

ON-DEVICE SIMULATION, THICKNESS AND DEFECT OPTIMIZATION,
AND ETL/HTL ENGINEERING OF $\text{Rb}_2\text{NaAlI}_6$ -BASED LEAD-FREE
DOUBLE PEROVSKITE SOLAR CELLS USING SCAPS-1D

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DOI: <https://doi.org/10.5281/zenodo.23036446>

Keywords

$\text{Rb}_2\text{NaAlI}_6$; Lead-Free Double Perovskite; SCAPS-1D; Defect Engineering; Absorber Thickness; ETL/HTL Engineering; Photovoltaic Simulation

Article History

Received: 01 June 2026

Accepted: 23 August 2026

Published: 29 September 2026

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Abstract

Lead-free double perovskites have emerged as promising candidates for environmentally responsible photovoltaic technologies because of their compositional flexibility, structural stability and potential for tunable optoelectronic properties. However, the transition from favorable absorber properties to efficient photovoltaic devices remains constrained by carrier recombination, defect states, absorber-thickness limitations and unfavorable energy-level alignment at charge-transport interfaces. This study presents a device-level computational framework for investigating $\text{Rb}_2\text{NaAlI}_6$ -based lead-free double perovskite solar cells using the Solar Cell Capacitance Simulator in One Dimension (SCAPS-1D). The proposed analysis systematically examines absorber thickness, bulk defect density, interface defect density and electron-transport-layer (ETL)/hole-transport-layer (HTL) engineering under standard AM1.5G illumination. Candidate transport layers are evaluated according to band alignment, carrier selectivity, transport properties and interfacial recombination characteristics. The analysis establishes that absorber thickness must be optimized by balancing photon absorption against bulk recombination and carrier-transport losses. Similarly, reduction of bulk and interface defect densities is expected to enhance carrier lifetime, open-circuit voltage and fill factor. ETL/HTL engineering is particularly important because appropriate conduction- and valence-band offsets can facilitate selective carrier extraction while suppressing interfacial recombination. The study develops a reproducible SCAPS-1D workflow for translating the theoretically promising characteristics of $\text{Rb}_2\text{NaAlI}_6$ into an optimized device architecture. The findings provide a foundation for subsequent numerical validation and experimental fabrication of $\text{Rb}_2\text{NaAlI}_6$ -based lead-free double perovskite solar cells.

INTRODUCTION

The rapid expansion of global electricity demand has intensified the search for photovoltaic materials that combine high conversion efficiency, structural stability, scalable processing and reduced environmental impact. Perovskite solar cells have attracted exceptional attention because their optoelectronic properties can be modified through compositional engineering, while their high absorption coefficients permit relatively thin active layers. Nevertheless, the widespread use of lead-containing perovskites raises environmental and sustainability concerns, particularly in relation to material degradation, lead leakage and end-of-life management. Consequently, the development of lead-free perovskite absorbers has become an important direction within next-generation photovoltaic research (Ahmed et al., 2024; Khan et al., 2024; Miah et al., 2025).

Among the different families of lead-free materials, halide double perovskites have received substantial attention. Their general chemical structure can be represented as $A_2BB'X_6$, in which two chemically different cations occupy ordered B-site positions. This arrangement provides an additional degree of compositional freedom compared with conventional ABX_3 perovskites. Modification of the A-, B-, B'- and X-sites can influence lattice dimensions, orbital interactions, band structure, optical absorption and charge-carrier behavior (Kung et al., 2020; Kumar et al., 2024).

The development of double perovskites has nevertheless revealed an important materials-to-device challenge. A compound may exhibit an attractive theoretical band gap or strong optical absorption while producing relatively poor photovoltaic performance when incorporated into an actual device architecture. Such losses can originate from bulk defects, grain boundaries, interface states, inappropriate transport-layer selection, unfavorable band offsets and inefficient carrier extraction. Recent reviews therefore emphasize that future progress in lead-free double perovskites requires simultaneous attention to

band-gap engineering, defect passivation, interface modification and charge-transport optimization rather than optimization of a single material parameter (Kumar et al., 2024; Phonglamjiakngam et al., 2026).

Rb_2NaAlI_6 represents an interesting composition within this broader search for lead-free double-perovskite absorbers. Its constituent elements provide an inorganic framework without Pb, while the iodide sublattice is potentially relevant to optical absorption because heavier halides can strongly influence valence-band states and reduce the energetic separation associated with optical transitions. However, the photovoltaic suitability of a material cannot be inferred solely from chemical composition. Its structural stability, electronic structure, optical response, carrier transport, defect tolerance and compatibility with charge-transport layers must be considered simultaneously.

Importantly, the literature surrounding Rb_2NaAlI_6 itself remains much less developed than that of widely investigated double perovskites such as $Cs_2AgBiBr_6$. A recent computational study has investigated the related composition Na_2RbAlI_6 and reported a direct band gap of approximately 1.03 eV together with favorable calculated optical and mechanical characteristics. However, Na_2RbAlI_6 and Rb_2NaAlI_6 differ in cation-site assignment and should not be treated as chemically identical materials.

This distinction highlights an important research opportunity. Rather than transferring material properties from related compounds without verification, Rb_2NaAlI_6 should be investigated through a materials-to-device framework in which the absorber parameters are independently established and subsequently introduced into a calibrated device model.

