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Transparent and semi-interpenetrating network P(vinyl alcohol)- P(Acrylic acid) hydrogels: pH responsive and electroactive application

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ABSTRACT

In this paper, poly(vinyl alcohol)-poly(acrylic acid) based transparent semi-interpenetrating network (semi-IPN) hydrogels were synthesized by using a solvent mixture of dimethyl sulfoxide and deionized water via free radical polymerization and subsequent freeze-thaw technique. The formation of the semi-IPN hydrogels was confirmed from FT-IR spectra. The acrylic acid concentration effect on the hydrogels was investigated in terms of transparency, crystalline structure and thermal stability by using UV-visible spectroscopy, X-ray diffraction and thermogravimetric analysis. Swelling behaviours of the semi-IPNPAP hydrogels were studied in deionized water and different pH solutions. The compression and electroactive behaviour was tested in fully hydrated stage by using compression test and by applying electrical voltage. The hydrogels showed displacements under the applying voltage and detailed experiment is illustrated.

Introduction

Every year, there is a considerable usage of hydrogels in numerous technological fields such as biomedical, drug delivery, tissue engineering and artificial muscles because of their structural and specific properties. Most of the hydrogels are prepared either by physical or chemical crosslinking techniques. Semi-interpenetrating network (semi-IPN) hydrogels are much attractive due to their content of the crosslinking techniques referred above. Generally, semi-IPN hydrogels are special class of hydrogels and the semi-IPN hydrogels are formed by at least polymerized/crosslinked one component in the presence of another material which means the first material is in free network state and act as primary network structure when the second one polymerized/crosslinked in the presence of primary network, to form semi-IPN hydrogels [1,2]. Usually, this hydrogel is prepared via blending, free radical, gamma radiation and UV-radiation [3–5]. Among the polymerized material, different type of materials which are used with acrylic acid were reported [6–9]. Based on the materials, polyvinyl alcohol (PVA) has been chosen as the primary polymer matrix in the preparation of transparent semi-IPN hydrogels.

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PVA is a water-soluble and semicrystalline synthetic polymer material, which has been studied broadly in technological fields due to its chemical stability, biodegradability, biocompatibility and eco-friendly nature \[10\]. Its various properties make it suitable for a variety of applications in industrial fields and more over PVA is used for contact lens due to excellent transparency \[11–13\]. Acrylic acid (AA) is a vinyl group of organic compound widely used in hydrogels preparation mainly in superabsorbent hydrogels. Since poly (acrylic acid) (PAA) polymers have negative charge in their structure due to the loss of their protons in aqueous solution, they shows pH, electrical and temperature-response \[14\]. Both PVA and PAA polymers have an excellent hydrogel-forming ability. In addition, they are easily compatible with each other to form semi-IPN hydrogels when they polymerized/blended together. Knowing this fact, the combination of hydrogels having the above properties, several methods are available for the preparation of PVA-PAA based semi-IPN hydrogels \[6,15–17\]. Recently, Lu et al. reported PVA/PAA hydrogels prepared for electrode-neural tissue applications and electrical responsive hydrogels in an electrolyte solution were reported \[15,17–19\]. However, there is no report on the synthesis of transparent semi-IPNPAP hydrogels yet. Transparency is very important for electroactive lens application.

This article reports the synthesis of semi-IPN PVA-crosslinked PAA (semi-IPNPAP) hydrogels using DMSO and water solvent mixture via free radical and subsequent freeze-thaw technique. AA is polymerized via free radical in the presence of PVA polymer to form a semi-IPNPAP hydrogel. Here, the PVA polymer can be a primary polymeric network and polymerized PAA polymer acts as a secondary polymeric network. In these two polymeric networks, a strong intermolecular bond can be formed between the hydroxyl groups of PVA and the carboxyl groups of PAA. Figure 1 shows the schematic formation of crosslinking/bond in the semi-IPNPAP hydrogel. The effect of different concentration of AA on the electroactive behaviour is studied. Recently, researchers reported transparent PVA hydrogels could be obtained by using the above solvent mixture (DMSO: H\(_2\)O) for optical lens applications \[20,21\]. The formation of the semi-IPNPAP hydrogels was confirmed from the Fourier transform infrared spectroscopy (FTIR), thermogravimetric analysis (TGA) and X-ray diffraction studies. Optical transparency of semi-IPNPAP hydrogels was measured using UV-vis spectroscopy and compression test and swelling studies were conducted. Actuation test of the synthesized semi-IPNPAP hydrogels was carried out by applying actuation voltage.

