Iranian Polymer Journal **15** (11), 2006, 891-900

Available online at: http://journal.ippi.ac.ir

Swelling Characterization of Anionic Acrylic Acid Hydrogel in an External Electric Field

Javad Tavakoli¹, Esmaiel Jabbari², Mohammad Etrati Khosroshahi^{1*}, and Mina Boroujerdi³

(1) Biomaterial Group, Bioengineering Faculty, Amirkabir University of Technology, P.O. Box: 15875/4413, Tehran, Iran

(2) Department of Chemical Engineering, South Carolina University, Columbia 29208, USA

(3) Bioelectric Group, Electrical and Computer Science Faculty, Tehran University, P.O. Box: 14395-515, Tehran, Iran

Received 22 May 2006; accepted 25 November 2006

A B S T R A C T

polymer gel is a cross-linked polymer which undergoes a reversible volume and/or sol-gel phase transition in response to physiological (temperature, pH and presence of ion in organism fluids) or other external (electric field, light) stimuli. In structurally soft gels, the motion of polymer network and the diffusion of ions easily take place by an external stimulus. A typical function of gel containing ionic groups is to swell under the influence of an electric field, making it useful for wide biomedical applications. In this study, poly(acrylic acid) (PAA) hydrogel network was prepared by free radical polymerization whose average molecular weight between crosslinkes was calculated as 18500 g/mol and its homogeneity assessment evaluated by comparing 30 samples swelling ratios after 48 h soaking in distilled deionized water. The swelling behaviour of PAA under an electric field application was also investigated as a function of cross-linking agent and electric field intensity variation. It is seen that equilibrium swelling ratio of PAA gel increases from 16 (no electric field) to 30 (by application of 300 V/m electric field), however this variation is dependent on the sample location relative to electrodes. The normalized swelling ratio of sample changes by 4.8 to 0.9 relative to its distance from positive electrode. Increasing the concentration of cross-linking agent (EGDMA) from 0.14 to 0.71 molar percent of monomer resulted in 45% decrease of gel volume fraction. It is proven that applying an external electric field can make an improvement in the time-response of the hydrogel expansion phase and swelling behaviour.

Key Words:

poly(acrylic acid); electric field; swelling ratio; cross-linking agent; gel volume fraction; swelling theory.

INTRODUCTION

Swelling of hydrophilic polymeric matrices has been the subject of significant research in the last few years. Polymer gels are widely used in biomedical applications such as drug delivery systems, immobilizing enzymes and living cells and artificial cartilages as passive applications [1-3]. In contrast to such passive uses, an increasing interest in polymer gels as active elements has raised new develop-

(*) To whom correspondence to be addressed. E-mail: Khosro@aut.ac.ir ment such as stimuli-responsive motion hydrogels and muscular system [4-5].

The microstructure of a hydrogel can affect solute diffusion. Some of the factors affecting solute diffusion in hydrogels are chain entanglement, chain mobility, cross-linking density and type, equilibrium degree of swelling, and the relaxational characteristics of the gel network. On a molecular level, the solute diffusion rate through swollen gels depends on the relative hydrodynamic radius of the solute, and the network mesh size formed by the macromolecular chains. A larger solute diffuses at a slower rate than a smaller one [6-7].

Stimuli-responsive hydrogels have been shown to be a natural choice for use in microfluidic systems because of their controllable response by diffusion processes. Hence, as hydrogel devices are made smaller, the time-response of their volume transitions becomes faster. Hydrogels have been shown to respond to a number of different stimuli, including pH [8], electric fields [9], temperature [10], light [11], even organic compounds [12] and antigens [13]. Although responsive hydrogels have been studied extensively, their application has been limited by their slow response times at larger scales. However, recently, many of these hydrogels have received a renewed attention as potential valves, filters, sensors, and actuators in micro fluidic systems, where small (on the order of tens of μ m) and fast (less than a second) responses are necessary.

