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Title

Architecting graphene oxide rolled-up micromotors: a simple paper-based manufacturing technology

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Keywords: Micromotors, wax printed membranes, graphene, rolled-up tubes

We report a graphene oxide rolled-up tubes production process using wax printed membranes for the fabrication of on demand engineered micromotors at different levels of oxidation, thicknesses and lateral dimensions. The resultant graphene oxide rolled-up tubes can show magnetic and catalytic movement within the addition of magnetic nanoparticles or sputtered platinum in the surface of graphene oxide modified wax printed membranes prior to the scrolling process. As a proof of concept, the as-prepared catalytic graphene oxide rolled-up micromotors are successfully exploited for oil removal from water. This micromotor production technology relies on an easy, operator-friendly, fast and cost-efficient wax-printed paper-based method and may offer a myriad of hybrid devices and applications.

Graphene oxide (GO) based rolled-up tubes are emergent materials that, in contrast to carbon nanotubes, provide full access to their exposed layers at the inner, outer and interlayer space among sheets; thus resulting in advanced electronic structure.^[1] These rolled-up structures mainly assume an Archimedean-type spiral form resulting from the scroll formation at one corner of a graphene square, for instance. Until now, GO rolled-up tubes were processed by strong sonication treatments, chemical microexplosion, nanomaterials modification among others.^[2-5] As a result of such outstanding configuration along with the recognized properties of GO, GO based rolled-up tubes have been explored in myriad of applications such as superlubricity, sensors, supercapacitors, batteries or catalysis.^[6-10] Theoretical studies predict the use of GO in electromechanical actuation applications, which could make a step forward in the implementation of these structures in advanced devices by coupling selective membranes, electronics fields and actuation.^[1,11-14]

Micro and nanomotors are tinny devices that self-propel in an aqueous media by converting physical and (bio)chemical energy into motion. The great potential of micromotors has been widely demonstrated during the last decade in multiple applications that go from the biomedical field, environmental monitoring and water remediation to diagnostics and operation in real-life environments.^[15-22] Micromotors have been fabricated utilizing a wide range of technologies, including microelectronic technology, electroplating, 3D-printing, and inkjet printing.^[23-26] Since the first thin solid films rolled-up into nanotubes, tubular micromotors have been the most applied.^[27-29] However, different efforts have been directed to build more cost-efficient and less laborious tubular machines that overcome the limitations of microelectronic technology related to the requirements of a clean room facility and trained personal.^[30] In this context, this is the first report on a simple, paper-based, wax-printed graphene rolled-up micromotors. Graphene have been already the structural material of some janus and electroplated tubular micromotors, but not in connection to the wax-printed

(‘stamping’) technology presented herein which is extremely advantageous in term of cost-efficiency and versatility for future applications.^[18,30-31]

Here, we report on a simple, cost-effective and straight forward paper-based, GO rolled-up process for the fabrication of on demand engineered micromotors. We stand the production of well-shaped GO based rolled-up tubes at different levels of oxidation, thicknesses and lateral dimensions for two different geometries (squares and rectangles). The resultant GO rolled-up tubes are further modified to show magnetic and catalytic movement. The reported micromotors have the ability to open and close reversibly as bubbles are formed/ejected from their internal cavities. Such cyclic process is easily modulated by the amount of fuel provided and can be observed by optical microscope due to GO optical transparency. The resultant rolled-up structures displace smoothly in water and oil environments. Indeed, as a proof of concept, the as-prepared GO rolled-up micromotors are successfully exploited for oil removal from water. Although janus and rolled-up microfabricated micromotors have been developed with graphene, this is the first report that relies on an easy, operator-friendly, fast and cost-efficient wax-printed paper-based method. Such method have been previously reported by our group for printed electronics into substrates, but not in connection to micromotors.^[18,32-33] A suitable modification of such process by inducing curvature on the GO sheet until rolled-up tube formation provides a novel route to build versatile on-demand engineered tubular GO structures. This technology allows for a mass production of GO rolled-up tubes which was further exploited for micromotors fabrication, thus expanding and boosting the use of GO tubular structures beyond the current limitations.

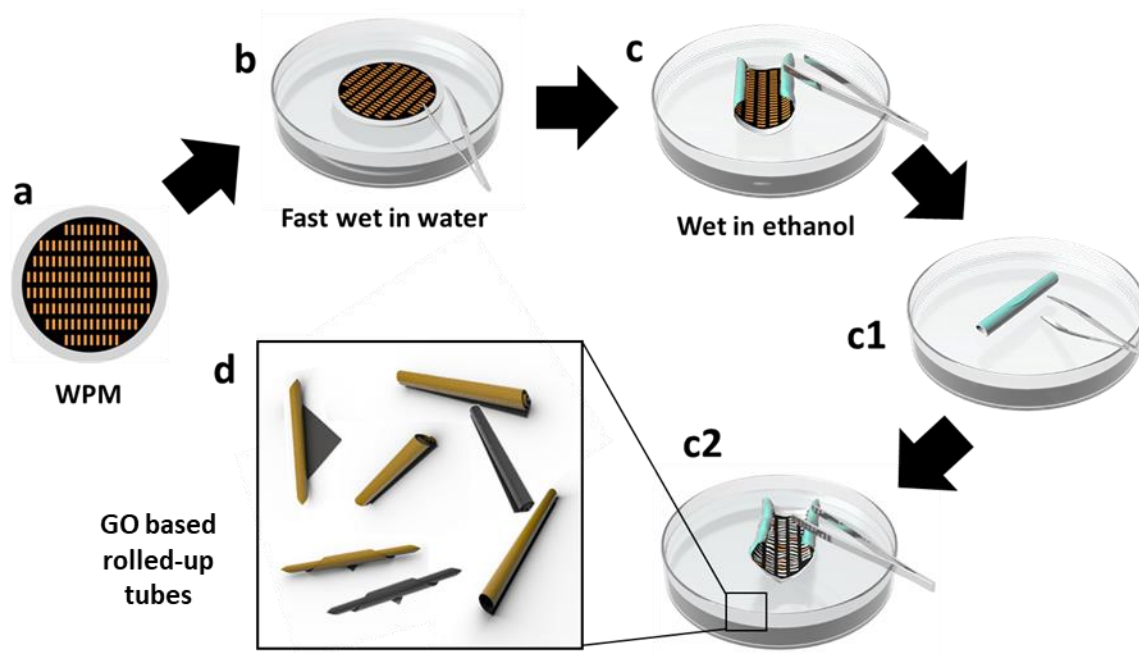


Figure 1. a) WPM modified with GO. b) WPM modified with GO being fast wetted in water and c) wetted in ethanol. c1) WPM modified with GO automatic rolled-up tubes upon contact with ethanol and c2) being manually unrolled. d) Resultant GO based rolled-up tubes.

The process to produce GO rolled-up tubes can be divided in three steps (see scheme in **Figure 1**): First a solution of GO sheets is filtered through a wax-patterned membrane (WPM) and dried in air for a least one hour (Figure 1a). After drying, the bottom part of the GO-coated WPM is wetted in water and the excess of water removed with a tissue (Figure 1b). In a third step, the membrane is transferred into ethanol (Figure 1c) and subjected to one minute of manual lateral shaking (Figure 1c1 and 1c2). Noteworthy, to form rolled-up materials the wetting time in water must be as short as one second. Such an instantaneous wetting step generates latter enough stress to produce rolled-up tubes when the GO-coated WPM is soaked in ethanol (Figure 1c and 1d). Higher wetting times, such as 1 minute for instance, enables to release much easier the filtered GO structures but hinders the rolled-up process. As a result, on demand suspended structures such as patterned letters or medusa-like structures can be

obtained (Figure S1, Supporting Information). A further discussion of this process is described below.

