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Research Article

Preparation and Property of Xylan/Poly(Methacrylic Acid) Semi-Interpenetrating Network Hydrogel

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Xylan/poly(methacrylic acid) semi-interpenetrating network hydrogels were synthesized, and these hydrogels were fabricated using various ratios of xylan and methacrylic acid and cross-linked by N,N′-methylenebisacrylamide. The chemical structure and morphology of the prepared hydrogels were characterized by FT-IR spectroscopy and SEM analysis. The porous structure of the prepared hydrogels was found, and the interconnected porous channels increased when the content of xylan was increased in the hydrogels. The hydrogels presented excellent pH sensitivity and swelling reversibility. The dynamic swelling kinetics were also studied, and all obtained results indicated that the prepared xylan/poly(methacrylic acid) semi-IPN hydrogel could be used in biomedical fields, especially for drug release.

1. Introduction

Hydrogel has a three-dimensional network structure, and it can absorb water but cannot dissolve in water [1, 2]. Because of its excellent swelling property and multiresponses on environmental stimuli, hydrogel has attracted considerable research interest in many fields, especially in drug controlled release [3–6], enzyme immobilization [7, 8], tissue engineering [9, 10], cell culture [11, 12], wastewater treatment, and so on [13–15]. Depending on the chemical composition of hydrogel, it is classified into two categories: one is synthetic polymer-based hydrogel that was mainly synthesized from monomers or synthetic polymers such as poly(methacrylic acid); the other is the hydrogel that was prepared from natural polymer, for instance, cellulose, chitosan, alginate, gelatin, konjac glucomannan, guar gum, and other natural polymers [4, 6, 8, 9, 16, 17]. Hydrogel made of poly(methacrylic acid) is an ionizable hydrogel which has been studied for controlled drug release [18], but it is not biodegradable. Because of the excellent biodegradability and biocompatibility of natural polymers, the preparation of these hydrogels becomes a hot research topic. In particular, the hydrogel based on natural polysaccharide can be used in the field of the controlled drug release. Interpenetrating polymer networks hydrogel based on konjac glucomannan and poly(methacrylic acid) had been prepared for controlled drug release [18].

Xylan is second only to cellulose as most prevalent polysaccharide in nature. It possesses β-(1,4) linked xylose units as the backbone and sugar- and *O*-acetyl groups as substitutes, and it accounts for 25–35% of the dry weight of plant cells; therefore, xylan is a rich, inexhaustible, and renewable polymer [19]. Xylan has biodegradable property and biocompatibility and possesses inhibitory actions on mutagenicity activity and antiphlogistic effects and immune functions [20–22]. In previous studies, we have studied the separation and chemical structure of xylan [23, 24], and these studies offer the foundation for the synthesis of xylan-based materials. In recent years, xylan has been used for the preparation of food packaging materials, baking, papermaking, and food additives [21]. Oliveira has extracted xylan from corn cobs and found that xylan is a promising raw material for the pharmaceutical industry [25]. Because of the excellent performance, xylan-based materials are expected to be widely used in the medical field, especially for drug delivery and tissue engineering. A novel xylan/PVA hydrogel had been synthesized, and the study indicated that the gel was noncytotoxic and had a potential for biomedical application [26]. Recently, we prepared hemicellulose-based,

pH-sensitive, and biodegradable hydrogel for controlled drug delivery [22].

In this paper, the semi-interpenetrating network hydrogels based on xylan and poly(methacrylic acid) (xylan/PMAA semi-IPN hydrogel) were synthesized and characterized. The chemical structure and surface morphology were analyzed using FT-IR and SEM, respectively. pH sensitivity, swelling reversibility, and swelling kinetics were also investigated. The exponential heuristic equation was used to analyze the swelling mechanism of the prepared hydrogels in different pH buffer solutions.

2. Experimental

2.1. Materials. The monomer methacrylic acid (MAA) was purchased from the Tianjin Kermel Chemical Reagent Company in China. The cross-linking agent N,N-methylenebisacrylamide (Bis), initiator ammonium persulfate (APS), and anhydrous sodium sulphite were purchased from the Tianjin Hongyan Chemical Reagent Factory in China. All of the reagents used were of analytical grade. Twice-distilled water was used for synthesis reactions and the preparation of buffer solutions.

