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Encapsulation of curcumin in polysaccharide-based hydrogel beads: Impact of bead type on lipid digestion and curcumin bioaccessibility

Zipei Zhang [a], Ruojie Zhang [a], Liqiang Zou [a], Long Chen [b], Youssri Ahmed [c, d], Widad Al Bishri [c, e], Khadija Balamash [c, e, f], David Julian McClements [a, c, *]

[a] Department of Food Science, University of Massachusetts Amherst, Amherst, MA 01003, USA
[b] State Key Laboratory of Food Science and Technology, Jiangnan University, Jiangsu Wuxi 213122, China
[c] Department of Biochemistry, Faculty of Science and Production of Bioproducts for Industrial Applications Research Group, P. O. Box 80203, Jeddah, 21589, Saudi Arabia
[d] Experimental Biochemistry Unit, King Fahd Medical Research Center, King Abdulaziz University, P. O. Box 80203, Jeddah, 21589, Saudi Arabia
[e] Department of Biochemistry, Faculty of Science for Girl's, P. O. Box 80203, Jeddah, 21589, Saudi Arabia
[f] Department of Biochemistry, Vitamin D Pharmacogenomics Research, P. O. Box 80203, Jeddah, 21589, Saudi Arabia

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ABSTRACT

Curcumin was incorporated into three different delivery systems: free lipid droplets; lipid-loaded alginate beads; and, lipid-loaded carrageenan beads. Hydrogel beads were fabricated from polysaccharides (alginate or carrageenan) using an injection method combined with ion gelation (calcium or potassium, respectively). The delivery systems were passed through a simulated gastrointestinal tract (GIT) that included mouth, stomach, and small intestine phases. Light scattering and microscopy indicated that carrageenan beads had a relatively fragile structure that was easily disrupted in the GIT and released the encapsulated lipid droplets and curcumin. Conversely, alginate beads had a robust structure that remained relatively intact throughout the GIT and retained the lipid droplets and curcumin. The rate and extent of lipid digestion decreased in the following order: free lipid droplets > carrageenan beads > alginate beads. Curcumin bioaccessibility followed a similar order: free lipid droplets (73%) > carrageenan beads (33%) > alginate beads (16%). These results suggest that lipid droplets must be digested by lipase in order to release the encapsulated curcumin and to form mixed micelles capable of solubilizing the released curcumin. Overall, our results show that delivery systems with different structures and compositions can be designed to control the release of lipids and hydrophobic nutraceuticals in the GIT, which may be advantageous for the development of certain functional food products.

1. Introduction

Turmeric is a commonly used pigment, spice, and nutraceutical in foods due to its intense yellowish color, unique flavor profile, and biological activities (Prasad, Gupta, Tyagi, & Aggarwal, 2014; Syed, Liew, Loh, & Peh, 2015). Curcumin is the major bioactive agent in turmeric, and it consists of three closely related lipophilic molecules that have a number of phenolic groups and conjugated double bonds (Heger, van Golen, Broekgaarden, & Michel, 2014; Prasad et al., 2014). The potential pharmaceutical activity of curcumin has been established for numerous diseases, including multiple myeloma, rheumatoid arthritis, cystic fibrosis, inflammatory bowel disease, and colon cancer (Heger et al., 2014; Prasad et al., 2014; Wilken, Veena, Wang, & Srivatsan, 2011). Previous research suggests that the health benefits of curcumin are associated with a number of biological activities, including anti-oxidative, anti-inflammatory, anti-microbial, anti-parasitic, anti-mutagenic, and anti-tumor activities (Ringman, Frautschy, Cole, Masterman, & Cummings, 2005; Sharma, Gescher, & Steward, 2005; Wilken et al., 2011). Another benefit of curcumin is that it has a low toxicity even when ingested at relatively high doses. Consumer demand for health-promoting foods containing fewer synthetic additives has made curcumin of particular interest as a natural nutraceutical ingredient (Anand, Kunnumakkara, Newman, & Aggarwal, 2007).

However, there are a number of physicochemical characteristics

* Corresponding author. Department of Food Science, University of Massachusetts Amherst, Amherst, MA 01003, USA.
E-mail address: mcclements@foodsci.umass.edu (D.J. McClements).

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of curcumin that currently limit its incorporation into many functional food products, such as its intense color, strong flavor, low water-solubility, and chemical instability (Letchford, Liggins, & Burt, 2008). Moreover, curcumin is rapidly metabolized within the gastrointestinal tract (GIT), which limits its potential beneficial biological effects (Anand et al., 2007; Sharma, Steward, & Gescher, 2007). Some of these limitations can be overcome by appropriate food product selection, *e.g.,* appearance and flavor problems can be overcome by incorporating curcumin into highly colored and spicy products. Other limitations can be overcome by encapsulating curcumin within food-grade colloidal delivery systems or by mixing it with colloidal excipient systems (Maiti, Mukherjee, Gantait, Saha, & Mukherjee, 2007; Margulis, Magdassi, Lee, & Macosko, 2014; Sahu, Kasoju, & Bora, 2008; Tapal & Tiku, 2012; Tiyaboonchai, Tungpradit, & Plianbangchang, 2007; Yu, Shi, Liu, & Huang, 2012; Zou, Liu, Liu, Xiao, & McClements, 2015a). A colloidal delivery system consists of small particles (typically comprised of lipids, phospholipids, surfactants, and/or biopolymers) that contain the curcumin (McClements, 2015b). On the other hand, a colloidal excipient system consists of small particles that are mixed with another food containing curcumin (Zou et al., 2015). These small particles are specifically designed to boost the bioavailability of curcumin. Colloidal delivery systems can be designed to facilitate the incorporation of curcumin into commercial food products, while both colloidal delivery and excipient systems can be designed to improve curcumin's chemical/biochemical stability and to control its fate within the GIT.