The rolled-up process here reported does strongly depend on several experimental parameters, such as thickness, shape and GO oxidation level. For a thorough study of the roll-up process, 350x700 μm GO films were prepared at different concentrations. For example, GO concentrations of 10, 5 and 2.5 $\mu\text{g/mL}$ lead to 350x700 μm GO films with thicknesses of 84.1 ± 7.1 , 60.7 ± 7.9 and 57.2 ± 1.7 nm respectively. After placing the WPM in ethanol and shaking the solution, rolled-up tubes were spontaneously formed. Ethanol was chosen in this work, among other solvents with higher apolarity constants (such as, isopropanol or 1-octanol), because tends to precipitate GO and reduced GO (RGO) sheets, and does not dissolve neither the membrane nor the wax, while inducing the enough stress or shrinkage over the membrane that enable it to roll-up. We also note that absolute ethanol (99%) was more effective in the scroll formation than ethanol 96% (Figure S2, Supporting Information). The GO and RGO rolled-up structures tend to precipitate and agglomerate in ethanol medium after one hour. This provides stability to the rolled-up structures in terms of morphology and avoids π - π stacking interactions in the case of RGO structures or dissolution of GO structures (as in case of water medium), being the best condition for long-term storage and further dispersion by simple hand shaking. The resultant structures were stable for 9 months at room temperature storage conditions with the possibility of resuspension. The results and discussion below are related to the scrolling processes done using absolute ethanol.

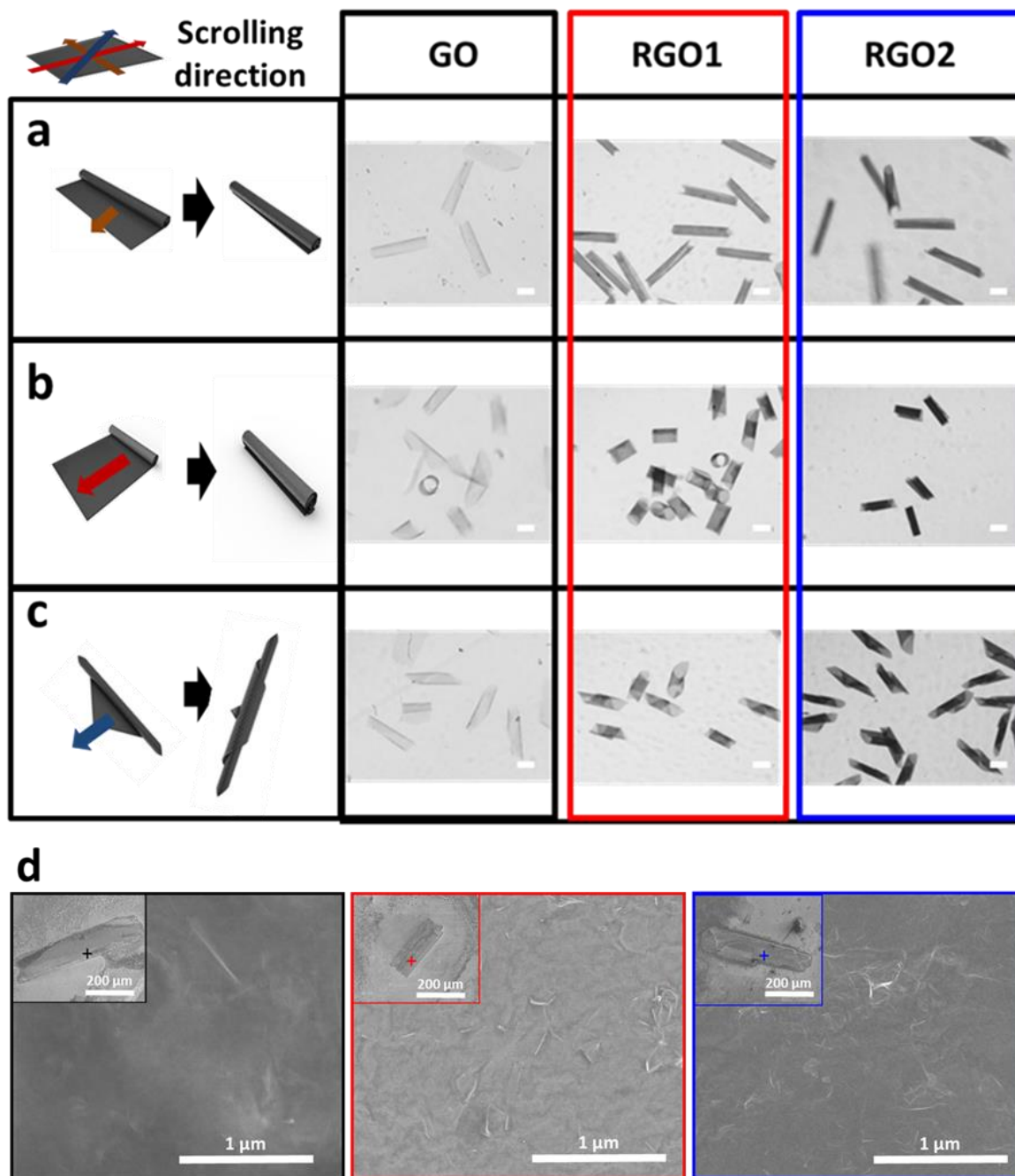


Figure 2. One-sided scrolling from a shortest path (a), from a largest path (b) and diagonal scrolling (c) of GO, RGO1 and RGO2, respectively (scale bar 200μm). (d) Surface SEM characterization images for GO, RGO1 and RGO2.

For sake of simplicity we will focus on the rolled-up process for standard rectangular 350x700μm GO sheets, of two different reduction levels of GO with a concentration of 10

$\mu\text{g/mL}$. **Figure 2** summarizes the different rolled-up events that take place on a rectangular shaped film depending on the scrolling direction for three different levels of oxidation. Note that due to symmetry, squares have only two rolling-up directions while rectangles have three. The rolling direction of the WPM when soaked into ethanol defines the type of GO based rolled-up tube obtained after hand shaking. The shaking direction is inverse to the scrolling direction and thus, defines the direction of the GO based rolled-up tubes as represented in **Figure 2**. This can be slightly controlled by the position of the tweezer in the desired corner of the WPM (**figure 1c**) and the direction of the rectangles upon wetting in ethanol. Additionally, we have also experimentally observed that the wax design can influence the WPM rolling process. Both strategies could be used in the future, for precise control of rolling direction and thus, control over the final rolled-up tube. On one hand, we observed laterally scrolled structures from the two different scrolling pathways; either rectangular shape but short-side or rectangular shape but long-side rolled-up (**Figure 2a** and **2b** respectively). On the other hand, diagonally rolled-up structures represent the longest tubes formed (**Figure 2c**). The efficiency of this process is approximately 82% which results in the formation of approximately 2492 rolled-up tubes per WPM. It is known that reduction methodologies (such as the experimentally used in our process) increase structural defects in the structure at the nanoscale, affecting the mechanical properties of GO based films and thereby the scrolling process.^[34] Yet, no visual defects on the structure, such as cracks, were visualized by SEM (**Figure 2d**).

At this point, it is worthy to underline that the WPM used for producing rolled-up tubes was custom designed to boost the scrolling process. The design consists on the formation of edges along the vertex introducing small triangles (see **Figure 3a**), which facilitate the release of the GO structures from the WPM.

