

## Summary of Phase 4 of the SEE 8 Project

At this stage, activities related to the manufacture and characterization of perovskite solar cells fabricated on both flexible and glass substrates were performed, in order to establish the deposition parameters leading to the highest power conversion efficiency. The most efficient perovskite cells (PCE > 15%) were obtained using the standard geometry and when both the compact and mesoporous TiO<sub>2</sub> layers were deposited by spray pyrolysis and spray coating respectively, using the PERPHECT printing equipment built during this stage. In order to achieve solar cells on a flexible substrate, we replaced the FTO conductive transparent electrode with metal nanowires on glass deposited by electrospinning. Using these substrates, standard solar cells were fabricated by spraying the TiO<sub>2</sub> layers over the Au wires. Thus, we obtained functional solar cells with a maximum PCE of 12.20% (mean value 10.2%). The typical J-V curve obtained for a cell with Au nanowires network is shown in Fig. 1. Flexible electrodes can be attached on top as well, resulting in a flexible solar cell with transparent electrodes on both sides. The technological problem we are currently facing is that it is technically difficult to contact the transparent top electrode outside of the solar cell area. During the entire project, we were able to measure all solar cells by directly contacting the top electrode on the active surface of the device. PERPHECT printing prototype was assembled and tested during this stage (see Figure 2).

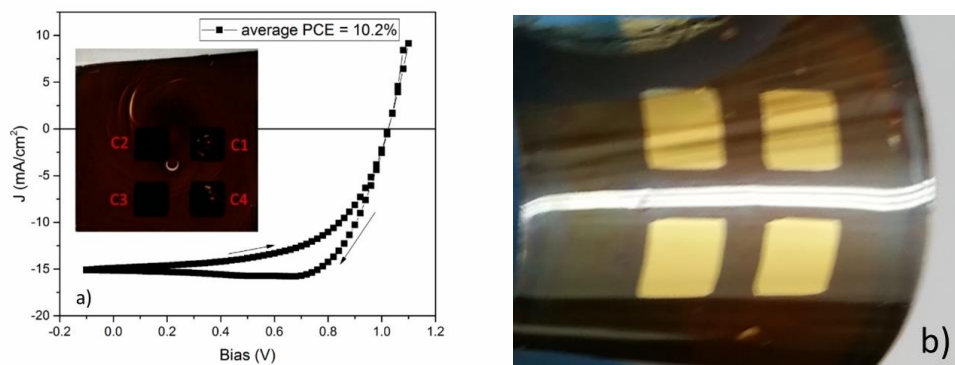


Fig 1. a) The typical J-V characteristic for perovskite solar cells in which the FTO electrode was replaced with a flexible one from Au nanowires network. The insertion is a photo taken through the transparent metallic Au nanowires mesh and the squares represents the shades from the top electrodes of Au (C1, C2, C3 and C4). The brown colour comes from the MAPI active layer; b) Top view of the flexible perovskite solar cell showing the top gold electrodes.

Using substrates of 10x10 cm<sup>2</sup> of glass covered with FTO or metallic web, compact and mesoporous layers of TiO<sub>2</sub> were deposited by spray coating, followed by the perovskite and Spiro-OMeTAD layers deposited by dr. blade. The PERPHECT prototype equipment was used in the project to manufacture perovskite solar cells. The prototype was designed as a modular printer offering the possibility of depositing successive layers of different types of materials with different viscosities - in liquid or paste form. Each of the deposited layers can be heated and maintained at a temperature up to 150°C, depending on each layer requirements.

