

## Supplementary Information

### **Electrochemical reduction of oxygen-functional-group-controlled graphene oxide for high carrier mobility**

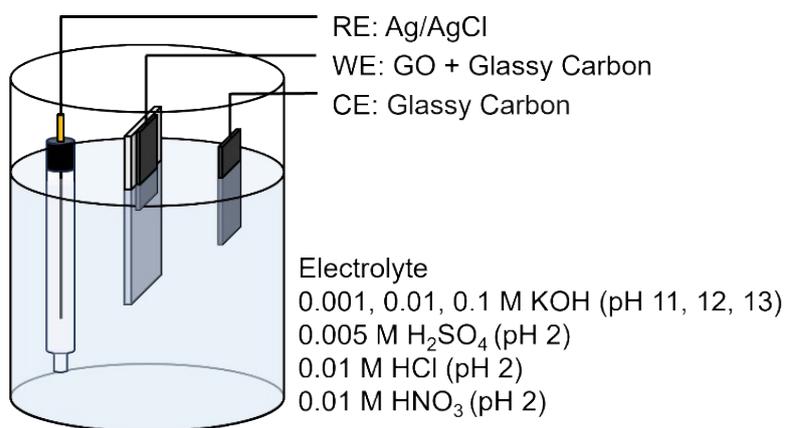
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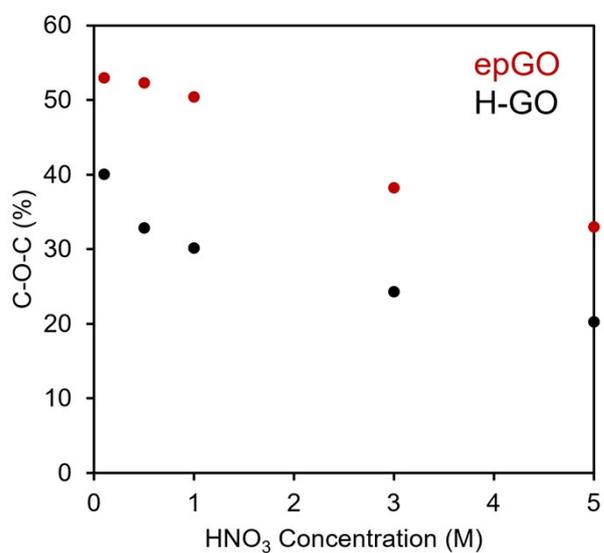
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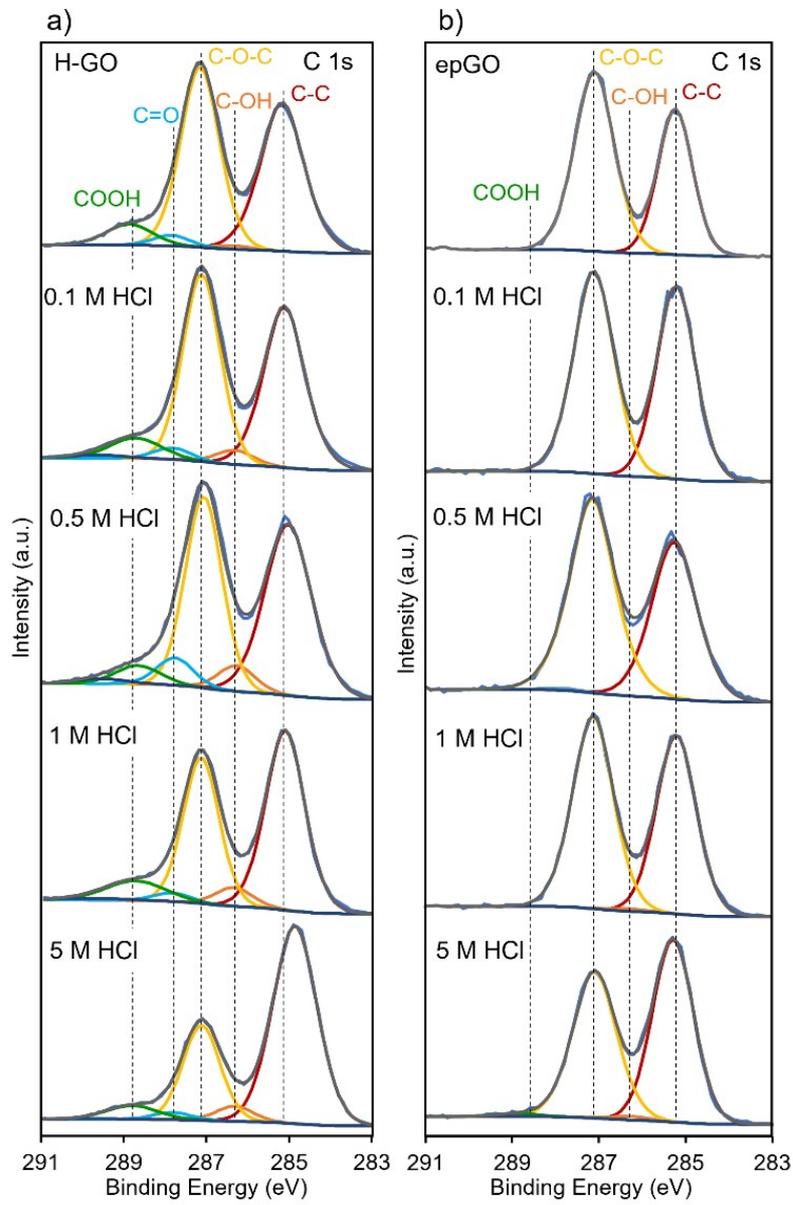
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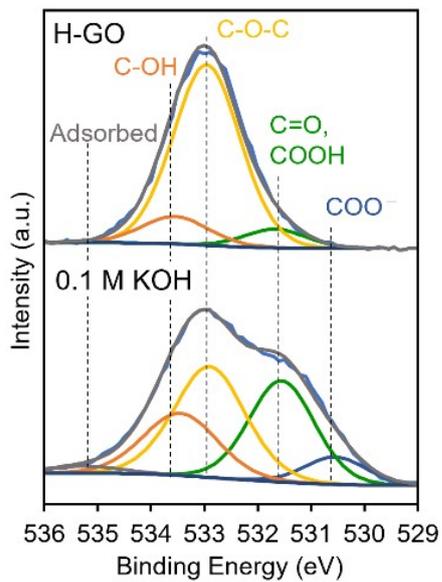
**Fig. S1** Schematic diagram of the electrochemical reduction setup.



