

Supplementary Information

**Boosting moisture induced electricity generation from graphene oxide through engineering oxygen-based functional groups**

Renbo Zhu, Yanzhe Zhu, Fandi Chen, Robert Patterson, Yingze Zhou, Tao Wan\*, Long Hu\*, Tom Wu, Rakesh Joshi, Mengyao Li\*, Claudio Cazorla\*, Yuerui Lu, Zhaojun Han, Dewei Chu

R. Zhu, Y. Zhu, F. Chen, Y. Zhou, Dr. T. Wan, Dr. L. Hu, Prof. T. Wu, Dr. R. Joshi, Dr. M. Li, Prof. D. Chu

School of Materials Science and Engineering, University of New South Wales, Sydney, NSW 2052, Australia

E-mail: tao.wan@unsw.edu.au; long.hu@unsw.edu.au; mengyao.li1@unsw.edu.au

Dr. R. Patterson

Australian Centre for Advanced Photovoltaics, University of New South Wales, Sydney, NSW 2052, Australia

Dr. Z. Han

School of Chemical Engineering, University of New South Wales, Sydney, NSW 2052, Australia

School of Mechanical and Manufacturing Engineering, University of New South Wales, Sydney, NSW 2052, Australia

CSIRO Manufacturing, 36 Bradfield Road, Lindfield, NSW 2070, Australia

Dr. C. Cazorla

Departament de Física, Universitat Politècnica de Catalunya, Campus Nord B4-B5, E-08034 Barcelona, Spain

E-mail: claudio.cazorla@upc.edu

Dr. Y. Lu

College of Engineering and Computer Science, Australian National University, Canberra, ACT 2601, Australia

Supplementary Figures

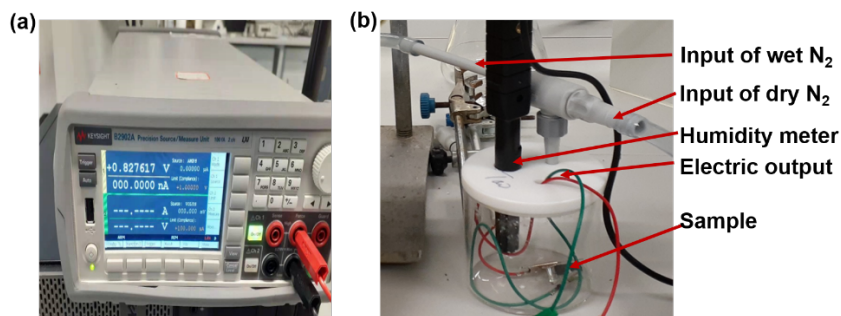


Fig. S1. Digital photo of experimental setup. (a) Keysight system for electric output measurement. (b) Sample chamber for moisture generation and electric connection.

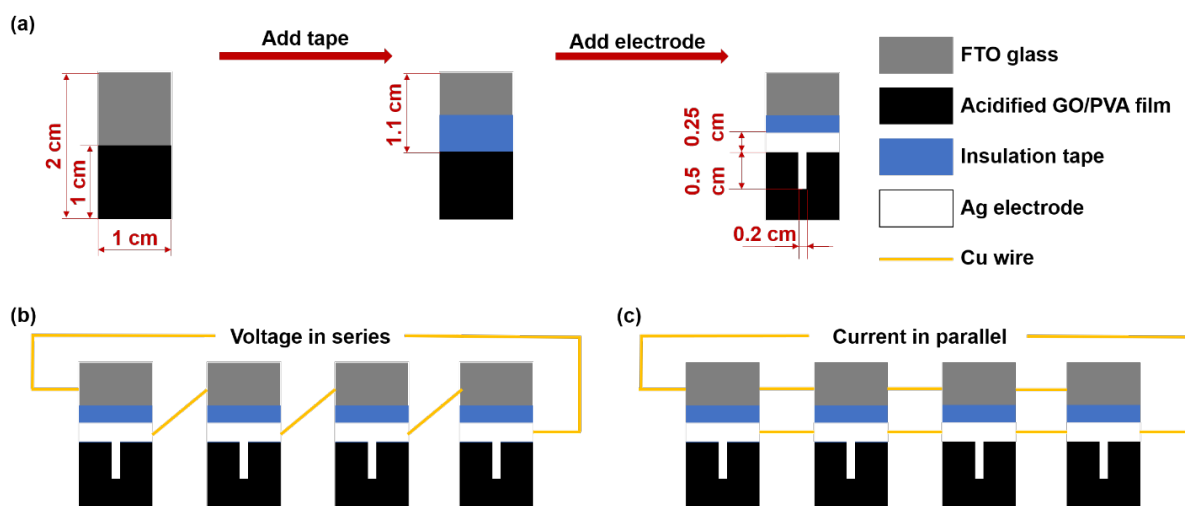


Fig. S2. Illustration of device fabrication. (a) Single MEG unit. (b) Four units in series for voltage measurement. (c) Four units in parallel for current measurement.

