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Journal Article as: peer-reviewed accepted version (Postprint)

DOI of this document*(secondary publication): 10.26092/elib/2609

Publication date of this document: 24/10/2023

* for better findability or for reliable citation

Recommended Citation (primary publication/Version of Record) incl. DOI:

Hang Jiang, Tong Zhang, Joeri Smits, Xiaonan Huang, Michael Maas, Shouwei Yin, To Ngai, Edible high internal phase Pickering emulsion with double-emulsion morphology, Food Hydrocolloids, Volume 111, 2021, 106405, ISSN 0268-005X, https://doi.org/10.1016/j.foodhyd.2020.106405

Edible high internal phase Pickering emulsion with double-emulsion morphology

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Keywords:
High internal phase pickering emulsion
Double emulsion
Zein nanoparticle
Lecithin
Interfacial rheology

A B S T R A C T

Food-grade high internal phase Pickering emulsions (HIPPEs) unify the stability of Pickering emulsions and the advantages of detergent-based high internal phase emulsions (HIPEs), making them attractive as nutritional products. However, as oral delivery systems, HIPPEs are usually prepared in the form of o/w emulsions, which are suitable mainly for oleophilic active ingredients and may suffer from leakage during gastric digestion. To better protect and deliver hydrophilic cargo molecules, we developed a HIPPE-based w/o/w double emulsion system. Zein nanoparticles and soybean lecithin are found to have a synergistic effect in stabilization — using both natural emulsifiers together results in the formation of w/o/w double emulsions with improved stability, which is further confirmed by the interfacial tension and rheology of zein- and/or lecithin-laden oil-water interfaces. A combination of zein nanoparticles and lecithin achieves the fastest interfacial tension decrease, indicating an improved interfacial activity. Besides, lecithin contributes to the strong surface elasticity of the interfacial films, which makes the formed emulsions even stabler. Simulated digestion experiments suggest that the inner aqueous droplets can be strongly protected from gastric fluids. This edible HIPPE with double emulsion morphology provides new ideas for designing healthy foods for nutrients delivery.

1. Introduction

Particle-stabilized emulsions, namely Pickering emulsions (Aveyard, Binks, & Clint, 2003), have attracted strong attention in the pharmaceutics, agriculture, food, and personal-care fields in recent years (Berton-Carabin & Schroën, 2015; Calabrese, Courtenay, Edler, & Scott, 2018; Frelichowska et al., 2009; Tang, Quinlan, & Tam, 2015; Xia et al., 2018). Compared with traditional emulsions stabilized by surfactants, Pickering emulsions are highly stable due to the nearly irreversible interfacial adsorption of particulate stabilizers (Binks, 2002), and can potentially exhibit improved biocompatibility (Wu & Ma, 2016). When the volume fraction Φ of the dispersed phase exceeds 0.74 (Lissant, Peace, Wu, & Mayhan, 1974), a unique type of emulsion, known as high internal phase Pickering emulsion (HIPPE), will be obtained. With a large interfacial area and tunable viscoelasticity, HIPPEs have attracted considerable attention especially in food products and templates for porous materials, and great efforts have been devoted to obtaining stable HIPPEs using various particles as stabilizers (Hu et al., 2016; Kimmins & Cameron, 2011; Yang, Hu, Wang, & Binks, 2017; Yuan et al., 2017). Menner (Menner, Ikem, Salgueiro, Shaffer, & Bismarck, 2007) and Ikem (Ikem, Menner, & Bismarck, 2009) reported the preparation of HIPPEs stabilized by rigid particles like titanium dioxide and silica. Ngai and co-workers (Li, Ming, Wang, & Ngai, 2009), on the other hand, chose the soft poly (*N*-isopropylamide)-*co*-(methacrylic acid) (PNIPAM-*co*-MAA) microgel particles as emulsifiers, and the formed HIPPEs had an internal phase fraction as high as 90%. With such advantages, however, further application of the obtained HIPPE is hindered by its poor biocompatibility because of the potentially toxic polymer component. Thus, new types of natural and nontoxic stabilizers are desired for the formation of HIPPEs, and one of the possible sources is protein. Such

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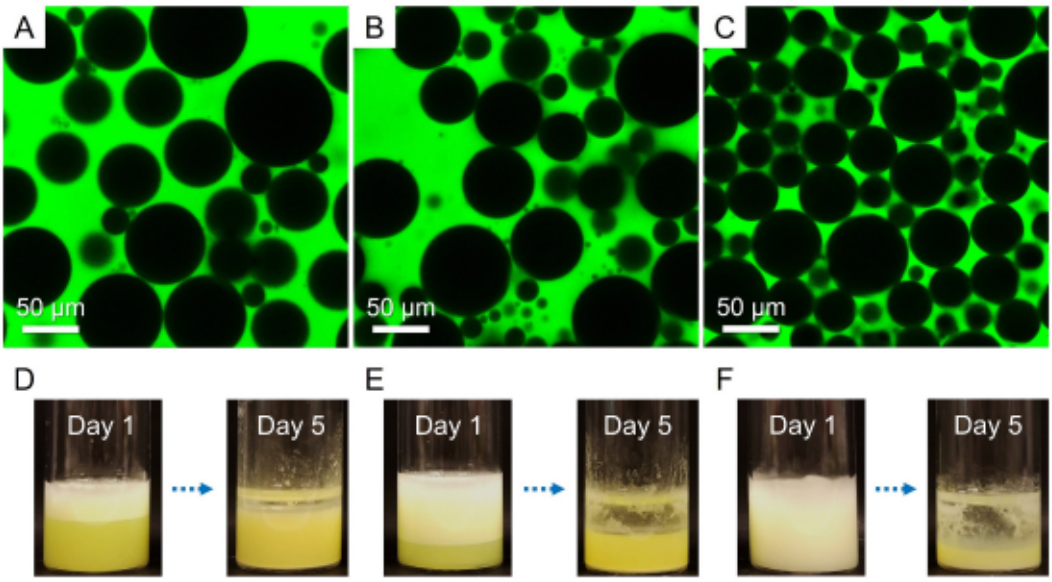

