

## Fabrication and characterization of DPPS-introduced perovskite solar cells

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## INTRODUCTION &amp; AIM

Various halide perovskite crystals based on  $\text{CH}_3\text{NH}_3\text{PbI}_3$  (abbreviated  $\text{MAPbI}_3$ ) are expected to become next-generation solar cell materials and are being researched and developed worldwide. However, despite its high photoelectric conversion efficiency,  $\text{MAPbI}_3$  has an unstable structure due to the presence of  $\text{CH}_3\text{NH}_3$  (MA) organic molecules, and the decomposition of  $\text{CH}_3\text{NH}_3$  from  $\text{CH}_3\text{NH}_3\text{PbI}_3$  in the atmosphere results in the formation of  $\text{PbI}_2$ . The expensive spiro-OMeTAD is widely used throughout the world as a hole transport material in perovskite solar cells. To enhance the hole transport, spiro-OMeTAD needs dopants such as lithium bis(trifluoromethane sulfonyl) imide and 4-*tert*-butylpyridine, which are highly hygroscopic and accelerate degradation of the perovskite layer when moisture is absorbed. Therefore, it is necessary to find alternative hole transport materials.

The aim of the present work is fabrication of perovskite photovoltaic devices in air and under unsealed conditions using decaphenylcyclopentasilane (DPPS) selected as a material to modify the perovskite surface, DPPS is stable in air at temperatures up to 300 °C and has the potential to suppress MA desorption, and high-temperature heat treatment can form dense cubic perovskite, which is a stable phase at high temperatures. Polysilane has also been reported to function as a hole transport material, which is also expected to improve photoelectric conversion properties by using the DPPS as a hole transport layer. The fabrication of perovskite photovoltaic devices with DPPS by the air blow method in air could greatly simplify the manufacturing process without using a glove box and stabilizes the solar cell characteristics.

## METHOD

A method for fabricating a perovskite solar cell is shown in Fig. 1(a). A compact  $\text{TiO}_2$  layer and a mesoporous  $\text{TiO}_2$  layer were prepared on the F-doped tin-oxide (FTO) substrates by spin-coating and annealing at 550 °C for 30 min in air. The basic  $\text{MAPbI}_3$  precursor solutions were prepared by mixing  $\text{CH}_3\text{NH}_3\text{I}$  (Tokyo Chemical Industry, 2.4 M) and  $\text{PbCl}_2$  (Sigma-Aldrich, 0.8 M) in *N,N*-dimethylformamide (Sigma-Aldrich) at 60 °C for 24 h. GA-, FA-, or EA-added precursors were prepared by adding guanidinium iodide (GAI), formamidinium iodide (FAI), or ethylammonium iodide (EAI) to control the molar ratio of 12.5 % at the MA site. A method for fabricating the perovskite and DPPS layers is shown in Fig. 1(b). First, the prepared perovskite precursor solutions were spin-coated (2000 rpm for 60 s) three times on the mesoporous  $\text{TiO}_2$  layer applying a hot air blowing method. Next, the MA-substituted perovskite precursor solutions added with 12.5 % organic cations (GA, FA, or EA), such as  $\text{MAPbI}_3$ ,  $\text{MA}_{0.875}\text{GA}_{0.125}\text{PbI}_3$ ,  $\text{MA}_{0.875}\text{FA}_{0.125}\text{PbI}_3$ , and  $\text{MA}_{0.875}\text{EA}_{0.125}\text{PbI}_3$ , were then spin-coated several times. On the third spin-coating, chlorobenzene solution with DPPS (SI-30-15, Osaka Gas Chemicals, 20 mg  $\text{mL}^{-1}$ ) were dropped on the perovskite layer. The deposition sequences such as  $\text{MAPbI}_3$ -3 layers,  $\text{MA}_{0.875}\text{GA}_{0.125}\text{PbI}_3$ -2 layers, and DPPS are denoted as MA3/GA2/DPPS in the present work. To separate the perovskite layers clearly between the standard  $\text{MAPbI}_3$  and GA-, FA-, or EA-added  $\text{MAPbI}_3$ , the double-DPPS device structures such as MA3/DPPS/GA2/DPPS were also fabricated. The prepared cells were annealed at 190 °C for 10 min in air to form the perovskite layer. Finally, a gold (Au) thin film was evaporated onto the DPPS layer, as top electrodes.

