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A Green Approach to Electromechanical Actuation Based on Lotus Leaf-inspired Nanocomposite Hydrogels

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ABSTRACT

Carbon nanotube composite hydrogels are interesting candidates for applications like flexible conductors, actuators and artificial muscles owing to their favorable characteristics such as ease of functionalization, propensity to modulate the interfacial characteristics of the hydrogels and tunable electrical conductivity. Here we report the preparation, characterization and electromechanical actuation properties of Multi-walled Carbon Nanotube/polyacrylamide (MWNT/PAM) composite hydrogels. Various amounts of MWNTs (0.002, 0.005 and 0.01 wt%) have been introduced into polyarylamide (PAM) gel matrix by *in situ* polymerization of acrylamide/bisacrylamide mixture in presence of MWNTs. The effect of various parameters such as nanotube content, method of drying (vacuum drying and freeze drying) and partial hydrolysis of the composite hydrogels have been investigated by following water-uptake and electro-response behavior, scanning electron microscopy and contact angle measurements.

Key words: Multi-walled carbon nanotube, actuator, polyacrylamide, hydrogel, nanocomposite

INTRODUCTION

Stimuli-sensitive smart systems are gaining more focus now-a-days owing to their wide variety of applications ranging from robotics to health care. These systems instantaneously respond to external stimuli such as electric field, temperature, pH, ionic strength or mechanical stress by changing their appearance in terms of colour or dimensions. Among the different types of stimuli used for actuation, electric field offers many advantages such as ease of controlling the magnitude of the stimulus, possibility of miniaturization and portability of devices. Electromechanical actuation refers to the direct conversion of electrical energy into mechanical energy and materials capable of exhibiting the phenomenon are known as electro-responsive materials, which are used for application in robotics, prosthetic devices, electrically controlled drug delivery systems, micro-pumps and optic fibre switches. Several categories of electro-responsive materials have been reported, some of which include Ionomeric Polymer-metal Composites (IPMCs), conducting polymer blends, Interpenetrating Polymer Networks (IPNs) and nanocomposites based on ionic liquids. The actuation mechanism, performance parameters and hence the application of electromechanical actuators depend on their respective category. For example, conducting polymers like polyaniline undergo reversible dimensional changes in response to an applied electric field through electrochemical oxidation and reduction by a mechanism known as 'redox cycling'

(Bajpai *et al.*, 2008). On the other hand, IPMC based actuators involve electro-osmotic water transport across the material driven by diffusion of solvated cations (Onishi *et al.*, 2001). As far as the performance parameters are concerned, air-operable electromechanical actuators require very high voltage (>1 kV) for their operation and hence cannot be used for *in vivo* applications (Shankar *et al.*, 2007a). But, actuators operating in an electrolyte medium require only very small voltage (~1 V) and are suitable for biomedical applications (De *et al.*, 2002). Other parameters for comparing the performance of electromechanical actuators include actuation strain/displacement, work density and electromechanical coupling efficiency (Shankar *et al.*, 2007b).

Electromechanical actuators based on polymer hydrogels are attractive systems due to their excellent displacement/strain on actuation (~30%) and lower voltage of operation (3-10 V) arising from the flexibility offered by the hydrophilic domains of the polymer matrix and confined water molecules. Owing to these attributes, electroresponse of a variety of hydrogels of polymers such as polyacrylonitrile (Schreyer *et al.*, 2000), alginate/poly(diallyldimethylammonium chloride) IPN (Kim *et al.*, 2003), chitosan/poly(ethylene glycol) blends (Sun and Mak, 2001), conductive hydrogels of polyaniline-polyvinylalcohol-polyacrylic acid blends (Bajpai *et al.*, 2008) and polyacrylamide (Shamsudeen *et al.*, 2006) have been reported. Among these, polyacrylamide hydrogel has many attractive properties like biocompatibility, ease of fabrication and tunable functionality suitable for biomedical and bioseparation applications (Peppas *et al.*, 2006; Parthasarathy *et al.*, 2011; Gunavathi *et al.*, 2012). However, the poor mechanical stability and the dependence of mechanical properties on crosslinking density limit the practical applicability of polyacrylamide hydrogels (Shamsudeen *et al.*, 2006). Nevertheless, mechanical properties of the hydrogels can be improved by adding some reinforcing fillers like high aspect ratio nanomaterials like carbon nanotubes without affecting their function.

