



The 25th Iranian Conference on
Optics and Photonics (ICOP 2019),
and the 11th Iranian Conference on
Photonics Engineering and
Technology (ICPET 2019).
University of Shiraz,
Shiraz, Iran,
Jan. 29-31, 2019



بررسی مقایسه‌ای اثر ضخامت لایه‌ی اکسید آلومینیوم در سلول‌های خورشیدی پروسکائیتی

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چکیده - در این مقاله اثر نشان دادن ضخامت‌های مختلف لایه‌ی اکسید آلومینیوم روی لایه‌ی متخلخل انتقال دهنده الکترون (mp-TiO₂) در خواص فتوولتاییکی سلول‌های خورشیدی پروسکائیتی بررسی شده است. در این کار از روش کندوپاش مغناطیسی چرخشی برای نشان دادن لایه‌ی اکسید آلومینیوم استفاده شده است. سلول‌های خورشیدی ساخته شده دارای ساختار رایج سلول افزایش می‌یابد؛ چگالی جریان کاهش می‌یابد. به دلیل عایق بودن اکسید آلومینیوم، کاهش جریان سلول با افزایش ضخامت اکسید، ولتاژ مدار باز سلول افزایش می‌دهد؛ چگالی جریان کاهش می‌یابد. به دلیل عایق بودن اکسید آلومینیوم، کاهش جریان سلول با افزایش ضخامت اکسید آلومینیوم دور از انتظار نیست. مقایسه سلول‌های خورشیدی دارای سه ضخامت مختلف از اکسید آلومینیوم، نشان می‌دهد؛ نمونه‌های حاوی ضخامت ۱۵ نانومتر بهترین مشخصات فتوولتاییکی را از خود نشان می‌دهند.

کلید واژه - اکسید آلومینیوم، خواص فتوولتاییکی، سلول خورشیدی پروسکائیتی

Comparative study of perovskite solar cells with different Al₂O₃ thicknesses

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Abstract- The effect of the Al₂O₃ thin film on photovoltaic characteristics of perovskite solar cells is studied. The thin film of the Al₂O₃ is deposited on mesoporous TiO₂ by a rotational angular reactive sputtering method. The considered perovskite solar cells have the normal architecture of FTO/c-TiO₂/mp-TiO₂/CH₃NH₃PbI₃/spiro-OMeTAD/Au. The increase of Al₂O₃ thickness leads to higher open circuit voltage. However, the current densities of the fabricated solar cells are reduced due to insulating characteristics of Al₂O₃. Here, solar cells with 15 nm thickness of Al₂O₃ present the best performance among the three different thicknesses studied.

Keywords: perovskite solar cells, reactive magnetron sputtering, photovoltaic characteristics

1. Introduction

Perovskite solar cells (PSCs) composed of organic-metal-halide structures have achieved an undeniable breakthrough in photovoltaic development. The power conversion efficiency (PCE) of the perovskite solar cells has rapidly jumped from 3.8% [1] to certified 23.3% [2]. The general formula of the commonly used perovskite is ABX_3 containing an organic cation A, such as methylammonium (MA) or formamidinium (FA), a divalent metal B, such as Pb or Sn, and a halide X, such as bromine or iodine.

The impressive PCE increase can be attributed to their superior optoelectronic properties, including strong absorption coefficient ($\sim 10^5 \text{ cm}^{-1}$), low exciton binding energy ($\sim 20 \text{ meV}$), and relatively long carrier diffusion length ($> 1 \mu\text{m}$). In addition, a wide range of solution processing techniques, including one-step deposition, two-steps sequential deposition, solvent-quenching, and other modified approaches based on these methods have been developed to fabricate uniform and high-crystalline perovskite films, resulting in the fast-rising in the device performance [3].

Although PSCs have made impressive progress in just a few years, there is still a long way to their commercialization. A key factor to further improve the device performance is to reduce nonradiative recombination processes. One of the commonly used structures for PSCs is FTO glass/ compact- TiO_2 (c- TiO_2)/ mesoporous- TiO_2 (mp- TiO_2)/Perovskite (MAPbI_3)/ hole transport material (HTM)/ Au. It is known that the surface passivation of potential recombination sites in mp- TiO_2 interface suppresses the interfacial recombination. Several metal oxides such as SiO_2 , ZrO_2 , MgO , Al_2O_3 are recognized as efficient capping materials in the solid-state dye-sensitized solar cells (DSCs) [4].

Here, we introduce a thin Al_2O_3 layer on mp- TiO_2 by the rotational angular reactive magnetron sputtering (RMS) deposition process in order to boost PSCs photovoltaic performance.

