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# Electroactive behavior of poly(acrylic acid) grafted poly(vinyl alcohol) samples, their synthesis using a Ce$^{(IV)}$ glucose redox system and their characterization

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## Abstract

Grafted copolymers of poly(acrylic acid) (PAA) and poly(vinyl alcohol) (PVA) were prepared using a Ce$^{(IV)}$ glucose redox initiator by free radical polymerization. Three grafted copolymers having 20%, 50% and 80% grafting were selected for this study. Thus-modified polymer was characterized by means of Fourier transform infrared spectra, $^1$H NMR, gel permeation chromatography, thermogravimetric analysis and universal testing machine approaches. The membranes were prepared by a solution casting method, where the cross-linking process was performed through the *in situ* addition of glutaraldehyde and hydrochloric acid as the cross-linking agent and catalyst respectively. The following four membranes were prepared: (i) pure PVA; (ii) 20% grafted PVA; (iii) 50% grafted PVA; (iv) 80% grafted PVA. The membranes obtained were employed in the electroactive behavior study under a DC electric stimulus in different concentrations of electrolyte. The equilibrium bending angles (EBA) of these polymers were studied with respect to time, poly(acrylic acid) content, electric voltage applied across the polymer and ionic strength of the electrolyte used. Experimental results show stable reversibility of the bending behavior of these polymers under an applied DC electric field. The EBA increased with increase in the applied electric voltage and poly(acrylic acid) content within the polymer.

(Some figures in this article are in colour only in the electronic version)

## 1. Introduction

Electroactive polymers (EAPs) have received much attention in recent years because they are considered to be smart materials and have variety of applications in artificial muscles [1], electrochemical devices [2], robotics [3], biomimetics [4], medical welfare instruments and separation techniques. Ionic hydrogels form one class of EAPs, which can change their volume and shape reversibly in response to external stimuli, such as temperature, pH, solvent, ionic strength and electric field [5–9]. These hydrogels do not dissolve in water at a physiological temperature and pH but swell considerably in an aqueous medium. This system can transform chemical free energy directly into mechanical work to give an isothermal energy conversion.

In 1880 Wilhelm Roentgen charged and discharged a rubber band, fixing its one end and attaching a mass to

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the other end, which led to the beginning of EAP [10]. The strain response to electric field activation was later formulated [11]. These beginnings were followed by the development of piezoelectric polymer (electret), which was prepared from carnauba wax, rosin and beeswax by solidifying and cooling them while subjecting them to a DC bias field [12]. More recently, half a century ago collagen filaments, which show the reversible behavior of contracting and expanding when dipped in acid and alkaline aqueous solutions, respectively, led to the discovery of environmentally sensitive or intelligent polymers [13]. To exploit such 'chemomechanical' actuators, researchers are developing synthetic polymers which can mimic biological muscles. In such efforts many polymers like poly(vinyl alcohol), poly(acrylic acid), poly(methacrylic acid), poly(*N*-isopropylacrylamide), polyaniline fibers, ionomeric polymer–metal composite (IPMC) and ion exchange membrane platinum composite (IMPC) are being studied.

In this study, grafted copolymers of PVA and PAA were prepared by free radical polymerization and their electroactivities were studied in NaCl electrolyte solutions under DC electric current. PVA is a nontoxic water-soluble synthetic polymer, which is widely used in biochemical and medical applications because of its compatibility with the living body [14]. PVA has good film forming and highly hydrophilic properties, and has been studied as a membrane in various applications. PAA is another important water-soluble polymer being studied as a hydrogel and pH sensitive material.

## 2. Experimental details

### 2.1. Materials

PVA was obtained from Kanto Chemicals Co. Inc., Japan. Ceric ammonium nitrate (CAN) was purchased from Sigma-Aldrich, glucose from Aldrich, acrylic acid monomer, glutaraldehyde (25% in water) from Junsei Chemical Co. Ltd, Japan. Hydrochloric acid and acetone were procured from Jin Chemical Pharmaceutical Co. Ltd, Korea. All the chemicals were used without any further purification. Double-distilled deionized water was used throughout this study.

