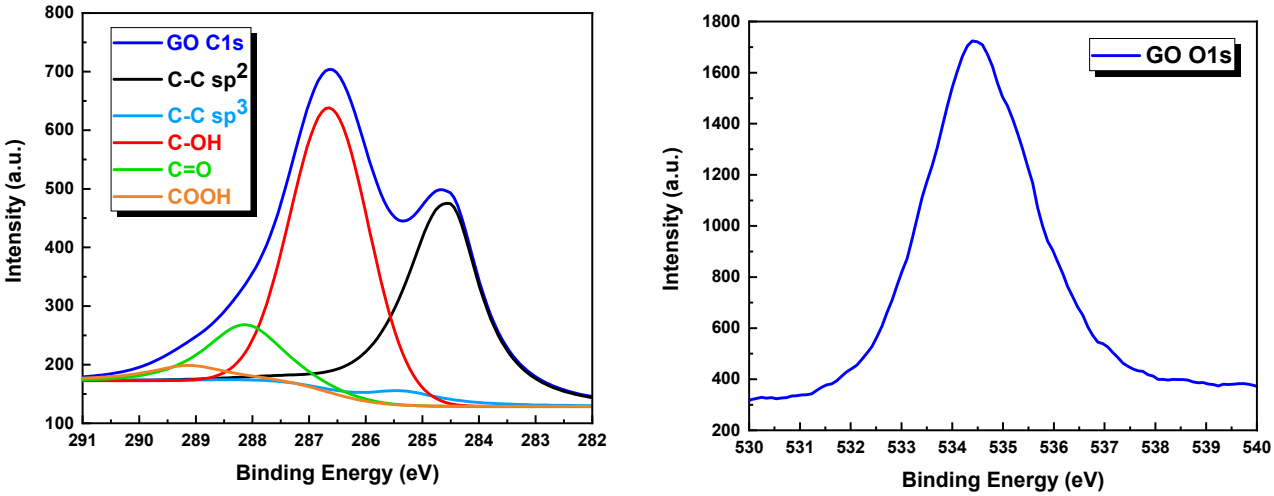
Supplementary Materials: Geometric Tuning for Enhanced Moisture-Driven Electricity Generation Enabled by Graphene-Oxide Flakes

Katerina Anagnostou [1,†], George Veisakis [1,†], Ioannis Kalogerakis [1], George Viskadouros [1], Konstantinos Rogdakis [1,2,*] and Emmanuel Kymakis [1,2,*]

[1] Department of Electrical & Computer Engineering, Hellenic Mediterranean University (HMU), 71410 Heraklion, Greece
[2] Institute of Emerging Technologies (i-EMERGE) of HMU Research Center, 71410 Heraklion, Greece
* Correspondence: krogdakis@hmu.gr (K.R.); kymakis@hmu.gr (E.K.)
† These authors contributed equally to this work.



(**a**) (**b**)

Figure S1. (**a**) Deconvoluted XPS C1s peak; and (**b**) XPS O1s peak of graphite oxide powder synthesised via improved Hummers' method.

From the overall Raman spectrum (Figure S2), it is evident that the as-prepared graphite oxide has a highly disordered structure due to the presence of many functional groups that are formed during the oxidation of graphite powder with the modified Hummers' method [43]. The Raman spectrum of the graphite oxide powder exhibits two prominent peaks at 1348 and 1594 cm^{-1} which correspond to the D and G bands, respectively [31,32,44]. The G peak originates from the primary in-plane vibrational mode of the sp^2 carbon atoms and corresponds to the first-order scattering of the E_{2g} phonon at the Brillouin zone centre [45–47]. The D peak derives from the breathing mode of A1g symmetry phonons around the K or K' points of the Brillouin zone and is activated only by defects within the graphene flakes, e.g., by the presence of oxygen functional groups and disruption of the sp^2-hybridised carbon system [47,48]. The intensity of the D peak to the intensity of the G peak ratio (I_D/I_G) is related to the number of defects present in a 2D material's structure [45]. The D band virtually has the same intensity as the G band which is indicative of structural disorder of the graphitic lattice [49]. Two lower intensity peaks appear at 2735 and 2929 cm^{-1} and are attributed to the 2D and D + D' peaks, respectively [49–52]. The 2D band originates from a two-phonon double resonance Raman process and is related to the number of layers within the 2D material. [45,49,53]. A 2D peak that appears at >2700 cm^{-1} is indicative of a multi-layer or bulk 2D sample [53]. The D + D' band requires defects for its activation [24].

