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REVIEW ARTICLE

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Advancing perovskite photovoltaic technology through machine learning-driven automation

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Abstract

Since its emergence in 2009, perovskite photovoltaic technology has achieved remarkable progress, with efficiencies soaring from 3.8% to over 26%. Despite these advancements, challenges such as long-term material and device stability remain. Addressing these challenges requires reproducible, user-independent laboratory processes and intelligent experimental preselection. Traditional trial-and-error methods and manual analysis are inefficient and urgently need advanced strategies. Automated acceleration platforms have transformed this field by improving efficiency, minimizing errors, and ensuring consistency. This review summarizes recent developments in machine learning-driven automation for perovskite photovoltaics, with a focus on its application in new transport material discovery, composition screening, and device preparation optimization. Furthermore, the review introduces the concept of the self-driven Autonomous Material and Device Acceleration Platforms (AMADAP) laboratory and discusses potential challenges it may face. This approach streamlines the entire process, from material discovery to device performance improvement, ultimately accelerating the development of emerging photovoltaic technologies.

KEYWORDS

automation, device acceleration platforms, machine learning, materials acceleration platforms, perovskite solar cells, self-driving laboratory

Jiyun Zhang and Jianchang Wu contributed equally to this study.

1 | INTRODUCTION

Perovskite photovoltaics have experienced remarkable development since their first application in solar cells in 2009. The power conversion efficiency (PCE) of these materials has surged from 3.8% to over 26%.[1–3] Perovskite materials, typically organic–inorganic metal halides with an ABX_3 structure, consist of organic and inorganic cations at the A-site (e.g., MA^+: methylammonium, FA^+: formamidinium, Cs^+: cesium), metal cations at the B-site (e.g., Pb^{2+}: lead, Sn^{2+}: tin), and halogen anions at the X-site (e.g., I^-: iodide, Br^-: bromide, Cl^-: chloride). This crystal structure imparts several advantageous photovoltaic properties, including a high absorption coefficient, long carrier lifetimes, and tunable band gaps.[4] These attributes enable efficient light absorption, effective carrier transport, and spectral response optimization.[5,6] Additionally, solution-processable fabrication techniques, such as spin-coating, doctor-blading, and spray-coating, provide cost-effective production at low temperatures compared with silicon-based photovoltaics.[7–9] However, perovskite solar cells face critical challenges, such as long-term material and device stability.[10–12] Addressing these issues requires efforts in material screening, interface engineering, device structure design, and process parameter optimization.[13–16] These processes often entail intricate multi-objective optimization challenges within large multidimensional composition and parameter spaces, sometimes involving millions of potential candidates to explore. Traditional trial-and-error approaches and manual analysis, constrained by inefficiency and labor-intensive processes, are inadequate for tackling these challenges.[17,18] Overcoming these bottlenecks requires advanced strategies capable of rapidly processing large experimental datasets and optimizing complex parameter spaces.[19]

Automated acceleration platforms have emerged as transformative tools for addressing high-dimensional optimization challenges across various disciplines, such as chemistry, physics, and drug synthesis.[20–28] The integration of automation into energy materials research, particularly for perovskite solar materials, has garnered increasing attention.[29–32] These platforms enable high-throughput experimentation, processing a large number of samples in parallel while improving efficiency, reducing errors, and ensuring reproducibility.[33–35] Furthermore, the introduction of machine learning (ML) algorithms into materials science has become increasingly pivotal, as evidenced by the increasing number of related publications (Figure 1).[36–44] The growing application of ML enhances the capabilities of these automated platforms.[19,45] ML algorithms can identify complex patterns within experimental data and predict the performance of untested materials and processes.[46–49] These algorithms extract key insights from large-scale datasets to guide experimental design and parameter optimization.[50] The predictive power of ML models shifts the optimization process from reliance on experience and intuition to a robust, data-driven approach.[51–53] The combination of ML and automation offers a powerful strategy for advancing perovskite photovoltaic technology. This integrated approach addresses the limitations of traditional methods by improving efficiency, accuracy, and optimization capabilities.[54–57] By accelerating material discovery and process development, ML-driven automation represents a significant leap forward, enabling the rapid advancement of perovskite photovoltaics and underscoring its transformative potential in renewable energy technologies.[58,59]

This review overviews the advancements in ML-driven automation for perovskite photovoltaics research

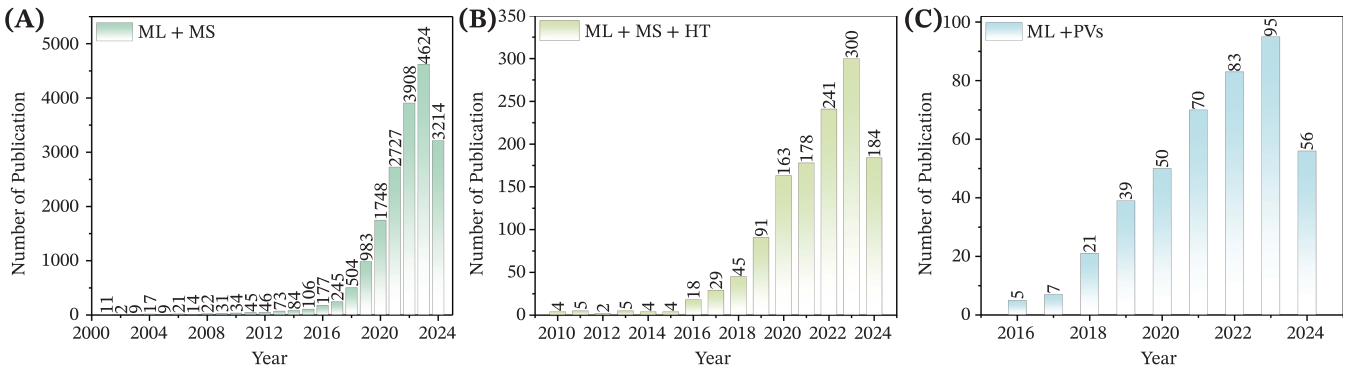

FIGURE 1 The increasing number of research publications (based on a Web of Science search) in the following topics: (A) "Machine Learning (ML) + Materials Science (MS)", (B) "ML + MS + High-throughput (HT)", and (C) "ML + Photovoltaics (PVs)". The data, collected from January 2000 to July 2024, underscores the growing utilization of automated platforms in material science research.

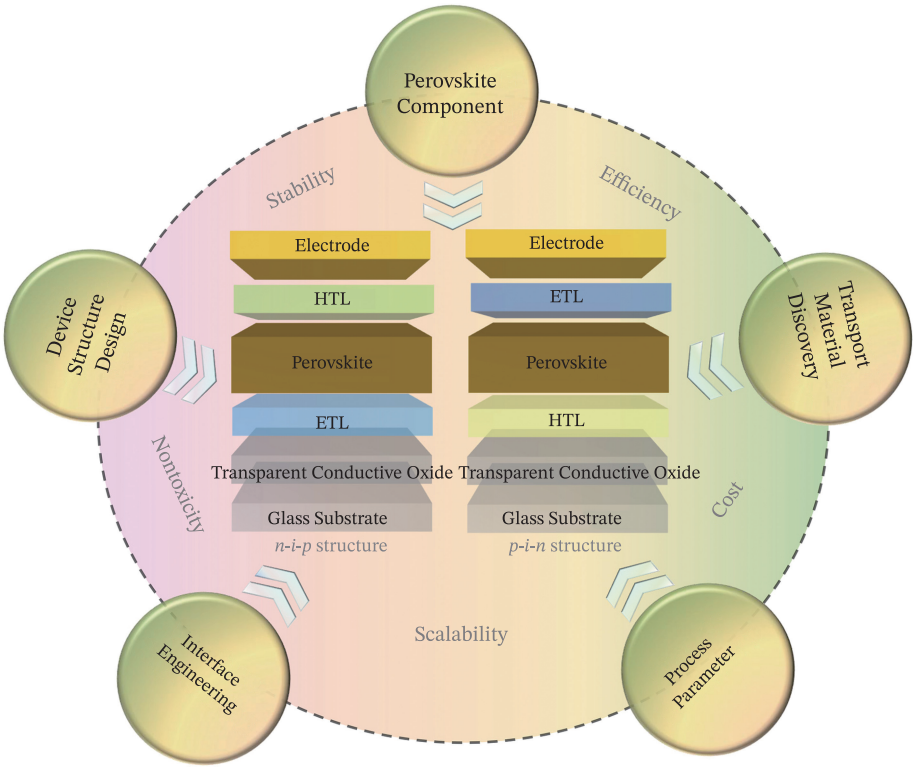

FIGURE 2 Schematic representation of the key factors influencing the performance of perovskite photovoltaic devices, including material selection, interface engineering, device structure design, and preparation process optimization.

and development. It explores autonomous methods for novel transport material discovery, rapid multicomponent screening, performance prediction, and process optimization. The review concludes by introducing the concept of the self-driven Autonomous Material and Device Acceleration Platforms (AMADAP) laboratory, which integrates autonomous material discovery with device optimization. Potential challenges in implementing such platforms are also discussed. By summarizing current achievements and proposing a strategic roadmap, this review provides valuable insights for advancing perovskite photovoltaic technology.

