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# Systems Approaches to Materials Design: Past, Present, and Future

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## Abstract

There is increasing awareness of the imperative to accelerate materials discovery, design, development, and deployment. Materials design is essentially a goal-oriented activity that views the material as a complex system of interacting subsystems with models and experiments at multiple scales of materials structure hierarchy. The goal of materials design is effectively to invert quantitative relationships between process path, structure, and materials properties or responses to identify feasible materials. We first briefly discuss challenges in framing process-structure-property relationships for materials and the critical role of quantifying uncertainty and tracking its propagation through analysis and design. A case study exploiting inductive design of ultrahigh-performance concrete is briefly presented. We focus on important recent directions and key scientific challenges regarding the highly collaborative intersections of materials design with systems engineering, uncertainty quantification and management, optimization, and materials data science and informatics, which are essential to fueling continued progress in systems-based materials design.

## 1. INTRODUCTION

Throughout history, the vast majority of technological advancements have been enabled by the discovery of materials with required properties (1–3). Unfortunately, the materials development cycle is often one of the most consequential bottlenecks to potentially transformative technologies (4–7). As technological development accelerates, the rate at which new materials must be discovered increases (4, pp. 1–18). Decades ago, Olson (8) framed the problem of materials design as one in which materials are considered as complex hierarchical systems; articulation of the (composition) process-structure-properties (PSP) relations is exploited to tailor a material so that it meets a specified performance requirement or requirements. Improved performance of modern materials can be achieved only through intentional engineering of their structure across a range of length scales. Ni-base superalloys, for example, are some of the most sophisticated engineered materials due to the highly complex chemistries and sophisticated thermo-mechanical treatments that result in their hierarchical microstructures (9).

The intent of this review is to present materials design as essentially a goal-oriented activity, viewing the material as a complex system of interacting subsystems. The design process effectively seeks to invert quantitative PSP relationships to identify the accessible chemistries and process paths necessary to realize a hierarchy of (micro)structures that may achieve application-specific performance metrics (8, 10). Materials design concepts have played a powerful role over the past two decades in motivating the integration of computational mechanics and materials science with engineering materials development [see the DARPA Accelerated Insertion of Materials Initiative (11) and the Integrated Computational Materials Engineering (ICME) initiative (7)]. While volumes of case studies have been compiled (7), the intent of the present overview is to focus on important recent directions at the highly collaborative intersection of materials design with systems engineering, optimization, and materials informatics (12) that we consider essential to fueling continued progress in systems-based materials design.

An important characteristic of advanced materials is the notion of structure hierarchy, ranging from interatomic/intermolecular spacing to relevant scales of microstructure to the scale of manufactured parts. Materials properties are affected by various levels of structure hierarchy. Prediction of mechanical behavior of materials based on concurrent (13, 14) or hierarchical (15) multiscale modeling involves the propagation of information between models across different length scales and timescales; multiscale information flow comes at significant computational cost as well as with potentially high levels of uncertainty. Multiscale modeling provides decision support for materials design by estimating the sensitivity of responses or properties to variation in the materials structure at each level of structure hierarchy. The role of materials structure is important for at least two reasons: (*a*) Sensitivity analyses are necessary to quantify dominant design variables among levels of materials structure hierarchy, as these variables control property/response variance, and (*b*) sensitivity of process-structure and structure-property relations are central to concepts of robust design (16), where the goal is to explore a range of candidate solutions to identify those that are least sensitive to process path, materials composition, target microstructures, and even range of service conditions (1).

The goal of simulation-assisted materials design is typically not to accurately predict mean properties at higher scales. Rather, the goal is to (*a*) understand their sensitivity to materials (micro)structures, (*b*) capture dominant mechanisms and their transitions that affect materials responses or properties, or (*c*) provide support to search for regions in the materials design space that may deliver optimal performance in some desired sense. Design of the materials structure hierarchy together with product-level performance compels the tracking of materials structure attributes (e.g., defect structure, grain or phase size, shape and orientation distributions) at various levels of hierarchy.

Consideration of uncertainty in multiscale materials simulation has important implications for providing decision support for materials design and affects the type of multiscale modeling scheme most suitable to this task. When one is addressing problems related to materials design, the more common approach is to explore causal relationships in the PSP relations in a hierarchical manner, as opposed to through concurrent modeling schemes, mainly because models of important mechanisms tend to be framed at specific levels of the materials structure hierarchy (17).

## 2. MATERIALS DESIGN WITHIN THE CONTEXT OF ENGINEERING SYSTEMS DESIGN

### 2.1. Simulation-Assisted Materials Design as a Goal-Oriented Activity

Engineering design is a decision-making process (18) in which design objectives are formulated and then the design space is explored through search methods appropriate to the specific problem at hand. Materials design thus implies a top-down-driven, simulation/model-assisted decision-making process whereby one seeks to determine the materials composition and processing necessary to tailor the material's hierarchical structure to meet performance requirements (16, 19). Our emphasis is on simulation-assisted—rather than wholly simulation-based—materials design, as this closely aligns with the premise of ICME and implicitly acknowledges the many limitations of materials simulations frameworks. The intent of simulation-assisted materials design is to reduce but not eliminate experimental routes to the establishment of PSP relationships (20). The models used in the process can be constructed from theory, simulation, or empirical evidence. They may be analytical, computational, or of surrogate character on the basis of reduced-order models. These models, regardless of their nature, are used to assist in the process of exploring the (multilevel) materials design space.

### 2.2. Materials Design as an Inverse Problem

Materials science tends to focus on the forward problem of establishing PSP relationships. This approach seeks answers to the following question: Given a set of materials descriptors that can be manipulated through materials synthesis and processing, what are the corresponding properties and performance characteristics? In contrast, materials design necessitates inversion of this process (8, 21): Given the desired performance requirements, what are the feasible and perhaps most desirable materials composition and processing routes to realizing structures that meet these requirements? Materials design seeks to effectively invert the PSP relationships to identify domains in the process-structure and structure-property space or spaces that meet specific performance targets, whether expressed as ranged sets of materials properties or in terms of product-level requirements. Microstructure-sensitive design (17, 22, 23) is an example, as it consists of the efficient mathematical representation of microstructures and use of homogenization techniques to relate microstructure (expressed in terms of low-dimensional representations) to performance, followed by the goal-oriented search of the microstructure space to identify microstructures that best achieve the design goals.

The idea of designing a material with targeted properties can be traced to works of the Steel Research Group at Northwestern University in the mid-1980s (**http://srg.northwestern.edu/**). Olson's Venn diagram (8) relating PSP relations to performance is foundational to engineering systems approaches to materials design. This systems-level framing clearly distinguishes the pursuit of top-down, goals/means, inductive systems engineering (design) from bottom-up, cause-and-effect, deductive linkages. The materials-by-design approach (8) rests on two principles: (*a*) the

treatment of materials as hierarchically organized systems and (*b*) the integration of computational models describing each of the scales/hierarchies in the materials system to assist in optimizing performance.

Because of the incredible complexity of process-structure relations, their exploration has been carried out almost exclusively—with some exceptions (24)—through experiments. Structure-property assessments are then carried out via experiments or computations once interesting candidate structures are realized in the laboratory. These processes are subsequently scaled up to the prototype level to determine whether the resulting property sets deliver the required performance. This is the conventional paradigm of iterative parametric design (8, 25), in which the design search is parameterized in terms of associated dominant variables, with the goal of minimizing the number of iterations.

A practical and compelling goal of a simulation-assisted systems strategy for design and development of materials is to replace an increasing fraction of otherwise experimentally driven decisions with those informed by computational simulation or by statistical models derived from high-throughput (HT) experimental exploration of the materials space. When using physically based multiscale models as aids in the decision-making process in materials design, we should consider that (*a*) they must be efficient and fast acting and (*b*) they must address uncertainty of models and experiments at each scale (26, 27). The first requirement is often difficult to realize, given the considerable cost associated with multiscale materials simulations. Current efforts are under way to make the forward evaluation of materials solutions more efficient by combining physically based models with efficient data-driven representations (28, 29). In any approach that relies on materials simulations, the importance of quantifying uncertainty is paramount; uncertainties arise in experiments, the models and model parameters (26), the cost of experimentation, the complexity of the models used to predict materials behavior, and the dimensionality of the design space. More importantly, the limitations inherent in materials models mean that uncertainty management is a critical part of any materials design problem (26, 30). McDowell et al. (1) and McDowell & Olson (25) address uncertainty by proposing design philosophies that favor designs that minimize sensitivity to variation of design variables on the basis of Taguchi's robust-design concept (31, 32). Such robust-design approaches have been extended to various application domains (33–37).

## 3. CONTRASTING MATERIALS DESIGN WITH MATERIALS DISCOVERY AND SELECTION

Since the advent of the Materials Genome Initiative (MGI) (4, pp. 1–18), much emphasis has been placed on the use of data science to enable the acceleration of materials discovery and the materials development cycle (38), although efforts to rapidly explore the materials design space (39) trace back to well before the MGI. By combining prior knowledge, brute force synthesis approaches, and, admittedly, a certain degree of serendipity, combinational materials discovery was established (40). It is facilitated by the development of rapid synthesis methods, HT screening techniques, and high-capacity information processing. Over time, combinatorial approaches have become one of the dominant paradigms in materials research (41). Although much progress has been made (42, 43), there exist limitations, particularly with regard to the ability to characterize structures and/or properties of samples at the same rate at which they are synthesized.

More recently, HT computational materials discovery (44) has followed a parallel track as its experimental counterpart. Most efforts on this front rely on the use of large computational capacity to carry out simulations that attempt to establish relationships between materials structure (almost

exclusively at the atomic level to date) and properties, including energetics and even functionality (45). Most computational materials discovery frameworks rely on quantum mechanical approaches such as density functional theory (DFT) (46). In fact, DFT-based HT materials discovery (47) has rapidly emerged as a powerful complement to experimental attempts to rapidly explore the space of materials structures and properties/responses, although the theory has important limitations (48).

Both computational and experimental materials discovery approaches have benefited from very recent developments in materials informatics (12, 49), which encompasses the systematic integration of data; correlation analysis; and, importantly, validation by empirical observations, theoretical insights, and mechanism-based models to develop insight and construct suitable predictions of PSP relationships (49). Materials informatics workflows consist of capturing data related to PSP relationships, identifying the dominant factors responsible for the structural features or materials properties of interest through feature selection or dimensional reduction approaches, and finally developing correlations and predictive models that establish quantitative connections along the PSP model chain (38).

