 2015a) with permission from Elsevier, Copyright 2015.

3.3 Encapsulation of flavors and essential oils

Under food processing conditions (e.g. heat and humidity), food products usually lost their original flavors (Belingheri et al., 2015a). Flavor ingredients have been widely used in food products to improve their sensory properties. Flavor compounds in a liquid state are especially vulnerable as they are easy to be lost via oxidation, evaporation, or interaction with other ingredients (Zeller,

887 Saleeb, & Ludescher, 1998). In this regard, flavor ingredients can be encapsulated so that they are
888 resistant to the environments (Pellicer et al., 2019). Besides, encapsulation of flavor ingredients
889 could allow the release of them in a controlled way. Due to the special structures that can be formed
890 by starch (e.g. the helical structure of amylose (Yeo, Thompson, & Peterson, 2016), microporous
891 structure (Belingheri et al., 2015a), and normal particles with high porosity (Zhu et al., 2018)), starch
892 has been used to develop flavor carriers (Zeller et al., 1998). Starch-based encapsulation systems
893 have been used to carry different flavors such as tomato flavor (Belingheri et al., 2015a), strawberry
894 flavor (Pellicer et al., 2019), vanilla oil (Zhu et al., 2018), and limonene (Yeo et al., 2016).
895 Belingheri et al. (Belingheri et al., 2015a) used a simple plating procedure to make liquid tomato
896 flavor encapsulated in a porous starch (StarrierR®) and the sensory properties of tomato sauce (fresh
897 or aged during storage for 3 and 6 months) flavored by these particles were analyzed. It was found
898 that the solvent used to disperse the liquid tomato flavor into the porous starch significantly affected
899 the flavor content in the porous particles after 6 months. For example, the particles prepared using
900 MCT (which is apolar) as the solvent led to the highest flavor content (0.974) of *p*-cymene (which is
901 apolar) than those using propylene glycol (0.051) and triacetin (0.360), both of which are polar
902 solvents (Belingheri et al., 2015a). Thus, to obtain the best flavor retention effect, solvents with
903 similar polarity to flavor molecules should be chosen for loading flavor molecules into porous starch
904 (Belingheri et al., 2015a).
905 In a study, Chen et al. (Chen, Guo, Wang, Yin, & Yang, 2016) prepared a structured flavoring
906 O/W emulsion (stabilized by OSA-modified waxy maize starch (Purity Gum 2000®)) by heat
907 homogenization, which could delay the release of volatile compounds. As β-sitosterol could form

plate- and needle-like crystals when being crystallized in sunflower oil (**Figure 6A**) or in the presence of OSA-modified waxy maize starch (**Figure 6B**), the release rate and maximum headspace concentration of structured emulsion loaded with volatile flavor compounds (hexanal, diacetyl, D-limonene, ethyl hexanoate, ethyl octanoate, and linalool) were both lower than that of the unstructured emulsion during 50 min (Chen et al., 2016). The obtained structured flavoring O/W emulsion had both good colloidal stability (volume-average emulsion size = 0.835) after storage for 90 days and the ability to delay the volatile release, as compared with the unstructured flavoring O/W emulsion (volume-average emulsion size = 6.5) (**Figure 6C-E**) (Chen et al., 2016).

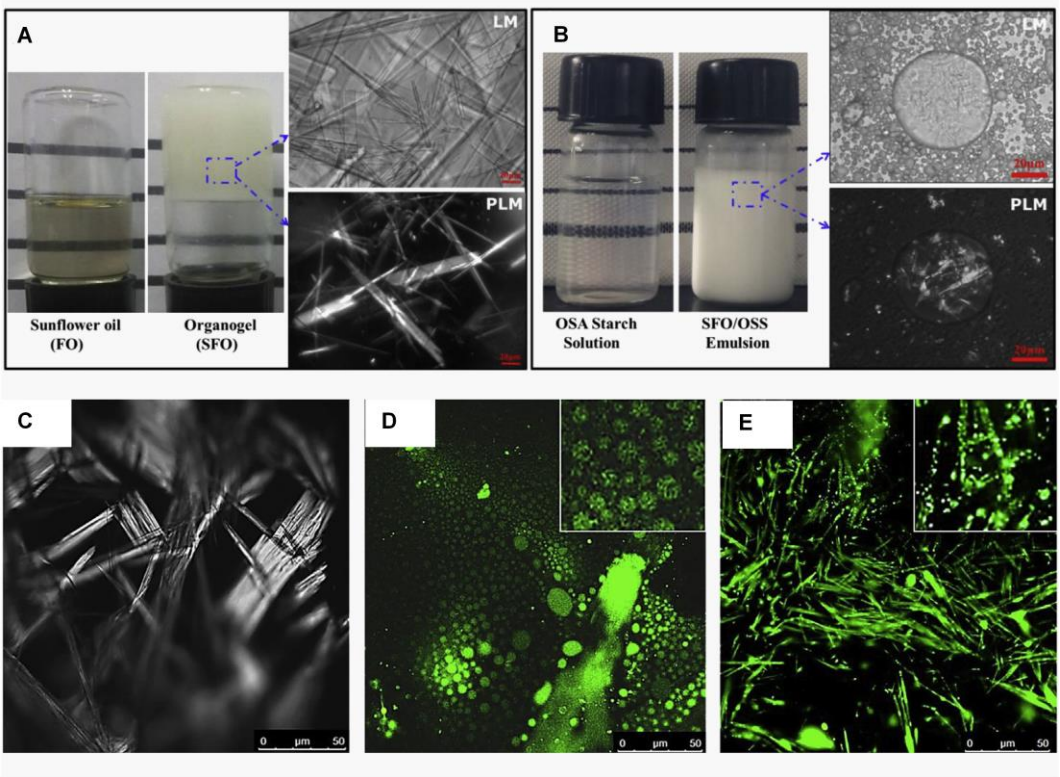


Figure 6 Crystallization behavior of β-sitosterol in A) sunflower oil and B) O/W emulsion stabilized by OSA-modified waxy maize starch (FO: sunflower oil; SFO: β-sitosterol structured sunflower oil, OSS: OSA-modified waxy maize starch; LM: light microscopy; PLM: polarized light micrograph);

C) PLM image revealing the microstructure of needle-like β-sitosterol crystals formed at the oil/water interface without surfactant; D,E) Confocal laser scanning microscopy (CLSM) image of the unstructured O/W emulsion interface stabilized by OSA-modified waxy maize starch and the structured O/W emulsion interface stabilized by OSA-modified waxy maize starch, respectively. Adapted from Ref. (Chen et al., 2016) with permission from Elsevier, Copyright 2016.

Encapsulation technology can also be used to encapsulate vanilla oil, as a kind of flavoring that is widely used in food products and the confectionery industry, since its application could be hindered by its volatility and instability (Zhu et al., 2018). Zhu et al. (Zhu et al., 2018) used jackfruit seed starch, β-cyclodextrin, and chitosan, respectively, as encapsulants to prepare normal particles loaded with vanilla oil via sonication and freeze-drying based on a simple solution mixing method, and the release performance of the normal particles during storage was studied by an electronic nose. As compared with the rapid release of flavor molecules encapsulated in β-cyclodextrin and chitosan at the beginning, the particles based on jackfruit seed starch exhibited a slower release of flavor molecules (Zhu et al., 2018). Specifically, the shelf time and encapsulation efficiency of the starch-based normal particles (250 days and 79.33%) was higher than those based on β-cyclodextrin (160 days, 77.92%) or chitosan (160 days, 76.64%). These results can be ascribed to the low crystallinity, stronger gel property, better plasticity, and better film-forming property of jackfruit seed starch than β-cyclodextrin or chitosan (Zhu et al., 2018).