Fig. 1. Single oil-in-water (o/w) emulsion stabilized only by zein nanoparticles with the oil/water ratios of 1:3 (A), 2:2 (B), and 3:1 (C), respectively; (D–F) Appearance of the freshly prepared o/w emulsions and emulsions after 5 days under room temperature, which are corresponding to (A), (B) and (C). The aqueous phase is dyed with FSS.

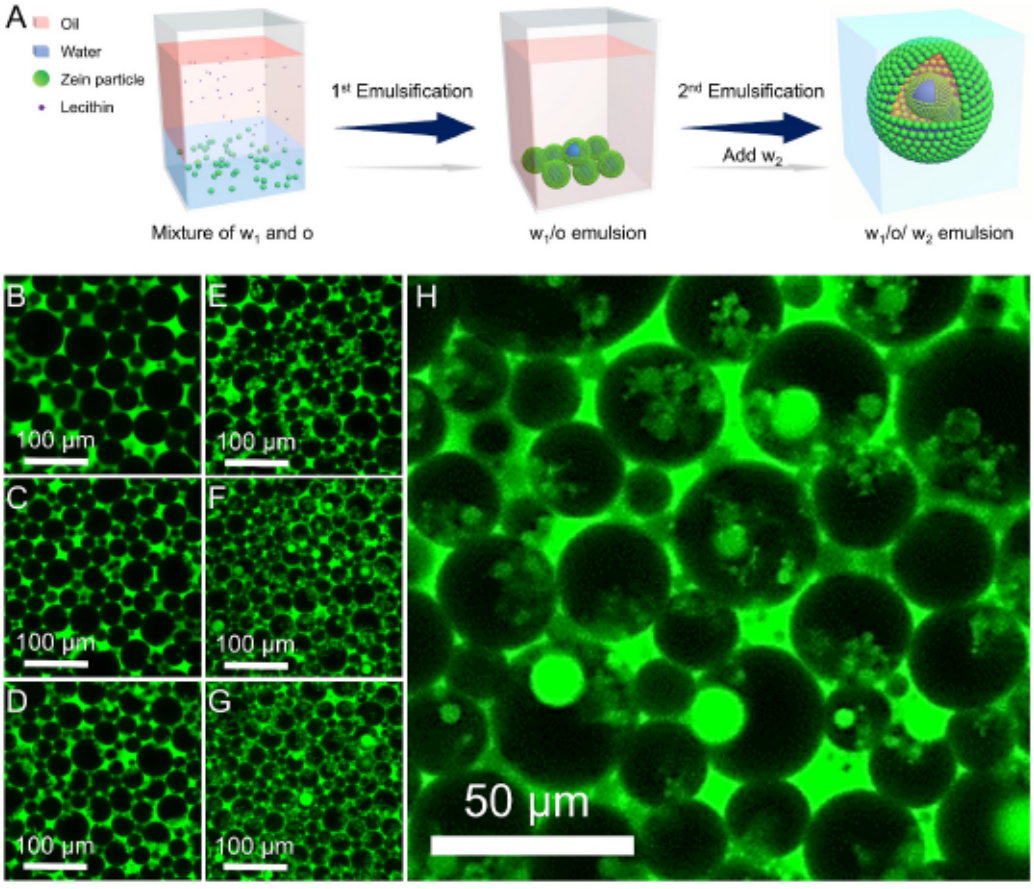

Fig. 2. (A) Schematic illustration of a two-step process for preparing w/o/w HIPPEs with the coordination of lecithin; (B–G) CLSM images of emulsions stabilized by zein nanoparticles and lecithin, and the lecithin concentrations are 0%, 0.1%, 0.25%, 0.5%, 1%, and 2%, respectively; (H) Magnification of section of (F).

protein-stabilized HIPPEs were first reported by Tan et al., in 2014 using colloidal gelatin particles as sole stabilizers (Tan, Sun, Lin, Mu, & Ngai, 2014). In recent years, whey protein isolate (WPI) (Wijaya, Van der Meeren, Wijaya, & Patel, 2017) and peanut protein isolate (PPI) (Jiao, Shi, Wang, & Binks, 2018) have also been utilized as stabilizers for HIPPEs.