Over time, materials that are discovered become well characterized, and their properties and performance metrics become part of the knowledge base that is used to select a material for a specific technology or application. Accordingly, materials selection (50) is concerned with the search for and eventual specification of already existing materials that fulfill the performance requirements of the specific application that must be supported. By merging phenomenological relationships in materials properties with data on specific materials characteristics, one can begin to develop classification schemes for materials behavior. The framework is very efficient and effective for establishing common structure-property relationships across seemingly different classes of materials. In contrast with materials discovery, materials selection requires that the materials design space be already well (if not fully) known.

Regardless of the means by which one interacts with or queries the materials design space, the key difference between materials discovery and materials design is the shift in emphasis from the outcome of exploration aimed at screening possibilities in the former, to the process by which the design space is systematically and comprehensively explored in the latter (51)—in this sense, materials discovery can be viewed as an early stage of design exploration or screening. In turn, materials selection is clearly differentiated from materials design in that in the former the materials design space is assumed to be already well known, while in the latter the main point is to augment the existing materials space to include materials that are yet to be designed.

## 4. BREAKING DOWN THE ELEMENTS OF MATERIALS DESIGN AS AN ENABLER OF ICME

ICME has emerged as a powerful conceptual framework, with the goal of integrating "science and engineering as well as the results of theory, experiments, and simulations into computational tools that can be used directly in engineering of new products or manufacturing processes" (52). To realize the goal of ICME, it is necessary to develop quantitative/predictive models capable of connecting chemistry and process route to microstructures and then to relate microstructures to properties and ultimately to performance, as shown in **Figure 1**. Considerable work has been carried out to address the simulation-assisted establishment of linkages across the PSP chain. In what follows, we present a brief overview of current, well-established efforts on this front, focusing on computational frameworks associated with the design of structural metallic alloys and related systems.

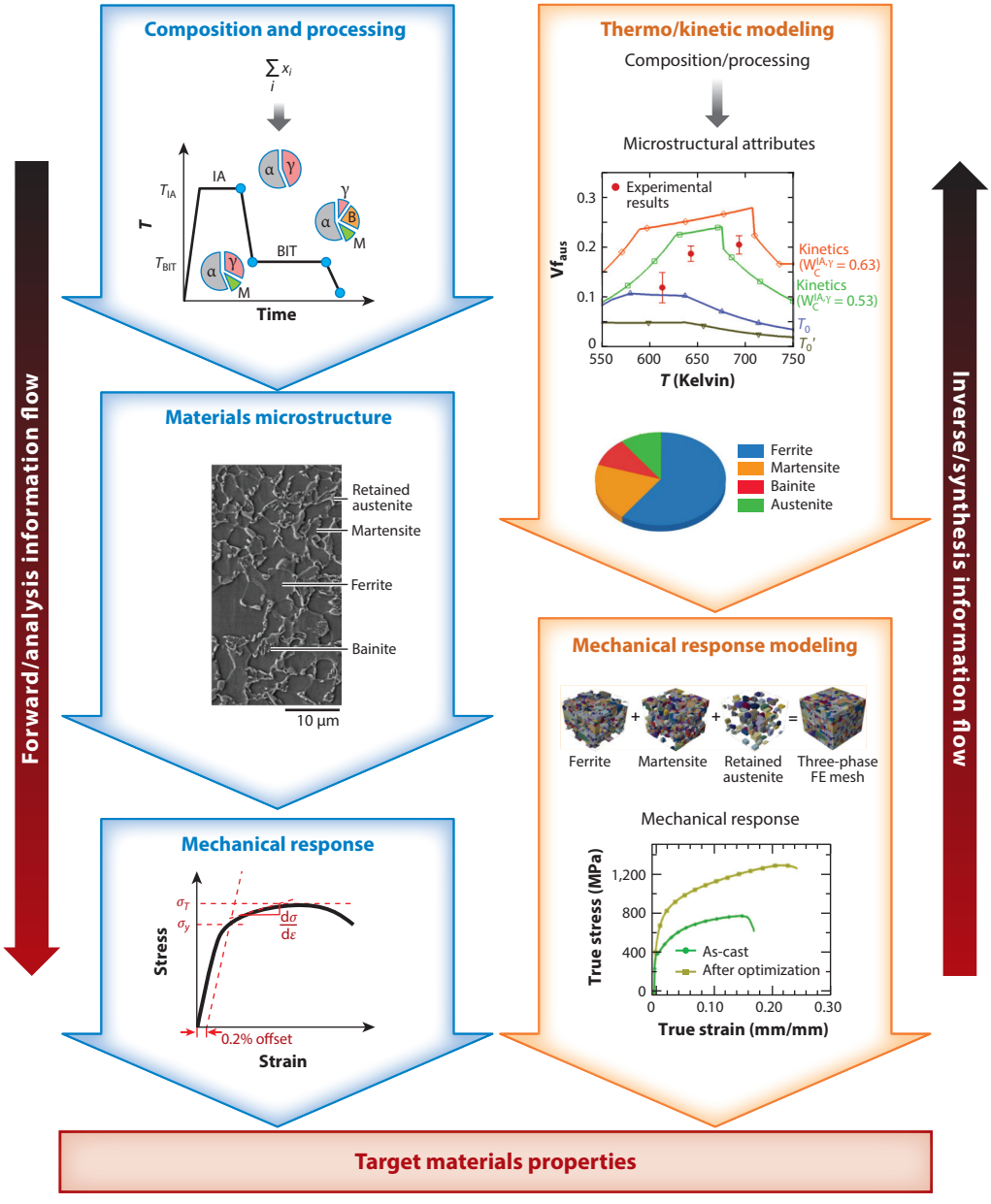


**Figure 1**

Forward models that predictively link chemical composition and processing to microstructure and alloy properties tackle the inverse problem of searching for candidate alloys that meet target design requirements in materials design. As an example, the process-microstructure-property relationships of a TRIP-assisted steels is presented. Abbreviations: α, ferrite; γ, austenite; B, bainite; BIT, bainite isothermal treatment; FE, finite element; IA, intercritical annealing; M, martensite. Elements of the figure adapted with permission from References 53 and 54.

## 4.1. CALPHAD-Based Phase Stability and Materials Design

The development of new materials—metallic alloys in this case—must start with an understanding of their phase stability, i.e., with a description of the relationship between thermodynamic conditions and the resulting phase constitution (equilibrium state). For decades, researchers have used the CALPHAD method (55) to develop self-consistent databases that encode the thermodynamic behavior of phases (56). In a so-called forward mode, thermodynamic conditions can be mapped to equilibrium states through minimization of the total Gibbs energy (56–58). CALPHAD resulted from a concerted effort toward a systematic encoding of alloy phase stability in terms of Gibbs energy functions assessed, with the use of experimental evidence for phase stability as well as thermo-chemical measurements. With the advent and popularization of DFT methods and software, CALPHAD assessments of phase stability have been enriched through the incorporation of so-called first-principles calculations of thermodynamic properties of phases (59, 60). The ability of the CALPHAD approach to encode thermodynamic information quickly led to extensions in which similar frameworks were used to assess and predict kinetic (e.g., atomic mobilities) properties of phases (57, 61). Recently, CALPHAD-inspired efforts toward encoding a much wider range of thermodynamic properties—such as molar volumes (62), thermal expansion (62), and elastic properties (63)—have gained much attention with the need to employ phase-level information in materials design.

Several groups have used CALPHAD-based methods to design new alloys. For example, Xu et al. (64) employed computational thermodynamics and basic models for the homogeneous nucleation of precipitates to design nanoprecipitate-strengthened steels. The solution implemented genetic algorithm–based optimization to evaluate candidate alloys in terms of specific alloy design criteria, identified the fittest candidates, and then let them pass on their "genes" to further generations to identify optimal regions in the composition space. Others have used similar approaches. Tancret (65) employed similar evolutionary approaches to navigate high-dimensional materials design spaces; this effort was recently expanded to address multiobjective design problems in which CALPHAD-based tools are supplemented by machine learning structure-property predictors in $\gamma'$-strengthened Ni-base superalloys (65) and so-called high-entropy alloys (66). Similar efforts across other regions in the alloy space have continued to demonstrate the power of CALPHAD-based phase stability predictions (66–71). Much work remains to be done, particularly as the community embarks on the exploration of the high-dimensional design space of high-entropy alloys.

## 4.2. Connecting Process Path to Structure (and Vice Versa)

The CALPHAD-based approaches to materials design outlined above have focused primarily on the impact of phase stability on phase constitution and its relationship with (some) relevant materials performance metrics. The ability to predict properties of alloys hinges strongly on the degree to which these properties are controlled by microstructural descriptors (72). To establish predictive models connecting structure to properties, a necessary (but insufficient) step is to go beyond the CALPHAD-based prediction of phase information and instead explicitly predict the effect of chemistry and processing on microstructure of alloys. The process-structure connection, however, is extremely challenging to quantify because of several factors. First, the final microstructure of an alloy system is generated from a very complex, process-specific, history-dependent sequence of transformations that is very difficult to specify a priori. Second, quantitative modeling of materials processing and resulting microstructure is largely underdeveloped. Third, inverting the process-structure linkages remains largely an unsolved problem due to the nonequilibrium, path-dependent nature of process-structure relations.

Perhaps the best-established examples of quantitative process-structure linkages have been established in the context of microstructure modification via controlled precipitation of strengthening phases (73). Modeling precipitation processes can be carried out at different levels of structure hierarchy, ranging from the explicit simulation of precipitate nucleation, growth, and evolution via microstructural phase-field methods (74–76), to mean-field approaches based on the implementation of the Kampmann-Wagner (K-W) (77) model of precipitation (78–80).

Such quantitative relationships between process route and (micro)structure have already been used in the design of alloy systems (81). Zhang et al. (82) investigated the precipitation of strengthening phases in the magnesium alloy AZ91. Lang et al. (83) investigated the coupling of thermodynamic modeling and precipitation simulations based on classical nucleation theory to investigate the effect of designed precipitate microstructures on hydrogen trapping behavior in martensitic steels. Recently, Martin and coworkers (84) provided a novel approach to controlling coarsening effects in 3D printing of Al alloys by focusing on control of nucleation during solidification via nanoparticle additions. While many similar works attempting to connect process to structure have been published, few works have attempted to arrive at quantitative PSP linkages. Recently, Galindo-Nava et al. (85) presented a physics-based modeling framework to describe the microstructure and mechanical properties of maraging steels. They explicitly considered the chemistry and treatment temperature effects on the hierarchical martensitic microstructure as well as the secondary precipitation of strengthening phases, arriving at the optimal chemistry and processing conditions for achieving the target mechanical performance metrics.