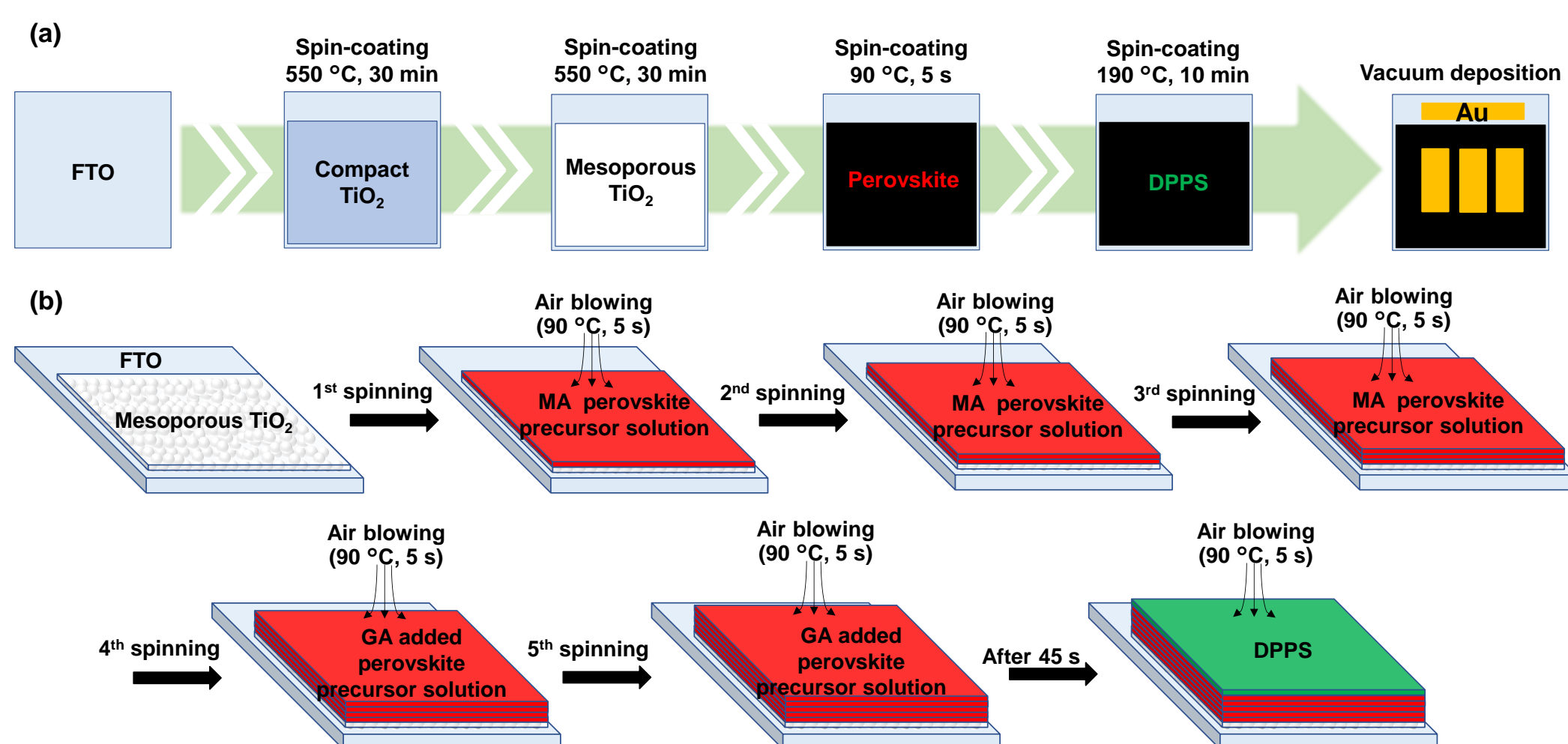


Fig. 1. (a) Schematic illustration of the process used to fabricate the perovskite solar cells. (b) Detailed processes of the perovskite and DPPS layers [1].

## RESULTS &amp; DISCUSSION

The changes in the  $\eta$  values of representative prepared perovskite solar cells are shown in Fig. 2. The  $\eta_{\text{ave}}$  values were plotted, where the error bars in Fig. 2 represent the maximum and minimum conversion efficiencies of the cells. Small increases in the  $\eta$  values were observed for the MA3 and MA3/GA2 devices after 50 days. Upon inserting the DPPS layers into the perovskites, the  $\eta$  values increased, especially for the MA3/GA2/DPPS device, and remained almost unchanged after 200 days.

