FINAL SCRIPT: APPROVED FOR FILMING

Submission ID #: 69354
Scriptwriter Name: Pallavi Sharma
Project Page Link: https://review.jove.com/account/file-uploader?src=21153018

Title: Spray-Coated Melanin/PEDOT:PSS Films for Sustainable Organic Electrochemical Transistors

Authors and Affiliations:
Natan L. Nozella[1,2], Gabriel L. Nogueira[1], João V. Paulin[1], Rafael F. de Oliveira[2,3], Carlos F. O. Graeff[1]

[1]School of Sciences, Department of Physics and Meteorology, São Paulo State University (UNESP)
[2]Brazilian Nanotechnology National Laboratory (LNNano), Brazilian Center for Research in Energy and Materials (CNPEM)
[3]Institute of Physics "Gleb Wataghin" (IFGW), University of Campinas (UNICAMP)

Corresponding Authors:
Carlos F. O. Graeff carlos.graeff@unesp.br

Email Addresses for All Authors:
Natan L. Nozella n.nozella@unesp.br
Gabriel L. Nogueira leonardo.nogueira@unesp.br
João V. Paulin jv.paulin@unesp.br
Rafael F. de Oliveira rafael.furlan@lnnano.cnpem.br
Carlos F. O. Graeff carlos.graeff@unesp.br

Author Questionnaire

1. Microscopy: Does your protocol require the use of a dissecting or stereomicroscope for performing a complex dissection, microinjection technique, or something similar? **No**

2. Software: Does the part of your protocol being filmed include step-by-step descriptions of software usage? **Yes,all done**

3. Filming location: Will the filming need to take place in multiple locations? **Yes**

100 m

4. Testimonials (optional): Would you be open to filming two short testimonial statements **live during your JoVE shoot**? These will **not appear in your JoVE video** but may be used in JoVE's promotional materials. **Yes**

Current Protocol Length
Number of Steps: 20
Number of Shots: 36

Introduction

Videographer: Obtain headshots for all authors available at the filming location.

INTRODUCTION:

~~What is the scope of your research? What questions are you trying to answer?~~

1.1. **Gabriel Nogueira:** In this research, we developed sustainable organic electronic devices through a scalable deposition approach using low-cost setup and solution-processable materials.

1.1.1. INTERVIEW: Named talent says the statement above in an interview-style shot, looking slightly off-camera. *Suggested B roll: 2.4.1*

~~What are the most recent developments in your field of research?~~

1.2. **João Paulin:** Recent advances involve solution-processing of biocompatible and green materials, like melanin, and their integration into functional electronic devices.

1.2.1. INTERVIEW: Named talent says the statement above in an interview-style shot, looking slightly off-camera.

CONCLUSION:

~~What advantage does your protocol offer compared to other techniques?~~

1.3. **Gabriel Nogueira:** We demonstrate that automated spray deposition enables controlled thickness increase, leading to higher volumetric capacitance and improved OECT performance compared to conventional deposition techniques.

1.3.1. INTERVIEW: Named talent says the statement above in an interview-style shot, looking slightly off-camera. *Suggested B roll: 2.17.1*

~~How will your findings advance research in your field?~~

1.4. **João Paulin:** Melanin dedopes PEDOT:PSS, enhancing transconductance and ionic-electronic coupling, while controlled deposition enables tuning of response times and memory effects.

1.4.1. INTERVIEW: Named talent says the statement above in an interview-style shot, looking slightly off-camera.

~~What new scientific questions have your results paved the way for?~~

1.5. **Gabriel Nogueira:** Now, we aim to understand the effects of spray-coating on OECT parameters and their implementation in neuromorphic and spintronic devices.

1.5.1. INTERVIEW: Named talent says the statement above in an interview-style shot, looking slightly off-camera. *Suggested B roll: Figure 5*

Videographer: Obtain headshots for all authors available at the filming location.