### 2.2. Preparation of grafted copolymers

PAA-*g*-PVA was prepared by using $Ce^{(IV)}$ glucose redox initiator [15–17] and the reaction mechanism is displayed in figure 1. PVA was soaked in the required quantity of acrylic acid (AA) monomer for 30 min in a three-necked round bottom (RB) flask and then 100 ml of 0.1 N sulfuric acid was added to the flask. The RB flask was fitted with a water condenser, thermometer and needle for purging nitrogen gas. This RB flask was kept in a water bath maintaining a temperature of 45 °C, with constant stirring by a magnetic stirrer. In order to maintain an inert atmosphere nitrogen gas was purged into the reaction mixture continuously. Glucose was added as a reducing agent to the RB flask under constant stirring; after 15 min CAN was added to the reaction mixture and kept for stirring in a water bath maintaining a constant temperature of 45 °C. The reaction process was arrested by quenching in an ice cold water bath after 3 h. The acidic reaction mixture was neutralized with sodium hydroxide
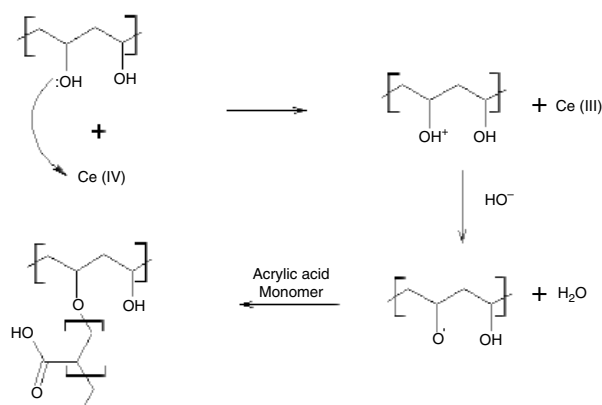
**Figure 1.** Reaction mechanism.

solution, checked by using pH paper. The reaction mixture was concentrated by evaporating the solvent water, by using the water bath. The mixture was cooled and acetone was added; the grafted polymer precipitates and the homopolymer dissolves in acetone. The grafted polymer was removed by filtration, washed with acetone repeatedly until a constant weight was obtained and dried in a vacuum oven at 45 °C. The percentage grafting was calculated by using the equation

$$\% \text{ grafting} = \left( \frac{W_f - W_i}{W_i} \right) \times 100 \tag{1}$$

where $W_f$ is final weight of the grafted copolymer and $W_i$ is the initial weight of the backbone polymer. Three grafted copolymers having percentages of grafting of 20, 50 and 80 were selected for this study.

### 2.3. Membrane preparation

Pure PVA and 20%, 50% and 80% grafted polymers (4 g each) were dissolved separately in 40 ml of distilled water at 60 °C with constant stirring under a slow stream of nitrogen gas; after complete hydration of polymer the solution was cooled to room temperature. To these polymer solutions, 100 $\mu$l glutaraldehyde and 200 $\mu$l HCl were added and the mixture stirred for 10 min to achieve an effective cross-linking of the copolymer. Films were cast on clean glass plates by uniformly pouring the polymer solutions under controlled humidity conditions. Membranes were dried in a vacuum oven at 45 °C for 24 h. The dried membranes were peeled off from the glass plate and washed with water repeatedly to remove excess glutaraldehyde and HCl and then allowed to dry at room temperature. The membranes obtained were denoted in this study as P-1, P-2, P-3 and P-4, prepared from pure PVA and 20%, 50% and 80% grafted copolymers respectively.