![Figure 1. The formation of semi-IPNPAP hydrogel.](image-url)
Materials and preparation method

Materials

Polyvinyl alcohol (PVA = 85,000–124,000 g mole⁻¹, 99% hydrolysed), acrylic acid (AA), N,N1-methylene bis-acrylamide (MBA), Potassium persulphate (KPS), N,N,N1,N1-tetramethylethylenediamine (TMEDA) were purchased from Sigma-Aldrich. Dimethyl sulfoxide was purchased from Dae Jung chemicals and metals Co. Ltd and used without any further purification to prepare the semi-IPNPAP hydrogels. Deionized (DI) water was used throughout the experiment.

Synthesis of semi-ipnpap hydrogels

Initially, an aqueous PVA solution (10 wt.%) was prepared under nitrogen atmosphere by dissolving PVA in a solvent mixture (4:1 ratio of Dimethyl sulfoxide: DI water) and stirred by using magnetic stirrer at 70 °C for 6 h. The particular amount of AA was dissolved in 4 mL of the solvent mixture. To this solution, 1 mL of MBA and 1 mL of KPS/1 mL of TEMDA were added and stirred at 60 °C using a magnetic stirrer a period of 30 min. The solution was then added to 15 g of 10 wt.% PVA solution and stirred at 70 °C for 60 min. The homogeneous mixture was poured into a petri dish and freeze dried at −20 °C for 12 h. The freeze-dried sample was brought to room temperature for 6 h and the process was repeated three times. After completion of the three cycles, a transparent semi-IPNPAP hydrogel was formed. The formed hydrogel was carefully immersed into a beaker containing 100 mL of DI water and the DI water was changed for every 6 h for 3 days in order to remove unreacted materials such as monomer, crosslinker, solvent and unreacted materials [22]. In a similar procedure, various concentrations of AA based hydrogels were prepared. Table 1 illustrates the various concentrations of PAP hydrogels.

Characterizations

Fourier transform infrared spectroscopy (FTIR) spectrophotometer was used to study the formation of the Semi-IPNPAP hydrogels. Before the analysis of FTIR spectra, the samples were completely dried in the oven at 60 °C for 6 h and the spectra recorded on a FTIR spectroscopy (Bruker Optics Inc.) using KBr pellet method in the spectral range of 4000–400 cm⁻¹. The optical transparency of the semi-IPNPAP hydrogels was measured by using a UV-visible spectroscopy (HP8452A, Agilent). The hydrogels were cut into the desired shape for the analysis of transparency and the spectra were recorded from 200–800 nm. The thermal behaviour of the PVA and semi-IPNPAP

| Hydrogel code | 10 wt% PVA (g) | AA (mM) | MBA (mM) | KPS (mM) | TMEDA (mM) | Swelling ratio (g/g) | Displacement (µm) |
|---------------|---------------|---------|----------|----------|------------|----------------------|------------------|
| PVA           | 15            | −−      | −−       | −−       | −−         | 3.697                | 4.5              |
| Semi-IPNPAP1  | 15            | 6.939   | 0.648    | 1.849    | 0.861      | 4.755                | 15.49            |
| Semi-IPNPAP2  | 15            | 13.877  | 0.648    | 1.849    | 0.861      | 5.2029               | 12.25            |
| Semi-IPNPAP3  | 15            | 20.816  | 0.648    | 1.849    | 0.861      | 5.2404               | 11.05            |
hydrogels were studied by a TGA (STA 409 PC, NETZSCH), at a constant heating rate of 10 °C/min under a constant nitrogen flow (20mL/min). The X-ray diffraction (XRD) studies of the PVA and semi-IPNPAP hydrogels were carried out on an X-ray diffractometer (Rotafl ex RU-200B, Rigaku) using Cu Ka. The diffraction data was collected from 5 to 70 °C with 300 mA and 40 kV, respectively. The compression mechanical test was conducted according to ASTM D-882–97 using a universal testing machine. The desired shape of the sample was fully hydrated in DI water for 24 h and kept between two parallel plates; the hydrated sample was compressed with upper plate until it reached a fixed value. The test was performed at room temperature and at the compressing rate of 0.0005 mm/s.

The electrical actuation test was carried out by using a high voltage amplifier connected to a function generator (33220A, Agilent) in an environmental chamber. Figure 2 shows the schematic of actuation test. A laser displacement sensor (LK-G85, Keyance) was used to measure the displacement along with a data acquisition system (Pulse, B&K) and Labview software on a personal laptop controls the data acquisition. Before conducting the actuation test, the desired shape of hydrogels (10 x 10 x 4 mm³) was equilibrated in DI water for 24 h. After reaching an equilibration, the hydrated hydrogels were kept in between two electrodes (polyimide tape attached on ITO glass) then an electric field was applied between two electrodes.

