Achieving efficiency above 30% with new inorganic cubic perovskites X_2SnBr_6 (X = Cs, Rb, K, Na) via DFT and SCAPS-1D

Md. Ferdous Rahman[1], Tanvir Al Galib[1], Md. Azizur Rahman[1], Md. Hafizur Rahman[1], Md. Harun-Or-Rashid[1], Md Al Ijajul Islam[1], Md. Monirul Islam[1], N. Dhahri[2], Ahmad Irfan[3]

[1]Advanced Energy Materials and Solar Cell Research Laboratory, Department of Electrical and Electronic Engineering, Begum Rokeya University, Rangpur 5400, Bangladesh

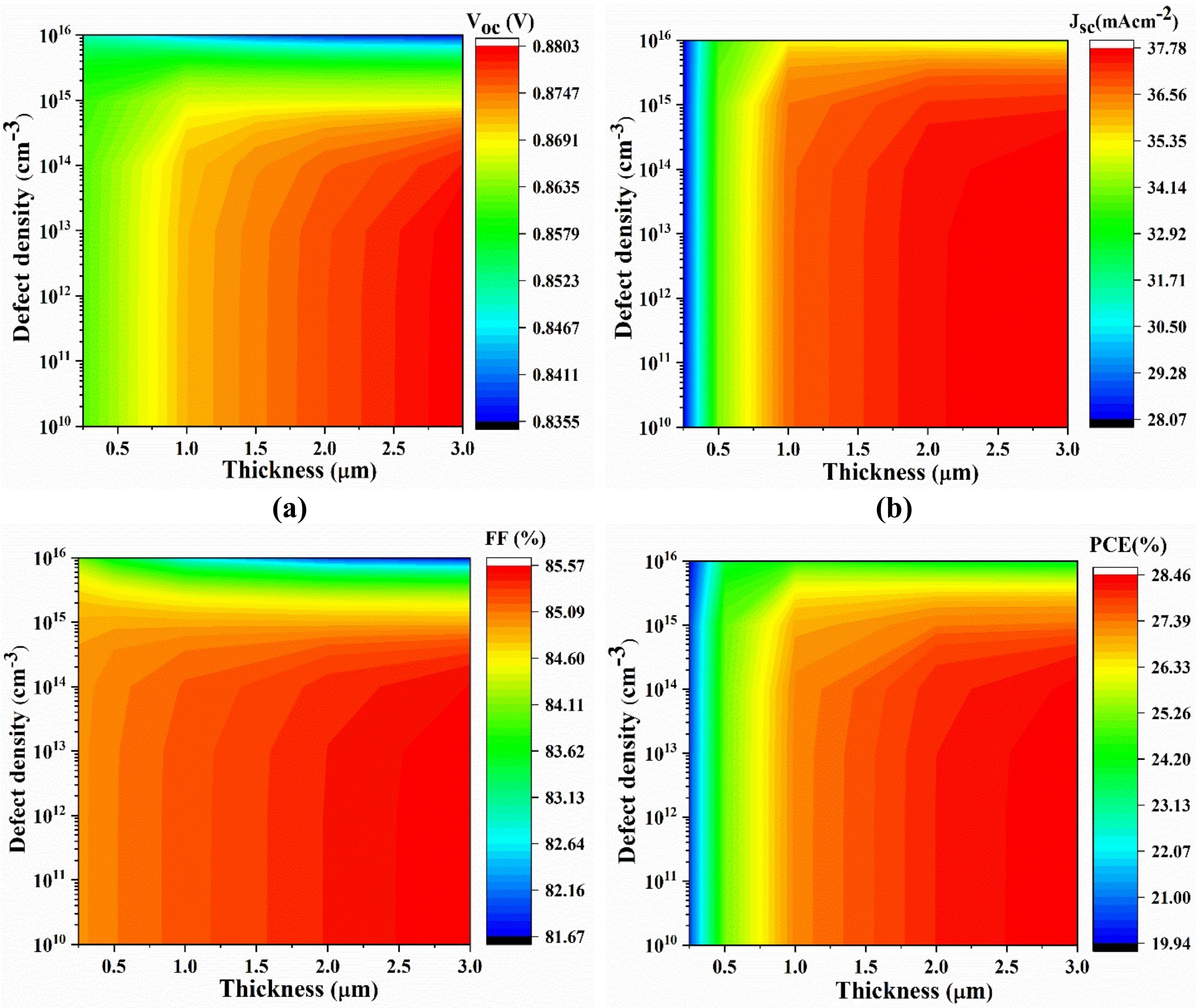
[2]Department of Physics, College of Science, Northern Border University, Arar, Saudi Arabia

