Atomic Layer Engineering of Aluminum-Doped Zinc Oxide Films for Efficient and Stable Perovskite Solar Cells

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Joanna Kruszyńska, J. Ostapko, V. Ozkaya, B. Surucu, O. Szawcow, K. Nikiforow, M. Hołdyński, M. M. Tavakoli, P. Yadav, M. Kot, G. P. Kołodziej, M. Wlazło, S. Satapathi, S. Akin, D. Prochowicz

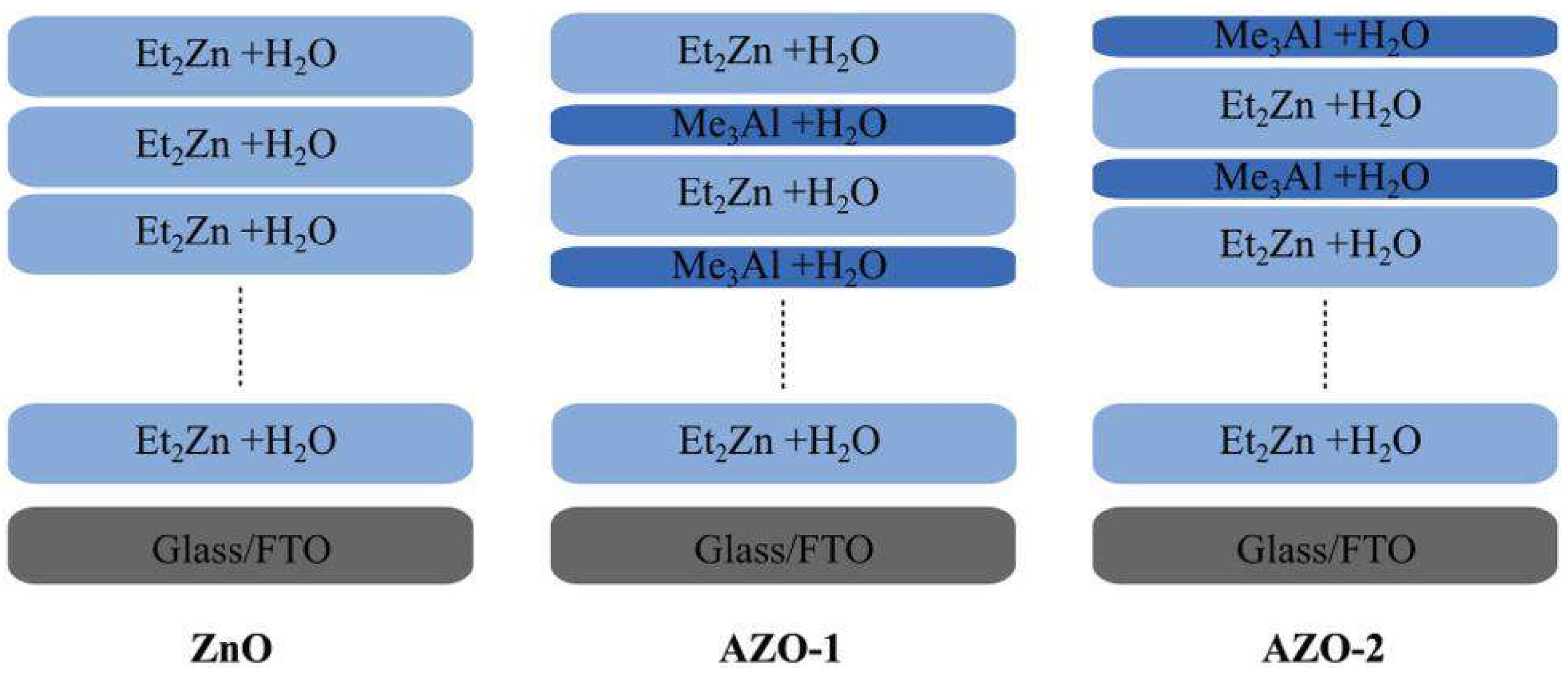
Institute of Physical Chemistry, PAS, Kasprzaka 44/52, 01-224 Warsaw, Poland, jkruszynska@ichf.edu.pl

Introduction

Atomic layer deposition (ALD) has been considered as an efficient method to deposit high quality and uniform thin films of various electron transport materials for perovskite solar cells (PSCs).