Nevertheless, the reported food-grade particle-stabilized HIPPEs are all single emulsions (oil-in-water, o/w or water-in-oil, w/o). For o/w

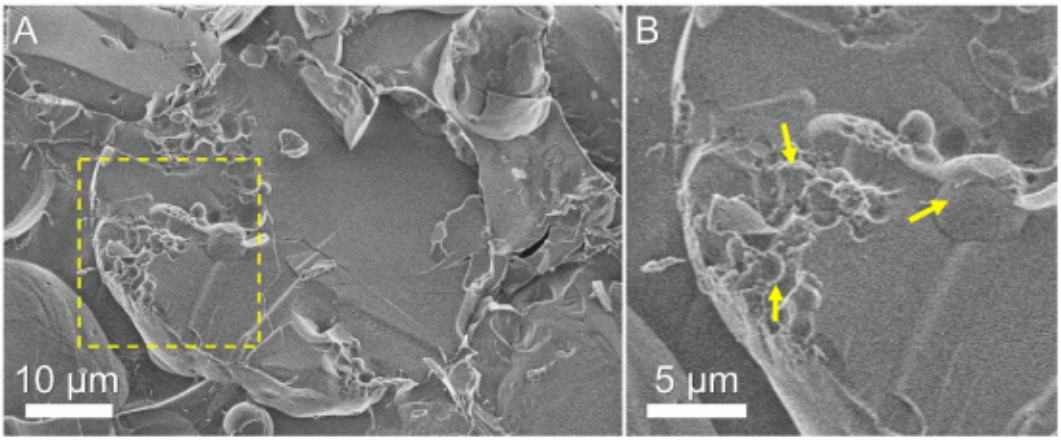

Fig. 3. Cryo-SEM images of double HIPPE stabilized by zein nanoparticles and lecithin (1%). The dashed rectangle in (A) is enlarged in (B). The yellow arrows in (B) indicate the location of inner aqueous droplets. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

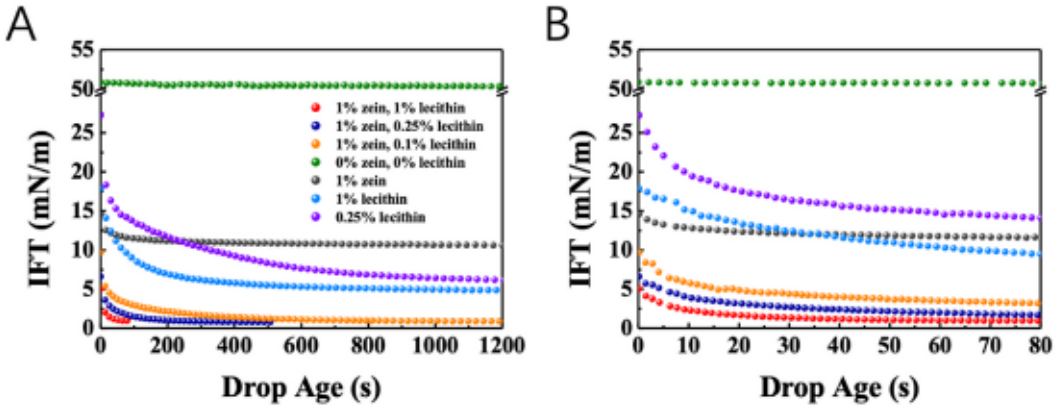

Fig. 4. (A) Dynamic interfacial tensions of squalane/water interface by adsorption of lecithin, zein nanoparticles, or both. (B) Magnified in the range of 0–80 s.

emulsions, only oil-soluble ingredients can be encapsulated, and most of them would be destroyed in the gastric juice environment, resulting in the leakage of the transported nutritional actives (Mantovani, Cavallieri, Netto, & Cunha, 2013; Tan et al., 2017). On the other hand, the development of w/o emulsions is rarely pursued due to their unpleasantly oily taste during oral delivery. Thus, double emulsions, in which dispersed droplets of one liquid phase contain smaller droplets of another phase (Dickinson, 2011; Muschiolik, 2007), are an ideal alternative for delivering water-soluble active ingredients, especially water-in-oil-in-water (w/o/w) emulsions (Chen et al., 2018). Besides good protection for nutrients, the advantages of dual-delivery, masking of off flavors, and reducing fat content also make double emulsions attractive (Aditya et al., 2015; Muschiolik, 2007; Sapei, Naqvi, & Rousseau, 2012; Serdaroğlu, Öztürk, & Urgu, 2016). However, the associative advantages of HIPPEs and double emulsions are not easy to achieve concurrently because it requires incorporating the morphology of a double emulsion into a HIPPE while avoiding catastrophic phase inversion at the same time. Lei et al. reported a HIPPE with double emulsion morphology recently, and poly (2-(diethylamino)ethyl methacrylate) (PDEA) microgel particles were used as the particulate stabilizers (Lei, Zhang, Shi, & Zhu, 2016). Although this example provides a versatile platform for fabricating porous scaffolds, it is far from application in the food area where all-natural materials are preferable. Hence, producing edible or food-grade HIPPE with double emulsion morphology is desirable.