[3]Department of Chemistry, College of Science, King Khalid University, Abha 61413, P.O. Box 9004, Saudi Arabia

Correspondence: *ferdousapee@gmail.com (Md. Ferdous Rahman); ORCID: 0000-0002- 0090-2384

1. **Impact of Performance due to Variation of Absorber Thickness and Absorber Layer Defect Density.**

Within the Al/FTO/TiO_2/Rb_2SnBr_6/Au structure, the PCE, FF, J_{SC}, and V_{OC} values exhibit a tendency to decrease with an increase in defect density and rise with the thickness of the absorber layer that's shown in Fig S1(a) to Fig S1 (d). For absorber layer thicknesses spanning from 250 to 3000 nm and defect densities ranging from 10^{10} to 10^{16} cm^{-3}, the alterations noted in PCE, FF, J_{SC}, and V_{OC} are as follows: 28.46% to 19.94%, 85.57% to 81.67%, 37.78 mA/cm^2 to 28.07 mA/cm^2, and 0.88V to 0.84V, respectively. Up to a thickness of 1000 nm, there is a noticeable upswing in these metrics; nevertheless, once the thickness exceeds 1000 nm, the increments become marginal. Similarly, these metrics experience a significant decline when the defect density is enhanced up to 10^{12} cm^{-3} [1].Yet, surpassing this limit, there is a minor downturn in the values. Thus, the most advantageous parameters are a thickness of 1000 nm and a defect density of 10^{12} cm^{-3}. Under these optimum conditions, the PCE, FF, J_{SC}, and V_{OC} stand at 27.29%, 85.21%, 36.73 mA/cm^2, and 0.87V, respectively.



(a) (b) (c) (d)

Fig. S1 Influence of variations in absorber thickness and defect density of Rb_2SnBr_6 on key PV performance parameters: (a) V_{OC}, (b) J_{SC}, (c) FF and (d) PCE.

Within the Al/FTO/TiO_2/K_2SnBr_6/Au structures, as the thickness of absorber layer varies from 250 to 3000 nm and the defect density fluctuates between 10^{10} and 10^{16} cm^{-3}, PCE, FF,J_{SC}, and V_{OC} experience a decline from 32.05% to 20.35%, 86.30% to 74.40%, 43.82 mA/cm^2 to 32.15 mA/cm^2, and 0.87 V to 0.73V, respectively. As depicted in the results shown in Fig. S2(a), the highest V_{OC} of 0.87V is achieved when the thickness is maintained below 1000 nm, and the defect density value of fewer than 10^{12} cm^{-3}. Nevertheless, surpassing the 10^{12} cm^{-3} threshold in defect density results in a substantial decrease in V_{OC} to 0.73V.

