

Enhancing absorption in thin film organometal trihalide perovskite solar cell by photon recycling

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Abstract

Over the past few years thin film planar heterojunctions solar cells have made much progress as a low cost with high power conversion efficiency photovoltaic devices. Among the materials used in fabricating such solar cells organometal trihalide perovskite (MAPbI_3) has proven to be a promising absorber material due to cheaper organic-inorganic perovskite compounds, abundantly available in nature, ease of fabrication and compatible with low temperature large scale processing. In addition to the efficient absorption in ultra-violet range the material possess intriguing optoelectronic properties such as high crystallinity, high carrier mobility and large carrier diffusion lengths. Currently, the highest power conversion efficiency achieved by such perovskite solar cells is only 23.9% as reported in 2017. In this work we demonstrate a thin film organometal trihalide perovskite solar cell with hybrid interfaces between different materials which are selected after extensive study to achieve reduced recombination and high performance. Further, the absorption of the incident solar spectrum is enhanced by incorporating a 1D photonic crystal at the bottom of the cell facilitating the photon recycling process. The proposed solar cell parameters are numerically computed using rigorous coupled wave algorithm through SYNOPSIS RSOFT CAD tool. The thickness of each layer of the structure is optimized using MOST scanning and optimization module of RSOFT CAD tool to achieve highest power conversion efficiency at minimum device thickness ($\sim 2 \mu\text{m}$). The power conversion efficiency thus obtained is 25.2% with a fill factor of 86.3% at AM 1.5, which is very promising. This demonstrates the remarkable potential of the proposed design to achieve efficiencies over 20% and compete with the existing crystalline silicon photovoltaic market. Copyright © 2018 VBRI Press.

Keywords: Perovskite, solar cell, thin film, photonic crystal, photon recycling.

Introduction

In recent years, the need for clean, cheap and abundant source of energy has stirred large research efforts in the field of photovoltaic (PV) to make low cost and high power conversion efficiency (PCE) solar cells, particularly in thin film solar cells (TFSCs). New materials are being explored that are capable of absorbing maximum range of the solar spectrum but must be cheap and promising PV candidate for large scale manufacturing. Among such materials organometal trihalide perovskite material (MAPbI_3) has captivated much attention due to its intriguing light harvesting and other opto-electronic properties such as large carrier diffusion lengths, high carrier mobility and high crystallinity. Perovskite refers to a class of crystalline materials that can be easily synthesized to mock the properties of the naturally occurring mineral perovskite. Perovskites are also incredibly cheap compared to silicon, and they are compatible with high volume, low cost manufacturing processes. Even without perovskites, the cost of solar power is already

competitive with fossil fuels in some markets. If and when perovskites break through commercially, costs will drop even further. The most appealing aspect of them is that they combine the characteristic of both thin-film and organics [1]. Perovskite solar cells (PSCs) have made a dramatic success that only in few years the PCE of PSCs has reached more than 20% (**Fig.1**). The record efficiency of PSCs till date is 23.9% as claimed by IMEC research institute at Belgium in 2017 [2] just after the few weeks of reporting second highest PCE of PSC by WS. Yang et al. at Ulsan National Institute of Science and Technology, Korea [3]. However, currently there are some major issues with the PSCs that are yet to be addressed such as, first, the operational lifetime of the PSC is much less as compared to other PV material solar cells such as silicon solar cells. Second major issue is the use of lead in perovskite compounds. Although very small quantity of lead is used, it's still a big problem as it is the possible threat for the contamination of rain water during rains. There are researches going on the above drawbacks to make it

