



Electromechanical performance of chitosan-based composite electroactive actuators



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ABSTRACT

A novel chitosan-based electroactive actuator was fabricated by using chitosan, *N, N'*-methylenebisacrylamide (MBA), poly (diallyldimethylammonium chloride), and gold metal. The constructive effect of crosslinker to the actuator performance was investigated by using various amounts of MBA. The chitosan-based film samples were characterized using Fourier transform infrared analyses, X-ray diffraction analysis, thermogravimetric analysis, scanning electron microscopy analysis, and tensile test. The viscoelastic properties of films were determined by dynamic mechanical analysis. The motion and force generation capabilities and the repeatability of the actuators were also investigated under electrical stimuli up to 21 V. The suitability of the prepared chitosan-based films to be used as actuator for soft robotic applications is verified experimentally.

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1. Introduction

Electroactive polymers (EAPs) are smart polymers that response to electrical stimulation [1]. Ionic polymer metal composite (IPMC) is an electro active polymer that has many application areas such as artificial muscles, humans catheter systems, underwater robotics fishes, micro pumps, transducers etc. IPMC has invaluable properties like exhibiting large tip displacement under low voltage, being flexible, easy actuation, light-weighted, and simple manufacturing [2]. Besides these properties, biocompatibility is important and desired property for low voltage actuatable materials [1]. Chitosan (Chi) is a natural, biocompatible and biodegradable polycationic polysaccharide. It is derived from chitin which is the second most abundant natural polymer on the earth [3,4]. Chi has many application areas such as drug delivery [5], biosensors [1], electroactive polymers [6] due to its non-antigenicity, bioactivity, and non-

toxicity [7]. Chi was investigated in many researches on electroactive polymers [8–11] to enhance the actuation performance [12].

In this study, a novel Chi-based actuator was prepared and actuation behaviour has been investigated in details. Chi was crosslinked with *N, N'*-methylenebisacrylamide (MBA) through free radical polymerization to improve mechanical properties and the electroactive properties. The crosslinking concentration affects the stiffness, number of functional group, etc., of the sample structure and hence affects the electroactive performance [13–17]. Poly (diallyldimethylammonium chloride) (pDADMAC), a linear positively charged polyelectrolyte [18], was used to increase the ion mobility in chitosan based actuator. The novelty and originality of this study is combining the unique properties of Chi, MBA, and pDADMAC in one structure, and as a result obtained actuators have invaluable electroactive properties.

The actuation performances of samples were investigated in terms of tip displacement and force generation capability of the actuators under various DC voltage excitations. Besides, Fourier transform infrared (FTIR), thermogravimetric analysis (TGA), scanning electron microscopy (SEM), X ray diffraction (XRD),

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dynamic mechanical analysis (DMA) and tensile test of the samples were carried out to characterize chitosan-based films.

2. Materials and methods

2.1. Materials

Chi (low-viscosity, 50494), acetic acid ($\geq 99.85\%$), *N, N'*-methylenebisacrylamide (146072-100G), *N,N,N',N'*-tetramethylethylenediamine (T22500) (TEMED), ammonium persulfate (248614) (APS), pDADMAC [20 wt% in H₂O], and polyethylene glycol (PEG) with average molecular weight of 1450 g/mol were purchased from Sigma–Aldrich, and gold leaf (thickness of 0.14 μm) was purchased from L.A. Gold Leaf.

2.2. Fabrication of chitosan-based films and actuators

The flow chart of the procedure to prepare the composites was shown in Fig. 1. The details of procedure are as follows: 2.3% (w/v) Chi solution was prepared by using 2% (v/v) acetic acid. The solution was stirred overnight at room temperature to obtain homogeneous solution. Afterwards, appropriate amounts of PEG and pDADMAC were added to the chitosan solution. On the other hand, 0.0123 g of MBA was dissolved in 1.87 mL distilled water (dw), and 0.230 mL of 4.34% (v/v) TEMED and 0.15 mL of 0.876 M APS solutions were added to the MBA solution. The mixture was allowed to stand in a water bath at 100 °C for 20 min, and then added to the Chi solution. Afterwards, the mixture was stirred for 24 h at room temperature and cast into petri dish, and dried at 80 °C for 16 h in a vacuum oven. The same method was repeated by using 0.032 g and 0.1 g of MBA as crosslinker. The films containing 0.0123 g, 0.0320 g, and 0.1000 g of MBA were coded as ChiPM-1, ChiPM-2, and ChiPM-3,

respectively. The films were coated with above-stated gold material to fabricate Chi-based actuators.