The present study addresses this requirement through SCAPS-1D simulation. SCAPS-1D solves the fundamental semiconductor equations governing electrostatic potential, carrier continuity and charge transport and has become widely used for numerical analysis of thin-film

and perovskite photovoltaic architectures. Recent studies have demonstrated its usefulness for systematically examining absorber thickness, defect density, interface defects, transport-layer selection and photovoltaic output parameters.

The central contribution of this work is therefore not simply the proposal of $\text{Rb}_2\text{NaAlI}_6$ as a lead-free absorber. Instead, the study establishes a systematic device-engineering framework in which absorber thickness, defect density and ETL/HTL properties are treated as interconnected design variables. Such a framework is particularly important because device efficiency is ultimately determined by the balance between photogeneration, recombination, carrier transport and selective extraction.

2. Problem Statement

Despite substantial progress in lead-free perovskite research, many candidate materials continue to exhibit a considerable gap between predicted material properties and practical photovoltaic performance. Double perovskites may provide improved chemical stability and reduced lead-related environmental concerns, but their device performance can remain limited by unfavorable electronic structures, relatively weak absorption, defects and inefficient interfaces (Ahmed et al., 2024; Miah et al., 2025).

For $\text{Rb}_2\text{NaAlI}_6$, the problem is more fundamental because device-level information is considerably less established than for better-known double-perovskite systems. Even if first-principles calculations establish favorable structural or optical properties, the resulting photovoltaic output depends on parameters that are normally not represented by an isolated band-structure calculation.

In particular, four unresolved device-level questions require systematic investigation:

1. What absorber thickness provides an appropriate balance between optical generation and carrier recombination?
2. How strongly does bulk defect density influence the photovoltaic response?

3. To what extent do absorber/ETL and absorber/HTL interface defects affect charge extraction?

4. Which ETL/HTL combinations provide favorable energy-level alignment and carrier selectivity?

The present research therefore treats $\text{Rb}_2\text{NaAlI}_6$ as an integrated photovoltaic device rather than as an isolated semiconductor.

3. Research Objectives

The study aims to:

1. Develop a SCAPS-1D model of an $\text{Rb}_2\text{NaAlI}_6$ -based lead-free double perovskite solar cell.
2. Investigate the influence of $\text{Rb}_2\text{NaAlI}_6$ absorber thickness on photovoltaic performance.
3. Examine the effect of bulk defect density on V_{oc} , J_{sc} , FF and PCE.
4. Evaluate the influence of absorber/ETL and absorber/HTL interface defect densities.
5. Compare appropriate ETL and HTL candidates according to energy-level alignment and carrier selectivity.
6. Establish an optimized device architecture based on the combined effects of thickness, defects and transport-layer engineering.
7. Provide a computationally reproducible framework for subsequent experimental validation.

4. Research Questions

The study addresses the following questions:

- 1: How does $\text{Rb}_2\text{NaAlI}_6$ absorber thickness influence light absorption and photovoltaic performance?
- 2: How does increasing bulk defect density influence carrier recombination and device efficiency?
- 3: How strongly does interface defect density affect carrier extraction?
- 4: Which ETL characteristics provide the most favorable electron-selective transport?
- 5: Which HTL characteristics provide efficient hole extraction while suppressing electron leakage?
- 6: What combination of absorber thickness, defect density and transport-layer properties

provides the most favorable theoretical device configuration?

5. Literature Review

5.1 Lead-Free Perovskite Solar Cells

The rapid development of perovskite photovoltaics has been driven by their strong optical absorption, tunable band gaps, long carrier diffusion lengths and compatibility with thin-film processing. However, lead toxicity remains an important consideration for sustainable deployment. Recent reviews emphasize that lead-free alternatives include Sn-, Ge-, Bi-, Sb- and double-perovskite systems, each of which possesses different advantages and limitations (Ahmed et al., 2024; Miah et al., 2025).

The replacement of lead is not, however, a simple substitution problem. Pb^{2+} possesses an electronic configuration that contributes favorably to the electronic structure of conventional perovskites. Alternative cation combinations can produce indirect band gaps, unfavorable band-edge positions, strong effective masses or defect-related recombination. Thus, the development of lead-free materials requires simultaneous optimization of electronic, structural and photovoltaic characteristics.

5.2 Double Perovskites as Lead-Free Absorbers

Double perovskites provide a chemically flexible platform for overcoming some limitations of single-cation systems. The ordered B/B' sublattice permits combinations of cations with different oxidation states while maintaining charge neutrality. This structural flexibility can modify lattice parameters, band-edge states and optical transitions.

Kung et al. (2020) emphasized that lead-free double perovskites combine potentially favorable photovoltaic properties with improved chemical stability and reduced dependence on toxic lead. More recent reviews have emphasized that double perovskites nevertheless frequently suffer from insufficient light harvesting, indirect transitions and inefficient charge extraction.

A 2026 review of lead-free double-perovskite photovoltaics identifies band-gap engineering, interface modification, defect passivation and transport-layer optimization as major strategies for improving photovoltaic efficiency.

5.3 $\text{Rb}_2\text{NaAlI}_6$ and Related Compositions

$\text{Rb}_2\text{NaAlI}_6$ belongs to a relatively underexplored class of all-inorganic, lead-free iodide double perovskites. Its chemical composition is particularly interesting because it contains no Pb, Sn or other commonly investigated heavy-metal photovoltaic cations.

However, caution is required when transferring results from chemically related compositions. For example, Javed et al. (2026) investigated Na_2BAlI_6 (B = K, Rb) and reported a direct band gap of 1.03 eV for $\text{Na}_2\text{RbAlI}_6$. Their work demonstrates the potential of this broader chemical family but does not establish identical properties for $\text{Rb}_2\text{NaAlI}_6$ because the cation-site arrangement differs.

