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Perspective

Self-Driving Laboratory for Polymer Electronics[†]

Aikaterini Vriza, Henry Chan, and Jie Xu*

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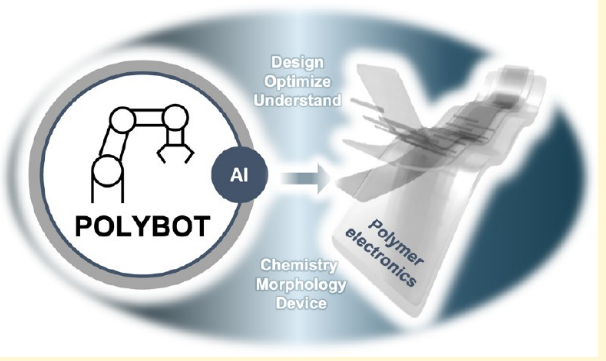
ABSTRACT: Owing to the chemical pluripotency and viscoelastic nature of electronic polymers, polymer electronics have shown unique advances in many emerging applications such as skin-like electronics, large-area printed energy devices, and neuromorphic computing devices, but their development period is years-long. Recent advancements in automation, robotics, and learning algorithms have led to a growing number of self-driving (autonomous) laboratories that have begun to revolutionize the development and accelerated discovery of materials. In this perspective, we first introduce the current state of autonomous laboratories. Then we analyze why it is challenging to conduct polymer electronics research by an autonomous laboratory and highlight the needs. We further discuss our efforts in building an autonomous laboratory, namely Polybot, for the automated synthesis and characterization of electronic polymers and their processing and fabrication into electronic devices. Finally, we share our vision in using a self-driving laboratory for different types of polymer electronics research.



1. INTRODUCTION

Polymers with (opto)electronic properties have enabled new capabilities in electronics, including mechanical flexibility/stretchability, biocompatibility, self-healing capability, degradability, and mixed electron−ion conductivity (Figure 1), which led to the development of new applications such as skin-like electronics, bioelectronics, large-area printed polymer photovoltaics, soft light-emitting diodes, brain-mimetic neuromorphic computing devices, and energy conversion.[1−7] Despite over 40 years of research on (opto)electronic polymers, there is still much to be done to improve their device performance for realizing commercialization.[8] One of the reasons is that the properties of (opto)electronic polymers typically depend on both their chemical structures as well as assembled multiscale morphologies, which are less common for small molecules.[9] Moreover, the fabrication of these polymers into electronic devices requires multiple processing steps and many of these processing conditions are intertwined, and therefore cannot be individually optimized.[10] As such, developing polymer electronics requires searching through a very large design space from a single polymer chain structure optimization to hierarchically assembled morphology and multilayer device structures via processing and fabrication, which is equivalent to performing thousands of experiments under the Edisonian trial-and-error experimentation approach and years of human labor.

Automated materials discovery and process optimization represents a new frontier in materials research and manufacturing. The concept is embodied in paradigm of the self-driving laboratory (SDL), i.e., autonomous or closed-loop experimentation, where robotics and artificial intelligence (AI) are seamlessly integrated, capable of evaluating and directing its own experimental inquiries based on data that is automatically analyzed by machine learning (ML) methods. Such a platform goes beyond standard laboratory automation in that it can solve a problem (e.g., finding formulations that give over 1000 S/cm^2 conductivity) as opposed to being programmed to simply execute a task (e.g., mix solution A and B with a ratio of 1:2). The AI is programmed to make unattended decisions about which experiments to run next based on data collected to date, prior knowledge, and physical constraints. The automated robotics then perform these experiments with precision and speed, enabling highly parallelized and multiplexed testing of materials synthesis/processing conditions and other variables. A SDL is especially useful for combinatorial experiments and multidimensional relationship problems, which has been demonstrated by many researchers for chemical synthesis,[16] mechanical design in 3D printed materials,[17] perovskite-type materials for photovoltaics,[18,19] carbon nanomaterials growth,[20] inorganic quantum dots,[21] and gold nanoparticles.[22] However, SDLs are mostly in their infancy and have so far almost been exclusively

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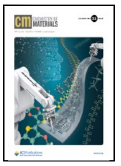

[†]This Perspective is part of the *Up-and-Coming* series.

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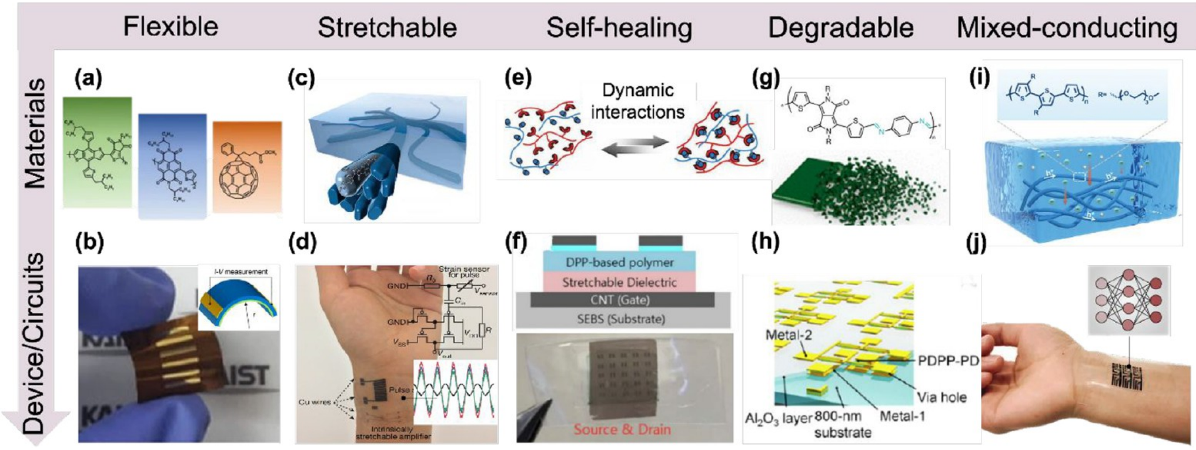

Figure 1. New capabilities enabled by polymer electronics. (a, b) Flexible solar cell made by conjugated polymers and PCBM. Portions of figures adapted with permission from ref 11 licensed under a Creative Commons CC BY license (http://creativecommons.org/licenses/by/4.0/). (c, d) Skin-like circuit made by stretchable semiconducting polymer. Adapted with permission from refs 1 (Copyright 2017 AAAS) and 10 (Copyright 2018 Springer Nature). (e, f) Self-healable transistor arrays made by conjugated polymers with dynamic bonds. Adapted with permission from refs 12 (Copyright 2018 American Chemical Society) and 4 (Copyright 2016 Springer Nature). (g, h) Degradable circuit made by conjugated polymers with degradable bonds.[13,14] Adapted with permission from refs 13 (Copyright 2019 American Chemical Society) and 14 (Copyright 2017 PNAS). (i, j) Neuromorphic array fabricated by mix-conducting polymer. Adapted with permission from refs 15 (Copyright 2022 Wiley-VCH) and 5 (Copyright 2022 Elsevier).

