Solar Energy Materials and Solar Cells 174 (2018) 206–213

Contents lists available at ScienceDirect

Solar Energy Materials and Solar Cells

journal homepage: www.elsevier.com/locate/solmat

Quantifying energy losses in planar perovskite solar cells

Yun Da[a], Yimin Xuan[a,b,*], Qiang Li[a]

[a] School of Energy and Power Engineering, Nanjing University of Science & Technology, Nanjing 210094, China
[b] School of Energy and Power Engineering, Nanjing University of Aeronautics and Astronautics, Nanjing 210016, China

ARTICLE INFO

Keywords:
Perovskite solar cells
Loss analysis
Optical absorption
Electrical recombination
Coupled optical and electrical modeling

ABSTRACT

Perovskite solar cells are considered as an up-and-coming substitute for the next generation solar cells. Despite of significant increase of its photon-electric conversion efficiency, a definitive direction for further increment remains ambiguous. In this paper, we quantitatively assess the energy losses in planar perovskite solar cells in terms of the underlying physical mechanisms. The coupled optical and electrical modeling is developed to explore the working principle of the perovskite solar cells. A comparison between simulation results and experimental data under different operating conditions is investigated to elucidate the reliability of the device modeling. With the aid of the accurate device modeling, we explore the energy loss mechanisms in planar perovskite solar cells. Five energy loss mechanisms are quantified, such as thermalization loss, below bandgap loss, optical loss, recombination loss, and spatial relaxation loss. The effects of the optical properties, carrier diffusion length, surface recombination velocity, and series resistance on the performance of the perovskite solar cells are analyzed to identify the dominant loss contributors limiting the power conversion efficiency of the perovskite solar cells. Our results indicate that more efforts should be paid to enhance the optical absorption of the perovskite layer, improve the surface passivation, and reduce the series resistance. Based on the theoretical analysis, a roadmap to promote the device performance of the perovskite solar cells is summarized. Our work provides a detail guideline for design and innovation of perovskite-based device.

1. Introduction

The development of inorganic-organic halide perovskite solar cells is booming in the past several years since it was firstly announced by Kojima et al. in 2006 [1]. The first hybrid lead halide perovskite solar cells appeared in the form of dye-sensitized solar cells (DSSCs) with the power conversion efficiency (PCE) of 2.2%. Three years later, the same group of Kojima et al. improved the PCE to the level of 3.8% [2]. Their work gained much attention and greatly promoted fascinating progresses in perovskite solar cells and resulted in an unprecedented rise rate of PCE. Currently, the record efficiency of perovskite solar cells reached to 22.1% [3]. The perovskite materials possess excellent optical and electrical characteristics, such as direct band-gap, high light absorption coefficient, high charge carrier mobility, long charge carrier diffusion length, and long electron-hole recombination lifetime [4–8], making them desirable for high efficiency thin-film solar cells. Hence, depending on these properties, perovskite solar cells become a star in the photovoltaic realm and attract tremendous research interests. It has been experimentally demonstrated that the PCE of the perovskite solar cells depends on the morphology of film, device configuration, interface losses, and crystal quality [9–15]. However, the key factors affecting the performance of the perovskite solar cells remain challenging. The theoretical model to clarify these influencing factors is still lacking. Thus, there is an urgent requirement to develop an accurate theoretical model to describe the working mechanisms of the perovskite solar cells and to examine the extent of the impact of these factors.

To correctly establish the device modeling, it is necessary to understand the operation principle of the perovskite solar cells. It is well known that the perovskite solar cells were initially derived from the DSSCs. The working mechanism may be similar to DSSCs or bulk heterojunction organic solar cells at first impression. But the subsequent investigations denied this conjecture [16,17]. Hu et al. provided the direct evidence to reveal the non-excitonic nature of the perovskites by comparing the exciton dissociation behavior of perovskite materials to conventional excitonic semiconductors [18]. Additionally, the small exciton binding energy of the perovskite crystal further confirmed the fact that the free charge carriers are formed in perovskite solar cells under operation conditions [19]. Therefore, the perovskite solar cells can be modeled as inorganic photovoltaic devices [20]. It is reasonable to apply the device physics modeling of inorganic cells to perovskite solar cells. Recently, many researchers have established one-dimensional (1D) device modeling and successfully explained the

