



Synthesis and characterization of retrograded starch nanoparticles through homogenization and miniemulsion cross-linking



Yongbo Ding^{a,b}, Jiong Zheng^{a,b}, Fusheng Zhang^{a,b}, Jianquan Kan^{a,b,*}

^a College of Food Science, Southwest University, Chongqing 400715, China

^b Laboratory of Quality & Safety Risk Assessment for Agro-products on Storage and Preservation Chongqing, Ministry of Agriculture, 400715, China

ARTICLE INFO

Article history:

Received 27 January 2016

Received in revised form 28 May 2016

Accepted 1 June 2016

Available online 2 June 2016

Keywords:

Box–Behnken design

Homogenization

RS3NPs

Physicochemical properties

ABSTRACT

A new and convenient route to synthesizing retrograded starch nanoparticles (RS3NPs) through homogenization combined with a water-in-oil miniemulsion cross-linking technique was developed. The RS3NPs were optimized using Box–Behnken experimental design. Homogenization pressure (X_1), oil/water ratio (X_2), and surfactant (X_3) were selected as independent variables, whereas particle size was considered as a dependent variable. Results indicated that homogenization pressure was the main contributing variable for particle size. The optimum values for homogenization pressure, oil/water ratio, and surfactant were 30 MPa, 9.34:1, and 2.54 g, respectively, whereas the particle size was predicted to be 288.2 nm. Morphological, physical, chemical, and functional properties of the RS3NPs were assessed. Scanning electron microscopy and dynamic light scattering images showed that RS3NP granules were broken down to size of about 222.2 nm. X-ray diffraction results revealed a disruption in crystallinity. The RS3NPs exhibited a slight decrease in T_o , but T_p and T_c increased and narrowest T_c – T_o . The solubility and swelling power were also increased. New peaks at 1594.84 and 1403.65 cm^{-1} were observed in the FTIR graph. However, homogenization minimally influenced the antidigestibility of RS3NPs. The absorption properties improved, and the adsorption kinetic described the contact time on the adsorption of captopril onto RS3NPs. In vitro release experiment indicated that the drug was released as follows: 21% after 2 h in SGF, 42.78% at the end of 8 h (2 h in SGF and 6 h in SIF), and 92.55% after 12 h in SCF. These findings may help better utilize RS3NP in biomedical applications as a drug delivery material.

© 2016 Elsevier Ltd. All rights reserved.

1. Introduction

Modern drug-carrier systems play an important role in the controlled delivery of pharmacological agents to its target at a therapeutically optimal rate and dose. Various nanoparticles (NPs) of colloidal drug delivery systems represent a very promising approach to this aim (Maincent et al., 1992; Kreuter, 1994; Nakada, Fattal, Foulquier, & Couvreur, 1996). Their protein protection and controlled release behavior by the polymeric matrix seem to be useful for drug delivery system. NPs are especially interesting because of their particle size that allows transport of carriers across biological membranes and specific targeting through a possible surface modification (Bourel, Rolland, Le Verge, & Genetet, 1988; Alyautdin et al., 1997).

Many authors (Blanco and Alonso, 1997; Iwata and McGinity, 1991) have previously shown that the double emulsion technique was the most appropriate method to produce microparticles. However, most of the particles prepared using the water-in-oil miniemulsion technique had a relatively large diameter of up to 1 μm . Many physical emulsification techniques such as high shear emulsification, sonicators and high pressure homogenization can be used to prepare mini-emulsions. As we reported in our previous study, we have prepared resistant starch type IV nanoparticles through ultrasonication and miniemulsion cross-linking (Ding, Zheng, Xia, Ren, & Kan, 2016a; Zheng, Xia, Ren, & Kan, 2016b). The high pressure homogenization has been developed recently as one of the most efficient emulsification techniques. Compared to the ultrasonication and high shear emulsification, the high pressure homogenization can produce smaller droplets and better homogeneity in emulsions. The productivity of the high pressure homogenization method is the best among the above mentioned emulsification methods (Asua, 2002). When the high pressure homogenization and mini-emulsion cross-linking reac-

* Corresponding author at: College of Food Science, Southwest University, Tiansheng Road 1, Chongqing, 400715, PR China.

E-mail address: kanjianquan@163.com (J. Kan).

Table 1
Variables used in the Box–Behnken experimental design.

Variable, unit	Factors	Level		
		x	Low (−1)	Middle (0)
Homogenization pressure, MPa	X ₁	30	40	50
Oil/water	X ₂	8:1	10:1	12:1
Surfactant, g	X ₃	1	2	3

tion are combined, starch submicron particles can be obtained in the emulsion droplets.

Over the last two decades, research works have been emphasizing on the health-promoting properties of retrograded starch and classified one form of retrograded starch (RS3) (Englyst, Kingman, & Cummings, 1992). RS3 escapes digestion in the gastrointestinal tract of a healthy man and has a health-promoting function (Wang and Copeland, 2013). Long amylopectin chains exhibit a substantial tendency to form insoluble, semicrystalline structures with high thermal stability and resistance to amylase activity (Englyst et al., 1992).

This study aimed to: first, prepare RS3NPs with lower size polydispersity using a pressure-homogenization device utilizing the water-in-oil miniemulsion cross-linking technique; second, to optimize RS3 NP formation by varying the homogenization pressure, oil/water ratio, and surfactant concentration; and third, to investigate the effect of homogenization on the physicochemical properties, in vitro starch digestibility, adsorption behaviors, in vitro release of RS3NPs.