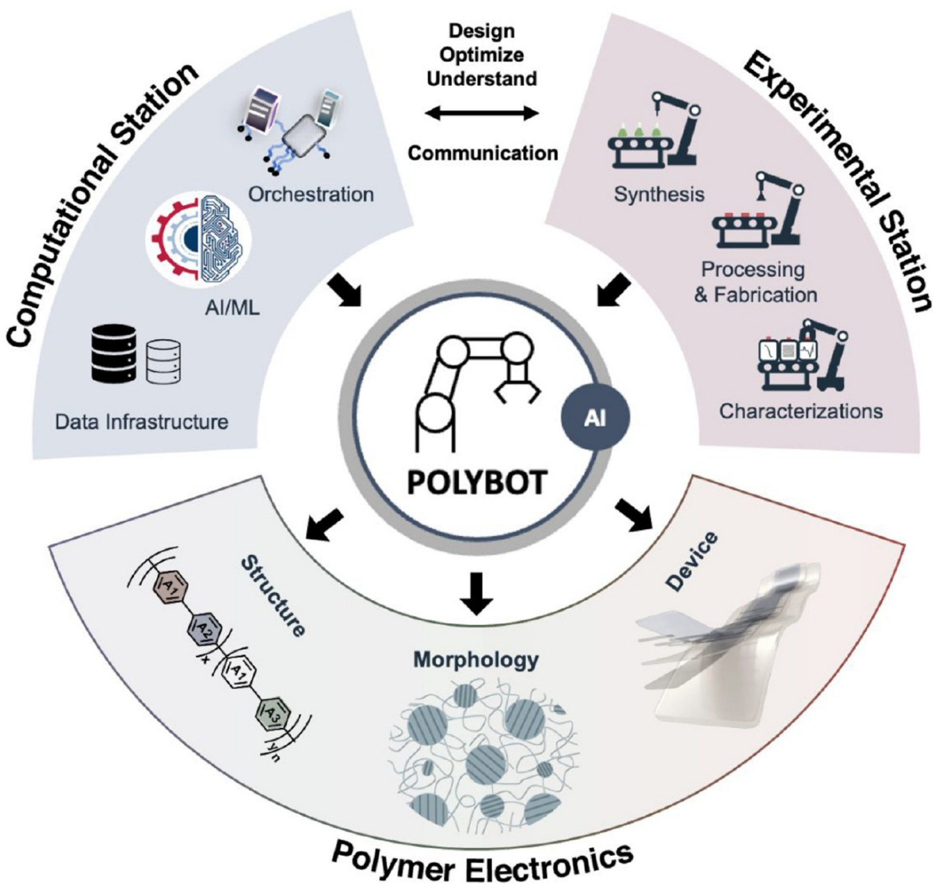

Figure 2. Schematic illustration of a SDLy that combines the power of AI/ML and automation for accelerating the development of polymer electronics.

demonstrated on hard materials and organic small molecules. Their use for both polymers and electronics is still in a nascent stage due to the complexity of experimentations, which requires controls across hypothesis, synthesis, processing, and characterization (Figure 2). In this perspective, we discuss the general design of a SDL, challenges for autonomous polymer electronics discovery, the building of a SDL for polymer electronics, and our near future vision.