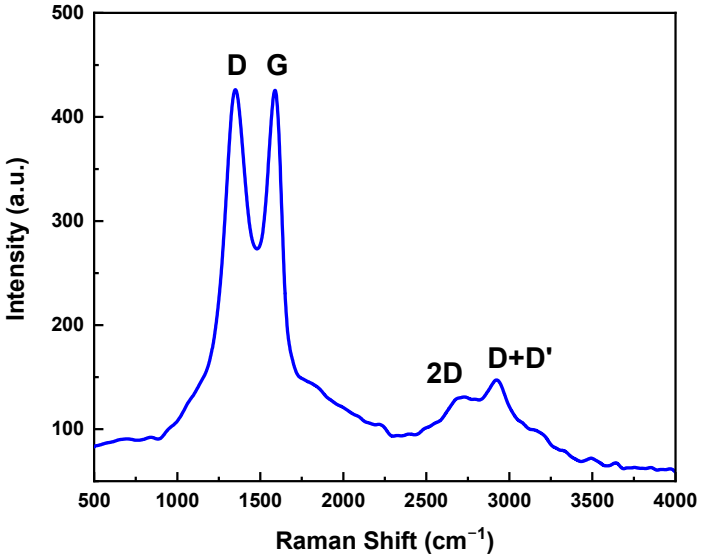
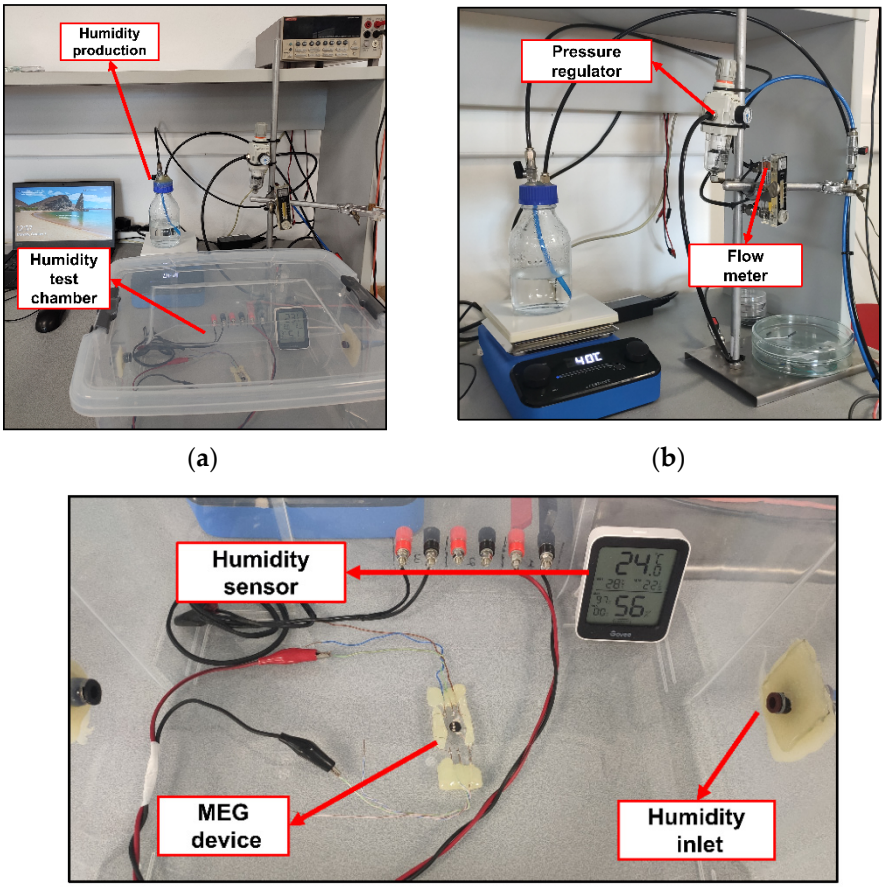


Figure S2. Raman spectrum of graphite oxide powder synthesised via improved Hummers' method.



