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## Design and Development of Mixed Perovskite Solar Cells with High Efficiency and Stability through DFT and AI-Based Design Approaches

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Compared to different solar technologies, Perovskite-based solar cells are preferred by many for their high PCE and cost-effectiveness. Still, building a market-ready solution requires handling various important issues related to stability, how the device is designed, and efficiency. The study presents comprehensive approaches involving Density Functional Theory (DFT), device modeling, and Machine Learning (ML) to improve and evaluate mixed perovskite materials. DFT was utilized to study the electronic structure, energy gap, and defect properties of perovskites. By using SCAPS-1D simulations, different optimization factors were studied. Furthermore, different ML algorithms were trained to find key device properties. The training involved both experiments and simulations to learn from the data and predict how each material would work, allowing for fast screening of various perovskite compositions. Because of this framework, researchers can identify new, efficient materials and learn more about how different compositions affect solar cell performance. This strategy uses DFT modeling, numerical simulation, and data analysis together to improve the speed of developing better perovskite solar cells.

### I. INTRODUCTION

The global transition toward sustainable energy systems has positioned solar energy as a cornerstone of renewable energy infrastructure, offering a viable pathway to mitigate climate change and reduce reliance on fossil fuels. Photovoltaic (PV) technologies, which convert sunlight directly into electricity, have evolved significantly over the past decades, with silicon-based solar cells dominating the market due to their maturity and reliability [1]. However, the quest for higher efficiency, lower production costs, and scalable manufacturing has spurred intensive research into next-generation solar materials. Among these, perovskite solar cells (PSC) have emerged as a revolutionary candidate, demonstrating unprecedented progress in power conversion efficiency (PCE)-from 3.8% in 2009 to over 25% in recent years [2, 3]. This rapid advancement underscores their potential to redefine the future of photovoltaics.

**The Rise of Perovskite Solar Cells:** Perovskite materials, characterized by their  $ABX_3$  crystal structure (where A is a cation, B a metal ion, and X a halide), exhibit exceptional optoelectronic properties, including high absorption coefficients, tunable bandgaps, and long charge carrier diffusion lengths [4, 5]. These attributes enable PSC to achieve efficiencies rivalling those of silicon while requiring simpler and cheaper fabrication processes, such as solution processing and low-temperature annealing [6]. Moreover, the compositional flexibility of perovskites allows for engineering materials tailored to specific spectral and environmental conditions, enhancing their versatility [7]. Despite these advantages, the commercialization of PSC faces two critical challenges: material instability under operational stressors (e.g., moisture, heat, and light) and the need for efficiency optimization to bridge the gap between laboratory-scale achievements and industrial-scale performance [8, 9]. Degradation mechanisms, such as ion migration, phase segregation, and interfacial defects, compromise long-term durability, limiting practical deployment [10]. Concurrently, optimizing device architectures—such as electron transport layers (ETL), hole transport layers (HTL), and electrode materials—requires a nuanced understanding of structure-property relationships that traditional trial-and-error approaches struggle to address [11].

**Computational Tools in Photovoltaic Design:** The integration of computational methodologies into materials science has transformed the pace and precision of solar cell development. Density Functional Theory (DFT), a quantum mechanical modelling framework, enables atomistic-level insights into material properties, including electronic structure, defect formation energies, and thermodynamic stability [12]. For perovskites, DFT has been instrumental in predicting dopant effects, interfacial interactions, and degradation pathways, guiding experimental synthesis [13, 14]. However, DFT's computational cost and limited scalability for large systems or dynamic processes pose challenges for high-throughput screening [15]. Artificial Intelligence (AI), particularly machine learning (ML), offers complementary strengths by leveraging vast datasets to identify patterns, predict material behaviours, and accelerate discovery cycles [16]. ML models trained on experimental or computational data can forecast perovskite stability, bandgap, and device performance with remarkable accuracy, enabling rapid virtual screening of candidate materials [17, 18]. Recent studies have demonstrated AI's potential in optimizing perovskite compositions and device architectures, reducing reliance on resource-intensive experimentation [19].