2.2. Isolation of Xylan from Wheat Straw. Xylan was isolated from wheat straw by the following steps: wheat straw was first cut and dried in an oven at 60°C and then dewaxed using toluene and ethanol in a 2 : 1 volume ratio in a Soxhlet apparatus for 12 h. After filtration, the residue was delignified using sodium chlorite in an acidic solution (pH 4.0) at 75°C for 2 h, in order to obtain holocellulose. The holocellulose was treated using 10% KOH at a 1 : 20 solid to liquid ratio at room temperature for 10 h and then filtered to separate the hemicellulose and cellulose. The filtrate was neutralized to pH 5.5, concentrated under a reduced pressure, and then precipitated into 3 volumes of ethanol to obtain hemicellulosic powder.

To prepare pure xylan from wheat straw hemicellulose, the obtained hemicellulosic powders were treated using 0.05 mol/L HCl at a 1 : 30 solid to liquid ratio at 50°C for 2 h. The obtained solution was neutralized to pH 5.5 and concentrated under a reduced pressure and then precipitated in 3 volumes of ethanol. Pure xylan powders were finally obtained for further use. The prepared xylan contained 93.0% xylose and 5.0% arabinose (related to the total sugar content), which was determined using gas chromatography.

2.3. Preparation of Xylan/PMAA Semi-IPN Hydrogels. Various amounts of xylan, MAA, and Bis were dissolved in distilled water, and the total concentration of both xylan and MAA was 20 wt.%. Next, 1 wt.% of APS and anhydrous sodium sulphite that related to the amount of methacrylic acid were added. After mixing, the mixture was sealed and placed in a water bath at 60°C for 2 h without stirring. After the gelation was achieved, the hydrogels were removed and cut into uniform sized pieces, and the cut samples were soaked in distilled water for 24 h. During this period, it was necessary to change the water regularly for washing away the unreacted monomer and initiator. Finally, the hydrogels were vacuum-dried in an oven at 60°C for 24 h. The feed composition for hydrogel preparation is presented in Table 1.

Table 1: Feed compositions for the preparation of xylan/PMAA semi-IPN hydrogels.

Component	Sample code			
	PMAA	Semi-IPN2	Semi-IPN5	Semi-IPN7
MAA (g)	2.0	1.8	1.5	1.3
Xylan (g)	0	0.2	0.5	0.7
Bis (g)	0.040	0.036	0.030	0.024

2.4. Characterizations. The dried hydrogel samples were analyzed using Fourier transform infrared spectroscopy within the frequency range of 4000–400 cm^{-1} after these samples were milled and mixed with KBr to press discs. The morphology of the hydrogels was observed using scanning electron microscope after swollen hydrogel samples were freeze-dried at −50°C and coated with gold.

2.5. pH Sensitivity and Swelling Reversibility. pH sensitivity of the prepared hydrogels was determined gravimetrically by measuring the swelling extent of the hydrogels in buffer solutions of different pH (2.0–10.0) value at room temperature. The ionic strengths of buffer solutions were adjusted to 0.05 M using NaCl. To ensure complete swelling, samples were allowed to swell for 48 h. The swollen hydrogels were weighed after the excess surface water was removed using a filter paper. The equilibrium swelling ratio was calculated using the following equation:

$$S_e\left(\frac{g}{g}\right) = \frac{(W_e - W_d)}{W_d}, \tag{1}$$

where $W_e(g)$ is the weight of the swollen hydrogel at equilibrium state; $W_d(g)$ is the weight of the dry hydrogel.

The hydrogels swollen in buffer solution of pH = 7.4 were placed into the pH 2.0 buffer solution. The hydrogels were removed and weighed after certain time intervals. The reswelling behaviors of the shrinking hydrogels were studied in pH 7.4 buffer solution. At predetermined time interval, the hydrogels were weighed after blotting the excess water on the surface of the hydrogels. The swelling and deswelling tests were carried out repeatedly with a period of 48 h. The swelling ratio at time t was defined from the following equation:

$$S_t\left(\frac{g}{g}\right) = \frac{(W_t - W_d)}{W_d}, \tag{2}$$

where W_t is the weight of the hydrogel at swelling time t and W_d is defined as stated above.

2.6. Dynamic Swelling Kinetics. The known mass of hydrogel samples was placed in 100 mL buffer solutions at room temperature. After certain time interval, the hydrogels were removed and weighed after removing the excess water on the surface. The swelling ratio at time t was calculated by (2).

