AI for Materials Science: Tuning Laser-Induced Graphene Production

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Abstract

AI has advanced the state of the art in many application domains, including ones not ordinarily associated with computer science. We present an application of automated parameter tuning to materials science, in particular, we use surrogate models for automated parameter tuning to optimize the fabrication of laser-induced graphene. This process allows to create microscopic conductive lines in thin layers of insulating material, enabling the development of next-generation nano-circuits. Optimizing the parameters that control the laser irradiation process is crucial to creating high-quality graphene that is suitable for this purpose. Through the application of state-of-the-art parameter tuning techniques, we are able to achieve improvements of up to a factor of two compared to existing approaches in the literature and to what human experts are able to achieve. Our results are reproducible across different experimental specimen and the deployed application can be used by domain scientists without a background in AI or machine learning.

1 Introduction

AI is increasingly pervasive in everyday life and has enabled advances that were unimaginable without it in many areas of science. One such application domain is materials science, where AI has enabled the discovery of new solar light absorbers [Bai *et al.*, 2018], new metallic glasses [Ren *et al.*, 2018], autonomous growth of carbon nanotubes [Nikolaev *et al.*, 2016], and the production of Bose-Einstein condensates [Wigley *et al.*, 2016], to name but a few examples. At the core of all of these successes lies the application of state-of-the-art AI techniques to processes that have traditionally been the domain of human experts, who base their work on theories that describe the underlying processes and were developed over decades. In many such cases, we find that AI can handle complex tasks better than the human experts given enough training examples, even though AI models do not usually have a semantic understanding of the fundamental laws.

In this paper, we present a new case where AI can help human experts to design materials – the transformation of graphene oxide into graphene through laser irradiation. This process creates electrically conductive areas (graphene) within an insulating surrounding environment (graphene oxide). The topical application of a laser allows precise control over where these conductive areas are created, enabling the creation of microscopic circuits, among other things. Graphene oxide can be deposited onto practically any surface in very thin layers, enabling for example flexible electronics and circuits on irregular surfaces.

Transforming graphene oxide into graphene through laser irradiation is not a straightforward process. The experimental parameters that control the irradiation process have to be set correctly to achieve the desired result. Such parameters include the power of the laser, the duration it is applied to a particular area, and the pressure in the reaction chamber. Incorrect settings may result in graphene oxide that is not sufficiently transformed into graphene and therefore not conductive, or even the destruction of the material.

Manual exploration of this parameter space is tedious and costly. Evaluating a particular parameter configuration requires time, the raw graphene oxide material, specialized equipment, and a skilled operator. There is little theory that could guide the exploration of the parameter space – while the underlying processes are understood in general, the high-level physical and chemical laws that govern the reduction of graphene oxide to graphene do not offer practical guidance for setting parameters.

Automated parameter tuning techniques are well-established in AI and have seen successful applications in many areas, for example combinatorial problem solving [Hurley *et al.*, 2014], continuous optimization [Xu *et al.*, 2011], and machine learning [Feurer *et al.*, 2015]. More specifically, modeling challenging parameter spaces with so-called surrogate models and using these models in iterative model-based approaches to optimize parameter configurations allows to explore large parameter spaces and efficiently while minimizing expensive evaluations of the underlying process.

We present the application of state-of-the-art AI and machine learning techniques to the production of laser-induced graphene, improving the properties of the material by up to a factor of two. This improvement translates directly into better advanced materials that will enable the next generation of advanced electronics and their cost-efficient production.

2 Background and Related Work

We first provide background on the AI techniques, in particular model-based optimization, and then present the materials science application domain.

2.1 Automated Parameter Tuning

In this paper, we consider self-adaptive learning systems that improve over time [Vilalta and Drissi, 2002]. In particular, we investigate the application of techniques for automated parameter tuning through model-based optimization. Automated parameter tuning, also called algorithm configuration, parameter control, or hyperparameter configuration, aims to find the best parameter configuration of an algorithm on a set of problem instances to solve [Hoos, 2011]. The output of an automated parameter tuning system is usually a configuration, i.e. a complete parameterization of the input algorithm that can be applied directly to solve new problem instances. Such a configuration may include the algorithm to run, chosen from a portfolio of available algorithms; i.e. a solution to the joint optimization problem that chooses the algorithm as well as its parameters [Kotthoff *et al.*, 2017].