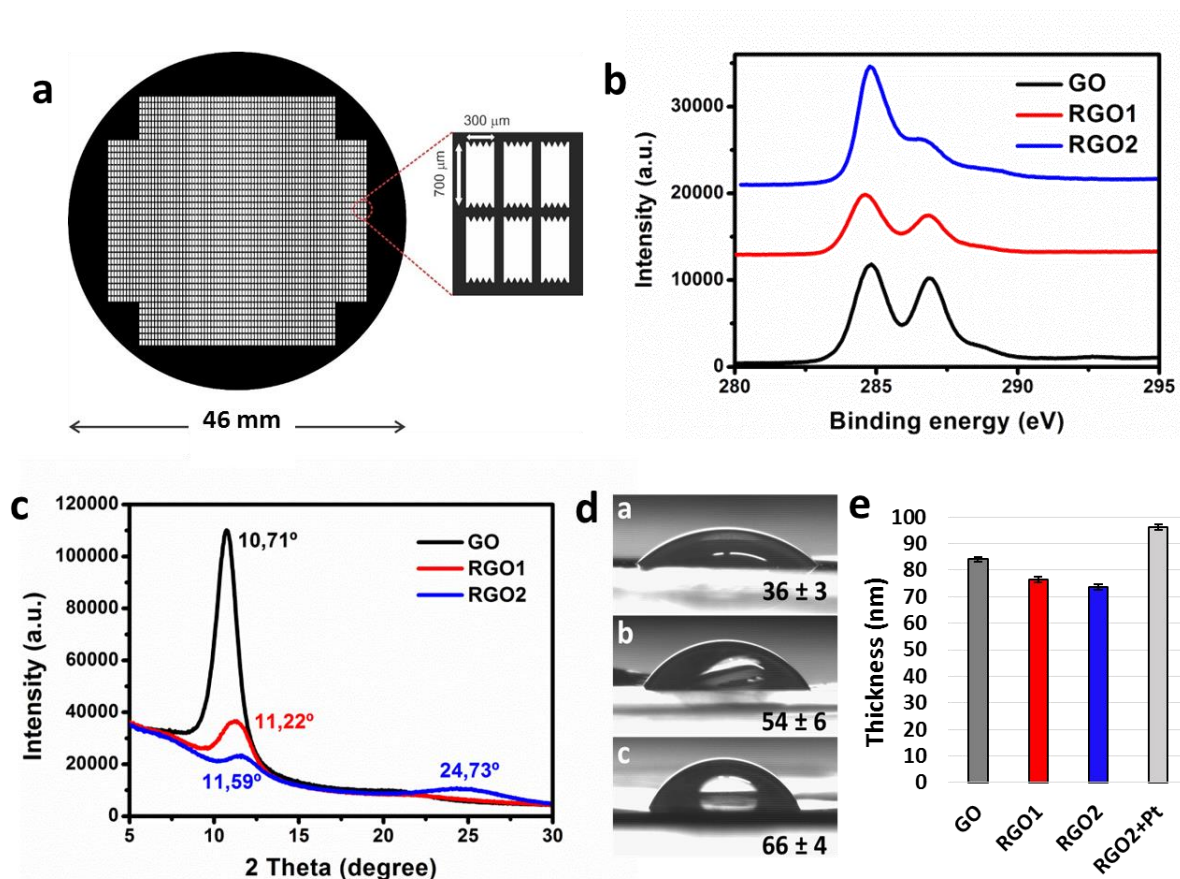


Figure 3. a) WPM used for scrolling GO with 3040 rectangles. X-ray photoelectron spectroscopy (XPS) (b) XRD (c) spectra of the rolled-up tubes based on the GO type. d) Hydrophobicity and e) thickness of the different films at different levels of reduction.

Figure 2 shows that we were able to roll-up GO films at two levels of reduction (RGO1 and RGO2) with C/O ratios of 2.97 and 3.37 respectively (note that GO has a C/O ratio of 2.38, see Figure 3b and c). The reduction was performed by placing the GO filtered through WPMs in 1mg/mL ascorbic acid for one (RGO1) and three (RGO2) days, respectively. X-ray diffraction (XRD) analysis showed how the film structure changes from a d-spacing of 0.82 nm of GO to 0.79 nm in RGO1, while two attributed peaks of 0.77 nm and 0.36 nm were observed for RGO2 (Figure 3c). Such decrease in d-spacing is in agreement with the GO reduction process, where the GO sheets spacing decreases as a result of the elimination of intercalated oxygen-carbon by carbon-carbon structure bonds.^[34] Such reduction process is

not complete and oxygen functional groups still remain. This evolution of the structure increases the hydrophobicity of the material (Figure 3d) alone and decrease the thickness from 84.1 ± 7.1 to 73.5 ± 1.7 nm for GO and RGO2, respectively (Figure 3e).

It is worthy to underline that the properties of RGO are considerably different compared to GO and hence the scrolling process. On one hand, GO-coated membranes have the ability to absorb water molecules on their structure by using the oxygen open venues, but they cannot absorb ethanol molecules.^[13,35-36] For this reason, and in order to have only absorption of water molecules on the bottom surface, the wetting process must be fast. In this way, the rolled-up tube formation is achieved by using the interior dried layers from the GO based films as elastic precursors for scrolling. On the other hand, RGO has less venues for the water adsorption process as the stacked and hydrophobic carbon-carbon layers prevent water adsorption inside the structure, thereby staying only at its surface. As a result, the initial water wetting step is not as crucial as for the GO sheets. This hydrophobicity seems to be a crucial issue for the formation of thinner rolled-up tubes, that is, the formation of rolled-up tubes with smaller radius in comparison to the rolled-up tubes produced with the pristine GO. In other words, reduced sheets appear easier to scroll than pristine GO sheets. We latter confirm this hypothesis studding the rolling-up process by filtering 5 mL (10 μ g/mL) of a GO solution thorough 350x350 μ m squares. Interesting, reduced squares (RGO1 and RGO2) form totally rolled-up structures unlike his pristine GO counterparts of same size and shape (Figure S3, Supporting Information). This control experiment highlights the role of the GO reduction on the scrolling process. Nonetheless, this is not only the parameter to be taken in consideration. A combination of thickness, lateral dimensions and oxygen content are the crucial parameters that enable or disable the rolled-up tubes to form at a given energy provided by the ethanol/water interface. We hypothesize that the above mentioned parameters directly define the mechanical properties of the sheet and hence the energy necessary to scroll them. We would like to highlight the relationship between the supplied energy and the mechanical

properties of the sheets. Rigid sheets (reduced sheets) are more feasible to be scrolled which means that the energy provided by the water to ethanol interface might be stored in the sheet as mechanical stress and later released during the scrolling process. Instead, flexible sheets (GO sheets) might absorb this energy for instance as a plastic deformation which will not be available for the roll-up process. Noteworthy, these rolled-up tubes can also be opened or unscrolled by simple releasing a drop of ethanol containing the rolled-up tubes into a large volume of water, or in the reverse way, by dropping a drop of water containing the structures into a large volume of ethanol. Finally, we observed a limitation on the geometry: large sheets are not able to be rolled-up probably due to the larger section and thus higher mechanical resistance of the sheet to be rolled-up (Figure S4, Supporting Information). Electrical features of the rolled-up tubes were studied by using cyclic voltammetry in 1M H₂SO₄ (Figure S5, Supporting Information). RGO showed a typical capacitive behavior, where the higher the scan rate higher the capacity, which opens a way to capacitive-based sensors and actuators worthy of being explored.