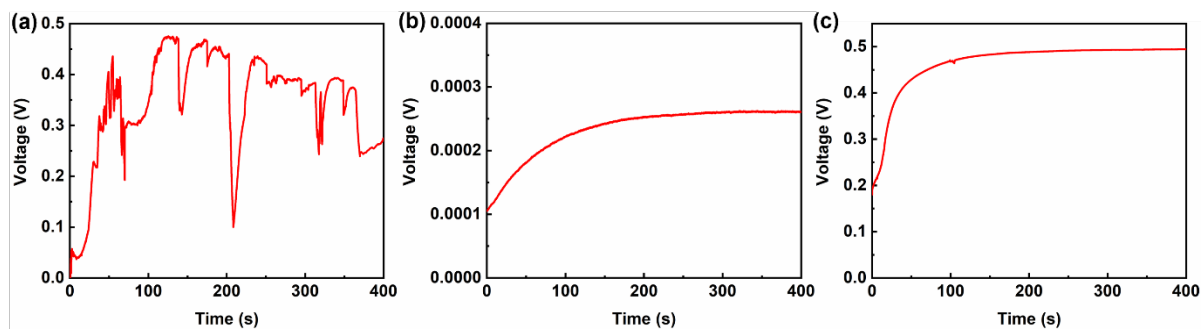


Fig. S3. Voltage output of MEGs fabricated with different functional materials at RH=75%. (a) GO. (b) PVA. (c) GO/PVA.

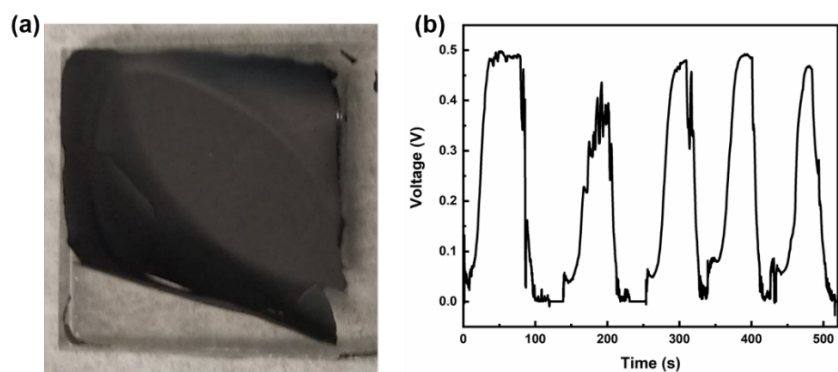


Fig. S4. MEGs fabricated without PVA.  $20 \text{ mg}\cdot\text{mL}^{-1}$  GO solution was dried on the FTO glass at  $50^\circ$  for 12 h. (a) Photo of GO films on the FTO glass. (b) Voltage cycles of GO films at  $\Delta\text{RH}=75\%$ .

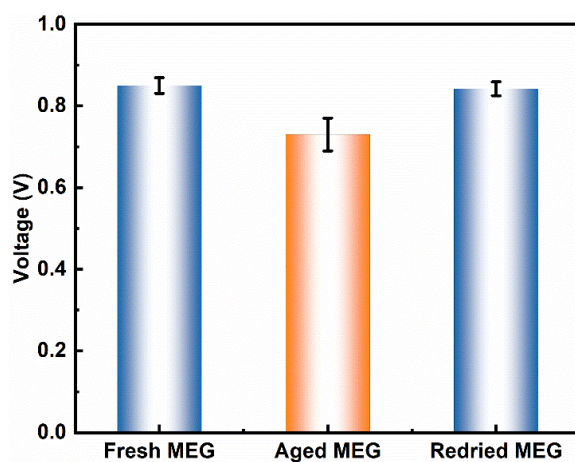


Fig. S5. The voltage comparison of fresh MEG, MEG placed for 1 week (aged MEG), MEG by drying aged MEG (redried MEG). The sample is placed in room humidity of 55% for 1 week and then gets dried at  $50^\circ\text{C}$  for 24 h. The voltages of all MEGs are tested at a relative humidity of 75%.