In addition, the source of starch also affects the encapsulation efficiency. Ades et al. (Ades et al., 2012) used starches with different amylose content (i.e. amylose from potato starch, normal maize

942 starch, waxy maize starch, and high-amylose maize starch) to encapsulate three aromas with different
943 hydrophobicity (i.e. limonene, menthol, and menthone). Particles were prepared by adding the aroma
944 compounds to the starch suspensions before freeze-drying, and the oral release of aroma compounds
945 in simulated salivary fluids was studied. Only menthol and menthone led to the formation of V-
946 amylose complexes to allow the controlled release of themselves. Increasing amylose content led to a
947 high complexation yield and encapsulation efficiency. These V-type amylose complexes could be
948 used as an efficient platform for the controlled release of aroma in the oral cavity (Ades et al., 2012).
949 To date, the spoilage and contamination during food processing, storage, and preservation due to
950 the presence of spoilage microorganisms remain to be problematic (Tao, Hill, Peng, & Gomes, 2014;
951 Wang et al., 2014b). To extend the shelf life of food and considering the limited antibacterial
952 spectrum and harmful side effects of traditional antibiotics, natural antibacterial agents such as plant
953 essential oils have attracted great attention (Hassan et al., 2019; Qiu et al., 2017; Wang et al., 2018b;
954 Zhou et al., 2016).
955 Porous starch encapsulating antimicrobial agents can be used for food preservation (Wang et al.,
956 2018b). Wang et al. (Wang et al., 2018b) used porous starch (a commercial product from Chongqing
957 Taiwei Bioengineering Ltd) and β-cyclodextrin as a composite material to prepare clove oil–loaded
958 microgel particles by emulsification and spray-drying, and suspensions of the particles (0.07% or
959 0.08% concentration) were used to treat cooked meat (i.e. chicken, pork, beef, and fish) without or
960 with heat treatment (being boiled for 30 min). Up to 96 h, the four meat products that were treated by
961 the microparticle suspensions did not show any mold (Wang et al., 2018b). This indicates the
962 microparticles have both good heat-resistance and antifungal activities.

963 Linalool has antimicrobial, insecticidal and pharmacological effects and can be used as an
964 antimicrobial additive for food products. However, linalool is hydrophobic, volatile, and easy to
965 oxidize, which hinder its application in food processing (Zhou et al., 2016). Zhou et al. (Zhou et al.,
966 2016) prepared linalool-loaded molecular aggregation particles by adding linalool into the saturated
967 solutions of amylose or oxidized amylose (OAM). The encapsulation ability of OAM towards
968 linalool decreased with increasing oxidation degree of OAM, owing to the depolymerization of
969 amylose. The antimicrobial activity of these two types of molecular aggregation particles against
970 *Staphylococcus aureus* (*S. aureus*) and *Escherichia coli* (*E. coli*) was evaluated. The broth that was
971 added with linalool-loaded molecular aggregation particles were clear when the concentration of
972 linalool was higher than the minimal inhibitory concentration (MIC) whereas the broth in the control
973 group was turbid (Zhou et al., 2016). In addition, the MICs of linalool encapsulated in amylose
974 against *S. aureus* and *E. coli* were 0.8 and 1.6 mg/mL respectively and those in OAM were 0.4 and
975 0.8 mg/mL respectively. This indicates the linalool encapsulated in OAM had better antimicrobial
976 performance than that in AM due to the solubilization effect of OAM on linalool and the limited
977 release of linalool caused by the fast aggregation and retrogradation of amylose–linalool molecular
978 aggregation particles in an aqueous solution (Zhou et al., 2016). However, amylose prevented the
979 volatilization of linalool more effectively than OAM, which might be explained by the loss of the
980 regularly packed crystal structure of OAM (Zhou et al., 2016).
981 Qiu et al. (Qiu et al., 2017) studied the *in vitro* release and the antimicrobial activity of menthone
982 as an essential oil encapsulated in molecular aggregation particles (diameter: 93–113 nm) based on
983 waxy maize starch. The obtained menthone-loaded molecular aggregation particles formed at 90 °C

had the highest encapsulation efficiency (86.6%) than the particles formed at 30°C (72.4%) or 60 °C (78.1%). Regarding this, the high process temperature for molecular aggregation particles may lead to a more ordered crystalline structure of starch and thus impart the starch with higher encapsulation capability with menthone (Qiu et al., 2017). The release of menthone from the molecular aggregation particles was slow and sustained, and the release contents of menthone encapsulated in particles formed at different temperature (30 °C, 60 °C, and 90 °C) during the first 10 h were less than 55%, 70%, and 62%, respectively (Qiu et al., 2017). While the antibacterial efficiency of the free menthone decreased slowly after 2 h, the antibacterial activity of the encapsulated menthone increased continuously during the test time range (12 h) (Qiu et al., 2017). In addition, the antioxidant activity of the menthone was also extended from 4 h to 12 h. Thus, encapsulation could improve the antioxidant activity and antimicrobial activity of menthone, enabling the application of this essential oil for food preservation (Qiu et al., 2017).

3.4 Encapsulation of probiotics

As live microorganisms, probiotics exert beneficial health effects only by passing the upper GIT with high viability maintained until reaching the gut (Ashwar et al., 2018). Probiotics have a significant role in maintaining the growth and balance of healthy flora in human intestines; however, the extreme conditions in the digestive system may negatively affect probiotics to play a role in the gut by reducing their activity before reaching the target site (Ashwar et al., 2018). As such, various encapsulation methods have been employed to protect probiotics against the extreme conditions in the digestive system and deliver them to specific sites (Ahmad, Gani, Hamed, & Maqsood, 2019;

Ashwar et al., 2018; Dafe et al., 2017; Zaeim, Sarabi-Jamab, Ghorani, & Kadkhodaee, 2019). Starch-based normal particles and microgel particles have been used to encapsulate probiotics to achieve the controlled release of these probiotics and target delivery (Ahmad et al., 2019; Ashwar et al., 2018).

Ahmad et al. (Ahmad et al., 2019) studied the encapsulation effects of native water chestnut starch (NWS) and water chestnut starch nanogranules (WSN, prepared by ball milling) on camel milk-derived probiotics under simulated gastrointestinal tract conditions, and two kinds of microgel particles were obtained using emulsification and freeze-drying. The microgel particles based on NWS had higher cell viability (85%) than those based on WSN (73%). Regarding this, WSN might have permeated to cell membranes of probiotics and thus not be able to protect probiotics (Ahmad et al., 2019). The cell viability in NWS particles was also higher (5.62 log CFU/g) than those (0 log CFU/g) in WSN for 15 min of heat treatment at 80 °C, as the nano-sized WSN may not be able to keep probiotics in its core due to their small size (Ahmad et al., 2019). Besides, NWS particles could protect the probiotic cells well under the SGF conditions and about 5.66 log CFU/g viable cells in the microgel particles reached the SIF. Thus, the encapsulation system based on NWS could achieve the target delivery of probiotics to the intestine (Ahmad et al., 2019).

Ashwar et al. (Ashwar et al., 2018) utilized the same fabrication method to prepare microgel particles loaded with three different *Lactobacilli* (*Lactobacillus casei*, *Lactobacillus brevis*, and *Lactobacillus plantarum*) using RS (crosslinked phosphorylated rice starch) as an encapsulant. Differential scanning calorimetry (DSC) results showed that compared to native rice starch, the microgel particles had a significantly increased thermal transition temperature and enthalpy change (ΔH). The enhanced thermal stability of the microgel particles was explained due to the crosslinking

of starch. The average losses of these three *lactobacilli*, free and encapsulated, after explosion at a temperature of 55 °C for 10 min were 5.93, 6.06, and 6.78 log CFU·g^{-1} and 0.12, 0.18, and 0.30 log CFU/g, respectively, and those after exposure at 65 °C for 10 min were 7.45, 5.61, and 6.56 log CFU/g and 5.04, 4.35, and 4.59 log CFU/g, respectively. The viability of the encapsulated *lactobacilli* (8.42, 8.61, and 7.29, log CFU/g) in the SIF was much higher than that of the free *lactobacilli* (3.67, 3.08, and 3.43, log CFU/g), and the viability of the encapsulated *lactobacillus* could remain high (> log 7 CFU/g) under 4 °C for 2 months (Ashwar et al., 2018). All these results suggesting that the encapsulation could well protect *Lactobacilli*.

By encapsulating probiotics (*Lactobacillus rhamnosus* GG), which are labile to fermentation, in normal particles using modified huauzontle's starch (acid hydrolysis followed by extrusion modification) and whey protein isolate (1.6:1, w/w) via spray-drying and adding these particles into green tea beverage, Hernández-Barrueta et al. (Hernández-Barrueta et al., 2020) found that the viability of the probiotics just reduced from 7.95 log CFU/mL to 7.33 log CFU/mL after storage at 4 °C for 5 weeks. After extrusion, the soluble components of huauzontle's starch were released and more hydroxyl groups were exposed. Hydroxyl groups could interact with molecules that were encapsulated in particles, and thus promoting the retention of these molecules for encapsulation purposes (Hernández-Barrueta et al., 2020). In addition, huauzontle's starch subjected to acid hydrolysis and extrusion can be used as an encapsulant for preparing normal particles of probiotics by spray-drying due to the increased RS content and reduced viscosity (Hernández-Barrueta et al., 2020). The particles prevented the fermentation of green tea beverage, and the color and antioxidant capacity of the beverage both had no significant changes (Hernández-Barrueta et al., 2020).

