

# Electromechanical Actuators Based on Graphene and Graphene/Fe<sub>3</sub>O<sub>4</sub> Hybrid Paper

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Exceptionally high specific surface area, mechanical strength, electrical conductivity, and a special two-dimensional structure make graphene a highly promising material for electromechanical actuators. Electromechanical actuators are fabricated using flexible graphene-based paper prepared via a filtration process, and the stroke of these graphene-based actuators is directly measured during electrochemical double-layer charge injection. Actuation strain up to 0.064% was obtained for pristine graphene paper in response to an applied potential of -1 V in 1 M NaCl solution. Double-layer charge injection in graphene sheets is believed to induce actuation strain through a combination of coulombic and quantum-chemical-based expansion. To increase electrochemical-double-layer capacitance and actuator performance, Fe<sub>3</sub>O<sub>4</sub> nanoparticles were used to partially prevent graphene sheets from restacking and allow the electrolyte ions to infiltrate the resulting magnetic graphene paper more easily. The magnetic graphene paper exhibits actuation strain as large as 0.1% at -1 V applied potential, which is about 56% higher than that of the pristine graphene paper.

### 1. Introduction

Electromechanical actuators, which can directly convert electrical energy into mechanical energy through changes in material dimensions, have been dubbed "artificial muscles" due to some similarities with biological muscles.<sup>[1,2]</sup> These actuators have been proposed for various applications, including as sensitive switches, microrobotics, optical displays, prosthetic devices, and microscopic pumps.<sup>[1]</sup> Though ferroelectrics, electrostrictive materials, and conducting polymers have been deployed as actuators, their practical application has been restricted by either the requirement for high driving voltages or limitations on cycle life and response rate.<sup>[1-3]</sup>

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As early as 1969, it was realized that the carbon-carbon bond length in graphite increases when potassium is intercalated between graphene sheets in graphite.<sup>[4,5]</sup> This dimensional change provides the foundation for a new generation of actuators using various carbon-based nanomaterials. In recent decades, because of their outstanding electrical and mechanical properties, extraordinarily high surface area, and charge-transfer features, carbon nanotube (CNT) based actuators have grabbed the attention of both scientists and engineers.<sup>[1,3,4,6]</sup> The first report in 1999 demonstrated that electrochemical actuators based on double-layer charging of sheets of single-walled carbon nanotubes (SWNTs) can generate much higher stresses than those of natural muscle and also generate useful strain at low applied voltages.<sup>[1]</sup> Subsequently, actuation of sheets of multiwalled carbon nanotubes

(MWNTs) was reported.<sup>[3]</sup> Recently, high-strength yarns made through twist-based spinning MWNTs have been developed, which displayed an actuation strain of up to 0.7%.<sup>[4,6]</sup> Since stacking graphene sheets provides graphite and CNTs comprise seamless cylinders of graphene,<sup>[7]</sup> it is interesting to conduct experimental investigations that enable comparison of these highly related different forms of carbon.

Graphene, the two-dimensional construction material for carbon materials of all other dimensionalities,<sup>[7]</sup> is emerging as a rising star in the field of material science owing to its prominent intrinsic properties<sup>[8-12]</sup> and structural features.<sup>[13,14]</sup> Both theoretical and experimental results have revealed that the electric conductivity,<sup>[8,9]</sup> mechanical strength,<sup>[11,12]</sup> and surface area (2,675 m<sup>2</sup> g<sup>-1</sup>)<sup>[15]</sup> for a perfect graphene sheet can rival or surpass that of individual CNTs of any type. Thus, it is expected that the combination of these remarkable properties could turn graphene into a highly promising material for actuation applications. For instance, an electromechanically driven cantilever-based microactuator was developed, based on a grapheneon-epoxy film hybrid bimorph, which provided large bending displacement and rapid response while operating at low input power.<sup>[16]</sup> Recently, Park et al. prepared a graphene oxide/MWNT bilayer stack that provided cantilever-based actuation as a result of changes in temperature or humidity.<sup>[17]</sup> Xie et al. investigated an interesting graphene-based electrochemical actuator based on a rationally designed graphene film with asymmetrically



surfaces modified by hexane and oxygen plasma and demonstrated the potential of graphene film for applications in various electromechanical systems.<sup>[18,19]</sup> Nevertheless, the mechanisms of graphene actuation are not yet fully understood, and may be quite complex, especially for graphene arrays. Furthermore, physical or chemical surface treatment of graphene can introduce defects that profoundly affect actuation.