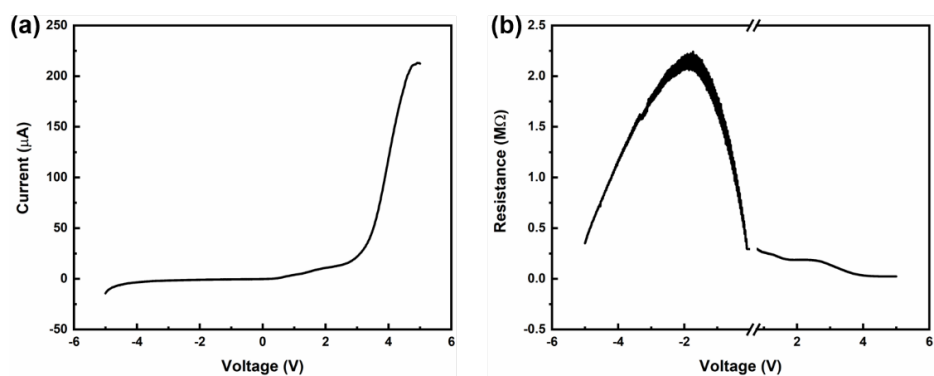


Fig. S6. MEG acidified with 32% HCl was conducted by sweep voltage. (a) Current output (b) resistance of acidified GO/PVA films in an external sweep voltage (RH=75%).

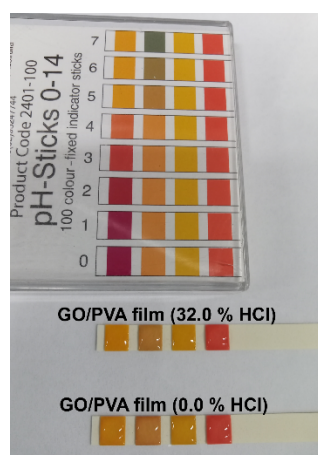


Fig. S7. The pH of water by pH indicator sticks after soaking GO/PVA film in distilled water for 10 mins. The residual water from GO/PVA film with and without acidification shows same pH and demonstrates that the mobilized protons originate from MEG instead of HCl acid.

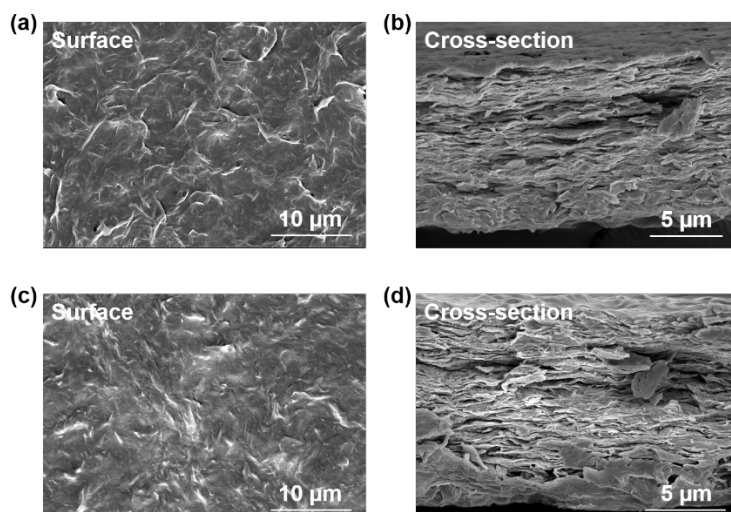


Fig. S8. Morphology of GO/PVA film. (a) Surface of pristine film. (b) Cross-section of pristine film. (c) Surface of film acidified with 32% HCl. (d) Cross-section of film acidified with 32% HCl.

