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Autonomous materials discovery and manufacturing (AMDM): A review and perspectives

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ABSTRACT

This article presents an overview of the emerging themes in Autonomous Materials Discovery and Manufacturing (AMDM). This interdisciplinary field is garnering a growing interest among the scientists and engineers in the materials and manufacturing domains as well as those in the Artificial Intelligence (AI) and data sciences domains, and it offers immense research potential for the industrial systems engineering (ISE) and manufacturing fields. Although there are a few reviews related to this topic, they had focused exclusively on sequential experimentation techniques, AI/machine learning applications, or materials synthesis processes. In contrast, this review treats AMDM as a cyberphysical system, comprising an intelligent software *brain* that incorporates various computational models and sequential experimentation strategies, and a hardware *body* that integrates equipment platforms for materials synthesis with measurement and testing capabilities. This review offers a balanced perspective of the software and the hardware components of an AMDM system, and discusses the current state-of-the-art and the emerging challenges at the nexus of manufacturing/materials sciences and AI/data sciences in this nascent, exciting area.

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1. Introduction

Discovery and development of new materials and manufacturing process-recipes for societal applications has been a constant human endeavor, almost since the ancient times (Sass, 1998; Miodownik, 2014; Wellmann, 2021). Although the discoveries of many historic materials and manufacturing processes were attributed to serendipity, materials discovery has become a goal-driven pursuit in the modern era. More recently, goal-oriented discovery and development of materials and processes have assumed an even greater impetus, especially towards addressing the following three important needs. Foremost, the industrial performance envelope and global competitiveness landscape, such as, in hypersonic systems and energy storage fields, are becoming increasingly dependent on new high-performance materials and their processing (Correa-Baena *et al.*, 2018). Second, the growing climate and sustainability challenges are fueling the innovations of novel green products and process technologies that can reduce total energy and carbon footprint (Tabor *et al.*, 2018; Zimmerman *et al.*, 2020). Additionally, materials and process discovery has been sought to address the growing shortage of critical materials, especially for the new energy and strategic applications (National Research Council, 2008; Chu, 2011; National Academies of Sciences, 2019).

A goal-oriented discovery of materials and manufacturing process-recipes requires the identification of the material composition and the manufacturing process history with which a component (i.e., a material sample or a product) is realized. A material-discovery goal is specified in terms of the structural features of the material spread over multiple spatial scales (e.g., morphology and microstructure) and/or the *desired* properties (e.g., a specific thermomechanical response characteristic of a shape-memory alloy) (Olson, 1997; Panchal *et al.*, 2013; Jha *et al.*, 2015). Discovering a material involves conducting a series of time-consuming and costly physical and/or computational experiments to search a high-dimensional Materials Design Space (MDS). The MDS spans the possible material compositions and processing condition-combinations under which a material can be synthesized. Each experiment consists of controlled synthesis of material samples with particular composition(s) under a certain process history, and subsequent determination of the structure, constitution, and/or properties of the realized material. Such a manual and iterative process of materials discovery and manufacturing, leading to their deployment takes place over multiple years, often extending beyond two or more decades.

Consequently, accelerated materials development has emerged as a major international imperative (National Research Council, 2008; Holdren, 2011; Jarvis, 2012; Schmitz and Prahl, 2014; Department of Energy, 2018; Hong *et al.* 2021). The development of effective strategies to integrate simulation and experimental data with expert knowledge on materials design is an active area of research (Drosback, 2014; Kalidindi and Graef, 2015; Agrawal and Choudhary, 2016). This problem of accelerating materials discovery is considered as one of conducting

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iterative physical and/or simulation experiments to search the MDS by striking a balance between *exploitation,* i.e., opportunistically and perhaps locally improving the search solutions, versus *exploration,* i.e., searching in the MDS regions that are "different" and have not been considered before. (Lookman, Alexander and Bishop, 2016; Lookman, Balachandran, Xue, Pilania, Shearman, Theiler, Gubernatis, Hogden, Barros, and BenNaim, 2016; Xue *et al.*, 2016) Olson (Olson 1997) made a pioneering attempt to towards accelerating materials discovery by combining simulations with experiments to develop quantitative mapping connecting the Process–Structure Properties (PSPs) of a material. Following this, different methods that incorporate high-throughput experimentation and computation have been deployed within conventional, human-centric, materials development frameworks (Glamm *et al.*, 2015; Luo, 2015; Robinson, 2015; Howe *et al.*, 2016). These high-throughput experimental (Chang *et al.*, 1998; Potyrailo *et al.*, 2011) and computational (Yang *et al.,* 2012; Curtarolo *et al.,* 2013) approaches have severe limitations for Autonomous Materials Discovery and Manufacturing (AMDM). First, the experimentation process to search the MDS, i.e., determining the settings to perform the experiments, is driven manually, employing human intuition and prior knowledge, or an *a priori* developed computational simulation or simpler statistical surrogate models. Such a search process is incapable of capturing the high-dimensional MDS (spanning all possible compositions, configurations, and microstructures) and complex causal relationships (e.g., multi-physics) underlying the MDS. Therefore, the search process invariably leads to a poor coverage of MDS and suboptimal results (Panchal *et al.*, 2013). Second, hardcoded workflows are typically employed to search the MDS, i.e., the choices of the experimental process chain and the testing methods employed are set *a priori.* They lack flexibility to iteratively learn and adapt based on the streaming measurements as well as knowledge acquired to assure balanced exploitation versus exploration of the MDS. Third, the search strategies preclude *expansion* of the MDS–i.e., increasing the dimensionality of the MDS based on mining new knowledge from literature and human inputs, preferentially searching in regions likely to contain superior material design solutions. An expansion can translate to the identification of alternative material components and extra processing steps as new information is received. Fourth, the current methods involve humans for routine analysis and decision making in the so-called human-in-control mode, limiting the repeatability, throughput rate, and autonomous capture of expert knowledge (Baker, 2010). Additionally, the MDS search strategies based on these methods tend to be suboptimal in resource allocation, as experimental decisions do not correctly assess the cost and the time duration of experimentation.

What manufacturing and materials sciences need instead are systems capable of efficiently navigating complex, multi-dimensional MDS. Recently, autonomous research systems have evinced significant interest across various scientific and engineering domains (Sparkes *et al.*, 2010; Chandran *et al.*, 2011; Graham *et al.*, 2013; Glauche *et al.*, 2017). Notably, engineering domains such as autonomous transportation and smart grids have reported some success in imbibing the two key elements of autonomy, namely, learning and adaptation as part of the real-time decision processes (Åström and Kumar, 2014). However, AMDM remains at a nascent stage (Nikolaev *et al.*, 2016). This is possibly due to challenges pertaining to the complex hierarchical nature of materials microstructure and behavior (Fasolka and Amis, 2007), a strong dependence of microstructure (and resulting properties) on the manufacturing process chain, as well as the elevated costs of experiments and simulations to realize sufficiently precise models, besides the challenges as outlined earlier.

Pertaining to the traditional human-driven materials discovery process, a common understanding is that a prospective discoverer should possess a "mental model" (or a representation) of the relationships connecting how process settings with material morphology and microstructure, and properties. Such PSP relationships constitute the central representation frame for materials discovery and manufacturing. A PSP relationship-based mental model is typically acquired via 5+ years of specialized training and experiences. Any materials discovery system, manual or automated, that does not encode PSP relationships within its learning framework will be very limited in its ability to predict the structure, and hence, the behaviors of the manufactured material components or samples. Also, as stated earlier, purely manual and automated approaches to materials discovery are impractical, due to the complexity and high dimensionality of the MDS, as well as the limitations of humans to consistently carryout elaborate repetitive tasks.

Although all materials discovery systems involve sequential experimentation, autonomous systems for materials discovery and manufacturing fundamentally differ from purely automated systems in that they incorporate at least two extra elements, namely, *learning*, i.e., the mental model of the experimentation system is regularly updated based on the new data, observations and evidences, and *adaptation*, i.e., the workflow of the MDS search process, as well as the experimentation procedures, can change based on the observations and updated mental models (Åström and Kumar, 2014). Besides these two elements, these systems can incorporate higher levels of autonomy such as cognition and reasoning, as pointed out in an National Academy of Engineering study (NAE, 2004).