2 | ADVANCING PEROVSKITE PHOTOVOLTAICS THROUGH ML-DRIVEN AUTOMATION

The development of high-performance perovskite photovoltaic devices requires a comprehensive consideration of various aspects such as composition selection, interface engineering, device structure design, and process optimization (Figure 2).[60–63] Each of these factors critically influences key performance metrics, such as PCE and stability.[64] However, optimizing these factors via traditional methods is often challenging due to the vast parameter spaces and interdependent nature of the variables involved.[65] ML-driven automation platforms have transformed these processes by improving efficiency and accuracy. These platforms enable rapid exploration of parameter spaces, uncover complex relationships, and optimize processes systematically. This section discusses how ML-driven automation accelerates transport materials discovery, perovskite component screening, and the optimization of device fabrication processes.

2.1 | New organic transport materials discovery

The rapid development of ML technology is transforming organic synthesis, with applications ranging from reaction optimization to automated synthesis and acceleration of material discovery. ML-driven automation holds great promise for identifying structure–property relationships and discovering new hole-transport materials for perovskite devices.[66–71] This section reviews the role of ML-driven automation in organic synthesis and hole-transport material discovery for perovskite devices from three perspectives: reaction optimization, structure–property relationships, and material discovery.

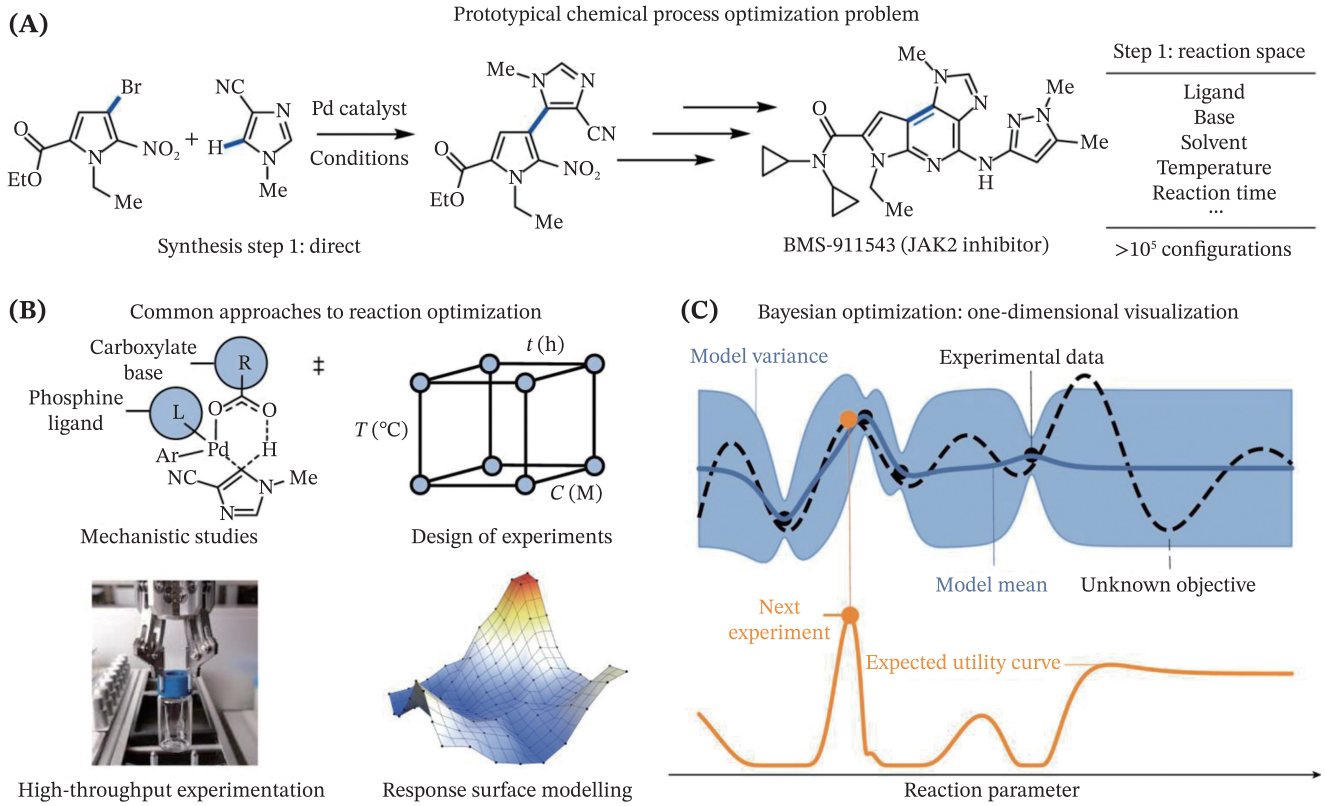

FIGURE 3 Bayesian reaction optimization. (A) Example of chemical process optimization: Synthesis of BMS-911543. (B) Typical reaction optimization approaches, including mechanistic studies, design of experiments, high-throughput experimentation, and response surface modeling. (C) Overview of Bayesian optimization with a Gaussian process surrogate model and an expected utility surface for experiment selection. Reprinted with permission.[73] Copyright 2021, Springer Nature.

2.1.1 | Reaction optimization

Organic semiconductors are commonly synthesized via metal-catalyzed coupling reactions to form conjugated chains, where catalyst selection plays a pivotal role.[72] Traditionally, catalyst design for asymmetric reactions has relied on empirical methods, where researchers qualitatively identify structural patterns to enhance selectivity. Integrating ML algorithms with cheminformatics offers a transformative approach by analyzing large datasets to uncover hidden patterns and optimize reaction conditions efficiently. Doyle et al. demonstrated the potential of Bayesian optimization (BO) in identifying optimal reaction conditions.[73] Using a benchmark dataset derived from a palladium-catalyzed direct arylation reaction, they systematically compared the performance of BO with traditional human decision-making (Figure 3). The BO framework showed its practical utility by being further applied to real-world scenarios, such as Mitsunobu and deoxyfluorination reactions. An innovative online game in this study linked expert decision-making with real experimental outcomes, which provided a novel way to evaluate optimization strategies in reaction development. Schoenebeck et al. addressed reaction optimization challenges by employing an unsupervised learning algorithm. This method successfully predicted and validated effective phosphine ligands using only five experimental data points.[74] The approach demonstrated remarkable predictive capabilities in predicting dinuclear Pd(I) complexes among 348 ligands. These examples illustrate the significant potential of ML-driven frameworks in accelerating reaction optimization for organic semiconductors.

In addition to catalysts, coupling reactions are influenced by factors such as substrates, ligands, bases, and additives. High-throughput experimentation combined with ML has proven effective in evaluating these factors comprehensively.[39] Ahneman et al. applied this approach to a palladium-catalyzed Buchwald–Hartwig cross-coupling between aryl halides and 4-methylaniline. Computational scripts calculated and extracted atomic, molecular, and vibrational descriptors from the reaction components. By using these descriptors as inputs and reaction yield as the output, a random forest algorithm was employed significantly outperforming traditional linear regression analysis. Its advantages included better

