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# Understanding of Open Circuit Voltage Loss Mechanism in Perovskite Solar Cells

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## Abstract

**In order to study the open circuit voltage (Voc) loss mechanism in the perovskite solar cells, the radiative open circuit voltage ($V_{oc}^{rad}$ = 1.313 V) was estimated by combining the detailed balance theory and Van-Roosbroeck and Schockely Relation. Relationships between the Voc and the carrier lifetime of the devices are revealed. Based on the lifetime estimated by Time-Resolved Photoluminescence (TRPL) measurement, the calculated ideal Voc (1.223 V), which limited by bulk recombination, is much higher than the measured Voc (1.068 V) under one sun illumination. It identified that the dominant Voc loss mechanism of the devices is non-radiative recombination in the interface.**

## 1. Introduction

Perovskite solar cells (PSCs) have attracted much attention due to the fast progress of their power conversion efficiency (PCE), which from 3.8% to 25.2% in the past 10 years.[1] However, the efficiency of the real perovskite solar cell devices (for example, $MAPbI_3$) is much lower than the Shockley–Queisser (SQ) limit. SQ limit is the efficiency limit of the single-junction solar cell based on the black body model in which only the radiative recombination loss included. The SQ limit of $MAPbI_3$ (Band gap Eg = 1.6 eV) perovskite solar cell is calculated to be 30.5%. However, the efficiency of perovskite solar cell devices in this work is only around 16%. The high open circuit voltage (Voc) loss in the PSC devices is one of the most severe problems which causes a high PCE loss. In order to improve the properties of the perovskite solar cell devices, the PCE loss mechanism needs to be understood. In this work, the Voc loss mechanism will be discussed based on the perovskite solar cell devices.

## 2. Device fabrication

In this work, the inverted structure perovskite solar cell is used for research. The structure of the solar cell devices is FTO/$NiO_x$/$MAPbI_3$/PCBM/BCP/Ag.

The hole transport layer (HTL) $NiO_x$ was prepared by a spray pyrolysis process. The $MAPbI_3$ layer was deposited on the NiOx deposited FTO substrate by one step anti-solvent dripping method. PCBM and BCP were deposited on the top by the spin coating process. At last, the Ag was deposited on the fabricated devices by thermal evaporation.

The PCE of the completed solar cell device is 16.64% (Jsc = 19.6 mA/cm$^2$, Voc = 1.068 V, FF = 0.79).

## 3. Results and discussion

The transmittance (T) and reflectance (R) were measured for the $MAPbI_3$ layer. The absorption coefficient ($\alpha$) of the $MAPbI_3$ can be transferred based on the followed function:[2]

$$\alpha = \frac{1}{d}\ln\left[\frac{(1-R)^2}{T}\right] \qquad (1)$$

d: Thickness of the materials

The absorption coefficient spectrum is shown in Fig. 1. Because the accurate Urbach tail can not be detected from the UV-Vis measurement, the Urbach tail (15 meV) was extracted from the fitting EQE data (inserted figure in Fig.1). The radiative Voc limit of the subsistent $MAPbI_3$ perovskite solar cell can be estimated from the followed equation:[3]

$$V_{oc}^{rad} = \frac{kT}{q} ln\frac{J_{sc}}{J_0^{rad}} = \frac{kT}{q} ln\frac{J_{sc}{n_r}^2}{qd{n_i}^2B} \qquad (2)$$

$n_i$: Intrinsic carrier density; B: Radiative recombination constant; $n_r$: Refractive index.

The radiative recombination rate is shown in Fig. 1 and the radiative recombination constant was calculated with the absorption coefficient based on the Van-Roosbroeck and Schockely Relation:[2]

$$B = \frac{R}{{n_i}^2} = \frac{g\cdot{n_r}^2}{{n_i}^2}\int_0^\infty \frac{\alpha(E)\cdot E^2}{e^{\frac{E}{kT}}-1}dE \qquad (3)$$

R: Integrated radiative recombination rate.

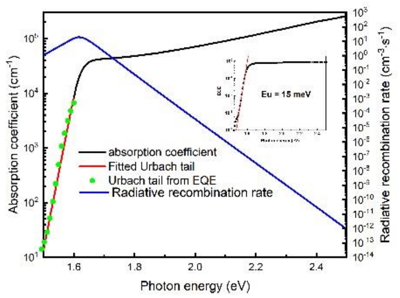


Fig. 1. Absorption coefficient and radiative recombination rate of the MAPbI3 materials

The radiative recombination constant and $V_{oc}^{rad}$ can be estimated to be $2.06 \times 10^{-10}\ cm^{-3} \cdot s^{-1}$ and 1.313 V, respectively, From the equation (2)-(3). The relationship between the radiative recombination constant and the radiative lifetime is shown as the followed equation:

$$\tau_{rad} = \frac{1}{B\Delta n} \qquad (4)$$

$\Delta n$: Photo generated carrier density.

