SCHOOL OF MECHANICAL ENGINEERING

UNIVERSITY OF LEEDS

MECH3890 – Individual Engineering Project

Using Machine Learning to Optimise the MW-PECVD Process for Polymer Substrates in Industry

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If the project is industrially linked, tick this box and provide details below

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Abstract

Thin film coatings are desired for materials because of their ability to alter and improve properties of materials such as increasing their hardness and Young's modulus. Within these coatings, diamond-like carbon is one of the most popular due to its versatility and wide range of desirable properties.

Typically, metal substrates have been used in thin film coating deposition processes due to their high operating temperatures allowing them to withstand the high temperatures involved in these processes. However, with the novel microwave plasma enhanced chemical vapour deposition technique, the temperature of the process can be kept as low as 70°C which allows polymer substrates with lower operating temperatures to be coated in thin films. By depositing thin film coatings on to polymer substrates, their hardness and Young's modulus can be increased to allow them to be used in high load applications which they wouldn't be suited for before and this would be desirable for industries as polymers are typically cheaper to produce and easier to shape than metals.

However, tuning the deposition parameters to ensure the desired properties of the deposited coating are obtained while also ensuring the temperatures remain below the maximum operating temperature of the polymer can be challenging and can act as a barrier to entry for manufacturing industries. For this reason, this project developed an optimisation tool which used a machine learning model to recommend the optimum deposition parameters for industries to obtain their desired hardness and Young's modulus values for their diamond-like carbon coatings for polymer substrates while taking into consideration economic and sustainability issues.

Two machine learning models were trained, tested, and validated against literature in this project using the scikit-learn machine learning library in Python: a traditional machine learning multi-linear regression model and a deep learning multi-layer perceptron regressor neural network model. These two models were compared and, despite being significantly more computationally expensive, the neural network model was found to be more accurate and identified non-linear relationships between the deposition parameters and the hardness and Young's modulus values and so it was used to develop the optimisation tool.

Chapter 1: Introduction

1.1. Introduction

Thin film coating deposition refers to the methods used to apply thin films on to substrate materials to give them desirable properties and the global thin film coating market is projected to reach $13.6 billion by 2024 due to its many manufacturing applications [1, 2]. In chemical vapour deposition, a common type of deposition technique, chemical reactions between gas reactants, called precursors, take place above the surface of a substrate to form the coating [3, 4]. In plasma enhanced chemical vapour deposition (PECVD), the reactions are activated with plasma (energised ions) generated from an unreactive gas, e.g. argon and in microwave PECVD (MW-PECVD), the plasma is generated using microwave (MW) power. The generated plasma means the need for the high temperatures (~700°C) required in typical chemical vapour deposition processes is circumvented and the temperatures in MW-PECVD can be kept as low as 70°C [3-5]; this allows for coatings to be deposited on to polymer substrates which wouldn't be possible with other techniques due to the high temperatures damaging the substrates [3, 5].

Polymers are a popular engineering material in industry as they are cheap to produce and easy to shape [3]. However, the use of polymers in high load applications, such as for automotive components or surgical medical equipment, is limited due to their relatively low hardness and Young's modulus; thin film coatings can be utilised to improve these properties to allow for polymers to be expanded into these applications [3, 4, 6]. In 2017, Carley undertook a study which found diamond-like carbon (DLC) thin film coatings deposited through MW-PECVD to adhere well to acrylonitrile-butadiene-styrene (ABS) and polyetherimide (PEI) polymer substrates, two common engineering polymers, and increased their hardness and Young's modulus up to 25x and 7x, respectively [3].

DLC, the most popular thin film coating within thin film coatings due to its versatility, is composed of carbon atoms which have undergone either sp2 or sp3 hybridization and, depending on the sp3:sp2 ratio, its structure can either more closely resemble graphite or diamond [4]. Having a higher sp3:sp2 ratio will yield higher hardness and Young's modulus values, resembling diamond, while a lower ratio will yield lower values resembling graphite [4]. The precursors commonly used to deposit DLC are hydrocarbons, e.g. acetylene, which also hydrogenate the DLC [3, 4]. The versatility of DLC coatings comes from the fact that the sp3:sp2 ratio and hydrogen percentage, and hence the properties, can be tailored by altering the deposition parameters used to deposit the DLC coating which allows it be suitable for a range of different applications such as for automotive components or surgical medical equipment [2-4].

However, in manufacturing industries, this versatility can act as a barrier to entry as many high-level employees don't have the necessary knowledge to alter the deposition parameters appropriately to obtain the mechanical properties they desire. In addition to this, especially for polymer substrates due to their far lower operating temperatures relative to the more commonly used metal substrates, care must be taken for the parameters used to not result in temperatures greater than the operating temperatures of the substrate and cause damage to the material. For this reason, industries must hire surface engineering experts for their deposition processes, but machine learning (ML) could be used to make MW-PECVD more accessible.

ML refers to models which use real-world data for a particular process to predict future outcomes for any input without explicit programming [7]. Deep learning (DL) is a subset of ML where neural networks, through the use hidden layers between the input and output layers, are used to allow computers to learn from data in a way similar to the human brain [8, 9]. Both traditional ML and DL have found applications in modelling and optimising many different manufacturing processes. For example, a study by Qi et al. demonstrated the use of neural networks in optimising additive manufacturing processes by using a neural network to carry out regression analysis on printing process parameters and mechanical properties of printed parts [9]. DL models, such as the aforementioned, can be used to develop tools which guide industries on the optimum parameters to use to achieve the results they require, such as for MW-PECVD of DLC coatings with specific hardness and Young's modulus values, which minimises waste due to unsuccessful attempts.

In regression analysis, neural networks benefit from being able to predict complex non-linear relationships, which traditional ML methods, e.g. multi-linear regression (MLR), are unable to. However, DL solutions require more computational power and time to be trained than traditional ML solutions and they both perform relatively similar when being used to predict simple, linear relationships or when being trained on small datasets; in these scenarios, due to them requiring less computational power, traditional ML solutions are better suited [7]. Therefore, it's important to evaluate both when deciding on an appropriate model. Typically, when assessing the accuracy of these models, an accuracy of 80% is considered 'good' and 90% is considered 'highly accurate' according to Lewis, 1982 with 80% being what is typically aimed for with ML models [10, 11].

1.2. Aims

This project aims to determine whether a traditional ML model or a DL neural network model is more suitable to predict the relationship between MW-PECVD parameters and the mechanical properties of DLC coatings deposited on to polymer substrates and to use the most suitable model to develop a tool to optimise the MW-PECVD process for polymers in industries.

1.3. Objectives

1. Obtain real-world data on the hardness and Young's modulus of DLC coatings deposited on polymers through MW-PECVD
2. Use obtained data to train a traditional ML MLR model to have an accuracy of >80% in predicting hardness and Young's modulus of DLC coatings deposited on to polymers through MW-PECVD
3. Use obtained data to train a DL neural network model to have an accuracy of >80% in predicting hardness and Young's modulus of DLC coatings deposited on to polymers through MW-PECVD
4. Compare MLR and neural network models to assess which is more appropriate by comparing accuracy and performance
5. Develop a MW-PECVD optimisation tool for industries which uses the most suitable model

1.4. Report Layout

Chapter 1 is intended to introduce the project's aims and relevant background information. Chapter 2 discusses the real-world data obtained on MW-PECVD parameters and the mechanical properties of DLC coatings on polymer substrates used to train the ML models of this project. Chapter 3 discusses the methodology behind training a traditional ML MLR model and the results from testing it and comparing it against literature. Chapter 4 discusses the methodology behind training a DL neural network model and the results from testing it, comparing it against literature and comparing it against the MLR model from Chapter 3 to determine which is more suited MW-PECVD of DLC for polymers. Chapter 5 discusses the methodology taken to develop an optimisation tool which uses the most suitable ML model to predict the optimum MW-PECVD parameters industries should use for their polymers. Finally, Chapter 6 covers the project's achievements and shortcomings/limitations along with possible future work to be carried out.

Chapter 2: Training/Testing Data Collection for ML Model

2.1. Introduction

To train any ML model to predict the outcomes of a process, an initial set of real-world data of the input variables and output variables is required [7]. In this study, the ML models are being trained to understand the relationship between MW-PECVD parameters and the mechanical properties of DLC coatings deposited on to polymer substrates and so data is required for mechanical properties of DLC coatings deposited on to polymer substrates using MW-PECVD and the values set for the deposition parameters. In a study by Zhang et al. on determining suitable sample sizes for applying ML models to materials science processes, they determined that to achieve an accuracy of 80%, a sample size of at least 100 should be used to train the model [11]. To test ML models, a small portion of the data, typically 20% due to the Pareto principle, is set aside to assess the accuracy of the trained model by comparing the model's predictions against the actual values in the testing data [12].

In 2017, Carley undertook a study into the effects of the MW power, working pressure and acetylene/argon gas flowrate ratio in MW-PECVD on the mechanical properties of DLC coatings deposited on to polymer substrates to help improve their performance in industry [3]. In this chapter, the data obtained from Carley's study will be judged to determine whether it's suitable for training and testing the ML models for this study based on the methodology employed during their study.