Herein, we demonstrate the effect of the ALD deposition sequence of aluminum-doped zinc oxide (AZO) film on its optoelectronic properties and the film quality of the $MAPbI_3$ perovskite layer. Particularly, we find that the ending of the ALD process with the TMA/H_2O pulse (AZO-2) lead to the formation of AZO film exhibiting superior electrical conductivity compared to the AZO ended with DEZ/H_2O pulse (AZO-1) or pristine ZnO ETL.

Scheme 1 shows the schematic representation of ALD processes for the formation of ZnO and AZO films.



Scheme 1. Schematic representation of growth process of ZnO and AZO films by ALD.

Results

First, we tested the thermal stability of the $MAPbI_3$ films deposited on different ETLs. The films were annealed at 100 ºC in argon glovebox at different times, and the color change in films was monitored as shown in **Figure 1**. The $MAPbI_3$ film deposited on the ZnO substrate starts to decompose after 10 min of annealing, whereas the perovskite films deposited on AZO-1 and AZO-2 remained black during the applied heating time. The presence of perovskite structure was confirmed by the analysis of the X-ray diffraction (XRD) patterns, as shown in **Figure 2**. A prolonged heating time (30 min) starts degradation of the $MAPbI_3$ film coated on top of AZO-1 as the presence of the characteristic peak around 12,5 º2θ in the XRD pattern suggests the formation of the PbI_2 phase. In turn, there is a small PbI_2 peak over this period for the $MAPbI_3$ film deposited on AZO-2 ETL, indicating an improved ETL/perovskite interface stability.

