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Original paper

Microencapsulation using Spray-drying: The Use of Fine Starch Solution for the Wall Material

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A fine waxy rice starch solution was spray-dried as a wall material for microencapsulation. For this process, the primary solution of mixed core and wall materials needs a low viscosity. Low molecular weight polysaccharides have been commonly used as the wall material. In the present study, a starch solution prepared hydrothermal process at 100 °C and fine starch solutions prepared by compressed hot water process at 140, 160, and 180 °C were investigated. The compressed hot water process reduced the viscosity of the fine starch solution and its particle size to produce a uniformly fine spray-dried powder. Finally, inulin hydrolysate including difructose anhydrides (DFAs) were microencapsulated by spray-drying. Previously, preparing inulin hydrolysate powder through spray-drying and freeze-drying has been difficult because it adheres to the inside surface of the drying chamber. Microencapsulation of inulin hydrolysate using a 160 °C fine starch solution as the wall material produced powder efficiently and enhanced its handling properties.

Keywords: viscosity, compressed hot water, particle size, crystallinity index, difructose anhydrides (DFAs)

Introduction

To contribute towards environmental conservation and a sustainable society, research into fundamental techniques is needed to widen the range of application of biomass. Starch, the second most abundant of these biomass materials, is stored in many plants as a source of energy from photosynthesis. The food industry commonly extracts starch from corn, wheat, and tapioca, in its native or modified forms, for use in various fields such as food, paper, and pharmaceuticals (Whistler and Paschall, 1965; Buléon *et al.*, 1998; Ono *et al.*, 2005).

To use biomass effectively, techniques for preparing nanoscale biomass have been studied, with nanoscale waxy rice starch particles being prepared using the compressed hot water process through hydrolysis. The smallest average particle size of 150.4 nm has been obtained using compressed hot water process at 180 °C, with a starch concentration of 0.1 % (w/w), and an initial pressure of 3.0 MPa (Yoshioka and Shimizu, 2014). Starch nanoparticles with diameter 20–50 nm was

produced from starch-butanol complex using α -amylase, however, most of starch was hydrolyzed and recovery was low (Kim and Lim, 2009). Starch nanoparticles prepared by acid hydrolysis is also low recovery because of the same reason and removing the salts produced by neutralizing and it needs long reaction time (Kim *et al.*, 2015). The compressed hot water process can be conducted without acid, enzyme or catalyst in a short time, therefore it can be applied as one of the useful biomass conversion reaction field. The conversion of inulin solution to inulin hydrolysate solution including difructose anhydrides (DFAs) using the compressed hot water process has also been investigated. DFAs are cyclic disaccharides consisting of two fructose units, with α -D-fructofuranose- β -D-fructofuranose 2',1:2,3'-dianhydride (DFAIII) being a functional disaccharide, which increases calcium absorption in the internal organs (Saito and Tomita, 2000). DFAs are prepared optimally from an inulin hydrolysate solution using the compressed hot water process at 160–165 °C, at an inulin

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concentration of 1.0 % (w/v), and an initial pressure of 3.0 MPa (Itoh and Shimizu, 2014). These studies have contributed towards studying the quantitative and qualitative use of biomass.

Using a fine starch solution as the wall material for microencapsulation by spray-drying has also been considered. Microencapsulation is the process where small particles, droplets, and substances are surrounded or entrapped by a relatively stable material to obtain small capsules. The interior material is the “core material”, and surrounding it is the “wall material”. This method of processing leads to many useful properties: reducing the reactivity of the core material to oxygen, light, and water; controlling the release rate of the core material to its surroundings; promoting easier handling of the core materials; masking the taste of the core material; and diluting the core material when it only needs to be used in small amounts (Shahidi and Han, 1993; Gharsallaoui *et al.*, 2007; Poshadri and Kuna, 2010, Badee *et al.*, 2012). The microencapsulation process has been widely studied in various fields with many techniques being established: spray-drying, spray-cooling, extrusion, fluidized-bed coating, cocrystallization, and coacervation. Spray-drying is the most common method used in food industry, because of its efficiency, low cost, flexibility and use of easily available units (Sébastien, 2004; Gharsallaoui *et al.*, 2007).

