Experimental report

25/10/2022

Proposal: **5-21-1144** **Council:** 10/2019

Title: Neutron diffraction experiment on structural ordering/degradation insight of hybrid perovskite solar cells

Research area: Materials

This proposal is a new proposal

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Samples: MAxFA(1-x)PbI3

Instrument	Requested days	Allocated days	From	To
D20	3	2	24/08/2020	26/08/2020
D2B	2	0		

Abstract:

The field of photovoltaics and energy harvesting has been deeply impacted recently by the appearance of perovskite solar cells (PSCs) as a new material for solar energy conversion. The unique combination of high performance and low-cost fabrication processes of PSCs make them the ideal candidate to be implemented in solar cell devices. For a full understanding of the underlying mechanisms leading to the optimal properties of PSCs, it is a requirement to investigate the structural phase transitions occurring in them, with the ultimate goal of establishing a direct relation between their efficiency and the perovskite symmetry, organic cation/halide mixture ordering/distribution within the inorganic lead-iodide net, as well as their degradation processes. In this context, we aim at studying the crystalline structures of mixed FA/MA halide PSCs with the objective of establishing a relation between the PCE and the crystal structure symmetry, as well as the FA/MA and halide mixture position and distribution within the crystal structure. In addition to this, we also want to study the effect of degradation on the PSCs.

EXPERIMENT N°: 5-21-1144 **INSTRUMENTS:** D20

DATES OF EXPERIMENT 24/08/2020-26/08/2020

TITLE: Neutron diffraction experiment on structural ordering/degradation insight of hybrid perovskite solar cells

The field of photovoltaics and energy harvesting has been deeply impacted recently by the appearance of perovskite solar cells (PSCs) as a new material for solar energy conversion. The unique combination of high performance and low-cost fabrication processes of PSCs make them the ideal candidate to be implemented in solar cell devices. Among the wide variety of organic-inorganic perovskite formulations, $CH_3NH_3PbI_3$ is the most widely investigated. The methylammonium (MA) cation (2.17 Å) satisfies perfectly the Goldschmidt tolerance factor (t=0.89), however, it presents an orientational disorder in the crystal which increases with the temperature. In this circumstance, three different crystal phases occur at different temperatures, depending on the organic cation ordering and inorganic structure distortion. In order to block the distortion of the inorganic network due to the organic cation mismatch, larger organic cations such as formamidinium (FA) (2.53 Å), which involves a tolerance factor close to one, have been used to stabilize highly symmetric perovskite structures at RT. However, depending thermal treatments such as annealing, FA perovskite could exhibit a corner sharing α-phase structure (black colour, tannealing < 150ºC) or a face-sharing δ-phase perovskite (yellow colour, , tannealing ≥ 150ºC), this last undesired for applications.

Nonetheless, the stabilization of the aforementioned phases in FA-based PSCs is still under research, and different approaches have been considered so far. In this context, Salado et al. demonstrated in a recent study that an addition of 20% MA in FA-based PSCs avoids the formation of the undesired δ-phase while maintaining the red-shifted band gap of FAPbI3. An increase of MA to 40%, keeping the FA cation content at 60%, shows the best performance results in MA-FA mixed cation PSCs [5].

Within this experiment we aimed at studying the effect of degradation on the PSCs on the aforementioned crystal phases of the studied PSCs, by acquiring diffractograms under temperature and humidity conditions. In particular, we measured three different PSCs at room temperature. 85ºC and 150ºC. On top of this, we also exposed similar samples to S1, S2 and S3 to humidity conditions and measured diffractograms on them at room temperature and 150ºC, to observe if there was any effect on the crystal structure both after temperature and humidity exposure. Representative obtained diffractograms are shown in Figures 1 and 2.

The results obtained with this experiment will be cross-correlated with neutron reflectometry experiments. The first one has been performed at D17 (experiment number 1-04-170), where we studied temperature and humidity degradation processes in PSCs with similar compositions to those studied here, but the conclusions from that experiment are still incomplete. We will perform a second neutron reflectometry experiment at ANSTO by the end of the year, and with the input of the three experiments we expect to be able to extract a publication on the degradation mechanisms of PSCs under temperature and humidity conditions.