Testimonial Questions (OPTIONAL):

Videographer: Please capture all testimonial shots in a wide-angle format with sufficient headspace, as the final videos will be rendered in a 1:1 aspect ratio. Testimonial statements will be presented live by the authors, sharing their spontaneous perspectives.

NOTE: Authors deleted testimonials

Protocol

2. Melanin Synthesis and Melanin/PEDOT:PSS Blends Preparation

Demonstrators: João Vitor Paulin, Gabriel Leonardo Nogueira

2.1. To begin, add 0.3 grams of 3,4-dihydroxy-DL-phenylalanine *(Three-four-Di-hydroxy-D-L-Phenyl-Alanine)* to a glass beaker **[1]**. Add 60 milliliters of deionized water to the beaker to dissolve the 3,4-dihydroxy-DL-phenylalanine under magnetic stirring **[2]**.

2.1.1. WIDE: Talent adding 0.3 grams of 3,4-dihydroxy-DL-phenylalanine powder to a clean glass beaker.

2.1.2. Talent pouring 60 milliliters of deionized water into the beaker and placing it on a magnetic stirrer.

2.2. Transfer the prepared solution to a polytetrafluoroethylene liner inside a stainless-steel reactor **[1]**. Add 400 microliters of ammonium hydroxide to start the oxidation process **[2]**. Then, close the reactor and increase the pressure to 6 atmospheres with industrial oxygen gas **[3]**. Keep the mixture under magnetic stirring for 6 hours **[4]**.

2.2.1. Talent pouring the solution into the polytetrafluoroethylene liner.

2.2.2. Talent pipetting 400 microliters of ammonium hydroxide into the liner.

2.2.3. Talent closing the stainless-steel reactor and adjusting the gas regulator to increase pressure to 6 atmospheres.

2.2.4. Shot of the closed reactor placed on a magnetic stirrer.

2.3. Transfer the synthetic mixture into a dialysis membrane **[1]**. Dialyze against deionized water to extract and purify the melanin **[2]**, replacing the water daily for 7 days or until it becomes colorless **[3]**. Dry the purified material in an oven at 90 degrees Celsius for 12 hours to obtain melanin powder **[4]**.

2.3.1. Talent transferring the synthetic mixture into the dialysis membrane.

Added shot:2.3.2C Shot of the dialysis setup in deionized water after day 0 (after 3 hours in the dialysis membrane in water).

2.3.2. A Shot of the dialysis setup in deionized water Day 7.

Added shot: 2.3.2 B Removing the synthetic mixture of the dialyze membrane and placing it in a beaker.

2.3.3. Talent placing the dialyzed material into an oven set at 90 degrees Celsius.

2.4. Then, place the substrate below a shadow mask patterned to obtain five pairs of source and drain electrodes on each substrate, separated by 1 millimeter in length and in width **[1]**. Afterwards, place the shadow mask with the substrates in the evaporation support **[2]**.

2.4.1. A Talent positioning the substrate carefully beneath the patterned shadow mask.

Added shot: 2.4.1. B Placing the shadow mask in the evaporation support.

2.5. Now, place the required amount of gold in the evaporation boat to ensure adequate film thickness and uniform deposition **[1]**. Place the masked samples into the sample holder inside the evaporation chamber **[2]**.

2.5.1. Talent placing gold pieces into the evaporation boat.

2.5.2. Talent inserting the masked substrates into the sample holder of the evaporation chamber.

2.6. Next, configure the deposition controller to begin gold deposition at a slow rate of 0.2 angstroms per second until a thickness of 5 nanometers is reached for controlled nucleation and uniform film formation **[1]**. Then, set the deposition rate to 1 angstrom per second until the final thickness of 100 nanometers is achieved **[2]**.

2.6.1. Show the interface of the deposition controller being set to 0.2 angstroms per second with a stop at 5 nanometers.

2.6.2. Show the deposition rate being adjusted to 1 angstrom per second and continue until reaching 100 nanometers.

2.7. Automatically evacuate the evaporation chamber until a vacuum of 5×10^{-6} millibar is reached and start the evaporation process **[1]**.

