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Influence of methylcellulose on attributes of β-carotene fortified starch-based filled hydrogels: Optical, rheological, structural, digestibility, and bioaccessibility properties

Saehun Mun [a,b], Shinjae Park [a], Yong-Ro Kim [a], David Julian McClements [b,*]

[a] Center for Food and Bioconvergence, and Department of Biosystems and Biomaterials Science and Engineering, Seoul National University, Seoul 151-742, Republic of Korea
[b] Department of Food Science, University of Massachusetts, Amherst, MA 01003, United States

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ABSTRACT

There is considerable interest in controlling the gastrointestinal fate of nutraceuticals to improve their efficacy. In this study, the influence of methylcellulose (an indigestible polysaccharide) on lipid digestion and β-carotene bioaccessibility was determined. The carotenoids were encapsulated within lipid droplets that were then loaded into rice starch hydrogels containing different methylcellulose levels. Incorporation of 0 to 0.2% of methylcellulose had little impact on the dynamic shear rheology of the starch hydrogels, which may be important for formulating functional foods with desirable textural attributes. The microstructure, lipid digestion, and β-carotene bioaccessibility of the filled hydrogels were measured as the samples were passed through simulated oral, gastric, and small intestinal phases. The lipid digestion rate and carotenoid bioaccessibility decreased with increasing methylcellulose. This effect was attributed to the ability of the methylcellulose to inhibit molecular diffusion, promote droplet flocculation, or bind gastrointestinal components thereby inhibiting triacylglycerol hydrolysis at the lipid droplet surfaces. This information may be useful for rationally designing functional foods with improved nutritional benefits.

1. Introduction

The food industry is attempting to fortify many types of food and beverage products with nutraceuticals. Nutraceuticals are bioactive agents found in foods that are not required for normal human functioning (unlike nutrients, minerals, and vitamins), but that may improve human health, wellbeing, and performance through their biological effects (McClements, 2013). Nutraceuticals include food components such as ω-3 oils, carotenoids, polyphenols, phytosterols, and curcumin. Many of these nutraceuticals cannot simply be introduced into functional foods in their pure form because of physicochemical constraints, such as limited solubility, chemical instability, and poor bioavailability (Liang, Shoemaker, Yang, Zhong, & Huang, 2013; McClements, 2013; Reboul, 2013; Tang & Zhong-Gui, 2007). These challenges can often be overcome using food-grade colloidal delivery systems, such as emulsions, nanoemulsions, microemulsions, solid lipid nanoparticles, and filled hydrogels (Mun, Kim, Shin, & McClements, 2015; Nik, Langmaid, & Wright, 2012; Tokle, Lesmes, Decker, & McClements, 2012; Verrijssen et al., 2014; Zhang et al., 2015). Lipophilic nutraceuticals can be encapsulated within lipid droplets that can readily be dispersed in water, that can protect the bioactives from degradation, and that are digested in the gastrointestinal tract (GIT) to form mixed micelles that enhance bioaccessibility (McClements, 2013).

Many commonly consumed food products have gel-like properties, such as some desserts, confectionary, and meat-substitutes. These products are often prepared using food-grade biopolymers that form a three-dimensional network that traps water and leads to a material with viscoelastic properties. In a previous study, our group showed that β-carotene could be encapsulated in lipid droplets and then dispersed in macroscopic starch hydrogels (Mun, Kim, & McClements, 2015; Mun, Kim, Shin, & McClements, 2015). This research showed that β-carotene bioaccessibility was higher in filled hydrogels (lipid droplets dispersed in hydrogels), than in lipid droplets or in hydrogels alone. This effect was attributed to the ability of the starch hydrogels to prevent the lipid droplets from aggregating in the gastrointestinal tract, which increased the accessibility of the lipase to the emulsified lipid phase.