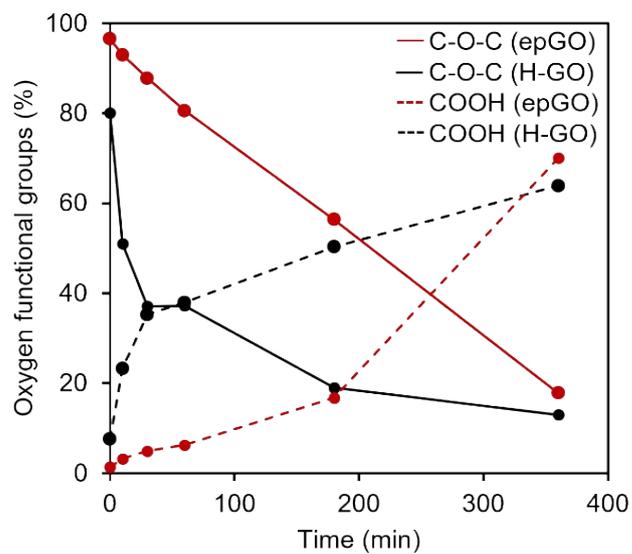
**Fig. S2** Relationship between the HNO<sub>3</sub> concentration used for 1-hour immersion and the amount of epoxy groups.



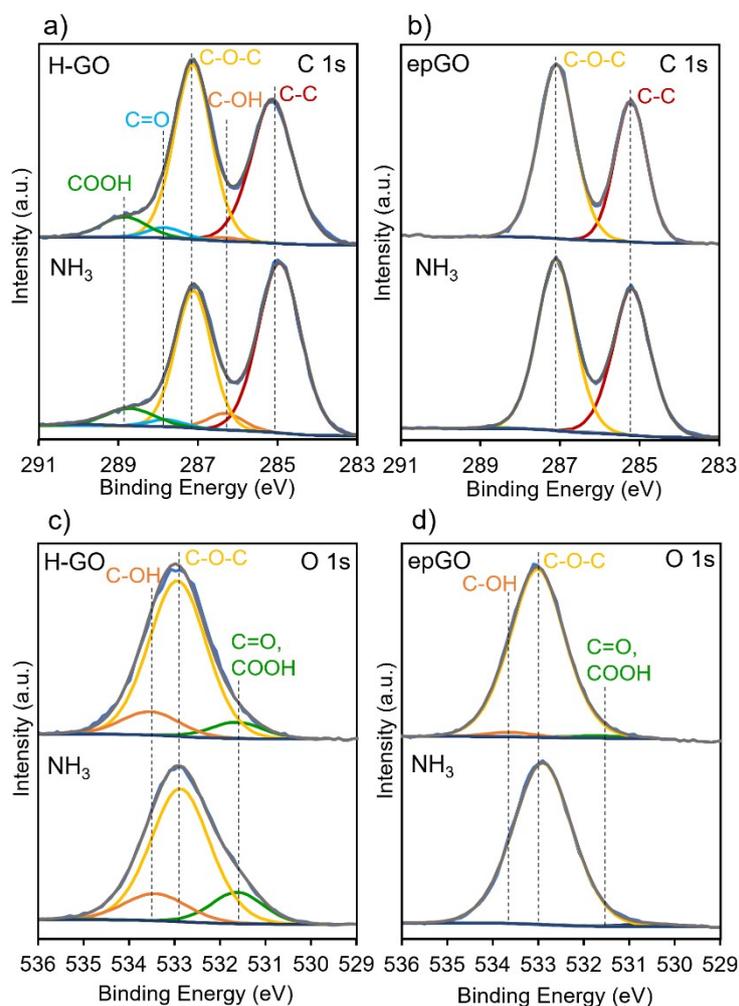
**Fig. S3** C 1s XPS spectrum of (a) H-GO and (b) epGO films before treatment and after immersion in 0.1, 0.5, 1, 5 M HCl solution for 1 h, respectively.