We here demonstrate electrochemically based actuation for self-supported sheets comprised exclusively of graphene flakes. As for the case of nanotube sheets made by a process like that of making ordinary paper, these graphene sheets are referred to as graphene paper. For the purpose of improving actuation by interfering with graphene assembly into stacks during the paper-making process, a series of magnetic graphene hybrid paper samples were fabricated and characterized. This hybrid paper, which comprised graphene flakes mixed with Fe<sub>3</sub>O<sub>4</sub> nanoparticles, was prepared through a simple yet effective solution process that is described in our previous work.<sup>[20]</sup> This magnetic graphene hybrid paper showed higher actuation strain than the pristine graphene papers. Based on these results, we also predict that the maximum strain of graphene paper actuators composed of non-restacked graphene sheets should be at least as high as for nonbundled SWNTs.<sup>[1]</sup>

#### 2. Results and Discussion

The graphene paper used in this work was fabricated by filtration of a controlled amount of graphene dispersion, followed by air drying and peeling the paper from the filter, according to the previous literature.<sup>[20–22]</sup> Since the graphene in the dispersion is derived by the dispersion-based reduction of exfoliated graphene oxide using hydrazine hydrate, it is not fully reduced. Subsequent annealing of the graphene sheets provided further reduction of the graphene. For comparison of actuation, two kinds of graphene paper sheets were produced: pristine graphene-1 (PG-1) paper, which was annealed for one hour at 250 °C, and pristine graphene-2 (PG-2) paper, which was annealed at 400 °C for one hour. The fabrication of magnetic graphene hybrid paper (MG) was from a mixed dispersion of water-soluble partially-oxidized graphene and 5–10 nm diameter Fe<sub>3</sub>O<sub>4</sub> nanoparticles, using our previously reported procedure.<sup>[20]</sup> The as-

fabricated magnetic graphene hybrid paper samples, which contained different weight percents of  $Fe_3O_4$ , were annealed at 400 °C under argon. The amount of  $Fe_3O_4$  nanoparticles in the magnetic graphene hybrid paper was derived from atomic absorption spectra to provide the results shown in **Table 1**.

The structure of pristine graphene and magnetic graphene hybrid papers were characterized by using scanning electron microscopy (SEM) to provide the micrographs shown in **Figure 1**a and b. The cross-sections of fracture-generated sheet edges show that both the pristine graphene and magnetic hybrid graphene paper have a well-ordered, layered structure, which contrasts with the highly entangled structure of CNT mats Table 1. The amount of  $Fe_3O_4$  nanoparticles loaded in the magnetic graphene paper. The results are calculated from the content of Fe as detected by atomic absorption spectra.

MG samples	MG-1	MG-2	MG-3	MG-4
Amount of Fe <sub>3</sub> O <sub>4</sub> (wt%)	7.5	15	18	21

produced by the paper-making process.<sup>[1,3]</sup> This graphene-based layered structure, with layers parallel to the paper surface, is also confirmed by X-ray diffraction (XRD) measurements.<sup>[20–22]</sup> The interlayer distance (d spacing) between stacked graphene sheets in graphene-based paper is around 0.35–0.40 nm,<sup>[20–22]</sup> which is slightly larger than that in graphite. As an expected result of interference of the Fe<sub>3</sub>O<sub>4</sub> nanoparticles in graphene stacking, the hybrid graphene/Fe<sub>3</sub>O<sub>4</sub> paper has a more porous structure (Figure 1b) than that of the pristine graphene paper (Figure 1a). In fact, we will show that this more porous structure increases electrochemical capacitance and corresponding actuator stroke. Surprisingly, these magnetic graphene hybrid paper strips have improved mechanical strength over that of the pristine paper strips,<sup>[20]</sup> which is useful for increasing actuator load capacity.

Electromechanical actuation was first studied for the pristine graphene paper strips (PG-1 and PG-2) immersed in 1  $\mbox{M}$  NaCl solution. Actuation results from applying a voltage between this working electrode of the graphene paper and a platinum mesh counter-electrode, so as to change the potential of the actuating graphene electrode with respect to a reference saturated calomel electrode (SCE). The length changes of the graphene-based paper electrodes were directly measured as a function of electrode potential using a PerkinElmer 7E dynamic mechanical analyzer.<sup>[1,3,23]</sup>

The configuration used for these measurements of electromechanical actuation is schematically pictured in Figure S1 (Supporting Information). One end of a vertical graphene paper strip was fixed to the bottom of the aqueous 1 M NaCl solution. The other end was attached to the muscle-analyzer arm. The PerkinElmer 7E dynamic mechanical analyzer can be programmed to keep the load on the paper strip constant and to measure changes in the strip length during actuation. To improve wetting with the electrolyte, the graphene paper strips were first conditioned by applying ten voltage cycles from



Figure 1. a) SEM image of the cross-section of PG-2 paper and b) MG-1 paper.



