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بهبود ولتاژ مدارباز در سلول‌های خورشیدی پروسکایتی با استفاده از لایه‌ی دی اکسید آلومینیوم به عنوان لایه خنثی الکتریکی

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چکیده – در این مقاله اثر نشاندن لایه‌ی دی اکسید آلومینیوم روی لایه‌ی متخلخل انتقال دهنده الکترون ($mp\text{-}TiO_2$) در خواص فتوولتاییکی سلول‌های خورشیدی پروسکایتی بررسی شده است. به دلیل عایق بودن دی اکسید آلومینیوم نیاز است نشاندن لایه به صورت غیر یکنواخت صورت بگیرد و به این منظوراز روش کندوپاش مغناطیسی چرخشی استفاده شده‌است. در این کار، ولتاژ مدار باز سلول‌های خورشیدی پروسکایتی با استفاده از لایه‌ی دی اکسید آلومینیوم و بدون آن مقایسه شده‌است. با استفاده از لایه نازک دی اکسید آلومینیوم، به دلیل کاهش مراکز بازترکیب در ساختار، ولتاژ مدار باز بزرگتری به دست می‌آید، که با نتایج حاصل از مشخصه‌های چگالی جریان-ولتاژ، افت ولتاژ مدار باز و مشخصه جریان تاریک سلول‌های خورشیدی مورد تایید است.

کلید واژه- بازترکیب، دی‌اکسید آلومینیوم، ولتاژمدار باز، سلول های خورشیدی پروسکایت

Enhanced Open-circuit Voltage Using Al_2O_3 Inert Layer in Perovskite Solar Cells

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Abstract- We investigate the effect of Al_2O_3 thin film deposited on an electron transfer layer (ETL: mp-TiO_2) in the perovskite solar cells with the conventional structure of FTO/c-TiO_2/mp-TiO_2/$CH_3NH_3PbI_3$/spiro-OMeTAD/Au. Using the rotational angular deposition method to deposit a nanolayer of insulating Al_2O_3 by the reactive magnetron sputtering (RMS), as a passivating layer, we compare the open-circuit voltage (V_{OC}) of the perovskite solar cells with and without Al_2O_3. The comparison shows the passivated cells has a higher V_{OC}. We observe the same effect for solar cells with and without the hole transfer layer (HTL: spiro-OMeTAD). The Al_2O_3 nanolayer decreases the recombination centers, leading to higher V_{OC} and cell efficiency.

Keywords: reactive magnetron sputtering, passivation, Al_2O_3, recombination, perovskite solar cell

1 Introduction

The organic-inorganic perovskite solar cells have attracted great deal of attention in recent years due to their low cost and high power conversion efficiency over 22%[1]. As a first attempt, Kojima et al. used methylammonium lead iodide ($CH_3NH_3PbI_3$) as a light absorbing material in dye-sensitized solar cells (DSCs) [2] and numerous researches were conducted on different perovskite materials, deposition methods and charge carrier dynamics of the cells, thereafter.
However, there are some deficiencies that limits their practical applications regarding device instability, hysteresis behaviour of the current-voltage characteristics and charge carrier extraction difficulties arising from the recombination centres at hole transport layer (HTL)/perovskite (PSK) and electron transport layer (ETL)/ PSK interfaces. There have been several metal oxides such as ZrO_2, MgO and mostly Al_2O_3 used in perovskite solar cells structure stack to address each of the aforementioned issues. In this work, a passivation Al_2O_3 layer was deposited on mp-TiO_2 layer in the conventional structure of FTO/c-TiO_2/mp-TiO_2/$CH_3NH_3PbI_3$/spiro-OMeTAD/Au for PSK solar cells in order to increase open-circuit voltage (V_{OC}) by suppressing the carriers recombination at the ETL/PSK interface.

2 Experimental Section

2.1 Device Fabrication

FTO-coated glass substrates were patterned by Zn powder and 2 M HCl etching solution. The patterned FTO substrates were cleaned in an ultrasonic bath with acetone and ethanol. A hole-blocking layer of TiO_2 was deposited by spin-coating and then annealed at 500^0C for 30 min to obtain a uniform compact layer. A mesoporous TiO_2 layer was spin-coated using a solution of TiO_2 paste and annealed after drying on a hotplate.[3]
A new rotational angular method, the (RMS) was used for non-conformal deposition of Al_2O_3 layer on mp-TiO_2 sublayer.