For microencapsulation using spray-drying, the primary solution consists of a mixture of the core and wall material. Therefore, these materials must be soluble in water or be prepared as dispersions or emulsions. In some cases, a primary solution with a low viscosity is desirable to prevent clogging of the spray-drying nozzle and adhesion, and to provide uniform spraying. When polysaccharides have been used as the wall material, those with a low molecular weight such as maltodextrin, dextran, and corn syrup solids, have been commonly used (Gharsallaoui *et al.*, 2007, Poshadri and Kuna, 2010). Waxy rice starch, which consists mostly of amylopectin, has a high molecular weight so the viscosity of its solution is also very high and therefore unsuitable as a wall material for microencapsulation by spray-drying.

In the present study, the microencapsulation of inulin hydrolysate including DFAs using spray-drying will be investigated using a fine starch solution as the wall material. Microencapsulation using dextrin hydrate as the wall material will be carried out as the control sample. The changes in viscosity of the fine starch solution will also be measured, as well as analyzing the changes in powder morphology through the compressed hot water and spray-drying processes.

Materials and Methods

Chemicals Waxy rice starch was obtained from Joetsu Starch Co. Ltd. (Niigata, Japan), inulin extracted from chicory (Orafti GR: average degree of polymerization, over 10) from DKSH Japan Co. Ltd (Tokyo, Japan), and dextrin hydrate from

Wako Pure Chemical Industries Ltd. (Osaka, Japan). Diffructose anhydride III from Wako Pure Chemical Industries Ltd was used as the standard sample in the mass spectrometry. Distilled water was used throughout the entire experiment.

Preparation of fine starch solution and inulin hydrolysate solution The starch solution was prepared by the hydrothermal process at 100 °C (100 °C starch solution), the fine starch solution by the compressed hot water process at 140, 160, and 180 °C (140 °C, 160 °C, and 180 °C fine starch solutions, respectively), and the inulin hydrolysate solution including DFAs by the compressed hot water process at 165 °C (inulin hydrolysate solution).

A glass reaction vessel was used for the hydrothermal process and a pressure vessel (stainless-steel vessel: $\phi 48$ mm, Teflon resin inner vessel: $\phi 36$ mm $\times$ 120 mm tall) for the compressed hot water process. For the hydrothermal process, starch (4.5 g) was placed in the glass reaction vessel with distilled water (150 mL) and a stirring chip, heated until the solution temperature was 100 °C using an organic synthesizer (Chemi-Station PPV-3000, Tokyo Rikakikai Co. Ltd, Tokyo Japan), then the vessel was cooled in a refrigerator at 5 °C. For the compressed hot water process, starch (1.5 g) was placed in the pressure vessel with distilled water (50 mL) and a stirring chip, heated until the solution temperature was 140 °C, 160 °C, or 180 °C using the same organic synthesizer, then the vessel was also cooled in a refrigerator at 5 °C. All solutions were then stored at 5 °C. The inulin hydrolysate solution was prepared by the compressed hot water process at 165 °C from the inulin solution then checked by electrospray ionization mass spectrometry (ESI-MS). The pressure vessel was filled with nitrogen gas at a primary pressure of 3 MPa and the stirring chip speed was set at 1000 rpm (Table 1). The solution temperature in the vessel during heating was recorded using a type K thermocouple connected to a data logger (Multichannel Temperature Recorder MCR-4TC, T&D Co., Nagano, Japan)

The primary solution supplied to the spray-dryer was the mixed inulin hydrolysate solution (core material) and the 160 °C fine starch solution (wall material) in a core: wall ratio of 1: 4. The control sample was prepared using the mixed inulin hydrolysate solution and dextrin hydrate solution as the wall material, in the same ratio. The concentration of all solutions used in these experiments was 3 % (w/w).