2. Materials and methods

2.1. Materials

RS3 was obtained from high-amylose corn starch (National Starch, USA). The pepsin and pancreatic elastase that underwent in vitro starch digestibility test were purchased from Sigma (USA). Potassium hydroxide, potassium persulfate, sodium bisulfite, *N,N*-methylene diacrylamide (MBAA), cyclohexane, acetic acid, Tween-80, and Span-80 were provided by Chongqing Chemical Company (Chongqing, China). All chemicals and solvents were certified to be of analytical reagent grade.

2.2. Optimizing RS3 nanoparticle synthesis using the Box–Behnken design

Three factors (homogenization pressure (X₁), the oil/water ratio (X₂), and surfactant concentration (X₃)) for the measured response (particle size (Y)) were established for the three-level Box–Behnken design. The factor levels were coded (−1, 0, and 1). The Box–Behnken design generated 17 experiments, and the code level ranges of levels of the independent variables are shown in Table 1. The design matrix in the results coded and predicted by the model is illustrated in Table 2. Variance analysis (Table 3) was used to determine the importance of each factor in the model and select the appropriate model for optimization.

2.3. RS3 nanoparticle synthesis

NPs were prepared using the w/o miniemulsion technique previously applied to prepare microparticles and NPs (Chang, Jian, Yu, & Ma, 2010; Chaudhuri & Paria, 2010; Ding et al., 2016a, 2016b; Wang, Smetana, Boeckl, Brown, & Wai, 2009). NPs were created in w/o miniemulsion system using emulsion cross-linking technique with MBAA as across-linking agent. K₂S₂O₈ and NaHSO₃ were used as inducing agent (Shi, Li, Wang, Li, & Adhikari, 2011). The adjustment was based on the use of a homogenizer in the emulsifi-

cation process, thus considerably reducing the size of the dispersed droplets. The water phase was briefly formed by dissolving 0.15 g of RS3 in 10.0 mL of 2 mol/L potassium hydroxide solution and stirring for 2 h until the starch was fully dissolved. Subsequently, about 0.05, 0.027, and 0.104 g of MBAA, K₂S₂O₈, and NaHSO₃ were added into the solution, respectively. This w/o emulsion was thereafter poured into cyclohexane containing Tween-80 and Span-80 (according to Table 2) to produce w/o emulsion. The emulsion containing submicron drops was then prepared using a high-pressure homogenizer (ATS® AH-100D, BVI, Canada) (according to Table 2). The RS3NPs were finally obtained after 12 h of crosslinking and solidification.

After washing with acetone twice, the starch submicron particles from 40 mL of miniemulsion was dispersed in 100 mL of DI water. The suspension containing starch submicron particles was stirred for 1 h to make sure all starch submicron particles completely dispersed and then left for spray drying.

2.4. Physicochemical properties of RS3 nanoparticle

2.4.1. Particle-size distribution

The particle diameters were measured using the Malvern Nano-Zetasizer ZS Instrument (Malvern, England) at 25 °C. The dispersions were diluted with water to 0.1% and transferred into a plastic disposable cuvette for measurement. Dynamic light scattering measurements provided the Z-average size.

2.4.2. Scanning electron microscopy (SEM)

The morphological characteristics of the dried samples were evaluated using SEM (ESEM, Japan) at an accelerating voltage of 20 kV. Dried and finely ground samples were sprinkled on a double-sided sticky tape placed on aluminum stubs and covered with thin gold film.

2.4.3. X-ray diffraction (XRD)

The XRD patterns of the dried samples were studied using an X-ray diffractometer (PERSEE, Beijing) according to the procedure described by Zobel (1964). The graphs were plotted at 2 theta within 10°–60°.

2.4.4. Thermal properties (DSC)

The thermal properties of the dried samples were measured and recorded using a differential scanning calorimeter (DSC, NET-ZSCH, Germany). The instrument was calibrated with indium, and an empty pan was used as the reference. The dried samples (10 mg) were sealed in aluminum pans and heated from 20 °C to 200 °C at 10 °C/min under nitrogen atmosphere (flow rate of 30 mL/min).

2.4.5. Fourier transform infrared (FTIR) spectroscopy

The FTIR spectra of samples were recorded using an FTIR spectrometer (NEXUS-870, Thermo Nicolet Corporation). About 2 mg-dried samples were mixed with 150 mg of KBr, pressed into pellets, and grounded. The FTIR spectra were obtained within the wave number range of 400–4000 cm^{−1}.

2.4.6. Solubility and swelling power (SP)

The solubility and SP of the samples were analyzed using the method described by Reddy, Haripriya, Mohamed, and Suriya (2014) and Liu et al. (2015). A sample (0.2 g) was dispersed in water (20 mL) to form a suspension, which was heated to 85 °C in a water bath for 30 min with vigorous shaking every 5 min. The starch gel was then centrifuged at 2200 rpm for 15 min. The weight of the sediment was used to calculate the SP. The supernatant was dried and weighed to measure the amount of dissolved starch in the super-

Download English Version:

<https://daneshyari.com/en/article/1383040>

Download Persian Version:

<https://daneshyari.com/article/1383040>

[Daneshyari.com](https://daneshyari.com)