### 2.4. Swelling experiments

Swelling experiments were performed in different ionic strength NaCl electrolyte solutions at 25 ± 0.5 °C in an electronically controlled oven (Heraeus, Germany) by following the procedures published earlier [18]. Circularly cut (surface area = 5.04 cm²) disk-shaped membranes were kept

in a desiccator over anhydrous calcium chloride maintained at 25 °C for 48 h before use. The initial mass of membranes was measured using a single-pan digital microbalance (CAS, South Korea), sensitive to ±0.01 mg. Polymer samples were placed inside the airtight test bottles containing electrolyte solution. Test bottles were placed in an oven maintained at 25 °C. After 24 h (i.e., after complete attainment of equilibrium), membranes were removed and surface adhering solvent drops were removed by using soft filter papers; the samples were then weighed immediately. The degree of swelling (DS) was calculated by taking the ratio of equilibrium mass, $W_\infty$, to that of dry mass, $W_0$, of the membrane by using

$$\text{DS} = \frac{W_\infty}{W_0}. \qquad (2)$$

### 2.5. Fourier transform infrared (FTIR) spectra

Polymer samples were crushed to make KBr pellets under a hydraulic pressure of 600 kg cm$^{-2}$. FTIR (Midac Corporation, Model M-2000, USA) spectra were recorded in the wavelength region of 400–4000 cm$^{-1}$.

### 2.6. NMR measurement

Proton NMR measurements were performed on a Bruker AMX-500 spectrometer operating at frequency of 500.13 MHz. 10 mg of each sample, i.e. pure PVA and 50% grafted PVA, were dissolved in 2 ml of D$_2$O solvent; the sample was heated slightly in order to attain complete solubility of the polymer in the solvent and measurements were performed at 300 K.

### 2.7. GPC analysis

A model Waters: Alliance 2000 analytical GPC (gel permeation chromatograph) was used for the molecular weight determination. The analytical columns ultrahydrogel (2) and ultrahydrogel 500 (1) having dimensions 7.8 mm × 300 mm were used. Samples of 0.05 g ml$^{-1}$ were prepared in distilled water and 100 $\mu$l of this sample was injected into the column at an operating temperature of 318 K. A flow rate of 0.6 ml min$^{-1}$ was maintained and RI (the refractive index) was used as a detector.

### 2.8. Thermogravimetric analysis (TGA)

Dynamic thermogravimetric analyses were carried out with a thermogravimetric analyzer (Scinco, S-1500, South Korea). Analyses were performed with 10–12 mg samples on a platinum pan under nitrogen atmosphere with a normal gas flow rate of 40 ml min$^{-1}$ and heating rate of 10 °C min$^{-1}$ from 30 to 800 °C.

### 2.9. Mechanical properties

The tensile strength and ultimate elongation were measured for all the prepared membranes. Tensile tests were performed with a universal testing machine (UTM; Lloyd, UK) according to the test method ASTM D 412 by using a 5.0 N load cell with a cross-head speed of 100 mm min$^{-1}$. Averages of five tests for each polymer membrane sample were considered.
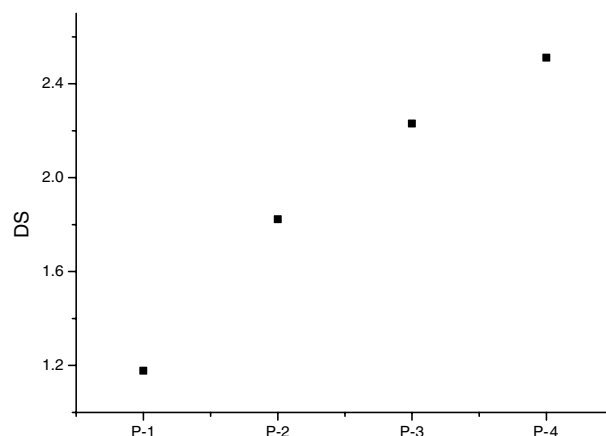


**Figure 2.** DS for all polymeric membranes in 0.1 N NaCl electrolyte solution at 7 pH.

### 2.10. Bending angle measurement under electric stimulus

Each sample was allowed to attain equilibrium swelling in the NaCl aqueous solution at room temperature before electrical stimulation. The same solution was taken in the glass case equipped with two parallel carbon electrodes fixed 30 mm apart at the center of the glass case. A fibrous specimen having dimensions of $15 \times 1 \times 1$ mm$^3$ was cut and placed vertically at the center of the two electrodes fixing one end of the specimen at the top. The DC voltage was applied across the solution between electrodes and the equilibrium bending angle was measured with a protractor [19–21].