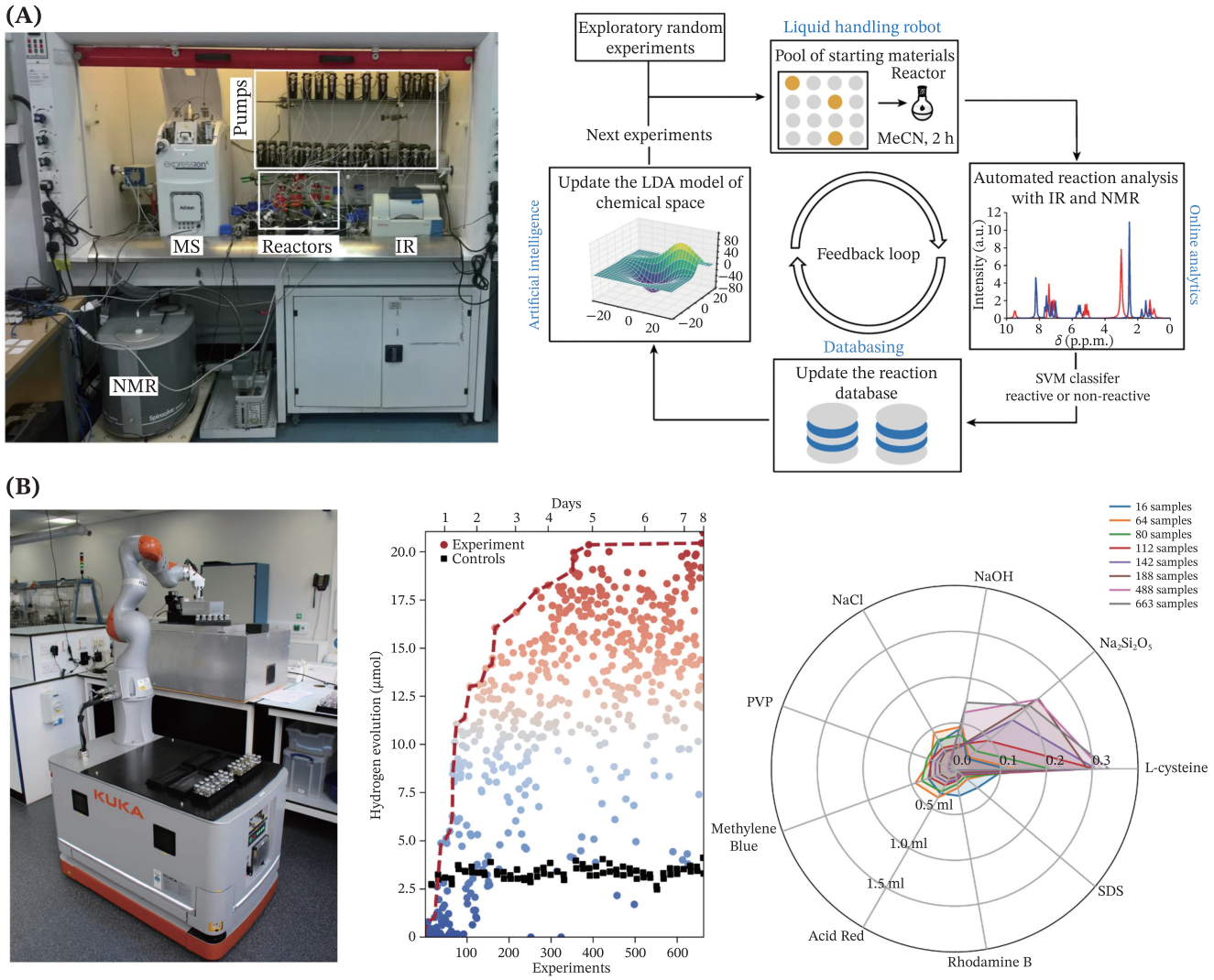

FIGURE 4 (A) Schematic and photograph of the chemical robot and the workflow of ML-guided exploration of chemical space. Reprinted with permission.[38] Copyright 2018, Springer Nature. (B) Photograph of the autonomous mobile robot and the output from the autonomous robotic search. Reprinted with permission.[77] Copyright 2020, Springer Nature.

handling of sparse training datasets and more robust out-of-sample predictions. The results highlight its potential for streamlining synthetic methodology optimization in chemical research. Similarly, Denmark et al. constructed an experimental dataset and utilized neural network models to design systematic experiments. This approach successfully predicted several new conditions for C–N coupling reactions with yields exceeding 85%.[75] Random forest algorithms further improved yield prediction accuracy using high-throughput experimental data. BO, an iterative response surface-based global optimization algorithm, has also shown exceptional capabilities in reaction tuning. Doyle et al. applied the BO-guided framework to optimize various coupling reactions, including C–C and C–N bond formations, significantly enhancing efficiency and consistency.[73] To make these tools accessible, they developed open-source software for chemists to incorporate optimization algorithms into daily workflows, which offers better performance compared with human decision-making.[76] These examples underscore the transformative role of ML in optimizing complex coupling reactions, advancing both experimental efficiency and predictive accuracy.

2.1.2 | Autonomous synthesis

Combining ML algorithms with automated platforms to autonomously design and execute experiments represents a transformative method in accelerating material iteration and reducing manual workloads for chemists. Cronin et al. demonstrated this approach by employing chemical computer-aided design (ChemCAD) to digitize reaction equipment and perform multi-step organic

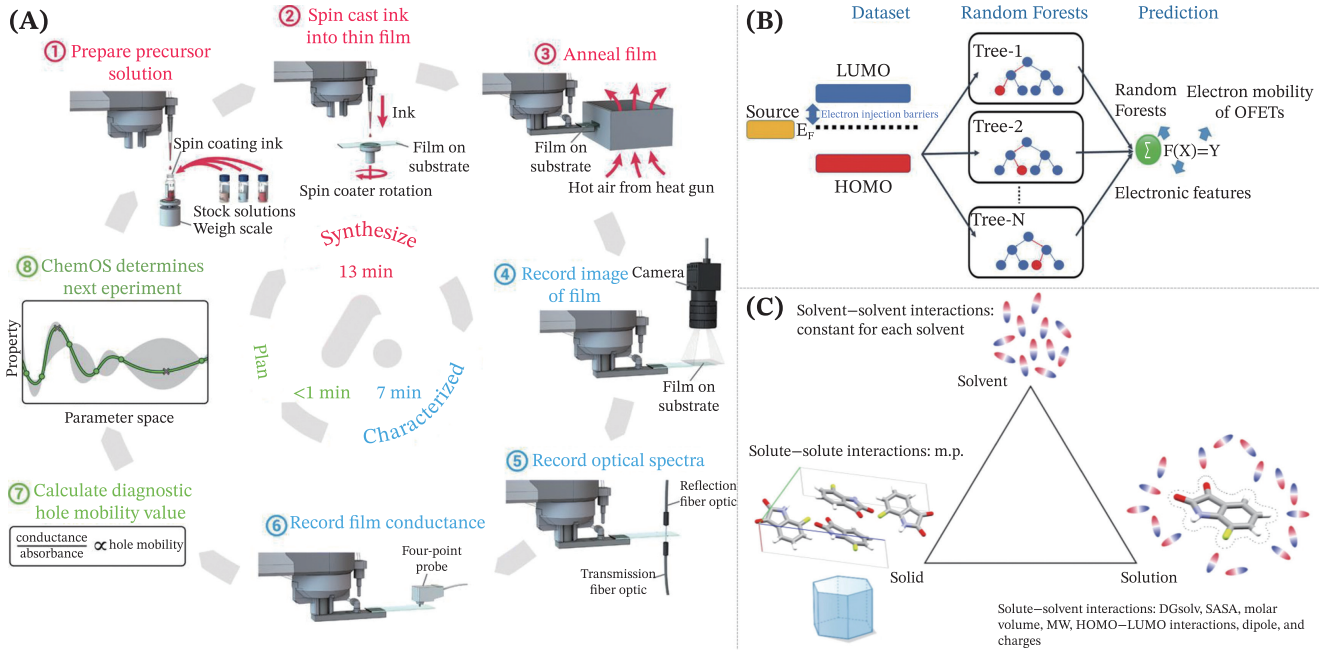

FIGURE 5 The application of ML in structure–property relationships discovery. (A) Ada uses an autonomous optimization workflow. Reprinted with permission.[83] Copyright 2020, AAAS. (B) Schematic of the machine-learning approach toward energy level modulation of *n*-type OFETs. Reprinted with permission.[84] Copyright 2019, John Wiley and Sons. (C) Concepts of solubility prediction and data availability. Physical aspects of the dissolution process of solids and corresponding descriptors. Reprinted with permission.[85] Copyright 2019, Springer Nature.

synthesis.[78] This method translated traditional bench-scale synthesis into platform-independent digital code, which guided the fabrication of 3D-printed devices capable of executing the entire synthetic route. For example, this approach successfully synthesized the γ-aminobutyric acid receptor agonist, (±)-baclofen. In the same year, Cronin's team developed another ML-controlled organic synthesis robot capable of performing chemical reactions and analyses faster than manual methods (Figure 4A).[38] The system encoded chemical inputs in binary and used real-time nuclear magnetic resonance (NMR) and infrared (IR) spectroscopy for assessment. Based on data from only 10% of the dataset, the system predicted the reactivity of approximately 1000 reaction combinations with over 80% accuracy. The model also discovered four new reactions, which were later manually validated by a chemist. Similarly, Bode et al. utilized a high-throughput photocatalysis platform to generate data for predicting molecular properties and reaction success rates using deep learning models.[79] This platform processed 1152 discrete reactions within a short period, significantly enhancing the efficiency of new molecule discovery. Cooper's team developed a mobile robotic chemist capable of autonomously planning synthetic routes and optimizing reaction conditions (Figure 4B).[77] This system iteratively optimized reaction parameters, successfully synthesizing multiple organic compounds with conversion rates far exceeding existing benchmarks. These advancements highlight the transformative potential of integrating ML with automated platforms. By enabling autonomous experimentation, data-driven predictions, and iterative optimization, this approach significantly accelerates material development and opens new avenues for discovering complex synthetic pathways and high-performance materials.

2.1.3 | Structure–property relationships

Understanding structure–property relationships is important for designing new molecules and advancing material development. Human cognition, limited by memory capacity and experiential biases, often provides limited insights into these complex relationships.[80–82] In contrast, ML excels at analyzing multivariate datasets to uncover comprehensive and intricate patterns. Berlinguette et al. developed a self-driving laboratory called the Ada platform to optimize thin-film material compositions and processing parameters for achieving enhanced material mobility (Figure 5A).[83] Min-Hsuan Lee applied ML techniques, such as Gradient Boosting and Random Forest regression, to model charge transport mobility in *n*-type organic field-effect transistors (OFETs). By optimizing HOMO and LUMO energy levels, this approach