Carbon nanotubes are known since several decades for their excellent mechanical, electrical and thermal properties. They have been identified as promising candidates for actuators, owing to their interesting properties such as optimum temperature of operation, optimum voltage and significant work density (mechanical work performed after electrical actuation per unit mass of the actuator) per cycle due to their high Young's modulus (Baughman *et al.*, 1999). Actuators made directly with bucky papers or CNT mats exhibit bidirectional motion only at low charge density and are suitable mainly for active noise and vibration damping applications at low frequencies (Baughman *et al.*, 1999; Levitsky *et al.*, 2004). CNTs incorporated in polymer electrolytes like Nafion (Chattopadhyay *et al.*, 2002), CNT sheets or bucky papers sandwiching in between a layer of Nafion (Levitsky *et al.*, 2004) and CNT/ionic liquid composites (Mukai *et al.*, 2009) are capable of operating in air but they require higher operating voltages (>1 kV) and suffer fatigue during multiple cycles of operation. Composite hydrogels of CNTs with polymers like gelatin have been reported to have better mechanical stability and good electromechanical coupling (Haider *et al.*, 2007). In this context, we report here the swelling and electroresponse behaviour of multi-walled carbon nanotube (MWNT) composites of polyacrylamide hydrogels. The effect of parameters such as MWNT loading, drying conditions viz., vacuum drying and lyophilisation, on the swelling and electroresponse behaviour of MWNT/PAM composite hydrogels are investigated in this study, which could be useful for practical application of these actuators in robotics and micropumps.

MATERIALS AND METHODS

Materials: Multiwalled carbon nanotubes (MWNT) of size 20 nm diameter, 5-15 μm length, 95% purity were purchased from Nanostructured and Amorphous materials Inc., Houston, Texas

USA. Acrylamide, Sodium Dodecyl Sulphate (SDS), Ammonium Persulphate (APS) and Sodium Hydroxide pellets were purchased from Merck chemicals, Bangalore, India. N,N'-Methylene Bisacrylamide was procured from Himedia Chemicals India Ltd. Tetramethylethylenediamine (TEMED) and Hydrochloric Acid were purchased from SD Fine Chemicals Pvt. Ltd., Mumbai, India. Carbon Electrodes of dimension 70 mm length, 10 mm thickness and 10 mm width were bought from RajKart Enterprises, India.

Methods

Preparation of pristine PAM and MWNT/PAM hydrogels: The monomer solution was prepared by dissolving 7.1 g of Acrylamide and 1.54 g of Bisacrylamide (cross-linking agent) in 100 mL deionised water. Pristine PAM was prepared from 10 mL monomer solution, 0.1 mL of 10% SDS, 0.01 g of APS (polymerization initiator) and 5 μ L of TEMED.

MWNT/PAM nanocomposite of various concentrations were prepared as described below. The monomer solution with 10% SDS and various CNT loadings was sonicated, for uniform dispersion of CNTs in the monomer solution. APS and 5 μ L of TEMED were added after sonication. The prepared composition was casted in gel casting unit. Gels of thickness 1 mm were obtained.

Partial hydrolysis of hydrogels: The as-synthesized gels were hydrolysed, by immersing them in 8.3 M NaOH solution for 48 h. One set of partially hydrolysed gels was freeze-dried and another set was vacuum-dried for swelling studies. The remaining NaOH solution was back titrated with 1 N HCl solution in order to compare the extent of hydrolysis of MWNT/PAM gels with pristine PAM gels.

Drying procedure: The as-synthesized gels of uniform size were dried by two different methods, viz., lyophilisation (or) freeze-drying and vacuum drying. One set of gels were deep frozen at -72°C in an ultra low temperature freezer (New Brunswick Scientific, USA) for three days prior to lyophilization. Lyophilization was carried out at -88°C and 0.001 m bar vacuum atmosphere for one day in Lyophilizer (Christ, Alpha 2-4 LD plus, Germany). The other set was vacuum dried by continuous purging of vacuum into vacuum chamber (Tarsons Vacuum Chamber) for 3 h, using a vacuum pump (Vdo Hivac Technology, Chennai, India). The gels were maintained in vacuum atmosphere for two days, prior to experimentation.

