Machine Learning Approach for Process Optimization of Black Nickel Electroplating

Yajuan Sun[1*], Yong Teck Tan[1], Yang Zhao[1], Aaron Zhi Hao Teo[1], Yujie Zhou[1] and Joseph Kee Seng Wong[1]

[1]Singapore Institute of Manufacturing Technology (SIMTech), Agency for Science, Technology and Research (A*STAR), 2 Fusionopolis Way, Singapore 138634, Republic of Singapore.

*Corresponding author(s). E-mail(s): Sun_Yajuan@simtech.a-star.edu.sg;
Contributing authors: tanyongt@simtech.a-star.edu.sg; Zhao_Yang@simtech.a-star.edu.sg; Aaron_Teo@simtech.a-star.edu.sg; yjzhou@simtech.a-star.edu.sg; Joseph_Wong@simtech.a-star.edu.sg;

Abstract

Electroplating enhances the mechanical, thermal, and tribological properties of components in industries like aerospace, computing, pharmaceuticals, and telecommunications. To achieve high-quality electroplated coatings, control of process parameters is essential. This study focuses on the challenges associated with the black nickel electroplating process, particularly its nonlinearity which makes traditional linear methods inadequate. We employed machine learning techniques to develop models capable of predicting defects, coating colour, and coating mass specifically for black nickel electroplating process adhering to the MIL-P-18317 specification, a boric acid-free method that is more environmentally friendly but more sensitive to bath conditions, hence process optimization more challenging. Our research addresses significant gaps in the literature, focusing on boric acid-free black nickel plating, which requires unique modelling approaches different from other electroplating processes. Unlike previous studies that mainly focused on predicting coating mass, our models also predict defects and colour, ensuring comprehensive quality control. The best-performing models achieved an F1 score of 0.9875 for defect prediction, an F1 score of 0.95 for colour prediction, and R^2

= 0.9561, MAE = 0.0039, MSE = 0.00001 for mass prediction. Additionally, we developed a novel optimization algorithm based on these models to fine-tune process parameters, ensuring that the resulting coatings meet strict standards for quality, appearance, and productivity. The validity of our approach was confirmed through experimental results. This research demonstrates how machine learning can help surface finishers, by providing effective strategies for optimizing processes in nonlinear scenarios, thereby improving product quality and productivity.

Keywords: Black nickel plating, Process optimization, Machine learning, Electroplating, Coating

Nomenclature

MSE Mean square error
MAE Mean absolute error
R^2 Coefficient of determination
E_1 Experiment setting 1
E_2 Experiment setting 2
$M_{def_{E1}}$ Machine learning model for defect prediction under E_1
$M_{clr_{E1}}$ Machine learning model for colour prediction under E_1
$M_{cm_{E1}}$ Machine learning model for coating mass prediction under E_1
$M_{def_{E2}}$ Machine learning model for defect prediction under E_2
$M_{clr_{E2}}$ Machine learning model for colour prediction under E_2
$M_{cm_{E2}}$ Machine learning model for coating mass prediction under E_2
X_p the range of initial pH defined as $X_p = [a_b, b_p]$
X_{cd} the range of current density defined as $X_{cd} = [a_{cd}, b_{cd}]$
X_z the range of zinc concentration defined as $X_z = [a_z, b_z]$
X_s the range of surface area defined as $X_s = [a_s, b_s]$
Δ_p the step size of initial pH
Δ_{cd} the step size of current density
Δ_z the step size of zinc concentration
Δ_s the step size of surface area
T_{des} the desired coating mass
η the tolerance parameter for coating mass
f_{def} the function of the best machine learning model for defect prediction
f_{clr} the function of the best machine learning model for colour prediction
f_{cm} the function of the best machine learning model for coating mass prediction
pH pH value of the electroplating solution
CD current density
AAH accumulated ampere-hour
$[Z_n]$ zinc concentration in the electroplating solution

1 Introduction

Electroplating is a technique where an electric current is used to coat an object with a thin layer of metal. This process is crucial for enhancing the mechanical, thermal, and tribological properties of various components. Electroplated coatings are widely employed in industries such as aerospace, computer, pharmaceuticals, and telecommunications to improve hardness, wear resistance, corrosion resistance, surface appearance, and conductivity, among other properties. In industrial applications, electroplated coatings must meet stringent requirements, including defect-free surfaces and precise thickness control. To achieve a high-quality coating, models to understand how process parameters link to coating quality (e.g. no defect) and coating thickness should be developed.