## 3. Results and discussion

### 3.1. Characterization

Figure 2 displays the degree of swelling (DS) for all polymeric membranes in 0.1 N NaCl electrolyte solution at 7 pH. The degree of swelling is higher for grafted polymeric membranes (P-2, P-3 and P-4) than pure PVA (P-1) membrane. The pKa of PAA is 4.28 [7, 22]; below this value of the pH the –COOH groups of PAA associate and above this value the same groups dissociate. In the dissociation state of –COOH groups there will be higher interactions between these groups and water molecules which lead to greater swelling of the grafted polymer. As can be seen in figure 2, the DS for pure PVA is least of all and increases in the order P-2, P-3, P-4.

Figure 3 represents Fourier transform infrared spectra of all the polymeric membranes scanned in the infrared range 400–4000 cm$^{-1}$. The characteristic peak at 1730 cm$^{-1}$ is due to the presence of C=O stretching vibration and the characteristic peak observed at 650 cm$^{-1}$ is due to the O–H out-of-plane vibration of the carboxylic groups of PAA which appear only in the spectra of grafted polymers, i.e. P-2, P-3 and P-4, which confirms the grafting reaction. The characteristic peak at 1100 cm$^{-1}$ is due to the formation of the acetal ring and ether linkage as a result of the cross-linking reaction between hydroxyl groups of PVA and aldehydic groups of glutaraldehyde.

Pure PVA and 50% grafted copolymer samples were selected for the proton NMR analysis and the results for these
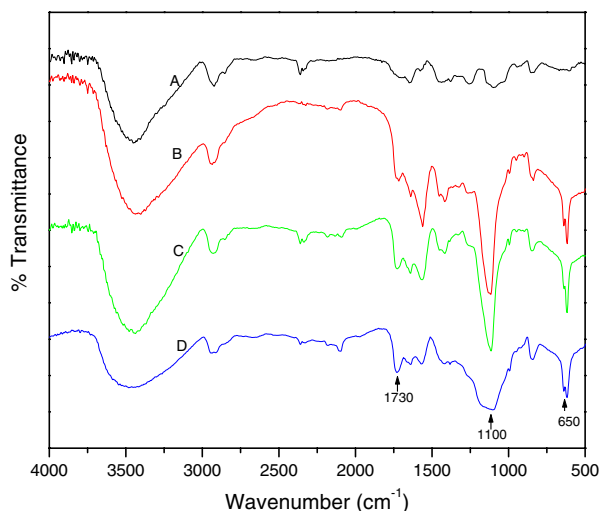
**Figure 3.** FTIR spectra of P-1 (A), P-2 (B), P-3 (C) and P-4 (D).

**Table 1.** Mechanical properties of all polymeric membranes.

| Polymer | Tensile strength MPa ± S.D. | Ultimate elongation ± S.D. |
|---------|------------------------------|-----------------------------|
| P-1 | 88.7 ± 0.4 | 86.3 ± 0.2 |
| P-2 | 63.0 ± 0.5 | 894.6 ± 0.5 |
| P-3 | 46.0 ± 0.3 | 929.0 ± 1.2 |
| P-4 | 36.7 ± 0.2 | 1290.6 ± 1.2 |

are displayed in figures 4(I) and (II) respectively. The solvent $D_2O$ shows a peak at around the chemical shift of 4.9 (d), which appears in both figures 4(I) and (II). The peaks (a) at the chemical shifts of 1.46–1.9 are due to the –CH$_2$– protons, the peak (b) at the chemical shift of 4.0 can be assigned to –CH– protons of PVA and peak (c) at the chemical shift of 2.2 is due to –CH$_3$ protons of the acetate group [23, 24]. The spectra of 50% grafted copolymer shown in figure 4(II) confirm the grafting of PAA on the backbone of PVA, as the results of GPC revealed that the molecular weight of the grafted polymer has increased with increase in percentage grafting; the peak (e) at 2.2 chemical shift has appeared, which may be due to the –CH protons of poly(acrylic acid).

