

## بهبود ولتاژ مدار باز 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 نشان دادن لایه‌ی دی اکسید آلومینیوم روی لایه‌ی متخلخل انتقال دهنده الکترون ( $mp-TiO_2$ ) در خواص فتوولتاییکی سلول‌های خورشیدی پروسکایتی بررسی شده است. به دلیل عایق بودن دی اکسید آلومینیوم نیاز است نشان دادن لایه به صورت غیر یکنواخت صورت بگیرد و به این منظور از روش کندوپاش مغناطیسی چرخشی استفاده شده است. در این کار، ولتاژ مدار باز سلول‌های خورشیدی پروسکایتی با استفاده از لایه‌ی دی اکسید آلومینیوم و بدون آن مقایسه شده است. با استفاده از لایه نازک دی اکسید آلومینیوم، به دلیل کاهش مراکز باز ترکیب 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- باز ترکیب، دی اکسید آلومینیوم، ولتاژ مدار باز، سلول‌های 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 ( $\text{CH}_3\text{NH}_3\text{PbI}_3$ ) 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  $\text{ZrO}_2$ ,  $\text{MgO}$  and mostly  $\text{Al}_2\text{O}_3$  used in perovskite solar cells structure stack to address each of the aforementioned issues. In this work, a passivation  $\text{Al}_2\text{O}_3$  layer was deposited on mp- $\text{TiO}_2$  layer in the conventional structure of FTO/c- $\text{TiO}_2$ /mp- $\text{TiO}_2$ / $\text{CH}_3\text{NH}_3\text{PbI}_3$ /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

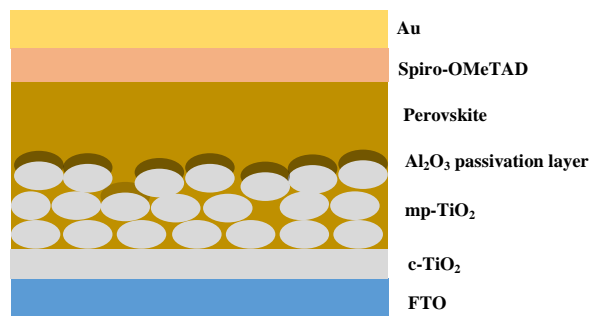
FTO-coated glass substrates were patterned by Zn powder and 2 M HCl etching solution. The patterned FTO substrates were cleaned in an ultrasonic bath with acetone and ethanol. A hole-blocking layer of  $\text{TiO}_2$  was deposited by spin-coating and then annealed at  $500^\circ\text{C}$  for 30 min to obtain a uniform compact layer. A mesoporous  $\text{TiO}_2$  layer was spin-coated using a solution of  $\text{TiO}_2$  paste and annealed after drying on a hotplate.[3]

A new rotational angular method, the (RMS) was used for non-conformal deposition of  $\text{Al}_2\text{O}_3$  layer on mp- $\text{TiO}_2$  sublayer.

### 2.2 Perovskite Deposition Method

The perovskite layer was prepared using a two-step spin-coating method. A 1 M  $\text{PbI}_2$  solution was spin coated. After drying on hotplate, the solution of  $\text{CH}_3\text{NH}_3\text{I}$  in 2-propanol was spin coated on  $\text{PbI}_2$  coated substrate. Immediately after preparing

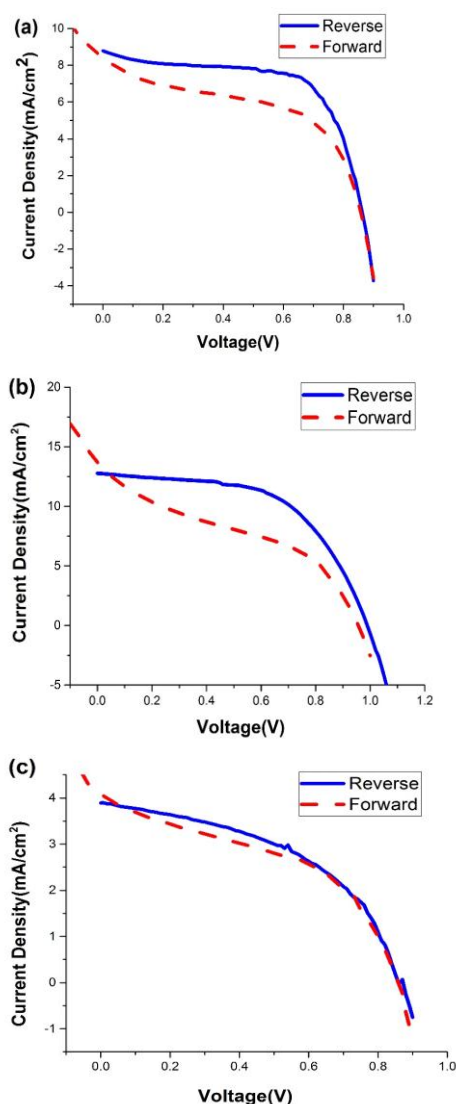
perovskite substrate, The spiro-OMeTAD layer, with LiTFSi and tBP additives was spin coated and the Au electrode with the thickness of 100nm was deposited using thermal evaporation method[4] (Figure 1).