Recent attempts towards developing autonomous materials discovery systems employ AI/machine learning methods to derive these mental models (Raccuglia *et al.*, 2016; Saal *et al.*, 2020; Suh *et al.,* 2020). These models are predominantly derived based on supervised learning. Due to the high costs of experiments in manufacturing and material sciences, conventional supervised learning would not be feasible. Alternatively, active learning methods have been attempted to provide an optimal balance between exploitation to rapidly hone in on the most promising regions of the MDS, and exploration to adequately cover the MDS and search unusual designs during sequential experimentation (Balachandran *et al.*, 2016). These active learning methods are at the heart of the emerging AMDM systems.

Pertinently, the incorporation of PSP relationships as part of sequential experimentation and active learning

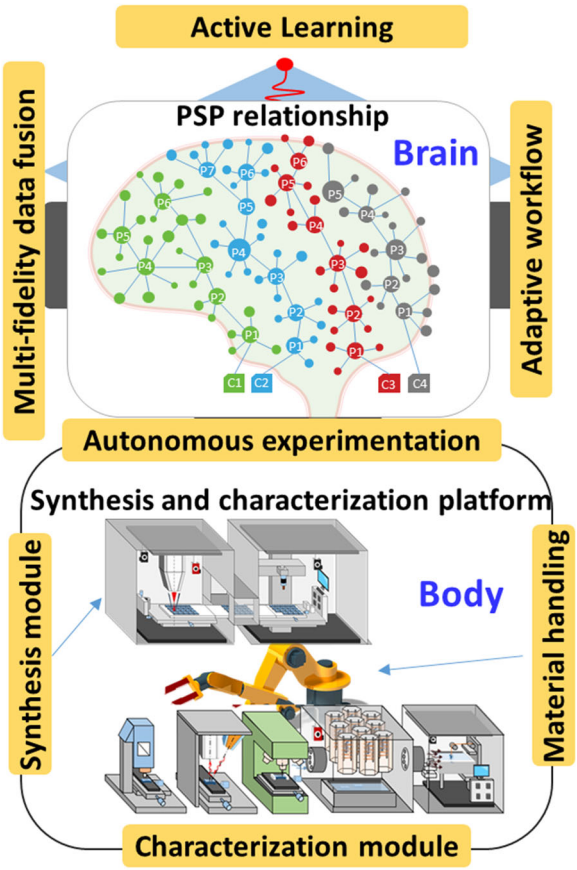

Figure 1. Schematic of an AMDM system consisting of a software brain and a hardware body.

frameworks is a key challenge that needs to be addressed in order to make autonomous materials discovery a reality. This aspect receives a particular attention in this review. Additionally, AMDM is an interdisciplinary challenge that connects materials and manufacturing sciences with AI/data sciences. Although AMDM systems are still in their infancy, multiple international efforts are underway to address the discovery of a variety of materials employing diverse AI/ML methods and synthesis platforms. This article, from this context, presents an overview of this growing volume of work as relevant to the manufacturing and data science/AI communities in general, and those within the Industrial and Systems Engineering and Operations Research (ISE/OR) professions in particular. Also, recent reviews on materials discovery have focused almost exclusively on the AI and machine learning efforts (Correa-Baena *et al.*, 2018; Saal *et al.*, 2020; Juan *et al.*, 2021), and sequential experimentation methods (Dehghannasiri *et al.*, 2017; Talapatra *et al.*, 2018a; Talapatra *et al.*, 2019). The current efforts towards closed-loop material synthesis platforms (Kusne *et al.*, 2020) largely overlook the critical challenges associated with materials discovery and manufacturing. Very few, if any, efforts have reviewed the various materials synthesis and experimentation platforms, together with their integration with AI and sequential experimentation methods for autonomous materials discovery.

Comparatively, this article brings a joint perspective that combines the review of experimentation platforms and manufacturing machine tools alongside the advances in the pertinent AI approaches. As such, it also aims to bridge the terminology across the relevant, well-established domains, and delineates challenges that can be of specific interest to the ISE/OR community. The remainder of this article is organized as follows. Section 2 provides an overview and a perspective of an AMDM system. Sections 3 and 4 present a review of the software "brain" and hardware "body" elements of an AMDM system. Section 5 presents a discussion of the state-of-the-art as well as possible future directions in this emerging area.

2. Overview of autonomous materials discovery and manufacturing systems

An AMDM system enables an iterative and adaptive execution of active learning of PSP relationships, as well as sequential experimentation to search an MDS through a closed loop interaction between a software (brain) and hardware (body) component (Figure 1), all with minimal human intervention. The software brain essentially consists of two modules. The first, *forward mapping module* aims to learn the PSP relationship based on fusing information from various data sources, and to predict the structure and the properties-of-interest of a material at a specified design and process setting (i.e., a point in the MDS). The data sources would include experimental observations and measurements of the material and the manufacturing process, simulation of multiple physical processes that capture the PSP relationships over multiple resolutions, as well as the domain expertise and experiential knowledge. Such multi-fidelity and multi-modal data sources are used for learning the PSP relationships. The second, *inverse mapping module* aims to prescribe the experimentation settings, and direct the search of MDS, based on the PSP relationship model, to discover a material and its manufacturing recipe. In effect, the software brain offers how to make the best predictions of the PSP relationship given the data, and at what settings to conduct the subsequent experiments (and thereby seek the next sets of data) towards achieving the desired materials discovery goal.

The hardware body is composed of two modules. A *synthesis module* aims to execute the brain signals (control input or a process recipe) autonomously to manufacture material sample(s) and adapt processing workflows based on the prior outcomes or observations, as with a reflex action. Additionally, a *characterization module* aims to probe the synthesized material, as well as measure and/or quantify the resulting properties (e.g., material hardness or thermal conductivity) and/or performance (e.g., the holistic indicators, such as energy efficiency, that depend on the entire structure of the manufactured component and the process, not just the material).

The execution of a process recipe in a synthesis module can be as simple as mixing the specified amounts of different chemical species in a test-tube, or as complicated as executing an elaborate chain of processes to transform the input precursors (e.g., raw materials and consumables) into

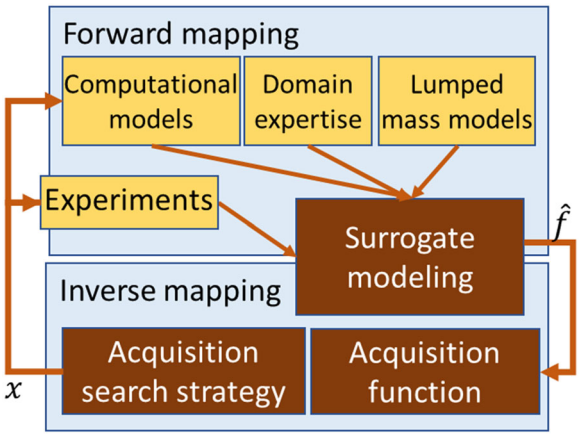

Figure 2. The overall structure of the brain executing iterative learning and experimentation process.

a product. Inherent to a synthesis module are multiple synthesis chambers (e.g., machine tools, reaction beds, test-tubes), material delivery and storage mechanisms to feed the desired raw materials into the synthesis chambers, and a material handling system to transport the material within and across the synthesis chambers.

The characterization module employs one or more measurement and testing stages to assess the properties and functionality of the synthesized material or part. The test stages aim to assess the property of interest, such as tensile strength, magnetic permeability and hardness of a synthesized functional alloy, or the energy conversion efficiency and rheology of a chemical synthesized in a test-tube or a petri dish.

