Contents lists available at ScienceDirect

## Nano Energy

journal homepage: www.elsevier.com/locate/nanoen

# Laser-processed graphene based micro-supercapacitors for ultrathin, rollable, compact and designable energy storage components

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#### ARTICLE INFO

Article history: Received 14 March 2016 Accepted 27 April 2016 Available online 27 April 2016

Keywords: Micro-supercapacitor Surface mountable supercapacitor Aluminum electrolyte capacitor Graphene Laser

#### ABSTRACT

With the development of wearable/flexible electronics, a formidable challenge is to integrate electronic components which were large in their original size into a flexible, thin, and arbitrary layout. As an indispensible component in electronics, commercial micro-supercapacitors are disadvantageous in their clumsy cuboid geometry and limited capacity, and are not promising for future applications. In comparison, film-like micro-supercapacitors are superior in miniaturized system integration since they can be folded to fit in restricted spaces while maintaining a high level of volumetric energy density. Here, we carried out a benchmark study of a state-of-the-art well-packaged thin film micro-supercapacitor toward commercial micro-supercapacitor and aluminum electrolyte capacitor. The micro-planar supercapacitor not only exhibits 3.75 times of a commercial micro-supercapacitor and 8785 times of an aluminum electrolytic capacitor in volumetric energy density under 1000 mV s<sup>-1</sup> scan rate, but can also be tailored into diversified shapes, rolled up, and plugged into tiny interstitial spaces inside a device. Such ultrathin  $(18 \,\mu\text{m})$  micro-supercapacitor component with high volumetric energy density (0.98 mWh cm<sup>-3</sup> in LiCl-PVA gel, 5.7 mWh cm<sup>-3</sup> in ionic liquid), can be integrated into an electronic device system and shows a series of superior performance characteristics over current commercial benchmarks, which may find vast applications.

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## 1. Introduction

With the bloom of wearable electronics, the technology of highly integrated electronic systems has significantly advanced. For example, Apple Co. recently launched the Apple Watch, which shows the most advanced system in package (SiP) technology that integrates most components into a single module. Without exception, this technological trend has intrigued the development of highly-miniaturized and arbitrarily-shaped energy storage units that can excellently fit into a system and still maintains the performance characteristics such as high energy and power densities.

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http://dx.doi.org/10.1016/j.nanoen.2016.04.045 2211-2855/© 2016 Published by Elsevier Ltd.

Besides the major power source such as the lithium ion battery pack, aluminum electrolytic capacitor (AEC) and electrochemically isolated energy device cells (i.e. micro-supercapacitors) are also important components for power supplementation in microelectronics [1-3], and they have been usually surface-mounted to flexible printed circuit (FPC) for various applications such as automatically shutting-down and driving flash-lights. Considering that AEC has a much larger size, surface mountable supercapacitor (SMS) has become more popular nowadays due to its more compact size and higher capacity. However, for its wider application, it is necessary to further shrink its size down to the level of resistors and ceramic capacitors, which still face significant technical challenges due to their intrinsic electrolytic characteristic and the corresponding packaging difficulties. For example, a commercial Seiko CPX3225A752D SMS (7.5 mF) has a dimension of  $3.2 \times$  $2.5 \times 0.9$  mm<sup>3</sup> (SMD type: 1210), which is much larger than a 0402 resistor (dimension:  $1.0 \times 0.5 \times 0.5$  mm<sup>3</sup>). Further miniaturizing the size of micro-supercapacitor and maintaining a comparable capacity to commercial SMSs (i.e. several mF) still remains a great







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challenge; for instance, Karolina U. Laszczyk et al. successfully developed the micro-supercapacitor with the component area down to about 1 mm<sup>2</sup> by using photolithography and ion etching techniques, yet the capacity of a single cell was only  $34.3 \,\mu\text{F}$  [4]. Considering the very precious surface area for a FPC and in order to maintain a high level of component capacitance, it would be a wise choice to develop ultrathin, flexible, integratable and designable micro-supercapacitor components which can be rolled-up or even folded and plugged into the seams between the other functional parts in a device [5]. From the system level point of view, the precious surface spaces on FPC can be saved, and those spaces which have been conventionally regarded useless can be utilized now. To be noted, there are readily plenty of rooms in a portable electronic for receiving such a new type of component. An analogous case is the commercial wireless charging module for smartphones, which can be attached on top of the lithium ion battery pack and beneath the back cover, which does not account for any increase of the whole device size.

