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Research Article

Development of Indium Tin Oxide Stack Layer Using DC and RF Sputtering for Perovskite Solar Cells

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Abstract: Indium tin oxide (ITO) films were used as transparent conductive oxide (TCO) layers in perovskite solar cells (PSCs). Generally, the commercial ITO film-coated glass substrate has good electrical properties and a film thickness of around 340 nm but low transmittance in long wavelength regions of 750-1200 nm. The enhancement of light transmittance by the ITO stack layer was developed. Direct current (DC) sputtering with the oxygen-argon (O_2-Ar) gas mixture deposited the first ITO layer on a soda-lime glass substrate. Radio frequency (RF) sputtering with the Ar gas deposited in the second ITO layer. By optimizing the deposition conditions, the ITO stack layer has a total film thickness of around 330 nm, with a higher light transmittance of about 24% in the wavelength range of 750-1200 nm compared to the reference ITO (commercial). By using the ITO stack layer as a substrate for perovskite solar cells, the ITO stack layer structure of the DC:RF (1:1) has the highest efficiency compared to ITO film deposited from DC, RF, and reference ITO, which mainly from the increasing short current density (J_{sc}) of the solar cell. Applying an ITO stack layer for a perovskite solar cell achieved a power conversion efficiency of 14.9%.

Keywords: Indium tin oxide, ITO stack layer, Perovskite solar cell

1. Introduction

Perovskite is an attractive structure for solar cell technology. One of the perovskite properties is a band gap tunability of around 1.5-2.0 eV, in which an absorption wavelength range is 300-800 nm depending on the chemical composition [1-4]. Thus, perovskite has the potential for single and tandem solar cell structures [5-6]. Perovskite film can be prepared by solution and a low-temperature process of around 150 °C. Perovskite solar cells can fabricate on glass and flexible substrate [7]. Besides perovskite solar cell fabrication, transparent conductive oxide (TCO) glass substrate is used for the electrode and substrate. The properties of TCO should have high transparency and low resistivity to enhance the short current density (J_{sc}) and solar cell performance. Indium tin oxide (ITO) is generally used as a TCO for perovskite solar cells. The commercial ITO film has a thickness of around 340 nm and sheet resistance of around 5 Ω/sq. The transmittance value is more than 75% at a wavelength of 550 nm, but the transmittance value trend decreases in long-wavelength regions. For the perovskite/silicon tandem solar cell structure, due to ITO film being used in perovskite solar cells as the top cell, the ITO film might affect low-light transmission from the perovskite top cell to the silicon bottom cell. Moreover, commercial ITO cannot improve or modify the optical or electrical

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properties. Usually, ITO films are prepared by using physical vapor deposition (PVD) sputtering techniques. The films' properties depend on the deposition conditions, such as temperature, power, and gas pressure. In this study, the ITO stack layer coated glass substrate was deposited and investigated properties for use as electrode and substrate in single-cell perovskite solar cells.

2. Methodology

Preparation of ITO film

The ITO stack layer was deposited on the soda-lime glasses thickness of 1.1 mm as substrate and using direct current (DC) and radio frequency (RF) 13.56 MHz magnetron sputtering technique with an ITO target of In_2O_3:SnO_2 (95:5 wt.%). The soda-lime glasses substrate was prepared with 2.5 cm × 2.5 cm, cleaned with deionized water (DI) water/acetone/IPA (isopropyl alcohol) in an ultrasonic cleaner, respectively, and dried with N_2. In the ITO stack layer deposition sequence, the first ITO layer was deposited using the DC sputtering method with an Ar-O_2 gas mixture, and the second layer was deposited by RF sputtering with Ar gas. The total thickness of the ITO stack layer film was around 330 nm. The structure of the ITO stack layer, the thickness of the first layer (DC) and second layer (RF) film was varied with DC:RF film thickness ratios of 1:3 (85 nm:255 nm), 1:1 (170 nm:170 nm), and 3:1 (255 nm:85 nm). Besides, the conventional ITO films were deposited from DC and RF sputtering as the same thickness to compare film properties. The details of the ITO film deposition condition are shown in Table 1. The properties of ITO films were analyzed by a UV-Vis-NIR spectrophotometer, a Hall effect measurement, and a field emission scanning electron microscope.