It is pertinent to note that the various initiatives were launched in recent years have largely focused on autonomous materials discovery (National Research Council, 2008; Tabor *et al.*, 2018; Boyce and Uchic 2019; de Pablo *et al.* 2019; Gomes *et al.*, 2019; National Academies of Sciences, 2019). Several AI methods were developed to search the MDS, and successful initial experimental platform implementations have been reported (National Research Council, 2008; Mies *et al.*, 2016; Alberi *et al.*, 2018; Aspuru-Guzik and Persson, 2018; Tabor *et al.*, 2018; Talapatra *et al.*, 2018a; Yager, 2018; Boyce and Uchic, 2019; de Pablo *et al.* 2019; Gomes *et al.*, 2019; Stein and Gregoire, 2019), including for digital and topological material structures (Hiller and Lipson, 2009; Yan and Felser, 2017) . However, manufacturing process planning, i.e., deriving and possibly discovering the processing recipes to synthesize targeted materials in bulk and into a product, poses significant additional challenges (Aspuru-Guzik and Persson, 2018; Tabor *et al.*, 2018; National Academies of Sciences, 2019). Peculiar to manufacturing is that successful recipes require consideration of the entire processing chain; a mere specification of the "static" parameter settings would not suffice. In other words, the evolution of the process state variables in time, space, and across the multiple stages of the process chain need to be measured or estimated to correctly predict the structure.

Additionally, process discovery often results from clever, "out of the box" process improvisations as opposed to just optimizing the process parameters. Also, a flexible workflow for searching and discovering manufacturing recipes is somewhat like a game of chess where we observe the boards at every step (analogously, from measurements and physical understandings of a multi-stage process chain), and decide the moves (here, recipes from among a continuum of possibilities as opposed to finite combinations) to achieve a specified goal (functionality).

With recent advances in 3D printing, artificial intelligence (AI)/data science, and materials genomics, success in AMDM could open an exciting opportunity of material-on-demand manufacturing (Iquebal *et al.*, 2020a; Zhao *et al.*, 2020). Here, the material composition and microstructure, not just the shape and morphology of a 3D-printed component, can be controlled to the resolutions that can ensure a component's functionality. A new generation of smart machine tools are being considered to serve as reactor beds for autonomous, bulk-scale, on-demand manufacturing of novel materials (Botcha *et al.*, 2020). Such a capability opens interesting possibilities to manufacture components with specified functionality via innovative processing recipes, without relying on expensive and scarce (e.g., rare earth) materials—thus addressing the critical materials challenge noted by the National Academy of Sciences (National Research Council, 2008; National Academies of Sciences, 2019). Advances in AMDM can be discussed in terms of the advances in computational AI/data science models towards enabling autonomy and achieving a discovery (brain) as well as advances in the hardware platforms to execute flexible workflows for autonomous experimentation for synthesis and discovery (body).

3. The brain: Computational AI/ML approaches and multi-fidelity learning

The brain of an AMDM system mainly consists of two types of computational AI/ML models, constituting a forward and an inverse mapping loop, respectively (Figure 2). The forward mapping loop focuses on establishing the PSP relationship. It seeks answers to the question: "*Given a set of material descriptors that can be manipulated through materials synthesis and processing, what are the corresponding properties and performance characteristics?*" On the other hand, the MDS search necessitates inversion of this map (Olson, 1997; Olson, 2000). The inverse mapping loop seeks answers to the question: "*Given the desired property and performance requirements, what material composition and processing routes are feasible, and well-suited to realize structures that meet these requirements?*" The inverse mapping thus seeks to identify domains in the process–structure and structure–property spaces that meet specific property or performance targets. These targets are often expressed based on the desired functionality and behaviors, such as ductile-to-brittle transition characteristic of refractory high-entropy alloys to improve their machinability.

Altogether, the brain embeds a sequential experimentation and active learning framework. The forward mapping fuses data from experiments and other sources, such as simulations, domain expertise and historical conventions to

establish a quantitative PSP relationship. The first step is to derive the models to predict the process state and maps $\hat{f}$ from the data. Subsequently, the process state predictions are used to estimate the resulting material (micro)structure (i.e., P→S mapping) and then the properties (i.e., S→P mapping). Here, the representation of the process state and the material structure becomes an important consideration. Also, the forward mapping effort can provide efficient initial priors that would be progressively updated as part of a Bayesian framework-driven sequential experimentation or an active learning strategy for AMDM.

An inverse mapping aims to optimize the experimental effort to achieve a specified material discovery objective. The inverse mapping methods employ an acquisition function and an acquisition strategy. The acquisition function in effect ascribes a value earned from conducting an experiment or a sequence of experiments at the specified points x in the MDS based on the predictions from the forward models and/or their updates during a sequential experimentation or active learning. A numerical scheme is then devised to determine a search strategy that optimizes the acquisition function.

3.1. Forward mapping via advanced representations and a computational modeling

PSP relationship maps provide a systematic framework to capture the material–manufacturing process interactions (Arróyave and McDowell, 2019; Ramakrishna *et al.*, 2019). Any materials discovery system that does not encode PSP relationships within its learning framework will be very limited in its ability to discern the underlying causal pathways and manufacturing recipes to realize targeted materials response and properties. Although significant efforts have been made towards developing structure-to-property (S→P) mapping, the process-to-structure (P→S) mapping efforts are relatively less developed.

S→P mapping: In the last two decades significant advancements have been made in computational models and simulations towards predicting the properties of synthesized materials by employing quantum chemical calculations that consider a material's electronic structure (Drosback, 2014; Kirklin *et al.*, 2015, Alberi *et al.* 2018). These efforts have led to fast, *ab initio,* first-principles density functional theory (DFT) implementation framework efforts such as AFLOW, MPDS, and JARVIS-DFT projects (Kirklin *et al.*, 2015; de Pablo *et al.,* 2019). However, these implementations are often computationally expensive, even for estimating simple mechanical properties and material behaviors over realistic scales (Narulkar *et al.*, 2008; Cheng *et al.*, 2012). They also pose significant issues to capture the effects of processing on the material structures (Mansouri Tehrani *et al.*, 2018). The potential functions and correlation functionals employed in these models do not capture the real-world behavior (e.g., elastic response) and can lead to aphysical outcomes (Raff *et al.*, 2012).

Towards addressing these limitations, various surrogate models derived based on machine learning approaches have been employed. These surrogate models can (a) be incorporated within a computational simulation model, such as for deriving more realistic interatomic potentials for molecular dynamics simulations (Raff *et al.*, 2012); (b) serve as a faster, possibly physics-informed, surrogate for an intensive computational model (Pukrittayakamee *et al.*, 2009, Correa-Baena *et al.,* 2018); or (c) fuse multiple computational simulation and experimental measurements, each possibly of a different fidelity and resolution, to enhance prediction accuracies (Patra *et al.*, 2020; Greenaway and Jelfs, 2021; Revi *et al.*, 2021). Saal *et al.* (2020) note that 70 machine learning surrogate models were reported in the literature for materials discovery. In parallel, efforts have also been made to integrate simulations and experimental data with expert knowledge (Kalidindi and Graef, 2015; Agrawal and Choudhary, 2016; Ramakrishna *et al.*, 2019). This active research area has led to the deployment of these integrated models (e.g., Talapatra *et al.*, 2018a; Talapatra *et al.*, 2018b; Talapatra *et al.*, 2019; Talapatra *et al.*, 2021) within conventional, human-centric, materials discovery and manufacturing platforms, some of which are reviewed in Section 4.

Data and material structure representation: Towards ensuring consistency and computational tractability of the PSP mappings, some prior efforts have addressed the derivation of a proper representation of the performance, properties, and the material structure. Representation of the performance is relatively simpler compared with the representation of a material structure or a process. The measurements from various instruments and test platforms, or the estimates derived from the measurements are used to represent the property or the performance.