Recently, the emergence of planar supercapacitor is regarded as an important member in the family of miniaturized energy storage devices, which has drawn unprecedented attentions in science community [6-9]. As compared with the conventional supercapacitors which have a sandwich structure, a planar layout can render the diffusion length independent to the electrode thickness, and thereby it allows more active material loaded per unit area while maintaining excellent rate performance and capacitance [10]. More importantly, owing to the planar layout and elimination of separator, the total thickness of supercapacitor device significantly decreases, showing great potential in microelectronic applications. Most recently, there have been remarkable advances regarding the fabrication of planar supercapacitors. For instance. John Chmiola et al. reported a planar supercapacitor based on carbide-derived carbon originated form sputtered TiC film with chlorination process, showing a high volumetric capacitance [7]. David Pech et al. deposited the onion-like carbon on integrated gold current collector, giving an ultrafast discharge rate  $(200 \text{ V} \text{ s}^{-1})$ , which was three orders of magnitude higher than conventional supercapacitors [11]. Wu et al. fabricated a graphene based planar supercapacitor on arbitrary substrates, which exhibited ultrahigh rate performance and excellent energy density [12]. However, a critical step toward this technology transfer to industry is to elaborate the detailed fabrication and packaging technology of the state-of-the-art planar micro-supercapacitor at electronic component level. Besides, there still lack a benchmark study regarding such planar micro-supercapacitor component with the commercial-available counterparts.

In the last decade, laser fabrication technology featured with high efficiency, ultrahigh resolution and low cost has presented great potential in miniaturized device fabrication. Recently, because of the interaction between laser beam and graphene oxide (GO), laser technology has been used to reduce GO into graphene (a two-dimensional carbon sheet with 2675  $m^2 g^{-1}$  of theoretical specific surface area (SSA,) and 550  $Fg^{-1}$  of theoretical specific capacitance), which has drawn much attention in miniaturized energy storage field [13–15]. For example, Yonglai Zhang et al. fabricated desired micrometer-sized graphene circuits on GO films by using femtosecond laser nano-writing, which brought about a new horizon for applications of graphene-based materials in electronic micro-devices [16]. El-Kady et al. reported a scalable fabrication of graphene based planar supercapacitors by laser writing on GO films using a commercial LightScribe DVD burner, showing excellent performance [17]. Zhiwei Peng et al. demonstrated that boron can be doped into graphene by direct laser treatment, which delivered higher areal capacitance [18]. In light of the superior performance for planar supercapacitor based on GO treated with laser, the combination between high performance planar supercapacitor and micro-electronics may play more important roles for next generation of consumer electronics [8].

Here, we report a simple and scalable fabrication technology for the pure laser-processed graphene based micro-planar supercapacitor (LPG-MPS) electronic component which has superior volumetric energy performance characteristics over the commercial micro-supercapacitor benchmark. Via tuning the output laser power, a 355 nm nanosecond laser machine can both reducing the electro-sprayed GO thin layer into the laser-processed graphene (LPG) and patterning the electrode arrays. A thin layer of nickel (Ni, 500 nm in thickness) sputtered on a piece of polyethylene terephthalate (PET) film (6 um in thickness) was used as the current collector to support the electrostatic-spray deposited GO laver [19]. Electrostatic spray deposition technique can ensure a homogeneous coverage of GO with adjustable thickness, which is beneficial to maintain a high quality of the micro-supercapacitor electrode. By laser scanning with different powers, this layered structure can be patterned with great convenience into: 1) the LPG area supported by the Ni layer and 2) the totally ablated area with only the PET film remained (Fig. 1). After the laser treatment, the LPG-MPS with excellent rate performance and cycle stability shows high volumetric energy density (0.98 mWh cm<sup>-3</sup> in LiCl-PVA gel,  $5.7 \text{ mWh cm}^{-3}$  in ionic liquid electrolyte), which is comparable to recently reported works (Fig. 3(f)). In term of packing process, a protective PET cover layer is applied on LPG-MPS by the polyurethane (PU) based hot melt glue printed with stencil printing method for encapsulation [20], and the total thickness of LPG-MPS device is only 18  $\mu$ m, which shows 3.75 and 8785 times to SMS and AEC in volumetric energy density in ionic liquid electrolyte at 1000 mV s<sup>-1</sup> scan rate. In order to demonstrate the potential application in micro-device, LPG-MPS devices connected to the FPC by the anisotropically conductive film (ACF) technique as the supplementary energy storage component can successfully drive three pieces of light emitting diodes (LEDs), and can be rolled-up and folded to be tucked into the seam in an iPhone 4 smart phone and a Jawbone UP 24 smart bracelet. Combining the mature technologies of electrostatic spray deposition, laser treatment and stencil printing, our technology shows obvious technical advantages and is readily scalable for production.

#### 2. Experimental

## 2.1. Fabrication of nickel film on PET

Poly(ethylene terephthalate) (PET, 6  $\mu$ m-thick) was chosen as the flexible substrate and the sputtering technique was employed. The nickel layer was controlled to be 500 nm in thickness by magnetron sputtering at ambient temperature, and the square resistance of the sample is about 0.15  $\Omega/\Box$ .