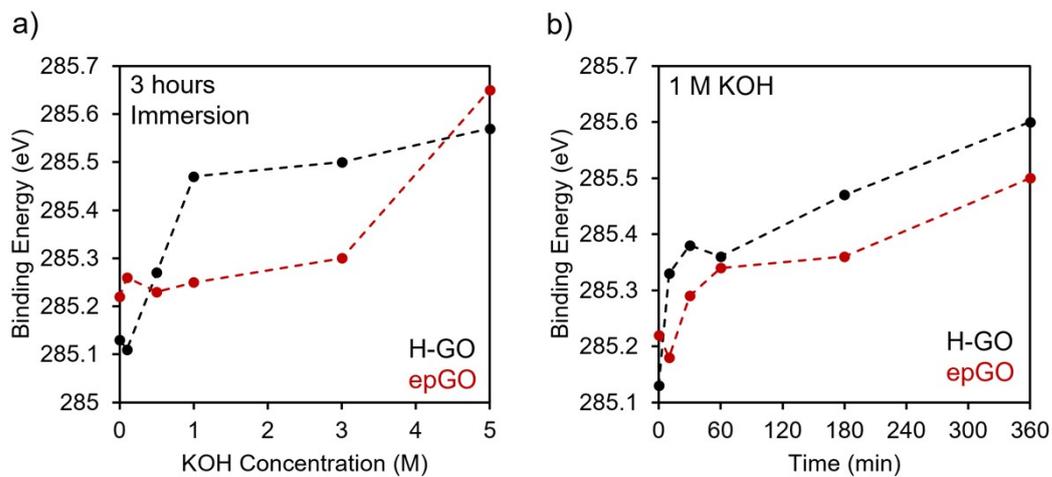
**Fig. S4** O1s XPS spectra of H-GO film before treatment and after immersion in 0.1 M KOH solution for 3h.



**Fig. S5** Correlation between C–O–C and COOH groups after immersing HGO and epGO films in 1 M KOH solution for various durations.

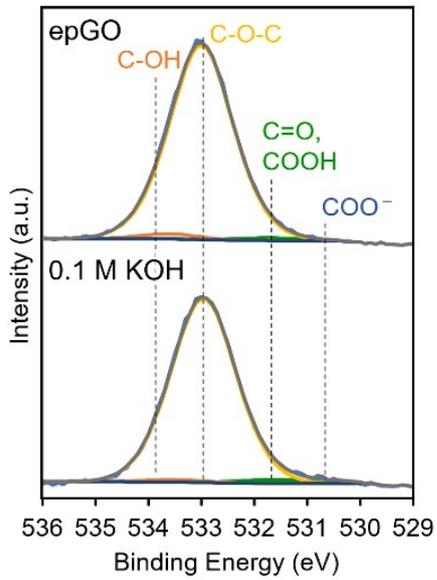


**Fig. S6** C 1s XPS spectrum of (a) H-GO and (b) epGO films, O 1s XPS spectrum of (c) H-GO and (d) epGO films before treatment and after immersion in 1 M NH<sub>3</sub> aqueous solution for 1 h.