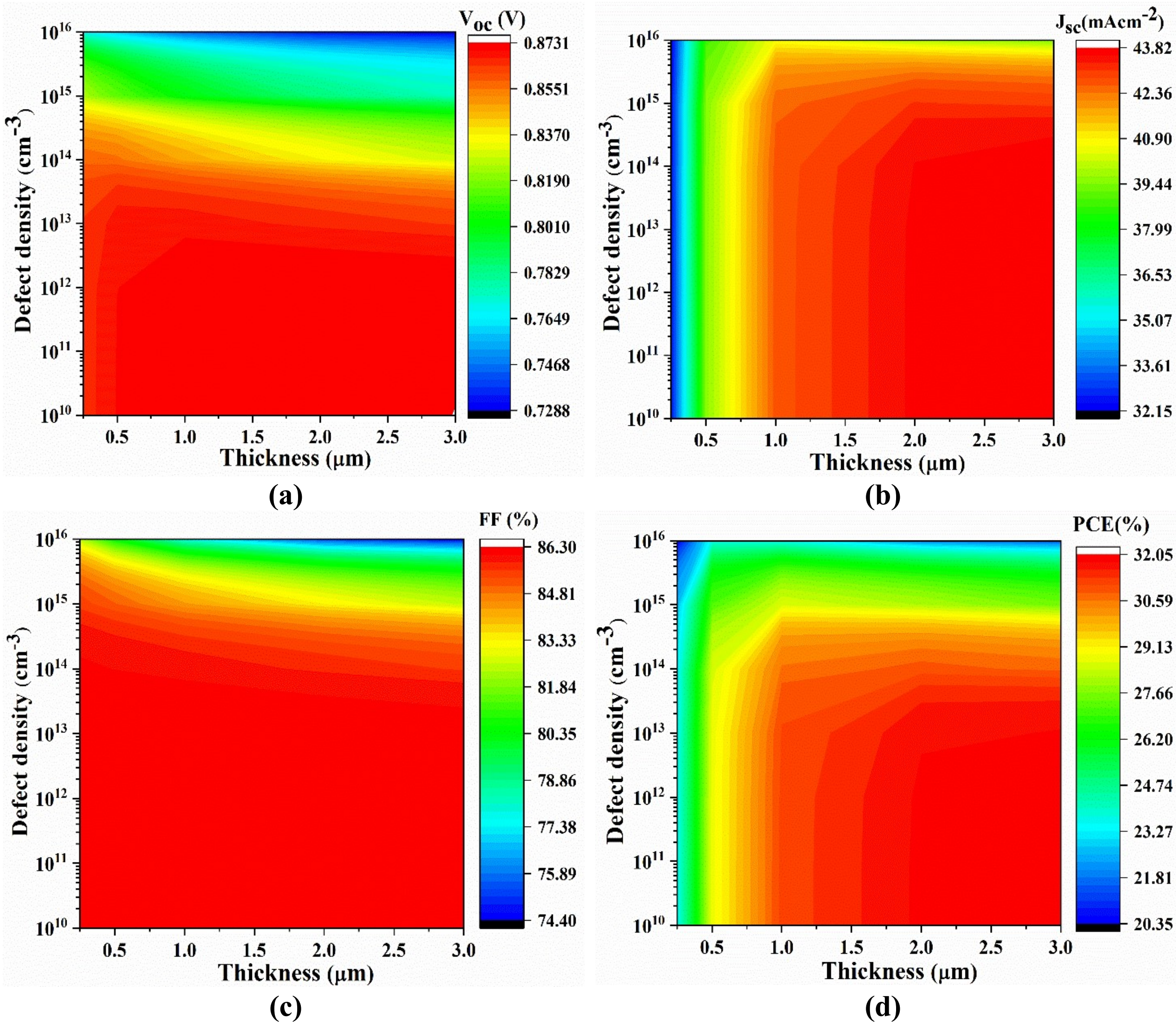


Fig. S2 Influence of variations in absorber thickness and defect density of K_2SnBr_6 on key PV performance parameters: (a) V_{OC}, (b) J_{SC}, (c) FF and (d) PCE.

According to the data in Fig. S2(b), reaching the maximum J_{SC} value of 43.82 mA/cm^2 necessitates a thickness exceeding 1000 nm, while keeping the defect density below 10^{12} cm^{-3}. Moreover, as depicted in Fig. S2(c), FF attains its highest value of 86.30% when the thickness is less than or equal to 1000 nm, and the defect density covers the entire range. Finally, as shown in Fig. S2(d), the highest conversion efficiency exceeding 32.05% is attained within the thickness range of 250 to 3000 nm and a defect density of up to 10^{12} cm^{-3}. The optimal parameters yielded a PCE of 31.75%, accompanied by a FF of 86.88%, a J_{SC} of 42.62 mA/cm^2, and a V_{OC} of 0.87V. In the Al/FTO/TiO_2/Na_2SnBr_6/Au structures, as the absorber layer thickness varies from 250 to 3000 nm and the defect density fluctuates between 10^{10} and 10^{16} cm^{-3}, PCE, FF, J_{SC}, and V_{OC} experience a decline from 29.90% to 11.40%, 82.60% to 43.11%, 46.60 mA/cm^2 to 33.65 mA/cm^2, and 0.78V to 0.66V, respectively.

