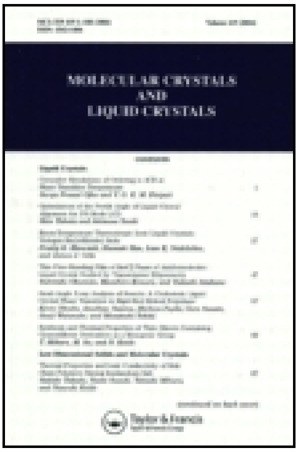

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Graphene Electrodes for Artificial Muscles

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Reduced graphene oxides (RGOs) thin films were fabricated in order to apply for the compliant electrodes of artificial muscles. The RGO was chemically prepared by chemical oxidation of natural graphite via Hummers method and following reduction using hydrazine. The RGO electrodes were coated on both sides of polydimethylsiloxane (PDMS) by using spray coater. The resulting PDMS dielectric elastomers with RGO electrodes had larger electromechanical strain response than those with conventional carbon paste electrodes. It might be caused by higher conductivity and higher flexibility of RGO than those of conventional electrodes.

Keywords Compliant electrode; dielectric elastomer; PDMS; reduced graphene oxide

1. Introduction

Dielectric elastomer actuators (DEA) being capable of converting electrical energy to mechanical force or movement have garnered a fast growing interest because they had a lot of advantages such as a high electromechanical strain, fast response, low cost, and facile processibility [1–5]. It has been considered as a good candidate material for the next generation artificial muscles.

DEAs consist of synthetic elastomer film and compliant electrodes coated on top and bottom surface of the elastomer film. When external electric field is applied, DEA is squeezed in the thickness direction and is expanded in area [6].

DEAs require compliant electrodes being capable of sustaining the large area expansion strain without damage and losing conductivity. A large variety of electrode materials have been explored such as thin metallic films, carbon paste, silver paste, carbon grease. Thin metallic films provide good electrical conductivity but maximum strain limited to 1% to 3% due to high stiffness. On the other hand, soft carbon grease or paste electrodes provide high maximum strain, making them used most commonly as an electrode of DEA [7–8]. The important requirements as an

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electrode of DEA are mechanical compliance, high electrical conductivity, and resistance to delamination and rubbing off.

Graphene consisting of a flat monolayer of carbon atoms with a two-dimensional lattice is an attractive material as an electrode owing to its good flexibility and its high electron mobility (2,000,000 cm^2/Vs) allowing extremely high-speed operation and low electric power consumption if graphene is used as a channel material, a transistor, and energy converter [9].

In this article, in order to investigate the effect of compliant electrode on the electric actuation performance of the dielectric elastomer, electromechanical strain responses of the PDMS coated with graphene electrodes and carbon grease electrodes are compared. Polydimethylsiloxane (PDMS) is used as a dielectric elastomer.

2. Experimental

2.1. *Materials*

The PDMS (Sylgard 3481) and sylgard oil (DC-561) was purchased from Dow Corning. Purified natural graphites, sulfuric acid, hydrazine, ethanol purchased from Sigma–Aldrich were used as received without additional purification. Carbon paste (FTU-20) was purchased from ASAHI Chem. Research Lab. Co. Ltd. to compare prepared electrode with carbon paste.

2.2. *Coating of Compliant Electrodes on the Dielectric Elastomer Films*

It was cured after mixing with 50 wt.% DC-561 sylgard oil and 10 wt% curing agent at room temperatures. PDMS film with thickness of 0.5 mm was made by bar-coating.

Graphene oxide (GO) was prepared from purified natural graphite (45 μm, Sigma–Aldrich) via the Hummers method [10]. Reduced graphene oxide (RGO) was prepared by reduction of GO. GO (20 mg) was loaded in a 250-ml round bottom flask and water (200 ml) was then added, yielding an inhomogeneous brown dispersion. This dispersion was sonicated using a Sonics INC. VCX750 ultra-sonicator until it became clear with no visible particulate matter. Hydrazine hydrate (0.05 ml, $N_2H_4 \cdot H_2O$ 98%) was then added and the solution heated in a water bath at 100°C for 2 h [11]. RGOs were gradually precipitated out as dark aggregates. RGOs were filtered and dried. And then RGOs were redispersed in ethanol. RGO electrodes were spray-coated on the surface of PDMS films. Circular carbon paste electrodes were spray-coated with each of carbon paste on both sides of the films.