Electrospray ionization mass spectrometry (ESI-MS) Mass spectrometry was used to confirm the presence of DFAs in inulin hydrolysate solution (Itoh and Shimizu, 2014). The DFAIII solution and inulin hydrolysate solution prepared by the compressed hot water process at 165 °C were analyzed using ESI-MS (Exactive, Thermo Fisher Scientific K.K., Kanagawa, Japan) in the mass range m/z 150–2000 (m/z : mass-to-charge ratio). Each solution was diluted with methanol.

Spray-drying The solutions (100 mL) were spray-dried in a laboratory-scale dryer using two-fluid nozzle. The drying conditions were: inlet temperature 200 °C, and exhaust

Table 1. Preparation of fine starch solution and DFAs solution.

Sample	Concentration %(w/w)	Pressure in vessel MPa	Stirring chip speed rpm	Temperature °C	Temperature at heater °C
100 °C starch solution		0.1(Atmospheric pressure)		100	140
140 °C fine starch solution	3		1000	140	180
160 °C fine starch solution		3		160	200
180 °C fine starch solution				180	200
DFAs solution	3	3	1000	165	200

temperature approximately 130 °C with a flow rate of approximately 3.5–3.75 mL/min. The solutions were atomized using air compressor (0.4LE-8S, Hitachi Ltd., Tokyo, Japan) at a pressure of over 0.25 MPa. The nozzle diameter in the atomizer was 0.5 mm, but because of clogging occurring when the 100 °C starch solution and the 140 °C fine starch solution were spray-dried, the ϕ 1 mm nozzle was used instead. Spray-drying was carried out twice then the powders were stored in zippered plastic bags, respectively.

Measurement of viscosity The viscosity of all the solutions used for microencapsulation by spray-drying were measured using a rotating cylinder viscometer (Model LV-2000, Spindle L1: ϕ 19 mm $\times$ 65 mm tall, Cannon Instrument Co., State College, PA, USA). More than 400 mL of the solutions were prepared to completely soak the spindles. During measurement, the solution temperature was maintained at 25 °C using a thermostat. The spindle speeds available were 0.3, 0.6, 1.5, 3, 6, 12, 30, and 60 rpm which in terms of the moving velocity of the side cylinder surface corresponded to approximately 12, 23, 58, 116, 233, 466, 1164, and 2328 cm²/min, respectively. The measurement was performed in triplicate.

Determination of crystallinity using X-ray diffractograms Diffractograms of the waxy rice starch, and the spray-dried powders from the 100 °C starch solution and the 140 °C, 160 °C, and 180 °C fine starch solutions were recorded using a powder X-ray diffractometer (X-ray generating apparatus: RINT 2000/PC, Horizontal Goniometer: Ultima+, Rigaku Co. Ltd, Tokyo, Japan) using Ni-filtered Cu-K α radiation (λ = 0.1541 nm) generated at 40 kV/30 mA. The powder samples were mounted on glass holders and flattened. The scan range was 3–40 degree (2 θ) with a scan speed of 2 degree/min in 0.02 degree steps. The divergence slit and scattering slit were 1 degree, and the receiving slit 0.3 mm. The crystallinity index, X_c , was calculated from the recorded diffractogram and was defined as the percentage of crystalline structure in the whole sample using the following formula (Hulleman *et al.*, 1999):

$$X_c = \frac{\text{Intensity of peak derived from crystalline structure}}{\text{Total intensity}}$$

Observation using SEM and measurement of particle diameter The waxy rice starch, and spray-dried powders from the 100 °C starch solution, 140 °C, 160 °C, and 180 °C fine starch solutions, and the microencapsulated inulin hydrolysate powder (160 °C fine starch solution with dextrin hydrate as the wall material) were observed using a scanning electron

microscope (JSM-6301F, JEOL Ltd., Tokyo, Japan). The samples were mounted on aluminum stubs using double-sided carbon tape then coated with a gold-palladium alloy using ion sputtering (E101 Ion Sputter, Hitachi Ltd., Tokyo, Japan) before SEM observation. The observations were carried out under high vacuum conditions at less than 1.0×10^{-3} Pa with an acceleration voltage of 10 kV. From the micrographs, the particle diameter was derived from the circle equivalent diameter using a tool in the open source software, ImageJ (imagej.net). For measuring the particle size distribution, 300 particles with distinct outlines were counted.