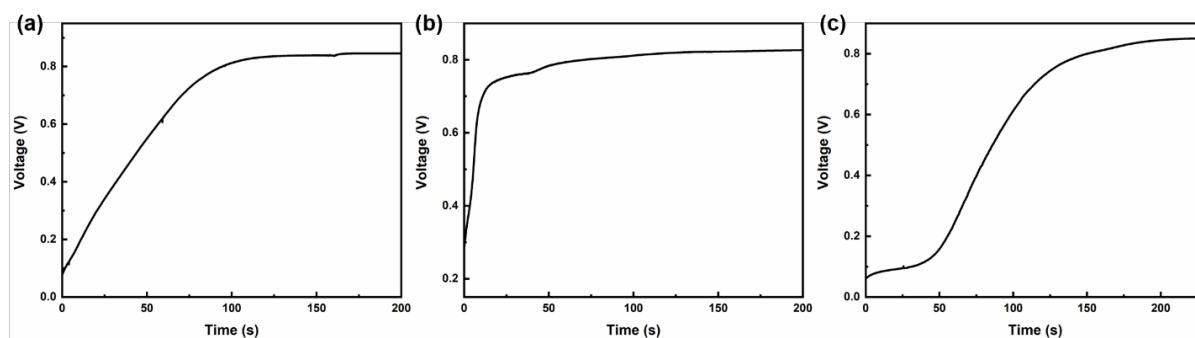


Fig. S9. Voltage output of acidified GO/PVA films with different areas at RH=75%. (a)  $0.5 \times 0.5 \text{ cm}^2$ . (b)  $1.0 \times 1.0 \text{ cm}^2$ . (c)  $1.5 \times 1.5 \text{ cm}^2$ .

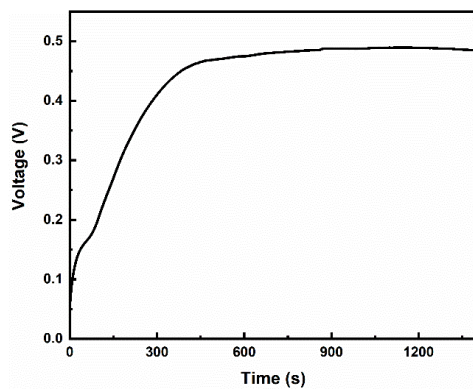


Fig. S10. Voltage output of acidified PVA at RH=75%. 20 mg·mL<sup>-1</sup> PVA solution was dried on the FTO glass at 50 ° for 12 h and acidified by 32.0% HCl to form acidified PVA films with a thickness of 16.41 μm.

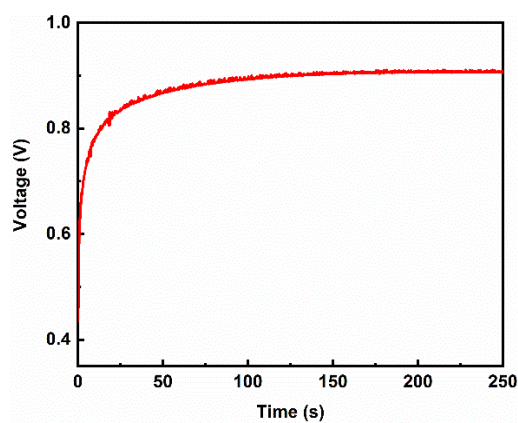


Fig. S11. Voltage output of GO/PVA film acidified by 32.0% HCl and tested with a RH of 85%.

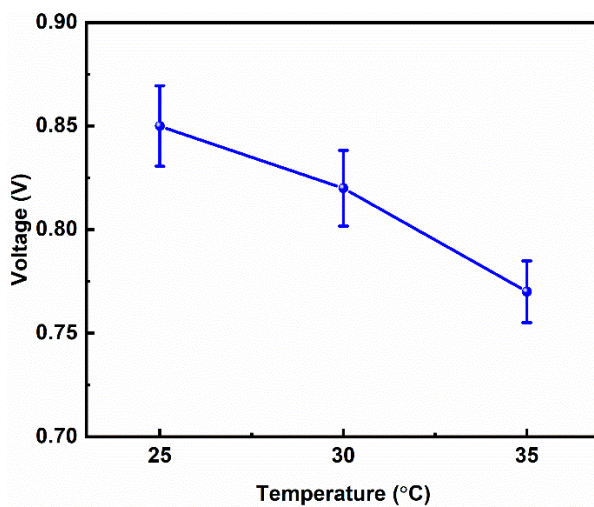


Fig. S12.  $V_{\max}$  of MEG acidified with 32.0% HCl and tested in different temperatures with a RH=75%.

