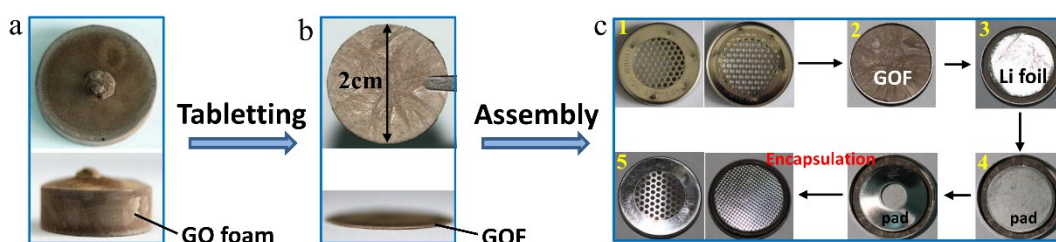


## Supporting Information

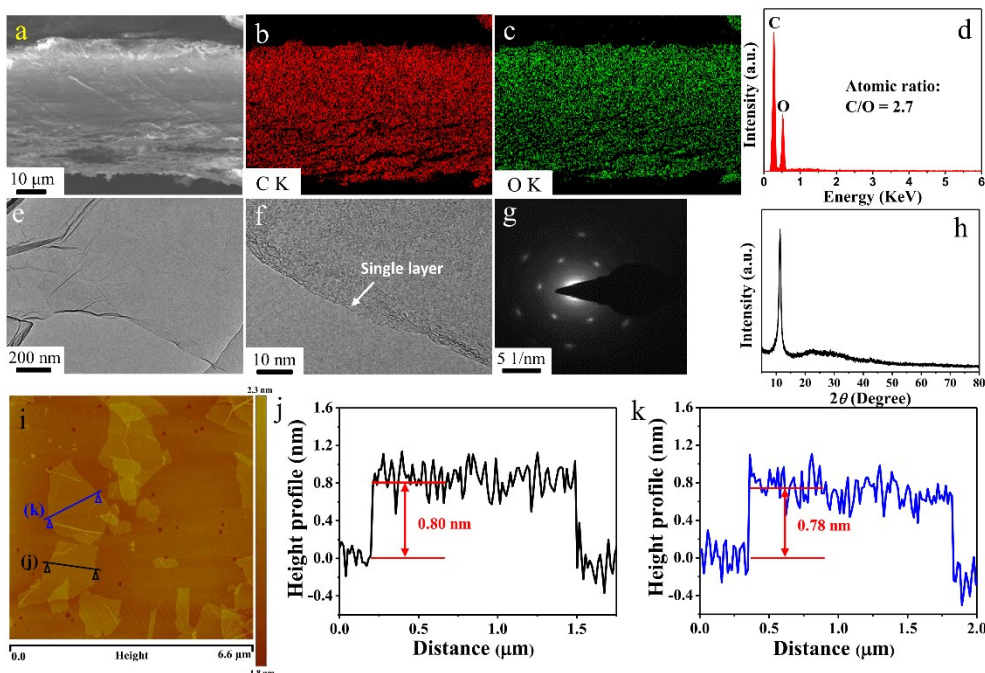
### A respiration-detective graphene oxide/lithium battery

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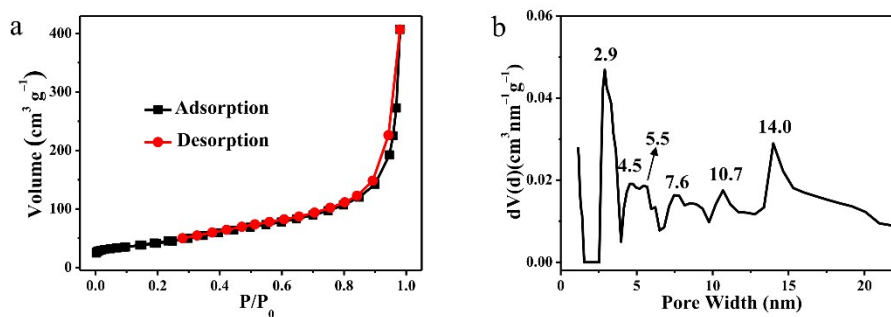
**Fig. S1.** Photographs of the preparation processes of Li-GOF battery, including three-step strategy: (a) The GO solution was freeze-dried to form porous 3D GO foam, (b) tableting GO foam to fabricate graphene oxide film (GOF) and (c) assembling lithium foil and GOF into a button-type cell without additional electrolytes, in which the encapsulation of Li-GOF battery was performed in an argon-filled glove box with a pressure of ca. 6 MPa. The top cap of the cell was punched to guarantee the entrance of water molecules (step 1) and the introduction of pads helped with the closed contact between GOF and Li foil (step 3–5).