In contrast, representations of a material structure or the process states are nontrivial. A structure representation should capture the spatial patterns (and chemistry) to the resolutions that inform the properties (e.g., hardness and machinability), and are influenced by the processing conditions (e.g., laser power, scan speed). The contemporary representations are largely statistical (e.g., n-point correlation functions), or simple geometric descriptors (e.g., columnar dendritic, equiaxial grains) (Parvinian *et al.*, 2020). For example, microstructure-sensitive design discussed in Fast *et al.* (2008) and Fullwood *et al.* (2010) employs efficient low-dimensional mathematical representation of microstructures, and uses homogenization techniques to relate microstructure to property/performance. Two major categories of AI/data science methods have been applied to derive representations based on the high-dimensional data in the materials synthesis and manufacturing contexts. A vast majority of these methods are based on supervised learning models that relate images and other signals with the physical material structure (e.g., phase composition) or property (e.g., hardness) (Lee and Yoo, 2008; Yang and Choe, 2010; Le Guen and Paul, 2014; Zhang *et al.*, 2015). The heterogeneity of the data sets, as well as the variable sizes and rates at which different data elements are realized pose significant challenges to the representation and learning of underlying variables and relationships. Systematic methods are necessary to fuse the diverse multimodal data streams for representation and

learning (Beyca *et al.*, 2015; Gahrooei *et al.*, 2021). An extensive treatment of how to handle various kinds of images, both snapshot as well as dynamic, can be found in a research monograph (Park and Ding, 2021). Recently, efforts have been made to classify and annotate 20,000 Scanning Electron Microscopy (SEM) images of various material structures (Aversa *et al.*, 2018). However, the costs to label large and diverse data sets are prohibitively high. They are somewhat unwieldy for AMDM applications where previously unknown and unlabeled structures can occur. Unsupervised learning methods, such as dynamic time warping (Baumes *et al.*, 2008), shape and textural features (Egelandsdal *et al.*, 1999; Xu *et al.*, 2015), and deep learning (de Vos *et al.*, 2019; Jung *et al.*, 2020) may be attempted to derive low-dimensional representations of the material structures.

P→S mapping: Prior efforts have attempted three major types of models to capture the process—material (structure) interactions. The first type, *computational models* are used to capture the effects of spatiotemporal distribution of process variables such as temperature and strain on the morphology and microstructure of a manufactured material sample (Karayagiz *et al.*, 2019; Karayagiz *et al.*, 2020; Yavari, 2020). Of recent interest is to develop computational models to determine the thermo-mechanical history in additive manufacturing processes for material synthesis under different process parameter settings (e.g., laser power, scan raster pattern, scan speed), and relate these to the morphology (e.g., voids, pores) and the microstructure (Tan *et al.*, 2020). The thermomechanical models predominantly employ finite-element or finite-difference schemes (Karayagiz *et al.*, 2019), or their refinements to simulate the heat flow or the thermal history without or with mass flow considerations (Tan *et al.*, 2020). An additional (tandem) computational model which uses the solution from a thermomechanical model (e.g., the thermal history) is employed to predict the microstructure. The microstructure prediction models have been predominantly based on one of phase-field (Karayagiz *et al.*, 2020), cellular automata (Liu and Shin, 2020; Teferra and Rowenhorst, 2021), and kinetic Monte Carlo (Mishra and DebRoy, 2004; Rodgers *et al.*, 2017) (listed in the order of decreasing computing overhead). All these computational models invariably require the use of high-performance computing environments. Also, the sharp thermo-mechanical gradients, such as those prevalent near the laser source in an additive manufacturing process, or at the shear localization regions in machining make the microstructure predictions vulnerable to the settings of the numerical scheme employed to solve the models.

The second, *lumped systems analytical models* aim to capture a specific, narrow aspect of a process multi-physics (Noori-Khajavi and Komanduri, 1993; Pratt and Nayfeh, 1999; Davies and Burns, 2001; Bukkapatnam and Clark, 2006), and relate certain process signatures extracted based on these models to specific material structures (Iquebal *et al.*, 2020a, Zhao *et al.*, 2020). Since 1990, significant efforts have been made in sensor fusion methods to capture the dynamics underlying manufacturing processes (Emel and Kannatey-Asibu, 1989; Dornfeld and DeVries, 1990; Azouzi and Guillot, 1997). A majority of the seminal methods have aimed to establish a static mapping between the sensor data (features) and the properties or the performance. In the past 20 years, research groups have attempted to derive reduced-order dynamic models that are often referred to as Process–Machine Interaction (PMI) models, to track the process state evolution. These models are based on combining dynamic systems principles with sensor fusion techniques (Bukkapatnam and Clark, 2006; Rao *et al.*, 2014; Beyca *et al.*, 2015). These low-dimensional analytical models can capture the active degrees of freedom in a process while ensuring a certain degree of consistency with the underlying physical phenomena. Attempts have also been made to capture the transient and nonlinear dynamics innate to manufacturing processes by employing nonparametric statistical modeling and network science methods (Wang and Bukkapatnam, 2018; Iquebal *et al.*, 2020a, b). Lumped mass models offer a promising direction for microstructure predictions. However, they currently remain at a nascent stage and offer significant potential for future research.

The third, *statistical surrogate models* seek to map the process settings to the structure, property, and performance. Representing individual linkages along a PSP chain through parametric or nonparametric statistical surrogate models is of recent interest. In particular, Bayesian modeling methods have been employed to learn PSP relationship towards significantly accelerating materials design (Ghoreishi *et al.*, 2018; Ghoreishi *et al.*, 2019; Talapatra *et al.*, 2019; Couperthwaite *et al.* 2020; Khatamsaz *et al.*, 2020). However, current process–structure mapping methods are insufficient for materials discovery and manufacturing. These methods make a strong (often unrealistic) assumption that the process evolution can be determined by the initial parameter settings and ignore the dynamic nature of the manufacturing processes.

Perspectives: Evidently, PSP representations are essential to incorporate materials scientist-like reasoning into materials discovery and material-on-demand manufacturing systems. Successful materials discovery systems must make full use of the PSP relationships *chains*. However, the current approaches take a rather static view, and overlook the dynamic aspect of the PSP relationships (Nikolaev *et al.*, 2014), i.e., *how properties of materials are determined by the underlying (micro)structural features, and how the features, in turn, are ultimately controlled by pursuing a specified thermodynamic pathway through a process chain*. Significant information loss occurs while capturing the solutions generated through the computational simulations and experimental observations using static surrogate models, especially to represent the manufacturing processes (Botcha *et al.*, 2021). Towards this end, it is important to reconcile the recent significant advances in computational materials science (Honarmandi and Arróyave, 2020) with sensor fusion and uncertainty quantification methods to provide a dynamic, as opposed to the current, largely static representations of process conditions (state) for PSP mapping. Such an approach can capture the thermodynamic pathways (i.e., trajectories)

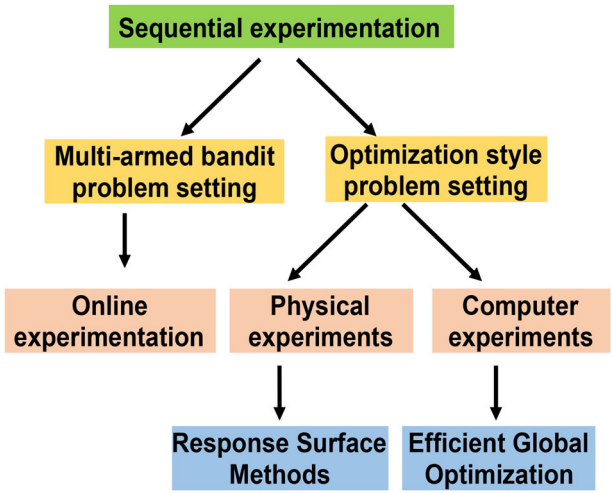

Figure 3. Major branches of sequential experimentation.

that a material undergoes during a process chain. Tracking the pathway leads to a more consistent PSP mapping as microstructure represents the end state of this trajectory. It will also offer the framework to learn the phase changes especially the influence of the composition, time and temperature variations on the material phases the microstructure, and hence, the properties.

3.2. Inverse mapping via sequential experimentation and active learning

A systematic, statistical framework for Design Of Experiments (DOE) was pioneered by R. A. Fisher (Fisher, 1949), with applications mainly in the biology and agriculture areas. Subsequently, George Box, Jeff Wu, and others, extended the DOE methodologies and popularized the methods to other applications and industry sectors (Box *et al.*, 2005; Wu and Hamada, 2011). In due course, researchers have realized the importance of sequential experiments, as it is untenable to understand a complex system fully through one time "batch" of experiments, *even in the high-throughput execution of parallel experiments*. The modern sequential experiment principles can be traced back to two possible moorings, one based on the Response Surface Methodology (RSM) by Box and Wilson (Box and Wilson, 1951), and the other on the sequential analysis work by Wald (Wald, 1947) (see Figure 3). In fact, in their formative years, sequential experiments were attempted to mimic the discovery process as practiced by a human scientist.

