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Effects of Hydroxypropylation on the Functional Properties of Psyllium

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The hydroxypropylated psyllium derivatives were successfully prepared with propylene oxide under the alkaline condition for the first time. Four hydroxypropylated psyllium derivatives, denoted as HP1, HP2, HP3, and HP4, were characterized for their hydroxypropyl content, molar substitution, and IR spectra. The hydroxypropyl derivatives were also evaluated for their surface structure, gelling properties, water uptake capacities, swelling volumes, and in vitro bile acid-binding abilities. The results showed that hydroxypropylation significantly reduced the gelling properties of psyllium. Psyllium derivatives with a relatively low hydroxypropyl substitution degree had greater in vitro binding capacities against cholic and chenodeoxycholic acids and higher swelling ability. The results from this study suggested that hydroxypropylation may be a possible approach for obtaining novel psyllium derivatives with improved physicochemical, functional, and biological properties for utilization in functional foods or supplemental and pharmaceutical products.

KEYWORDS: Hydroxypropylation; psyllium; gelling properties; bile acid-binding capacity

INTRODUCTION

Psyllium is a mucilaginous material derived from seed husks of *Plantago* ovata and has been used for the treatment of constipation, diarrhea, irritable bowel syndrome, inflammation bowel diseases, ulcerative colitis, colon cancer, obesity in children and adolescents, high cholesterol, and diabetes in the functional food, dietary supplemental, and folk medicine products (*1*−*4*). However, it is still a challenge to incorporate adequate amount of psyllium in a selected food product for the health claim because of its physicochemical properties such as the strong gelling capacity.

A few physical, mechanical, enzymatic, and chemical approaches have been developed to improve the physicochemical properties of psyllium and consequently to promote its utilizations in food or other consumer products (*1*, *4*−*7*). The enzymatic and chemical approaches have been shown to be able to modify the chemical and molecular structures of psyllium, and may effectively improve its physicochemical properties and biological activity (*4*, *6*). The enzymatic approaches are limited by the availability of food grade enzymes and the overall cost, although they are generally considered green since no organic solvents or chemicals are involved or generated (*7*). However, chemical modification is less expensive and may produce psyllium derivatives with more diverse chemical and molecular structures, and becomes an important approach to modify the properties of natural biopolymers including psyllium.

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Recently, a few psyllium derivatives have been prepared using chemical methods and investigated for their functionalities (*2*, *7*−*10*). Carboxymethylation was shown to enhance the water solubility of the arabinoxylan from psyllium (*2*), and the ethylation was able to alter the intrinsic viscosity of the arabinoxylan derivatives (*8*). Our recent study indicated that sulfation may reduce the gelling property of psyllium and enhance its bile acid-binding capacity (*10*). These research activities have indicated the potential of improving psyllium functionality through chemical modifications.

Hydroxypropylation is a widely accepted method to improve the functionality of polysaccharides. Several edible polysaccharides such as starch, chitin, and cellulose have been hydroxypropylated to improve shelf life, freeze−thaw stability, cold water swelling, and reconstituting properties (*11*, *12*). Particularly, among starch ethers, hydroxypropyl starch with a reduced gelatinization property is unique for food use (*11*, *13*). To date, hydroxypropylation has not been evaluated for its potential in improving physicochemical properties and functionalities of psyllium.

As part of our continued effort in promoting the application of psyllium in food and supplemental products to improve human health, this research was conducted to evaluate the effects of hydroxypropylation on physicochemical and biological properties of psyllium. The preparation of hydroxypropylated psyllium derivatives using propylene oxide under alkaline conditions was reported. The chemical structures of the hydroxypropylated psyllium derivatives were characterized using FT-IR, hydroxypropyl content determination, and scanning electron microscope (SEM). In addition, the effects of hydroxypropylation on the gelling properties, water uptake capabilities, swelling behaviors,

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and in vitro bile acid binding capacities of psyllium were investigated.