Herein, we develop a w/o/w HIPPE based on all-natural materials, including the oil phase, aqueous phase, and stabilizers. In this work, zein protein nanoparticles serve as Pickering emulsifiers, and soybean lecithin is demonstrated to have a synergistic effect with zein protein nanoparticles for the stabilization of w/o/w HIPPEs. Based on that, the interfacial behavior of the zein protein particles and lecithin, as well as the interfacial rheology induced by them are studied. Moreover, the materials for the formulation of the emulsions, e.g. squalane, lecithin, zein protein, are all nutrients that are known for a variety of healthcare functions, and the squalane is a kind of high-purity oil compared with vegetable oils for avoiding influence of impurity from the oil, while the specially designed two-compartment structure equips the emulsion with multiple protections against simulated gastric juice. In this work, we try to utilize the synergy of lecithin and zein colloidal particles for designing HIPEs with double emulsion structure, as the HIPEs have the desired rheology and sensory properties, while the double emulsion structure provides the emulsions with alternative encapsulation. Our work provides a green and sustainable concept for preparing HIPPEs with double emulsion morphology and will open up a new and healthy platform for nutraceutical delivery and cosmetics.

2. Materials and methods

2.1. Materials

Zein (Z3625), fluorescent sodium salt (FSS), porcine lipase (L3126, 100–500 U/mg, type II), porcine pepsin (P7000, ≥250 U/mg), porcine pancreatin (P7545, 8 × USP specifications), and bile bovine (B3883) were purchased from Sigma-Aldrich (USA). Hydrochloric acid (HCl) (37%) and absolute ethanol were purchased from VWR (Fontenay-Les-

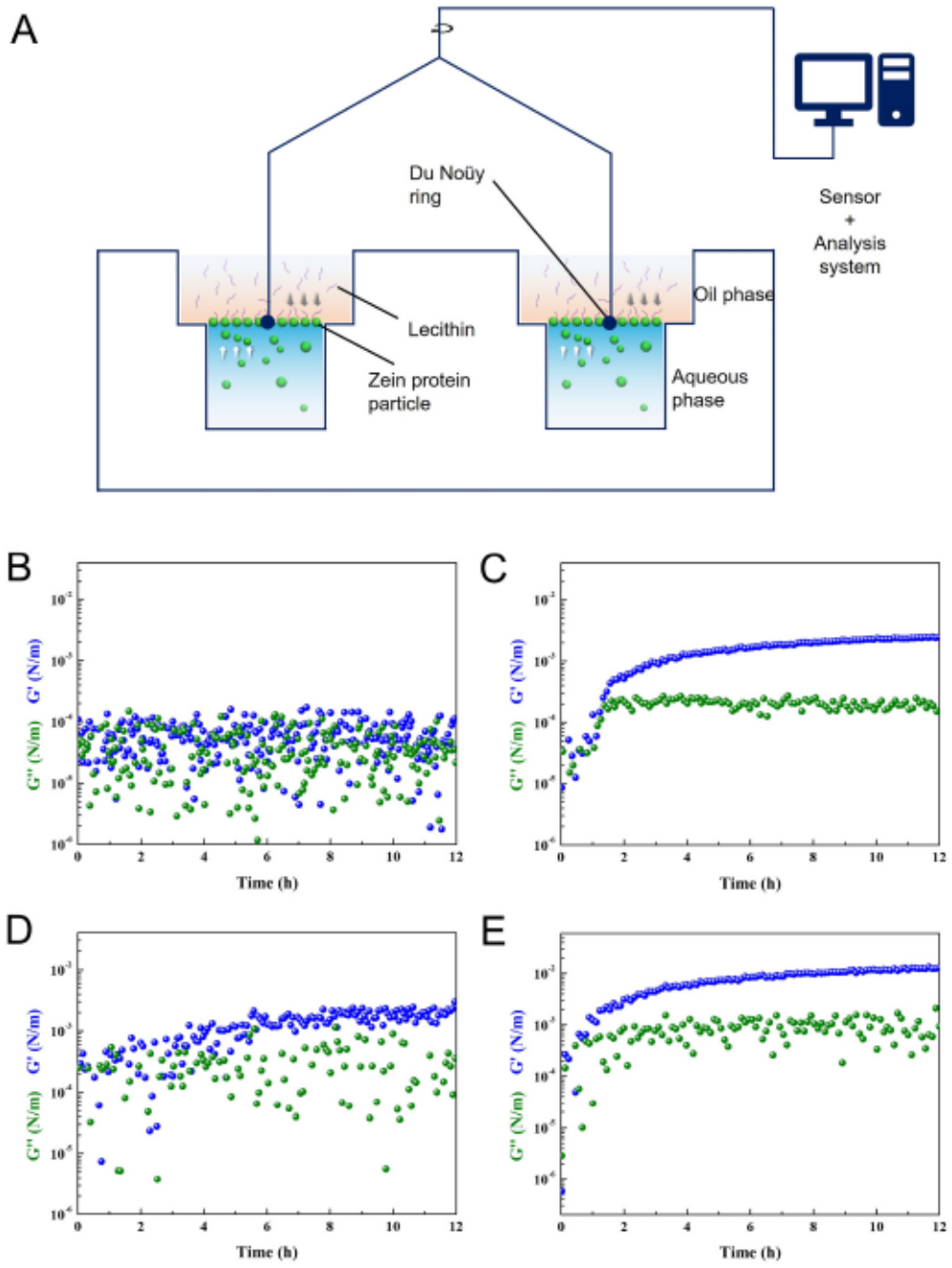

Fig. 5. (A) Scheme of interfacial shear rheology setup. Interfacial shear rheology of squalane/water interface by adsorption of lecithin, zein nanoparticles, or both: (B) 1% zein, (C) 1% zein and 0.25% lecithin, (D) 0.25% lecithin, (E) 1% lecithin, respectively.