To demonstrate the feasibility of using this process to produce rolled-up micromotor structures, we modified the WPM coated with GO sheets surface to induce catalytic movement and magnetization, thus transforming the rolled-up tubes into micromotors. For this purpose, 350x700 μ m rectangles (see Figure 3c and **Figure 4a** and 4b) and 350x350 μ m squares of RGO2 (Figure S3, Supporting Information) with a concentration of 10 μ g/mL GO were coated with a 20 nm platinum layer by sputtering (detailed conditions in the experimental section). After deposition of platinum, the scrolling process was even more efficient resulting in tighter rolled-up tubes with smaller radius ($69.3 \pm 5.9 \mu$ m) in comparison with the RGO2 sheets without the platinum layer ($94.8 \pm 6.5 \mu$ m) (Figure S6, Supporting Information). Interestingly, these rolled-up in all directions either one-sided or diagonal in the same WPM. In contrast, the same platinum coverage over GO did not allow an efficient rolling-up process as platinum tends to be released/lifted-off/peeled-off from the GO sheet

and form fractured small rolled-up structures (Figure S7, Supporting Information). This fact reinforces the influence of the reduction level of the GO sheets on their scrolling capabilities. RGO2 based rolled-up tubes can be transferred into aqueous solutions by slow addition of the solution and further ethanol evaporation. The sputtering process could be easily replaced by filtration of Platinum nanoparticles through the GO-coated membranes, analogously to that used to provide the structures with magnetic properties, as will be shown below. It demonstrates one more time the simplicity and versatility of the fabrication process reported herein.

After forming the tubular structures with a platinum layer on the inner cavity, catalytic motion was tested by using hydrogen peroxide as fuel. Figure 4c shows a diagonal flexible scroll which can autonomously and cyclically go from a completely rolled-up structure, with formation of big bubbles, to the corresponding opened initial rectangular shape upon bubbles release. This behavior, thus far never observed for graphene based rolled-up tubes, reassembles the features of shape memory alloys or stimuli-responsive shape memory polymers/composites (see movie 1).^[37-38] Structures of short-side scrolled rectangular shapes, opened and closed rather fast. Such actuation mechanism enables a mechanical energy to displace the structures in addition to the catalytic one (see figure 3d and movie 2). These soft micromotor actuation systems were observed when navigated in a solution containing 2.5% sodium cholate (NaCH) and 1% H₂O₂ moving at a speed of ~400 μm/s. NaCH is a surfactant added to the solution to change its surface tension. Such a change helps to modulate the size and frequency of the O₂ bubbles ejected from one of the microtubular structure sides, which in the last stay is responsible for its motion.^[24]

By changing the H₂O₂ concentration, the fold and un-fold process can be tuned up. Completely irreversible or more rigid rolled-up tubes demonstrated to be typical tubular micromotors with potential for creating a micro-vortex effect that accelerate the kinetics of a variety of chemical reactions as reported³⁹ (figure 3e and movie 3).^[39] As a result of the

catalysis on top of the platinum layer, the wax-assisted micromotors self-propelled with a rocket-like configuration for diagonal scrolled structures.^[40] The formation of oxygen bubbles at the inside of the RGO rolled-up tubes provides a directional thrust at a speed of $\sim 50 \mu\text{m/s}$ when navigated in a solution containing 2.5% NaCH and the very low concentration of 0.2% H_2O_2 . Square based RGO Platinum-modified based rolled-up tubes show similar features as discussed above (Figure S8, Supporting Information and movie 4).

Further work could be developed in order to determine the mechanical stability of the RGO rolled-up micromotors at higher thicknesses and their behavior under bubble formation. It is known that the flexibility of the RGO is lower at thicker films, which could be interesting for the production of more stacked tubes, more resistant to bubble formation at higher H_2O_2 concentrations, for specific applications.

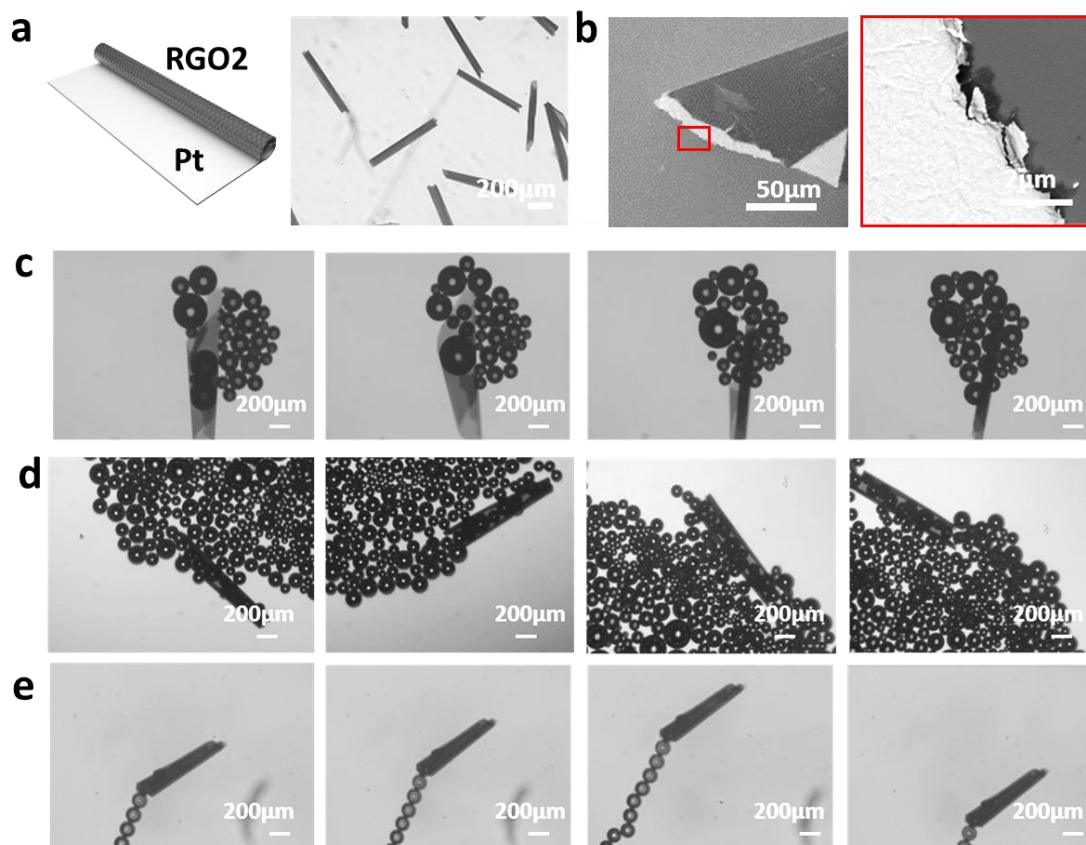


Figure 4. a) RGO2 modified with Platinum schematics, b) optical microscope image and b') closed-up image. Catalytic displacement of 350x700 μ m RGO2 platinum-modified rolled-up tubes. Displacement of c) cyclically opening and closing of a diagonal flexible structure, d) a semi-opened and e) a tight closed structure, respectively. Motion conditions are 2.5 % NaCh and 1% H₂O₂ for c) and d) and 0.2% for e).

To further expand the potential of the as-prepared micromotors, we filtrate magnetite nanoparticle (NP) (5 mL at a 0.1mg/mL) solutions over filtered GO in order to produce GO-NP hybrid rolled-up tubes. Once the GO layer is formed at the top of the WPM, NPs can be filtered through, thus resulting in a NPs thin layer only on the top face of the membrane (above GO). Such a simple filtration process provides rolled-up tubes with magnetic properties, which can be exploited as a steering system to guide micromotors over large volumes or pre-concentrate target analytes (Figure S9, Supporting Information and movie 5). Due to the packed GO structure upon drying, the filtration is slow and can comprise agglomerated islands of NP on top of the GO film in the WPM, which can affect micromotors movement and directional properties. Simple layer-by-layer self-assembly or advanced binding chemistry could be innovative strategies for the production of layered films of nanoparticles at the surface of GO, compatible with our proposed method. Filtrated NPs are a major step-wise in the production of rolled-up tubes with nanoparticles based solutions and could be extended to other nanomaterials such as Pt, Au or CdSe nanoparticles, among others.