Hydrogels sensitive to electric current are usually made of polyelectrolytes such as the pH-sensitive hydrogels [14]. Electro-sensitive hydrogels undergo shrinkage or swelling in the presence of an applied electric field. In evaluation and characterization the use of chitosan gels as matrices for electrically modulated drug delivery, release-time profiles for neutral (hydrocortisone), anionic (benzoic acid) and cationic (lidocaine hydrochloride) drug molecules from hydrated chitosan gels were monitored in response to different milliamperages of current as a function of time [15]. Likewise, chondroitin 4-sulphate hydrogels were examined as potential matrices for the electrocontrolled delivery of peptides and proteins [16].

In our research, swelling behaviour of anionic acrylic acid gels was studied in an external electric field with respect to the effect of variation of electric field intensity and cross-linking agent on gel swelling characteristics. The novelty of this research is an equation which describes the electric field contribution to equilibrium swelling of polyelectrolyte gel. Also we showed that the relation between normalized swelling ratio and electric field variation is almost linear which was not considered in the previous research works. The results of this study can be used for swelling controlled drug delivery from hydrogels under electric field stimuli. Loading drugs into polymer gel network can be directly controlled by increasing the electric field as its equilibrium swelling ratio changes almost linearly with the applied electric field. On the other hand drug or biological active agents being released to medium can also be under control by electric field.

EXPERIMENTAL

Materials and Method

Acrylic acid (AA) hydrogel was prepared by free radical polymerization of the distilled monomer (Merck, Germany) with ammonium persulphate and sodium disulphide (Merck, Germany) as initiators, each 2 mol percent of monomer. Monomer purification was performed by vacuum distillation under 30°C temperature and 5 mmHg pressure. Distilled monomers were kept at -5°C in order to prevent spontaneous polymerization. Ethylene glycol dimethacrylate (EGDMA) (Hochst, Germany) was used as cross-linking agent with concentration of 0.14, 0.28, 0.43 and 0.71 mol percent of monomer. Polymerization was carried out at 50°C for 2 h in 1 cm diameter and 2 mm depth, disk shape moulds. Prior to any measurement, samples were dialyzed against distilled water for 8 h in order to extract unreacted materials from the gel network. Gel swelling measurement conducted by weight using a precise balance (Sartorious, Swiss). For each gravimetric measurement the sample was removed from its place in an electric field, the surface of swollen gel was dried by tissue paper and immediately situated in their place after weighing.

Samples Homogeneity Evaluation

30 Disk shape PAA hydrogels were weighed in dry state, then they were soaked in separate dishes with

Sample	Dry weight (g)	Swelling ratio
1	0.133	6.75
2	0.152	6.73
3	0.154	9.95
4	0.164	6.98
5	0.148	6.87
6	0.152	7.01
7	0.159	6.83
8	0.154	7.12
9	0.161	6.93
10	0.166	6.87
11	0.145	6.93
12	0.161	7.05
13	0.154	6.89
14	0.142	6.99
15	0.166	7.11

 Table 1. PAA Homogeneity evaluation by swelling ratio

 measurement.

500 mL distilled deionized water (DDW) for 48 h. Swelling ratio indicated homogeneity of samples. Table 1 shows some related data.

Preparation of Buffer Solutions

For determining the average molecular weight between cross-links in gel network, it was necessary to study the swelling behaviour of gel in different pH buffer solutions. To achieve buffer solutions with pH of 5 and 7, 50 mL of potassium hydrogen phthalate (0.1 molar) with 22.6 and 29.1 mL sodium hydroxide (0.1 molar) were mixed. For buffer solution with pH of 10, 50 mL of borax (0.025 molar) was mixed with 18.3 mL of sodium hydroxide (0.1 molar).

Analysis of Cross-linked Structure of PAA Gel

For an anionic hydrogel in distilled deionized water at thermodynamic equilibrium the total chemical potential gradient between the gel and the aqueous phase should be equal to zero. The molecular weight between cross-links, M_c , can be determined from swelling pressure at equilibrium state which is described by eqn (2).



