

Cellulose/polyvinyl alcohol based hydrogels for reconfigurable lens

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ABSTRACT

Electroactive hydrogels are attractive for soft robotics and reconfigurable lens applications. Here we describe the design and fabrication of cellulose-poly vinyl alcohol based hydrogels. The fabricated hydrogels were confirmed by Fourier transformer spectroscopy, swelling studies, thermal analysis, surface morphology of fabricated hydrogel was study by using scanning electron microscopy. The effect of poly vinyl alcohol concentration on the optical and electrical behavior of hydrogels was studied.

Key words: Polyvinyl alcohol, Cellulose, Dimethylacetamide, Electroactive hydrogel.

1. INTRODUCTION

During the past few decades hydrogels have received great consideration in numerous applications such as biomedical, tissue engineering, drug delivery, bioactive electrode coatings actuators biomimetic field.^{1,2,3,4} Electroactive hydrogels are much interested in soft robotic applications due to soft and flexible properties and these materials exhibit bending/longitudinal deformation by applying low voltages.⁵ Generally hydrogels are hydrophilic three dimensional network structures which are formed either physical or chemical crosslinking by using the synthetic or natural materials.⁶ Electroactive hydrogels are prepared by adding various electrical nature materials, for example, conducting metal nanoparticles, electrically conductive polymer nanoparticles and electroactive polymers in the preparation of electroactive hydrogels network.^{7,8,9} Among them, we choose Poly vinyl alcohol (PVA) and cellulose for preparation of electroactive hydrogels. In general, these materials have electroactive nature.^{10,11}

Cellulose is one of the most abundant natural resource organic material in nature. Now a days, cellulosed based materials are considered for large number of applications due to the advantages, such as biodegradability, biocompatibility, low cost, mechanical, readily availability and chemical structure.^{12,13,14} Based on these properties and applications of cellulose, its derivatives and combination of other synthetic or natural materials are used for various fields. Recently cellulose with PVA based hydrogels were prepared science cellulose have large amount of hydroxyl groups and these groups can easily react with hydroxyl groups of PVA so as to form intermolecular hydrogen bond.¹⁵

The aim of this study is to enhance the deformation of PVA based electroactive hydrogels by physical blending PVA with cellulose. The fabricated hydrogels network structure was confirmed from FTIR, TGA, studies and the transparency was measured using UV-vis spectroscopy. The actuation performance test was carried out by using AC voltage.

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2. MATERIALS AND METHODS

2.1 Materials

Cotton pulp (MVE, DP-4580) was supplied by Buckeye Technologies Co. Ltd. (USA) and Poly vinyl alcohol (PVA = 89 000-98 000 g mole⁻¹, 99% hydrolyzed). Extra pure lithium chloride (LiCl) was carefully dried by using an oven at 110 °C, 2 h before use, N,N-dimethylacetamide (DMAc) were purchased from Sigma-Aldrich. The DI water was used throughout experiment.

2.2 Preparation of PVA-cellulose hydrogel

Cellulose/PVA (CP) hydrogels were prepared by blending method. Before blending cellulose and PVA solutions were prepared. Here a common solvent was used for both solution preparations. Preparation of the cellulose solution was described in our previous paper¹⁶ and here is brief explanation. The pure cotton pulp was dispersed in 8.5 % LiCl/DMAc solvent and stirred by using magnetic stirrer at room temperature over a period of 5 days after that completion of this process obtained 1.5 wt % of cellulose solution and also 1.5 wt% of PVA solution was obtained by above similar process at 60 °C. After getting the both solutions were mixed with different ratios 75/25, 50/50 and 25/75, stirred at room temperature until homogeneous mixture getting after that the solution was poured in Petridis and kept in oven at 30°C for a period of 3 days. After 3 days transparent hydrogel is formed and the formed hydrogel was immersed in 500 mL beaker containing 250 mL of DI water. The water was repeatedly changed every 6 h up to 5 days to remove LiCl/DMAc and unreacted materials from the hydrogel.

2.3 Characterization of PVA- Cellulose hydrogels

The FTIR spectra were used to study the formation of CP hydrogel. Before, analysis of the FTIR spectra the samples were completely dried in the oven at 60 °C for 6 h and the samples were examined between 500-4000 cm⁻¹ on Bruker Optics, Billerica, MA using the KBr pellet method. Transparency analysis of the CP hydrogels was performed by using UV-visible spectroscopy (HP8452A, Agilent). The hydrogels were cut into a desire shape for transparency measurement and the spectra recorded from 200-800 nm. The thermal behavior of the Cellulose and CP hydrogels were studied by thermogravimetric analysis (TGA) (NETZSCH, STA 409 PC), at a heating rate of 10 °C/min under a constant nitrogen flow (20mL/min).