Results and discussion

Swelling and ph swelling studies

The swelling studies play a significant role in hydrogels technology and it was calculated using an analytical method [22]. For pre-weighed dry hydrogels were immersed in DI water at room temperature until to reach equilibrium. The swollen hydrogels were
weighed after removing the excess water with a moister observed tissue paper. The equilibrium swelling ratio was calculated using the following equation [23].

\[
\text{Eq. swelling ratio } (S_{g/g}) = \frac{(W_s - W_d)}{W_d}
\]

(1)

Here, \( W_d \) and \( W_s \) correspond to the weight of dry and swollen states of the hydrogels. The swelling experiment was carried out three times to minimize errors.

Figure 3(a) shows the swelling ratio of the PVA and semi IPNPAP hydrogels with different concentration of AA. The swelling ratio of the synthesized hydrogels depends on the concentration of AA. It can be seen that the swelling ratio of the semi-IPNPAP hydrogels increased with increase in the AA concentration and reached maximum swelling ratio of 5.24 in the semi-IPNPAP3 hydrogel, respectively. This behaviour is attributed due to the increase of hydrophilic nature of the AA in the semi-IPNPAP hydrogels and it was improving the three-dimensional network structure of the hydrogel, leading to improve the affinity of the water molecules, thus resulting increased the swelling ratio. Furthermore, the swelling studies were carried out in different pH solutions and these results signify the pH sensitivity of the semi-IPNPA hydrogels prepared. For this purpose, different pH solutions (2 to 12) were prepared and adjusted by 0.1 M

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**Figure 3.** Swelling behaviour of the PVA and semi-IPNPAP hydrogels (a) DI water, (b) Different pH solutions and (c) FTIR spectra of the PVA and semi-IPNPAP hydrogels.
HCl/0.1 M NaOH solutions and determine using a pH meter (Orion Star A211 pH Benchtop Meter, Thermo Scientific) [24,25]. Figure 3(b) shows the pH swelling studies of the PVA and semi-IPNPAP hydrogels with various concentration of AA. From the Figure 3(b) the pH swelling ratio of the hydrogels prepared increased noticeably from pH 2 to 12, it was observed in all the hydrogels with different swelling ratio. Overall, the maximum and minimum swelling ratio was observed at pH 12 and 2. For example, the swelling ratio of semi-IPNPAP3 at 24 h were 5.43 and 6.46 for pH 2 and 12, respectively. This behaviour occurred due to the ionization and protonation of the carboxylic functional groups (-COOH) present in the semi-IPNPAP3 hydrogels. The most of the carboxylic groups were protonated under the acidic medium (lower pH), that means hydrogen bond formed between the carboxylic groups in semi-IPNPAP3, which causes to the interactions between the polymer chains in the hydrogel network. Meanwhile, the electrostatic repulsion between the carboxylic groups was restricted and thus the diffusion of water molecules curtailed into the hydrogel network. So, the semi-IPNPAP3 hydrogel absorbed less water molecules and consequently shows the less swelling ratio value in acidic medium [25]. As increasing the pH, the hydrogen bonding interaction decreased in between the carboxylic groups due to the ionization effect and also electrostatic repulsion increased between these groups due to this both reasons the swelling ratio value higher in higher pH [26]. A similar trend was observed in semi-IPNPAP1, semi-IPNPAP2 and PVA hydrogel.

**Fourier transforms infrared spectroscopy**

Figure 3(c) shows the FTIR spectra of the PVA and semi-IPNPAP hydrogels. In the PVA hydrogel, the peak observed at 3436 cm$^{-1}$ corresponds to the stretching vibration of -OH group. The characteristic peaks at 1095 cm$^{-1}$ and 2925 cm$^{-1}$ are related to stretching vibrations of the C-O and C-H and the peak at 1647 cm$^{-1}$ is related to an acetyl group (C = O) of PVA [27]. The characteristic peaks at 917 cm$^{-1}$ and 854 cm$^{-1}$ are peaks that confirm the skeletal vibration of PVA and peaks at 1436 cm$^{-1}$ and 1382 cm$^{-1}$ correspond to deformation vibrations of C-H [28]. While the semi-IPNPAP hydrogels show all the above characteristic peaks with slight changes in frequency (3423 cm$^{-1}$ corresponding to – OH group, 1093 cm$^{-1}$ and 2920 cm$^{-1}$ related to stretching vibrations of the C-O, C-H, and the acetyl group (C = O) at 1636 cm$^{-1}$), an interesting peak was observed at 1714 cm$^{-1}$ in the semi-IPNPAP hydrogels. This is related to carbonyl groups of the AA [29]. Also, the peak intensities of semi-IPNPAP hydrogels lessened than the PVA hydrogel, which is due to the intermolecular bond formation between the hydroxyl group of the PVA and carboxylic group of the PAA.