2.7.1. Show the vacuum gauge decreasing to 5×10^{-6} millibar and the controller initiating the evaporation sequence.

2.8. Wait for the deposition process to complete **[1]**. Once the evaporation chamber has cooled for 1 hour, refill the chamber until 1 atmosphere and remove the substrates **[2]**.

2.8.1. Shot of the completed deposition process, with display showing completion.

2.8.2. Talent carefully removing the cooled substrates from the evaporation chamber.

2.9. Then, cover the gold electrodes with an adhesive mask that has openings for the channel deposition **[1]**.

2.9.1. Talent applying the adhesive mask onto the substrate.

2.10. Select a robust three-dimensional printer with suitable dimensions to use as the base platform for the spray-coating system **[1]**.

2.10.1. Talent showing compatible three-dimensional printer, clearing the platform for setup.

2.11. Then, modify the three-dimensional printer framework to mount the airbrush for mechanical adaptation. Remove the extruder **[1]** and place the airbrush on a custom holder **[2]**.

2.11.1. Talent disassembling the extruder from the three-dimensional printer.

2.11.2. Talent installing the airbrush into a custom-designed holder mounted on the printer.

2.12. Use the X-, Y-, and Z-axis stepper motors to control the motion and height of the airbrush **[1]**. Adapt the extruder to actuate the airbrush trigger, enabling software-controlled spray release **[2]**.

2.12.1. SCREEN: 69354_Screenshot_1.mp4 00:00-00:19

2.12.2. Talent connecting the extruder control system to the airbrush trigger mechanism.

2.13. Then, center the airbrush in the XY plane directly above the hot plate, maintaining a vertical distance of 15 centimeters from the airbrush to the hot plate surface **[1]**.

2.13.1. Talent aligning the airbrush above the center of the hot plate and measuring 15 centimeters vertically.

2.14. Now, set the hot plate temperature to 100 degrees Celsius **[1]** and adjust the compressed air pump to a pressure of 1 bar **[2]**.

2.14.1. Talent programming the hot plate to 100 degrees Celsius on its digital interface.

2.14.2. Talent setting the pressure regulator on the compressed air pump to 1 bar.

2.15. Program the G-code to move the airbrush 10 centimeters along the X-axis at a speed parameter of F8000 *(F-Eight-Thousand)* **[1]**. Set the spray opening width using the **E1.5** *(E-One-Point-Five)* command and define a dwell time of **P5000** *(P-Five-Thousand)* between forward and return strokes **[2]**.

2.15.1. SCREEN: 69354_Screenshot_2.mp4. 00:00-00:14

2.15.2. SCREEN: 69354_Screenshot_3.mp4. 00:00-00:18

2.16. Then, place the masked substrate, with the source and drain electrodes already deposited, at the center of the hot plate **[1]**.

Added shot: 2.16.1. A Talent aligning the source-drain gaps and a shadow mask.

2.16.1. Talent positioning the prepared substrate carefully at the center of the hot plate.AUTHOR'S NOTE: Use file 2.16.1 B for this step

2.17. Add 250 microliters of the melanin and PEDOT:PSS *(Pe-Dot-P-S-S)* solution into the airbrush reservoir **[1]**. Edit the desired number of spray cycles in the G-code, typically setting it to 10 cycles, and run the program **[2-TXT]**.

2.17.1. A Talent pipetting 250 microliters of melanin/PEDOT:PSS solution into the reservoir of the airbrush.

2.17.2. SCREEN:69354_Screenshot_4.mp4. 00:07-00:22
TXT: Repeat until a total of 2 mL solution has been deposited

AND
Added shot: 2.17.1B Running of spray cycles to deposit the film in the delimited area.
Video Editor: Please play both shots side by side in a split screen

2.18. Clean the airbrush reservoir with deionized water after completing the deposition **[1]**.

2.18.1. Talent rinsing and cleaning the airbrush reservoir using deionized water.

2.19. To measure the output curves of OECTs, sweep the drain voltage from 0 volts to minus 0.6 volts ~~while keeping the gate voltage fixed between minus 0.5 volts and 0.5 volts~~ **[1-TXT]**. Record the drain current at each step **[2]**.

