



DETERMINATION OF RESWELLING PROPERTIES AND WATER DIFFUSION MECHANISM OF HYDROGEL COMPOSITES

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Highlights

- Hydrogel composites were prepared by free radical polymerization technique
- The composites had higher equilibrium water content in water and at different pHs
- Water diffusion of the composites was governed by less Fickian diffusion mechanism



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ABSTRACT: This study focused on acrylamide/*N*-vinyl-2-pyrrolidone chemically cross-linked hydrogel composites. As fillers, sepiolite and alkyl ammonium salt modified sepiolite were used in the preparation of the hydrogel composites. Characterization of the hydrogel composites was carried out with X-ray diffraction (XRD), Fourier transform infrared spectroscopy (FTIR) and scanning electron microscopy (SEM) analyses. Swelling of the hydrogel composites as a function of time was investigated with tea-bag method. Reswelling ability of the hydrogel composites was revealed after the three cycles. In the third swelling test, the swelling percentage of the NVP hydrogel was about 1690%. The swelling percentage of the NVP+MSP hydrogel composite increased from approximately 1610% to 1760% after the three repeated swelling tests. The hydrogel composite including modified sepiolite showed higher equilibrium water content (EWC) in the distilled water and at different pHs compared to other samples. The highest EWC value was obtained for the hydrogel composite including modified sepiolite (0.9637) in alkali conditions. Water diffusion mechanism of the hydrogel composites was examined based on the Fickian diffusion index (*n*). *n* values of the hydrogel composites were lower than 0.5 which is indication of water diffusion governed by less Fickian diffusion mechanism. The results showed that hydrogel composites can be used in various applications required the reswelling ability and high EWC value.

Keywords: *Alkyl Ammonium Salt, Hydrogel, Reswelling Ability, Sepiolite, Water Diffusion Mechanism*

1. INTRODUCTION

Hydrogels are defined as 3D polymer networks which uptake a large quantity of water or various biological fluids [1]. The main advantages of hydrogels such as high water uptake, responsiveness to external factors (light, pH, temperature, electrical and magnetic field *etc.*), biocompatibility and biodegradability make hydrogels smart materials [2, 3]. Due to their desired properties, hydrogels are utilized in drug delivery, tissue engineering, contact lenses, hygiene materials, electrochemical applications, separation processes, food industry and agriculture [4, 5].

In general, hydrogels are synthesized with chain growth polymerization or step growth polymerization. Owing to hydrophilic segments of hydrogels, cross-linking step is required to prevent the hydrophilic segment dissolution in an aqueous medium. Hydrophilic polymers exhibit Newtonian behavior in the absence of cross-linker. When cross-linkers are used, hydrogels exhibit visco-elastic or non-Newtonian behavior [6]. Based on cross-linking methods, hydrogels are separated into: chemically and physically cross-linked hydrogels. Chemically cross-linking is conducted through chemical reactions of complementary groups, high energy radiation, free radical polymerization and cross-linking with enzymes. On the other hand, hydrogen bonding, chain entanglements, π - π stacking, metal-ligand coordination, ionic interactions, hydrophobic interactions, crystallization, Van der Waals forces and protein interactions are effective for physically cross-linking [7, 8].

Swelling is one of the most important properties for hydrogel applications. The swelling process takes place in three stages: water diffusion into hydrogel structure, polymer chain loosening and hydrogel expansion, respectively [9]. Swelling ratio of hydrogels generally changes depending on monomer type and concentration, cross-linking degree, external stimuli, solvent type and concentration,

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initiator content as well as surface functionalization of hydrogels [10, 11]. For example, low amount of cross-linker can hinder formation of hydrogel network effectively which adversely affects water uptake. Using high amount of cross-linker can result in high cross-linking degree, so leading limited water diffusion into hydrogel structure [12]. Moreover, type of monomer including hydrophilic or hydrophobic groups alters swelling properties of hydrogels. Hydrophilic groups have synergistic effects on the swelling ratio of hydrogels. However, hydrophobic groups prevent interactions between water and hydrogel network that decreases swelling capacity [13].

