

Supplementary Material

1 MATERIALS SYNTHESIS

1.1 Graphene Oxide

Graphite underwent oxidation following a modified Hummer's method, as referenced in Mehl et al. (2014). In a 500 mL round-bottom glass flask, 46 mL of concentrated sulfuric acid - H_2SO_4 - (Anidrol, 98 %) was introduced, and the flask was placed in an ice bath. The acid was agitated at 1000 rpm, followed by the addition of 1 g of sodium nitrate NaNO_3 (Vetec, 99 %) and 2 g of graphite (Graflake 99580 from Nacional de Grafite, 99.84 %). The mixture was stirred for 10 minutes, after which 6 g of potassium permanganate KMnO_4 (Synth, 99 %) was gradually added, a few crystals at a time, over the first 30 minutes, with an approximate 30-second interval between each addition. Subsequently, potassium permanganate KMnO_4 was added in larger portions until the mixture showed green coloration. The addition frequency was increased until all potassium permanganate had been incorporated, taking approximately 1 hour in total. The round-bottom flask was then covered, and stirring continued for 75 minutes.

After this period, 92 mL of distilled room-temperature water were added in small portions, poured slowly along the glass flask's wall. The ice bath was removed. Ten minutes later, 280 mL of deionized water at 353 K was gradually introduced into the glass flask, and the mixture was stirred for 30 minutes. To halt the oxidation process, 10 mL of hydrogen peroxide H_2O_2 (Vetec, 30 %) was slowly added. The resulting mixture was transferred to a 1 L beaker, washed with a 1 mol L^{-1} aqueous hydrochloric acid HCl (Neon, 37 %) solution. Approximately 330 mL of the acid solution was added to the beaker, stirred for 15 minutes, and the supernatant was discarded. This acid washing procedure was repeated three times. The solid was then filtered using a Büchner funnel with a paper filter, washed with distilled water until the pH reached approximately 7. The material was removed from the paper filter and dried in an oven at 343 K for 24 hours.

For the exfoliation of graphite oxide and the preparation of graphene oxide dispersions, 25 mg of graphite oxide was combined in a 100 mL round-bottom glass flask with 50 mL of deionized water and subjected to 90 minutes of sonication in an ultrasound bath, which was periodically cooled with small ice portions. The resulting mixture was subsequently centrifuged at 3500 rpm for 90 minutes, and the resulting supernatant was carefully transferred to another container and centrifuged at 3500 rpm for an additional 90 minutes. The resulting dispersion of graphene oxide (GO) had a concentration of approximately $5.0 \times 10^{-2} \text{ mg mL}^{-1}$ and was further diluted to $2.1 \times 10^{-2} \text{ mg mL}^{-1}$ using deionized water.

1.2 Aqueous PEDOT ink

PEDOT was synthesized via the oxidative polymerization of EDOT using FeCl_3 as the oxidizing agent as referenced in das Neves et al. (2021). In summary, 50 mL of an iron(III) chloride solution in acetonitrile (with a concentration of 8.742 mol L^{-1}) was introduced into a 100 mL round-bottom glass flask and agitated at 1000 rpm. Subsequently, 200 μL of EDOT was added, resulting in the immediate formation of a dark blue solid. The mixture was stirred for a duration of 1 hour, after which it was transferred to 50 mL propylene tubes and centrifuged at 3500 rpm for 30 minutes. The supernatant was then carefully discarded, and the remaining solid was washed one time with 40 mL of acetonitrile and two times with 40 mL of deionized water, each washing was followed by centrifugation at 3500 rpm for 10 min and supernatants were discarded.

The solid sample obtained was designated as PEDOT. For solid characterizations, PEDOT was dried in an oven at 100°C for approximately 1 hour. An aqueous ink of PEDOT was prepared using the wet solid obtained immediately after the second washing process with water, as described earlier. Specifically, wet PEDOT (1.25 g) was placed in a 100 mL round-bottom glass flask along with 50 mL of deionized water (this ratio was consistently maintained). The resulting mixture underwent sonication for 1 hour in a bath sonicator, with periodic addition of small ice portions every 10 minutes to maintain the temperature. After the ultrasound treatment, the aqueous PEDOT ink was deemed ready for deposition and was identified as PEDOT-H₂O. This ink was stored in a glass vial.