**Figure 2.** a) The applied voltage, current, and resulting actuation strains of PG-1 paper in 1  $\,$ M NaCl solution. b) Actuation strain of PG-1 paper as a function of applied voltage in 1  $\,$ M NaCl solution. c) Actuation strain of PG-2 paper as a function of applied voltage, in 1  $\,$ M NaCl solution. The cycle times used to collect this data were all 1 min.

-1.0 V to +1.0 V at 100 mV s<sup>-1</sup>. All measurements of tensile actuation were performed using the same conditions.

Figure 2a displays the actuation strain and interelectrode current of the PG-1 paper, in response to a series of applied

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square-wave voltage pulses between -1 V to +1 V, using a cycle time of 1 minute. Similar to for CNT actuators, creep of strain with potential cycling is clearly observed for the PG-1 paper. The observed actuator strain depends approximately quadratically upon the applied potential (Figure 2b and c), which is also the case for carbon nanotube paper evaluated using similar conditions.<sup>[1,3,24,25]</sup> Importantly, while Xie et al. showed that their O<sub>2</sub>plasma-treated graphene strips expanded when electrons were injected and contracted when holes were injected,<sup>[18,19]</sup> our pristine graphene paper electrodes expanded for both large applied negative and positive voltages.

As depicted in the schematic illustration of Figure 3a, the graphene-paper electrodes act as supercapacitor capacitors that are charged and discharged during actuator cycling. The corresponding changes in charge density on the carbon atoms provide changes of C-C bond length that at least in part result in actuation.<sup>[1,3]</sup> Therefore, increasing the electrochemical-double-layer capacitance of the graphene-based paper is expected to improve actuator performance. The tendency of graphene flakes to restack during paper fabrication dramatically decreases the surface area available for charge injection, and therefore decreases the gravimetric electrochemical capacitance compared with that for unstacked graphene sheets.<sup>[26]</sup> The Fe<sub>3</sub>O<sub>4</sub> nanoparticles in the graphene/Fe<sub>3</sub>O<sub>4</sub> hybrid paper interfere with this restacking, and can potentially act as pillars that separate individual graphene sheets, thereby facilitating electrochemical accessibility between sheets, as suggested schematically in Figure 3b.

As can be seen in Figure 4a and b, the specific capacitances of PG-1 and PG-2 are about 95 F  $g^{-1}$  and 31 F  $g^{-1}$  (at a scan rate of 10 mV  $\ensuremath{\mathrm{s}^{-1}}\xspace$  ), respectively. One possible explanation for this capacitance decrease is that increasing annealing temperature from 250 °C for PG-1 to 400 °C for PG-2 could increase interlayer proximity and thereby make it more difficult for the electrolyte ions to infiltrate into the graphene paper. However, this interpretation seems inconsistent with the higher actuation strokes observed for the PG-2 paper than for the PG1 paper. While PG-1 paper produced a maximum strain about 0.042%, PG-2 paper had a maximum strain of about 0.064% when a -1 V potential was applied (Figure 2b and c). Hence, we believe that the increased capacitance for the lower-temperature-annealed PG-1 is likely due to pseudocapacitance associated with residual oxygen functionalities in the graphene. This interpretation is consistent with the lower electric conductivity ( $\sigma \approx 2,300$  S m<sup>-1</sup>) of the PG-1 paper compared with that for the PG-2 paper (ca. 3,500 S m<sup>-1</sup>), as shown in Figure S2 in the Supporting Information. Additionally, the shape of the cyclic voltammetry curve for PG-1 is less rectangular than that of the higher-temperatureannealed PG-1 and graphene/Fe3O4 hybrid paper samples (Figure 4a and c), which is consistent with a higher pseudocapacitance contribution for PG-1 sheets. Consequently, in the remaining discussions we will compare actuation and capacitances for PG-1 and graphene/Fe<sub>3</sub>O<sub>4</sub> hybrid structures that were annealed at the same temperature (400 °C for one hour).

