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# Preparation and electromechanical properties of the chitosan gel polymer actuator based on heat treating

Gang Zhao [a], Junjie Yang [a], Yujian Wang [a], Honghao Zhao [b,*], Zhijie Wang [a]

[a] Institute of Intelligent Manufacturing and Robotics, College of Mechanical and Electrical Engineering, Harbin Engineering University, Harbin 150001, People's Republic of China
[b] Department of Decision Science, School of Business, Macau University of Science and Technology, Macau, People's Republic of China

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ABSTRACT

As a new kind of smart materials for fabricating ionic electric actuator, Chitosan Gel Polymer (CGP) known as bionic artificial muscle, has broad application prospects and high academic value with advantages of simple structure, good biocompatibility and large deflection under low driving voltage (≤5 V). In this paper, effects and enhancement mechanism of the heat treating optimization technology on response speed performance of the CGP actuator were mainly investigated. Furthermore, its preparation process including material selection, membrane fabrication and the assembly along with fabrication principle were deeply researched. The CGP actuator consisted of two parts; electric actuating membrane which is sandwiched between non-metallic electrode membranes. On one hand, high viscosity chitosan inside the actuating membrane served as framework which gives access to anions and cations within the electrode membranes. On the other hand, Multi-walled Carbon Nanotube (MCNT) was adopted to modify chitosan in the electrode membrane for obtaining good conductivity and stable chemical character. Moreover, response speed of the CGP actuator that underwent repeated heat treating for a short time, would be continuously improved with the increase of heat treating times. But as the heat treating time prolonged, electromechanical properties of the CGP actuator began to degrade. In addition, since conduction velocity of microscopic ions inside the CGP actuator was presented as macroscopic current magnitude, the experimental results revealed that the changing trend of response speed was almost in accord with the current trend of the CGP actuator. Both of these are fitted in a quadratic function relationship.



## 1. Introduction

At present the CGP actuator, a kind of emergingionic Electro Active Polymer (EAP), can change its shape and size under electric field with a green actuating method. It is called "artificial muscle" because of its soft texture, high elasticity, embrace quiet operation and biological-like actuating performance [1,2], which can replace many machine parts like bearings and gears for bio-mimetic robots development. The CGP actuators are flexible, compact and able to promote energy efficiency of complex actuating mechanisms by eliminating their power trains [3,4]. Meanwhile, the CGP with self-sensing ability is capable of contributing to develop a efficient and compact inchworm inspired micro-robot for underwater applications [5]. Generally, the CGP is applied as shanks and thighs for underwater robots, such as a Lobster robot with multi-functionality, cheap monolithic fins with complicated deformations and jellyfish robot with accurate bell deformation. Besides a natural biopolymer, chitosan can be made into an electrically sensitive hydrogel that has significant deflection under applied voltage. It has many good characteristics such as easy preparation, low cost and high biodegradability which makes it a potential research candidate in mechanical engineering and intelligence material fields [6,7]. Specifically, a chitosan-based electric actuator is manufactured with gold electrode, which is cross-linked with methylene bis propyl amide [8–11]. Then modification of electrode film is further studied to improve its performance, with silver nanoparticles plated on the surface of graphene [12]. But at this stage, their fabrication methods are still immature and the raw materials are expensive heavy metal and ionic liquid with little commercial value [13,14]. In particular, they often utilize a coupling agent, glutaraldehyde that is highly toxic to human body [15]. Thus, in this work, an innovative and reliable preparation process by optimizing traditional preparation processes for the CGP actuator is provided in detail and its principles are analyzed comprehensively. In the proposed process, all the materials are biocompatible and had no toxic side effects, which fits the environmental protection concern with

* Corresponding author.
*E-mail address:* hhzhao@must.edu.mo (H. Zhao).

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high efficiency and low cost. The twice dispersed MCNT is used to make the electrode membrane with superior mechanical and conductivity performance [16]. Each part of the process is researched theoretically and experimentally to ascertain the optimal primary preparation process parameters.

Moreover, the slow response speed of the CGP actuator is still a big hindrance in widening its applications scope in bionic engineering, Micro-electromechanical Systems (MEMS) and Bio-science [17,18]. It is important to find effective techniques to elevate its unstable response characteristics. Unfortunately, investigating the key factors of response speed, output force and life cycle usually brings much workload for researchers due to the complexity on measuring different performance indexes and the long time for preparation process. Currently, displacement behavior of the folded dielectric elastomer actuator is studied and then response lifetime of the single walled carbon nanotube based bioinspired actuator is tested under different voltages [19,20]. So scientists usually attempted to improve the electromechanical properties of actuators by fabrication method, doping technology or organics cross-linking [21,22]. However, they often neglect the effects of further heat treating of newly developed CGP actuator which may improve its response capability. In this paper, heat treating technology of the CGP actuator is explored earnestly to investigate its effect and reinforcement mechanism on the response speed characteristic just like the metal heat treating process where mechanical properties could be greatly improved [23,24]. Moreover, this technology is green and its fine results on the CGP actuator structure could ameliorate the tangled crosslinking network structure because hydrolyzing of chitosan in acid solution raises ion motion rate inside it. An experimental setup is established to measure the CGP actuator deflection and its electrical conductivity so that its electromechanical properties can be characterizes and the proposed preparation process can be validated.