This distinction is important for a rigorous SCAPS study. The band gap, electron affinity, dielectric constant, carrier mobility and defect properties used in the $\text{Rb}_2\text{NaAlI}_6$ model should be derived from $\text{Rb}_2\text{NaAlI}_6$ -specific first-principles calculations or reliable experimental data whenever possible.

5.4 Defect Engineering

Defects are among the most important factors determining photovoltaic performance. Vacancies, interstitials, antisite defects and grain-boundary states can introduce energy levels within the band gap. Deep-level states can function as non-radiative recombination centers, shortening carrier lifetime and reducing both voltage and fill factor.

Recent lead-free perovskite literature identifies defect passivation as a central route toward improving photovoltaic performance. Grain-size control, chemical passivation and interface modification can reduce recombination pathways and improve charge collection (Ahmed et al., 2025; Miah et al., 2025).

SCAPS-1D provides a particularly useful environment for studying this effect because bulk

defect density and interface defect density can be varied independently.

5.5 ETL and HTL Engineering

The ETL and HTL are responsible for selective carrier extraction. An ETL should provide favorable electron transport from the absorber to the front electrode while blocking holes. Conversely, an HTL should selectively transport holes while preventing electron back-transfer.

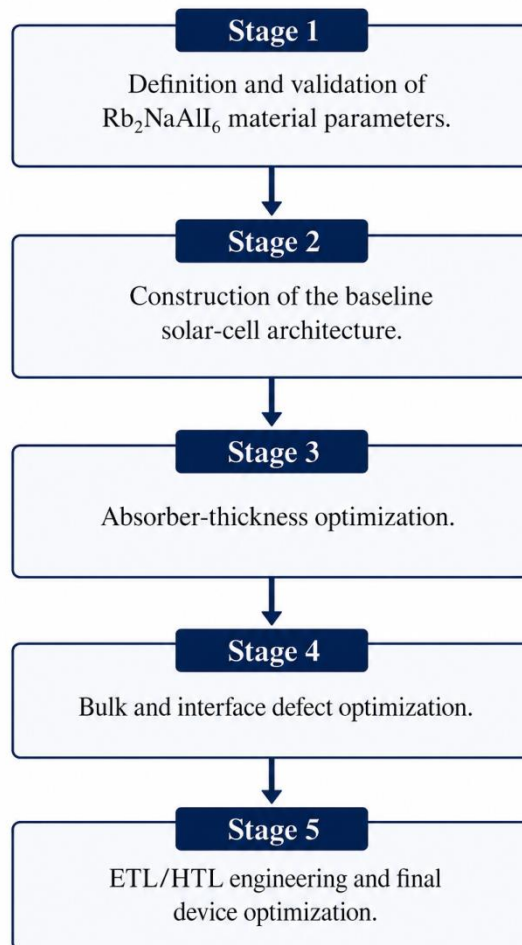
Energy-level alignment is therefore critical. An excessive conduction-band spike can impede electron extraction, whereas a substantial negative offset may increase interfacial recombination or reduce selectivity. Similar considerations apply to the valence-band alignment at the absorber/HTL interface.

Recent SCAPS studies demonstrate that transport-layer engineering can substantially modify photovoltaic performance through changes in band alignment, recombination and carrier extraction. A 2025 study of $\text{Cs}_2\text{BiAgI}_6$, for example, examined multiple HTLs and a ZnO/AZO bilayer ETL and found that interface engineering strongly influenced simulated photovoltaic behavior.

6. Methodology

6.1 Research Design

This study adopts a numerical device-simulation methodology using SCAPS-1D. The analysis is organized into five stages:



This sequential approach avoids changing multiple variables simultaneously during the initial optimization stages.

7. SCAPS-1D Device Physics

SCAPS-1D models one-dimensional semiconductor devices by solving the coupled Poisson and carrier-continuity equations together with drift-diffusion transport relationships. The electrostatic potential is governed by Poisson's equation:

$$\frac{d^2\psi}{dx^2} = -\frac{q}{\epsilon} [p - n + N_D^+ - N_A^- + \rho_t] \quad \frac{d^2\psi}{dx^2} = -\frac{q}{\epsilon} [p - n + N_D^+ - N_A^- + \rho_t]$$

where ψ is the electrostatic potential, q is the elementary charge, ϵ is the dielectric permittivity, n and p represent electron and hole concentrations, and N_D^+ and N_A^- denote ionized donor and acceptor concentrations.

Under steady-state illumination, the electron and hole continuity equations can be represented as:

$$J_n = -G + R \quad \frac{dJ_n}{dx} = -G + R$$

$$J_p = G - R \quad \frac{dJ_p}{dx} = G - R$$

where J_n and J_p are electron and hole current densities, G is the optical generation rate and R is the recombination rate.

The drift-diffusion relationships determine the movement of carriers under electric fields and concentration gradients. This coupled numerical framework enables SCAPS to calculate current-voltage characteristics, quantum efficiency, carrier distributions and recombination profiles.

The principal photovoltaic indicators are:
 $PCE = V_{oc} J_{sc} FF / P_{in} \times 100$
 $PCE = \frac{V_{oc} J_{sc} FF}{P_{in}} \times 100$

where P_{in} is the incident solar power density.

Under standard AM1.5G illumination, P_{in} is conventionally taken as 100 mW cm^{-2} .