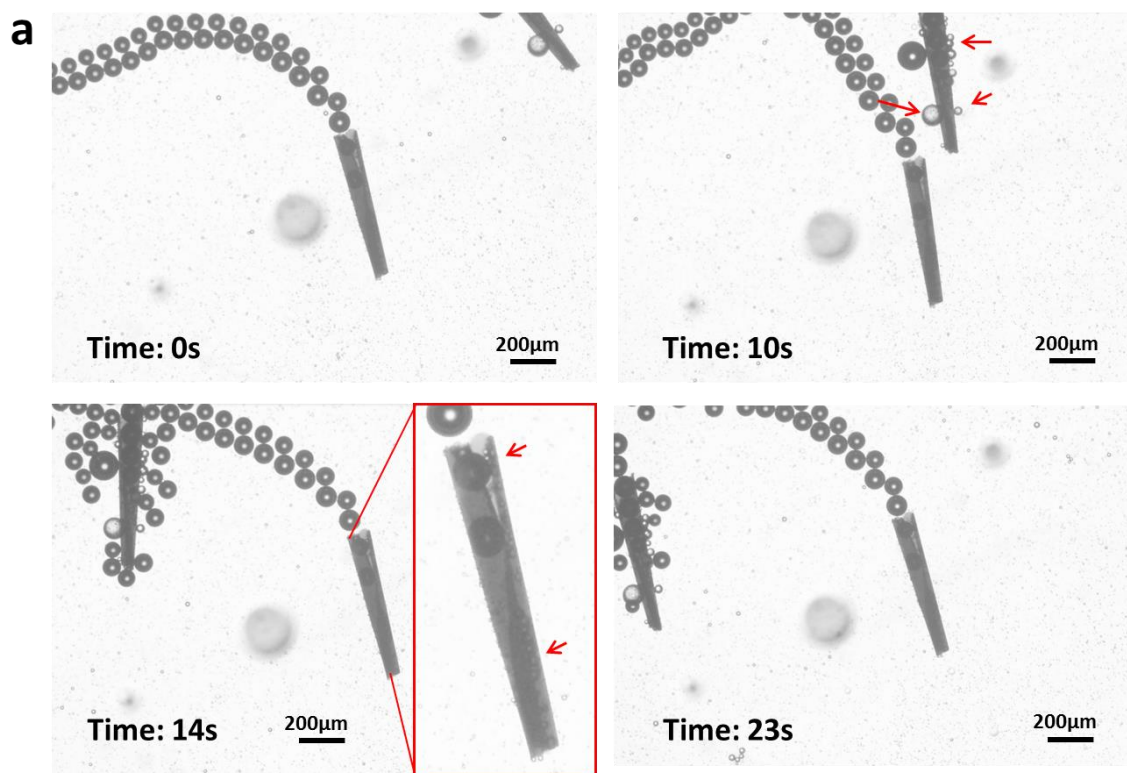


Figure 5. a) Proof-of-concept of oil removal using 300x700µm RGO2 platinum-modified rolled-up tubes. Micromotor navigating at initial conditions a), oil collection after b) 10 s (attached oil indicated in red narrows), c) 14 s (closed-up image in red with bubble forming in the inside), and d) 23 s. Motion conditions are at 2.5 % NaCH and 0.2 % H₂O₂.

To demonstrate a practical application of the wax-printed rolled-up RGO micromotors, they were successfully applied for oil collection from water. Taking advantage of the hydrophobic nature of RGO tubular-shaped micromotors, oil droplets were removed from water. The high surface to volume ratio associated with the motor material and their self-propelled movement offer favorable conditions to collect oil droplets present in water through hydrophobic interactions. After 5 min incubation of RGO2 platinum-modified micromotors in a solution

containing 2.5 % NaCH, 0.2 % H₂O₂ and 50% oil, successfully capture and transport of oil droplets was observed (**Figure 5** and movie 6). Considering the capability of RGO to adsorb and release oil, the micromotors open a door for renewable cleaning features.^[41] This result highlights the great potential of such micromotors for the dynamic removal of large amounts of pollutants from water. Herein, the oil clean up application was presented only as a proof-of-concept. In a practical scenario, a judicious scaling up of the technology must be necessary. GO sheet-based micromotors have demonstrated to have potential to perform remediation tasks. For example, persistent organic pollutants and heavy metals got efficiently stacked and adsorbed on GO sheet-based micromotors, whereas, they were able to carry and transport cargo, even several times higher than their size, owing to their powerful towing force. ^[18,30,41] Such well-demonstrated properties along with the easiness of the fabrication process open-up a myriad of opportunities in the environmental field. Overall, this work showed a simple and cost-effective way to produce on demand GO-based structures. Among them, those modified with platinum and magnetite nanoparticles holds great promise as micromotors with potential for the transport of different cargos. It opens an avenue for their use not only in cleaning processes but also in release applications such as drug delivery or self-repairing/healing; both in the environmental and biomedical fields.

In conclusion, we have developed on demand GO-based rolled-up tubular structures, by using a very simple and cost-effective WPM-assisted approach. We studied the roll-up tubes formation conditions and probed the ability to produce on-demand shaped structures either one-sided or diagonal rolled-up tubes of two different geometries and three levels of GO oxidation. The planar structures were successfully decorated with platinum and NPs and the resultant modified rolled-up structures self-propelled either by catalysis or magnetization. Upon bubble formation resulting from the reaction of platinum and H₂O₂, rolled-up tubes were able to move in a typical rocket-like architecture or to open and close rapidly with shape

memory. As a proof-of- concept, we used RGO platinum-modified rolled-up tubes for oil removal in water. Such wide range of actions can be exploited for instance to capture or encapsulate different cargos for its further release or elimination. Overall, this work shows for the first time a user-friendly and cost-effective process to produce flat or rolled-up tubes structures of GO-based flexible composite for a wide range of applications. Optical properties of graphene quantum dots, amenable with paper-based manufacturing technology, are worthy of being explored for the development of rolled up tubular micromotors. The presented structures combined with their electrical features can be of profit for enhanced micromotors, soft micromachines, biomimetics, kirigami-like structures and developing of micro-opto-electro-mechanical devices. ^[42-45]

Experimental section

Filtration of GO on wax-printed membranes. Nitrocellulose membranes (pore size 0.025 μ m, Merck Millipore Ltd) were printed in a wax printer (Xerox ColorQube 8580) into the desired shape to produce WPM. 5mL of GO (Angstrom Materials) with the desired concentration was filtered through the open pores unprinted by the wax using vacuum filtration. For producing GO Platinum-modified rolled-up tubes firstly the GO coated WPM were left to dry overnight and the sputtering procedure was done on top of the WPM coated with GO. For the production of GO-NP rolled-up tubes, firstly the GO coated WPM were left to dry overnight and a 5mL of a solution of NPs were filtered over the top of GO coated WPM for 5min. The remaining solution was removed using a pipette and the WPM coated with GO-NP left to dry.

Reduction of the membranes and contact angle measurements. GO WPMs were reduced by using a 1 mg/mL ascorbic acid (Sigma Aldrich) solution. After filtration, membranes were left to dry at least for one hour and placed in the ascorbic solution for 24h (RGO1) or for 72 hours (RGO2). After reduction, membranes were washed with Milli-Q water to remove any trace of ascorbic acid.

