Deep Learning Image-based Design of Graphene-reinforced Polyurethane Foams

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Abstract

Machine (Deep-) learning techniques have been rapidly progressing in the past decade and have become an important tool for discovery within the scientific community. The adoption of these techniques within the material science community has been critical in enabling high throughput research through rapid characterization and discovery of materials. Machine (Deep-) learning techniques are being used not only for characterizing materials but also for extrapolating structure-property relationships as well as for generating new structures from existing material structure data. More broadly, the use of these techniques has allowed the quantitative use of images as a powerful information source for both material analytics and discovery. In this report we discuss two case studies where we demonstrate the use of deep learning techniques for modeling and optimizing the design of graphene-reinforced polyurethane foams. Specifically we demonstrate both forward and inverse design techniques through the use of image regression and Generative Adversarial Networks applied to SEM images of graphene-reinforced polyurethane (PU) Foams. This may be the first such investigation into the microstructure of Foams.

1 Introduction

While AI has had major successes in fields such as computer vision, natural language processing (NLP) and autonomous driving to name a few, AI approaches are still in their infancy when applied to materials science (1). The application of Artificial intelligence (AI) to material science problems is an emerging area of active research as indicated in Figure(1). Primary research directions include the use of AI techniques for better understanding of material properties, for accelerating material discovery and for enabling efficient development of advanced materials. These are often collectively termed as the area of high throughput materials research, where, in contrast to the traditional trial-and-error approach to material discovery, AI enabled methods offer the promise of a much reduced turnaround time for material discovery and commercialization of novel materials. In a recent IBM report (2) it was projected that the time for new material discovery was anticipated to be reduced by 90%, from 10 years to 1 year. The efficiency gains in the IBM process begins with the use of NLP for the extraction of information/insights at an ingestion rate of 6000 papers/patents per hour. This has also been demonstrated by (6) where they show that NLP techniques can extract material synthesis insights from scientific literature and predict synthesis parameters and outcomes on new materials systems, vastly outperforming heuristic strategies. The need to incorporate AI in material research and development has become especially evident as material systems become exceedingly complex and the search spaces become extremely large. These systems are often too complex to develop accurate physical models which, even if developed, are computationally expensive and very sensitive to changes in system composition. Additionally, given the large parameter space defining these systems, the model identification burden remains high. Searching for and designing new micro-structures with desired properties is another challenge especially given the multi-criteria optimization objectives and high

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dimensional, and often non-convex, design optimization space. For such systems manual exploration, based on empirical methods, is simply beyond human bandwidth. AI methods provide powerful means for understanding and for predicting material behaviors, functions and engineering properties and for automating the material discovery process. Recently, a new paradigm (the 5^{th} paradigm) is projected to accelerate material science discoveries, in addition to AI, by utilizing advances in quantum computing, autonomous testing and hybrid-cloud platforms (2).

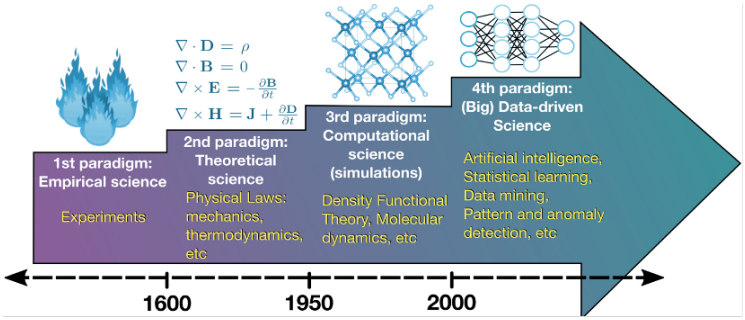

Figure 1: The fourth paradigm in material science will be driven in large by AI techniques (7).

There are multiple opportunities for using AI methods for progressing material science and material discovery. Fundamentally AI applications in material science can be classified into material characterization, selection and discovery techniques. Understanding material behavior and structure-property relationships are key to material discovery. AI models can be designed to predict material properties as a function of composition and process parameters. This is often referred to as the forward approach and is based on regression or classification techniques. Conversely, given material property and/or response the material composition may be predicted. This is the inverse approach and relies on the use of generative models. Some published examples of AI techniques applied to material science include AI-based screening and experimental validation of thermoplastic polymers (3), inorganic phosphor host materials for solid-state lighting (4), and high-entropy alloys for structural applications (5). AI methods require substantially more data than conventional methods, unfortunately, material scientists have traditionally worked with small data sets that are also noisy and sparse. Additionally, data is also often available in many forms including analog/digital sensor data, various image types and texts from logs. Some of these data concerns may be alleviated by modernizing material science labs with fast sensing and data acquisition systems (11), but given the large design space for material candidates, synthesis and processing parameters, and a wide range of properties. Additionally AI based techniques such as data augmentation, transfer learning (8) and generative models (9; 10) are also very effective. Together these approaches are necessary to alleviate the severe data bottleneck problem faced by material scientists seeking to apply AI techniques.