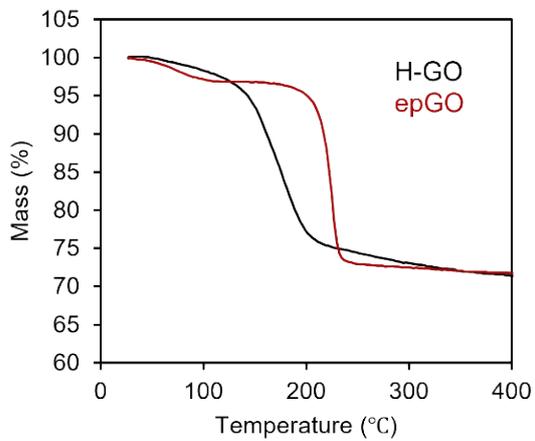


**Fig. S7** Correlation plot of bond energy of peaks originating from C-C bonds versus (a) KOH

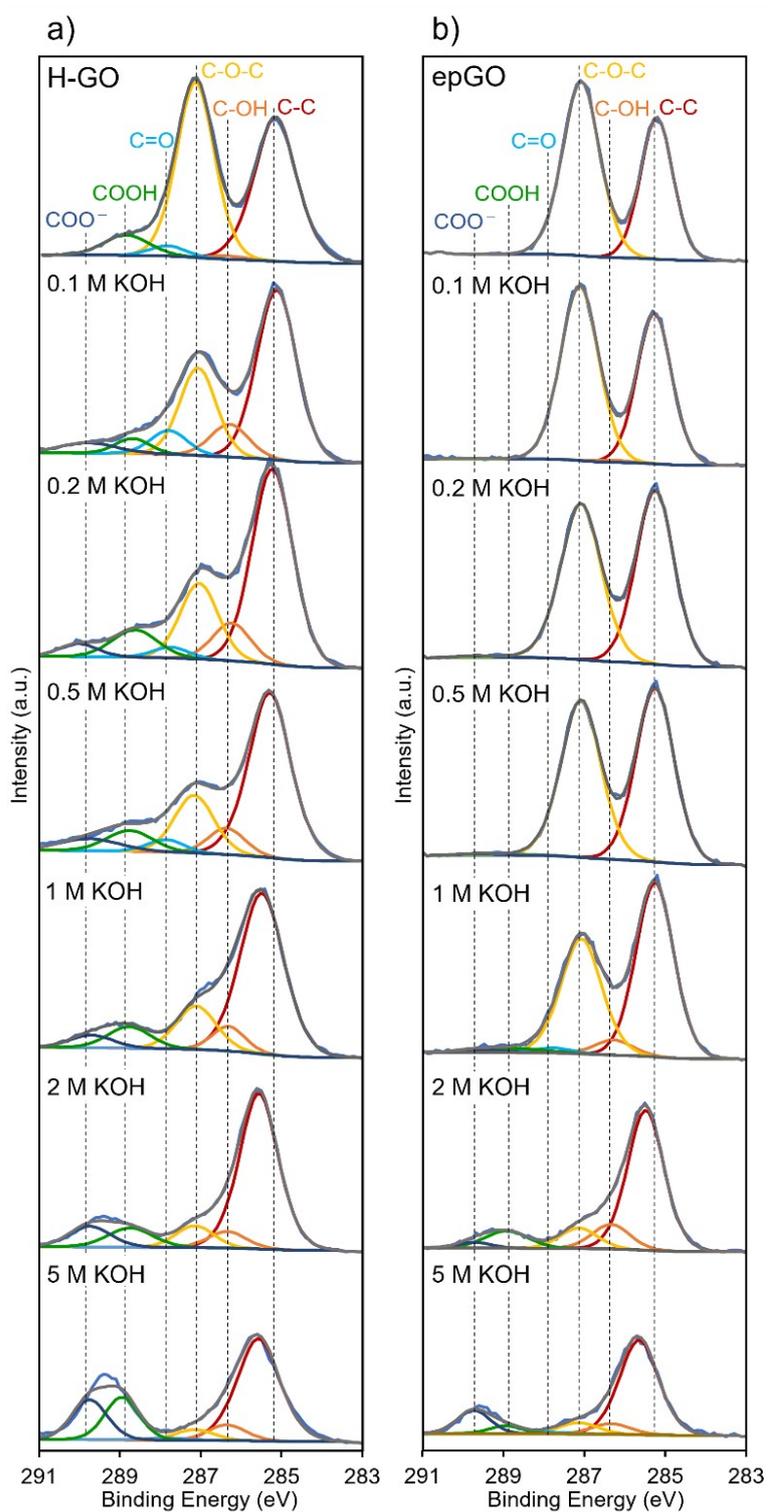
concentration and (b) immersion time in 1M KOH solution.



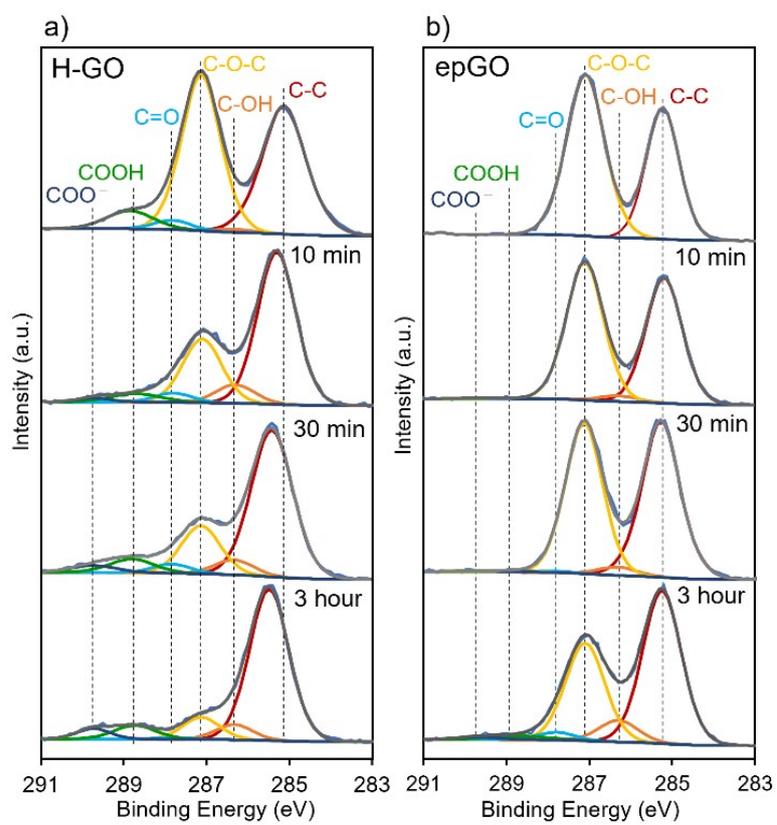
**Fig. S8** O 1s XPS spectra of epGO film before treatment and after immersion in 0.1 M KOH solution for 3h.