Figure 1. Schematic diagram of experimental set up.

Electric Field

To study the effect of electric field on swelling of poly(acrylic acid) gels, a two-liter vessel containing distilled deionized water, two platinum plates of 144 cm² surface area as electrodes with adjustable distance between were placed vertically. A DC power source (Afzarazma, Iran) was used to generate the electric field. The disk shape gels were allowed to reach equilibrium swelling in distilled deionized water for at least 24 h, then they were placed between the two platinum electrodes such that the direction of electric field lines were perpendicular to disk surface. All the experiments with electric field were carried out in distilled deionized water. The schematic diagram of experiment set up is shown in Figure 1.

Theory

The average molecular weight between cross-links, M_c , was calculated from the swelling data in equilibrium state using eqn (2) as discussed by Brannon and Peppas [17] followed by eqn (1):

$$\Delta \mu_{t} = \Delta \mu_{mix} + \Delta \mu_{el} + \Delta \mu_{ion} = 0 \tag{1}$$

 $\Delta \mu_{mix}$, $\Delta \mu_{el}$, and $\Delta \mu_{ion}$ are chemical potential gradient due to mixing, elastic retractile force and ionic interaction, respectively.

$$\begin{bmatrix} \text{Ln} (1 - \upsilon_{2,s}) + \upsilon_{2,s} + \chi \ \upsilon_{2,s}^2 \end{bmatrix} + \begin{bmatrix} (V/4I) \ [k_a/(10^{-pH} + k_a)^2] \ (\upsilon_{2,s}/\upsilon) \end{bmatrix} + \begin{bmatrix} (\upsilon_{2,r}\upsilon/2VM_c)(1 - 2M_c/M_n) \ [(\upsilon_{2,s}/\upsilon_{2,r})^{1/3} - 0.5((\upsilon_{2,s}/\upsilon_{2,r})] \end{bmatrix} = 0$$
(2)

where M_n is the number average molecular weight of

Iranian Polymer Journal / Volume 15 Number 11 (2006) 893

polymer before cross-linking (g/mol), υ is specific volume of PAA (cm³/g), V is molar volume of water (cm³/mol), $\upsilon_{2,r}$ is the polymer volume fraction in the gel in the relax state, $\upsilon_{2,s}$ is the polymer volume fraction in the gel in the swollen state, χ is the interaction parameter of polymer and water, and I is ion strength of buffer.

The average molecular weight between crosslinks, M_c , was measured by swelling the 27 PAA gel disks in buffered solutions of pH 5, 7 and 10. All the samples were cross-linked by concentration of 0.5 mol percent of monomer of EGDMA. To estimate M_c from swelling data, M_n was considered as 10⁵ g/mol, which according to manufacturer description and the value of polymer-solvent interaction parameter, χ is about 0.45.

Initial experiments with hydrogels in electric fields showed that fast and repeatable volume changes are possible by taking advantage of the osmotic pressurebased mechanism discussed earlier [18-19]. This mechanism is driven by mobile ion concentration differences in and around a hydrogel in an aqueous solution. In the absence of an electric field, the distribution of ions follows the principle of Donnan equilibrium [20], whereby the ionic concentration inside a hydrogel membrane having fixed charges (i.e., acidic or basic groups bound to the polymer matrix) is not necessarily equal to the external bath concentration. For instance, if the hydrogel considered is anionic, the concentration of cations inside the gel is higher than in the surrounding bath and the concentration of anions is lower in the gel than in the bath before application of any electric field. Electroneutrality of gel was maintained constant by having positive charges in the hydrogel matrix. When an electric field is applied, mobile ions inside and outside the hydrogel are rearranged and concentration gradients are established. Figure 2 shows the effect of an applied electric field on mobile cations in and around the anionic hydrogel disk.