Previous studies have demonstrated the potential of improving the bioavailability of curcumin using colloidal delivery systems (Ahmed, Li, McClements, & Xiao, 2012; Anand et al., 2010; Patel & Velikov, 2011; Sun et al., 2012; Ting, Jiang, Ho, & Huang, 2014; Yu & Huang, 2012) or colloidal excipient systems (Zou, Liu, Liu, Xiao, & McClements, 2015b; Zou et al., 2015a; Zou et al., 2015). Emulsion-based systems are particularly useful for this purpose because their composition and structures can be designed to alter the bioaccessibility, absorption, and transformation of lipophilic bioactives (McClements & Xiao, 2014). In particular, the lipid phase in emulsions can be designed to rapidly digest within the small intestine and form mixed micelles capable of solubilizing and transporting lipophilic bioactives (Porter, Pouton, Cuine, & Charman, 2008; Yao, Xiao, & McClements, 2014). In addition, emulsions are already integral parts of many commercial food and beverage products (McClements, 2015a), and so emulsion-based delivery or excipient systems can easily be incorporated into a wide range of foods and beverages. One of the main limitations of conventional oil-in-water emulsions for this purpose is that they only have limited scope for controlling the stability and release of any encapsulated bioactive agents because the lipid droplets are only coated by a thin layer of emulsifier molecules. This limitation can be overcome by trapping the lipid droplets inside hydrogel beads ("microgels").

Hydrogel beads suitable for utilization in the food industry are usually fabricated from food-grade biopolymers, such as proteins and/or polysaccharides (Chen, Remondetto, & Subirade, 2006; Joye & McClements, 2014; Shewan & Stokes, 2013). These beads can be fabricated using numerous approaches, including injection, coacervation, thermodynamic incompatibility, antisolvent precipitation, templating, and molding methods (Matalanis, Jones, & McClements, 2011). The injection-gelation method is one of the simplest and most widely used approaches for the encapsulation, protection, and delivery of food-grade bioactive components such as nutrients, nutraceuticals, and vitamins. In this method, a biopolymer solution containing the bioactive component is injected into another "hardening" solution under conditions that promote the gelation of the injected biopolymer. This procedure results in the formation of a hydrogel bead with the bioactive components trapped inside. The nature of the hydrogel matrix surrounding the bioactive can be designed to improve its physical and chemical stability, as well as to control its GIT fate.

In the present study, curcumin was initially solubilized within the lipid phase of an oil-in-water nanoemulsion. The curcumin-loaded lipid droplets were then used as delivery systems themselves, or they were incorporated into hydrogel beads fabricated from either carrageenan or alginate. The filled hydrogel beads were formed by mixing the lipid droplets with a polysaccharide solution (alginate or κ-carrageenan), and then injecting the resulting mixture into a solution of positively charged ions (Ca^{2+} or K^{+}, respectively). The cations act as salt bridges to promote gelation of the anionic polysaccharides molecules. Carrageenan and alginate were used to form the hydrogel beads because they are already widely utilized as food-grade ingredients. Furthermore, these two polysaccharides were used because they are expected to form hydrogel beads with different functional attributes. Alginate is an anionic block copolymer that is composed of 1–4-linked β-D-mannuronic acid and α-L-guluronic acid units, with the negative charged originating from carboxyl groups (Lee & Mooney, 2012). Carrageenan is an anionic polymer that is compoared of galactan monomers linked together by alternating (1 → 3)- and (1 → 4)-β-d-glycosidic bonds with the negative charge originating from sulfate groups (Li, Ni, Shao, & Mao, 2014). We hypothesized that the hydrogel beads formed by these two different polysaccharides would have different structural and physicochemical properties, which would result in differences in the gastrointestinal fate of the encapsulated curcumin. The potential gastrointestinal fate of the different curcumin delivery systems was studied using a simulated GIT (mouth, stomach, and small intestine). In particular, the influence of the different hydrogel beads on the bioaccessibility of encapsulated curcumin was compared. The information obtained in this study may facilitate the rational design of hydrogel beads to modulate the bioavailability of encapsulated lipophilic nutraceuticals.

2. Materials and methods

2.1. Materials

Corn oil was purchased from a local supermarket and used without further purification. The following chemicals were purchased from the Sigma Chemical Company (St. Louis, MO): alginic acid (sodium salt) (Lot# 180947, viscosity of 1% alginic acid in water is 15–20 mPa s); curcumin (C1386-10G, purity ≥65%); mucin from porcine stomach (M2378-100G); pepsin from porcine gastric mucosa (P7000-25G); lipase from porcine pancreas pancreatin (L3126-100G); porcine bile extract (B8631-100G); Tween 80 (P1754-1L); and Nile Red (N3013-100MG). Carrageenan was kindly donated by FMC Biopolymer (Viscarin SD 389, Philadelphia, PA). All chemicals used were analytical grade. Double distilled water was used to make all solutions.

2.2. Preparation of oil-in-water emulsions

Initially, an aqueous phase was prepared by mixing 1% (w/w) Tween 80 with a buffer solution (5 mM phosphate buffer saline (PBS), pH 6.5) and stirring for at least 2 h. The emulsifier solution were then stored overnighted at 4 °C to ensure complete dissolution. Excipient emulsions stabilized by Tween 80 were prepared by homogenizing 10% (w/w) corn oil with 90% (w/w) aqueous emulsifier solution using a high-speed blender for 2 min (M133/1281-0, Biospec Products, Inc., ESGC, Switzerland). The droplet size in this coarse emulsion was reduced by passing it through a high-pressure