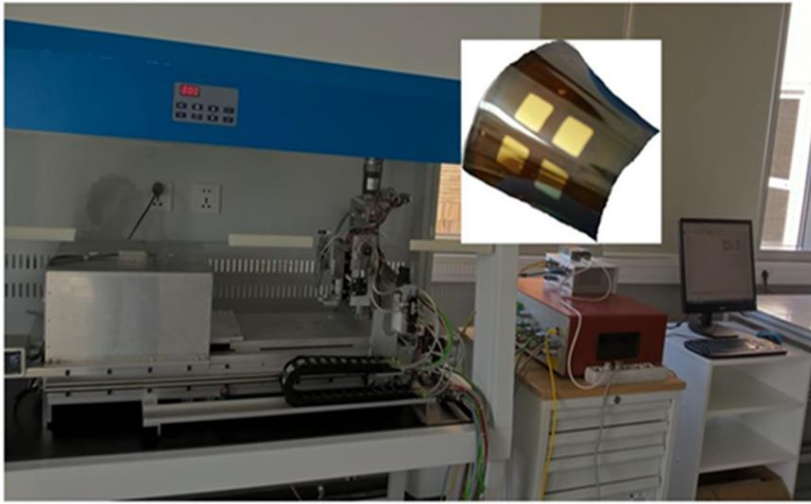


Fig. 2 Manufacturing Perovskite Solar Cells (PSC) on a flexible and transparent substrate with the prototype equipment mounted in an fume hood at the Coordinator headquarters (NIMP)

One of the main conclusions demonstrated both theoretically and experimentally is that different forms of J-V hysteresis can occur by varying the initially applied polarization voltage. The presence of both types of normal (NH) and inverted (IH) hysteresis combines apparently contradictory results reported in recent publications. The experimental data fits the dynamic electrical model (DEM), showing the existence of both NH and IH on the same sample depending on the J-V curves measurement procedure and the voltage applied initially to the cell before measurement. Examples of NH and IH predicted by DEM and obtained from real measurements are shown in Figure 3.

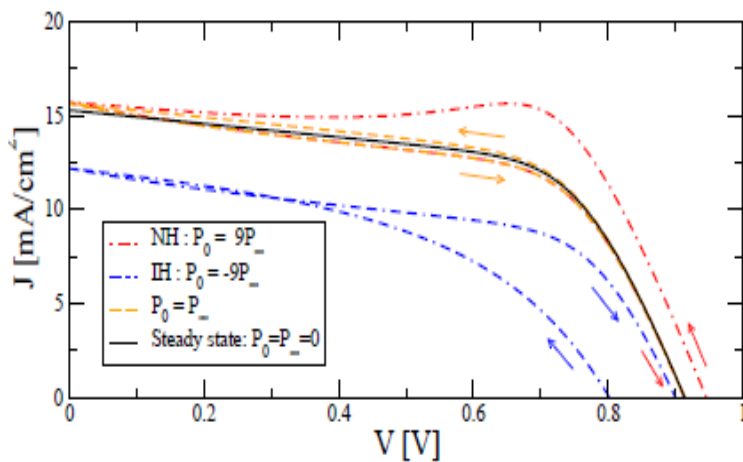


Fig 3. NH si IH in comparison to the steady state characteristics on the same sample. The figure shows the presence of NH ( $P_0 = 9 P_\infty$ ), IH ( $P_0 = -9 P_\infty$ ) and minimal polarization. ( $P_0 = P_\infty$ ) for a scanning rate of 20 mV / s and steady state solution ( $P_\infty = 0$ ) in voltage mode  $[0, V_{oc}]$ .

PCE varies drastically in direct and reverse scanning for NH and IH with a large deviation in the reverse versus steady state: 27% overestimation for NH and 48% underestimation for IH. In the direct scanning mode of the NH with minimal pre-polarization (in  $V_{oc}$ ), there is a deviation from the actual PCE of only 2.3%, while for IH a PCE value is obtained by 28% lower. Thus, DEM reproduces the NH and IH behaviours due to the initial polarization, highlighting the importance of controlling the PSC preconditions, the state induced by the previous measurements that may affect the subsequent measurement. The three-step measurement protocol, including the stages of stabilization, pre-polarization and measurement, is appropriate to obtain accurate and reproducible experimental data. Atomistic simulations of the structural and electronic properties of  $\text{CH}_3\text{NH}_3\text{Pb}(\text{I},\text{Cl})_3$  layers deposited on ferroelectric surfaces have been extended to the use of  $\text{BaTiO}_3$  and the main conclusion is that one should use a ferroelectric with a strong enough polarization, in a balance with a low lead concentration.