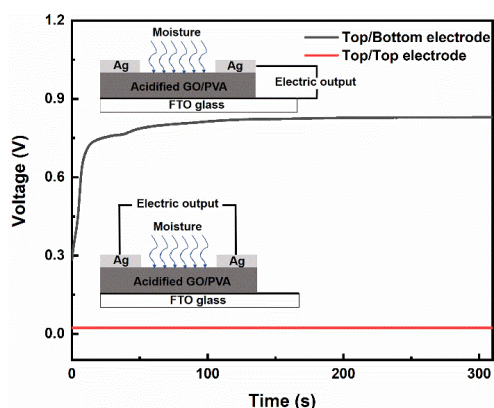


Fig. S13. Voltage output of MEGs with different electrodes at RH=75%. The MEG with top/bottom electrodes shows a high voltage of 0.85 V, and MEG with top/top electrodes shows no obvious voltage output.

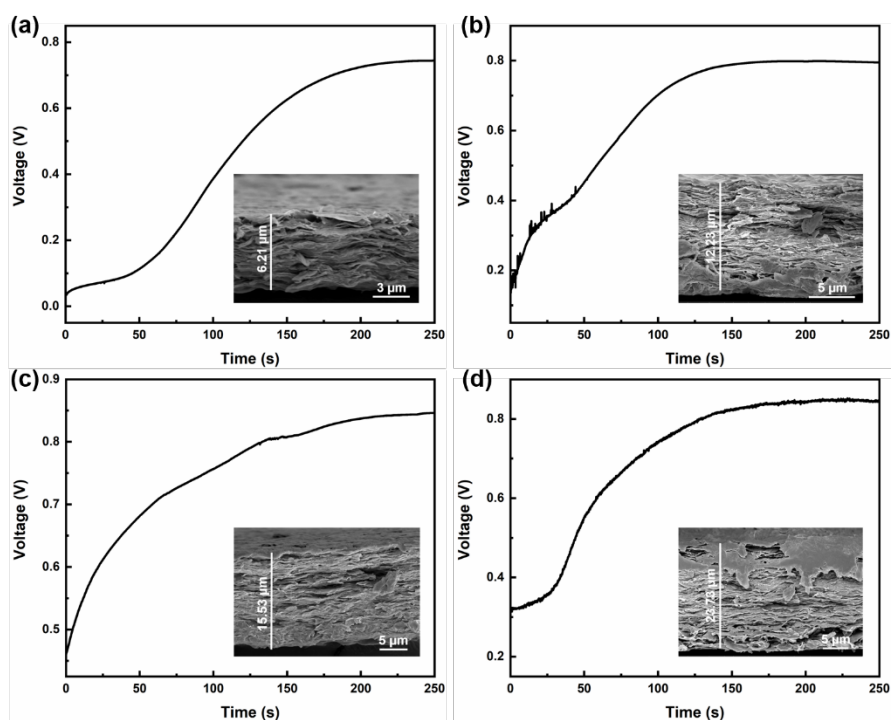


Fig. S14. Voltage output of GO/PVA films with different thickness after 32.0% HCl acidification. (a) 6.21  $\mu\text{m}$ . (b) 12.23  $\mu\text{m}$ . (c) 15.53  $\mu\text{m}$ . (d) 23.73  $\mu\text{m}$ .

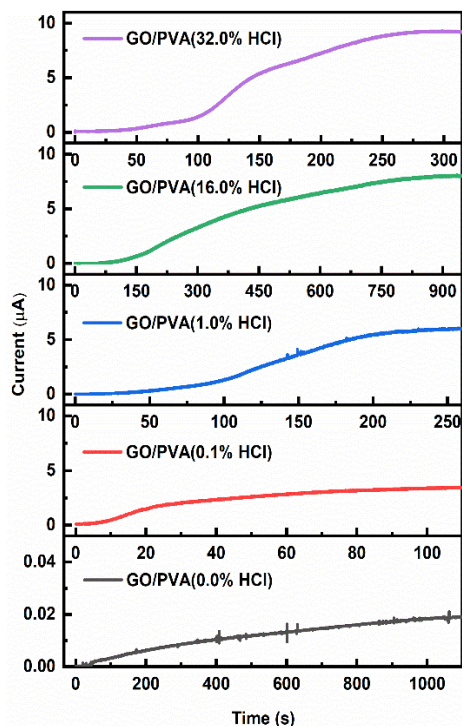


Fig. S15. Current output of GO/PVA films acidified with different HCl concentration. The MEGs are tested at RH=75% until the current output reaches highest value. The Ag electrode area of a single MEG is  $0.5 \times 0.2 \text{ cm}^2$ .