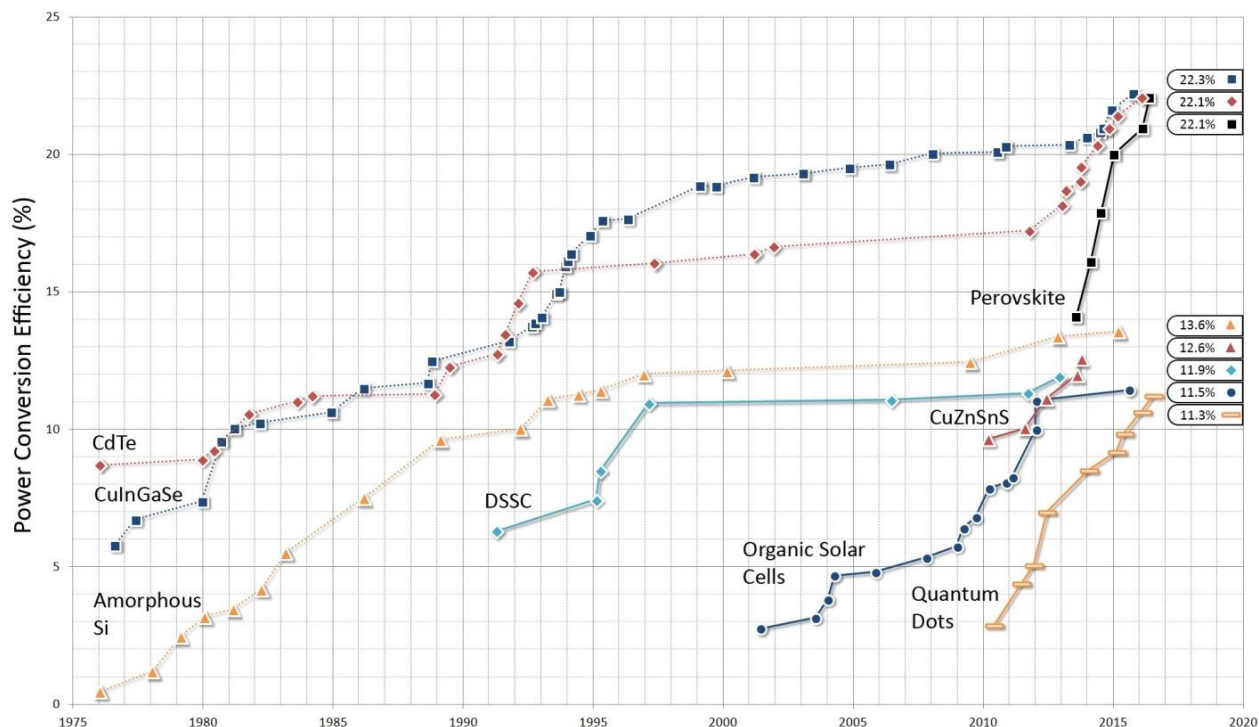


Fig. 1. Rapid growth in PCE of perovskite solar cells compared to other types of photovoltaic materials [41].

able to compete with the existing silicon market. Seon Joo Lee *et al.* in 2016 demonstrated one such perovskite compound (Formamidinium tin halide) replacing lead (Pb) with tin (Sn) and methylammonium (CH_3NH_3) with formamidinium ($\text{CH}(\text{NH}_2)_2$) which showed better stability for a period of 100 days under standard conditions and humidity level of $\sim 25\%$ [4, 5]. The efficiency of such cells can be further increased by using metamaterials such as photonic crystals. Photonic crystals are artificial structures characterized by alternate arrangement of dielectrics that can be used to control the light as desired and hence there are widely used as light trapping structures in solar cells [6]. In this paper we have proposed a hybrid perovskite cell consisting of photonic crystal acting as a wavelength selective reflector at the back of the cell which helps in recycling the photons back into the cell to enhance the overall absorption of the thin film perovskite solar cell. We have implemented a 1D PhC to keep the device thin with ease of fabrication for large scale production.

Device structure

The proposed solar cell structure (Fig. 2) consists of methylammonium lead iodide (MAPbI_3) as a perovskite absorber layer (~ 600 nm), Molybdenum tri-oxide (MoO_3) as a transparent efficient hole transport layer (HTL) (50 nm). A bi-layer of Phenyl-C61-butyric acid methyl ester (PCBM) and titanium di-oxide (TiO_2) is used as efficient electron transport layer (ETL) (~ 100 nm). Also, TiO_2 here acts as a buffer layer to reduce the strain and defects at the interface between the electron transport layer and photonic crystal. Fluorine doped tin oxide (FTO) is used as a transparent conductive oxide

(TCO) layer (~ 306 nm) serving three purposes at a time as a protective layer, anti-reflection coating (ARC) and transparent conductive electrode for charge collection [7]. Finally, a 1D photonic crystal is incorporated at the back of the solar cell which enables us to reflect back the incident solar spectrum (longer wavelengths) which was not absorbed by the absorber layer due to its higher band gap and thickness limitation. The total thickness of the device is only 1.7 microns.