Solution of the inverse problem of connecting the desired precipitate microstructure to the required chemistry and thermal history in the case of precipitation-strengthened alloys remains an important challenge, with many possible approaches. Johnson & Arróyave (24) recently presented a framework whereby the precise temperature-time schedule necessary to arrive at the target precipitate structures [achieved through a mean-field K-W model (77)] was determined through black-box multiobjective optimization.

Other recent efforts have been directed at modeling more general process-structure relations. Yabansu et al. (28) proposed the use of low-dimensional representations of materials structure [principal-component analysis (PCA) of two-point spatial correlations] that are simulated via phase-field methods as a way to connect (computational) process parameters and microstructure states. Similar strategies in which a data-centric layer connects process and microstructure descriptors will likely accelerate the development of process-structure linkages, particularly when physics-based models are unable to provide quantitative predictions. Indeed, this is an area in which many contributions are required to advance the field of materials design.

## 4.3. From Structure to Properties (and Back)

The key to successful extraction of high-value PSP linkages lies in the identification of the salient descriptors of the materials hierarchical structure, including chemical composition. While it is abundantly clear that the rich hierarchical structures of most advanced materials demand an unimaginably high-dimensional description, it is also clear from past experience that only a few salient features dominate the materials response of relevance to any particular application. In most cases, these salient structure descriptors are not known a priori, and experts conduct numerous laborious and time-intensive trials (employing both measurements and models) for each application. Since this process is often nonstandard and leads to a highly customized, designer-dependent, set of structure descriptors, the knowledge gained in the process has only limited transferability. To dramatically improve the efficacy, scalability, and transferability of the above process to realize

the disruptive acceleration envisioned in ICME and the MGI, it is imperative to formulate and adopt a consistent framework for the description of the hierarchical materials structure.

Kalidindi and coworkers (17, 86, 87) recently developed one such promising framework, which has been demonstrated on a variety of materials structures at vastly different length scales. The Materials Knowledge System (MKS) (88, 89) employs digital representations, *n*-point spatial correlations (90, 91), and PCA (92) for dimensionality reduction to arrive at data-driven measures of the salient materials structure descriptors. Once such measures are established, they can be utilized to establish efficient surrogate hierarchical PSP linkages in problems of both homogenization (87, 93) (i.e., information flowing from lower scales to higher scales) and localization (94, 95) (i.e., information flowing from higher scales to lower scales). A distinctive feature of this framework is that it aims to harmoniously combine well-established micromechanical theories for heterogeneous materials (96, 97) with established systems theories (98, 99) and emerging concepts in data science (38) to take full advantage of their respective strengths. The MKS approach was recently extended to apply to extreme value distributions of driving forces to high cycle fatigue in alloy microstructures based on localization schema, supporting rapid parametric design of complex textured hcp alloy microstructures for fatigue response (100, 101). Much work remains to be done to extend the MKS framework to more general nonequilibrium, history-dependent structure-property relations (e.g., diffusional creep and relaxation, grain growth, load history dependence of defect structures), but the method might be used in combination with direct numerical simulation or with metamodels for such processes with varying degrees of coupling.

## 5. MODERN STRATEGIES FOR MATERIALS DESIGN

We next turn our attention to some contemporary methodologies employed in the top-down search for feasible materials design solutions once the PSP relations are established. Materials design approaches that predated the 2008 release of the ICME report (4–7) largely made use of iterative design approaches commonly employed in the design of other complex engineering systems (e.g., prototyping, testing, analyzing, and refining) (8, 21). By adding to this iterative process known information regarding analytical or computational models for key phenomena across the PSP relations, one is able to parametrize the responses in terms of input variables, which supports design optimization approaches (102). There is an increasing infusion of multiscale modeling and decision-based design strategies into ICME workflows (see Reference 15). This latter direction has been a key point of discussion between the materials and multidisciplinary design optimization communities, with a heavy focus on design optimization under uncertainty that respects the complexity of real materials. These strategies have also come to increasingly rely on modern data science approaches. We therefore focus on materials design strategies that have followed in the wake of the launch of ICME (2008) and more recently that of the MGI (2011).

### 5.1. The Inductive Design Exploration Method

The inductive design exploration method (IDEM) (1, 19, 103) is an example of a practical strategy for formally organizing top-down design searches based on bottom-up PSP relations in a way that emphasizes decision support; it may be regarded as composing an ICME PSP workflow. IDEM (**Figure 2**) provides a step-by-step strategy to facilitate the combined bottom-up modeling and simulation and/or experiments, leading to consideration of top-down mappings that satisfy high-level performance requirements while enabling uncertainty propagation across model chains. In the first step, IDEM employs parallelized bottom-up simulations, experiments, and/or associated reduced-order models or correlations to establish process-structure and structure-property

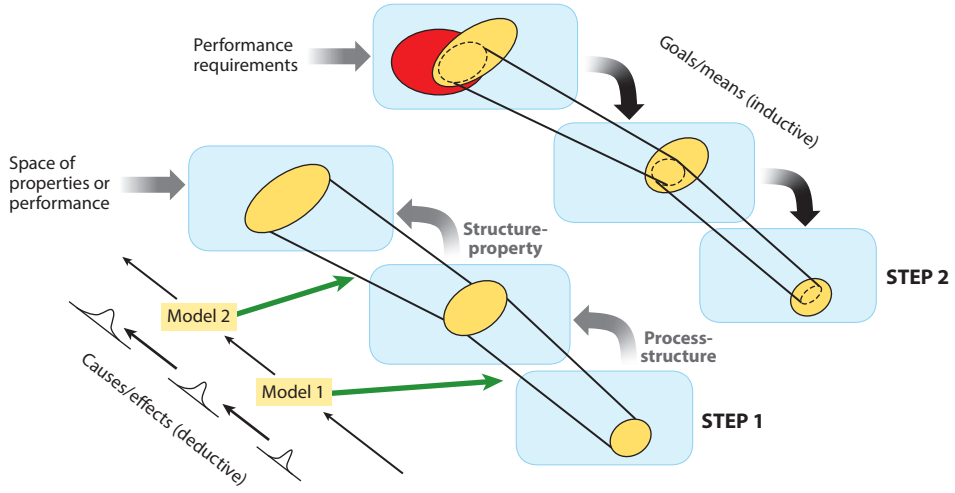


**Figure 2**

Schematic of Steps 1 and 2 in the inductive design exploration method (1). Step 1 involves bottom-up simulations or experiments for process-structure and structure-property relations, typically conducted in parallel fashion, to map composition into structure and then into properties, with regions in yellow showing the feasible ranged sets of points from these mappings. Step 2 involves top-down evaluation of points from the ranged set of specified performance requirements that overlap with the feasible regions established by bottom-up simulations in Step 1. Adapted with permission from McDowell & Kalidindi (2).

relations. The goal is, starting with a ranged set of performance requirements, to identify candidate ranged sets of solutions that jointly satisfy both top-down requirements (given performance, then resulting properties, then structure, then process route) and bottom-up (experimental- or model-based) mappings of the process-structure-property-performance (PSPP) relations, along with any imposed constraints. IDEM's second step involves a search within the ranged sets of performance requirements or associated property sets (for example, obtained via materials selection) backward for feasible sets of structures and associated process paths that overlap with design requirements.

Design decisions can be made on the basis of user-specified preferences within the Pareto frontier—the set of nondominated solutions to a multiobjective optimization problem. As discussed elsewhere in detail (1), the Pareto frontier for acceptable solutions can then be selected to maximize a multiobjective function or to express insensitivity of target responses with regard to changes in process or microstructure design variables; this notion of insensitivity appeals to principles of robust design as laid out by Taguchi for process control (31). To accommodate robustness of design solutions, IDEM employs hyperdimensional error margin indices to measure the distance from a given feasible solution in the space of interest (e.g., composition, structure, property) to the nearest boundary of feasible solutions and makes use of Gaussian pseudolikelihood measures of variance to define the probability of overlap. In this way, bottom-up information flow is combined with top-down search based on performance requirements in the presence of uncertainty. Moreover, this approach indicates how sensitive designer-selected solutions might be to changes in materials process path or microstructure, which is of practical interest to materials suppliers in terms of quality control/assurance. IDEM may also indicate which elements of the PSP framework require more investment to reduce uncertainty and facilitate execution of more detailed design.

Quality experimental information can be factored in as desired to fill gaps in physics-based modeling. IDEM is essentially just a template for a balanced decision-making process that has

been employed in design for many years, biased toward quality information and insights (25). Applications of IDEM to robust-design methods have been described elsewhere (1, 19, 104). The IDEM template for instantiating PSP relations followed by guided top-down design search was recently written in Python script format (pyDEM), and its capabilities were extended to support complex feasible design spaces and robust search algorithms for a range of materials design and selection problems (105). Recently, Wang et al. (106) presented an update to this class of approaches by developing a template-based ontological method for the exploration of the design space.

We illustrate IDEM via a representative case study [Ellis & McDowell (104)] aimed at concurrent design of the hierarchical structure of a complex structural material, ultrahigh-performance concrete (UHPC), together with design of a blast panel required to withstand a specified blast wave impulse. This example employed hierarchical multiscale computational modeling, analytical models, and associated metamodels to construct a set of bottom-up, deductive PSP mappings, instantiated within IDEM to propagate various sources of uncertainty. It was then employed to pursue top-down inductive design exploration for application-specific design objectives. A set of PSPP mappings were constructed across micro-, meso-, and macrolength scales by using analytical expressions and hierarchical multiscale finite element models at the single-fiber, multiple-fiber, and structural length scales. The set of PSPP deductive mappings considered seven design variables—panel thickness; fiber pitch; ratio of water to cementitious materials; curing temperature; and volume fractions of fibers, cement, and silica fume—across four levels of materials hierarchy.

Starting with the highest and next-to-highest hierarchical levels as the output and input spaces, respectively, IDEM was implemented via application of three steps: discretization of input variables, projection of discretized sets of input variables with account of uncertainty to a range in the output space, and determination of which sets of discrete input values satisfy the output space requirement or requirements. By recursively applying these three steps, PSPP relations were explored in a robust manner for properties, structures, and processes that satisfied the performance requirement or requirements. This approach has the advantage of identifying ranged sets of values of design variables that account for propagated uncertainty. By defining additional mass and cost objectives, the feasible input space was then searched to find the preferred combination of values of design variables that minimized mass and minimized cost while maintaining a robust materials and structural design.