#### 2.2. Preparation of GO film

In order to obtain a homogeneous GO film, electrostatic spray deposition method was used. Briefly, GO was synthesized using a modified Hummer's method [21], and then subjected to strong sonication in deionized water for 2 h. The precursor (1.5 mg mL<sup>-1</sup>) containing equal volume GO solution and ethanol was stirred for 30 min. Subsequently, the precursor was electro-sprayed under 18 kV operating voltage onto the surface of sputtered nickel layer at 0.5 mm min<sup>-1</sup> injection speed. In addition, the temperature of spray deposition process was 50 °C, aiming at a fast evaporation of the solvent (ethanol–water mixture), which contributed to form a uniform GO film. In this process, the thickness of GO film can be well-controlled.



**Fig. 1.** Schematic illustration of ultrathin laser-processed graphene based micro-planar supercapacitors. **The PET film (6 µm) sputtered with Ni (500 nm) was homogenously** deposited with GO layer by electrostatic spray deposition. After the different powers of laser treatment, the reduction and ablation of GO film were completed, resulting in the formation of LPG and micro-electrode layout, respectively. With a thin PET film printed with crosslinkable PU cofferdams used as cover layer, the LPG-MPS coated with electrolyte as an integration to apply in micro-electronics.

#### 2.3. Fabrication of LPG-MPS

A typical LPG-MPS was fabricated within two steps. Firstly, the GO film was exposed to the laser beam (wavelength: 355 nm, model: Han's Laser EP-15-DW, nanosecond frequency, scanning line speed 5 m s<sup>-1</sup>), which enables the GO to turn into LPG. Afterward, GO area was also subjected to the laser beam with more power so that GO and nickel layer can be removed, which can prevent short circuit between the two adjacent electrodes without damaging the underlying PET film. In this work, the area of a single LPG-MPS is 15.0–17.6 mm<sup>2</sup>, and the width of integrated electrode and interspace were both 100  $\mu$ m and these configurations can be easily revised.

#### 2.4. Packaging of LPG-MPS

The LPG-MPS was assembled with ultrathin PET film (6  $\mu$ m) printed with polyurethane (PU) cofferdams (Henkel Loctite 3542). The regular PU cofferdams can be prepared on the surface of PET film by simple stencil printing. Because of the sensitivity to humidity, PU glue should be hot-pressed in 2 min. For the preparation of LiCl-PVA gel electrolyte, LiCl (12.75 g, Adamas) and PVA (poly(vinyl alcohol), 6 g, Adamas) were mixed with deionized water (60 mL, purified by Millipore Milli-DI water purification system), which were heated to 90 °C with stirring until the mixture became clear. Subsequently, after nitrogen plasma treatment (550 V, 170 mA, model: PDC-MG, 150 W, Ming heng company) for 2 min, the LPG array were uniformly casted by the LiCl-PVA gel electrolyte, and carefully laminated with PET (printed with PU cofferdams) under ~20 Pa at 80 °C for 5 min. LPG-MPS casted with ionic liquid electrolyte (1-ethyl-3-methylimidazolium)

tetrafluoroborate, EMIMBF<sub>4</sub>) should be operated in glove box filled with argon. Control samples are the commercial SMS (ROSWIN, RW, AD102, 2.5 V, 25 mF) and AEC (SANYO, 47  $\mu$ F, 6.3 V).

#### 2.5. Materials characterizations and electrochemical measurements

The morphological characteristic of LPG and LPG-MPS were investigated by scanning electron microscopy (SEM, ZEISS SUPRA 55, 5 kV, Germany) and high-resolution transmission electron microscopy (HR-TEM, FEI G2 spirit, 300 kV, USA). Additionally, water contact angle (CA) measurement (Kruss DSA 30, Germany), Fourier transformation infrared spectroscopy (FTIR, Nicolet iS50R, U. S.), X-ray photoelectron spectroscopy (XPS) measurement (ES-CALAB 250Xi, Thermo Fisher Scientific, U. S.), and Raman spectroscopy (LabRAM HR800, HORIBA Jobin Yvon, Japan) were involved for materials characterizations.