2.3. *Characterization*

UV/Visible spectral analysis of GO and RGO was carried out via Hewlett Packard UV/Visible Spectrophotometer (Model 8453A). Surface morphologies of coated graphene electrodes were observed by FE-SEM, using a Hitachi S-4100 with an acceleration voltage of 10 keV. The electric resistance of the electrode was measured by four probe method, Mitubishi Loresta-GP MCP-T610. The electromechanical thickness strain was measured by using two laser displacement sensors (Keyence LK-G80). The electric voltage was delivered to the film by using a function generator, Agilent 33250A amplified by a factor of 1000 through a high voltage lock-in amplifier, Trek 10/10B. Incident two lasers beams were exposed perpendicular to the film surfaces [12].

Results and Discussions

Figure 1 show the UV/Visible absorption spectra (normalized the result using the absolute value of the maximum) and pictures of the graphene oxides and reduced graphene oxides solution. In the picture of diluent GOs and RGOs solution, GOs solution looked orange, while RGOs appeared to be black. In the UV/Visible absorption spectra, GOs had a maximum absorption peak at 230 nm and a shoulder at around 300 nm, which are attributed to $\pi \rightarrow \pi^*$ transitions of the aromatic C–C bonds of graphene oxide and $n \rightarrow \pi^*$ transitions of C=O bonds, respectively. After reduction of GO with hydrazine, the maximum peak at 230 nm was redshifted to 260 nm and the shoulder at 300 nm disappeared. That is, the oxidation products, graphene oxides had a layered morphology with –OH functionality disrupting the hexagonal carbon basal planes on the interior of graphene oxide; –COOH and carbonyl groups decorated the periphery of the planes [13]. After hydrazine treatment, the absorption peak exhibited a redshift to around 260 nm, because electronic conjugation of aromatic C–C bonds has been restored. Meanwhile, the shoulder at 300 nm disappeared because C=O functionality was removed.

Figure 2a and 2b show photographs of PDMS films coated with RGOs and carbon pastes electrodes, respectively. Circular electrodes were spray-coated on both top and bottom sides of PDMS films. Spray coating method achieved quite uniform carbon paste and RGO electrodes. At the same surface resistance level, PDMS film with RGOs electrodes had transmittance of 65%. PDMS film with carbon paste electrodes had zero transmittance. Figure 2c and 2d show SEM images of RGOs and carbon pastes electrodes on the PDMS films, respectively. Reduced graphene oxide sheets were observed on the surface of RGOs electrodes. Distortions and wrinkles appeared because they experienced harsh chemical oxidation and reduction processing. Each sheet was about 10 nm thick. It indicated that RGOs was a stack of

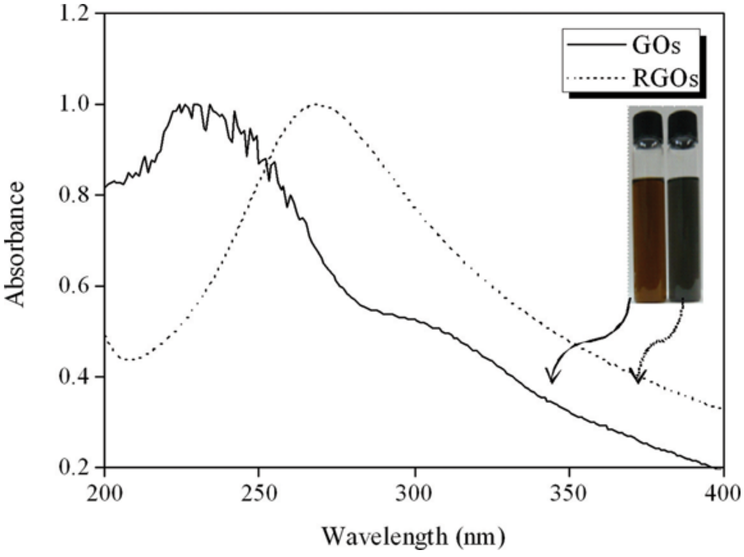

Figure 1. UV/Visible absorption spectra of GOs and RGOs solution. Inset shows the photographs of the GOs and RGOs dispersions.