3.5 Encapsulation of other food ingredients

The long-term iron deficiency may cause anemia. Efforts have been put to enhance the supplementation and absorption of iron for humans. While the fortification of foods is seen as an effective route to address iron deficiency, food fortification directly with iron may cause sensory change and lead to unwanted interactions between food ingredients and iron (Bryszewska et al., 2019; Gupta et al., 2015). Moreover, the lack of other nutrition substances such as vitamins and coenzymes can also cause human diseases (Cheuk et al., 2015; Liu, Green, & Kitts, 2015; Mun et al., 2015b). At present, some researchers (Bryszewska et al., 2019; Cheuk et al., 2015; Gupta et al., 2015; Hategekimana, Masamba, Ma, & Zhong, 2015; Hoyos-Leyva et al., 2018e; Liu et al., 2015; Morozova et al., 2020) have made efforts to solve these issues by encapsulating these substances into starch-based encapsulation systems which can be added into food products.

Bryszewska et al. (Bryszewska et al., 2019) prepared iron-loaded normal particles using a modified starch (TMS) as an encapsulant, and the obtained particles loaded with ferrous sulphate or ferrous lactate were added into breads prepared by conventional fermentation or sourdough fermentation. The encapsulant based on TMS could prevent interaction between iron and chelating agents or ligands (Bryszewska et al., 2019). In this study, the bioavailability of iron was assessed by calculating two iron absorption parameters (transport, and transport efficiency) of iron by Caco-2 cells after the SGF and the SIF digestion. The transport efficiency of iron in traditional yeast bread (6.09%) was higher than that in sourdough bread (1.79%) (Bryszewska et al., 2019).

Folate is an essential vitamin B to humans and a lack of folate may give rise to anemia and pregnancy-related issues (Liu et al., 2015). Folate is vulnerable in the presence of oxygen and heat,

especially during cooking and storage. Liu et al. (Liu et al., 2015) used modified waxy starch (Hi-Cap® 100) as an encapsulant and sodium ascorbate as a stabilizer to encapsulate L-5-methyltetrahydrofolate with a ratio of 0.1:91:9 (folate/starch/sodium ascorbate, w/w/w), which was added into flour to make noodles. The recovery of free L-5-methyltetrahydrofolate and the encapsulated one in cooked noodles was 345 μg/100 g and 162 μg/100 g, respectively. Thus, this encapsulation system can improve the intake of folate in food (Liu et al., 2015). In another study, Hoyos-Leyva et al. (Hoyos-Leyva et al., 2018e) used taro starch as an encapsulant and prepared starch granule aggregation particles (**Figure 7**) loaded with L-ascorbic acid (vitamin C) by spray-drying. The effective encapsulation efficiency of the normal particles was 20.9%. Nonetheless, the porous cavities of normal particles could lead to the exposure of L-ascorbic acid to adverse conditions and further contributed to the degradation of L-ascorbic acid (Hoyos-Leyva et al., 2018e).

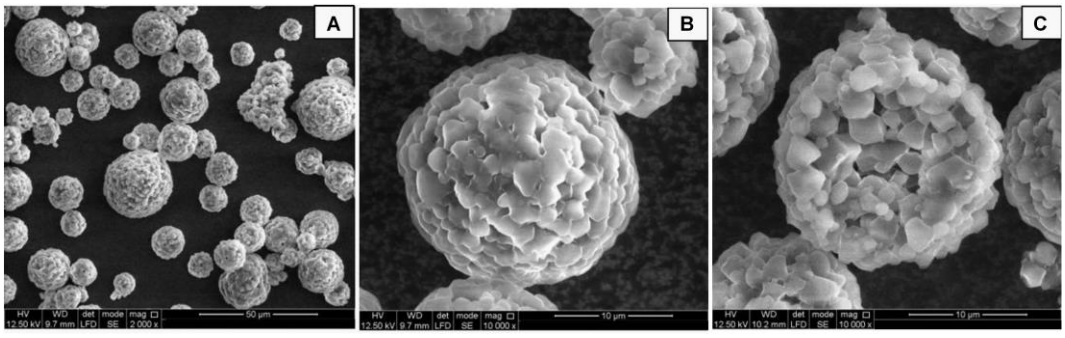


Figure 7. SEM images of starch granule aggregation particles loaded with L-ascorbic acid. Adapted from Ref. (Hoyos-Leyva et al., 2018e) with permission from Elsevier, Copyright 2018.

Caffeine has a strong irritation effect on the central nervous system and keeps one's spirit excited (Melocchi et al., 2020), the fast absorption of caffeine caused a short-lived stimulative effect

that lasted for only 2–3 h. The slow release of caffeine can help its role in promoting excitement. To control the release of caffeine after ingestion, Noor et al. (Noor et al., 2018) explored the release behavior of caffeine encapsulated in normal particles prepared using RS (rice starch subjected to HMT) in simulated gastrointestinal conditions. The free caffeine was fully released after 1.5 h whereas the release content of the encapsulated caffeine was 24 μg/mL and 29 μg/mL after 60 min and 180 min of digestion, respectively (Noor et al., 2018). The slow release of the encapsulated caffeine in RS could be attributed to the RS evading gastric digestion and finally slowly digested in the colon under the action of fermenting bacteria (Noor et al., 2018).

While nisin has a wide range of antibacterial activity on gram-positive bacteria, it can be easily affected by food ingredients and proteolytic enzymes (Hassan et al., 2019). Hassan et al. (Hassan et al., 2019) used a method of extrusion combined with vibration to prepare microgel particles loaded with nisin, where the encapsulants were mixtures of gelatinized or non-gelatinized Hi-Maize® RS and sodium alginate, and the antimicrobial activity and the release of the encapsulated nisin were determined. With the ratio being 0.5:1, 1:1 (sodium alginate/gelatinized starch, w/w), and 1:0.5 (sodium alginate/non-gelatinized starch, w/w), the release of nisin in skim milk after 26 days still kept at a high level (2.84 μg/mL) as compared with the original level (3.97 μg/ml). In addition, the count of *C. tyrobutyricum*, a harmful microorganism, in cheddar cheese that incorporated with the encapsulated nisin could be significantly reduced by approximately 1.4 log CFU/g (Hassan et al., 2019). The incorporation of RS in the complex encapsulant could form compact, spherical microgel particles with no cracks, which was in favor of the encapsulation of nisin and could extend the

release and activity of nisin during storage, thus reducing harmful microorganisms in cheddar cheese (Hassan et al., 2019).

Starch-based encapsulation systems can also be used in increasing the shelf-life of food products. For example, using fresh tiger nut milk as the core material and the mixture of inulin and OSA-modified tiger nut starch as an encapsulant, lyophilized microspheres with an average particle size of 1.01 μm were fabricated (da Costa Neto et al., 2019). The encapsulated tiger nut milk had pH stable in the range of 6.88–6.99 during 60 days of storage, showing good microbiological stability. In contrast, the pH of fresh tiger nut milk decreased from 7.00 to 4.03 within less than 10 days, and the acidity was probably a result of fungus growth and can further cause the decomposition of fresh tiger nut milk (da Costa Neto et al., 2019). Normal particles with a continuous surface without cracks or interruptions could be formed when inulin was combined with the OSA-modified tiger nut starch. This complex encapsulant had low permeability and a good protection effect and the smooth surface of particles could hinder the release of active substances (da Costa Neto et al., 2019).

Coenzyme Q10, as a nutritional supplement entrapped in microcapsules, was also used in food fortification (Cheuk et al., 2015). Cheuk et al. (Cheuk et al., 2015) developed microgel particles based on OSA-modified starch (Hi-Cap® 100) via high-pressure homogenization and freeze-drying to entrap coenzyme Q10. The obtained microgel particles in deionized water (0.4 g/mL) could keep stable overnight at temperatures up to 100 °C and were also stable at low-pH (3 and 5) citrate–phosphate buffers (1 mg/mL) after storage for 20 days (Cheuk et al., 2015). The microgel particles could be added to fruit juices and baked goods to improve their nutritional values (Cheuk et al., 2015).

4 Summary and future perspectives

The recent progress towards starch-based encapsulation systems (including porous starch, microgels, molecular aggregates, starch granule aggregates, and normal particles) has demonstrated their great potential in the design and processing of foods with enhanced functionality, flavors, sensory properties, and nutrition. Porous starch particles, molecular aggregation particles, starch granule aggregation particles, and microgel particles have been widely studied to deliver bioactive substances such as fatty acids, antioxidants (phenolic compounds and carotenoids), flavor ingredients, essential oils, iron, vitamins, probiotics, bacteriocins, co-enzymes and caffeine. Starch-based encapsulation systems have been used in various food products (e.g. cooked meat products, milk, beverage, bread, noodles, among others) and allow for the controlled release and target delivery of bioactives or nutrients and the extension of the shelf life of food products.

