Aerosol-printed MoS$_2$ ink as high sensitivity humidity sensor

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1) Characterization of MoS2 ink by UV-Vis

The UV-Vis image of the MoS$_2$ ink is shown below. The signature absorption maxima due to the exciton states of MoS$_2$ was observed in the liquid-phase exfoliated MoS$_2$ at 453, 611, and 670 nm, and they are assigned to C, B, and A exciton states of molybdenum disulfide, respectively. According to Backes and collaborators, the observed “A” exciton maximum at 670 nm in MoS$_2$ indicates that the obtained suspension has mostly thin layers of around six sheet thickness.
2) Preparation of copper-plated phenolite and Au/Cr interdigital electrodes (IDEs)

MoS$_2$ ink sensor was prepared in the form of interdigitated electrodes fabricated on copper-plated phenolite substrate, with dimensions of 65 mm in length, 35 mm in width and 0.7 mm of separation between the interdigitated contacts. The mask used to prepare the interdigitated contacts is show in figure S2a and the device illustration is depicted in figure S2b.

The sensor also was fabricated on flexible substrate of polyethylene terephthalate (PET), where Au/Cr interdigital electrodes (IDEs) were used as electric contact made with via thermal evaporation of a 1 nm chromium layer thick followed by a 50 nm thick gold layer. The dimension of the mask employed to perform the Au/Cr interdigital electrodes were 25 mm long, 10 mm wide, and 1 mm separation between the interdigitated fingers (Figure S2c). MoS$_2$ ink device deposited on a PET is showed in figure S1d.

The measurement system is formed by a Keithley K2400 DC source, a DL Instruments/Ithaco Model 1211 Current Preamplifier (which measures current with full scale sensitivity ranging from $10^{-2}$ to $10^{-12}$ amperes) and an HP34401A multimeter. The control and collection of measurements were performed by a homemade code for the Matlab software. Electronic mass flow controllers (MFC) regulated by MKS Type 247D Four-Channel Readout controller were used to control the gas volume at the chamber inlet. A bubbler under heating was added to one argon stream to increase the chamber's
humidity and another stream of argon gas was used as purging gas and dehumidification. MoS₂ ink sensor and the commercial humidity sensor AM2302 DHT22 were put in the camera to measurements of relative humidity. Figure S3 shows a schematic of the measurement system of humidity sensor with all components.

**Figure S2.** (a) Image of the mask that was applied during the deposition process to build the device on copper-plated phenolite substrate. (b) Illustration of the device with the MoS₂ film deposited on the interdigitated contacts. (c) Image of the mask that was applied during the deposition process to build the device on flexible PET substrate. (d) Illustration of the device based on a MoS₂ film sensor on a flexible substrate.
3) Influence of time of the exfoliation on the lateral size by DLS

The size profile distribution of MoS$_2$ particles in liquid suspension was determined by dynamic light scattering (DLS) on an Anton Paar Litesizer 500 particle analyzer. The particle size distributions obtained from DLS measurements for MoS$_2$ nanoparticles prepared with different ultrasonic exfoliation times are shown in Fig. S4. One can observe that the increase of ultrasonic exfoliation time led to a change of the DLS peak position from 385 nm to 275 nm, and 180 nm, respectively, reducing the size of the MoS$_2$ flakes. The high energy generated by the effect of cavitation and micro-jet with the formation, growth, and implosive collapse of bubbles is the main driving force for the exfoliation of MoS$_2$ in the ultrasound process.$^{3,4}$

The hydrodynamic diameter is influenced by the thickness of the electrical double layer (solvation shell) and by any substance adsorbed on the flake surface (e.g., stabilizers) that move along with the nanoparticle.$^5$ Consequently, the size measured in the DLS technique could be higher in comparison with other techniques. Besides that, during DLS measurements, small objects can also be suppressed by larger ones and will not even be seen.$^6$ In summary, one can note from the discussion above that careful consideration needs to be taken when providing results of size and thickness of 2D
materials in suspension. We believe that the AFM method seems to provide a more straightforward and reliable approach to determine the thickness and size of 2D dispersion solutions.

