



Pioneering Advances in Materials

Journal homepage: <http://piadm.sbu.ac.ir>

ISSN: 3115-7114

Research paper

High-Resolution Perovskite Light-Emitting Touch-Sensitive Device via Trowel-Coating Technique

Talebifar Faezeh, Taheri Ghahrizjani Reza, Sadeghian Mohamadreza, Rostami Aliakbar, Mohajerani Ezeddin*

Laser and Plasma Research Institute, Shahid Beheshti University, Tehran, Iran.

*e-mohajerani@sbu.ac.ir

Article info:

Article history:

Received: 21/05/2025

Accepted: 05/07/2025

Keywords:

light emitting,
perovskite, touching
devices, coating method,
silver nanowires.

Abstract

There is a growing demand for touch devices, including displays, fingerprint sensors, and other pattern recognition systems. Recent advances in light-emitting diode (LED) materials have opened up new possibilities for developing emissive touch devices. In such systems, touching the LED causes a visible glow in a fine pattern, which can be utilized in various applications. Here, a perovskite LED (PeLED) is turned into a touch device. Such a device, on one hand, needs a uniform and high-resolution emitting layer, and on the other hand, a flexible and thin top electrode. The uniformity, and hence the resolution of the device, is achieved by an innovative coating method. Instead of the conventional spin-coating technique, the emitting layer solution is dropped onto a glass substrate and then uniformly distributed by troweling another glass substrate across it. This process forms a thin, uniform perovskite layer with fine and small microcrystals, which is well-suited for high-resolution touch applications. The emissive material used was methylammonium lead bromide (MAPbBr_3) and the structure of the top flexible electrode was polydimethylsiloxane/ silver nanowires/ poly(3,4-ethylenedioxythiophene): Poly-(Styrenesulfonate) (PDMS/AgNWs/PEDOT: PSS). Also, a thin layer of Poly(methylmethacrylate) (PMMA) was applied after the perovskite layer to improve the performance of the device. With this device, we succeeded in recording the pattern of a fingerprint with a resolution of about "100 nm", which was stable after repeatedly touching the device. However, further work is needed to evaluate the reproducibility and scalability of the proposed fabrication method for industrial applications.

1. Introduction

Touching devices have a very important role in recent technology and will be more in demand in

the future. These devices in different forms have found applications in display, robotics, medicine, security, and many more for identification, data collection, and other



applications[1-4]. These devices have been commercialized for over 3 decades and are growing in science and engineering. Different techniques based on optics[5], capacitor[6], resistance[7], and audio[8], are examples of mechanisms used to develop such devices[9, 10].

These devices usually require additional electronics to receive and display output readings[11].

Light-emitting touching devices (LETD) have recently attracted some attention [12]. In such devices, in order to display details of touching objects such as fingerprints, on the one hand, the resolution in the emitting layer is important, and on the other hand, the use of flexible touching electrodes is essential[13, 14].

Resolution is determined by the structure of the emitting layer materials and also the way the material is coated on the substrate in a way that, by switching one point of the device on, a similar pattern as the contact is emitted [15].

Such property is also important for other applications, such as micro-displays and producing fine emissive patterns. In these applications where resolution matters, organic light-emitting diodes (OLEDs) have shown good potential due to the molecular nature of the emitting elements[16].

Devices based on OLEDs have shown good resolution, luminance, and lifetime[17]. However, due to many potential advantages of PeLEDs, researchers are trying to use these materials for LETDs [18-21].

There are still some challenges for the perovskite layer for such devices. Apart from the material, the coating method strongly affects the properties of the emitting layer and hence the emission[22].

As mentioned for such devices a uniform, almost similar size crystals and dense but high-resolution layer is crucial.

To achieve this, the crystals must be small, with little overlap but dense enough to cover all the surface with no empty space is required[23, 24].

There are different coating methods from solution such as dip, spin, blade, spray coating and other methods, with different effect on the properties of the layer[25].

Thickness, morphology, grain size, crystal size, uniformity, and overlap of the species are different in different methods.

For laboratory production of PeLEDs, spin-coating which makes thin uniform layer of micro-crystals of perovskites, is very popular[26, 27]. However, for perovskite which micro-crystals will be formed by solvent evaporation, Spin-coating usually, does not give required uniformity and morphology.

This results in non-uniform emission and normally many defects are visible by eye[28].

To improve the result researchers tried different solvents, adding different acids or other materials to speed the crystallization and other ways although the final results were not successful especially for high-resolution applications[29].