In the current study, the influence of the addition of methylcellulose on the microstructure, lipid digestibility, and β-carotene bioaccessibility of the filled starch hydrogels was examined using a simulated GIT. Methylcellulose is an indigestible water-soluble polysaccharide that is widely used in the food industry for its physicochemical and

* Corresponding author.
E-mail address: mcclements@foodsci.umass.edu (D.J. McClements).

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physiological properties (Chawla & Patil, 2010). Methylcellulose is commonly used as a thickener and emulsifier in foods, as well as to prevent constipation in supplements (Li & Nie, 2016). Previous studies have shown that the microstructure of starch hydrogels can be appreciably altered by the addition of indigestible polysaccharides such as guar gum, xanthan gum, and cellulose derivatives (Gladkowska-Balewicz, Norton, & Hamilton, 2014; Ptaszek et al., 2009; Techawipharat, Suphantharika, & BeMiller, 2008). Other studies have shown that interactions between starch and indigestible polysaccharides may appreciably alter the functional properties of starch hydrogels, such as their appearance, rheology, and stability (Baranowska, Sikora, Krystyjan, & Tomasik, 2011; Krystyjan, Adamczyk, Sikora, & Tomasik, 2013; Samutsri & Suphantharika, 2012; Sikoral, Tomasik, & Krystyjan, 2010). We therefore hypothesized that the addition of methylcellulose to filled starch hydrogels would alter their microstructure, rheology, and gastrointestinal fate, which could alter the bioaccessibility of any encapsulated nutraceuticals. This information may be useful in the design of functional foods with enhanced health benefits, such as controlled lipid digestion or nutraceutical bioaccessibility.

2. Materials and methods

2.1. Materials

The rice starch used for this study was isolated from native rice (Ilmi byeo, Korea) in a laboratory using a traditional alkaline method (Lumdubwong & Seib, 2000). β-Carotene, pancreatin (from porcine pancreas) and bile extract (porcine) were purchased from Sigma Aldrich (St. Louis, MO). Whey protein isolate (WPI) was kindly provided by Davisco Foods International Inc. (BiPRO, Le Sueur, MN, USA). Corn oil was purchased from a local supermarket. All other chemicals were of analytical grade. Double-distilled water was used to prepare all solution and emulsions. Most concentrations are reported as wt.%, i.e., g of specified component per 100 g of sample.

2.2. Viscoelastic behavior

The dynamic viscoelastic properties of the samples were determined using a dynamic shear rheometer (AR 1500 ex, TA instruments Ltd., New Castle, DE, USA) operating in oscillatory mode with parallel plate geometry (20 mm diameter, 1 mm gap). Starch pastes were prepared by dispersing rice starch powder in distilled water (10 wt.% starch) and then heating at 90 °C for 10 min in the absence and presence of emulsion and methyl cellulose. After thermal treatment, the resulting hot paste was loaded between the parallel plates of the rheometer that had previously been equilibrated at 4 °C and a thin layer of paraffin oil was applied to the outer edges of the sample to prevent evaporation during measurement. A dynamic frequency sweep test was conducted by applying a constant strain of 0.5%, which was within the linear viscoelastic region (established in a preliminary test), over a frequency range between 0.63 and 63 rad/s. The storage (G′) and loss (G″) moduli of selected samples were then reported over a range of frequencies, and the moduli of all samples were reported at a constant frequency of 10 rad/s.

2.3. Emulsion preparation

An oil phase was prepared by dispersing β-carotene (0.3%, w/w) in corn oil using a sonicating water bath (Model 250, Ultrasonic cleaner, E/MC RAI research, Long Island, New York) for 5 min and then heating (60 °C for 30 min) to ensure complete dissolution. An aqueous emulsifier solution was prepared by dispersing WPI (1.06 wt.%) in 10 mM phosphate buffer (pH 7.0). A stock emulsion was prepared by homogenizing 6 wt.% corn oil and 94 wt.% emulsifier solution (1.06 wt.% WPI, pH 7.0) using a high speed blender for 2 min (M133/1281-0, Biospec Products, Inc., ESGC, Switzerland) and then passing through a microfludizer four times at 10 kpsi (Model 110L, Microfluidics, Newton, MA).