Implementation of the materials design process proceeded by defining a set of PSPP mappings (see **Figure 3**); determining which analytical and empirical relations from literature could be employed, with emphasis on process-structure mappings; developing computational models to complete the set of PSPP mappings; validating the analytical, empirical, and numerical models; generating the response surfaces and estimating error associated with each response function; determining ranged sets of design variable values within the feasible domain via IDEM; defining mass and cost objective functions; and determining the preferred materials designs for mass and cost. By using this strategy, feasible solutions were explored in the materials design space (maximum tensile strength, fiber pitch, panel thickness, and fiber volume fractions) for a desired blast impulse.

This case study example is significant for three reasons. First, it demonstrates the utility of a bottom-up, hierarchical multiscale model involving both the UHPC materials structure and panel subjected to blast loading. Second, it demonstrates the simultaneous design of UHPC materials and structures subject to blast loading. Third, this work demonstrates a materials design process that can be employed for the concurrent design of other materials and structures, which could be subject to a variety of loading and service conditions. Essentially, it represents the integration of systems design of both materials and manufactured product forms.

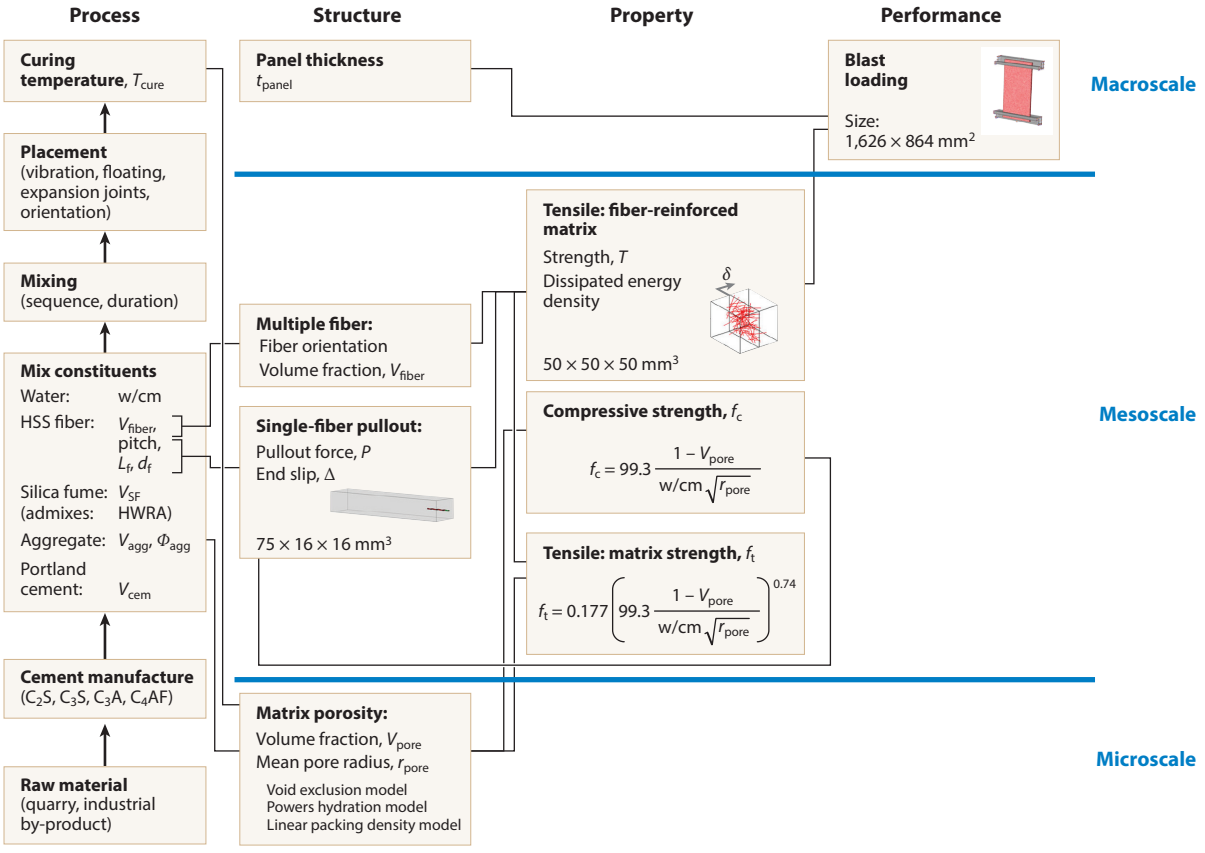


**Figure 3**

A set of process-structure-property-performance mappings for concurrent design of ultrahigh-performance concrete and a panel subjected to blast loading. HSS denotes high-strength steel. Adapted with permission from Ellis & McDowell (104).

## 5.2. Optimal Materials Discovery and Design

The discovery, design, and development of new materials are resource limited. Moreover, exclusively experimental approaches, due to their very resource-intensive nature, preclude the systematic exploration of the typically vast design space. An approach that has received increasing attention is the HT exploration of the materials space, via either computation (44) or experiments (42). The data generated are then used to populate databases of features/response sets that are in turn analyzed via machine learning approaches to establish structure-property relationships, and these machine learning predictions are then used to identify materials with the optimal characteristics (107). HT process-structure determinations, even those based on data science methods, remain scarce. Some recent works have employed direct projections from process path to properties for a given alloy system of interest on the basis of extensive experimental assays (108), although the ramifications of skipping the associated accessible microstructures are not addressed; this approach may create challenges in defining methods for quality control assurance and inspection of manufactured parts. Moreover, absence of materials structure information complicates any inverse search operations that one may wish to conduct in materials design, since data science–based

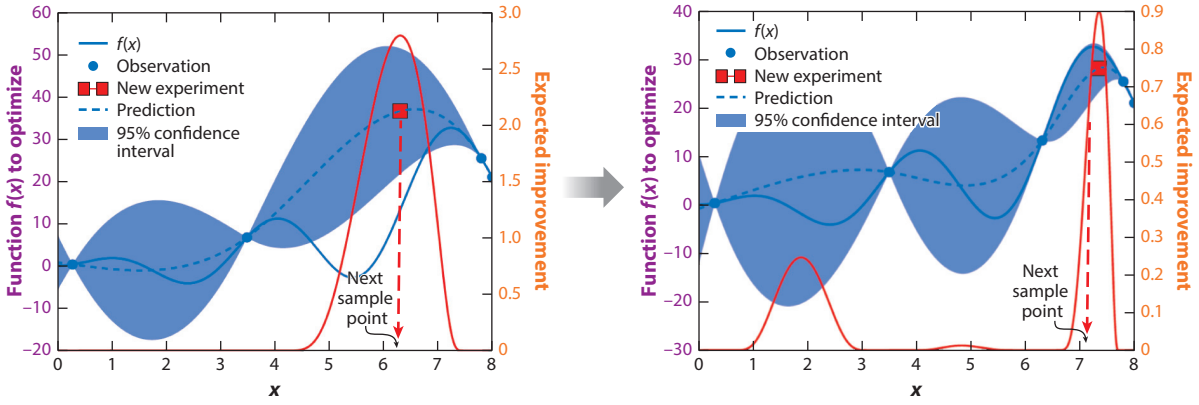


**Figure 4**

Schematic illustration of Bayesian optimization (BO). From a limited number of observations on a system (*blue solid line*), a stochastic model (*dashed blue line* and *shaded area*) is built. The next observation is determined by accounting for the trade-off between the exploitation of the current knowledge and the exploration of the unknown regions of the design domain $x$. In this case, expected improvement is the metric used, and thus the policy falls within the efficient global optimization framework (112).

methods for direct process-property relations are often of black-box nature, lacking guidance from physics-based models.

HT methods are often limited in that they do not account for either (*a*) constraints in the available resources to carry out the exploration of the materials design space of interest or (*b*) bottlenecks in the scientific/research workflow that necessarily prevent the parallel execution of specific experimental or computational tasks. For example, one can imagine the development of a full complement of experimental synthesis platforms to carry out the HT synthesis of thousands of compounds at once, only for this effort to be throttled by the low-throughput nature of the characterization operations necessary to establish structure-property linkages.

Given the limitations of conventional HT approaches, several approaches to the efficient exploration of the materials design space have begun to emerge (109). The most efficient methods for materials discovery are based on variants of Bayesian optimization (BO) (110) (see **Figure 4**). Bayesian methods provide the mathematical framework to update prior knowledge upon acquisition of new information (111). General BO methods are used to find global optima of expensive (black-box) objective functions (112) by combining a hypothesized model (prior) describing possible outcomes of the expensive function to be optimized with actual evaluations of the expensive objective function (data) to construct an update model (posterior) that predicts the outcome of evaluating the objective function over the entire optimization search space. The posterior model is orders of magnitude less computationally expensive than the black-box function and contains information about its mean response and variance. BO methods provide the framework to combine the mean and variance of the posterior model into a utility function that can then be used to select the next-best point to evaluate in the search space (112). Generally, the utility function is constructed to balance the exploration (sampling regions with high degree of uncertainty) with the exploitation (sampling regions likely to yield locally optimal results) of the design space. While BO is typically framed in the context of global optimization of expensive black-box functions, one could easily make the connection to materials discovery/optimization, as experiments and/or simulations can be considered to be expensive black-box functions that need to be optimized to achieve the desired performance objectives. The key advantage of BO-based approaches to materials discovery over more traditional HT frameworks is the optimal allocation of resources. To date,

several approaches have employed BO in a materials science context (113, 114) and have shown dramatic improvements in the efficiency with which experimental/computational resources are utilized to find materials with optimal properties/performance, even within exclusively experimentally based frameworks (115).

Most BO efforts to date have focused on single-objective optimization. In materials science, however, multiple objectives (along a Pareto frontier) must be optimized at once, as pointed out in the IDEM discussion above. The problem then consists of identifying the optimal sequence of observations (via experiments or simulations) that is most efficient at identifying the Pareto frontier of candidate solutions. In common BO approaches, the search for the global optimum of the black-box function is always assumed to be sequential: The function is evaluated one step at a time, regardless of the number of objectives to optimize. Thus, even in multiobjective BO, it is necessary to quantify the utility of a potential experiment as a scalar quantity. A powerful scalar utility metric used in multiobjective optimization is the so-called expected hypervolume improvement (EHVI) (116) as a utility metric. Similar to the utility functions used in single-objective BO, EHVI is constructed by balancing the exploration and exploitation of the design space to efficiently locate the Pareto frontier. Recently, Arróyave and collaborators presented a multiobjective (up to three objectives) optimal materials discovery framework (117) and demonstrated its efficacy by identifying regions in the microstructural space that yielded optimal performance in a precipitation-strengthened NiTi-based shape memory alloy (see **Figure 5**). The microstructure design space was explored via a computationally expensive micromechanical model, and using the EHVI as a utility function was much more efficient than any other potential strategy for the solution to this multiobjective optimization problem.