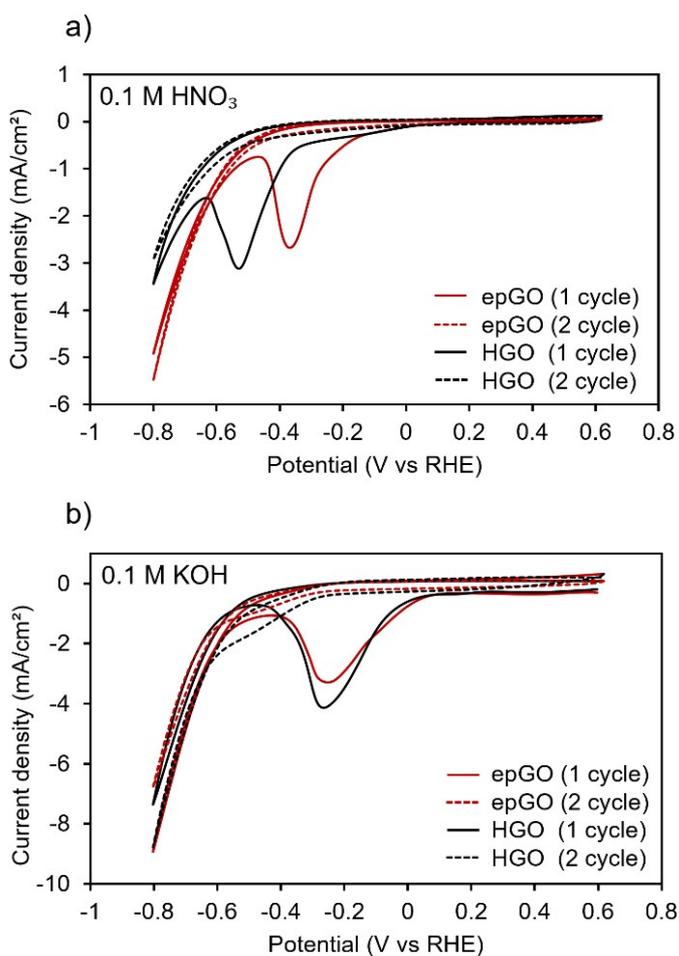
**Fig. S9** TG profiles obtained by thermogravimetric analysis conducted under vacuum condition.



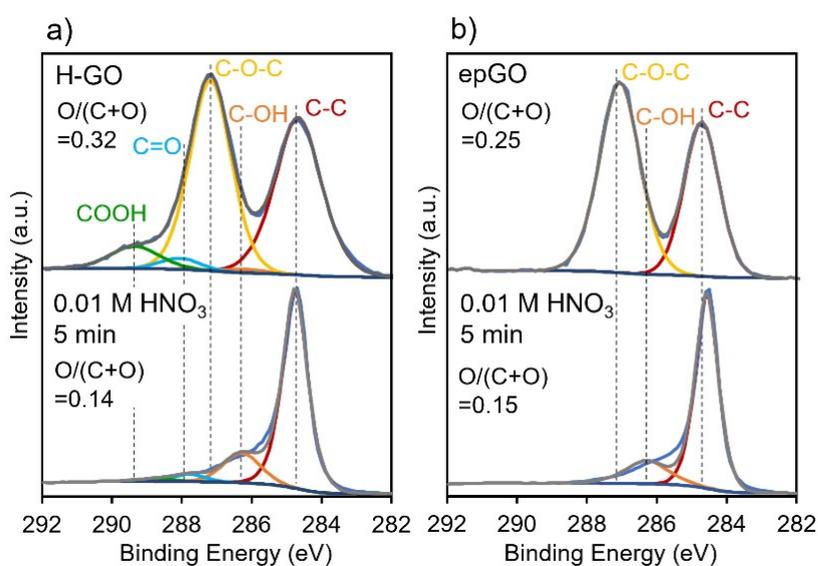
**Fig. S10** C 1s XPS spectrum of (a) H-GO and (b) epGO films before treatment and after immersion in 0.1, 0.2, 0.5, 1, 2, 5 M KOH for 3 h, respectively.



**Fig. S11** C 1s XPS spectra of (a) H-GO and (b) epGO films before treatment and after immersion in 1 M KOH solution for 10 min, 30 min and 3 h, respectively.

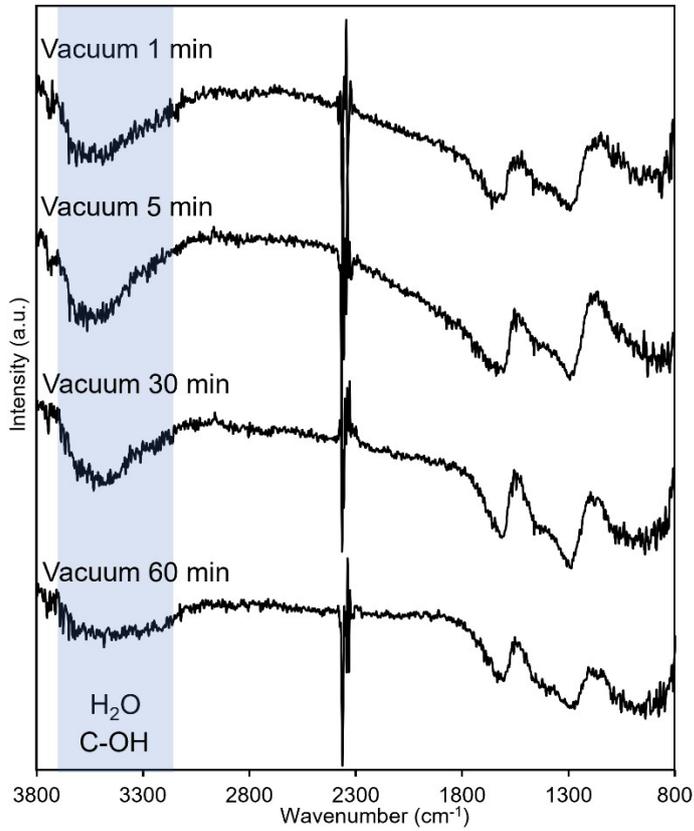


**Fig. S12** Cyclic voltammograms of H-GO and epGO films in (a) 0.1 M HNO<sub>3</sub> and (b) 0.1 M KOH aqueous solution.