2.2. Methodology

Carley used the Hauzer Flexicoat 850 Coating System at the University of Leeds to deposit DLC coatings, through MW-PECVD with 10 different variations of the MW power, working pressure, precursor (acetylene) flowrate and carrier gas (argon) flowrate, on to two polymer substrates, ABS and PEI, so that 20 deposition processes were run in total [3]. The data obtained by Carley's study (see Appendices A and B) was used to obtain the training/testing datasets for the ML and DL models developed in this project.

Carley concluded the values recorded for the mechanical properties of the DLC coating of the ABS substrate at Leg 8 to be anomalous after plotting a graph of the Young's modulus values against the hardness values and finding those values to not fit the positive correlation trend [3]. For this reason, these values were not included in the training/testing datasets for the ML models.

When splitting the data into training and testing data, 1 leg was randomly selected for each the ABS and PEI substrate so that the rule of thumb of 20% of the raw dataset being separated for testing was followed. Leg 6 of the ABS depositions and Leg 3 of the PEI depositions were selected and were separated to form the training dataset and the rest of the data was used to train the ML models.

2.3. Results

The training and testing datasets extracted from the data obtained by Carley's study and used for the ML models are shown in Table 2.1 and Table 2.2, respectively.

Table 2.1: Training data obtained from Carley's study to train ML models [3]

MW Power (W)	Working Pressure (mbar)	Gas Flowrate Ratio (%C2H2)	Hardness (GPa)	Young's Modulus (GPa)
1032	0.012	94	4.9	21.9
1087	0.011	92	4.1	19.8
1032	0.011	85	3.4	19.2
1087	0.009	40	2.1	14.0
1178	0.009	74	3.9	20.2
1178	0.011	91	4.2	18.5
1032	0.011	66	3.3	16.9
1087	0.013	67	4.7	23.1
1032	0.012	94	4.9	24.6
1087	0.011	92	4.1	22.4
1087	0.009	40	2.2	15.3
1178	0.009	74	3.6	22.1
1178	0.011	42	2.7	16.9
1178	0.011	91	4.8	25.0
1032	0.013	43	3.0	18.3
1032	0.011	66	4.5	24.2
1087	0.013	67	4.3	24.7

Table 2.2: Testing data obtained from Carley's study to test ML models [3]

MW Power (W)	Working Pressure (mbar)	Gas Flowrate Ratio (%C2H2)	Hardness (GPa)	Young's Modulus (GPa)
1178	0.011	42	2.5	13.5
1032	0.011	85	4.1	24.8

2.4. Discussion

For their study, Carley varied the MW power, working pressure, acetylene flowrate and argon flowrate of the MW-PECVD process in a Hauzer Flexicoat 850 Coating system [3]. The results obtained by Carley may vary for other coating machines and precursor and carrier gas combinations which means any ML models trained on this data would have the limitation of giving inaccurate results for industries using different combinations. For this reason, the final tool developed in this project should inform the user of the machine and gasses the ML model has been trained using.

Although the Flexicoat coating system did have the ability to apply a bias voltage across the substrate, the substrates used in Carley's study were electrically insulating and bias voltages across them would have had no effect on the deposition process and so Carley didn't apply one [3]. However, many studies have shown that altering the bias voltage across an electrically conductive substrate in PECVD processes affects the coating's mechanical properties which is why industries commonly apply bias voltages to tune these properties [13]. Although a self-bias was applied by Carley across the plasma to accelerate ions to the substrate, the coating machine had no feature to measure this and so this parameter wasn't varied during Carley's study [3]. A study by Gou et al. showed that the self-bias during PECVD processes is a factor in the mechanical properties of the DLC coating deposited and so it's important for industries to know what self-bias to use when running their MW-PECVD processes but, since Carley's study was unable to record the self-bias used, not providing a suitable self-bias value was another limitation of the final tool developed in this project [3, 14]. For these reasons, the final tool should inform users that the ML model has been constructed based on MW-PECVD processes carried out with a constant self-bias of unknown voltage and without a bias voltage across the substrate.

Substrate temperature was another parameter which is a factor in the mechanical properties of DLC coatings that wasn't varied by Carley in their study due to the low operating temperatures of ABS and PEI [3, 15]. ABS has a lower maximum operating temperature than PEI of 80°C so Carley kept the substrate temperature at 70°C as a higher temperature would damage the substrate while a lower one would cause the MW-PECVD process to not be carried out successfully due to a plasma not being maintained [3, 6]. The final tool should display the substrate temperature the ML model had been trained on to ensure industries don't allow the temperature to differ from this as this would make the optimisation tool inaccurate and also to warn them to not use engineering polymers with maximum operating temperatures below this value as the recommended parameters could damage these.

Before selecting the values for the deposition parameters, Carley ran MW-PECVD processes in a trial-and-error approach where the parameters were varied to find the extreme limits which would allow a stable plasma discharge in the deposition to be maintained and ensured the temperature would be kept below the maximum operating temperature of the polymer substrates used [3]. Once these bounds were determined, the parameters used for the actual experiments were kept within these bounds so that the DLC coatings would be deposited successfully on to the substrates [3]. Because setting the parameters outside the boundaries set by Carley would not allow for the deposition process to occur or could damage common polymer substrates, it was decided that the optimisation tool should not suggest parameters outside the ranges used by Carley. Furthermore, the model would have to be extrapolated to be able to suggest parameters outside the ranges used by Carley which would reduce the accuracy of the predictions [7].

Although 19 is too small a sample size (<100) to train/test a ML model to be 80% accurate according to Zhang et al., Carley's study economised greatly on the experiments run by following a design of experiments (DOE) approach which allowed for several variables to be changed per trial rather than a conventional approach of only changing a single variable per experiment [3, 11]. By using this DOE approach, Carley maximized the information obtained on the relationships between the deposition parameters and the mechanical properties of the DLC coatings which suggests the data obtained is suitable for training a ML model despite its small size.

After the MW-PECVD processes were carried out, nanoindentation tests were carried out on the DLC coatings by Carley to find their hardness and Young's modulus values [3]. These indentations were taken at 5% of the depth of the film thickness which followed the Oliver and Pharr method of taking readings of the properties of DLC coatings at less than 10% of the film thickness to ensure there was minimal effect from the substrate on the coatings' recorded properties [16]. This means the ML models trained on this data will be valid for other polymer substrates which ensures the robustness of the final optimisation tool.

Although Carley's study varied the gas flowrates in the experiments, the gas flowrate ratio values were used for this project due to the actual flowrates and the working pressure being interdependent [3]. During MW-PECVD, a high vacuum is created in the deposition chamber and the gases are added which increases the working pressure [17]. Since most traditional models, such as MLR have an assumption that the independent variables aren't interdependent, flowrate ratio was used to train the models in this project as these values are independent of the pressure and would therefore reduce the risk of inaccuracies in the model [7].

Chapter 3: Training Multi-Linear Regression Model

3.1. Introduction

This chapter aims to detail the steps taken to train a traditional ML MLR model to identify the relationships between the MW-PECVD parameters and the hardness and Young's modulus of DLC coatings deposited on to polymer substrates at an accuracy of >80%. This chapter also aims to validate this model by comparing the results of its testing against literature to assess how suitable traditional machine learning is to optimise the MW-PECVD process of DLC coatings for polymers in industries.

MLR models are a traditional ML method used for continuous, tabular data which assume linear relationships between variables [7]. These models utilise the ordinary least squares method which calculates the coefficients and intercepts of lines of best fit for the data which minimise the sum of squared differences between values predicted by the line and the actual values [18]. The assumption of a linear relationship means this model has the limitation of not recognising non-linear relationships between the deposition parameters and the mechanical properties of the DLC coatings. However, since this model only provides a single coefficient to each input variable for every output variable, the model can easily be validated by assessing whether these coefficients are reasonable and can be explained by literature; this would be difficult with a more complex neural network which is commonly called a 'black-box' since it assigns multiple coefficients to each input variable [7, 19]. Furthermore, the model's relatively simple nature means it requires less computational power to run which would be desirable for industries [7].

To assess the accuracy of ML models, including MLR models, metrics such as mean squared error (MSE) or mean average percentage error (MAPE) are used which are calculated from comparing the predictions of the model to actual values in the testing data [8, 10, 20]. Since the outputs of this model, hardness and Young's modulus, have different scales, it was deemed more appropriate to assess using percentage. The accuracy of a model can be calculated by subtracting the MAPE from 100% and for the accuracy of this model to be 'good', it should be 80% [10].

Python is a commonly used programming language for ML programs due to its variety of dedicated libraries [20]. Libraries refer to a group of modules which contain pre-written code to allow developers to carry out tasks without having to write the code from scratch. One such library is scikit-learn, written by Pedregosa et al. which is a ML library for Python which contains various ML algorithms, including MLR, which was used for this project [8, 20].