2.2 Perovskite Deposition Method

The perovskite layer was prepared using a two-step spin-coating method. A 1 M PbI_2 solution was spin coated. After drying on hotplate, the solution of CH_3NH_3I in 2-propanol was spin coated on PbI_2 coated substrate. Immediately after preparing perovskite substrate, The spiro-OMeTAD layer, with LiTFSi and tBP additives was spin coated and the Au electrode with the thickness of 100nm was deposited using thermal evaporation method[4] (Figure 1).

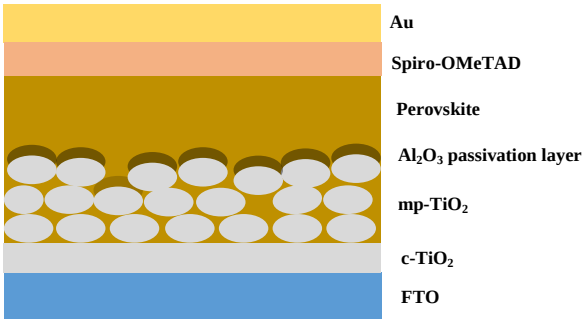

Figure 1 Schematic of perovskites solar cell with Al_2O_3.

3 Results and Discussion

Figures 2a and 2b show the current density-voltage characteristics of the solar cells without and with the deposited layer of Al_2O_3 on mp-TiO_2 sublayer, respectively. The observed increase of V_{OC} and current density (J) in the devices with deposition of Al_2O_3 was attributed to the decreased carriers recombination. Taking into account the V_{OC} enhancement, it is possible to interpret that by using the passivation layer, the number of recombination sites at the ETL/PSK interface decreases, increasing the efficiency. Normally, perovskite layers have pinholes through which ETL layer is in intimate contact with HTL or Au electrode resulting in higher recombination sites, lower V_{OC}, and hence lower efficiency. However, the deposited Al_2O_3 passivation layer with the insulator band gap of 8.8 eV (for bulk crystalline) [5], suppresses charge carrier recombination through the perovskite pinholes. At the same time, there is still enough surface contact between mesoporous ETL layer and perovskite for effective charge extraction. The RMS used for Al_2O_3 deposition leads to a non-conformal layer making no difficulties for perovskite material to diffuse into the mesoporous TiO_2 layer. This provides an appropriate contact between the perovskite layer and ETL. Thus, with a constant photo generation, the thermal recombination rate is decreased due to the passivation layer, leading to a higher cell efficiency. Considering reduced recombination, less charge dissipation, higher optical efficiency, higher V_{OC} and more current density is expected as shown in Figure 2b.

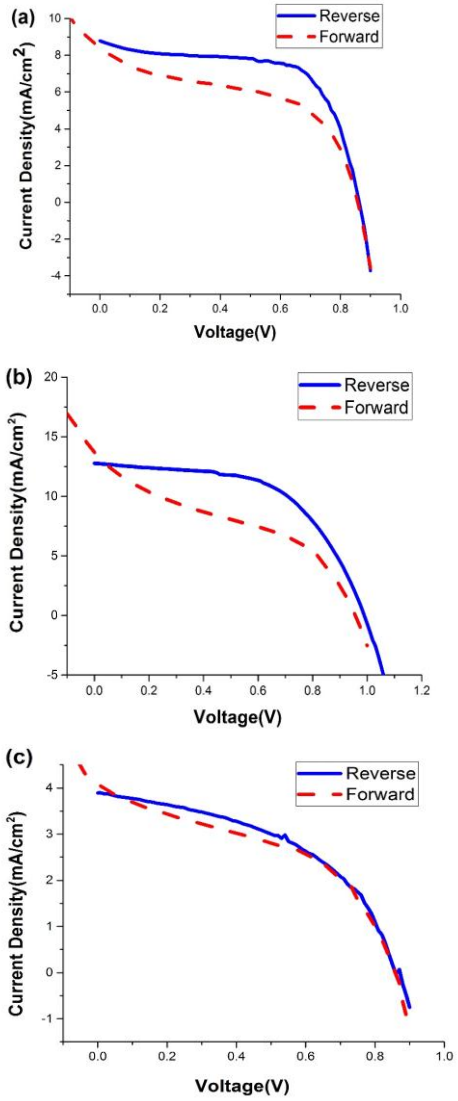

Figure 2 Current-voltage characteristics of solar cell (a) without Al_2O_3 (b) with Al_2O_3 (c) with Al_2O_3/ without spiro-OMeTAD