Equation (5) defines the relationship between Voc and effective lifetime of devices. It can be derived from equation (2) and (4) in which the radiative lifetime can be replaced by effective lifetime because of consideration of the non-radiative recombination.

$$V_{oc} = \frac{kT}{q} ln\left(\frac{J_{sc}\Delta n {n_r}^2}{qd{n_i}^2} \cdot \tau_{eff}\right) \qquad (5)$$

$\tau_{eff}$: Effective lifetime

Under 1 sun illumination, the photogenerated carrier density of the $MAPbI_3$ is estimated to be around $10^{15}\ cm^{-3}$.[4] The radiative recombination lifetime of $4.84\ \mu s$ can be obtained. In ideal cases, the Voc of these solar cell devices can reach the radiative limit of 1.313 V because non-radiative recombination does not exist. The carrier lifetime of the solar cell should be $4.84\ \mu s$. The carrier lifetime of the real devices is shorter because of the existence of non-radiative recombination. Typically, the non-radiative recombination happens in the bulk and interface. Time-Resolved Photoluminescence (TRPL) measurement employed for $MAPbI_3$ thin film for studying the influence of the non-radiative recombination mechanism to the Voc loss. From the TRPL spectrum (shown in Fig.2), the lifetime which was fitted by the slow component of 149 ns is assumed to be the bulk lifetime ($\tau_{bulk}$) of $MAPbI_3$. In the condition that there is no interface recombination in the solar cell devices, the Voc limited by bulk recombination so-called bulk limit ($V_{oc}^{bulk}$) can be estimated to be 1.223 V from equation (5).

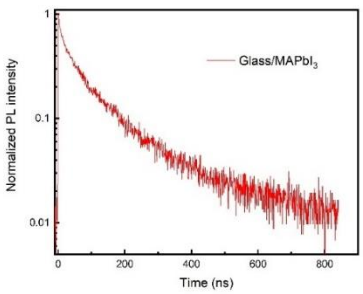


Fig. 2. TRPL spectra of the MAPbI3 film.

The effective lifetime ($\tau_{eff} = 0.37\ ns$ corresponding Voc of 1.68 V from the experimental result) can be estimated from equation (5). The effective lifetime which including bulk recombination and interface recombination can be described as the equation (6):

$$\frac{1}{\tau_{eff}} = \frac{1}{\tau_{bulk}} + \frac{1}{\tau_{interface}} \qquad (6)$$

The interface lifetime ($\tau_{interface}$) of 0.37 ns can be estimated. Fig. 3 shows the variation of the IV curve in different conditions. In SQ model, the $V_{oc}^{SQ}$ is estimated to be 1.324 V. Based on the real devices which were used in this work, the $V_{oc}^{rad}$ of 1.313 V can be calculated. The Voc loss of 0.09 V was estimated from $V_{oc}^{rad}$ to $V_{oc}^{bulk}$, corresponding with external luminescence efficiency ($\eta_{ext}$) of around 3 %. The Voc of the real device was measured to be 1.068 V in which the $\eta_{ext}$ of around 0.01 % was estimated. It can identify that most of the Voc loss is coming from non-radiative recombination in the interface, which means the recombination in the interface is dominant in Voc loss mechanism in this device.

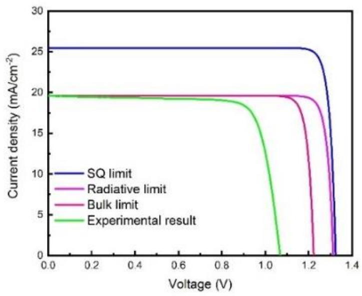


Fig. 3. IV curve in different conditions.

## 4. Conclusions

In this work, the radiative recombination constant of $2.06 \times 10^{-10}\ cm^{-3} \cdot s^{-1}$ was calculated from the experimental absorption coefficient spectrum and the radiative lifetime of $4.84\ \mu s$ was estimated. The radiative Voc limit of 1.313 V and bulk Voc limit of 1.223 V were estimated based on the real devices. From the calculation result, Voc loss is dominant by interface recombination can be identified. To improve the Voc of the PSC devices approaching the bulk limit of 1.223 V, reducing the interface recombination is the main objective. The interface passivation is one of the most important processes to improve Voc of the devices. The reduction of the bulk recombination should be considered to improve the Voc to approach the radiative limit (1.313 V). In such a way, the passivation of the bulk defect will be very important for improving the Voc approaching the SQ limit of the PSC devices.

## References

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