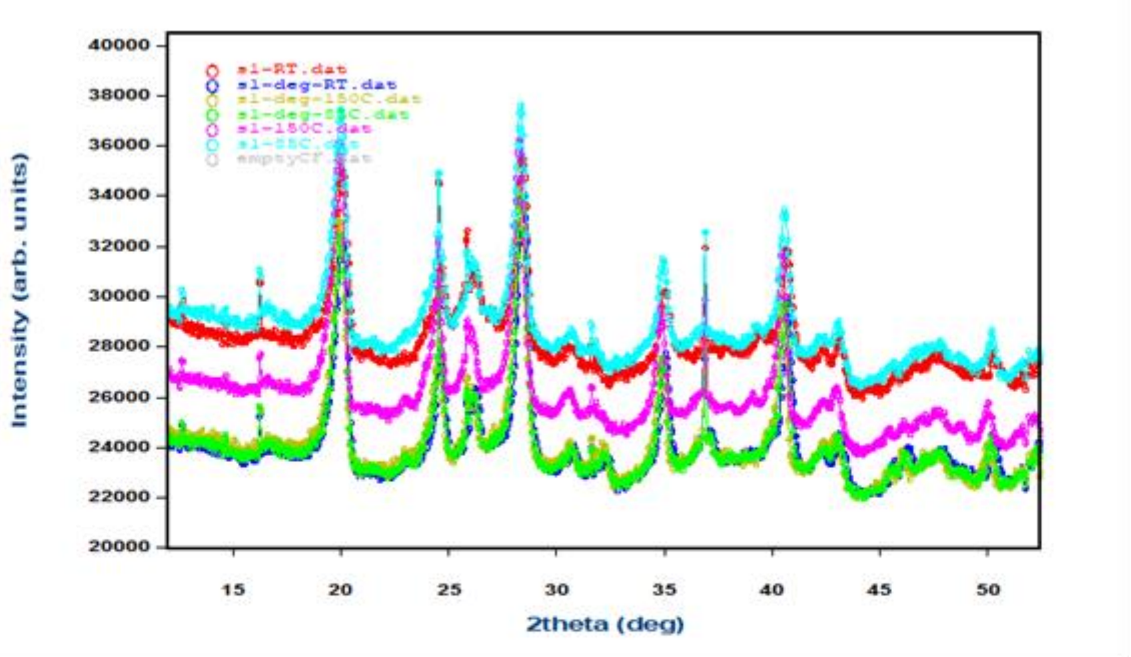

Figure 1: Powder neutron diffraction data obtained from the S1 PSC at different temperatures. Dark blue, cyan and magenta plots correspond to diffractograms acquired after exposure to humidity conditions at the temperatures labelled in the legend.

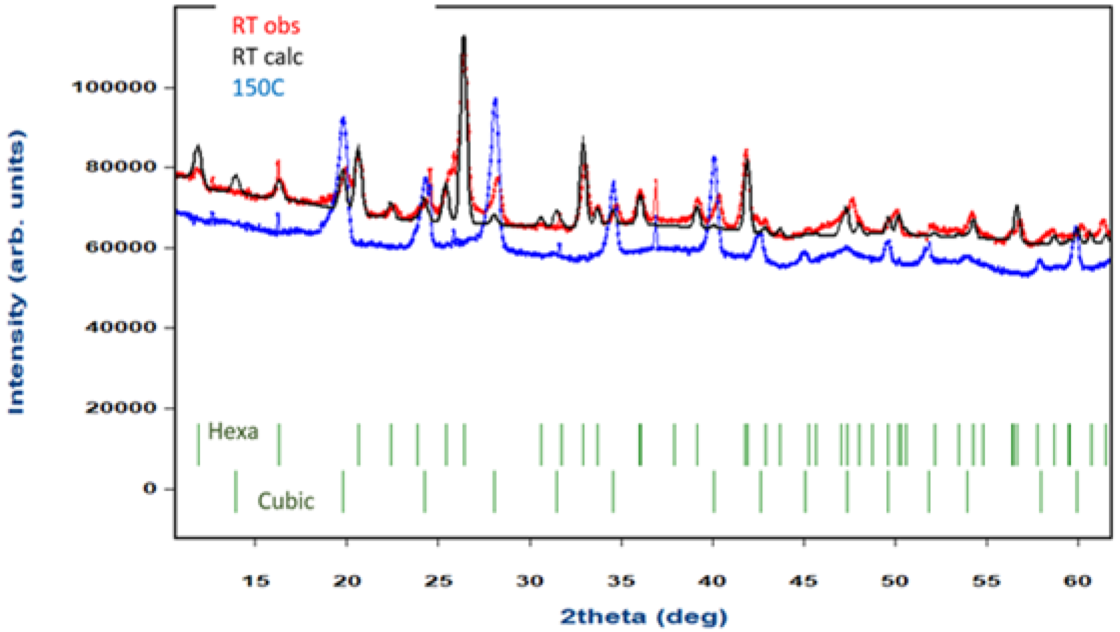

Figure 2: Powder neutron diffraction data obtained from the S3 PSC at different temperatures. The preliminary data analysis infers that, while at RT the sample presents a mixture of hexagonal and cubic phases, at 150ºC the sample presents a fully cubic crystal structure.