Materials Letters 309 (2022) 131353

Contents lists available at ScienceDirect

Materials Letters

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

2D-MXene as an additive to improve the power conversion efficiency of monolithic perovskite solar cells

Satish Bykkam [a], Arti Mishra [a], D.N. Prasad [a], Muni Raj Maurya [a,b], John-John Cabibihan [b], Zubair Ahmad [c], Kishor Kumar Sadasivuni [a,*]

[a] Center for Advanced Materials, Qatar University, P.O. Box. 2713, Qatar
[b] Department of Mechanical and Industrial Engineering, College of Engineering, Qatar University, P.O. Box. 2713, Qatar
[c] Qatar University Young Scientists Centre, P.O. Box. 2713, Doha, Qatar

ARTICLE INFO

Keywords:
Monolithic perovskite solar cells
2D-MXene
Additive
Power conversion efficiency

ABSTRACT

In this study, two-dimensional (2D) material-MXene has been utilized as an additive in perovskite ($MAPbI_3$) from 0 to 20 vol.% with an increment of 5 vol.%. The effect of different vol.% of 2D-MXene on monolithic perovskite solar cells (mPSCs) has been investigated in detail through various characterization techniques such as X-ray diffraction (XRD), Raman Spectroscopy, and Field emission scanning electron microscopy (FE-SEM). The best device, perovskite with 5 vol.% 2D-MXene has shown a 13.62% power conversion efficiency (PCE) compared to the perovskite without 2D-MXene PCE of 11.35%.

1. Introduction

Perovskite solar cells (PSCs) have received a lot of attention due to their high-power conversion efficiency (PCE) and low fabrication cost. Different methods like composition engineering [1,2], charge carriers transport layers modification [3] and heterostructure [4] have been adopted for enhancing the PCE and stability of PSCs. Recently, carbon-based monolithic perovskite solar cells (mPSCs) with novel architecture have emerged as one of the most promising designs for commercializing large-area perovskite solar cells (PSCs) at reasonable costs. Furthermore, the carbon-based design eliminates the need for hole-transporting materials (HTMs) like Spiro-OMeTAD. Since manufacturing costs are also less, low-cost photovoltaic systems can be developed. To further increase the performance, the additive engineering method is adapted. mPSCs are made up of four successive layers, as shown in Fig.S1 (Supporting Information), which includes glass/FTO/compact-TiO_2/mesoporous-TiO_2/mesoporous-ZrO_2/carbon. These mPSCs are filled with perovskite, thereby acting as the light-absorbing layer respectively. In this design, perovskite behaves as a hole transport layer (HTL) as well as an absorber layer at the same time [5]. To enhance the performance of mPSCs, different techniques have been explored, including antisolvent optimization [6], post-treatment [7], and additive engineering [8]. From the methods mentioned above, additive engineering is extremely promising and simple to use, and it has played a key role in numerous mPSCs for creating breakthroughs. In addition, various 2D materials like WS_2, MoS_2, and black phosphorous have been used as additives to improve the performance of PSCs [9]. It has been observed that the addition of 2D materials with perovskite blocks the unrequired ion movements, which has been the major bottleneck towards the PSCs stability [5]. MXene (Ti_3C_2) is a new 2D material with the general formula $M_{n+1}X_nT_x$ (n = 1,2,3), where M represents an early transition metal, X represents carbon or nitrogen, and T represents surface functionalization groups ($^{-}F, ^{-}O, ^{-}OH$). High electrical conductivity, mobility, charge carrier density, and hydrophilicity are all the unique characteristics of a typical MXene, which allows it to be processed from aqueous solutions. MXene is also utilized as an additive in the layers of PSCs [10]. However, in this work, we used 2D-MXene material as an additive with varying vol.% in the range of 0 to 20 vol%, into a 3D $MAPbI_3$ perovskite for infiltration. The impact of varying vol.% on the structural, morphological, and electrical characteristics of the mPSCs has been studied.