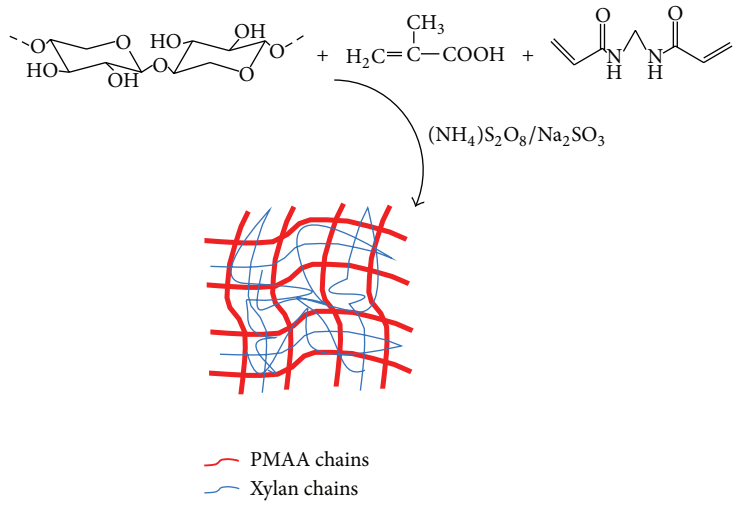

Figure 1: The proposed mechanism for the synthesis of xylan/PMAA semi-IPN hydrogel.

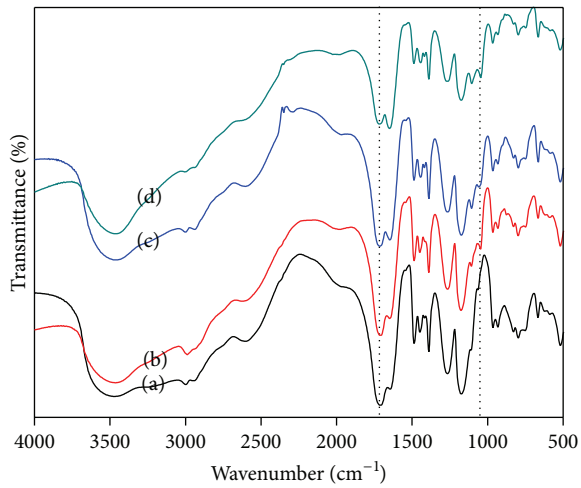

Figure 2: FT-IR spectra of PMAA (a), semi-IPN2 (b), semi-IPN5 (c), and semi-IPN7 (d).

3. Results and Discussion

3.1. Preparation of Xylan/PMAA Semi-IPN Hydrogels. The proposed mechanism for the synthesis of xylan/PMAA semi-IPN hydrogels is shown in Figure 1. The redox initiator system, APS-anhydrous sodium sulphite, generated sulfate radical anion ($SO_4^{\bullet -}$) from APS, which initiated the polymerization of MAA, and poly(methacrylic acid) (PMAA) would be cross-linked by cross-linker Bis to form the three-dimensional hydrogel. Xylan was incorporated into the three-dimensional network of PMAA hydrogel mainly with hydrogel bonds formed through the hydroxyl groups and carbonyl groups on xylan and PMAA chains. Xylan/PMAA semi-IPN hydrogel was obtained as above.

3.2. FT-IR Analysis of Xylan/PMAA Semi-INP Hydrogels. FT-IR spectra of semi-IPN2, semi-IPN5, semi-IPN7, and PMAA hydrogels are shown in Figure 2. The two absorption bands at 3430 and 2930 cm^{-1} were assigned to the stretching of O-H and the stretching of C-H, respectively. The absorption bands at 1710 cm^{-1}, 1390 cm^{-1}, and 965 cm^{-1} were the characteristic absorptions of -COOH groups, which are attributed to the C=O stretching, the C-O stretching vibration, and the –OH in-plane vibration of carboxyl groups [27, 28]. The band at 1487 cm^{-1} was assigned to the bending vibration of C-H of methyl groups [29]. The characteristic absorption peak for typical xylan was located at 1044 cm^{-1}, which was attributed to the C-O, C-C stretching, and the glycosidic linkage ν(C-O-C) contributions [30]. The sharp absorption peak at 894 cm^{-1} was assigned to C1 group frequency of vibration and frequency of vibration of the ring, which was the characteristic absorption of the glycosidic bond between sugar units [30]. More importantly, the intensity of the peak at 1044 cm^{-1} was enhanced and that of the peak at 1710 cm^{-1} declined with an increase of the content of xylan.