2.19.1. SCREEN: 69354_Screenshot_5.mp4. 00:00-00:14
TXT: Keep gate voltage fixed between - 0.5 V to 0.5 V

2.19.2. SCREEN: 69354_Screenshot_6.mp4. 00:04-00:14

2.20. To measure the transference curve, fix the drain voltage at minus 0.5 volts **[1]**. Sweep the gate voltage from minus 0.5 volts to plus 0.6 volts and record the drain current **[2]**.

2.20.1. SCREEN: 69354_Screenshot_7.mp4. 00:00-00:17

2.20.2. SCREEN: 69354_Screenshot_8.mp4. 00:04-00:12, 00:30-00:38

Results

3. Results

3.1. UV–Visible spectra confirmed successful incorporation of melanin into PEDOT:PSS *(P-E-Dot-P-S-S)* films by the appearance of characteristic melanin absorption bands **[1].** FTIR analysis showed interactions between melanin and PEDOT:PSS, demonstrated by the disappearance of characteristic PEDOT:PSS absorption bands **[2].**

3.1.1. LAB MEDIA: Figure 3A. *Video editor: Highlight the curve labelled "Pristine Melanin" and Pristine PEDOT:PSS*

3.1.2. LAB MEDIA: Figure 3B. *Video editor: Highlight the "Pristine PEDOT:PSS" spectrum curve.*

3.2. Profilometry revealed that pristine PEDOT:PSS films had the highest thickness of 961 nanometers **[1],** which decreased to 342 nanometers upon addition of 10 percent melanin **[2],** then increased to 493 nanometers at 30 percent **[3],** and 562 nanometers at 50 percent melanin **[4].**

3.2.1. LAB MEDIA: Table 1. *Video editor: Highlight the cell for thickness of "Pristine PEDOT:PSS" showing 961 ± 48 nanometers.*

3.2.2. LAB MEDIA: Table 1. *Video editor: Highlight the thickness value for "10Mel" showing 342 ± 33 nanometers.*

3.2.3. LAB MEDIA: Table 1. *Video editor: Highlight the thickness value for "30Mel" showing 493 ± 27 nanometers.*

3.2.4. LAB MEDIA: Table 1. *Video editor: Highlight the thickness value for "50Mel" showing 562 ± 41 nanometers.*

3.3. Atomic force microscopy showed that the surface roughness of pristine PEDOT:PSS films was 4.4 nanometers **[1],** decreased to 3.3 nanometers with 10 percent melanin **[2],** and increased to 7.8 nanometers and 15.6 nanometers with 30 percent and 50 percent melanin, respectively **[3].**

3.3.1. LAB MEDIA: Figure 4A. *Video editor: Highlight the "Pure PEDOT" image showing RMS: 4.4 ± 0.4 nanometers.*

3.3.2. LAB MEDIA: Figure 4B. *Video editor: Highlight the "10Mel" image showing RMS: 3.3 ± 0.3 nanometers.*

3.3.3. LAB MEDIA: Figure 4C and D. *Video editor: Highlight the "30Mel" and "50Mel" image*

3.4. Output curves showed that pristine PEDOT:PSS devices produced high drain currents, which decreased upon addition of 10 percent melanin **[1].** Transfer characteristics for 10 percent melanin devices displayed a pronounced hysteresis loop and an ON and OFF current ratio of approximately 150 **[2].**

3.4.1. LAB MEDIA: Figure 5A.

3.4.2. LAB MEDIA: Figure 5B. *Video editor: Show the two red curves and mark the looped shape.*

3.5. Transconductance measurements revealed that the maximum gm *(G-M)* value shifted to more negative gate voltages as melanin concentration increased **[1].**

3.5.1. LAB MEDIA: Figure 5C. *Video editor: Highlight the peaks of each curve and show their progressive shift leftward on the x-axis*