**Optical transparency**

The semi-IPNPAP hydrogels were prepared by using a solvent mixture 4:1 ratio of DMSO: H$_2$O. Few, researchers revealed transparent PVA hydrogels which are prepared by using this solvent mixture [20,21]. Transparency of the synthesized hydrogels was measured by taking UV-vis spectroscopy in the swollen state and the spectral range from 400 nm to 800 nm. Transparency of the synthesized semi-IPNPAP hydrogels depends on the AA concentration. Figure 4(a) shows UV-vis spectra of the PVA and semi-IPNPAP hydrogels...
Figure 4(b) represents their photographs. Semi-IPNPAP hydrogels are quite transparent. It was observed that the transparency of the semi-IPNPAP hydrogels was decreased with increasing the AA concentration and also depending on the hydrogel thickness [20]. The transparency of the PVA hydrogel measured at 500 nm is shown to be 74 %, and those of semi-IPNPAP hydrogels are 62, 58 and 47 %, respectively depending on the AA concentration.

**X-ray diffraction study**

Figure 5 shows the XRD spectra of the PVA and semi-IPNPAP hydrogels. The PVA hydrogel shows sharp and greater intensity diffraction peaks at $2\theta = 19.58^\circ$, and a shoulder peak at $2\theta = 22.89^\circ$ [30,31]. It is known that PVA has a semi-crystalline nature and freeze-thaw technique increases the crystalline structure of PVA [32]. The XRD spectra of semi-IPNPAP hydrogels were different from that of the PVA hydrogel: with the increase of AA concentration, the diffraction peak corresponding to $2\theta = 19.58^\circ$ of the PVA hydrogel gradually decreased. This peak was slightly shifted from 19.58$^\circ$ to 19.17$^\circ$ and the shoulder peak (22.89$^\circ$) disappeared in the semi-IPNPAP hydrogels as increasing AA concentration. This means that the overall crystallinity of the semi-IPNPAP hydrogels is slightly lower than the crystallinity of the PVA hydrogel due to the formation of amorphous PAA hydrogel. The diffraction intensity of the semi-
IPNPAP hydrogels depends strongly on the concentration of AA and the diffraction intensity peak decreases with increasing the AA concentration.

**Thermogravimetric analysis**

The thermal stability of synthesized hydrogels were analysed by taking TGA. Figure 6(a) represents the TGA graphs of the PVA and semi-IPNPAP hydrogels. All hydrogels exhibit two degradation stages. A minor weight loss (0.5 wt.%) was observed around 103 °C which might be due to loss of absorbed water molecules, beginning at nearly 82.9 °C. However, above 170 °C a significant weight loss was observed in all hydrogels. The degradation temperature of the PVA hydrogel was found to be approximately 180 °C and the weight loss of up to 9 % was observed in the range of 190–290 °C. When the temperature increased, the weight loss rate rapidly decreased, and the maximum weight loss of 96.4 wt.% was observed at around 600 °C [33]. In the case of semi-IPNPAP hydrogels, 5% weight loss was observed in the range of 190–290 °C, and 95.0% weight loss was seen at 600 °C. The initial degradation temperature of the semi-IPNPAP hydrogels increased to 195 °C from 180 °C of the PVA hydrogel. This might be due to the strong intermolecular bond formation [34]. This can be conformed from derivative thermogravimetric analysis (DTA) spectra. Thus, DTA curves of the PVA and semi-IPNPAP hydrogels are shown in Figure 6(b-e). The PVA hydrogel shows the initial degradation temperature (IDT) around 185.3 °C. Obviously, this value was increased in the semi-IPNPAP hydrogels due to the formation of strong intermolecular bond. Also the first DTA peak was shifted to higher temperature with increasing the AA concentration. Because AA could easily consume the hydroxyl groups of PVA, it can form strong intermolecular bond, which was conformed from the FTIR and XRD spectra. Thus, AA can effectively inhibit the dehydration of hydroxyl groups and increase the thermal stability. The DTA values of semi-IPNPAP hydrogels were 194.9, 209.7 and 233.1 °C, respectively. Figure 6(f) shows the AA concentration effect on IDT of the semi-IPNPAP hydrogels.
IDT values increase with the AA concentration. The overall semi-IPNPAP hydrogels show that the thermal stability and IDT values are higher than that of the PVA hydrogel.