2 EVALUATION OF RESISTIVITY AFTER TREATMENT

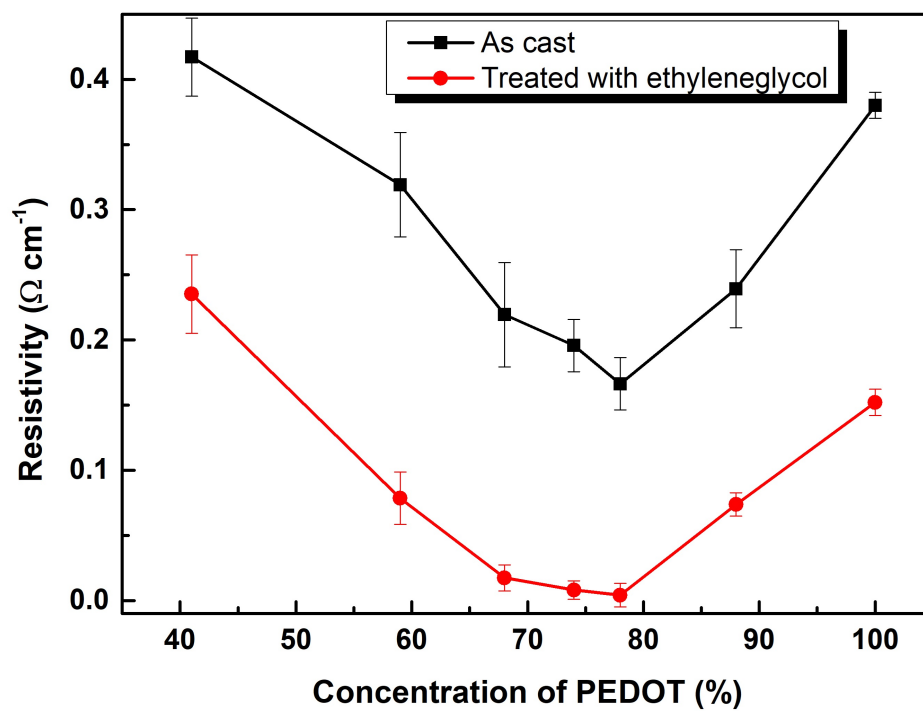


Figure S1. Resistivity analysis of GO:PEDOT:PSS samples after the treatment with ethyleneglycol, resulting in the same spot for the lowest value of resistivity.

3 DYNAMIC LIGHT SCATTERING ANALYSIS

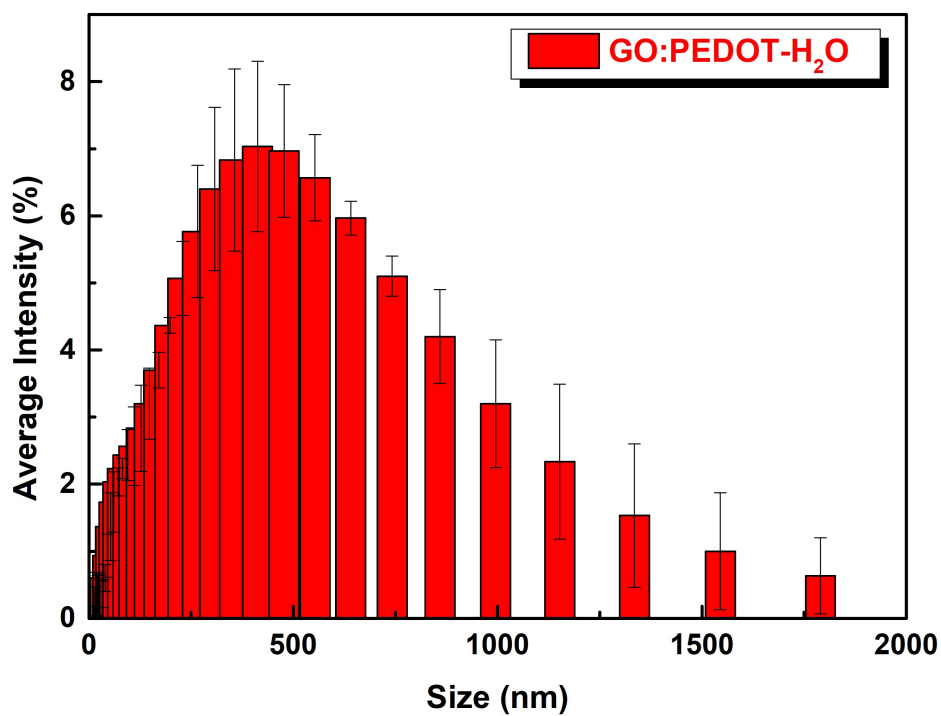


Figure S2. Dynamic Light Scattering analysis for GO:PEDOT samples.

