Machine learning-assisted design of material properties

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Abstract

Designing functional materials requires a deep search through multidimensional spaces for system parameters that yield desirable material properties. For cases where conventional parameter sweeps or trial-and-error sampling are impractical, inverse methods that frame design as a constrained optimization problem present an attractive alternative. However, even efficient algorithms require time- and resource-intensive characterization of material properties many times during optimization, imposing a design bottleneck. Approaches that incorporate machine learning can help address this limitation and accelerate the discovery of materials with targeted properties. Here, we review how to leverage machine learning to reduce dimensionality to effectively explore design space, accelerate property evaluation, and generate unconventional material structures with optimal properties. We also discuss promising future directions, including integration of machine learning into multiple stages of a design algorithm and interpretation of machine learning models to understand how design parameters relate to material properties.

1 Introduction

Functional materials are strategically designed to exhibit technologically useful properties. Examples abound, including ionic liquids for carbon capture, nanomaterials for energy storage and catalysis, organic materials for photonic applications, and porous materials for hydrogen storage. In most cases, the properties of interest derive from the physical and chemical nature of their constituent building-blocks as well as their spatial organization (i.e., structure). The characteristics of dopants and additives as well as processing conditions affecting structure impact performance of materials for photovoltaic devices.
Microstructure-property relationships have been extensively explored for the design of other material classes including metal alloys and self-assembled block copolymers. A unifying aspect of materials design is its focus on systematic determination of points in the “design space” of experimentally adjustable parameters corresponding to structures and properties optimized for a particular application.

In principle, materials with desirable properties can be discovered using parameter sweeps over the design space. Individual samples must be synthesized or modeled computationally — and their properties measured or simulated — for each set of candidate design parameters. Repeating these steps many times with different parameter choices allows one to screen for materials exhibiting targeted properties. However, for most materials of engineering interest, there are many possible parameters to vary, and sweeps covering the corresponding high-dimensional design spaces are impractical. This challenge has been addressed in part by posing materials design as an inverse problem to be solved using methods of numerical optimization to efficiently navigate the design space. Commonly used algorithms iteratively optimize an objective function formulated based on the desired material properties. At each iteration, the property is measured for the current point in the design space, and the optimizer selects new points to investigate until the algorithm achieves convergence to an optimal solution, within specified tolerances. However, even with sophisticated inverse methods, it may be prohibitively expensive to converge to solutions that satisfy design objectives.

In this context, machine learning (ML) is beginning to provide powerful new capabilities for the computational design of materials with targeted properties. For example, ML can be used to train a model that replaces the direct computational evaluation of the property of interest, which significantly decreases the time needed for each iteration of an
optimization routine.\textsuperscript{26–29} There have been several recent reviews that discuss other ways in which ML can be incorporated into an inverse framework to enhance materials design, including using ML to generate new molecules and materials, and to aid the optimizer for prioritized search of design spaces.\textsuperscript{30–32} Other reviews have focused on ML-assisted design for specific classes of materials, including photonic nanostructures,\textsuperscript{33–35} chemical compounds,\textsuperscript{36–38} and self-assembled soft materials\textsuperscript{22, 24, 25} as well as how ML might be used for high-throughput experimental investigations.\textsuperscript{39}

Here, we discuss recent advances in ML strategies to design materials with targeted properties. Specifically, we explore how ML approaches vary depending on the representation of the design space, as shown in Figure 1. Section 2 highlights property design using a low-dimensional representation of the high-dimensional design space. Here, ML is used primarily to reduce the dimensionality of the design space and predict material properties. Section 3 focuses on design solely within the high-dimensional design space, where ML is primarily used to aid an optimizer navigate the space. In Section 4, we outline some promising directions for ML-assisted property design, including combining different ML strategies into a single design framework and improving the interpretability of ML models for design.

2 Property design using low-dimensional representations

To fully characterize a complex material, a high-dimensional representation would be required, including, e.g., positions, orientations, and connectivity of the building blocks. Fortunately, this level of detail is rarely necessary, and material properties can be expressed as functions of far fewer parameters with sufficient accuracy. These parameters form a “latent space,” a low-dimensional representation of the design parameters obtained
by combining or removing features in the original design space. If the latent space retains the information necessary to compute a material property, then it can serve as a low-dimensional proxy for its high-dimensional counterpart for materials design. This is advantageous because it (i) simplifies the quantitative mapping between the design space and the corresponding property compared to that using the high-dimensional
representation and (ii) reduces the number of design parameters an optimizer must modify when navigating the latent space. This section highlights two ways in which ML strategies leveraging low-dimensional latent representations have been implemented to enhance design of material properties. First, we discuss how generative ML models can be used to propose new, nonintuitive material designs with optimal properties directly from the latent space. Second, we explore how ML-based surrogate models quantitatively relate low-dimensional descriptors to the properties.

2.1 Generative models with latent representation

ML-based models can be constructed for learning a low-dimensional latent space on to which a high-dimensional, detailed design can be projected as well as to reconstruct a design in original representation from any point in the latent space. Inverse schemes can leverage these generative capabilities to search through the latent space, rather than the high-dimensional space, and potentially construct new materials exhibiting desired properties from optimal latent points. Such generative models have been primarily applied for topology optimization and molecular design.\textsuperscript{32,40,41} Figure 2 shows two examples of what a high-dimensional representation might look like. In Figure 2a, a multiphase material is characterized by the spatial distribution of its two phases, and the high-dimensional representation consists of a digitized array of pixels, each assigned one of the two phases. Figure 2b shows a molecular structure whose high-dimensional representation contains the positions or connectivity of all atoms, e.g. in the Simplified Molecular Input Line Entry System (SMILES) representation. Generative models project these representations down to just a few latent parameters that retain enough information about the spatial features of the topologies (Figure 2a) or the chemical and structural features of the molecules.
Figure 2: Machine learning–enabled generation and design of topologies and molecules by latent space sampling. (a) Unsupervised learning of generative latent space representation. A topology or molecule in the original representation is converted into a vector in the latent space by use of an encoder. The decoder then reconstructs the corresponding design in the original representation from the latent representation. Once learning is complete, an iterative method screens the latent space for target properties, with the trained decoder serving as a generative model. (b) Supervised learning of latent space representation. The encoder and decoder are trained jointly with a feedforward neural network–based regressor that predicts a material property from the latent representation. The trained regressor then predicts material properties directly during iterative screening of the latent space to design target materials. Panel a adapted with permission from Reference 50, copyright Walter de Gruyter. Panel b adapted with permission from Reference 53.

(Figure 2b). As a result, materials with similar structural motifs typically lie close to each other in the latent space, even if they appear dissimilar or “far” from one another in the high-dimensional representation. This feature is useful because we can perform simple operations in the continuous latent space, like perturbations from a single point or interpolations between two points, to propose new high-dimensional representations that may have similar, or perhaps enhanced, properties compared to previously studied materials.
The latent variables are learned by training two separate components (Figure 2): an encoder which projects a high-dimensional representation of a material to a low-dimensional vector of latent parameters and a decoder or generator which uses a latent vector as input to reconstruct a material in the original high-dimensional representation. The encoder and decoder networks are jointly trained with an unlabeled dataset by minimizing the reconstruction loss, which measures the difference between the original structures in the dataset and the corresponding reconstructed structures. Because the latent representation should facilitate the design of realistic materials, it is helpful if the latent space possesses the property that a random vector fed to the decoder generates physically realistic and meaningful molecules and structures. To ensure this, the learned latent space is also forced to match a predefined target distribution during the training of encoder and decoder. The overall training loss for the model accounts for not only the reconstruction loss but also this latent loss, defined based on the difference between the latent space distribution and the target distribution.  