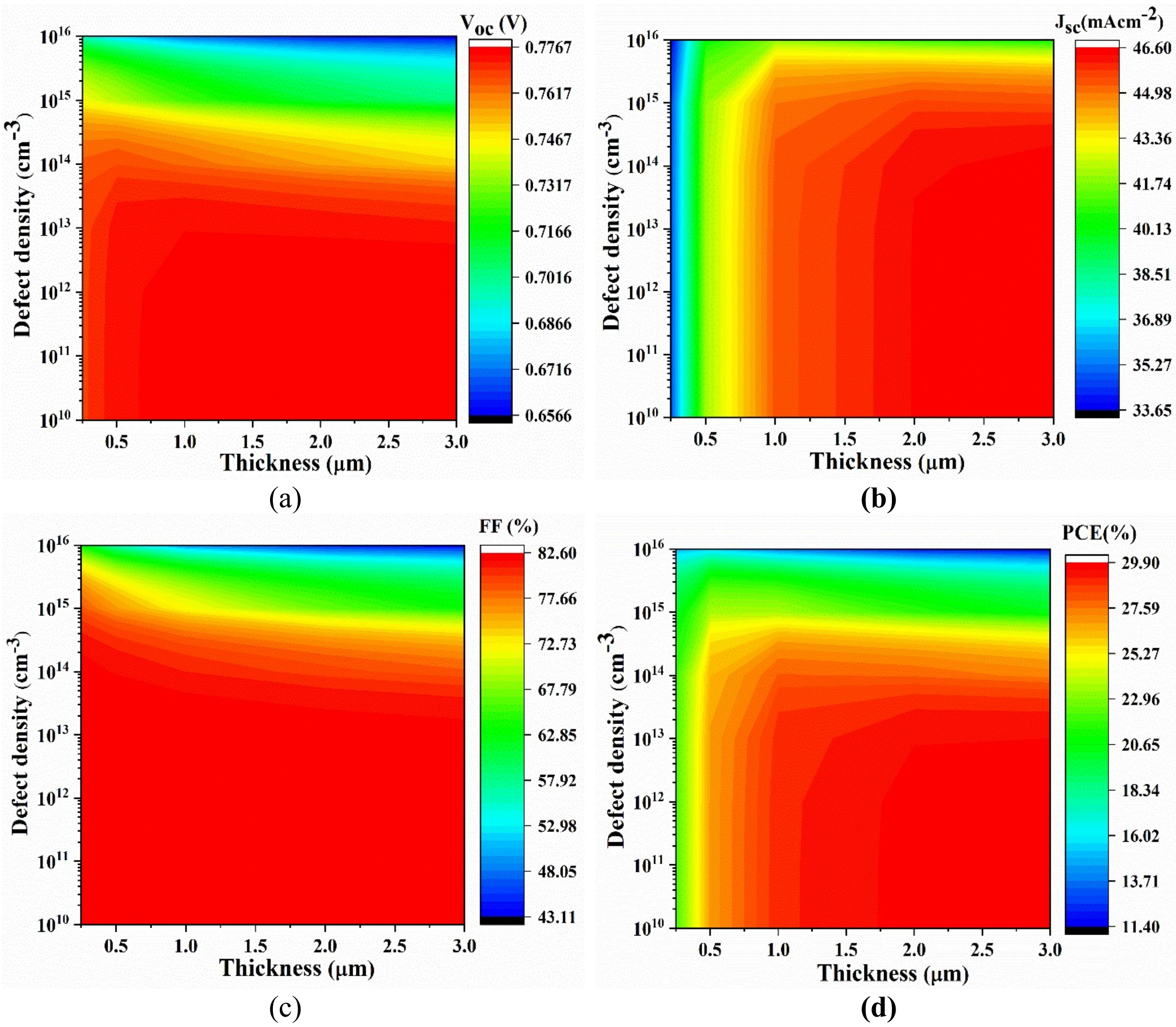


Fig. S3 Influence of variations in absorber thickness and defect density of Na_2SnBr_6 on key PV performance parameters: (a) V_{OC}, (b) J_{SC}, (c) FF and (d) PCE.

As per the results presented in Fig. S3(a), the highest V_{OC} of 0.78V is achieved when the thickness is maintained below 1000 nm, and the defect density remains under 10^{12} cm^{-3}. However, exceeding the defect density threshold of 10^{12} cm^{-3} causes a considerable fall in V_{OC} (0.66V). As demonstrated in Fig. S3(b), achieving the highest J_{SC} value of 46.60 mA/cm^2 requires a thickness surpassing 1000 nm while maintaining the defect density below 10^{12} cm^{-3}. Moreover, as depicted in Fig. S3(c), the

FF attains its highest value of 82.60% when the thickness is less than or equal to 1000 nm, and the defect density covers the entire range. Finally, as shown in Fig. S3(d), the most substantial conversion efficiency exceeding 29.90% is attained within the thickness range of 250 to 3000 nm and a defect density of up to 10^{12} cm^{-3}. The optimal parameters led to a PCE of 29.01%, accompanied by a FF of 82.49%, a J_{SC} of 45.35 mA/cm^2, and a V_{OC} of 0.77V.