(**a**) (**b**)

(**c**)

Figure S3. Digital photographs of custom experimental set-up built for moisture-induced voltage measurements **(a)** entire set-up which consists of the humidity test chamber, MEG device, humidity production and regulation components and data acquisition equipment; **(b)** humidity is produced by mildly heated water and regulated using a pressure regulator and a flow meter; **(c)** Close-up view of the interior of the test chamber.

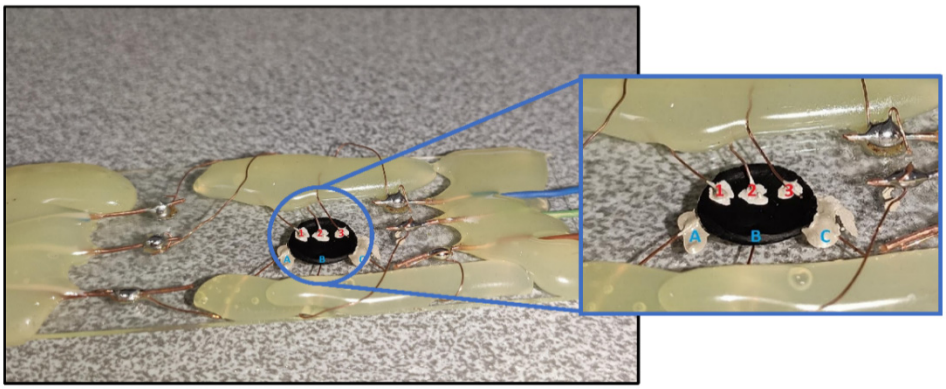


Figure S4. Digital photograph of fabricated GOP-1 device for evaluation of multiple contact configurations.

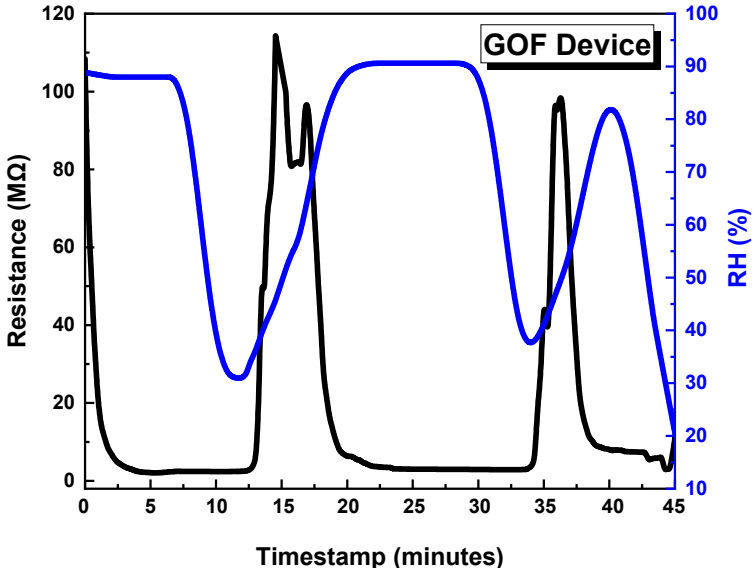


Figure S5. Resistance (MΩ) of GOF device as a function of RH (%).