2.3. Characterization methods

FTIR analyses of Chi-based films were conducted by using in Perkin Elmer Spectrum BX-II FTIR spectrometer. FTIR spectra were recorded in the range 400–4000 cm^{-1} with 25 scans at a resolution of 2 cm^{-1} . The morphologies of Chi-based films were examined by using FEI Quanta FEG 250 SEM instrument, with an operating voltage of 5 kV. The samples were cut in transverse section and coated with gold by means of plasma sputtering method. Thermogravimetric analyses of the samples were carried out using TGA (Shimadzu, TGA 50). Analyses were performed under the under nitrogen atmosphere with a flow rate of 1.0 mL/min. The samples were heated from 30 to 600 °C with a rate of 10 °C/min. XRD analyses of Chi-based films were done via a Bruker D2 Phaser desktop diffractometer with a Lynx Eye detector. A Cu-K α radiation ($\lambda = 1.54 \text{ \AA}$) generated at a 30 kV voltage, and a 10 mA current was used. The XRD instrument was set to operate in continuous PSD fast scan mode. The samples were scanned from 0° to 70° (2 θ). The scan step size and duration were set to be 0.012° and 14.4 s, respectively. The crystallinity values were obtained by the DIFFRAC.EVA V3.0 software. The tensile strength and Young's modulus were measured by using tensile testing machine with a 50N load cell at a crosshead speed of 0.1 mm/min. DMA analyses of the samples were conducted by using TA Instrument Q800 Dynamic Mechanical Analyzer. Specimens (13 mm \times 6.40 mm \times 0.10 mm) were measured in film tension mode at a frequency of 1 Hz. The samples were heated from 20 to 200 °C at a heating rate of 10 °C/min. Water uptake ratios of film samples were also determined according to the procedure given elsewhere [19]. Water uptake values for ChiPM-1, ChiPM-2, and ChiPM-3 were obtained to be 81, 80, and 67%, respectively. The lowest water uptake ratio belongs to the film having the greatest MBA ratio.

2.4. Experimental setup for actuator characterization

For electroactive characterization of actuator samples, their capability to move and generate force was analyzed experimentally. The tip displacement of the actuator samples was observed via machine vision system, which consists of a camera (Basler acA2040-180 km camera) and a frame grabber (NI PXIe-1435) running on industrial PC (Computer M0814-MP – 8 mm – F1.4 lens). The blocking force of the actuator was measured by a precision balance (Precisa 225SM-DR). For both of the experiments, the input signals were generated via analog output card (NIPXLe-9133pc) and amplified with a buffer (TDA2040). All the input/output and control codes were developed by ourselves. The vision data were analyzed using Kinovea. The experimental setups are shown in Fig. 2. All tip displacement and blocking force experiments were carried out in air at room temperature.

3. Results and discussions

3.1. FTIR analysis

Fig. 3 represents FTIR spectra of Chi powder and Chi-based films. A broad band from 3600 to 2800 cm^{-1} in the spectrum of Chi corresponds to the associated –OH, –NH, and –CH stretching vibrations of Chi. The vibration of amine group of Chi is observed at 1562 cm^{-1} in the spectrum of Chi [20]. Bending vibrations of methylene and methyl groups of ChiPM films are seen at about 1400 cm^{-1} [21]. The absorption bands at 1150–1000 cm^{-1} are associated with vibrations of C–O group [22]. OH stretching

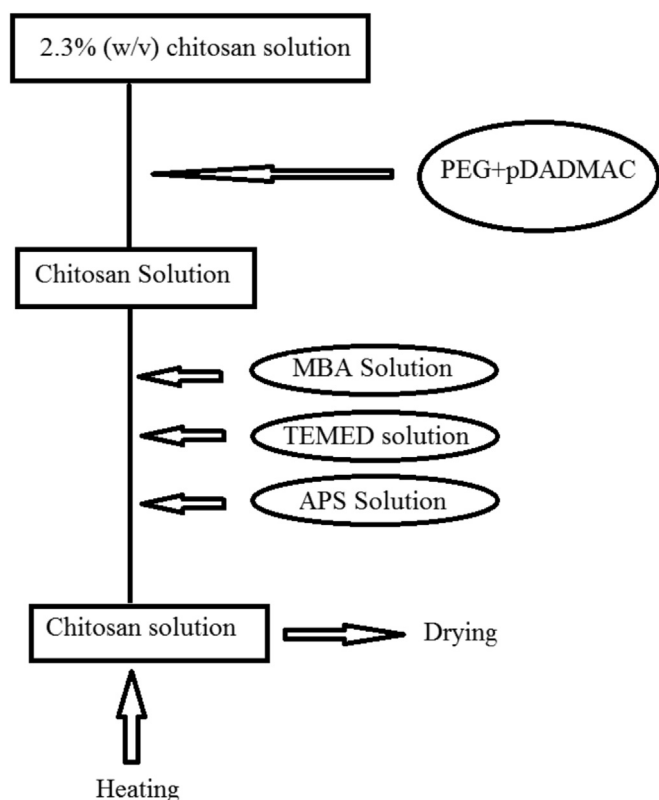


Fig. 1. The flow chart of the procedure.

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