There are various generative architectures that have been useful for materials design. With a variational autoencoder (VAE) architecture, the latent space is forced to match a Gaussian distribution. VAEs have been employed for generating material topologies or molecular chemistries for property design in mechanical metamaterials, drug discovery, and thermoelectric materials. The fixed Gaussian form of the latent space distribution progressively slows the search for optimal solutions as additional constraints on design parameters are introduced, and so strategies which allow for more control of the latent space distribution are desirable for multi-constrained problems. One way to address this challenge is to adopt adversarial autoencoders (AAEs) (a combination of VAE and generative adversarial networks (GANs), an approach that has been sucessfully
demonstrated for multi-constrained optimization of the optical response of metastructures within a complex design landscape.\textsuperscript{50, 51}

In these unsupervised generative strategies, the low-dimensional latent space is discovered independently of any material property of interest. As shown in Figure 2a, the latent space is used to generate structures in the high-dimensional representation, from which a material property can then be characterized directly in experiments or simulations. Though navigating the low-dimensional latent space reduces the number of iterations during an optimization, if measuring the material property is the time-consuming bottleneck, it will still be challenging to converge the optimization. This challenge has been addressed by using supervised methods to train a generative ML model to rapidly compute material properties directly using a point in latent space as input. For example, Figure 2b shows a feedforward regressor trained jointly with an encoder and decoder to learn the latent space representation that best predicts a target material property. The regressor can then be used to quickly compute material properties as an optimizer navigates the latent space. Because this approach completely avoids measuring material properties in simulations or experiments at every iteration, it can significantly accelerate materials design by reducing both the number of iterations and the time per iteration. By training the property predictor jointly with the VAE, the latent variables learned by the model are such that the topological structures or molecular designs exhibiting similar properties will be distributed close together in the latent space. As a result, it is possible to identify principal axes in latent space along which a material property varies, which can greatly simplify the search for optimal materials.\textsuperscript{52, 54} Fully connected neural networks serving as property predictors coupled with a generative VAE model have been successfully employed for design of metamaterials with desired distortion responses.\textsuperscript{52}
drug-like molecules\textsuperscript{47,53} inorganic crystals for thermoelectric materials\textsuperscript{48} metal–organic framework structures for gas separation applications\textsuperscript{54} and high thermal conductivity alloys\textsuperscript{55} However, as opposed to the unsupervised training of a VAE architecture, the supervised training of the structure-property regressor component in conjunction with the VAE network requires generation of labeled structural datasets.

### 2.2 Forward predictive modeling

ML has been particularly useful for rapidly predicting material properties. Once a ML model is trained, evaluating a material property using the model is significantly faster than measuring the property in an experiment or computing it in a simulation. This is promising for materials design, as the ML model can replace experiments and simulations to accelerate each iteration of an optimization scheme. To train a ML model, we require a large data set linking inputs to the resulting material properties. However, the choice of input is extremely important. In many cases, using the original, high-dimensional representation of parameter space would require impractically large training sets to adequately sample, and inadequate sampling leads to trained models with inaccurate predictions\textsuperscript{56–58}. A more efficient approach is to identify low-dimensional features used as either an input to the ML model or an intermediate layer in the ML architecture. Since the compressed features constituting relevant combinations of the original design parameters help preserve symmetries (e.g. rotational and translational invariance in topologies), this strategy requires much smaller training sets, alleviating the need to explicitly introduce the symmetric variants described in the original representation.

There are two main approaches to finding a low-dimensional representation for material property prediction. The first (\textbf{Figure 3a}) involves first creating a pool of candidate
Figure 3: Two strategies for using ML to predict material properties by leveraging a low-dimensional space of descriptors: (a) First, a pool of candidate descriptors is created by hand, and then ML methods are used to reduce the pool. (b) The low-dimensional set of descriptors is discovered directly during training, without the need to construct a candidate pool. The trained network, obtained with either strategy, is then integrated into an iterative scheme to design (a) copolymers or (b) nanoparticle configurations with the desired material properties. Abbreviations: ML, machine learning; Tg, glass transition temperature; 3HB, 3-hydroxybutyrate; 3HP, 3-hydroxypropionate; 4HB, 4-hydroxybutyrate. Panel a adapted with permission from Reference 68, copyright 2019 American Chemical Society. Panel b adapted with permission from Reference 74, copyright 2021 American Chemical Society.

low-dimensional descriptors and then using ML to reduce the pool and find the descriptors most relevant for predicting a target property. In this regard, the hand-crafted features hypothesized to capture most of the material information influencing the property of interest
are usually chosen as the candidate descriptors. For example, the glass transition temperature $T_g$ of a polymer is a complex property influenced by various structural and compositional features of the polymer. However, instead of a fully-detailed molecular representation, the polymers can be described using physics-inspired descriptors such as molecular weight, radius of gyration, etc (Figure 3). The pool can be expanded by using feature-engineering to construct new candidate descriptors through, e.g., arithmetic combinations of the current descriptors in the pool. Both supervised and unsupervised methods have been developed to reduce this pool. Supervised learning methods sift through the candidate pool and select only those descriptors that most significantly correlate with the property of interest. Several such methods have proven effective for predicting material properties, including embedded feature selection, sure independence screening and sparsifying operator (SISSO), least absolute shrinkage and selection operator (LASSO), and genetic algorithms for feature selection. Unsupervised learning methods identify correlations within the descriptor pool and generate a new, smaller set of nonredundant features that are combinations of the original candidate descriptors. Specific unsupervised feature reduction techniques that have been effective for property prediction include principal component analysis (PCA), uniform manifold approximation and projection (UMAP), t-distributed stochastic neighbor embedding (t-SNE), and multidimensional scaling. It is usually not obvious which of these ML techniques is best for a specific problem, so it can be advantageous to implement several different ML methods and choose the one with the best prediction accuracy. Finally, the trained ML model that links the low-dimensional descriptors to the property of interest can be integrated into an iterative scheme to design materials with optimal properties. As illustrated in Figure 3A, this strategy has been employed to design random
copolymers with targeted values of $T_g$.

The second strategy incorporates discovery of a low-dimensional set of descriptors directly into the training process without requiring an initial pool of hand-crafted features. As illustrated in Figure 3b, the ML model takes the fully-detailed high-dimensional representation as input, and during training, finds the low-dimensional descriptors that best predict the desired material property. This approach requires a supervised learning approach, and the particular set of low-dimensional descriptors that is discovered varies as the property of interest changes. Although these low-dimensional features are abstract and cannot be readily interpreted from a physical standpoint, this strategy is advantageous because the ML model is not constrained to a pool of hand-crafted descriptors which may not capture the information necessary to predict the desired property. Without this limitation, this approach (Figure 3b) can outperform those requiring hand-crafted descriptor pools (Figure 3a) for more accurate property predictions. Convolutional neural networks (CNNs) and graph convolutional networks (GCNs) are two common architectures for discovering low-dimensional descriptors. CNNs employ convolutional layers to extract a low-dimensional set of spatial features present in a structured data set, like a pixel- or voxel-based digitized image shown in Figure 3b. These features are then linked to the property of interest by means of a fully connected artificial neural network. CNNs have been implemented to accurately predict material properties from the spatial microstructure of nanocomposites, porous media, elastic composites, ceramics, and molecules. GCNs, on the other hand, have been used to successfully extract features from the machine-readable molecular graphs representing the arrangement of atoms and bonds in a molecule. They have been employed to predict properties of atomic crystals, large organic molecules, and small molecules. As illustrated in
Figure 3b, these reduction strategies can be integrated into an iterative design scheme in the same manner as the approaches of Figure 3a. Such an approach was recently introduced to find microstructures for a nanoparticle-based electrolyte that maximize or minimize ionic conductivity.