Significantly, all the graphene/ $Fe_3O_4$  hybrid paper samples had higher gravimetric electrochemical capacitance than that of PG-2 paper, as seen in Figure 4c and 4d. However, MG-1 paper (7.5 wt% loading of  $Fe_3O_4$ ) had the highest specific capacitance and a further increase in content of  $Fe_3O_4$  nanoparticles

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**Figure 3.** Schematic illustration of charge injection in graphene-based electromechanical actuators. a) Charge injection in the outer layers of stacks of graphene in pristine graphene paper, which is balanced by the pictured surface layers of electrolyte ions. The used electrolytes are unable to access the inner layers of graphene stacks. b) Charge injection at the outer surface and inside stacks of graphene, which are partially separated by magnetic nanoparticles in graphene paper. This electrode charge is balanced by the pictured surface layers of electrolyte ions.

decreased the specific capacitance. We postulate that two main factors may result in this decrease: 1) Though the Fe<sub>3</sub>O<sub>4</sub> nanoparticles play an important role in forming a porous structure for the graphene/Fe<sub>3</sub>O<sub>4</sub> hybrid paper, the surface area of these magnetic nanoparticles contributes little to specific paper capacitance (about 5 F g<sup>-1</sup>, when normalized to Fe<sub>3</sub>O<sub>4</sub> weight).<sup>[27]</sup> Hence, after the effect of Fe<sub>3</sub>O<sub>4</sub> nanoparticles on increasing the graphene surface area saturates, the effect of further increase in Fe<sub>3</sub>O<sub>4</sub> concentration for Fe<sub>3</sub>O<sub>4</sub> nanoparticles could be to

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decrease the specific capacitance of the hybrids (which are normalized to total weight of the hybrids). 2) Increase in the coverage of graphene sheet area by  $Fe_3O_4$  nanoparticles would decrease the graphene sheet area that is available for electrochemical double-layer formation.

Figure 5 provides results for actuation of graphene/Fe<sub>3</sub>O<sub>4</sub> hybrid paper and for PG-2. Figure 5d shows the dependence of actuator strain and capacitance on the weight percent of Fe<sub>3</sub>O<sub>4</sub> nanoparticles, which demonstrates that percent actuator stroke increases with increased specific capacitance of the graphene-based electrodes. The largest percent actuator stroke was obtained for MG-1 paper, which provided 0.1% expansion for -1 V applied potential (versus SCE), which is about 56% higher than for the PG-2 paper (0.064%). This result demonstrates that introduction of Fe<sub>3</sub>O<sub>4</sub> nanoparticles into graphene-based sheets increases performance.

As for CNT paper and pristine graphene paper, mechanical creep during potential cycling was also found for MG-1 paper (Figure 5a) and for the other hybrid paper samples. Also, as for the PG-1 and PG-2 samples, actuator strain (an expansion) increases with increasing large negative or positive charge injection, and the minimum length of the paper is likely at the potential of zero charge. The minimum length of the paper occurs at a value between 0 and 0.2 V (versus SCE), which is reasonably close to the potential of zero charge for the basal plane of graphite (-0.2 V versus SCE), especially since potential scans of length changes versus potential depend upon whether potential is increasing or decreasing.<sup>[1]</sup> Strain versus potential is quasiquadratic for all samples, though there is asymmetry with respect to the potential minimum, with actuator strains versus potential (relative to the potential minimum, which increases more slowly with increasing potential upon hole injection than for electron injection), as shown in Figure 2b and c for the PG-1 and PG-2 papers, Figure 5b for the MG-1 sheet, and Figure 5c for the MG-1, MG-2, and PG-2 sheets.

The likely explanation for this asymmetry with respect to the minimum length of the paper is that actuator strain results from a combination of Coulombic and quantum-mechanical effects. Coulombic effects are, to first-order approximation, quadratic in applied potential, though the difference in the counter-ions for hole and electron injection may cause asymmetry with respect to the potential of zero charge injection. Importantly, the quantum-mechanical contribution to actuation can break hole-electron symmetry. More specifically, the results of Kertesz et al.<sup>[28]</sup> showed that expansion of graphite sheets occurs for negative charge injection and contraction occurs for positive charge injection, due to the electron-hole symmetry-breaking effect of second-neighbor antibonding interactions. An additional contribution to actuation can also result from volume changes for the ion layer needed to compensate electrode charge, which will generally also be asymmetric with respect to the sign of injected charge if the volume of anions and associated solvation sphere for hole injection differs from the corresponding cation-associated volume for electron injection.