3.2. Methodology

The training and testing datasets from Table 2.1 and Table 2.2, respectively, were imported into a Python script (see script in Appendices C and D). These datasets were then divided into the input and output variables with the input being the MW power, working pressure and gas flowrate ratio and the output being the hardness and Young's modulus.

For linear regression, it's important to scale the input data if they have different scales to ensure all the input variables are treated equally when the model is calculating their coefficients [20]. For the deposition parameters, this is especially important as they're all on very different scales; for example, the MW power values have a magnitude of 10^3 and the pressure values have a magnitude of 10^{-2}. For this reason, each parameter value in the training and testing datasets was min-max scaled, using Equation 1, in the range [0,1] before being used to train/test the model.

$$x_{scaled} = \frac{x - \min(x)}{\max(x) - \min(x)} \tag{1}$$

An MLR model was brought into the Python script from the scikit-learn library and was trained on the training datasets. During this fitting, a timer was set in the Python script to measure the time taken for this process to complete to compare with the time taken for the same process for the neural network model in Chapter 4 to assess how computationally expensive each model is relative to each other. Due to this time varying between runs, the model was trained 3 times to get an average value.

Once the model had been trained, its coefficients were extracted to be compared with literature to assess the accuracy of the model. The model was also assessed by using the hardness and Young's modulus predicted by the model and the actual values in the testing data to calculate a mean average percentage error (MAPE) score for the model following Equation 2.

$$MAPE = \frac{1}{n}\sum_{t=1}^{n}\frac{A_t - P_t}{A_t} \tag{2}$$

where:

n is the number of values in the testing data

A_t is the actual value from the testing data

P_t is the value predicted by the model

3.3. Results

The scaled input parameters used to train the MLR model are shown in Table 3.1.

Table 3.1: Scaled input data for training MLR model

MW Power	Working Pressure	Gas Flowrate Ratio
0.00	0.75	1.00
0.38	0.50	0.96
0.00	0.50	0.83
0.38	0.00	0.00
1.00	0.00	0.63
1.00	0.50	0.94
0.00	0.50	0.48
0.38	1.00	0.50
0.00	0.75	1.00
0.38	0.50	0.96
0.38	0.00	0.00
1.00	0.00	0.63
1.00	0.50	0.04
1.00	0.50	0.94
0.00	1.00	0.06
0.00	0.50	0.48
0.38	1.00	0.50

The average time taken to train the MLP regressor model was 0.0056s and the coefficients assigned to the parameter variables by the MLR model are shown in Table 3.2.

Table 3.2: MLR model coefficients for validating against literature

Deposition Parameter	Coefficient for Hardness	Coefficient for Young's Modulus
Power	0.273	1.329
Working Pressure	1.139	4.409
Gas Flowrate Ratio	1.727	4.863

Table 3.3 show the results obtained from testing the MLR model.

Table 3.3: Actual and predicted values from testing of MLR model

Hardness (GPa)		Young's Modulus (GPa)	
Actual	Predicted	Actual	Predicted
2.5	3.0	13.5	18.5
4.1	4.1	24.8	21.1

The MAPE calculated from the values in Table 3.3 is shown in Equation 3.

$$MAPE = \frac{1}{4}\left(\left|\frac{2.5-3}{2.5}\right| + \left|\frac{4.1-4.1}{4.1}\right| + \left|\frac{13.5-18.5}{13.5}\right| + \left|\frac{24.8-21.1}{24.8}\right|\right) = 18\% \tag{3}$$

3.4. Discussion

In their study, Carley ranked the MW-PECVD parameters on the effect they had on the hardness and Young's modulus of the DLC coatings deposited on to the ABS and PEI substrates. Gas flowrate ratio was ranked as having the most significant effect, followed by working pressure and then MW power, which was deemed to have minimal effect on the properties [3]. The assigned coefficients in the linear regression model agree with these findings from Carley's study as they show that the gas flowrate ratio has the biggest effect on the hardness and Young's modulus values of the deposited DLC coatings, then working pressure and then MW power. This suggests the MLR model was successful in modelling the MW-PECVD process.

The coefficients assigned to the flowrate ratio variable in the MLR model state that increasing it, i.e. increasing the percentage of acetylene, increases the hardness and Young's modulus of the DLC coatings. A study by Bi et al. into the influence of the acetylene content on the properties of DLC coatings deposited by PECVD found that increasing the percentage of acetylene from 50% to 75% in a mixture with argon increased the sp3:sp2 ratio, hence increasing the hardness and Young's modulus, in the DLC coatings [21]. Bi et al. speculated this was due to argon ion bombardment breaking the unstable sp3 bonds in the coating, causing sp2 bonds to form and, therefore, by increasing the acetylene percentage and reducing the argon percentage, the likelihood of this occurring decreased [21]. These findings support the coefficient assigned to gas flowrate ratio in the linear regression model and suggest the model is suitable for the MW-PECVD process.

On the other hand, the study by Bi et al. also found that increasing the acetylene percentage from 75% to 100% decreased the hardness and Young's modulus, and Bi et al. speculated this was due to the lack of argon ions meaning ion bombardment was unable to break the C-H bonds in the acetylene and encourage sp3 bonds to form [21]. This suggests that the relationships between the gas flowrate ratio and the mechanical properties of the coating aren't linear and that a neural network would be more suitable as it can identify non-linear relationships.

Similarly, the coefficients assigned to the working pressure variable are supported by a study carried out by Wang et al. into the effect of PECVD pressure on DLC coatings. The study found that increasing the deposition pressure from 50mTorr (0.067mbar) to 70mTorr (0.093mbar) increased the mechanical properties of the DLC coatings due to the increased plasma density breaking more C-H bonds in the acetylene and promoting sp3 bonds which supports the positive coefficients assigned to working pressure in the MLR model [22].

However, similar to the study by Bi et al., in Wang et al.'s study, increasing the pressure past 70mTorr (0.093mbar) caused a decrease in the hardness and Young's modulus due to the increased pressure causing gas molecules to collide with the coating and break sp3 bonds which are more unstable than the sp2 bonds [22]. This critical pressure value depends on the power and temperature used when the deposition is carried out as this plays a role in the energy of the gas molecules which means the critical pressure may not be possible to reach when keeping the substrate temperature low enough to not damage a polymer substrate [3, 22].

The coefficients assigned to MW power in the MLR model state that increasing the power would increase both the hardness and the Young's modulus of the DLC. This relationship can be explained by the fact that increasing the MW power generates a greater plasma density which is able to break the weak C-H bonds in the acetylene and promote more sp3 bonds to be formed, therefore increasing the hardness and Young's modulus in the deposited DLC coating – this is supported by a study carried out by Han et al which found that increasing the MW power in a MW-PECVD process from 750W to 1000W increased the sp3/sp2 ratio of the deposited DLC coating [23]. However, the same study found that increasing the MW power from 1000W to 1250W caused a decrease in the sp3/sp2 ratio due to the more energetic ions in the plasma bombarding the coating and breaking the less stable sp3 bonds in the DLC coating which suggests a neural network model is more appropriate for this process [23].

The model took an average of 0.0056s to train which is very insignificant and supports the statement that the MLR model requires very little computational power to train which makes it more desirable for industries. This value will be compared with the time taken for the DL model trained in Chapter 4 to assess how much more computationally expensive it is compared to this model. Since these times can vary depending on the computer used to train the model, both models will be trained using the same hardware and software.

Finally, from the MAPE score obtained of 18%, it can be stated that the model's accuracy is 'good' [10]. However, the studies discussed in this section have suggested the relationships between the MW-PECVD parameters and the DLC properties to be non-linear, this suggests a neural network model which can identify these non-linearities would reduce this error. The MAPE was compared to the score obtained for DL model tested in Chapter 4 to assess how much more accurate it is.

Chapter 4: Training Multi-Layer Perceptron Regressor Model

4.1. Introduction

This chapter aims to detail the steps taken to train a DL multi-layer perceptron (MLP) regressor neural network model to identify the relationships between the MW-PECVD parameters and the hardness and Young's modulus of DLC coatings at an accuracy of >80%. This chapter also aims to validate this model by comparing the results of its testing against literature and against the previously trained and tested MLR model to assess how suitable it is to optimise the MW-PECVD process of DLC coatings for polymers in industries and whether DL is more suitable for this than traditional ML.

Compared to other neural network models, the MLP regressor model is ideal for continuous, tabular datasets which makes it suitable for the data obtained for this project [8, 20]. Furthermore, the study by Qi et al. demonstrated the high accuracy of using a MLP regressor model in identifying relationships between printing process parameters and mechanical properties of printed parts [9]. For these reasons, the MLP regressor model was selected as the DL model for this project.

