A Research on Perovskite Solar Cells' Tolerance under Proton Radiations

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Perovskite solar cells (PSCs) have been getting significant attention in the photovoltaic community because of their high power conversion efficiency, solution processability, large-area device fabrication, and low production cost [1-3]. Recently, PSC technology is being studied for space applications to overcome its disadvantages in instability in normal terrestrial ambient air conditions containing moisture and oxygen [4]. The superiors combining with their high specific power (power per weight) [5] as well as the facile low-temperature solution processibility for in-situ fabrication in spaceships make PSCs a promising replacement for currently used PV technologies in space applications. However, the space radiation hardness of PSCs should be further studied before they can be applied in space missions.

To the date, PSCs were evaluated at a few specific particle energies and fluences such as: 1MeV electron [6-7], 50keV [4&7], 300keV [8], 7MeV [9], 68MeV [10-11] proton. This is most likely due to the testing facility, cost or time limitations (or all three), but it limits the reliability of the data and makes inter-study comparisons difficult. Furthermore, the main purpose of these evaluations is to develop optimized cells for space mission. It is a fact that there are a wide range of perovskite compositions, charge transport materials, and device architectures, which makes it impractical to experimentally evaluate radiation tolerance of all the variations of PSCs. Therefore, at this early stage of investigating PSCs for space applications, it is important to identify what parameters are most important when choosing materials and device architectures for better radiation tolerance.

In this work, we introduce a new simulation-based method to predict the degradation of perovskite solar cells under proton radiation and then confirm our approach via a radiation test. Depth-dependent defect profiles (create by proton radiation) is generated by ion scattering simulations as a function of proton energy and fluence, which are then incorporated into an optoelectronic simulation to predict solar cell degradation of 2 kinds of perovskite compositions.

Firstly, to see the effect of A-cation side to the radiation tolerance of perovskite materials: all inorganic perovskite ($CsPbI_2Br$) and hybrid perovskite ($FA_{0.86}MA_{0.14}PbI_3$), we simulate the perovskite layers only to isolate other layers' impacts. It is note that Br and I have the same threshold displacement energy in the simulation model and also, they have similar atomic mass (comparing to organic counterparts). Therefore, the differences between 2 selected perovskites are mainly from A-cation head. They are simulated using the freely available Stopping and Range of Ions in Matter (SRIM)/Transport of Ions in Matter (TRIM) software tools, and the results are shown in Figure 1. From the results, we observe that low proton energies (<50keV) cause more damage on the light-weight materials while higher energy ones are more harmful to materials with higher average atomic mass. It is notice that the proton radiation damage in space is mainly contributed by protons in the range from 10keV to 1MeV with abundant fluences, and it is also the range cause highest damage on photovoltaic systems on space missions. Within this range, we clearly see that materials with higher atomic mass receive more damage than lighter materials.

Before going further, we would like to clarify our assumptions in this work that we only consider the defects within the active layer and assume that all other layers remain the same for a fair comparison between different perovskite components. So, the defect profiles within the active layers are added into the photoelectronic model (COMSOL simulation) [12]. With different energies and fluence of protons, we obtain different degradation levels for the studying PSCs. We applied the method for fully inorganic perovskite ($CsPbI_2Br$) and hybrid perovskite ($FA_{0.86}MA_{0.14}PbI_3$) with experiment-based are 17.4% and 24.1% [13-14], respectively. The results are shown in Figure 2 with a bit better tolerance for organic-cation one. Overall, the simulations predict that both types of PSCs retain >60% of their initial efficiencies after a 100 keV proton radiation at fluence of 1e14

particle/cm2, which are far higher than that of III-V solar cells – currently used for space mission, which degrades at the same level for proton fluences 3-4 order of magnitude lower.