MATERIALS AND METHODS

Chemicals and Reagents. Acid treated psyllium (Psy) was obtained from psyllium husks by acid hydrolysis as described previously (*7*). Propylene oxide, sodium hydroxide, isopropanol, and hydrochloric acid were purchased from Thermo Fisher Scientific Inc. (NY, USA). Nitroblue tetrazolium chloride (NBT), individual bile acids (cholic and chenodeoxycholic acids), diphorase, nicotinamide adenine dinucleotide, and 3-α hydroxysterol dehydrogenase were purchased from Sigma-Aldrich (St. Louis, MO). All other chemicals used were of analytical grade and without further purification.

Hydroxypropylation. Hydroxypropylation procedure was developed according to previously reported methods with minor modifications (*12*, *14*). Briefly, psyllium (5.0 g) was mixed with isopropanol and followed by the addition of 50 mL of sodium hydroxide solution. After stirring for 30 min, different amounts of propylene oxide were added to the reaction mixture. The reaction was carried out with continuous stirring at 40 °C for 36 h. On completion of the reaction, we neutralized the solution with 5 M HCl and dialyzed it against distilled water for 96 h. The dialyzates were concentrated under reduced pressure and lyophilized to obtain the hydroxypropyl derivatives.

Determination of Hydroxypropyl Content. The method described by Jones and Riddick (*15*) was used for the determination of the hydroxypropyl groups. The hydroxypropyl derivative (100 mg) was weighed accurately into a 100 mL volumetric flask and 25 mL of 0.5 M H_2SO_4 was added. The mixture was heated in a water bath (60 °C) to dissolve the sample. The dissolved sample solution was cooled and diluted to 100 mL with distilled water. One milliliter aliquot of the solution was transferred into 25 mL graduated test tubes and immersed in a cold water bath, and 8 mL of concentrated H_2SO_4 was added dropwise. After mixing well, the tube was placed in a boiling water bath for 20 min and transferred immediately to an ice bath. Then, 0.6 mL of ninhydrin reagent (3% ninhydrin in 5% $Na_2S_2O_5$) was added, and the tube was shaken thoroughly before being kept in a 25 °C water bath for 100 min. The solution was made up to 25 mL with concentrated H_2SO_4 and mixed well. The absorbance was measured at 590 nm after keeping still for 10 min. A calibration curve was prepared with a 1 mL aliquot of standard aqueous solution containing 10, 20, 30, 40, or 50 μg of propylene glycol per mL. The factor to convert the weight of propylene glycol into the hydroxypropyl group was 0.7763. The numbers of moles of propylene oxide per anhydroxylose (MS) was calculated using the following equation (*11*):

$$\mathrm{MS} = \frac{132 \times \%C_3H_7O}{59 \times (100 - \%C_3H_7O)}$$

Infrared Spectral Analysis. The IR spectra were recorded on a Nicolet-170X spectrophotometer (Nicolet, USA) between 400 and 4000 cm^{-1}.

Scanning Electron Microscope (SEM). The morphological information of Psy and hydroxypropyl derivatives was investigated using an SU-70 Analytical Ultra-High Resolution Scanning Electron Microscope (Hitachi, Tokyo, Japan). Samples were placed directly onto conductive carbon tapes on a specimen stub and coated with a 10 nm layer of gold.

Gelling Property Determination. Gelling properties of modified psyllium samples were measured and compared with that of the starting material and the control according to a previously described laboratory method (*7*). The control sample went through the hydroxypropylation procedure without the presence of propylene oxide. A TA. XT plus texture analyzer with a 10 mm diameter probe (Texture Technologies Corp., Scarsdale, NY) was used for the determination. Gel samples were prepared by mixing 1.50 g of psyllium sample in 30 mL of pure water in a 50 mL beaker. After setting for 24 h at ambient temperature, the gel samples were subjected to a double compression test. Measurements were performed with a pretest speed of 2.0 mm/s, a test speed of 5.0 mm/s, a post-test speed of 5.0 mm/s, and a distance of 6.0 mm. The maximal force to break the gel was determined as gel hardness, and the force of gel sample to hold the probe was determined as the adhesiveness. Triplicate measurements were taken for each sample.

