Solution-processed silver nanowire (AgNW) thin films were designed to function as transparent electrode to replace indium tin oxide (ITO) electrode. Various times spin-coated AgNW films on substrate were fabricated and optimized according to transmission and sheet resistance $R_s$ of electrode film. The optimized solution-processed AgNWs were further transferred and imprinted on flexible substrate (PEN) which can improve elastic modulus and mechanical properties of the electrode. Moreover, the modified AgNW electrode will possess strong endurance ability against resonant cleaning process and preprocess work. The power conversion efficiency (PCE) of flexible ITO-free perovskite solar cells based on AgNW electrode can reach 9.15%.

The development of highly conductive transparent Conductive Oxide (TCO) is a goal for future optoelectronics technologies, such as wearable electronic devices, organic light-emitting diodes (OLEDs), and solar cells. ITO electrodes are widely used in a variety of applications due to its excellent conductivity and high transparency. However, these electrodes are not cost-effective due to its limited flexibility and high price. Therefore, other effective techniques for ITO alternative have become intense investigation for applications as transparent electrodes in these areas.1–4

In the past, highly conductive PEDOT:PSS, thin metals, metal grids, metal oxide thin films, graphene, carbon nanotubes, nanotube–polymer composites, and silver AgNW meshes are being developed.5–17 Although some research groups have successfully demonstrated well-performed devices by substituting the ITO with other transparent conducting electrode on hard substrate (glass), the performance of devices on flexible substrate are generally low.6–10 This study develops a cost-effective and convenient approach to fabricate ITO-comparable transparent electrodes by utilizing nanoimprint process to combine AgNWs with PEN substrate. PEN has good chemical stability of the organic solution, and has excellent thermal properties better than other plastic substrates due to the increase of the aromatic ring of naphthalene molecules. Furthermore, PEN has stable mechanical properties, even at high temperature and high pressure conditions. The imprinted AgNW electrode was further applied in the renewable solar cells. In this work, we have demonstrated a flexible ITO-free perovskite solar cells based on imprinted AgNW electrode with the PCE of 9.15%.

### Experimental

First, the AgNW thin film (Blue Nano, Inc., NC) with diameters (~35 nm) and lengths (15–30 μm) was spin coated on the silicon wafer at 2000 rpm for 30 seconds, multiple times. Depending on the number of times the thin AgNW layer was spin-coated. Therefore, the five times spin-coated AgNW thin film was chosen for the next step due to the higher Haacke’s figure of merit (F.O.M.) value of 16.6 (10−8Ω−1), which is close to that of ITO.

To improve the overlap regions of the electrode, AgNW thin film was annealed at 120°C for 10 minutes. Finally, after utilizing the nanoimprint surface treatment, the five times AgNW thin film layer was transferred to the PEN substrate. The detailed parameters for nanoimprinting transfer process is shown in Fig. S1. To observe the surface condition and morphology of the AgNW thin film, scanning electron microscope (SEM) and atomic force microscopy (AFM) top view images of AgNW transparent electrodes were taken.

Fig. 1a shows the SEM top view images of the five times spin-coated AgNW electrode used in this work. In Fig. 1a, AgNWs have almost covered the whole substrate, and therefore the density of AgNWs in PEN substrate is very high. It can be also observed from the AFM image as shown in Fig. 1b. It is noticed that the roughness of the electrode has decreased as we increased the number of times the thin AgNW layer was spin-coated. Therefore, the five times spin-coated AgNW imprinted electrode shows the best uniformity in our experiments.

The layer structures of the flexible ITO-free perovskite solar cells consisted of PEN substrate /AgNWs /PEDOT:PSS / PbI2 + MAI /PC60BM/C60 /BCP /Al as shown in Fig. 2a. The detailed fabrication process for perovskite solar cells is shown in Fig. S2. For comparison, another flexible perovskite solar cell based on ITO electrode (PET substrate) was also prepared. Fig. 2b is the side view photo of the flexible ITO-free perovskite solar cell. It can be observed that the device based on AgNW electrode possess high flexibility, which is due to the great mechanical properties of the AgNW electrode.