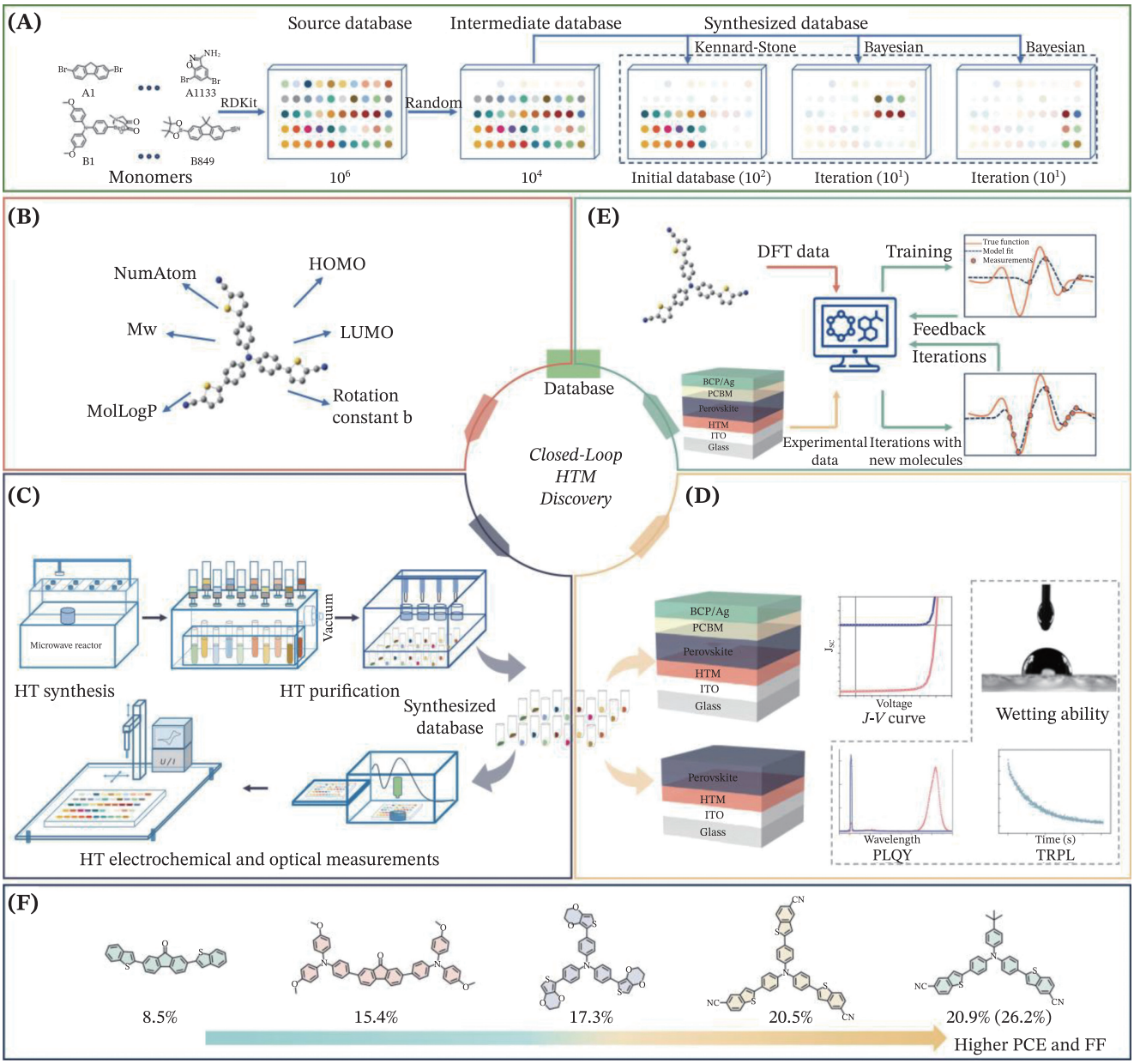

FIGURE 6 Overview of the ML-driven workflow for high-performance hole-transport material (HTM). (A) Databases used in the study include the source database of virtual combinations of commercial monomers via Suzuki coupling, the intermediate database of randomly selected molecules for DFT calculations, and the synthesized database of experimentally synthesized molecules for model training and validation. (B) Molecular descriptors for the intermediate database were obtained through DFT calculations. (C) Synthesized molecules were processed, purified, and characterized using an in-house high-throughput platform. (D) Synthesized molecules were utilized as HTMs in PSCs, and devices were characterized using ITO and BCP/Ag configurations. (E) The model trained on HTM descriptors and device parameters iteratively predicted, synthesized, and measured new molecules, leading to the discovery of the best HTM. (F) Molecular iterations were summarized and analyzed to refine the process. Reprinted with permission.[88] Copyright 2024, AAAS.

deepened the understanding of *n*-type organic materials, informed material and electrode selection, and quantified trade-offs between charge transport mobility and electronic energy levels (Figure 5B). This work advanced the design of next-generation organic circuits.[84] For solution-processed perovskite solar devices, understanding the relationship between solubility and molecular structure is vital. Nguyen et al. introduced a method combining ML algorithms—ANN, SVM, RF, ExtraTrees, Bagging, and GP—with computational chemistry to predict solubility in organic solvents and water (Figure 5C).[85] Their models effectively captured the physicochemical relationship between solubility and molecular properties across different solvents. This approach not only reproduced

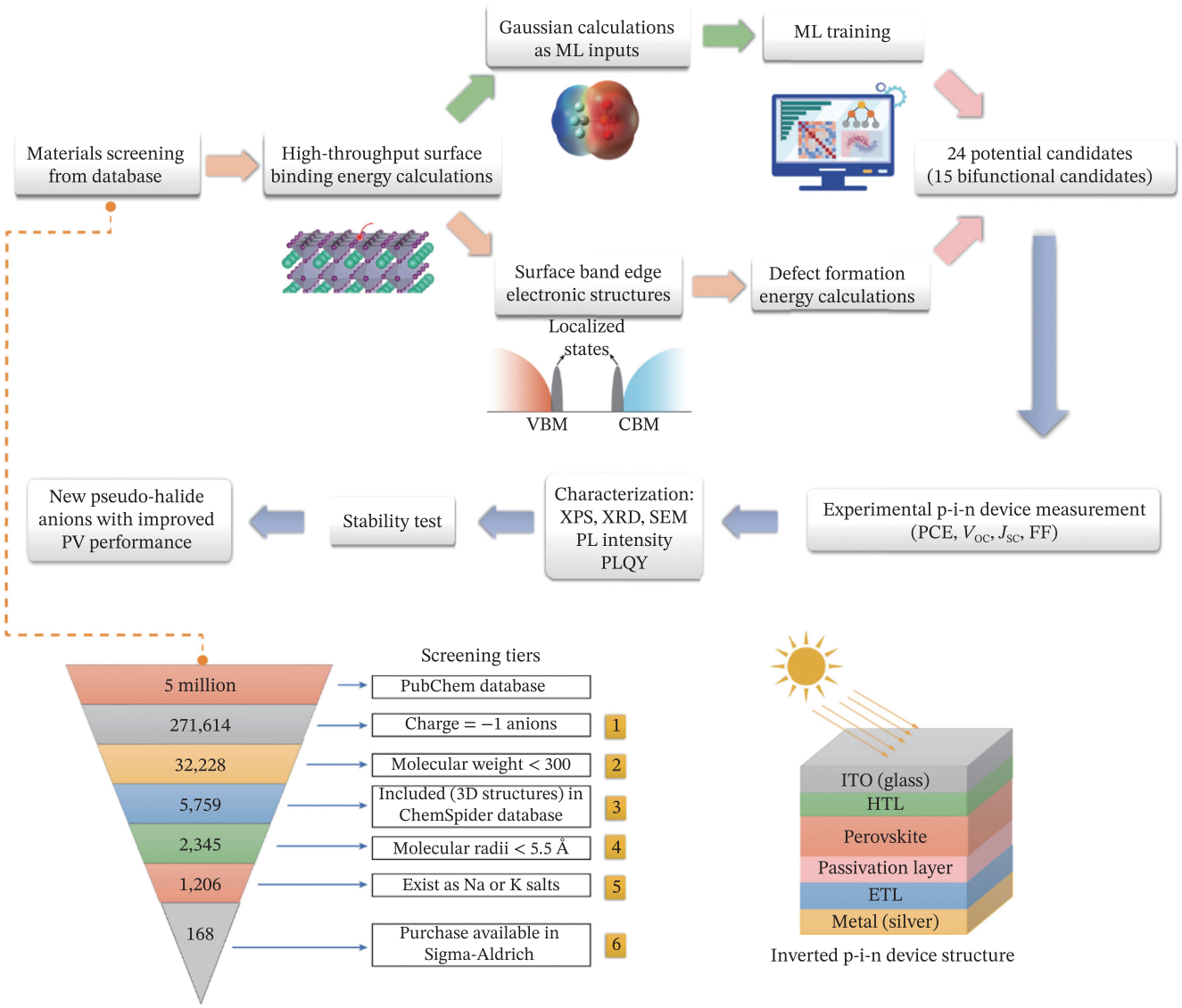

FIGURE 7 ML-guided workflow used to identify candidate PH anions as passivants to improve PSCs performance. Reprinted with permission.[89] Copyright 2023, Springer Nature.

experimental trends but also proposed rational strategies to improve model accuracy, providing valuable insights for optimizing materials for perovskite solar devices. These examples demonstrate how ML-driven insights into structure–property relationships accelerate material discovery and enable the design of advanced materials for diverse applications.

2.1.4 | Material discovery

Perovskite devices require organic semiconductors with specific properties, including energy level alignment, solubility, interaction with perovskites, defect passivation, and stability.[86] Combining automated synthesis with structure–activity relationship analysis within a single system provides an effective approach to material discovery (Figure 6). Automated synthesis delivers a continuous stream of experimental data to ML algorithms. High-throughput experiments and in situ material characterization ensure robust data consistency and repeatability. The iterative interaction between ML and automated synthesis facilitates the discovery of unexpected and high-performing materials. Recently, our team developed a high-throughput synthesis and purification platform for hole-transporting materials used in perovskite devices.[87,88] This automated platform directly evaluated performance by integrating synthesized materials into devices. ML algorithms modeled the relationship between molecular structure and device performance, iteratively refining the model during the discovery process.