Swelling Capacity Assay. Swelling capacities of psyllium samples were determined following the laboratory procedure described by Zhou and others (*16*). Briefly, 0.1 g of each sample was suspended in 7 mL of simulated intestinal fluid without enzyme according to U.S. pharmacopeia (*17*). After keeping the suspensions at 37 °C for 8 h with occasional shaking, the mixture was kept for another 16 h at ambient temperature (22 °C) for the sediment to settle. Then the volumes of the sediments were recorded. The swelling capacity was calculated as mL of sediment/g of each psyllium sample. Duplicate assays were conducted.

Water-Uptake Capacity Assay. Water-uptake capacities of Psy and the hydroxypropyl derivatives were determined and compared gravimetrically according to a previously reported laboratory protocol (*6*) with minor modifications. Briefly, the samples were equilibrated in a 15% relative humidity chamber at ambient temperature (22 °C) for 72 h, and their weights were measured. The samples were then transferred to a 90% relative humidity chamber and exposed for 30 min, and measured for total weight. The weight change was calculated and reported in milligram of water taken by per gram of the sample per min. Duplicate tests were conducted for each psyllium sample.

Bile Acid Binding Capacity Assay. The bile acid-binding capacities were determined following a previously reported laboratory procedure (*7*, *16*). Briefly, a 50 mg sample was treated with 0.5 mL of 0.01 mol/L HCl, which simulated gastric conditions, and was incubated at 37 °C for 60 min with continuous shaking (50 rpm). The solution was adjusted to pH 7.0 by adding 0.05 mL of 0.1 mol/L NaOH and mixed with 5.0 mL of 400 μmol/L bile acid stock solution prepared in 0.01 mol/L phosphate buffer (pH 7.0), which simulated the intestinal condition. The mixture was incubated for another 60 min at 37 °C and centrifuged for 10 min at 6000 rpm, and the supernatant was collected for bile acid determination. Quantification of the unbound bile acids was carried out using a commercial kit from Sigma-Aldrich (St. Louis, MO). The final assay mixture was added with 100 μL of supernatant or bile acid standards, 125 μL of 1.22 mmol/L nicotinamide adenine dinucleotide, 125 μL of 5 mmol/L NBT, 100 μL of 625 units/1 diphorase, and 100 μL of 625 units/1 3-α hydroxysterol dehydrogenase. The mixture was incubated for 60 min at ambient temperature. After incubation, 100 μL of 1.33 mol/L phosphoric acid was added to stop the reaction, and the absorbance of each reaction mixture was measured at 530 nm. The levels of unbound bile acids were obtained using a standard curve prepared with each of the two pure bile acids, which were cholic and chenodeoxycholic acids. The bile acid-binding capacity (mg/g sample) was calculated against a reagent blank. Triplicate tests were performed for each sample against each bile acid.

Statistical Analysis. Data are reported as the mean ± SD for all determinations. ANOVA and Tukey's tests were performed (Minitab for Windows, Version 13, Minitab Inc., PA) to identify differences among means. Statistical significance was declared at $P \leq 0.05$.

RESULTS AND DISCUSSION

Characterization of Hydroxypropyl Psyllium Derivatives. Hydroxypropylation of psyllium is an etherification process achieved by using propylene oxide as the etherifying reagent. The reaction introduced hydroxypropyl groups onto the free hydroxyl group of the polymeric xylan chain (*18*, *19*). Four hydroxypropylated psyllium products, HP1, HP2, HP3, and HP4, were obtained using the different reaction conditions with a yield of 60.24−84.45% (**Table 1**). The hydroxypropyl content of the four hydroxypropylated psyllium ranged from 6.33 to 15.26%, and the molar substitution (MS) of these derivatives ranged from 0.15 to 0.40 (**Table 1**). The hydroxypropylated psyllium with higher hydroxypropyl content had greater MS (**Table 1**). Increase in either the NaOH concentration or the volume of propylene oxide resulted in a greater hydroxypropyl content or MS in the hydroxypropylated psylliums (**Table 1**).

