



Actuation of a novel Pluronic-based hydrogel: Electromechanical response and the role of applied current



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ABSTRACT

A novel Pluronic-based hydrogel copolymer was synthesized and actuated. The combined properties of improved biocompatibility and response to electrical excitation of this cross-linked gel make it a potential candidate for actuators such as electrically controllable occlusion devices and non-surgical implants. The electrical response of the copolymer was verified using bending tests performed in potassium chloride (KCl) solutions of different concentrations and the novel material was characterized using equilibrium swelling experiments, environmental scanning microscopy (ESEM) imaging, and a potentiometric method for estimating the fixed charge. We considered the influence of current, pH and concentration on the hydrogel actuator behavior, which was manifested in the degree of bending and in direction of bending. A reversal of the bending direction was observed twice in a single system under DC bias (without reversal of electric field polarity). We described this phenomenon as representing several different types of electric-responsive behavior that become dominant at different actuation stages of the polymer system. The distinction between the different actuation effects is important for the development of hydrogel actuators.

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1. Introduction

There is an increasing interest in the use of stimuli responsive gels as actuators [1]. The implications of this are that a new class of bio-medical technologies, facilitated by the unique mechanical and chemical characteristics of these materials, can be developed. The soft elastomeric nature of hydrogels, serves to minimize mechanical and frictional irritation to the tissue bed, suggesting applications in tissue engineering [2,3], artificial muscles [4–7], and biomimicry [8–11]. Their swelling capacity results in high permeability for certain drug molecules and their reversible mesh sizes enables on/off diffusion behavior [12], making them ideal materials for drug delivery. They are “smart” materials due to their ability to sense external stimuli and respond in real time [13], performing dramatic volume transitions in response to external environmental stimuli such as the pH [12] and ionic strength of the solution, temperature, and electrical field [14]. Electrical stimulation is the most practical method for controlling the hydrogel dimensions because it allows for a high degree of controllability that can potentially be realized through integration with electrical circuits. A prime example is the

field of microfluidics where hydrogels have recently been utilized as responsive microvalves [15,16]. In order to realize the potential of these materials as actuators in vivo, two conditions must be fulfilled. The first is to develop a biocompatible hydrogel that displays electro-active behavior. The second is to find a suitable model for the electro-mechanical response of the hydrogel, which adequately reflects its behavior as an actuator.

Biocompatibility is crucial for in vivo applications such as arterial occlusions, tissue engineering, and drug delivery. Many of the studies on electric-responsive hydrogels have used polyacrylic acid gels [17], although the monomers are known to be toxic [13]. Pluronic F127 (PF127) is the least toxic of all commercially available polymers [18]. Because of its low toxicity and reversible thermal gelation, it has been investigated for drug delivery [19] and a cross-linked version was proposed as a material implantation [20]. Only in the past two years has Pluronic been used as a base for electric-responsive hydrogel synthesis [21]. A cationic electric responsive polymer based on PF127 was UV synthesized and shown to swell reversibly as a response to electrodes that are in contact with the gel sample [21]. However, preceding this work, no experiments using anionic electric responsive polymers based on Pluronic have been conducted using any configuration of electrochemical cell.

The different mechanisms responsible for the bending of hydrogels in response to an applied bias are reviewed by Shiga [22] and in

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the more recent work of Glazer et al. [17]. The main theory describing the bending of hydrogel beams that are not in contact with the electrodes is the osmotic pressure differences between gel and solution as presented in the comprehensive model by Doi et al. [14]. Asymmetry in the swelling of the two sides of the beam results in a bending of the beam.

Because osmotic pressure is a function of ion concentration [14], it is not surprising that pH changes in the electrolyte as a result of reactions at the electrodes have been considered as a factor influencing ion concentration in the hydrogel. This is indeed a major factor in determining gel behavior when electrodes are in direct contact with the gel sample [22], but it was recently determined that when the gel is not in contact with the electrodes, pH is not the trigger for initial bending in response to electrical bias [17]. It is often difficult to isolate the driving force behind the bending in a given system due to the many coupled effects found in most testing systems (i.e. temp, pH, current, electrical bias). The parameters that are considered important in the majority of the experimental work reported on hydrogel electro-actuation are the electrolyte concentration and the applied voltage.