Bois, France). Squalane (> 98.0%) and lecithin from soybean (L-α-Phosphatidylcholine) were purchased from TCI (Tokyo, Japan). Sodium hydroxide (NaOH, > 98.5%) was purchased from Scharlau (Sentmenat, Spain). Perylene was purchased from Acros Organics (USA). All products were used without purification unless otherwise stated. Deionized water (Milli-Q grade, 18.2 MΩ cm) was used in all experiments.

2.2. Synthesis of zein protein nanoparticles

Zein protein nanoparticles were fabricated by the anti-solvent method (de Folter, van Ruijven, & Velikov, 2012). Briefly, 1.0 g zein protein powder was dissolved in 40 mL 80% (v/v) ethanol aqueous solution under magnetic stirring to form the stock solution. Then the zein stock solution was added dropwise into 120 mL deionized water under vigorous stirring in 2–3 min. After that, the ethanol and part of water in the zein solution were removed under reduced pressure by rotary evaporation which resulted in a concentrated zein particle dispersion. The particle dispersions were stored at 4 °C.

2.3. Preparation of squalane-in-water emulsions stabilized by zein protein nanoparticles

Squalane-in-water Pickering emulsions with different oil/water ratios were solely stabilized by zein protein nanoparticles. Briefly, the total volume of the squalane and zein dispersion was kept as 4 mL, with the oil/water volume ratio varied from 1:3 to 2:2 and 3:1. The pH of the zein particle dispersion was tuned to 9 using 2 M NaOH solution beforehand,

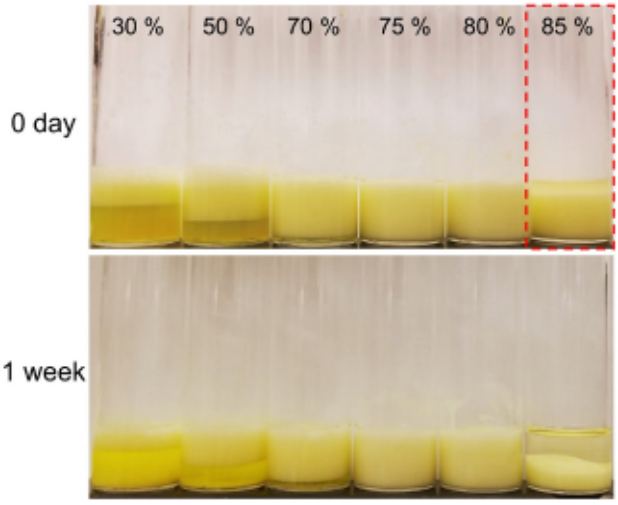

Fig. 6. Emulsions stabilized by zein nanoparticles and lecithin with varied squalane/water ratio through a two-step emulsification.

and then the zein dispersion was diluted with aqueous phase of pH 9 to a final concentration of around 1% (w/v) and stained with FSS. Then squalane oil was mixed with the zein dispersion, followed by homogenizing with a 10 mm head at 10,000 rpm for 30 s.

2.4. Preparation of w/o/w high internal phase Pickering emulsions stabilized by zein protein nanoparticles and lecithin

A two-step emulsification method was used to prepare w/o/w double emulsions. Both the inner and external aqueous phases (w_1 & w_2) were the dispersions of zein protein nanoparticles. The middle oil phase (o) was squalane containing soybean lecithin (0, 0.1, 0.25, 0.5, 1, 2% (w/v)). Typically, the primary w_1/o emulsion was prepared by mixing 0.3 mL of w_1 and 3 mL of oil phase, followed by homogenizing with a 10 mm head at 20,000 rpm for 1 min. After that, 0.7 mL of the zein dispersion (phase w_2) was added into the primary w_1/o emulsion and then homogenized at 10,000 rpm for 30 s. The percentage of double droplets was calculated by counting the emulsion droplets containing double emulsions and the total number of emulsion droplets from the confocal microscopy images.

2.5. Inversion of emulsion type by changing the oil fractions

To investigate the phase inversion of $w_1/o/w_2$ double emulsions, the above two-step emulsification process was adopted, but the total volume fractions of squalane were altered as 0.3, 0.5, 0.7, 0.75, 0.8, and 0.85. The ratio of w_1/o was kept as 1:10.

2.6. Measurement of dynamic interfacial tension of squalane/water interface

Measurement of dynamic interfacial tensions were conducted by an all-purpose contact angle measuring & contour analysis system (OCA 25, Dataphysics) at ambient temperature (25 °C). The interfacial tension was automatically calculated from the shape of a pendant zein dispersion drop in a bulk phase of squalane. For monitoring the adsorption of zein protein nanoparticles at the interface, 1% (w/v) zein particle dispersion (pH 9) was used as the pendant droplet. Pure water (pH 9) was used as blank control. Different amounts of lecithin (0, 0.1, 0.25, and 1% w/v) were added into the oil phase prior to the measurement.