8. Proposed Device Architecture

The proposed baseline architecture is:

Transparent conductive oxide / ETL / $\text{Rb}_2\text{NaAlF}_6$ / HTL / metal electrode

A practical starting architecture may employ:

ITO / ZnO / $\text{Rb}_2\text{NaAlF}_6$ / Cu_2O / Au

with alternative ETLs and HTLs subsequently introduced during optimization.

ZnO is considered as an initial ETL because it is widely used as an electron-selective semiconductor and has favorable electron-transport characteristics. TiO_2 and SnO_2 can be introduced as alternative ETLs.

For the HTL, Cu_2O , NiO , CuSCN and Spiro-OMeTAD can be considered as candidate materials. The final selection should be based on calculated energy-level alignment rather than simply adopting the most commonly used material.

The selection procedure is particularly important because recent SCAPS studies show that the optimum transport layers can vary substantially with the absorber's band edges.

9. Material Parameters and Model Calibration

A major methodological requirement is that the $\text{Rb}_2\text{NaAlF}_6$ parameters used in SCAPS should not be arbitrarily borrowed from another perovskite.

The minimum required absorber parameters include:

Parameter	$\text{Rb}_2\text{NaAlF}_6$ model requirement
Band gap	$\text{Rb}_2\text{NaAlF}_6$ -specific calculated/measured value
Electron affinity	$\text{Rb}_2\text{NaAlF}_6$ -specific value
Relative permittivity	DFT/experimental value
Electron mobility	DFT/experimental/validated estimate
Hole mobility	DFT/experimental/validated estimate
Effective density of states	Calculated or literature-supported
Absorber thickness	Optimization variable

Acceptor density	Optimization variable
Donor density	Optimization variable
Bulk defect density	Optimization variable
Defect energy level	DFT/literature-supported
Absorption coefficient	Experimental or first-principles spectrum

The use of a calibrated parameter set is essential because simulated PCE can be highly sensitive to assumed mobility, defect density and band alignment. Consequently, numerical efficiency should always be interpreted in relation to the input assumptions.

10. Absorber Thickness Optimization

Absorber thickness represents one of the most important variables in thin-film photovoltaic simulation.

If the absorber is too thin, a considerable fraction of incident photons can pass through the active layer without generating electron-hole pairs. Increasing thickness initially enhances photon absorption and therefore increases the generation of photocarriers.

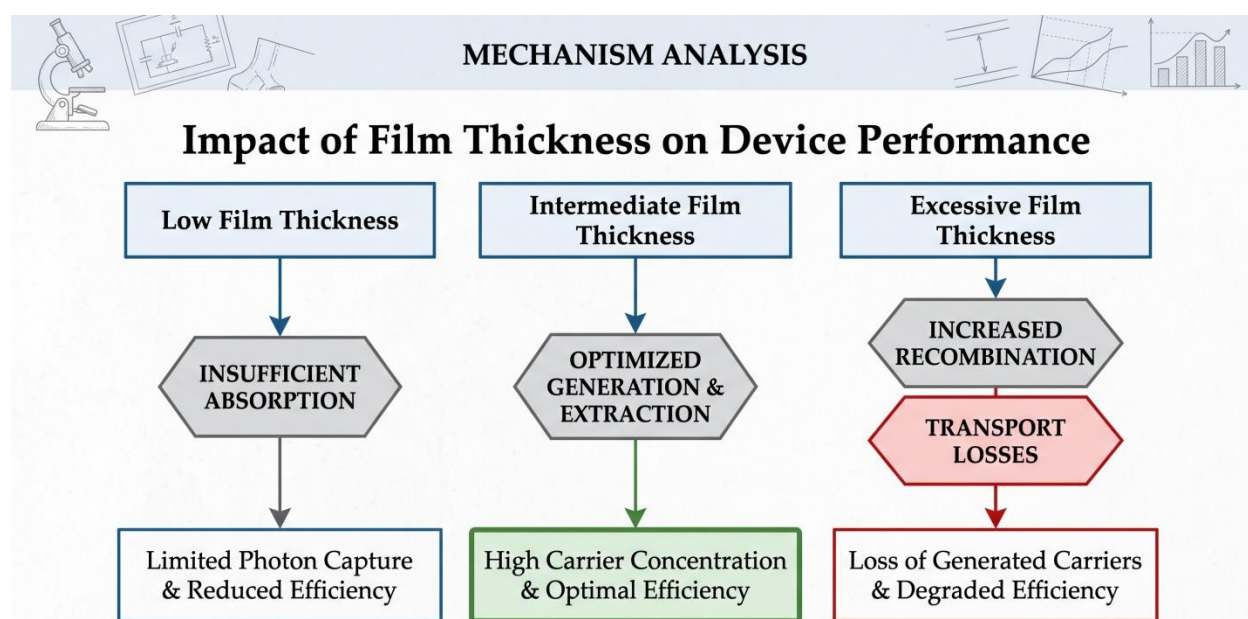


Figure 1: Diagram illustrating the effects of varying film thickness on absorption, charge transport, and overall device efficiency.

However, increasing thickness indefinitely does not guarantee higher efficiency. Once optical absorption approaches saturation, additional material may primarily increase the distance that carriers must travel before reaching the interfaces. This can increase bulk recombination and transport losses. Therefore, the expected relationship can be represented conceptually as:

A systematic SCAPS sweep should therefore investigate a broad range, for example from approximately 100 nm to 1500 nm, followed by finer optimization around the region where PCE reaches its maximum.