2. Experimental

Solaronix SA (Aubonee, Switzerland) produced printed electrodes (glass/FTO/compact-TiO_2/mesoporous-TiO_2/mesoporous-ZrO_2/monolithic carbon) were annealed at 400°C for 30 min and cooled down. 0.1 M of 2D-MXene (vol.% from 0 to 20) was dispersed in 500 μL DMF (N, N-

* Corresponding author.
E-mail address: kishorkumars@qu.edu.qa (K.K. Sadasivuni).

https://doi.org/10.1016/j.matlet.2021.131353
Received 25 August 2021; Received in revised form 1 November 2021; Accepted 19 November 2021
Available online 25 November 2021

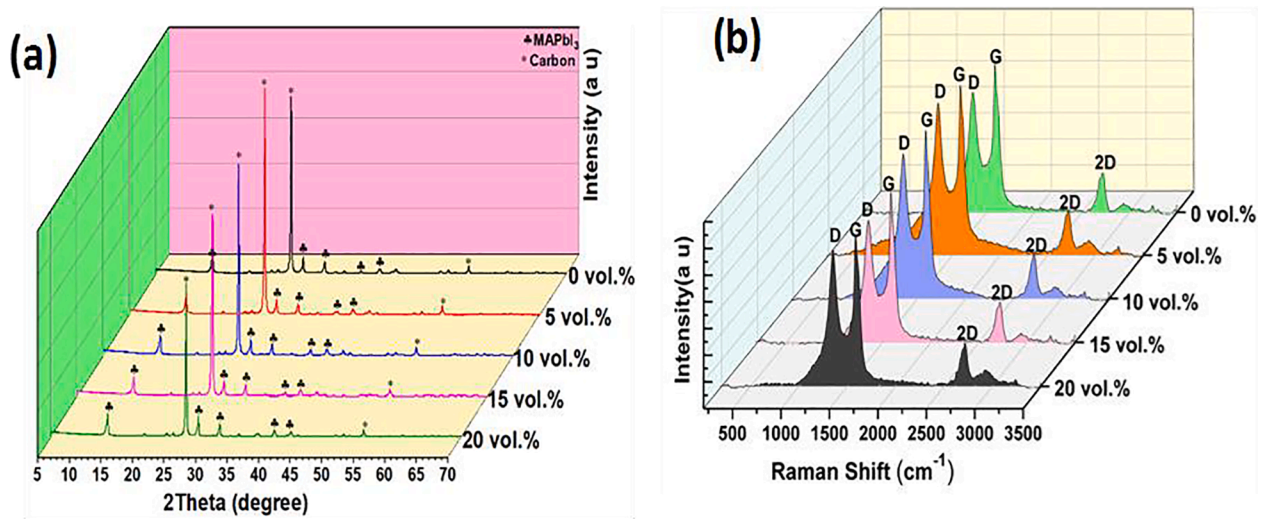

Fig. 1. (a and b) XRD and Raman Spectra of perovskite films containing various vol.% of 2D-MXene additive.