The first branch has its roots in framing the experimental design problem as an optimization task, in which the experimental platform (physical or simulation) itself serves as the black-box function to optimize (Myers *et al.*, 2016). Since experiments are costly, sequential experimentation approaches that minimize the number of evaluations are preferred. One classic approach is the RSM of Box and Wilson (Box and Wilson, 1951). It uses at most second-order polynomial models to capture the underlying relationships. As such, these models have rather limited capability to represent and learn the high-dimensional response surfaces of a MDS from simulations and experimental data as part of autonomous materials discovery.

The introduction of Gaussian Process Regression (GPR) in spatial statistics (Cressie, 1991) and their subsequent adoption to the model the relationships from computer experiments (Sacks *et al.*, 1989, Santner *et al.*, 2013) brought a paradigm shift. Gaussian process models are nonparametric in nature, and they provide a great degree of flexibility and adaptivity in modeling complex response surfaces. Subsequently, GPR has emerged as a popular general-purpose method for broad machine learning applications (Williams and Rasmussen, 2006). A GPR model offers flexibility, numerical stability, and the capability for uncertainty quantification (Matheron, 1963; McKay *et al.*, 1979;, Shewry and Wynn, 1987; Sacks *et al.* 1989; Fang *et al.*, 2005; Williams and Rasmussen, 2006; Santner *et al.*, 2013; Molina *et al.*, 2015; Sun *et al.*, 2019). More pertinently, GPR models with Bayesian Optimization (BO) techniques enable simultaneous learning of the complex functional relationship as well as searching for the best design/process (input) settings (Mockus, 2012) employing a sequential experimentation strategy. Every BO algorithm defines a sequential sampling procedure, which successively generates new input points, based on optimizing an acquisition function—sort of an objective function that quantifies MDS search and materials discovery goal—by employing a specified acquisition search strategy as noted earlier.

Jones *et al.* (1998) introduced a sequential experimentation strategy popularly referred to as Efficient Global Optimization (EGO) by leveraging GPR. As with other BO methods, the next sampled point(s) in EGO is also decided by optimizing an acquisition function by executing an acquisition or a search strategy. They also proposed an acquisition function, known as *Expected Improvement* (EI) to guide where to sample the next data point, as part of EGO. Unlike a simple *Maximum Mean* (MM) value criterion that is adopted to find the maximal or minimal response, the EI criterion tries to balance between sampling the next data point with the highest expected value, usually within a specified local region (exploitation), versus sampling the point with the highest uncertainty (exploration). The success of EGO in many applications is attributed to the ability of EI to balance between exploitation and exploration.

Implementations of a basic EGO method involve the use of a space-filling design (Santner *et al.*, 2013) to perform an initial sampling of the design space, GPR for learning using these samples and predict the response of the system, and EI as an acquisition function to balance exploitation and exploration of the MDS. Such a BO approach has been employed widely for hyperparameter tuning, combinatorial optimization and reinforcement learning (Greenhill *et al.*, 2020).

Over the past decade, several variants of EGO have been developed by advancing the BO principles (MacKay, 1992; Cohn *et al.*, 1996; Chan *et al.*, 2010). This is because EI, as a traditional BO acquisition function, is criticized for over-exploiting the fitted model and under-exploring the design space (Bull, 2011; Chen *et al.*, 2019). Balancing exploration

and exploitation is an important step for reaching the global optimum of a continuous function or approximating the whole response surface using limited samples. The "balance" should nonetheless be driven by the search or discovery goal. Such goals can be one of the three kinds: (a) improve a possibly multivariate response, as in the hysteresis characteristic of a shape memory alloy material, or machinability of a refractory high-entropy alloy (exploitation); (b) increase the knowledge of the P→S or S→P relationship connecting the input design parameters of the MDS and the manufacturing recipe, the material structure and the properties (exploration); or (c) simultaneously understanding the underlying space while improving the response (balance).

Multiple works have investigated the achievement of this balance during a sequential design (Bull, 2011; Qin *et al.*, 2017; Chen *et al.*, 2019; De Ath *et al.*, 2021). Improvements to EI (Mockus *et al.*, 1978; Jones *et al.*, 1998; Huang *et al.*, 2006; Picheny *et al.*, 2013), as well as alternative acquisition functions, such as the Probability of Improvement (PI) (Kushner, 1964), Gaussian process Upper Confidence Bound (UCB) (Srinivas *et al.*, 2009; Azimi *et al.*, 2010; Contal *et al.*, 2013; Desautels *et al.*, 2014), Predictive Entropy (PE) of search (Hernández-Lobato *et al.*, 2014), Entropy Search Portfolio (ESP) (Shahriari *et al.*, 2014), and Knowledge Gradient (KG) (Scott *et al.*, 2011; Wu and Frazier, 2016; Wu *et al.*, 2017) were considered. Among these, the MM and EI criteria are typically employed when the goal is to exclusively improve the response (both under a local and a global sense). Information theoretic criteria including the Shannon entropy, KL-Divergence, PE and ESP are employed to enhance exploration of the input space and reduce the risk of the MDS search process from yield suboptimal local optima. Criteria such as PI and UCB, as well as a few EI improvements (e.g., ε-EI) have been suggested when the goal is to learn the input space while improving the response.

The second branch, starting with Wald's sequential analysis (Wald, 1947), uses random, sequential sampling from an unknown distribution with the sample size and constitution decided on-the-fly based on prior observations. This line of research led to a Multi-Armed Bandit (MAB) framework (Burtini *et al.*, 2015), in which an agent (software entity) seeks to acquire new knowledge (*exploration*) and optimize decisions––i.e., how to carry out new experiments––based on the existing knowledge (*exploitation*). The agent balances these tasks by maximizing the expected value of information in the sequence by choosing the optimal experiment set. Many MAB problems are formulated into a reinforcement learning setup where such trade-off between exploration and exploitation can be quantifiably stated (Besbes *et al.*, 2019). Reinforcement learning-based approaches are beginning to be applied for optimizing the molecular structures for nanomaterials (Whitelam and Tamblyn, 2020) and certain chemical reactions (Zhou *et al.*, 2017; Zhou *et al.*, 2018). Unlike MAB or those problems to which reinforcement learning is typically considered, autonomous experimentation platforms are constrained by limited sampling resources (Lovell *et al.*, 2011), rendering the basic versions of MAB and reinforcement learning approaches not well-suited. Newer classes of reinforcement learning methods and algorithms are beginning to be investigated for high-dimensional design spaces and sparse data (Hao *et al.*, 2021).

Interestingly, the machine learning community refers to the sequential data sampling problem as active learning (Settles, 2009). In actuality, active learning and sequential experiments have profound connections and they often aim to address almost identical problems. Although sequential experimentation and active learning methods have garnered considerable attention in recent years for advanced manufacturing and materials synthesis/discovery, few of the current approaches target the central challenges in this domain (Burtini *et al.*, 2015; Wu *et al.*, 2017; Butler *et al.*, 2018; Besbes *et al.*, 2019; Castillo *et al.*, 2019; Kalidindi, 2020; Whitelam and Tamblyn, 2020; Zhang *et al.*, 2020). Nonetheless, a major boost to enhancing the autonomy as well as the performance of a materials discovery system would result from developments in the sequential experimentation and active learning approaches.

Perspectives: Prior sequential experimentation methods rely more on exploitation and less on exploration of design spaces and can easily be trapped in local optima. Most critically, they are not autonomous. Instead, they rely heavily on human experts to steer the direction of the experimental sequence. A materials scientists' success hinges on harnessing the intuition and expertise to adaptively fuse diverse data sources, including prior knowledge, materials theories, experimental observations (which often include surprises), and simulations; autonomously generate, pivot, and test parallel and competing hypotheses; execute complex experiments to discover the materials composition and manufacturing recipe to meet or enhance the functionality. Recent developments in active learning promise a totally different approach to impart such an enhanced autonomy. Here, the learner optimally seeks out experiments in a way that balances the exploration and exploitation of experimental spaces.