It should be noted that the acidic groups bound to the polymer chains are assumed to be immobile under an electric field [21], and therefore the concentration of the bound groups are considered constant and uniform inside the hydrogel. These fixed charges retard the migration of free ions inside the hydrogel to some extent. As a result, the ionic concentration gradient



Figure 2. Cationic profile before and after application of an electric field.

inside the gel is smaller than in the solution. In addition, other experimental conditions such as flow and bubbles formation will also affect ion migration.

For an anionic hydrogel in distilled deionized water placed in an external electric field at thermodynamic equilibrium, the total chemical potential gradient between the gel and the aqueous phase should be equal to zero.

$$\Delta \mu_{\rm mix} + \Delta \mu_{\rm el} + \Delta \mu_{\rm ion} + \Delta \mu_{\rm ef} = 0 \tag{3}$$

 $\Delta \mu_{mix}$, $\Delta \mu_{el}$, $\Delta \mu_{ion}$, and $\Delta \mu_{ef}$ are chemical potential gradient due to mixing, elastic retractile force, ionic interaction and external electric field, respectively. Flory-Huggins, rubber elasticity and Donnan theories were used to estimate $\Delta \mu_{mix}$, $\Delta \mu_{el}$ and $\Delta \mu_{ion}$, respectively. The chemical potential gradient due to electric field can be estimated by eqn (4) derived by authors.

$$\Delta \mu_{\rm ef} = (27B\omega/\rho\pi D^2 L_0) (3\beta^2 + 2B\beta)^{-1}$$
(4)

where:

 ω = number of moles of network chain

D = initial diameter of the disk shape sample

 L_0 = distance between the electrodes

 β = relative change in disk thickness due to swelling in an electric field

B = reduced electric field potential

The relation between B and β has been derived before [5].

Derivation of eqn (4) is achieved by considering the electric field theory. According to this theory, a force equal to E.q is exerted on a charge q which is located in an electric field with intensity, E. So the force against a gel network located in an electric field could be explained by eqn (5) as follows:

$$\mathbf{F} = (\boldsymbol{\omega}.\mathbf{f}.\mathbf{e}). \mathbf{E}$$
 (5)

where ω is number of gel chain, f is the ionized fraction, e is electron charge (= 1.9×10^{-19} C) and E is electric field intensity. Application of electric field on a disk shape gel with ΔZ_0 thickness and D_0 diameter which is located at distance Z from negative electrode causes an increase of $\beta \Delta Z_0$ and αD_0 in thickness and diameter, respectively. In this state, as the gel thickness is small and its deformation occurs in the direction of electric field line, the total deformation of gel chains due to swelling is $\Delta Z_0(\beta - \beta_{ne})$, where β_{ne} is change in thickness due to swelling without any external stimuli. In another word swelling behaviour of gel is influenced by its location relative to electrodes. So multiplying deformation equation in non-dimensional parameter Z/L₀, presents relative deformation (volume change) to electrode. Eqn (6) shows the change of free Gibbs energy due to electric field application.

$$\Delta G = (\omega.f.e). \ E \ \Delta Z_0 \ (\beta - \beta_{ne}) \ (Z/L_0) \tag{6}$$

In eqn (6), (f.e.Z)KT could be replaced by B which is reduced electric field potential. So ΔG could be rewritten as eqn (7):

$$\Delta G = BKT\omega \left(\beta - \beta_{ne}\right) \left(\Delta Z_0 / L_0\right) \tag{7}$$

Chemical potential energy could be calculated through derivation of eqn (7) by the number of solvent molecules, N_s, which is equal to V_s(ρ/M_s) N_{ave}. As the volume of diffused solvent to gel network, V_s is described by $(\pi/4)(\alpha D)^2(\beta \Delta Z_0)$ -V_p, number of solvent molecules governing equation could be rewritten as eqn (8):

$$N_{s} = (\rho/M_{s})N_{ave}[(\pi/4)(\alpha D)^{2}(\beta \Delta Z_{0})-V_{p}]$$
(8)

where ρ and M_s are the density and molecular weight of solvent, respectively, N_{ave} is Avogadro number and V_p is network volume in dry state. It is proved that the sum of free energies of gel swelling is minimized when the relation $\alpha^2 = \beta^2 + B\beta$ is governing [5]. So the final relation between number of solvent molecules and gel dimensions and swelling parameters is described by eqn (9):

$$N_{s} = (\rho/M_{s})N_{ave}[(\pi/4)(\beta^{2}+B\beta)D^{2}(\beta\Delta Z_{0})-V_{p}]$$
(9)