2.4 Actuation test

The actuation test was carried out by using a laser displacement sensor (Keyence LK-G85, Tokyo, Japan) a high voltage amplifier connected to a function generator, (33220A, Agilent), Lab view software on a personal computer. Before conducting the actuation test the desired shape of the hydrogels (10 x 10 x 0.4 mm) were equilibrated in DI water for 24 h after reaching of equilibration, the swollen hydrogels were kept in between two electrodes (polyimide tape attached on ITO glass) and an electric field was applied between two electrodes. Displacement of the hydrogel was measure by the laser displacement sensor along with a data acquisition system (PULSE, B&K) as shown in Fig.1.

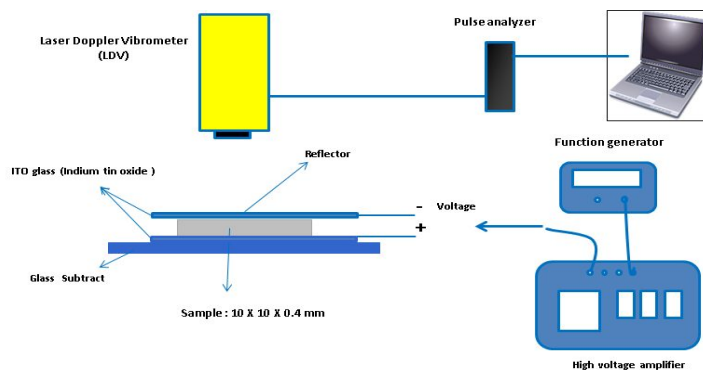


Figure 1. Schematic setup of actuation test.

3. RESULTS & DISCUSSION

The cellulose/PVA (CP hydrogel) hydrogels were developed by physical blending method and in this method PVA react with cellulose by intermolecular interaction so as to form transparent hydrogels. Fig. 2 shows the schematic formation of the hydrogels. The formation of the hydrogels are confirmed from FTIR spectra (Fig.3)