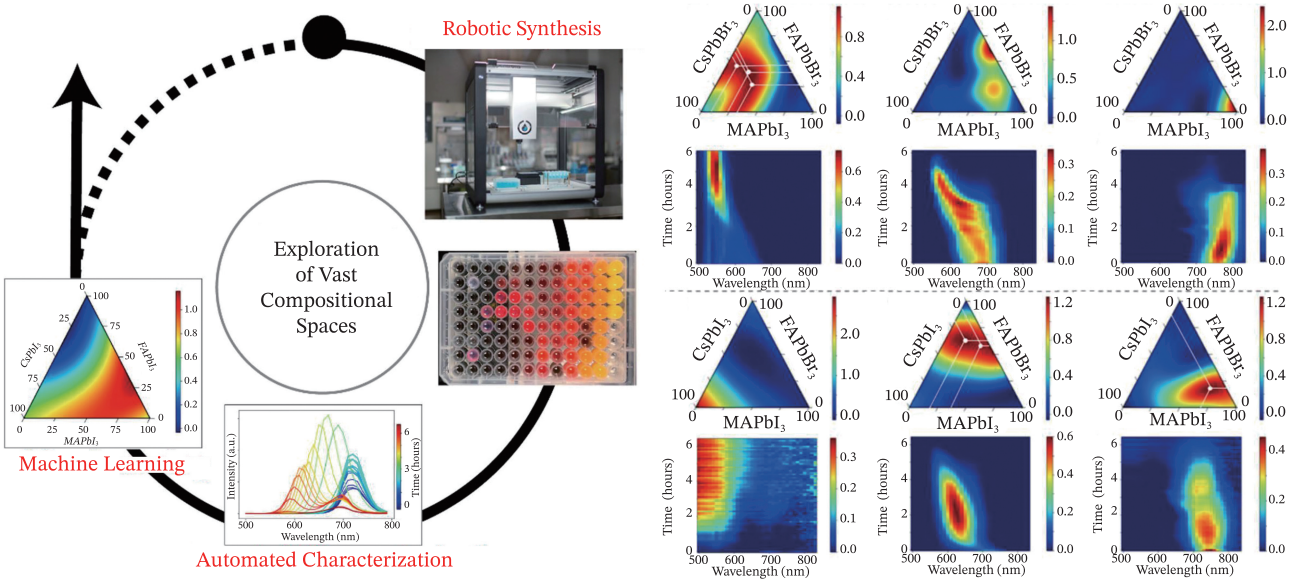

FIGURE 8 Schematic of the automated workflow for combinatorial discovery and multivariate analysis of stability in quasi-ternary metal-halide perovskite systems. Reprinted with permission.[91] Copyright 2020, American Chemical Society.

From a pool of 1 million candidate molecules, a series of high-performance hole-transport materials was successfully identified. The discovered materials achieved top efficiencies of up to 26.2%. This result highlights the transformative potential of combining ML and automated synthesis to accelerate the discovery and development of advanced materials for perovskite photovoltaics. Sargent's group developed a ML workflow to accelerate the discovery of anions by using full-density functional theory calculations to train their ML model (Figure 7).[89] This physics-informed model identified promising molecules with head groups designed to prevent lattice distortion and anti-site defect formation, paired with tail groups optimized for robust surface attachment. Using this approach, they screened 15 potential anions and successfully incorporated them into high-performance perovskite solar cells, achieving a maximum PCE of 24.56%.

2.2 | Perovskite component screening

Perovskite materials encompass a wide variety of chemical compositions and structural forms, with each combination offering unique performance characteristics.[90] Identifying the optimal material configuration for specific applications requires navigating a vast parameter space of chemical elements and their combinations. Traditional experimental methods, however, often struggle to efficiently identify optimal combinations in a short timeframe. In this context, ML-driven automation platforms have emerged as powerful tools to address these challenges. These intelligent systems can efficiently prepare hundreds or even thousands of samples and analyze extensive experimental datasets within a short period. By uncovering patterns, discovering new material combinations, and predicting their properties, ML-driven platforms significantly accelerate the pace of innovation and discovery.

2.2.1 | Metal–halide perovskite microcrystals

Perovskite materials are pivotal in determining the stability of photovoltaic devices, making careful screening for stable compositions important to ensure device longevity and reliability.[5,92] The anti-solvent method is widely used for producing high-quality perovskites.[93] Leveraging high-throughput automation enables rapid, efficient, and precise material screening. Higgins et al. developed a framework that integrated chemical robotics and ML to evaluate the stability of multicomponent halide perovskites across a multidimensional combinatorial space (Figure 8).[91,94] This approach involved an integrated automated experimental workflow, including a pipetting robot programmed to synthesize diverse perovskite combinations in a 96-well plate, a multi-mode microplate reader for automated photoluminescence (PL) spectra measurements, and data analysis employing non-negative matrix factorization (NMF) and Gaussian process regression (GPR). Their study revealed that Br-

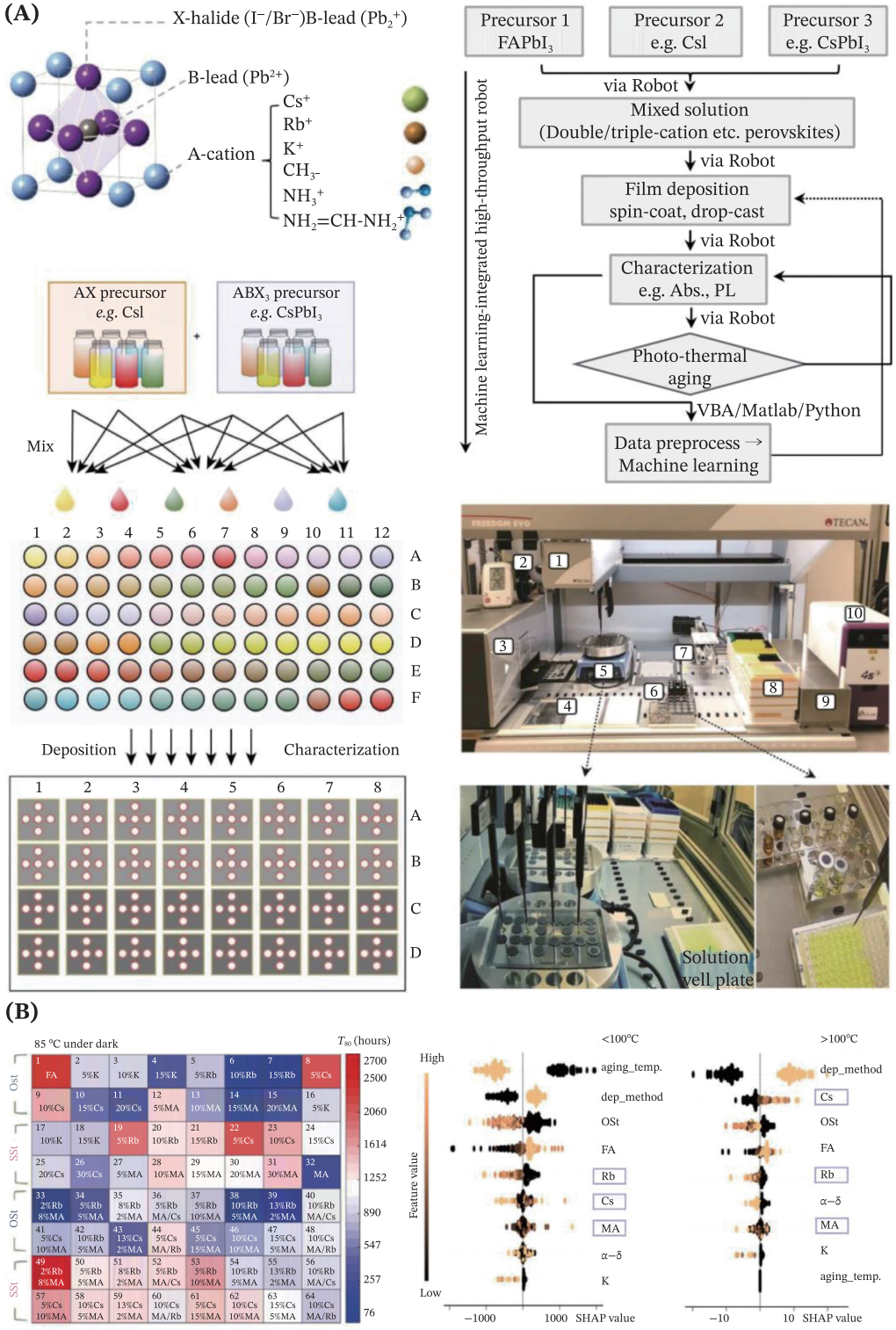

FIGURE 9 (A) Schematic of a high-throughput workflow used for automatic synthesis and characterization of multiple-cation perovskites. (B) GBT regression and SHAP-based feature importance analysis for T_{80} lifetime of perovskites. Reprinted under the terms of the CC-BY 4.0 license.[97] Copyright 2021, Springer Nature.

rich mixed halide systems exhibited excellent stability, whereas I-rich systems showed lower stability. Notably, materials within the $CsPbBr_3$ and $FAPbBr_3$-rich domains demonstrated enhanced stability under ambient conditions. This study illustrates the powerful application of ML-guided automation in rapidly screening and characterizing perovskite materials for developing stable perovskite photovoltaic technologies.