Modelling the black nickel electroplating process studied in this paper is inherently challenging due to its nonlinearity, rendering traditional linear methods inadequate. Recent advancements have employed machine learning techniques [1, 2] for manufacturing process modelling. In particular, for black nickel electroplating process , some machine learning models have been built to predict microhardness [3–5], tribological properties [6], surface roughness [7], coating appearance [8], and coating mass and thickness [9–13]. For example, Shozib et al. [3] explored various applications of machine learning in surface coating. Different machine learning algorithms such as artificial neural network (ANN), gradient boosted regression tree (GBRT), and extra trees (ET) were used to predict the microhardness of electroless Ni-P-TiO_2 composite coating. H. Beygi et al. [13] used ANN to design a high efficiency electroless nickel bath to achieve maximum coating mass with minimum received materials. Wu et al. [10] developed a neural network for the simulation and prediction of coating mass and phosphorus content (P%) in the coatings of electroless nickel plating. P. J. G. Nieto et al. [9] applied a hybrid gradient boosted regression tree (GBRT) methodology for hard chromium layer thickness prediction and hyperparameter of GBRT was optimized by differential evolution algorithms.

However, the existing literature has several limitations: 1. None of the processes mentioned pertain to black nickel coatings, which are used to reduce the surface reflectivity of parts for optical devices and as solar absorption coatings for solar panels. Since the processes differ, the models will also be significantly different. 2. Even within different electroplating processes, most models focus on tribological properties, surface roughness, microhardness, coating mass/thickness prediction or general appearance prediction (good surface or not). No models have been developed for detailed defect and colour prediction, which is critical for understanding coating quality in industrial applications. 3. The mentioned literature does not consider process optimization. Process optimization, i.e., process tuning and control, is crucial to adapt to changing bath conditions and ensure consistent results.

To address the aforementioned gaps, this study focuses on the black nickel electroplating process, specifically adhering to the MIL-P-18317 specification [14] which outlines a boric acid-free electroplating process. We developed three

machine learning models to predict defect appearance, coating colour, and coating mass, with the best models achieving an F1 score of 0.9875 for coating defect prediction, an F1 score of 0.95 for coating colour prediction, and $R^2 = 0.9561$, MAE = 0.0039, MSE = 0.00001 for coating mass prediction. Additionally, we devised an optimization algorithm based on these machine learning models to select process parameters, ensuring that the resulting coating meets all three requirements: quality (i.e. no coating defect), appearance (i.e. sufficiently black), and productivity (i.e. desired coating mass achieved in a single run without having to re-work) simultaneously. The effectiveness of the models has been validated through experiments.

This paper presents several novel contributions compared to existing literature: 1. We focused on a black nickel plating process to achieve black nickel coating, utilizing a boric acid-free process, which is more environmentally friendly. However, due to the lack of boric acid acting as a pH buffer, this process is highly sensitive to bath and process conditions, making process modelling and optimization more challenging. 2. For process modelling, we not only developed a coating mass model but also models for predicting defects and colour, which is a significant advancement over existing literature. 3. Based on the developed models, we created an optimization algorithm that ensures a flawless coating with the desired colour and mass. This is crucial for real industrial applications, where coatings must meet stringent quality (no defects), appearance (black colour), and productivity requirements.

The paper is organized as follows. Section 1 is the introduction while Section 2 presents the experimental methods such as the machine learning approaches used, electroplating experiments and coating characterization. Section 3 gives the coating experimental results and the outcomes of the machine learning models for predictions of coating defects, coating colour and coating mass, plus an overall process optimization algorithm. Section 4 provides a discussion on the new insights obtained from machine learning models, the strengths of the machine learning models and their limitations. The conclusions of this work are given in Section 5.

2 Experimental Methods

2.1 Machine Learning Approaches

Scikit learn library supports many machine learning (ML) models. Here we introduce 5 different models which will be used in our work: multi-layer perceptron (MLP), random forest (RF), AdaBoost, support vector machine (SVM), and logistic regression. Some of these models cover both classification algorithm and regression algorithm. The main difference between classification algorithm and regresssion algorithm is that classification algorithm is used to predict different classes, e.g. class 0 (no defect) or class 1 (presence of defect); while regression algorithm is used to predict a continuous value such as the value of coating mass in our work.