Results and Discussion

Mass spectra of inulin hydrolysate solution Fig. 1 shows the ESI-MS spectra (negative ion mode) of DFAlII and inulin hydrolysate prepared by the compressed hot water process at 165 °C (inulin hydrolysate solution). The m/z ratios of 323 and 647 correspond to the DFAs and DFAs dimer signals respectively, therefore, it was confirmed that the inulin hydrolysate solution prepared by the hot compressed water process contained DFAs. In addition, the m/z 179 signal indicated the presence of monosaccharides contributing to fructose and glucose (Barclay *et al.* 2012). The m/z 359 and m/z 1006–1883 signals indicated the monosaccharides dimer and completely unreacted inulin, respectively.

Viscosity changes in primary starch solution caused by the compressed hot water process Fig. 2 shows the viscosity of the solutions. The range of viscosity measurement varied according to the spindle speed, so some points could not be measured. And furthermore measurements below 10 mPa·s was unmeasured. The viscosity of the 100 °C starch solution was approximately 2700 mPa·s (58 cm²/min) to 9300 mPa·s (12 cm²/min). This value decreased by approximately 380 mPa·s (466 cm²/min) to 2000 mPa·s (12 cm²/min) in solutions processed by compressed hot water at 140 °C with those at 160 °C and 180 °C being 22–24 and under 10 mPa·s, or immeasurably small, respectively. The viscosities of the mixed solution of inulin hydrolysate and 160 °C fine starch solution were 13–14 mPa·s and those of the inulin hydrolysate and dextrin hydrate solutions under 10 mPa·s.

The starch solution, prepared at 100 °C and originally exhibiting a high viscosity, was a non-Newtonian, pseudoplastic fluid, because of its characteristics and molecular structure in solution. The compressed hot water process