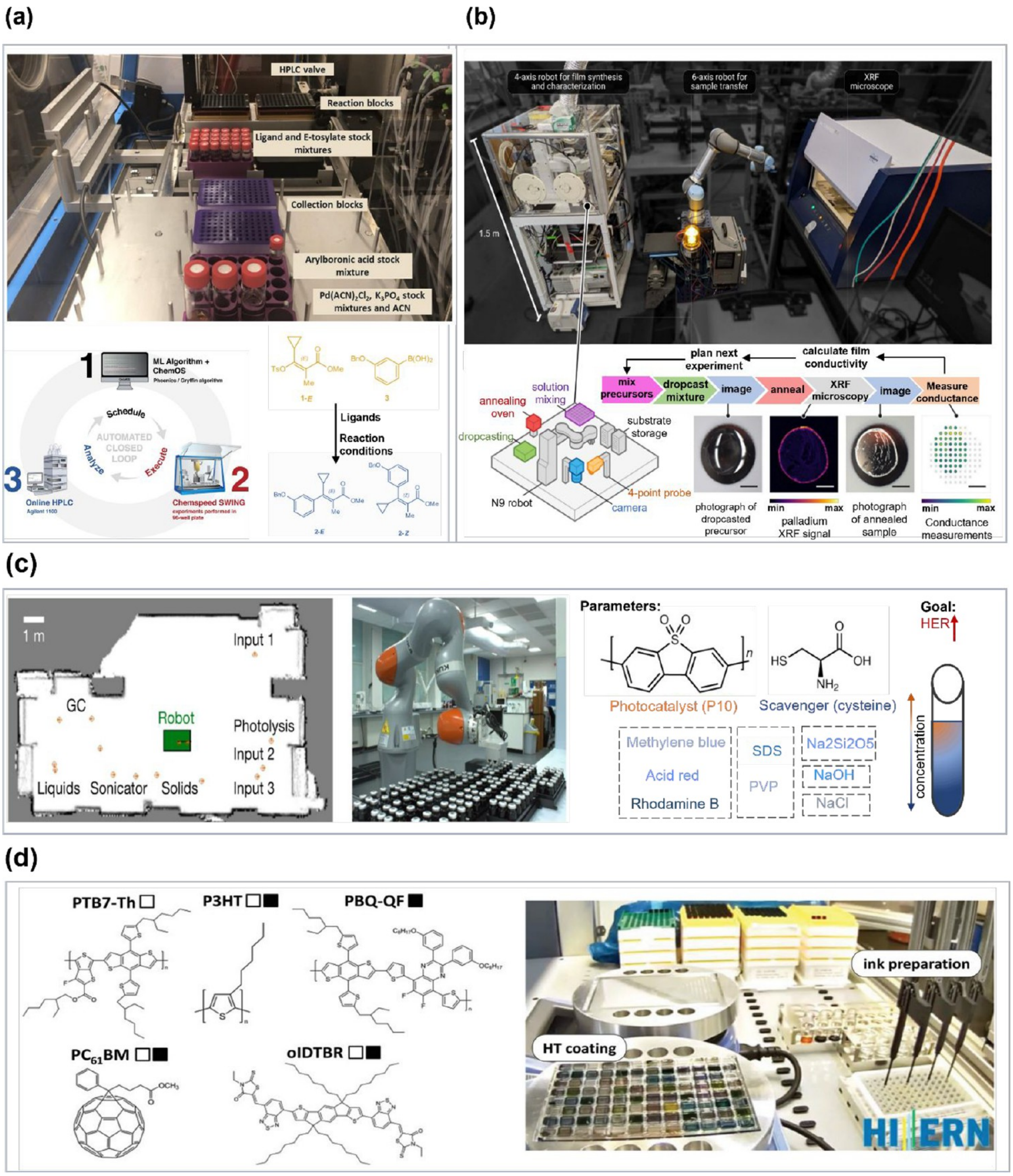

Figure 3. Self-driving laboratories developed for different materials. (a) A Chemspeed robot for the optimization of a stereoselective Suzuki−Miyaura coupling reaction. Within a budget of 192 experiments, the robot identified the conditions that maximize both the yield and the selectivity of the desired product. Adapted with permission from ref 23, licensed under a Creative Commons CC BY license (http://creativecommons.org/licenses/by/4.0/). (b) Two robots (N9 and UR5e) working together for the synthesis and characterization of palladium films. The robot is able to handle several processing and fabrication conditions and optimize the annealing temperature and conductivity within a budget of 50 experiments. Adapted with permission from ref 24, licensed under a Creative Commons CC BY license (http://creativecommons.org/licenses/by/4.0/). (c) A mobile KUKA robot designed to screen photocatalysts for hydrogen production from water. The robot searched 10 different parameters and maximized the hydrogen evolution rate (HER) within 688 autonomous experiments. Adapted with permission from ref 25. Copyright 2020 Springer Nature. (d) SDL for the fabrication of organic photovoltaic (OPV) films optimized for photostability. Following the self-driving approach, the group was able to identify the optimal materials within a budget of 60 samples. Adapted with permission from ref 26. Copyright 2020 Wiley-VCH.

2. GENERAL DESIGN OF A SELF-DRIVING LABORATORY

A SDL consists of three building blocks: (*i*) the equipment (hardware), (*ii*) the processes (software), and (*iii*) the data. Hardware includes equipment components such as reactors, coaters, characterization instruments, and robots. Software refers to orchestration tools and algorithms that control the hardware, as well as learning algorithms that contribute to automatic decision-making. Learning algorithms, such as Bayesian optimization, are often coupled with an automated system to enable materials property predictions and to propose next trial parameters for experimentation that can efficiently explore a materials search space. A less constrained search space

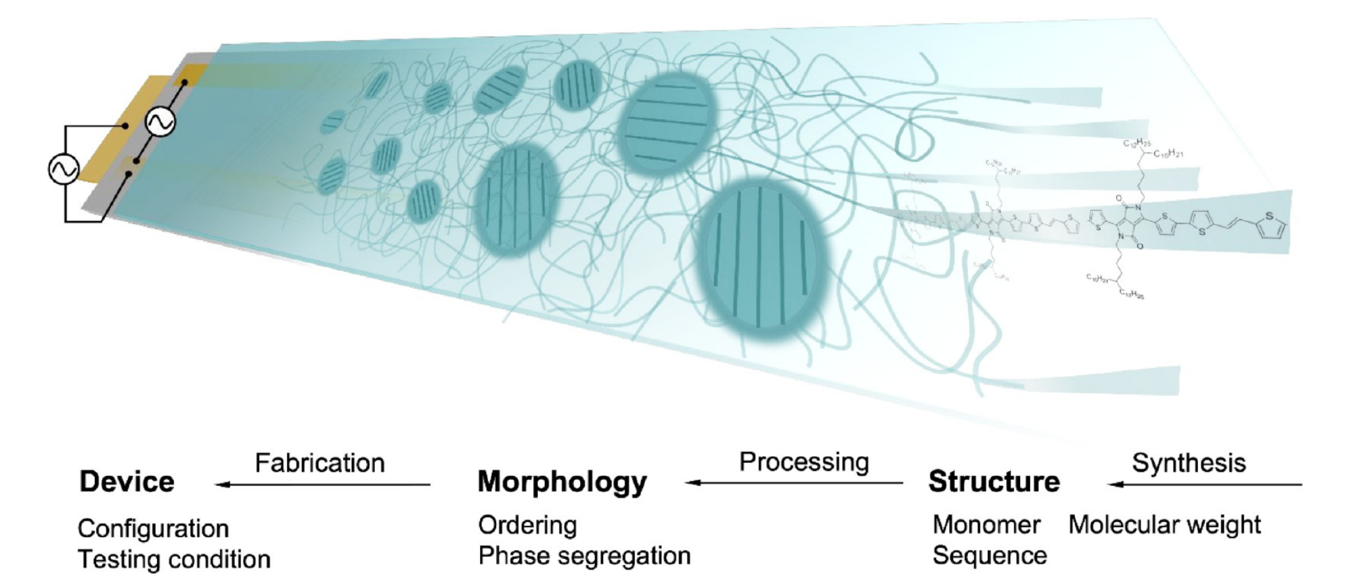

Figure 4. Device to morphology to polymer chemical structure designs in the development of polymer electronics.

and a more flexible automation platform allows for a higher possibility of new discoveries. Lastly, the sources of data can include knowledge extracted from past experiments or literature, information measured by instruments (e.g., temperature in a reactor throughout the experiment), and data analyzed by ML methods. In short, a SDL performs experimental procedures using robotics/high-throughput instruments and responds to feedback provided by data to adjust a search or handle any component failures and failed experiments.