2. Experimental Section

FTO coated glass substrates are patterned by Zn powder and 2M HCl etching solution. The substrates are brushed vigorously without scratching FTO surface and they are cleaned ultrasonically for 10 minutes in detergent, deionized water, and isopropanol. After cleaning, UV ozone or plasma cleaning is used for 15 min right before depositing the TiO_2 . The hole blocking compact and mesoporous TiO_2 layers are deposited by spin-coating respectively and then annealed at 500°C for 30 min [5]. Mesoporous thin film Al_2O_3 layer is deposited on mp- TiO_2 using rotational angular reactive DC magnetron sputtering. The chamber is evacuated to $\sim 4 \times 10^{-4}$ Torr. Prior to sputter deposition, Ar and O_2 gas are introduced via separate mass-flow-controlled inlets. The perovskite layer is prepared by a two-step spin-coating method. A 1 M PbI_2 solution is dropped wisely spin-coated. After drying on a hotplate, the solution of $\text{CH}_3\text{NH}_3\text{I}$ in isopropanol was spin coated on PbI_2 coated substrate. Spiro-OMeTAD solution doped with tBP and LiTFSI is spin-coated at 4000 rpm. The devices are left sealed overnight and then finished with 80nm thick gold contacts deposited on top via thermal evaporation.

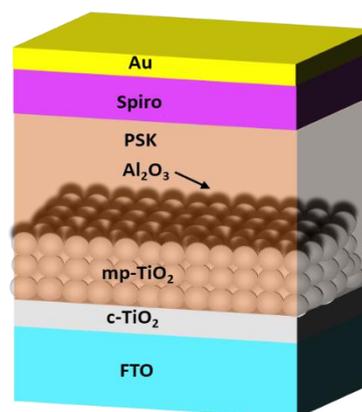


Fig. 1: Perovskite solar cells Schematic

3. Results and Discussion

The FE-SEM image of thin Al_2O_3 layer sputtered on mp- TiO_2 is shown in Fig. 1. Comparing the two top layer images, it is possible to interpret that the porosity of the upper layer is conserved after the deposition of Al_2O_3 layer. Thus, the perovskite solution still can penetrate the mp- TiO_2 layer,

reserving high surface contact with mp-TiO₂ that facilitates the electron extraction in mesostructure perovskite solar cells.

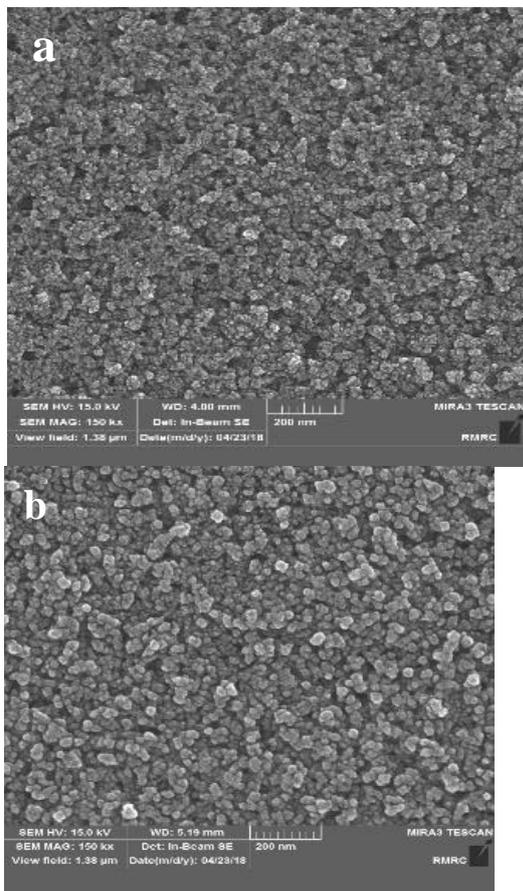


Fig. 1: FE-SEM image of a) mp-TiO₂ b) mp-TiO₂/Al₂O₃

The sunlight illuminates the cell from the FTO side, through the wide bandgap Al₂O₃ layer. Hence, no optical absorption takes place by wide bandgap Al₂O₃. As a result, the aforementioned passivation layer causes no disturbance in the photogeneration of electron-hole pairs, while enhancing the photovoltaic characteristics of the cells.

Table 1 illustrates the photovoltaic properties of solar cells consisting of Al₂O₃ with different thicknesses of 5, 15, 25 nm. By increasing the thickness of insulating Al₂O₃ with the band gap of 6.6-8.8 (eV) (for bulk crystalline), the amount of open circuit voltages increases, on average. Al₂O₃ layer in the solar cell stack passivates possible recombination centers in the mp-TiO₂ interface.