3.6. The normalized maximum transconductance was highest for the 10 percent melanin blend, which had the lowest film thickness **[1].**

3.6.1. LAB MEDIA: Figure 5D. *Video editor: Highlight the peak in the black curve labelled "Normalized gm,max"*

Pronunciation Guide:

1. Melanin
   Pronunciation link: https://www.merriam-webster.com/dictionary/melanin
   IPA: /ˈmɛl.ə.nɪn/
   Phonetic Spelling: meh-luh-nin
2. PEDOT:PSS
   Pronunciation link: No confirmed link found
   IPA: /ˈpiː.dɒt ˌpiːˌɛsˈɛs/
   Phonetic Spelling: pee-dot pee-ess-ess
3. Polytetrafluoroethylene
   Pronunciation link: https://www.merriam-webster.com/dictionary/polytetrafluoroethylene
   IPA: /ˌpɒl.iˌtɛt.rəˌflʊə.roʊˈɛθ.əˌliːn/
   Phonetic Spelling: pah-lee-tet-ruh-floo-roe-eth-uh-leen
4. Ammonium hydroxide
   Pronunciation link: https://www.merriam-webster.com/dictionary/ammonium%20hydroxide
   IPA: /əˈmoʊ.ni.əm haɪˈdrɒk.saɪd/
   Phonetic Spelling: uh-moh-nee-uhm hy-drok-side
5. Atmospheres
   Pronunciation link: https://www.merriam-webster.com/dictionary/atmosphere
   IPA: /ˈæt.məˌsfɪrz/
   Phonetic Spelling: at-muh-sfeerz
6. Dialysis
   Pronunciation link: https://www.merriam-webster.com/dictionary/dialysis
   IPA: /daɪˈæl.ə.sɪs/
   Phonetic Spelling: dye-al-uh-sis
7. Substrate
   Pronunciation link: https://www.merriam-webster.com/dictionary/substrate
   IPA: /ˈsʌb.streɪt/
   Phonetic Spelling: sub-strayt
8. Evaporation
   Pronunciation link: https://www.merriam-webster.com/dictionary/evaporation
   IPA: /ɪˌvæp.əˈreɪ.ʃən/
   Phonetic Spelling: ih-vap-uh-ray-shun
9. Nanometers
   Pronunciation link: https://www.merriam-webster.com/dictionary/nanometer
   IPA: /ˈnæn.əˌmiː.tərz/
   Phonetic Spelling: nan-uh-mee-terz
10. Vacuum
   Pronunciation link: https://www.merriam-webster.com/dictionary/vacuum

IPA: /ˈvæk.juːəm/
Phonetic Spelling: vak-yoo-uhm

11. Three-dimensional
    Pronunciation link: https://www.merriam-webster.com/dictionary/three-dimensional
    IPA: /ˌθriː.dɪˈmɛn.ʃən.əl/
    Phonetic Spelling: three-dih-men-shuh-nuhl
12. Transconductance
    Pronunciation link: No confirmed link found
    IPA: /ˌtræns.kənˈdʌk.təns/
    Phonetic Spelling: trans-kun-duhk-tens
13. Spectra
    Pronunciation link: https://www.merriam-webster.com/dictionary/spectrum
    IPA: /ˈspɛk.trə/
    Phonetic Spelling: spek-truh
14. Profilometry
    Pronunciation link: No confirmed link found
    IPA: /ˌproʊ.fɪˈlɒ.mə.tri/
    Phonetic Spelling: proh-fih-lah-muh-tree
15. Atomic force microscopy
    Pronunciation link: No confirmed link found
    IPA: /əˈtɒm.ɪk fɔːrs maɪˈkrɒs.kə.pi/
    Phonetic Spelling: uh-tom-ik fors my-kros-kuh-pee
16. Hysteresis
    Pronunciation link: https://www.merriam-webster.com/dictionary/hysteresis
    IPA: /ˌhɪs.təˈriː.səs/
    Phonetic Spelling: his-tuh-ree-sis