Over other encapsulation materials (e.g. proteins, lipids, and other polysaccharides), the advantages of starch-based encapsulation systems may be attributed to the following points: RS can escape the digestion in the small intestine and could be used for the target delivery of bioactive substances like probiotics; Amylose tends to form V-type inclusion complexes with hydrophobic substances and the formed complexes have good thermal stability and digestion resistance; Gelatinized starch can be easily transformed into a paste and have good film-forming ability; Starch itself is an important ingredient in food with a wide range of sources and low price and thus using it for the encapsulation of other food additives could receive high consumer acceptance.

Starch-based encapsulation systems in the form of microgel particles, normal particle, molecular aggregation particles and starch granule aggregation particles are currently the three most widely used particle forms. Besides, OSA modification and enzyme modification are also the most widely used modification methods for starch to achieve obtain encapsulation systems with suitable characteristics. Among various starch cultivars, normal maize starch, taro starch, and high-amylose starch have been mostly studied for developing encapsulation systems due to their wide availability, being easy to form molecular aggregates, starch granule aggregates, and normal particles, or excellent digestion resistance.

Despite the advantages and promising results achieved for starch-based functional micro-/nano-encapsulation systems, encapsulants based on only starch may not be capable enough of offering all functional characteristics. For example, OSA-modified starch as an encapsulation material has been widely used in starch-based encapsulation systems due to its amphiphilicity, non-toxicity, and excellent emulsification ability. However, encapsulating particles based on only OSA-modified starch usually have more wrinkles and lower oxidative stability (Wang et al., 2020c). To overcome this drawback, researchers tend to combine OSA-modified starch with other polysaccharides (e.g. insulin, maltodextrin, alginate, chitosan, xanthan gum, and β-cyclodextrin) or protein (e.g. gelatin and whey protein isolate) to prepare encapsulation systems. In addition, OSA-modified starch with too large a molecular mass might cause over-aggregation and low encapsulation efficiency due to the high viscosity and poor gravitational stability (Xiang et al., 2020).

The particle size of starch-based encapsulation systems prepared by existing techniques are usually uneven (Beirão-da-Costa et al., 2011; Benavent-Gil et al., 2018; Gomes et al., 2019; Hasani

et al., 2018; Noor et al., 2018; Wang et al., 2020a). Although there have been some studies (Al nuumani, Vladisavljević, Kasprzak, & Wolf, 2020; He et al., 2020; Jeong & Kim, 2011) on using the new microfluidic emulsification technique to achieve good mono-dispersity of microgel particles in liquids based on starch, chitosan, or gelatin, only a few studies involved the use of starch as an encapsulation material without solidifying the emulsion droplets into microgel particles. More research is deserved to manufacture starch-based micro- or nanoparticles with uniform size for encapsulation purpose.

Moreover, it is worth noting that the controlled release and target-delivery performance of starch-based encapsulation systems were mainly based on simulation media but there is a lack of *in vivo* data to support the delivery efficacy. Hence, research to further verify the controlled release and target-delivery behavior of starch-based encapsulation systems in more complex *in vivo* environments could be highly necessary. Besides, we should consider the safety of modified starch to the human body such as OSA-modified starch, and the safe dosage of modified starch needs further evaluation (Gomes et al., 2019).

Declarations of interest

none

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Tables

Table 1 Types of starch-based encapsulation systems

Type	Fabrication methods/formation mechanism	Further process
Porous starch	- Enzymatic modification - Gelatinization + atomization - Solvent exchange - Heat-moisture treatment + enzymatic modification - Combination of enzymatic and ultrasonic treatments - Sacrifice template approach	- Freeze-drying - Oven-drying - Microwave-drying

Microgels	- Syringe-type extrusion and ionic gelation (e.g. to form hydrogels by electrostatic interaction between carboxyl groups and calcium ions) - Chemical cross-linking (e.g. oxidized starch or carboxymethyl starch with trisodium metaphosphate) - Emulsification (e.g. O/W, W/W, and multiple) to obtain droplets and gelation via emulsion cross-linking or mixing with starch hydrogel or heating the emulsion- starch dispersions	- Freeze-drying - Spray-drying - Encapsulating drugs through electrostatic interaction (for cross-linking microgels) - Supercritical fluid extraction of emulsions + freeze-drying

Molecular aggregates	- Hydrophobic interaction and electrostatic interaction of the polymer (self-assembly) - Complexation of amylose and hydrophobic ligands	- Freeze-drying - Spray-drying
Starch granule aggregates	- Spray-drying a starch dispersion to form starch spherical aggregates with or without a bonding agent - Spray-drying the mixture of starch dispersion and encapsulated materials	- Supercritical solvent impregnation
Normal particles	- Mixing the encapsulant and the encapsulated material in a solvent (which may be treated by ultrasonication)	- Freeze-drying - Spray-drying - Hot-melt extrusion

	- Mixing the encapsulant and the encapsulated material in a highly concentrated state	

1721

1722 **Table 2** Fabrication methods for porous starch

Method	**Starch source**	**Formulation and preparation**	**Major findings**	**Reference**
Enzymatic modification	Normal maize starch, intermediate-amylose rice starch	AMG/starch (16.5:1 U/g), AM/starch (11:1 U/g)	The pore size and pore frequency of porous maize starch (0.59 μm^2, 4.47%) and porous intermediate-amylose rice starch (0.19 μm^2, 3.69%) obtained via AMG modification were higher than those of the AM-modified starches (0.13 μm^2, 1.57%; 0.03 μm^2, 0.43%)	(Benavent-Gil et al., 2018)
	Purple sweet potato starch	Starch/CADHPBS (1:3 g/mL), temperature (35–60 °C), pH (4.0–5.5), stirring (10 min), AA/GA (3:1 g/g)/starch (0.2%–1.4% w/w total enzymes), reaction (8–14 h), termination	- Optimal reaction conditions: enzymes/starch (0.6%), reaction time (12 h), temperature (45 °C) and pH (5); - Maximum adsorption capacity	(Lei et al., 2018)

		reaction (0.1 M NaOH), adjusting pH (7), centrifugation (3000 rpm, 5 min), washing (distilled water, three times), drying	(~43.84%)	
	Native maize starch (22.2% amylose)	Starch/PBS (5% w/v), enzyme/starch (0.5 U/mg), enzyme (PA, P, AM), stirring (250 g, 37 °C, 30 min, 120 min), centrifugation (4000 g, 5 min), washing (ethanol, three times), drying (40 °C, overnight)	- The surface pores and pore sizes were similar among porous starches treated with three different enzymes; - Larger pores and deeper cavities were formed with the increased enzyme treatment duration.	(Li et al., 2016a)
	Normal maize starch	Starch/AASAB (20 wt%, pH 4.2), AMG/starch (10 U/g), incubation (50 °C, 350 rpm, 0.5–24 h),	- The pores were mostly circular with various depths; - The starch granules remained	(Yang et al., 2019)

		centrifugation (2000 g, 5 min), washing (Mill-Q water, three times), drying (air-drying, 45 °C)	intact after 15 h of reaction but broken after 24 h of reaction.	
	Normal maize starch	Starch/water (1:8 g/mL), AA/GA (6:1 g/g)/starch (0.02:1 w/w total enzymes), reaction (50 °C, pH 5.5, 12 h), inactivating enzymes (0.1 M NaOH), adjusting pH (10), filtering and washing (distilled water), drying	The mass ratios of AA/GA and total enzymes/starch, ratio of starch/water, reaction pH, temperature and time affected the adsorption capacity of the porous starch.	(Zhang et al., 2012)
	Normal maize starch	Starch/PBS (1:5 g/mL, pH 6.0) or starch/SAB (1:5 g/mL, pH 4.0), enzyme (AMG, AM), enzyme/starch (4:1 or 5:1 U/g), incubation (50 °C, 24 h, 50 rpm), homogenization (1 min), centrifugation	The porous starch treated by AMG possessed more and bigger pores than the porous starch treated by AM.	(Dura et al., 2014)