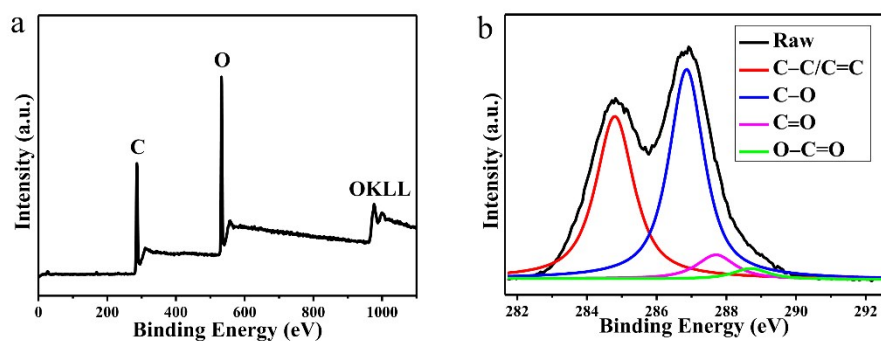
**Fig. S2.** (a) The side-view SEM image of the raw GOF and (b,c) the corresponding C and O maps and (d) EDS. (e,f) TEM images of GO with different magnification and (g) the selected-area electron

diffraction pattern. (h) XRD pattern of GOF. (i–k) AFM image and its height profiles of GO nanosheets.

As shown in Fig. S2a†, the side-view SEM image of GOF revealed that it had a thickness of around 50  $\mu\text{m}$ . The composition and elemental distribution of GOF was analyzed using energy-dispersive X-ray spectroscopy (EDS). Typical EDS mapping analysis showed the uniform distribution of C and O elements (Fig. S2b,c†). The GOF possessed a C/O ratio of 2.7 (Fig. S2d†). The XRD pattern of GOF displayed a sharp peak with  $2\theta$  value of about  $11.3^\circ$  with an interlayer spacing of ca. 0.78 nm that can be indexed to carbon (001) diffraction (Fig. S2h†).<sup>S1</sup> It can be noted that there were many intrinsic wrinkles and ripples on the surface of GO nanosheet (Fig. S2e†). Both the high-resolution TEM image with clearly visible crystalline (Fig. S2f†) and the selected area electron diffraction (SAED) pattern (Fig. S2g†) with a hexagonal structure revealed a single sheet of GO.<sup>S2</sup> Fig. S2i depicted the atomic force microscopy (AFM) images of GO sheets, and the thickness were ca. 0.8 nm (Fig. S2j†) and 0.78 nm (Fig. S2k†) respectively, as marked in Fig. S2i†,<sup>S3</sup> which implied the single-layered GO nanosheet in good agreement with TEM results.

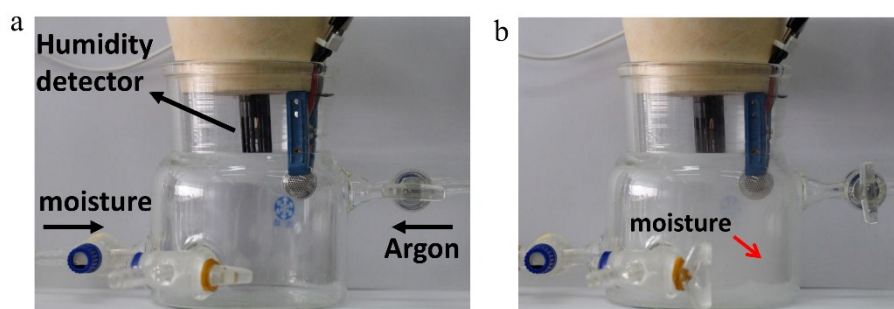


**Fig. S3.** (a)  $\text{N}_2$  adsorption-desorption isotherms of GOF and (b) the corresponding pore-size distributions.