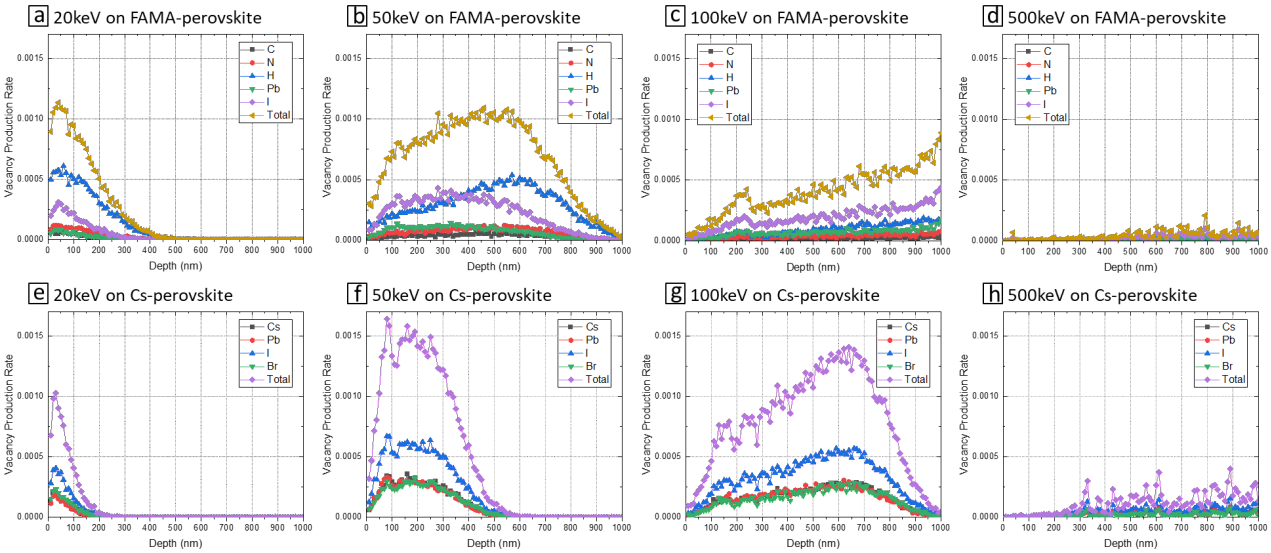

Figure 1. Vacancy production rate within perovskite layers ($CsPbI_2Br$ and $FA_{0.86}MA_{0.14}PbI_3$) with 100nm of gold on top

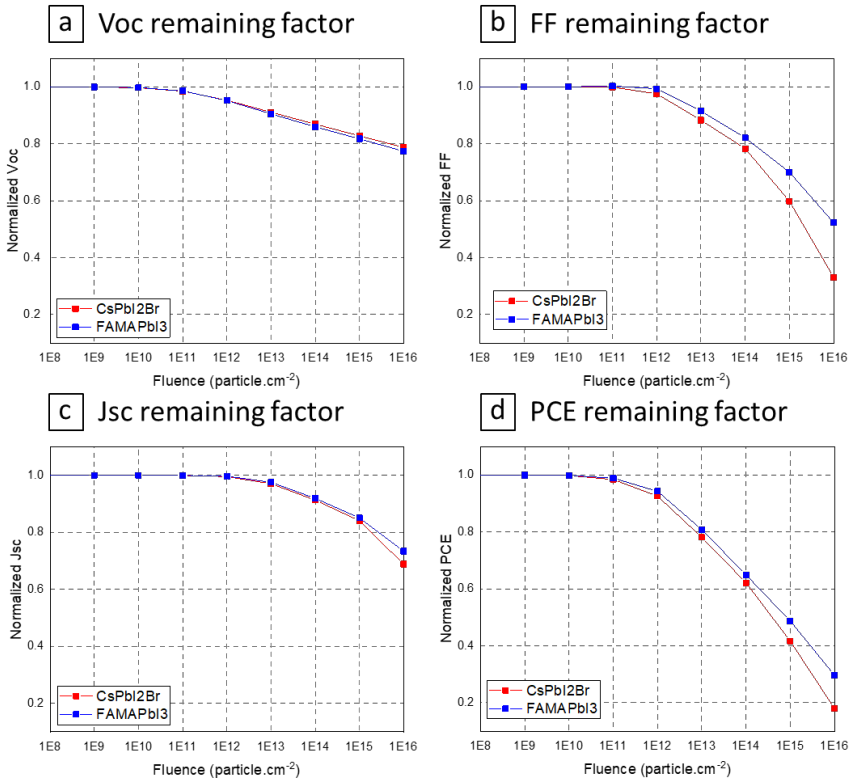

Figure 2. PSC device performance comparison of inorganic and organic perovskites: $CsPbI_2Br$ vs. $FA_{0.86}MA_{0.14}PbI_3$ under 100keV proton radiation

Finally, we confirm our approach by conducting a radiation test. However, due to facility limitation, the chosen proton energy is 10MeV with 3 different fluences of 1e12, 1e13, and 1e14 particle/cm2. The PSC structure is Glass/ITO/SnO_2/$Cs_{0.1}FA_{0.9}PbI_3$/Spiro-OMeTAD/Au which is well established in our lab with the initial efficiency >24% at AM1.5G. The J-V characterization of the samples

before and after 10MeV proton radiations are shown in Figure 3a while the remaining factor of all opto-electronic parameters for both simulation and experiment results are compared in Figure 3b.

After exposure to 10MeV proton radiation at fluence of 1e14 particle/cm^2, the PSC still remains its 89% initial PCE. It's noted that recently published works show quite low initial efficiency in range from 4.7% to 18% [6-11] which are far lower than the state-of-the-art PSC (>25%). The initial efficiencies show the initial conditions of the samples without radiation-induced defects; thus, low performance samples (possibly with higher level of defects) might show less contribution of additional defects on the degradation of the cells during irradiation. As far as we know, the initial efficiency of PSC in this work is the highest one among published works. The comparison with recent published works is shown in Table 1.