2.1.1 Multi-Layer Perceptron (MLP)

Multi-Layer Perceptron (MLP) is a supervised learning algorithm which learns a function through a training data set. Let $X = x_1, x_2, ..., x_n$ be the input and $y{=}y_1, y_2, ..., y_n$ be the output. Usually, an MLP includes several layers, such as an input layer, multiple hidden layers and an output layer. Given a set of features and a target, MLP can learn a nonlinear function approximator by using transfer function $F(X)$ as below. And the output of y is given by Eq. 1

$$y = F(x) = F(\sum_{i=1}^{n}(W_{ij}x_i + b_j)), \tag{1}$$

where W_{ij} represents the weight and b_j represents the bias with respect to the inputs x_i. MLP classifier implements an MLP algorithm that trains using back-propagation. That is, it trains using gradient descent which are calculated using backpropagation. For classification problem, MLP classifier in Scikit learn minimizes the cross-entropy loss function. For regression problem, Multi-Layer Perceptron Regressor (MLPR) minimizes square error loss function.

2.1.2 Random Forest (RF) and AdaBoost

Decision tree is a supervised learning method by creating a prediction model with tree-like structure. Usually, a decision tree includes branches representing decision-making steps which can lead to a desired result. A typical example of decision tree is shown in Fig. 1.

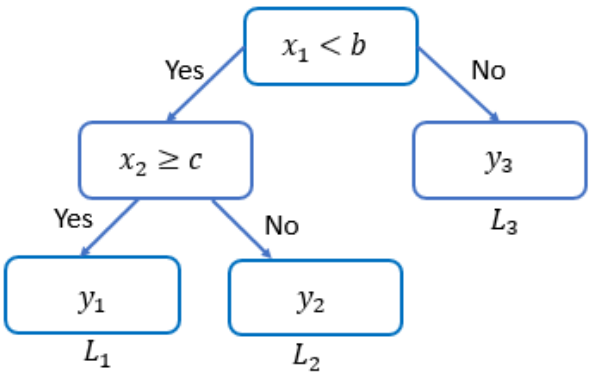

Fig. 1 An example of decision tree (DT) model.

The decision tree in Fig. 1 includes 2 input variables x_1 and x_2. Based on the condition/internal node, e.g. $x_1 < b$, the tree splits into 2 branches/edges. Here L_1, L_2 and L_3 are the leaf nodes with the target value y_1, y_2 and y_3, respectively.

A representative algorithm for training decision trees is the CART (Classification and Regression Trees) algorithm. This algorithm uses metrics such as Gini impurity, information gain, and mean square error as cost functions. The split that results in the lowest cost is selected, which helps in creating the most informative branches in the tree. Random forest is an ensemble learning method that consists of many individual decision trees that operate together. Each tree is trained on a bootstrap sample of the data, and a random subset of features is used for splitting nodes. This randomness helps to create diverse trees whose combined predictions are more accurate and robust. The final prediction of the random forest is determined by averaging the predictions of all trees (in regression) or by majority voting (in classification). Mathematically, this ensemble approach reduces overfitting and variance, as the final model, $f_{rf}(x) = \frac{1}{T}\sum_{t=1}^{T} f_t(x)$, benefits from the collective strength of multiple trees, where T is the number of trees and $f_t(x)$ is the prediction of the t-th tree. AdaBoost, short for Adaptive Boosting, is another powerful ensemble learning technique. Unlike random forest, which builds trees independently, AdaBoost builds a series of weak models sequentially. Each model in the sequence focuses on the errors made by the previous ones. Initially, all data points are given equal weight. After each iteration, the weights of misclassified points are increased, forcing the next model to focus more on these harder cases. The final model is a weighted sum of all weak models, emphasizing those that performed well.

Mathematically, AdaBoost minimizes the exponential loss function, $\sum_{i=1}^{n} \exp(-y_i f(x_i))$, where y_i is the true label and $f(x_i)$ is the predicted output. This approach improves the model's performance by reducing both bias and variance, making it particularly effective for imbalanced datasets. Both random forest and AdaBoost enhance the performance of simple decision trees by aggregating multiple models, but they do so in fundamentally different ways: random forest relies on averaging the predictions of many independently built trees, while AdaBoost combines weak learners sequentially, focusing on correcting errors from previous iterations. This makes them robust and versatile tools for various machine learning tasks.

2.1.3 Support Vector Machine (SVM)

Support vector machine is used for classification problem by constructing hyper-planes in a high dimensional space. Usually, a desired hyper-plane achieves the largest distance to the nearest training data points of any class. This distant is called to be functional margin. A typical example of SVM solves the following equations from a training data set $\{x_i, y_i\}_{i=1}^{n}$ with $x_i \in R^N$ and label $y_i \in \{-1, +1\}$,