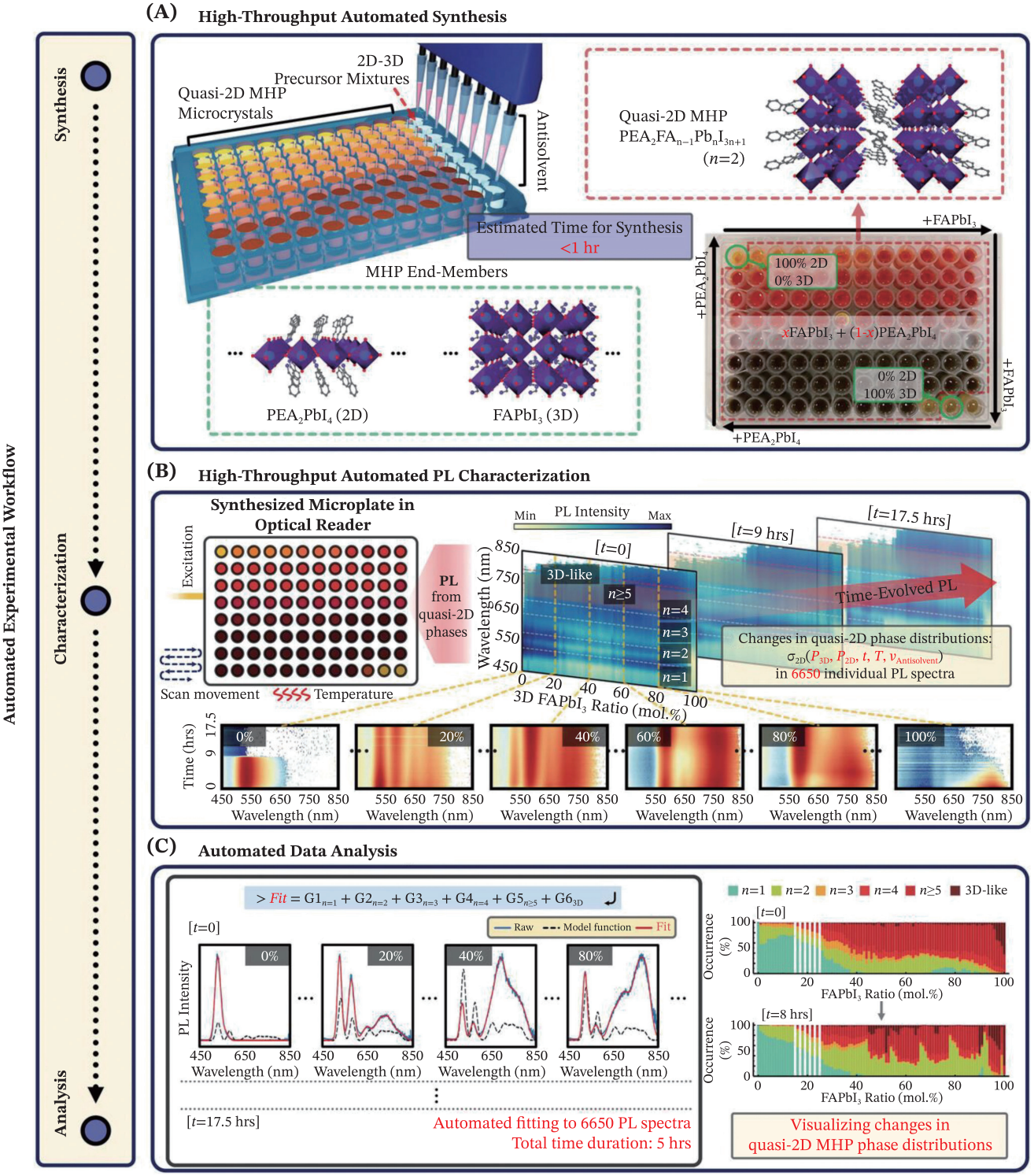

FIGURE 10 Automated synthesis-characterization-analysis workflow for quasi-2D perovskite phase growth. (A) illustration of high-throughput automated synthesis. (B) High-throughput PL characterizations. (C) Automated data analysis of the PL dataset using a custom peak-fitting in a Python interface to evaluate each sample. Reprinted with permission.[103] Copyright 2023, John Wiley and Sons.

2.2.2 | Metal–halide perovskite thin films

Perovskite composition is crucial for device performance and stability. It is highly necessary to systematically study and understand how different compositions influence these metrics. Traditional manual methods are inefficient, time-consuming, and difficult to unravel complex component–property relationships. Automated platforms

offer a solution by synthesizing and characterizing large numbers of samples to generate FAIR datasets, while machine learning can mine these data to extract key features and determine optimal solutions. Our group developed a robotic platform for high-throughput precursor preparation, thin-film deposition, and stability testing under various aging conditions.[95,96] The automated system synthesized and tested over 1000 perovskite samples within a few days, which enabled the generation of large datasets (Figure 9A).[97] Gradient boosting decision tree (GBT) regression models, interpreted through SHapley Additive exPlanations (SHAP), revealed critical stability determinants such as cation composition, stoichiometry, and deposition methods (Figure 9B). One key finding was the temperature-dependent stability reversal phenomenon, where organic cations, like MA, enhanced stability below 100°C but caused degradation at higher temperatures, whereas inorganic cations, like Cs, exhibited the opposite trend. These findings highlight the intricate interplay of thermodynamic and kinetic factors in perovskite degradation, which aligns with the theoretical study on phase stability and miscibility in mixed-cation perovskites.[98] This approach provides insightful guidelines for compositional design engineering toward durable perovskite solar materials.

2.2.3 | Quasi-2D metal halide perovskites

Quasi-2D metal halide perovskites exhibit promising optoelectronic properties and enhanced environmental stability, which makes them suitable for optoelectronic applications.[99–101] However, the multiphase mixing phenomenon during fabrication remains a challenge.[102] It is crucial to investigate their phase growth behavior to understand the initial phase distribution and the principles governing it. Yang et al. utilized ML-guided high-throughput automated synthesis and characterization platform to study the phase formation and growth dynamics of FA-based quasi-2D perovskites (Figure 10).[103] A micropipette robot synthesized 95 quasi-2D perovskite samples with varying 2D:3D component ratios. High-throughput PL characterization and automated data analysis via Python tracked phase distribution changes over time. The analysis revealed that the $n = 2$ phase is the most stable, for it formed preferentially across all 3D perovskite ratios and remained dominant even when 3D $FAPbI_3$ content reached 95%. Higher n-phases and 3D-like phases appeared as the 3D ratio increased, with thermal annealing further enhancing their formation but also increasing the likelihood of unwanted PbI_2 byproducts. Their findings emphasize the need for precise compositional optimization toward desired quasi-2D perovskite materials. The research demonstrated the effectiveness of high-throughput experimentation combined with machine learning algorithms in accelerating the discovery and optimization of functional energy materials.

2.2.4 | Perovskite-like materials

Despite achieving remarkable conversion efficiencies, current perovskite-based photovoltaic devices benefit from exploring perovskite-like materials as complementary options.[104,105] Using a combination of high-throughput synthesis and ML diagnostics, Sun et al. synthesized and characterized 75 perovskite-inspired compositions within 2 months. Among these, 87% exhibited band gaps between 1.2 and 2.4 eV.[106] A deep neural network classified the synthesized compounds into 0D, 2D, and 3D structures based on experimental X-ray diffraction (XRD) data; the accuracy rate was 90%. This approach proved 10 times faster than traditional manual analysis and successfully clarified the nonlinear band gap behavior and dimensional transitions in the $Cs_3(Bi_{1-x}Sb_x)_2(I_{1-x}Br_x)_9$ a lead-free alloy series. By integrating accelerated experimental cycles with ML diagnostics, this study demonstrated significant progress in the discovery and development of functional energy materials.

2.3 | Device preparation process optimization

Optimizing process parameters is critical for achieving high-performance perovskite solar cells. The complex interdependencies among these parameters form a multidimensional parameter space, where varying even a few parameters across multiple values results in thousands or millions of parameter combinations. Traditional single-variable methods cannot address such high-dimensional optimization challenges effectively.[33] While high-throughput platforms enable accelerated experiments and data collection, their efficiency is limited by resource constraints and experimental timelines. Integrating ML models into high-throughput platforms offers an efficient and intelligent solution to explore these complex parameter spaces.