3.3. Morphology Analysis of Xylan/PMAA Semi-IPN Hydrogels. The morphology of the hydrogels prepared using different feed compositions is shown in Figure 3. The morphology of PMAA hydrogel presented a porous structure, which is typical morphology of PMAA-based hydrogel. The porosity of the xylan/PMAA semi-IPN hydrogels evidently increased and the pore wall became thin, and this resulted in the formation of the interconnected porous channel. The interconnected porous channels provide paths for the diffusion of water molecules, which is important to improve the sensitivity and swelling ratio of the hydrogel. The porous structure of the xylan/PMAA semi-IPN hydrogel would extend its application in some fields, such as wastewater treatment and the drug release control.

3.4. pH Sensitivity of Xylan/PMAA Semi-IPN Hydrogels. The effect of pH on the equilibrium swelling ratio is presented in Figure 4. The equilibrium swelling ratio increased at first and then decreased when the pH value increased. This is primarily due to -COOH groups in hydrogel, and the dissociation of the

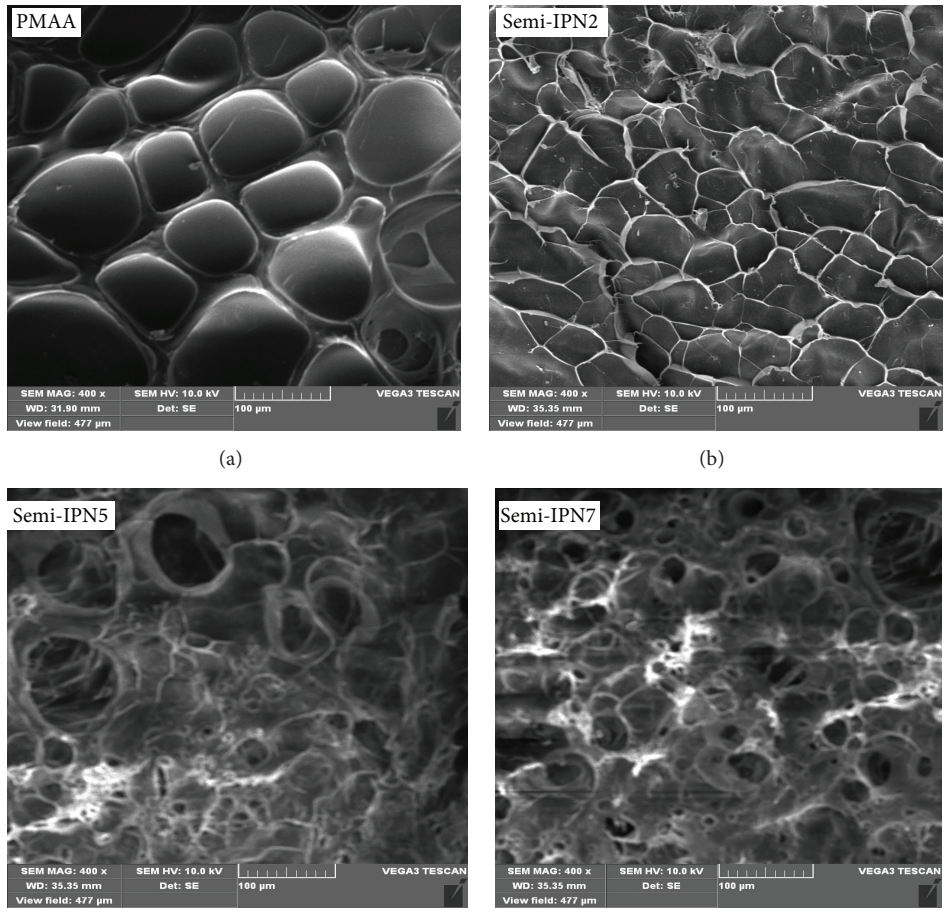

Figure 3: SEM images of PMAA (a), semi-IPN2 (b), semi-IPN5 (c), and semi-IPN7 (d).