Sputtering of Platinum layer. Sputtering of Platinum was carried out on a Leica EM ACE600. Current intensity (deposition rate) *i.e.* 30, 60 and 90 mA (0.1, 0.23 and 0.46 nm s⁻¹), respectively and thickness of the layer (20, 40 and 60 nm) were tuned to get optimal motion of the micromotors. Optimal conditions were set at 30 mA current, 20 nm Platinum layer and a rotation speed of ~18 rpm.

Synthesis of magnetic NPs. Magnetite nanoparticles were prepared by the co-precipitation method mixing 0.05 mol of $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ and 0.025 mol $\text{FeCl}_2 \cdot 4\text{H}_2\text{O}$ in 250 mL of ultrapure water, using a 500 mL three neck flask. The mixture was stirred under nitrogen atmosphere and heated until 80 °C, and then 20 mL of NH_4OH from the stock solution, was added (drop wise). The reaction remained under reflux for 40 min and was cooled down at room temperature. The black product was separated and washed with ultrapure water three times.

Graphene oxide rolled-up tubes characterization. The X-ray Photoelectron Spectroscopy (XPS) measurements were performed with a Phoibos 150 analyzer (SPECS GmbH, Berlin, Germany) in ultra-high vacuum conditions, pressure 1E-10mbar) with a monochromatic aluminium Kalpha x-ray source (1486.74eV). The energy resolution as measured by the FWHM of the Ag 3d5/2 peak for a sputtered silver foil was 0.58 eV. Scanning Electron Microscopy (SEM) was done on a FEI Quanta FEG (pressure: 70Pa; HV: 20kV; and spot: four). Thickness measurements were done using Profiler KLA-Tencor P15. The samples were electrically characterized using Autolab302 potentiostat/galvanostat/frequency-response analyzer PGST30, controlled by GPES/FRA Version 4.9. Optical microscopy images were obtained on a Nikon Eclipse LV100 microscope with a 20x/0.45 objective lens.

XRD measurements were performed with a X'pert MPD diffractometer (Multipurpose Diffractometer) at room temperature using a $\text{Cu } K_\alpha$ radiation ($\lambda=1.540 \text{ \AA}$). This diffractometer has a vertical theta-theta goniometer (240 mm radius), where the sample stages are fixed and do not rotate around omega axis as in omega-2theta diffractometers. The detector used is an X'Celerator which is an ultra-fast X-ray detector based on Real Time Multiple Strip (RTMS) technology. The diffraction pattern was recorded between 4 and 30° using an step size of 0,03° and a time per step of 1000s.

Supporting Information

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

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Graphene oxide rolled-up tubes production process using wax printed membranes for the fabrication of on demand engineered micromotors is presented. This micromotor production technology relies on an easy, operator-friendly, fast and cost-efficient wax-printed paper-based method and may offer a myriad of hybrid devices and applications.

Keyword

Micromotors, wax printed membranes, graphene, rolled-up tubes.

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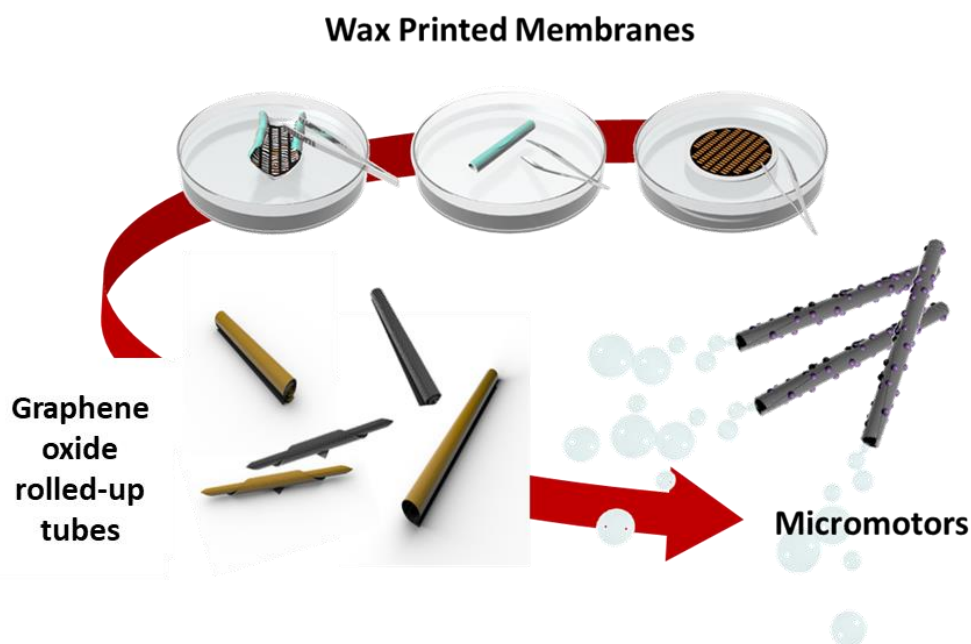
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Title

Architecting graphene oxide rolled-up micromotors: a simple paper-based manufacturing technology

TOC

Supporting Information

Architecting graphene oxide rolled-up micromotors: a simple paper-based manufacturing technology

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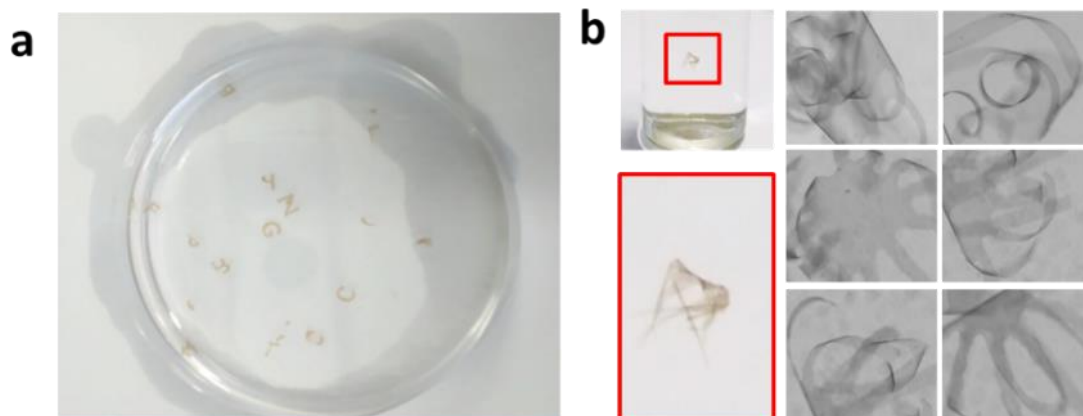


Figure S1. a) GO patterned letters released from the WPM in ethanol medium. b) GO medusa-like structure released from the WPM in ethanol medium.