Techniques for automated parameter tuning usually proceed in an iterative fashion – they predict the configuration to evaluate, and the result of this evaluation informs the predictions for the configuration to evaluate next. At the heart of these techniques are so-called surrogate models, which approximate and model the process whose parameters are to be tuned. This underlying process is expensive to evaluate, i.e. it is infeasible to exhaustively explore the parameter space and we are interested in keeping the number of evaluations as small as possible. The approximate surrogate model on the other hand is cheap to evaluate and allows for a targeted exploration of the parameter space, identifying promising configurations that available resources for evaluations of the underlying process should be directed towards.

This family of techniques is often referred to as model-based optimization (MBO), as the optimization process is based on the predictions of the surrogate models that serve as a replacement for the underlying process. Surrogate models are induced using machine learning, taking an increasing amount of ground-truth data into account between subsequent iterations. State-of-the-art MBO approaches often use Gaussian Processes or random forests to induce surrogate models, depending on the nature of the parameter space. The interested reader is referred to [Jones *et al.*, 1998] for an overview.

2.2 Laser-Induced Graphene

Graphene is a two-dimensional single layer of carbon atoms with extraordinary properties, such as strength higher than any other material, high conductivity, and near transparency. To produce graphene, natural sources of carbon, e.g. graphite, coal, and biochar, can be converted into graphene oxide inks that can be printed directly onto substrates as thin films. Irradiating this precursor material with a laser heats and anneals the graphene oxide selectively to reduce the oxygen, ultimately converting it into pure graphene. This reduction of graphene oxide into graphene allows for the rapid and chemical-free manufacturing of advanced electronic devices such as sensors [Luo *et al.*, 2016], supercapacitors [Lin *et al.*, 2014], and solar cells [Sygletou *et al.*, 2016], to name but a few examples. There are industrial motivations to use graphene oxide as the initial material for creating electronically active surfaces – in contrast to graphene or graphite, graphene oxide is soluble in water and can be easily cast onto films using liquid-phase chemistry.

The targeted irradiation allows the reduced graphene oxide to be patterned onto solid substrates without pre-patterned masks in only a few minutes. While graphene is electrically conductive, graphene oxide is not – patterns of graphene in an insulating material form electric circuits. The laser irradiation process enables the scalable and cost-efficient fabrication of miniaturized electronic devices [El-Kady and Kaner, 2013].

Raman spectroscopy is a common technique for determining the quality of the laser-irradiated graphene oxide through observing how laser photons scatter after they interact with the vibrating molecules in the sample probe. The intensities of the characteristic D and G bands in the Raman spectra can be used to judge to what extent the graphene oxide has been reduced to graphene, i.e. the quality of the resulting material. The D and G bands result from the defects and in-plane vibrations of sp^2 carbon atoms. In particular, the degree of reduction of the graphene oxide, and thus the conductivity of the irradiated area, can be quantified through the ratio of the intensities of the D and G bands – the smaller this ratio, the more the precursor material has been reduced.

The main factors affecting the quality of the reduced material are the experimental parameters of the irradiation process. A recent study emphasizes the need to optimize these parameters to achieve good results [Wan *et al.*, 2019]. The authors focus on the understanding of the sub-processes involved in reduction process and how their interplay can be affected by changing the laser power and the irradiation duration. Their ultimate goal, as in this paper, is to produce graphene with desired properties. However, they consider only a subset of the parameters we take into account here and rely solely on manual tuning of the values of those parameters.

An in-depth description of ways of reducing graphene oxide to graphene is beyond the scope of this paper; the interested reader is referred to a recent survey on laser-induced graphene for more background [Wang *et al.*, 2018]. While there have been applications of model-based optimization in other areas of materials science (e.g. [Häse *et al.*, 2018; Talapatra *et al.*, 2018]), we present the first application to laser-induced graphene, to the best of our knowledge.