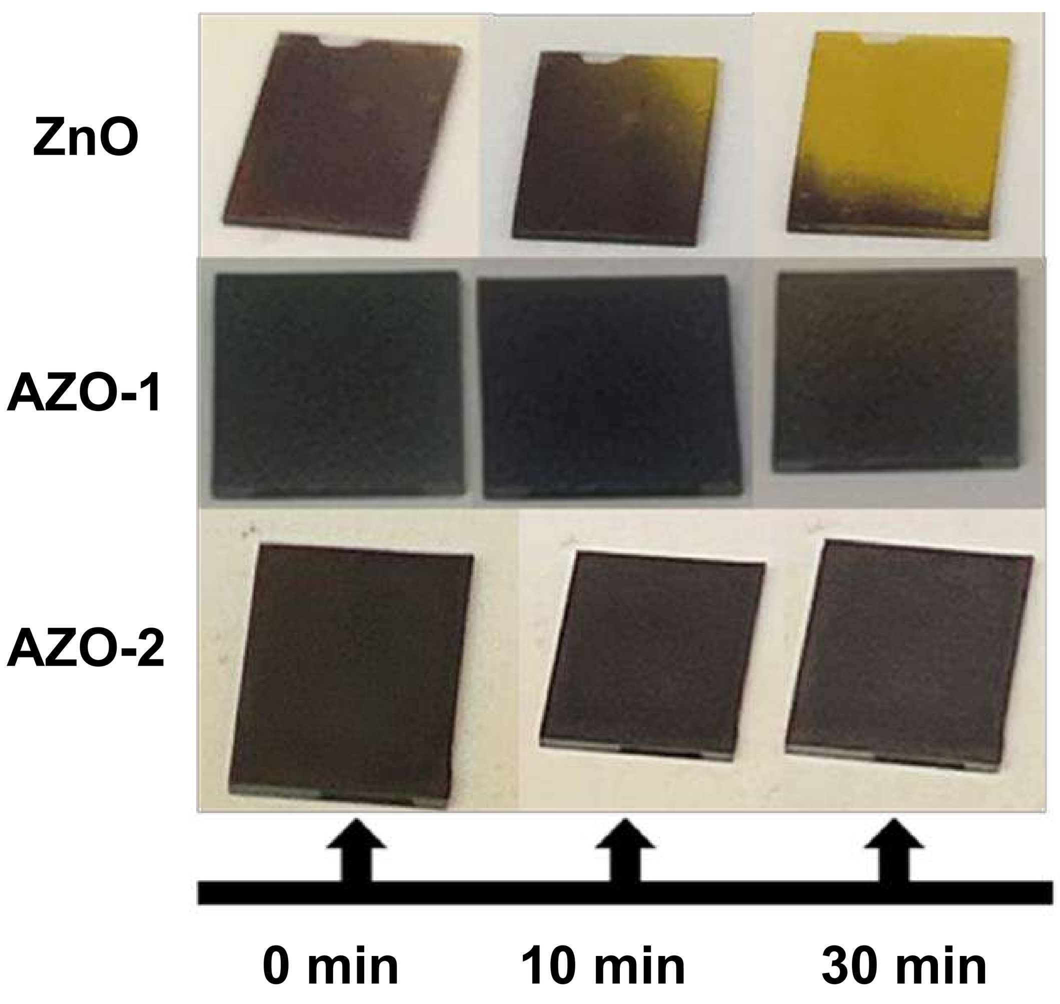


Figure 1. The optical images of perovskite films deposited on different ETLs and annealed at 100 ºC for 10 and 30 minutes.