A MLP regressor model consists of an input layer, an output layer and at least one hidden layer in between which contains neurons [7]. During the training phase, each neuron in the first hidden layer receives a sum of each of the inputs multiplied by a randomly set weight (similar to a coefficient in linear regression) [20]. Each neuron also contains a randomly selected bias (similar to intercept in linear regression) which is added to the weighted sum of inputs [20]. This sum of weighted inputs and bias is passed through an activation function which determines if the sum is significant enough to be passed through to the next layer [7]. This process is repeated for each hidden layer of neurons until the output layer is reached [20]. These outputs are compared to the expected outputs from the training data by calculating the mean squared error (MSE) and this error is propagated back through the network and the coefficients and biases in each neuron are updated accordingly to minimise the error and this process is iterated until the minimum MSE value is reached; the specific method of this backpropagation is determined by the solver used for the model [24]. The parameters of the model are tuned by developers to select the appropriate number of neurons, number of hidden layers, activation functions and back-propagation methods for their specific use-cases [7].

Having multiple layers and neurons with different weightings and biases means this model can identify non-linear relationships between the deposition parameters and the DLC properties which is an advantage it has over the MLR model used in Chapter 4. However, this also essentially makes the model a 'black-box' as the weightings and biases are difficult to interpret and validated against literature. Additionally, this complexity also means additional computing power is needed, which may be detrimental to the industries using it.

4.2. Methodology

The training and testing datasets from Table 2.1 and Table 2.2, respectively, were imported into a Python script (see in Appendices E-G). These datasets were then divided into the input and output variables with the input being the MW power, working pressure and gas flowrate ratio and the output being the hardness and Young's modulus. While scaling is not as necessary for training data for MLP models since they're not as affected by scaling as MLR models are, it was decided to min-max scale the input data, in the same way as for the MLR model, to ensure the two models were trained on the exact same data so that there were no differences in performance due to data scaling [7].

A blank MLP regressor model was brought into the Python script from the scikit-learn library and the parameters of the model were set as follows. The number of hidden layers was set to 1 since many studies state that 1 hidden layer, with a sufficient number of neurons, can approximate any function and that adding more hidden layers can result in overfitting and requires extra computational power [8, 20, 25]. Although different studies have found that this configuration isn't sufficient for more complex relationships, since studies on PECVD processes, discussed in Chapter 3, have stated the relationships between deposition parameters and their mechanical properties to either be linear or follow a simple curve, 1 hidden layer was deemed sufficient to accurately identify these relationships.

The number of neurons in the hidden layer was set to 2 as many studies have usually followed a general rule of thumb of setting the number of neurons to 2/3 of the size of the input layer, 3 in this case, and be between the input layer size and the output size, 2 in this case [25]. When setting the number of neurons in the hidden layer, setting it too high causes the model to be overfit to the training data which is where the relationships predicted by the model are too specific to the peculiarities of the training data and not a general representation of the actual real-world process [26]. On the other hand, setting it too low would cause the model to not accurately recognise the relationships between the deposition parameters and the mechanical properties and this rule of thumb is designed to balance between these two [25].

The activation function in modern MLP networks is commonly set to the rectified linear unit (ReLU) function rather than the previously popular hyperbolic tangent function [27]. The ReLU activation function checks if the output from a neuron is positive; if positive, the function outputs the output value and, if negative, outputs 0 [27]. The hyperbolic tangent function maps the inputs it receives to an output in the range of -1 to 1 which is an issue because this causes the 'vanishing gradient problem' which is where the derivative of the output activation function becomes increasingly small and is unable update the weight values effectively to minimise error during backpropagation; the ReLU function doesn't suffer from this issue as it outputs the actual whole value or 0 which is why it's the most widely used activation function currently which is also why it was selected for this project [27].

The solver of the model was set as the Adaptive Moment Estimation (Adam) optimisation algorithm which is a stochastic gradient-based optimiser written by Kingma et al. in 2017 which has become increasingly popular due to its training time and validation scores. The parameters of the MLP regressor model were set so that, when back-propagating through the network following the Adam algorithm, the model would stop iterating when the change in the loss values is below the tolerance, 0.0001, for 10 iterations at which point, convergence has occurred; these are the values recommended by the scikit-learn library [8, 20]. However, if these values don't allow for the model to reach convergence, the tolerance can be increased.

Since the model initially assigns random weights and biases to each neuron, the performance of the model can vary each time the model is trained. For this reason, the 'random_state' parameter was set to a constant, arbitrarily selected number, 70, which allowed the results of this experiment to be reproducible as this parameter controls the starting values of the weights and biases [8, 20].

This model was then trained on the training data and a timer was set in the Python script to measure the time taken for this process to compare with the time recorded for the MLR model in Chapter 4 to assess how much more computationally expensive it is. Once the MLP model had been trained, the model was assessed by comparing the hardness and Young's modulus values predicted by the model and the actual properties in the testing data to calculate an MAPE score for the model, using Equation 2, which was compared to the score for the MLR model to assess whether the performance improvement was worth the extra computational power.

Since neural network models assign multiple coefficients and biases to the input variables, it's difficult to extract and interpret these values. For this reason, partial dependence plots are typically used to understand the relationships predicted by them [19]. Partial dependence plots display the relationship between an output variable and an individual variable with all other independent variables set to be constant [19]. To validate the model, these plots were outputted using the scikit-learn library in Python and compared to literature to ensure they could be explained.

4.3. Results

The average time taken to train the MLP regressor model was 0.57s. The partial dependence plot of hardness on the MW-PECVD parameters is shown in Figure 4.1 and the plot for Young's modulus is shown in Figure 4.2.

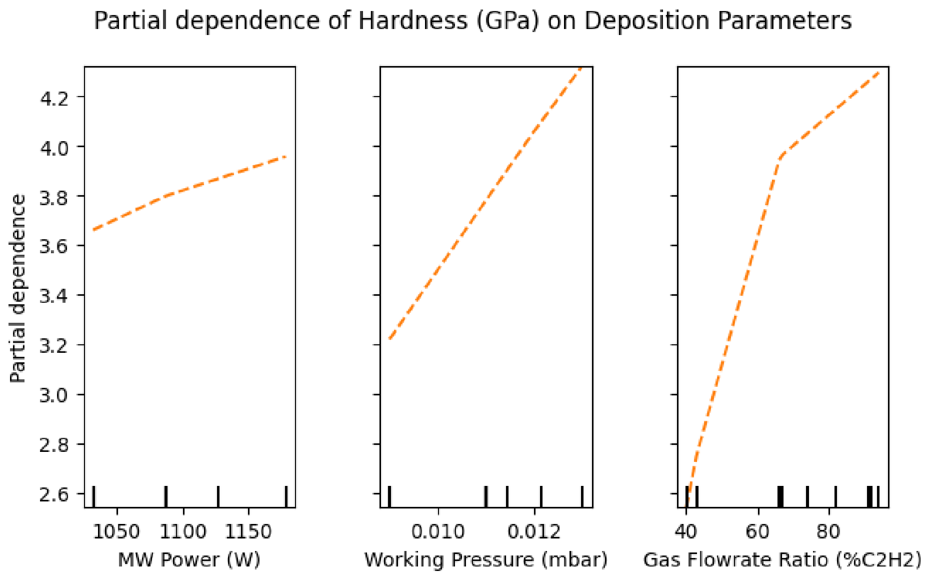

Figure 4.1: Partial dependence of Hardness (GPa) on parameters in MLP model

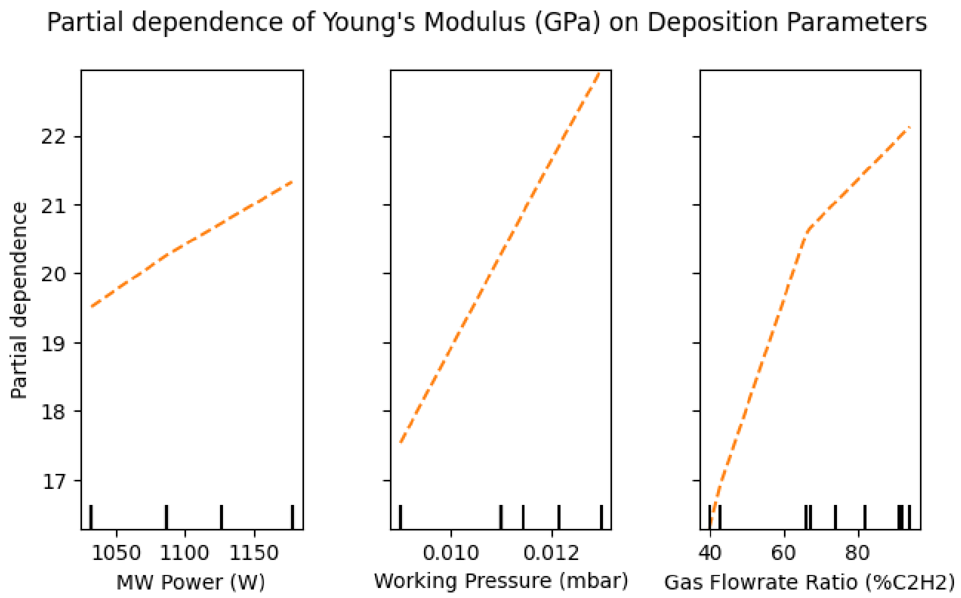

Figure 4.2: Partial dependence of Young's Modulus (GPa) on parameters in MLP model

Table 4.1 shows the results obtained from testing the MLP model using the test data.