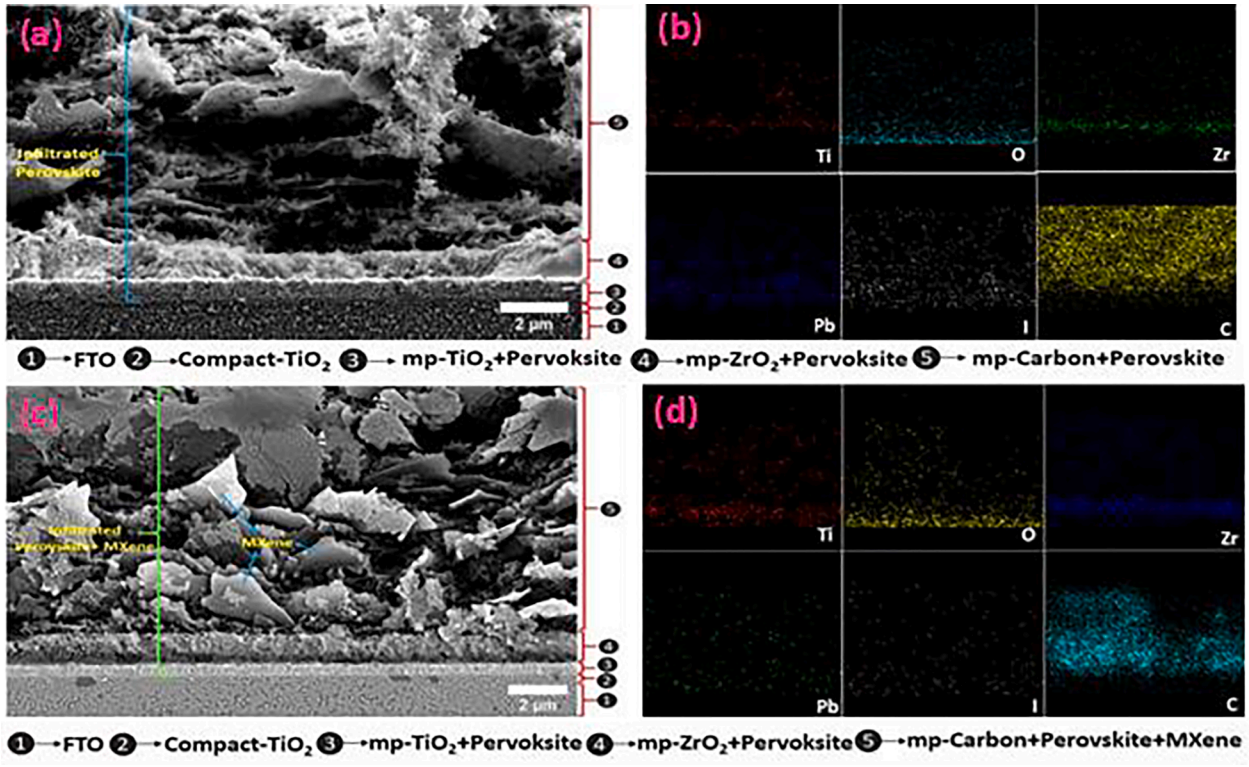

Fig. 2. Cross-sectional FE-SEM images and EDX elemental mapping of perovskites with (a and b) 0 vol.% of 2D-MXene additive, (c and d) 5 vol.% of 2D-MXene additive.

Dimethylformamide) solvent and was added to the perovskite solution; $MAPbI_3$ (mixture of lead iodide, methylammonium iodide and 5-aminovaleric acid hydroiodide in Ɣ-butyrolactone). 0.5 μL of the prepared precursor solution was infiltrated into the mesoporous structure. The prepared films were annealed at 100°C for 30 min in ambient conditions.

3. Results and discussion

To evaluate the crystallinity of samples, XRD (Model No; EMPYREAN) plots were obtained for pure perovskite and different vol.% of 2D-MXene added perovskite films. Fig. 1(a) depicts the XRD patterns of the perovskite and 2D-MXene perovskite films with various vol.% of 0, 5, 10, 15, and 20. Irrespective of the additive vol.%, all the samples apprehend a sharp peak at an angle of 14.2°, representing (1 1 0) crystal plane of $MAPbI_3$ phase, indicating the cubic phase of the perovskite. A sharp diffraction peak detected in all samples at $2\theta = 26.2°$ corresponds to (0 0 2) plane of carbon. The characteristic diffraction peaks observed at an angle (2θ) of 28.3°, 31.6°, 40.5°, and 43.1°, which are indexed to the (0 0 4), (2 2 0), (3 1 0), (2 2 4), and (3 1 4) reflections, of $MAPbI_3$, respectively. The effect of adding different vol.% of the 2D-MXene as an additive can be observed by analyzing the main perovskite peak at $2\theta = 14.2°$. Fig. S2 shows the magnified view of the peak centered at $2\theta = 14.2°$. From Fig. S2 it can be observed that with an increase in the vol.%