Considering eqns (9) and (7) chemical potential energy can be derived as eqn (10):

$$\Delta \mu_{ef} = (\partial G / \partial N_s) = (\partial G / \partial \beta) (\partial \beta / \partial N_s)$$

= B\oked KT(72/\rho\pi D^2 L_0) (3\beta^2 + 2B\beta)^{-1} N_{ave} (10)

As K N_{ave} is equal to R the final equation of chemical potential energy due to electric field could be written as it described before in eqn (4) and the total free energies in equilibrium state could be presented in eqn (11):

$$\begin{split} &RT\{[ln(1 - \upsilon_{2,s}) + \upsilon_{2,s} + \chi \upsilon_{2,s}^2] + [(V/4I)[k_a/(10^{-pH} + k_a)^2](\upsilon_{2,s}/\upsilon)] + [(\upsilon_{2,r}V/2\upsilon^*M_c)(1 - 2M_c/M_n) [6\beta + 2B - (3\beta + 2B)(\beta^2 + B\beta)^{-1}](3\beta^2 + 2B\beta)^{-1}]] + (72B\omega/\rho\pi D^2L_0) \\ &(3\beta^2 + 2B\beta)^{-1} = 0 \end{split}$$

In the presented model, solvent molar volume (18 cm³/mol), specific volume of gel in dry state (0.83 cm³/g), molar volume of polymerized monomers (60 cm³/mol), molecular weight of linear polymer (100,000 g/mol) and average molecular weight of cross-linked chains (18,500 g/mol) are considered in order to follow equilibrium swelling behaviour. To calculate B as a function of electric field intensity, distance between electrodes (10 cm), location of gel relative to negative electrode (5 cm), Boltzman constant (1.38×10^{-23}) , and fraction of hydrolyzed group in gel network (0.68) were considered. Fraction of hydrolyzed groups (f) in gel network was calculated by measuring the pH environment. The experiment was carried out at 30°C. Ion strength, I, was calculated by equation $I = 0.5(\Sigma C_i Z_i^2)$ according to buffer element concentration.

RESULTS AND DISCUSSION

The average molecular weight between cross-links, M_c , was 18500 g/mol (approx.), as measured by swelling the 28 gel disks in buffered solutions of pH 5, 7 and 10. All the samples were cross-linked by con-

Iranian Polymer Journal / Volume 15 Number 11 (2006) 895



(◆) No electric field (▲) 300 (V/m) **Figure 3.** Swelling ratio as a function of time for PAA gel equilibrated in DDW.

centration of 0.5 mol percent of monomer of EGDMA.

Figure 3 shows the swelling ratio as a function of time for PAA gel equilibrated in DDW, with and without electric field. The gel was placed at the centre with equal distance of 5 cm from the two electrodes. In the presence of electric field (300 V/m), the equilibrium swelling ratio was increased from 16 to 30, indicating the electric field on the swelling of PAA gels.

The variation of PAA swelling ratio under different electric fields as a function of time was also studied. As it is seen from Figure 4, variation in an electric field intensity from 50 to 100, 150, 200 and 250 V/m has caused an increase of equilibrium normalized swelling ratio (NSR) by 1.4, 1.8, 2.9, and 4.1, respectively. The equilibrium normalized swelling ratio is defined as the swelling ratio in the presence of electric field which can be calculated by eqn (12).

$$NSR = (W_e - W_{ne})/W_{ne}$$
(12)

where W_e and W_{ne} are the weight of swollen gel in equilibrium state in the presence of electric field and equilibrium state in the absence of it, respectively.