Bending toward the anode of polyacrylic gels with fixed negative charges has been noted by a number of researchers [6,23,24], while other groups have reported bending toward the cathode [5,8,22]. It is difficult to reconcile these seemingly contradictory observations because in the overwhelming majority of the research, there is no investigation of the critical conditions required to generate bending on one direction or the other. Despite the existence of models predicting bending in the gel, it is not always easy to predict in which way the gel will respond due to the many parameters that determine the bending direction such as the type of moving ions in the solution and in the gel [4], the type of fixed charge, the density of crosslinks [3], and the concentration of the solution and the pH of the bathing solution [22]. Glazer et al. showed bending of anionic gels toward the cathode in low salt conditions, and bending toward the anode in high salt conditions [17]. Lim et al. showed bending toward the cathode for highly crosslinked samples and bending toward the anode for samples of low crosslink density [3]. There have been two cases of dynamic reversal of bending direction under DC bias published in the past year where the direction is first cathode and then anode [3,25]. This implies either a redistribution of ions under the DC field, or the presence of a number of bending mechanisms acting in the same system. A number of researchers have reported on the reversal of the field polarity or the application of an alternating current as a ways to dynamically change the bending direction of a hydrogel beam, however, in these cases the hydrogel beam is actually consistently bending toward either anode or cathode [1].

The purpose of the present work is to study the actuation behavior of a novel anionic Pluronic-based electroactive copolymer (PF127-BMA-MAA). In order to better understand the phenomena governing the hydrogel's behavior under electrical bias, the current was monitored along with other parameters such as the voltage across the cell, the electrolyte concentration, the pH and the pre-history of the hydrogel. In addition, we performed an evaluation of the fixed charge density along with basic material characterization of the material. We report on the copolymer's response to electrical activation and note the degree and direction of its bending behavior as compared with the widely exploited anionic hydrogel, poly sodium acrylate. We observed the phenomenon of bending reversal of the Pluronic based gel, not caused by a change in the polarity of the field, and suggest an explanation for this dynamic reversal. An understanding of the different effects present in the system is critical for use of this new soft material as an actuator. Dynamic reversal under DC bias may impair reliability, but it also implies a new aspect of functionality for hydrogel actuators, namely, that the same system can be used to generate different

kinds of controllable behavior in the same sample under different condition.

2. Materials and methods

2.1. Hydrogel synthesis

2.1.1. PF127-BMA-MAA

The novel hydrogel copolymer was developed by Gent University and details on the process and will be published in a separate paper. Cross-linked copolymer samples were prepared by free-radical polymerization. Cross-linkable modified poly(ethylene oxide-co-propylene oxide) (Pluronic) based monomers (PF127-BMA), and hydrolyzed methacrylic acid (MAA) were supplied by Gent University. 2.5 g of hydrolyzed MAA and 0.8 g of PF127-BMA were dissolved into 5.2 ml of nitrogen-flushed deionized water (DIW) to create a monomer solution. The monomer solution was mixed on ice to prevent thermal gelation of the PF127 and then refrigerated overnight. Ammonium persulfate (APS, Sigma-Aldrich) was used as an initiator for cross-linking. 4.56 g of APS were dissolved in 20 ml of nitrogen gassed DIW and then 0.75 ml of the mixture was added to the monomer solution. Tetramethylethylenediamine (TEMED, Sigma) was used as an accelerator. 2.98 ml of TEMED was diluted in 20 ml of nitrogen gassed DIW and then 0.75 ml of the mixture was added to the monomer solution.

The solution was poured into cylindrical glass molds, covered, and refrigerated for 1 h. The glass molds were then placed in a water bath at 37 °C for three hours. A thin glass mold with a 9 mm diameter was used for gel samples that underwent electroactuation. This was to promote even heating of the solution during curing so that a homogeneously crosslinked sample can be obtained. After curing, the gel samples were extracted from the molds and rinsed in DIW for a number of days to remove unwanted reagents. The water was refreshed until the gel reached a constant mass.

2.1.2. Poly(sodium acrylate)

Following the work of Tasaki, we synthesized a widely exploited polyacrylate hydrogel for comparison with the novel copolymer [26]. Briefly, 55 g of a 30% NaOH solution (by mass) were added 50 g of an 80% solution of acrylic acid (99%, Alpha Aesar). 20 mg of N-N'-methylbisacrylamide (Alpha Aesar), used as a cross-linker, were then added along with 90 mg of potassium persulfate, which was used as an initiator. The solution was poured into a mold and heated in an oven at 70 °C for 90 min. After curing, extraction and rinsing were performed as in the previous section.

2.2. Electrical testing

Although there exists no consensus regarding testing methods for quantifying the electric-response of hydrogels, two types of tests are typically performed when characterizing the electric response of gel samples in electrolytic solutions: bending and swelling measurements. The electrodes can either be in contact or not in contact with the gel at the time of actuation and this will have a profound effect on the mechanism responsible for the volumetric changes undergone by the gel. In the present work, bending tests using electrodes that are not in contact with the hydrogel were performed.

A home-made testing tank was designed for this purpose (see Fig. 1). The Perspex™ tank contains two parallel 30 mm × 30 mm × 0.17 mm Pt foil electrodes that are connected to a DC power source. An amperometer was connected in series in order to monitor the current. The effective size of the electrodes is dependent on the amount of solution filling the tank during experimentation. For the present study, this area was approximately

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