Virtually all BO-based materials discovery/design approaches first require the identification of the most important features that control the outcome of the experiments/simulations in question. Feature selection, however, requires a considerable amount of information, and this step may be prohibitive if the cost of acquiring this information is high. Unfortunately, most materials design/discovery problems involve the evaluation of costly experiments/simulations, and thus in many cases it may not be practical to carry out a feature selection step before using BO. To address this issue, Arróyave and collaborators (118, 119) proposed a framework that is capable of adaptively selecting competing models that connect materials features to performance metrics through Bayesian model averaging, followed by optimal experimental design. The framework, termed Bayesian optimization under model uncertainty (BOMU), essentially carries out feature selection and BO simultaneously. BOMU thus enables BO even when very few initial data are available and there is a lack of understanding of which are the most important features responsible for influencing the outcome of the experiments.

In BOMU (see **Figure 6**), the entire feature space (which could contain tens and even hundreds of features) is subdivided into feature sets that correspond to different models or theories that connect experimental degrees of freedom to experimental outputs. Each model is then used to predict the next-best experiment to carry out given the data, as in conventional BO. While at the beginning all models are considered to have an equal probability of being correct, the probability of each model is updated—via Bayesian methods—by comparing its performance against the observations. The utility of every potential experiment/calculation is then computed by considering all the models at once, weighed by their individual probabilities, and the loop is repeated until a certain termination criterion is met. Talapatra et al. (118, 119) found that BOMU not only was extremely effective at finding the solution to single-objective and multiobjective materials discovery problems but also identified the model that most precisely predicted experimental outcomes, thus helping in the discovery of the materials features most associated with the properties of interest.

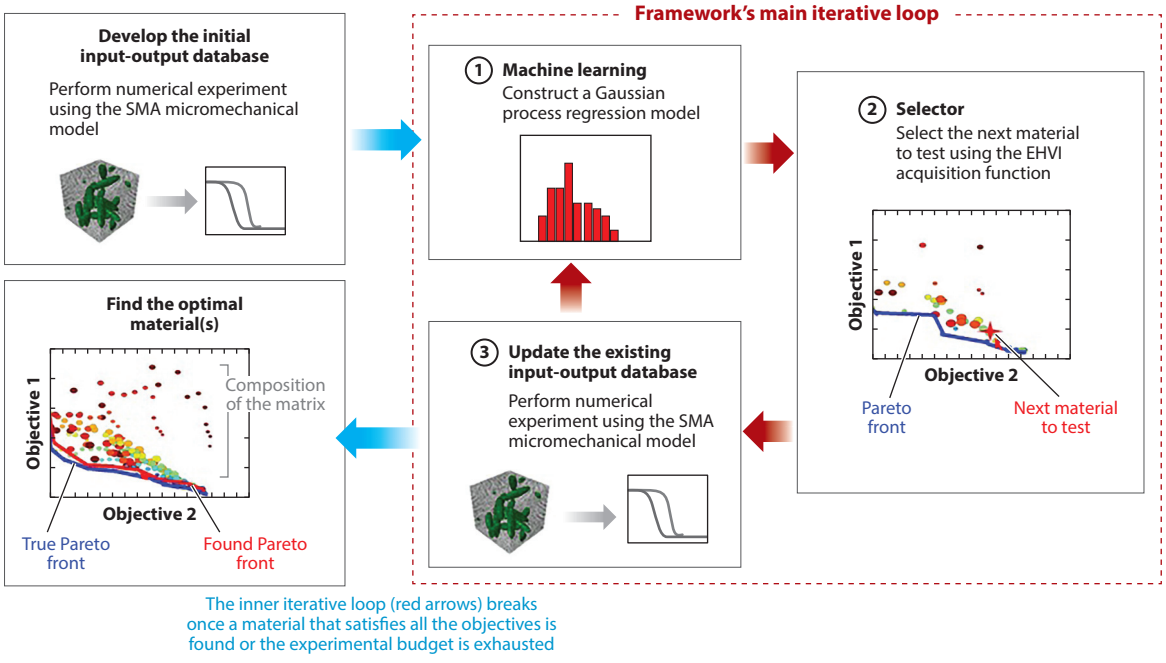


**Figure 5**

Multiobjective Bayesian optimization experimental design framework. The workflow for Bayesian optimization consists of distinct ① machine learning, ② selector, and ③ update steps. The balanced exploration-exploitation of the multiobjective problem was measured using the expected hypervolume improvement (EHVI), which is a scalar quantity that measures the utility of carrying out an experiment that balances the needs to explore and exploit the design space. As in single-objective Bayesian optimization, one starts with initial data that are then used to develop a model of the experiments that accounts for the mean response and variance of observations. By using this model, the EHVI acquisition function is computed, and experiments are selected on the basis of how much the Pareto frontier can be improved. SMA denotes shape memory alloy. Adapted with permission from Solomou et al. (117).

## 5.3. Multi-Information Source Materials Optimization: Model/Information Source Fusion

Frameworks for model-based ICME workflows focus on integrating tools at multiple levels under the assumption that there is a single model, tool, or source of information or metadata relevant to each level of the PSP relations and for each level of hierarchy of materials structure. ICME design and development applications often preclude the use of multiple models for each level or process, which tends to ignore uncertainty of the model form. We regard uncertainty of the model form (regardless of source) as a key ongoing challenge in materials design that has received too little attention from both the materials design and multidisciplinary design optimization communities. Data-centric approaches, in contrast, tend to focus on brute-force exploration of the materials design space, without accounting for the considerable cost associated with such exploration. However, they do tend to mitigate model form uncertainty to some extent since they often do not force preexisting conceptions of models or mechanisms onto the solution and/or they admit a learning process that reduces such constraints.

Recently, one of the authors and collaborators (120) put forward a framework for the multi-information source Bayesian optimization (MISBO) of materials design spaces. By exploiting the statistical correlations between multiple sources of information connecting structure features to properties/performance and by employing knowledge gradient policies (121) over the fused

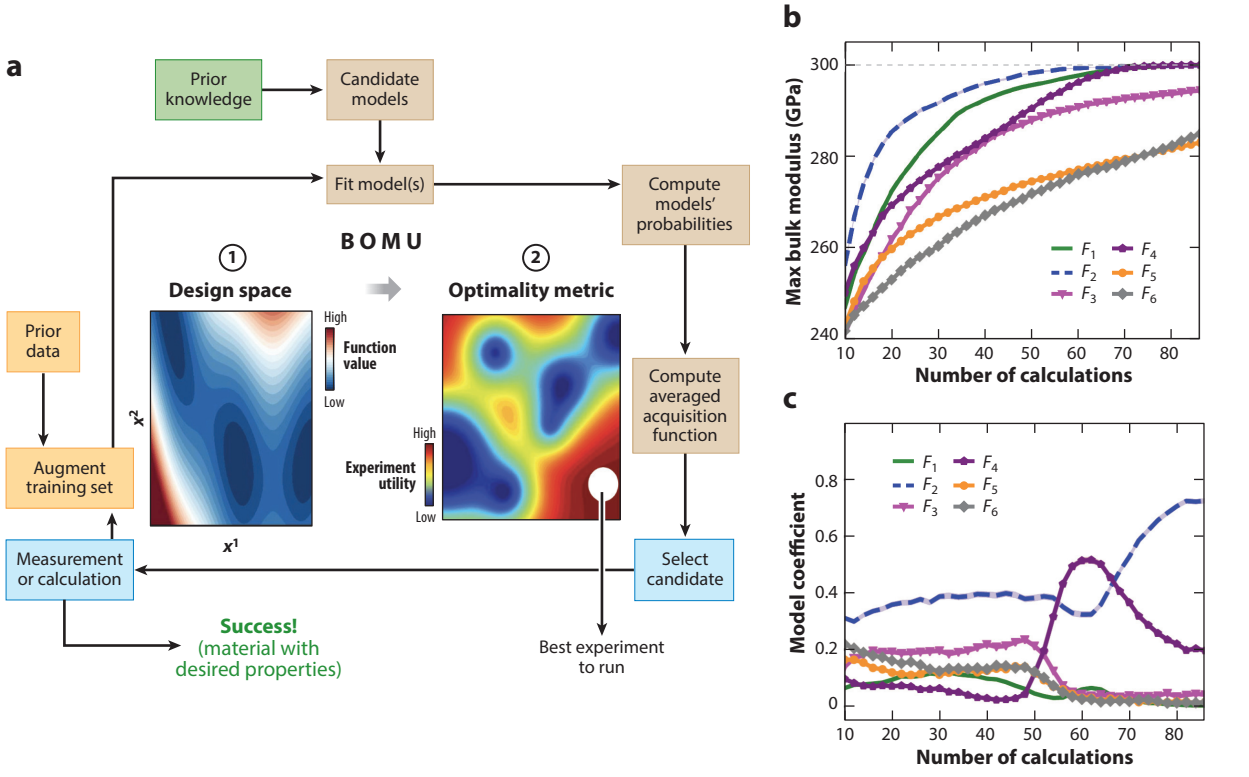


**Figure 6**

Bayesian optimization under model uncertainty (BOMU). (*a*) Initial data and a set of candidate models are used to construct a stochastic representation of an experiment/simulation. Each model is evaluated in a Bayesian sense, and its probability is determined. By using the model probabilities, an effective acquisition function is computed, which is then used to select the next point in the materials design space that needs to be queried. The process is continued iteratively until target is reached or budget is exhausted. The green block represents prior knowledge, orange blocks represent data, brown blocks are related to models, and blue blocks denote actions. (*b*) The performance of each individual model (labeled as $F_i$) against the objective of identifying the crystal structure that has the maximum bulk modulus. (*c*) Individual probabilities of different models and their evolution during the Bayesian optimization sequence. Adapted with permission from Talapatra et al. (118).

models, the framework addresses two problems at once: (*a*) the next-best point in the design space to use and (*b*) the source from which that point should be queried in the (materials) design space. We note that MISBO is different from BOMU mainly because in the latter it is assumed that the different models being considered are statistically independent, while in the former the opposite assumption is made. The framework was demonstrated on the optimization of a dual-phase steel in which different reduced-order physics-based models were fused to discover the optimal microstructure configurations as evaluated by a (ground truth) microstructure-sensitive micromechanical model. In that work, the performance of MISBO given multiple inexpensive information sources in addition to the ground truth was compared against that of conventional BO approaches operating exclusively on the expensive ground truth, and it was found that MISBO was significantly more efficient.