2.7. Simulated digestion of water-in-squalane-in-water high internal phase Pickering emulsion

For the simulated digestion experiments, the $w_1/o/w_2$ emulsion was prepared as above with the middle oil phase dyed with perylene instead. Simulated gastric fluid (SGF) was prepared according to the method of Minekus et al. (Minekus et al., 2014). The emulsion (2 mL) was mixed with 12 mL of 1.25x SGF, and the pH of the mixture was adjusted to 2 by 1 M HCl, followed by the addition of 4 mL of pepsin solution (64 mg of pepsin). The resulting mixtures were incubated in 37 °C for 2 h under continuous stirring. A Nikon Eclipse Ti inverted microscopy (Nikon, Japan) was utilized to observe the morphology of emulsions during the digestion process.

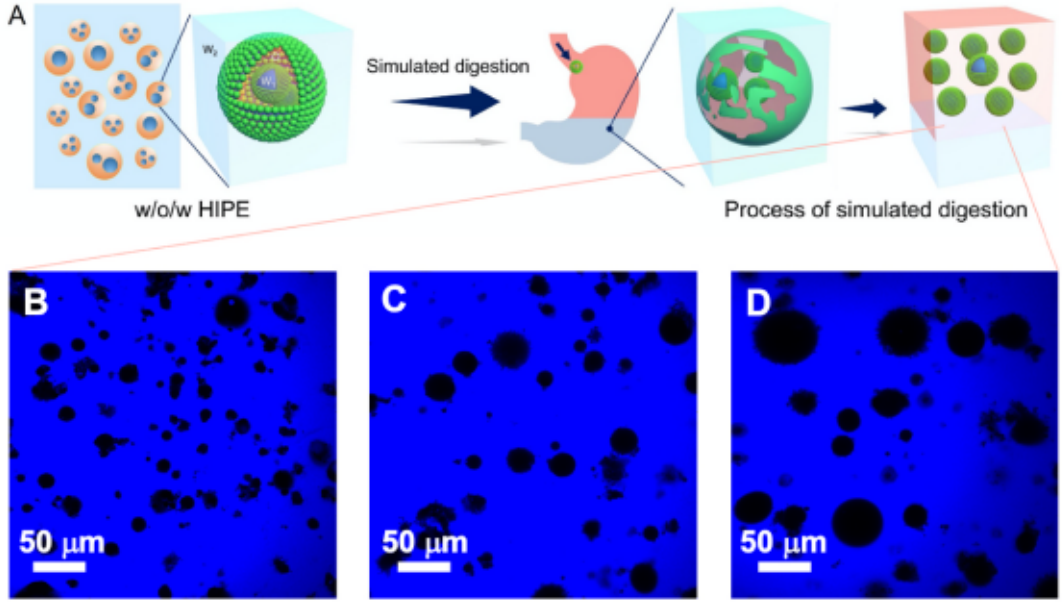

Fig. 7. (A) Schematic description of water-in-squalane-in-water double HIPPE in the digestion process; (B–D) CLSM image of the remaining w/o emulsion after digestion for 30, 60, and 120 min at 37 °C, respectively. The oil phase is stained with pyrene and appears blue color. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

2.8. Interfacial shear rheology of the zein-laden squalane/water interface in the presence of lecithin

Interfacial shear rheology was measured using the Double Wall Du Noüy ring geometry attached to a Discovery HR-3 hybrid rheometer (TA instruments) (Bollhorst et al., 2015). The Du Noüy ring was flame-cleaned before each measurement and brought into contact with the air-water interface before adding the oil phase. The volumes of aqueous phase and oil phase were kept at 5.8 mL and 3 mL, respectively. Zein nanoparticles were dispersed in the aqueous phase, while soybean lecithin was dissolved in squalane beforehand. The pH of the aqueous phase was maintained at 9, around which zein nanoparticles are highly interfacially active. Oscillatory measurements with defined frequency, amplitude and duration were carried out from which the interfacial viscoelastic shear moduli were calculated. Time sweep measurements were started right after forming a fresh oil-water interface. During these measurements the Du Noüy ring oscillated at 0.8% strain and 0.1 rad/s. The interface was monitored for 12 h. Amplitude (fixed frequency at 0.1 rad/s) and frequency (fixed strain at 0.8%) sweeps were conducted after the time tests to verify the viscoelastic threshold and independency of the inertia of the measurement system, respectively.

2.9. Characterization

Confocal laser scanning microscopy (CLSM) images of the emulsions were taken with a Nikon Eclipse Ti inverted microscope (Nikon, Japan). Cryo-SEM images were obtained with a S-4800 cold-cathode field-emission scanning electron microscope (Hitachi) equipped with a PP3010T Cryo-SEM preparation system (Quorum). Dynamic light scattering (DLS) measurements were conducted with a Zetasizer Nano ZS90 (Malvern).

