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بررسی مقایسهای اثر ضخامت لایهی اکسید آلومینیوم در سلولهای خورشیدی پروسکایتی

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چکیده – در این مقاله اثر نشاندن ضخامتهای مختلف لایهی اکسید آلومینیوم روی لایهی متخلخل انتقـال دهنـده الکتـرون (-mp) در خواص فتوولتاییکی سلولهای خورشیدی پروسکایتی بررسی شده اسـت. در ایـن کـار از روش کنـدوپاش مغناطیسـی چرخشی برای نشاندن لایهی اکسید آلومینیوم استفاده شدهاسـت. سـلولهـای خورشـیدی سـاخته شـده دارای سـاختار رایـج چرخشی برای نشاندن لایهی اکسید، ولتـاژ مـدارباز FTO/c-TiO2/mp-TiO2/CH3NH3PbI3/spiro-OMeTAD/Au اند. علی رغم این که با افزایش ضخامت لایهی اکسید، ولتـاژ مـدارباز 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 Al₂O₃ thicknesses

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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 organicmetal-halide structures have achieved 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 ABX₃ 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-TiO₂ (mp-TiO₂)/Perovskite (MAPbI₃)/ hole transport material (HTM)/ Au. It is known that the surface passivation of potential recombination sites in mp-TiO₂ interface suppresses the interfacial recombination. Several metal oxides such as SiO₂, ZrO₂, MgO, Al₂O₃ are recognized as efficient capping materials in the solid-state dye-sensitized solar cells (DSCs) [4].

Here, we introduce a thin Al₂O₃ layer on mp-TiO₂ 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 TiO₂. The hole blocking compact and mesoporous TiO2 layers are deposited by spin-coating respectively and then annealed at 500°C for 30 min [5]. Mesoporous thin film Al₂O₃ layer is deposited on mp-TiO2 using rotational angular reactive DC magnetron sputtering. The chamber is evacuated to $\sim 4 \times 10^{-4}$ Torr. Prior to sputter deposition, Ar and O2 gas are introduced via separate mass-flow-controlled inlets. The perovskite layer is prepared by a two-step spincoating method. A 1 M PbI2 solution is dropped wisely spin-coated. After drying on a hotplate, the solution of CH₃NH₃I in isopropanol was spin coated on PbI2 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.

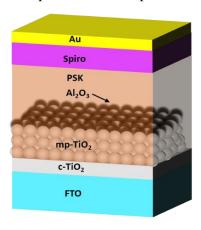


Fig. 1: Perovskite solar cells Schematic

3. Results and Discussion

The FE-SEM image of thin Al_2O_3 layer sputtered on mp-TiO₂ 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-TiO₂ layer,

reserving high surface contact with mp- TiO_2 that facilitates the electron extraction in mesostructure perovskite solar cells.

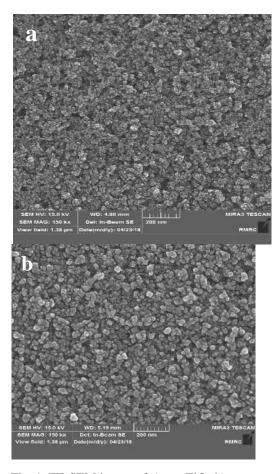


Fig. 1: FE-SEM image of a) mp-TiO₂ b) mp-TiO₂/Al₂O₃

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-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 In addition. devices. 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 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.

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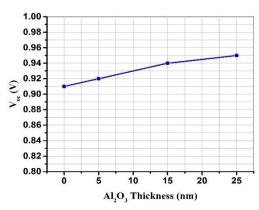


Fig. 2: Open-circuit voltage of the cells with different Al₂O₃ thicknesses

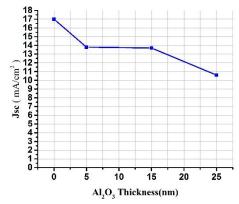


Fig. 3: Current density of the cells with different Al₂O₃ thicknesses

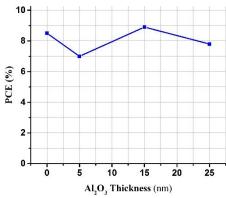


Fig. 4: Power conversion efficiency of the cells with different Al₂O₃ thicknesses

Current-voltage characteristics of solar cells with 15 nm 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.

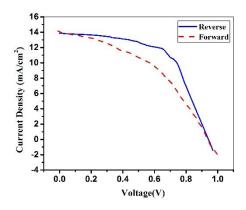


Fig. 5: Current-voltage characteristics of solar cells containing 15nm Al₂O₃

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.

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