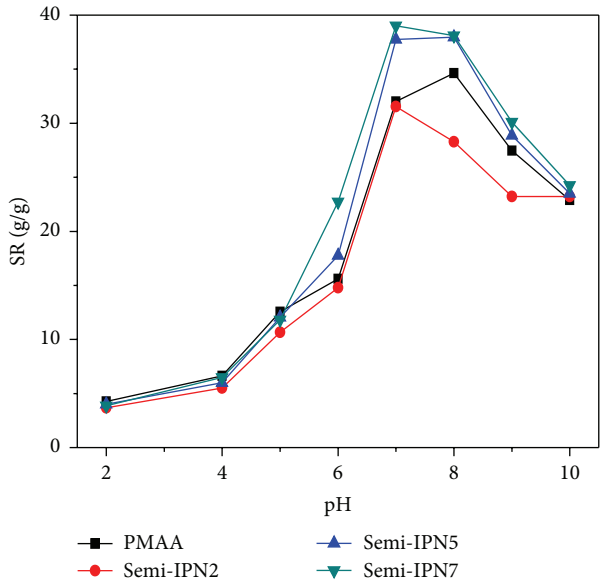

Figure 4: The swelling ratio of the hydrogels in different pH value buffer solutions.

-COOH group did not occur in a strongly acidic condition (pH < pK_a), and there were strong hydrogen bonds present within the hydrogels. When pH value is higher than pK_a of the carboxyl group (4.28), the -COOH group changed into $-COO^-$ group which enhanced the electrostatic repulsion forces in the hydrogels network; therefore, the swelling ratio increased. But the interactions between the carboxylic acid anions and cations occurred with an increase of pH value, resulting in a slight decrease of the swelling ratio [16].

pH sensitivity is closely related to the morphology of the swollen hydrogels. To clearly analyze the sensitivity of the xylan/PMAA hydrogels, the surface morphology of the semi-IPN7 was observed, and the SEM micrographs are shown in Figure 5. As shown in Figure 5(a), the surface of the semi-IPN7 swollen in pH 2.0 buffer solutions presented a little porous structure and the interconnected porous channels could not be observed, and this observation could be attributed to the hydrogen bond's effect in acidic solution. Meanwhile Figures 5(b) and 5(c) showed obviously the porous structure and the interconnected porous channels, which may be due to the higher swelling ratio in neutral and alkaline solutions. The porous size of the hydrogel sample

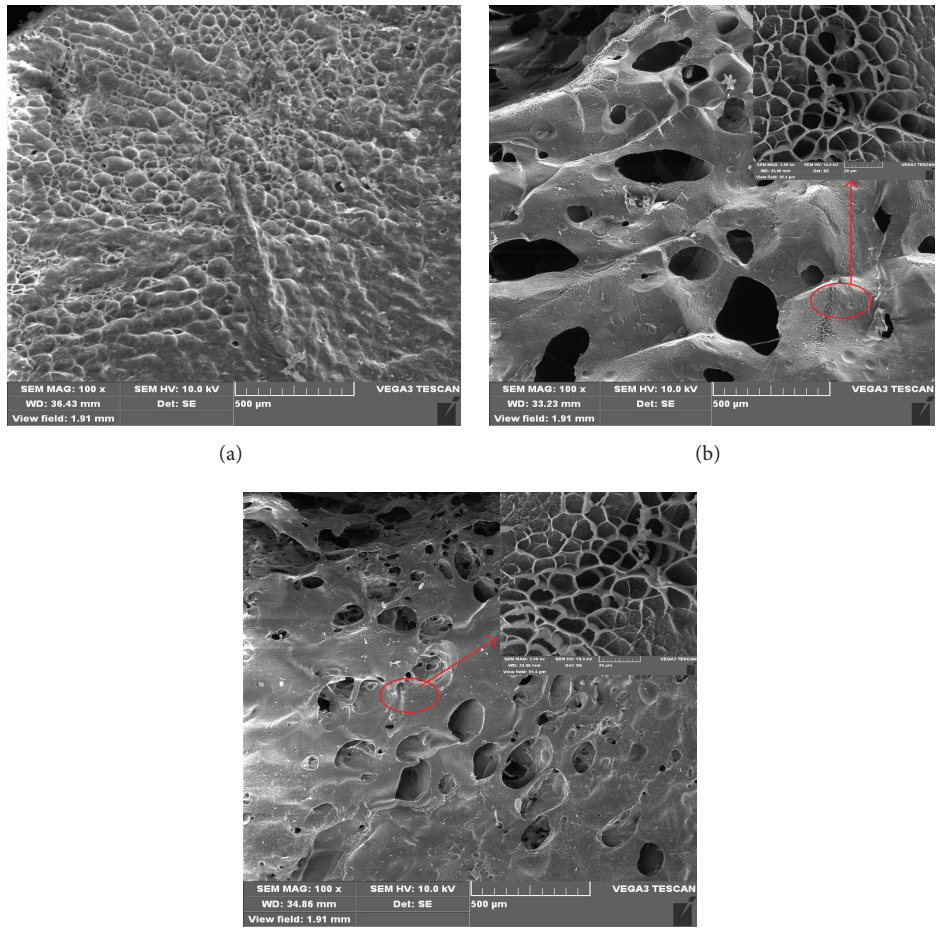

Figure 5: SEM images of the semi-IPN7 swollen in pH 2.0 (a), pH 7.4 (b), and pH 10.0 (c) buffer solutions.