We believe that further optimizing the mechanical strength, electrical conductivity, and surface area for graphene-based papers should result in much more impressive actuator performance. More specifically, if graphene-based papers composed of

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**Figure 4.** a) Cyclic voltammetry at a scan rate of 100 mV s<sup>-1</sup> for PG-1 and PG-2 paper in 1 M NaCl solution. b) Specific capacitance as a function of scan rate for PG-1 and PG-2 paper in 1 M NaCl solution. c) Cyclic voltammetry at a scan rate of 10 mV s<sup>-1</sup> for PG-2, MG-1, MG-2, MG-3, and MG-4 paper in 1 M NaCl solution. d) Specific capacitance at various scan rates for PG-2, MG-1, MG-2, MG-3, and MG-4 paper in 1 M NaCl solution.

completely nonstacked graphene sheets could be obtained for application as electromechanical actuators, up to the entire surface area (2,675 m<sup>2</sup> g<sup>-1</sup>) <sup>[15]</sup> of graphene could be utilized for maximizing actuator strain (and corresponding actuator stroke), which we will predict for a potential of -1 V (with respect to SCE).

The maximum actuation strain for this applied potential of -1 V (versus SCE) can be predicted using  $S_V = C_A A_G M (\Delta L/L\Delta \gamma)/N$ ,<sup>[1]</sup> where  $S_V$  is the strain–voltage coefficient (i.e., the ratio of the change in actuator strain to the change in applied electrode potential). The data in Figure 2c yield an  $S_V$  value for the PG-2 paper of 0.064%/V for a potential of -1 V. M is the molecular weight of carbon and N is the number of coulombs of charge in a mole of electrons (about 96,500 C mole<sup>-1</sup>).  $C_A$  and  $A_G$  are the areal capacitance and surface area, respectively. The factor  $\Delta L/L\Delta \gamma$  is the strain–charge coefficient, in which  $\Delta L/L$  is the fractional change in length caused by a  $\Delta \gamma$  change in the concentration of injected charge per carbon atom.<sup>[1]</sup> Thus, by substituting the specific capacitance for PG-2 in 1 M NaCl solution ( $C_A A_G = 30$  F/g), a value of  $\Delta L/L\Delta \gamma = 0.17$  at -1 V is derived from the  $S_V$  for PG-2 paper.

Accordingly, actuation performance for graphene-based papers composed of nonstacked graphene flakes can be predicted from this strain–charge coefficient. Graphene has a remarkably high specific surface area ( $C_A$ ) up to 2,675 m<sup>2</sup> g<sup>-1</sup> and the intrinsic capacitance ( $A_G$ ) of graphene was recently found to be  $21 \mu F \text{ cm}^{-2}$ ,<sup>[15]</sup> which are both higher than for nonbundled SWNTs. Combining this surface area, the specific interfacial capacitance for graphene and the strain–charge coefficient ( $\Delta L/L\Delta \gamma = 0.17$  at -1 V), derived using results from charge injection in



1 M NaCl electrolyte for PG-2, a value of  $S_V = 1.2\%/V$  is obtained for an electrode potential of -1 V for sheets comprising unstacked graphene flakes. As long as the main contributions to actuation are the same as for the present experimentally characterized partially stacked graphene PG-2 paper structure, the graphene sheet capacitance is dominant, and sheet elastic properties are unchanged, this is the predicted actuation obtainable at -1 V applied potential for graphene sheets.

#### 3. Conclusions

In summary, the electromechanical actuation of free-standing graphene-based paper is reported as a function of applied potential and compared with that for graphene/Fe<sub>3</sub>O<sub>4</sub> nanoparticle hybrids. The pristine graphene paper, annealed at 400 °C, has an actuation strain of about 0.064%. This actuation is increased by using Fe<sub>3</sub>O<sub>4</sub> nanoparticles to reduce the aggregation of graphene sheets into stacks, which decreases electrochemical doublelayer capacitance. The magnetic graphene/ Fe<sub>3</sub>O<sub>4</sub> hybrid papers provide increased actuation strain over the pristine graphene papers,

thereby providing a maximum strain of 0.1% at a potential of -1 V versus SCE, which is about 56% higher than for pristine graphene paper. The roughly quadratic dependence of actuation strain on potential indicates that coulombic contribution to actuation dominates. However, higher strains are obtained for electron injection than for hole injection, which indicates that quantum-mechanically induced strains (which change sign with potential with respect to the potential of zero charge), and possibly volumetric effects that depend on solvated cation and solvated ion volumes, are important. By using experimental results and the surface area and area-normalized capacitance of single graphene sheets, we predict the dramatically increased actuator strokes that might be obtainable for graphene-flake structures in which sheet stacking does not occur.