Blade-coating of perovskite layer also have been reported to reduce the problems of Spin-coating. Apart from touchable emitting devices, resolution is important in micro-displays, and making fine emitting patterns like reticles [30, 31]. Here in this article, a new coating method is introduced. With this innovative method, we achieved fabrication of a PeLED with an emitting layer suitable for high-resolution applications such as touch-emitting devices.

So-called Trowel-coating is used to make the perovskite the emissive layer. This method has many more advantages as compared to Spin-coating, apart from making a uniform emitting layer. In this way, the consumption of the solution was much less than Spin-coating, and the required uniformity of the emitting layer was achieved. Also, in order to resolve the touching pattern, the size of the crystals is important. Uniformity and size of micro-crystals make this novel coating method attractive for such applications where the resolution matters. The device was optimized by applying a layer of PMMA in the structure for better performance[32, 33]. At the end, a flexible electrode made of silver nanowires was designed to perform the touching function[34]. The final device was capable of recording the pattern of a fingerprint.

2. Experimental section

The structure *ITO/PEDOT: PSS/PVK/MAPbBr₃/PMMA/Ag* shown in Figure 1a, b, is used to make the perovskite light-emitting diode (PeLED). This structure is chosen based on experiences and works reported by others.

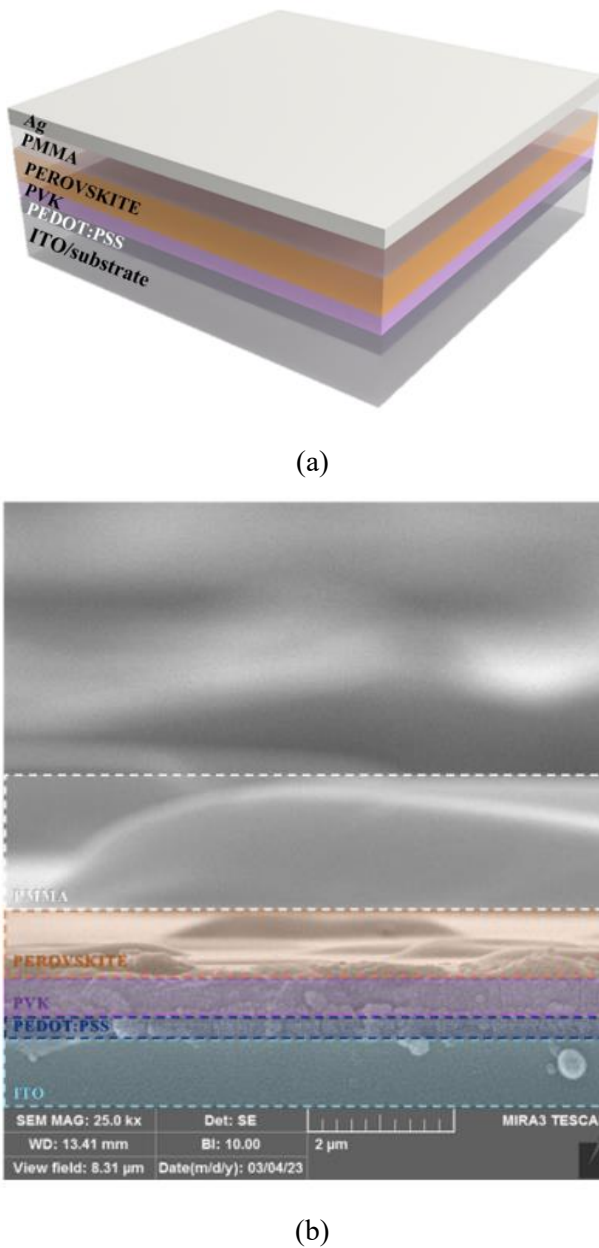


Figure 1. (a) Structure schematic of the PeLED used in this study and (b) layers cross-sectional SEM image.

3. Materials

N, N-Dimethylformamide (DMF) (anhydrous, 99.8%), Poly(9-Vinylcarbazole) (PVK), Poly(methylmethacrylate) (PMMA), poly(3,4-ethylenedioxythiophene): Poly-(Styrenesulfonate) (PEDOT: PSS), lead (II) bromide (99.999%) ($PbBr_2$), Chlorobenzene and polydimethylsiloxane (PDMS) were purchased

from Sigma-Aldrich. The methylammonium bromide (MABr) was purchased from 1-Material Inc. Silver nanowires (AgNWs) were purchased from AMINBIC Advanced Materials CO. All materials are used directly without any further purification.

3.1. Perovskite synthesis

The perovskite precursor solution was prepared by dissolving $PbBr_2$ and MABr with a 1:1.15 molar ratio in anhydrous DMF to give weight ratios of 20%, 30%, and 40%. The resulting mixture was then stirred for 2h at 60°C.

