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

Shiraz University

بررسی مقایسه‌ای اثر ضخامت لایه‌ی اکسید آلومینیوم در سلول‌های خورشیدی پروسکایتی

عاطفه قربانی کل‌تپه، بهرام عبدالهی نژند، محمدکاظم مروج فرشی[1]

[1]دانشکده مهندسی برق و کامپیوتر، هسته پژوهشی نانو پلاسموفوتونیک، دانشگاه تربیت مدرس، تهران، ایران

چکیده- در این مقاله اثر نشاندن ضخامت‌های مختلف لایه‌ی اکسید آلومینیوم روی لایه‌ی متخلخل انتقال دهنده الکترون (mp-TiO2) در خواص فتوولتاییکی سلول‌های خورشیدی پروسکایتی بررسی شده است. در این کار از روش کندوپاش مغناطیسی چرخشی برای نشاندن لایه‌ی اکسید آلومینیوم استفاده شده‌است. سلول‌های خورشیدی ساخته شده دارای ساختار رایج FTO/c-TiO_2/mp-TiO_2/$CH_3NH_3PbI_3$/spiro-OMeTAD/Au اند. علی رغم این‌که با افزایش ضخامت لایه‌ی اکسید، ولتاژ مداربار سلول افزایش می‌یابد؛ 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 پروسکایتی

Comparative study of perovskite solar cells with different Al_2O_3 thicknesses

A. Ghorbani Koltapeh , B. Abdollahi Nejand , M. K. Moravvej-Farshi*

Faculty of Electrical and Computer Engineering, Nano Plasmo-Photonic Research Group, Tarbiat Modares University, Tehran, Iran
*moravvej@modares.ac.ir

Abstract- The effect of the Al_2O_3 thin film on photovoltaic characteristics of perovskite solar cells is studied. The thin film of the Al_2O_3 is deposited on mesoporous TiO_2 by a rotational angular reactive sputtering method. The considered perovskite solar cells have the normal architecture of FTO/c-TiO_2/mp-TiO_2/$CH_3NH_3PbI_3$/spiro-OMeTAD/Au. The increase of Al_2O_3 thickness leads to higher open circuit voltage. However, the current densities of the fabricated solar cells are reduced due to insulating characteristics of Al_2O_3. Here, solar cells with 15 nm thickness of Al_2O_3 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 (~105 cm^{-1}), low exciton binding energy (~20 meV), and relatively long carrier diffusion length (>1 μm). 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 $500^{\circ C}$ 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 ~ 4×10^{-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 CH_3NH_3I 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.

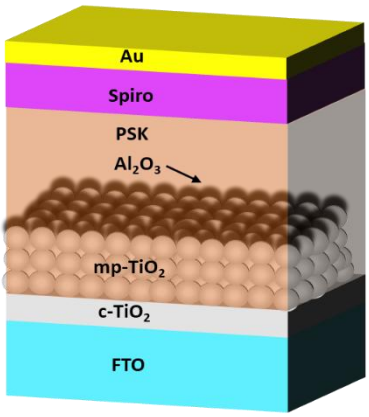

Fig. 1: Perovskite solar cells Schematic

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-TiO_2 that facilitates the electron extraction in mesostructure perovskite solar cells.

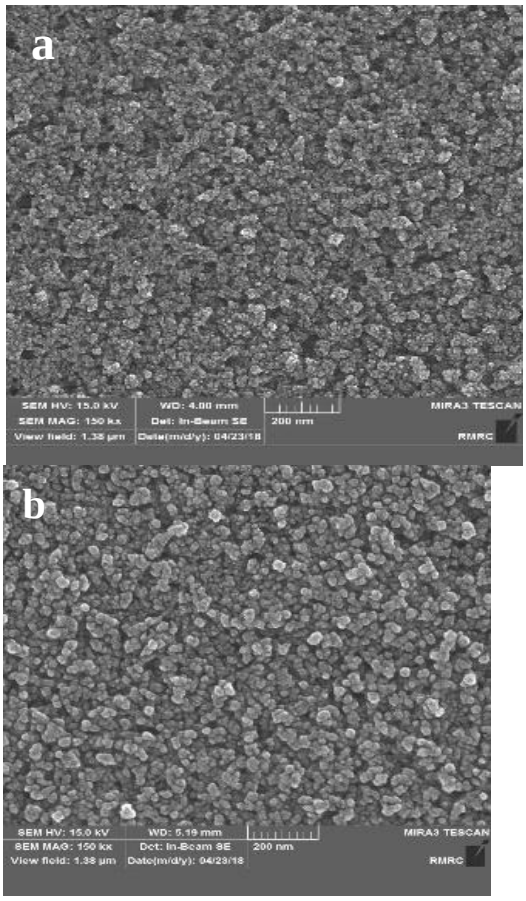

Fig. 1: FE-SEM image of a) mp-TiO_2 b) mp-TiO_2/Al_2O_3

The sunlight illuminates the cell from the FTO side, through the wide bandgap Al_2O_3 layer. Hence, no optical absorption takes place by wide bandgap Al_2O_3. 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 Al_2O_3 with different thicknesses of 5, 15, 25 nm. By increasing the thickness of insulating Al_2O_3 with the band gap of 6.6-8.8 (eV) (for bulk crystalline), the amount of open circuit voltages increases, on average. Al_2O_3 layer in the solar cell stack passivates possible recombination centers in the mp-TiO_2 interface.