One approach to reducing the data dependency of AI models is to use materials domain knowledge to inform AI models such as by embedding physics within neural network models (i.e. physics informed neural networks - PINNs) (12). This guarantees that symmetries and conservation laws, present in the physical system being studied, are not violated in the models. In addition to reducing the data needs, physics aware AI models also improve computational efficiency and prediction accuracy. In most AI models, the way inputs are transformed into outputs are often opaque resulting in black box solutions (18; 19; 20). As such, to increase the transparency and robustness of AI models for scientific users, interpretability and explainability have become key criteria in model development. Some techniques are Grad-CAM (21) which produces coarse localization heatmaps highlighting the important regions in the image used for the predictions and an ablation study where, by removing some "feature" of the model or algorithm, one quantifies the effects on model performance.

In this study, we implemented an integrated approach that combines material science experimentation and artificial intelligence techniques. In particular we use deep learning (DL) methods of image regression and generative adversarial networks (GANs) to provide mechanistic and physical insights from multi-modal, in-situ characterization data and SEM microstructure images. These methods are applied for accelerating material discovery and optimizing physical properties in graphene-reinforced polyurethane foam composites.

2 Materials and Experimental Methods: Synthesis and Characterization

Polyurethane (PU) foams are a well-known class of polymers with a wide variety of automotive applications including seat cushions, headliners, engine covers etc. due to their excellent mechanical properties, lightweight nature, high levels of acoustic dampening, thermal stability and reduced cost to name a few (22). The customizable nature and versatility of PU foams such as in the choice of catalysts, fillers, isocyanate component, and nature of degree of cross-linking sites, polyol molecular weight, viscosity, etc. has led to the development of various polyurethane materials tailored to meet specific industry needs. Recent studies (23; 24; 25) have shown changes in foam microstructural

Table 1: Polyurethane foam materials and formulations summary.

	Component Type	**Weight Percentage**
Voranol 4701	Petroleum Polyol	62.2 %
Lumulse POE (26) GLYC	Cell Opener	0.6%
Tegostab B4690	Surfactant	0.3%
Diethanolamine (DEA)	Cross Linker	0.9%
Niax A300	Catalyst	0.4%
Niax A1	Catalyst	0.2%
Deionized Water	Blowing Agent	1.9%
Rubinate 7304	Diisocyanate	33.5%
Graphene	Additive	(0, 0.01, 0.025, 0.05)%

features such as cell size, density, morphology, etc. accompany enhancement in mechanical, electrical, and thermal properties. By synthesizing PU foams reinforced with varied amounts of graphene, we aim to develop here, an image-based AI solution for foam material design and property prediction from SEM images and physical property multi-modal data, as well as, generate realistic synthetic microstructure images based on user-input physical/processing parameters.

PU foams were synthesized by reacting polyols with isocyanate in a lab scale production. The polyol is first combined with a few additives including a cell opener, a surfactant, a crosslinker, two catalysts, and a blowing agent before reacting with a diisocyanate. The reader is referred to (22) for the detailed synthesis conditions, preparation, and mixing of the polyols and additives The PU foams were cut to comply with ASTM standard testing procedures to obtain physical and mechanical properties of each foam formulation.

Table 2: A summary of the tests and their standards.

Test	**Standard**
Compression Force Deflection	ASTM D3574 Test C
Compression Modulus	ASTM D3574 Test C
Tensile Stress at Maximum Load	ASTM D3574 Test E
Tensile Modulus	ASTM D3574 Test E
Tear Resistance	Ford Specification WSS-M15P20-B1, 3.3.5

Foam was analyzed based on mechanical testing. Density was measured, and tensile, tear, and compression properties were tested using an Instron 3366 machine. Six samples were selected and tested from each foam formulation. A summary of the tests and their standards are summarized in Table 3 [1]. Glass transition temperature and storage modulus were examined using DMA. Three foam samples were analyzed at 10 Hz and 1% strain using Rheometric Scientific Dynamic Mechanical Thermal Analyzer IV.