**Figure S4.** Intensity particle size distribution for MoS$_2$ nanoflakes exfoliated with different ultrasonic times. Time of 3 hours, 4 hours, and 5 hours for 385 nm (blue curve), 275 nm (red curve), 180 nm (black curve).

4) **Characterization of the devices with MoS$_2$ ink**

Figure S5 shows the device sensitivity $S$ versus RH in linear scale for a range of 10-20%.

**Figure S5.** Device sensitivity $S$ versus RH in linear scale for the range of 10-20%.
The MoS$_2$ ink was also tested for reproducibility. Five different devices were built and they were tested in the range between 10 and 95% RH to compare their response and sensitivity. Figure S6a shows conductance versus relative humidity measurements of the MoS$_2$ ink sensors, evidencing the repeatability of the MoS$_2$ ink devices as relative humidity sensors. The figure S6b shows sensitivity S versus RH in log scale for 10%-95%, evidencing the repeatability of the MoS$_2$ ink devices and that $S$ changes from 0 to $10^3$-$10^6$ % in this humidity range.

![Figure S6](image)

**Figure S6.** Repeatability of three different devices of MoS$_2$ ink sensor a) Conductance versus RH; b) Sensitivity S versus RH in log scale; inset: Sensitivity in log linear scale for humidity values between 10% and 20% RH.

5) Test of MoS$_2$ ink on flexible PET substrate

Figure S7a shows the relationship between resistance and relative humidity to on copper-plated phenolite substrate and the flexible MoS$_2$ ink sensor on PET, showing similar sensing behavior between the sensors, which would provide broad prospects for applications in multifunctional flexible electronics. The sensitivity of flexible sensor (inset) varied from 0% to 1.9x10$^5$% (85% RH). There is a variation of five orders of magnitude in the current value, whose value approaches from that presented for the sensor on copper-plated phenolite substrate. We consider this value excellent, since it is only a preliminary study of the application of MoS$_2$ ink on a flexible substrate.

To compare the time of response of the flexible MoS$_2$ ink sensor with the response of the commercial sensor, we investigated the response and recovery dynamics of the MoS$_2$ (black) and commercial (red) sensors to sudden changes in relative humidity
(Figure S7b). The flexible MoS$_2$ sensor shows an equal response to the commercial sensor, with fast and reversible variation between the two states. Moreover, it is possible to verify that the flexible MoS$_2$ ink sensor responds faster than the commercial sensor, whose response time is 2 seconds. Thus, we can conclude that the flexible MoS$_2$ ink sensor is also an excellent candidate for application as a relative humidity sensor.

Figure S7. (a) Relationship between resistance and relative humidity of MoS$_2$ ink sensor on PET and on copper-plated phenolite substrate. Inset: Sensitivity S versus RH of MoS$_2$ ink flexible sensor. (b) Comparison of the time response of the flexible MoS$_2$ ink sensor with the response of the commercial sensor.

6) Response and Recovery times for a large switching the humidity levels

Extracting the intrinsic response and recovery times for different changes in humidity can be a difficult issue. This is so because those times are usually dependent on the measurement system and on how fast one can change the RH% in the chamber. In our case, due to our setup experimental limitations, the test chamber is not able to quickly change the humidity from low to high levels (nor the opposite). For this reason, we defined the response and recovery times as switching the humidity levels between 87% and 85% to eliminate any misleading data due to the fact of the chamber response time being longer than the sensors’ response and recovery times to reach (or decrease) a relative humidity value. In figure S8, we present the current variation (left axis) of our MoS$_2$ sensor and relative humidity (right axis) collected by sensor model DHT22 as a function of time. In the datasheet of this sensor (https://datasheetspdf.com/pdf/792211/Aosong/DHT22/1) there is the following
information: “only get new data from it once every 2 seconds”, therefore the response and recovery times are around 2 seconds. However, figure S8 depicts a response time for the commercial sensor to be about 2.67 min, indicating that the chamber is not able to change the humidity fast enough in the range of 38.5-73 %RH. Figure S8 also shows that our sensor responds to changes in humidity as fast as the commercial sensor.

Figure S8- Response and recovery dynamics of the MoS$_2$ (black) and commercial (blue) sensors to 38.5-73% changes in relative humidity.

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