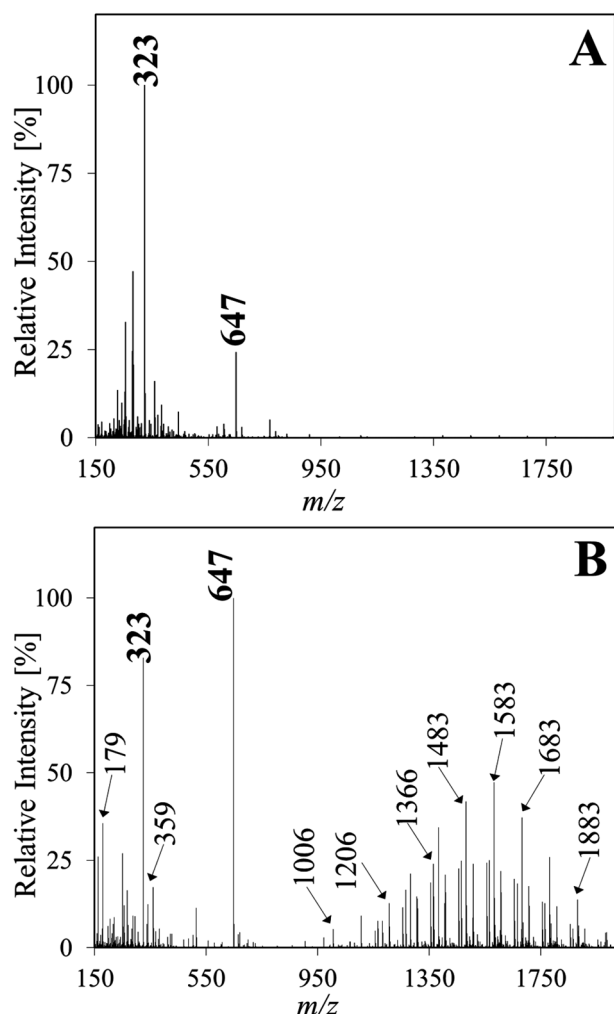


Fig. 1. ESI-MS spectra (negative ion mode) of DFAIII (A) and inulin hydrolysate solution prepared by the hot compressed water process at 165 °C (B). m/z 323 correspond to the DFAs signals [M-H]⁻ and m/z 647 correspond to the DFAs dimer [2M-H]⁻.

through hydrolysis led to this peculiar starch structure gradually disappearing and its viscosity gradually decreasing.

The fine starch solution became less viscous and the starch particles smaller because of the compressed hot water process while the solution concentration remained the same. Therefore, the fine starch solution was deemed suitable to use as a wall material for microencapsulation (Yoshioka and Shimizu, 2014).

Powder X-ray diffraction Fig. 3 shows the results from this procedure where the starch powder exhibited a peak derived from its crystalline form with an X_c value of approximately 0.5. After the hydrothermal and spray-drying processes, the crystals in the starch particles had broken down to the amorphous form with an X_c value of 0.

SEM micrographs and particle size distribution Fig. 4 shows the SEM micrographs of each powder. The particle morphology of the waxy rice starch (Fig. 4 (A)) showed an

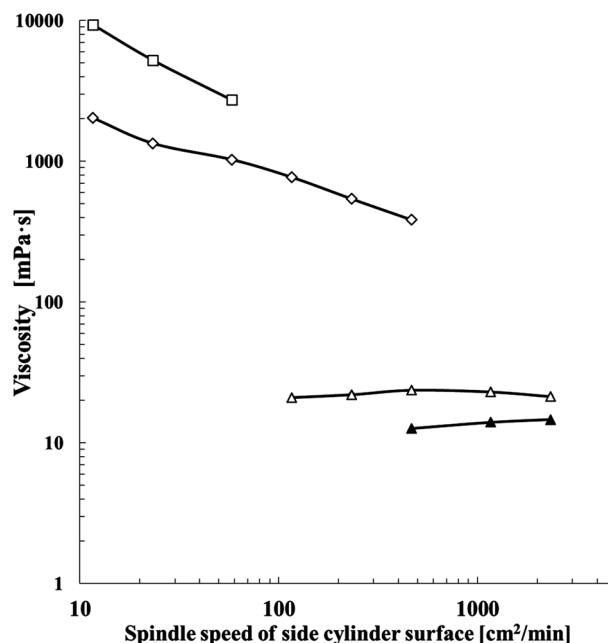


Fig. 2. The viscosity change of 100 °C starch solution (□), 140 °C (◇), and 160 °C (△). The viscosity change of mixed solutions of core (DFAs) and 160 °C fine starch solution (▲) as wall material using a rotating cylinder viscometer.

angular shape and a uniform material, but the spray-dried particles (Fig. 4 (B)–(E)) showed an amorphous structure with many wrinkles or dimples. The spray-dried powder from the fine starch solution prepared by the compressed hot water process at a higher temperature led to small particles of a uniform size (Fig. 5 (A)).

During the spray-drying process, the high viscosity of the solution interfered with the atomization process. This led to the solution adhering to the inside surface of the drying chamber, to forming large droplets that decreased the drying rate, and to producing a non-uniform distribution of particle sizes. In microencapsulation by spray-drying, the appropriate particle size is obtained through the selection of the core and wall materials (Gharsallaoui *et al.* 2007). In the present study, the particle size of the spray-dried powder became uniform as the viscosity of the fine starch solution decreased. The particle size can also be controlled by changing the diameter of the nozzle in the spray-dryer, and other aspects of the operation.