swollen in pH 10.0 solutions was smaller than that of the hydrogel sample swollen in pH 7.4 solutions, and this could be attributed to the change of the swelling ratio when the pH value was increased, and the swelling behavior of the hydrogel in alkali solution was restrained because of the electrostatic screen effect. In addition, the pore density of the hydrogel sample swollen in pH 2.0 and pH 7.4 buffer solutions was less than that of the hydrogel sample swollen in pH 10.0 buffer solution. To analyze this phenomenon, the nonporous region of the hydrogel sample in Figures 5(b) and 5(c) was magnified by 2000 times, and the wealth of porous structure was observed. Because -COOH groups would dissociate into $-COO^-$ in a higher pH (pH > pK_a) solution, which could generate the electrostatic attraction forces with cations and reduce the repulsion force in hydrogel network, the porous size of the hydrogel sample decreased. Through the analysis of the morphology of the hydrogel, it is found that the trend of the pore size and the swelling degree change with pH change was the same.

3.5. Swelling Reversibility of Xylan/PMAA Semi-IPN Hydrogels. Figure 6 presents the swelling reversibility of the

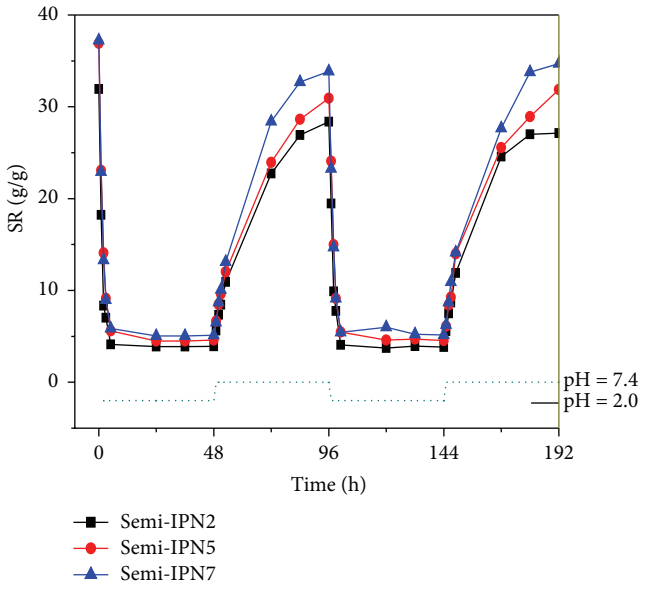

Figure 6: Swelling reversibility of the xylan/PMAA semi-IPN hydrogels in pH 2.0 and pH 7.4 buffer solutions.