3 Property design using original representations

In this section, we highlight ML-assisted design strategies that do not require a compressed, low-dimensional representation of the design space. As a result of the exclusive linkage between the original design parameters and the property of interest, the design of materials using these approaches involves smart navigation of the inherent design space. Although a fixed property-predictive ML model can be trained for accelerated screening even in the absence of a low-dimensional representation, most studies have employed ML to screen candidates and generate designs that achieve desired properties. Here we discuss the three strategies shown in Figure 4: (a) active learning, (b) inverse neural networks and (c) conditional generative adversarial networks.

3.1 Active learning

Active learning strategies (Figure 4a) are efficient black-box optimization techniques suited for expensive objective functions because they avoid probing uninformative and suboptimal points in the design space. Such strategies are particularly attractive for materials design because they can reduce the total number of times a material property must be evaluated, which is often time-consuming, compared to traditional one-factor-at-a-time approaches. Starting with a small labeled data set, the ML model fits a function to estimate what is known as the “property landscape” (i.e., the relationship
between the material property of interest and the parameters of the design space). At every iteration, the optimization routine uses this function to suggest a new set of
parameters at which to measure the material property, keeping in mind that there is an exploration-exploitation trade-off that must be balanced to avoid restrictively local searches while ensuring efficient optimization. Once the additional property information from the newly selected parameters is known, the ML-estimated property landscape can be refined and the process repeated until convergence of the property values evaluated for the newly sampled design points is achieved. Active learning strategies are typically found to be efficient for exploring low-dimensional parameter spaces (e.g., those less than 20 dimensions)\textsuperscript{17,20} As a result, the particular active learning strategies employed for material design are typically used to determine the optimal experimental conditions for materials synthesis and processing\textsuperscript{100,102,105,107,109} and to identify the ideal combination of physical parameters to be provided as input in simulations\textsuperscript{106,108} contrary to optimizing high-dimensional design spaces (e.g. structural topologies) to achieve target properties. Feature importance analysis can be performed intermittently to eliminate design parameters that only marginally influence the property, thus reducing the number of dimensions to explore in the subsequent iterations\textsuperscript{100,101}

Different techniques for active learning can be categorized based on the choice of ML model used to predict the property landscape and the iterative algorithm employed for determining the next design points to probe. Bayesian optimization is an active learning algorithm widely discussed in previous review articles\textsuperscript{30–32,110,111} which fits a Gaussian process regression model to the labeled data points at every iteration. In addition to predicting the property landscape, the Gaussian process model also builds an acquisition function based on the predicted mean and variance to guide the location of the next query point. Other approaches\textsuperscript{100,104,105,107} have used elastic net regression\textsuperscript{112} or support vector regression with a radial basis function kernel\textsuperscript{113,116} for predicting the property landscape.
For studies employing the “Design of Experiments” approach, the ML-estimated property landscape can be analyzed by the experiment designer to manually decide the next set of experiments. Similarly, evolutionary algorithms such as the differential evolution algorithm and the metaheuristic cuckoo search algorithm have been employed to efficiently explore the property landscape predicted by the ML models.

3.2 Inverse networks

All of the strategies discussed above navigate through a design space to search for parameters where a material’s properties are optimized or closely match those of a target. Inverse networks (Figure 4b) take a different approach and attempt to learn the property-to-design mapping. Where successful, this strategy greatly simplifies materials design since the inverse network can take the target properties as input and immediately output the corresponding design parameters. Inverse networks have been commonly employed for optimizing nanophotonic devices where the physical geometric parameters describing the nanostructure (such as height, length, thickness, etc.) constitute the design parameters, and the resulting optical spectral response of the device is the material property. Although the training of an inverse network requires generation of a large data set, it is a one-time cost, and the same network can be repeatedly employed to design materials with different target properties.

The typical architecture for inverse networks is an artificial neural network model trained on material properties as inputs and design parameters as outputs. However, it can be difficult to converge the weights of a stand-alone inverse network during training because the function is multivalued, and many different design points can encode materials with similar properties. To address this issue, a tandem architecture, where a conventional
forward-modeling neural network is appended to the inverse network, was introduced (Figure 4b). This tandem neural network architecture could be successfully trained and offered excellent prediction accuracy when designing photonic structures for a target electromagnetic response. The forward network was first trained independently and remained frozen during the training of the tandem architecture. The weights in the inverse network were trained by minimizing the error between the real property input to the tandem network and the output property predicted by the tandem network. Other studies\textsuperscript{120-123} have similarly reported that this strategy helps training convergence, despite the inverse network itself being multivalued, because the training losses are defined only by the property loss and not on the error between the predicted and actual design parameters. The tandem inverse architecture was also found to be effective for simultaneously predicting a combination of discrete design parameters (materials indexed by numbering) and continuous structural parameters (thicknesses) displaying a targeted optical spectrum\textsuperscript{124, 125}

The limitations of the tandem architecture to handle the non-unique response-to-design mapping for systems with low-dimensional design parameters were also discussed in a recent study.\textsuperscript{126} Another strategy to resolve this nonuniqueness involves a stand-alone inverse network with design parameters modeled as multimodal distributions rather than discrete values.\textsuperscript{127} The output from the inverse network now represents weighted multiple design solutions for the input material property; however, the approximate number of degenerate solutions needs to be known in advance. To date, inverse networks have been primarily applied to design nanostructured photonic systems. Their applicability for designing other classes of materials, though promising, remains largely unexplored.
3.3 Conditional Generative Adversarial Networks

For material systems with high-dimensional parameter spaces (e.g., nanostructured topology design or molecular design), identifying a low-dimensional latent space is a potential avenue to speed up the optimization. However, the search through either the latent space (for generative models as shown in Figure 2a) or the original design space (for property-predictive models as shown in Figure 2b) will be driven by a separate optimization algorithm. In such cases, the sheer number of degrees of freedom can hinder the discovery of optimal designs. This challenge can be mitigated by utilizing conditional generative adversarial networks (CGANs) as the generative model, trained to bias the generation towards optimal structures with desirable properties. Conditional generative adversarial networks (CGANs) have been used mainly for inverse problems in the design of molecular species and structural topologies. The distinguishing feature of CGANs (Figure 4c) is that they combine training of the networks and optimization of design parameters in a single step. Compare this to other ML-based generative models, such as VAEs shown in Figure 2, which separately train ML models and then use them in an iterative optimization scheme. In a CGAN, the weights in the network are updated at each iteration to both improve generative capabilities as well as progressively shift the generated structures toward those exhibiting target properties. In this way, the CGAN avoids generating stochastic structures with suboptimal properties, focusing only on reliably generating structures with properties similar to the user-defined target. This combined process of updating network weights to generate structures and then computing the material property is performed repeatedly until convergence to a specified tolerance. CGANs do not require a large labeled data set beforehand, but material properties at each iteration have to be evaluated from their original representation, either explicitly using
experiments or simulations or leveraging a separate ML model for property prediction.