Cyclic voltammetry (CV), galvanostatic charging/discharging (GCD) and electrochemical impedance spectroscopy (EIS) of the as-prepared samples under a two-electrode configuration with LiCl-PVA gel electrolyte or ionic liquid electrolyte were investigated on an electrochemical station (VMP3, Bio-Logic, France). The applied potential window of CV and GCD was in the range from 0 V to 0.9 V with LiCl-PVA gel electrolyte (0–2.5 V, ionic liquid electrolyte). The EIS was conducted in the frequency range between 0.01 Hz and 100 kHz with amplitude of 5 mV at an opencircuit potential. The areal capacitance (C<sub>s</sub>), volumetric energy density ( $E_v$ ) and power density ( $P_v$ ) were calculated from the GCD curves according to the equations below.

$$C_{s} = \frac{I \cdot \Delta t}{\Delta U \cdot S}$$



**Fig. 2.** Materials characterizations of LPG. (a) SEM image showing the contrast of LPG (top) and GO (bottom). (b) SEM image giving the open structure of LPG at high magnification. (c) Cross-view SEM image of LPG (height ~4 µm). (d) A TEM image of LPG (inset: SAED of LPG). (e) Optical microscopic image of the electrode arrays (dark: LPG electrodes, bright: ablated bare PET surface). (f). Photographic image of LPG-MPS arrays. (Inset is a single unit).

$$C_{v} = \frac{C_{s}}{h} = \frac{\int i(V) dV}{V \cdot v \cdot \Delta U}$$
(2)

$$E_{v} = \frac{C_{v} \cdot (\Delta U)^{2}}{2 \cdot 3600}$$
(3)

$$P_{v} = \frac{E_{v}}{\Delta t}$$
(4)

Where  $C_s$ ,  $C_v$  is the areal and volumetric capacitance of LPG-MPS. I is the applied current, i (V) is the voltammetric current, v is the scan rate and  $\Delta t$  is the discharge time. S is the total area of device, including all interspaces and h and V is thickness and total volume of LPG-MPS, respectively;  $\Delta U$  is the operating voltage window.

#### 3. Results and discussion

Fig. 1 shows the fabrication process of LPG-MPS. A conductive substrate (PET film sputtered with Ni layer) was homogenously loaded with GO film by electrostatic spray deposition that was reported in our previous work [19]. After that, laser processing technique was employed to efficiently fabricate high resolution micro-electrode pattern. It's worth noting that two power settings were used in the fabrication of LPG (Fig. S1), which play two roles: one is to provide adequate photon energy to reduce GO into LPG with a low power level (1 W, beam size 10  $\mu$ m); the other is to ablate/evaporate both the GO and Ni layers to pattern the electrodes with a high power level (2 W, beam size 15  $\mu$ m). By controlling the laser scanning program, the fabrication of a large piece of LPG array becomes very convenient, as compared to recently reported planar supercapacitor preparation technologies, which involved multiple steps within limited fabrication area [22–24].

As shown in Fig. 2(a), after laser beam treatment, the LPG greatly expanded and exfoliated as compared with the flat and

untreated GO region, which facilitates ions transport to electrode surface. Detailed inspection in both Fig. 2(b) and Fig. S2a clearly indicates the regular open structure of LPG. From the cross-sectional observation of LPG (Fig. 2(c), Fig. S2b), the height of the electrodes was about 4 um. Transmission electron microscopy (TEM) analysis (Fig. 2(d)) also confirmed that LPG has the open structure as well, which is conducive for ions to transport in the electrode. The selected area electron diffraction (SAED) pattern (inset of Fig. 2(d)) of LPG displays a bright diffraction ring corresponding to the (100) graphene plane, suggesting that the conjugated structure of graphitic domains in GO was recovered [25]. Taking the advantages of laser-scribing technology, complex and high resolution patterns can be conveniently fabricated in largescale. To prevent short circuit within LPG-MPSs, the Ni conductive layer can be removed by increasing the power of laser beam. Fig. 2 (e) gives magnified optical photograph of the integrated LPG electrodes (dark parts) separated by the bright PET substrate, with a clear interface. The width and distance of each LPG electrode were as low as 100  $\mu\text{m},$  which is conducive to accelerate ion transports and results in superior rate performance [17,18,26]. Additionally, combining computer aided design technique and laser technology, LPG-MPSs with various geometries can be obtained and connected in series or parallel, which help cater to the electronic device with various operating voltages and currents. Fig. 2(f) shows the array of LPG-MPSs, and the inset is the magnification of single supercapacitor with high resolution.

When the GO layer was exposed to the laser beam, a deoxygenation reaction is triggered instantaneously via both photochemical and photothermal reduction pathways [13,27]. It can be observed that the color of sample changed from gray to black (Fig. S3). This reduction from GO to LPG accompanied with the change of wettability. Fig. S4a, b shows the contact angles (CA) of both GO and LPG for deionized water droplets. The CA of GO was 91.8° showing moderate wettability; where the CA of LPG was up to 147.9° reflecting a hydrophobic surface, which manifested the successful conversion from GO to LPG. Moreover, this conversion can be confirmed by X-ray photoelectron spectroscopy (XPS) and Fourier Transform infrared spectroscopy (FTIR) characterizations. As shown in Fig. S5a, b, both GO and LPG have signals of carbon