Table 1 Deposition conditions for ITO films

Parameters	Value
Power of radio frequency 13.56 MHz (W)	250
Power of direct current power supply (W)	250
Substrate temperature (°C)	180
O_2/Ar	3.4%
Thickness (nm)	330

Fabrication of perovskite solar cells

The perovskite solar cells were fabricated on the ITO stack layer coated glass as the substrate and front electrode. The ITO stack layer coated glass substrate was prepared and cleaned by DI water/acetone/IPA/UV-ozone. The schematic structure of the perovskite solar cells used to verify the effect of the ITO stack film on the photovoltaic (PV) parameters is shown in Figure 1.

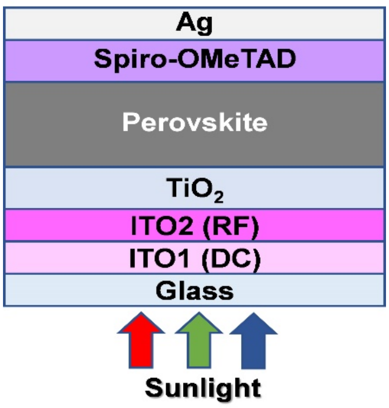

Figure 1 Schematic structure of the perovskite solar cells with the ITO stack layer.

The perovskite solar cells were fabricated by the spin coating method. The TiO_2 (0.3 M Titanium diisopropoxide bis (acetylacetonate) in butanol solution was coated on ITO stack layer glass substrate and heated at 450 °C for 30 min. Perovskite solution $Cs_{0.05}(FA_{0.83}MA_{0.17})_{0.95}Pb(I_{0.83}Br_{0.17})_3$ was coated on a TiO_2 layer, then heated at 100 °C for 30 min. The Spiro-OMeTAD layer was deposited onto the perovskite layer. Finally, the deposition of silver (Ag) metal back contact was deposited by thermal evaporation. The total solar cell area was 0.18 cm^2. The current-voltage (I-V) characteristics and photovoltaic (PV) parameters of the solar cells were measured under standard conditions (AM 1.5, 100 mW/cm^2, 25 °C) using a xenon and halogen light source solar simulator.

3. Results and discussion

The optical and electrical properties of the ITO stack films were studied. The first ITO layer was deposited by DC sputtering with an O_2/Ar gas mixture ratio of 3.4%, and the second ITO layer was deposited by RF sputtering with an Ar gas. Figure 2 shows the transmittance of the ITO stack layer with various DC:RF film thickness ratios of 1:3, 1:1, and 3:1. At wavelengths below 400 nm, the ITO stack layer at DC:RF ratio of 1:3 has higher transmittance than at ratios of 1:1 and 3:1. Besides wavelengths of 400-1200 nm, ITO stack layer film with various DC:RF ratio show fluctuation transmittance values between 75-90%.

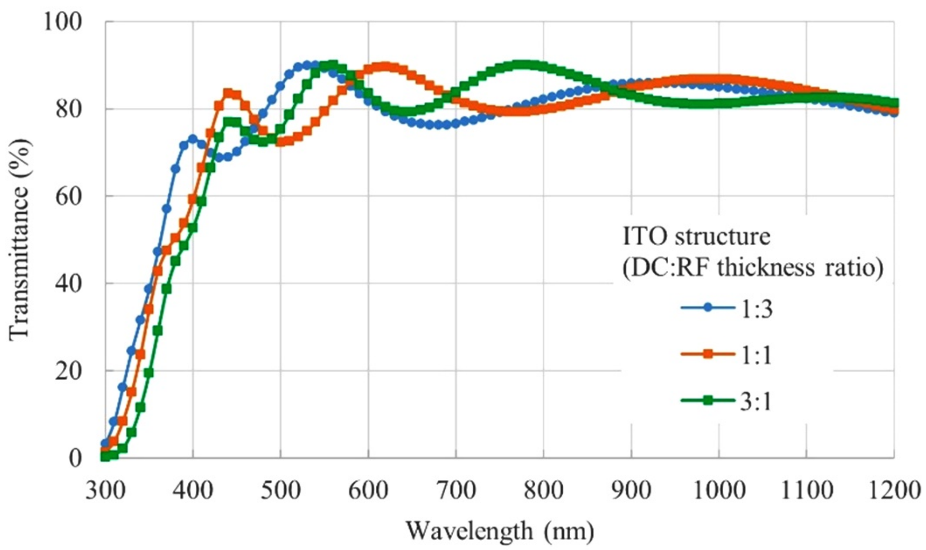

Figure 2 The transmittance of the ITO stack layer with various DC:RF film thickness ratios of 1:3, 1:1, and 3:1.