The appeal of a SDL lies in its potential to increase efficiency via high-throughput autonomous experimentation and data analysis. In an end-to-end system, it would be possible to perform experimentation at scale with very limited human interactions. As such, manual labor can be reduced and researchers can focus more on higher level scientific tasks such as formulating a hypothesis, designing experimental campaigns, and interpreting results. The health and safety of researchers will also be improved, as harmful chemicals or difficult experimental sequences can be performed with reduced human exposure. Furthermore, AI/ML algorithms are better at handling high dimensional data than a human, and data collected by a SDL can be more information rich, e.g., contain precise details of the experimental conditions and other relevant metadata. Such data can be monitored and displayed live on dashboards or shared across platforms for a variety of scenarios such as experiments that are performed under a similar setup.

In the past few years, several academic and industrial research groups have worked on accelerating the discovery of a wide range of materials using platforms that integrate automated experimentations and AI/ML algorithms to guide experiments without human intervention. Abolhasani and co-workers developed the Artificial Chemist for automated continuous flow synthesis of inorganic quantum dots and simultaneously optimized three objectives using information extracted from the emission spectra of their formulations.[27] Hein et al. developed an automated high-throughput screening workflow for multi-objective optimization of the Suzuki−Miyaura coupling by integrating a Chemspeed Technologies SWING system with online HPLC analysis (Figure 3a).[23] Cooper et al. developed a mobile robotic chemist for autonomously screening active photocatalyst mixtures of various reagent types and concentrations (Figure 3c).[25] Cronin and co-workers developed a robotic system capable of autonomously performing multistep organic synthesis and optimizing the yield and purity of pharmaceutical compounds.[28] Berlinguette and co-workers demonstrated thin film manufacturing that takes into account materials solid state performance (Figure 3b).[24,29] Brabec et al. developed an automated platform for the optimization of multicomponent polymer blends for OPVs (Figure 3d).[26] Aspuru-Guzik et al. conceptualized an autonomous robotic workflow for inorganic thin-film materials with solid state control capabilities.[30] Despite the significant advancements made by these research groups, a transition from traditional benchtop experimentation to a fully autonomous experimental paradigm is still not yet achieved. This is mainly due to the high complexity in automated experimental workflow for materials discovery and the requirement for a well-trained team of researchers with expertise that span across interdisciplinary fields (e.g., material, automation, AI/ML). Geographic decentralization of the equipment also makes it difficult for different research groups with specific automation capabilities to closely collaborate. The cost of an autonomous lab can vary widely depending on the research requirements. Individual Principal Investigator led basic research lab groups could be very good in building SDL for their domain science after considering the capital and expertise required. User facilities could develop libraries of computational and ML methods, automated workflows including unique facilities (X-ray and supercomputer), and data sets of different applications that could be widely available to the community. Some companies (e.g., IBM, DeepMind) are also developing SDL and AI systems for scientific research.[31,32] A recent attempt in developing SDL involves the Acceleration consortium[33] which tries to connect the academia, government, and industry to join forces and inspire the new generation of researchers. Currently, we are still at the early stage of SDL development, and there is still so much to do and to learn. In the future, the efficiency could be further improved through innovations such as improvements in workflow tools and data infrastructure, corporative work of many robots, and better prediction models that leverage physics-informed machine learning methods and multiscale simulations.

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3. CHALLENGES IN CONDUCTING POLYMER ELECTRONIC RESEARCH USING AN AUTONOMOUS LABORATORY

The properties of electronic polymers typically depend on both the chemical structure and the morphology of the material, which requires codesign of synthetic and processing techniques in order to achieve the final electronic polymer material with desired properties in the device. The fabrication of electronic polymers into electronic devices involves ensembles of multiple layers rather than single layer material, which complicates their processing and characterization. In this section, we highlight several unique challenges of polymer electronic research with regard to polymer synthesis and preparation, device fabrication, and (opto)electronic performance characterization (Figure 4).

Electronic Polymer Synthesis. Unlike inorganic materials or small molecules, polymers are large molecules made of strong covalently bonded organic monomers that are interpenetrated or entangled with each other, which leads to requiring polymer specific experimentations and procedures for their synthesis. For example, the chemistry employed in a step-growth polymerization typically requires extremely high conversion to reach suitable molar mass and thus has high requirements in the control of its reaction conditions and operations.[34] In addition, the mixing method of a reaction station should be designed for a viscous system resulting from polymer products. Different than small molecules, the general method for purifying polymers is precipitation, not recrystallization, when considering all common purification methods for small molecules including column chromatography. For these reasons, a suitable automated modular polymerization system needs to be specifically designed for polymerization reactions and proper methods are needed to purify polymers.

Electronic Polymer Thin Film Processing and Device Fabrication. Electronic polymers are usually solution-processed into a thin film and serve as active layers in different types of functional devices. While solution processing offers the advantages of being additive, low-temperature, and scalable, it remains a central challenge to control the assembly of electronic polymers from the molecular scale to the device scale due to highly nonlinear growth processes.[35] The final morphology and properties of electronic polymer thin films are a direct consequence of the formulation (e.g., concentrations, additives) and processing conditions (e.g., coating speed, coating temperature, annealing conditions). Therefore, an automated modular processing platform that can simultaneously tune the formulations and processing conditions is essential for achieving desirable morphology in electronic polymer thin films. Automatically fabricating electronic polymers into multilayer devices is also challenging, where process compatibility and patterning are the major issues.[10] Selecting the suitable device structures or fabrication methods is the key to enable high-throughput device fabrication. In general, for polymer electronics, the metrics of both material and device comprise of a high dimensional design space and require very complicated experimental procedures. For example, optimizing the performance of a polymer electrochemical transistor can require tuning of 4-dimensional formulation-processing design space (formulation, processing, postprocessing, and substrate) and measurements in a 5-dimensional property space (charge carrier mobility, ionic conductivity, capacitance, transconductance, threshold voltage).[15] The complexity of experimental procedures and degrees of freedom associated with the exploration space is particularly challenging to incorporate in an autonomous experimental workflow.