2. Influence of thickness and interface effect on solar energy efficiency

The most effective setup to attain the highest attainable PCE of 28.59% has been identified, entailing a 1000 nm thickness for the Cs_2SnBr_6 absorber layer and an interface defect density of 10^{12} cm^{-3}. These configurations resulted in a solar cell with an V_{OC} of 1.02 V, a J_{SC} of 31.89 mA/cm^2, and a FF of 88.28%. Within the Al/FTO/TiO2/Rb_2SnBr_6/Au structure, V_{OC}, J_{SC}, FF, and PCE tend to decrease with an increase in defect density and rise with the absorber layer thickness that's shown in Fig S4(a) to Fig S4(d).

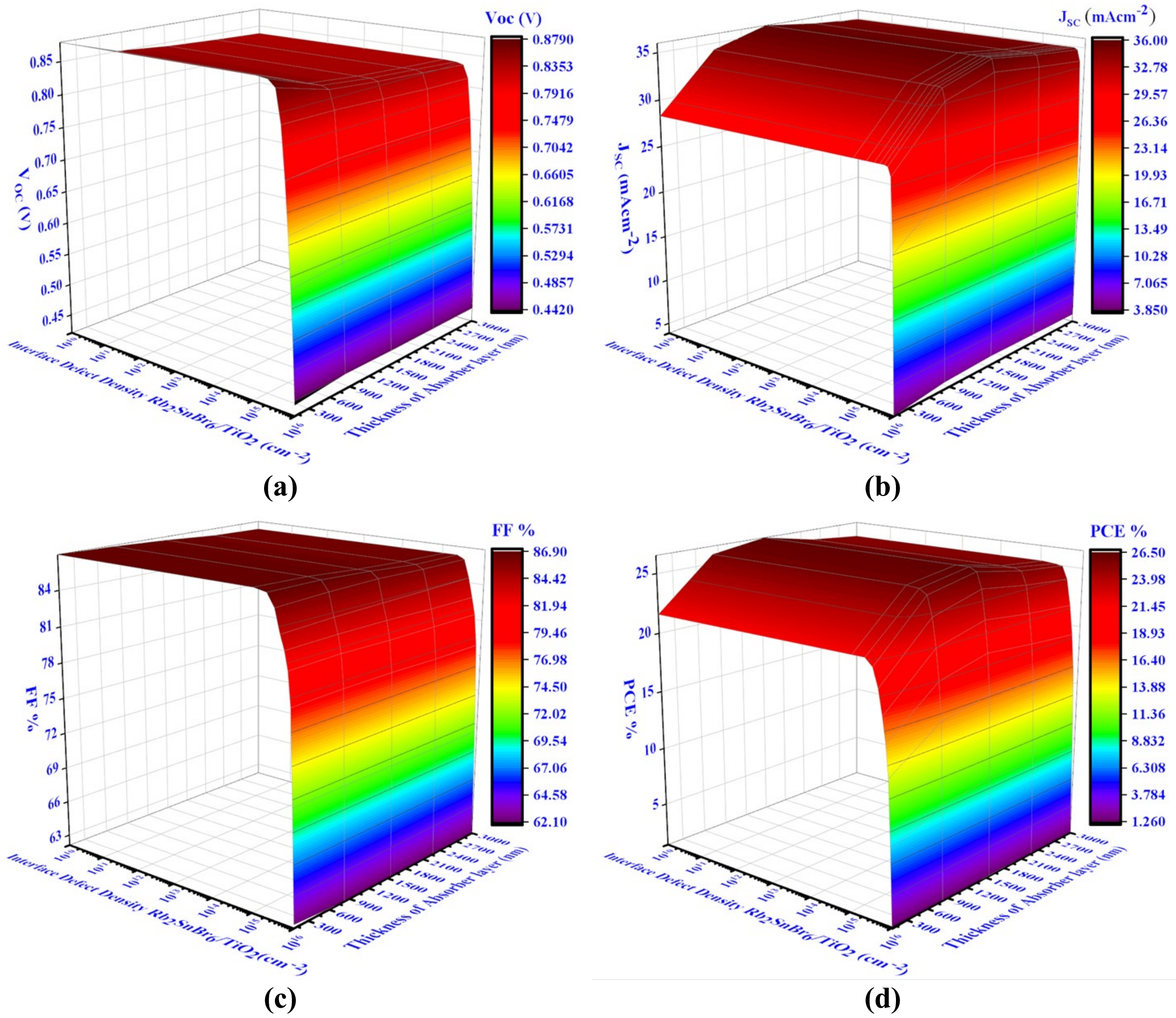


Fig. S4 Influence of variations in absorber thickness and defect interface density for Rb_2SnBr_6 on key PV performance factors: (a) V_{OC}, (b) J_{SC}, (c) FF and (d) PCE.