**Figure 1** Schematic of perovskites solar cell with  $\text{Al}_2\text{O}_3$ .

## 3 Results and Discussion

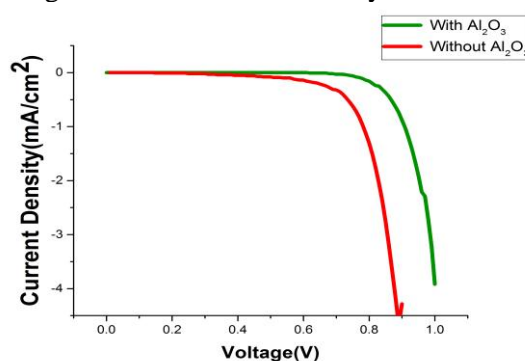
Figures 2a and 2b show the current density-voltage characteristics of the solar cells without and with the deposited layer of  $\text{Al}_2\text{O}_3$  on mp- $\text{TiO}_2$  sublayer, respectively. The observed increase of  $V_{oc}$  and current density ( $J$ ) in the devices with deposition of  $\text{Al}_2\text{O}_3$  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  $\text{Al}_2\text{O}_3$  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  $\text{Al}_2\text{O}_3$  deposition leads to a non-conformal layer making no difficulties for perovskite material to diffuse into the mesoporous  $\text{TiO}_2$  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  $\text{Al}_2\text{O}_3$  (b) with  $\text{Al}_2\text{O}_3$  (c) with  $\text{Al}_2\text{O}_3$ /without spiro-OMeTAD

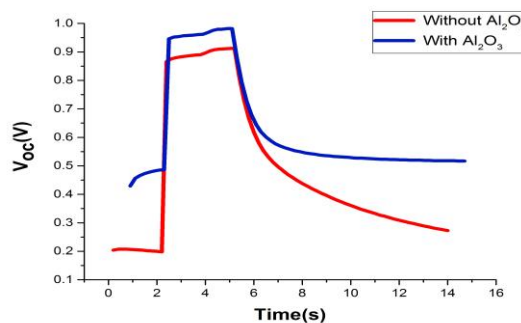
Figure 2c indicates that in HTL-free devices with  $\text{Al}_2\text{O}_3$  layer, there was no considerable voltage drop ( $V_{OC} = 0.87 \text{ V}$ ) compared to that of devices with HTL ( $V_{OC} = 0.98 \text{ V}$ ). However, low current density in HTL-free devices was observed due to the lack of HTL layer that helps charge extraction with high hole mobility. High  $V_{OC}$  with low current density is ascribed to the  $\text{Al}_2\text{O}_3$  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  $\text{Al}_2\text{O}_3$  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  $\text{Al}_2\text{O}_3$  layer. Zero current density was observed at higher voltages using  $\text{Al}_2\text{O}_3$  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  $\text{Al}_2\text{O}_3$  layer hindering carrier transition through mp-TiO<sub>2</sub>, resulting in low dark current density.



**Figure 3.** Dark current-voltage characteristic of solar cells with/without  $\text{Al}_2\text{O}_3$

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  $\text{Al}_2\text{O}_3$  present voltage drop with lower speed and reserve more voltage with the passage of time compared to the devices without  $\text{Al}_2\text{O}_3$  layer, revealing less possibility for charge recombination in agreement with the effective role of  $\text{Al}_2\text{O}_3$  in decreasing charge recombination sites at mp-TiO<sub>2</sub>/perovskite interface resulting in  $V_{OC}$  enhancement.



**Figure 4.** Photovoltage decay characteristics of devices with/without  $\text{Al}_2\text{O}_3$

## 4 conclusion

We used the rotational angular RMS method, for deposition of insulator  $\text{Al}_2\text{O}_3$  layer on mp- $\text{TiO}_2$  in order to enhance  $V_{\text{oc}}$  in perovskite solar cells. The metal oxide deposited layer had no negative influence in fabrication process. Using  $\text{Al}_2\text{O}_3$ , the great surface contact between perovskite and mp- $\text{TiO}_2$  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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