		(7000 g, 5 min, 4 °C, two times), inactivating enzymes (boil in water, 10 min), freeze-drying		
	Normal maize starch	Starch/PBS (1:5 g/mL, pH 6.0) or starch/SAB (1:5 g/mL, pH 4.0), enzyme (AMG, AM, CGTase, BE), enzyme/starch (5.5:1–55:1 for AMG/AM, 0.1:1–1:1 for CGTase, 500:1–5000:1 for BE U/g), incubation (50 rpm, 50 °C, 2 h), centrifugation (7000 g, 15 min, 4 °C), inactivating enzymes (boiled in water, 10 min), washing (water, two times),	- Porous starches were obtained via the enzymatic treatment of normal maize starch at sub-gelatinization temperature; - The type and level of enzymes significantly affected the pore size and pore area distribution of porous starch; - The largest or smallest holes could be obtained with AMG or CGTase, respectively.	(Benavent-Gil & Rosell, 2017)

		homogenization (1 min), centrifugation again, freeze-drying		
	Potato starch (19.89% amylose), normal maize starch (22.01% amylose), wheat starch (25.14% amylose) and sweet potato starch (16.74% amylose)	- Starch/SAB (30 wt%, pH 6.8), AA/starch (1500:1 U/g dry weight of starch), incubation (48 °C, 8 h), stopping reaction (1 mol/L NaOH), adjusting pH to 5.5 (0.02 mol/L CADHPBS), GA/AA-modified hydrolysate (1200:1 U/g), incubation (50 °C, 4 h), stopping reaction (1 mol/L NaOH), precipitation (100% alcohol), washing (distilled water), freeze-drying	- The four starches treated with GT-BE generated more pores than the starches treated with GA-AA; - The type of pores depended on the enzyme treatment.	(Guo et al., 2020)

		- Starch/SAB (30 wt%, pH 5), GT/starch (1500:1 U/g dry weight of starch), incubation (45 °C, 3 h), stopping reaction (1 mol/L NaOH), adjusting pH to 5.5 (0.03mol/L SAB), BE/GT-modified hydrolysate (300:1 U/g dry weight of starch), incubation (50 °C, 4 h), stopping reaction (1 mol/L NaOH), precipitation (98% alcohol), washing (distilled water), freeze-drying		
Gelatinization + atomization	High-amylose maize starch (Hylon VII)	Starch/water (8% w/w), heating (140 °C, 4 °C/min), stirring (330 rpm), heat	The pore structure of porous microspheres collapsed due to the high	(Glenn et al., 2010)

		preservation (140 °C, 10min), cooling (85 °C), atomization (100 mL/min, 0.55 MPa), collection (95% ethanol)	surface tension caused by the evaporation of the water trapped within the pores when partially dehydrated (70% ethanol), whereas the pore structure was preserved during air-drying when dehydrated in 100% ethanol.	
Solvent exchange	Normal maize starch	Starch/water (5% w/v, 90 °C, 0.5h), cool (5 °C, 48h) cut into cylinders (1 × 1 cm), frozen (−10 °C), solvent exchange (100% ethanol, three times, 1 h each time), freeze-drying	The honeycomb structure of the porous starch resulted in high encapsulation efficiency (94.05%) and increased the absorbability of bioactive compounds.	(Oliyaei et al., 2020)
	Normal maize starch	Starch/water (1:20 g/mL, 90 °C, 0.5 h), cooling (5 °C, 48 h), cutting into	The freeze-drying porous starch at a high ratio of ethanol (ethanol/water,	(Oliyaei et al., 2019)

		cylinders (1 × 1 cm), frozen (−10 °C), ethanol/water (100:0, 80:20, 60:40, 40:60), solvent exchange (three times, 1 h), drying (50 °C, 6 h + 105 °C, 2 h) or freeze-drying or microwave (2450 MHz, 6 min)	100:0) had larger pores and the highest adsorption capacity (4.75, g/g) compared with that obtained by drying (3.83, g/g) and microwave treatment (1.29, g/g)	
Repeated heat-moisture treatment + enzymatic modification	Native wheat starch (24.45% amylose), A-type wheat starch (31.45% amylose), B-type wheat starch (31.21% amylose)	Starch/PBS (1:3 g/mL, pH 5.8), equilibration (50 °C, 10 min), AA+GA (1:3 U/U), incubation (50 °C, 8 h, 170 rpm), deactivating enzymes (100% ethanol), centrifugation (2500 g, 10 min), washing (distilled water, three times), freeze-drying	- Compound enzymatic hydrolysis initiated from the starch granule surface and penetrated into the starch granule interior; - Repeated heat-moisture treatment promoted the enzymatic hydrolysis and formed more pores compared	(Xie et al., 2019)

			with the porous starch only modified by compound enzymes.	
Combination of enzymatic and ultrasonic treatment	Pure wheat starch (obtained from Fars-Glucosin Co.)	Starch/water (20% w/w, pH 6.2), AM/starch suspension (0.2%, 0.4%, 0.6% w/v), incubation (45 °C, 24 h), sonication (35 kHz, 240 W, 100%), duration (20, 40 and 60 min), centrifugation (3000 g, 20 min), washing (distilled water, three times), drying (80 °C, 12 h)	- Combination of AM and ultrasound treatment increased the size and quantity of the micropores of the porous starch; - However, sonication (especially 40 min and 60 min) after enzyme treatment destroyed some starch granules.	(Majzoobi et al., 2015)
Sacrifice template approach	Potato starch (21.2% amylose)	Ethanol/water (15 wt%), $CaCO_3$ NPs/ ethanol solution (1:400, 1:200, 1:100, 1:66.7, 1:50 g/mL), sonication (5 min),	Maximum adsorption capacity (86.7%) was obtained under the optimal reaction	(Fang et al., 2020)

		starch/water (1:15 g/mL), heating (100 °C, 30 min, 200 rpm), mixing (1200 rpm, 30 min), gelation (4 °C, 12 h), centrifugation (4000 rpm, 5 min), washing (distilled water), dispersion in EDTA solution (0.5 M, pH 7.4), stirring (100 rpm, 40 min), centrifugation and repeated washing	condition ($CaCO_3$ NPs/starch, 3:2, w/w).	

Abbreviations: Pancreatic α-amylase (PA), pancreatin (P), fungal α-amylase (AM), amyloglucosidase (AMG), acetic acid-sodium acetate buffer (AASAB), citric acid–disodium hydrogen phosphate buffer solution (CADHPBS), sodium acetate buffer (SAB), cyclodextrin glycosyltransferase (CGTase), branching enzyme (BE), glycosyltransferase (GT), α-amylase (AA), glucoamylase (GA), nanoparticles (NPs), ethylenediaminetetraacetic acid (EDTA)

1728 **Table 3** Fabrication methods for starch-based particle-type encapsulation systems

Fabrication methods	**Forms**	**Particle size**	**Advantage/disadvantage**	**Substances to be encapsulated**	**References**
Emulsification	Pickering emulsion colloid	–	Advantage: reduce lipid oxidation; superior storage stability	β-carotene, lutein	(Li, Zhang, Li, Fu, & Huang, 2020b; Li et al., 2020c)
Emulsification + oven drying	Microgel particles	10–40 μm	Advantage: high encapsulation efficiency	Protein	(Yang et al., 2020a)
Spray-drying	Normal particles	1.6–250 μm	Advantage: maintain antioxidation capability; good thermal stability; resistant to high temperature during spray-drying and gastrointestinal conditions; high retention ability and	Probiotics, vitamins, flavors, essential oils, xanthophylls, anthocyanins, polyphenols,	(Alfaro-Galarza et al., 2020; Avila-Reyes et al., 2014; Bamidele, Duodu, & Emmambux, 2019; Beirão-da-Costa et al., 2011; Das, Goud,

			encapsulation efficiency; controlled release and target delivery; digestion resistance; Disadvantage: low water stability	lipophilic carotenoids, ascorbyl palmitate	& Das, 2019; Ding et al., 2020; Gonzalez-Soto, de la Vega, García-Suarez, Agama-Acevedo, & Bello-Pérez, 2011; Hong et al., 2019; Hoyos-Leyva et al., 2018a; Hoyos-Leyva, Bello-Pérez, Agama-Acevedo, & Alvarez-Ramirez, 2018b; Hoyos-Leyva, Bello-Pérez, & Alvarez-Ramirez,

					2018c; Hoyos-Leyva et al., 2018e; Hoyos-Leyva et al., 2019; Marinopoulou et al., 2019; Pérez-Masiá et al., 2015; Reyes, Chotiko, Chouljenko, & Sathivel, 2018; Santiago et al., 2016; Souza et al., 2018; Subpuch et al., 2016)
Solution mixing + freeze-drying	Normal particles	182–255.05 nm	Advantage: good water solubility; good color stability	β-carotene, curcumin	(Li et al., 2016b; Spada et al., 2012)