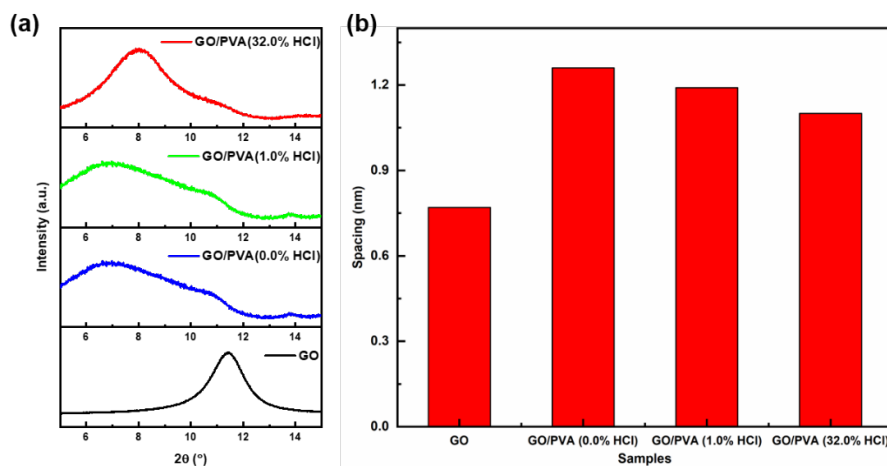


Fig. S16. XRD of GO/PVA films acidified by HCl with different concentration. (a) XRD pattern of GO films and GO/PVA films. (b) Interlayer spacing of GO films and GO/PVA films.

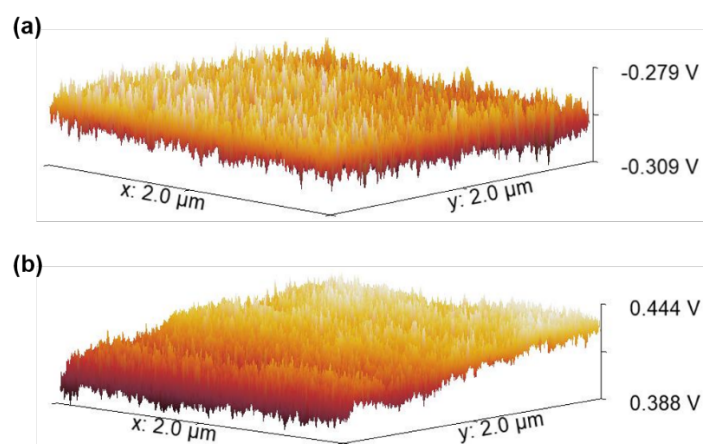


Fig. S17. Surface potential of film surface by KPMF at room humidity RH=55%. (a) Pristine GO/PVA film. (b) Acidified GO/PVA film. The acidified film exhibits a higher surface potential, indicating higher work function in the surface of acidified GO/PVA film.