A key hallmark of many historical scientific discoveries has been an element of *surprise* that preceded a discovery by a scientist. A surprise is noted whenever an outcome defies what's considered the realm of possibilities. The ability to discern the surprise based on fusing information from multiple data sources and models, and thereby pivot to newer hypotheses is an innate hallmark of human discoverers. It is therefore important to endow an autonomous materials discovery system with the capability to be surprised and to adapt the MDS search process based on the surprise. Although alternative definitions of a surprise exist, research into adopting these for materials discovery can pose interesting challenges in terms of how the various quantitative formulations of a surprise affect the exploration–exploitation balance (Ahmed *et al.*, 2021). Additionally, while reacting to a surprise, one may need to not just exploit and explore but expand the MDS itself (think in terms of the MDS dimensions). Such an expansion should draw from the copious unstructured domain knowledge and intuition available in the literature, as well as among the domain experts.

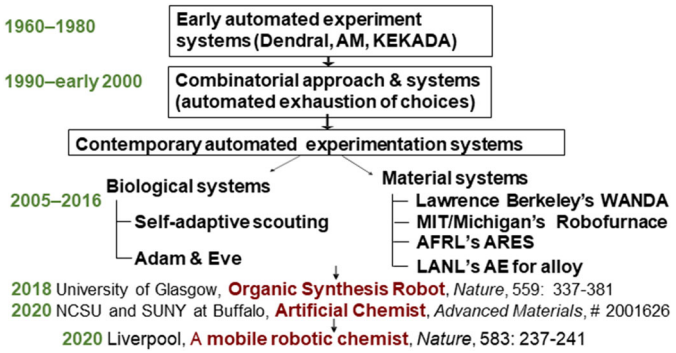

Figure 4. Evolution of autonomous experimentation setups.

Approaches to combine such unstructured and abstract domain knowledge can enhance the level of autonomy and the performance of materials discovery systems.

4. The body: Autonomous discovery platforms

The body of an AMDM system mainly consists of hardware platforms comprising: (a) one or more manufacturing and material handling equipment to process and transport material precursors through a process chain to synthesize materials; and (b) multiple sensors and measurement instruments to characterize the materials and estimate properties. The hardware platforms should support the execution of flexible workflow to synthesize materials per the direction of the brain. More specifically, the hardware platforms should be capable of synthesizing materials at conditions that amount to a comprehensive coverage of the MDS, and they should accommodate possible expansion of the MDS. Additionally, they should provide the data needed in a timely fashion, and to the desired resolution to update the models in the brain. The state-of-the-art and the emerging trends in these components are detailed in the following subsections.

4.1. Autonomous experimentation and material synthesis

The quest for autonomous experimental systems began in the 1960s and evolved, rather slowly, in the 1970s and 1980s (Ritchie and Hanna, 1984; Kulkarni and Simon, 1988). The early systems had few commonalities with the sequential experiment research (Figure 4). One of the first platforms was Dendral, developed in the 1960s (Lindsay *et al.*, 1993). Subsequently, Automated Mathematician (AM) (Ritchie and Hanna, 1984) and KEKADA (Kulkarni and Simon, 1988) platforms were developed in the 1980s. Sparkes *et al.* (2010) note they, "… *had limited background knowledge when compared to human scientists, and … needed more heuristics in order to continue its discoveries.*"

Researchers soon realized the drawbacks of heuristics to handle the ever-increasing complexity of design spaces (Lindsay *et al.*, 1993; Cassell *et al.*, 2001; Agrafiotis *et al.*, 2002; Noda *et al.*, 2005). Researchers instead resorted to brute-force enumerations of design solutions, employing automatic

Table 1. A summary of the recent autonomous materials discovery platforms.

Platform	AFRL-ARES	Alamos - Adaptive Design	Glasgow - Synthesis robot	NCSU&SUNY - Artificial chemist	Nature - Robotic chemist
Year	2016	2016	2018	2020	2020
Problem	Synthesize single-walled carbon nanotubes w/ targeted growth rate	Search for SMAs with targeted hysteresis properties	Predict the reactivity of organic reagents via a liquid handling robot	Synthesize quantum dots with targeted peak emission energy	Discover bioderived scavenger with a maximizing hydrogen evoluted
Experimental batch size	100	4	6	1	1
Input	continuous	continuous	binary	continuous	continuous
Output	continuous	continuous	binary	3-dim continuous	continuous
Surrogate Model	Random Forest	SVR_{rbf} out of {GP, SVR_{rbf}, SVR_{lin}}	1. SVM 2. NN	NN with Adaboost	GP_{matern}
Acquisition strategy	Genetic Algorithm (objective function)	KG out of {EI, KG, greedy}	1. LDA score 2. Greedy	UCB out of {EPLT, MV, EI, UCB}	UCB

machines such as computer clusters. The next wave of systems focused on specifying large parameter-combinations for automated gene sequencing (Agrafiotis *et al.*, 2002) and combinatorial search, particularly to discover and synthesize nanomaterials (Cassell *et al.*, 2001; Noda *et al.*, 2005).

Ever since, automated experimentation systems have become more sophisticated. Robofurnace developed by the MIT/Michigan group (Oliver *et al.*, 2013) is a semi-automated system for nanomaterial synthesis. Lawrence Berkeley Lab developed WANDA (Chan *et al.*, 2010) to enable high-throughput synthesis of colloidal nanocrystals.

After 2005, advancements in data science and ML methods started to influence the automatic or autonomous material experimentation systems, especially for reducing the number of experiments and for making the search process more efficient and effective. These systems have begun to include data science models that are built based on experimental data and, in some instances, these models could be updated on-the-fly as experiments and data collection are carried out sequentially (Chan *et al.*, 2010; Oliver *et al.* 2013; Nikolaev *et al.*, 2014; Lookman, Alexander and Rajan, 2016; Nikolaev *et al.*, 2016; Xue *et al.*, 2016).

Table 1 summarizes some of the prominent autonomous discovery platforms developed in recent years. Nikolaev *et al.* (2014) at AFRL developed an automated experimentation platform for the discovery of carbon nanotubes. Subsequently, they demonstrated a first of its kind, Autonomous Research Experiment System (ARES) for materials discovery (Nikolaev *et al.*, 2016). Contemporaneously, Lookman's group proposed an adaptive design framework for materials discovery and applied the framework to address a demonstration challenge of discovering shape memory alloys with specified hysteresis characteristic (Balachandran *et al.*, 2016; Lookman, Alexander and Bishop, 2016, Lookman, Balachandran, Xue, Pilania, Shearman, Theiler, Gubernatis, Hogden, Barros, and BenNaim, 2016; Xue *et al.*, 2016).

The efforts in the past 4 years have seen further advancements both in terms of incorporating the elements of sequential experimentation and workflow adaptation into the materials discovery loops, as well as in the use of robotic systems to implement these flexible experimental workflows. In 2018, Granda *et al.* (2018) sought to map out the chemical reaction space to predict the reactivity of various reagent pairs based on learning from a small number of experiments. The experiments were conducted by a wet-lab robot experimenter and the reactivity was assessed in real-time using nuclear magnetic resonance and infrared spectroscopy. In 2020, Epps *et al.* (Epps *et al.*, 2020) pursued an on-demand synthesis of quantum dots with specified peak emission energies. As part of this effort, they investigated a few contemporary surrogate models to learn the underlying MDS. In 2021, Burger *et al.* (Burger *et al.*, 2020) developed a *robotic chemist* that operated without human intervention for 8 days, performing some 688 experiments to discover photocatalysts for hydrogen production from water by learning the underlying MDS and the PSP relationships.

These recent efforts have garnered particular attention, and some of these efforts were published in high-profile journals like *Nature* (Granda *et al.*, 2018; Burger *et al.*, 2020; Epps *et al.*, 2020). However, the data science and machine learning methods employed for deriving surrogate models in these systems are mostly well-established, and their application is rather straightforward. Advanced machine learning approaches that can have a transformative potential for both forward and inverse mapping are yet to be fully explored, if at all. Among the multiple acquisition functions that have been considered, the UCB, which aims to promote more exploration of the MDS space compared with EI, seemed to perform well in terms of mapping out the underlying relationships. Nonetheless, most of these implementations use a rather low-dimensional input space (i.e., MDS), with at most a few tens of dimensions.