For absorber layer thicknesses spanning from 100 to 3000 nm and defect densities ranging from 10^{10} to 10^{16} cm^{-3}, the alterations noted in PCE, FF, J_{SC}, and V_{OC} are as follows: 25.5% to 1.26%, 86.90% to 62.10%, 36.00 mA/cm^2 to 3.85 mA/cm^2, and 0.88V to 0.44V, respectively. Up to a thickness of 1000 nm, there is a noticeable upswing in these metrics; nevertheless, once the thickness exceeds 1000 nm, the increments become marginal. Similarly, these metrics experience a significant decline when the interface defect density is elevated from 10^{12} cm^{-3}. Therefore, the ideal parameters consist of a thickness of 1000 nm and an interface defect density of 10^{12} cm^{-3}. With these optimum values, PCE, FF, J_{SC}, and V_{OC} are 26.51%, 86.49%, 36.02 mA/cm^2, and 0.84 V, respectively.

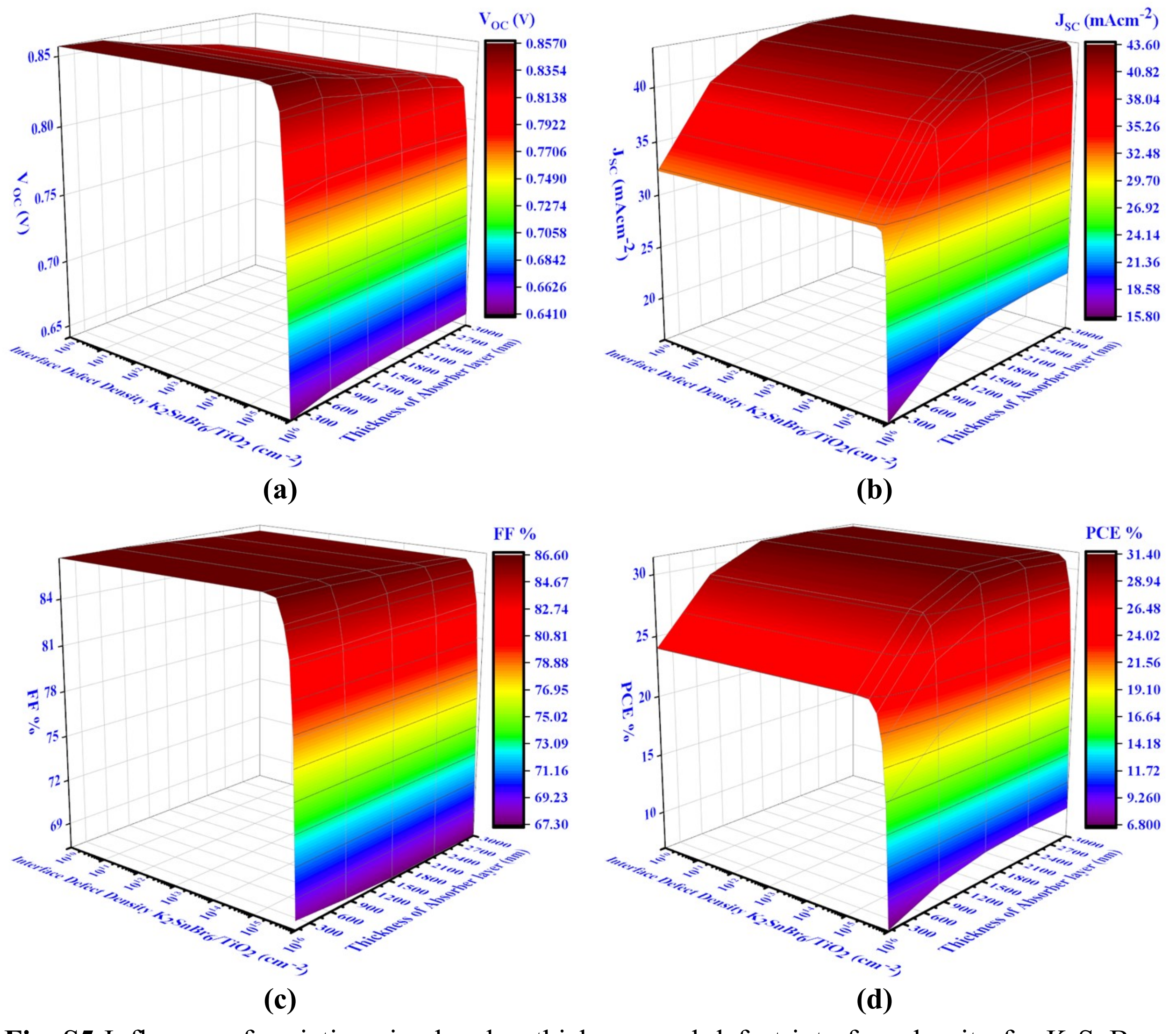


Fig. S5 Influence of variations in absorber thickness and defect interface density for K_2SnBr_6 on key PV performance factors: (a) V_{OC}, (b) J_{SC}, (c) FF and (d) PCE.