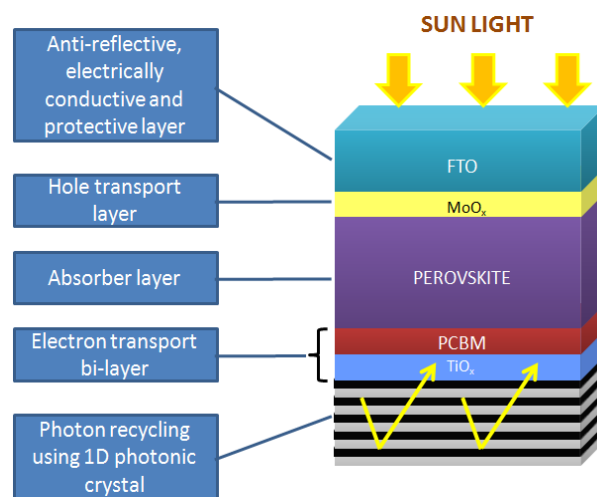


Fig. 2. Proposed thin film organometal trihalide perovskite solar cell structure.

MAPBI₃ as perovskite absorber material

Perovskite material has a general formula of ABX_3 (A and B are cations, X= carbon, oxygen or halide) where A is occupied in a cubo-octahedral while B in

Octahedral (Fig. 3). A and B are monovalent and divalent in case of perovskite with halides. C. Li *et al.* build a model for perovskite formability depending upon the Goldschmidt's tolerance factor and the octahedral factor [8, 9].

$$t = (r_A + r_X) / (2^{1/2}[r_B + r_X]), \quad (1)$$

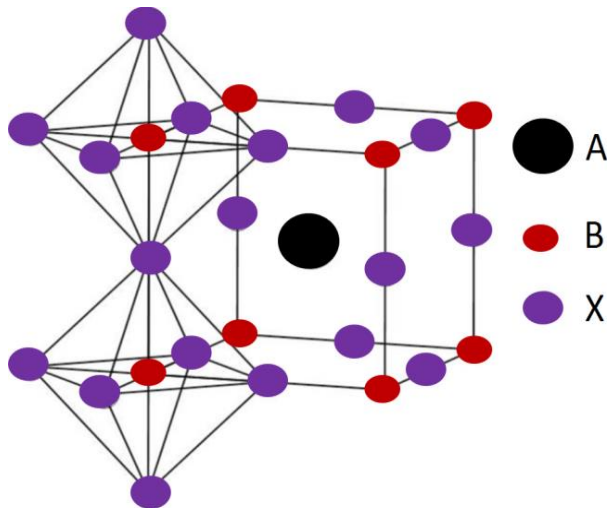


Fig. 3. Structure representing perovskite of form ABX_3 where A denotes the methylammonium, B is lead cation and X is halide anion.

where r_A , r_B and r_X are the effective radii for A, B and X ions, respectively. When $t=1$ the perovskite attains a perfect cubic structure which is the ideal case while octahedral distortion is expected when $t < 1$. For perfect stability the value of t should lie in between 0.75 – 1.0. The study of formability of perovskite material is important as it may affect the electronics properties. This criterion of Goldschmidt was widely accepted for the formation of perovskites [4, 9]. Another important parameter is the ratio of the ionic radius of the metal cation B and halide anion X called as octahedral factor ' μ '.

$$\mu = r_B / r_X \quad (2)$$

Goldschmidt realized that the tolerance factor is not enough to completely construct the structural map of the perovskite. Thus both the parameters are equally important to study the regularities of formability of cubic and general perovskite material [8-11].

Salient Features of MAPbI₃

One of the salient features of MAPbI₃ is its extended lifetime and diffusion lengths of generated excitons. Jacques E. Moser in 2017 experimentally demonstrated that the lifetime of the photo-generated carriers exceeds beyond the Langevin limit for direct recombination by several magnitude. This is attributed to the existence of additional dark states in MAPbI₃ and the fast radiative recombination is forbidden even in the presence of defects which makes it a defect tolerant material [12].

The case is much similar like indirect semiconductors [12, 13]. Thus the material exhibit slow recombination which is particularly important to avoid recombination of the generated charge carriers before they get collected at terminals in a solar cell device.

Second salient feature of MAPbI₃ is its high electronic dimensionality. A good electronic dimensionality is defined by the connectivity of the electronic orbitals of upper valence band and lower conduction band [14]. This explains a lot about the superior photovoltaic properties, bandgap, carrier mobilities and defects in a perovskite absorber. The higher the electronic dimensionality the higher the effective carrier transport [15-17]. Third feature is the efficient charge screening seen in perovskite material. Non-Langevin bimolecular recombination is practically seen in almost all the efficient solar cells technology. However, in MAPbI₃ the large dielectric constant shields holes and electrons such that they do not feel the presence of each other reducing the second order charge recombination [18-19]. Another feature is the low doping concentration in perovskites. Doping semiconductors are required in order to enhance the conductivity. However, in perovskites only a moderated doping is preferred to reduce the internal resistance. Also, external doping in perovskites are difficult to achieve due to its defect tolerant property. The low doping concentration in perovskites may contribute to large carrier diffusion lengths [20-22].