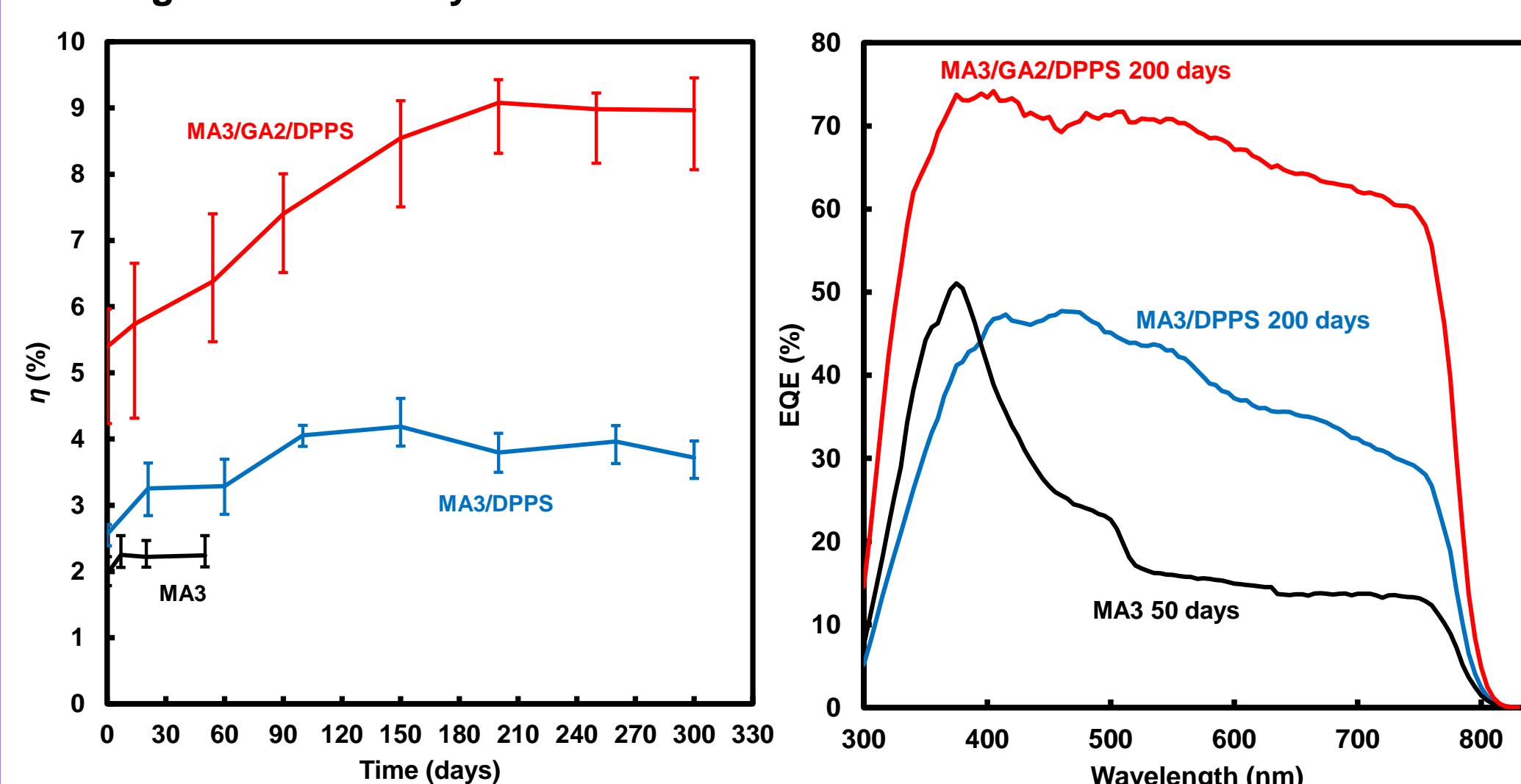


Fig. 2. Changes in the conversion efficiencies and EQE of the present cells.

The XRD patterns of the devices stored for 50 and 200 days in a dark room at ~22 °C are shown in Fig. 3. The diffraction intensity of the 100 and 200 peaks were high, indicating the formation of (100)-oriented perovskite crystals. The energy-level diagram shows carriers generated in the perovskite layer were separated into holes and electrons, and the holes and electrons flowed to the gold electrode via DPPS and the titanium dioxide layer, respectively.