Hydroxypropylation of psyllium was also confirmed by IR spectra. **Figure 1** presents the IR spectra of psyllium (Psy), the control which went through the procedure without the presence of propylene oxide, and HP1, HP2, HP3, and HP4 in the 400−4000 cm^{-1} region. The IR spectrum of all psyllium samples

Table 1. Reaction Parameters, Hydroxypropyl Content, Molar Substitution (MS), and Yield[a]

product code	NaOH concentration	propylene oxide (mL)	hydroxypropyl content (%)	MS	yield (w/w, %)
HP1	30%	15	6.33 ± 0.001	0.15	78.72
HP2	50%	15	10.47 ± 0.003	0.26	84.45
HP3	30%	30	12.63 ± 0.007	0.32	69.97
HP4	50%	30	15.26 ± 0.006	0.40	60.24

[a] HP1, HP2, HP3, and HP4 represent different hydroxypropyl psylliums, respectively.

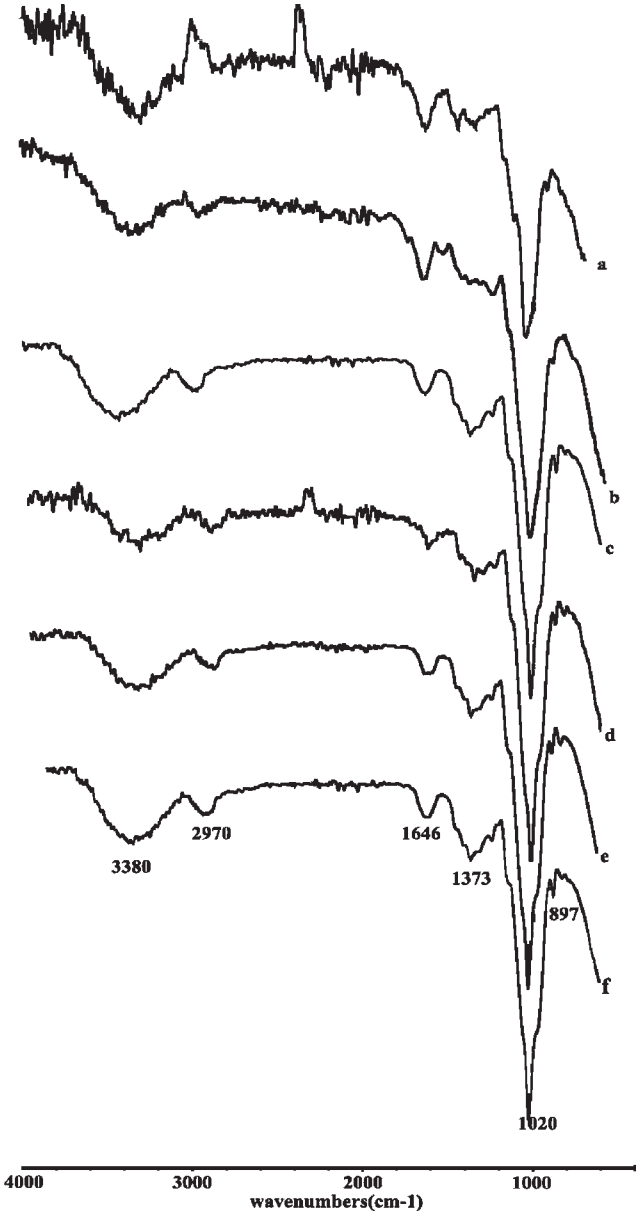

Figure 1. IR spectra of psyllium samples. a, represents the starting psyllium preparation; b, represents the control sample which went through the hydroxypropylation procedure without the hydroxypropylating reagent; c−f, represent different hydroxypropylated psylliums HP1, HP2, HP3, and HP4, respectively.

had the typical absorption peaks assigned to the saccharide moiety at 3300−3400, 1646, 1020, and 897 cm^{-1}. In comparison with Psy, two new characteristic absorption bands appeared in the IR spectra of the four hydroxypropyl derivatives, one appeared at 2970 cm^{-1} (which overlapped the band at 2920 cm^{-1}) corresponding to the C−H stretching and the other at 1379 cm^{-1}, representing the bending of the CH_3 group. These absorption peaks indicated that the CH_3 group was introduced into the psyllium molecule after reacting with propylene oxide under the experimental conditions (*9*, *20*).