Other groups (122) recently presented multi-information source sequential optimization. For example, Pilania et al. (123) showed that it is possible to combine low-fidelity approximate predictions for materials properties to supplement high-fidelity, expensive computational simulations,

arriving at accurate predictive models at a relatively low cost. The framework in Ghoreishi et al. (120) differs from that of classical multifidelity methods (124) in that in the former there is no assumed hierarchy of information sources and all sources are considered on an equal basis at the outset; what matters is the statistical correlations among them as well as between them and the ground truth. This allows, for example, for the seamless combination of experiments, models, and theories into unified frameworks for materials discovery.

Yet another important practical aspect of ICME PSP workflows involves fusion of information from models and other sources to arrive at a statistically meaningful process for calibration of model parameters. As an example, consider the hierarchy of models of dislocation plasticity. It is desirable not only to exercise models appropriate for different levels of hierarchy as demanded by the properties or responses required for design (e.g., atomistic → elastic constants, discrete dislocation dynamics → work hardening, polycrystal plasticity → effective strength and elastic stiffness of polycrystals) but also to achieve some measure of consistency of model forms and model parameters across scales. To this end, recent work of Talman and collaborators (125) has introduced an approach in which bottom-up atomistic modeling information constrains top-down experimental information to inform the connection of each source of information to mesoscale model parameters in crystal plasticity at the grain scale.

## 6. SOME THOUGHTS REGARDING FUTURE WORK AND PRIORITIES

Much of the attention of the scientific community with regard to the MGI has been drawn toward combinatorial first-principles methods and HT synthesis strategies to facilitate materials discovery, whereas the broader stream of materials development should involve the integration of manufacturing processes and product realization. Materials development has been clearly articulated by ICME as pertaining to the so-called valley of death (126) that inhibits the translation of early-stage materials developments into product deployment. In addition to designing materials with targeted property sets, another major implication of concurrent materials and product design is the capability to design location-specific properties that vary throughout the part in a manner that in some way optimizes performance for a given set of system requirements. Location-specific materials design requires an intimate link between systems design and materials synthesis and processing to achieve the necessary heterogeneity of materials structure as a function of position. Indeed, part shapes and product functionality can change dramatically in this scenario relative to design with homogeneous materials structure and properties manufactured using traditional machining technologies. Additive manufacturing is an application domain in which this concurrency of materials processing and part manufacturing is clear, although it is certainly not the only one.

We have some thoughts on the difference between the next generations of the MGI and ICME and the initial stages (the past decade) of these initiatives in terms of areas of scientific/research focus in materials discovery and design. At the beginning of Section 5, we discuss the increasing role of systems design optimization strategies that may incorporate substantial elements of data science in pursuing materials discovery and design. In our view, much of the materials engineering community has focused heavily this past decade on the issue of digital materials data, including various aspects related to the construction of PSP relations and how to standardize related schema for handling these data. While this will likely continue as a foundational pursuit in the MGI and ICME, we see these initiatives broadening substantially to involve research communities in uncertainty quantification and propagation, optimization methods, design exploration, sensor fusion, advanced data science correlations, metamodeling, and statistical learning approaches.

In addition, the principles of manufacturing cannot be dissociated from materials discovery and development. As they largely lie outside of traditional materials science, physics and chemistry

academic disciplines, and their associated curricula, it is incumbent upon the MGI and ICME communities (university, federal agencies and research laboratories, and industry) to design new types of materials-centric curricula and future workforce development approaches that embed enabling fields such as mathematics, computer science, manufacturing, signal processing, and multidisciplinary design optimization. These new developments will require thoughtful consideration of branching materials science toward (even) more interdisciplinarity (51, 127–129), and there is some indication of universities starting to move in these directions (**http://engineering.buffalo.edu/materials-design-innovation.html**, **http://www.materials.gatech.edu/**, **https://d3em.tamu.edu/**).

### FUTURE ISSUES

Some of the key scientific challenges to be fully addressed to move forward in materials design across various materials classes include the following, which were emphasized in a 2014 MGI workshop (130):

1. Fuse materials PSP linkages with computer-aided design and design for manufacture;
2. Link downstream materials certification requirements more closely to screening protocols in the materials discovery stage;
3. Couple multiscale modeling of hierarchically structured materials more closely with uncertainty quantification and multiobjective systems design strategies;
4. Couple experimental observations at multiple length scales and timescales with modeling and simulation through data science in mutually beneficial ways, particularly with regard to improving understanding of kinetics of unit processes at the nanoscale via in situ measurements;
5. Provide new capabilities to quantify outcomes of materials synthesis and processing by quantifying spatial statistics of microstructures using *n*-point spatial correlations, forming a natural linkage of materials data to modeling and simulation; and
6. Develop HT materials synthesis routes, as well as structure and performance characterization protocols, that provide the critical information needed for simulation-assisted, concurrent design of materials and manufacturing processes.

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## LITERATURE CITED


1. McDowell DL, Panchal J, Choi H-J, Seepersad C, Allen J, Mistree F. 2010. *Integrated Design of Multiscale, Multifunctional Materials and Products*. Oxford, UK: Butterworth-Heinemann
2. McDowell DL, Kalidindi SR. 2016. The materials innovation ecosystem: a key enabler for the Materials Genome Initiative. *MRS Bull.* 41:326–37
3. Hale J. 2006. Boeing 787 from the ground up. *AERO* 4:7
4. Holdren JP. 2011. *Materials Genome Initiative for global competitiveness*. Rep., Natl. Sci. Technol. Counc., Washington, DC
5. Breneman CM, Brinson LC, Schadler LS, Natarajan B, Krein M, et al. 2013. Stalking the materials genome: a data-driven approach to the virtual design of nanostructured polymers. *Adv. Funct. Mater.* 23:5746–52
6. White A. 2012. The Materials Genome Initiative: one year on. *MRS Bull.* 37:715–16
7. Pollock TM, Allison JE, Backman DG, Boyce MC, Gersh M, et al. 2008. *Integrated computational materials engineering: a transformational discipline for improved competitiveness and national security*. Rep. 0-309-12000-4, Natl. Acad. Sci., Washington, DC
8. Olson GB. 1997. Computational design of hierarchically structured materials. *Science* 277:1237–42
9. Pollock TM, Tin S. 2006. Nickel-based superalloys for advanced turbine engines: chemistry, microstructure and properties. *J. Propuls. Power* 22:361–74
10. McDowell DL. 2007. Simulation-assisted materials design for the concurrent design of materials and products. *JOM* 59:21–25
11. Backman DG, Wei DY, Whitis DD, Buczek MB, Finnigan PM, Gao D. 2006. ICME at GE: accelerating the insertion of new materials and processes. *JOM* 58:36–41
12. Rajan K. 2015. Materials informatics: the materials "gene" and big data. *Annu. Rev. Mater. Res.* 45:153–69
13. Lu G, Tadmor EB, Kaxiras E. 2006. From electrons to finite elements: a concurrent multiscale approach for metals. *Phys. Rev. B* 73:024108
14. Curtin WA, Miller RE. 2003. Atomistic/continuum coupling in computational materials science. *Model. Simul. Mater. Sci. Eng.* 11:R33
15. Horstemeyer MF. 2012. *Integrated Computational Materials Engineering (ICME) for Metals: Using Multiscale Modeling to Invigorate Engineering Design with Science*. Hoboken, NJ: Wiley
16. Allen JK, Seepersad C, Choi H, Mistree F. 2006. Robust design for multiscale and multidisciplinary applications. *J. Mech. Des.* 128:832–43
17. Fullwood DT, Niezgoda SR, Adams BL, Kalidindi SR. 2010. Microstructure sensitive design for performance optimization. *Prog. Mater. Sci.* 55:477–562
18. Thompson SC, Paredis CJ. 2010. *An introduction to rational design theory*. Presented at ASME 2010 International Design Engineering Technical Conferences and Computers and Information in Engineering Conference, Aug. 15–18, Montreal
19. Choi H, McDowell DL, Allen JK, Rosen D, Mistree F. 2008. An inductive design exploration method for robust multiscale materials design. *J. Mech. Des.* 130:031402
20. Voorhees P, Spanos G. 2015. *Modeling across scales: a roadmapping study for connecting materials models and simulations across length and time scales*. Rep., TMS
21. Olson GB. 2000. Designing a new material world. *Science* 288:993–98
22. Adams BL, Kalidindi SR, Fullwood DT. 2012. *Microstructure Sensitive Design for Performance Optimization*. Oxford, UK: Butterworth-Heinemann
23. Fast T, Knezevic M, Kalidindi SR. 2008. Application of microstructure sensitive design to structural components produced from hexagonal polycrystalline metals. *Comput. Mater. Sci.* 43:374–83
24. Johnson L, Arróyave R. 2016. An inverse design framework for prescribing precipitation heat treatments from a target microstructure. *Mater. Des.* 107:7–17
25. McDowell DL, Olson G. 2008. Concurrent design of hierarchical materials and structures. In *Scientific Modeling and Simulations*, ed. S Yip, T Diaz de la Rubia, pp. 207–40. Dordrecht, Neth.: Springer
26. Panchal JH, Kalidindi SR, McDowell DL. 2013. Key computational modeling issues in integrated computational materials engineering. *Comput. Aided Des.* 45:4–25