3 Methodology and Experimental Setup

The graphene oxide samples investigated in this paper were prepared from graphite powder. The powder was completely oxidized in solution via Hummers' method [Hummers and Offeman, 1958]. After centrifugation, the oxidized residue was freeze-dried into powder, dispersed in isopropanol, and sonicated twice. The resulting ink supernatant was separated; an isopropanol solution with a suspended graphene-oxide-like material was collected for each sample. The ink supernatant was used to deposit a thin graphene oxide film onto quartz substrates via ultrasonic spray deposition. The prepa-

ration of a sample to be irradiated requires about one week to produce the graphene oxide powder and 1-2 days to create the ink and deposit it onto the substrate.

Deposited graphene oxide films were placed in a sample chamber containing Argon gas. The graphene patterns were made using a continuous-wave diode-pumped solid-state laser system with a wave length of $532\,\mathrm{nm}$. The laser beam was focused with a 50x microscope lens to a spot size of $20\,\mu\mathrm{m}$ on the sample surface. Irradiated beam spots were positioned $500\,\mu\mathrm{m}$ apart from each other to ensure pristine precursor material for each experiment. Each sample is approximately $1\,\mathrm{cm} \times 1\,\mathrm{cm}$, allowing for 361 spots. In practice, the number of effective evaluations this allows for is lower because of repeated experiments and inconsistencies in the material that need to be avoided.

The quality of the laser-induced graphene spots was determined using a Raman spectrometer. As the sample is irradiated, the laser beam is backscattered and filtered through a long-pass filter to increase the sensitivity of the signal. Using the same laser source for patterning and Raman spectroscopy allows to characterize the identical spot. The Raman data for each spot were averaged over 10 measurements with a collection time of $3\,\mathrm{s}$ at laser power $<10\,\mathrm{mW}$ for each measurement. The Raman spectra were post-processed with a linear background subtraction to 0 and normalization of the maximum peak to 1. The D and G bands were fitted using a Lorentzian function and the ratio of their intensities computed as the ratio of the areas under the fitted functions.

A skilled human operator can perform about 30 experiments per day, including irradiating the sample with a given parameter configuration, performing the Raman analysis, and computing the ratio of the D and G band intensities. We are currently working on automating this process fully to increase experimental throughput.

To determine the reliability of our results, we performed three replicates each for five different parameter settings and recorded the ratio extracted from the measured Raman spectra. The median standard deviation was $\approx 6\%$ – significantly smaller than the improvements we demonstrate.

3.1 Parameter Space

We consider the following three experimental parameters that control the irradiation process.

- The power applied to the laser used to irradiate the sample. We considered a power range of $1\,\mathrm{mW}$ to $4400\,\mathrm{mW}$, the maximum power supported by this laser. The power accuracy was $\pm 0.25\%$.
- The duration a particular spot was irradiated by the laser. We varied this parameter from $710\,\mathrm{ms}$ to $20\,210\,\mathrm{ms}$. The lower value is determined by the time it takes to move the mirror used to direct the laser towards the reaction chamber in our experimental setup.
- The pressure in the reaction chamber. The values for this parameter ranged from $10\,\mathrm{psi}$ to $100\,\mathrm{psi}$. Both minimum and maximum values are the regulatory minimum and maximum safe pressures for our experimental setup, which allows the pressure to be set with an accuracy of ± 2 psi.

The parameters and their ranges give rise to a total of 7 808 200 400 possible configurations. The difficulty of exploring this parameter space, apart from its size, comes from the high cost of gathering experimental data, which is very time-consuming and labor-intensive, much more so than for computational applications. While humans are very good at identifying and exploiting patterns, in this case there is simply not enough data for them to do so. Our machine learning models, on the other hand, are able to capitalize on even small amounts of data. This is particularly relevant here as a single graphene oxide sample supports at most 361 evaluations and takes a long time to prepare.