Scanning electron micrographs of Psy, the control, and HP1 are shown in **Figure 2**. The micrograph of only one derivatized psyllium is presented because no pronounced differences were observed among the micrographs of all four hydroxypropylated psylliums. The surfaces of the control and Psy are different, suggesting that the method of lyophilization might have induced significant change to the surface structure of psyllium particles. Furthermore, the filamentous gel network seemed to be replaced by small pieces with smooth surface in the SEM picture of HP1. It was reported earlier that particle size and specific surface structure might influence the hydration behavior of gel forming polymers (*21*, *22*). Therefore, the SEM results also suggested that hydroxypropylation might alter the gelling properties and solution behavior of psyllium.

Effects of Hydroxypropylation on Gelling Properties. No significant change in both hardness and adhesiveness of the starting material Psy and the control indicated that the lyophilization procedure did not alter the gelling properties of psyllium (**Figure 3**). **Figure 3a** showed that less force was required to break the gels prepared from the hydroxypropylated psyllium than that to break the gels prepared from the starting material Psy or the control. Significant reductions were also observed in the adhesiveness of the gels prepared from the hydroxypropyl derivatives compared to that for the starting material Psy or the control (**Figure 3b**). HP1 and HP2 with a hydroxypropyl substitution degree of 0.15−0.26 showed a reduction of 31.96% and 49.72% in gel hardness and a reduction of 31.66% and 56.48% in gel adhesiveness as compared to those of the starting material, while HP3 and HP4 with a hydroxypropyl substitution degree of 0.32−0.40 formed a very weak gel requiring very low forces of 1.9 and 1.85 g to break the gel and very weak forces of 0.90 and 0.78 g to hold the compressing probe, respectively. Taken together, these data indicate that hydroxypropylation had significantly reduced the gelling capacities of psyllium (**Figure 3**). The reduced gelling capacity might be explained by the reduced capacity for junction zone formation due to the presence of the bulky hydroxypropyl substitutions along the xylan backbone (*6*, *7*, *11*). This statement was supported by the previous observation that waxy maize and potato starch preparations with higher hydroxypropylation degree formed very soft gels (*23*). These data also indicated the possibility of obtaining psyllium derivatives with desirable gelling properties through the introduction of selected degrees of hydroxypropyl groups into the molecule, and derivatives with different hydroxypropyl content might have different rheological behaviors.

Effects of Hydroxypropylation on Swelling Capacity. Swelling volume measures the water-holding capacity of polymers, and it is also an important parameter for predicting the health beneficial properties of psyllium (*4*). The swelling volume of

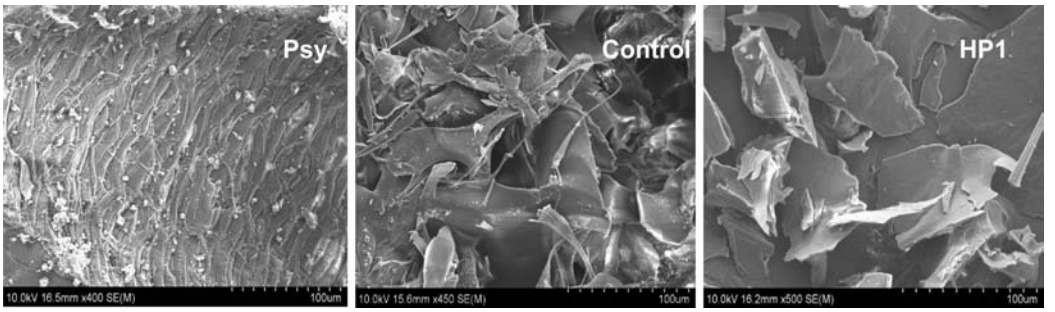

Figure 2. SEM measurement of psyllium samples. Left panel represents the starting psyllium preparation, the middle panel represents the control sample which went through the hydroxypropylation procedure without the hydroxypropylating reagent, and the right panel represents hydroxypropylated psyllium HP1.