Swelling studies: The prepared samples were dried by two methods, viz., lyophilization and vacuum drying, until a constant weight was obtained. Rectangular samples with the dimensions of 10 mm [length] \times 5 mm [width] \times 5 mm [thickness] were used for swelling experiment. Both hydrolysed and non hydrolysed or as-synthesized samples were used for swelling studies. Thus the studies were performed for four sets of samples, viz., Hydrolysed/Lyophilized, Lyophilized/non-hydrolysed, hydrolysed/vacuum dried and vacuum dried/non-hydrolysed. The dry weight of each sample was determined. Later the gels were immersed in 10 mL of deionised water at room temperature. Each sample, was taken out of water at regular intervals of 1 h and was thoroughly wiped using filter paper, to remove excess surface water and was weighed. Thus the wet weight was determined, until the gels attained a uniform equilibrium weight. The swelling ratio was calculated using the following equation:

$$\text{Swelling ratio(\%)} = \frac{W_o - W}{W} \times 100 \quad (1)$$

where, W_o is the wet weight and W is the dry weight of the sample.

Electro-response studies: The bending studies were performed for hydrolysed samples. Non-hydrolysed samples did not show bending phenomenon, due to the lack of ionic conductivity. In a home-made electrolysis tank of dimensions 100×100 mm, gels of uniform dimension (30×2×1 mm) were suspended between two parallel graphite electrodes (70×10×10 mm). The electrodes were placed 30 mm apart in 0.01 M NaOH solution. At a voltage of 6 V, the bending behaviour of the samples viz., Pristine PAM and MWNT/PAM composites with varying CNT concentrations were studied for 5 cycles.

Contact angle measurements: The contact angle measurements of vacuum dried pristine PAM and 0.01% MWNT/PAM composite were made in order to study the hydrophobicity of the gels. A standard goniometer (Ramehart, Model No. 250-F1, Serial No. 805193) with Drop Image Advanced software was used to measure the contact angle. Sessile drop method was employed.

Scanning electron microscopy: The gel morphology of the vacuum dried, non-hydrolysed samples, viz., pristine PAM, 0.002, 0.005 and 0.01% MWNT/PAM composites were characterized using Field Emission Scanning Electron Microscope (JSM 6701F, JEOL, Japan). The samples were mounted on a brass stub using carbon tape and made conductive by sputter coating it with Platinum. The samples were then loaded into the specimen chamber and were imaged at 3 kV, at a working distance of 7.3 mm.

RESULTS AND DISCUSSION

Water-uptake capacity of polymer hydrogels depends on several factors like hydrophilicity of the polymer domains, elasticity of the gel matrix, method of drying and temperature. Accordingly, swelling studies were conducted on as-prepared and hydrolyzed gel samples of polyacrylamide and compared with the behaviour of MWNT/PAM composite hydrogels containing different amounts of MWNTs viz., 0.002, 0.005 and 0.01 wt%. Swelling studies are generally performed on dry gels. However, the method of drying could affect the gel morphology especially the pore size and in turn might affect the water uptake capacity. Hence, in this investigation, one set of samples was subjected to freeze-drying at -72°C for three days and then lyophilizing the same at -88°C and 0.001 m bar vacuum and another set of samples was subjected to vacuum drying using a 10⁻¹ Pa diffusion pump before swelling. Photographs of gels prepared under various conditions are compiled in Fig. 1. Figure 2 compares the swelling kinetics of four types of pristine PAM gels: As-prepared, hydrolyzed, vacuum dried and lyophilized. The swelling behaviour of hydrolyzed gels was found to be more affected by the method of drying than the as-prepared non-hydrolyzed gels, which could be attributed to the increase in local hydrophilicity imparted by the partial hydrolysis of amide groups to carboxyl groups. Among the hydrolyzed gels, those dried under vacuum swelled to a lesser extent than the lyophilized gels. This could be understood based on the mechanism of lyophilization, in which water present in the pores of the hydrogel is frozen to ice and then sublimed to vapour. Freezing of water inside the pores increases the pore size, which remain as voids when the ice is sublimed. As a result, lyophilized samples have bigger pore size and hence can