Interfacial carrier transport layers

TiO₂ and PCBM is employed as ETL or n-type collection layer in the proposed device. Selection of charge transport layer (CTL) is important in a perovskite solar cell as to extract the photo-generated electron-hole pairs effectively while keeping them away from recombination [23]. By carefully selecting the CTL material and sandwiching the perovskite layer between CTLs the photocurrent hysteresis can be eliminated and PCBM/TiO₂ here fulfils the role effectively. Although, the exact reason for the photocurrent hysteresis is not known till date. The most important characteristic of an ETL is the band alignment must be satisfied with perovskite absorber that is the highest occupied molecular orbital (HOMO) and the lowest unoccupied molecular orbital (LUMO) must be higher than the perovskite absorber layer. Recent researches show that diffusion lengths of the charge carriers in perovskite exceed even 1000 nm for MAPbI₃ [24, 26]. For PCBM the thickness must be carefully optimized as it has been observed that thickness below <25nm of PCBM the performance of the solar cell degrades and shows hysteresis behavior [23-26]. In our structure we have kept the thickness of the PCBM ETL to ~50nm. While TiO₂ here is acting both as ETL and buffer layer reducing trap assisted recombination and strain at PCBM and PhC interface. TiO₂ is most commonly used as ETL because of its hole blocking ability due to favourable valence band maximum for perovskite materials [27].

For the efficient hole extraction we have used crystalline molybdenum trioxide (MoO_3) as HTL as crystalline MoO_3 is more efficient in hole extraction than amorphous MoO_3 . It has also been demonstrated that MoO_3 is effective as an insertion layer [28]. However, shot circuit current may be improved more and leaves much room for further research due to low transmittance of MoO_3 [28-29].

Photon recycling using photonic crystal

Photonic crystals (PhCs) are being used since past decade in solar cells for efficient light trapping. PhCs were first implemented in solar cell in 2006 by L. Zeng *et al.* as a structure to control light inside the solar cell by reflecting back the desired incident photons back into the cell [30]. Since the TFSCs, suffers from low absorption of incident solar spectrum due to limited thickness which depends upon the absorption coefficient of the absorber material and also with the proposed device thickness absorption starts decreasing around 580 nm, structures like PhCs becomes important to facilitate excitons generation process. The photonic crystal implemented in our structure is a 1D photonic crystal consisting of alternate layers of silicon-di-oxide (SiO_2) and germanium (Ge). The combination of the two materials is selected keeping in mind the availability, fabrication, optical and electrical parameters. The dielectric contrast of the materials used results in index modulation and reflects back the desired wavelength. The modes of prohibition and propagation giving rise to photonic band gap for the PhC structure can be computed by solving the master equation derived from the Maxwell's equations as:

$$\Delta \times 1/\epsilon(\mathbf{r}) \nabla \times \mathbf{H}(\mathbf{r}) = (\omega/c)^2 \mathbf{H}(\mathbf{r}) \quad (3)$$

where, ' ϵ ' is the permittivity of the structure, ' ω ' is the frequency of operation, ' c ' is the velocity of light in free space and ' \mathbf{H} ' is the magnetic field in the region. Here, Plane Wave Expansion method (PWE) is used to solve the Eq. (3). In our structure we have optimized the PhC structure using RSOFT CAD tool to find the optimal thickness of the alternate layers, so as to reflect the desired range of wavelengths (**Fig. 4**). The center wavelength is kept at 900nm. Thus, this reflects back the desired portion of the incident solar spectrum back into the solar cell (increasing the optical path length of the photons) to enhance absorption. This process is known as photon recycling [31]. Conventionally, a metallic layer of Aluminium (Al) or Silver (Ag) is used as back reflector having reflectance more than 90%. However, the semiconductor-metal interface introduces unwanted losses due to surface plasmon resonance. Metallic surfaces also have limited diffraction capabilities and high degradation rate as they are corrosive in nature [32, 33].