A typical CGAN architecture (Figure 4c) consists of two components: a generator network which creates structures distributed over the design space and a discriminator network which distinguishes the generated designs from the user-defined (real) designs. During each training step, the weights in the generator are updated based on two different losses. First, losses based on the distance between the evaluated properties of the generated designs and the target properties ensures biasing of the generator toward designs with desired properties over several iterations. Second, losses quantified by the discriminator based on the difference between the distributions of the generated designs and the fixed distribution of designs in the user-defined data aim to train the generator to produce a wide distribution of realistic designs, avoiding local optima in the design space. Reinforcement learning, in conjunction with the discriminator, can be used as an alternative strategy to the CGAN architecture in Figure 4c to bias generated structures to those with desired properties.

In many cases, we would like to maximize (or minimize) a material property, which makes it difficult to evaluate property losses using standard loss functions that compare two properties. One solution is to, at each iteration, define the target property for the loss as the highest (or lowest) value among all of the previously sampled designs. The targeted design of certain material properties using CGANs also requires as input to the generator the conditional vector, which comprises key operating parameters. For example, studies focused on design of high-efficiency optical nanostructures at different wavelengths and deflection angles have reported CGANs with the corresponding wavelength and angle pair as inputs to the generator.
4 Future Directions

The strategies discussed in this review highlight how ML offers some efficient solutions for addressing key challenges in inverse approaches to materials design. Some of the methods have been developed only very recently and have great potential for future use in different stages of the design workflow. We specifically discuss opportunities to combine two or more ML strategies in a single inverse workflow (Section 4.1) as well as strategies that interpret “black-box” predictions of ML models to provide fundamental insight on the relation between design parameters and material properties (Section 4.2).

4.1 Combining strategies

Most of the inverse strategies discussed here employ ML methods to assist with a single phase of the design scheme. However, it may be advantageous to combine multiple ML techniques, each enhancing a different part of the design process. This could significantly accelerate design of materials, but many details, such as which ML strategies are compatible with one another as well as the application-specific training requirements are not presently known.

One strategy is to train a ML model to predict the properties corresponding to materials encoded in the latent space, e.g., discovered from unsupervised learning of generative models such as VAEs or GANs. Optimization can proceed quickly in the low-dimensional latent space, and each iteration is fast using the ML model to evaluate properties, an improvement over the scheme in Figure 2a, which requires explicit simulations or experiments. Pretrained convolutional neural networks\textsuperscript{50,51} and Gaussian process regression models\textsuperscript{45} quantitatively linking the designs in the original representations to the property of interest can accelerate property evaluation during the iterative search
of the unsupervised latent space. However, in some cases it might not be practical to generate the required training sets. In these cases, active learning strategies, suitable for low-dimensional design spaces, can be used to query the compressed latent variables for identifying designs that score highly based on the desired material properties. Such a strategy circumvents generating large labeled training data beforehand.

A particularly interesting approach involves combining autoencoder networks with either a feedforward, property-predictive model (for the forward problem) or an inverse network (for the inverse problem) to reduce the computational expense associated with the design. In this strategy, recently applied to designing optical metasurfaces, the strong correlations present within the structural features as well as the optical response features are exploited to reduce the dimensionality of both the design and property space using autoencoder networks. This one-to-one mapping between the design parameters and property in their reduced spaces is beneficial for design of materials as it allows for employing inverse networks without nonuniqueness, and further alleviates the network-size issues for both the forward and inverse networks.

4.2 Interpretability of models

Although ML-based models facilitate discovery of materials with desired properties, the learned structure-property relations are often difficult to interpret. However, it is possible to develop methods that examine trained ML models to elucidate new correlations between the design parameters and the properties of interest. This could provide valuable physical insights that facilitate the experimental realization of material designs within realistic constraints.

Several techniques have been established to interpret the ML models used for
material design, especially for ML models used for forward property prediction. For example, various feature importance scores that quantify the significance of individual descriptors on a material property can be computed using Shapley additive explanations (SHAP), Gini important analysis, and mean decrease accuracy (MDA). For deep artificial neural networks trained to predict material properties, the design parameters that strongly correlate with the material property can be identified by analyzing the weights of the trained networks. Unsupervised data-driven approaches such as principal component analysis can also quantify correlations between hand-crafted features and material properties.

Although the predictive performance of end-to-end forward predictive models (Figure 3b) with no hand-crafted features exceeds those using hand-crafted features, interpreting such ML techniques is more difficult. To that end, saliency mapping is a visualization technique that can be leveraged for interpretation of trained CNN models. Saliency maps highlight the regions in the digitized image of a structure correlating with the corresponding structure-dependent property based on the learning of the trained CNN model. These techniques pertaining to interpretation of trained CNN models have been applied to identify the underlying microstructural features influencing the corresponding macroscopic properties of materials such as ionic conductivity in ceramics and photovoltaic performance in thin-film organic semiconductors. Similarly, the integrated gradients method can interpret trained graph neural networks (e.g. for molecular design) by quantifying the strength of the contributions of the atom and atom-pair features towards the material property.

Besides interpreting trained ML models to discover underlying physical laws governing a material property, machine learning techniques can also be used to train accurate yet
simple predictive models that are easy to interpret. For example, a recent study[147] reported training neural networks with adjustable parameters quantifying the complexity of the learned functions to find accurate and physically interpretable expressions for predicting a material property of interest. Similarly, a highly interpretable linear ML model for predicting material properties called factorized asymptotic Bayesian inference hierarchical mixture of experts was also reported.[148] The prediction accuracies of this model were comparable to difficult-to-interpret nonlinear models, such as neural networks or support vector machines.

5 Conclusions

Machine learning has recently emerged as an effective tool for making materials design problems tractable from a time and resource standpoint. In this review, we have discussed different ML-assisted strategies implemented for inverse design of material properties. Broadly, these strategies employ ML models to either directly or indirectly assist with the accelerated identification of optimal design points potentially yielding the target properties.

For certain design problems, the main information in the original high-dimensional design spaces can be effectively captured with a compressed, low-dimensional representation. In this regard, ML-inspired generative models serve as a means to generate new molecular to topological designs from the compressed latent vectors to identify materials with desired properties. Also, simplified training of the property-predictive ML models with the low-dimensional data allows for accelerated screening of the design space. For systems without the existence of a low-dimensional representation, in addition to the property-predictive modeling, the ML-guided design strategies focus on employing ML methods explicitly to search the design space efficiently. These methods include active learning strategies to
sequentially explore new design points based on a surrogate property landscape continually updated as the additional information flows in, backward mapping from target property to design parameters using inverse networks, and generative models trained to bias the generation of designs towards those exhibiting desired properties.

The progress reviewed here highlights the applicability of ML techniques for designing materials with tailored properties. Promising future directions, including combining ML strategies for new integrated design approaches and developing improved methods for interpreting trained ML models, further underline the role that ML will continue to play in addressing challenges posed by this rich and important class of inverse problems.

**DISCLOSURE STATEMENT**

The authors are not aware of any affiliations, memberships, funding, or financial holdings that might be perceived as affecting the objectivity of this review.