4 SCANNING ELECTRON MICROSCOPY IMAGES

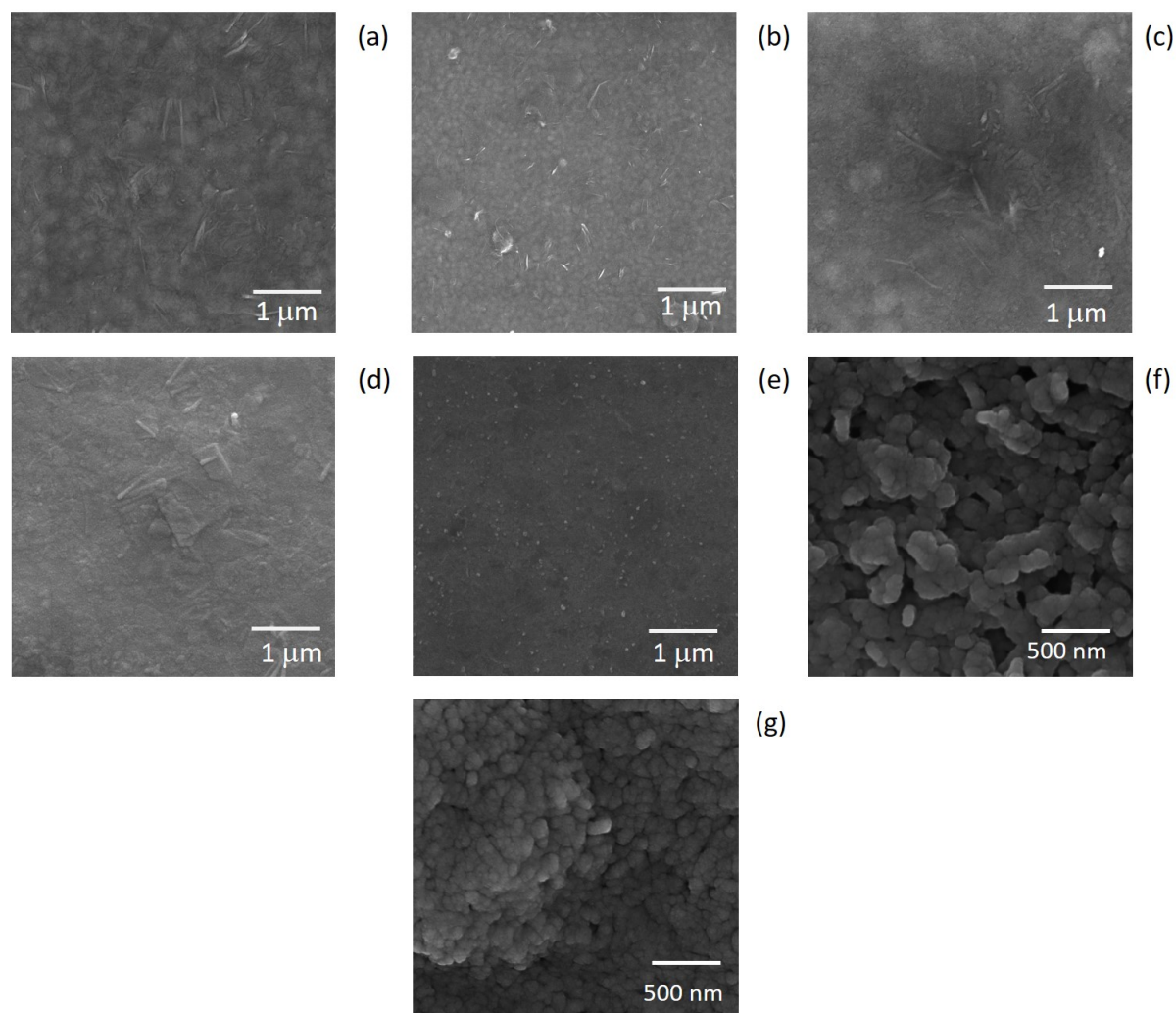


Figure S3. Scanning Electron Microscopy for GO:PEDOT:PSS (a) 40 % (b) 59 % (c) 74 % (d) 78 % and (e) 88 % thin films. (f) and (g) are referent to 500 nm scale for PEDOT and GO:PEDOT, respectively.