**Compression test**

Mechanical properties of the PVA and semi-IPNPAP hydrogels with various concentration of AA were studied by using the universal testing machine and the
compressed samples were at hydrated states. Figure 7(a) Shows stress-strain curves of the hydrogels. The AA concentration has an effect on the mechanical properties of the semi-IPNPAP hydrogels. The compression modulus of the PVA hydrogel is 0.341 MPa. Contrarily, various AA concentrations of the semi-IPNPAP hydrogels exhibit lower Young’s modulus than the PVA hydrogel. This might be due to the PAA polymer chains formed in the presence of PVA polymer, which eventually form the semi-IPNPAP hydrogel. Usually, PVA hydrogel is formed with intra or inter molecular bonds between PVA chains. When the PAA polymerized in the presence of PVA, the carboxylic groups of the PAA react with the hydroxyl groups of the PVA so as to form strong inter molecular bond between these groups. Due to this reason, the semi-IPNPAP hydrogel shows lower Young’s modulus than PVA. When the AA concentration increased from 6.6939 mM to 20.816 m, the compression modulus decreased from 0.147 to 0.112 MPa, respectively (Figure 7(c)). In summary, Young’s modulus of the semi-IPNPAP hydrogels descend in the sequence of PVA>semi-IPNPAP1>semi-IPNPAP2>semi-IPNPAP3 (0.341,0.147,0.128 and 0.112MPa). This was conformed from the swelling studies shown in Figure 3(a). Increasing the AA concentration of the semi-IPNPAP hydrogels results in increase the swelling ratio.
**Electrical actuation test**

The electrical displacement test was conducted and the displacement output was measured with the laser displacement sensor. The influence of actuation voltage on the displacement behaviour of the semi-IPNPAP hydrogels was compared with the PVA hydrogel under the same experimental condition. Figure 7(c) shows the displacement output of the hydrogels as a function of actuation voltage at 0.1 Hz and Figure 7(c) does the displacement output with the actuation frequency at 1.6 kV. The displacement increases with increasing the voltage. The semi-IPNPAP hydrogels mostly showed higher displacement output at each actuation voltage and the displacement depends on the AA concentration. The semi-IPNPAP1 hydrogel shows higher displacement than the other semi-IPNPAP hydrogels, due to its lower Young's modulus, which means softer, as shown in Figure 7(a). Note that the maximum displacement output of 15.49 µm was achieved with the lower AA concentration case of semi-IPNPAP1 hydrogel under 1.6 kV/cm at 0.1 Hz. Figure 7(c) shows the frequency dependence of the displacement output of the hydrogels at 1.6 kV/cm. The displacement output decreases with increasing the frequency. The semi-IPNPAP hydrogels shows higher displacement output than the PVA hydrogel due to the negative charged ionic (COO-) groups present in the semi-IPNPAP hydrogels. In the semi-IPNPAP hydrogels, the AA plays a significant role in the displacement of semi-IPNPAP hydrogels because these hydrogels have negative charged ionic groups (COO-) within the hydrogel network [35–37]. Generally, these groups contained hydrogels show pH, temperature and electric filed sensitivity [15,29].

**Conclusions**

This paper reported the transparent semi-IPNPAP hydrogels, prepared with polyvinyl alcohol and poly(acrylic acid), by using the solvent mixture of dimethyl sulfoxide and DI water followed by free radical polymerization and subsequent freeze-thaw technique. This study provides a simple synthesis process of the semi-IPNPAP hydrogels. Thus, these hydrogels are more applicable where transparent hydrogels are required. The synthesized semi-IPNPAP hydrogels conformed from the FTIR spectra, revealing the formation of the intermolecular bond between the hydroxyl groups of the PVA and carboxylic groups of PAA. The AA concentration influenced the transparency and crystalline structure of the semi-IPNPAP hydrogels. Increasing the AA concentration resulted in decreased transparency and crystallinity of the hydrogels, whereas increased thermal stability and initial degradation temperatures of them. The swelling ratio and pH-responsive behaviour increase with increasing the AA concentration. The pH-responsive behaviour shows due to the formation of hydrogen bond between the functional groups, which led to decreased swelling ratio in low pH medium and increase in higher pH due to the ionic repulsion between the functional groups of PAA and PVA. The electrical actuation displacement increased with increasing the actuation voltage and higher displacement output was observed with lower AA concentration. The semi-IPNPAP hydrogels showed higher electrical displacement output than the PVA hydrogel. The synthesized hydrogels can be applied for biomimetic actuators, artificial muscles and electroactive reconfigurable lens.
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