Figure 2c indicates that in HTL-free devices with Al_2O_3 layer, there was no considerable voltage drop (V_{OC} = 0.87 V) compared to that of devices with HTL (V_{OC} =0.98V). However, low current density in HTL-free devices was observed due to the lack of HTL layer that helps charge extraction with high hole mobility. High V_{OC} with low current density is ascribed to the Al_2O_3 passivation layer, indicating that the number of recombination sites was decreased, although there was still poor charge extraction leading to lower current density.

The sun light illuminates the cell from the FTO side, through the wide bandgap Al_2O_3 layer. Hence, no optical absorption take place before the light being absorbed by the perovskite layer. As a result, the aforementioned passivation layer causes no disturbance in the photo generation of electron-hole pairs, while enhancing the cell efficiency.

Considering dark current-voltage characteristics as an indicator of intrinsic recombination of the structure, Figure 3 shows less recombination in devices using Al_2O_3 layer. Zero current density was observed at higher voltages using Al_2O_3 as a passivation layer, demonstrating the role of the insulator metal oxide layer in suppressing the possible contacts between Au electrode and ETL or HTL and ETL through the perovskite layer pinholes. There is not much charge carrier generation in dark condition and the intrinsic charge carriers face the insulating Al_2O_3 layer hindering carrier transition through mp-TiO_2, resulting in low dark current density.

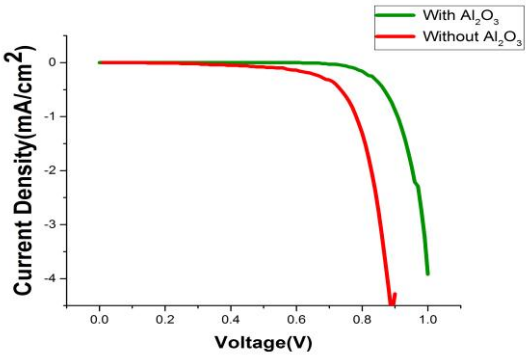

Figure 3. Dark current-voltage characteristic of solar cells with/without Al_2O_3

Solar cells with higher recombination rate indicate faster voltage drop to minimum magnitudes in V_{OC} decay characteristics. According to the V_{OC} decay analysis shown in Figure 4, the fabricated devices with Al_2O_3 present voltage drop with lower speed and reserve more voltage with the passage of time compared to the devices without Al_2O_3 layer, revealing less possibility for charge recombination in agreement with the effective role of Al_2O_3 in decreasing charge recombination sites at mp-TiO_2/ perovskite interface resulting in V_{OC} enhancement.

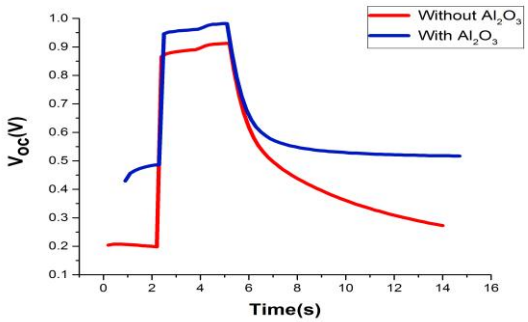

Figure 4. Photovoltage decay characteristics of devices with/without Al_2O_3

4 conclusion

We used the rotational angular RMS method, for deposition of insulator Al_2O_3 layer on mp-TiO_2 in order to enhance V_{OC} in perovskite solar cells. The metal oxide deposited layer had no negative influence in fabrication process. Using Al_2O_3, the great surface contact between perovskite and mp-TiO_2 was reserved along with hindering the likely HTL/ETL or Au electrode/ETL contacts which lead to less recombination sites in the structure. This was verified with current-voltage, photovoltage decay and dark current-voltage characteristics.

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