To enhance swelling properties, incorporation of fillers such as metals, metal oxides, polymers, clays and carbonaceous materials is a promising strategy [14]. Clays (kaolin, vermiculite, montmorillonite, bentonite, sepiolite, and so on) are widely used as filler owing to their abundance, low-cost, hydrophilic structure and easiness of preparation [15]. In case of preparation of hydrogel composites, hydrophobic structure of polymers can limit homogeneous dispersion of clays. So, clay modification is generally applied using alkyl ammonium salts such as tetramethyl ammonium bromide, hexadecyl ammonium chloride and octadecyl trimethyl ammonium bromide to provide uniform dispersion of clays in polymer network [16, 17].

This study focused on acrylamide/*N*-vinyl-2-pyrrolidone chemically cross-linked hydrogel composites. Reswelling in water and equilibrium water content of the hydrogel composites were determined based on data collected by tea-bag method. Water diffusion mechanism of the hydrogel composites was investigated with Fick's law of diffusion. Effects of presence of sepiolite in the hydrogel network and sepiolite modification with an alkyl ammonium salt on the swelling/reswelling properties, equilibrium water content and water diffusion were studied in detail. Characterization of the hydrogel composites was carried out X-ray diffraction (XRD), Fourier transform infrared spectroscopy (FTIR) and scanning electron microscopy (SEM) analyses. When compared to literature studies related to hydrogel composites including clays, significant advantages of the hydrogel composites prepared in this study were discussed.

2. MATERIAL AND METHODS

2.1. Materials

Acrylamide (Merck) and *N*-vinyl-2-pyrrolidone (Sigma-Aldrich) as a monomer were used. Ammonium persulfate (Sigma-Aldrich) was utilized as reaction initiator. Ethylene glycol dimethacrylate (Sigma-Aldrich) and *N,N,N',N'*-tetramethylethylenediamine (Sigma-Aldrich) were used as cross-linker and accelerator, respectively. Sepiolite with the molecular formula of $Mg_8Si_{12}O_{30}(OH)_4(H_2O)_{4.8}H_2O$ was evaluated as a filler. Tetramethyl ammonium chloride (Merck) was used to modify sepiolite. Distilled water was used for synthesis and swelling studies.

2.2. Hydrogel Synthesis

Free radical polymerization technique was used for hydrogel synthesis [18]. Acrylamide solution and *N*-vinyl-2-pyrrolidone were mixed with the mole ratio of 1:1. Cross-linker (0.05 mL) was added to monomer solution under the reflux. After the addition of 0.01 g initiator and 100 μ L accelerator into the solution, gel was obtained at 50 °C. Removal of impurities from the gel was provided with water washing at room temperature and the gel was dried at 50 °C until reaching to constant mass. The hydrogel was called as *N*-vinyl-2-pyrrolidone (NVP) hydrogel.

2.3. Preparation of Hydrogel Composites

Following to the cross-linker mixing with monomer solution, sepiolite with 1 wt% was added to the solution. After the homogeneous dispersion of sepiolite, initiator and accelerator was added to suspension. Gel formation was observed at 50 °C, and then similar washing and drying were applied to

prepare hydrogel composite as in the hydrogel synthesis. The hydrogel composite was called as *N*-vinyl-2-pyrrolidone + sepiolite (NVP+SP) hydrogel composite.

Hydrogel composite was also prepared by using modified sepiolite as mentioned above. For sepiolite modification, sepiolite was dispersed in distilled water (1:5, w:v) and tetramethyl ammonium chloride solution was added to suspension. The suspension was stirred for 24 h at room temperature. Modified sepiolite was washed with distilled water until removal of Cl⁻ ions which was controlled with 0.1 N AgNO₃ solution. The modified sepiolite was dried at 60 °C. The hydrogel composite was called as *N*-vinyl-2-pyrrolidone + modified sepiolite (NVP+MSP) hydrogel composite.

2.4. Characterization of Samples

XRD analyses were performed by Bruker D8 Advance equipment (Cu-K α radiation, λ : 1.54 Å). FTIR analyses were conducted on Bruker Vertex 70 spectrometer in the range of 4000-1000 cm⁻¹. SEM analyses were carried out by SM Zeiss LS-10 device.