As it is seen, the application of electric filed makes a stepwise swelling trend in PAA gel, hence, increasing of electric field intensity makes this phenomena much more obvious. It is estimated that increasing the electric field intensity causes higher gel swelling, so that some cross-linked chains with short length may



Figure 4. Variation of normalized swelling ratio versus time for different electrical field intensities.

be ruptured under osmotic pressure, π , due to external electric field. So in each step the specification of hydrogel network varies as an average length of cross-linked chains in the gel bulk change.

It must be noted that, anionic hydrogels swollen in an aqueous electrolyte solution show three types of deformation under the influence of an external D.C. electric field: swelling, shrinking and bending. The type of deformation mainly depends on the gel shape and the position of the gel between the electrodes. When a polyelectrolyte gel is placed perpendicular to the electric field lines, bending is the most probable observation. This bending behaviour takes place towards a positive electrode for anionic poly(acrylic acid) gel. The bending mechanism could be explained with osmotic pressure phenomena. The volume of a gel is controlled by osmotic pressure. The osmotic pressure is given as the sum of π_1 , π_2 and π_3 which corresponds to the osmotic pressures due to the solubility of the solvent in the gel chain, the rubber elasticity and ion concentration differences between the inside and outside of the gel respectively defined by eqn (13) [22]:

$$\pi = [\ln(1-\upsilon) + \upsilon + \chi \upsilon^2] \operatorname{RT/V} + (\upsilon^{1/3} - \upsilon/2) \operatorname{RT} \upsilon_e / \upsilon_0 + (\Sigma c_i - \Sigma c_j) \operatorname{RT}$$
(13)

where, υ is the volume fraction of the gel network, χ is solubility parameter, υ_0 is the volume of gel network under the dry condition, υ_e is the number of the



Figure 5. Schematic diagram of gel bending in an external electric field application.

chains, V is the molar volume of the solvent, c_i and c_j are the ion concentration inside and outside of the gel, respectively, R is gas constant and T is the temperature.

At equilibrium, the value of π is equal to that of the surrounding aqueous solution, π_0 . Therefore π_1 , π_2 and π_3 have their definite values. When an electric field is applied to gel in the solution, the counterion and the free ion can drift to the positive or negative electrode, whereas the polyion cannot move. Consequently, π_3 varies and the value of π deviates from π_0 . The swelling and shrinkage of the gel occur until the gel reaches its new equilibrium state. Since the ion drift at the different speeds depends on their size and valency, the osmotic pressure of the positive side is unequal to the negative side and the gel bending occurs.

But in our experiment the gel sample placed in parallel to electric field lines, so buckling and nonhomogeneous swelling observed during the use of electric field. Symmetric shape of the samples (circular shape samples) with small thickness may cause some bending negligible compared to non-homo geneous swelling. Due to non-homogeneous swelling of the gel in an electric field, the positive side of the gel (the part of the gel close to positive electrode) swelled, while its negative side shrank. It must be noted that the above conditions are difficult to be diff-



(♦) Negative electrode (■) Center (▲) Positive electrode

Figure 6. Swelling behaviour of PAA gel in an electric field relative to electrodes.

erentiated. Schematic diagram of what explained is described in Figure 5.

The position of the gel relative to electrodes had a significant effect on swelling, as shown in Figure 6. When the position of the gel was increased relative to positive electrode from 1 to 5 and 9 cm, the equilibrium normalized swelling ratio decreased from 4.8 to 2.9 and 0.9, respectively under 300V/m. When the gel was placed in the electric field, the positive hydrogen ions of the -COO⁻H⁺ groups of acrylic acid migrated towards the negative electrode. The most arguable reason for gel swelling in the electric field is the presence concentration gradient of mobile cations of the gel/surrounding interface at the anode electrode side. It is important to notice that the contribution of fixed negative charges to ionic swelling pressure of PAA gel is negligible compared with mobile counterions. This fact is also depicted in Figure 1. When placed near the positive electrode, repulsion of hydrogen ions from the electrode, made more hydrogen ions to migrate towards the negative electrode and produced an increase in the swelling ratio.