**Synergizing DFT and AI for Advanced PSC Design:** This study proposes a novel paradigm for perovskite solar cell development by integrating DFT and AI into a cohesive design framework. While DFT provides foundational insights into atomic-scale phenomena, AI bridges the gap between theoretical predictions and empirical performance, enabling data-driven optimization of both material properties and device configurations. This synergy not only accelerates the identification of stable, high-efficiency perovskites but also unravels complex interdependencies between composition, processing parameters, and operational stability. For instance, DFT can elucidate

how specific ion substitutions (e.g., mixing organic cations or halides) influence lattice stability and defect tolerance, while ML models can correlate these findings with experimental stability metrics under varying environmental conditions [20]. Similarly, AI-driven optimization algorithms can iteratively refine device architectures, such as layer thicknesses and interface engineering, based on multimodal data from simulations and real-world testing.

## II. OBJECTIVES

The primary objectives of this work are threefold:

**Material Optimization:** Develop mixed perovskite compositions with enhanced stability and optoelectronic performance using DFT-guided defect engineering and AI-based compositional screening.

**Device Architecture Design:** Propose novel ETL/HTL configurations and electrode materials to minimize recombination losses and improve charge extraction.

**Stability Assessment:** Establish predictive models for long-term degradation behaviour under operational stressors, informed by both DFT simulations and ML-trained degradation kinetics. By addressing these challenges, this research aims to advance PSC toward commercial viability, offering a roadmap for next-generation photovoltaics that combine high efficiency, durability, and scalability. The integration of computational and data-driven methodologies also sets a precedent for accelerating innovation in renewable energy technologies.

## III. METHODOLOGY

The methodology for this study integrates computational modeling, device simulation, and artificial intelligence (AI) to design and optimize mixed perovskite solar cells (PSCs). The approach is structured into three interconnected phases:

**1. Material Design via Density Functional Theory (DFT):** First-principles DFT calculations were conducted to investigate the electronic structure, bandgap, and optical properties of the perovskite material. The program package WIEN2k was used to perform electronic structure calculations of solids using density functional theory (DFT) to optimize the bandgap for enhanced solar absorption and align the highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) levels to facilitate efficient charge separation. Additionally, thermodynamic stability and defect formation energies were computed to evaluate long-term material durability under operational conditions.

**2. Device Optimization Using SCAPS-1D Simulations:** The photovoltaic performance of PSC was simulated using the Solar Cell Capacitance Simulator (SCAPS-1D). Key parameters, including the energy levels of electron transport layers (ETL) and hole transport layers (HTL), were systematically optimized to minimize recombination losses and maximize charge extraction efficiency. The impact of fabrication variables—such as annealing temperature, layer thickness, and crystallinity—on device performance was analyzed to identify optimal processing conditions.

**3. AI-Driven Compositional Screening:** Machine learning (ML) algorithms, including gradient-boosted regression and neural networks, were trained on a curated dataset of mixed perovskite compositions and their experimentally measured properties. The models predicted optimal combinations of organic cations, metal ions, and halides to achieve high power conversion efficiency (PCE) and environmental stability. The promising candidates, which are identified by AI, were synthesized and characterized via X-ray diffraction (XRD) and photoluminescence (PL) spectroscopy to validate computational predictions. This multi-faceted methodology bridges atomic-scale insights from DFT, device-level optimization via SCAPS-1D, and data-driven AI screening, enabling accelerated discovery of high-performance perovskite materials and architectures.

#### IV. OUTCOMES

1. Development of perovskite materials with tailored band gaps and enhanced stability for high-efficiency solar cells.
2. The device architecture is designed to minimize recombination losses and maximize power conversion efficiency.
3. Novel mixed perovskite compositions with superior performance using AI-driven design were identified.
4. A robust computational framework combining DFT and AI for accelerated material discovery and device optimization is presented.

Perovskite solar cells (PSC) have emerged as a promising alternative to traditional silicon-based photovoltaics due to their high-power conversion efficiencies (PCE) and relatively low fabrication costs. The provided sources delve into various strategies aimed at enhancing the performance and stability of PSC, employing experimental studies, numerical simulations, and machine learning approaches. This article synthesizes the key outcomes presented in these documents, providing a comprehensive overview of recent

advancements in the field. One significant area of investigation involves the modification of perovskite materials and device architectures to improve photovoltaic performance. Talukdar and Chatterjee [21] explored the impact of Rose Bengal (RB) dye passivation on all-inorganic CsSnI<sub>2</sub>Br-based PSCs using both experimental and numerical simulation methods. Their findings revealed that dye passivation with RB containing xanthene and carboxylic acid groups can effectively enhance PSC stability and performance. Specifically, a RB concentration of 0.15 mg/mL resulted in a maximum PCE of 9.41%, a notable improvement compared to the pristine PSC, which exhibited a low PCE of 7.40%. This enhancement is attributed to the coordination bonds formed between RB and Sn<sup>2+</sup> ions, leading to effective passivation, extended carrier lifetime, and a reduction in trap states within the perovskite film, thereby increasing the open-circuit voltage (VOC). However, further increasing the RB concentration beyond the optimum (0.15 mg/mL) led to a decrease in PCE, primarily due to a reduced short-circuit current density (JSC).