		297.553 μm	–	Caffeine	(Noor et al., 2018)
Emulsification + freeze-drying	Microgel particles	0.3–27.3 μm	Advantage: good color stability and emulsion ability; high encapsulation efficiency; good thermal stability; Disadvantage: particles may have damaged structure	Fatty acids, pigments, essential oils, fish oil, probiotics	(Anwar & Kunz, 2011; Bilenler et al., 2017; Hasani et al., 2018; Marefati et al., 2015; No & Shin, 2019; Yildiz et al., 2018)
Dispersion-inverse gelation + freeze-drying	Microgel particles	1.71–1.84 mm	Advantage: high loading efficiency; maintain antioxidant activity; controlled release and target delivery	polyphenols	(Wang et al., 2020b)
Emulsification + spray-drying	Microgel particles	0.2–14.2 μm	Advantage: good thermal stability; good water	Pigments, β-carotene, lutein,	(Álvarez-Henao et al., 2018; Fang et al., 2019;

			dispersibility; good protection against oxidation; target delivery	conjugated linoleic acid, algal oil, phenolic compounds, essential oils	Fernandes et al., 2014; Gomes et al., 2019; He et al., 2016; Liang et al., 2013; No & Shin, 2019; Sharif et al., 2017a; Wang et al., 2020c)
Self-assembly	Molecular aggregation particles	10–1000 nm	Advantage: improved solubility of encapsulated material	Citrus flavonoids	(Xiang et al., 2020)
Electrospraying + fluidized bed coating	Normal particles	450 μm	Advantage: protect thermosensitive microorganisms; high encapsulation efficiency	Heat-sensitive probiotics	(Pitigraisorn et al., 2017)

Fluidized bed coating	Normal particles	462 μm	Disadvantage: poor storage stability	Strawberry flavor	(Pellicer et al., 2019)
Coacervation	Molecular aggregation particles	–	Advantage: reduced degradation rate of encapsulated material	Astaxanthin	(Zhao et al., 2019)
Complexation	Molecular aggregation particles	0.063–40 μm	Advantage: target delivery; high thermal stability	Menthol, *p*-coumaric acid, ferulic acid, fatty acids, ascorbyl palmitate	(Di Marco et al., 2020; Dries et al., 2017a; Dries et al., 2017b; Fanta et al., 2015; Gao, Zhang, Qiu, Fu, & Huang, 2020; Kenar, Compton, Little, & Peterson, 2016; Marinopoulou et al.,

					2019; Shi et al., 2019; Wang et al., 2019; Yeo et al., 2016; Zhang, Gladden, Guo, Tan, & Kong, 2020a)
Solvent evaporation	Normal particles	6.84–33.42 μm	Advantage: high encapsulation efficiency; good storage stability	Essential trace elements	(Gupta et al., 2015)
Complexation + freeze-drying	Molecular aggregation particles	–	Advantage: improved stability of bioactive ingredients; Disadvantage: particles may have damaged structure	Tangeretin	(Wang et al., 2020a)
Electrospraying	Normal particles	0.1–0.6 μm	–	–	(Pérez-Masiá et al., 2014)

Supercritical solvent impregnation	Normal particles	<10 μm	Advantage: maintain high antioxidant activity	Essential oils	(Almeida et al., 2013)
Supercritical fluid extraction of emulsions	Microgel particles	<20 μm	Advantage: improved antioxidant stability; good color stability; high solubility and stability in aqueous media; high encapsulation efficiency; reduced thermal or oxidative degradation	Yacon leaf extract, astaxanthin, carotenoid, capsaicinoids	(Aguiar et al., 2016; Cruz et al., 2020; Mezzomo et al., 2012; Santos et al., 2012)
Hot-melt extrusion	Normal particles	415.4–498.8 μm	Advantage: controlled release of microparticles	Resveratrol	(Chen et al., 2020)
Syringe-type extrusion + external ionic gelation	Microgel particles	106.6–1490 μm	Advantage: resistant to harsh conditions in the gastrointestinal	Probiotics	(Dafe et al., 2017; Poletto et al., 2019)

			tract and adverse storage conditions		
Complexation + sonication	Molecular aggregation particles	201.5–307 nm	Advantage: obtain smaller particles size	Conjugated linoleic acid	(Seo, Kim, & Lim, 2016)

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– Supplementary Material –

Starch-based materials encapsulating food ingredients: Recent advances in fabrication methods and applications

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Table S1 Starch-based encapsulation systems in food applications

Type of starch	**Core material**	**Fabrication methods**	**Type of particles**	**Function**	**Application**	**Reference**
For fatty acids						
Taro starch	Almond oil	Spray-drying	Starch granule aggregation particles	Improve oxidative stability at 65 °C or 120 °C	–	(Hoyos-Leyva, Bello-Perez, Agama-Acevedo, Alvarez-Ramirez, & Jaramillo-Echeverry, 2019)
OSA-modified starch (Capsul®)	Algal oil	Emulsification + spray-drying	Microgel particles	Increase solubility in cold water; improve oxidative and thermal stability at 120 °C or 200 °C; particles based on the starch/CS/IN composite had the	–	(Wang et al., 2020b)

				highest encapsulation efficiency (98.57%) and oxidative stability		
Porous starch (StarrierR®)	Sunflower oil	Plating	Porous particles	Improve oxidation stability during storage at 40 °C and under exposure to 600 Klux light	–	(Belingheri, Giussani, Rodriguez-Estrada, Ferrillo, & Vittadini, 2015)
OSA-modified starch (Hi-Cap® 100)	Fish oil	Spray granulation, spray-drying, freeze-drying	Microgel particles	Improve oxidative stability during storage at 21 °C and 30% RH without light; reduce unpleasant flavor; particles prepared via spray granulation had the best oxidative stability	–	(Anwar & Kunz, 2011)
Porous starch (obtained via enzymatic modification of purple potato starch)	Olive oil	Plating	Porous particles	Improve oxidative stability during storage at 60 °C or 90 °C and under exposure to oxygen (6 bar)	–	(Lei et al., 2018)

Normal maize starch, resistant maize starch (Hi-Maize® 260)	Maize oil	Emulsification + heating the emulsion-starch dispersions	Microgel particles	Delay lipid digestion in the SGF; hydrogel (formed in PBS, 10 mM, pH 7.0) containing 35 wt% normal maize starch as an encapsulant had the lowest initial digestion rate	–	(Tangsrianugul, Suphantharika, & McClements, 2015)
OSA-modified waxy maize starch (a commercial product from Cargill)	Conjugated linoleic acid	Emulsification + spray-drying	Microgel particles	Enable controlled release in the stomach of rats and target delivery to the small intestine; the composite encapsulant (starch/XG, 100:1, w/w) had the highest encapsulation efficiency (99.51%)	–	(Yang et al., 2020)
OSA-modified waxy maize starch (a commercial product from Fonovo Food Ingredients Co., Ltd)	Conjugated linoleic acid	Emulsification + spray-drying	Microgel particles	Improve thermal stability at 100–160 °C; improve oxidative stability during storage at 50 °C for 70 h; enable controlled release in the small intestine; the starch/XG (60:1, w/w)	–	(He et al., 2016)

				composite for encapsulation provided the best antioxidant protective effect		
Hydroxypropylated Starch (a commercial product from Sigma Chemicals)	Omega-3-rich canola oil	Emulsification + freeze-drying	Microgel particles	Improve the oxidative stability during storage (room temperature, 30 days); enable controlled release in the SGF and the SIF	–	(Yildiz et al., 2018)
For phenolic compounds						
High-amylose maize starch	Cocoa polyphenol extract	Complexation + spray-drying	Molecular aggregation particles	Enable target delivery to the lower digestive tract; mask unwanted bitterness	Mask the bitter taste of cocoa-nut creams	(Vitaglione et al., 2013)
OSA-modified starch (Capsul®)	Brazil nut residue extract	Spray-drying	Normal particles	Improve stability and maintain antioxidant capacity during storage at 27 °C for 120 days without light; and the starch/IN (1:1, w/w) composite for	–	(Gomes et al., 2019)

				encapsulation provided the most effective antioxidant capacity		
Debranched maize starch (obtained via enzymatic modification)	Tea polyphenols	Solution mixing + spray-drying	Normal particles	Enable controlled release in the SGF and the SIF; particles based on a composite of debranched maize starch (34% DB) and xanthan gum (40:1, w/w) had better control release performance	–	(Hong et al., 2019)
Modified maize starch (Pregeflo® CH 20)	Anthocyanin-rich Iranian borage extract	Solution mixing + spray-drying	Normal particles	Improve storage stability during storage (5 °C and 40 °C, 15 days and 60 days); maintain antioxidant capacity during storage (40 °C, 60 days); enable controlled release in the SGF and the SIF; particles based on MD/starch (1:1, w/w) had the	–	(Mehran, Masoum, & Memarzadeh, 2020)