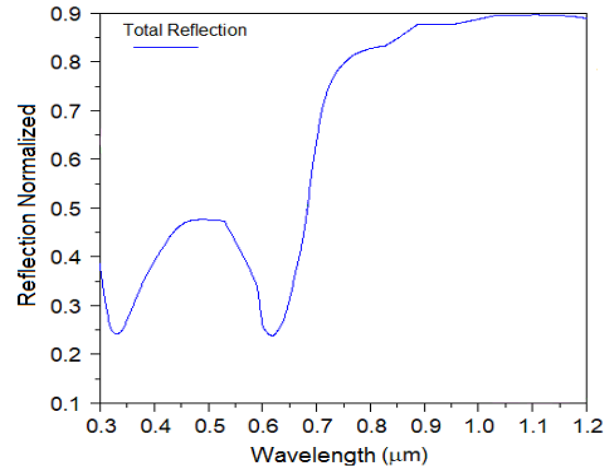


Fig. 4. Reflection from the PhC after optimizing the thickness of the layers.

Computing power conversion efficiency

The total number of incoming photons at incident solar spectrum $S(\lambda)$ is given as:

$$n_s(\lambda) = \frac{S(\lambda)}{E(\lambda)} = \frac{\lambda}{hc} S(\lambda) \quad (4)$$

where, $E(\lambda)$ is the energy of the incident photon. The sum of absorption spectra of each layer thus gives the total absorption spectrum of the device as:

$$A(\lambda) = \sum A_i(\lambda) \quad (5)$$

The absorption spectrum is thus computed by RSoft's CAD tool. The photons absorbed by each layer can be calculated as:

$$n_i(\lambda) = \frac{S(\lambda) A_i(\lambda)}{E(\lambda)} = \frac{\lambda}{hc} S(\lambda) A_i(\lambda) \quad (6)$$

Finally, the PCE of the solar cell can be computed by the expression as:

$$\eta(\%) = \frac{J_{sc} \times V_{oc} \times FF}{P_{(in)}} \times 100 \quad (7)$$

The structure is then simulated using "RSOFT's DiffractMOD and Solar Cell Utility" based on RCWA algorithm to analyze the behavior of the solar cell and to compute the PCE. RCWA algorithm provides fast and efficient technique to solve Maxwell's equations. The method is a semi-analytical technique, meaning it picks one direction (longitudinal) to solve analytically and the other two (transverse) to be solved numerically and efficiently determines the transmission and reflection from periodic structures. It uses a discrete Fourier transform to discretize the fields in the transverse direction. This means it represents the fields and materials as a set of plane waves (or planar gratings), so RCWA is extremely fast and efficient for devices with low to moderate index contrast where the waves are more like pure plane waves. The longitudinal direction is solved analytically so the layers can be of any thickness, without affecting the computational burden [34-36].

In this computation the number of spatial field harmonic plays an important role to obtain accurate results. In Fourier space, the expansion of the refractive index and field is determined by the number of harmonics. More the number of harmonics more accurate will be the simulated results. However, this consumes large memory and simulation time. By performing convergence study and running several simulations to see how the results are varying with increasing number of harmonics it is found that the results become almost stable at harmonics equal to 5 or more. Thus, we have chosen number of harmonics equal to 5 for our simulation [37]. The designed device is a multilayered structure in vertical direction (z direction). The algorithm considers lateral Bloch periodic boundary condition and decomposes the solar cell structure into thin layers or slices for which transmission and reflection is calculated. This approach not only provides a physical insight into the problem but also the computational advantages [38].

Results and discussion

The structure is simulated at 100% collection efficiency, considering only optical losses. The resulted PCE is ' η ' = 25.2% with ' J_{sc} ' = 32.1 mA/cm², ' V_{oc} ' = 0.81 V and a fill factor of 'FF' = 86.3 % (Fig. 5).

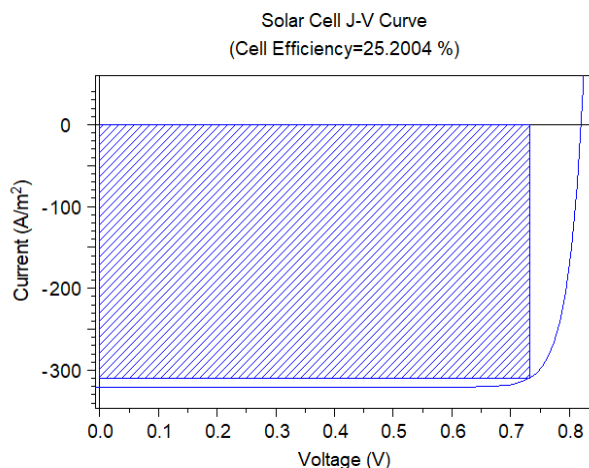


Fig. 5. J-V curve of the proposed thin film perovskite solar cell with PCE = 25.2 %, J_{sc} = 32.1 mA/sq-cm and a fill factor of 86.3%.