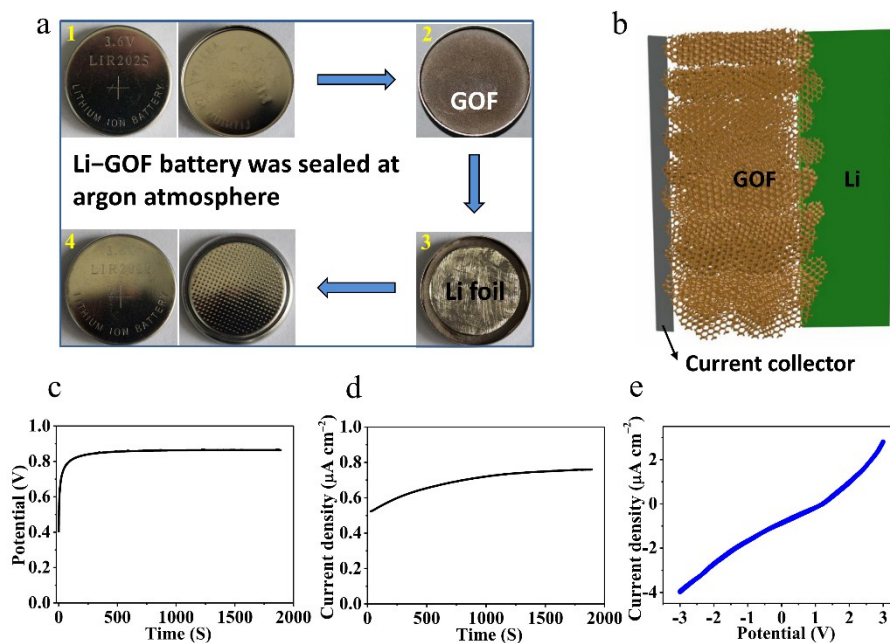


**Fig. S4.** (a) The survey XPS spectrum of GOF and (b) the high-resolution C1s spectrum.

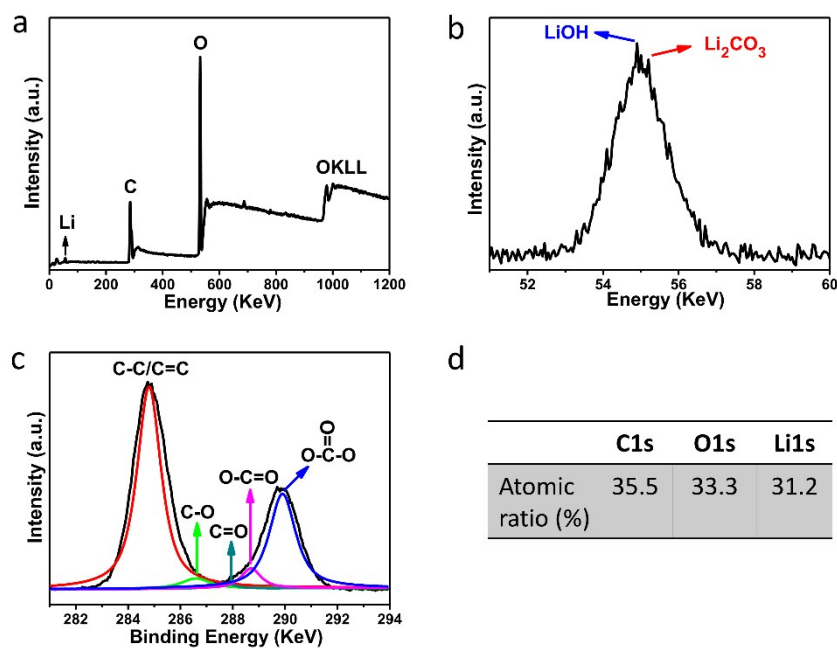
The chemical structure of GOF was investigated by X-ray photoelectron spectroscopy (XPS). As shown in Fig. S4a†, the predominant two peaks were attributed to C1s peak ( $\sim 284.8$  eV) and O1s peak ( $\sim 532.0$  eV). The XPS C1s spectra could be deconvoluted into four peaks and attributed to C–C/C=C (284.8 eV), C–O (286.6 eV), C=O (287.7 eV), and O–C=O (288.7 eV) (Fig. S4b†).<sup>S4</sup>