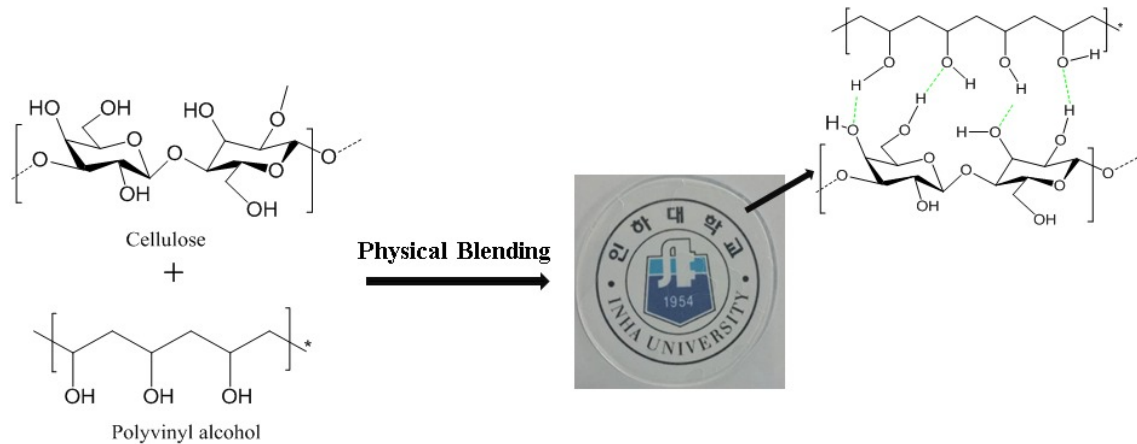


Figure 2. Schematic diagram of CP hydrogels formation

3.1 Fourier transform Infrared Spectroscopy

The FTIR spectra of the PVA, cellulose and CP hydrogels are showed in Fig.3. From Fig. 3, the CP hydrogels show the similar peaks of PVA, which demonstrates that the cellulose is uniformly dispersed in PVA. The characteristic peaks at 1098 cm^{-1} and 2923 cm^{-1} are related to stretching vibrations of the C-O and C-H. A broad absorption peak at 3438 cm^{-1} corresponds to the -OH stretching vibration of PVA. While these peaks were slightly shifted to 1098 cm^{-1} and 2923 cm^{-1} to 1063 cm^{-1} and 2917 cm^{-1} in CP hydrogel and also the -OH group was slightly shifted from 3438 cm^{-1} to 3425 cm^{-1} in CP hydrogel. The intensity of this peak was slightly decreased and appeared to be a bit broader than the -OH of the PVA. This is due to the strong intermolecular bond formation between the PVA and cellulose matrix.¹⁷ An interestingly absorption peak was absorbed at 899 cm^{-1} this the β -glucan group of the cellulose and also the fingerprint region peaks intensity increased due to the crystalline structure of the cellulose.^{18,19,20} The pure cellulose hydrogel shows the above characteristic peaks of the CP hydrogel peaks with slightly changes (3427 cm^{-1} , 2920 cm^{-1} , 1069 cm^{-1} and 896 cm^{-1}).

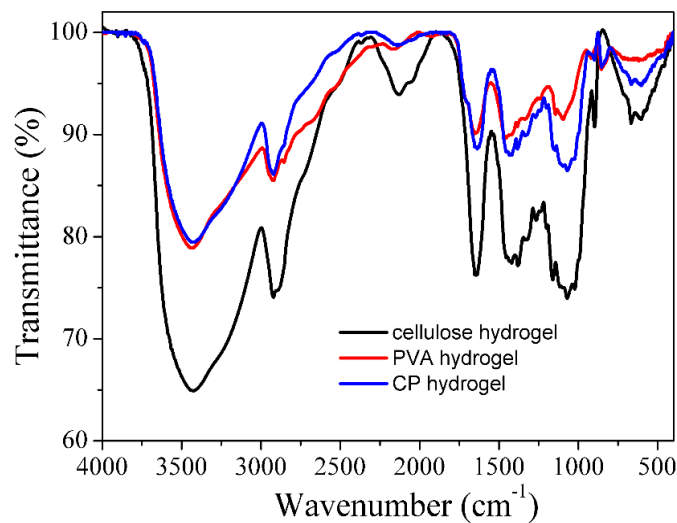


Figure 3. FTIR spectra of PVA, Cellulose and CP hydrogels

3.2 UV-Vis transmittance

The prepared CP hydrogel transparency was measured by using UV-vis spectroscopy. The transparency depends on the concentration of PVA in the prepared hydrogels. Figure 4 shows the transparency of the CP hydrogel. The transparency was decreased with increasing the PVA concentration in the hydrogel and also it depends on the thickness of the hydrogel. As increasing the thickness of the hydrogel the transmittance decreases. However the overall transparency was followed in the sequence of CP75<CP50<CP25<Cellulose hydrogel at 500 nm wavelength.

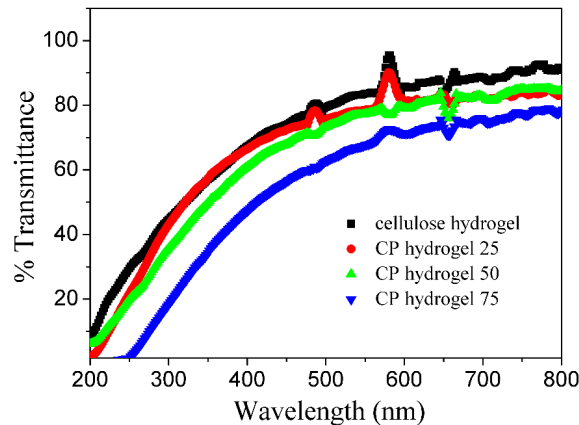


Figure 4. UV-vis spectra of Cellulose and CP hydrogels

3.3 Thermogravimetric analysis

The thermal stability of the PVA, cellulose and CP hydrogels were measured by using thermogravimetric (TGA) analysis. Figure 5 shows the thermal stability of the PVA, cellulose and CP hydrogels. All hydrogel curves exhibited mainly two degradation stages. A minor weight loss was observed below 200 °C due to the water molecules present in the samples. The PVA hydrogel shows the initial and final degradations at 310 °C and 408 °C and cellulose hydrogels shows the initial and final degradation at 264 °C and 329 °C. whereas the CP shows at 274 °C and 346 °C.²¹ The overall CP hydrogel shows the highest thermal stability than the PVA and cellulose hydrogel.