Figure 3 shows the transmittance of the ITO stack layer film (DC:RF 1:3) with various ITO film structures (reference, DC, and RF). At wavelengths below 400 nm, the reference ITO (commercial) film had the highest transmittance. Meanwhile, the ITO film stack layer (DC:RF 1:3) has a high transmittance as the ITO film deposited by RF sputtering. Furthermore, the transmittance of a wavelength around 800-1200 nm is higher than the ITO film deposited from other methods. Figure 4 shows variations in the resistivity, carrier density, and Hall mobility of the ITO films with various structures. The resistivity of the ITO stack layer was increased from 5.23×10^{-4} Ω·cm to 1.20×10^{-3} Ω·cm with the DC:RF ratio from 1:3 to 3:1. While the ITO film deposited by DC sputtering technique has the highest resistivity of 1.20×10^{-3} Ω·cm. Meanwhile, the resistivity of the ITO film deposited by RF sputtering was 4.42×10^{-4} Ω·cm, and the reference ITO film had the lowest resistivity of 1.33×10^{-4} Ω·cm. The carrier density and Hall mobility of the reference ITO films show the highest value. Figure 5 presents the surface morphology of the ITO film with various film structures. The ITO film deposited by DC and RF has a rough surface with a grain size of about 0.1 μm and 0.2 μm, as shown in Figure 5(d-e), respectively. The ITO film stack layer in Figure 5(a-c) has surface roughness, and grain size is the same as that of the DC sputtering film because the ITO second layer deposited by RF sputtering has a thin layer and a small columnar structure, while the reference ITO surface has a large grain size of about 0.4 μm, as shown in Figure 5(f).

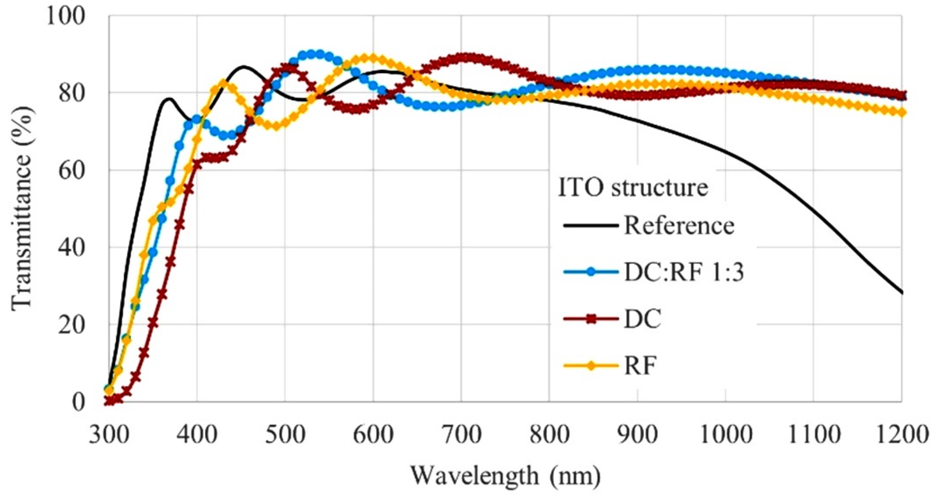

Figure 3 Transmittance of the ITO stack layer film (DC:RF 1:3) with various ITO film structures.