2.5. Swelling and Diffusion Studies of Samples

To examine water uptake of the samples through tea-bag method, the samples were put in distilled water at room temperature. The samples were taken out from the water and wiped excess water with a tissue paper. The mass of the samples were recorded and swelling percentage (*S*, %) was determined by the following equation [19]:

$$S = \frac{m_1 - m_0}{m_0} \times 100 \quad (1)$$

where m_1 and m_0 indicate mass of the samples at time t and initial, respectively.

Reswelling behaviour of the samples was investigated in distilled water with repeating swelling experiment three times at room temperature. After each swelling test, the swollen samples were dried at 50 °C. The dried samples were used again for the next swelling test.

Equilibrium water content (EWC) of the samples in distilled water and at different pHs was determined by the following equation [20]:

$$EWC = \frac{m_1 - m_0}{m_1} \quad (2)$$

Water diffusion mechanism of the samples in distilled water and at different pHs was explained based on the following equation [21]:

$$F = \frac{m_1}{m_\infty} = kt^n \quad (3)$$

where m_∞ shows mass of the sample at equilibrium point. k is swelling rate constant and n is diffusion index which is used to determine type of the water diffusion. This equation can be applied in case of $F < 0.6$ [22].

3. RESULTS AND DISCUSSION

Figure 1 shows XRD pattern of the samples. With the incorporation of 1 wt% sepiolite or 1 wt% modified sepiolite, no significant change was specified XRD pattern of the samples. A common broad diffraction peaks at about $2\theta = 13^\circ$ and 22° in the XRD pattern of the samples indicated that the samples were in the amorphous state [23].

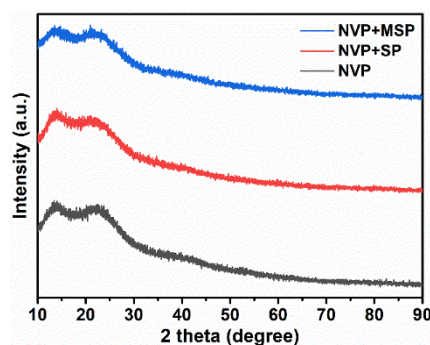


Figure 1. XRD pattern of the samples

Figure 2 shows FTIR spectrum of the samples. The band related to stretching of -OH was determined at the centered of 3400 cm^{-1} . The peak at 3166 cm^{-1} was attributed to N-H stretching vibrations. The peaks at 2927 cm^{-1} and 2880 cm^{-1} were contributed to asymmetric and symmetric stretching vibrations of C-H, respectively. Formation of C=O groups between acrylamide and *N*-vinyl-2-pyrrolidone resulted in appearance of the peak at 1670 cm^{-1} . The peaks at 1460 cm^{-1} and 1280 cm^{-1} were assigned to C-H deformation of CH_2 groups and C-N stretching, respectively [24]. The peak at 1172 cm^{-1} was originated from C-C stretching of *N*-vinyl-2-pyrrolidone ring [25].

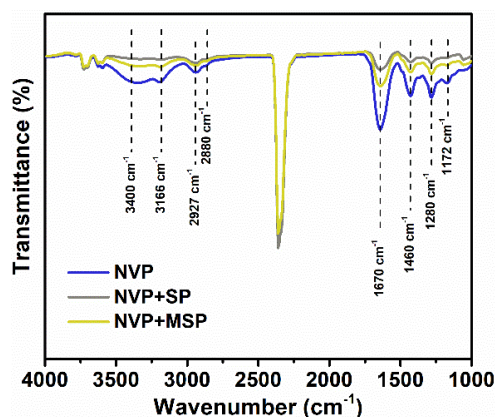


Figure 2. FTIR spectrum of the samples

Figure 3 shows morphological structure of the samples. In the SEM image of the NVP hydrogel (Figure 3a), regular layers which are effective in water diffusion were observed. While using unmodified sepiolite, uneven surface belonged to the NVP+SP hydrogel composite was obtained (Figure 3b). And also, formation of the agglomerated structure was determined that has generally adverse effects on the water uptake of the materials [26, 27]. Whereas, homogeneous dispersed NVP+MSP hydrogel composite including regular layers was prepared with the incorporation of modified sepiolite into the hydrogel network as shown in Figure 3c. It is known that interlayer distance between the clay plates increase in case of mixing with monomer molecules providing the formation of layered structure that makes diffusion of water molecules easy into the hydrogel network [28].