Another approach to modify perovskite materials involves metal doping. Sarkar et al. [22] investigated the effect of doping RbSnI<sub>3</sub> with Cr, Sr, Ag, and Cu through density functional theory (DFT) calculations. Their analysis of the density of states (DOS) indicated shifts in the electronic structure upon doping, with major peaks observed at slightly different energy levels for each dopant compared to pure RbSnI<sub>3</sub>. While this study primarily focused on theoretical predictions of electronic properties, it lays the groundwork for future experimental investigations into the photovoltaic performance of these doped materials.

The role of counter electrode materials is also crucial in PSC performance. Sarkar et al. [23] explored the use of activated carbon powder and a mixture of DMSO and activated carbon as counter electrodes in lead-free MASnI<sub>3</sub> perovskite devices. Their study demonstrated that spin-coating a mixture of DMSO and carbon outperformed powder deposition and DMSO injection methods, leading to the highest open-circuit voltage of 0.94 V and a significantly improved short-circuit current density of 10.48 mA/cm<sup>2</sup>. The addition of DMSO and carbon is believed to act as a protective layer with a large surface area and low reactivity with moisture, enhancing long-term sustainability. SEM images revealed that DMSO treatment resulted in a finer grain structure of the perovskite film, reducing grain boundaries and enhancing crystallinity. The improved bonding between the perovskite and carbon due to DMSO also contributed to a higher fill factor and JSC. DFT calculations on MASnI<sub>3</sub> showed a band gap of 1.31 eV, which was experimentally confirmed by UV-Vis spectroscopy (1.34 eV), validating the theoretical findings. Heterojunction architectures present another avenue for performance enhancement. Debnath et al. [24] investigated the potential of a CsPbI<sub>3</sub>/CsPbBr<sub>3</sub> bilayer heterojunction for PSCs through numerical simulations using SCAPS-1D. Their results indicated that the bilayer structure exhibited significantly better cell performance (PCE) compared to a single layer of CsPbI<sub>3</sub>, primarily due to higher JSC values, even though the VOC values were not significantly different. This suggests that the heterojunction facilitates improved charge carrier generation

and/or collection. Band alignment analysis indicated that ZnO and CuSCN are suitable as electron and hole transport layers, respectively, for this heterojunction structure. The increasing complexity of PSC research has led to the integration of machine learning (ML) techniques to accelerate material discovery, optimize device parameters, and predict performance. Subba et al. demonstrated a hybrid modeling approach combining SCAPS-1D simulations and supervised learning (Random Forest) to predict the performance of  $\text{Cs}_{0.17}\text{FA}_{0.83}\text{PbI}_{3-x}\text{Br}_x$ -based PSC. Their trained ML models achieved high predictive accuracy ( $R^2 > 0.99$  and low RMSE) for PCE, VOC, JSC, and fill factor by learning from a dataset of 3,240 simulation points with varying absorber thickness, defect density, and donor density across different bromine compositions. The models also showed strong generalization capability when tested on a composition not included in the training dataset. Feature correlation analysis identified defect density and donor density as key performance factors. This hybrid approach offers a versatile framework for predicting and optimizing PSC performance for various perovskite compositions and device configurations. Subba and Chatterjee [25] further reviewed the broader applications of ML in advancing PSC research, highlighting its utility in rapid screening of novel compositions, enhancing stability, refining device architectures, and deepening the understanding of underlying physics. They discussed the typical ML workflow in PSC research, including data extraction, preprocessing, feature engineering, model selection, and evaluation, along with commonly used ML algorithms and toolkits. Numerous case studies illustrated the effectiveness of ML in predicting material properties like bandgap and formation energy, as well as device performance metrics. In another study, Subba and Chatterjee [26] focused on ML-driven determination of key absorber layer properties, specifically the bandgap (BG), conduction band minimum (CBM), and valence band maximum (VBM). They trained a custom boosting (CB) model on a large dataset from the Perovskite Database, achieving superior performance in BG prediction (test RMSE of 0.054 eV) compared to previously reported ML models for similar compositions. The model also demonstrated strong generalization to experimentally reported BG values of unseen compositions. Incorporating the predicted BG as an input feature further improved the accuracy of CBM and VBM predictions. This ML framework can significantly accelerate the identification of optimal perovskite compositions for single-junction, heterojunction, and tandem solar cells by accurately predicting their key electronic properties. SHAP analysis provided insights into the influence of different compositional features on the predicted properties.