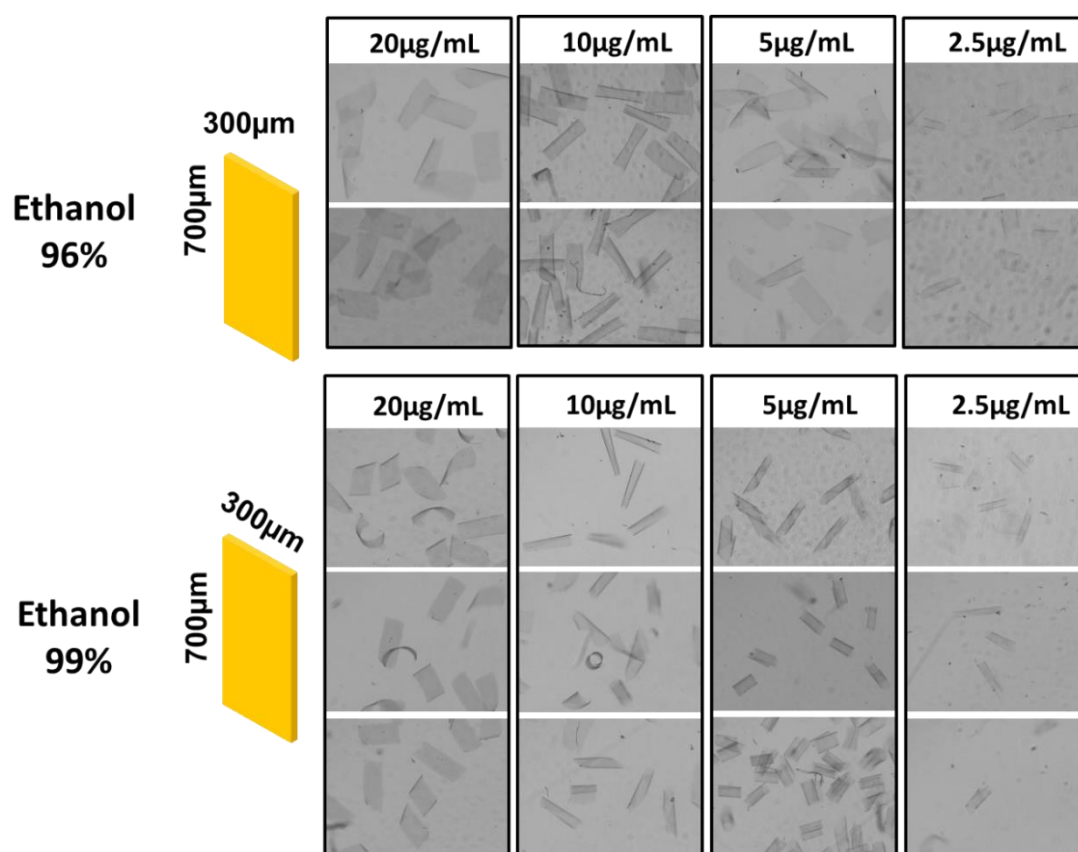


Figure S2. GO rolled-up structures released from the WPM at different concentrations in ethanol 96% and ethanol 99%.

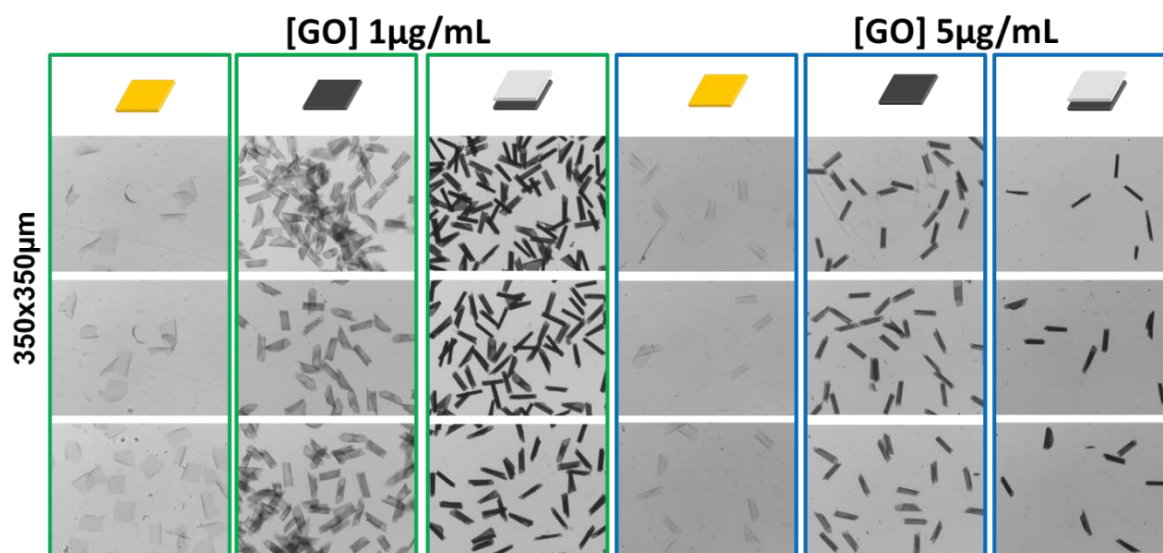


Figure S3. GO, RGO and RGO Platinum-modified rolled-up squares at two different concentrations.

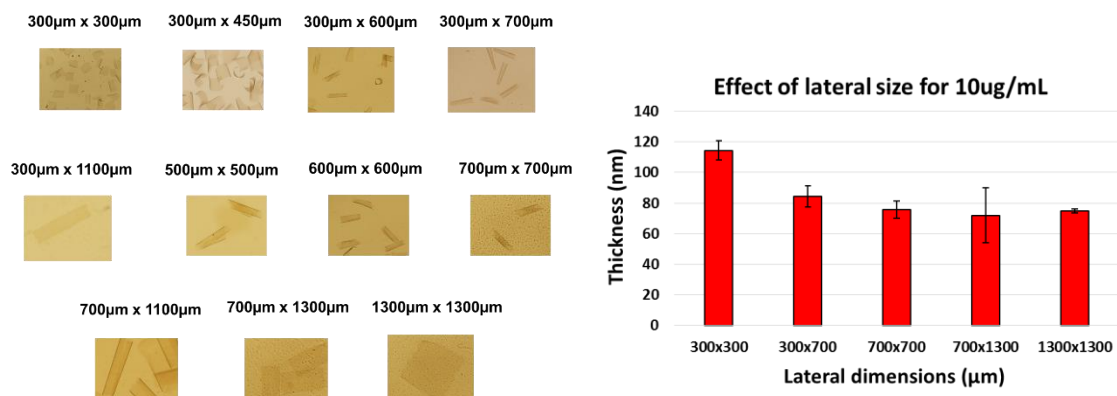


Figure S4. Rolled-up GO tubes at different lateral dimension and effect of lateral size on the sheet thickness for the same concentration.

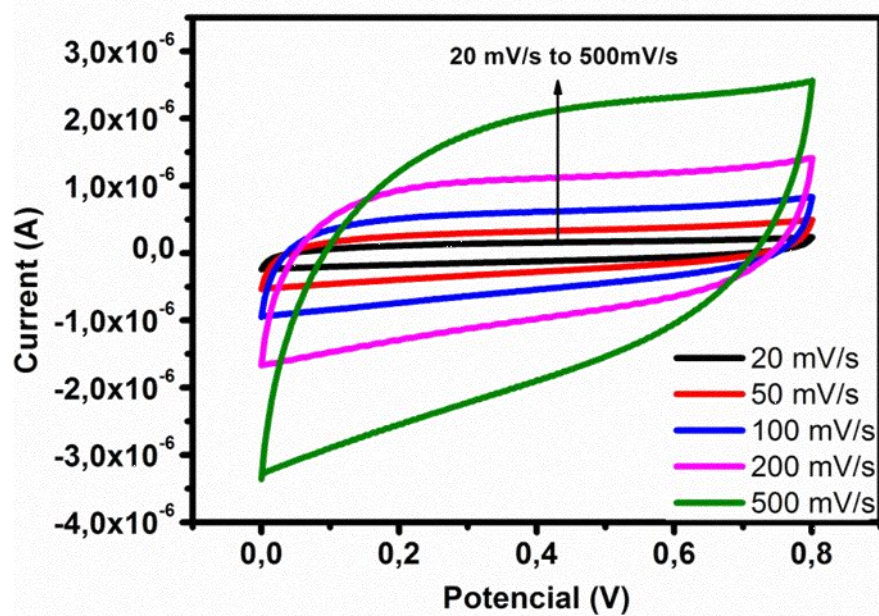


Figure S5. Electrical properties of the RGO rolled-up tubes using CV measurement in H₂SO₄ (1M).