Electronic Polymer and Device Characterizations. To study the performance of polymer electronics, a suite of selected automated characterization tools needs to be tailored for both electronic polymer and device characterizations. On the materials side, obtaining the structural and morphological characteristics of electronic polymers could be challenging. For instance, studying the polymer morphology (e.g., domain size) may require characterization techniques that are time-consuming, expensive, and not easily available in the laboratory (e.g., X-ray scattering in high energy light source facilities).[36] Finding alternative characterization methods, such as UV−vis spectroscopy or optical microscopy, could be a solution for fast screening. On the device side, characterization of polymer electronics also presents unique challenges since more than one parameter is typically needed to measure and determine the device performance. In addition, data sets generated from different measurements are vast and require dedicated methods for online data analysis and automatically extracting the inputs/outputs for the closed-loop experiment.[37] Different types of polymer electronics might require different methods for property characterizations.

4. TECHNICAL REQUIREMENTS OF A SELF-DRIVING LABORATORY FOR POLYMER ELECTRONICS

Automated Experimental Workflow. The first step of an autonomous research study for polymer electronics is to identify the scientific questions and formulate the experimental procedure. The experimental procedure is then translated into hardware sequences that can be executed by a robotic platform. Since a user who is familiar with an experiment is not necessarily knowledgeable about robotics hardware or computer programming, a large number of trials-and-errors is often needed to adapt an experimental procedure designed for human experimenters to an automated workflow. In order to handle a complex experimental workflow which is typical of electronic polymer synthesis, processing ,and characterization, a SDL should provide a flexible yet simple interface for setting up the autonomous processes so that the time and human effort involved can be reduced. Most existing commercially available synthesis and characterization instruments do not come with an Application Programming Interface (API) that allows for automated operations and direct communication with the control system of a SDL and are not designed to be easily reconfigurable.[38] For example, a mechanical probe station, a commonly used electronic characterization platform for polymer electronics, does not come with an automated mode that can be controlled by programming codes. Despite the existence of data management platforms like ChemOS,[39] the integration between vendor-provided hardware and custom laboratory codes remains a major challenge. Open sourcing of hardware and software among the research community can be beneficial for researchers who are deploying similar systems that can be used for polymer electronic research.

AI/ML Frameworks. Machine learning has become a powerful tool in laboratory automation for experimental planning and optimization of the manufacturing conditions. Bayesian optimization (BO) is among the most common optimization strategies used as an alternative to the traditional design of experiments (DOE) approach. Relating to the practical application of these approaches, there are still many questions to answer: (1) how to effectively navigate and sample from

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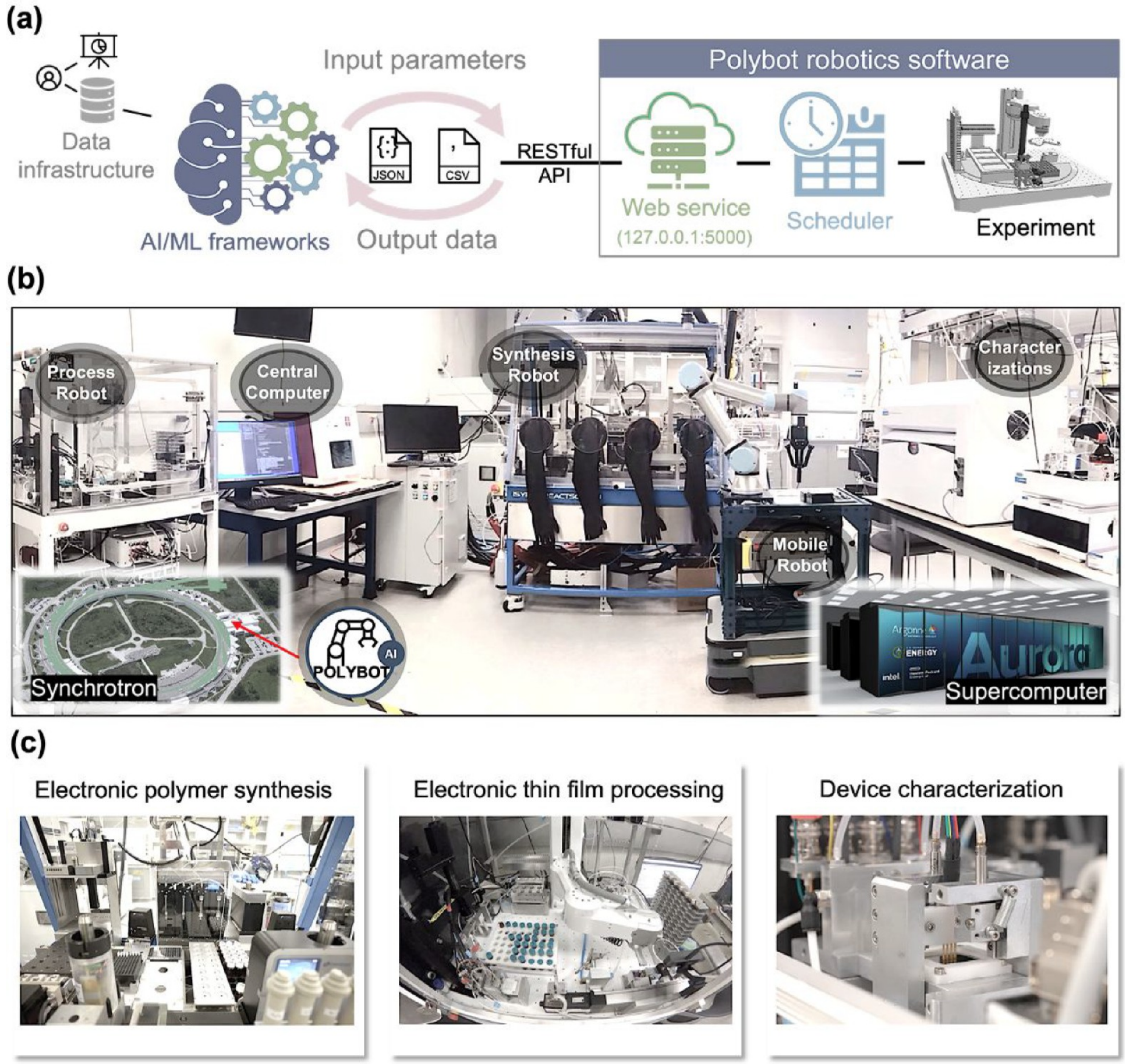