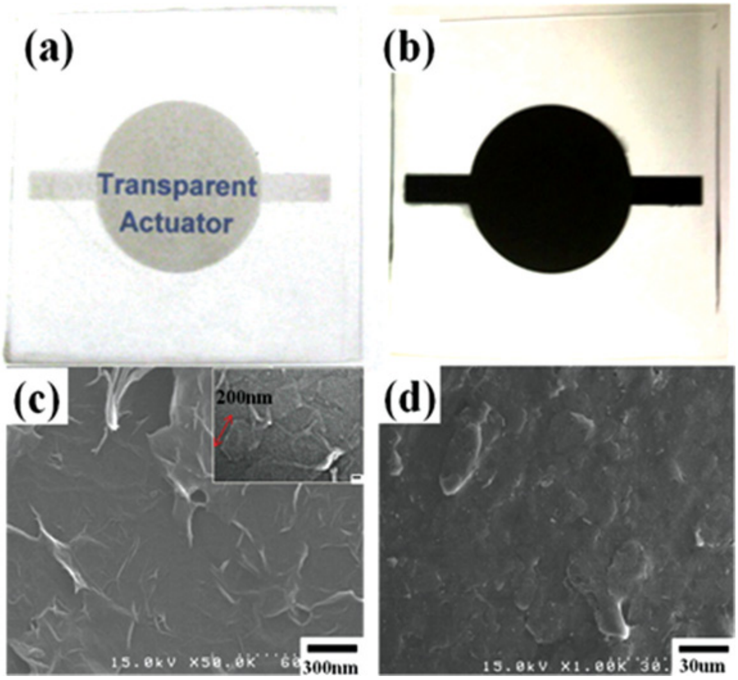

Figure 2. Photographs and SEM images of PDMS films coated with RGOs and carbon pastes electrodes photographs: (a) photograph of RGO electrodes, (b) photograph of carbon paste electrodes, (c) SEM image of RGO electrodes, and (d) SEM image of carbon paste electrodes.

several tens reduced graphene oxide sheets. Carbon paste electrodes showed a homogeneous morphology without any significant carbon aggregation.

Figure 3 shows the surface resistivity of RGOs and carbon pastes electrodes as a function of thickness. RGO electrodes had much smaller surface resistivity than carbon paste electrodes. RGOs electrodes with the thickness of 67 nm had transmittance of 70% and surface resistivity of $5.24 \times 10^3\ \Omega/\square$. In contrast, carbon paste electrodes with the thickness of 3.2 μm had zero transmittance and surface resistivity of $4.52 \times 10^4\ \Omega/\square$. Surface resistivity decreased with increase in thickness of electrodes.

Figure 4 shows electromechanical thickness strain of PDMS films with RGO electrodes and carbon paste electrodes at various applied electric field. Thickness strain (s_z) defined as a thickness of the sample film divided by an initial thickness. Higher thickness strain indicates higher electromechanical deformation change. In order to use same electrical conductivity, RGOs electrodes had the thickness of 15 nm and 67 nm. Carbon paste electrodes had the thickness of 530 nm and 4.89×10^3 nm. The measured strains increased with the electric field as expected. Theoretically, thickness strain is known to be exponentially increased with electric field strength [14]. Additionally, the thickness strain was significantly affected not only by type of electrodes, but also thickness of electrodes. RGO electrodes produced higher thickness strain response than carbon paste electrodes. At around 20 V/μm, especially, RGOs electrodes exhibited 100% higher strain than carbon paste electrodes. For both electrode systems, thin electrode exhibited higher