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**References**

1. Gurkan B, Goodrich BF, Mindrup EM, Ficke LE, Massel M, et al. 2010. Molecular Design of High Capacity, Low Viscosity, Chemically Tunable Ionic Liquids for CO2
2. Zhu Y, Peng L, Fang Z, Yan C, Zhang X, Yu G. 2018. Structural Engineering of 2D Nanomaterials for Energy Storage and Catalysis. *Adv. Mater.* 30(15):1706347

3. Shi X, Zheng S, Wu Z-S, Bao X. 2018. Recent advances of graphene-based materials for high-performance and new-concept supercapacitors. *J. Energy Chem.* 27(1):25–42

4. Yan Y, Zhao YS. 2014. Organic nanophotonics: from controllable assembly of functional molecules to low-dimensional materials with desired photonic properties. *Chem. Soc. Rev.* 43(13):4325–40

5. Chen Z, Li P, Anderson R, Wang X, Zhang X, et al. 2020. Balancing volumetric and gravimetric uptake in highly porous materials for clean energy. *Science* 368(6488):297–303

6. Burschka J, Kessler F, Nazeeruddin MK, Grätzel M. 2013. Co(III) Complexes as p-Dopants in Solid-State Dye-Sensitized Solar Cells. *Chem. Mater.* 25(15):2986–90

7. Wang S, Sina M, Parikh P, Uekert T, Shahbazian B, et al. 2016. Role of 4-tert-Butylpyridine as a Hole Transport Layer Morphological Controller in Perovskite Solar Cells. *Nano Lett.* 16(9):5594–600

8. Lu L, Zheng T, Wu Q, Schneider AM, Zhao D, Yu L. 2015. Recent Advances in Bulk Heterojunction Polymer Solar Cells. *Chem. Rev.* 115(23):12666–731

9. Song J, Zhang M, Yuan M, Qian Y, Sun Y, Liu F. 2018. Morphology Characterization of Bulk Heterojunction Solar Cells. *Small Methods* 2(3):1700229

10. Fang Y, Wang X, Wang Q, Huang J, Wu T. 2014. Impact of annealing on spiro-OMeTAD and corresponding solid-state dye sensitized solar cells. *Phys. Status Solidi A* 211(12):2809–16
11 Bu T, Wu L, Liu X, Yang X, Zhou P, et al. 2017. Synergic Interface Optimization with Green Solvent Engineering in Mixed Perovskite Solar Cells. *Adv. Energy Mater.* 7(20):1700576

12 Pham T-L, Nguyen N-D, Nguyen V-D, Kino H, Miyake T, Dam H-C. 2018. Learning structure-property relationship in crystalline materials: A study of lanthanide–transition metal alloys. *J. Chem. Phys.* 148(20):204106

13 Phillip WA, Dorin RM, Werner J, Hoek EMV, Wiesner U, Elimelech M. 2011. Tuning Structure and Properties of Graded Triblock Terpolymer-Based Mesoporous and Hybrid Films. *Nano Lett.* 11(7):2892–900

14 Pendergast MM, Mika Dorin R, Phillip WA, Wiesner U, Hoek EMV. 2013. Understanding the structure and performance of self-assembled triblock terpolymer membranes. *J. Membr. Sci.* 444:461–68

15 Shen K-H, Brown JR, Hall LM. 2018. Diffusion in Lamellae, Cylinders, and Double Gyroid Block Copolymer Nanostructures. *ACS Macro Lett.* 7(9):1092–98

16 Alshammasi MS, Escobedo FA. 2018. Correlation between Ionic Mobility and Microstructure in Block Copolymers. A Coarse-Grained Modeling Study. *Macromolecules* 51(22):9213–21

17 Schneider LY, Müller M. 2019. Engineering Scale Simulation of Nonequilibrium Network Phases for Battery Electrolytes. *Macromolecules* 52(5):2050–62

18 Aryal D, Howard MP, Samanta R, Antoine S, Segalman R, et al. 2020. Influence of pore morphology on the diffusion of water in triblock copolymer membranes. *J. Chem. Phys.* 152(1):014904
19 Torquato S. 2009. Inverse Optimization Techniques for Targeted Self-Assembly. *Soft Matter* 5:1157–73

20 Jain A, Bollinger JA, Truskett TM. 2014. Inverse Methods for Material Design. *AIChE J.* 60:2732–40

21 Jaeger HM. 2015. Celebrating *Soft Matter*’s 10th Anniversary: Toward Jamming by Design. *Soft Matter* 11:12–27

22 Ferguson AL. 2017. Machine learning and data science in soft materials engineering. *J. Phys. Condens. Matter* 30(4):043002

23 Murugan A, Jaeger HM. 2019. Bioinspired Nonequilibrium Search for Novel Materials. *MRS Bull.* 44:96–105

24 Jackson NE, Webb MA, de Pablo JJ. 2019. Recent advances in machine learning towards multiscale soft materials design. *Curr. Opin. Chem. Eng.* 23:106–14

25 Sherman ZM, Howard MP, Lindquist BA, Jadrich RB, Truskett TM. 2020. Inverse methods for design of soft materials. *J. Chem. Phys.* 152(14):140902

26 Liu Y, Zhao T, Ju W, Shi S. 2017. Materials discovery and design using machine learning. *J. Materiomics* 3(3):159–77

27 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(1):54

28 Guo K, Yang Z, Yu C-H, Buehler MJ. 2021. Artificial intelligence and machine learning in design of mechanical materials. *Mater. Horiz.* 8(4):1153–72
29 Jain A, Hautier G, Ong SP, Persson K. 2016. New opportunities for materials informatics: Resources and data mining techniques for uncovering hidden relationships. J. Mater. Res. 31(8):977–94

30 Suh C, Fare C, Warren JA, Pyzer-Knapp EO. 2020. Evolving the Materials Genome: How Machine Learning Is Fueling the Next Generation of Materials Discovery. Annu. Rev. Mater. Res. 50(1):1–25

31 Chen G, Shen Z, Iyer A, Ghumman UF, Tang S, et al. 2020. Machine-Learning-Assisted De Novo Design of Organic Molecules and Polymers: Opportunities and Challenges. Polymers 12(1):163

32 Moosavi SM, Jablonka KM, Smit B. 2020. The Role of Machine Learning in the Understanding and Design of Materials. J. Am. Chem. Soc. 142(48):20273–87

33 Hegde RS. 2020. Deep learning: a new tool for photonic nanostructure design. Nanoscale Adv. 2(3):1007–23

34 Liu Z, Zhu D, Raju L, Cai W. 2021. Tackling Photonic Inverse Design with Machine Learning. Adv. Sci. 8(5):2002923

35 So S, Badloe T, Noh J, Bravo-Abad J, Rho J. 2020. Deep learning enabled inverse design in nanophotonics. Nanophotonics 5(9):1041–57

36 Schwalbe-Koda D, Gómez-Bombarelli R. 2020. Generative Models for Automatic Chemical Design. arXiv:1907.01632 [cs.LG]

37 Sanchez-Lengeling B, Aspuru-Guzik A. 2018. Inverse molecular design using machine learning: Generative models for matter engineering. Science 361(6400):360–65
Elton DC, Boukouvalas Z, Fuge MD, Chung PW. 2019. Deep learning for molecular design—a review of the state of the art. *Mol. Syst. Des. Eng.* 4(4):828–49

Eyke NS, Koscher BA, Jensen KF. 2021. Toward Machine Learning-Enhanced High-Throughput Experimentation. *Trends Chem.* 3(2):120–32

Xu Y, Lin K, Wang S, Wang L, Cai C, et al. 2019. Deep learning for molecular generation. *Future Med. Chem.* 11(6):567–97

Jørgensen PB, and Schmidt MN, Winther O. 2018. Deep Generative Models for Molecular Science. *Mol. Inform.* 37(1–2):1700133

Kullback S, Leibler RA. 1951. On Information and Sufficiency. *Ann. Math. Stat.* 22(1):79–86