Figure 5. Polybot: an AI-integrated robotic laboratory that is suitable for polymer electronics discovery. (a) Polybot software framework for the setup of AI-guided robotic experimental workflows. (b) Layout of Polybot with synthesis robot platform, processing robot platform, a suite of automated cauterization modules that are connected by a mobile robot. Through a web application, integrated autonomous workflows can be enabled across facilities (e.g., synchrotron and supercomputers). (c) Modular automated experimental features in Polybot for enabling the experimental features of synthesis, processing, and characterizations in polymer electronics.

constraint spaces which are very common in chemical research,[40] (2) how to incorporate advanced algorithms that can handle multiobjective problems,[41,42] (3) which guidelines to use for selecting the appropriate optimization strategy based on the materials case,[43] (4) how to make better use of materials descriptors in the cases when we have correlated categorical variables),[44] (5) how to benchmark the available algorithms and select the best optimization models,[45] and (6) whether we can use prior knowledge from related problems to enable knowledge transfer across optimization campaigns.[46] Finally, looking beyond BO, are there better algorithms and learning strategies that can be used as experimental guiding tools in fully autonomous workflows while being interpretable and explainable that can incorporate physics and domain knowledge?[47,48]

Orchestration Software. The orchestration of experimental procedures, equipment, and the AI/ML framework is crucial for the realization of a closed-loop process without human intervention. A typical workflow for polymer electronics involves the use of multiple synthesis/processing and characterization tools that often require transporting samples across different robotic platforms and instruments. Since polymer electronic samples can be in different form factors such as dimensions, hardness, and even solid−liquid hybrid, mobile robots or track-based sample transfer systems with a properly designed gripper and sample handling method is required for handling the loading and unloading of samples across different equipment. To efficiently utilize multiple automated tools in a SDL, the orchestrator should leverage warehouse-like logistics and concurrency/parallelization strategies in the planning of work-flow operations and the pick-up sequence and location of samples, to optimize the costs, time, and other objectives and constraints such as energy consumption and task dependencies.

Data Infrastructure. A well-established data infrastructure is fundamental to the handling of the volume, integrity, and

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accuracy of data generated from an autonomous experimental workflow.[49,50] The major requirements are (1) choosing a standardized file format that is flexible enough to store a variety of data types (e.g., images, tables, spectra, chemical structures, $I-V$ curves), (2) defining a data schema that provides consistent description and organization of information including input/output of instruments as well as other context-specific data such as experimental process details, (3) selecting a file transfer protocol or data management service that is scalable for a large number of files and sizes, (4) engineering workflow procedures that utilize statistical approaches to provide insights on the validity and reproducibility of data, and (5) leveraging the FAIR (Findability, Accessibility, Interoperability, and Reuse) principles[51] to ensure high quality reproducible data of both successful and failed experiments are properly stored and are easily accessible by AI/ML frameworks.

Database Creation and Data Mining. Information, such as formulation conditions, characterization data, and physical properties, that are calculated or extracted from well-established public or proprietary data sets or similar experiments can be used to improve the performance of a closed-loop experiment and reduce the number of required iterations. An effective gathering of data requires both the extraction of literature reported data and the submission of data to an organized database. With regards to data extraction from literature, several data mining tools have emerged to automate the incorporation of relevant information such as reaction recipes to materials acceleration platforms.[52,53] However, even the most well-trained algorithms can struggle with older or newer literature papers due to periodic changes in journal formatting and the way data are stored. Regarding the data deposition, there are still authors and journals that do not directly incorporate the data set within the publication and might not provide the data when requested.[54] Moreover, the quality of the reported data might be inconsistent between sources or lacking "negative" data from unsuccessful synthetic/manufacturing attempts. Although there are currently significant attempts to encourage data sharing and deposition from journals and there are already some examples of high-quality FAIR databases for materials, e.g., the Cambridge Structural Database (CSD) for crystal structure deposition,[55] a standardized database for polymer electronics has not yet been realized.

Human Interactions. A SDL that operates in the presence of human scientists should be programmed for effective communication and exchange of information during the robot−equipment and robot−human interactions. For example, a mobile robot that shares a work area with a human should leverage computer vision and other sensors to adjust its speed and distance to handle a sudden appearance of obstacles. Furthermore, in the event of a collision or emergency stop, the orchestrator should notify the user, record the status of all equipment, and present relevant information so that a human can troubleshoot and roll the system back to a safe point to restart or resume the autonomous experiment.

5. POLYBOT—A SELF-DRIVING LABORATORY FOR POLYMER ELECTRONICS

Taking into consideration the above-mentioned experimentation challenges and technical requirements, we developed a SDL, namely Polybot, to target the development of polymer electronics (Figure 5). In this section, we highlight and discuss selected features of Polybot.

Workflow and Orchestration Software. Polybot features a software framework for the setup of common experimental workflows for polymer electronics research.[56] The framework is built using the Python programming language for its readability, flexibility, and wide adoption in the scientific communities. To handle complex workflows, we represent hardware modules as objects and steps of an experimental procedure as functions, which further organize the autonomous workflow into high-level entities written in a single script that are composed of frequently used intermediate steps encapsulating low-level robotics and instrument control sequences. The workflow software module also provides a means to define the search space of input parameters associated with every workflow step, which can comprise of discrete and/or continuous variables. A web application is developed to parse the workflow script and facilitate server-client type communication via RESTful API, which provides a way to integrate autonomous platforms across facilities (e.g., The Advanced Photon Source and Aurora supercomputer at Argonne National Laboratory). The web application also provides a graphical user interface and the ability to generate a web form for novice users to cross check the workflow steps and execute the autonomous workflow with specific input parameters.

The Polybot software also provides a scheduler that coordinates the execution of sample processing and robotic sequences of hardware equipment. The scheduler aims to be a hardware scheduling system that can orchestrate modules within a robotic platform and leverages AI/ML to improve the efficiency of their interactions and sequence of actions, e.g., robot arm movement, solution stirring, and temperature annealing. This feature is particularly useful for a serial robot station that can only process one sample at a time. With the execution time predicted by AI/ML, strategies can be applied to concurrently process multiple materials samples, which effectively make use of waiting time of time-consuming steps to perform actions on other samples, e.g., prepare the next few sample solutions during the spin coating process of the current sample on a substrate.

