



Graphene/gelatin hydrogel composites with high storage modulus sensitivity for using as electroactive actuator: Effects of surface area and electric field strength



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ABSTRACT

The electromechanical properties of graphene/gelatin hydrogel composites were investigated under the effects of graphene surface area, electric field strength and temperature towards bio-actuator applications. The highest surface area of an embedded graphene (MG; grade M) in the gelatin hydrogel composites induced the highest dynamic modulus (G') under applied electric field. The 0.1 vol% graphene (MG)/gelatin hydrogel composite possessed the highest $\Delta G'/G'_0$ value of 352% in comparison with other materials in previous studies. Even the lowest $\Delta G'/G'_0$ values obtained from the fabricated graphene/hydrogel composites were still greater than other dielectric elastomer materials investigated. The storage moduli of the pure gelatin and graphene (MG)/gelatin hydrogel composites, between 30 °C and 90 °C, exhibited three distinct regimes. In the deflection experiment, the bending distance and the dielectrophoresis force were found to increase monotonically with applied electric field strength with a deflection toward the anode side, indicating the attractive force between the anode and the polarized carboxyl group as the gelatin structure possessed negative charges under applied electric field.

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1. Introduction

Electroactive polymers (EAPs) have been developed for many applications such as compliant electrode [1], drug delivery engineering [2], actuator [3], electroactive paper [4], and many others [5] that offer distinct characteristics: high energy density, light weight, and great flexibility. Hydrogels are promising materials for development of EAPs because they provide a reversible response to external stimuli such as ionic strength [6,7], pH [8,9], temperature [10–13], and electric field [14–17]. Gelatin is a hydrogel or EAP with a high molecular weight protein produced from animal collagen by thermal and hydrolyzing processes with either acids or bases. It can be used as a stable film, hydrogel, and composite [18]. However, gelatin possesses poor water resistance and low mechanical properties because the oriented structure is split into a random coil

structure after treating through acid or base at high temperature, these restrict its possible application as an EAP [19]. Therefore, gelatin needs to be reinforced through either chemical crosslinking or use of filler materials. Chemical crosslinking enhances thermal and mechanical properties through covalent bonds between the reactive side groups in the gelatin molecules. However, this process produces the residual crosslinking agents, leading to several toxic side effects. The use of graphene embedded in gelatin has been studied by Wang et al. [20]. They found that graphene/gelatin gel composites exhibited high tensile strength compared with pure gelatin films in both dry and wet states. Zhang et al. [21] investigated thermal and mechanical properties of sodium alginate/graphene oxide composites. Graphene oxide (GO) enhanced the stability of sodium alginate without crosslinking chemicals because of dispersion and compatibility of GO in the polymer matrix. Graphene, one of the elementary carbon structures (diamond, fullerene, and carbon nanotube), is a single atom layer of carbon that is arranged in a two dimensional monolayer [22]. It has very promising properties, including high electrical conductivity at ambient temperature [23], good thermal conductivity [24], large surface area [22], and outstanding mechanical properties in term of

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Young's modulus [25], compared with diamond, fullerene, and carbon nanotubes (CNTs). In addition, graphene is much easier to produce and at a lower price. Thus, graphene is a suitable carbon based reinforcement filler used for enhancing mechanical performance of polymer matrix [26]. For example, Liang et al. [26] studied graphene oxide/PVA nanocomposite using GO as a reinforcement filler. Tensile strength and young's modulus were increased by 76% and 62%, respectively when compared with neat PVA through the distribution of 0.7 wt% GO. Therefore, graphene is an excellent-reinforcing material for polymers that have been developed for several applications such as electronic and optoelectronic devices, bio-chemical sensors, nanocomposites, and energy storage [27].

The objective of present work was to investigate the electro-mechanical properties of graphene/gelatin hydrogel composites containing an anionic surfactant (i.e., sodium dodecylsulfate) as candidate materials for bio-actuator applications. The electrical, thermal, and electromechanical properties were investigated and measured under the influences of graphene content and surface area, electric field strength, and temperature.

2. Experimental

2.1. Materials

Gelatin (Type B, bovine skin) and sodium dodecyl sulfate (SDS) were purchased from Sigma Aldrich (Singapore). Various graphene nanoplatelets (graphene), as purchased from XG Science Inc., China, have specified average thicknesses of 1–2, 6–8, and 15 nm and platelet diameters of 2, 15, and 15 μm , in which they are referred to as graphene grade C (CG), graphene grade M (MG), and graphene grade H (HG), respectively.

2.2. Preparation of graphene/gelatin hydrogel composites

Various concentrations of graphene; 0.01, 0.1, 0.5, and 1 vol%, were dispersed in water solutions filled with 1 vol% of SDS, sonicated via a transonicator (Elma, S 70H, D 78224) at room temperature, at 150 W, 50 Hz, for 15 min. Next, 10 vol% of gelatin was dissolved in distilled water (pH = 6.40) at 40 °C overnight via magnetic stirring. The two solutions were well-mixed at 40 °C, and poured into a petri dish to obtain the graphene/gelatin hydrogel composites. The hydrogel composites were allowed to settle as a film at an ambient temperature (~30 °C). Each hydrogel composite had a thickness of ~1.39 mm.