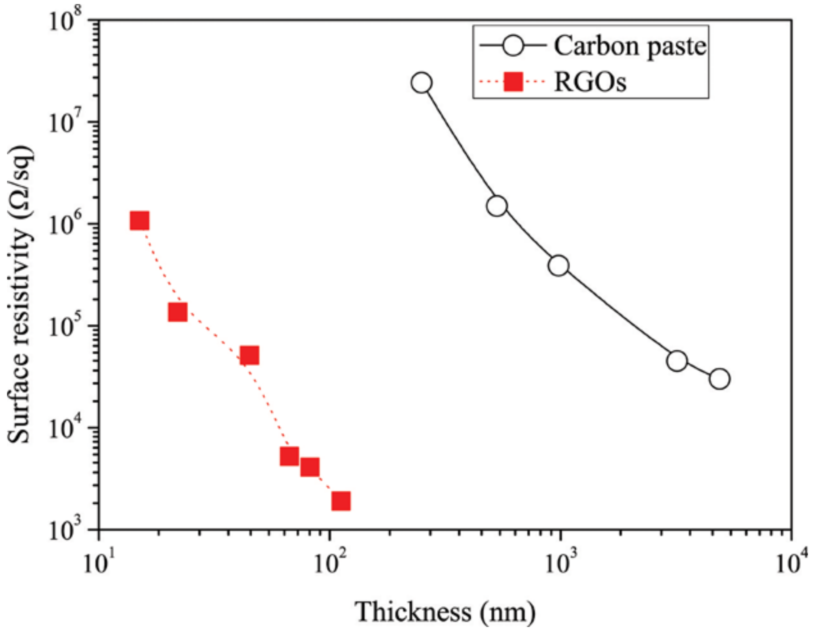

Figure 3. Surface resistivity of RGOs and carbon pastes electrodes as a function of thickness.

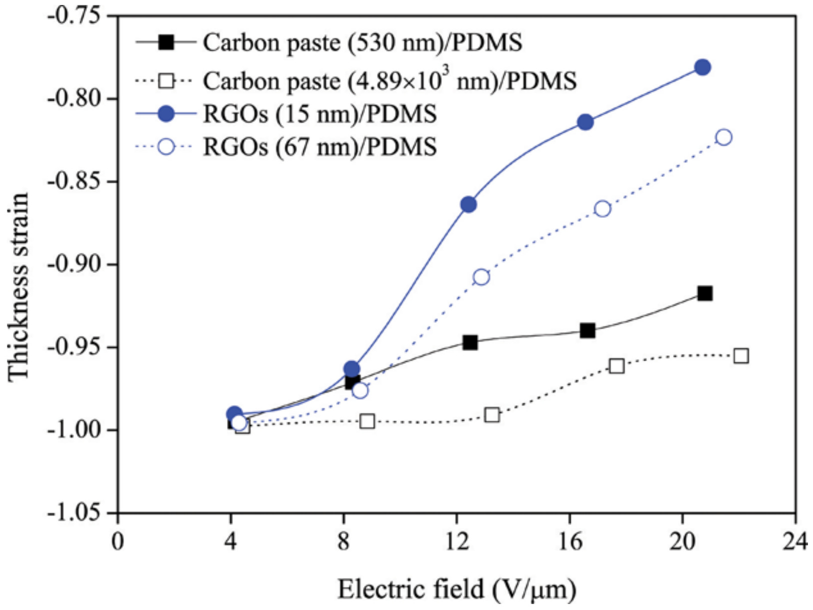

Figure 4. Thickness strain, s_z of the PDMS films with RGO electrodes and carbon paste electrodes as a function of an applied electric field. RGO electrode with the thickness of 15 nm ($1.07 \times 10^6\,\Omega/\square$) and 67 ($5.24 \times 10^3\,\Omega/\square$) nm and carbon paste electrodes with the thickness of 530 nm ($1.50 \times 10^6\,\Omega/\square$) and 4.89×10^3 nm ($3.01 \times 10^4\,\Omega/\square$) were compared.

thickness strain than thick electrode, even though thin electrode had much higher resistivity than thick electrode.

Thus, the better electromechanical performance of the RGO electrodes might be caused by higher conductivity and higher flexibility of RGO electrodes than those of conventional carbon paste electrodes. RGO had much higher electrical conductivity than carbon paste at the same thickness level. Therefore, electrode thickness can be reduced to several tens nanometer, while carbon paste electrode had to have several micron scale thickness for the electromechanical actuation test due to poor conductivity. The thin electrodes were able to minimize the constraint effect of the electrode during the electric actuation of dielectric elastomer. Hence, better electromechanical actuation performance could be achieved.

Conclusion

Homogeneous RGOs ethanol dispersion was chemically prepared by chemical oxidation of natural graphite via Hummers method and following reduction using hydrazine. The RGO electrodes with uniform morphology were successfully spray-coated on both sides of polydimethylsiloxane (PDMS). We have shown that chemically synthesized reduced graphene oxides can be used as potential compliant electrode of dielectric elastomer actuators.

Acknowledgment

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