Table 4.1: Actual and predicted values from testing of MLP model

Hardness (GPa)		Young's Modulus (GPa)	
Actual	**Predicted**	**Actual**	**Predicted**
2.5	2.5	13.5	16.9
4.1	3.9	24.8	20.6

The MAPE calculated from the values in Table 3.3 is shown in Equation 4.

$$MAPE = \frac{1}{4}\left(\left|\frac{2.5-2.5}{2.5}\right| + \left|\frac{4.1-3.9}{4.1}\right| + \left|\frac{13.5-16.9}{13.5}\right| + \left|\frac{24.8-20.6}{24.8}\right|\right) = 12\% \tag{4}$$

4.4. Discussion

As shown in Figures 4.1 and 4.2, the partial dependence plots for the gas flowrate ratio's and MW power's influence on the DLC coating's hardness and Young's modulus show non-linear relationships which suggests that the MLP model is more suited for modelling the MW-PECVD process than the MLR model.

For gas flowrate ratio, the partial dependence plots show that increasing the acetylene percentage increases the hardness and Young's modulus of the DLC coating, similar to the relationship predicted by the MLR model. However, after ~60%, increasing the acetylene percentage gives diminishing returns. This relationship is supported by the study carried out by Bi et al., as discussed in Chapter 3, which showed that increasing the acetylene percentage from 50% to 75% caused an increase in the hardness and Young's modulus of the DLC, due to less argon ions being present to disrupt the DLC coating, while increasing from 75% to 100% caused a decrease since, at 100% acetylene, there's no argon ions present to break the C-H bonds in the acetylene and promote sp3 bonds in the coating [21]. Although the MLP regressor model doesn't show a decrease in the hardness and Young's modulus, this may be due to the fact that in Carley's study, the gas flowrate ratio wasn't set to 100% as this would result in temperatures above the operating temperature of the polymer substrates used so the model didn't have information on the effect having no argon present in the chamber would have on the DLC's mechanical properties. Nonetheless, the findings from Bi et al.'s study support the statement that the MLP regressor model is better suited to model the MW-PECVD process than the MLR model as it has identified the non-linear relationship between the gas flowrate ratio and the DLC mechanical properties.

Similar to the relationship predicted by the MLR model, the partial dependence plots for MW power state that increasing it increases the DLC hardness and Young's modulus. However, the MLP model differs in that the relationship isn't linear and that increasing the power past ~1100W gives diminishing returns on the hardness and Young's modulus. This relationship is supported by the study carried out by Han et al., discussed in Chapter 3, which stated that increasing the MW power in a PECVD process from ~750W to ~1000W caused an increase in the hardness and Young's modulus due to there being a greater plasma density generated to break bonds in the acetylene, and then increasing it past ~1000W decreased these properties due to the increased plasma disrupting the DLC coating [23].

On the other hand, the partial dependence plots for working pressure show a linear relationship which goes against the study carried out by Wang et al. which found that increasing the pressure past a certain point in PECVD of DLC caused a decrease in the hardness and Young's modulus due to the gas molecules disrupting the sp3 bonds in the coating. However, the explanation for this could be that, in Carley's study, the pressure was kept low to keep the substrate temperature below 70°C to prevent any heat damage to the polymer substrate and so the pressure wasn't increased past the point at which it would cause disruption to the coating.

The MLP regressor model took an average of 0.57s to train while the MLR model took only 0.0030s which means the MLP regressor model which is a very significant 102x increase and demonstrates that the MLP model is significantly more computationally expensive to train. Despite this, the MLP regressor model scored a MAPE of 12% (88% accuracy) after testing whereas the MLR model had a MAPE of 18% (82% accuracy) which shows that the MLP regressor model is more accurate than the MLR model. From this accuracy increase and validation of the non-linear relationships displayed in the partial dependence plots, the MLP regressor model is better suited to modelling the MW-PECVD process than the MLR model.

Although the MLP model is more accurate than the MLR model, it's still classed as being 'good' according to Lewis and not 'highly accurate' which is accuracy is above 90%. The model's accuracy could be improved with a greater sample size to train on and, according to the study by Zhang et al., a sample size of ~150 should be used to achieve a 10% MAPE. Increasing the training dataset size to 150 would also cause the sharp turns apparent in the partial dependence plots in Figures 4.1 and 4.2 to be smoother curves and be more realistic representations of the MW-PECVD process.

Chapter 5: Developing MW-PECVD Optimisation Tool

5.1. Introduction

This chapter aims to develop an optimisation which uses the MLP regressor model developed in Chapter 5 to guide industries on the optimum MW-PECVD parameters to use to obtain their desired DLC mechanical properties to optimise the process for their polymer substrates. After comparing the traditional ML MLR model and the DL MLP regressor model, the MLP model, despite being more computationally expensive, was found to be more accurate than the MLR model when predicting the mechanical properties of DLC coatings deposited by MW-PECVD. For this reason, the MLP regressor model was selected to be used to develop this optimisation tool.

The optimum parameters recommended by the tool should remain within the boundaries stated by Carley in their study to ensure the plasma can be maintained within the deposition chamber and that the temperature is kept within safe operating temperatures of polymers commonly used in industry. Additionally, these optimum parameters should ensure energy and material usage is minimised to ensure the MW-PECVD process is economic and sustainable for industries. Keeping the MW power low reduces the energy usage of the process which improves the sustainability of the process and the energy costs involved [28]. Moreover, the lower MW power reduces the temperatures involved in the process which reduces the risk of damage to polymer substrates with low melting temperatures [3, 28]. The working pressure should also be kept as low as possible to reduce the amount of acetylene and argon gas used for the process and therefore, reducing the cost spent on them [3]. Additionally, the percentage of acetylene used should also be kept low as it's a flammable gas and a study on improving sustainability of CVD states to reduce the amount of hazardous precursor used in processes [28, 29]. This tool should also inform the user of the limitations of the model used so that industries are aware of possible inaccuracies.

5.2. Methodology

As discussed in Chapter 5, training the MLP regressor model was significantly more computationally expensive than the MLR model which could become an issue for industries repeatedly using the tool. For this reason, it was decided to have one program which would train the MLP model and output a lookup table which contained all the possible MW-PECVD parameter combinations within the boundaries of Carley's study and their corresponding hardness and Young's modulus values as predicted by the MLP model, called Program A, and another program which would search through this lookup table to find the optimum parameters to recommend to users for their desired properties, Program B. As this method meant the MLP model didn't need to be trained each time the tool was used, the issue of training it being computationally expensive was minimised which makes this tool more efficient.

Program A was written to train the MLP model from Chapter 4 and output a comma-separated values (csv) file containing all the possible unscaled parameter combinations, with their predicted hardness and Young's modulus values, in order of ascending MW power values then ascending pressure values and then ascending flowrate ratio values (see Appendices E-H). The unscaled values were used as users would be able to understand these rather than the scaled values used to train the model. The minimum and maximum values for the parameters in the lookup table were set as the boundaries used in Carley's study because parameters outside of this would not be able to maintain a plasma or would cause the temperature to be too high for polymer substrates. The flowchart in Figure 5.1 was used to guide development of Program A.