Microencapsulation of inulin hydrolysate Preparing inulin hydrolysate powder by spray-drying or freeze-drying can be difficult because the inulin hydrolysate solution strongly adheres to the inside surface of the drying chamber. For microencapsulation by spray-drying using the 160 °C fine starch solution as the wall material, this strong adhesion disappeared and the microencapsulated inulin hydrolysate powder could be obtained (Fig. 4 (F)). For the control treatment using dextrin hydrate as the wall material, microencapsulated inulin hydrolysate powder was also obtained (Fig. 4 (G)). The

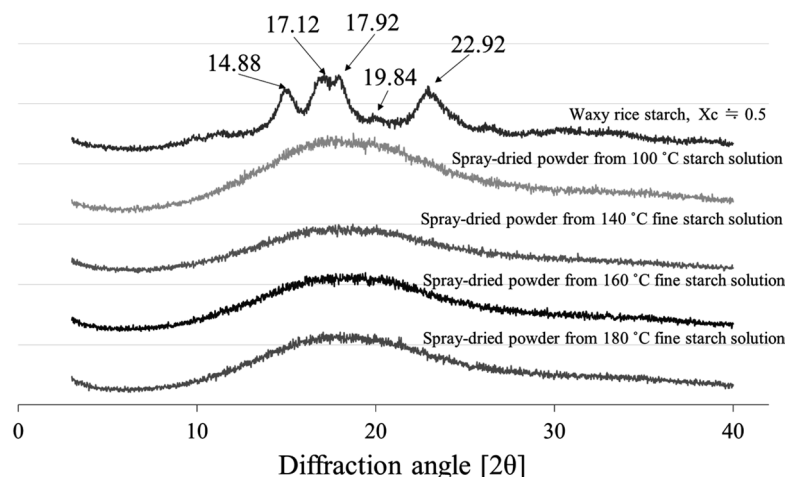


Fig. 3. X-ray diffraction of the waxy rice starch, and the spray-dried powders from the 100 °C starch solution and the 140 °C, 160 °C, and 180 °C fine starch solutions.

average size of the microencapsulated particles using fine starch and dextrin hydrate was 4.1 and 3.2 μm , respectively. It can be suggested that the particle size was influenced by the viscosity of the primary solution (Fig. 5 (B)).

Dextrin or maltodextrin used as wall material has generally low weight-average molecular weight: $M_w < 20000$ (Sun *et al.*, 2010). Fine starch prepared compressed hot water process was appeared to have large molar mass compared to dextrin and maltodextrin from the results of viscosity and the previous study (Yoshioka and Shimizu, 2014). Conversely, fine starch appeared to have a peculiar amylopectin structure including branching. Branching in macromolecule affects polymer properties including glass transition temperature, chemical resistance, release rate of core materials, so on (Podzimek, 2011). Fine starch is expected to have different properties from dextrin and maltodextrin.

However, although the DFAs content of the microencapsulated powder was not known, the convenience of handling inulin hydrolysate including DFAs was enhanced so that this treatment should contribute to the wider application of DFAs, including tablets. From these results, the fine starch solution functioned adequately as a wall material.

By using microencapsulation, deterioration in the quality of the core material through the reaction of heat, oxygen, and volatilization can be prevented. Inulin hydrolysate are not a material which can easily deteriorate and volatilize. To investigate how the fine starch prepared using the compressed hot water process led to this distinctive wall function, further experiments on microencapsulation using unstable materials as the core material are needed.

In the present study, the pressure vessel used for the compressed hot water process meant a batch-type process. Pressure vessels operating continuously do exist so the continuous production of powder may be possible when the compressed hot water process is linked to the cooling and

spray-drying processes. In this study, the core material solution (inulin hydrolysate solution) and fine starch solution (wall solution) were prepared separately then mixed. This preparation work could be simplified by first mixing the starch and inulin solutions, and then prepared by the compressed hot water process which would provide a more efficient production of microencapsulated inulin hydrolysate powder than previously possible.