#### 4. Experimental Section

Preparation of Pristine Graphene Paper: Graphene paper was fabricated by vacuum filtration of a graphene dispersion according to the literature.<sup>[20,21]</sup> In a typical procedure, 20 mg graphene oxide (GO, prepared from natural graphite by the modified Hummer's method<sup>[29,30]</sup>) was first completely exfoliated down to individual graphene oxide sheets in 80 mL distilled water to form a stable dispersion of graphene oxide solution (0.25 mg mL<sup>-1</sup>). Afterwards, 40  $\mu$ L hydrazine hydrate (80%) and 280  $\mu$ L ammonia solution (25%) were added to the homogeneous dispersion and vigorously stirred for a few minutes. The dispersion was then subjected to reduction by heating to 95 °C for an hour. Pristine graphene paper was obtained by filtration of the black solution through an Anodisc membrane filter (47 mm in diameter, 02  $\mu$ m pore size; Whatman), followed by air drying and peeling the paper from the filter. PG-1 paper was obtained



**Figure 5.** a) The applied voltage, current, and resulting actuation strains of MG-1 paper in 1 mu NaCl solution. b) Actuation strain of MG-1 paper as a function of applied voltage in 1 mu NaCl solution. c) Comparison of the voltage dependence of actuation strain for PG-2, MG-1, and MG-2 paper in 1 mu NaCl solution. d) Actuation strain at +1 V (left axis) and specific capacitance at a scan rate of 10 mV s<sup>-1</sup> as a function of wt% Fe<sub>3</sub>O<sub>4</sub> for MG-1, MG-2, MG-3, and MG-4 paper.

by subsequent annealing at 250  $^\circ C$  in argon for 1 h, and PG-2 paper was annealed at 400  $^\circ C$  in argon for 1 h.

Preparation of Magnetic Graphene Paper. Magnetic graphene/Fe<sub>3</sub>O<sub>4</sub> hybrid papers were fabricated according to our previous work.<sup>[20]</sup> Watersoluble Fe<sub>3</sub>O<sub>4</sub> nanoparticles were first prepared using known methods.<sup>[31,32]</sup> Then, the needed amount of Fe<sub>3</sub>O<sub>4</sub> nanoparticles aqueous solution was added to the GO dispersion (in 80 mL distilled water, 0.25 mg mL<sup>-1</sup>) under vigorous stirring or ultrasonication, and then the mixture was reduced with 40  $\mu$ L hydrazine hydrate (80%) at 90 °C for 1 h. Afterward, a homogeneous black dispersion with a small quantity of black precipitate was obtained. Graphene/Fe<sub>3</sub>O<sub>4</sub> hybrid paper was obtained by filtration of the black supernatant through an Anodisc membrane filter (47 mm in diameter, 0.2  $\mu$ m pore size; Whatman), followed by air drying and peeling from the filter. The magnetic graphene papers were all annealed at 400 °C in argon for 1 h for further reduction. In this work, a series of four magnetic graphene papers (from MG-1 to MG-4) was prepared with different concentrations of Fe<sub>3</sub>O<sub>4</sub> nanoparticles (see Table 1).

Electromechanical Actuator Testing: Tensile actuation measurements were performed using PerkinElmer 7E dynamic mechanical analyzer, according to previously described methodology.<sup>[1,3,23]</sup> Both pristine graphene and magnetic graphene papers were evaluated. The samples were cut into strips about 5 mm wide and 6–10 mm long. Measurements were carried out in the three-electrode cell in aqueous 1 M NaCl solution with the paper as the working electrode, Pt mesh as counter-electrode and SCE as a reference electrode. To improve wetting of the material, all

samples were first conditioned at voltage –0.5 V to 0.5 V for 10 cycles at 100 mV  $\rm s^{-1}.$ 

*Electrochemical Measurements*: A three-electrode cell system was used to evaluate the cyclic voltammetry (CV) performance for the pristine graphene and magnetic graphene papers on a LK98BII microcomputer-based electrochemical analyzer. The measurements were performed in aqueous 1  $\,$  M NaCl solution with the graphene-based papers as working electrodes, a Pt mesh counter-electrode, and SCE as a reference electrode. To improve wetting of the material, all samples were conditioned at voltage –0.5 V to 0.5 V for 10 cycles at 100 mV s<sup>-1</sup> before actuation measurements.

#### **Supporting Information**

Supporting Information is available from the Wiley Online Library or from the author.

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