It has been observed in the final results that there is a huge enhancement in the absorption of the incident solar spectrum when PhC is incorporated as a back reflector in the design. The overall cell absorption, with and without using PhC as back reflector shows that the cell with PhC as back reflector is now efficiently absorbing photons of the incident spectrum ranging from ~580 nm to ~1200 nm wavelength. We compared the conversion efficiency and short circuit current of the device with and without PhC and it is found that the respective parameters have nearly doubled when using PhC as a selective wavelength reflector (Table. 1 and Fig. 6). Advantage of using dielectric photonic crystal

structure over metal back reflector is that it produces the lossless diffraction of photons unlike surface plasmon resonance losses at metal semiconductor interface [39, 40].

Table I. Comparison of the power conversion efficiency and short circuit current of proposed thin film perovskite solar cell with and without PhC as a back reflector.

Device Structure	PCE (%)	J_{sc} (mA/cm ²)
Thin film PSC without PhC	16.8	21.3
Thin film PSC with PhC	25.2	32.1

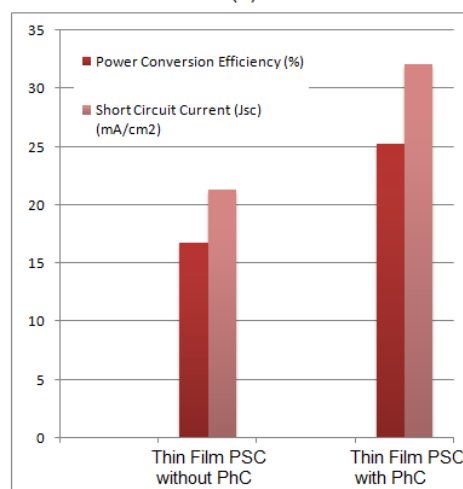
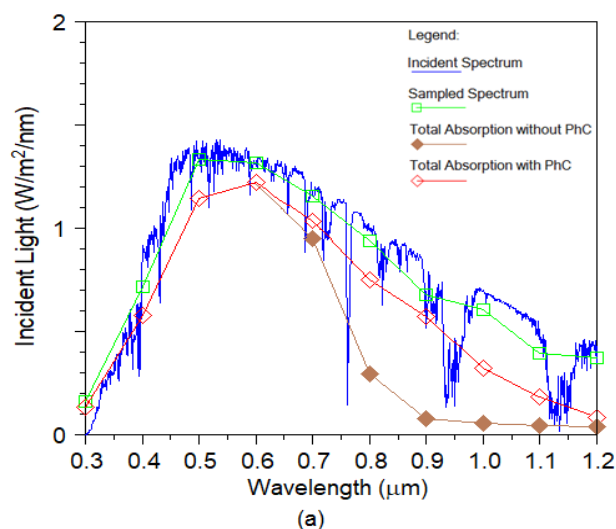


Fig. 6. (a) Graph displaying increased absorption of incident solar spectrum as a result of photon recycling on incorporating PhC as light trapping structure, (b) Comparison of the enhancement in final PCE and short circuit current with and without PhC.

Conclusion

Thus, from the results we see that using PhCs in solar cells as light trapping structures can greatly enhance the PCE of solar cells in addition to that perovskites solar cells already have high potential in competing the existing silicon market making it possible to fabricate cheaper and efficient solar cells. These organic-

inorganic perovskites can be easily prepared by HX salts of lead halides and organic amines. MAPbI₃ has lower defect state related to dangling bonds due to its high ionic nature and high diffusion length to absorption length ratio responsible for generating large photo-current. Using tuned 1D PhC's which is nothing but a distributed bragg reflector enables us to propose simple thin film solar cell design viable for large scale industrial fabrication. The large J_{sc} here clearly indicates a huge increase in the photo-generated charge carriers because of photon recycling process. However, the above results are obtained by conducting numerical simulation study. In practical scenario there may be contact shading and shunting losses depending upon the fabrication process and environmental conditions which has to be considered when fabricating such device. Since the results are promising the future work will be to fabricate the structure and analyze the output in the real world conditions.

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Author's contributions

Conceived the plan, demonstrated efficiency enhancement through simulations.

Supporting information

Supporting informations are available from VBRI Press.

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