Machine learning and atomic layer deposition: predicting saturation times from reactor growth profiles using artificial neural networks

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In this work we explore the application of deep neural networks to the optimization of atomic layer deposition processes based on thickness values obtained at different points of an ALD reactor. We introduce a dataset designed to train neural networks to predict saturation times based on the dose time and thickness values measured at different points of the reactor for a single experimental condition. We then explore different artificial neural network configurations, including depth (number of hidden layers) and size (number of neurons in each layer) to better understand the size and complexity that neural networks should have to achieve high predictive accuracy. The results obtained show that trained neural networks can accurately predict saturation times without requiring any prior information on the surface kinetics. This provides a viable approach to minimize the number of experiments required to optimize new ALD processes in a known reactor. However, the datasets and training procedure depend on the reactor geometry.

I. INTRODUCTION

Machine learning and in particular artificial neural networks have revolutionized how we think and work with data. While artificial neural networks are not new, and they have long been explored as a way of modeling complex, non-linear relationships between different variables, the development of powerful tools capable of implementing and optimizing large networks combined with increasingly powerful computing capabilities has lead to a veritable explosion in the range of applications and types of architectures.

At their core, feedforward artificial neural networks are universal function approximators, capable of modeling connections between inputs and outputs of arbitrary complexity. This makes them a useful tool to develop surrogate models without having to carry out complex calculations. One of the challenges, at least in the context of physical sciences, is that they require large amounts of data for training. For instance, the MNIST dataset, one of the most commonly used entry-level machine learning datasets, is composed of 60,000 pictures of handwritten digits for training, plus an additional 10,000 samples for testing.

In this work we explore the application of deep neural networks in the context of the optimization of atomic layer deposition processes. In particular, we focus on a very practical question: given thickness measurements from a set of samples distributed inside a reactor, can we predict the dose time that would lead to saturation everywhere inside the reactor? The experimental measurement of thickness profiles using a set of samples or witness coupons is a common approach used in research labs and in industry as part of the qualification of a new ALD process or reactor. By training artificial neural networks to develop a surrogate model capable of predicting saturation times from growth profiles we can minimize the number of experiments required.

In particular, we introduce datasets designed to train neural networks to predict saturation times based on the dose time and thickness values measured at different points of the reactor for a single experimental condition. We then explore different artificial neural network configurations, including depth (number of hidden layers) and size (number of neurons in each layer) to better understand the size and complexity of that neural networks should have to achieve high predictive accuracy. Finally, we evaluate the minimum number of experimental data points required to achieve high classification accuracies. The dataset and the code implementing the networks and the training and evaluation processes have been made available upon publication of this work, and they can be found online at https://github.com/aldsim/saturationdataset.

II. METHODOLOGY

A. Dataset

In order to explore the application of neural networks to ALD process optimization, we have created a simple dataset connecting growth profiles and precursor dose times with saturation times.

As mentioned in the introduction, one of the challenges of neural networks is that they usually require large datasets for training. This is a challenge from an experimental standpoint. In a prior work we demonstrated that computational fluid dynamic models provided excellent quantitative agreement with experimental growth profiles in our own ALD reactors. Therefore, in this work we have used these models to generate a large dataset that covers a representative range of experimental parameters expected in an ALD process.

For each sample in the dataset, we randomly select values for the model input parameters. These include precursor pressure, molecular mass, process temperature, sticking probability, growth per cycle, and the corresponding dose time. We then simulate the reactive transport of the precursor inside the reactor both to the selected dose time, and we calculate the dose time required to achieve full saturation. The result is a set of thickness values at specific locations, the dose time, and the saturation time: \((x, t_d, I_{sat})\).

By repeating this process, we have constructed a series of
datasets comprising 100,000 independent samples for training, plus 10,000 independent samples for testing. For this work, we have chosen a cylindrical horizontal viscous flow reactor configuration analogous to the custom-built reactors in our laboratory. We have created independent datasets for the following number of samples in the reactor (in parenthesis the separation between consecutive points): 20 (2 cm), 16 (2.5 cm), 10 (4 cm), 8 (5 cm), 5 (8 cm), and 4 (10 cm).

B. Model

We have used the datasets described in Section II A to explore the application of deep neural networks to learn the functional relationship:

\[(x, t_d) \rightarrow t_{\text{sat}} \quad (1)\]

between growth profiles and dose time (the experimental observables) and the saturation dose time (our optimization target) that is central to any ALD process optimization.

To this end, we have explored three different networks, shown in Figure 1, a shallow network and two different deep networks with one and two hidden layers. All networks use the vector of thickness values and the dose time as input values, providing the predicted saturation time as output. In order to achieve high accuracies over a range of times spanning more than two orders of magnitude, we used the logarithm of the dose and saturation times in seconds as inputs and predicted targets.

In all cases, layers are connected with all-to-all connections, so that the output for each layer is given by:

\[a_i = \text{ReLU}(W a_{i-1} + b) \quad (2)\]

where ReLU(·) represents the rectified linear function [Figure 1(d)]. One of the motivations to use all-to-all connectivities instead of convolutional layers is that it encompasses the case where samples inside the reactor are not equidistant from each other, but may be located downstream or upstream to a substrate of interest. For the deep networks, the size of each of the hidden layers, \(M\) for the case of a single hidden layer and \(M_1\) and \(M_2\) for the two hidden layer network, are additional adjustable parameters.