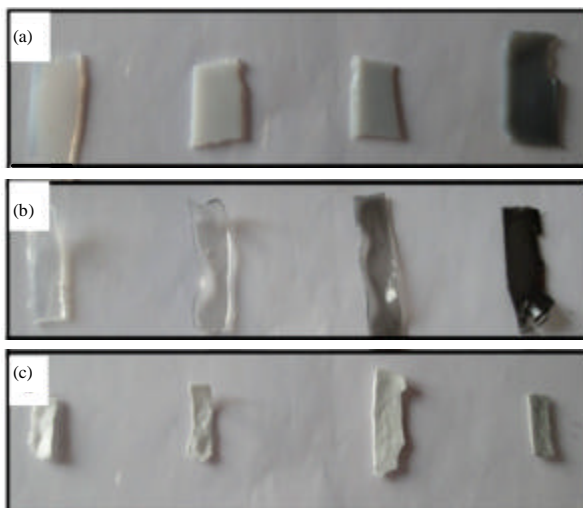


Fig. 1(a-c): Photographs of gels with different loadings of MWNTs (0, 0.002, 0.005 and 0.01 wt%) (left to right) (a) Before drying, (b) After vacuum drying and (c) After freeze-drying

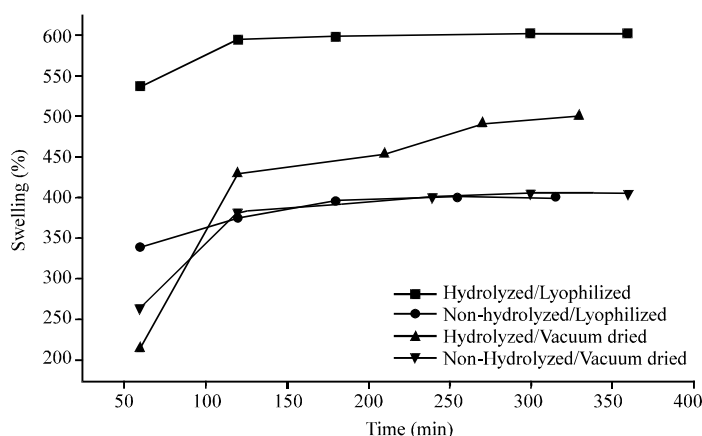


Fig. 2: Effect of hydrolysis on the swelling behavior in de-ionized water of vacuum-dried and freeze-dried pristine polyacrylamide hydrogels

uptake more water, swelling to a greater extent than vacuum dried gels. The effect of drying method on pore size has been reported for even cement based materials (Galle, 2001). Further, swelling behaviour of all the gels viz., hydrolyzed and non-hydrolyzed, vacuum dried and lyophilized, with varying loading of MWNT is compared in Fig. 3. In all the four conditions, pristine PAM gels were found to swell more than MWNT/PAM gels probably because of an increase in stiffness of the gels due to the incorporation of nanotubes in the matrix. This is also supplemented by contact angle measurements, in which the composite gels absorbed less water and equilibrated quickly when compared to the pristine gel (Fig. 4). From the contact angle measurements, the lyophilized gels were found to be highly hydrophilic and hence snapshots of water absorption could not be recorded. In the case of vacuum dried non-hydrolysed gels, pristine PAM swells faster than MWNT/PAM. This may be due to higher rigidity of PAM gel conferred by the nanotubes. At the