* Corresponding author at: School of Energy and Power Engineering, Nanjing University of Science & Technology, Nanjing 210094, China.
E-mail address: ymxuan@mail.njust.edu.cn (Y. Xuan).

http://dx.doi.org/10.1016/j.solmat.2017.09.002
Received 24 June 2017; Received in revised form 22 August 2017; Accepted 4 September 2017
Available online 12 September 2017

fundamental role of the influencing factors in perovskite solar cells [21–24]. Unfortunately, 1D simulation is restricted by its simplified process, such as lacking the ability to entirely characterize the optical properties, failing to consider the influence of microstructure in the solar cells. Thus, it is required to develop an sufficiently accurate device modeling for the perovskite solar cells.

Meanwhile, energy loss analysis is an impressive procedure to understand the working principle of the solar cells as well. It can help us to comprehend where the lost energy went and how to improve the PCE of the photovoltaic cell. At present, two typical device architectures including mesoporous scaffold structure and planar junction configuration have already succeeded in obtaining high efficiency for perovskite solar cells. In terms of the simple structure and facile reproducibility, planar heterojunction configuration is preferred to mesoporous scaffold structure because it has prospect of industrial application due to roll-to-roll production [25]. It is generally known that there exist intrinsic loss and extrinsic loss processes in the solar cells. Due to the non-excitonic nature of the planar perovskite solar cells, five kinds of energy loss processes occurs inside the device including thermalization loss, below bandgap loss, optical loss, recombination loss, and spatial relaxation loss. Nevertheless, till today, the energy loss in planar heterojunction perovskite solar cells is not yet quantified and the dominant energy loss mechanisms remain ambiguous. Of particular note is that the current PCE of the perovskite solar cells is far less than the theoretical maximum value. For example, the radiative efficiency of the $CH_3NH_3PbI_{3-x}Cl_x$ solar cell, which is a typical perovskite material and has the bandgap of 1.55 eV, is 31.5% according to Shockley-Queisser limit under AM1.5G illumination conditions [26,27]. The energy loss analysis can quantitatively illustrate the gap between the current PCE and the fundamental efficiency limit, pointing out the specific direction for targeted improvement of the performance.

In this work, we perform a comprehensive analysis of energy losses affecting the performance of the $CH_3NH_3PbI_{3-x}Cl_x$ perovskite-based solar cells under standard AM1.5G sunlight irradiance conditions by means of coupled optical and electrical modeling. Our theoretical model is validated by comparing the simulation results with the experimental data. On the basis of the accurate device modeling, each energy loss mechanism is properly considered in the planar perovskite solar cells and the value of each loss is quantitatively given. Extrinsic losses including optical losses and electrical losses, which can be determined by the cell configuration and the fabrication process, have been carried out in detail. Firstly, the device modeling is described in brief and the simulation results are compared with the experimental data to validate the device modeling. Then, the category of the energy losses is clarified and the corresponding analytical expression is presented. Subsequently, each energy loss mechanism is visibly visualized in a novel graphic representation, which involves all the incident solar radiation energy. We systematically analyze the extrinsic energy loss mechanisms in perovskite solar cells, finding out the dominant energy loss channel. Finally, we issue the guidelines on how to design and optimize for realization of the high efficiency perovskite solar cells and promote a roadmap to extend the PCE to 23%.

2. Device modeling and validation

In this work, the coupled optical and electrical modeling is carried out to investigate the optical and electrical performance of the perovskite solar cells. This numerical method intends to separately address the optical and electrical properties of the solar cells. More details about this method can be found in our previous publications [28–30]. For the optical simulation, Maxwell's equations are the foundation to describe the light-matter interaction and enable to calculate the spectral features.