27. Chernatynskiy A, Phillpot SR, LeSar R. 2013. Uncertainty quantification in multiscale simulation of materials: a prospective. *Annu. Rev. Mater. Res.* 43:157–82
28. Yabansu YC, Steinmetz P, Hötzer J, Kalidindi SR, Nestler B. 2017. Extraction of reduced-order process-structure linkages from phase-field simulations. *Acta Mater.* 124:182–94
29. Senn M. 2015. An integrated surrogate modeling approach for materials and process design. In *Proceedings of the 3rd World Congress on Integrated Computational Materials Engineering (ICME 2015)*, Vol. 35, ed. W Poole, S Christensen, S Kalidindi, A Luo, J Martin, et al., pp. 331–38. Hoboken, NJ: Wiley
30. Seepersad CC, Kumar RS, Allen JK, Mistree F, McDowell DL. 2004. Multifunctional design of prismatic cellular materials. *J. Comput. Aided Mater. Des.* 11:163–81
31. Taguchi G. 1993. *Taguchi on Robust Technology Development: Bringing Quality Upstream*. New York: ASME
32. Taguchi G. 1986. *Introduction to Quality Engineering: Designing Quality into Products and Processes*. White Plains, NY: Qual. Resour.
33. Chen W, Allen JK, Tsui K-L, Mistree F. 1996. Procedure for robust design: minimizing variations caused by noise factors and control factors. *J. Mech. Des.* 118:478–85
34. Nair VN, Abraham B, MacKay J, Box G, Kacker RN, et al. 1992. Taguchi's parameter design: a panel discussion. *Technometrics* 34:127–61
35. Ng KK, Tsui K-L. 1992. Expressing variability and yield with a focus on the customer. *Qual. Eng.* 5:255–67
36. Murphy TE, Tsui K-L, Allen JK. 2005. A review of robust design methods for multiple responses. *Res. Eng. Des.* 15:201–15
37. Tsui K-L. 1999. Robust design optimization for multiple characteristic problems. *Int. J. Prod. Res.* 37:433–45
38. Agrawal A, Choudhary A. 2016. Perspective: materials informatics and big data: realization of the "fourth paradigm" of science in materials science. *APL Mater.* 4:053208
39. Xiang X-D, Sun X, Briceno G, Lou Y, Wang K-A, et al. 1995. A combinatorial approach to materials discovery. *Science* 268:1738–40
40. McFarland EW, Weinberg WH. 1999. Combinatorial approaches to materials discovery. *Trends Biotechnol.* 17:107–15
41. Engstrom JR, Weinberg WH. 2000. Combinatorial materials science: paradigm shift in materials discovery and optimization. *AIChE J.* 46:2–5
42. Koinuma H, Takeuchi I. 2004. Combinatorial solid-state chemistry of inorganic materials. *Nat. Mater.* 3:429–38
43. Xiong Z, He Y, Hattrick-Simpers JR, Hu J. 2017. Automated phase segmentation for large-scale X-ray diffraction data using a graph-based phase segmentation (GPhase) algorithm. *ACS Comb. Sci.* 19:137–44
44. Curtarolo S, Hart GL, Nardelli MB, Mingo N, Sanvito S, Levy O. 2013. The high-throughput highway to computational materials design. *Nat. Mater.* 12:191–201
45. Pulido A, Chen L, Kaczorowski T, Holden D, Little MA, et al. 2017. Functional materials discovery using energy–structure–function maps. *Nature* 543:657–64
46. Hautier G, Jain A, Ong SP. 2012. From the computer to the laboratory: materials discovery and design using first-principles calculations. *J. Mater. Sci.* 47:7317–40
47. Saal JE, Kirklin S, Aykol M, Meredig B, Wolverton C. 2013. Materials design and discovery with high-throughput density functional theory: the Open Quantum Materials Database (OQMD). *JOM* 65:1501–9
48. Cohen AJ, Mori-Sánchez P, Yang W. 2008. Insights into current limitations of density functional theory. *Science* 321:792–94
49. Rajan K. 2013. *Informatics for Materials Science and Engineering: Data-Driven Discovery for Accelerated Experimentation and Application*. Oxford, UK: Butterworth-Heinemann
50. Ashby MF, Cebon D. 1993. Materials selection in mechanical design. *J. Phys. IV* 3:C7-1–C7-9
51. Arroyave R, Shields S, Chang C-N, Fowler D, Malak R, Allaire D. 2018. Interdisciplinary research on designing engineering materials systems: results from a National Science Foundation workshop. *J. Mech. Des.* 140:110801

52. Allison J. 2011. Integrated computational materials engineering: a perspective on progress and future steps. *JOM* 63:15–18
53. Srivastava A, Ghassemi-Armaki H, Sung H, Chen P, Kumar S, Bower AF. 2015. Micromechanics of plastic deformation and phase transformation in a three-phase TRIP-assisted advanced high strength steel: experiments and modeling. *J. Mech. Phys. Solids* 78:46–69
54. Li SY. 2013. *A computational-based approach for the design of TRIP steels*. PhD Thesis, Texas A&M Univ.
55. Saunders N, Miodownik AP. 1998. *CALPHAD (Calculation of Phase Diagrams): A Comprehensive Guide*. Oxford, UK/New York: Pergamon Press. 479 pp.
56. Cao W, Chen S-L, Zhang F, Wu K, Yang Y, et al. 2009. PANDAT software with PanEngine, PanOptimizer and PanPrecipitation for multi-component phase diagram calculation and materials property simulation. *CALPHAD* 33:328–42
57. Andersson J-O, Helander T, Höglund L, Shi P, Sundman B. 2002. Thermo-Calc & DICTRA, computational tools for materials science. *CALPHAD* 26:273–312
58. Sundman B, Kattner UR, Palumbo M, Fries SG. 2015. OpenCalphad: a free thermodynamic software. *Integr. Mater. Manuf. Innov.* 4:1
59. Turchi PE, Abrikosov IA, Burton B, Fries SG, Grimvall G, et al. 2007. Interface between quantum-mechanical-based approaches, experiments, and CALPHAD methodology. *CALPHAD* 31:4–27
60. Bigdeli S. 2017. *Developing the third generation of Calphad databases: What can ab-initio contribute?* Dr. Thesis, KTH R. Inst. Technol.
61. Campbell CE, Boettinger WJ, Kattner UR. 2002. Development of a diffusion mobility database for Ni-base superalloys. *Acta Mater.* 50:775–92
62. Lu X-G, Selleby M, Sundman B. 2005. Assessments of molar volume and thermal expansion for selected bcc, fcc and hcp metallic elements. *CALPHAD* 29:68–89
63. Kim D, Shang S-L, Liu Z-K. 2009. Effects of alloying elements on elastic properties of Ni by first-principles calculations. *Comput. Mater. Sci.* 47:254–60
64. Xu W, Rivera-Díaz-del-Castillo P, Van der Zwaag S. 2008. Designing nanoprecipitation strengthened UHS stainless steels combining genetic algorithms and thermodynamics. *Comput. Mater. Sci.* 44:678–89
65. Tancret F. 2012. Computational thermodynamics and genetic algorithms to design affordable γ′-strengthened nickel–iron based superalloys. *Model. Simul. Mater. Sci. Eng.* 20:045012
66. Menou E, Toda-Caraballo I, Rivera-Díaz-del-Castillo PEJ, Pineau C, Bertrand E, et al. 2018. Evolutionary design of strong and stable high entropy alloys using multi-objective optimisation based on physical models, statistics and thermodynamics. *Mater. Des.* 143:185–95
67. Jha R, Dulikravich G, Colaco M, Fan M, Schwartz J, Koch C. 2017. Magnetic alloys design using multi-objective optimization. In *Properties and Characterization of Modern Materials*, ed. A Öchsner, H Altenbach, pp. 261–84. Singapore: Springer
68. Lu Q, Xu W, van der Zwaag S. 2014. The design of a compositionally robust martensitic creep-resistant steel with an optimized combination of precipitation hardening and solid-solution strengthening for high-temperature use. *Acta Mater.* 77:310–23
69. Gheribi AE, Harvey J-P, Bélisle E, Robelin C, Chartrand P, et al. 2016. Use of a biobjective direct search algorithm in the process design of material science applications. *Optim. Eng.* 17:27–45
70. Lu Q, van der Zwaag S, Xu W. 2017. Charting the 'composition–strength' space for novel austenitic, martensitic and ferritic creep resistant steels. *J. Mater. Sci. Technol.* 33:1577–81
71. Li S, Kattner UR, Campbell CE. 2017. A computational framework for material design. *Integr. Mater. Manuf. Innov.* 6:229–48
72. Toda-Caraballo I, Galindo-Nava EI, Rivera-Díaz-del-Castillo PEJ. 2013. Unravelling the materials genome: symmetry relationships in alloy properties. *J. Alloys Comp.* 566:217–28
73. Xiong W, Olson GB. 2015. Integrated computational materials design for high-performance alloys. *MRS Bull.* 40:1035–44
74. Zhu J, Liu Z, Vaithyanathan V, Chen L. 2002. Linking phase-field model to CALPHAD: application to precipitate shape evolution in Ni-base alloys. *Scr. Mater.* 46:401–6
75. Kim K, Roy A, Gururajan M, Wolverton C, Voorhees P. 2017. First-principles/phase-field modeling of θ′ precipitation in Al-Cu alloys. *Acta Mater.* 140:344–54

76. Wen Y, Wang B, Simmons J, Wang Y. 2006. A phase-field model for heat treatment applications in Ni-based alloys. *Acta Mater*. 54:2087–99
77. Kampmann R, Wagner R. 1984. Kinetics of precipitation in metastable binary alloys: theory and application to Cu–1.9 at % Ti and Ni–14 at % Al. In *Decomposition of Alloys: The Early Stages (Proceedings of the 2nd Acta-Scripta Metallurgica Conference)*, ed. P Haasen, V Gerold, R Wagner, MF Ashby, pp. 91–103. Oxford, UK: Pergamon Press
78. Kozeschnik E, Svoboda J, Fischer F. 2004. Modified evolution equations for the precipitation kinetics of complex phases in multi-component systems. *CALPHAD* 28:379–82
79. Kozeschnik E, Svoboda J, Fratzl P, Fischer F. 2004. Modelling of kinetics in multi-component multi-phase systems with spherical precipitates. II. Numerical solution and application. *Mater. Sci. Eng. A* 385:157–65
80. Svoboda J, Fischer F, Fratzl P, Kozeschnik E. 2004. Modelling of kinetics in multi-component multi-phase systems with spherical precipitates. I. Theory. *Mater. Sci. Eng. A* 385:166–74
81. Ghosh G, Olson G. 2007. Integrated design of Nb-based superalloys: ab initio calculations, computational thermodynamics and kinetics, and experimental results. *Acta Mater*. 55:3281–303
82. Zhang C, Cao W, Chen S-L, Zhu J, Zhang F, et al. 2014. Precipitation simulation of AZ91 alloy. *JOM* 66:389–96
83. Lang P, Rath M, Kozeschnik E, Rivera-Díaz-del-Castillo PEJ. 2015. Modelling the influence of austenitisation temperature on hydrogen trapping in Nb containing martensitic steels. *Scr. Mater.* 101:60–63
84. Martin JH, Yahata BD, Hundley JM, Mayer JA, Schaedler TA, Pollock TM. 2017. 3D printing of high-strength aluminium alloys. *Nature* 549:365–69
85. Galindo-Nava E, Rainforth W, Rivera-Díaz-del-Castillo PEJ. 2016. Predicting microstructure and strength of maraging steels: elemental optimisation. *Acta Mater*. 117:270–85
86. Houskamp JR, Proust G, Kalidindi SR. 2007. Integration of microstructure-sensitive design with finite element methods: elastic-plastic case studies in FCC polycrystals. *Int. J. Multiscale Comput. Eng.* 5:261–72
87. Kalidindi SR. 2015. Data science and cyberinfrastructure: critical enablers for accelerated development of hierarchical materials. *Int. Mater. Rev.* 60:150–68
88. Kalidindi SR. 2012. Computationally efficient, fully coupled multiscale modeling of materials phenomena using calibrated localization linkages. *ISRN Mater. Sci.* 2012:305692
89. Wheeler D, Brough D, Fast T, Kalidindi S, Reid A. 2014. *PyMKS: Materials Knowledge System in Python*. **https://doi.org/10.6084/m9.figshare.1015761**
90. Niezgoda S, Fullwood D, Kalidindi S. 2008. Delineation of the space of 2-point correlations in a composite material system. *Acta Mater*. 56:5285–92
91. McDowell D, Ghosh S, Kalidindi S. 2011. Representation and computational structure-property relations of random media. *JOM* 63:45–51
92. Niezgoda SR, Kanjarla AK, Kalidindi SR. 2013. Novel microstructure quantification framework for databasing, visualization, and analysis of microstructure data. *Integr. Mater. Manuf. Innov.* 2:3
93. Gupta A, Cecen A, Goyal S, Singh AK, Kalidindi SR. 2015. Structure–property linkages using a data science approach: application to a non-metallic inclusion/steel composite system. *Acta Mater*. 91:239–54
94. Fast T, Niezgoda SR, Kalidindi SR. 2011. A new framework for computationally efficient structure–structure evolution linkages to facilitate high-fidelity scale bridging in multi-scale materials models. *Acta Mater*. 59:699–707
95. Yabansu YC, Kalidindi SR. 2015. Representation and calibration of elastic localization kernels for a broad class of cubic polycrystals. *Acta Mater*. 94:26–35
96. Kröner E. 1977. Bounds for effective elastic moduli of disordered materials. *J. Mech. Phys. Solids* 25:137–55
97. Kröner E. 2001. Benefits and shortcomings of the continuous theory of dislocations. *Int. J. Solids Struct.* 38:1115–34
98. Korenberg MJ, Hunter IW. 1996. The identification of nonlinear biological systems: Volterra kernel approaches. *Ann. Biomed. Eng.* 24:250–68