**Fig. 3.** Electrochemical characterization of LPG-MPS. (a) CV curves under different scan rates ranging from  $0.1 \text{ V s}^{-1}$  to  $2.0 \text{ V s}^{-1}$  with the LiCl-PVA gel electrolyte. (b) GCD curves with various current densities ranging from  $0.3 \text{ mA cm}^{-2}$  to  $5.0 \text{ mA cm}^{-2}$ . (c) Nyquist plot of LPG-MPS; (inset) magnified plot of the high-frequency region. (d) Cycle stability of device undergone 20,000 cycles; (inset) the CV curves recorded in cycling process. (e) CV curves of device observed in different bending state. (f) Ragone plot of LPG-MPS and some reported supercapacitors.

and oxygen. After treatment with laser beam, O1s peak intensity of LPG was dramatically decreased compared with that of GO, indicating the elimination of oxygen. The C1s peak of both GO and LPG could be deconvoluted into three peaks that corresponding to C–C, C–O and C=O, respectively (insets of Fig. S5a, b). It's clear that the intensity of C–O and C=O bands were significantly weaker after reduction, and the C/O ratio drastically increased from 2.43 to 17.38, higher than some recently-reported works, strongly suggesting the successful reduction from GO into LPG [3,16,28]. To be noted, UV laser (Nd: YAG, 355 nm) delivers a high photon energy over the threshold for GO reduction (390 nm, 3.2 eV), which involves less oxidative decomposition of GO when processing in ambient condition [29].

Since the oxygenated functional groups are IR sensitive, attenuated total reflectance Fourier transform infrared spectroscopy (ATR-FTIR) spectra (Fig. S5c) can qualitatively measure the deoxygenating reactions. In the spectrum of GO film, the broad peak located at around 3300 cm<sup>-1</sup> is associated with isolated hydroxyl groups and adsorbed water. An intense peak at approximately 1500–1730 cm<sup>-1</sup> can be attributed to carbonyl and carboxylic groups. The presence of O-H bending in tertiary alcohol groups was indicated by a peak at  $1360 \text{ cm}^{-1}$ , which was reported to be located on the basal plane of GO [30]. The peak at 1060  $\text{cm}^{-1}$  corresponds to phenol groups. After the laser treatment, it's evident that the intensity of the characteristic peaks ascribed to most of the oxygencontaining functional groups were significantly reduced, indicating that most of the oxygen-containing functional groups in GO can be effectively eliminated. In addition, Raman spectroscopy (Fig. S5d) showed that the intensity ratio  $I_D/I_G$  increased from 0.99 to 1.07 after the reduction process, which are attributable to the increased amount of small conjugated domains [31].

In order to fabricate a LPG-MPS, LiCl-PVA gel electrolyte or ionic liquid electrolyte was carefully dispensed onto the surface of the LPG electrode arrays for testing at room temperature. Before applying the gel electrolyte, the LPG electrode array was exposed in a nitrogen plasma chamber for two minutes, aiming at improving both the electrical conductivity [32] and wettability of the electrodes; correspondingly, the contact angle dropped significantly from 147.9° to 9° (Fig. S4b, c). To evaluate the electrochemical performance of LPG-MPS, CV, GCD and EIS analyses were used. The LPG-MPS with LiCl-PVA gel electrolyte showed excellent capacitive characteristic and rate performance, as indicated by the almost ideal symmetric rectangular shape of CV curves at various scan rates ranging from 100 mV s<sup>-1</sup> to 2000 mV s<sup>-1</sup> (Fig. 3(a), CV curves under 5000 mV  $s^{-1}$  is displayed in Fig. S6). The GCD test was performed with different current densities, within the voltage window of 0–0.9 V. As shown in Fig. 3(b), the typical triangular shape of GCD plot exhibited negligible IR drop of 0.002 V at 0.3 mA cm<sup>-2</sup>, and 0.060 V at 5.0 mA cm<sup>-2</sup>, by virtue of the small Ohmic contact resistance between Ni layer and LPG [33]. Additionally, the electrochemical performance of LPG-MPS covered with ionic liquid electrolyte was also investigated, and the CV and GCD curves reflected good capacitive feature (Fig. S7). In order to accurately calculate the areal specific capacitance, the CV curves of pure Ni layer and Ni layer deposited with LPG were recorded under the same scan rate (  $1000 \text{ mV s}^{-1}$ ), showing negligible contribution of the Ni current collector to the total capacitance of LPG-MPS (Fig. S8) [34]. Fig. S9 summarizes the areal specific capacitance versus current density based on the entire device area (17.6 mm<sup>2</sup>) including the interspaces between integrated electrodes. In Fig. S9a, the maximum areal specific capacitance of the LPG-MPS with LiCl-PVA gel electrolyte was up to  $3.9 \text{ mF cm}^{-2}$ , which was higher than recently reported results [3,17]. The superior specific capacitance are related to the open porous structure, high surface area and high electrical conductivity etc. of the LPG electrode rendered by the efficient laser reduction technology [13]. Besides, it appears that the current technology is conducive to form a good graphene electrode structure. Because the laser