	Diameter (μm)	Size (μm)		Diameter (μm)	Size (μm)		Diameter (μm)	Size (μm)
GO_1 	84 \pm 13.5	455 \pm 29.1	RGO1_1 	95.2 \pm 5.63	580.8 \pm 13.64	RGO2_1 	100 \pm 10.9	604.8 \pm 23.50
GO_2 	101.1 \pm 12.3	256.1 \pm 28.2	RGO1_2 	145.1 \pm 4.5	267.3 \pm 15.75	RGO2_2 	92.3 \pm 2.51	285.3 \pm 16.02
GO_3 	110.6 \pm 18.4	425.3 \pm 60.5	RGO1_3 	112.6 \pm 2.08	515 \pm 27.09	RGO2_3 	92.1 \pm 8	438.3 \pm 10.41
						RGO2_Pt 	69.28 \pm 5.93	576.3 \pm 36.15
						RGO2_MB 	126.6 \pm 20.63	295.6 \pm 44.95

Figure S6. Diameter and diagonal size of different rolled-up tubes (GO, RGO, RGO Platinum-modified and RGO-NP).

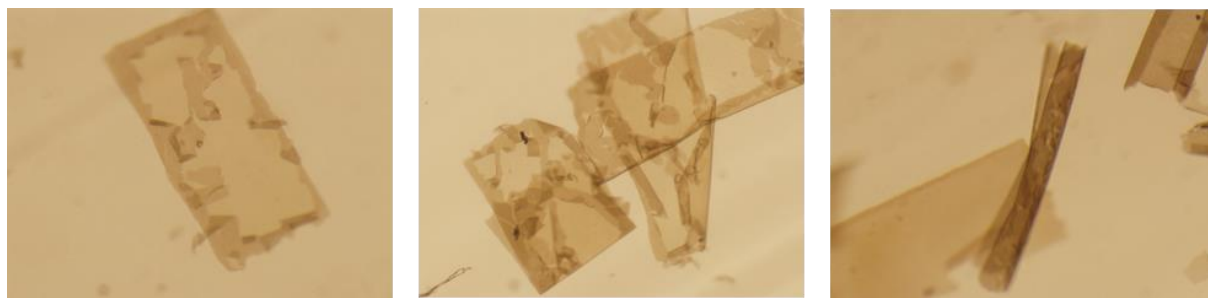


Figure S7. Rolled-up GO Platinum modified tubes.

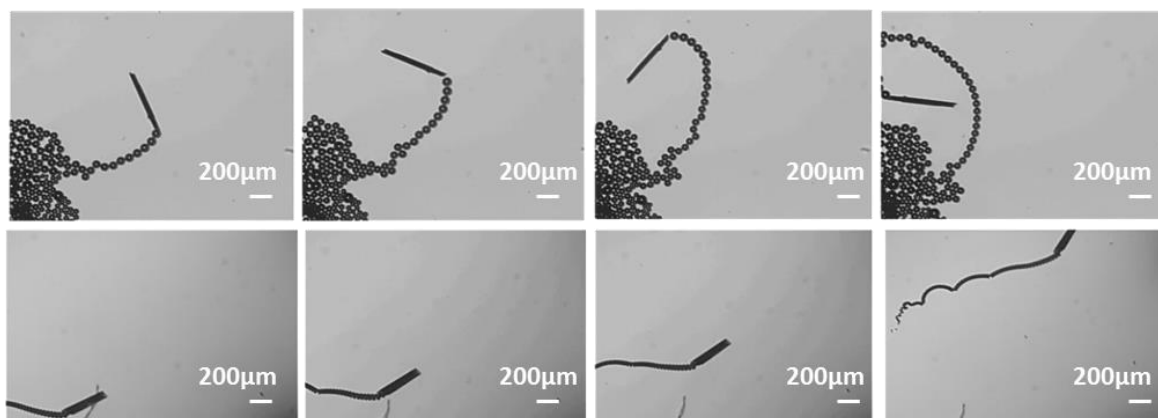


Figure S8. Catalytic movement of rolled-up RGO platinum-modified squares with initial lateral dimension of 300x300μm.

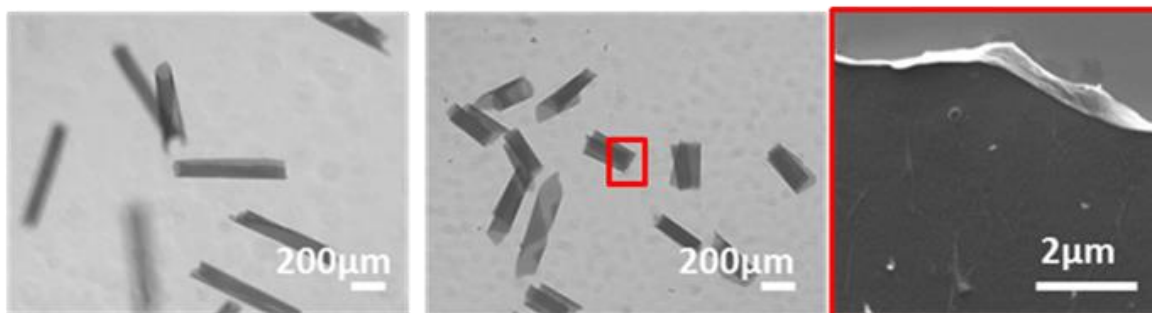
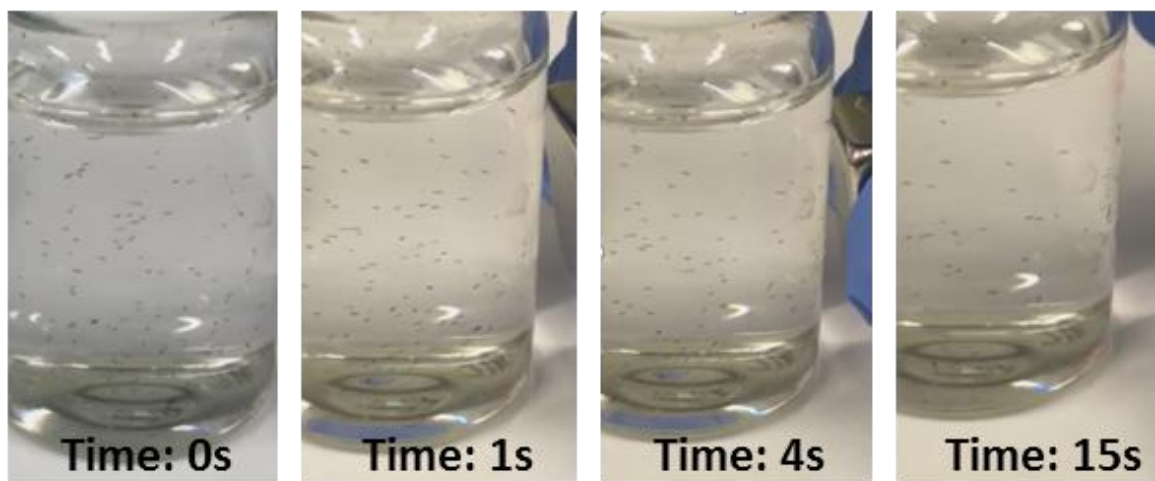
a**b**

Figure S9. RGO-NP rolled-up tubes. a) optical microscope images and SEM images. b) displacement of the RGO-NP rolled-up micromotors upon the proximity of a magnet.