99. Rudd RE, Broughton JQ. 2000. Concurrent coupling of length scales in solid state systems. *Phys. Status Solid. B* 217:251–91
100. Paulson NH, Priddy MW, McDowell DL, Kalidindi SR. 2017. Reduced-order structure-property linkages for polycrystalline microstructures based on 2-point statistics. *Acta Mater.* 129:428–38
101. Priddy MW, Paulson NH, Kalidindi SR, McDowell DL. 2017. Strategies for rapid parametric assessment of microstructure-sensitive fatigue for HCP polycrystals. *Int. J. Fatigue* 104:231–42
102. Horstemeyer M, Wang P. 2003. Cradle-to-grave simulation-based design incorporating multiscale microstructure-property modeling: reinvigorating design with science. *J. Comput. Aided Mater. Des.* 10:13–34
103. Choi H-J. 2005. *A robust design method for model and propagated uncertainty*. PhD Diss., Sch. Mech. Eng., Georgia Inst. Technol.
104. Ellis BD, McDowell DL. 2017. Application-specific computational materials design via multiscale modeling and the inductive design exploration method (IDEM). *Integr. Mater. Manuf. Innov.* 6:9–35
105. Kern PC, Priddy MW, Ellis BD, McDowell DL. 2017. pyDEM: a generalized implementation of the inductive design exploration method. *Mater. Des.* 134:293–300
106. Wang R, Nellippallil AB, Wang G, Yan Y, Allen JK, Mistree F. 2018. Systematic design space exploration using a template-based ontological method. *Adv. Eng. Inform.* 36:163–77
107. Ramprasad R, Batra R, Pilania G, Mannodi-Kanakkithodi A, Kim C. 2017. Machine learning in materials informatics: recent applications and prospects. *NPJ Comput. Mater.* 3:54
108. Forsmark JH, Zindel JW, Godlewski L, Zheng J, Allison JE, Li M. 2015. Using quality mapping to predict spatial variation in local properties and component performance in Mg alloy thin-walled high-pressure die castings: an ICME approach and case study. *Integr. Mater. Manuf. Innov.* 4:6
109. Lookman T, Balachandran PV, Xue D, Hogden J, Theiler J. 2017. Statistical inference and adaptive design for materials discovery. *Curr. Opin. Solid State Mater. Sci.* 21:121–28
110. Brochu E, Cora VM, De Freitas N. 2010. A tutorial on Bayesian optimization of expensive cost functions, with application to active user modeling and hierarchical reinforcement learning. arXiv:1012.2599 [cs.LG]
111. Good IJ, Hacking I, Jeffrey R, Törnebohm H. 1966. The estimation of probabilities: an essay on modern Bayesian methods. *Synthese* 16:234–44
112. Jones DR, Schonlau M, Welch WJ. 1998. Efficient global optimization of expensive black-box functions. *J. Glob. Optim.* 13:455–92
113. Xue D, Balachandran PV, Hogden J, Theiler J, Xue D, Lookman T. 2016. Accelerated search for materials with targeted properties by adaptive design. *Nat. Commun.* 7:11241
114. Xue D, Balachandran PV, Yuan R, Hu T, Qian X, et al. 2016. Accelerated search for $BaTiO_3$-based piezoelectrics with vertical morphotropic phase boundary using Bayesian learning. *PNAS* 113:13301–6
115. Xue D, Xue D, Yuan R, Zhou Y, Balachandran PV, et al. 2017. An informatics approach to transformation temperatures of NiTi-based shape memory alloys. *Acta Mater.* 125:532–41
116. Emmerich MT, Deutz AH, Klinkenberg JW. 2011. *Hypervolume-based expected improvement: monotonicity properties and exact computation*. Presented at 2011 IEEE Congress on Evolutionary Computation (CEC), June 5–8, New Orleans
117. Solomou A, Zhao G, Boluki S, Joy JK, Qian X, et al. 2018. Multi-objective Bayesian materials discovery: application on the discovery of precipitation strengthened NiTi shape memory alloys through micromechanical modeling. *Mater. Des.* 160:810–27
118. Talapatra A, Boluki S, Duong T, Qian X, Dougherty E, Arróyave R. 2018. Autonomous efficient experiment design for materials discovery with Bayesian model averaging. *Phys. Rev. Mater.* 2:113803
119. Talapatra A, Boluki S, Duong T, Qian X, Dougherty E, Arróyave R. 2018. Towards an autonomous efficient materials discovery framework: an example of optimal experiment design under model uncertainty. arXiv:1803.05460
120. Ghoreishi SF, Molkeri A, Srivastava A, Arroyave R, Allaire D. 2018. Multi-information source fusion and optimization to realize ICME: application to dual-phase materials. *J. Mech. Des.* 140:111409

121. Frazier P, Powell W, Dayanik S. 2009. The knowledge-gradient policy for correlated normal beliefs. *INFORMS J. Comput.* 21:599–613
122. Chen S, Jiang Z, Yang S, Chen W. 2016. Multimodel fusion based sequential optimization. *AIAA J.* 55:241–54
123. Pilania G, Gubernatis JE, Lookman T. 2017. Multi-fidelity machine learning models for accurate bandgap predictions of solids. *Comput. Mater. Sci.* 129:156–63
124. Qian PZ, Wu CJ. 2008. Bayesian hierarchical modeling for integrating low-accuracy and high-accuracy experiments. *Technometrics* 50:192–204
125. Tallman AE, Swiler LP, Wang Y, McDowell DL. 2017. Reconciled top-down and bottom-up hierarchical multiscale calibration of bcc Fe crystal plasticity. *Int. J. Multiscale Comput. Eng.* 15:505–23
126. McDowell DL. 2014. Critical path issues in ICME. In *Models, Databases and Simulation Tools Needed for Realization of Integrated Computational Materials Engineering*, ed. SM Arnold, TT Wong, pp. 31–37. Materials Park, OH: ASM Int.
127. Chang C-N, Semma B, Fowler D, Arroyave R. 2017. *An interdisciplinary graduate education model for the materials engineering field*. Presented at American Society of Engineering Education Conference and Exposition, June 25–28, Columbus, OH
128. Chang C-N, Semma B, Pardo ML, Fowler D, Shamberger P, Arroyave R. 2017. Data-enabled discovery and design of energy materials (D3EM): structure of an interdisciplinary materials design graduate program. *MRS Adv.* 2:1693–98
129. Fowler D, Arroyave R, Ross J, Malak R, Banerjee S. 2017. Looking outwards from the "central science": an interdisciplinary perspective on graduate education in materials chemistry. In *Educational and Outreach Projects from the Cottrell Scholars*, Vol. 1, ed. R Waterman, A Feig, pp. 65–89. Washington, DC: Am. Chem. Soc.
130. Georgia Inst. Technol., Univ. Wisconsin, Univ. Michigan. 2014. *Building an integrated materials genome initiative accelerator network*. Rep. of Workshop, June 5–6, Atlanta, GA. **http://acceleratornetwork.org/wp-uploads/2014/09/MAN-MGI-REPORT-2015.pdf**

## NOTE ADDED IN PROOF

In Section 5.1 we highlight IDEM as an example of a practical design optimization strategy or framework for formally organizing top-down design searches on the basis of bottom-up PSP relations. Another strategy has been developed recently in this regard, namely goal-oriented inverse design (GoID). GoID is a requirements-driven, top-down strategy for designing a system and associated subsystems by taking a goal-oriented, inverse approach (131). Using this strategy, designers start with the end goals for both the product and material PSP relations and then design the system to satisfy the specified end goals as closely as possible by exploring the design space. The GoID strategy offers an increase in problem size, improved flexibility in the design of the various processes involved, the capability to identify robust satisficing solutions for multiple conflicting goals (132), and the capability to visualize and explore solutions that are relatively insensitive to uncertainty.

131. Nellippallil AB, Rangaraj V, Gautham BP, Singh AK, Allen JK, Mistree F. 2018. An inverse, decision-based design method for integrated design exploration of materials, products, and manufacturing processes. *ASME J. Mech. Design* 140(11):111403
132. Nellippallil AB, Mohan P, Allen JK, Mistree F. 2018. *Robust concept exploration of materials, products and associated manufacturing processes*. Presented at ASME Design Automation Conference, Pap. DETC2018-85913, Quebec City, Can.