**Fig. 4.** Comparison of AEC, SMS and LPG-MPS. (a) Photographic image showing the thickness (18 μm, as shown from a caliper) of a piece of LPG-MPS array containing 6 units. (b) Photographic images of LPG-MPS, SMS and AEC. (c) CV curves of LPG-MPS, SMS and AEC obtained at 1000 mV s<sup>-1</sup>. (LPG-MPS: 694 mF cm<sup>-3</sup>; SMS: 185 mF cm<sup>-3</sup>; AEC: 79 μF cm<sup>-3</sup>).

reduction process here involves with photochemical reduction pathway (photon energy > 3.2 eV, wavelength < 390 nm) rather than with only the photothermal one; and thus it renders less oxidative decomposition effect of GO compared with other laser sources with a higher wavelength when processing in ambient condition [29]. It should be noted that the areal specific capacitance loss was only 20.5% when the current density was increased by a factor about 17 (from 0.3 to 5.0 mA cm<sup>-2</sup>), strongly indicating the outstanding rate performance. The relatively small capacitance in ionic liquid electrolyte can be attributed to poor wettability between LPG and ionic liquid and low mobility of ion (Fig. S9b).

Electrochemical impedance spectroscopy also indicates superior performance of the LPG-MPS. An equivalent circuit model (Fig. S10) was used to fit the EIS data. In Fig. 3(c), the charge transfer resistance (R<sub>ct</sub>, caused by Faradic reactions and the double-layer capacitance on the grain surface) was only 1.01  $\Omega$ , suggesting the good contact between nickel substrate and successful reduction of LPG [35]. In low frequency range, the Nyquist plot showed a vertical line closed to -Z" axe, giving a pure capacitive behavior of supercapacitor. Fig. 3(d) shows the cycle stability of LPG-MPS. After 20,000 cycles (scan rate:  $1000 \text{ mV s}^{-1}$ ), the capacitance retention still remained 93%, suggesting excellent cycle stability. From the CV curves recorded during the cycling process, it's clear that the CV curves kept nearly rectangular shape, and the integrated area of the curves showed tiny variation, once again indicating exceptional cycle performance. In order to evaluate the mechanical property of LPG-MPS sample, it was tested under various bending conditions at 1000 mV s<sup>-1</sup> scan rate. From Fig. 3 (e), there were negligible changes of the CV curves under different bending states (bending radius is about 1.5 cm), which greatly suggests excellent flexibility of LPG-MPS. This superior mechanical stability enables LPG-MPS to apply in flexible electronics, such as roll up displays, smart sensors, and radio frequency identification devices [36,37].

As pointed out by Gogotsi and Simon et al., volumetric energy density and power density can provide much more reliable performance metrics for supercapacitor devices compared to gravimetric capacitance [38]. The Ragone plot shown in Fig. 3 (f) suggests that the energy density of this LPG-MPS with LiCl-PVA gel electrolyte achieved 0.98 mWh cm<sup>-3</sup> at the power density of 300 mW cm<sup>-3</sup>, which was almost 3 orders of magnitude higher compared with an AEC and was comparable to some recently-reported planar supercapacitors [4,12,17,18,23]. In addition, with the power density of LPG-MPS had no significant decline, showing excellent rate performance. More importantly, using the ionic

liquid electrolyte can greatly improve the energy density of LPG-MPS, and the max energy density was up to 5.7 mWh cm<sup>-3</sup> at 830 mW cm<sup>-3</sup>, which was comparable with lithium thin-film battery but provided more than 2 orders of magnitude higher power density. The superior electrochemical performances above can be mainly attributed to two aspects: 1) planar micro-electrodes eliminate the separator and minimize the ion diffusion paths, which help to improve the rate performance. 2) The unique open structure of LPG provide more surface area and benefit to a high mobility of the Li<sup>+</sup> ions, both of which help deliver high energy and power density.

For practical applications, assembling LPG-MPSs in series and parallel configuration can help control output voltage and current [39]. Based on the electrospray and laser-scribing technique, the LPG-MPSs with excellent consistency can be easily connected in series and parallel. Herein, the consistency tests were demonstrated by connecting two LPG-MPSs together both in series and parallel configurations under  $1.0 \text{ mA cm}^{-2}$  current density. As shown in Fig. S11, the CV and GCD curves clearly indicated that the supercapacitors connected in series or parallel exhibited almost ideal symmetrically rectangular and triangular shapes, which were consistent with the individual supercapacitor. Meanwhile, the operating voltage window and capacity doubled under the same testing condition, suggesting excellent consistence of each LPG-MPS [40]. Thus, LPG-MPSs with versatile output voltages and capacity can be achieved. The self-discharge behavior of the LPG-MPS was also studied in this work. The device was charged to V<sub>max</sub> (0.9 V) and held for 30 min, and then it took nearly 5000 s for the open-circuit voltage to decrease from V<sub>max</sub> to 1/2 V<sub>max</sub> (Fig. S12).