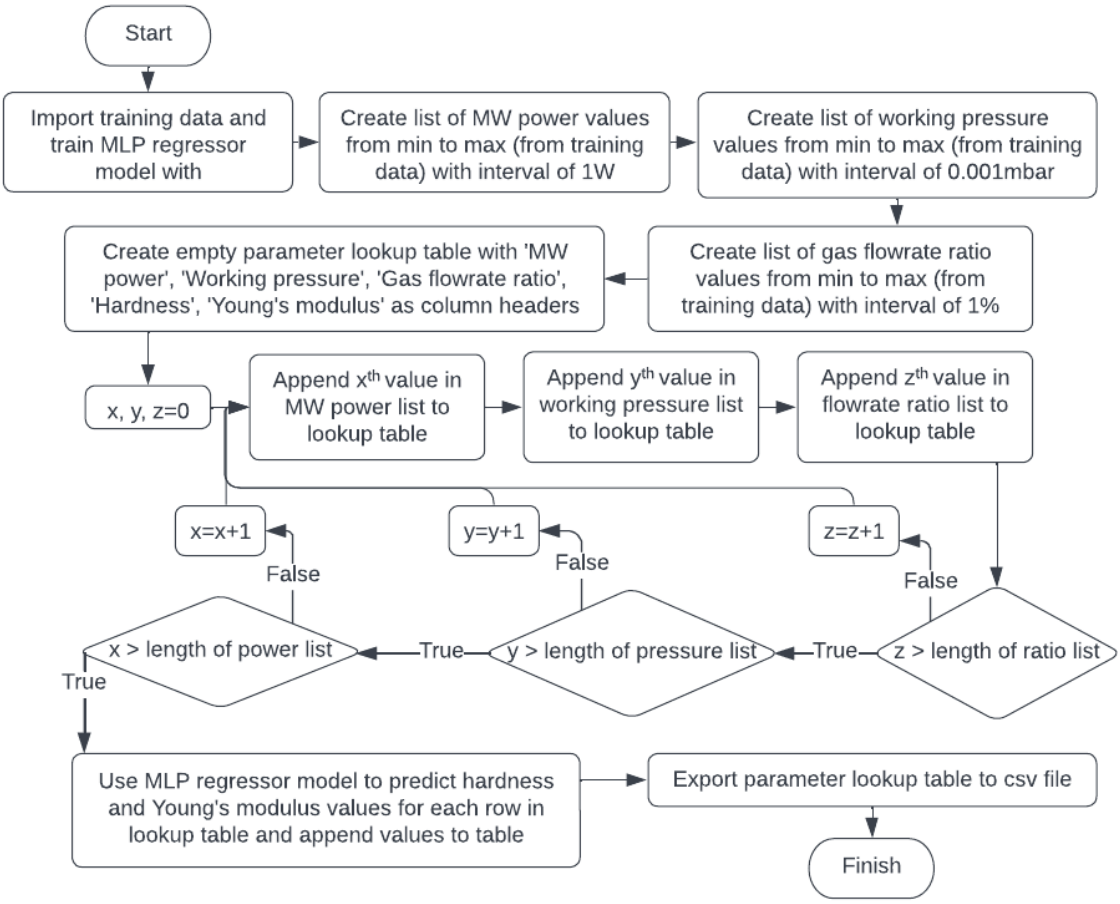

Figure 5.1: Program A flowchart

Program B was written to take a user input on desired hardness and Young's modulus and then iteratively search through each row in the lookup table until the hardness and Young's modulus values in the row were equal to or greater than the desired values, and then output the parameters from this row to the user (see Appendices I and J). The program was written to accept property values greater than the ones desired by the user in the scenario that the specific values they desire aren't possible to deposit (e.g., low hardness of 2.9GPa and high Young's modulus of 24.1GPa), in which case the program would search for the parameters which can deposit the higher property. It was deemed acceptable for the program to find values higher than the ones desired by the user in the scenario their desired selection wasn't possible as industries typically require a minimum hardness or Young's modulus to ensure their products can withstand the conditions they're intended for [3].

By beginning to iterate from the row of the lookup table, Program B recommends the lowest suitable MW power which can deposit DLC coatings with the properties required to minimise energy usage, the lowest pressure to minimise resources spent, and lowest flow rate ratio, to minimise risk from acetylene, which makes the tool more useful for industries by ensuring the process is economic and sustainable. Finally, it was important for the program to be written to output the limitations of the model used to create the lookup table so that the user is aware of potential inaccuracies in the recommended parameters. To guide development of the Program B, a flowchart was constructed, as shown in Figure 5.2.

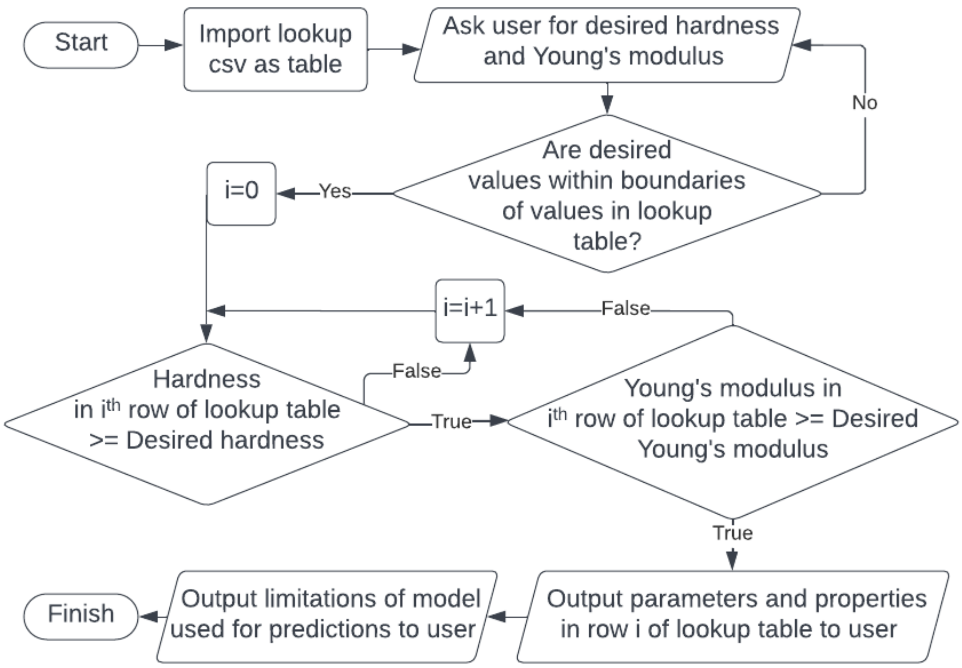

Figure 5.2: Program B flowchart

5.3. Results

Figure 5.3 displays a sample of the lookup table csv file outputted by Program A which contains all the possible MW-PECVD parameter combinations and their corresponding predicted mechanical properties.

MW Power (W)	Working Pressure (mbar)	Gas Flowrate Ratio (%C2H2)	Hardness (GPa)	Young's Modulus (GPa)
1033	0.009	89	4	21.6
1033	0.009	90	4	21.7
1033	0.009	91	4	21.8
1033	0.009	92	4.1	21.9
1033	0.009	93	4.1	22.1
1033	0.009	94	4.2	22.2
1033	0.01	40	2.2	15
1033	0.01	41	2.2	15.1
[illegible]	[illegible]	[illegible]	[illegible]	[illegible]

Figure 5.3: Sample from lookup table csv file generated by Program A

Figure 5.4 displays the ouput from Program B which displays the optimum parameter combination to the user for their desired mechanical properties along with the limitations of the data used to train the MLP regressor model.

```
Enter desired Hardness of DLC coating in GPa between 2.1GPa and 5.1GPa:10
Hardness value entered is above range of this tool
Enter desired Hardness of DLC coating in GPa between 2.1GPa and 5.1GPa:4
Enter desired Young's Modulus of DLC coating in GPa between 15.0GPa and 25.3GPa:20.5

Most economic settings to achieve a Hardness of 4.0GPa and a Young's Modulus of 21.5GPa
Power (W): 1032.0
Working Pressure (mbar): 0.009
Gas Flowrate Ratio (%C2H2): 89.0

These predictions are made based on data obtained using a Hauzer Flexicoat 850 Coating
system with no bias applied across the substrates used and no measurements taken of the
self-bias applied
```

Figure 5.4: Output of optimum MW-PECVD parameters and model limitations

5.4. Discussion

As shown from Figure 5.3, Program A was successful in outputting a lookup table which contained all the possible parameter combinations and their corresponding mechanical properties as predicted by the trained MLP regressor model. From Figure 5.3, it’s also evident that the intervals selected for the parameter values in Program A were appropriate as the mechanical property values in the outputted tables generally increase in increments of 0.1GPa which is the typical resolution of hardness and Young’s modulus values of DLC coatings in industry [3, 30]. Setting the intervals to be smaller would’ve caused the lookup table csv file to be larger than necessary which would’ve made it use unnecessary storage resources and increased the time take for Program B to search through it to find the optimum parameters.

However, the highest hardness and Young's modulus in the lookup table were 5GPa and 24.5GPa, respectively, which are far below the typical properties of DLC coatings used in industries. Diamolith DLC coatings produced for medical instruments and piston pins in automotive applications have a minimum hardness of 10GPa and Young's modulus of 170GPa [31]. Similarly, CERTESS DLC coatings for automotive purposes have a minimum hardness of 2000HV (19.61GPa) [30]. Since the properties in the lookup table don't reach these values, the scope of this tool is limited which reduces its use for polymer substrates in industry. These relatively low mechanical properties in the lookup table could be attributed to the fact that Carley's study, from which the training data was obtained, was carried out on a coating machine without an ability to monitor the self-bias voltage across the plasma [3]. As demonstrated by the study by Gou et al., self-bias plays a key role in the structure of DLC coatings, and its properties and so higher mechanical properties may have been possible if the self-bias could be tuned. Furthermore, the substrate temperature was kept below 70°C but a higher value could increase the mechanical property values, but this would also increase risk of heat damage to the polymers [3, 4].

From Figure 5.4, it's also shown that Program B finds the most economic settings to obtain the user selected mechanical properties. Program B does this by iteratively going through each row of the lookup table, beginning from the first, to find the properties which fulfil the ones desired by the user and then outputs the corresponding parameter values. By searching from the first row, Program B ensures that the lowest combination of MW power, working pressure and acetylene percentage is recommended to the user. This is beneficial to the user as lower power reduces energy usage, lower working pressure means less acetylene and argon gas is required in the deposition chamber hence less resources used and a lower acetylene percentage reduces the risks involved in the deposition process.