2.4. Preparation of filled hydrogels containing methylcellulose

Initially, the desired amount of methylcellulose was dissolved in 10 mM phosphate buffer (pH 7.0) and then stock WPI-stabilized emulsion and rice starch were added into the methylcellulose solution. The final concentrations of lipid, rice starch, and methylcellulose contained in the mixtures were 4 wt.% lipid, 10 wt.% of rice starch, and 0, 0.05 wt.%, 0.1 wt.%, or 0.2 wt.% of methylcellulose. These levels of methylcellulose were selected so as to cover a range of values that might be used in commercial products to modify their properties (Dar & Light, 2014). These mixtures were then heated at 90 °C for 10 min to gelatinize the starch granules. The heated mixtures were loaded into a flat, cylindrically shaped vessel, and then stored at 4 °C overnight to allow the hydrogels to set.

2.5. Simulated gastrointestinal tract model

Samples were passed through an in vitro GIT model consisting of mouth, stomach, and small intestine phases, which has been described in detail in previous studies (Lopez-Pena et al., 2016; Mun, Kim, & McClements, 2015; Salvia-Trujillo, Qian, Martin-Belloso, & McClements, 2013a, 2013b). Hence, the method is only briefly described here. This method has been widely used by our laboratory in previous studies, and is fairly similar to the standardized method proposed recently (Minekus et al., 2014).

For the mouth phase, the samples were mixed with simulated saliva fluid (SSF) containing mucin and various salts (Sarkar, Goh, & Singh, 2009) at a 50:50 vol/vol ratio. The pH of the mixture was then adjusted to pH 6.8 and then incubated at 37 °C for 10 min with continuous agitation. This incubation period is longer than the time a food would typically spend in the mouth, but was used to ensure consistency from sample to sample. For the stomach phase, the sample was mixed with simulated gastric fluid (SGF), which was prepared by dissolving 2 g NaCl, and 7 mL of HCl (37%) in 1 L of water and then adding 3.2 g of pepsin (Sarkar, Goh, Singh, & Singh, 2009). The sample was then adjusted to pH 2.5 and incubated at 37 °C for 2 h with continuous agitation at 100 rpm. For the small intestine phase, 30 mL of samples from the gastric phase was placed in a temperature-controlled (37 °C) chamber and the system was set at pH 7.0. Then 3.5 mL of bile extract solution (187.5 mg/3.5 mL) and 1.5 mL of salt solution (10 mM of calcium chloride and 150 mM of sodium chloride) were added to the samples and the mixture was adjusted to pH 7.0. Afterwards, 2.5 mL of freshly prepared pancreatin suspension (187.5 mg/2.5 mL) dissolved in phosphate buffer was added into the mixture and incubated at 37 °C for 2 h. The pH of the sample was monitored using a pH-stat automatic titration unit (Metrohm USA Inc., Riverview, FL) and the solution was maintained at pH 7.0 by adding 0.25 M NaOH to neutralize any free fatty acids (FFA) released due to lipid digestion. The percentage of FFAs released was calculated from the volume of alkaline solution required to neutralize the samples using the following equation:

$$\%\mathrm{FFA} = 100 \times \frac{V_{NaOH} \times m_{NaOH} \times M_{lipid}}{W_{lipid} \times 2}$$

where V_{NaOH} is the volume of titrant in liters, m_{NaOH} is the molarity of sodium hydroxide, M_{lipid} is the molecular weight of corn oil (872 g/mol), and w_{lipid} is the weight of oil in the digestion system in grams. Blanks (samples without oil) were run, and the volume of titrant used for these blank samples was subtracted from the corresponding test samples that contained oil.