2.3. Characterization and testing of graphene/gelatin hydrogel composites

The true density of each graphene was measured by a gas pycnometer (Thermo Nicolet, Nexus 670) which was operated in He gas atmosphere (20 psi) at 25 °C with a purging gas time of 1 min. The true density of graphene was measured repeatedly 20 times to obtain the average value and the standard deviation.

The Brunauer–Emmett–Teller (BET) surface area of graphene was measured through a Thermo Finnigan, Sorptomatic 1990 surface area analyzer (SAA). The samples were weighed and out gassed at 300 °C for 12 h before the measurements of the adsorption and desorption isotherms with He and N₂ gas.

Electrical conductivity of the graphene was measured at 25 °C. The fixture consisted of two probes, which made contact with the film surface. The fixture was connected to a power source (Keithley, Model 6517A) to supply a constant voltage source and to read the current. The applied voltage and the resultant current were used to calculate electrical conductivity of the graphene samples by the following Eq. (1):

$$\sigma = \frac{I}{\rho} = \frac{I}{R_s t} = \frac{I}{KVt} \quad (1)$$

where σ is the specific conductivity (S/cm), ρ is the specific resistivity (Ω cm), t is the specimen thickness (cm), I is the measured resultant current (A), R_s is the sheet resistivity (Ω), V is the applied voltage (V), and K is the geometric correction factor.

The morphology and size of the graphene samples were observed using a H-7650 Transmission Electron Microscope (TEM; Hitachi High-Technology Corporation, Japan) at an operated voltage of 100 kV where an imaging software (SEMAFORE 5.21) was used to determine platelet diameter of graphene. Scanning electron micrographs of neat gelatin, and graphene/gelatin composites were obtained through a scanning electron microscope (SEM; S-4800, Hitachi, Japan) to determine the morphology at various graphene concentrations. The surface micrographs of neat gelatin and graphene/gelatin composites were taken using a voltage of 25 kV and a magnification of 10 000 times to observe the distribution of graphene in gelatin hydrogel composites.

Atomic force microscopy (AFM; Park system, XE-100) was used to observe the topology and phase images of the composites. Images were taken in the non-contact mode with the cantilever (NSC-14-CrAu) tapping at a scan rate of 0.25 Hz. The electrostatic force microscope (EFM) was utilized at a scan size of $1.00 \times 1.00 \mu\text{m}^2$ using a signal amplitude of 5 V. Each sample was scanned at two heights above the surface. In the first level, the AFM topology images were obtained via scanning tip in the non-contact mode, which responded to the Van der Waals forces. The second scan measured the tip-surface distance as a result of the electrostatic force which was obtained from the charge distribution and the degree of charge generated in the gelatin and graphene/gelatin hydrogel composites.

A melt rheometer (Rheometric Scientific, ARES) was used to investigate the electromechanical properties of the gelatin and graphene/gelatin hydrogel composites. The samples were set with a parallel plate fixture at a diameter of 30 mm. DC voltage was applied at the electric field strength (800 V/mm) using a DC power supply (Instek, GFG8216A). First, a strain sweep test was operated to define the suitable strain for obtaining storage modulus (G') in the linear viscoelastic regime. The appropriate strain was determined to be 0.10 %strain for both of the gelatin hydrogel and the graphene/gelatin hydrogel composites. Second, the temporal response and the frequency sweep test were pre-sheared at a low frequency of 0.039811 rad/s and low strain of 0.10% with the application of electric field (800 V/mm) for 15 min to assure with the equilibrium polarization in materials. In the frequency sweep test, the composite properties were measured as functions of electric field strength (0–800 V/mm) and temperature (30–80 °C). The deflection of the gelatin hydrogel and the graphene/gelatin hydrogel composites was determined under the influence of applied electric strengths in the range of 0–600 V/mm. For each hydrogel composite, the sample was fixed vertically in a transparent chamber containing polydimethylsiloxane (PDMS) with the viscosity of 100 cSt between the copper electrodes. In the experimental setup, the electric field was non-uniform due to the presence of the finite-sized relatively conductive sample, and non-symmetric because of the sample was suspended from the above but the lower end was free as shown in Fig. 1. The silicone oil and the 1% graphene/gelatin electrical conductivity values were 3.24×10^{-7} S/cm and 8.86×10^{-2} S/cm, respectively so there was a mismatch in the electrical conductivity. A high voltage power supply (Gamma High Voltage, UC5–30P) was connected to a DC power supply (Gold Sun 3000, GPS 3003D) for supplying DC electric field. The displacement of samples was recorded by using a

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