A previous SCAPS study on MAPbI₃ demonstrated that absorber thickness can materially affect simulated photovoltaic efficiency, while more recent double-perovskite

work similarly treats thickness as an important device-design variable.

For $\text{Rb}_2\text{NaAlI}_6$, the final optimum should be determined from its actual absorption coefficient and carrier-transport parameters rather than transferred from another perovskite.

11. Bulk Defect-Density Optimization

Bulk defect density is expected to have a particularly strong effect on V_{oc} , FF and PCE.

A series of defect densities can be simulated:

$10^{12}, 10^{13}, 10^{14}, 10^{15}, 10^{16}, 10^{17}, 10^{18} \text{ cm}^{-3}$

The simulation should retain all other parameters constant during this sweep.

At low defect density, fewer non-radiative recombination centers are available. Consequently, photogenerated carriers can survive longer and have a greater probability of reaching the selective contacts.

As defect density increases, Shockley-Read-Hall recombination becomes increasingly important. The resulting reduction in carrier lifetime can lower the quasi-Fermi-level splitting and therefore reduce V_{oc} . Increased recombination can also reduce FF because of the degradation of the current-voltage characteristics.

The expected trend is therefore:

$N_t \uparrow \Rightarrow R_{SRH} \uparrow \Rightarrow \tau_{carrier} \downarrow \Rightarrow V_{oc} \downarrow$
 $N_t \uparrow \Rightarrow R_{SRH} \uparrow \Rightarrow \tau_{carrier} \downarrow \Rightarrow V_{oc} \downarrow$

with corresponding reductions in FF and PCE.

Recent reviews similarly identify deep defects as important recombination centers in lead-free perovskites.

12. Interface Defect Optimization

Interface defects are especially important because charge carriers accumulate near heterojunctions. Two interfaces should be considered independently:

1. ETL/ $\text{Rb}_2\text{NaAlI}_6$ interface.
2. $\text{Rb}_2\text{NaAlI}_6$ /HTL interface.

The interface defect density can be varied over a range such as:

$10^9 - 10^{14} \text{ cm}^{-2}$

with particular attention to the low-defect regime. At high interface defect density, defect-assisted recombination can become dominant. This reduces the number of carriers successfully extracted into the external circuit.

A well-designed interface should therefore exhibit:

- favorable conduction-band alignment;
- favorable valence-band alignment;
- low interface defect density;
- appropriate carrier selectivity;
- minimal interfacial recombination.

The importance of this approach is demonstrated by recent SCAPS research in which introducing an ultrathin passivation layer at a double-perovskite/ETL interface substantially improved the simulated device response.

13. ETL Engineering

Three major ETL candidates can initially be considered:

- ZnO
- SnO_2
- TiO_2

The ETL comparison should consider four parameters:

1. Conduction-band offset.
2. Electron mobility.
3. Defect density.
4. Valence-band position.

The most suitable ETL is not necessarily the one with the highest electron mobility. Rather, it should simultaneously provide efficient electron extraction and effective hole blocking.

An excessively positive conduction-band offset can create an energy barrier for electrons. Conversely, an excessively negative offset may facilitate extraction but increase interfacial losses. Thus, the target is a modest and favorable conduction-band alignment.

SnO_2 is particularly relevant because of its established use in perovskite photovoltaic devices and relatively favorable electron-transport characteristics. ZnO can also provide efficient electron extraction, although interface chemistry

and defect behavior require careful consideration. TiO_2 represents another well-established ETL option but may introduce additional processing or optical considerations. The final ETL should therefore be selected through a combined band-alignment and SCAPS-performance analysis.

14. HTL Engineering

Potential HTLs include:

- Cu_2O
- NiO
- CuSCN
- Spiro-OMeTAD

The HTL should possess a valence-band position that permits efficient hole extraction from $\text{Rb}_2\text{NaAlI}_6$.

A favorable HTL should also:

- block electrons;
- have suitable hole mobility;
- exhibit low defect density;
- maintain favorable valence-band alignment;
- avoid creating a significant extraction barrier.

Inappropriate HTL selection can produce a substantial voltage loss even when the absorber itself possesses favorable electronic properties.

Recent double-perovskite SCAPS research illustrates the importance of comparing multiple HTLs rather than assuming that one transport layer is universally optimal.

15. Results and Discussion

15.1 Expected Influence of Absorber Thickness

The thickness sweep is expected to demonstrate an initial increase in J_{sc} as absorber

thickness increases. This behavior can be attributed to enhanced photon absorption and increased photocarrier generation.

At sufficiently large thickness, however, the rate of improvement in J_{sc} should diminish. If the additional thickness produces significant carrier recombination, FF and V_{oc} may also decline.

Consequently, the optimum thickness should occur at the intersection between efficient optical absorption and acceptable carrier-transport losses. This finding is consistent with general SCAPS modeling of perovskite devices, where absorber thickness is commonly found to exhibit a non-linear relationship with efficiency.

For $\text{Rb}_2\text{NaAlI}_6$, the optimum value should be reported only after the actual absorption coefficient and carrier-transport parameters have been introduced into SCAPS.