Perspectives: Although the aforementioned systems are important steps towards autonomous experimentation, they fall short of the needs of autonomous materials discovery, and materials-on-demand manufacturing. The complexity and costs in running experiments, learning the forward and inverse mappings, as well as conducting the MDS search increase drastically, as researchers move from wet-chemistry to materials synthesis, and ultimately to their bulk-scale manufacturing. Even the successful prototypes, including of a robotic chemist, are still far distant from making autonomous discovery and materials-on-demand manufacturing a reality, because of the following reasons. First, many of them are at best semi-automatic. Their human-in-the-loop framework *runs counter to what an autonomous system should be*, since humans are not adept at navigating complex, multi-dimensional manufacturing process spaces. Second, they use methods that are rather mature and well-evaluated in the data science communities. However, they are known to be easily entrapped in local optima when searching in a design space that embeds a complicated manifold. Also, none of the previous systems can adaptively expand and learn MDS by fusing measurements, computational models, and experiential knowledge innate to manufacturing practice. Despite these limitations, these early experimentation platform implementations offer significant scope to create truly autonomous platforms for materials discovery and manufacturing. Another, more practical issue needs to be considered while leveraging manufacturing machine tools and other automated systems for materials synthesis. The architectures of most of the controllers employed in materials synthesis and manufacturing platforms are often *closed*. In effect, they may not lend themselves to autonomously receive, let alone execute the instructions and recipes from the "brain." Although workarounds exist in the form of open platform communication modes, synthesis platforms with an open architecture controller or a more ground up-developed synthesis platforms should be preferred for AMDM.

4.2. Rapid characterization of material microstructure and property

Although AMDM has been noted as an exciting grand challenge (National Research Council, 2008; Tabor *et al.*, 2018;

de Pablo *et al.*, 2019; Gomes *et al.* 2019; National Academies of Sciences 2019), and several significant international research efforts have been launched in the last 3 years (Yager, 2018; Gomes *et al.*, 2019; Stein and Gregoire, 2019), very few of the millions of new materials and structures identified in computational model predictions and crystallographic analyses have been synthesized at bulk scales. Far fewer were characterized, and their properties estimated (Alberi *et al.*, 2018; Maier, 2019; Iquebal *et al.*, 2020a) . Despite these international initiatives, *characterization and property estimation* remain critical bottlenecks (Ding and Bukkapatnam, 2015; Alberi *et al.*, 2018; Maier, 2019; Iquebal *et al.*, 2020a).

Also, as noted earlier, AMDM would evolve from the current state of discovering chemical structures through accelerated wet-chemistry experiments, to discovering and manufacturing bulk-scale material structures on demand. For example, recent advances in multi-material 3D-printing open possibilities to synthesize large portfolios of material in bulk-scales in a single experimental batch; dozens of material samples can be created with an hour, thereby accelerating the discovery process. Although technology for rapid synthesis of materials is advancing, characterization now becomes a critical bottleneck. Typically, it takes several hours to comprehensively map out the morphology and microstructures of every 3D printed sample. A 1-3 order speed up is essential for *impeding autonomous materials discovery, and the materials-on-demand manufacturing paradigm*. Characterization of the structure and properties of the synthesized materials, such as the microstructure of metallic and ceramic materials and composites over the desired resolutions (including the crystalline grain structure) is also important for the qualification of a material, a manufacturing process, or a product for industrial applications (Weaver *et al.*, 2016; Boyce and Uchic, 2019; National Academies of Sciences, 2019). Characterization is equivalent to the estimation of the relevant subset (subspace) of the process state (in laser-based 3D printing, the dendritic microstructure to a specific resolution), which in turn allows the prediction of the outputs (here, material hardness, machinability, and similar properties).

Recent advances in instrumentation and imaging technologies allow quantitative characterization of, not just the shape and surface morphology, but also the microstructure and material composition over multiple scales, at short cycle times. For example, the surfaces were imaged within a few minutes over an mm-scale field of view via multi-beam SEM (Burnett *et al.*, 2014; Malloy *et al.*, 2015; Slater *et al.*, 2017), a material phase diagram was constructed based on X-ray diffraction (XRD) and fluorescence spectroscopy (Gregoire *et al.*, 2009) to discover material phases (Xing *et al.*, 2018). Experimentation platforms that combine sample preparation with imaging, such as RoboMet.3D of Airforce Research Lab (AFRL) (Uchic *et al.*, 2012) have been developed to capture grain microstructure and morphological features (e.g., pores) in 3D-printed alloy samples over $\sim$1 mm^3 scales. Sandia Labs' platform integrates 3D printing with an electromechanical load-frame, imaging, and X-ray computed tomography capabilities (Boyce and Uchic, 2019) for automated testing of a large batch of 3D printed tensile specimens, saving significant transport and staging efforts. However, these technologies are a few orders of magnitude slower to comprehensively map out the underlying microstructures over bulk scales compared with the rates at which material samples can be synthesized (Oses *et al.*, 2018; Oertel, 2019).

Alternatively, direct property estimation methods, such as rapid hardness testing instruments, have been developed, circumventing the need for elaborate microstructure estimation (Baker, 1991; Baker, 2010). These methods, however, are confined to *post facto* analysis and are generally not suited for *in-situ* rapid characterization. This is because most of these methods employ the underlying microstructure as ground-truth to ensure repeatability (Zhao *et al.,* 2020).

Novel test protocols and setups that leverage advanced instruments, computational mechanics techniques, and sample preparation methods have also been developed. For example, digital image correlation (DIC) techniques were used to find the stress–strain characteristic of materials that were stacked together to form a tensile test specimen (Knoll *et al.*, 2017). *Instrumented indentation* techniques have also garnered notable attention. These techniques open pathways to compress time scales for estimating the morphological characteristics, and properties of many microstructures realized from DED and other 3D printing processes (Bukkapatnam *et al.*, 2018; Jin *et al.*, 2020; Jin *et al.*, 2022). Recent innovations have also addressed the challenges with speed (e.g., achieving $\sim$100 indents/min rate (Hintsala *et al.* 2018) of estimating properties over nano- through meso-scales (Iquebal *et al.*, 2020a)). However, ensuring the consistency of the estimates surmounting the interaction and sampling effects remain open issues (Weaver *et al.*, 2016). To address this challenge, multiple research groups advocate employing data science/AI techniques (Dimiduk *et al.*, 2018).

Perspectives: Thus far, very few of the current efforts focus on addressing specific challenges at the nexus of synthesis and characterization, especially leveraging recent advances in smart 3D-printing and other manufacturing platforms (Botcha *et al.*, 2020; Kaufmann *et al.*, 2020; Zhao *et al.*, 2020). In this context, one cannot just "deep learn" the relationships with the limited data, gathered from expensive experiments. Almost all purely supervised learning methods suffer from this limitation. A fundamentally new data science and AI paradigms are necessary (Aspuru-Guzik and Persson, 2018; Tabor *et al.*, 2018; Boyce and Uchic, 2019; Gomes *et al.*, 2019; Kauffman, 2019; National Academies of Sciences, 2019). New data science/AI principles that can combine mechanistic understanding of 3D printing and other manufacturing processes, and can fuse manufacturing process signals and images with offline measurements (Wolff *et al.*, 2017; Botcha *et al.*, 2020; Wolff, Wang, Gould, Parab, Wu, Zhao, Greco and Sun, 2021; Wolff, Webster, Parab, Aronson, Gould, Greco, and Sun, 2021) are necessary to accelerate characterization and property estimation one leap forward. Research in this front is in early stages. For example, automation of nano-material

characterization has been discussed in a recent article (Park and Ding, 2019). Some work has also been reported to connect the *in-situ* measurements including from acoustic emission, forces and temperature sensors to morphology, microstructure and intrinsic material properties (Iquebal *et al.*, 2020a; Nakkina *et al.*, 2020).

An additional growing trend is the integration of rapid characterization and property estimation as part of material synthesis platforms (Carroll *et al.*, 2021). Many offline instruments, such as surface profilometers and XRD are being adopted for *in-situ* measurements so that characterization can happen almost concurrently with material synthesis (Wolff, Wang, Gould, Parab, Wu, Zhao, Greco and Sun, 2021). New class of platforms integrated with these measurement capabilities and can execute complex manufacturing recipe (workflow), opening interesting possibilities to enhance the modeling and inferring of PSP relationship, and probing of MDS (Botcha *et al.*, 2020; Zhao *et al.*, 2020; Karthikeyan *et al.*, 2022).