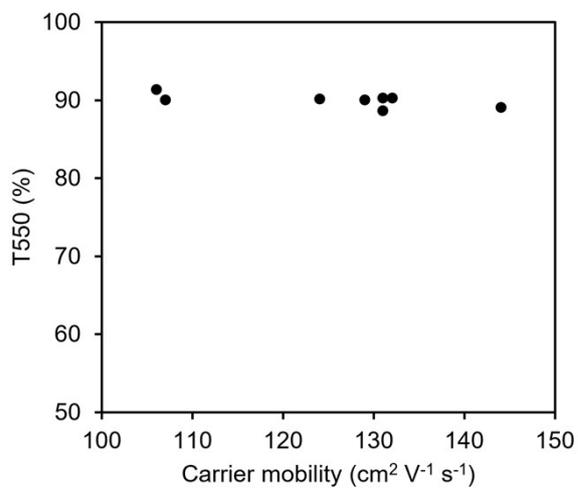


**Fig. S13** XPS C 1s spectra and O/(C+O) ratios of H-GO and epGO films before and after electrochemical

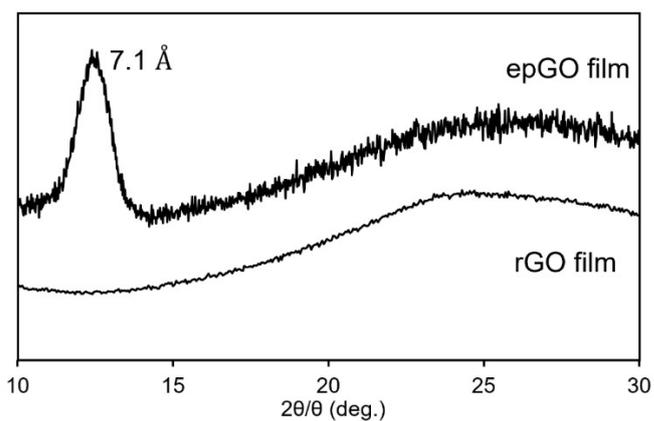
reduction in 0.01 M HNO<sub>3</sub> for 5 min.



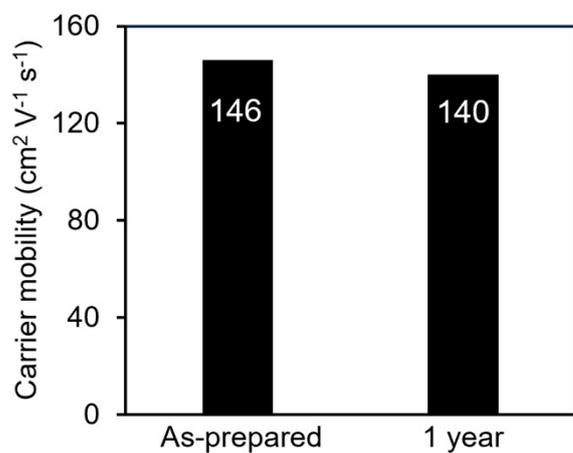
**Fig. S14** FT-IR spectra of the electrochemically reduced epGO film obtained after electrochemical reduction of epGO in 0.005 M H<sub>2</sub>SO<sub>4</sub> aqueous solution, measured under vacuum.



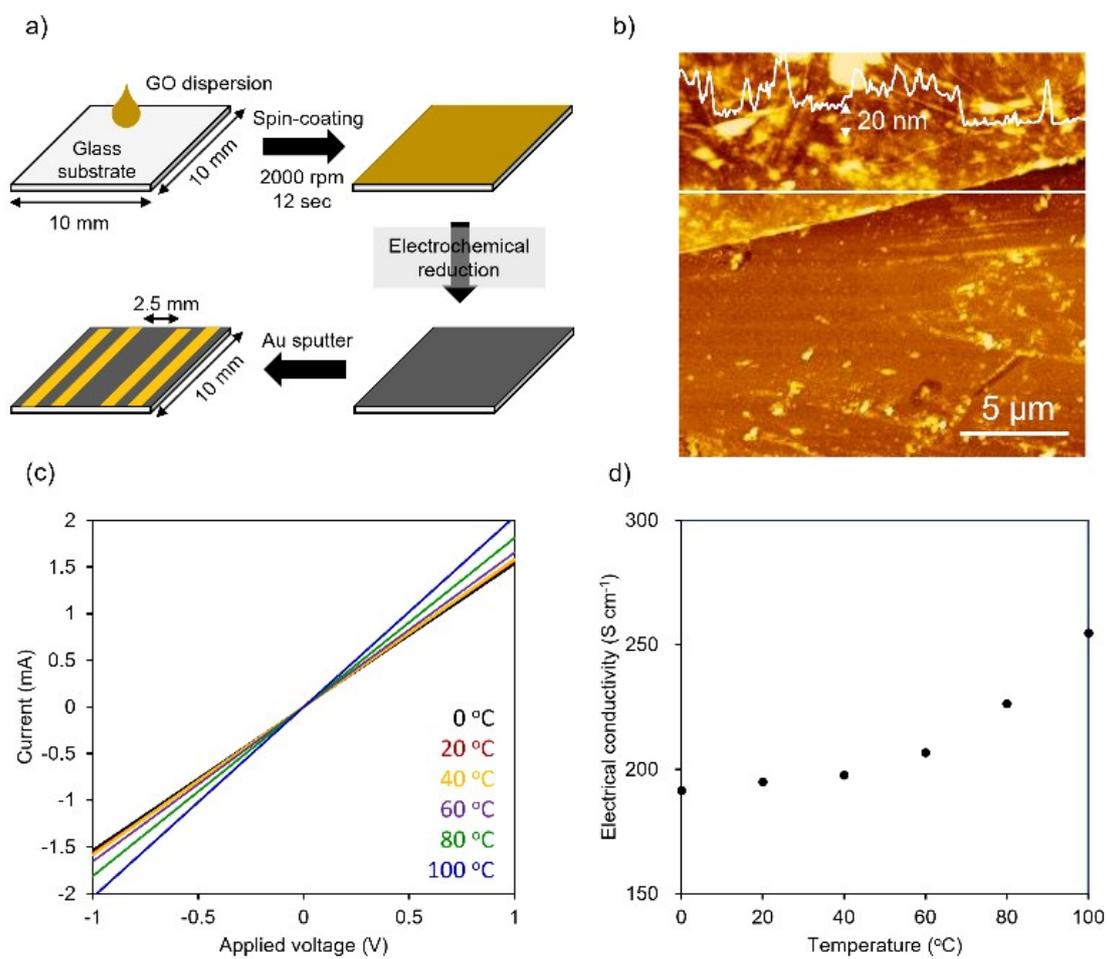
**Fig. S15** Optical transmittance at 550 nm (T<sub>550</sub>) of rGO films used for mobility measurement.