2.3.1 | Metal-halide perovskite devices

Simultaneously optimizing the processing parameters of solution-processed perovskite solar cells remains a

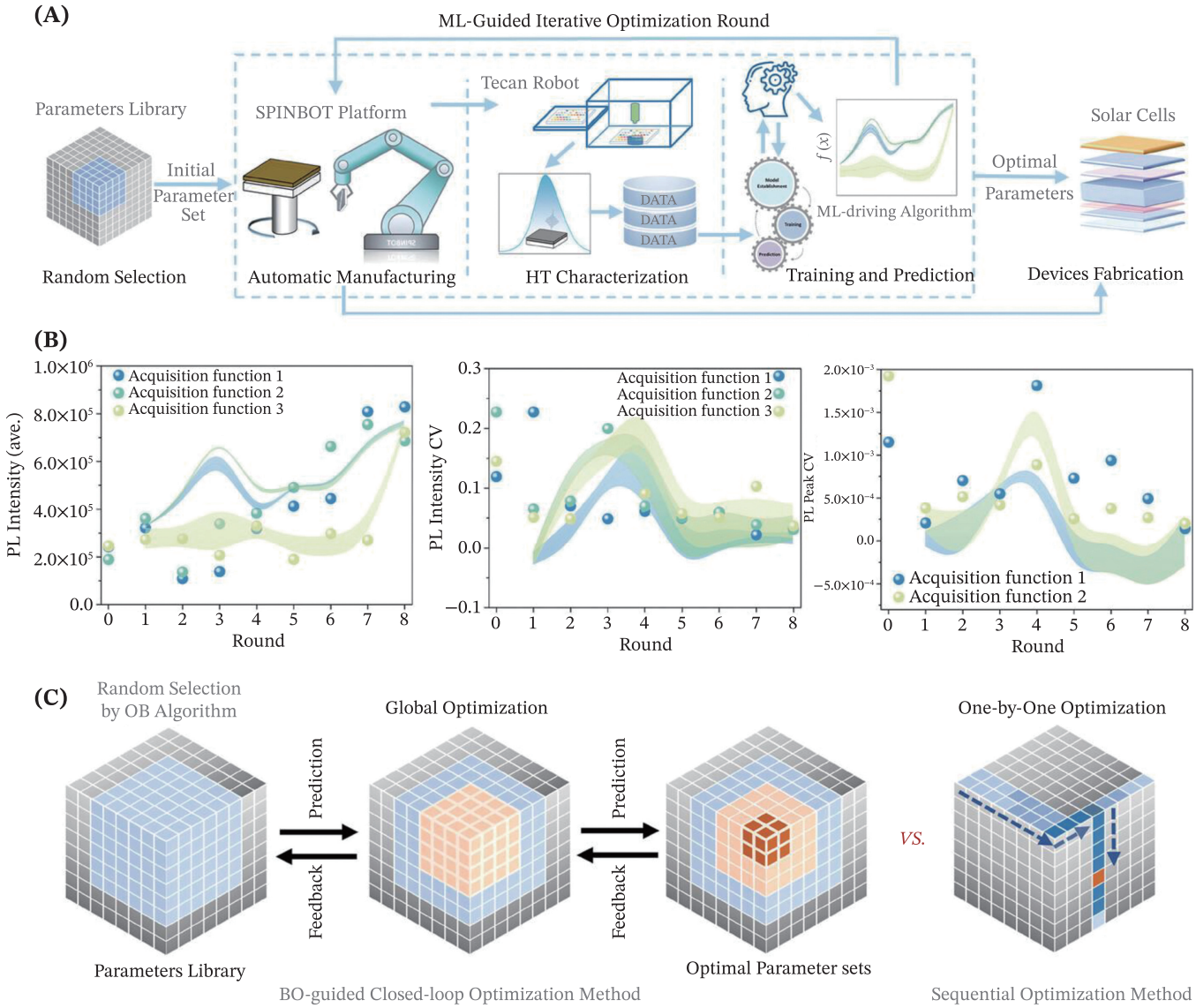

FIGURE 11 ML-guided closed-loop optimization for high-performance perovskite solar devices. (A) Schematic of the BO-guided experimentation workflow. (B) Evolution of experimental PL peak coefficient of variation (CV), PL intensity, and PL intensity CV values across iterative rounds. (C) Schematic illustrating the comparison of the ML-guided globalized optimization method and the step-by-step method. Reprinted under the terms of the CC-BY license.[56] Copyright 2023, the Authors.

significant challenge.[107] Our group developed a fully automated platform, SPINBOT, to optimize solution-processed semiconductor materials, particularly perovskites.[108] The SPINBOT system integrated a BO algorithm to achieve closed-loop optimization of perovskite devices in ambient conditions.[56,109] The flowchart in Figure 11A illustrates the process: the algorithm initially selects random parameter combinations for experimentation, evaluates film quality and uniformity through PL spectroscopy, and recommends predicted parameter sets for the next rounds based on acquired data. This iterative cycle systematically explored the parameter space; the film quality and device performance improved gradually. The optimization process considered both conventional process parameters and those challenging to control manually, such as tip height and solvent dripping speed. This holistic approach optimized perovskite processing parameters with only simple PL characterization (Figure 11B). The optimized films achieved a maximum PCE of 21.6% immediately after fabrication in ambient air. The unsealed devices maintained 90% of their initial efficiency over 1100 h under 60°C–65°C photothermal aging. This study proved the capability of the ML-guided autonomous platform in effectively exploring multidimensional parameter spaces for accelerated discovery of performance-optimized perovskite solar

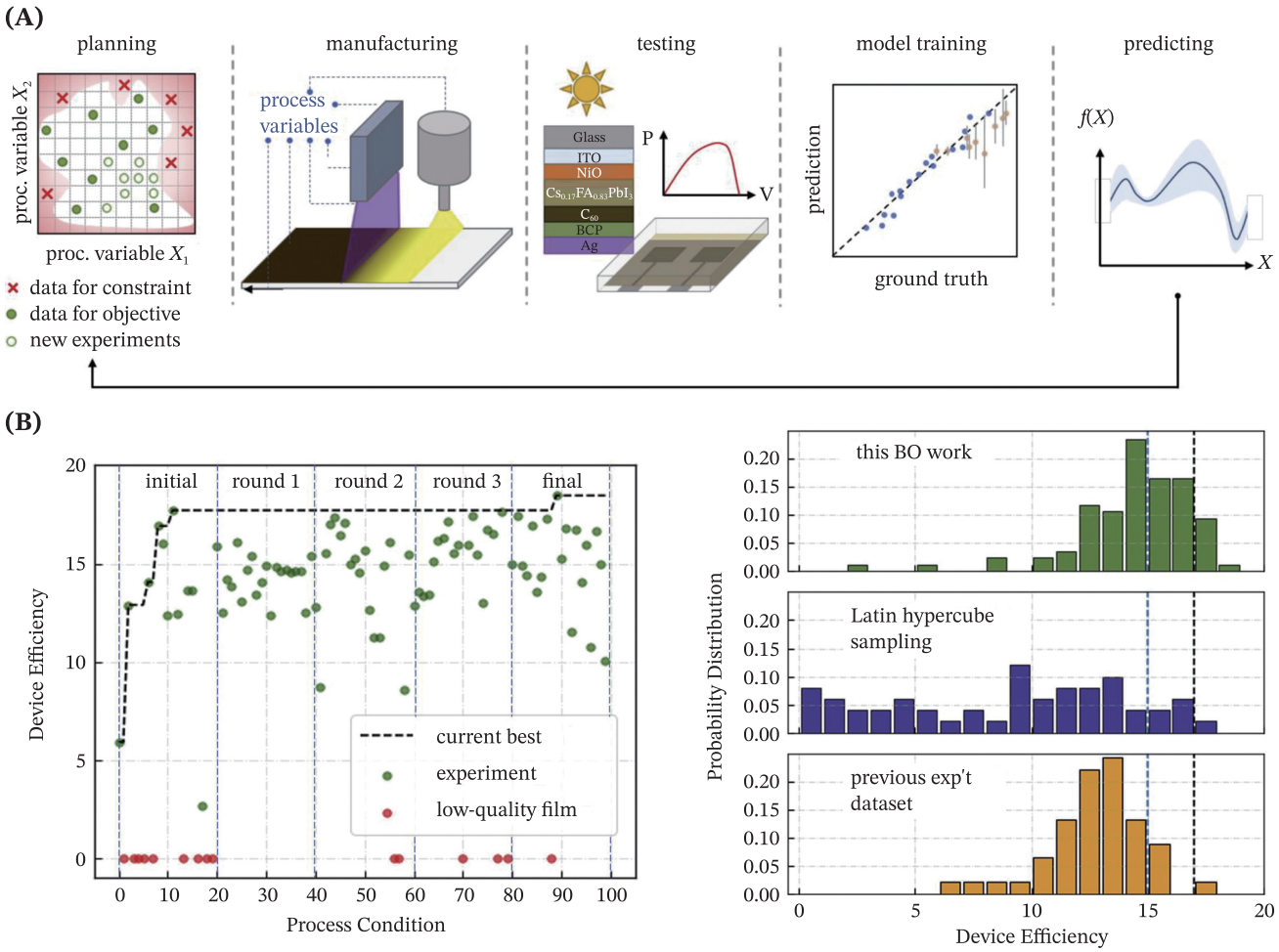

FIGURE 12 (A) Schematic of sequential learning optimization for perovskite solar cells with probabilistic constraints. The five-step iterative workflow includes planning, manufacturing, testing, model training, and prediction. This process iterates until the target efficiency is achieved or the maximum experimental budget is reached. (B) Visualization of the experimental PCE data of solar cells with BO and other methods. Reprinted with permission.[18] Copyright 2022, Elsevier.

cells (Figure 11C). This research paradigm offers a powerful tool to accelerate the discovery and optimization of functional thin films. This innovation could transform fields such as electronics, energy, and optoelectronics by enabling rapid prototyping, efficient process development, and exploration of novel materials and device architectures.