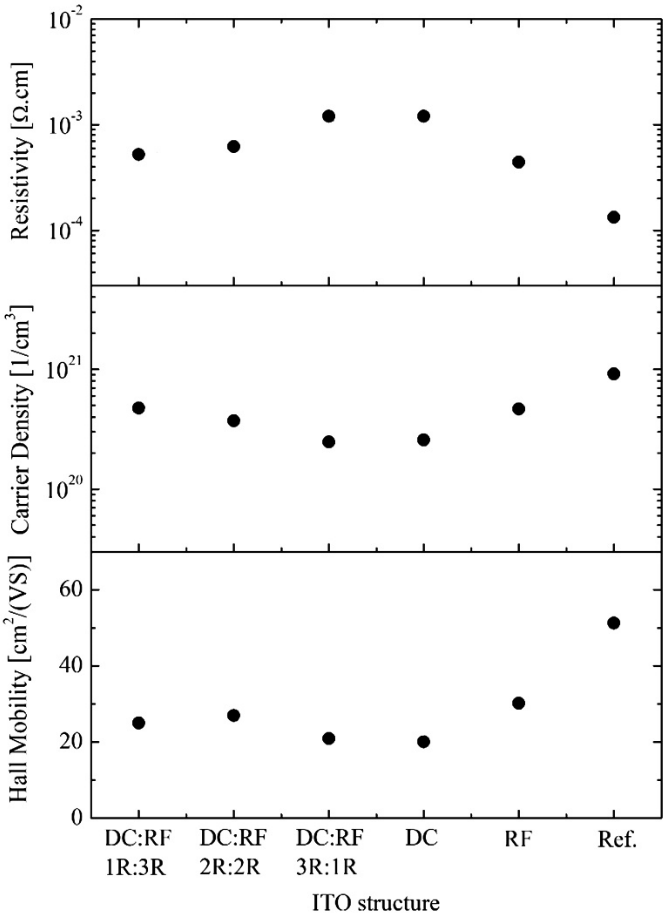

Figure 4 Variation of the resistivity, carrier density, and Hall mobility of the ITO films as a function of film structures.

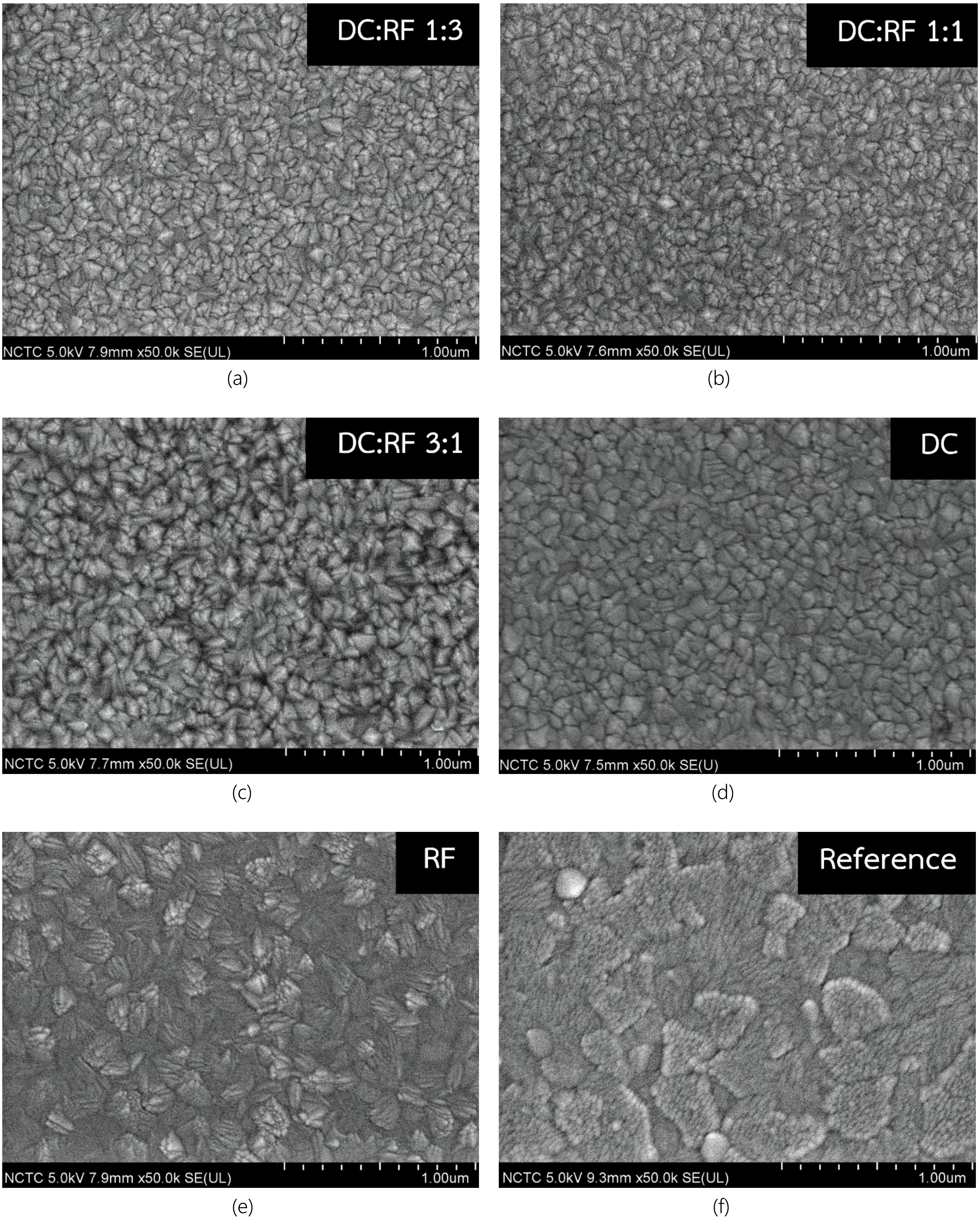

Figure 5 SEM images of ITO films with various O_2/Ar ratios of (a) 0, (b) 3.4, (c) 7.1, (d) 11.1, and (e) stack layer.