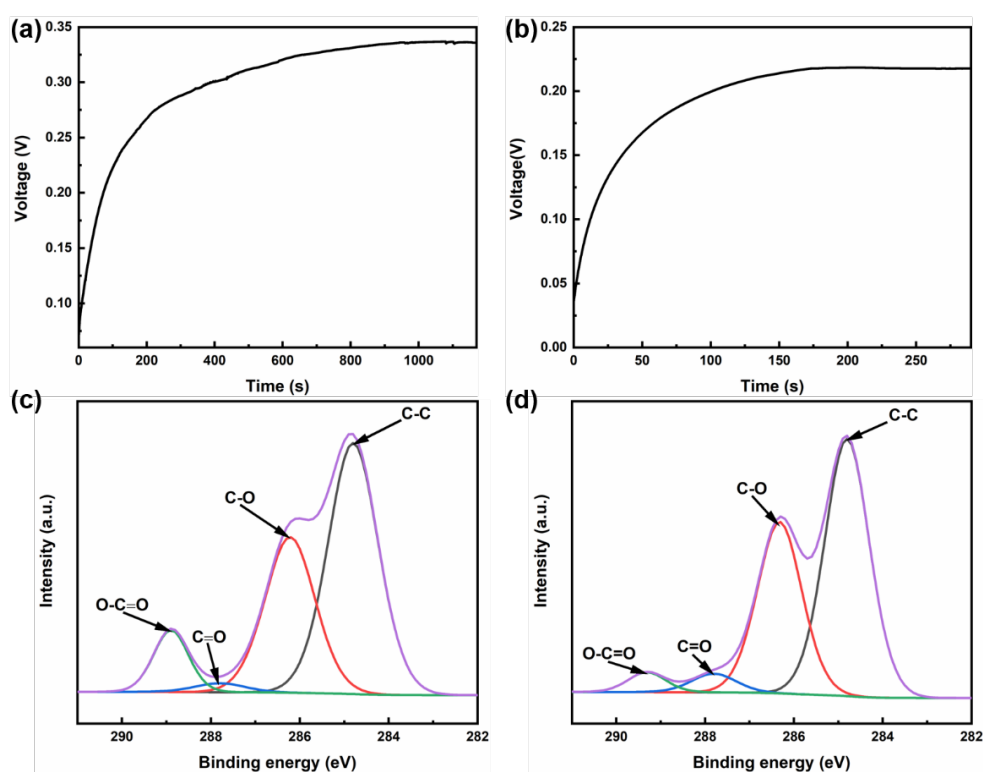


Fig. S18. GO/PVA films washed with different reagents. (a) Voltage of GO/PVA films washed with 80 wt.% acetic acid and (b) Voltage of GO/PVA films washed with 20 wt.% NaOH. (c) XPS spectra of GO/PVA films washed with 80 wt.% acetic acid and (d) XPS spectra of GO/

PVA films washed with 20 wt.% NaOH. GO/PVA films washed with acetic acid and NaOH exhibit lower voltage output and lower ratio of C=O bonds.

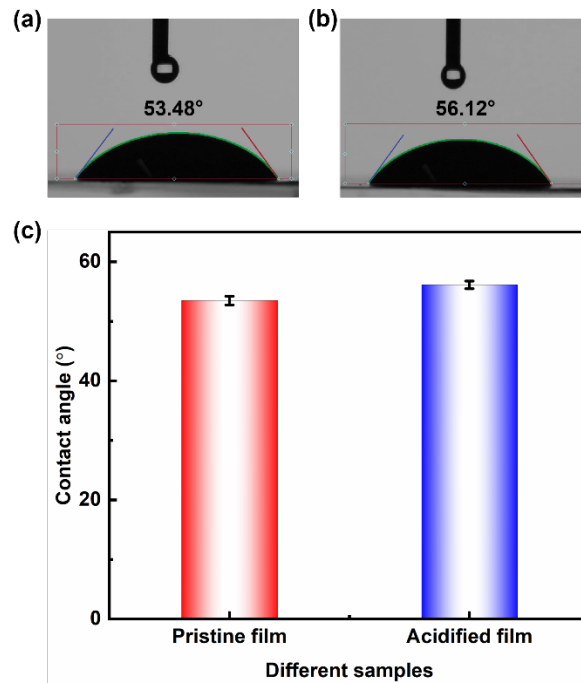


Fig. S19 The contact angle of GO/PVA with and without 32.0% HCl acidification. (a) Contact angle photo of films without acidification. (b) Contact angle photo of films with 32.0% HCl acidification. (c) Comparison of contact angle for the films with and without 32.0% HCl acidification.

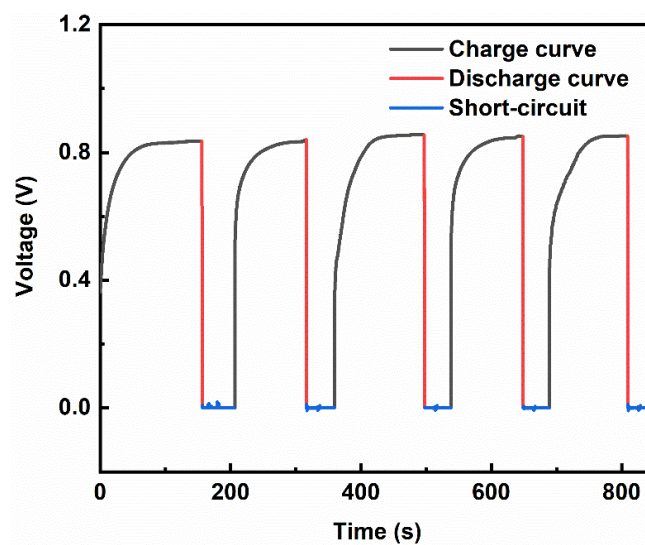


Fig. S20. Charge and discharge curves of MEG. The device was charged by controlling RH=75% and was discharged at  $20 \mu\text{A}\cdot\text{cm}^{-2}$ .