Data Quality and Infrastructure. There are two main data sources in Polybot, (*i*) data gathered from literature resources and (*ii*) data generated in the laboratory through autonomous experimentation. In polymer electronic research even very small changes in the experimental conditions can significantly affect the final product. As such, the quality of the data (e.g., device performance) being created during a high-throughput experiment needs to be established. Statistical analysis tools have been incorporated in our experimental workflow to ensure data reliability and reproducibility. While it is still a challenge for Polybot to operate for an extended period of time without human supervision, major improvements in hardware reliability are expected in the near future with the gradual development and implementation of an intelligent sensory network, computer vision, and sophisticated error handling procedures. Moreover, Polybot leverages advanced data transfer and management tools and is connected to materials data repositories, including the Materials Data Facility. In that way the knowledge produced in the laboratory can be shared with different groups and data facilities.

Robotic Stations. Polybot has two main robotic stations: a synthesis robot station (ISYNT from Chemspeed) and a processing and fabrication robot station (N9 from North Robotics). These stations are equipped with a suite of modular easily reconfigurable automated characterization modules for

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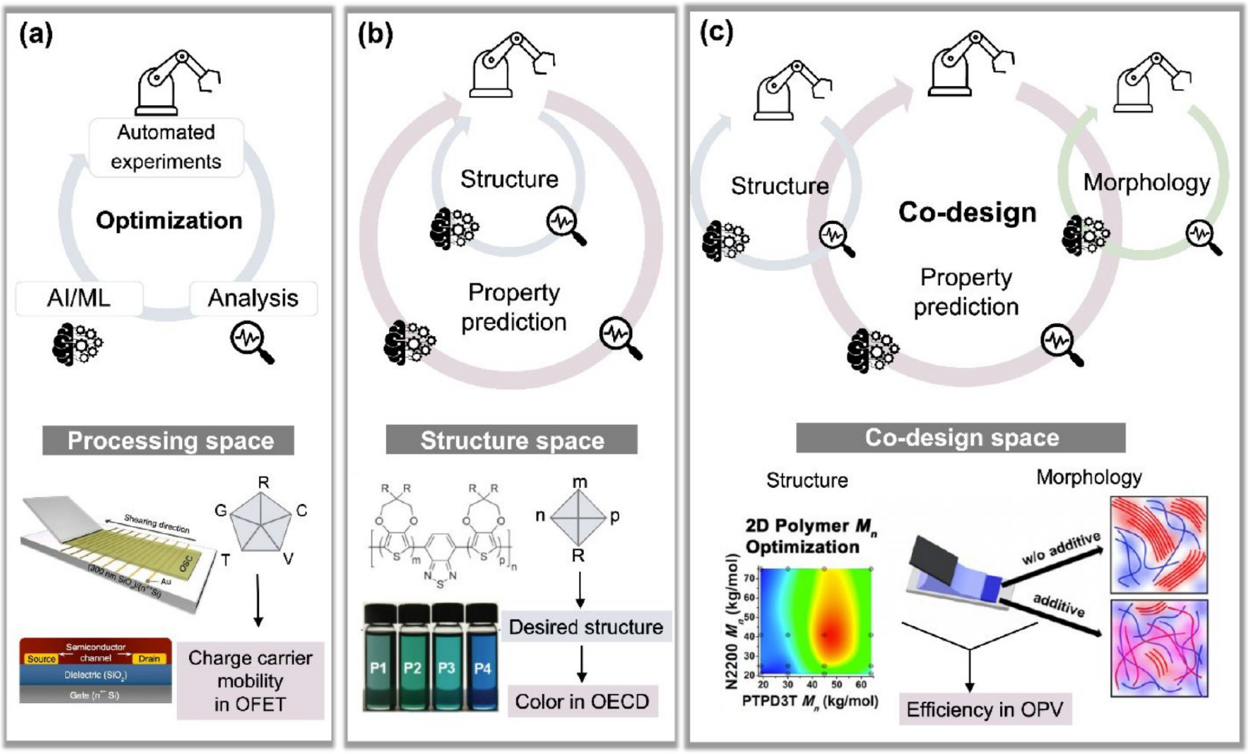

Figure 6. Perspectives of the near future self-driving platform for accelerating polymer electronics discovery. (a) Single-loop research. Bottom: possible research that optimizes the charge carrier mobility in a printed organic field effect transistor (OFET) via tuning the processing conditions (R: polymer blends ratio, C: ink concentration, V: coating velocity, T: coating temperature, G: printing blade design). Adapted with permission from ref 57. Copyright 2019 Springer Nature. (b) Two-loop research. Bottom: possible research that modulates the color in organic electrochromic device (OECD) via chemical structure design.[58] Adapted with permission from ref 58. Copyright 2010 Wiley-VCH. (c) Co-design research. Bottom: possible research that improves the efficiency of organic photovoltaic (OPV) cells via structure design and morphology control.[59] Adapted with permission from ref 59. Copyright 2021 American Chemical Society.

electronic polymer processing and device fabrication, and they are complemented by a mobile robot (MIR200 wheel robot, UR5e robot arm) that has finger and vacuum grippers for transferring materials samples across the robot stations and other characterization instruments.

The synthesis robot station is an enclosed system for conducting chemical reactions under a controlled temperature and gas environment. For polymer synthesis, it is customized with a fully automated modular polymerization system that includes a liquid and powder handing system, a parallel reactors, a purification system (e.g., filtration, centrifuge), a chemical storage system, and analytical instruments (e.g., gel permeation chromatography (GPC), viscometer). Batch synthesis can be performed using vial-based parallel reactors, which enables low-waste and rapid identification and screening of monomers as well as suitable polymerization conditions.

The processing and fabrication platform (N9, North Robotics) is also an enclosed system, which has a constrained frame design that facilitates environment management. It has a fully automated modular processing system that is developed for the electronic polymer, which involves controlling assembly in three major steps: solution formulation, thin film processing, and thin film postprocessing. The platform includes a pipetting system for solution transfer, a liquid handling system, a substrate handling system, different coating stations with controlled temperature and coating rate for film deposition, and a heating stage for film annealing and sample storage racks. To fabricate thin films into devices for electrical property evaluation, a simple method is to print the electronic polymer films on standard device substrates. For example, a semiconducting polymer thin film can be coated on top of a rigid device stack (e.g., predeposit source and drain electrodes on 300 nm SiO_2/Si) to fabricate a bottom gate/bottom contact organic field effect transistor (OFET). For other types of devices, such as polymer photovoltaics (PV) and polymer electrochemical transistors (PECT), multiple layers can be coated on a standard device stack to complete the device fabrication.