Effect of increasing cross-linking agent on swelling behaviour of PAA gel was also studied. Changing the concentration of EGDMA from 0.14 to 0.28, 0.43 and 0.71 (mol percent of monomer), decreased equilibrium swelling ratio by 32, 21, 17 and 14, respectively in application of 300 V/m electric field intensity. Adding more cross-linking agents makes the hydrogel network dense and hence, affects



Figure 7. Variation of normalized swelling ratio and gel volume fraction versus cross-linking agent.

the swelling of the gel. The equilibrium swelling ratio is also nonlinearly proportion to the inverse of polymer volume fraction in the gel network. Figure 7 indicates the variation of swelling ratio and polymer volume fraction versus cross-linking agent change. Increasing the concentration of EGDMA from 0.14 to 0.71 (mol percent of PAA) resulted in 54 percent decrease in polymer volume fraction.

It was also observed that at the equilibrium normalized swelling ratio changes almost linearly as electric field intensity increases. As it is indicated in Figure 8, increasing electric field intensity from 50 to 300 V/m increased the normalized equilibrium swelling ratio



Figure 8. Equilibrium normalized swelling ratio change versus electric field intensity.



Figure 9. Comparison of theoretical and experimental equilibrium PAA volume fraction as a function of electric field intensity.

from 0.5 to 3.2, respectively.

Figure 9 compares the theoretical equilibrium PAA volume fraction as a function of electric field intensity with experimental findings. The discrepancy between the experiment and theory reduces as the PAA volume fraction decreases. This is believed to be due to the assumption of dilute electrolyte solution used in the derivation of eqn (2). Also at lower values of the electric field strength, the electrochemical potential of swelling is dominated by ionic osmotic pressure which is overestimated by considering ionic network as an ideal solution. But as electric field strength increases, the contribution of the ionic osmotic pressure for the swelling of the hydrogel with respect to electric field driven osmotic pressure decreases and hence the deviation between experimental and theoretical reduces.

There are a number of parameters that must be controlled in carrying out an experiment with electrically triggered hydrogels. For instance, hydrogels swelling ratios differ with changing the location of hydrogel in relation to the electrodes, fluid medium, amount of cross-linking agent, and temperature. Most of the shape change occurs in the portion of the hydrogel nearest to the anode, as explained earlier. As the applied voltage increases, an improvement in the time-response of the hydrogel expansion phase can be observed. Electric field intensity increases with higher applied voltage, and as a result of which a larger ion concentration gradients are established. This, in turn, leads to a higher osmotic stress and a more rapid swelling.

CONCLUSION

Application of electric field significantly increases the swelling of the anionic poly(acrylic acid) (PAA) hydrogels. For anionic PAA gels, the extent of swelling was higher when the electric field intensity increased or placed near the positive electrode. The relation between normalized swelling ratio and electric field intensity was approximately linear. It is also believed that increasing the concentration of crosslinking agent decreases the swelling ratio of gel in the presence of electric field.

REFERENCES

- Lowman A.M., Morishita M., Kajita T., Peppas N.A., Oral delivery of insulin using pH responsive complexation gels, *J. Pharm. Sci.*, 9, 933-937, 2000.
- Yonese M., Baba K., Murabayashi H., Polymer Gels: Fundamentals and Biomedical Application, DeRossi, Kajiwara, Osada, Ymauchi (Eds.), Plenum, New York, Ch. 4, 319-340, 1991.
- Suzuki M., Polymer Gels: Fundamentals and Biomedical Application, DeRossi, Kajiwara, Osada, Ymauchi (Eds.), Plenum, New York, Ch. 2, 224-279, 1991.
- Micheal J.B., Aveek N., Aluru R., David J., Development and modeling of electrically triggered hydrogels for microfluidic application, *J. Microelectromech. Syst.*, 14, 1198-1207, 2005.
- Tavakoli J., Jabbari E., Effect of electric field on swelling of anionic acrylic acid hydrogels, *Proc. Int. Symp. Control. Rel. Bioact. Mater.*, Paris, 682-684, 2000.
- Mohammadi Y., Asghar Nejad N., Jabbari E., Hydrophobicity effects of crosslinking agent on swelling behavior of poly(acrylic acid) hydrogels in electric fields, *Proc. 6th Iran. Seminar Polym. Sci. Technol. (ISPST 2003)*, Tehran, 346, 2003.