The electrical characteristics of the devices were measured in the glove box. To measure the efficiency of the solar cells, the devices were illuminated at 100 mW/cm² from a 150W Oriel solar simulator, using an air mass 1.5 global (AM 1.5G) filter to obtain the current density–voltage (J–V) curve.

### Results and Discussion

Table I summarizes the optical and electrical performance of the ITO and AgNW thin film, such as the transmission at the wavelength of 550 nm, sheet resistance ($R_s$), and figure of merit. Compared to the properties of commercial flexible ITO electrode, five times spin-coated AgNW (x5) thin film has lower F.O.M. value but better $R_s$ value.

Table II lists the optimized device performance of perovskite solar cell based on AgNW thin film and ITO electrode including short circuit current $J_{sc}$, open circuit voltage $V_{oc}$, fill factor (FF), and conversion efficiency $\eta$. The flexible ITO-free perovskite solar cells based on five times spin-coated AgNW (x5) electrode exhibit the highest PCE of 9.15% which is close to that of devices based on ITO with the PCE value of 9.29%. It is noteworthy that Mechanical bending of the devices had also been measured and the device performance has decreased by 8% when a bending radius of 1 mm was used.

To replace ITO electrode, transmission and $R_s$ of electrode layer are very critical. Although the AgNW thin film exhibits relatively low transmission compare to ITO electrode, the $R_s$ of AgNW electrode...
Figure 1. (a) SEM top view images of the five times spin-coated AgNW electrode. (b) AFM image of the five times spin-coated AgNW thin film.

Figure 2. (a) Schematic diagrams of the flexible ITO-free Perovskite device structures. (b) The side view images of the perovskite solar cell.

Figure 3. J–V curve of the flexible perovskite solar cells with different electrodes.

Figure 4. EQE spectra of flexible AgNW-based flexible perovskite solar cell and flexible ITO-based perovskite solar cell.

Summary

Imprinted AgNW transparent electrode that possesses high transmission and low Rs value has been developed. Flexible perovskite solar cell based on five times spin-coated AgNW imprinted electrode was fabricated. The PCE of the flexible ITO-free perovskite solar cells is also relatively lower than ITO layer. Hence, the performances of both devices are similar. To further clarify the optical effect of the transparent electrodes, the external quantum efficiency (EQE) of the devices was also measured as shown in Fig. 4. The theoretical Jsc of the devices can be calculated via the EQE spectra. The slight difference between Jsc obtained from the EQE and PCE results is due to the difference in the light source used in the measurement equipment.

It is observed that the ITO-based device exhibits higher EQE at shorter wavelengths. Since the ITO transparent electrode shows higher transmission below the wavelength of 600 nm, this allows the device to absorb more light in the shorter-wavelength region. This result is well match with our previous work, which is using AgNW thin films as electrode to fabricate polymer solar cells.

## Table I. Figure of merit of ITO and AgNW film.

| film            | Transmission at 550 nm (%) | Rs (Ω/sq) | F.O.M. (10^{-2}Ω^{-1}) |
|-----------------|---------------------------|-----------|------------------------|
| AgNW (x3)       | 91.4                      | 24.35     | 16.70                  |
| AgNW (x4)       | 85.3                      | 15.45     | 13.19                  |
| AgNW (x5)       | 83.2                      | 9.54      | 16.64                  |
| ITO-PET         | 93.8                      | 20.11     | 24.06                  |

## Table II. The device performance of perovskite solar cells.

| Device | V_{oc} (V) | J_{sc} (mA/cm²) | FF | Rs (Ω/cm²) | Rsh (Ω/cm²) | PCE | η (%) | bending radius (mm) |
|--------|------------|-----------------|----|------------|-------------|-----|-------|---------------------|
| AgNW   | 14.90      | 63.9            | 7.6| 792        | 1088        | 9.15| 8.51  | 1mm                 |
| ITO-PET| 16.07      | 69.6            | 4.3| 1088       | 929         | 8.51| 8.73  |                     |
can reach 9.10%, which is close to ITO based perovskite solar cells with efficiency of 9.29%.

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ORCID

Ming Yi Lin https://orcid.org/0000-0002-2229-7074

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