5 RANDOM NETWORK

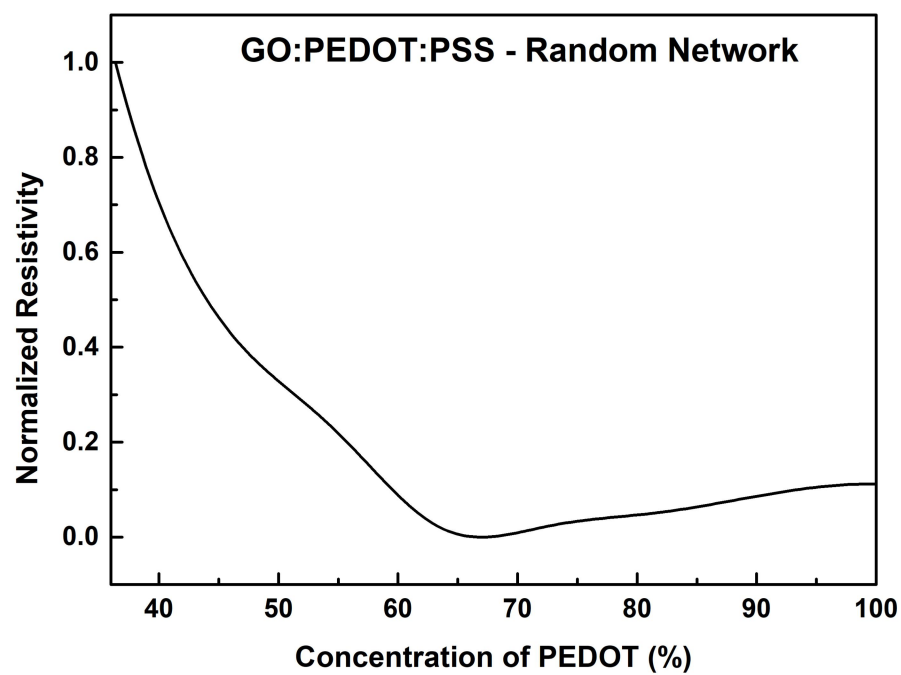


Figure S4. Normalized resistivity of GO:PEDOT:PSS random network representation.

6 6. QUALITATIVELY RESULTS

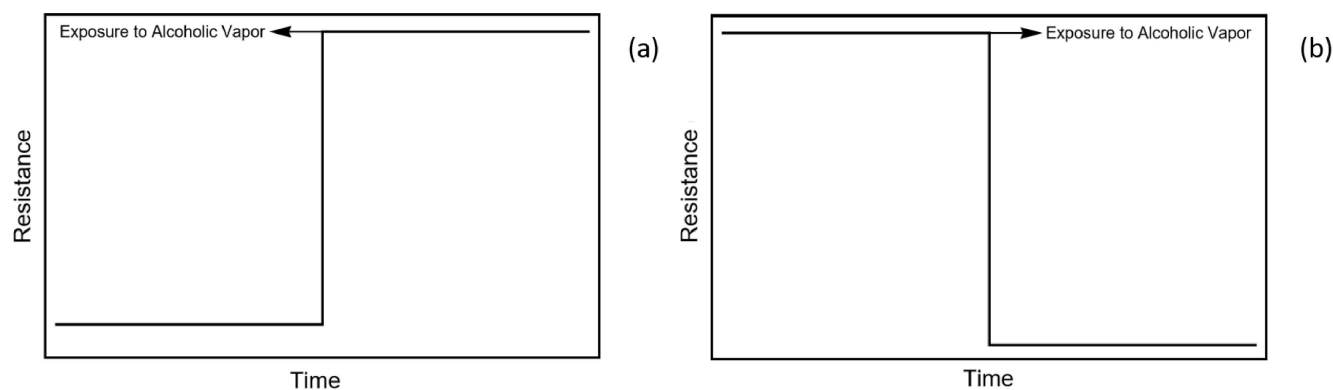


Figure S5. Step function as a result from modelling methanol sensor devices with (a) samples with PSS and (b) without PSS indicating the signal inversion during exposure to alcoholic vapor.

7 REFERENCES

REFERENCES

- das Neves, M. F., Damasceno, J. P. V., Junior, O. D., Zarbin, A. J., and Roman, L. S. (2021). Conductive ink based on pedot nanoparticles dispersed in water without organic solvents, passivant agents or metallic residues. *Synthetic Metals* 272, 116657
- Mehl, H., Matos, C. F., Neiva, E. G., Domingues, S. H., and Zarbin, A. J. (2014). The effect of variation of reactional parameters in the preparation of graphene by oxidation and reduction of graphite. *Química Nova* 37, 1639–1645