5. Discussion and concluding remarks

Materials discovery has traditionally been a highly manual and iterative endeavor. It had largely relied on the intuition and experiences of the experimenter. Many of the material discoveries have taken years, often a couple of decades of painstaking iterative efforts of material synthesis under various design conditions till a qualified material is discovered and manufactured for an industrial application. Recent advances in manufacturing technologies, robotics and AI unlock the possibilities to create autonomous materials manufacturing and measurement platforms endowed with advanced computing, inference, and control capabilities, besides capturing human scientists' high levels of logical reasoning, experimentation, and adaptations to shrink these cycles dramatically.

Additionally, the materials genome initiative has brought a significant progress since early 2000s towards the identification of millions of materials in computational model predictions and crystallographic analyses. Although very few of these identified materials and structures have been physically realized (Alberi *et al.*, 2018; Maier, 2019; Iquebal *et al.*, 2020a) these model predictions and analyses provide impetus to discover and manufacture many of these materials for targeted applications. These discoveries can lead to new products that are optimized not just in terms of the shape and topology, but also have frugal material structures that bring enhanced combination of properties previously thought impossible, and can dramatically reduce material footprint.

A few years ago, autonomous materials discovery was recognized as an exciting grand challenge that is at interdisciplinary confluence of AI/data sciences and manufacturing/materials sciences (National Research Council, 2008; Tabor *et al.*, 2018; de Pablo *et al.*, 2019; Gomes *et al.*, 2019; National Academies of Sciences, 2019). Multiple international efforts have brought together researchers and experts from these diverse disciplines to address this grand challenge. The autonomous discovery initially was a mere computational exercise that employed purely computational simulations instead of experimental (hardware) platforms. As summarized in the foregoing, several hardware platforms are being developed in recent years. Many of the current platforms focus on executing wet-chemistry processes. Significant scope exists for AMDM platforms to manufacture truly bulk-scale materials and parts, and integrate advanced capabilities for the "brain" and the "body" of an autonomous materials discovery system. A few additional trends in autonomous materials discovery and synthesis are as follows:

1. As data science and machine learning principles evolve, leading to advancements in and beyond encoding and learning methods, autonomous materials discovery can offer the so-called "challenge case studies" to develop, assess and adopt these newer classes of data-driven surrogate models. These newer classes of surrogate models should train and predict well by fusing unstructured domain knowledge and human experiences, and diverse snapshot and steaming signals and images, albeit with limited labeling (e.g., material property measurements, representations of microstructure, thermodynamic process states).
2. Recent sequential experimentation systems have shown some capability to execute flexible workflows (processing and measurement recipes) based on the outcomes from the earlier experiments and the prescribed MDS search strategy. In future, these systems should be endowed with a higher level of autonomy, emulating more human-like ability to develop and act upon the intuition, fuse multi-fidelity measurements and unstructured data, ability to be surprised and accordingly pivot the hypotheses, adapt the workflow *on-the-fly* based on the observations without waiting for the outcomes, synthesize deeper knowledge of the underlying relationships to develop "out of the box" thinking (i.e., expansion, not just exploitation and exploration) to develop recipes and material compositions.
3. The emerging trends also point to the development of manufacturing machine tools serving as material synthesis platforms. These platforms are being integrated with *in-situ* measurement and characterization capability, as well as advanced robotic arms to enable the handling of the material precursors and manufacturing tool heads (Shukla *et al.*, 2020; Thien *et al.*, 2022). Recent wire-fed hybrid manufacturing platforms are initial archetypes of these configurations. They can offer enhanced flexibility and adaptability of the workflow, i.e., the synthesis and MDS search process and react in real-time to more efficiently identify, and possibly discover and manufacture the desired material.
4. The majority of model materials investigated in the literature for automated and autonomous discovery are metallic materials, and much fewer are polymeric, or ceramics. Among metals, multiple research groups addressed the discovery of shape memory alloys (SMAs)

with specific thermo-mechanical response characteristic, as well as high-entropy alloy (HEA) materials with a specific phase configuration and thermo-mechanical response (Koželj *et al.* 2014; Toda-Caraballo and Rivera-Díaz-del-Castillo, 2015; Chaudhary *et al.*, 2017, Tapia *et al.*, 2017, Solomou *et al.*, 2018; Miracle, 2019; Vlassak and Arroyave, 2019; Wen *et al.*, 2019; Kaufmann and Vecchio, 2020; Li *et al.*, 2020; Meisenheimer and Heron, 2020; Wang *et al.*, 2020; Revi *et al.*, 2021; Witman *et al.*, 2021; Zhuang, 2021, Arróyave, 2022). These material systems are attractive from a materials discovery standpoint because the extreme sensitivity of their microstructure and properties to the processing conditions and workflow pose interesting challenges both in terms of learning the underlying PSP relationships, as well as executing suitable MDS search strategies. Among polymeric (Arora *et al.*, 2016; Audus, 2016; Chandrasekaran *et al.*, 2020) and ceramic (Roy, 1990; Rajan, 2001; Pullar, Zhang, Chen, Yang, Evans, and Alford 2007; Pullar, Zhang, Chen, Yang, Evans, Petrov, Salak, Kiselev, Kholkin, and Ferreira, 2007; Rajan and Tan, 2011; Pullar, 2012; Kaufmann *et al.*, 2020) materials, discoveries for energy storage, high temperature operations, and battery applications have received notable attention. The future "challenge case studies" could include the discovery of material structures that exploit the coupling across the spatial scales, such as between the microscale (micromechanics) and the macroscale (product geometry) features to derive enhanced, targeted properties.

5. Consequent to the current growing interest to integrate the synthesis platforms with a broader variety of testing and characterization setups (to assess material structure, property and performance), future autonomous materials discovery systems would open interesting possibilities for the identification and manufacturing of components with specified functionality via innovative processing recipes, without relying on expensive and scarce (e.g., rare earth) materials—thus addressing the critical materials challenge noted by the National Academy of Sciences (National Research Council, 2008; National Academies of Sciences, 2019).

The current trends and imperatives of an AMDM system, taken together, present some unique challenges that few other autonomous systems domains can offer. These challenges make AMDM an attractive area to develop the next wave of AI, data science, and optimization methodologies to address the innate unique challenges. Advances to the theory and methods of data-driven, dynamic decision-making form a central piece towards the realization of truly closed loop AMDM systems that can exhibit high levels of autonomy. In this sense, AMDM would present application challenges of interest to the ISE/OR fields. For example, the type of balance one needs to strike between the precision of the underlying models and timeliness of the estimates and decisions, the typically high volumes, velocity and uncertainty associated with the diverse data streams in an AMDM system, together with the computational constraints and experimentation costs impose interesting challenges for optimizing the MDS search process (to conduct experiments and/or simulations) as well as for learning the PSP relationships from the experiments and data. It is possible to investigate newer forms of acquisition functions, along with distributed learning and search strategies that can help achieve a more efficient global optimization in these scenarios.

As noted earlier, recent growing interest in reinforcement learning, especially to address the computationally efficiency issue, and also data scarcity in dynamic decision processes hold immense future potential for AMDM. Additionally, the PSP relationships of diverse materials and manufacturing processes are bound by the commonalities among underlying governing physical laws, as reflected in the structure and responses of the computational models, not just the similarities in the spatio-temporal characteristics of data from *in-situ* and *ex-situ* measurements. This offers some very interesting avenues to develop methods to distribute and/or transfer the PSP relationship learning procedures, and enhance the efficiency of the search process. Recent advances in distributed learning theory and methods would offer interesting untapped potential in this regard.

Also, as noted in the foregoing, advancements in AMDM could open the possibilities for a material-on-demand manufacturing paradigm. Newer classes of machine tools that can customize materials in addition to the geometry and morphology of the products can emerge. These advances provoke a rethink of the entire edifice of process planning to include the materials design and discovery on-demand aspects. New generations of production planning and control systems that can leverage these advances and are well suited for the emerging materials-on-demand manufacturing paradigm would be an important future research direction. The challenges towards realizing AMDM systems can thereby spur deep and sustained collaborations among the manufacturing/materials scientists together with AI/data scientists, including those from the ISE/OR fields, as the challenges that are precisely at the nexus of these disciplines.

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