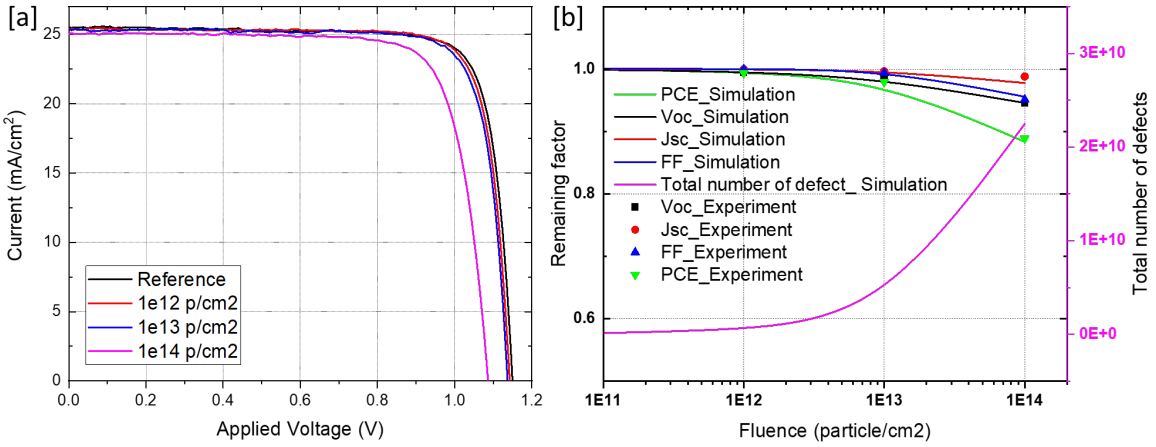

Figure 3. a) Experimental degradation of the PSC under 10MeV proton radiation. b) Comparison between simulation and experiment results for the degradation of the PSC under 10MeV proton radiation.

Table 1 Comparison between published data for proton radiation tests

Structure	Energy	Fluence	Initial effi.	Remaining	Ref.
ITO/TiO_2/$MAPbI_3$/P3HT/Au	150keV	1e12	4.8%	95%	6
		1e13		80%	
		1e14		70%	
ITO/c-TiO_2/mp-TiO_2/$MAPbI_{3-x}Cl_x$/P3HT/Au	50keV	1e12	4.8%	100%	7
		1e13		104%	
		1e14		110%	
ITO/polyTPD/PFN-Br/$FA_{0.8}Cs_{0.2}PbI_{2.4}Br_{0.6}Cl_{0.02}$/C60/$SnO_x$/ZTO/ITO/$Al_2O_3$	50keV	1e12	5.6%	77%	8
	300keV		5.8%	147%	
	2.5MeV		5.4%	117%	
ITO/SnO_2/$Cs_{0.15}FA_{0.85}PbI_3$/PTAA:Spiro/Au	7MeV	1e13	16.3%	48%	9
ITO/SnO_2/$Cs_{0.15}FA_{0.85}PbI_3$/PTAA/Au			14.9%	57%	
ITO/SnO_2/$Cs_{0.15}FA_{0.85}PbI_3$/PTAA:C8BTBT/Au			18.0%	57%	
ITO/polyTPD/PFN/$FA_{0.6}Cs_{0.3}DMA_{0.1}PbI_{2.4}Br_{0.6}$/LiF/C60/PEIE/AZO/ITO/PEDOT:PSS/$FA_{0.75}Cs_{0.25}Sn_{0.5}Pb_{0.5}I_3$/C60/BCP/Au	68MeV	2e12	15.6%	97%	10
		1e13		94%	

ITO/PEDOT:PSS/$MAPbI_3$/PC61BM/BCP/Ag	68MeV	1e12	4.7%	121%	11
$ITO/SnO_2/Cs_{0.1}FA_{0.9}PbI_3$/Spiro/Au	10MeV	1e12	24.1%	99%	This
		1e13		98%	
		1e14		89%	

It can be clearly seen from Table 1 that samples with low initial efficiencies (~5%) show a strange behaviour as their PCEs even increase after radiation. This might be a side effect of radiation – self-annealing which eliminates existing defects in the samples during irradiation. In this test, to avoid heating, we chose the flux as low as possible within the machine limitation (1e9p/cm^2.s). The final results show a well fit with the simulation results earlier as shown in Figure 3b.

With this result, we confidently conclude that our simulation approach could predict the degradation under proton radiation based on the combination of ion scattering and optoelectronic device simulations with a good agreement with experiment results. This method could save a lot of time and avoid a need of long conducting time and extremely expensive resources for radiation tests. Additionally, the PSCs remains 89% of its initial efficiency after exposure 10MeV proton radiation at a fluence of 1e14 particle/cm^2.

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