3.2. Flexible cathode fabrication

First, a solution of 10mg of AgNWs in 1 cc of ethanol was prepared. Then, a layer of $19 \Omega sq^{-1}$ of AgNWs was made using a capillary tube on a glass substrate. After that, a thin layer of PDMS was coated on the layer of AgNWs with a capillary tube. Now the glass substrate with two layers of AgNWs and PDMS was dried at 100 °C for 5 minutes. Layers of AgNWs and PDMS were pulled off the glass and mounted on another glass substrate with a PDMS layer at the bottom. PEDOT: PSS was spin-coated on the AgNWs layer at 3500 rpm, followed by 30 minutes drying on the hot plate at 80°C. The flexible electrode is ready and can be separated from the glass. The thickness of the flexible cathode at the end was about 60μm (about 40 μm PDMS, 15 μm AgNWs and 5 μm PEDOT: PSS) and the surface resistance was about 10 Ω (Figure 2).

3.3. Device fabrication

Here, the fabrication of the device is described step by step, briefly (Figure 2).

ITO-coated glasses ($14 \Omega sq^{-1}$), which are used as substrate, were washed with distilled water, de-ionized water, and isopropanol, in an ultrasonic bath for 20 minutes.

The substrates were dried with nitrogen, and then UV-ozone was applied for 30 minutes to remove any impurities and to make better layers. PEDOT: PSS was spin-coated at 3500 rpm on the cleaned substrates. The substrates were then annealed for 20 minutes at 120°C.

A solution of 10mg PVK in 1 cc of chlorobenzene was then spin-coated at 3000 rpm and then annealed at 120°C for 20 minutes. To make the emitting perovskite layer, a solution of

MAPbBr_3 in DMF was made by dissolving PbBr_2 and MABr in a 1:1.15 ratio at 20, 30 and 40 weight percent.

The emitting layer was made by spin-coating and Trowel-coating.

For Spin-coating, 3000 rpm was applied, and then the sample was annealed at 60°C for 20 minutes. In our novel coating method, so-called Trowel-coating, the idea is similar to blade coating, but instead of applying a blade, here the layer is formed by sliding another glass, such as a microscope slide, just like troweling the droplet of the coating material over the previous layers. To do this, the coated substrate is mounted over another microscope slide to make the operation easier.

$10\mu\text{l}$ of MAPbBr_3 solution was dropped over another microscope slide, and like a trowel, this glass was put on top of the coated substrate and moved with a speed of about 1 cm/s.

After moving the glass apart, nitrogen at 0.25 MPa pressure was applied for 1 minute to remove the solvent, followed by drying over a hot plate at 60°C for 20 minutes (Figure 3).

After making the emitting layer with Spin-coating and Trowel-coating, a layer of PMMA was added using dip coating from the solution of 20 mg polymer in 1 cc of chlorobenzene at a speed of 1 cm/s.

Finally, to study the PeLED itself without touching, silver paste was used as to electrode, which was cast using a capillary tube to make a smooth and uniform layer.

In order to make a touching PeLED, the already prepared PDMS/AgNWs/ PEDOT: PSS flexible electrode was placed over the device using a $100\mu\text{m}$ spacer.

All the fabrication steps and characterization tests were performed at room temperature.

3.4. Characterization

Scanning electron microscope images were taken by a MIRA3TESCAN-XMU.

The crystalline properties of the perovskite were determined by a Shimadzu XRD-7000 X-ray diffractometer.

Photoluminescence spectrum, absorption spectrum, and electroluminescence spectra were taken using the spectrometer AvaSpec-ULS2048CL-EVO.

The I-V and current switching tests were performed using a Keithley 2400 device.