GPC results are represented graphically in figure 5. It is clear that as the percentage grafting increases the weight average ($\bar{M}_w$) and number average ($\bar{M}_n$) molecular weights increase linearly from pure PVA to 50% grafted copolymer and the highest molecular weight is observed for 80% grafted copolymer; this may be due to the higher branches and longer chain lengths of PAA on the backbone of PVA in grafted copolymers.

Thermogravimetric analyses are displayed in figure 6. As polymers are hydrophilic in nature, 2–5% weight loss is observed at around 100 °C, which may be due to the absorbed moisture. Pure PVA has started decomposing at around 200 °C whereas grafted copolymers have shown still higher stability towards temperature. However, there is little increase in the decomposition temperature of 20% grafted copolymer observed when compared with pure PVA, which may be due to the lower grafting of the copolymer and as its chemical composition lies near to that of PVA. But
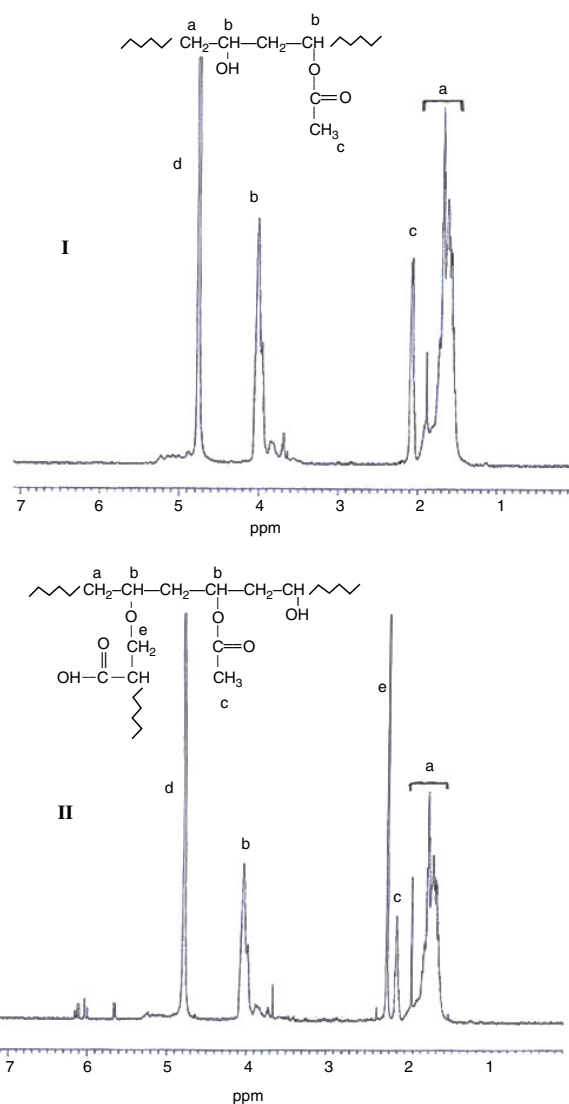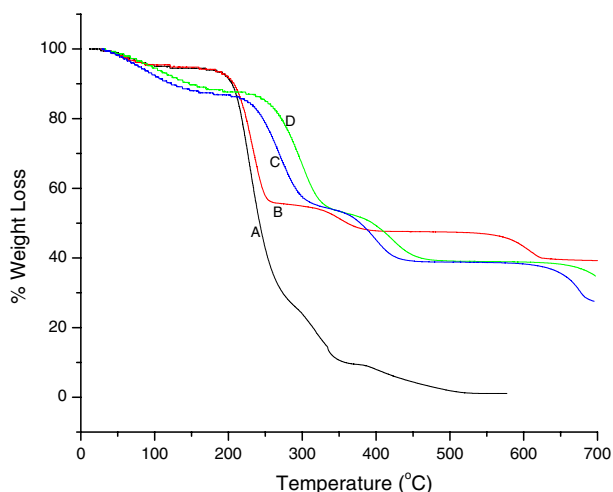