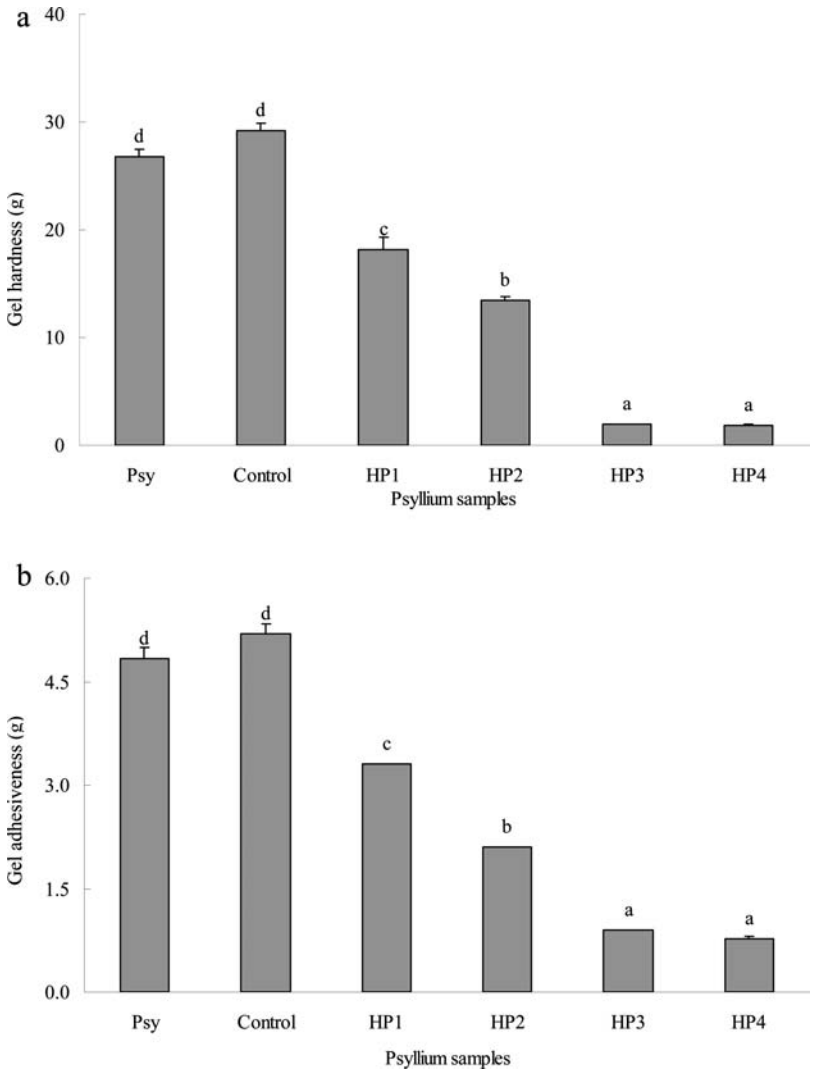

Figure 3. Effects of hydroxypropylation on gelling properties of psyllium: (**a**) hardness and (**b**) adhesiveness. Psy represents the starting psyllium preparation; Control represents the sample which went through the hydroxypropylation procedure without the hydroxypropylating reagent, while HP1, HP2, HP3, and HP4 represent different hydroxypropyl psylliums, respectively. Data are expressed as mean ± SD. Vertical bars represent the SD. Values carrying the same letters are not significantly different ($P > 0.05$).

the starting material, control, and hydroxypropyl psyllium preparations are presented in **Figure 4**. HP1 had significantly higher swelling volume than Psy and the control psyllium, and HP3 and HP4 had significantly lower swelling volumes than Psy and the control, whereas there was no difference in swelling volume between HP2 and the control (**Figure 4**), indicating the potential influence of hydroxypropylation on the swelling behavior of psyllium. The about 1.5-fold increase in swelling volume of HP1 compared to that of the control might be due to the introduction of bulky hydroxypropyl groups into the psyllium molecule, which might have enhanced the repulsion among the molecules, thereby facilitating water percolation into the psyllium molecule networks (*24*). Further increase in the hydroxypropyl content might have disrupted the intramolecular hydrogen bonds and other associations, and weakened the gel network (*25*).