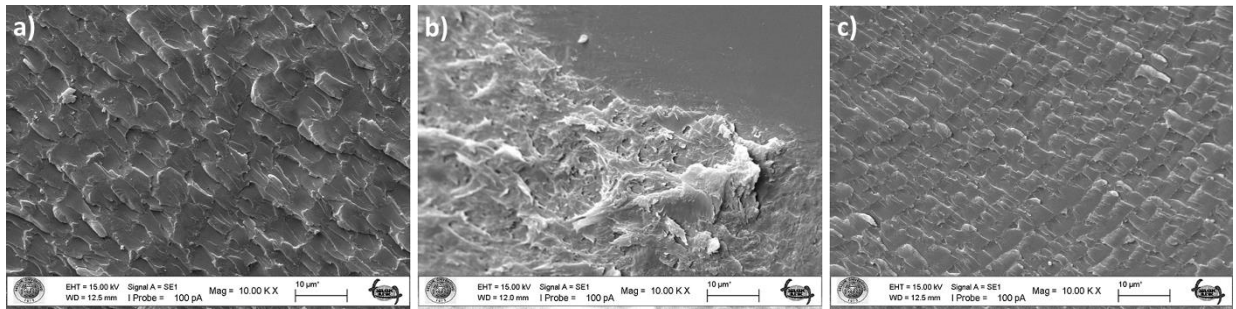


Figure 3. SEM image of the a) NVP hydrogel, b) NVP+SP hydrogel composite and c) NVP+MSP composite hydrogel

Figure 4a shows swelling percent of the samples in time. In the beginning, swelling percentage of the samples rapidly increased, and then swelling rate slowed down. In case of using neat sepiolite, a slight decrease in the swelling percentage of the NVP hydrogel was observed which was attributed to nonhomogeneous dispersion of the sepiolite in the hydrogel structure. However, the swelling percentage of the NVP hydrogel increased with the incorporation of modified sepiolite. By the help of modification, separation of layers of the sepiolite as well as uniform dispersion of the sepiolite had synergistic effects on the water uptake of the NVP+MSP hydrogel composite.