**Fig. S5.** (a) Photograph of the RH controlling system. (b) The system was filled with moisture (RH = 100 %).



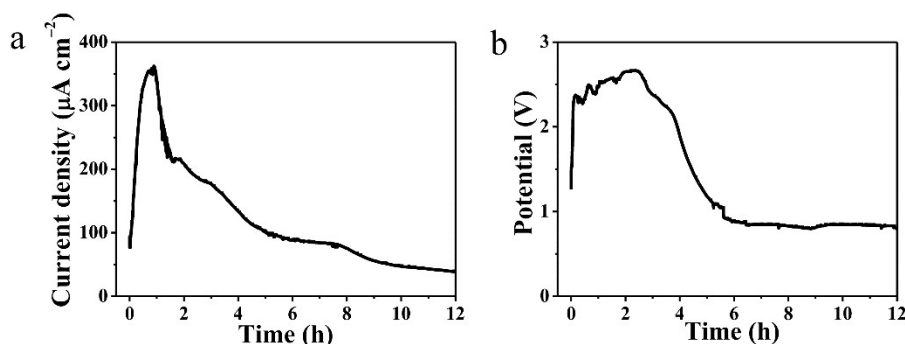
**Fig. S6.** (a) The Li-GOF battery was sealed at argon atmosphere, which was performed in argon-filled glove box. Both sides of the cell were not punched. (b) Schematic illustration of the Li-GOF battery. (c) The potential and (d) current output of Li-GOF battery. (e) I-V characteristics of Li-GOF battery.



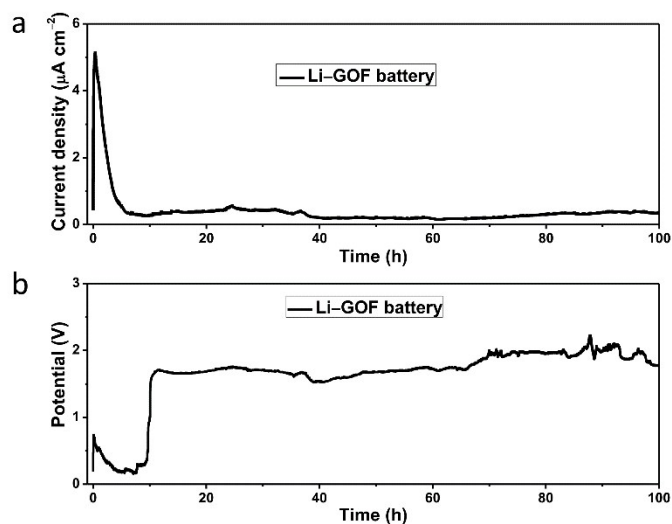
**Fig. S7.** (a) The survey XPS spectrum, (b) Li 1s and (c) C 1s of GOF. (d) The component distribution of GOF.

After a series of respiratory tests, the Li-GOF battery was decomposed and the surface component of GOF was investigated by XPS. The XPS results demonstrated that the resultant GOF

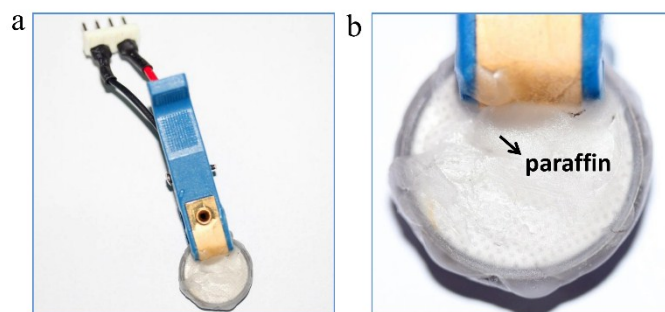
was composed of Li (~ 55 eV), C (~ 284.8 eV) and O (~ 532.0 eV) elements (Fig. S7a†). The Li 1s spectra could be deconvoluted into two peaks at ~ 54.9 eV and ~ 55.3 eV, which were assigned to LiOH and Li<sub>2</sub>CO<sub>3</sub>, respectively (Fig. S7b†). Among them, the formation of Li<sub>2</sub>CO<sub>3</sub> may be related to the interreaction of Li<sup>+</sup> and carboxyl (–COOH) groups within GOF. In contrast to the pristine GOF (Fig. S4b†), the newly appeared peak located at ~ 289.9 eV in C 1s spectra can be attributed to Li<sub>2</sub>CO<sub>3</sub> (Fig. S7c†), which was in accordance with Li 1s spectra (Fig. S7b†).