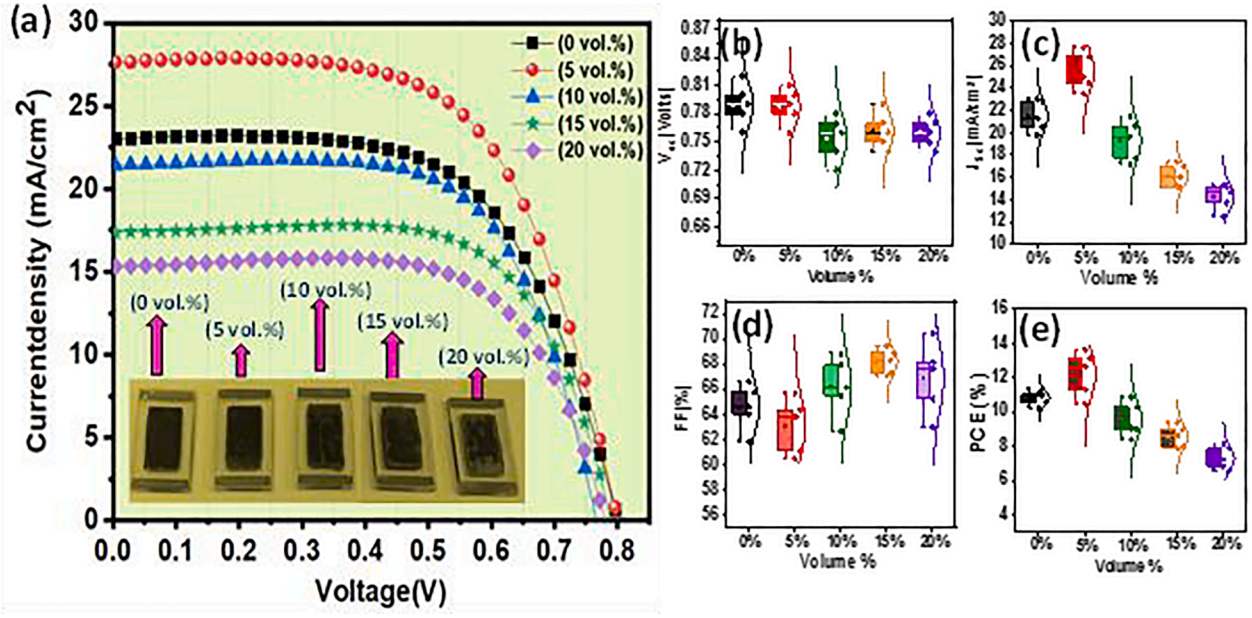

Fig. 3. (a) J-V characteristics of the fabricated mPSCs devices. (b-e) The statistical distribution of V_{oc}, J_{sc}, FF, and PCE of mPSCs with various vol.% of 2D-MXene additive.

of 2D-MXene as an additive in perovskite, the peak at $2\theta = 14.2^{\circ}$ exhibits a slight shift towards the lower angle. The consistent shift in the peak position indicates the stress induced by the 2D-MXene additive and attributes to the homogenous distribution of the strain during the growth of perovskite crystals. Further characterization was done by Raman spectra (Model No; Jobin-Yvon HR800) as shown in Fig. 1(b). Table S1 summarizes the estimated I_D/I_G ratio of the perovskite film with different vol.% 2D-MXene. And it is observed that perovskite film with 2D-MXene additive of 5 vol.% apprehends the highest I_D/I_G (~0.91), which suggests a significantly lower degree of disorder in the film than other samples. Moreover, when 2D-MXene in perovskite is above 5 vol. %, a decrease in the I_D/I_G value is observed, indicating increasing disorders during the film growth process. From the results, it can be inferred that the 5 vol.% 2D-MXene additive perovskite film offers fewer defects and helps in improving the photoresponse of the solar cell.

To elucidate the perovskite infiltration and investigate the change in morphology after adding 2D-MXene, FE-SEM (Model No; JEOL 7600) analysis was carried out. Fig. 2(a) represents cross-sectional FE-SEM images of mPSCs fabricated, and it is evident that the perovskite is successfully infiltrated across multilayers. The corresponding energy dispersive X-ray (EDX) elemental mapping (Fig. 2(b)) confirms the presence of elements such as Ti, O, Zr, Pb, I, and C. Fig. 2(c), represents the cross-sectional FE-SEM image of the mPSCs fabricated by infiltrating perovskite with 5 vol.% of 2D-MXene additive. The bright flake-like structure represents 2D-MXene. The corresponding EDX elemental mapping is shown in Fig. 2(d) and confirms the presence of Ti, O, Zr, Pb, I, and C elements. In comparison to Fig. 2(b), a significant increase in the count of the Ti element across the length of the mPSCs can be observed in Fig. 2(d). These additional Ti elements correspond to the Ti present in the 2D-MXene. Further increase in the vol.% of 2D-MXene results in stacking of the 2D-MXene flakes in the perovskite photo-absorbing layer. The related cross-sectional FE-SEM images and the corresponding EDX results were shown in Fig. S3 (f, h and i) and Fig. S3 (g, i and k), respectively. The above investigation suggests that adding 2D-MXene above 5 vol.% leads to stacking of the 2D-MXene flakes. The origin of stacking morphology may hinder the light propagation across the complete length of the perovskite layer and decrease the photogeneration of charge carriers.