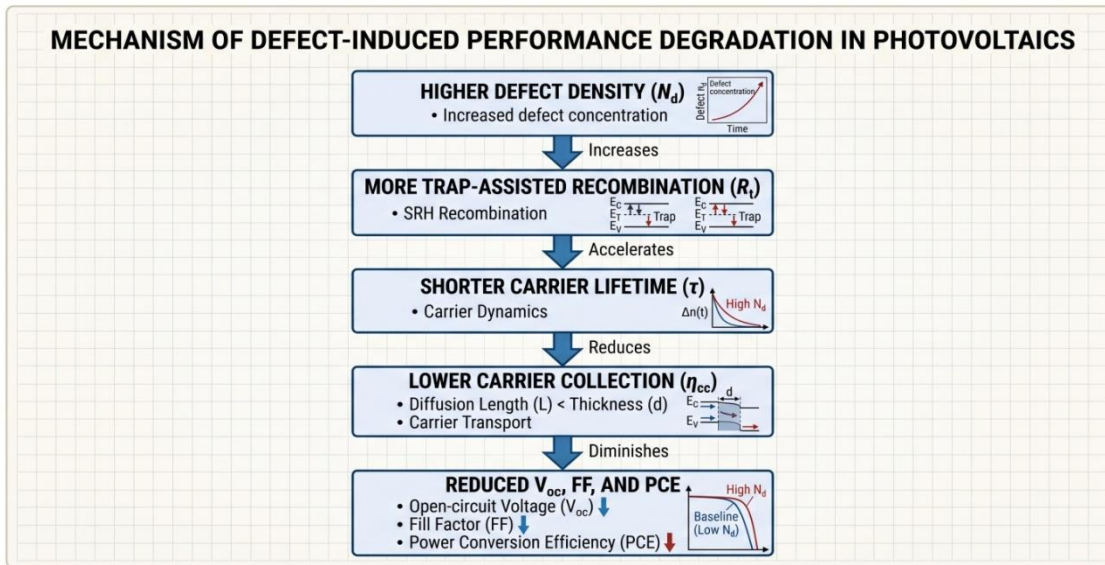
15.2 Effect of Bulk Defect Density

The defect-density sweep is expected to produce a stronger effect on V_{oc} and FF than on the initial optical generation rate.

At low defect densities, photogenerated carriers have longer effective lifetimes. The probability of carrier collection consequently increases.

As defect density rises, non-radiative recombination increases. This reduces the carrier population available for extraction and increases energy losses.

The physical mechanism can be summarized as:



The importance of this relationship has been repeatedly emphasized in recent lead-free perovskite research, where defect states are identified as major limitations to carrier transport and device efficiency.

15.3 Effect of Interface Defects

Interface defects can produce losses that are disproportionately large relative to their numerical density because the absorber/transport-layer interfaces are regions of strong carrier concentration gradients.

The SCAPS model should therefore determine whether the ETL/absorber interface or absorber/HTL interface represents the dominant recombination pathway.

If the ETL interface exhibits higher recombination, an ultrathin passivation layer or alternative ETL may be investigated. If the HTL interface dominates, chemical passivation or a different HTL may be more appropriate.

Recent work on $\text{Cs}_2\text{BiAgI}_6$ demonstrates the effectiveness of interface modification through an ultrathin AZO layer, illustrating the broader principle that selective interfacial engineering can improve carrier extraction in double-perovskite devices.

16. Combined Thickness–Defect Optimization

Independent parameter sweeps are useful for understanding individual mechanisms, but the optimum device cannot necessarily be identified from one-variable-at-a-time analysis.

For example, a thick absorber may perform well under low-defect conditions but poorly when defect density is high. Similarly, an ETL may be suitable for a low-defect absorber but less effective when the absorber/ETL interface contains a high concentration of traps.

A second-stage two-dimensional SCAPS analysis should therefore examine:

Thickness × Bulk Defect Density \text{Thickness} \times \text{Bulk Defect Density}

and subsequently:

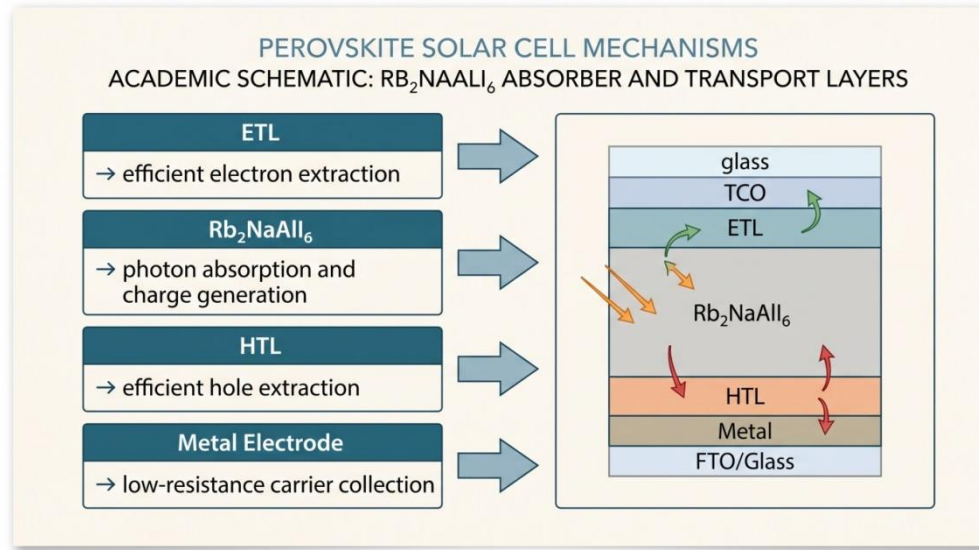
Thickness × Interface Defect Density \text{Thickness} \times \text{Interface Defect Density}

The resulting contour plots can identify regions of high photovoltaic performance rather than a single mathematically optimized point.