Another limitation of Program B is that it doesn't provide actual acetylene and argon gas flowrate values and instead gives the optimum ratio. This is an issue as coating machines require the flowrates to be entered as sccm values and so industries would need to experiment with the acetylene and argon flowrates until they found a combination which matched the pressure and ratio recommended by Program B which is inefficient [3]. To calculate the gas flowrate values from the percentage and working pressure, the temperature of the deposition chamber would be required but this would vary greatly depending on factors such as the MW power used and so it would be difficult to calculate and so, for this reason, the flowrate was kept as a ratio.

Chapter 6: Conclusion

6.1. Achievements

The main achievements of this project were as follows:

- Training a traditional ML MLR model to be >80% accurate in identifying the relationship between MW-PECVD parameters and the hardness and Young's modulus of DLC coatings on polymer substrates
- Training a DL MLP regressor neural network model to be >80% accurate in identifying the relationship between MW-PECVD parameters and the hardness and Young's modulus of DLC coatings on polymer substrates
- Using the trained DL model to develop an optimisation tool for industries to obtain the optimum MW-PECVD parameters needed to achieve their desired hardness and Young's modulus for the DLC coatings of their polymers

6.2. Discussion

This motivation behind this project was to make tuning the deposition parameters of the MW-PECVD of DLC for polymer substrates more accessible to high-level employees in manufacturing industries, reducing the need for surface engineering experts.

As shown in Figure 5.4, the optimisation tool developed in this project can find MW-PECVD parameters needed to obtain a user's desired mechanical properties for their DLC coating of a polymer substrate while ensuring the recommended parameters are as economic and sustainable as possible. Furthermore, the recommended parameters stay within a range which reduces the likelihood of the substrate temperature during the deposition process to reach levels which could cause heat damage to polymer substrates while also ensuring the deposition process can be carried out successfully.

However, the tool can only give parameter combinations for hardness and Young's modulus values of up to 5GPa and 24.5GPa, respectively, which limits the usefulness of the tool as DLC coatings typically used in industries have minimum hardness and Young's modulus values significantly greater than this. Nonetheless, these values still improve the mechanical properties of polymer substrates and allow them to be used in areas which they wouldn't have been suitable for before.

The optimisation tool recommends parameters based on the MLP regressor neural network trained in this project which was found to be 88% accurate which was above the accuracy this project aimed to achieve. A traditional ML MLR model was also trained and found to be 82% accurate and was significantly less computationally expensive to train due to its relatively simple nature compared to the MLP model. However, although the MLR model would make the optimisation tool more efficient due to its shorter training times, the MLP model was used due to its robustness in identifying non-linear relationships which is evident through its greater accuracy and when validated against literature.

Furthermore, using the MLP regressor model makes the optimisation tool more robust as it can be trained to include more MW-PECVD parameter variables, such as self-bias and substrate temperature, which could also have non-linear relationships with the mechanical properties of DLC coatings and this would reduce the limitations of the current model. The current MLP regressor model was trained on data where the substrate temperature wasn't varied, and the self-bias wasn't measured. Since both these variables have been shown to influence the mechanical properties of DLC coatings deposited through MW-PECVD, inaccuracies in the predictions of the model can stem from this model not considering these variables.

Additionally, the sample size used to train this model, 19, was found to be far below the recommended sample size for modelling surface engineering processes which was found to be 150 based on other studies taken [11]. However, as the optimisation tool is used more and more in industry, the model's accuracy will improve as it can learn from the MW-PECVD processes run using it which is a key benefit of using machine learning.

Although this optimisation tool isn't suitable for metal substrates due to it not having bias voltage as a variable and being trained on data with a substrate temperature far below the operating temperature of metal substrates commonly used for DLC coatings, the basic methodology detailed for training the ML models and developing the optimisation programs in this project can be applied to MW-PECVD of DLC for metal substrates or for other thin film coatings. Furthermore, the model can be trained to be optimised for other properties such as surface roughness which is an important property for products intended to be used as lubricants [3].

6.3. Conclusion

This project has succeeded in developing an optimisation tool which makes the MW-PECVD of DLC coatings for polymers more accessible by recommending the optimum parameters for their required hardness and Young's modulus. Although the

MLP regressor model trained in this project and used for the optimisation tool had limitations relating to not including all the MW-PECVD variables which would affect the hardness and Young's modulus of DLC coatings deposited on to polymers, the model was still shown to have an accuracy of 88% which is deemed 'good' by typical standards. The project has demonstrated that DL is more suitable for the MW-PECVD of DLC for polymer substrates than traditional ML due to its higher accuracy and ability to recognise more complex relationships than those recognisable by traditional ML which suggests the methodology behind this report could also be followed for the MW-PECVD of other thin film coatings for other substrates.

6.4. Future Work

For future work, the effect of the number of neurons and hidden layers in the MLP regressor model used to model the MW-PECVD process could be measured to ensure the optimum configuration is used.

References

1. Olayinka Oluwatosin, A., et al., *Overview of thin film deposition techniques.* AIMS Materials Science, 2019. **6**(2): p. 174-199.

2. Research and Markets, *Thin Film Coating Market Report: Trends, Forecast and Competitive Analysis*. 2019.

3. Carley, S.M., *Diamond-like Carbon Coatings for Improved Mechanical Performance of Polymer Materials*, in *School of Mechanical Engineering*. 2017, University of Leeds.

4. Austin, L.B., *Evaluation and optimisation of diamond-like carbon for tribological applications*, in *School of Mechanical Engineering*. 2014, University of Leeds.

5. Batey, J. and E. Tierney, *Low‐temperature deposition of high‐quality silicon dioxide by plasma‐enhanced chemical vapor deposition.* Journal of Applied Physics, 1986. **60**(9): p. 3136-3145.

6. Polymer Shapes. *ABS*. 2022 [cited 2022 10th January]; Available from: https://www.polymershapes.com/product/abs/.

7. Sarker, I.H., *Machine Learning: Algorithms, Real-World Applications and Research Directions.* SN Computer Science, 2021. **2**(3): p. 160.

8. Pedregosa, F., et al., *Scikit-learn: Machine Learning in Python.* J. Mach. Learn. Res., 2011. **12**: p. 2825–2830.

9. Qi, X., et al., *Applying Neural-Network-Based Machine Learning to Additive Manufacturing: Current Applications, Challenges, and Future Perspectives.* Engineering, 2019. **5**(4): p. 721-729.

10. Lewis, C.D., *Industrial and business forecasting methods : a practical guide to exponential smoothing and curve fitting*. 1982, Butterworth Scientific: London.

11. Zhang, Y. and C. Ling, *A strategy to apply machine learning to small datasets in materials science.* npj Computational Materials, 2018. **4**(1): p. 25.

12. Eranpurwala, A., S.E. Ghiasian, and K. Lewis, *Predicting Build Orientation of Additively Manufactured Parts With Mechanical Machining Features Using Deep Learning*. 2020.

13. Yang, W.J., et al., *Microstructure and tribological properties of SiOx/DLC films grown by PECVD.* Surface and Coatings Technology, 2005. **194**(1): p. 128-135.

14. Gou, W., et al., *Effect of negative self-bias voltage on microstructure and properties of DLC films deposited by RF glow discharge.* Surface and Coatings Technology, 2007. **201**(9): p. 5043-5045.

15. Takadoum, J., et al., *Comparative study of mechanical and tribological properties of CNx and DLC films deposited by PECVD technique.* Surface and Coatings Technology, 2003. **174-175**: p. 427-433.

16. Pharr, G.M. and W.C. Oliver, *Measurement of Thin Film Mechanical Properties Using Nanoindentation.* MRS Bulletin, 1992. **17**(7): p. 28-33.

17. Kumar, S., et al., *Versatile microwave PECVD technique for deposition of DLC and other ordered carbon nanostructures.* Vacuum, 2001. **63**(3): p. 433-439.

18. Hutcheson, G.D., *Ordinary Least-Squares Regression*, in *The Multivariate Social Scientist*. 1999, SAGE Publications, Ltd. p. 56-113.

19. Wright, R.E. *Interpreting Black-Box Machine Learning Models Using Partial Dependence and Individual Conditional Expectation Plots*. 2018.

20. Raschka, S., *Python Machine Learning*. 2015: Packt Publishing Ltd.

21. Bi, J., J. Peng, and L. Li, *Effect of acetylene content in working gas on microstructure and thermal stability of DLC coating.* E3S Web of Conferences, 2020. **206**.

22. Wang, X., et al., *Effect of deposition pressures on uniformity, mechanical and tribological properties of thick DLC coatings inside of a long pipe prepared by PECVD method.* Surface and Coatings Technology, 2019. **375**: p. 150-157.

23. Han, M.-K., et al., *Hydrophobic and Mechanical Characteristics of Hydrogenated Amorphous Carbon Films Synthesized by Linear Ar/CH 4 Microwave Plasma.* Applied Science and Convergence Technology, 2017. **26**: p. 34-41.