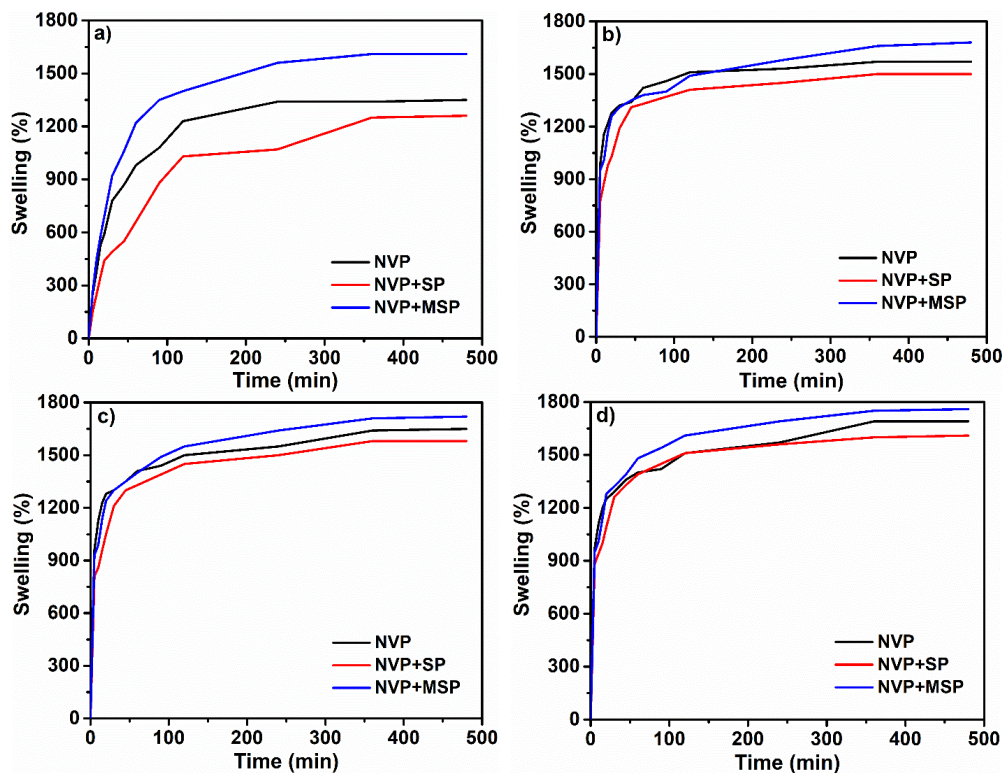


Figure 4. a) Original swelling, b) first reswelling, c) second reswelling and d) third reswelling of the samples

Figure 4b-d show ability of the samples to swell in the water over several cycles. It was clear that all the samples exhibited similar reswelling behaviour in the water. Original swelling percentage of the NVP hydrogel was determined as about 1350%. In the third swelling test, the swelling percentage of the NVP hydrogel was about 1690%. The swelling percentage of the NVP+MSP hydrogel composite increased from approximately 1610% to 1760% after the three repeated swelling tests. The results revealed that the swelling percentage of the samples was higher than that of their original swelling percentage based on the repeated swelling tests. No defect in the hydrogel network such as crack and

degradation was observed. So, it was obvious that the samples can be used for many times in various applications including reswelling ability.

EWC of the samples is given in Table 1. The samples had considerably high EWC which is associated with good biocompatibility of the materials in general [29]. EWC of the NVP hydrogel and NVP+SP hydrogel composite was about 0.95 in distilled water and at different pHs. Due to the utilization of modified sepiolite, NVP+MSP hydrogel composite exhibited higher EWC compared to other samples. At low pH, EWC of the NVP+MSP hydrogel composite was lower than that assigned to packed form of polymer chains due to hydrogen bonds between $-\text{COOH}$ groups [30]. Ionization of $-\text{COOH}$ groups into $-\text{COO}^-$ ions with increasing pH leads to repulsion forces between polymer chains that contribute higher swelling capacity [31]. The lowest EWC of the NVP+MSP hydrogel composite was 0.9561 at pH 2, while the highest EWC was specified as 0.9637 at pH 9 in this study.

Table 1. EWC of the samples

Samples	Distilled water	pH 2	pH 7	pH 9
NVP	0.9547	0.9545	0.9559	0.9539
NVP+SP	0.9523	0.9519	0.9555	0.9523
NVP+MSP	0.9609	0.9561	0.9593	0.9637

Figure 5 shows $\ln F$ vs. $\ln t$ curves of the samples. Water diffusion mechanism is generally specified based on the slope of the curves (n value). Depending on n value, diffusion mechanism is classified as less Fickian diffusion, Fickian diffusion and non-Fickian diffusion. While n value equals to 0.5, water diffusion is rate limiting [32]. In case of $n < 0.5$, less Fickian diffusion takes place which means that water diffusion rate is lower than relaxation rate of the polymer chains [33]. Non-Fickian diffusion is observed when $0.5 < n < 1.0$ that is indication of dominance of water diffusion rate or comparable relaxation and diffusion rate [34]. In this study, n values were lower than 0.5 which is evidence of less Fickian diffusion.

4. CONCLUSIONS

This work was related to reswelling properties and water diffusion mechanism of the hydrogel composites. Hydrogel composites were prepared by using sepiolite and alkyl ammonium salt modified sepiolite by free radical polymerization technique. Amorphous structure of the hydrogel composites was observed by XRD analyses. Characteristic peaks in the FTIR spectrum of the hydrogel composites were indication of successful preparation of the samples. Dispersion of the filler in the polymer chains was investigated by SEM analyses. Uniform dispersion of the modified sepiolite in the hydrogel network was specified in the SEM image in contrast to sepiolite.