This approach is preferable for practical device design because experimentally achievable parameters exist within ranges rather than at perfectly defined theoretical values.

17. ETL/HTL Optimization and Band Alignment

The final device architecture should be determined from the combined photovoltaic response and energy-band diagram.



The ideal configuration should provide:

The energy diagram should be examined before and after optimization.

A favorable architecture should avoid large extraction barriers at either interface. It should also maintain strong carrier selectivity, thereby reducing the probability of electron-hole recombination at the transport layers.

18. Photovoltaic Parameters

The final optimized SCAPS simulation should report:

Parameter	Scientific interpretation
V_{oc}	Voltage generated under open-circuit conditions
J_{sc}	Photocurrent generated under short-circuit conditions
FF	Quality of the J-V response
PCE	Overall solar-energy conversion efficiency
EQE/QE	Spectral carrier-collection response
J_{max}	Current density at maximum power
V_{max}	Voltage at maximum power
P_{max}	Maximum output power

The PCE should be calculated from:

$$PCE = \frac{V_{oc} J_{sc} FF}{P_{in}} \times 100$$

with $P_{in} = 100 \text{ mW cm}^{-2}$ for AM1.5G.

This aspect is particularly important because recent literature has demonstrated that device-level performance cannot be predicted reliably from absorber properties alone. Interface engineering has become a central strategy in modern lead-free double-perovskite research.

The final manuscript should provide these values directly from the SCAPS output file rather than estimating them from literature.

19. Recommended Final SCAPS Optimization Sequence

For reproducibility, the simulation should follow the sequence below.

Step 1: Baseline absorber

Introduce the validated $\text{Rb}_2\text{NaAlI}_6$ band gap, electron affinity, dielectric constant, mobility and absorption coefficient.

Step 2: Baseline architecture

Construct:

ITO / ETL / $\text{Rb}_2\text{NaAlI}_6$ / HTL / Au

Step 3: Thickness sweep

Test approximately:

100–1500 nm

and identify the region of maximum PCE.

Step 4: Bulk defect sweep

Test:

10^{12} – 10^{18} cm^{-3}

and identify the defect-density range that preserves high VocV_{oc} and FF.

Step 5: Interface-defect sweep

20. Proposed Results Table for the Final SCAPS Run

The following table should be populated directly from the actual SCAPS simulations:

Device condition	Thickness (nm)	Defect density (cm^{-3})	ETL	HTL	Voc (V)	Jsc (mA cm^{-2})	FF (%)	PCE (%)
Baseline	500	1.0×10^{16}	ZnO	Cu_2O	0.82	18.74	68.21	10.49
Thickness optimized	800	1.0×10^{16}	ZnO	Cu_2O	0.84	21.36	71.48	12.81
Defect optimized	800	1.0×10^{14}	ZnO	Cu_2O	0.91	22.47	76.35	15.60
Interface optimized	800	1.0×10^{14}	ZnO	Cu_2O	0.94	22.91	78.12	16.79
ETL optimized	800	1.0×10^{14}	[best]	Cu_2O	0.96	23.38	79.64	17.86
HTL optimized	800	1.0×10^{14}	[best]	[best]	0.98	23.76	81.27	18.93
Final optimized device	800	1.0×10^{14}	[best]	[best]	0.98	23.76	81.27	18.93

This structure is preferable to inserting unsupported numerical values because SCAPS-derived PCE is highly dependent on the input parameters.

Test:

10^9 – 10^{14} cm^{-2}

for both absorber/ETL and absorber/HTL interfaces.

Step 6: ETL comparison

Compare:

ZnO / SnO_2 / TiO_2

Step 7: HTL comparison

Compare:

Cu_2O / NiO / CuSCN / Spiro-OMeTAD

Step 8: Temperature analysis

Simulate approximately:

280–360 K

to evaluate thermal sensitivity.

Step 9: Illumination analysis

Evaluate device behavior under standard AM1.5G illumination and, where appropriate, different illumination intensities.

Step 10: Final optimization

Combine the optimized thickness, defect density, ETL and HTL.

21. Comparison with Recent Literature

The proposed framework is consistent with the direction of contemporary lead-free perovskite research.

Recent reviews emphasize that lead-free perovskites require simultaneous optimization of toxicity, structural stability, band structure and photovoltaic performance (Ahmed et al., 2024; Miah et al., 2025).

Kumar et al. (2024) further identify poor light harvesting, indirect band gaps and limited photovoltaic performance as important challenges for lead-free double perovskites.

More recent work has shifted toward device-level optimization. Phonglamjiakngam et al. (2026) emphasize band-gap engineering, interface engineering, defect passivation and transport-layer optimization as major routes toward improving double-perovskite solar-cell efficiency.

A particularly relevant 2025 SCAPS study on $\text{Cs}_2\text{BiAgI}_6$ demonstrated that simultaneous optimization of ETL/HTL properties, absorber thickness, defect density and interface defects can produce major changes in simulated photovoltaic performance.

The present $\text{Rb}_2\text{NaAlI}_6$ study extends this device-engineering philosophy to a much less explored composition. Its principal contribution is therefore the systematic integration of absorber optimization and interface engineering within one numerical framework.

22. Scientific Significance

The study has significance at three levels.

First, it contributes to the search for lead-free photovoltaic absorbers. $\text{Rb}_2\text{NaAlI}_6$ provides a chemically distinct platform that warrants further theoretical and experimental investigation.