Conclusion

In the present study, inulin hydrolysate including DFAs powder has been microencapsulated using spray-drying using a fine starch solution as the wall material, an improvement in the previous situation. In general, starch, especially amylopectin, has a large average molecular weight, so starch solutions have a high viscosity. This makes starch solutions unsuitable to use as wall materials in microencapsulation using spray-drying. When the starch solution is treated by the hot compressed water process, the particle size in solution and the solution viscosity decreases through hydrolysis. As the viscosity of the fine starch solution decreases, the spray-dried powder becomes uniformly fine. Inulin hydrolysate powder was also obtained using a 160 °C starch solution with dextrin hydrate as the wall material. The differences in the particle size and appearance of these microencapsulated powders are thought to be affected by the viscosity of the primary solution. Finally, starch is the second most abundant material in biomass and already widely used, so it has the great advantage of being inexpensive. Overall, a fine starch solution is a promising new wall material for microencapsulation by spray-drying.

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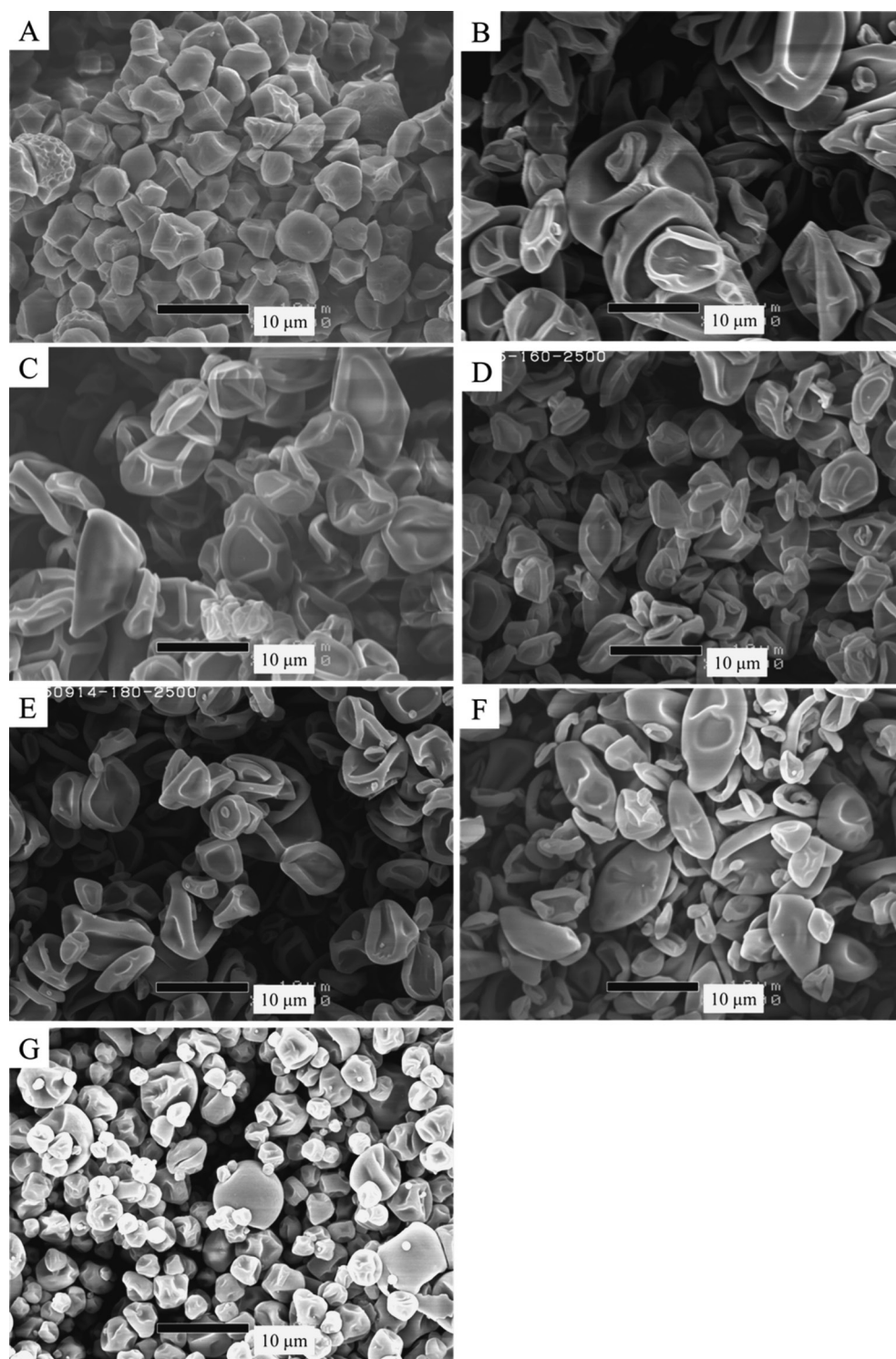