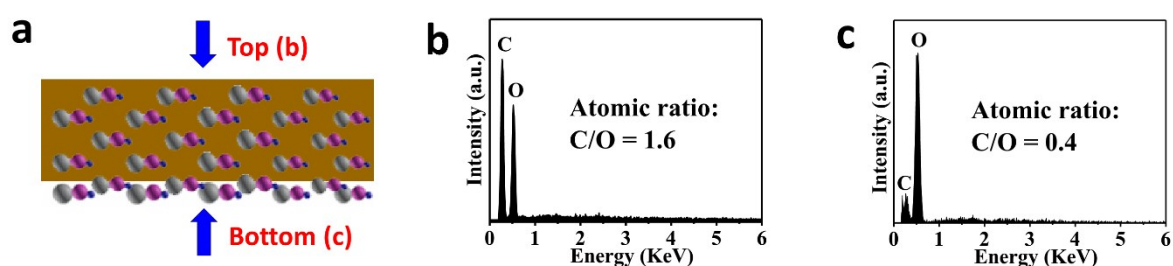
**Fig. S8.** (a) The current and (b) potential output of Li–GOF battery under moisture. The moisture was introduced into the RH controlling system to keep Li–GOF battery in a high humidity environment, which can be seen in Fig. S5b†.



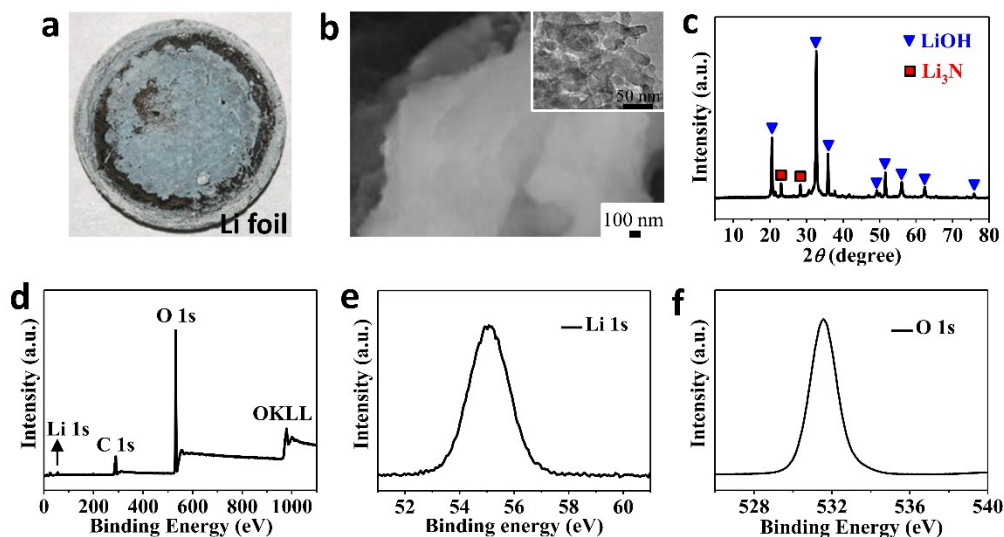
**Fig. S9.** (a) The current and (b) potential output of Li–GOF battery for 100 hours when exposed in air.