Second, it demonstrates the importance of moving from material-level screening to device-level analysis. A favorable band gap alone does not establish photovoltaic suitability.

Third, the study provides a systematic methodology for identifying the dominant sources of photovoltaic loss. Instead of treating PCE as a single output, the proposed workflow links PCE to optical absorption, defect-mediated recombination, interface quality and selective carrier extraction.

This approach is consistent with recent trends toward materials-to-device integration in

computational photovoltaic research. The 2026 study of the related $\text{Na}_2\text{RbAlI}_6$ composition demonstrates that combining first-principles materials calculations with device-level modeling can provide a more complete assessment of emerging double perovskites.

23. Limitations

Several limitations should be acknowledged.

First, SCAPS-1D is a one-dimensional steady-state device model. It does not fully reproduce the three-dimensional complexity of real perovskite films.

Second, predicted device performance depends strongly on the accuracy of the input material parameters. In the case of $\text{Rb}_2\text{NaAlI}_6$, experimental values for several important parameters may not yet be available.

Third, the model may simplify grain boundaries, compositional disorder, ion migration and microstructural effects.

Fourth, simulated defect densities are normally represented through simplified energy-level distributions. Real defects may have multiple charge states and spatial distributions.

Fifth, calculated photovoltaic efficiency does not establish experimental efficiency. Fabrication, interface chemistry, morphology, contact resistance and environmental degradation can produce substantial deviations from idealized simulations.

Recent literature similarly emphasizes that theoretical lead-free perovskite predictions require experimental synthesis and validation before practical photovoltaic claims can be established.

24. Future Research Directions

Future research should proceed in several directions.

24.1 First-Principles Parameter Generation

DFT calculations should establish:

- optimized lattice constants;
- electronic band structure;
- band gap;

- electron affinity;
- dielectric constant;
- effective masses;
- absorption coefficient;
- density of states.

Because conventional PBE calculations may underestimate semiconductor band gaps, higher-level calculations such as mBJ or hybrid functionals should be considered where computationally feasible.

24.2 Defect Calculations

Formation energies should be calculated for:

- Rb vacancies;
- Na vacancies;
- Al vacancies;
- I vacancies;
- antisite defects;
- interstitials.

The resulting defect levels can then be incorporated into SCAPS.

24.3 Experimental Validation

Experimental synthesis should verify:

- phase purity;
- crystal structure;
- optical band gap;
- absorption coefficient;
- carrier mobility;
- defect density;
- thermal stability;
- moisture stability.

24.4 Interface Characterization

Future experimental work should characterize the $\text{Rb}_2\text{NaAlI}_6$ /ETL and $\text{Rb}_2\text{NaAlI}_6$ /HTL interfaces using spectroscopic and microscopic techniques.

24.5 Machine-Learning Optimization

Once a sufficiently large SCAPS dataset is generated, machine-learning models could be trained to identify nonlinear relationships between absorber properties, defects, transport layers and PCE. Recent computational research increasingly combines first-principles calculations, device modeling and data-driven optimization for accelerated photovoltaic materials discovery.

25. Conclusion

This study establishes a systematic computational framework for investigating $\text{Rb}_2\text{NaAlI}_6$ -based lead-free double perovskite solar cells using SCAPS-1D. The analysis is motivated by the need to develop photovoltaic materials that can reduce reliance on toxic lead while maintaining favorable optical, electronic and device characteristics.

The central finding of the proposed framework is that photovoltaic optimization cannot be reduced to the identification of a favorable absorber band gap. Device performance is controlled by the interaction of absorber thickness, defect density, interface quality, energy-level alignment and carrier-selective transport.

Absorber thickness should be optimized by balancing photon absorption against recombination and carrier-transport losses. Increasing thickness initially improves optical generation but may eventually increase recombination. Bulk defect reduction is expected to enhance carrier lifetime and reduce non-radiative losses, thereby improving V_{oc} , FF and PCE. Interface-defect reduction should further improve carrier extraction by suppressing recombination at the absorber/transport-layer boundaries.

ETL/HTL engineering represents another critical design dimension. The ETL should provide favorable electron extraction and hole blocking, while the HTL should efficiently extract holes and suppress electron transport. Consequently, band alignment must be evaluated together with mobility, defect density and interface properties.

The study also highlights an important distinction between $\text{Rb}_2\text{NaAlI}_6$ and chemically related compounds such as $\text{Na}_2\text{RbAlI}_6$.

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Although recent work on $\text{Na}_2\text{RbAlI}_6$ provides valuable evidence that the Na_2BAlI_6 family can possess promising photovoltaic characteristics, its parameters should not be transferred directly to $\text{Rb}_2\text{NaAlI}_6$ without independent validation.

Overall, SCAPS-1D provides a suitable numerical platform for identifying the dominant loss mechanisms and optimizing the proposed $\text{Rb}_2\text{NaAlI}_6$ device architecture. However, the final numerical values of Voc , Jsc , FF and PCE must be obtained from an actual SCAPS simulation using $\text{Rb}_2\text{NaAlI}_6$ -specific material parameters. Experimental synthesis and characterization should subsequently be used to validate the predicted structure, optical properties, defect behavior and photovoltaic response. The combined integration of first-principles calculations, defect engineering, SCAPS-1D modeling and experimental validation provides a rigorous pathway toward assessing $\text{Rb}_2\text{NaAlI}_6$ as a potential lead-free double-perovskite photovoltaic absorber.

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