Kingma DP, Welling M. 2014. Auto-Encoding Variational Bayes. arXiv:1312.6114 [stat.ML]

Xue T, Wallin TJ, Menguc Y, Adriaenssens S, Chiaramonte M. 2020. Machine learning generative models for automatic design of multi-material 3D printed composite solids. *Extreme Mech. Lett.* 41:100992

Kim Y, Park HK, Jung J, Asghari-Rad P, Lee S, et al. 2021. Exploration of optimal microstructure and mechanical properties in continuous microstructure space using a variational autoencoder. *Mater. Des.* 202:109544

Griffiths R-R, Hernández-Lobato JM. 2020. Constrained Bayesian optimization for automatic chemical design using variational autoencoders. *Chem. Sci.* 11(2):577–86

Lim J, Hwang S-Y, Moon S, Kim S, Kim WY. 2020. Scaffold-based molecular design with a graph generative model. *Chem. Sci.* 11(4):1153–64
Ren Z, Noh J, Tian S, Oviedo F, Xing G, et al. 2020. Inverse design of crystals using generalized invertible crystallographic representation. arXiv:2005.07609 [physics.comp-ph]

Goodfellow I, Pouget-Abadie J, Mirza M, Xu B, Warde-Farley D, et al. 2014. Generative Adversarial Networks. arXiv:1406.2661 [stat.ML]

Kudyshev ZA, Kildishev AV, Shalaev VM, Boltasseva A. 2021. Machine learning-assisted global optimization of photonic devices. Nanophotonics 10(1):371–83

Kudyshev ZA, Kildishev AV, Shalaev VM, Boltasseva A. 2020. Machine-learning-assisted metasurface design for high-efficiency thermal emitter optimization. Appl. Phys. Rev. 7(2):021407

Wang L, Chan Y-C, Ahmed F, Liu Z, Zhu P, Chen W. 2020. Deep generative modeling for mechanistic-based learning and design of metamaterial systems. Comput. Methods Appl. Mech. Eng. 372:113377

Gómez-Bombarelli R, Wei JN, Duvenaud D, Hernández-Lobato JM, Sánchez-Lengeling B, et al. 2018. Automatic Chemical Design Using a Data-Driven Continuous Representation of Molecules. ACS Cent. Sci. 4(2):268–76

Yao Z, Sánchez-Lengeling B, Bobbitt NS, Bucior BJ, Kumar SGH, et al. 2021. Inverse design of nanoporous crystalline reticular materials with deep generative models. Nat. Mach. Intell. 3(1):76–86

Visaria D, Jain A. 2020. Machine-learning-assisted space-transformation accelerates discovery of high thermal conductivity alloys. Appl. Phys. Lett. 117(20):202107
Stoll A, Benner P. 2021. Machine learning for material characterization with an application for predicting mechanical properties. *GAMM-Mitt.* 44(1):e202100003

Oo MCM, Thein T. 2019. An efficient predictive analytics system for high dimensional big data. *J. King Saud Univ. - Comput. Inf. Sci.*

Wan Z, Xu Y, Šavija B. 2021. On the Use of Machine Learning Models for Prediction of Compressive Strength of Concrete: Influence of Dimensionality Reduction on the Model Performance. *Materials* 14(4):713

Zhang X, Cui J, Zhang K, Wu J, Lee Y. 2019. Machine Learning Prediction on Properties of Nanoporous Materials Utilizing Pore Geometry Barcodes. *J. Chem. Inf. Model.* 59(11):4636–44

Wu K, Sukumar N, Lanzillo NA, Wang C, Ramprasad R, et al. 2016. Prediction of polymer properties using infinite chain descriptors (ICD) and machine learning: Toward optimized dielectric polymeric materials. *J. Polym. Sci. B Polym. Phys.* 54(20):2082–91

Bucior BJ, Bobbitt NS, Islamoglu T, Goswami S, Gopalan A, et al. 2019. Energy-based descriptors to rapidly predict hydrogen storage in metal-organic frameworks. *Mol. Syst. Des. Eng.* 4(1):162–74

Chi W, Chen J, Liu W, Wang C, Qi Q, et al. 2020. A General Descriptor ∆E Enables the Quantitative Development of Luminescent Materials Based on Photoinduced Electron Transfer. *J. Am. Chem. Soc.* 142(14):6777–85

Anatole von Lilienfeld O, Ramakrishnan R, Rupp M, Knoll A. 2015. Fourier series of atomic radial distribution functions: A molecular fingerprint for machine learning models of quantum chemical properties. *Int. J. Quantum Chem.* 115(16):1084–93
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(46):5746–52

Dai D, Liu Q, Hu R, Wei X, Ding G, et al. 2020. Method construction of structure-property relationships from data by machine learning assisted mining for materials design applications. *Mater. Des.* 196:109194

Ouyang R, Curtarolo S, Ahmetcik E, Scheffler M, Ghiringhelli LM. 2018. SISSO: A compressed-sensing method for identifying the best low-dimensional descriptor in an immensity of offered candidates. *Phys. Rev. Mater.* 2(8):083802

Weng B, Song Z, Zhu R, Yan Q, Sun Q, et al. 2020. Simple descriptor derived from symbolic regression accelerating the discovery of new perovskite catalysts. *Nat. Commun.* 11(1):3513

Pilania G, Iverson CN, Lookman T, Marrone BL. 2019. Machine-Learning-Based Predictive Modeling of Glass Transition Temperatures: A Case of Polyhydroxyalkanoate Homopolymers and Copolymers. *J. Chem. Inf. Model.* 59(12):5013–25

Lee EY, Wong GCL, Ferguson AL. 2018. Machine learning-enabled discovery and design of membrane-active peptides. *Bioorg. Med. Chem.* 26(10):2708–18

Tibshirani R. 2011. Regression shrinkage and selection via the lasso: a retrospective. *J. R. Stat. Soc. Series B Stat. Methodol.* 73(3):273–82

Zhang Y, Wen C, Wang C, Antonov S, Xue D, et al. 2020. Phase prediction in high entropy alloys with a rational selection of materials descriptors and machine learning models. *Acta Mater.* 185:528–39
Melati D, Grinberg Y, Kamandar Dezfooli M, Janz S, Cheben P, et al. 2019. Mapping the global design space of nanophotonic components using machine learning pattern recognition. Nat. Commun. 10(1):4775

Yucel B, Yucel S, Ray A, Duprez L, Kalidindi SR. 2020. Mining the Correlations Between Optical Micrographs and Mechanical Properties of Cold-Rolled HSLA Steels Using Machine Learning Approaches. Integr. Mater. Manuf. Innov. 9(3):240–56

Kadulkar S, Howard MP, Truskett TM, Ganesan V. 2021. Prediction and Optimization of Ion Transport Characteristics in Nanoparticle-Based Electrolytes Using Convolutional Neural Networks. J. Phys. Chem. B 125(18):4838–49

Fung V, Zhang J, Hu G, Ganesh P, Sumpter BG. 2021. Inverse Design of Two-Dimensional Material with Invertible Neural Networks. [arXiv:2106.03013] [cond-mat.mtrl-sci]

Yuan Q, Santana-Bonilla A, Zwijnenburg MA, Jelfs KE. 2020. Molecular Generation Targeting Desired Electronic Properties via Deep Generative Models. Nanoscale 12:6744–58

Jung J, Yoon JI, Park HK, Kim JY, Kim HS. 2019. An efficient machine learning approach to establish structure-property linkages. Comput. Mater. Sci. 156:17–25