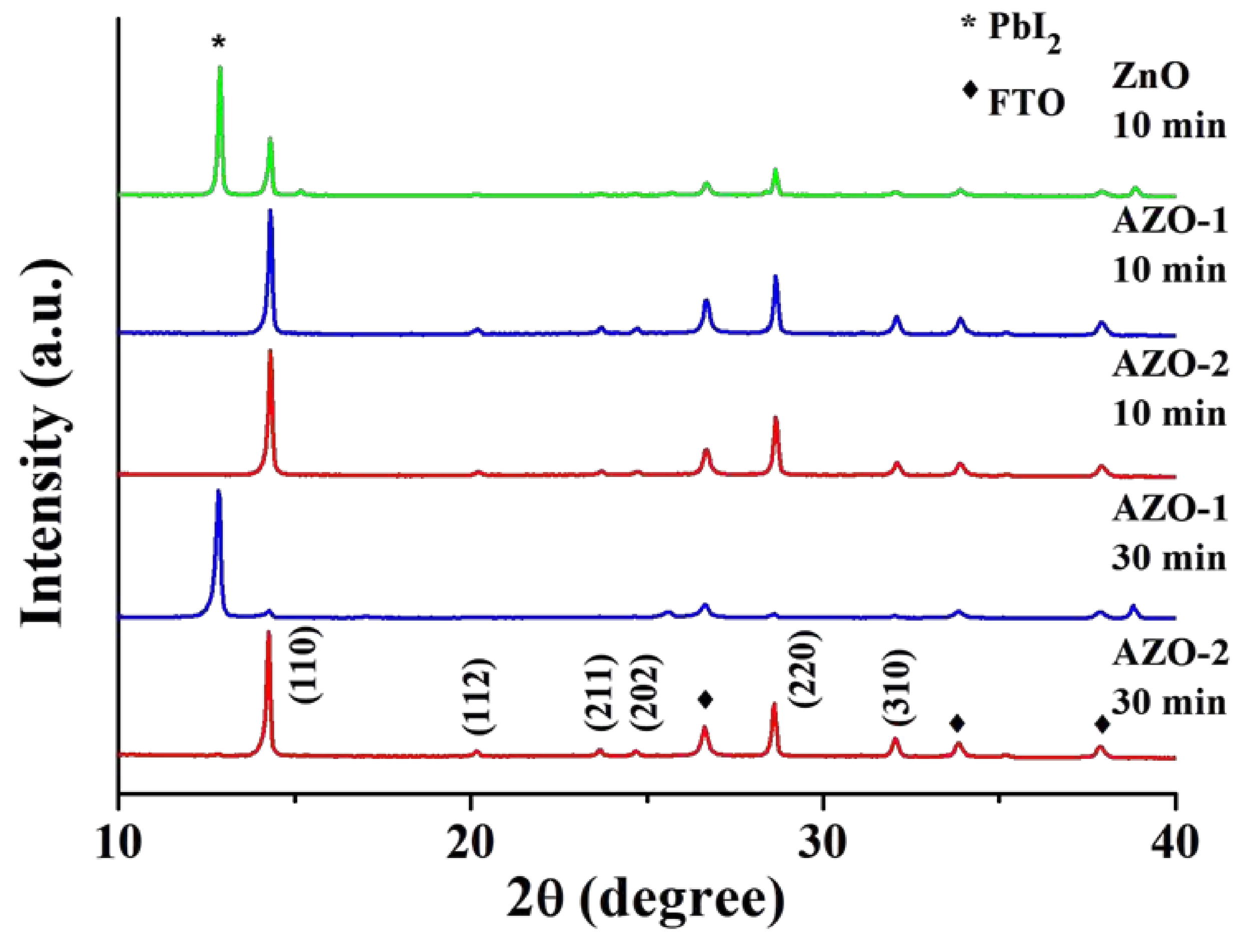


Figure 2. XRD patterns of the perovskite films deposited on different ETLs and annealed at 100 ºC at different times.

Photovoltaic performance

The effect of ALD-deposited ETLs on the performance and stability of PSCs was further investigated by the fabrication of planar devices consisting of FTO/ETL/$MAPbI_3$/spiro-OMeTAD/Au. The perovskite films were spin-coated on different ETLs and annealed at 100 ºC for only 10 min to avoid degradation. **Table 1** lists the photovoltaic (PV) parameters of the corresponding PSCs. The average data were collected from 8 devices for each ETL.

The ZnO-based device yields a champion PCE of 16,23% with a short-circuit current density (JSC) of 22,82 mA cm^{-2}, an open-circuit voltage (VOC) of 0,99 V, and a fill factor (FF) of 72%. In contrast, superior photovoltaic performance is observed for both AZO-based PSCs. While the AZO-1 device gives a PCE of 17,99% with a JSC of 23,20 mA cm^{-2}, VOC of 1,04 V, and FF of 74%, the device with AZO-2 offers the best PCE of 18,09% with a JSC of 23,16 mA cm^{-2}, VOC of 1,05 V, and FF of 74%.

ETL		V_{OC} (V)	J_{SC} (mA cm^{-2})	FF (%)	PCE (%)
ZnO	champion	0,99	22,82	72	16,23
	average	0,98 ± 0,01	22,80 ± 0,08	69 ± 1,5	15,45 ± 0,41
AZO-1	champion	1,04	23,20	74	17,99
	average	1,04 ± 0,01	23,17 ± 0,07	73 ± 1,1	17,65 ± 0,30
AZO-2	champion	1,05	23,16	74	18,09
	average	1,05 ± 0,01	23,11 ± 0,05	74 ± 0,7	18,01 ± 0,20

Table 1. The photovoltaic (PV) parameters of the corresponding PSCs.

Conclusions

The controlled termination of the ALD process with TMA/H_2O pulse not only offers high conductivity for the AZO film but also leads to the favorable band alignment concerning the perovskite layer, which facilitates charge transfer at the interface. In addition, the modification of a surface chemical state of AZO film prevents the degradation reaction at the ETL/perovskite interface and improves the thermal stability of the $MAPbI_3$ film compared to the control perovskite films. Consequently, our best planar device yielded a high PCE of 18,09%. These results demonstrate the superiority of the developed atomic layer engineering approach that may open up new possibilities for the design of efficient AZO-based ETLs to tackle the thermal instability issue of the ZnO/perovskite interface and offer a new direction for the fabrication of highly efficient PSCs.

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References

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