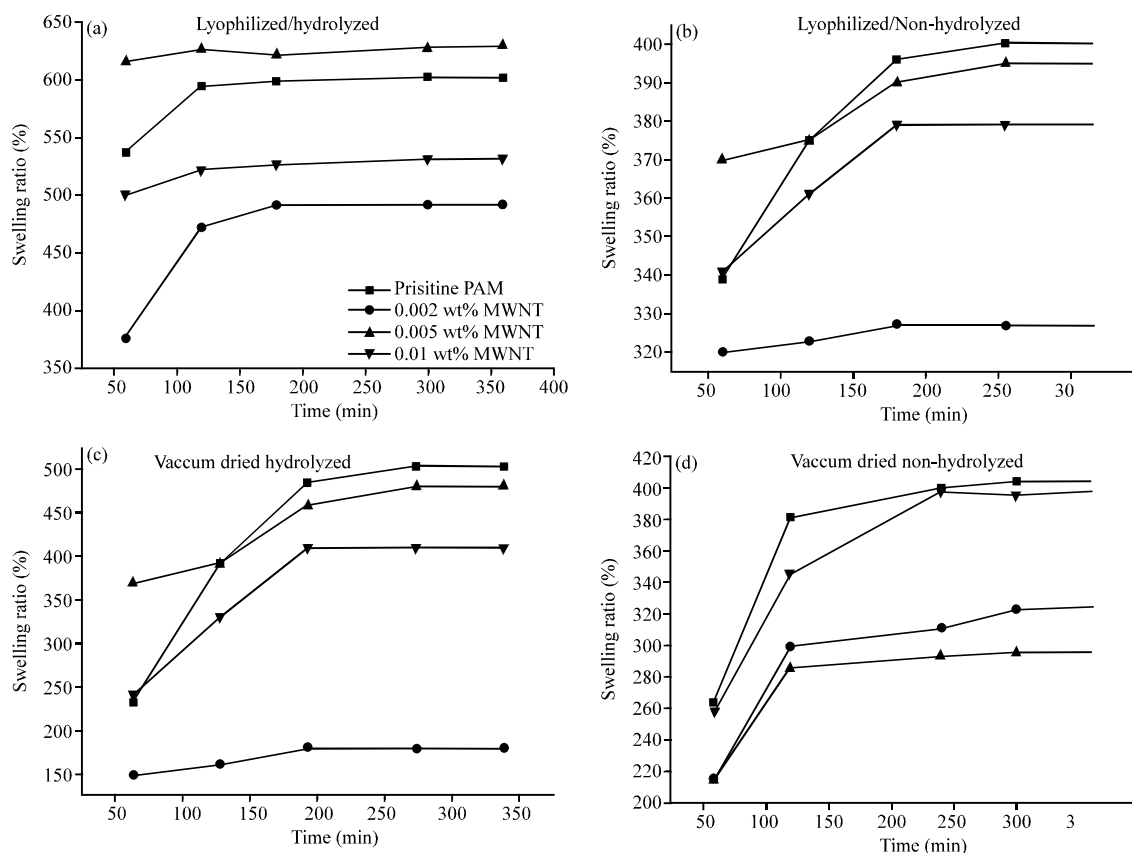


Fig. 3(a-d): Effect of hydrolysis on the swelling behavior in de-ionized water of vaccum-dried and freeze-dried polyacrylamide hydrogels with varying loading of multi-walled carbon nanotubes (0, 0.002, 0.005 and 0.01 wt%)



Fig. 4(a-b): Photographs showing the absorption of a water droplet with time (a) By a dry pristine PAM gel and (b) By a 0.01% MWNT/PAM composite gel

same time, MWNT/PAM composite attains equilibrium faster than pristine PAM. In addition, back titration of NaOH solution after hydrolysis with HCl indicated that the extent of hydrolysis was