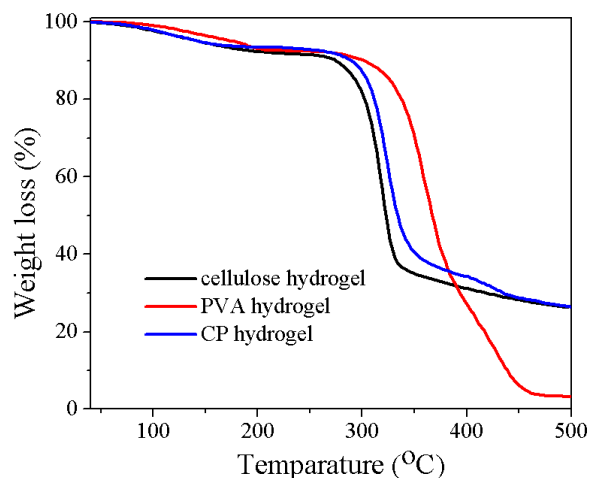


Figure 5. Thermogravimetric analysis of cellulose and CP hydrogels

3.4 Actuation test:

The CP hydrogels were prepared using physical blending method and the actuation test was performed. 4- 8 μm displacement output was achieved with 1000 V/cm at 0.1 Hz. Figure 6 shows the displacement of the physical hydrogels. Figure 6A shows the electric voltage dependent horizontal displacement, at 0.1 Hz. Here the displacement increased with increasing the voltage. Figure 6B shows the frequency dependent horizontal displacement at 1000 V. The displacement decreased with increasing the frequency.²²

Note that neat cellulose hydrogel shows higher displacement than the CP hydrogel due to the ion migration. Cellulose has some remnant ions caused by DMAC/LiCl solvent. When the PVA concentration increased in CP hydrogels the displacement decreased since intermolecular bond formation is increased, which results in weak ion injection.^{23,24}

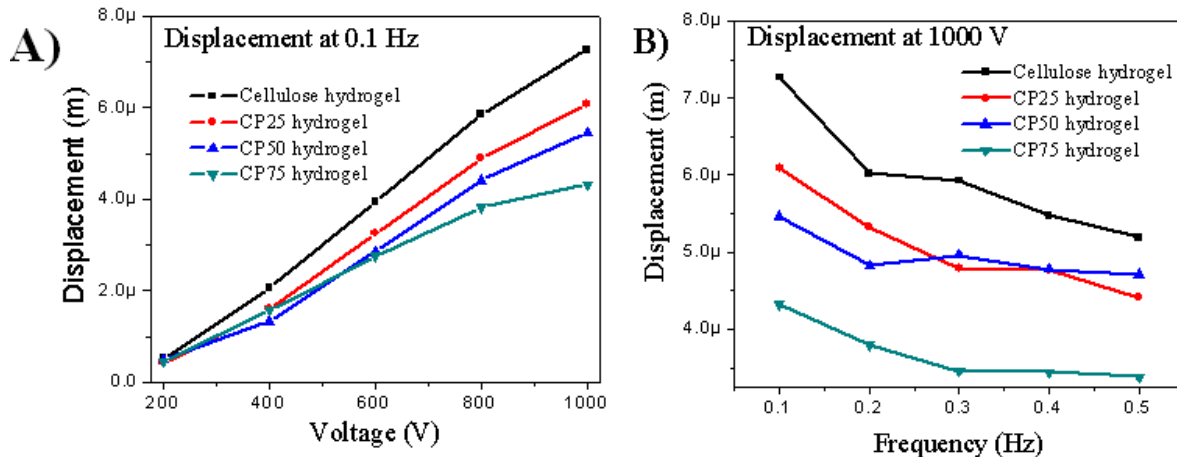


Figure 6. Actuation test of fabricated hydrogels: A) electric voltage dependent displacement at 0.1 Hz, B) Displacement output with frequency change.

4. CONCLUSION

The PVA-cellulose (CP) hydrogels were prepared by physical blending method. The formation of the CP hydrogels was conformed from FTIR, UV-vis. From the TGA analysis CP hydrogel showed highest thermal stability than PVA and cellulose hydrogels. Cellulose percentage influenced the displacement output of the fabricated CP hydrogels. The displacement increased with increasing the voltage and it increased by increasing the cellulose concentration.

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