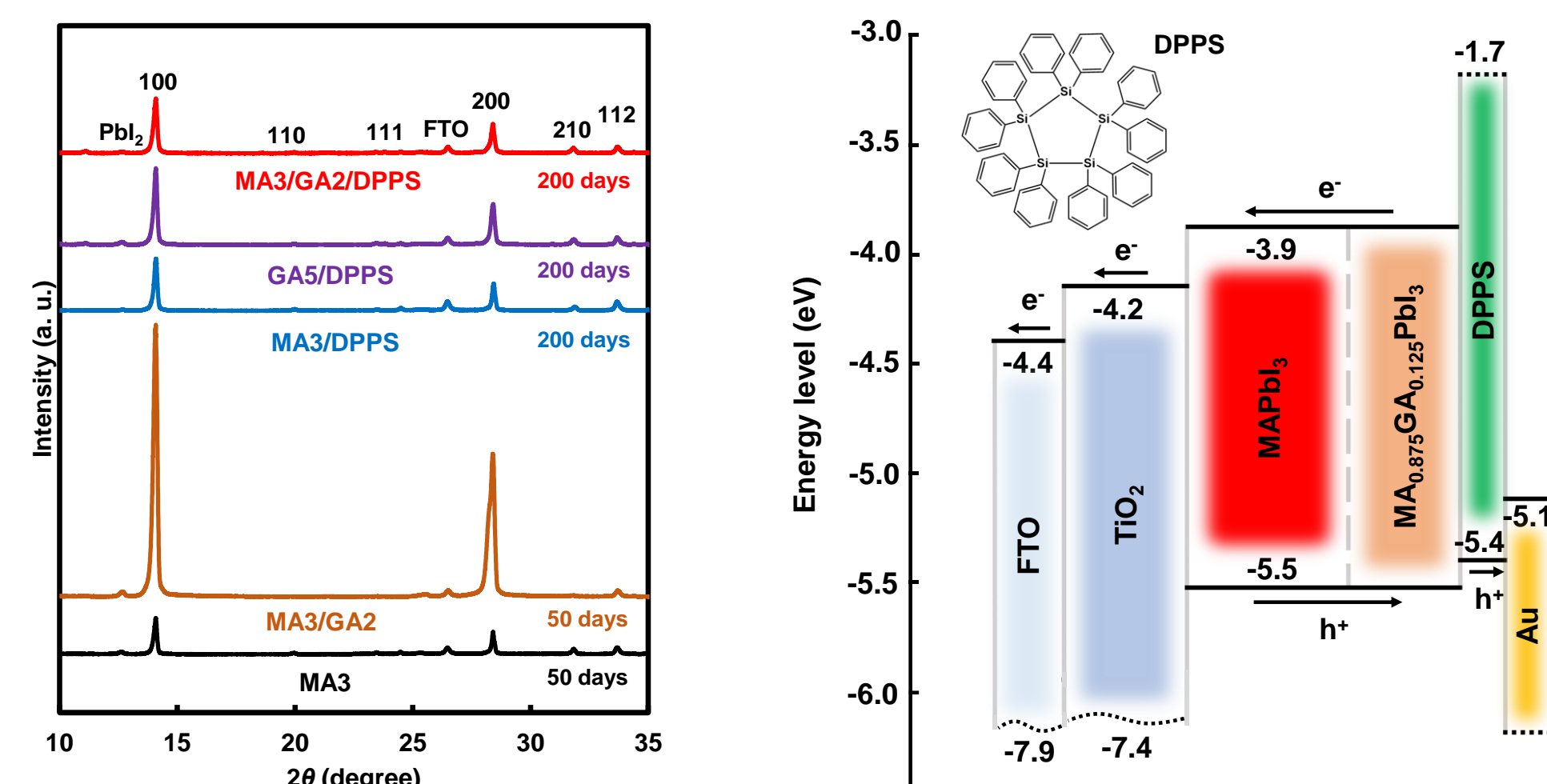


Fig. 3. XRD patterns and energy level diagram of the present cells [1].

## CONCLUSION

Stable DPPS hole transport layers were developed for guanidinium-, formamidinium-, or ethylammonium-added double-stacked  $\text{MAPbI}_3$  perovskite photovoltaic devices without using spiro-OMeTAD, which is a typical hole transport layer. Stacking two types of perovskite compounds inserted within the DPPS layer enabled the fabrication of stable perovskite solar cells under ambient atmosphere at 190 °C, which reduced the trap density and improved the photovoltaic properties of the devices. As expected, the DPPS-inserted layers functioned as a stable hole transport layer for perovskite solar cells.

## REFERENCES

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