**Figure 4.** $^1$H NMR spectra of pure PVA (I) and 50% grafted copolymer (II).
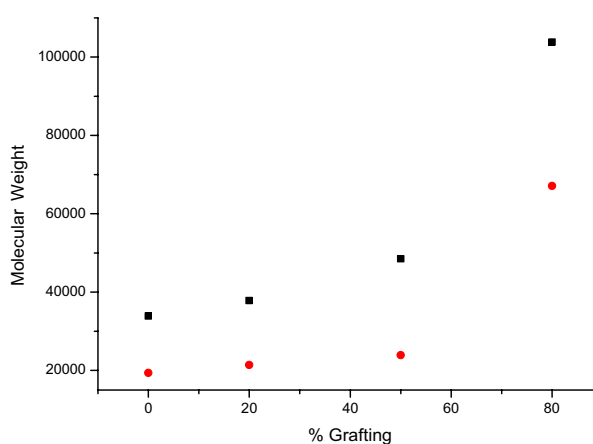


**Figure 5.** Weight average (■) and number average (●) molecular weights measured by GPC.

remarkable stability is observed in the cases of 50 and 80% grafted copolymers. The highest stability is observed for 80%

**Figure 6.** Thermogravimetric analysis of pure PVA (A) and 20% (B), 50% (C) and 80% grafted copolymer (D).



**Figure 7.** Plot of EBA versus percentage grafting of PAA in an electrolyte at 10 V in 0.05 N electrolyte solutions.

grafted copolymer, which has started decomposing at around 250 °C and 50% grafted copolymer has started decomposing at around 230 °C. This indicates that, as the percentage grafting increases, stability towards temperature increases, which may be due to the chemical branched structure of the grafted copolymers.

Mechanical properties were recorded for all the polymeric membranes as per ASTM D 412 and are tabulated in table 1. The tensile strength is highest for P-1 and decreases in the order P-2, P-3, P-4, while on the other hand the ultimate elongation is least for P-1 and increases in the order P-2, P-3, P-4. This behavior may be due to increased flexibility of the membrane materials as the percentage grafting increases in the membrane.

### 3.2. Polymer bending in an electric field

A constant electric voltage across two parallel carbon electrodes can cause bending of polymer hydrogel in NaCl electrolyte solution towards the cathode. It is generally thought that bending of the polymer is due to the voltage induced motion of the ions and the concomitant expansion of one side and contraction of the other side of the polymer. When an electric voltage is applied to the negatively charged polymer in the electrolyte solution, the counter-ions of the polyion remain immobile. Also, the free ions in the surrounding solution move toward their counter-electrode and come into the gel. Therefore the osmotic pressure of the gel polymer network near the positive electrode increases and becomes larger than that of the negative electrode side. Consequently, an osmotic pressure difference occurs within the gel and is the driving force that controls bending toward the negative electrode.