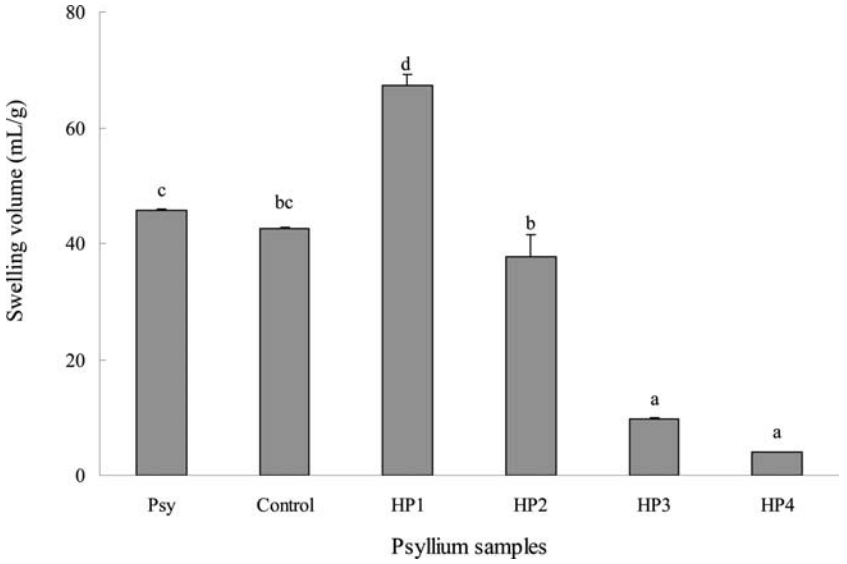

Figure 4. Effects of hydroxypropylation on the swelling volume of psyllium. Psy represents the starting psyllium preparation. Control represents the sample which went through the hydroxypropylation procedure without the hydroxypropylating reagent, while HP1, HP2, HP3, and HP4 represent different hydroxypropyl psylliums, respectively. Data are expressed as mean ± SD. Vertical bars represent the SD. Values carrying the same letters are not significantly different ($P > 0.05$).

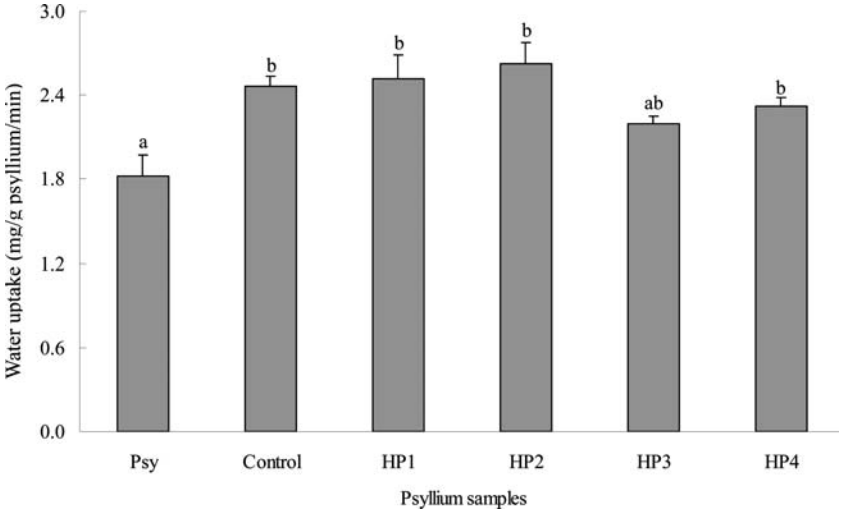

Figure 5. Effects of hydroxypropylation on water uptake capacities of psyllium. Psy represents the starting psyllium preparation. Control represents the sample which went through the hydroxypropylation procedure without the hydroxypropylating reagent, while HP1, HP2, HP3, and HP4 represent different hydroxypropyl psylliums, respectively. Data are expressed as mean ± SD. Vertical bars represent the SD. Values carrying the same letters are not significantly different ($P > 0.05$).