The photocurrent density–voltage (J-V) features were measured by Abet sunlight solar simulator (Oriel 3A) using Air Mass (AM) 1.5G filter at an illumination intensity of 100 mW/cm^2. The active area of the cell was accurately regulated by using a 0.16 cm^2 black metal mask. Fig. 3(a) displays the current density–voltage (J-V) characteristic of the devices fabricated with pure perovskite and perovskite with various vol.% of 2D-Mxene additive. The highest PCE of 13.62% for the 5 vol.% 2D-MXene devices was achieved with open-circuit voltage (V_{oc}) ~ 0.81 V, current–density (J_{sc}) ~ 27.64 mA/cm^2, and fill factor (FF) ~ 61.12. Adding 2D-MXene beyond 5 vol.%, the J_{sc} of the devices were below the pristine perovskite device and continuously kept on decreasing. The devices with 2D-MXene additive with 10, 15, and 20 vol.% displayed a limited J_{sc} of 21.04, 17.36, and 15.27 mA/cm^2, resulting in PCE of 10.85, 9.41, and 8.11%, respectively. The remarkable improvement of PCE by the addition of 2D-MXene is attributed to the relevant improvement in the device J_{sc}. Notably, 2D-MXene additive flakes serve as a conductive link at the perovskite/electron transport layer (ETL) interface and offer an improved electron transport path, resulting in efficient electron collection. The results clearly show that when the appropriate amount of additive was added to the perovskite film, the PCE of the device was improved. The detailed statics of device parameters are presented in Table 1. The statistical distribution (V_{oc}, J_{sc}, FF and PCE) of devices with various vol.% of 2D-MXene additive is reported in Fig. 3(b, c, d and e). The devices with 5 vol.% of 2D-MXene additive displayed good repeatability. The high electrical conductivity of the 2D-MXene additive significantly improves the charge transfer and reduces the charge

Table 1
Photovoltaic parameter champion device fabricated from mPSCs with perovskite precursor solution with various vol.% of 2D-MXene additive.

Device		V_{oc} (V)	J_{sc} (mA/cm^2)	FF (%)	PCE (%)
0 vol%-2D-MXene	Average	0.79	21.45	64.54	10.82
	Best	0.80	23.03	61.81	11.35
5 vol%-2D-MXene	Average	0.78	25.48	63.60	12.24
	Best	0.81	27.64	61.12	13.62
10 vol%-2D-MXene	Average	0.75	19.31	66.18	9.49
	Best	0.78	21.04	65.49	10.85
15 vol%-2D-MXene	Average	0.76	16.11	68.17	8.55
	Best	0.79	17.36	69.43	9.41
20 vol%-2D-MXene	Average	0.76	14.26	66.87	7.32
	Best	0.78	15.27	68.17	8.11

transfer resistance. Therefore, 5 vol.% 2D-MXene added in the perovskite effectively improved the power conversion efficiency in mPSCs.

4. Conclusion

In summary, we elucidated the promising role of 2D-MXene as an additive for the $MAPbI_3$ perovskite structure to improve the performance of mPSCs. The addition of 5 vol.% 2D-MXene in perovskite improved the crystallinity of the fabricated perovskite layer. The facile strategy resulted in obtaining high-quality photoactive films with fewer defect states. Such a high-quality film formation is a missing achievement in most research based on mPSCs. Through additive engineering, a champion efficiency of 13.62% was obtained for the 5 vol.% 2D-MXene loaded perovskite in mPSCs device structure, higher than the pure perovskite mPSCs with an efficiency of 11.35%. This work introduces an effective method for improving the charge transport by incorporating 2D-MXene into perovskite and thus a simple modification technique to fabricate high-performance mPSCs.