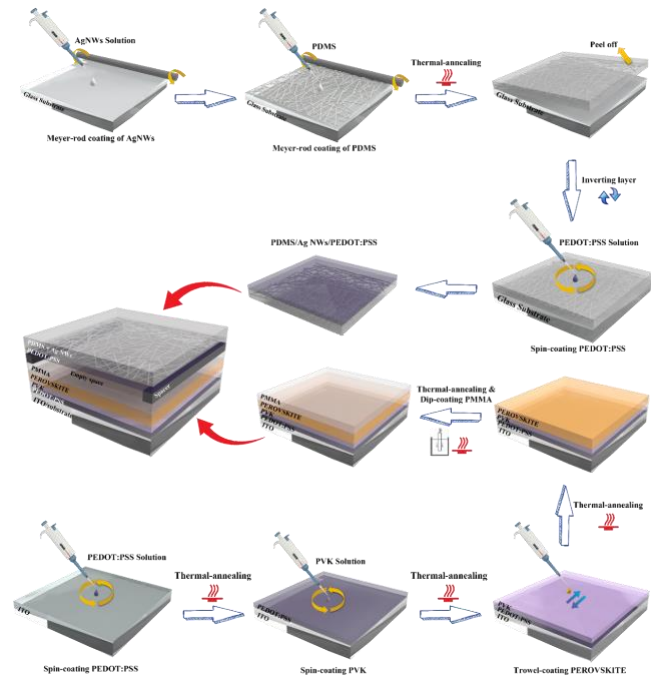


Figure 2. Schematic of the fabrication process of LETD.

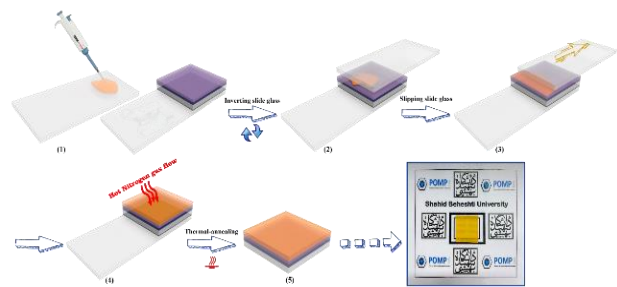


Figure 3. Trowel-coating steps.

4. Results and discussions

In order to achieve a LETD, we need an LED with uniform light and with the ability to give high-resolution light. Also, we need a flexible electrode to complete the touching action. Most of the results describe the parameters involved in making the necessary light-emitting layer suitable for this application. Spin-coating has some restrictions in making such an LED from perovskite. We start to review the results comparing the structure and morphology of the emitting layer between Spin-coating and Trowel-coating.

4.1. The effect of the concentration of perovskite in the Spin-coating and Trowel-coating methods. It was expected that the concentration of the perovskite and the solvent would affect the quality of the coated layer using either Spin-coating or Trowel-coating. $PbBr_2:MABr$ at the ratio of 1:1.15 and concentrations of 20, 30, and 40 weight percent were coated by both methods. Figure 4 show the top-down SEM image for the perovskite layer made at different concentration for both coating methods.

As expected, when the layer was coated by Spin-coating, the micro-crystals were scattered, and by increasing the concentration, the crystal size increased.

This could be due to the empty space between seeds, which can make larger crystals at higher concentrations. The density of micro-crystals will be increased when the perovskite layer is made using Trowel-coating.

It is interesting that by increasing the concentration of the perovskite solution, the micro-crystals grow more uniformly and denser to make a more defect-free emitting layer.

It seems that here the initial crystal growth centers will be more uniformly scattered across the layer, and crystal size will not increase too much as compared to Spin-coating at higher concentrations. In Spin-coating, the free space around crystal centers allows the crystals to grow at different sizes or even different shapes.

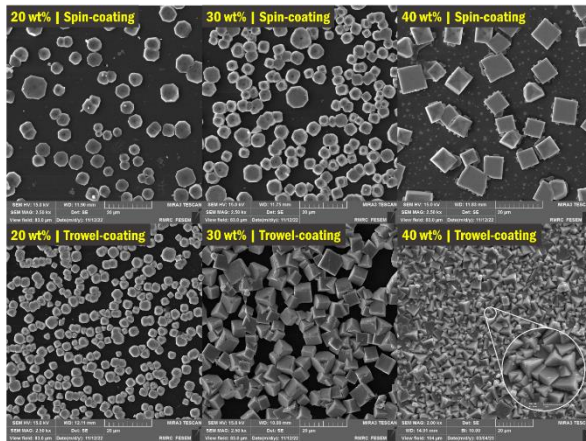
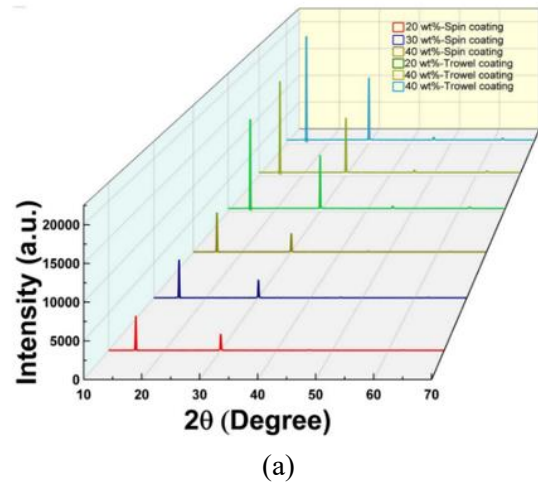


Figure 4. Perovskite layer top-down SEM images in different concentrations and two coating methods.