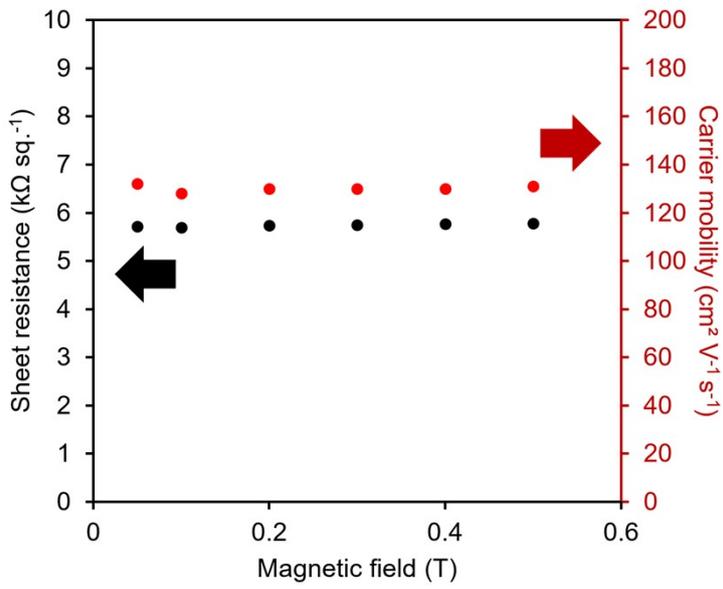
**Fig. S16** XRD profiles of epGO thin films and electrochemically reduced epGO thin films on glass substrates after electrochemical reduction.



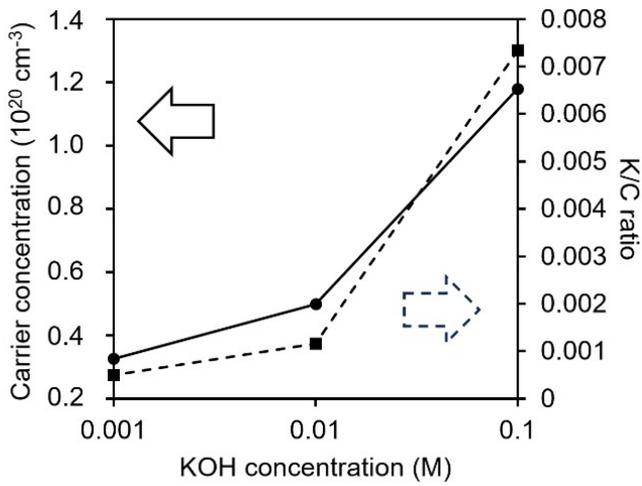
**Fig. S17** The carrier mobility of rGO thin films prepared by electrochemical reduction in 0.005 M  $\text{H}_2\text{SO}_4$  immediately after preparation and after 1 year.