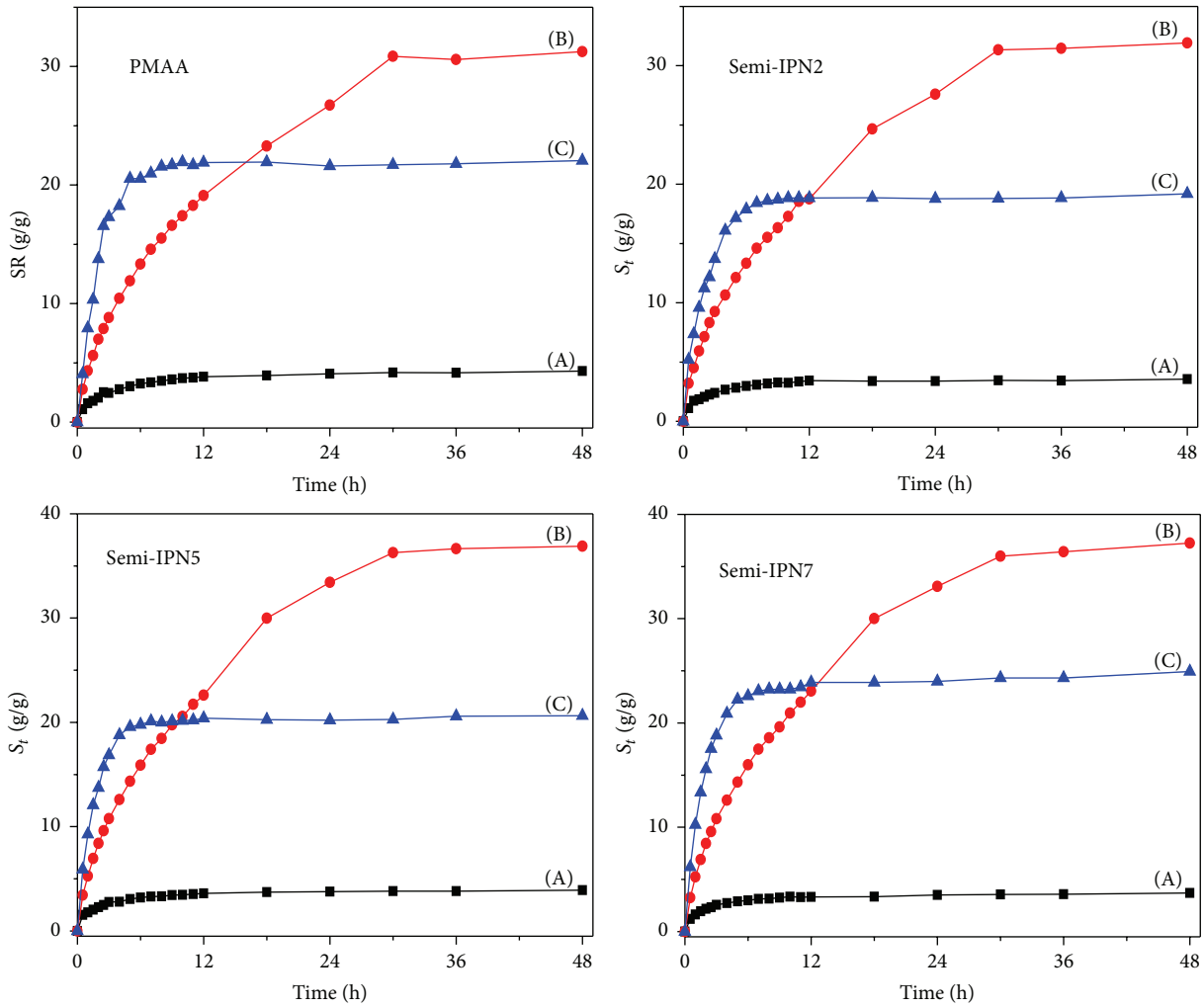

Figure 7: Time-dependent swelling ratio of the hydrogels in buffer solutions of pH 2.0 (A), 7.4 (B), and 10 (C).

xylan/PMAA semi-IPN hydrogels in buffer solutions of pH 2.0 and pH 7.4. The hydrogel samples swollen in pH 7.4 buffer solution were placed into pH 2.0 buffer solution and then transferred into the pH 7.4 buffer solution. Samples rapidly shrank in pH 7.4 buffer solution and reached the equilibrium. But the duration of the swelling was longer than the deswelling duration in pH 7.4 buffer solution. The swelling-deswelling studies revealed that the swelling-deswelling behaviors of the xylan/PMAA semi-IPN hydrogels had excellent reversibility.

3.6. Dynamic Swelling Kinetics of Xylan/PMAA Hydrogels. The ionization degree of -COOH groups is different in various pH solutions, and this could result in the difference of hydrogen bond's effect in hydrogels, and hydrogels would have different swelling process in various buffer solutions. Figure 7 presents the time-dependent swelling ratio of the PMAA hydrogel and xylan/PMAA semi-IPN hydrogel in buffer solutions of pH 2.0, 7.4, and 10.0. As shown in Figure 7, the hydrogels prepared with different components exhibited similar dynamic swelling curves and swelling behavior on pH sensitivity. In the initial swelling process, the water adsorption rate of the hydrogels increased with a rise of pH value of buffer solutions, and this is related to the morphology of the hydrogels in buffer solution. The hydrogels swollen in pH 7.4 buffer solution had the maximum swelling ratio and needed the longest time for achieving swelling equilibrium. The water adsorption rate of the hydrogels swollen in pH 10.0 buffer solution was obviously higher than that of the hydrogels swollen in pH 2.0 and 7.4 solutions. According to the Donnan equilibrium theory, the swelling behavior of the hydrogel is due to the difference of the free-ion concentration osmotic pressure in inside and outside of the hydrogel. When the pH value of the buffer solution was high, the concentration of -COO^- groups in the hydrogels increased, and this resulted in a higher osmotic pressure and electrostatic repulsion forces in the hydrogel; therefore, it led to faster swelling rate of the hydrogels. However, the electrostatic repulsion forces decreased because of the swelling of the hydrogels, which is attributed to the charge screening effect by cations in buffer solution, and the hydrogels rapidly absorbed water in pH 10.0 buffer media and achieved the swelling equilibrium.