In term of packaging the LPG-MPS, as shown in Fig. 1, stencil printing technique involved with a thermal-crosslinkable polyurethane (PU) hot-melt glue (Henkel Loctite 3542) was employed, which is simple and reliable. With stencil printing, the regular PU cofferdams can be easily constructed on the surface of the ultrathin PET film (6  $\mu$ m). When the PET film with PU cofferdams was laminated with LPG-MPSs, PU cofferdams not only act as the binder, but also form many isolated cell units that can effectively limit the leakage of electrolyte during shears, strains, and compressions [20]. It should be noted that PU exhibited ultra-strong binding force and superior reliability due to cross-linking reaction, which provided LPG-MPS with excellent mechanical property. As shown in Fig. 4(a), the total thickness of ultrathin LPG-MPS device was only 18 µm which included the carrier substrate, current collector, PU cofferdams and electrolyte. A photographic image of an LPG-MPS, a SMS and an AEC shows the significant difference in size (Fig. 4(b)). The volume of a single piece of LPG-MPS is 27 and



**Fig. 5.** Demonstrations of LPG-MPS device. (a) Photographic image of FPC (front) connected with a LPG-MPS array (behind). (Inset: The LPG-MPS array is wrapped.) (b, c) FPC with three LEDs was driven by LPG-MPS array to deliver dazzling light even after the LPG-MPS array was tailored. (Inset: the tailored LPG-MPS array can drive the FPC work when bended.) (d) Ultrathin flexible LPG-MPS array was tucked into the seam in iPhone 4. (e) The magnified image of location of rolled-up LPG-MPS. (f) The rolled-up LPG-MPS can be inserted into the unused spaces inside a Jawbone UP24 smart bracelet.

321 times smaller than a SMS and AEC, respectively (Table S1), which suggests the great potential in miniaturized device integration. It's clear that supercapacitor plays an important role as power supplementation in an electronic device when an instant and high power is needed. Therefore, high scan rate ( $1000 \text{ mV s}^{-1}$ ) aiming at simulating high power output was employed to compare the performance of LPG-MPS (a single unit), SMS (ROSWIN, RW, AD102, 25 mF) and the AEC (SANYO, 47 µF, 6.3 V). From Fig. 4(c), the CV curve of LPG-MPS (ionic liquid electrolyte) exhibited rectangular shape under the same potential window, while the control SMS showed a fusiform one. Meanwhile, the volumetric capacitance of one LPG-MPS was 3.75 times to a SMS and 8785 times to an AEC (LPG-MPS, 694 mF cm<sup>-3</sup>; SMS: 185 mF cm<sup>-3</sup>; AEC: 79 µF cm<sup>-3</sup>, calculated from Eq. (2)), which demonstrated the ultrahigh power output ability of LPG-MPS.

Besides electrochemical capacitive property, an outstanding mechanical strength of the device is equally important for practical device applications. LPG-MPS devices connected both in series and parallel configurations with PU cofferdams were assembled to an ultrathin flexible LPG-MPSs strip, which exhibited excellent mechanical property (inset of Fig. 5(a)). To demonstrate its application in micro-electronics, as compared to the conventional reflow soldering technique, the flexible LPG-MPSs strip were attached to a FPC test vehicle surface mounted with as simple as three red LEDs by a heat transfer process (Fig. 5(a)). This process has been widely used in industry over decades for the purpose of FPC to rigid board interconnection, and the two parts are connected by a piece of commercial anisotropic conductive film (ACF). ACF technique has been widely used for microchip bonding for its low cost, simple process, small pitch size and excellent mechanical robustness (Fig. S13). The bonding is via a heat transfer process, which can avoid the involvement of a high temperature process yet can provide adequate mechanical and electrical interconnection. After charging at 3 V for 5 s, the LPG-MPSs successfully drove three LEDs to deliver a dazzling light (Fig. 5(b)). Interestingly, the

PU cofferdams as mechanical scaffold enabled the LPG-MPSs strip with tailorable property, which is demonstrated in Fig. 5(c). When tailored by scissors, both the involvement of gel electrolyte and cofferdams can reduce the probability of leakage of electrolyte, guaranteeing the functionality of the other small cells of the LPG-MPSs. Therefore, each piece of the LPG-MPS unit is a completely packaged component and is readily applicable for various electronic applications; as it can be tailored into alien shapes and fit into various microelectronic systems. Due to excellent mechanical property and ultrathin merit (only 18  $\mu$ m for total thickness), the LPG-MPSs strip can be rolled-up, folded, and even tailored at ease (Fig. S14). For demonstration, a piece of LPG-MPS strip was rolledup and plugged into the seam of an iPhone 4 smartphone and a Jawbone UP smart bracelet, giving a way to further exploit the redundant spaces of a device and promote the integration of micro-electronics (Fig. 5d-f). Considering the irregular layout of most of the wearable electronics from ergonomic aspect, there will be vast future market for this technology.