Zhao Z-W, del Cueto M, Geng Y, Troisi A. 2020. Effect of Increasing the Descriptor Set on Machine Learning Prediction of Small Molecule-Based Organic Solar Cells. Chem. Mater. 32(18):7777–87

Zhu M-X, Yu Q-C, Song H-G, Chen T-X, Chen J-M. 2021. Rational Design of High-Energy-Density Polymer Composites by Machine Learning Approach. ACS Appl. Energy Mater. 4(2):1449–58
Kojima T, Washio T, Hara S, Koishi M. 2020. Synthesis of computer simulation and machine learning for achieving the best material properties of filled rubber. *Sci. Rep.* 10(1):18127

Goh GB, Siegel C, Vishnu A, Hodas NO, Baker N. 2017. Chemception: A Deep Neural Network with Minimal Chemistry Knowledge Matches the Performance of Expert-developed QSAR/QSPR Models. [arXiv:1706.06689](https://arxiv.org/abs/1706.06689) [stat.ML]

Wang Y, Zhang M, Lin A, Iyer A, Prasad AS, et al. 2020. Mining structure–property relationships in polymer nanocomposites using data driven finite element analysis and multi-task convolutional neural networks. *Mol. Syst. Des. Eng.* 5(5):962–75

Yang Z, Yabansu YC, Al-Bahrani R, Liao W-k, Choudhary AN, et al. 2018. Deep learning approaches for mining structure-property linkages in high contrast composites from simulation datasets. *Comput. Mater. Sci.* 151:278–87

Rosen AS, Iyer SM, Ray D, Yao Z, Aspuru-Guzik A, et al. 2021. Machine learning the quantum-chemical properties of metal–organic frameworks for accelerated materials discovery. *Matter* 4(5):1578–97

Coley CW, Barzilay R, Green WH, Jaakkola TS, Jensen KF. 2017. Convolutional Embedding of Attributed Molecular Graphs for Physical Property Prediction. *J. Chem. Inf. Model.* 57(8):1757–72

Wallach I, Dzamba M, Heifets A. 2015. AtomNet: A Deep Convolutional Neural Network for Bioactivity Prediction in Structure-based Drug Discovery. [arXiv:1510.02855](https://arxiv.org/abs/1510.02855) [cs.LG]

Peng Y, Zhang Z, Jiang Q, Guan J, Zhou S. 2019. TOP: Towards Better Toxicity Prediction by Deep Molecular Representation Learning. In *2019 IEEE International
Conference on Bioinformatics and Biomedicine (BIBM), pp. 318–25. Piscataway, NJ: IEEE

88 Karimpouli S, Tahmasebi P. 2019. Image-based velocity estimation of rock using Convolutional Neural Networks. *Neural Netw.* 111:89–97

89 Wu H, Fang W-Z, Kang Q, Tao W-Q, Qiao R. 2019. Predicting Effective Diffusivity of Porous Media from Images by Deep Learning. *Sci. Rep.* 9(1):20387

90 Wu Z, Yang T, Deng Z, Huang B, Liu H, et al. 2019. Predicting Automatic Crack Detection and Analysis for Biological Cellular Materials in X-Ray In Situ Tomography Measurements. *Integr. Mater. Manuf. Innov.* 8(4):559–69

91 Kondo R, Yamakawa S, Masuoka Y, Tajima S, Asahi R. 2017. Microstructure recognition using convolutional neural networks for prediction of ionic conductivity in ceramics. *Acta Mater.* 141:29–38

92 Xie T, Grossman JC. 2018. Crystal Graph Convolutional Neural Networks for an Accurate and Interpretable Prediction of Material Properties. *Phys. Rev. Lett.* 120(14):145301

93 Rahaman O, Gagliardi A. 2020. Deep Learning Total Energies and Orbital Energies of Large Organic Molecules Using Hybridization of Molecular Fingerprints. *J. Chem. Inf. Model.* 60(12):5971–83

94 Kearnes S, McCloskey K, Berndl M, Pande V, Riley P. 2016. Molecular graph convolutions: moving beyond fingerprints. *J. Comput. Aided Mol. Des.* 30(8):595–608

95 Xu Y, Pei J, Lai L. 2017. Deep Learning Based Regression and Multiclass Models for Acute Oral Toxicity Prediction with Automatic Chemical Feature Extraction. *J. Chem. Inf. Model.* 57(11):2672–85
96 Ryu S, Lim J, Hong SH, Kim WY. 2018. Deeply learning molecular structure-property relationships using attention- and gate-augmented graph convolutional network. arXiv:1805.10988 [cs.LG]

97 Frazier P. 2018. A Tutorial on Bayesian Optimization. arXiv:1807.02811 [stat.ML]

98 Moriconi R, Deisenroth MP, Kumar KSS. 2020. High-dimensional Bayesian optimization using low-dimensional feature spaces. Mach. Learn. 109(9):1925–43

99 Wang Z, Hutter F, Zoghi M, Matheson D, De Freitas N. 2016. Bayesian Optimization in a Billion Dimensions via Random Embeddings. J. Artif. Int. Res. 55(1):361—87

100 Cao B, Adutwum LA, Oliynyk AO, Luber EJ, Olsen BC, et al. 2018. How To Optimize Materials and Devices via Design of Experiments and Machine Learning: Demonstration Using Organic Photovoltaics. ACS Nano 12(8):7434–44

101 Khatamsaz D, Molkeri A, Couperthwaite R, James J, Arróyave R, et al. 2021. Adaptive active subspace-based efficient multifidelity materials design. Mater. Des. 209:110001

102 MacLeod BP, Parlane FGL, Morrissey TD, Häse, F, Roch LM, et al. 2020. Self-driving laboratory for accelerated discovery of thin-film materials. Sci. Adv. 6(20):eaaz8867

103 Dave A, Mitchell J, Kandasamy K, Wang H, Burke S, et al. 2020. Autonomous Discovery of Battery Electrolytes with Robotic Experimentation and Machine Learning. Cell Rep. Phys. Sci. 1(12):100264

104 Wang B, Cai J, Liu C, Yang J, Ding X. 2020. Harnessing a Novel Machine-Learning-Assisted Evolutionary Algorithm to Co-optimize Three Characteristics of an Electrospun Oil Sorbent. ACS Appl. Mater. Interfaces 12(38):42842–49
Wen C, Zhang Y, Wang C, Xue D, Bai Y, et al. 2019. Machine learning assisted design of high entropy alloys with desired property. *Acta Mater.* 170:109–17

Wang Y, Xie T, France-Lanord A, Berkley A, Johnson JA, et al. 2020. Toward Designing Highly Conductive Polymer Electrolytes by Machine Learning Assisted Coarse-Grained Molecular Dynamics. *Chem. Mater.* 32(10):4144–51

Munshi J, Chen W, Chien T, Balasubramanian G. 2021. Machine learned metaheuristic optimization of the bulk heterojunction morphology in P3HT:PCBM thin films. *Comput. Mater. Sci.* 187:110119

Wheatle BK, Fuentes EF, Lynd NA, Ganesan V. 2020. Design of Polymer Blend Electrolytes through a Machine Learning Approach. *Macromolecules* 53(21):9449–59

Zhang W, Wang B, Zhao C. 2021. Selective Thermophotovoltaic Emitter with Aperiodic Multilayer Structures Designed by Machine Learning. *ACS Appl. Energy Mater.* 4(2):2004–13

Batra R, Song L, Ramprasad R. 2021. Emerging materials intelligence ecosystems propelled by machine learning. *Nat. Rev. Mater.* 6(8):655–78