For absorber layer thicknesses ranging from 100 to 3000 nm and interface defect densities varying between 10^{10} and 10^{16} cm^{-3}, PCE, FF, J_{SC}, and V_{OC} of $Al/FTO/TiO_2/K_2SnBr_6/Au$ structures decline from 31.40% to 6.80%, 86.60% to 67.30%, 43.60 mA/cm^2 to 15.80 mA/cm^2, and 0.86 V to 0.64 V, respectively. As indicated in the results from Fig. S5(a), achieving the highest V_{OC} of 0.86 V requires maintaining the thickness below 1000 nm and ensuring the interface defect density remains below 10^{12} cm^{-3}.Nevertheless, surpassing the 10^{12} cm^{-3} defect density threshold results in a substantial drop in the V_{OC} to 0.71 V. As indicated by the data in Fig. S5(b), attaining the highest J_{SC} value of 43.60 mA/cm^2 necessitates a thickness exceeding 1000 nm while keeping the defect density below 10^{12} cm^{-3}. Furthermore, as shown in Fig. S5(c), the FF attains its highest value of 85.20% when the thickness is less than or equal to 1000 nm, and the interface defect density does not surpass 10^{12} cm^{-3}. Finally, Fig. S5(d) illustrates that the most substantial conversion efficiency, exceeding 31%, is reached within the range of thickness from 100 to 3000 nm and a defect density of up to 10^{12} cm^{-3}. The optimal configurations yielded a PCE of 31.12%, accompanied by a FF of 86.34%, a J_{SC} of 42.59 mA/cm^2, and a V_{OC} of 0.85 V.