C. Implementation, training and testing

We implemented the neural networks and carried out the training and testing in Pytorch, a free open source framework for deep learning. Each network was trained against the training dataset using using stochastic gradient descent with a Mean Square Error (MSE) loss function and the Adam optimizer using a learning rate of \(10^{-3}\). Each network was trained for 100 epochs using batches of 64 samples. The implementation and training script is provided in the Supporting Information and can be found online at https://github.com/aldsim/saturationdataset.

The resulting networks were then tested against the testing dataset. While the MSE was directly used as a loss function, for analysis and visualization we used the relative difference of the predicted saturation time, defined as:

\[\varepsilon = \frac{t_{\text{pred}} - t_{\text{sat}}}{t_{\text{sat}}} \quad (3)\]

For a highly performing, unbiased network we expect this error to have an average close to zero. The variance of \(\varepsilon, \sigma_\varepsilon\), therefore provides a good estimator of the prediction error.

III. RESULTS

In Figure 2 we show a sample of the prediction errors of three different neural networks: a shallow network, a network with one hidden layer and a network with two hidden layers. The networks are trained to predict saturation times from the testing dataset comprising 20 thickness values. Data points are colored based on how close was the dose time of the profile to the actual saturation dose, with darker points being closer to the saturation conditions.

It is apparent that the shallow network is not capable of accurately predicting the saturation times, with the predicted times diverging \(\pm 20\%\) from the true saturation value. In contrast, the deep network with one hidden layer [Fig. 2(b)] shows a much smaller dispersion and an excellent agreement with the predicted saturation times. It is interesting to note that the error seems to increase for the deep network with two hidden layers [Fig. 2(c)]. We attribute this to an overfitting of the training dataset due to the larger number of free parameters available in the network with two hidden layers. This emphasizes the importance of having separate training and testing datasets.

As mentioned in Section II C we can use the mean and standard deviation of the relative error \(\varepsilon\) to quantify the network’s accuracy. In the case of the profiles shown in Figure 2 the standard deviation values are 0.10, 0.007, and 0.015, respectively.
FIG. 2. Prediction errors in the testing dataset for one-shot prediction of saturation times from a single growth profile for three different neural networks: A) Shallow network; B) 1 hidden layer ($N = 30$); C) 2 hidden layers $N_1 = 30$, $N_2 = 10$. Data is shown for datasets comprising 20 independent thickness values. The data is color-coded according to how far the dose time used for prediction is to the saturation time (lighter means shorter dose times).

A. Impact of network size and number of experimental points

A fundamental question when measuring growth profiles is how many experimental points are needed to accurately capture the change in film thickness inside a reactor. In order to understand the impact that the number of points has on the ability to accurately predict saturation times, we trained our networks against a collection of datasets comprising different number of homogeneously distributed points. In Figure 3 we show the mean error and the standard deviation $\sigma_e$ of the predicted saturation time for the three networks shown in Figure 2 when trained on different numbers of inputs. The results show that the networks with one and two hidden layers can accurately predict the saturation time with as few as 8 thickness values. Using fewer values still produces results that are much more accurate than those obtained using a shallow neural network, but the standard deviation in the predicted saturation times start to significantly increase. In Figure 4 we show a visualization of the dispersion in the predicted saturation values for the datasets comprising growth profiles with 4, 5, and 10 points.

Finally, we have explored the impact of network size on its performance. In Figure 5 we show the classification performance of a neural network with one hidden layer as a function of the number of independent points in the growth profile for different hidden layer sizes. The results show how at least 20 neurons are needed in the hidden layer to maximize the network’s ability to predict saturation times. This corresponds to $21 \times n + 61$ free parameters, where $n$ is the number of input data points.
FIG. 4. Saturation time prediction accuracy using a 1 layer deep neural network ($M = 30$) for different numbers of data points in the growth profile: A) $N = 4$, B) $N = 5$ and C) $N = 10$. The data are color-coded according to how far the dose time used for prediction is from the saturation time (lighter means shorter dose times).

IV. CONCLUSIONS

These results demonstrate how deep neural networks can be used to predict future behavior of reactive transport systems without any prior knowledge of the surface kinetics, something that could help accelerate the optimization of manufacturing processes based on thin film deposition and surface modification technologies. Compared to the networks used in traditional machine learning domains such as image classification, the number of free parameters required to achieve a good agreement is significantly smaller, 691 for the case of a network with a single hidden layer with $N = 30$ neurons.

The trained neural networks can be interpreted as surrogate models capturing the underlying physics of the reactive transport of precursors inside an ALD reactor. While the behavior far into the future of the differential equations modeling precursor transport cannot be expressed as closed expressions, the training process is able to capture this functional relation from the pre-existing dataset, sidestepping the need to solve the transport models in real time.

Finally, it is important to mention that the results of the training process are reactor-specific. Consequently, specific datasets should be generated for each type of experimental reactor. However, a single trained model can be used to predict the saturation times of arbitrary processes, including those for which no experimental information on the surface kinetics is available. Based on the results presented in this work, the accuracy seems to be limited primarily by how accurately the kinetics of a given precursor are captured in the training dataset. Therefore, departures with respect to the expected behavior could also be used to identify non-idealities in the underlying surface kinetics.

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DATA AVAILABILITY STATEMENT

The data that support the findings of this study has been made openly available in github: https://github.com/aldsim/saturationdataset.

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