Ability of the hydrogel composites to swell in the water over several cycles was examined. The swelling percentage of the hydrogel composites was higher than that of their original swelling percentage, after the repeated swelling tests without crack or degradation. Based on the EWC of the samples, the lowest swelling percentage belonged to NVP+SP hydrogel. However, NVP+MSP hydrogel composite exhibited higher swelling percentage by the help of regular layered structure. The highest EWC of the NVP+MSP hydrogel composite was determined at pH 9 due to the ionization of $-\text{COOH}$ groups. n values lower than 0.5 obtained from $\ln F$ vs. $\ln t$ curves revealed that water diffusion mechanism of the hydrogel composites can be explained with lower water diffusion rate than relaxation rate of the polymer chains.

The results showed that hydrogel composite especially including the modified sepiolite can be used in various applications required the reswelling ability and high EWC value.

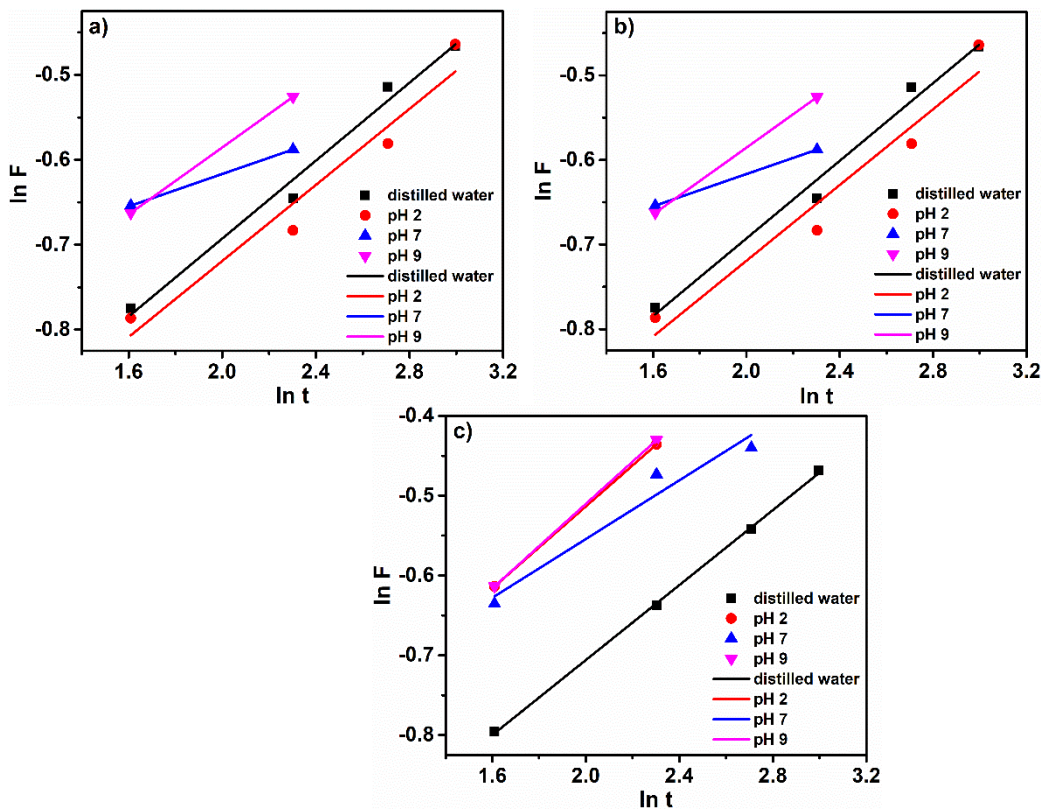


Figure 5. $\ln F$ vs. $\ln t$ curve of a) NVP hydrogel, b) NVP+SP hydrogel composite and c) NVP+MSP hydrogel composite

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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