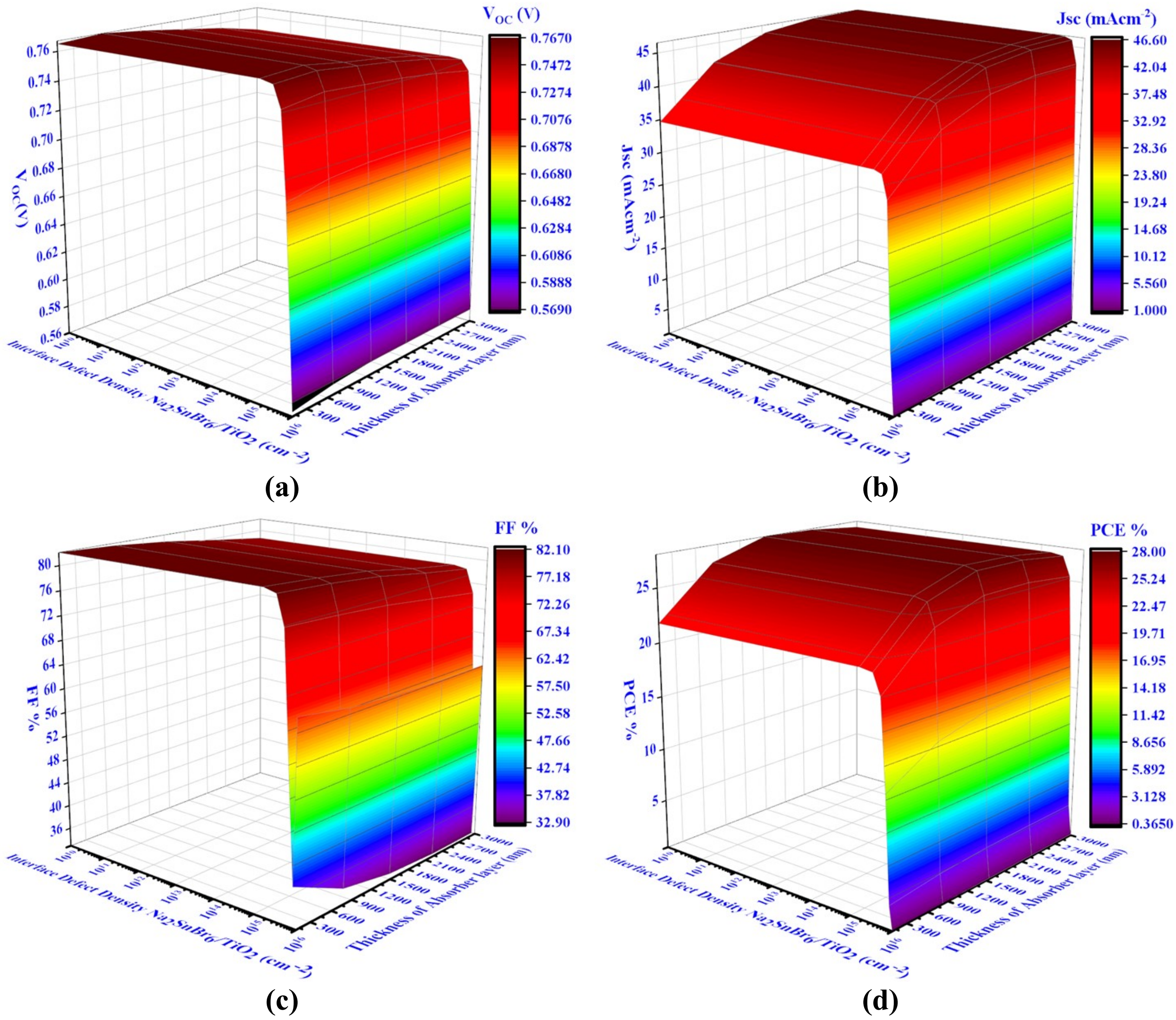


Fig. S6 Influence of variations in absorber thickness and defect interface density for Na_2SnBr_6 on key PV performance factors: (a) V_{OC}, (b) J_{SC}, (c) FF and (d) PCE.

Within the Al/FTO/TiO_2/Na_2SnBr_6/Au structure, there is a tendency for PCE, FF, J_{SC}, and V_{OC} values to decline with an increase in defect density and rise with the thickness of the absorber layer that's shown in Fig S6(a) to Fig S6(d). For the absorber layer thickness spanning from

100 to 3000 nm and the defect density ranging from 10^{10} to 10^{16} cm^{-3}, the alterations observed in PCE, FF, J_{SC}, and V_{OC} are as follows: from 28.00% to 0.37%, 82.20% to 32.90%, 46.60 mA/cm^2 to 1.00 mA/cm^2, and 0.78V to 0.57V, respectively. Up to a thickness of 1000 nm, there is a noticeable rise in these values; nevertheless, once the thickness exceeds 1000 nm, the increases become marginal. Similarly, these metrics experience a significant decline with the elevation of the interface defect density from 10^{12} cm^{-3}. Nevertheless, surpassing this threshold results in a slight decrease in the metrics. Therefore, the ideal conditions involve maintaining a thickness of 1000 nm and an interface defect density of 10^{12} cm^{-3}. With these optimized values, PCE, FF, J_{SC}, and V_{OC} are 27.74%, 80.38%, 45.27 mA/cm^2, and 0.76 V, respectively[2].

References

1 D. . Sarkar, A. K. Mahmud Hasan, M. Mottakin, V. Selvanathan, K. Sobayel, M. Ariful Islam, G. Muhammad, M. Aminuzzaman, M. Shahiduzzaman, K. Sopian and M. Akhtaruzzaman, *Solar Energy*, 2022, **243**, 215–224.

2 M. S. Uddin, M. A. Al Mashud, G. F. I. Toki, R. Pandey, M. Zulfiqar, O. Saidani, K. Chandran, M. Ouladsmane and M. K. Hossain, *Journal of Optics*, 2024, **53**, 3726–3742.