				best storage stability and antioxidant capacity		
Normal maize starch, dafozhi starch, damaling starch, daguo starches	Ginkgo biloba extracts	Solution mixing + freeze-drying	Normal particles	Enable controlled release in the SGF and the SIF	–	(Wang et al., 2018a)
Oxidized potato starch (30% degree of oxidation)	Quercetin	Emulsion cross-linking	Microgel particles	Specific adhesion to the intestine; enable controlled release in the SIF and target delivery to the small intestine	–	(Li et al., 2020)
OSA modified starch (Capsul®)	Polyphenols	Spray-drying	Normal particles	Improve thermal stability during heating (100 °C, water) and color stability at different pH (1–10)	–	(Zanoni, Primiterra, Angeli, & Zoccatelli, 2020)
Oxidized tapioca starch (oxidized by hydrogen peroxide)	Polyphenol from coffee residue	Coacervation	Molecular aggregation particles	Maintain antioxidant activity at room temperature without light; the starch/ALG (1:3, w/w) composite with a ratio of encapsulant/ polyphenol (5%,	–	(Palupi & Praptiningsih, 2016)

				w/v) for encapsulation had the best antioxidant activity		
Normal maize starch	Resveratrol	Hot-melt extrusion	Normal particles	Improve stability under light (irradiation energy, 30 $W \cdot m^{-2}$); enable controlled release in water during storage at 5 °C for 5 days	–	(Chen et al., 2020)
Starches from horse chestnut, water chestnut and lotus stem	Catechin	Solution mixing + sonication + freeze-drying	Normal particles	Maintain bioactive properties during in-vitro digestion; enable controlled release in the SIF; particles based on water chestnut starch had the best controlled release performance	–	(Ahmad et al., 2019b)
Modified starch (a commercial product from ShangHai YuanYe Biotechnology Co., Ltd)	Fingered citron extract (rich in phenolic compounds)	Spray-drying	Normal particles	Improve storage stability and particles wettability in distilled water at 25 °C; particles based on the GA/MD/starch (1:1:1, w/w/w) composite showed best storage ability and wettability	–	(Mahdi et al., 2020)

Native banana starch, acetylated banana starch (modified by acetic anhydride)	Curcumin	Solution mixing	Normal particles	Enable controlled release in the SGF and the SIF; particles based on acetylated banana starch achieved less release in the SGF	–	(Acevedo-Guevara, Nieto-Suaza, Sanchez, Pinzon, & Villa, 2018)
Soluble starch	Curcumin	Solution mixing + sonication + freeze-drying	Normal particles	Improve solubility in water; improve stability in water (at least 2 weeks) or under irradiation (UV, 24 h) at room temperature; maintain antioxidant ability at room temperature without light; starch particles containing 3 wt% curcumin had the best antioxidant ability and stability in water	-	(Li, Shin, Lee, Chen, & Park, 2016)
Pea starch (40% amylose content), mung bean starch	Catechin, epicatechin, proanthocyanin	Dispersion-inverse gelation + freeze-drying	Microgel particles	Enable controlled release in the SGF and the SIF; improve antioxidant activity	–	(Wang et al., 2020a)

(37.5% amylose content)						
Modified starch (Capsul® 0800)	Anthocyanins	Spray-drying	Normal particles	Improve stability during storage at 25 °C for 90 days without light; particles based on the GA/starch (1:1, w/w) composite provided the best protection	Enhance food color	(Santiago et al., 2016)
For carotenoids						
Mung bean starch, rice starch, OSA-modified starch (a commercial product from Ingredion)	β-carotene	Emulsification, emulsification + heating the emulsion-starch dispersions	Hydrogel particles, emulsion colloid	Enable target delivery to the small intestine	Nutrition fortification for lipophilic nutraceuticals	(Lin, Liang, Ye, Singh, & Zhong, 2017; Mun, Kim, & McClements, 2015; Mun, Kim, Shin, & McClements, 2015)
OSA-modified waxy maize starch	β-carotene	Emulsification	Emulsion colloid	Enable controlled release in the SGF and the SIF	–	(Lin, Liang, Zhong, Ye, & Singh, 2018)

Modified starch (Capsul®)	Lycopene	Spray-drying	Normal particles	Reduce degradation during storage and improve antioxidant activity; particles based on the MD/starch (1:1) composite had the best antioxidant activity	–	(Souza et al., 2018)
Modified starch (Clear-gum® CO 01)	Saffron and beetroot extracts	Solution mixing + Freeze-drying	Normal particles	Improve stability during storage at 40 °C for 10 weeks; particles based on the GA/starch (1:1, w/w) composite had the best color stability in chewing gum	Color enhancement for chewing gum	(Chranioti, Nikoloudaki, & Tzia, 2015)
OSA-modified kudzu starch	Astaxanthin	Complex coacervation	Molecular aggregation particles	Improve stability during storage at 25 °C for 10 days; particles formed at pH 6 and based on the gelatin/starch (1:1) composite retarded astaxanthin degradation most effectively	–	(Zhao et al., 2019)
OSA-modified waxy rice starch	Paprika pigment	Emulsification + freeze-drying or spray-drying	Microgel particles	Improve color stability and thermal stability after heating (boiling water bath, 30 min);	–	(No & Shin, 2019)

				particles prepared via freeze-drying had better thermal stability		
Modified starch (Capsul® MFY -212)	Lycopene	Emulsification + spray-drying	Microgel particles	Improve stability during storage at 10 °C and 25 °C for 73 days; particles contain 5 wt% lycopene had the best storage stability	Cake pigmentation for cake	(Rocha, Fávaro-Trindade, & Grosso, 2012)
Modified starch (Capsul®)	Lutein	Spray-drying	Normal particles	Enhance storage stability during storage (25 °C and 40 °C, 22 days, RH 20%); enhance thermal stability during storage (70 °C and 90° C, 2h, RH 20%); particles based on the starch/IN (1:1) composite had the highest encapsulation efficiency (80%) and retention value (87.9%)	Extend food shelf-life	(Ding et al., 2020)

Porous maize starch (obtained via solvent exchange)	Fucoxanthin	Solvent exchange + freeze-drying	Normal particles	Improve stability during storage (4 °C, 25 °C and 50 °C, 4 weeks); enable controlled release in the SGF and the SIF	–	(Oliyaei, Moosavi-Nasab, Tamaddon, & Fazaeli, 2020)
For flavor ingredients						
Porous starch (StarrierR®)	Liquid tomato flavor	Plating	Porous particles	Improve stability during storage at room temperature without light (22 °C, 6 months) and heat treatments (up to 120 °C)	Extend favor shelf-life of tomato sauce	(Belingheri, Ferrillo, & Vittadini, 2015)
Modified starch (Hi-Cap® 100)	Strawberry flavor	Spray-drying, freeze-drying, fluidized bed coating	Normal particles	Improve stability during storage without light (4 °C and 25 °C, 5 months); particles prepared via spray drying achieved the best results	–	(Pellicer et al., 2019)
Amylose from potato starch (Type III), normal maize starch (25% amylose	Menthone, menthol	Solution mixing + freeze-drying	Normal particles	Improve stability at different pH conditions (3, 5, 7.2 and 8), different temperatures (4–80 °C, 4h), and different storage time	–	(Ades, Kesselman, Ungar, & Shimoni, 2012)

content), waxy maize starch (<1% amylose content, high-amylose maize starch (70% amylose content)				(0–120 days); enable controlled release the aromas in the SSF		
Oxidized amylose	Linalool	Complexation + freeze-drying	Molecular aggregation particles	Enhance antimicrobial activity during storage at 37 °C for 24 h; enable controlled release at 20 °C and 40°C during storage; particles based on amylose had better controlling release performance whereas those based on oxidized amylose had better antimicrobial activity	–	(Zhou et al., 2016)
For essential oils						
Rice starch	Oregano essential oil	Spray-drying + supercritical solvent impregnation	Starch granule aggregation particles	Improve antioxidant activity during storage	–	(Almeida et al., 2013)

Short amylose (obtained by enzymatic modification of native waxy maize starch)	Menthone	Complexation + freeze-drying	Molecular aggregation particles	Improve antioxidant activity during incubation (room temperature, without light, 12 h) and thermal stability during incubation (25–100 °C, 1 h or 80 °C, 0–8 h); extend antimicrobial activity during incubation (37 °C, 0–8 h); enable controlled release during incubation (37 °C, 72 h); particles prepared at 90 °C had the best thermal stability	–	(Qiu et al., 2017)
Modified starch (Hi-cap® 100)	Lemon essential oil	Emulsification + freeze-drying	Microgel particles	Improve thermal stability (74.1–151.9 °C) and enable controlled release during storage (room temperature); particles based on the CS/starch (1.5:8.5, w/v) composite showed the highest encapsulation efficiency	–	(Hasani, Ojagh, & Ghorbani, 2018)