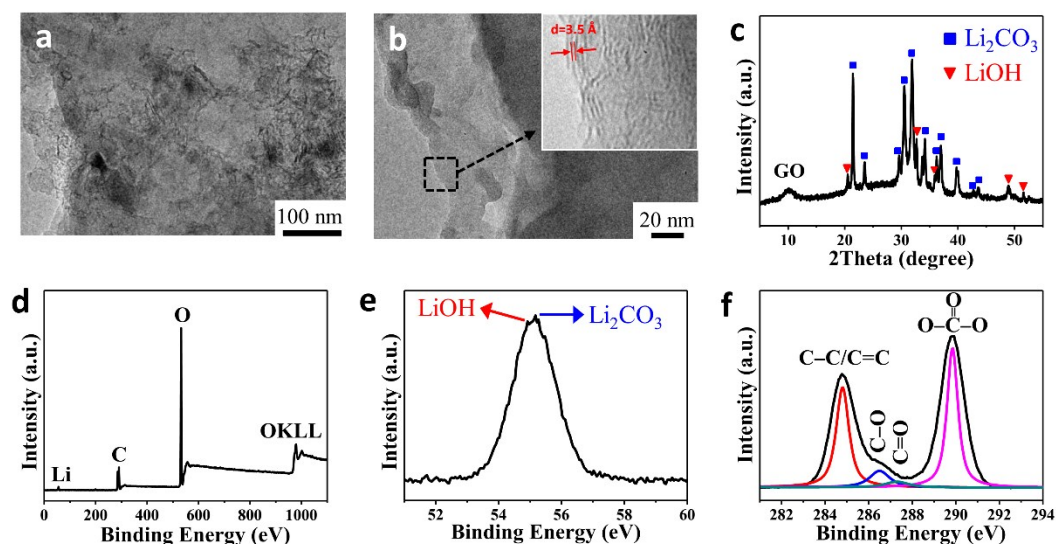
**Fig. S10.** (a,b) The photographs of Li-GOF battery, in which its bottom side is sealed by paraffin to avoid short-circuit induced by water.



**Fig. S11.** (a) Schematic illustration of fully covered GOF by LiOH. (b) The top and (c) bottom EDS of GOF.

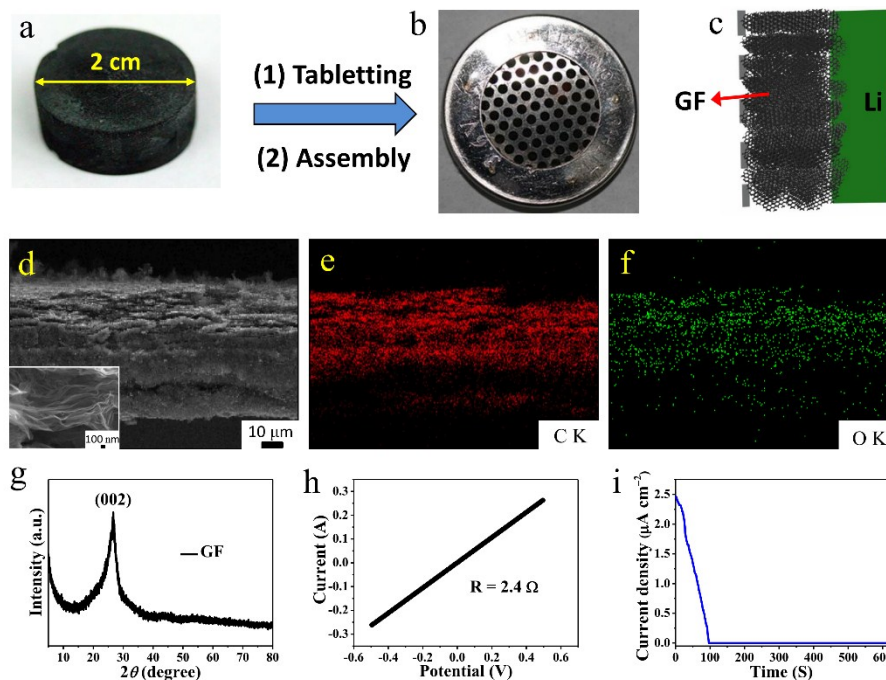


**Fig. S12.** After kept under moisture for about 10 h, the Li-GOF battery was decomposed. (a) The photograph of the resultant Li foil, (b) SEM image of LiOH and the corresponding TEM image (inset). (c) XRD pattern, (d) the survey XPS spectrum, (e) Li 1s spectrum and (f) O 1s spectrum of LiOH.



