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Effect of the degree of substitution of octenyl succinic anhydride-banana starch on emulsion stability

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ABSTRACT

Banana starch was esterified with octenylsuccinic anhydride (OSA) at different degree substitution (DS) and used to stabilize emulsions. Morphology, emulsion stability, emulsification index, rheological properties and particle size distribution of the emulsions were tested. Emulsions dyed with Solvent Red 26 showed affinity for the oil phase. Backscattering light showed three regions in the emulsion where the emulsified region was present. Starch concentration had higher effect in the emulsification index (EI) than the DS used in the study because similar values were found with OSA-banana and native starches. However, OSA-banana presented greater stability of the emulsified region. Rheological tests in emulsions with OSA-banana showed $G' > G''$ values and low dependence of G' with the frequency, indicating a dominant elastic response to shear. When emulsions were prepared under high-pressure conditions, the emulsions with OSA-banana starch with different DS showed a bimodal distribution of particle size. The emulsion with OSA-banana starch and the low DS showed similar mean droplet diameter than its native counterpart. In contrast, the highest DS led to the highest mean droplet diameter. It is concluded that OSA-banana starch with DS can be used to stabilize specific emulsion types.

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1. Introduction

Emulsions are heterogeneous systems consisting of droplets of a liquid dispersed in another non-miscible or partly miscible liquid (IUPAC, 2001). Stability is essential for emulsions to maintain their properties as long as possible. Emulsions can be stabilized not only by surfactants but also by solid particles, such as laponite (Whitby & Corbi, 2014), native starches (Chen, Yunxing, Peidong, & Cheng, 2013) and modified starches (Yussoff & Murray, 2011). The mechanism whereby solid particles protect emulsion droplets against coalescence by interfacial action is known as Pickering stabilization. The effectiveness of the mechanism relies on the fact that once a particle has become attached to the oil–water interface, it can be regarded as being irreversibly adsorbed (Tan, Xu, Liu Ch Li, Lu, & Wang, 2012). Depending on their surface hydrophobicity, particles can stabilize oil in water, water in oil or multiple emulsions.

Esterification of starch with octenyl succinic anhydride (OSA) produces an amphiphilic biopolymer that can be used to stabilize emulsions in foods, cosmetics and pharmaceutical products (Sweedman, Tizzotti, Schäfer, & Gilbert, 2013). Different starches, including quinoa, rice, maize as well as waxy and high-amylose starches have been tested in their native and OSA counterpart to stabilize emulsions (Timgren, Rayner, Dejmek, Marku, & Sjöö, 2013). The results of these tests suggested that the chemical modification, shape and size of the starch granules have a large impact on the stabilization of Pickering-type emulsions; small starch granules (quinoa) showed the best emulsifying properties. The use of OSA-starch as an emulsifier involves homogenization that affects its properties. The turbulent flow during homogenization produces degradation of OSA-starch and changes in the molecular shape and conformation of starch components in the granular structure, which could influence its emulsifying properties. However, these changes are related to the degree modification of OSA-starch, and they can be determined by its molar mass distribution. However, this information is rarely reported in the scientific literature (Dokić, Krstonošić, & Nikolić, 2012). Recently, Bello-Flores, Nuñez-Santiago, San Martín-González, BeMiller, and Bello-Pérez (2014)

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studied the effect of the degree of substitution (DS) of OSA-banana starch on the structural and molecular features. They concluded that the insertion of OSA groups in the starch induced disorder in the granular structure, specifically in the molecular size of amylopectin. An increase in the OSA concentration produced a decrease in molecular characteristics (e.g., molecular weight and radius of gyration).

The aim of the present study was to evaluate the effect of the DS of OSA-banana starch on the stability of emulsions for a better understanding of the stabilization mechanisms of OSA-starch in Pickering-type emulsions.

2. Materials and methods

2.1. Materials

Unripe plantain (*Musa paradisiacal* L.) was purchased at a local market in Cuautla, Morelos State, México. Starch was isolated according to the procedure of Flores-Gorosquera et al. (2004). OSA-banana starch was prepared according to the method reported by Bello-Flores et al. (2014). 2-Octen-1-ylsuccinic anhydride (OSA, purity of 97%) was purchased from Sigma-Aldrich Co. (St. Louis, MO, USA). The continuous phase was a 5 mmol/L phosphate buffer with pH 7 and 0.2 mol/L NaCl (Timgren et al., 2013). The disperse phase was liquid paraffin.

2.2. Preparation of starch granule-stabilized emulsions

Emulsions were prepared using the method of Timgren et al. (2013) with some modifications. The continuous phase of the emulsions was made of 5 mmol/L phosphate butter at pH 7 and 0.2 mol/L NaCl, and the dispersed phase was liquid paraffin. Four milliliters of continuous phase, 2 mL of dispersed phase and starch at varying amounts (0–250 mg/mL) were emulsified in glass test tubes by high shear mixer in a SilentChruser M mixer (Heidolph, Germany) at 22,000 rpm for 60 s.