The ML-guided device optimization strategy has proven to be effective and reliable across various fabrication techniques. Liu et al. introduced a method to accelerate the scale-up of perovskite manufacturing by using ML techniques.[18] They incorporated domain knowledge into the optimization process to form a customized BO framework. This framework was specifically designed to enhance rapid spray plasma processing (RSPP) and maximize perovskite devices PCE under open-air conditions. As shown in Figure 12, the framework carried out an iterative process that began with model-free sampling, followed by device fabrication and evaluation under standard test conditions. Data-driven insights and qualitative feedback, including visual assessments and historical data, guided the refinement of experimental parameters. This strategy reduced the number of required experimental iterations, increased efficiency, and minimized experimental overhead compared with traditional methods. By incorporating qualitative feedback and a sequential learning framework, this approach significantly streamlined the optimization process. Their work highlights the advantages of the ML-driven framework in reducing resource expenditure and enhancing predictability, advancing the development of high-performance solar technologies.

2.3.2 | Quasi-2D perovskite devices

Quasi-2D perovskite devices, such as those with Ruddlesden–Popper structures, offer improved photothermal stability and environmental resistance compared with traditional three-dimensional (ABX_3) perovskites.[110–112] Despite these

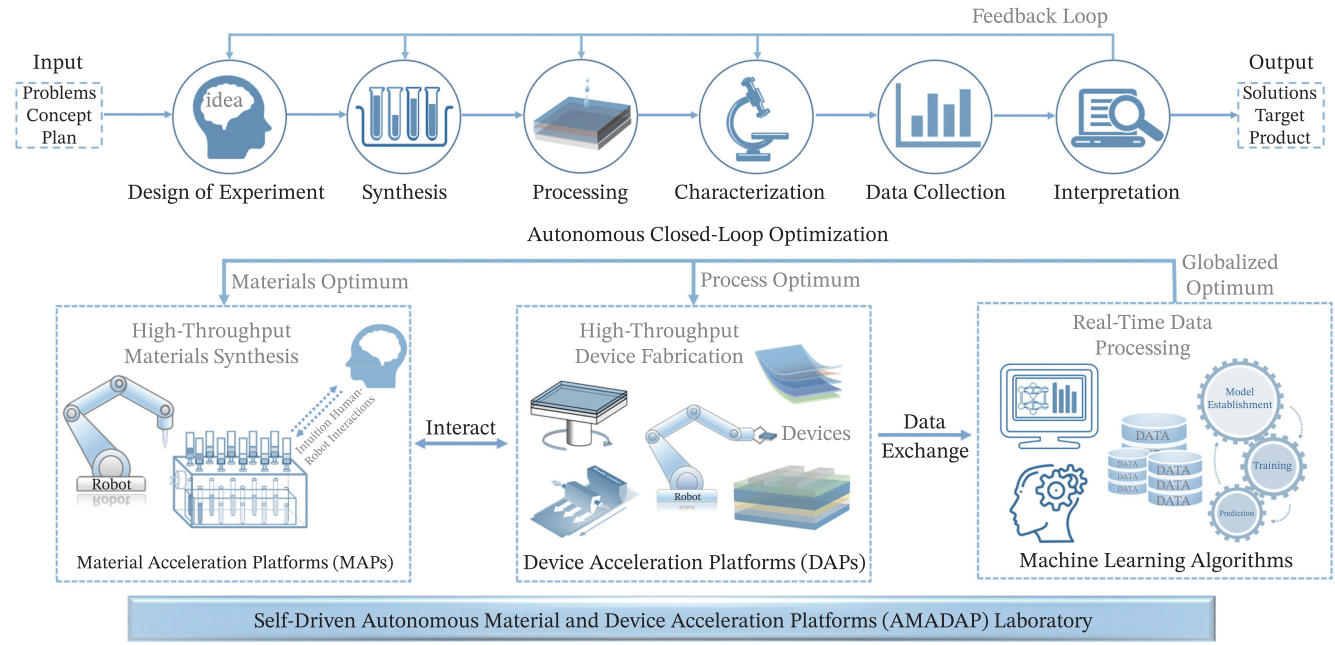

FIGURE 13 Schematic of a holistic technology platform: The self-driven AMADAP laboratory.

advantages, their fabrication involves greater complexity and requires optimizing multiple parameters. To address these challenges, a combinatorial high-throughput screening strategy integrated with a ML model was applied. Benchmark materials were selected based on prior research and experimental findings. A parameter space comprising 16 000 potential—spanning 2D spacer cations, 3D cations, halide ratios, additives, *n*-values, and annealing temperatures—was explored.[113] Automation of the fabrication process using microfluidic devices and programmable robotic platforms ensured consistent composition and minimized human errors. Optoelectronic properties of the films were systematically evaluated, and the resulting data trained the ML model. The model predicted the performance of new combinations and refined the optimization process. Specific parameter combinations were found to have significant effects on the optoelectronic properties of the films. Recommendations derived from the ML model substantially improved device performance. By combining automation with ML-driven optimization, this approach enhanced experimental repeatability and efficiency while accelerating the discovery of high-performance perovskite material combinations.

3 | TOWARD SELF-DRIVING AMADAP LABORATORY

The introduction of the AMADAP laboratory represents a significant advancement in establishing "self-driving laboratories" for emerging perovskite photovoltaic technology.[24,114,115] By integrating high-throughput autonomous Materials Acceleration Platforms (MAPs) with Device Acceleration Platforms (DAPs), the AMADAP laboratory enables simultaneous advancement in material discovery and device optimization (Figure 13).[31,116] Optimized communication, data exchange, and feedback loops between these autonomous platforms allow the effective transformation of promising materials into high-performance devices, streamlining research workflows and paradigms. The incorporation of ML algorithms and digital twin technology improves the efficiency and reliability of autonomous operations through enhanced real-time monitoring, analysis, and experimental simulation. The AMADAP laboratory advances the process of autonomous optimization while establishing a self-driven laboratory environment. This innovative approach provides comprehensive solutions for the development of perovskite photovoltaic technology and marks a paradigm shift in research methodologies.

4 | CHALLENGES

While self-driving laboratories have greatly improved research efficiency, reduced manual trial and error, and lowered experimental costs by combining automation with intelligent optimization, several bottlenecks remain.

4.1 | Construction costs and resource investment

Building a self-driving AMADAP laboratory requires high-precision automated equipment and significant computational resources, and the initial investments are

therefore high. Long-term operational and maintenance costs, as well as system upgrades, add to this burden. However, advancements in software and hardware technologies and the emergence of scale economies are expected to reduce these costs over time.

4.2 | Adaptability and customization of platforms

Ensuring automation platforms to accommodate diverse material systems and research objectives remains a challenge. Different experimental tasks often involve unique preparation processes and parameter spaces. Developing flexible hardware and software modules tailored to specific research needs is critical for broader applicability.

4.3 | Analysis of high-dimensional, multivariate data

High-throughput experiments generate vast amounts of high-dimensional, multivariate data. Efficiently analyzing, mining, and extracting valuable insights from the datasets is a significant challenge. Advanced data analysis tools and machine learning algorithms must be developed to identify optimal solutions and uncover meaningful relationships.

4.4 | Lack of "self-thinking" capability

Despite their efficiency in performing experiments and optimizing data, self-driving platforms cannot independently propose new scientific questions or discoveries. Creativity and scientific insight from the researchers remain crucial for the platform's success; that is, human intellectual output and experimental design play a decisive role in the whole process.

4.5 | Achieving full automation

Although significant progress has been made, some experimental tasks still require direct human participation and intervention, which limits the realization of fully autonomous operation. Tasks such as precise material preparation, equipment maintenance and calibration, and initial data screening often depend on researchers. Enhancements in automatic material management, self-diagnosing equipment, automated anomaly handling, and intelligent experimental design are needed to achieve higher degrees of automation.

4.6 | Interdisciplinary collaboration

Building and operating an AMADAP laboratory requires seamless cooperation among experts from various disciplines, including chemistry, material science, engineering, software development, and data science. Effective communication and collaboration between these interdisciplinary teams remain challenging and are essential for the successful implementation of the platforms.

Addressing these challenges will be essential for further advancing the capabilities and adoption of self-driving laboratories in materials science research.

5 | CONCLUSION

Perovskite photovoltaic technology is on the verge of achieving higher energy conversion efficiencies, lower production costs, and extended service lifetimes, positioning itself as a leading renewable energy solution. As this technology approaches mainstream utilization, the integration of ML-driven automation promises to revolutionize the field by accelerating new material discovery and optimizing multi-component device fabrication. This review highlights the transformative influence of ML-driven automation on the research and development of perovskite photovoltaics. It demonstrates the application of automated strategies for rapid multi-component screening, precise material discovery, performance prediction, and device preparation optimization. The concept of the self-driving AMADAP laboratory represents a significant step toward realizing "self-driving laboratories" in this domain. This approach integrates and streamlines the processes of material innovation and device performance enhancement, expediting the advancement of emerging photovoltaic technologies. By providing a comprehensive analysis of current achievements and challenges, this review not only emphasizes the potential of ML-driven automation but also outlines a promising roadmap for the future development of perovskite photovoltaic technology.

AUTHOR CONTRIBUTIONS

J.Z., Y.Z., and C.J.B. conceptualized this manuscript outline. J.Z. and J.W. made the figures and wrote the draft. J.A.H., Y.Z., and C.J.B. supervised the project. All authors contributed to discussing and approving the final version of the manuscript.

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CONFLICT OF INTEREST STATEMENT

The authors declare no conflict of interest.

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