- Dimitri R., Kofinas P., Characterization of anion diffusion in polymer hydrogels used of wastewater remediation, *Polymer*, 46, 9342-9347, 2005.
- Beebe D.J., Moore J.S., Bauer J.M., Yu Q., Liu R.H., Devadoss C., Functional hydrogel structures for autonomous flow control inside microfluidic channels, *Nature*, **404**, 588-590, 2000.
- Tanaka T., Nishio I., Sun S., Ueno-Nishio T., Collapse of gels in an electric field, *Science*, 218, 467-469, 1982.
- Richter A., Kuckling R., Howitz S., Gehring T., Arndt K., Electronically controllable microvalves based on smart hydrogels: Magnitudes and potential applications, *J. Microelectromech. Syst.*, 12, 748-753, 2003.
- Suzuki A., Tanaka T., Phase transition in polymer gels induced by visible light, *Nature*, **346**, 345-347, 1990.
- Kataoka K., Miyazaki H, Bunya M., Okano T., Sakurai Y., Totally synthetic polymer gels responding to external glucose concentration: Their preparation and application to on-off regulation of insulin release, *J. Am. Chem. Soc.*, **120**, 12694-12695, 1998.
- Miyata T., Asami N., Uragami T., A reversibly antigen-responsive hydrogel, *Nature*, **399**, 766-769, 1999.
- Qiu Y., Park K., Environment-sensitive hydrogels for drug delivery, *Adv. Drug Deliv. Rev.*, **39**, 321-339, 2001.
- 15. Ramanathan S., Block L.H., The use of chitosan gels as matrices for electrically-modulated drug delivery, *J. Control. Rel.*, **23**, 109-123, 2001.
- Jensen M., Hansen P.B., Murdan S., Frokjaer S., Florence A.T., Loading into electro-stimulated release of peptides and proteins from chondroitin 4-sulphate hydrogels, *Eur. J. Pharm. Sci.*, **15**, 139-148, 2002.
- Brannon M.L., Peppas N.A., Solute diffusion in swollen membranes: Part VIII. Characterization of and diffusion in asymmetric membranes, J. *Membrane Sci.*, **32**, 125-138, 1987
- Bassetti M.J., Moore J.S., Beebe D.J., Development of electrically triggered hydrogels for microfluidic applications, *Proc. 2nd Annual. Int. IEEE-EMBS Spec. Topic Conf. Microtechnol., In Medicine and Biology*, 410-413, 2002.

Iranian Polymer Journal / Volume 15 Number 11 (2006) 899

- 19. Bassetti M.J., Beebe D.J., Demonstration of hydrogel volume control using pulse width modulation, *Proc. MicroTAS 2002 Symp.*, 718-720, 2002.
- 20. Malmuvio J., Plonsey R., *Bioelectromagnetism: Principles and Applications of Bioelectric and Biomagnetic Fields*, Oxford University, New York, 1995.
- Hirose Y., Giannetti G., Marquardt J., Tanaka T., Migration of ions and pH gradients in gels under stationary electric fields, *J. Phys. Soc. Japan*, 61, 4085-4097, 1992.
- 22. Kurauchi K., Shiga T., Hirose Y., Okada A., Polymer Gels: Fundamentals and Biomedical Application, DeRossi, Kajiwara, Osada, Ymauchi (Eds.), Plenum, New York, Ch. 4, 237-247, 1991.