XRD of the perovskite layers is shown in Figure 5a. It is clear that the general structure of the X-ray diffraction patterns is almost similar to previously reported structures, and validates the presence of a perovskite micro-crystal structure. However, for Trowel-coating, the intensity of peaks at 15.01 and 21.23 is considerably increased, which seems to be due to the more intense number of crystals, which is due to the increasing number of crystal seeds[35, 36].

Figure 5b shows the absorption spectra of the perovskite layer coated by two methods. Trowel-coating makes the layer more absorbing as expected due to a denser crystal distribution. Photoluminescence (PL) from the coated layer at different concentrations and different coating methods is shown in Figure 5c. The PL spectra also show a stronger peak for trowel coating at 40% concentration. A stronger PL peak means fewer traps and more recombination, which is the result of more dense micro-crystals. In Trowel-coating more materials are crystallized as compared with Spin-coating which makes a denser crystal structure. Also, a blue shift in PL peak of the layers made by Trowel-coating is observed, which seems to be due to different crystal structures of two coating methods (Figure 5d). The SEM images of the crystal structure for both coating methods show that the structures and distribution of crystals are different in both coating methods, which might be due to the changes in pressure, temperature, and which result in a shift in PL. Parameters such as temperature, pressure, and, more importantly, coating method, affect the size and crystal structure, which affects the photoluminescence.



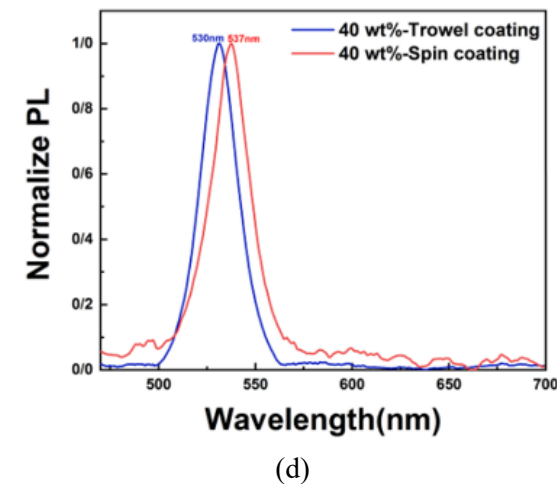
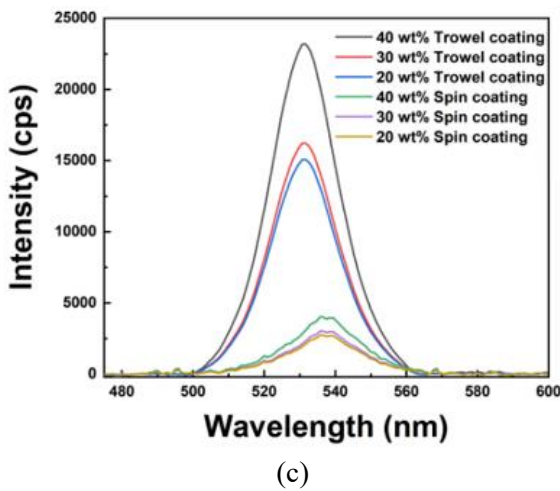
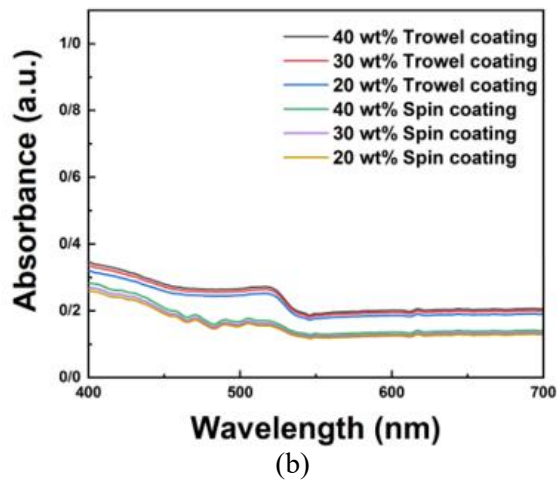
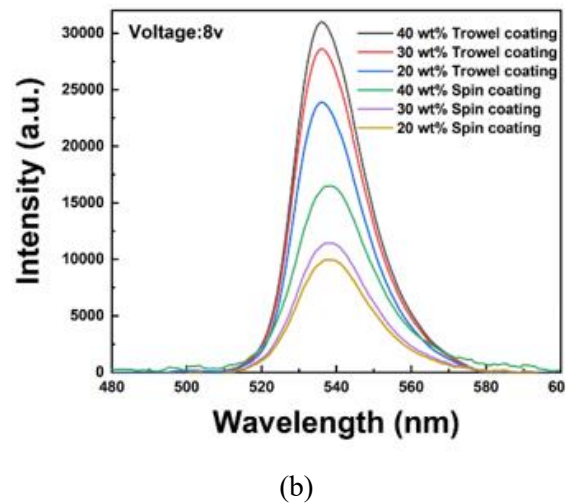
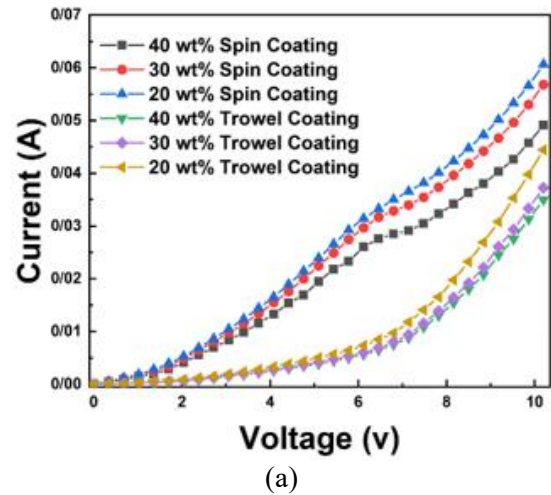
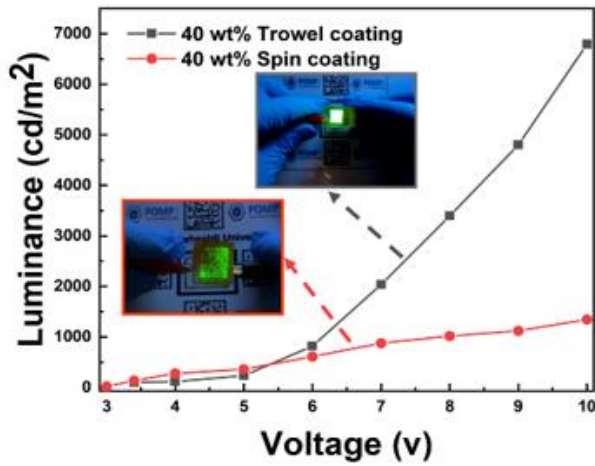