## 2. Preparation process

### 2.1. Materials and equipment

Main materials used in our experiment included MCNT dispersion liquid (10 wt%), acetic acid (analytical reagent), chitosan powder (high viscosity and 85% deacetylation degree), distilled water (homemade) and glycerin (AR). In addition, key instruments required were analytical balance (FA1004), ultrasonic cleaner (LT-05C), magnetic stirrer (MS300), ultrasonic cell disruptor (JY92-11 N), water distiller (10 L), hot press (4V210-08), vacuum drying oven (DZF-6020) and home-made glass mould (50 mm × 50 mm × 5 mm).

### 2.2. Process parameters analysis

#### 2.2.1. The twice dispersed MCNT

MCNT is easy to intertwine to form large aggregates and precipitate during storage because of its small diameter, big specific surface area, strong adsorptivity and Van der Waals'force between molecules [25]. Thus before adding MCNT dispersion liquid to the chitosan solution, the dispersion liquid needed to be dispersed for the second time in order to improve the uniformity and deaggregation, which was beneficial to the surface quality of electrode membrane, as shown in Fig. 1. It was illustrated that the surface quality of electrode membrane before MCNT secondary dispersion was much more rough and chapped than that after the dispersion. In this work, MCNT dispersion liquid was dispersed for the second time by using ultrasonic cell disruptor for 30 min.

**Table 1**
Drying process parameters.

| Drying temperature (°) | $\delta$ | |
|---|---|---|
| | The actuating membrane solution | The electrode membrane solution |
| 60 | 0.08 | 0.08 |
| 70 | 0.07 | 0.07 |
| 80 | 0.05 | 0.05 |

#### 2.2.2. Vacuum degree

The readings from vacuum gauge of vacuum drying oven used in experiment were relative to vacuum degree rather than to the absolute vacuum. According to the numerical range of vacuum gauge, they are related by the following formula:

$$P = 1 \times 10^5 \left(1 - \frac{\delta}{0.1}\right), \quad (1)$$

where $P$ (Pa) was the absolute value of vacuum degree and $\delta$ was the absolute value of indication scales on vacuum meter.

On the basis of vacuum working chamber of the drying oven, water could be evaporated rapidly from the membrane solution at a low drying temperature which avoided the damages to chitosan structure and the membrane surface quality under high temperature condition. Since the concentration of acetic acid solution was relatively low (about 3%), the relationship between saturated vapor pressure and drying temperature of the membrane solution could be approximately analyzed by that of water [26]. Then based on the relationship, the corresponding $\delta$ could be calculated by Eq. (1) to design experiments.

#### 2.2.3. Determination of drying temperature

Experimental results revealed that drying temperature and vacuum degree had great influences on membrane-forming performances of the actuating and electrode solutions. Their membrane formation was not a simple process of water evaporation, but was a sequential crystallization process under controlled temperature [27]. At lower drying temperature, Chitosan crystallinity promoted with small degradation but its grains obtained were too small in size to successfully form a membrane with good mechanical properties. When the drying temperature was 100°, chitosan was almost not crystallized with poor mechanical properties [28]. By considering available technique and technological error, the actuating membrane with few bubbles and surface defects was unavoidable, which had little influence on its conductivity and response speed. Furthermore, during drying process of the membrane solution, owing to high concentration of chitosan in acetic acid solution, the vacuum degree should be low enough to avert rapid water evaporation; otherwise large numbers of bubbles produced would seriously damage the membrane surface quality and electromechanical characteristic. The level of vacuum degree was chosen according to the observed performance of the CGP actuator. When the membrane was dried under a lower vacuum, evaporation of water was slow, and chitosan molecules moved slowly which was not conducive to the formation of ordered structure, which resulted in declining of the mechanical strength. Besides, the actuators started to behave differently for a value near to the lowest vacuum degree and the surface topography was illustrated clearly in a value slightly higher than the highest vacuum degree. These discrete degrees were typical values in various stages of the CGP characteristics and could greatly optimize experiment times but would not reduce the test feasibility. Accordingly, based on the parameters shown in Table 1, three groups of control experiments were executed respectively for the actuating and electrode membranes.

As shown in Fig. 2, the surface of electric actuating membrane involved lots of bubbles under 60°, small amounts of bubbles under