Above studies suggest that: 1) by use of two different laser power settings, both reduction and ablation of graphene-based patterns were successively rendered in one single process, showing great efficiency and convenience. Considering the high quality of both electrostatic spray deposition and laser processing can achieve, the current process can be easily scaled-up for mass production of well-defined, highly uniform graphene electrode arrays for LPG-MPS. 2) LPG-MPS device packaged with PU cofferdams can be conveniently integrated into electronic devices due to the excellent mechanical property and reliability. Table 1 shows performance characteristics comparison of AEC, SMS and LPG-MPS. AEC and SMS were widely employed in practical electronic with reflow soldering technique, while LPG-MPS was integrated into FPC with another mature technique, i.e. heat transfer process, which not only avoided high temperature, but also guaranteed adequate mechanical and electrical interconnection. In addition, the ultrathin LPG-MPS device delivered 3.75 times to a SMS and

Table 1						
Performance characteristics	comparison	of AEC,	SMS	and	LPG-MPS	5.

Device	Connection technique	Operating voltage (V)	Volume (mm <sup>3</sup> )	Volumetric capacitance (mF cm <sup>-3</sup> )	Flexibility	Main application
AEC SMS LPG- MPS	Reflow soldering Reflow soldering Heat transfer process*	6.3 2.5 2.5	86.736 7.200 0.270	0.079 185 694	None None Excellent	Line filtering Power management Power management

\* By choosing thermally stable packaging polymers and ionic liquid electrolytes, LPG-MPS can be further developed to be compatible with conventional reflow soldering process.

8785 times to an AEC in volumetric energy density (under the same operating voltage, 2.5 V), demonstrating the potential application in power management. More importantly, as compared with both the rigid AEC and SMS, the excellent flexibility of tailorable LPG-MPS assembled with PU cofferdams was highlighted, which enabled LPG-MPS to be tailored into versatile shapes and roll-upped to be plugged into tiny gaps to improve the integration of electronics.

## 4. Conclusion

In summary, we presented the ultrathin, flexible and high performance LPG-MPS as a versatile and scalable platform technology, which overcomes the size/performance limitations for current micro-supercapacitor related in flexible/wearable electronic devices. By adjusting the laser power output, the reduction and patterning of LPG electrode arrays can be fabricated in just one batch, and LPG-MPS arrays connected in series and parallel can be easily realized as well. Additionally, benefited by the planar layout and unique open structure of LPG, these LPG-MPSs show outstanding rate performance, excellent cycle stability and high volumetric energy density (0.98 mWh cm<sup>-3</sup> in LiCl-PVA aqueous gel, 5.7 mWh cm<sup>-3</sup> in ionic liquid). Besides, the ultrathin LPG-MPS device (18 µm) was packaged with PU cofferdams and PET film cover layer, which can effectively prevent device failure when LPG-MPSs array was tailored for desirable applications. In addition, LPG-MPS prepared with ionic liquid electrolyte can not only withstand a high processing temperature but deliver volumetric energy density under high scan rate with 3.75 and 8785 times to commercial SMS and AEC, manifesting significant advantage for high performance electronic device integrations. More importantly, that ultrathin and rollable LPG-MPS arrays as the supplementary energy storage and power management component can be plugged into the seams or integrate on FPC with ACF by heat transfer process, which demonstrates their great potential in wearable/flexible electronic applications.

Although the energy density of LPG-MPS with current design was limited by the use of LPG, recent reports have demonstrated that LPG can be mixed with pseudocapacitance material by direct laser treatment, which greatly improved the energy density of LPG-MPS [41–43]. Therefore, we envisage that introducing materials with high-volumetric capacitance and novel electrolytes with a wider operating voltage may sharply promote the energy density of MPS, and benefit the development of novel wearable and miniaturized devices [44,45,46].

## Acknowledgements

This work is financially supported by the National Key Basic Research Program of China (Project No. 2014CB932400), Guangdong Province Science and Technology Department (Project No. 2014B090917002 & 2014B090915002 & 2014A010105002 & 2015A030306010), Hong Kong government TRS Grant no. T23– 407/13-N and Nanshan District "Rising Stars" (Project No. KC2014JSQN0010A) for financial supports.

#### Appendix A. Supplementary material

Supplementary data associated with this article can be found in the online version at http://dx.doi.org/10.1016/j.nanoen.2016.04.045.

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