Figure 5. (a) XRD patterns of the $MAPbBr_3$ thin films, (b) absorption spectra, (c) Photoluminescence spectra in different concentrations and, (d) shift peak PL in 40 wt% concentration in two coating methods.

Figure 6a compares the I-V diagram of the PeLED made by both coating methods. It seems that the surface resistance in the layer formed by Trowel-coating is larger due to the denser distribution of microcrystals. Also, the figure shows that the current at a fixed voltage is less in this layer as compared with the layer formed by Spin-coating. Figure 6b shows the electroluminescence from complete devices fabricated by both methods with the fixed top electrode. Due to empty spaces and scattered micro-crystals made in spin-coated samples, the voltage cannot be increased enough and certainly the efficiency is restricted. When the layer is coated by Trowel-coating, a denser crystal structure allows applying higher voltage without any short-circuit, and hence, much more intense light can be achieved. Figure 6c shows the device with the luminosity of about 7000cd/m² at 10 volts.





(c)

Figure 6. (a) graph for I–V characteristic measured, (b) electroluminescence spectrum and (c) luminance on an $ITO/PEDOT:PSS/PVK/MAPbBr_3/PMMA/Ag$ device.

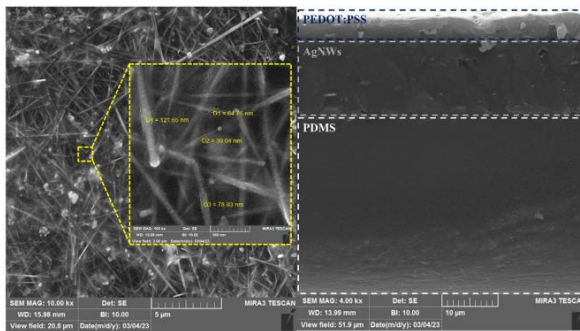
4.2. Flexible electrode

The flexible and ultra-thin electrode is essential for a touch device to make a high-resolution contact. Figure 7a shows cross SEM and top SEM of the electrode.