Table 1. Photovoltaic properties of solar cells with a different thickness of Al_2O_3

Al_2O_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-TiO_2 through perovskite pinholes that cause non-radiative recombination sites. An optimum thickness of Al_2O_3 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 Al_2O_3 thickness.

Figures 2-4 show the open circuit voltage, short circuit current density and the power conversion efficiency versus the Al_2O_3 thickness. One can see that as the Al_2O_3 thickness increases, the open circuit voltage increases while the current density decreases and PCE fluctuates. Nonetheless, these data show that the 15-nm Al_2O_3 thickness is the best choice for cells, benefiting from higher open circuit voltages, while not losing much of the current density and PCE.

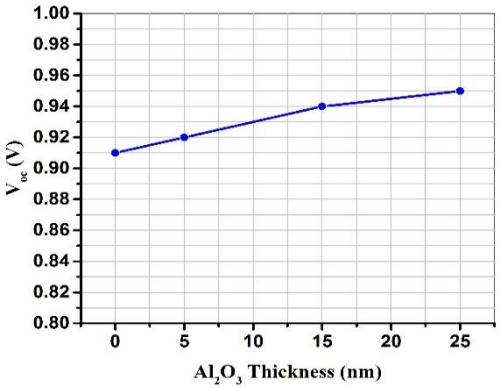

Fig. 2: Open-circuit voltage of the cells with different Al_2O_3 thicknesses

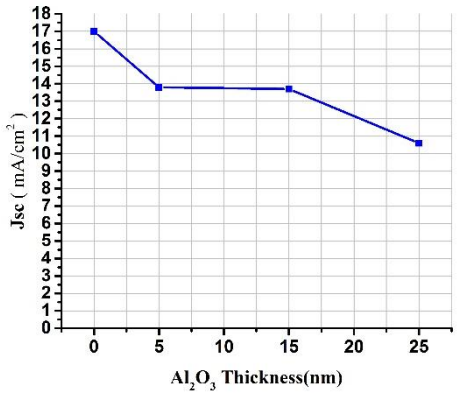

Fig. 3: Current density of the cells with different Al_2O_3 thicknesses

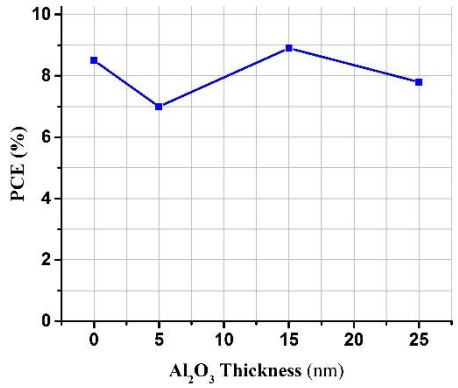

Fig. 4: Power conversion efficiency of the cells with different Al_2O_3 thicknesses

Current-voltage characteristics of solar cells with 15 nm Al_2O_3 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.

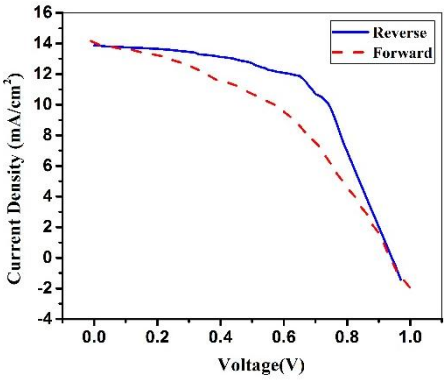

Fig. 5: Current-voltage characteristics of solar cells containing 15nm Al_2O_3

Conclusion

We used the reactive magnetron dc sputtering method for deposition of Al_2O_3 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 Al_2O_3 thicknesses were discussed.

Acknowledgment

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

References

[1] A. Kojima, K. Teshima, Y. Shirai, and T. Miyasaka, "Organometal Halide Perovskites as Visible-Light Sensitizers for Photovoltaic," *J. Am. Chem. Soc*, vol. 131, pp. 6050–6051, 2009.

[2] https://www.nrel.gov/pv/assets/pdfs/pv-efficiencies-07-17-2018.pdf

[3] F. Wang and S. Bai, "Defects engineering for high-performance perovskite solar cells," *npj Flex. Electron.*, vol. 2, no. 22, 2018.

[4] Y. H. Lee *et al.*, "Enhanced Charge Collection with Passivation Layers in Perovskite Solar Cells," *Adv. Mater.*, vol. 28, no. 20, pp. 3966–3972, 2016.

[5] S. Gharibzadeh *et al.*, "Two-Step Physical Deposition of a Compact CuI Hole-Transport Layer and the Formation of an Interfacial Species in Perovskite Solar Cells," *ChemSusChem*, vol. 9, no. 15, pp. 1929–1937, 2016.