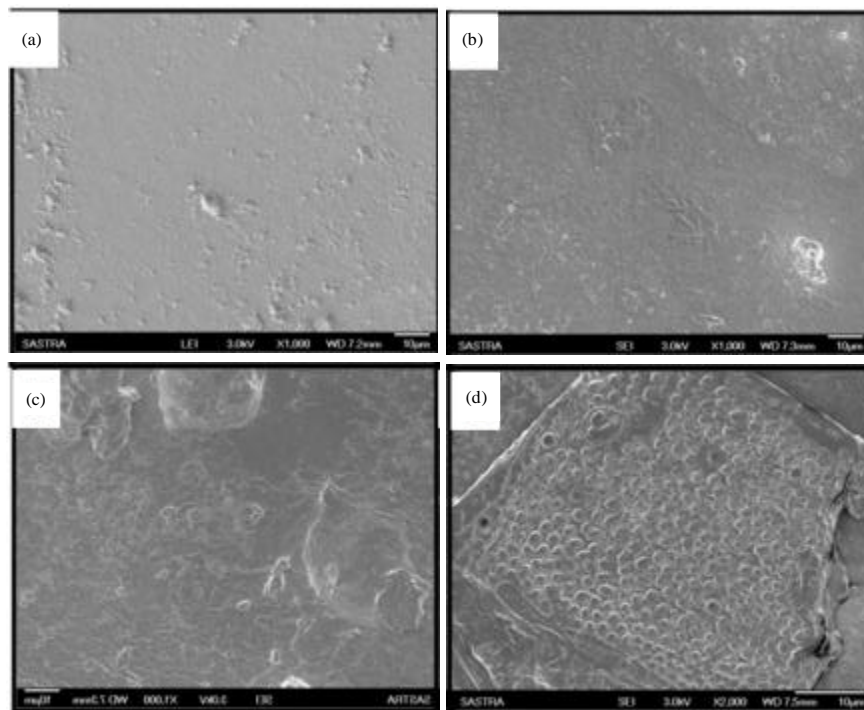


Fig. 5(a-d): Scanning electron micrographs of vacuum-dried hydrogels, (a) Pristine PAM, (b) 0.002 wt% MWNT/PAM, (c) 0.005 wt% MWNT/PAM and (d) 0.01 wt% MWNT/PAM

more for pristine PAM gel than the nanocomposite gels. This could be compared with a similar observation reported earlier, in which introduction of MWNTs into the polymer matrix reduced the water uptake of Nafion membrane (Kannan *et al.*, 2009). In addition, the surface of the gels were found to become rougher on increasing the nanotube content as evident from the SEM images shown in Fig. 5.

Figure 3 further indicates that, in the case of lyophilized gels, whether hydrolyzed or non-hydrolyzed, there was no significant difference in swelling kinetics with varying nanotube loading, as indicated by the slope of the swelling curves. A possible reason for the invariance could be that lyophilisation often results in large pore sizes ($\sim 1-10 \mu\text{m}$) which reduces the probability of the presence of nanotubes at the gel/water interface in order to show any local hydrophobicity effect (Dinu *et al.*, 2007). Unlike the case of pristine PAM shown in Fig. 1, vacuum-dried non-hydrolyzed samples behave differently from lyophilized non-hydrolyzed samples with increasing nanotube content in the gel matrix. This observation augments the interpretation of the previous observation in terms of the large pore size and lack of local hydrophobicity effects in the case of lyophilized gels. On the other hand, vacuum dried non-hydrolyzed gels did not exhibit significant changes in the swelling kinetics with varying MWNT content while vacuum dried hydrolyzed gels exhibited noticeable difference in swelling kinetics with nanotube content. However, a linear plot of t/w versus time, where t/w = time/weight of solvent absorbed per unit weight of the gel was obtained for all the MWNT/PAM composite gels like pristine PAM gel, indicating similarity in swelling mechanism resulting in a second order swelling kinetics (Haider *et al.*, 2007).

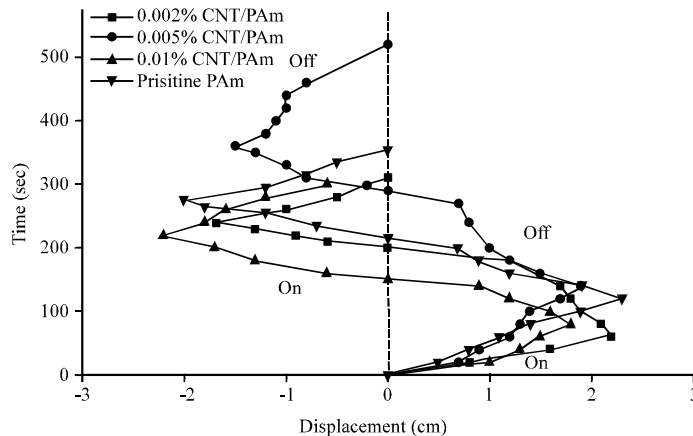


Fig. 6: Electro-response of partially hydrolyzed hydrogels with varying loading of MWNT (0, 0.002, 0.005 and 0.01 wt%) suspended in one end in 0.01 M NaOH solution between two parallel graphite electrodes