Deep learning methods for images frequently require datasets in the order of thousands of images. However, a careful selection of labelled images, data augmentation and pre-designed prototypical neural networks can reduce the amount of data needed. Here we mainly impalement geometric data augmentation techniques that include rotation, flips, etc. as the properties of the SEM images under study are invariant to these operations.

[1] Parenthesis indicates standard deviation.

Table 3: Polyurethane foam mechanical test results data.

Measured Property	Units	0%	0.01%	0.025%	0.05%
Mechanical					
Comp. Stress @ 25% strain	kPa	2.794 (0.163)	3.965 (0.632)	3.889 (0.225)	4.151 (0.569)
Comp. Stress @ 50% strain	kPa	4.102 (0.252)	6.520 (1.201)	7.182 (0.515)	7.109 (1.178)
Comp. Stress @ 65% strain	kPa	6.178 (0.407)	10.957 (2.885)	13.030 (1.832)	12.247 (2.894)
Comp. Modulus	MPa	0.0309 (0.0018)	0.0565 (0.0218)	0.0676 (0.0180)	0.0736 (0.0076)
Tensile Stress @Max Load	kPa	117.97 (6.400)	77.450 (6.402)	88.675 (15.863)	105.325 (38.298)
Tensile Modulus	MPa	0.197 (0.029)	0.190 (0.020)	0.200 (0.005)	0.227 (0.033)
Tear Resistance	N/mm	0.502 (0.045)	0.512 (0.040)	0.539 (0.026)	0.508 (0.070)
Thermal					
Tg	°C	-41.633 (1.293)	-40.805 (2.794)	-43.250 (1.964)	-43.480 (0.078)
Tg Max Point	°C	0.375 (0.023)	0.438 (0.022)	0.374 (0.031)	0.372 (0.028)
SM at RT	Pa	114,340 (1,2200)	98,600 (12,500)	123,440 (24,000)	144,923 (23,400)
SM at Tg	Pa	581,496 (85,900)	935,327 (129,000)	962,087(384,000)	1,545,173 (599,000)

3 Image Regression for Physical Properties

Material science image data processing and analysis often require prior subject matter expertise and are labor and time intensive. Recently, various techniques leveraging computer vision and machine learning techniques have been implemented as an efficient alternative for high throughput study of microstructural data such as in (26). Herein we implement a state-of-the-art AI based image regression technique to predict the amount of GNP additive (wt %) and selected mechanical properties of tensile stress and tensile modulus, based solely on given SEM images of GNP reinforced PU foams. The mechanical properties of PU foams can vary with changes in foam cell microstructure such as cell size, density, surface morphology and texture which in turn are correlate to their physical properties.

As such scanning electron microscope (SEM) images of the top surface of the above synthesized foams (c.f. Table 1) were obtained using JEOL JSM-6610 scanning electron microscope instrument at Ford Research and Advanced Engineering. The SEM images were taken under SE2 secondary electron detector at fixed beam energy and working distance. . The brightness and contrast levels were held constant across all images and samples; see also Table 3 . We collected 200 grayscale SEM images from four samples with varied amounts of additive (Control, 0.01%, 0.025%, 0.05%) as shown in Table 4. We consider an end-to-end deep learning (DL) image regression technique with raw

Table 4: SEM image dataset summary for GNP-reinforced PU foam.

Samples (GNP reinforced Foam)	SEM Image Dataset Size	Comments
Control (0.0%)	50	• high resolution fixed depth of focus, SED mode
0.01%	50	• high magnification (25x, approx. 10.3 pix/micron)
0.025%	50	• data augmented by rotation, flips etc.
0.05%	50	• physical property data measured for each sample

SEM image pixels as input and a supervised training procedure using the mechanical measurements as labels of the SEM images. The DL image regression technique comprises a ResNet34 network (27) pretrained on ImageNet (28) using a PyTorch implementation. The SEM images are normalized with respect to ImageNet stats and the number of outputs is set to three corresponding to additive %, tensile stress and tensile modulus. The loss function is chosen to be mean-squared error loss (MSE) and the model is trained for 100 iterations with resulting R-squared greater than 0.98 for each of the outputs.

The performance of the model is tested by predicting additive % and tensile properties on previously unseen examples (20% of the dataset) as shown in Fig. 3. Even though the model currently can be a black-box solution, it performs with high accuracy. The mechanism and explainability of the model shall be investigated further as in (15) to gain previously unknown insights or for other materials applications such as in multi-functional polymer composite materials design.