3. Results and discussion

3.1. Oil-in-water (o/w) emulsions stabilized only by zein nanoparticles

To explore the possibilities of squalane-in-water HIPPE stabilized by zein nanoparticles, the oil/water ratio is gradually increased from 1:3 to 3:1 while keeping the concentration of the zein nanoparticles constant. The diameters of the zein protein nanoparticles are around 100 nm, and the globular morphology is clearly observed from SEM (Fig. S1). As shown from confocal laser scanning microscopy (CLSM) in Fig. 1A, when the oil volume fraction is 0.25, an oil-in-water (o/w) emulsion is easily prepared. Increasing the oil fraction to 0.5, as anticipated, also results in o/w emulsions (Fig. 1B), and the creaming layer of the o/w Pickering emulsion (Fig. 1E) is more obvious compared with the one with oil/water ratio of 0.25 (Fig. 1D). Noticeably, the emulsion droplets in Fig. 1B are not too crowded, and there are excess zein nanoparticles left in the continuous phase as the subnatant is not transparent, indicating the amount of zein nanoparticles is sufficient for adsorption at the emulsion interface. Further raising the oil/water ratio to 3:1 exceeds the critical oil fraction of HIPE (0.74). Interestingly, squalane-in-water HIPPE with oil fraction of 0.75 is successfully obtained, and the emulsion droplets tend to be in contact with each other (Fig. 1C), which is similar to reported HIPEs (Huang et al., 2019). However, serious phase separation is found in all three groups of emulsions within 5 days under room temperature. In this case, zein particles fail as sole stabilizers for HIPPEs. Complexation with other emulsifiers might be helpful.

3.2. Formation of w/o/w high internal phase emulsions: synergistic performance of lecithin and zein nanoparticles

It has been reported that lecithin can interact with zein nanoparticles and has synergistic effects with many protein-based particles for stabilization of emulsions (Chuacharoen & Sabliov, 2016; Dai, Sun, Wang, & Gao, 2016; Xue & Zhong, 2014). On accounting of this, lecithin is introduced to improve the stability of the zein-stabilized HIPPEs. Moreover, a double emulsion morphology can be surprisingly generated by a two-step emulsification process (Fig. 2A). Specifically, in the first step lecithin is dissolved in the squalane oil, while zein nanoparticles are initially dispersed in the water (w_1). After emulsification for the first time, a w_1/o emulsion is produced (Fig. S2). Then, a second dispersion of zein nanoparticles (w_2) is added to the primary w_1/o emulsion followed by emulsification, giving a $w_1/o/w_2$ double emulsion. By keeping the total oil/water ratio as 3:1, a HIPPE with double emulsion morphology is obtained.

Seeking to study the influence of lecithin on the two-compartment structure, varied concentrations of lecithin are used in the preparation of emulsions, ranging from 0 to 2%. Not surprisingly, HIPPE with no lecithin still results in single o/w emulsions via the two-step emulsification, as shown in Fig. 2B, which may be attributed to the difficulty of stabilizing w/o emulsion by zein nanoparticles alone. With 0.1% lecithin, inner aqueous droplets can be observed in several oil emulsion droplets, as seen in Fig. 2C. Further increasing the concentration of lecithin leads to more and more w/o/w double emulsion droplets, as demonstrated in Fig. 2D and E. It is worth noting that over 90% of emulsion droplets have the two-compartment structure with clear inner aqueous droplets if the lecithin concentration is as high as 1% (Fig. 2E & F). The results reveal that lecithin can help to form the double-emulsion structure, which is probably due to the fact that lecithin is an oleophilic surfactant and can stabilize w/o emulsions on its own (Knoth, Scherze, & Muschiolik, 2005).

In addition, the droplet size of the o/w_2 emulsion is decreased with increasing lecithin concentration, and the percentage of double droplets is increased with increasing lecithin concentration, as shown in Fig. S3. There seems to be no obvious difference of the morphology of emulsion droplets when the lecithin concentration is further increased from 1% to 2%, indicating that 1% lecithin is sufficient for producing double HIPPEs. The inner aqueous droplets are also directly observed by Cryo-SEM characterization (Fig. 3). Similar to the confocal images, the inner aqueous droplets stick to each other and they tend to stay near the o/w_2 interface, which is probably due to the density difference and hydrophobic interaction.

Moreover, we find that the addition of lecithin can not only help form HIPPEs with a high degree of double emulsion, but also improve the stability of the prepared double HIPPEs. Compared with HIPPE stabilized solely by zein nanoparticles, the addition of even 0.1% of lecithin can avoid demulsification and phase separation for at least a week, as shown in Fig. S4.

For conventional Pickering emulsions stabilized by hard particles, the three-phase contact angle between oil, water and particle is an important parameter as it indicates the preferred emulsion type while the oil-water interfacial tension is usually not significantly changed (Smits, Vieira, Bisswurn, Rezwan, & Maas, 2019). However, the latter is usually significant for soft particles as they have much richer interfacial phenomena to be exploited (Kwok, Sun, & Ngai, 2019; Li et al., 2015). In this work, we measure the squalane-water interfacial tension as a function of time using pendant drop tensiometry, by which the adsorption of lecithin or zein nanoparticles at the interface can be dynamically monitored.

Dynamic interfacial tension results show a cooperative interaction between zein and lecithin. The interfacial tension reaches the lowest value when both zein and lecithin are present (Fig. 4). A small amount of lecithin (0.1%) together with 1% zein results in a strong decrease of interfacial tension. Adding more lecithin (0.25%) increases the initial adsorption kinetics, but the IFT reaches a similar value before the IFT becomes so low that the droplet detaches. However, in the presence of zein, more lecithin (1%) can lead to an even lower interfacial tension compared with 0.25% lecithin, which means zein and lecithin cooperate at the interface, achieving a better interfacial activity than either zein or lecithin alone.