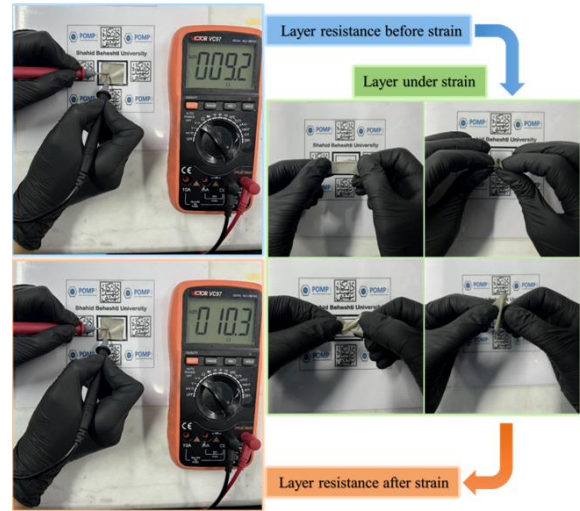
The flexibility and conductivity of the electrode was examined by Scotch adhesive, which showed good stability.

The conductivity remained unchanged after 50 times applying cello tape.

Also, the conductivity showed no changes after applying 15% stress and after bending many times. The thickness of the electrode was about $60\mu m$ which was thin enough for transferring the details of the touching objects. Figure 7b shows the results of the conductivity tests.



(a)



(b)

Figure 7. (a) AgNWs layer top-down SEM images and PDMS/AgNWs/PEDOT:PSS cross-sectional SEM image, and (b) Testing changes in electrode resistance under different strains.

4.3. The final touching device performance

As shown previously in Figure 2, the final device, including the optimized PMMA layer and installed flexible electrode, with the emitting layer coated by Trowel-coating, was tested for touching performance.

As mentioned, this LETD is made like a normal device according to the schematic of Fig., with the difference that the upper electrode is separated from the lower part (radiating layer) of the device by a spacer. The working mechanism of the LETD is shown in the schematic of Figure 8a. When an appropriate pressure is applied to this stretchable layer, a convex deformation is created in it, which enables the AgNWs/PEDOT:PSS to contact the light-emitting layer. The voltage applied between the ITO and the composite electrode causes the injection of electrons from the AgNWs and holes from the ITO into the perovskite layer and emission of green light. When the pressure is removed, the connection is broken, and the electrode returns to its original state, thus stopping light production. As you can see in Figure 8b, the touch device shows instantaneous light emission only in the areas where pressure is applied and an electrical connection is

established. for example, a Fingerprint is used as a touching object with a spacing of about $100\mu\text{m}$.

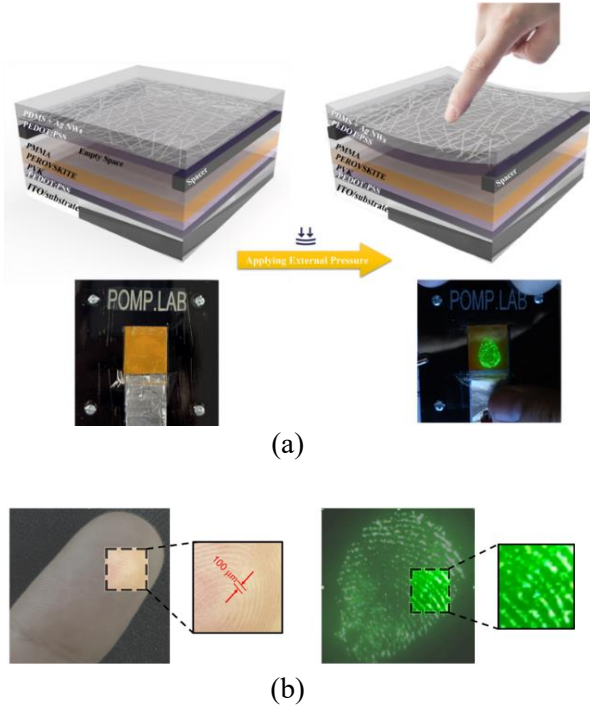


Figure 8. (a) Schematic illustration of LETD working mechanism and bar graphs of materials involved and, (b) An image of the resolution of the LETD.

The device was tested for repeatability, and required applied voltage and the clearance of the pattern. The fingerprint pattern was clearly visible by applying a voltage of 7 volts. By increasing the voltage, the intensity becomes so high to make the pattern unclear, and lower voltages reduce the visibility of the pattern (Figure 9a). By touching the flexible electrode gradually, the current was increased after any touch, resulting in a slight increase in current after 50 times touching (Figure 9b).