3.2 Model-Based Optimization

We use the mlrMBO package [Bischl *et al.*, 2017] to model the parameter space, build the surrogate models, and determine the most promising configuration for the next evaluation of the underlying process. As all of our parameters are numeric, we use Gaussian Processes as the surrogate model, and a focused search with at most 10 iterations evaluating not more than 10 000 points in the parameter space using the surrogate model to select the most promising configuration with respect to the expected improvement over the best configuration discovered so far. All other mlrMBO parameters were left at their default values.

We obtained training data for the initial surrogate model by evaluating parameter settings known to perform well from previous experiments and augmenting them with evaluations of random parameter values within ranges known to yield good results. The number of configurations and the cost of evaluating them makes obtaining training data in a grid that is not too coarse prohibitive, and hence we are unable to compare to results a grid search approach would achieve.

In each iteration of the tuning process, the next configuration to evaluate is predicted by mlrMBO. The human operator sets these values and performs the experiment. The resulting D to G ratio of the irradiated spot is added to the data used to train the surrogate model, which is retrained before predicting the configuration to evaluate for the next iteration.

4 Results

We performed a series of three sets of experiments. The experimental setup, considered parameters, and objective were the same, but different graphene oxide films (manufactured using the same process) and different training data points were used. The aim of running a series of experiments was to show that our method works with slight variations in the precursor material, as two thin films of graphene oxide are not exactly the same even if they are manufactured using the same process, and to show that we can achieve improvements with different training data.

In the first set of experiments, we used 19 different parameter configurations to train the initial surrogate model and performed 8 iterations of our automated parameter tuning approach. In the second set, we used 44 training data points and 48 tuning iterations, while in the third set, there were 20 training data points and 20 tuning iterations. The total experimental effort was more than a week of human operator time, in addition to the effort of preparing the samples.