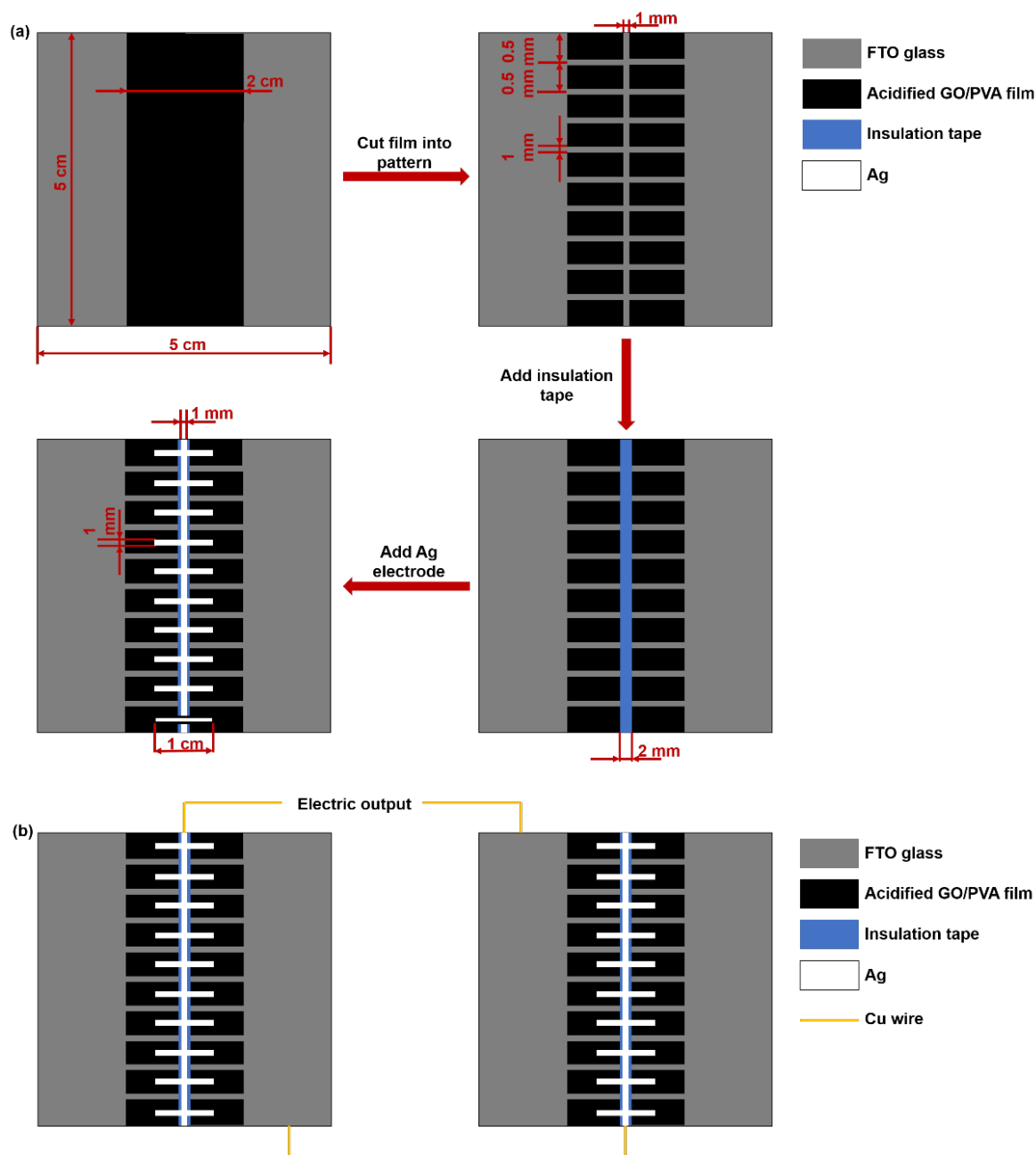


Fig. S21. Illustration of pattern fabrication. (a) Fabrication of GO/PVA pattern after acidification with 32 % HCl. (b) Illustration of two MEGs in series for powering practical device.