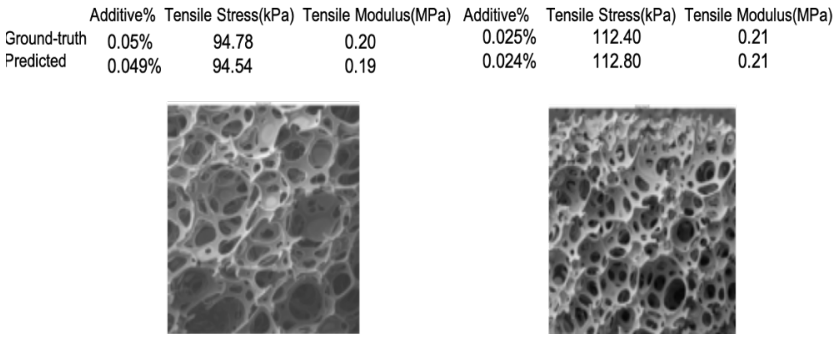

Figure 2: GNP-PU foam SEM image regression of additive percentage and mechanical properties.

4 Synthesizing Microstructures from Experimental Parameters: GANs for Inverse Materials Design

In the previous section, we implemented an image regression method where given an SEM image of a material sample, we are able to accurately predict material attributes such as the mechanical performance or graphene additive percent. In this section we implement a generative model that outputs a hypothetical SEM image corresponding to a given material attribute such as synthesis/processing condition or physical properties. This can significantly help domain scientists visualize and understand how changes in material attributes, synthesis/processing conditions impact the sample microstructure features which in turn determine its mechanical, thermal and electrical properties. This is all done without actually performing tests in the lab as well as to discover materials with optimal target properties through investigation of counterfactual relationships (15) (i.e. determining material features that control changes in the prediction).

Specifically, we use an image editing generative adversarial network, AttGAN (29), to synthesize realistic SEM images by varying a synthesis parameter, e.g. graphene additive weight percent. Different from other types of GANs which generate images starting from a noise vector, AttGAN is trained on input images along with the attributes encoding the desirable changes, thus reducing the heavy computational resources requirement during training as well as amount of image data. The reader is referred to the original work on AttGAN (29) for details on model architecture. As shown in Figure 3, the AttGAN contains three main components at training, i.e. the attribute classification constraint (correct attribute manipulation on the generated image), the reconstruction learning (preserving the attribute-excluding details) and the adversarial learning (for visually realistic generation). The overall objective function for the encoder and decoder (G_{enc}, G_{dec}) shown in the figure is formulated as below,

$$\min_{G_{enc},G_{dec}} \mathcal{L}_{enc,dec} = \lambda_1 \mathcal{L}_{rec} + \lambda_2 \mathcal{L}_{cls_g} + \mathcal{L}_{adv_g}, \tag{1}$$

while the objective function for the discriminator and the attribute classifier (D, C) is formulated as below,

$$\min_{D,C} \mathcal{L}_{dis,cls} = \lambda_3 \mathcal{L}_{cls_c} + \mathcal{L}_{adv_d}, \tag{2}$$

with λ_1, λ_2 and λ_3 are the hyperparameters for balancing the losses. We trained the AttGAN in the PyTorch implementation with the following model hyperparameters compared to the original implementation: learning rate of 0.002, batch size of 32, number of epochs = 6000; for the Generator: attribute loss weight = 10.0; reconstruction loss weight = 100.0; for the Discriminator: gradient penalty weight = 10.0; attribute loss weight = 1.0 and using an Adam optimizer. The results of the training are shown in 4 where a realistic visualization of the SEM image appearance of the material and subtle effect of the synthesis parameter change is shown given the previously unseen image (from the 20% of the dataset) for 0.01 % graphene additive reinforced PU foam. This can be further extended to demonstrate attribute/property-driven image generation process so as to design materials with target optimal properties such as high peak tensile modulus etc.

