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***In-situ & ex-situ* study of protons and electrons irradiations of perovskite solar cells**

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Abstract

The need for low cost photovoltaic solutions is becoming more and more important with the ongoing NewSpace revolution. In this context, alternative solar cell technologies are under the spotlight, in particular perovskites which can reach high specific power. In this study, we investigate the electron and proton radiation hardness of multi-cation mixed halide perovskite cells $\text{Cs}_x\text{FA}_{1-x}\text{Pb}(\text{I}_y\text{Br}_{1-y})_3$. The proton irradiations demonstrate an excellent radiation hardness of the perovskite material but also highlight the degradation of the HTL layer. And the *in-situ* IV measurements under vacuum following the electron irradiations reveals a self-healing phenomena.

I. INTRODUCTION

A wind of change is blowing through the space industry. Low earth orbit satellites constellations are emerging with new needs & objectives: develop / use low cost materials & technologies with sufficient performances for the space environment. With this new approach, the so-called “thin-film cells”, and in particular Perovskites Solar Cells (PSCs), have aroused great interest. Indeed, the latter have great potential: simplified manufacturing, low cost approach and an active material thickness $< 1\mu\text{m}$, which leads to record specific power of 30W/g [1]. In addition, perovskite solar cells went through fast developments in the last 10 years: power conversion efficiency has improved from 3.8% in 2009 [2] to 25.5% in 2021 [3], [4]. On top of that, initial studies show a good radiation hardness for this cell technology against both electron and proton irradiation [5]–[8]. However, the path for bringing this technology out of the lab towards powering real space missions requires developments and in turn comprehension of the detailed degradation mechanisms.

In this context, results presented here focus on the radiation hardness against protons and electrons of multiple cations perovskite solar cells $\text{Cs}_x\text{FA}_{1-x}\text{Pb}(\text{I}_y\text{Br}_{1-y})_3$. Through experimental investigations, the impact of 1 MeV protons until doses up to 5×10^{14} protons/cm² and 1 MeV electrons until 2×10^{15} electrons/cm² are assessed based on microstructural, optical and electrical characterisations. It will be presented here

ex-situ measurements (IV, sheet resistance) which were performed in begin-of-life (BOL) and End-Of-Life (EOL). *In-situ* I(V) measurements were also performed in order to follow the evolution of the cell behaviour under vacuum just after irradiations and therefore in the closest conditions to the space environment.

II. RESULTS AND DISCUSSIONS

To perform this irradiation study, the space environment was simulated with a proton & electron accelerator located at ONERA. In this study, samples were irradiated and measured in ONERA's MIRAGE set up [9] which allows the measurement of solar cells in AM0 after irradiation and under vacuum without venting and thus by avoiding some healing phenomena in presence of oxygen.

At first, a study of PSCs radiation hardness under 1MeV proton irradiations was conducted; Figure 1.a shows the normalised (to the initial value) power conversion efficiency (PCE), measured *in-situ* in the irradiation facility, as function of fluence, up to 5×10^{14} protons/cm². An excellent performance stability is observed up to 1×10^{14} protons/cm², which corresponds roughly to 10 years in LEO environment. However, a clear degradation of the efficiency was observed from 5×10^{14} protons/cm² (roughly 50% losses). This degradation is mostly linked to a pronounced diminution of J_{sc} and FF (not shown here). Thus, non-complete solar cells stacks were first manufactured and irradiated, Glass/ITO/SnO2/Pk and Glass/ITO/SnO2/Pk/PTAA, and then completed with non-irradiated layers : PTAA + Au in the first case and Au electrode in the second case. With this approach, the completed solar cells performance could be measured by *ex-situ* IV measurements, with only a subset of layers from the device being irradiated.

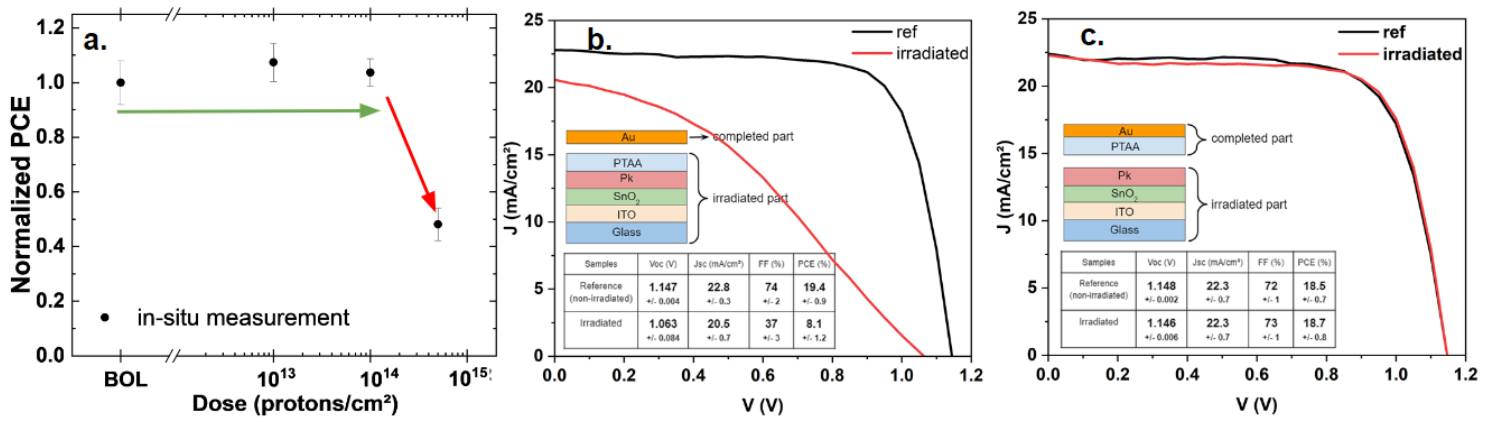


Figure 1. a. evolution of the performances under proton irradiations for several fluences (average of 6 samples under AM0 and under vacuum) : Comparison of JV characteristics for PSCs before and after irradiation @1 MeV 5×10^{14} protons/cm² of selected layers for b. Glass/ITO/SnO₂/Pk/ PTAA c. Glass/ITO/SnO₂/Pk and (averaged on 6 samples)