### 3.3. Effect of PAA content on the bending behavior

Figure 7 displays the dependence of the PAA content in the bending of the polymer in an electrolyte at 10 V in 0.05 N electrolyte solutions at pH 7. The effective bending angle (EBA) is least in the case of P-1, compared to P-2, P-3 and P-4. This is because of the presence of –COOH groups in the grafted polymer which are in a dissociated state and responsible for the increased ionicity of the polymeric gel which leads to
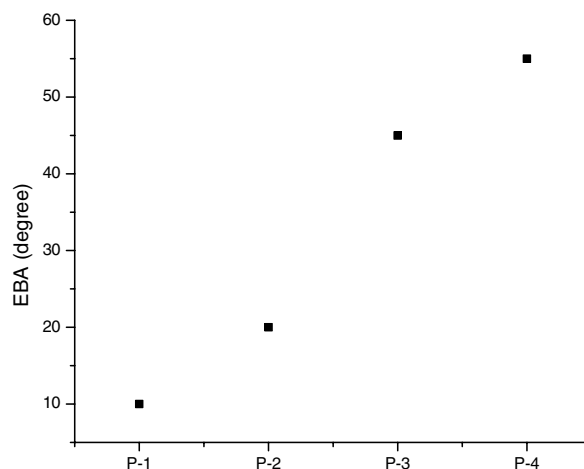
the increase in the swelling of the gel on the anode side. So, as the PAA content increases in the polymeric hydrogel, the EBA increases linearly [25, 26]. As, observed from swelling results from figure 2 it is clear that the EBA depends on the swelling of the membrane in the electrolyte solution, so there is a direct relation between the swelling behavior of the polymeric membrane in electrolyte solution and their electroactivity (bending) in the same electrolyte solution. It is observed from figures 2 and 7 that the higher the swelling, the better the bending behavior.

### 3.4. Effect of electrolyte concentration on the bending behavior

The dependence of EBA on the ionic strength of the electrolyte at 10 V is represented by figure 8. As the ionic strength of the electrolyte increased, EBA also increased up to 0.1 N, and further increase in the ionic strength resulted in the decrease of EBA. The dependences of EBA on the ionic strengths of 0.05, 0.1 and 0.15 N are graphically represented in figure 8, where the highest bending is observed in 0.1 N electrolyte solutions for all the polymers. If the concentration of the electrolyte solution is greater than its critical concentration, a shielding effect of the polyions, caused by the other ions in the electrolytic solute, occurs [20, 27], leading to the electrostatic repulsion of the polyions and a decrease in the degree of bending. According to Flory's theory [28], an increasing amount of ions could reduce the electrostatic repulsion of the polyions by the screening of fixed charges and bring about a decrease in the degree of bending. In such a situation, the network could shrink into a more compact structure, making ion diffusion through the gel more difficult. Because of this, the ionic gradient and osmotic pressure may not occur above the critical concentration of the electrolyte, due to which a decrease in EBA is observed above 0.1 N ionic strength of the electrolyte.

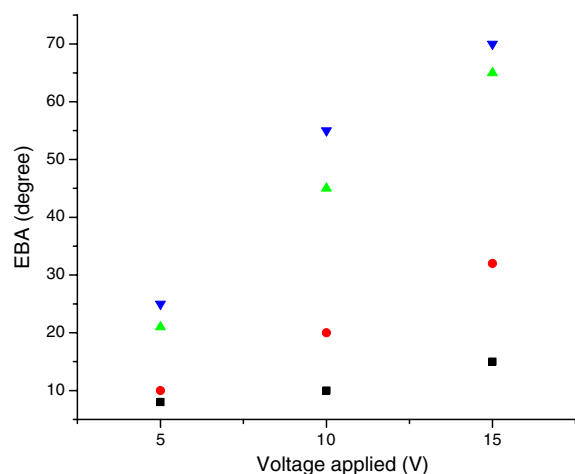### 3.5. Effect of electric voltage on the bending behavior