Table 2: Kinetic parameters of the PMAA hydrogel and xylan/PMAA semi-IPN hydrogels in buffer solutions of pH 2.0, 7.4, and 10.

Sample	pH 2.0		pH 7.4		pH 10.0	
	n	R^2	n	R^2	n	R^2
PMAA	0.471	0.976	0.610	0.998	0.903	0.994
Semi-IPN2	0.438	0.922	0.568	0.997	0.561	0.995
Semi-IPN5	0.279	0.963	0.549	0.998	0.649	0.999
Semi-IPN7	0.425	0.999	0.609	0.996	0.702	0.998

Because of the excellent swelling property, the xylan/PMAA semi-IPN hydrogels are expected to be a good carrier for colon-specific delivery. As the pH values in the human stomach and the colon are 2.0 and 7.0, respectively [31], the hydrogels, as drug carriers for colon-specific delivery, not only reduce the drug release in the gastric juice but also have high release efficiency and control release properties in the intestinal fluid.

The studies on the dynamic swelling kinetics of the hydrogels are important for their applications, especially for the drug release. To clearly understand the swelling mechanism of the hydrogels, the exponential heuristic equation was used to study the diffusion mechanism of water molecule in hydrogel network. It can be described as the following equation [32]:

$$F = \frac{W}{W_\infty} = kt^n. \quad (3)$$

Equation (3) can be deformed into the following equation:

$$\ln \frac{W}{W_\infty} = \ln k + n \ln t, \quad (4)$$

where F is the fractional uptake at time t (min); k is constant incorporating characteristics of the polymer network and the solvent, whereas n is the diffusion exponent which is indicative of the transport mechanism. The value of n indicates the swelling mechanism of the hydrogels: the swelling process follows Fick's law when n is less than 0.5; when $0.5 < n < 1$, it belongs to non-Fick's law, which is the result of the combined effects of solvent diffusion and relaxation of macromolecular chains; the swelling process is mainly decided by the relaxation of macromolecular chains when n is greater than 1 [3, 4, 32].

The diffusion exponent n obtained from the profile of $\ln F$-$\ln t$ is listed in Table 2, where n indicated the slope of curve. As can be seen from Table 2, the exponent n trended to increase with a rise of pH value, and Fickian behavior was observed in pH 2.0 buffer solution, and the swelling behavior in pH 7.4 and 10.0 buffer solutions followed non-Fickian behavior. The data of diffusion exponent n of the xylan/PMAA semi-IPN hydrogels was lower than that of the PMAA hydrogels. As drug carriers for colon-specific delivery, the lower n value is an advantage for the hydrogels, which not only reduces the drug release in the gastric juice but also could achieve the controlled release for drug in the intestinal fluid. These results indicated that the xylan/PMAA semi-IPN hydrogels can be considered a potential carrier for colon-specific and sustained drug release applications.

4. Conclusions

The xylan/PMAA semi-IPN hydrogels were fabricated successfully from xylan and methacrylic acid using N,N-methylenebisacrylamide as cross-linker. The SEM images demonstrated the existence of a porous honeycomb-like structure in the hydrogels. The swelling kinetic experiments showed that Fickian behavior was observed in pH 2.0 buffer solution. The swelling behavior of hydrogels in pH 7.4 and 10.0 buffer solutions followed non-Fickian behavior which demonstrated that the water transport was controlled by hydrogel relaxation and water diffusion in pH 7.4 and 10.0 buffer solutions.

Xylan plays a significant role in the morphology and swelling ratio of the prepared hydrogels, and the interconnected porous channels increased when the content of xylan was increased. The studies on pH sensitivity and dynamic swelling kinetics of hydrogels revealed that the semi-IPN xylan/PMAA hydrogels may serve as potential carriers for colon-specific and sustained drug release applications.

Competing Interests

The authors declare that they have no competing interests.

Acknowledgments

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