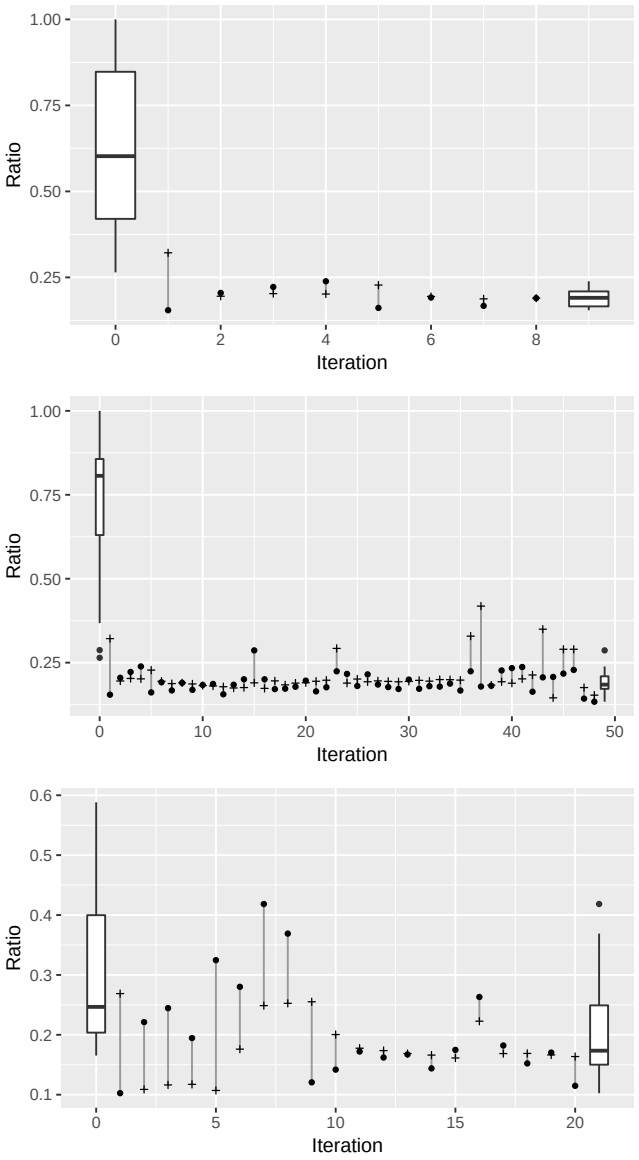

Figure 1: Performance values (ratio of D to G intensities) for three sets of experiments. On the left of each graph, the distribution of ratios for the training data is shown (iteration 0). The circles show ground-truth data, crosses values predicted by the surrogate model (connected to the corresponding ground-truth value with a grey line). The boxplot on the right of each graph shows the distribution of ratios of the ground-truth values of the configurations that the surrogate model explored. In all cases, this distribution is better than the distribution from the training data.

Figure 1 shows the performance successive tuning iterations achieve. We note that performance improves significantly as soon as we explore configurations that were predicted by the model-based parameter tuning approach. Predictions become more accurate (the distance between actual and predicted values becomes smaller) as more experiments are performed and the surrogate model is trained on more data, with a few exceptions. While we achieve significant performance improvements in all three sets of experiments, they are particularly large in the first and second set, where the distributions of ratios from the training set and the from predicted configurations barely overlap. The results in the third set of experiments are not as good because we intentionally included a few very good configurations from the first two sets of experiments to get a stronger baseline. We still achieve significant improvements over the best result in the training data, and improve on the best configuration discovered in our previous automated parameter tuning experiments.

The best ratio of D to G intensities we achieve is ≈ 0.1, with a base ratio of ≈ 1 for the precursor material. The best ratio our human experts were able to achieve was ≈ 0.2 – introducing automated parameter tuning with surrogate models improves performance by a factor of two in a small number of iterations with relatively little experimental effort.

Figure 2 shows what parts of the parameter space the automated tuning explores and the results achieved. The graphs are qualitatively similar for all three sets of experiments – while the first few iterations propose and evaluate configurations that are similar to the ones in the training data with respect to the ranges of the parameter values, the automated tuning approach quickly moves into parts of the parameter space that was not considered by the human experts. For example, in the third set of experiments, while the first few iterations (iterations 1 through 7) remain within or close to the range explored by the human experts, the predictions move into an unexplored area of the space (iteration 8), only to find that performance does not improve there, and jump into a different part of the space at the edge of the region explored by humans (iteration 9). Performance improves, and the surrogate model spends most of the remaining iterations exploring this part of the space. This demonstrates the power of our approach – not only do we achieve performance improvements, we also explore parts of the parameter space that human experts did not think to explore.

The main reason for this advantage of the automated parameter tuning approach is that we consider large ranges of values that humans tend to explore in "natural" steps. As an example, the configuration that achieved the best performance had a laser power of $486\,\mathrm{mW}$, an irradiation duration of $4377\,\mathrm{ms}$, and a pressure of $53\,\mathrm{psi}$. A human would never think of trying these particular values, but round them instead.

Inspection of the parameter tuning results revealed that irradiation time was the most important parameter, closely followed by laser power. Pressure played only a small role.

5 Discussion

There are numerous approaches in the literature that reduce graphene oxide to graphene. [Zhang *et al.*, 2010] report D to G intensity ratios of up to 0.89 for their laser-induced graphene, while [Guan *et al.*, 2016] achieve ratios of 0.27. [Sokolov *et al.*, 2013] achieve 0.29 and, with an experimental setup that is closest to ours, [Tao *et al.*, 2012] achieve 0.96.