				(85.44%) and the longest release time		
Porous starch (a commercial product from Chongqing Taiwei Bioengineering Ltd)	Clove oil	Emulsification + spray-drying	Microgel particles	Enhance antifungal activity during storage (37 °C, RH 75%) for different times (up to 96 h); improve heat resistance during heat treatment (boiled for 30 min)	Extend the shelf-life of cooked meat products (fish, chicken, pork and beef)	(Wang et al., 2018b)
Jackfruit seed starch	Vanilla oil	Solution mixing + freeze-drying	Normal particles	Improve storage stability during storage at 60 °C for 250 days; enable controlled release during storage	–	(Zhu, Zhang, Tian, & Chu, 2018)
OSA-modified starch (commercial modified starch from Ingredion)	Rose essential oil	Emulsification + spray-drying	Microgel particles	Enable controlled release during storage at different temperature (4–50 °C, RH 32%, 5 days) and relative humidity (32–85%, 25 °C, 5 days); particles based on the starch/MD (2:1) composite with an	–	(Xiao, Kang, Hou, Niu, & Kou, 2019)

				encapsulatnt/encapsulated material ratio of 35:30 (w/w) had the highest loading capacity (45.27%)		
OSA-modified starch (Capsul®)	Rosemary essential oil	Emulsification + electrospraying	Microgel particles	–	–	(Biduski et al., 2019)
For probiotics						
Normal maize starch	*Lactobacillus plantarum*, *Staphylococcus xylosus*	Emulsification + freeze-drying	Microgel particles	Improve viability during heat treatment (70 °C, 20 min) and storage of sausages; enable controlled release in sausages	Extend the shelf-time of heat-treated sausages	(Bilenler, Karabulut, & Candogan, 2017)
Water chestnut starch	Camel milk-derived probiotics	Emulsification + freeze-drying	Microgel particles	Improve thermal stability during heat treatment (up to 80 °C, 5 min and 15 min); enable controlled release in the SGF and the SIF	–	(Ahmad, Gani, Hamed, & Maqsood, 2019a)

Modified huauzontle's starch (obtained via acid hydrolysis + extrusion)	*Lactobacillus rhamnosus* GG	Spray-drying	Normal particles	Improve stability during storage at 4 °C for 5 weeks	Extend the shelf life of probiotic green tea	(Hernández-Barrueta et al., 2020)
Porous starch (obtained from normal maize starch and intermediate-amylose rice starch via enzymatic modification)	*Lactobacillus plantarum* cells	Plating + freeze-drying	Porous particles, normal particles	Improve thermal stability during heat treatment (55 °C, 20 min and 35 min)	–	(Benavent-Gil, Rodrigo, & Rosell, 2018)
Crosslinked phosphorylated rice starch	*Lactobacillus casei*, *Lactobacillus brevis*, *Lactobacillus plantarum*	Emulsification + freeze-drying	Microgel particles	Improve storage stability during storage at 4 °C for 2 months; enhance thermal stability during heat treatment (up to 75 °C, up to 10 min); increase viability in the SGF and the SIF	–	(Ashwar, Gani, Gani, Shah, & Masoodi, 2018)

Normal maize starch	*Lactobacillus plantarum*	Extrusion + external ionic gelation method	Microgel particles	Improve viability in the SGF and the SIF; enable controlled release in the SGF and the SIF; improve storage stability during storage at 4 °C for 1 month; particles based on the PC/starch (1:3, w/w) composite had the best encapsulation efficiency and controlled release performance	–	(Dafe, Etemadi, Dilmaghani, & Mahdavinia, 2017)
Potato resistant Starch (provided by Harbin Institute of Technology)	Lactobacillus plantarum	Spray-drying	Normal particles	Improve viability during storage at 25 °C for 42 days; enhance viability in the SGF and the SIF	–	(Muhammad, Ramzan, Huo, Tian, & Bian, 2017)
For other food ingredients						
OSA-modified waxy maize starch (Capsul®, Hi-Cap® 100), OSA-	Vitamin E	Emulsification + spray-drying	Microgel particles	Enable suspension in water; improve storage stability during storage (4–35 °C, RH 73%, 60	–	(Hategekimana, Masamba, Ma, & Zhong, 2015)

modified tapioca starch (Capsul® TA)				days); particles based on Capsul® or Capsul® TA had better storage stability		
Enzymatically modified maize starch	Vitamin C	Spray-drying	Normal particles	Improve storage stability during storage (55 °C, RH 52.86%, 9 weeks); enable controlled release in the SSF, the SGF and the SIF; particles based on the GA/starch (16 h of hydrolysis) (17.5%, w/w) composite provided better protection during storage and in-vitro digestion	–	(Leyva-López et al., 2019)
Native taro starch	Vitamin C	Spray-drying	Starch granule aggregation particles	Improve stability during storage without light (55 °C, RH 13–72%, 6 weeks)	–	(Hoyos-Leyva, Chavez-Salazar, Castellanos-Galeano, Bello-Perez, &

						Alvarez-Ramirez, 2018)
Modified food starch	Vitamin A acetate	Emulsification + spray-drying or fluidized bed coating	Microgel particles, normal particles	Maintain oxidative stability	Extend food shelf-life	(Morozova et al., 2020)
Normal maize starch, high amylose maize starch (Hylon® VII)	Ascorbyl palmitate	Complexation	Molecular aggregation particles	Improve antioxidant activities during storage for 12 weeks under UV light (16 W/cm^2) or without light (40 °C)	–	(Bamidele & Emmambux, 2019)
Modified waxy maize starch (Hi-Cap® 100)	L-5-methyltetrahydrofolate	Spray-drying	Normal particles	Improve stability during cooking (boiling, frying)	Nutrition fortification for noodles	(Liu, Green, & Kitts, 2015)
Modified waxy maize starch (Hi-Cap® 100)	L-5-methyltetrahydrofolic acid	Spray-drying	Normal particles	Improve stability during baking and storage for 3–7 days	Nutrition fortification for breads	(Liu, Green, Wong, & Kitts, 2013)
A modified starch	Ferrous sulphate or ferrous lactate	Spray-drying	Normal particles	Improve the bioavailability in the SGF and the SIF	Nutrition fortification for breads	(Bryszewska et al., 2019)

Modified Starch (Hi-Cap® 100)	Ferrous sulphate + ascorbic acid	Solvent evaporation	Normal particles	Improve the bioavailability in the SGF and the SIF; particles based on the GA/MD/starch (4:1:1, w/w/w) composite with an encapsulant/absolute alcohol ratio of 1:10 (w/v) had the highest encapsulation efficiency (91.58%)	Nutrition fortification for milk, extend the sensory shelf-life of milk	(Gupta, Chawla, Arora, Tomar, & Singh, 2015)
Esterified tiger nut starch	Tiger nut milk	Freeze-drying	Normal particles	Improve stability during storage at 25 °C for 60 days without light	Extend the shelf-life of tiger nut milk	(da Costa Neto et al., 2019)
Resistant starch from rice starch (obtained via heat-moisture treatment)	Caffeine	Emulsification + freeze-drying	Microgel particles	Enable controlled release in the SGF and the SIF	–	(Noor, Shah, Gani, Gani, & Masoodi, 2018)
Porous starch (a commercial product from Chongqing	Antimicrobial lipopeptide	Emulsification + spray-drying	Microgel particles	Improve antimicrobial potency; particles based on the starch/MD (1:9, w/w) composite with an	–	(Wang et al., 2014)

Taiwei Bioengineering Ltd)				encapsulant/encapsulated material ratio of 5% had the best antimicrobial potency (25711.3 IU/g)		
OSA-modified starch (Hi-Cap® 100)	Coenzyme Q10	Emulsification + freeze-drying	Microgel particles	Improve stability during storage (4 °C and 25 °C, 3 weeks); good thermal stability at 90 °C; good stability at low pH (3 and 5)	–	(Cheuk et al., 2015)
Hi-maize resistant starch	Nisin	Extrusion method + vibrating + external ionic gelation	Microgel particles	Enable controlled release during incubation in skim milk media for 2 months; particles based on the ALG/non-gelatinized starch (1:0.5, w/w) composite had the highest encapsulation efficiency (33%) and the best controlled release performance	Extend the shelf-life of cheddar cheese	(Hassan et al., 2019)

Abbreviations: Simulated saliva fluid (SSF), simulated gastric fluid (SGF), simulated intestinal fluid (SIF), Gum Arabic (GA), Maltodextrin (MD), Inulsin (IN), sodium alginate (ALG), chitosan (CS), xanthan gum (XG), pectin (PC), relative humidity (RH), degree of branching (DB)

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