Automated Characterization Tools. Within and outside of the two robot stations, Polybot features a suite of automated characterization tools tailored for the study of electronic polymers and their electronics performance. The list of characterization tools is for (*i*) electronic polymer structure (e.g., GPC, UV−vis), (*ii*) morphology of electronic thin films (e.g., optical techniques), (*iii*) electrical properties (e.g., automated and reconfigurable probe station, electronic characterization system), and (*iv*) mechanical properties (e.g., indentation). Many of these tools come with their own vendor specific control software with various degree of openness and are written in different programming languages. In Polybot, we utilized a combination of approaches to integrate these tools into the centralized control software, which includes vendor provided APIs, our own written codes, open-source codes (e.g., Seabreeze for UV/vis spectrometer), and wrappers for library files.

AI/ML Guided Experiments. The first step in running a polymer electronic experiment using Polybot is to design the searching space of a target of interest. For example, we begin with the selection of processing parameters or the recipe for the

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materials synthesis and identify boundaries of the corresponding search space based on hardware limitation, priorities, and budget on resources. Next, we define the objectives of the experiment, i.e., which are the properties we want to achieve, and include the appropriate characterization equipment. Polybot then utilizes an integrated active learning system that can leverage this information and guide the experimentation by suggesting the experimental conditions which are expected for each or the manufacturing goal in as few iterations as possible. The Active learning tool incorporates widely used optimization algorithms, e.g., Gaussian processes and Random Forest regression. The orchestration of the whole closed loop process is facilitated by the communication between the Scheduler and the Active learning module via an iterative process by the exchange of files (e.g., JSON, CSV file formats) which provides all the information about each individual experiment in a consistent data format.

6. FUTURE PERSPECTIVES

Today's electronics are rigid and brittle and cause a lot of environmental issues. We envision a future where a polymer can enable new capabilities for electronics that can be merged into what we wear and what we implant inside our bodies in a sustainable manner. To accelerate this field, the way that we are developing polymer electronics today needs to be changed. Cars are built using robots. Why are we not using robots to do lab experiments? Why are robots not "inventing and making" new polymer electronics?

In this Perspective article, we have described the main challenges and current progress toward the realization of a SDL for polymer electronics development. In this context, we are introducing our platform, namely, Polybot, designed for closed-loop discovery. Our ultimate goal is to combine the advantages of laboratory robotics with the power of machine learning algorithms to accelerate the development of polymer electronics with reduced cost, improved efficiency, and reliability.

We are currently exploring various approaches to extend self-driving components of our laboratory, taking into account several important features that a "laboratory of the future" should possess. Our vision in using a SDL for polymer electronics development can be summarized in three main types of research (Figure 6): (1) single-loop research with the possible goals to produce an electronic polymer with desirable chemical structures (e.g., monomer composition, molar mass, and distribution) with optimized reaction conditions, to process an electronic polymer thin film with favored morphologies (e.g., domain size, alignment, crystallinity) via tuning processing conditions, or just to do formulation/processing for best properties; (2) two-loop research that explores the desirable properties by precisely tuning the chemical structures/morphologies; and (3) three-loop or codesign research where the performances are determined by both chemical structure (synthesis) and morphology (processing). Herein, the synthesis cycle aims to drive discovery through chemical synthesis, the processing cycle to drive discovery via morphology control, and the third cycle to enable the integration and communication between the two previous cycles. That will lead to the creation of a polymer electronics database where all the generated information will be standardized and stored such that the relations and connections of materials and properties could be easily identified. With that knowledge at hand, we can further aim for inverse materials design to be able to synthesize on demand polymers with the desired properties for polymer electronics. Beyond this main mission, this new experimental framework is open to continuous improvement, such that sophistication can be added over years so that libraries of computational and ML methods, automated workflows, and data sets of different applications can be built to continually redefine the cutting-edge of autonomy for polymer electronic discovery.

AUTHOR INFORMATION

Corresponding Author

Jie Xu − *Nanoscience and Technology Division, Argonne National Laboratory, Lemont, Illinois 60439, United States; Pritzker School of Molecular Engineering, The University of Chicago, Chicago, Illinois 60637, United States;* orcid.org/0000-0001-7842-5496; Email: xuj@anl.gov

Authors

Aikaterini Vriza − *Nanoscience and Technology Division, Argonne National Laboratory, Lemont, Illinois 60439, United States*

Henry Chan − *Nanoscience and Technology Division, Argonne National Laboratory, Lemont, Illinois 60439, United States;* orcid.org/0000-0002-8198-7737

Complete contact information is available at: https://pubs.acs.org/10.1021/acs.chemmater.2c03593

Author Contributions

The manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript.

Notes

The authors declare no competing financial interest.

Biographies

Aikaterini Vriza (Orcid: 0000-0002-5663-8703) is a postdoctoral appointee at Argonne National Laboratory. She obtained her Ph.D. from the Material Innovation Factory at the University of Liverpool in 2022 and a Master in Green Chemistry and Sustainable Industrial Technology from the University of York. Prior to that she was an Aviation engineer in the Hellenic Airforce. Her research expertise lies between AI/ML, "green" chemistry, and laboratory automation, and she has worked on several related projects in both industrial and academic settings.

Henry Chan is an Assistant Scientist at Argonne National Laboratory. He received his Ph.D. in Computational Chemistry from the University of Illinois at Chicago in 2016. Prior to his current position, he was a postdoctoral researcher at both Argonne and UIC. His research interests focus on nanostructured materials for a circular economy and energy applications. He develops and applies AI/ML methods in areas including multiscale simulations, diffraction imaging, autonomous experimentation, and inverse design.

Jie Xu is an Assistant Scientist at Argonne National Laboratory. She received a Ph.D. from Nanjing University in 2014 and pursued postdoctoral training at Stanford until 2018. Her research focuses on developing an AI-integrated automated laboratory to accelerate the discovery of a new class of polymer-based electronic materials that are flexible, stretchable, durable, degradable, and easy-to-manufacture for our future electronics. She received the MRS Postdoctoral Award and is named to the MIT TR35 and the Newsweek list of America's Greatest Disruptors as a budding disruptor.

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