24. Tutunov, R., et al., *Compositional ADAM: An Adaptive Compositional Solver.* ArXiv, 2020.

25. Heaton, J., *Introduction to Neural Networks for Java, 2nd Edition*. 2008: Heaton Research, Inc.

26. Dietterich, T., *Overfitting and undercomputing in machine learning.* ACM computing surveys (CSUR), 1995. **27**(3): p. 326-327.

27. Pomerat, J., A. Segev, and R. Datta. *On Neural Network Activation Functions and Optimizers in Relation to Polynomial Regression*. in *2019 IEEE International Conference on Big Data (Big Data)*. 2019.

28. Pedersen, H., S.T. Barry, and J. Sundqvist, *Green CVD—Toward a sustainable philosophy for thin film deposition by chemical vapor deposition.* Journal of Vacuum Science & Technology A, 2021. **39**(5).

29. Miller, S.A. and E. Penny. *Hazards in Handling Acetylene in Chemical Processes Particularly under Pressure*. in *Proceedings of the Symposium on Chemical Process Hazards with special Reference to Plant Design*. 1959.

30. HEF Group. *CERTESS® DLC (Diamond-like Carbon) Coating*. 2022 [cited 2022 14th February]; Available from: https://www.hefusa.net/pvd_coating/certess-DLC-tribological-PECVD-coating.html.

31. Wallwork. *Diamolith DLC*. 2022 [cited 2022 14th February]; Available from: https://www.wallworkht.co.uk/content/diamolith_dlc/.

Appendix

Appendix A: Hardness and Young's modulus data obtained by Carley's study

	ABS		PEI	
Leg	**Hardness (GPa)**	**Young's Modulus (GPa)**	**Hardness (GPa)**	**Young's Modulus (GPa)**
1	4.9	21.9	4.9	24.6
2	4.1	19.8	4.1	22.4
3	3.4	19.2	4.1	24.8
4	2.1	14.0	2.2	15.3
5	3.9	20.2	3.6	22.1
6	2.5	13.5	2.7	16.9
7	4.2	18.5	4.8	25.0
8	2.2	19.5	3.0	18.3
9	3.3	16.9	4.5	24.2
10	4.7	23.1	4.3	24.7

Appendix B: MW-PECVD parameters used for Carley's study

Leg	Microwave Power (W)	Working Pressure (mbar)	C2H2 Flowrate (sccm)	Ar Flowrate (sccm)	Gas Flowrate Ratio (%C2H2)
1	1032	0.012	722	50	94
2	1087	0.011	720	60	92
3	1032	0.011	610	110	85
4	1087	0.009	200	300	40
5	1178	0.009	490	175	74
6	1178	0.011	250	350	42
7	1178	0.011	722	70	91
8	1032	0.013	300	400	43
9	1032	0.011	450	235	66
10	1087	0.013	520	260	67

Appendix C: Training of MLR model

```
#initialise timer
t1 = time.perf_counter()

#bring in linear regression model from scikit-learn library
model = Pipeline(steps=[('sca', MinMaxScaler()),
                        ('reg', linear_model.LinearRegression())])

#train model with training data
model.fit(x,y)

#output time taken for model to train
t = (time.perf_counter() - t1)
print(t)
```

Appendix D: Testing of MLR model

```
#output values predicted by model and actual value
pred = model.predict(test_x)
print(pred)
print(test_y)

#output coefficients from model
coefficients = pd.concat([pd.DataFrame(features),
                          pd.DataFrame(np.transpose(model.named_steps['reg'].coef_))],
                         axis = 1)
print(coefficients)
```

Appendix E: Optimisation Tool Program A – Training of MLR model

```
#bring in MLP regressor model from scikit-learn library
nn=Pipeline(steps=[('sca', MinMaxScaler()),
                   ('mlp', MLPRegressor(random_state=70,
                                        max_iter=34000,
                                        solver = 'adam',
                                        hidden_layer_sizes=(2)))])

#train model on training data
nn.fit(x,y)

#output time taken to train
t = (time.perf_counter())-t1
print(t)
```

Appendix F: Optimisation Tool Program A – Testing of MLR model

```
#output values predicted by model and actual values from testing data
parameters_pred = nn.predict(test_x)
print(parameters_pred)
print(test_y)
```

Appendix G: Optimisation Tool Program A – MLR model partial dependence plots

```
#hardness Partial dependence plots
display1 = PartialDependenceDisplay.from_estimator(
    nn,
    x,
    features,
    target = 0,
    grid_resolution=20,
    ice_lines_kw={"color": "tab:blue", "alpha": 0.2, "linewidth": 0.5},
    pd_line_kw={"color": "tab:orange", "linestyle": "--"},
)

display1.figure_.subplots_adjust(wspace=0.4, hspace=0.3)
display1.figure_.suptitle("Partial dependence of Hardness (GPa) on Deposition Parameters")

#ym Partial dependence plots

display2 = PartialDependenceDisplay.from_estimator(
    nn,
    x,
    features,
    target = 1,
    grid_resolution=20,
    ice_lines_kw={"color": "tab:blue", "alpha": 0.2, "linewidth": 0.5},
    pd_line_kw={"color": "tab:orange", "linestyle": "--"},
)

display2.figure_.subplots_adjust(wspace=0.4, hspace=0.3)
display2.figure_.suptitle("Partial dependence of Young's Modulus (GPa) on Deposition Parameters")

plt.show()
```

Appendix H: Optimisation Tool Program A – Writing look-up table csv file

```
#create lists of parameter values with appropriate interval for each
power_values = np.arange(min(power),
                         (max(power)+1),1).tolist()
pres_values = np.arange(min(working_pres),
                        (max(working_pres)+0.001), 0.001).tolist()
ratio_values = np.arange(min(flow_ratio),
                         (max(flow_ratio)+1), 1).tolist()

parameters_dict = {'MW Power (W)':[],
                   'Working Pressure (mbar)':[],
                   'Gas Flowrate Ratio (%C2H2)':[]}

#create dataframe of all possible combinations from lists
for power in power_values:
    for pres in pres_values:
        for ratio in ratio_values:
            parameters_dict['MW Power (W)'].append(power)
            parameters_dict['Working Pressure (mbar)'].append(pres)
            parameters_dict['Gas Flowrate Ratio (%C2H2)'].append(ratio)

parameters_df = pd.DataFrame(parameters_dict)

#add predicted property values to dataframe
predict_array = nn.predict(parameters_df)
predict_df = pd.DataFrame(predict_array, columns = targets)

#round up property values to 1 d.p.
predict_df = predict_df.round(1)

#output csv file
model_df = parameters_df.join(predict_df)
model_df.to_csv('model.csv', index=False, header=True)
```

Appendix I: Optimisation Tool Program B – Taking User Input

```
#get lookup table from csv file
model_df = pd.read_csv('model.csv')

#find boundaries stated in lookup table
hardness_min = model_df['Hardness (GPa)'].min()
hardness_max = model_df['Hardness (GPa)'].max()
ym_min = model_df['Young\'s Modulus (GPa)'].min()
ym_max = model_df['Young\'s Modulus (GPa)'].max()

hardness = 0
ym = 0

#ensure users desired properties are withing bounds from lookup table
while hardness_valid == False:
    hardness_target = float(input('Enter desired Hardness of DLC coating in GPa between\
                            '+str(hardness_min)+'GPa and '+str(hardness_max)+'GPa:'))
    if hardness_target<hardness_min:
        print('Hardness value entered is below range of this tool')
        continue
    if hardness_target>hardness_max:
        print('Hardness value entered is above range of this tool')
        continue
    else:
        hardness_valid = True


while ym_valid == False:
    ym_target = float(input('Enter desired Young\'s Modulus of DLC coating in GPa between\
                        '+str(ym_min)+'GPa and '+str(ym_max)+'GPa:'))
    if ym_target<ym_min:
        print('Young\'s Modulus value entered is below range of this tool')
        continue
    if ym_target>ym_max:
        print('Young\'s Modulus value entered is above range of this tool')
        continue
    else:
        ym_valid = True
```

Appendix J: Optimisation Tool Program B – Output optimum parameters and limitations of model

```
#iterate through lookup table from top to find optimum parameters
for i, row in model_df.iterrows():
    hardness = row['Hardness (GPa)']
    ym = row['Young\'s Modulus (GPa)']
    if hardness >= hardness_target and ym >= ym_target:
        power = row['MW Power (W)']
        working_pres = round(row['Working Pressure (mbar)'], 3)
        flow_ratio = row['Gas Flowrate Ratio (%C2H2)']
        break

#output optimum parameters and limitations of model
print(f'\nMost economic settings to achieve a Hardness of {hardness}GPa and a Young\'s Modulus of {ym}GPa \n\
Power (W): {power} \n\
Working Pressure (mbar): {working_pres} \n\
Gas Flowrate Ratio (%C2H2): {flow_ratio} \n\
\nThese predictions are made based on data obtained\
using a Hauzer Flexicoat 850 Coating system with no bias\
applied across the substrates used and no measurements taken\
of the self-bias applied')
```