Figure 2.b and c show the electrical responses of these two irradiated and completed stacks. In the first case, where the PTAA hole transport layer (HTL) has been irradiated, a major degradation of the electrical properties is observed between the irradiated cells and the non-irradiated references which were completed in the same way and at the same time. In the second case, where the stack stopping to Pk layer has been irradiated, no significant change is observed. These experiments thus suggests that the origin of the observed PSCs performance loss for high protons fluences is not linked to the Pk layer degradation but rather to the PTAA HTL that strongly impacts the opto-electronic properties of cells. In addition, transmission line measurement characterizations (not shown here) on the irradiated PTAA layer has revealed an increased resistance of this layer, which also confirms its degradation.

This first study has highlighted the radiation hardness of the perovskite material to this protons environment, and also the sensitivity of PTAA HTL: most of the performance decrease observed at high protons doses is linked to the PTAA degradation.

As a second study, we have investigated the radiation hardness of the same type of PSCs under electron flux. Figure 2 shows the *ex-situ* JV characteristics of PSCs with electrons particles at 1 MeV energy and for doses up to 2×10^{15} electrons/cm². As on can see, the cell performance is mostly impacted by Jsc & Voc losses, while the FF remains similar. However unlike the first protons irradiation study presented above, the 1MeV electrons here go through the all glass substrate (non space qualified), and optical transmission losses are happening. Indeed, BOL & EOL measurements of glass substrate transmission (not shown here) for these same doses have shown losses due to darkening effect. Thus, the observed Jsc decrease is at least partially related to electron degrading the glass substrate optical properties.

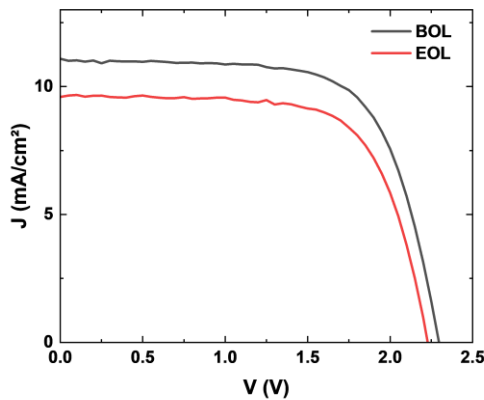


Figure 2. *Ex-situ* J(V) curves before and after irradiation with 2×10^{15} electrons/cm² @ 1 MeV under AM1.5G

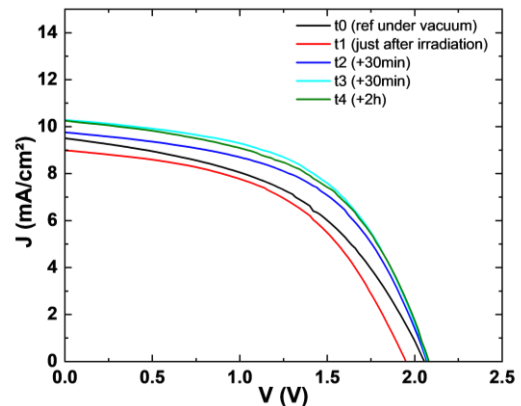


Figure 3. *In-situ* J(V) curves after irradiation 2×10^{15} electrons/cm² @ 1 MeV under AM0

The comparison with protons irradiation results reveals two interesting points. The first is that the general degradation behavior under the tested electron irradiation conditions for these PSCs differs from the one observed with the tested proton irradiations. Indeed, with electrons irradiation, no significant FF degradation is observed. Thus the whole stack of active layers (except glass) seems to resist better to electron flux.

The second point concerns the evolution of the electrical properties of cells under vacuum just after the irradiation. Figure 3. shows the measurements of J(V) carried out under vacuum just before and after irradiation at 2×10^{15} electrons/cm² @ 1 MeV. First, a degradation of the electrical properties (black curve to red curve)- is observed, with an overall decrease of Jsc & Voc during the first measurement after irradiations. The following measurements then clearly show a self-healing phenomenon under vacuum: the performance increases until a efficiency stabilisation after ~3 hours. These phenomena, monitored *in-situ* under vacuum is not be linked to air exposure and thus might also happen in real space conditions. Moreover, these self-healing phenomena were not observed during proton irradiation in the same conditions (environment & time between measures). In addition, it is observed that performances after irradiation are better than the initial performance under vacuum. It should be noted that it would be interesting to carry out SEM images before and after irradiation in order to see if a macroscopic rearrangement of the material is visible.

III. CONCLUSION

In conclusion, stacks of PSCs were irradiated with electrons and protons particles in order to monitor and compare their radiation hardness.

Firstly, the proton irradiations has shown an excellent resistance of the perovskite material itself, but also to a pronounced degradation of the PTAA HTL. It will be therefore interesting to investigate new HTL materials in similar conditions.

Secondly, electron irradiations have shown that the cells and its photoactive materials do not degrade significantly for this electrons irradiations conditions. In fact, no significant degradation of the HTL was observed in this case, and most of the Jsc decrease observed is linked to glass substrate optical darkening. Moreover, cells self-healing phenomena were

observed *in-situ*, with final performances surpassing those initially obtained under vacuum. Further work could focus on investigating microstructural and lifetime properties modification with irradiations.

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