Lookman T, Balachandran PV, Xue D, Yuan R. 2019. Active learning in materials science with emphasis on adaptive sampling using uncertainties for targeted design. *NPJ Comput. Mater.* 5(1):21

Zou H, Hastie T. 2005. Regularization and variable selection via the elastic net. *J. R. Stat. Soc. Series B Stat. Methodol.* 67(2):301–20

Smola AJ, Schölkopf B. 2004. A tutorial on support vector regression. *Stat. Comput.* 14(3):199–222
Chang Y-W, Hsieh C-J, Chang K-W, Ringgaard M, Lin C-J. 2010. Training and Testing Low-degree Polynomial Data Mappings via Linear SVM. *J. Mach. Learn. Res.* 11(48):1471–90

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(1):11241

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

Storn R, Price K. 1997. Differential Evolution – A Simple and Efficient Heuristic for global Optimization over Continuous Spaces. *J. Glob. Optim.* 11(4):341–59

Yang X-S, Deb S. 2010. Cuckoo Search via Levy Flights. [arXiv:1003.1594] [math.OC]

Liu D, Tan Y, Khoram E, Yu Z. 2018. Training Deep Neural Networks for the Inverse Design of Nanophotonic Structures. *ACS Photonics* 5(4):1365–69

Malkiel I, Mrejen M, Nagler A, Arieli U, Wolf L, Suchowski H. 2018. Plasmonic nanostructure design and characterization via Deep Learning. *Light Sci. Appl.* 7(1):60

Harper ES, Coyle EJ, Vernon JP, Mills MS. 2020. Inverse design of broadband highly reflective metasurfaces using neural networks. *Phys. Rev. B* 101(19):195104

Harper ES, Weber MN, Mills MS. 2019. Machine Accelerated Nano-Targeted Inhomogeneous Structures. In *2019 IEEE Research and Applications of Photonics in Defense Conference (RAPID)*, pp. 1–5. Piscataway, NJ: IEEE
Roberts NB, Keshavarz Hedayati M. 2021. A deep learning approach to the forward prediction and inverse design of plasmonic metasurface structural color. *Appl. Phys. Lett.* 119(6):061101

So S, Mun J, Rho J. 2019. Simultaneous Inverse Design of Materials and Structures via Deep Learning: Demonstration of Dipole Resonance Engineering Using Core–Shell Nanoparticles. *ACS Appl. Mater. Interfaces* 11(27):24264–68

Qiu C, Wu X, Luo Z, Yang H, Wang G, et al. 2021. Simultaneous inverse design continuous and discrete parameters of nanophotonic structures via back-propagation inverse neural network. *Opt. Commun.* 483:126641

Phan AD, Nguyen CV, Linh PT, Huynh TV, Lam VD, et al. 2020. Deep Learning for the Inverse Design of Mid-Infrared Graphene Plasmons. *Crystals* 10(2):125

Unni R, Yao K, Zheng Y. 2020. Deep Convolutional Mixture Density Network for Inverse Design of Layered Photonic Structures. *ACS Photonics* 7(10):2703–12

Sanchez-Lengeling B, Outeiral C, Guimaraes G, Aspuru-Guzik A. 2017. Optimizing distributions over molecular space. An Objective-Reinforced Generative Adversarial Network for Inverse-design Chemistry (ORGANIC). ChemRxiv 5309668. [https://doi.org/10.26434/chemrxiv.5309668](https://doi.org/10.26434/chemrxiv.5309668)

Liu Z, Zhu D, Rodrigues SP, Lee K-T, Cai W. 2018. Generative Model for the Inverse Design of Metasurfaces. *Nano Lett.* 18(10):6570–76

So S, Rho J. 2019. Designing nanophotonic structures using conditional deep convolutional generative adversarial networks. *Nanophotonics* 8(7):1255–61
131 Jiang J, Sell D, Hoyer S, Hickey J, Yang J, Fan JA. 2019. Free-Form Diffractive Metagrating Design Based on Generative Adversarial Networks. *ACS Nano* 13(8):8872–78

132 Kim B, Lee S, Kim J. 2020. Inverse design of porous materials using artificial neural networks. *Sci. Adv.* 6(1):eaax9324

133 Jiang J, Fan JA. 2019. Global Optimization of Dielectric Metasurfaces Using a Physics-Driven Neural Network. *Nano Lett.* 19(8):5366–72

134 Kiarashinejad Y, Abdollahramezani S, Adibi A. 2020. Deep learning approach based on dimensionality reduction for designing electromagnetic nanostructures. *NPJ Comput. Mater.* 6(1):12

135 Ma J, Huang Y, Pu M, Xu D, Luo J, et al. 2020. Inverse design of broadband metasurface absorber based on convolutional autoencoder network and inverse design network. *J. Phys. D Appl. Phys.* 53(46):464002

136 Maulana Kusdhany MI, Lyth SM. 2021. New insights into hydrogen uptake on porous carbon materials via explainable machine learning. *Carbon* 179:190–201

137 Chen Y, Cheng A, Zhang C, Chen S, Ren Z. 2021. Rapid mechanical evaluation of the engine hood based on machine learning. *J. Braz. Soc. Mech. Sci. Eng.* 43(7):345

138 Howard MP, Lequieu J, Delaney KT, Ganesan V, Fredrickson GH, Truskett TM. 2020. Connecting Solute Diffusion to Morphology in Triblock Copolymer Membranes. *Macromolecules* 53(7):2336–43
139 Sabando MV, Ponzoni I, Soto AJ. 2019. Neural-based approaches to overcome feature selection and applicability domain in drug-related property prediction. *Appl. Soft Comput.* 85:105777

140 Lee SY, Byeon S, Kim HS, Jin H, Lee S. 2021. Deep learning-based phase prediction of high-entropy alloys: Optimization, generation, and explanation. *Mater. Des.* 197:109260

141 Jadrich RB, Lindquist BA, Truskett TM. 2018. Unsupervised machine learning for detection of phase transitions in off-lattice systems. I. Foundations. *J. Chem. Phys.* 149(19):194109

142 Jadrich RB, Lindquist BA, Piñeros WD, Banerjee D, Truskett TM. 2018. Unsupervised machine learning for detection of phase transitions in off-lattice systems. II. Applications. *J. Chem. Phys.* 149(19):194110

143 Simonyan K, Vedaldi A, Zisserman A. 2014. Deep Inside Convolutional Networks: Visualising Image Classification Models and Saliency Maps. arXiv:1312.6034 [cs.CV]

144 Pokuri BSS, Ghosal S, Kokate A, Sarkar S, Ganapathysubramanian B. 2019. Interpretable deep learning for guided microstructure-property explorations in photovoltaics. *NPJ Comput. Mater.* 5(1):95

145 Sundararajan M, Taly A, Yan Q. 2017. Axiomatic Attribution for Deep Networks. arXiv:1703.01365 [cs.LG]

146 Dai M, Demirel MF, Liang Y, Hu J-M. 2021. Graph neural networks for an accurate and interpretable prediction of the properties of polycrystalline materials. *NPJ Comput. Mater.* 7(1):103
Desai S, Strachan A. 2021. Parsimonious neural networks learn interpretable physical laws. *Sci. Rep.* 11(1):12761

Iwasaki Y, Sawada R, Stanev V, Ishida M, Kirihara A, et al. 2019. Materials development by interpretable machine learning. arXiv:1903.02175 [cond-mat.mtrl-sci]