Approximately 1 mg of the oil-soluble dye Solvent Red 26 was added to the top of the emulsions, and the test tubes were gently turned three times after 1 h. The emulsions were shaken with a vortex mixer for 5 s to distribute the color in the entire sample. The color change in the emulsion was observed for 48 h. The color of the emulsion is a measure of the orthokinetic stability of the formed drops. Stable drops do not have an exchange with the lipophilic dye; hence, the emulsion phase remains white.

2.3. Light microscopy of starch granule-stabilized emulsions

A polarized light microscope (Eclipse 80i, Nikon, Japan) was employed with a 4x objective lense and equipped with a digital camera (Digital imaging Head, DC330 camera MTI, Japan). Individual emulsions were placed on a microscopic slide, and the drops were studied immediately after their preparation at room temperature.

2.4. Emulsion stability

Emulsion stability was measured by a technique based on multiple light scattering. The detection head is composed of a pulsed near-infrared light source (Λ = 850 nm) and two synchronous detectors. The optical characterization of the samples was evaluated using a Turbiscan Lab Expert (Formulation, Tolouse, France) (Mengual, Meunier, Cayre, Puech, & Snabre, 1999). An aliquot of recently prepared emulsion (6 mL) was poured in a glass test tube. The detection head scanned the entire length of the sample (approximately 60 mm), acquiring transmission and backscattering data every 40 μm, so the transmitted and backscattered light was

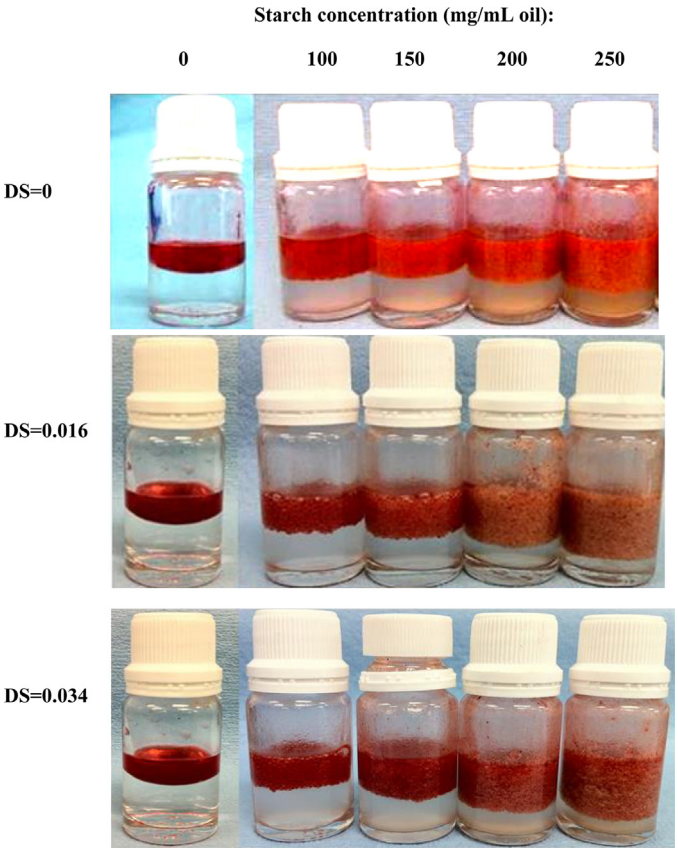

Fig. 1. Effect of the starch concentration and degree of substitution (DS) on the emulsification with octenyl succinic anhydride (OSA)-modified banana starch and continuous phase with 0.2 mol/L NaCl. Test sample images of emulsion are immediately after vortexing.

presented as a function of the sample height. A reference mode function was used, which subtracts the first curve (T_0) from the subsequent ones (Panaras, Moatsu, Yanniotis, & Mandala, 2011). Samples were monitored from 0 to 48 h. All studies were made by triplicate.

2.5. Emulsification index (EI)

The emulsification index of the samples was expressed as the volume of the emulsion to the total volume of the sample (Timgren et al., 2013). In this work, the transversal area of the glass test tubes was considered constant along the vials, so the EI was calculated as follows:

$$\text{EI} = \frac{\text{height of cream layer}}{\text{total height of emulsion}}$$

2.6. Rheological properties

To determine the rheological properties of the emulsion, a controlled-stress rheometer (AR-1500ex, TA Instruments, USA) was employed using a cone-plate geometry (40 mm in diameter, 80 mm in gap, and 2° in angle) and previously stabilized at 25 °C. The variation of G' and G'' with frequency (0.1 to 100 rad/s) was determined in the zone of linear viscoelasticity previously found from the strain sweep. All measurements were made by triplicate.

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