To assess the mechanical stability of the thin interfacial films

comprising the emulsion systems, interfacial shear rheology was performed at the oil/water interface. A simplified scheme of the device set-up is shown in Fig. 5A. Zein nanoparticles and lecithin were dispersed in the aqueous and oil phases, respectively, beforehand. The Du Noüy ring was placed exactly at the oil-water interface and oscillated sinusoidally as programed and the elastic (G′) and viscous (G″) moduli were recorded. It is found that zein alone cannot form an interfacial film strong enough to be detected (Fig. 5B), which is probably the reason why emulsions stabilized by zein alone destabilize within 5 days. Adding 0.25% lecithin results in an interfacial film with increased G′ and G'' (Fig. 5C). Without any zein nanoparticles, the pure lecithin-laden interface is barely detectable considering the signal-to-noise ratio of these measurements (Fig. 5D). Thus, a combination of zein and lecithin helps to form a strong and stable interface. However, increasing the lecithin concentration to 1% in the absence of zein, the strong increase in G′ and G″ shows good film stability (Fig. 5E). Thus, in the case of pure lecithin, the interface with 1% lecithin is slightly stronger than that formed by 0.25% lecithin and 1% zein. Considering that the strength is mainly contributed by lecithin, it can be deduced that more lecithin results in higher G′ and G″ regardless of the presence of zein, which is consistent with the emulsion results where the inner aqueous droplets of double emulsions are maintained the best when lecithin concentration is high (Fig. 2F & G), since the lecithin is dissolved in the middle phase, the synergetic effect contributes both to the w_1/o and o/w_2 interfaces.

3.3. Phase inversion of w/o/w double emulsions stabilized with zein nanoparticles and lecithin

The w/o/w emulsion is not always obtained if we keep increasing the oil fraction, as demonstrated in Fig. 6. By keeping the concentrations of zein nanoparticles and lecithin both unchanged as around 1% while changing the total squalane oil fraction from 0.3 to 0.85, we found w/o/w emulsions are fabricated only when the oil fraction is below 0.8. When the oil fraction is less than 0.7, creaming becomes apparent. Further increasing the fraction of oil to 0.85 leads to w/o emulsions instead, as shown in Fig. 6. This means that the maximum fraction of squalane for preparation of w/o/w HIPPE should be below 0.85. Since lecithin is added, all the emulsions are stable without phase separation in the process of storage at room temperature for one week.

3.4. Potential application in nutrients delivery protected against gastric juice

Normally, food-grade emulsions are o/w or w/o emulsions, and the challenge lies in the digestion process, as most of the o/w emulsions would be unstable in the gastric juice, and the resulting leakage can lead to loss of nutritional effect for inner actives. A w/o emulsion can effectively protect the encapsulated actives by the oily layer, but this kind of emulsion may have an unpleasant taste in the mouth because the continuous phase is oil liquid. Our water-in-squalane-in-water HIPPE system solves this problem by introducing an outermost aqueous phase. To verify the potential of the w/o/w HIPPE as a delivery system, we conducted digestion experiments in simulated gastric fluid, as shown in Fig. 7A. Upon being delivered to the stomach, although the outermost aqueous phase may be destroyed by the gastric juice, the remaining w/o emulsion can still protect the inner nutrients, ensuring a more successful delivery to the intestine. As shown in Fig. 7B–D, the simulated digestion results show that the inner w/o morphology remains intact after 120 min digestion in the simulated gastric juice.

4. Conclusions

In summary, w/o/w high internal phase Pickering emulsion can be successfully prepared via a two-step emulsification process, in which squalane can be used as the oil phase. The synergistic effect with lecithin improves the stability of zein protein particle stabilized double HIPPEs, and the double emulsion morphology is more obvious with increasing amount of lecithin. Moreover, the lecithin dramatically strengthens the elasticity of the oil/water interface. All materials or reagents involved in the fabrication of HIPPEs are from natural, renewable and nontoxic sources, and the simulated digestion results reveal that the double-emulsion structure provides strong protection for the interior droplets. Moreover, the structure of these HIPEs provide a better appearance, sensory quality, and unique rheology compared to conventional w/o/w double emulsions. We are convinced that this kind of edible w/o/w HIPPEs will have great potential to serve as a novel nutritional agents and carriers for personal care and healthcare applications.

CRediT authorship contribution statement

Hang Jiang: Conceptualization, Data curation, Investigation, Methodology, Resources, Writing - original draft, Visualization, Formal analysis. **Tong Zhang:** Conceptualization, Data curation, Investigation, Resources, Writing - review & editing, Formal analysis. **Joeri Smits:** Data curation, Software. **Xiaonan Huang:** Data curation, Methodology. **Michael Maas:** Supervision, Resources, Project administration, Writing - review & editing. **Shouwei Yin:** Supervision, Methodology, Project administration. **To Ngai:** Conceptualization, Supervision, Project administration, Writing - review & editing.

Declaration of competing interest

The authors declare no competing financial interest.

Acknowledgments

The authors gratefully acknowledge the financial support from the Hong Kong Special Administration Region (HKSAR) General Research Fund (CUHK14306617, 2130535) and the Germany/Hong Kong Joint Research Scheme Sponsored by the Research Grants Council of Hong Kong and the German Academic Exchange Service of Germany (G-CUHK410/18).

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.foodhyd.2020.106405.

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