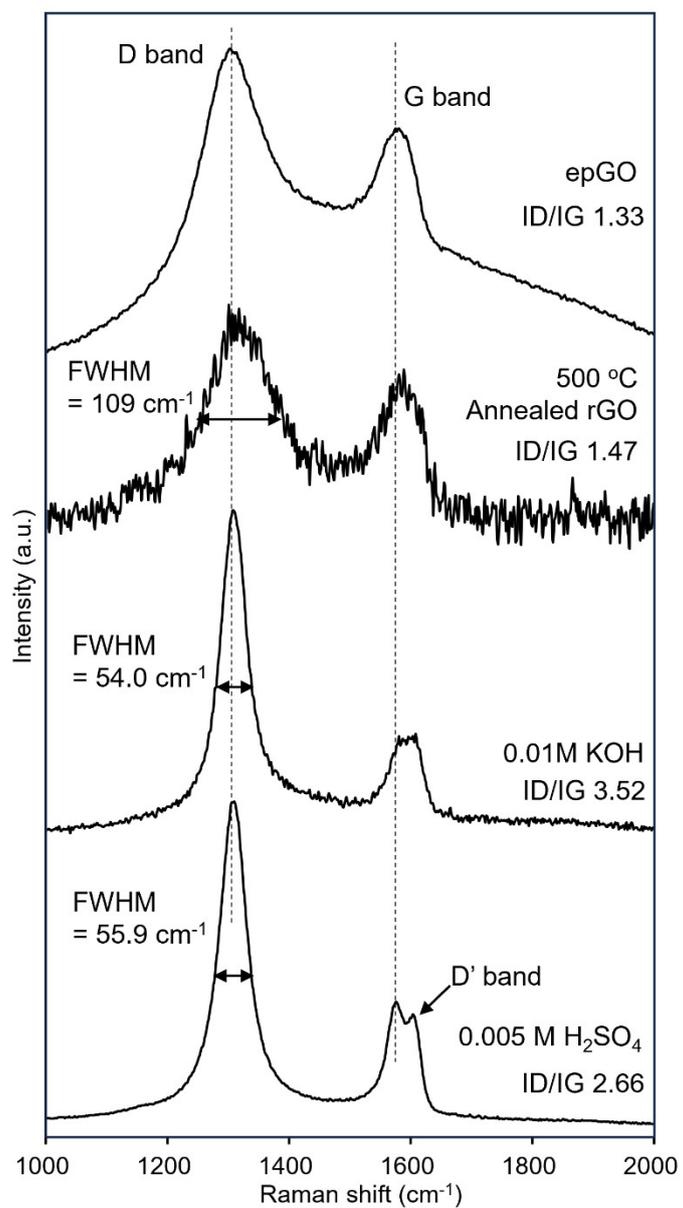
**Fig. S18** (a) Fabrication flow of electrochemical reduced GO thin films for electrical conductivity measurement. (b) AFM image of rGO thin film, temperature dependence of (c) I-V characteristics and (d) electrical conductivities.



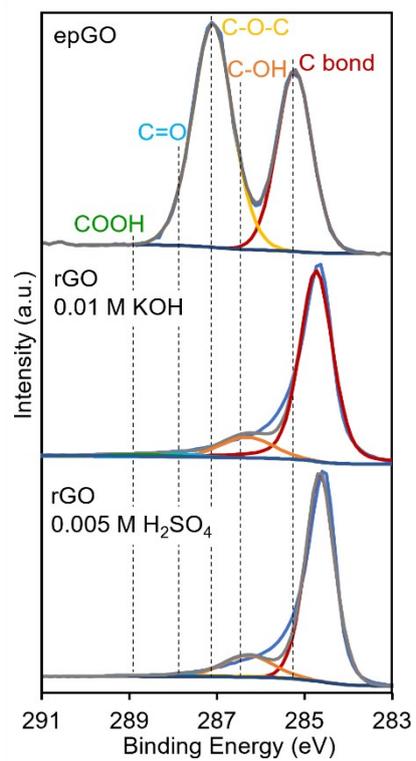
**Fig. S19** Correlation between sheet resistance and carrier mobility as a function of magnetic field.



**Fig. S20** Relationship diagram between KOH concentration and carrier concentration, and K/C ratio obtained by XPS analysis.



**Fig. S21** Raman spectra of epGO thin film, 500 °C annealed-rGO thin film and rGO thin films electrochemically reduced in 0.005 M H<sub>2</sub>SO<sub>4</sub> and 0.1 M KOH aqueous solution.



**Fig. S22** C1s XPS spectra of pristine epGO and epGO films electrochemically reduced in 0.01 M KOH aqueous solution and 0.005 M H<sub>2</sub>SO<sub>4</sub>

**Table S1.** Summary of C-C bonds peak positions and full width at half maximum (FWHM) in the electrochemical reduction of H-GO and epGO in 0.1 M HNO<sub>3</sub> and KOH aqueous solution.

| Samples   | Peak top (eV) | FWHM (eV) |
|---|---------------|-----------|
| H-GO  | 285.1         | 1.4       |
| Reduced H-GO (-0.44 V vs. RHE in 0.1 M HNO <sub>3</sub> ) | 284.7         | 0.9       |
| Reduced H-GO (-0.22 V vs. RHE in 0.1 M KOH)               | 285.0         | 1.0       |
| epGO  | 285.2         | 1.1       |
| Reduced epGO (-0.44 V vs. RHE in 0.1 M HNO <sub>3</sub> ) | 284.6         | 0.7       |
| Reduced epGO (-0.22 V vs. RHE in 0.1 M KOH)               | 284.6         | 0.9       |

**Table S2** The carrier mobility and transmittance at 550 nm (T550) values of rGO thin films.

| Sample                                 | Carrier mobility (cm <sup>2</sup> V <sup>-1</sup> s <sup>-1</sup> ) | T550 (%) |
|--|---|----------|
| 0.005 M H <sub>2</sub> SO <sub>4</sub> | 146   | 75.0     |
| 0.005 M H <sub>2</sub> SO <sub>4</sub> | 129   | 90.1     |
| 0.01 M HNO <sub>3</sub>                | 144   | 89.1     |
| 0.01 M HCl                             | 132   | 90.3     |
| 0.01 M KOH                             | 61.5  | 87.2     |