Most of the results in the literature are not directly comparable to ours, as the experimental setup differs. Other approaches use different kinds of lasers, pattern graphene oxide on different substrates and with different thicknesses, and use

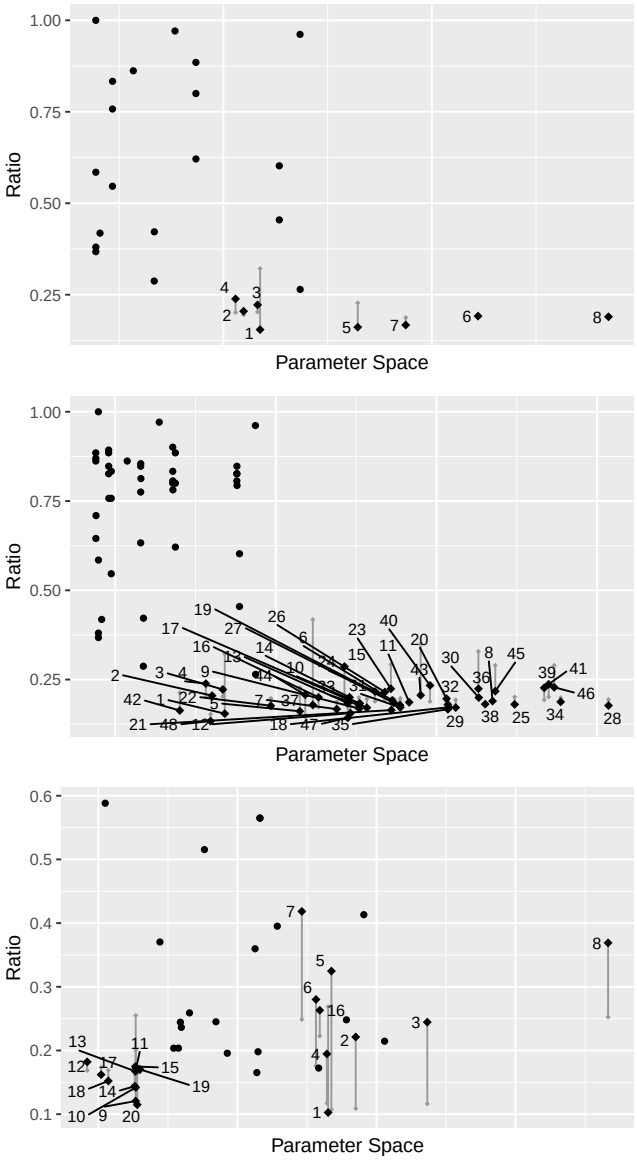

Figure 2: Performance values (ratio of D to G intensities) in parameter space, projected into a single dimension (x axis) using principal component analysis. Circles denote training data that was used to build the initial surrogate model, diamonds data obtained during the parameter tuning. Black diamonds denote ground-truth data, grey diamonds the values predicted by the surrogate model (connected to the corresponding ground-truth value with a grey line). The numbers denote the tuning iteration, i.e. the point labeled 1 was predicted by the surrogate model using only the training data, point 2 was predicted from the training data and the ground truth from point 1, etc.

different gases in the reaction chamber. Furthermore, they usually irradiate larger areas or lines. The reason we chose to irradiate spots in this paper is that this allows us to perform many more experiments on a graphene oxide sample of the same size in the same time. Given the large cost of running experiments, we preferred this approach. However, the techniques we present here are applicable for the approaches we reference as well. While minor modifications may be required, for example replacing the duration for irradiating a spot with the velocity for irradiating a line, we are confident that similar performance improvements to what we have shown here can be achieved.

Even though our experimental setup is not directly comparable to other setups in the literature, the large improvement we have been able to achieve (almost a factor of three) gives us confidence that we have indeed identified a new state of the art for laser-induced graphene production.

6 Conclusions and Outlook

We have applied state-of-the-art AI techniques, in particular surrogate-model-based optimization for automated parameter tuning, to the real-world problem of reducing graphene oxide to graphene through laser irradiation. By using AI to optimize the experimental parameters, we were able to improve over results previously reported in the literature by almost a factor of three, and over results achieved by human experts by up to a factor of two.

While the surrogate models we develop here achieve very good results, our understanding of the underlying processes is not improved. Leveraging these models for such purposes, for example through techniques that fall under the umbrella of explainable AI, would allow us to close the scientific loop and have AI work in tandem with human researchers to not only achieve better experimental results, but also to develop new theories in future work.

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