Fig. 4. Scanning electron microscope micrographs of waxy rice starch (A), spray-dried powders from 100 °C starch solution (B), 140 °C (C), 160 °C (D), and 180 °C (E) fine starch solution, microencapsulated inulin hydrolysate powders using the 160 °C fine starch solution (F) and dextrin hydrate (G) as wall material.

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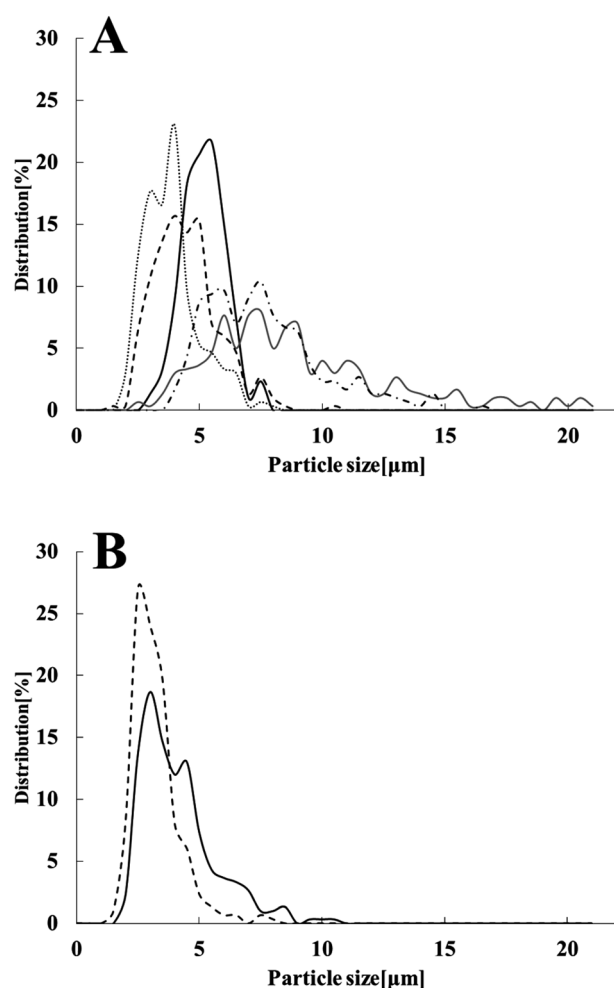


Fig. 5. Particle size distribution of waxy rice starch (—) with average particle size 5.2 μm, spray-dried powders from 100 °C starch solution (—), 140 °C (-----), 160 °C (----), and 180 °C (.....) fine starch solution with average particle size 9.0 μm, 7.6 μm, 4.5 μm, and 3.7 μm, respectively (A). Particle size distribution of microencapsulated inulin hydrolysate powders using 160 °C fine starch solution (—) and dextrin hydrate (-----) as wall material with average particle size 4.1 μm and 3.2 μm, respectively (B).

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