Figures 9, 10 and 11 represent the dependence of EBA on the voltages applied across the carbon electrodes in 0.05, 0.1 and
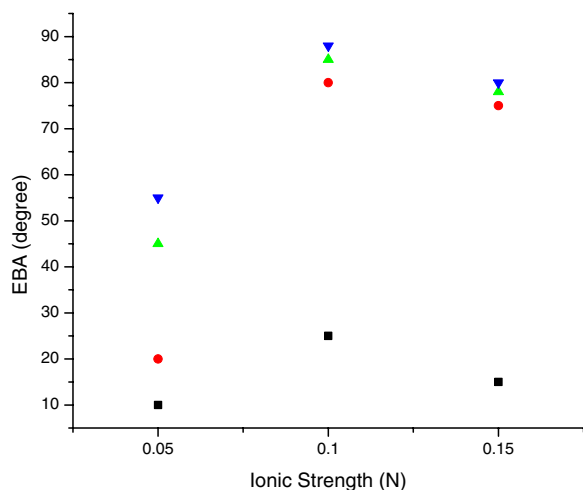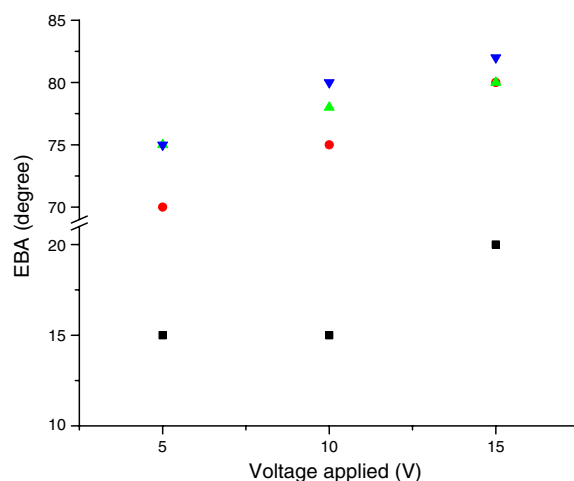
**Figure 8.** Plot of EBA versus ionic strength for the electrolyte at 10 V for P-1 (■), P-2 (●), P-3 (▲) and P-4 (▼).
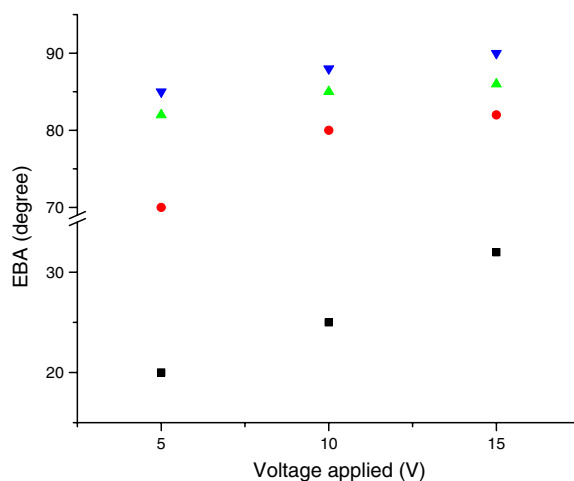


**Figure 10.** Plot of EBA versus voltage applied in 0.1 N electrolyte; the symbols are the same as in figure 8.



**Figure 9.** Plot of EBA versus voltage applied in 0.05 N electrolyte; the symbols are the same as in figure 8.



**Figure 11.** Plot of EBA versus voltage applied in 0.15 N electrolyte; the symbols are the same as in figure 8.

0.15 N NaCl electrolytes respectively. EBA is increased for all the polymeric hydrogels linearly with increase in electric voltage. It is observed that equilibrium bending is attained in the time range of 2–5 min by all hydrogels. In 0.05 N NaCl solution the increase in EBA with respect to voltage is remarkable for P-4 and P-3, which can be seen in figure 9. The increases of EBA in 0.1 and 0.15 N NaCl solutions as a function of voltage are not great because of the availability of enough free ions for movement.

## 4. Conclusions

In this paper, bending behavior studies were performed by using poly(acrylic acid) grafted poly(vinyl alcohol) prepared by Ce$^{(IV)}$ initiated free radical polymerization. The performance of pure PVA is compared with those of the grafted polymers. The grafted polymers have shown higher equilibrium bending angles in NaCl electrolyte solutions than pure PVA. The electroactivity of the grafted copolymer is very much dependent on the percentage grafting, ionic strength of

the electrolyte used and DC voltage applied across the polymer. The highest EBA is observed for P-4 in 0.1 N NaCl electrolyte at 15 V, which is 90°.

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