**Fig. S13.** (a,b) The TEM images of GOF/LiOH-Li<sub>2</sub>CO<sub>3</sub>. The inset was the high-resolution TEM image of GO nanosheet edge originated from the rectangular mark. (c) The XRD and (d-f) XPS of GOF/LiOH-Li<sub>2</sub>CO<sub>3</sub>.

As shown in Fig. S13a†, the TEM image revealed that the flower-like Li-based products were supported onto the 2D GO nanosheets. Nevertheless, the Li-based product was not steady but easily shrank even disappeared under the powerful X-ray of TEM (Fig. S13b†). The high-resolution TEM can clearly present the lattice fringe of GO nanosheet with a interlayer spacing of *ca.* 3.5 Å. The powder X-ray diffraction (XRD) pattern can be readily indexed to LiOH (JCPDS card No. 32-0564) and Li<sub>2</sub>CO<sub>3</sub> (JCPDS card No. 87-0729) (Fig. S13c†). XPS analysis showed the predominant three peaks were attributed to Li 1s peak (~ 55 eV), C 1s peak (~ 284.8 eV) and O 1s peak (~ 532.0 eV) (Fig. S13d†). The Li 1s spectra could be deconvoluted into two peaks at ~ 54.9 eV and ~ 55.3 eV, which were assigned to LiOH and Li<sub>2</sub>CO<sub>3</sub>, respectively (Fig. S13e†).<sup>S5</sup> More importantly, the C 1s spectra offered valid evidence, in which the newly appeared peak located at ~ 289.9 eV can be attributed to Li<sub>2</sub>CO<sub>3</sub> (Fig. S13f†).<sup>S6</sup>



**Fig. S14.** (a,b) Photographs of graphene foam and Li–GF junction. (c) Schematic illustration of Li–GF junction. (d–f) The side view SEM images of GF and the corresponding EDS mapping of C and O. (g) The XRD pattern of GF. (h) The I–V curve and (i) the current output of Li–GF junction.

The black graphene foam (GF) with a diameter of ca. 2 cm revealed that graphene oxide was reduced to graphene after 1000 °C annealing under the protection of argon gas (Fig. S14a†). The Li–GF junction was prepared by the direct contact of Li foil and GF (Fig. S14b,c†). The side-view SEM image of GF showed it possessed a thickness of ca. 50 μm, and consisted of lots of graphene nanosheets (Fig. S14d†). Typical EDS mapping analysis implied the uniform distribution of C and O atoms (Fig. S14e,f†). The XRD pattern of GF revealed that a sharp peak of ~ 26.7° with an interlayer spacing of ca. 0.33 nm could be indexed to carbon (002) diffraction (Fig. S14g†).

### Captions for Supporting Movies

**Movie S1.** The Li–GOF battery can reversibly control the on/off of a light emitting diode (LED) by adsorbing/desorbing moisture, respectively.

**Movie S2.** Once submerged in water, the Li–GOF battery can quickly light up a red LED.

**Movie S3.** When the Li–GOF battery was submerged in water, substantial H<sub>2</sub> bubbles released from the battery.



## References

- S1 D. A. Dikin, S. Stankovich, E. J. Zimney, R. D. Piner, G. H. B. Dommett, G. Evmenenko, S. T. Nguyen, R. S. Ruoff, *Nature*, 2007, **448**, 457–460.
- S2 D. R. Dreyer, S. Park, C. W. Bielawski, R. S. Ruoff, *Chem. Soc. Rev.*, **2010**, 39, 228–240.
- S3 Z. H. Tang, S. L. Shen, J. Zhuang, X. Wang, *Angew. Chem. Int. Ed.*, 2010, **49**, 4603–4607.
- S4 Y. N. Meng, Y. Zhao, C. G. Hu, H. H. Cheng, Y. Hu, Z. P. Zhang, G. Q. Shi, L. T. Qu, *Adv. Mater.*, 2013, **25**, 2326–2331.
- S5 A. C. Kozen, A. J. Pearse, C. F. Lin, M. A. Schroeder, M. Noked, S. B. Lee, G. W. Rubloff, *J. Phys. Chem. C*, 2014, **118**, 27749–27753.
- S6 M. H. Ye, Z. L. Dong, C. G. Hu, H. H. Cheng, H. B. Shao, N. Chen, L. T. Qu, *Small*, 2014, **10**, 5035–5041.