The effect of ITO film structure on the perovskite solar cells' performance was investigated. Perovskite solar cells were fabricated on ITO-coated glass with various film structures compared to the reference ITO glass substrate. The condition for fabricating all perovskite solar cells was soda-lime glass/ITO/TiO_2/perovskite/Spiro-OMeTAD/Ag. Figure 6 plots the J_{sc} and the open-circuit voltage (V_{oc}). The ITO stack layer of DC:RF 1:1 has a higher J_{sc} value than other structural ITO films and results in the highest solar cells efficiency. Detailed photovoltaic parameters are shown in Table 2. However, the efficiency of the solar cell depends on the optimization conditions for each ITO glass substrate.

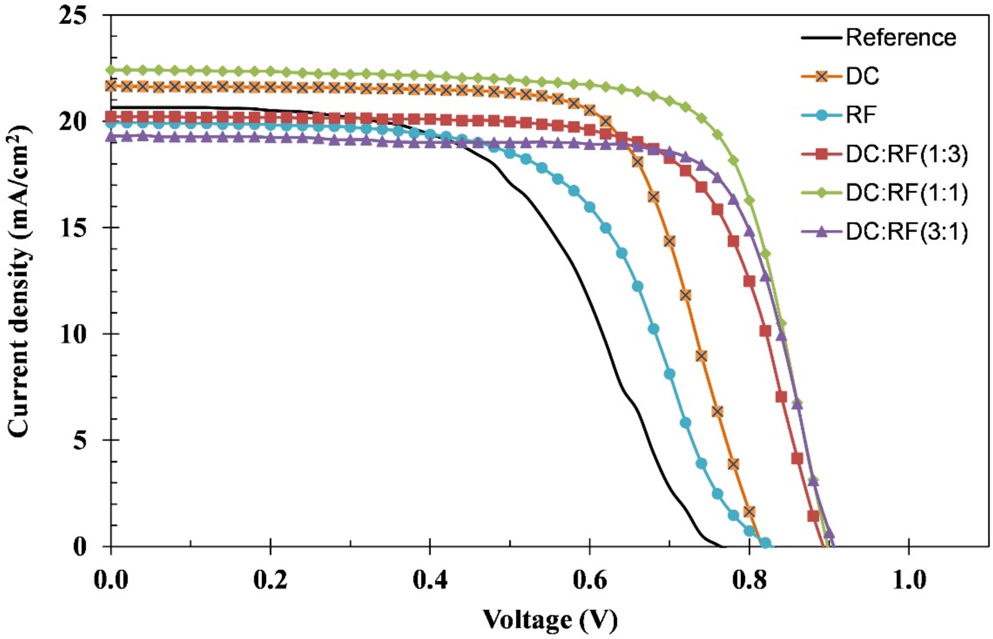

Figure 6 I-V characteristics of c-Si-HJ solar cells with different ITO film deposition.

Table 2 Parameters of perovskite solar cells with different ITO film structures

ITO glass substrate	V_{oc} (V)	J_{sc} (mA/cm^2)	FF	Eff (%)
Reference	0.76	20.6	0.55	8.6
DC	0.82	21.7	0.70	12.4
RF	0.83	20.0	0.59	9.7
DC:RF (1:3)	0.89	20.2	0.71	12.8
DC:RF (1:1)	0.90	22.4	0.74	14.9
DC:RF (3:1)	0.91	21.7	0.70	13.3

4. Conclusion

The ITO stack layer coated glass can increase the light transmittance in the wavelength range of 750-1200 nm compared to the reference ITO (commercial) with a thickness of about 330 nm. The ITO stack layer film can be used as substrate and transparent electrodes for perovskite solar cells. The perovskite solar cells using the ITO stack layer (DC:RF 1:1) have higher efficiencies than DC, RF, and reference ITO (commercial) under the same fabrication conditions due to the increasing J_{sc}, corresponding to the increased light transmittance value. The perovskite solar cells that use the ITO stack layer (DC:RF 1:1) showed the highest efficiency of 14.9% (V_{oc} = 0.9 V, J_{sc} = 22.4 mA/cm^2, FF = 0.74).

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