Image processing is used to compare changes between patterns after each touch. At first, the following function was introduced, which calculates the degree of similarity numerically by taking two histograms from the reference image (H_1) and the desired image (H_2) and comparing them with each other. Figure 9c shows that the device shows high stability after 40 times of fingerprinting, so that we see a high similarity of 70% between the 40th image and

the reference image, which indicates the high accuracy of the device in successive touches.

$$d(H_1, H_2) = \frac{\sum(H_1 - \bar{H}_1) \times (H_2 - \bar{H}_2)}{\sqrt{\sum(H_1 - \bar{H}_1)^2 \times (H_2 - \bar{H}_2)^2}}$$

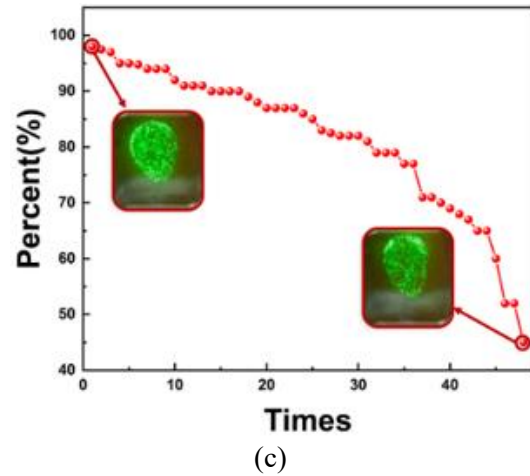
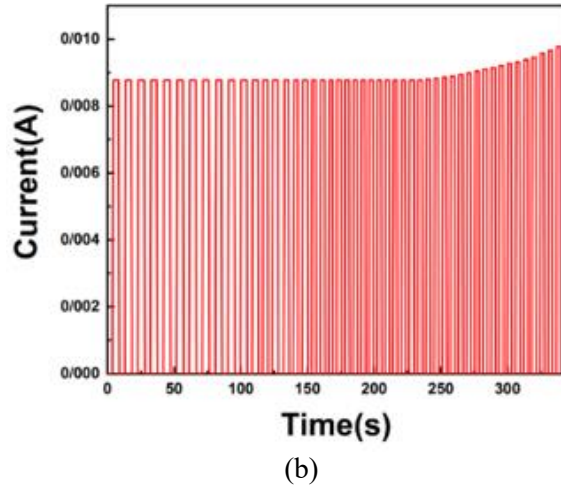
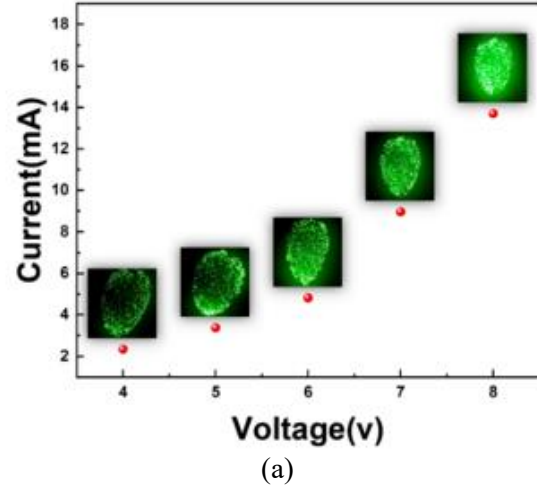


Figure 9. (a) the resolution of the device under different voltages, (b) LETD repeatability diagram under 7V voltage and, (c) Similarity percentage diagram of fingerprint images.

5. Conclusion

Recently the perovskite family of materials has shown a promising role in optoelectronic devices due to their interesting properties. An important characteristic of this class of materials is the possibility of controlling the properties of the fabricated device. Control over device parameters is crucial for innovative and unconventional applications. Resolution in LEDs is important for applications such as micro-displays, fabrication of fine illuminating patterns, and also emissive touch devices. The resolution of the LED mostly depends on the grain size of the emissive parts. In perovskite materials, these emissive parts are microcrystals. This study shows that the shape and size of these crystals can be controlled by the coating method and the parameters used during deposition of the emissive layer. The novel trowel-coating method offers several advantages when high-density, low-defect, and high-resolution LEDs are required. In addition, it minimizes material consumption, which is important for large-scale production. The main idea in this research was to take advantage of such a coating method and demonstrate its ability to fabricate an LED suitable for emissive touch-sensitive applications.

However, it should be noted that while the trowel-coating method demonstrated excellent control over film uniformity and resolution at the laboratory scale, challenges may arise in scaling up this technique for industrial production. Issues such as reproducibility, process automation, and integration into roll-to-roll manufacturing lines need to be further investigated. Future work should address these aspects to assess the commercial viability of the proposed approach.

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