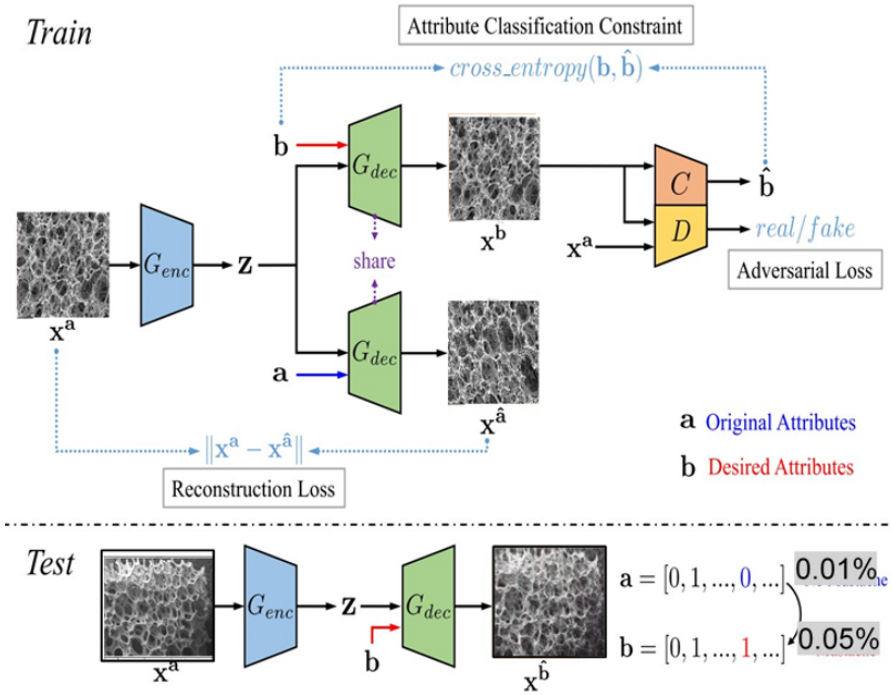

Figure 3: AttGAN model architecture for synthesizing microstructures with specified additive percentage attributes, adapted from (29).

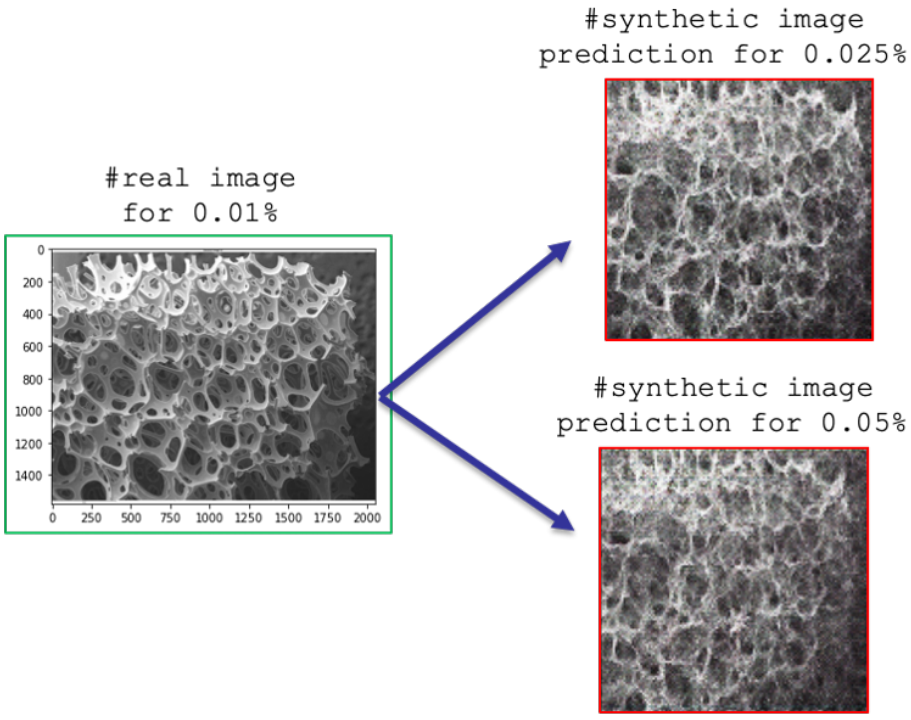

Figure 4: Synthetic image generation of SEM foam microstructure based on varied additive percentages; scale units in μm.

5 Conclusion

In conclusion, we demonstrated that state-of-the-art machine learning and deep learning techniques can be leveraged to model and optimize the design of graphene-reinforced polyurethane foams. A deep-learning image regression technique is used to predict mechanical properties of various graphene-reinforced polyurethane foams (PU) based solely on scanning electron microscopy (SEM) images with image regression $R^2 \sim 0.99$. An image editing generative adversarial network, AttGAN, in the PyTorch implementation was implemented to generate realistic synthetic microstructure images showing subtle effect of the varied synthesis parameter of graphene additive % from 0.01% to 0.025% and 0.05%. This allows domain scientists to visualize and understand how changes in material attributes, synthesis/ processing conditions impact the sample microstructure features which in turn determine its properties and thus can further be optimized. Further applications of AI coupled with multi-criteria multi-objective property optimization, optimal design of experiments and active learning as well as investigation of the explainability of the models built will be subjects of future research.

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