$$\begin{cases} \nabla \times \overrightarrow{H} = \frac{\partial \overrightarrow{D}}{\partial t} \\ \nabla \times \overrightarrow{E} = -\frac{\partial \overrightarrow{B}}{\partial t} \\ \overrightarrow{D} = \varepsilon \overrightarrow{E} \\ \overrightarrow{B} = \mu \overrightarrow{H} \end{cases} \quad (1)$$

where $\overrightarrow{H}$ is the magnetic field, $\overrightarrow{D}$ is the electric displacement field, $\overrightarrow{E}$ is the electric field, $\overrightarrow{B}$ is the magnetic flux density, ε is the complex permittivity, and μ is the complex permeability. The optical performance of the perovskite solar cells can be obtained via the Finite Difference Time Domain (FDTD) algorithm, which is a rigorous electromagnetic calculation for solving the Maxwell's equations. By assuming that each absorbed photon with energy greater than the bandgap of the semiconductor can generate an electron-hole pair, the photogeneration rate can be presented as

$$G(r) = \int_{E_g}^{\infty} \frac{\omega \mathrm{Im}(\varepsilon) |\overrightarrow{E}(r, E_\lambda)|^2 PFD_{AM1.5}(E_\lambda)}{2} dE_\lambda \quad (2)$$

where E_g is the bandgap of the photovoltaic material, ω is the angular frequency of the incident photon, $\mathrm{Im}(\varepsilon)$ is the imaginary part of the permittivity, E_λ is the photon energy, and $PFD_{AM1.5}$ is the photon flux density under AM1.5G conditions.

For the electrical simulation, the device characteristics are governed by the semiconductor equations including Poisson, continuity, and drift-diffusion equations.

$$\begin{cases} -\nabla \bullet (\varepsilon \nabla \phi) = q(p - n + N_D - N_A) \\ \frac{\partial n}{\partial t} = \frac{1}{q} \nabla \bullet J_n + G - R \\ \frac{\partial p}{\partial t} = -\frac{1}{q} \nabla \bullet J_p + G - R \\ J_n = -q\mu_n n \nabla \phi + qD_n \nabla n \\ J_p = -q\mu_p p \nabla \phi - qD_p \nabla p \end{cases} \quad (3)$$

where ϕ is the electrostatic potential, ε is the dielectric constant of the semiconductor, $n(p)$ is the electron (hole) concentration, $N_D(N_A)$ is the donor (acceptor) doping concentration, q is the elementary charge, $J_n(J_p)$ is the current density of electron (hole), $\mu_n(\mu_p)$ is the electron (hole) mobility, $D_n(D_p)$ is the electron (hole) diffusion coefficient, G is the photogeneration rate extracted from optical simulation, and R is the total carrier recombination. There are four types of recombination mechanisms considered in the electrical simulation with the expression of

$$\begin{cases} R = R_{rad} + R_{Aug} + R_{SRH} + R_{surf} \\ R_{rad} = B(np - n_i^2) \\ R_{Aug} = (C_n n + C_p p)(np - n_i^2) \\ R_{SRH} = \frac{np - n_i^2}{\tau_p(n + n_t) + \tau_n(p + p_t)} \\ R_{surf} = \frac{np - n_i^2}{\frac{1}{S_p}(n + n_{ts}) + \frac{1}{S_n}(p + p_{ts})} \end{cases} \quad (4)$$

where R_{rad} is the radiative recombination, B is the radiative recombination coefficient, R_{Aug} is the Auger recombination, $C_n(C_p)$ is the electron (hole) Auger recombination coefficient, R_{SRH} is the Shockley-Read-Hall recombination, $\tau_n(\tau_p)$ is the electron (hole) lifetime, R_{surf} is the surface recombination, $S_n(S_p)$ is the surface recombination velocity of electron (hole), and n_i is the intrinsic carrier concentration. Therefore, the coupled optical and electrical modeling can accurately elucidate the device performance of the solar cells.

In order to validate our theoretical model, the experimental data reported by Liu et al. [31] was used as a comparison. The device configuration reported by Liu et al. is a planar heterojunction p-i-n structure with thin film stacks composed of glass/FTO(fluorine-doped tin oxide, 500 nm)/ compact TiO_2(titanium dioxide, 50 nm)/ $CH_3NH_3PbI_{3-x}Cl_x$(methylammonium lead tri-iodide perovskite with