Table 1. Photovoltaic properties of solar cells with a different thickness of Al₂O₃

Al ₂ O ₃ Thickness (nm)	V _{oc} (V)	J _{sc} (mA/cm ²)	PCE (%)
0	0.91	17.0	8.5
5	0.92	13.8	7.0
15	0.94	13.7	8.9
25	0.95	10.6	7.8

Although perovskite deposition methods have been modified to make a perfect pinhole free perovskite layer, they lead to layers with pinhole and roughness undeniably. Therefore, it is likely for HTM to be in direct contact with mp-TiO₂ through perovskite pinholes that cause non-radiative recombination sites. An optimum thickness of Al₂O₃ can block the intimate contact of charge transfer layers and enhances open circuit voltage of devices. In addition, with a constant photogeneration, the thermal recombination is decreased due to the passivation layer, leading to a reasonable optical efficiency for the optimum thickness.

Since the used metal oxide layer has insulating characteristics, current density decreases with the increase of the metal oxide thickness. A trade-off is made between open circuit voltage enhancement and Al₂O₃ thickness.

Figures 2-4 show the open circuit voltage, short circuit current density and the power conversion efficiency versus the Al₂O₃ thickness. One can see that as the Al₂O₃ thickness increases, the open circuit voltage increases while the current density decreases and PCE fluctuates. Nonetheless, these data show that the 15-nm Al₂O₃ thickness is the best choice for cells, benefiting from higher open circuit voltages, while not losing much of the current density and PCE.

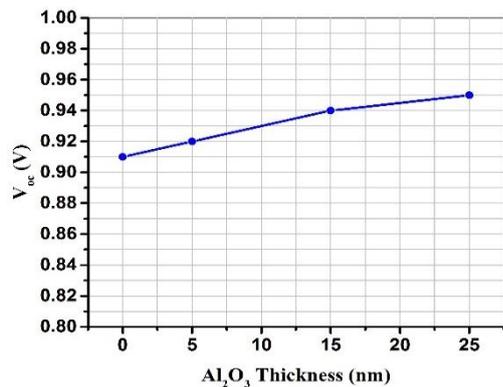


Fig. 2: Open-circuit voltage of the cells with different Al₂O₃ thicknesses

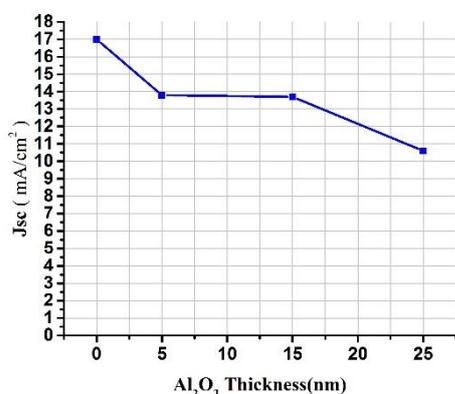


Fig. 3: Current density of the cells with different Al₂O₃ thicknesses

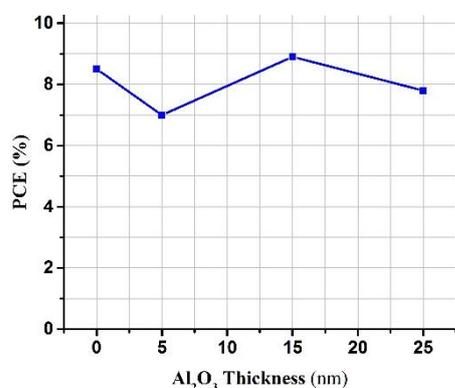


Fig. 4: Power conversion efficiency of the cells with different Al₂O₃ thicknesses

Current-voltage characteristics of solar cells with 15 nm Al₂O₃ thick is shown in Fig. 5. Using the metal oxide the open-circuit voltages improve. At the same time, it reduces the current density. There should be an efficient point in the increasing process of the passivation layer which results in higher voltages and a reasonable amount of current density decrease as well.

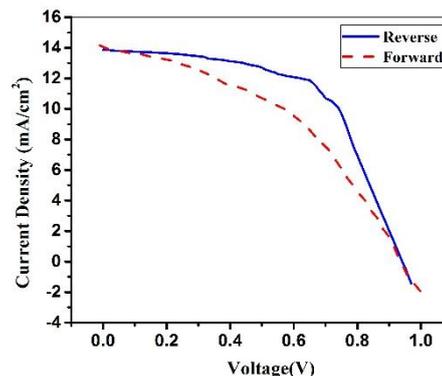


Fig. 5: Current-voltage characteristics of solar cells containing 15nm Al₂O₃

Conclusion

We used the reactive magnetron dc sputtering method for deposition of Al₂O₃ layer in order to enhance V_{OC} in perovskite solar cells. The top layer FE-SEM of the layer is shown and the photovoltaic characteristics of the cells with different Al₂O₃ thicknesses were discussed.

Acknowledgment

The authors would like to acknowledge the financial support received from Tarbiat Modares University through grant #IG-39703.

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