CRediT authorship contribution statement

Satish Bykkam: Writing – original draft, Conceptualization, Methodology. **Arti Mishra:** Methodology. **D.N. Prasad:** Formal analysis. **Muni Raj Maurya:** Investigation, Writing – review & editing. **John-John Cabibihan:** Investigation, Writing – review & editing. **Zubair Ahmad:** Validation, Visualization. **Kishor Kumar Sadasivuni:** Investigation, Writing – review & editing, Supervision.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

This work was supported by Qatar National Research Fund under the grant no. NPRP12S-0131–190030. The statements made herein are solely the responsibility of the authors. Open Access funding provided by the Qatar National Library

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.matlet.2021.131353.

References

[1] T. Ozturk, E. Akman, A.E. Shalan, S. Akin, Composition engineering of operationally stable $CsPbI_2Br$ perovskite solar cells with a record efficiency over 17%, Nano Energy 87 (2021), 106157.
[2] E. Akman, A.E. Shalan, F. Sadegh, S. Akin, Moisture-resistant $FAPbI_3$ perovskite solar cell with 22.25 % power conversion efficiency through pentafluorobenzyl phosphonic acid passivation, ChemSusChem 14 (2021) 1176–1183.
[3] G. Nagaraj, M.K.A. Mohammed, M. Shekargoftar, P. Sasikumar, P. Sakthivel, G. Ravi, M. Dehghanipour, S. Akin, A.E. Shalan, High-performance perovskite solar cells using the graphene quantum dot–modified SnO_2/ZnO photoelectrode, Materials Today Energy 22 (2021), 100853.
[4] M.K.A. Mohammed, A. Esmail Shalan, M. Dehghanipour, H.R. Mohseni, Improved mixed-dimensional 3D/2D perovskite layer with formamidinium bromide salt for highly efficient and stable perovskite solar cells, Chem. Eng. J. 428 (2022), 131185.
[5] L. Lin, L. Jiang, Y.u. Qiu, Y. Yu, Modeling and analysis of HTM-free perovskite solar cells based on ZnO electron transport layer, Superlattices Microstruct. 104 (2017) 167–177.
[6] M. Xiao, F. Huang, W. Huang, Y. Dkhissi, Y.e. Zhu, J. Etheridge, A. Gray-Weale, U. Bach, Y.-B. Cheng, L. Spiccia, A fast deposition-crystallization procedure for highly efficient lead iodide perovskite thin-film solar cells, Angewandte Chemie Int. Ed. 53 (37) (2014) 9898–9903.
[7] L.i. Hong, Y. Hu, A. Mei, Y. Sheng, P. Jiang, C. Tian, Y. Rong, H. Han, Improvement and regeneration of perovskite solar cells via methylamine gas post-treatment, Adv. Funct. Mater. 27 (43) (2017) 1703060, https://doi.org/10.1002/adfm.v27.4310.1002/adfm.201703060.
[8] Y. Wu, F. Xie, H. Chen, X. Yang, H. Su, M. Cai, Z. Zhou, T. Noda, L. Han, Han, thermally stable MAPbI3 perovskite solar cells with efficiency of 19.19% and area over 1 cm^2 achieved by additive engineering, Adv. Mater. 29 (28) (2017) 1701073, https://doi.org/10.1002/adma.v29.2810.1002/adma.201701073.
[9] A.S.R. Bati, M. Batmunkh, J.G. Shapter, Emerging 2D Layered Materials for Perovskite Solar Cells, Advanced Energy Materials 10(13) (2020) 1,902,253.
[10] Z. Guo, L. Gao, Z. Xu, S. Teo, C. Zhang, Y. Kamata, S. Hayase, T. Ma, High electrical conductivity 2D MXene serves as additive of perovskite for efficient solar cells, Small 14 (47) (2018) 1802738, https://doi.org/10.1002/smll.v14.4710.1002/smll.201802738.