Effects of Hydroxypropylation on Water-Uptake Properties. **Figure 5** showed significantly increased water-uptake capacity of hydroxypropylated psylliums and the control compared to that for the starting material. The water-uptake properties may be affected by many factors such as lyophilization process, introduction of hydrophilic hydroxypropyl groups, free hydroxyl groups at surfaces, and changes of surface structure and chain conformation of psyllium (*26*−*28*). The hydrophilic nature of the hydroxypropyl groups could enhance the water uptake ability of the polymers, while lyophilization also contributed to the increased water uptake capacity through an increase of the particle surface area (**Figure 2**). The multichip surface structure also provided more chances for the interaction between polymer and water molecules.

Effects of Hydroxypropylation on Bile Acid-Binding Properties. Bile acids are steroid carboxylic acids synthesized in the liver from cholesterol. Binding bile acids to nondigestible polymers may enhance their elimination and promote the conversion of cholesterol to bile acids, which may reduce the plasma total and LDL cholesterol levels and the risk of cardiovascular diseases (*29*). HP1, HP2, HP3, and HP4 were evaluated and compared to Psy and the control for their in vitro binding capacities against cholic acid (CA) and chenodeoxycholic acid (CDCA), which are the primary bile acids synthesized in the liver (**Figure 6**). **Figure 6** shows that HP1 with the lowest hydroxypropyl content had a 1.8-fold CA binding capacity than that of Psy, while both HP1 and HP2 with lower hydroxypropyl content exhibited no difference in their CDCA binding capacity compared to that for Psy. Furthermore, HP3 and HP4 with higher hydroxypropyl content lost binding capacities against both CA and CDCA (**Figure 6**). In addition, the trend of bile acid binding capacity agreed well with the order of swelling volume for the psyllium samples, indicating the potential link between the swelling behavior and bile acid

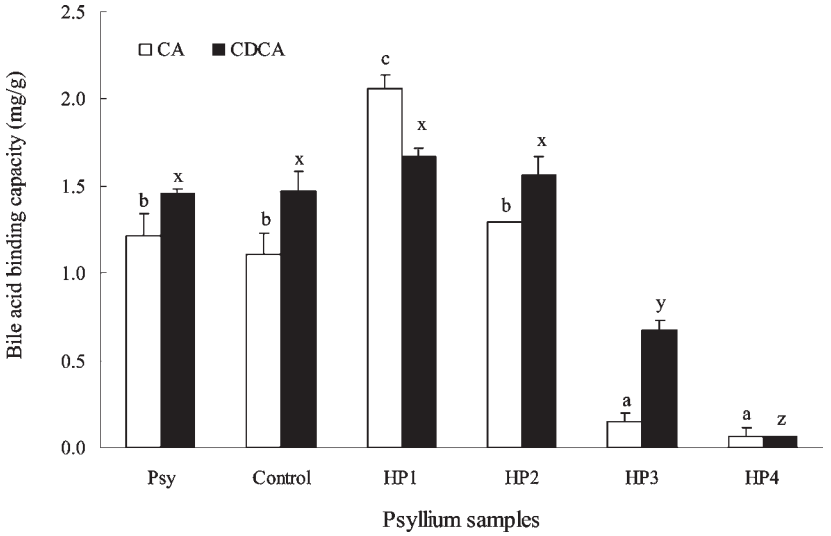

Figure 6. Effects of hydroxypropylation on bile acid-binding capacities of psyllium. Psy represents the starting psyllium preparation. Control represents the sample which went through the hydroxypropylation procedure without the hydroxypropylating reagent, while HP1, HP2, HP3, and HP4 represent different hydroxypropyl psylliums, respectively. Data are expressed as mean ± SD. Vertical bars represent the SD. Values carrying the same letters are not significantly different ($P > 0.05$).

binding property for hydroxypropylated psyllium samples. These data suggested that hydroxypropylation may alter its bile acid-binding capacity and that the substitution degree may be critical to its overall bile acid-binding property.

In summary, the results from this study indicate that hydroxypropylation may serve as a possible approach for improving physicochemical and functional properties of psyllium without reduction on its beneficial effects. Further in vivo bioactivity and toxicological evaluations of hydroxypropylated psyllium are recommended to promote their food, supplemental, and pharmaceutical applications.

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