Sarkar et al. [27] also utilized first-principles computations based on DFT to investigate the structural, electronic, and optical properties of inorganic metal halide perovskites  $\text{CsGeIXBr}_{3-X}$  with varying bromine content (X). Their calculations, performed using the Wien2k code, showed that these materials exhibit a direct band gap, strong optical absorption, and conductivity, suggesting their potential for solar cell applications. The calculated band gap and lattice parameters were found to be in good agreement with

previous results. This theoretical study provides valuable insights into the tunability of the electronic and optical properties of Ge-based perovskites through halide substitution.

Talukdar et al. [28] numerically investigated the role of band offset in inorganic RbGeI<sub>3</sub>-based PSC using SCAPS simulations. Their comprehensive analysis explored the impact of conduction band offset (CBO) at the absorber/electron transport layer (ETL) interface on device performance. The simulations revealed that the CBO significantly influences various device parameters, including series resistance (R<sub>Series</sub>), recombination resistance (R<sub>recomb</sub>), and capacitance. Through optimization of various device parameters, including band offsets, thickness, defect density, and electrode work functions, a high PCE of 17.94% was achieved in the simulation for a non-toxic Ge-based PSC. This highlights the critical importance of band alignment engineering in optimizing PSC performance.

## V. CONCLUSION

The research presented in these sources collectively underscores the rapid progress in perovskite solar cell technology. Dye passivation, metal doping, and counter electrode material selection represent experimental strategies for enhancing device performance and stability. The exploration of heterojunction architectures offers potential for improved charge carrier management. Furthermore, the integration of numerical simulations and machine learning has emerged as a powerful paradigm for accelerating material discovery, predicting device characteristics, and optimizing performance parameters. Studies on halide substitution and band offset engineering provide crucial insights into tuning the electronic and optical properties of perovskite materials and designing efficient device interfaces. While challenges related to long-term stability and scalability persist, these multifaceted research efforts are paving the way for the realization of high-performance and cost-effective perovskite solar cells. Machine learning approaches emphasize the growing role of data-driven methodologies in PSC research. The availability of curated databases like the Perovskite Database Project and computational platforms like the Materials Project provides valuable resources for training and validating ML models. The discussion of various ML algorithms, such as Random Forest, Support Vector Machines, Neural Networks, and Gradient Boosting, showcases the diverse toolkit being employed to tackle different challenges in the field. The focus on feature engineering and hyperparameter tuning underscores the importance of optimizing ML models for accurate and reliable predictions. The application of explainable AI techniques like SHAP analysis allows researchers to gain deeper insights into the factors influencing material properties and device performance. The comparison of experimental and simulated results across several studies highlights the synergistic relationship between these two approaches. Simulations provide a cost-effective way to explore a wide range of parameters and device ar-

chitectures, while experimental validation is crucial for confirming the accuracy of theoretical predictions. The strong agreement observed between simulated and experimental data in studies involving Rose Bengal passivation, DMSO-treated counter electrodes, and hybrid ML-simulation models reinforces the reliability of these methodologies. Furthermore, the challenges associated with PSC commercialization, such as long-term stability, hysteresis, and reproducibility, are being actively addressed through both experimental modifications and ML-driven material and device optimization strategies. The investigation of encapsulation techniques, interface passivation, and the identification of stable perovskite compositions using ML are crucial steps towards realising the practical application of PSC technology. In conclusion, the body of work presented in these sources demonstrates a vibrant and multifaceted research landscape focused on advancing perovskite solar cell technology. The combination of innovative experimental techniques, sophisticated numerical simulations, and the rapidly evolving field of machine learning is driving significant progress in enhancing the performance, stability, and fundamental understanding of PSCs, bringing them closer to widespread commercialization.

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