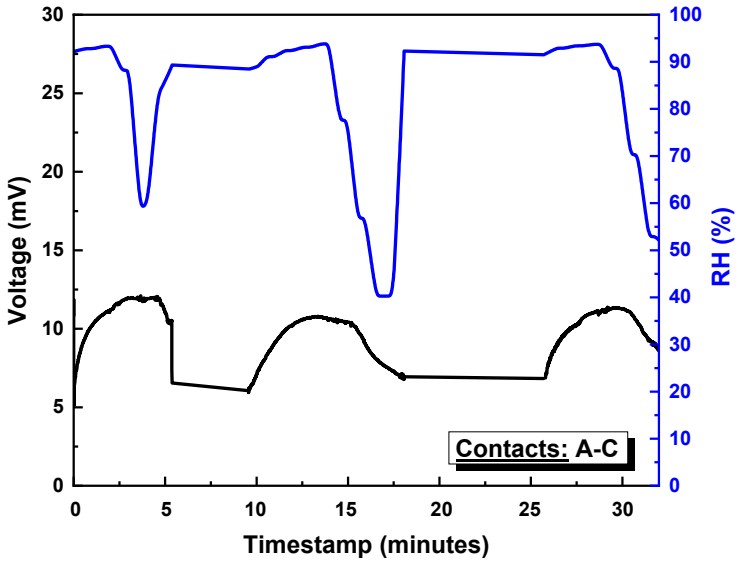
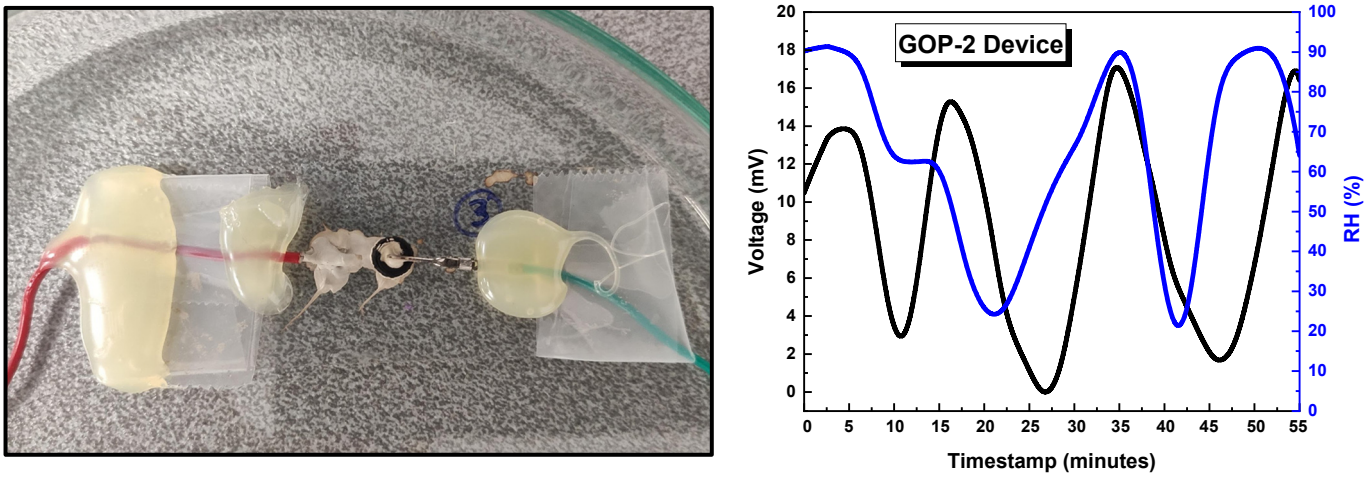


Figure S6. Voltage output as a function of RH when measuring contacts A-C of GOP-1 device. Since contacts A-C are practically concealed between the glass substrate and the GOP-1 sample, the bottom surface exposure to humidity is very limited, leading to suppressed V_o values.



(**a**) (**b**)

Figure S7. (**a**) Digital photograph of MEG device constructed using a GOP-2 as the moisture absorbing layer and large surface-area contacts in a vertical configuration (**b**) Moisture induced voltage (V_o) generated by the pristine GOP-2 device, i.e. without V_o enhancement by performing multiple RH cycles or by applying a DC bias in high humidity environments.

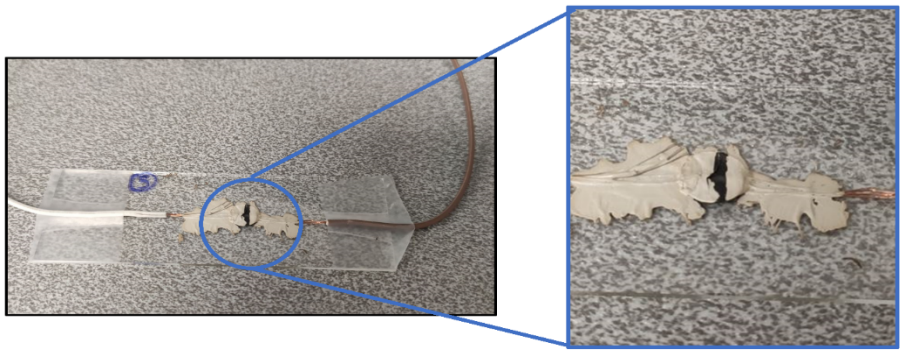


Figure S8. Digital photograph of MEG device constructed using GOF as the moisture absorbing layer and large surface area contacts in the horizontal configuration.