Actuation properties of the nanocomposite hydrogels in presence of an applied electric field were investigated in a home-made set up by suspending the gel (one-end fixed) in between two parallel graphite electrodes. Electromechanical actuation was noticed only in the case of partially hydrolyzed gels but not in the case of non-hydrolyzed gels. In addition, when a dc voltage was applied across the electrodes, the gel bent toward the cathode initially, came back to the equilibrium position when power supply was switched off and then bent in the opposite direction i.e., again toward the cathode on inverting the polarity of the electrodes as shown in Fig. 6. However, gelatin hydrogels have been reported to bend initially toward the anode and then self-oscillate in both the directions without the need for inverting the polarity of the electrodes (Haider *et al.*, 2007). It should be noted that despite both gelatin and partially hydrolyzed polyacrylamide having negative charges, the former bends toward the anode while the latter toward the cathode; the former operates in a bimorph configuration while the latter does so only when the polarity is inverted manually. These observations provide an interesting insight into the mechanism of electro-response. Gelatin actuators have been reported to work based on differences in osmotic pressure across the gel created by the diffusion of ions present in the electrolyte toward the oppositely charged electrodes; as gelatin is negatively charged, positively charged sodium ions in the electrolyte will accumulate first in the anode side, as they tend to move toward the cathode and as a result, the gel bends toward the anode. However, in the present case of partially hydrolyzed polyacrylamide gels, protons present in the carboxyl groups (formed during hydrolysis of amide groups) are extracted toward the cathode and the gel bends toward the cathode probably due to anisotropic swelling of gels. This could be compared with reports in the literature in which local pH changes during electro-response of hydrogels have been demonstrated. At the applied voltage of 6 V, water electrolysis occurs and there is a natural pH gradient in the system which will be 2.1 around the anode, 7.9 around the cathode and 3.8 between the two electrodes (Yuk *et al.*, 1992; Kaetsu *et al.*, 1992). Hence the initial bending of PAM gel toward the cathode could be partly due to the extrusion of protons from partially hydrolyzed gels due to alkaline pH generated around the cathode. A similar mechanism of electro-response has been reported for Single-walled carbon

nanotube/Nafion/Single-walled carbon nanotube type actuators (Levitsky *et al.*, 2004). Nevertheless, Nafion-based actuators suffer from irreproducibility of results due to the humidity-dependent proton conductivity of the polymer electrolyte.

Electro-response of partially hydrolyzed pristine PAM gel is compared with that nanocomposite PAM gels prepared with different loadings of MWNT. Nanocomposite PAM gels were observed to respond more quickly when compared to pristine PAM gel as evident from the graph plotted for pristine PAM, 0.002 and 0.01 wt% MWNT. However, the behaviour of 0.005 wt% MWNT/PAM off the trend, in all the three replicate measurements, could not be explained at the present level of investigation. On the other hand, only 0.01 wt% MWNT/PAM showed some difference in displacement when compared to pristine PAM gel, on reversal of electrode polarity. These results obtained here with pristine PAM are comparable with the reported data (Shamsudeen *et al.*, 2006). Yang *et al.* (2008) have also prepared SWNT/poly (N-isopropyl acrylamide) composites by infiltrating the acrylic monomer and crosslinker through an array of pre-fabricated SWNTs followed by polymerization (Yang *et al.*, 2008). This approach enabled the observation of conductivity differences between pristine polymer gel and nanocomposite gels as the nanotubes were protruding on the surface of the gels. However, the conductivity of the composite gels was found to be sensitive to changes in humidity and temperature, which might limit their application in artificial muscles.

In summary, the present report demonstrates the electromechanical actuation properties of multi-walled carbon nanotube/polyacrylamide composite hydrogels for applications like robotics and microfluidics. While previous reports involved significantly higher loading of carbon nanotubes (upto 8 wt%), the present report employs very low loading ranging from 0.002 to 0.01 wt% to improve the operational life and decrease the response time of polyacrylamide hydrogel actuators.

CONCLUSION

Cross-linked polyacrylamide hydrogels modified with Multi-walled Carbon Nanotubes have been demonstrated to show electromechanical actuation. Unlike previous reports on CNT composite hydrogel actuators, the present report involves very less amount of nanotube loading ranging from 0.002 to 0.01 wt% and a simple and cost-effective fabrication procedure. Increasing nanotube loading within the above range was found to affect various properties of the gel like water uptake, electro-response behaviour and surface roughness as evident from SEM. However, significant variations in electrical conductivity with varying nanotube loading was not observed due to the lack of alignment of nanotubes within the gel matrix, which would otherwise affect the actuation behaviour in terms of anisotropic and humidity/temperature-sensitive conductivity effects. In addition, the method of drying was observed to affect the water uptake behaviour of the gels with lyophilized gels taking up more water than vacuum-dried gels. The results of the present investigation imply the use of very small amounts of carbon nanotubes to modulate the properties of polyacrylamide gels which could find applications including artificial muscles, drug delivery, robotics and micropumps.

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