$$Min_{\{w,b\}} \frac{1}{2}||w||^2 + C\sum_{i=1}^{n} \xi_i, \tag{2}$$

such that

$$y_i(w^\top x_i + b) \leq 1 - \xi_i, \tag{3}$$

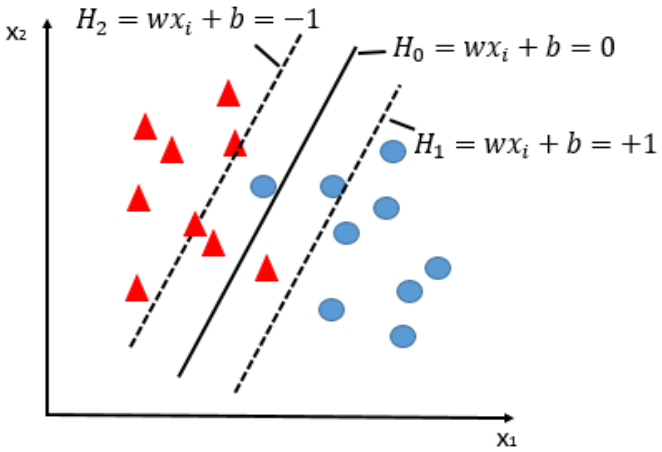

Fig. 2 An example of support vector machine (SVM) model to identify 2 classes where red triangle represents one class and blue circle represents another class.

$$\xi_i \leq 0, \tag{4}$$

where w and b describe a linear discriminate function $y = wx + b$ seen as Fig. 2, and ξ_i are positive slack variables which allows certain misclassification.

2.1.4 Logistic Regression

Logistic regression is an extension of linear regression for classification problems. The formula of the logistic regression is described as below:

$$F(x) = \frac{1}{1 + e^{-(\beta_0 + \sum_{i=1}^{m} \beta_i x_i)}}. \tag{5}$$

where $\beta_0 + \sum_{i=1}^{m} \beta_i x_i$ is similar to a linear model. It employs a sigmoid function as logistic function to restrict the value within the range of $[0, 1]$. Within machine learning, the negative log likelihood used as the loss function, logistic regression supports different optimization methods, e.g. large-scale bound-constrained optimization and stochastic average gradient optimization, to find the global maximum.

2.1.5 Pros and Cons of the ML Models

The general pros and cons of the above mentioned machine learning models are described in Table 1. However, each classifier/regressor has its own strengths and weaknesses, and the best choice depends on the specific characteristics of the dataset and the problem at hand.

Table 1 Comparison of Machine Learning Models

Machine Learning Models	Pros	Cos
AdaBoost (Classifier and Regressor)	Improves accuracy by combining weak learners. Versatile with different base models.	Sensitive to outliers. Risk of overfitting with many weak learners. Requires careful selection of base model.
Random Forest (Classifier and Regressor)	Handles large, high-dimensional datasets. Reduces overfitting.	Provides feature importance. Slow training and prediction. Less interpretable. High memory usage.
Support Vector Machine (SVM) (Classifier)	Effective in high dimensions. Robust to overfitting.	Computationally expensive. Complex to tune. Less effective with noisy data.
Multi-layer Perceptron (MLP) (Classifier and Regressor)	Models complex nonlinear relationships. Flexible architecture. Supports both tasks.	Data-hungry. Slow training. Prone to overfitting. Requires hyperparameter tuning.
Logistic Regression (Classifier)	Simple, efficient, interpretable. Works well with linearly separable classes	Assumes a linear decision boundary. Limited with nonlinear class boundaries.
Linear Regression (Regressor)	Simple, efficient, interpretable. Works well with linear data.	Assumes linear relationships. Sensitive to outliers. Limited with complex data.

2.1.6 Evaluation Criteria for Machine Learning Models

This subsection studies the evaluation of machine learning models. For classification problem, e.g. defect prediction, the performance is measured using the confusion matrix. A confusion matrix has four quadrants, i.e. true positives (TP), true negatives (TN), false positives (FP), and false negatives (FN). It is obtained a comparison between the class predicted by the model and the actual class in the testing set. Based on this these concepts, the following classification performance measures, i.e. accuracy, precision, recall and F1 score are defined as below. Accuracy measures the proportion of correctly predicted instances (both true positives and true negatives) out of the total instances. It is a suitable metric when the class distribution in the dataset is balanced, providing a general sense of how well the model is performing overall.

$$Accuracy = \frac{TN + TP}{TN + TP + FN + FP}, \tag{6}$$

Precision measures the proportion of true positive predictions out of all positive predictions made by the model. It is crucial in scenarios where the cost of false positives is high, such as in spam detection or disease diagnosis, where a false

positive could lead to unnecessary treatments or actions.

$$Precision = \frac{TP}{TP + FP}, \tag{7}$$

Recall, on the other hand, measures the proportion of true positive predictions out of all actual positives in the dataset. This metric is particularly important in situations where missing a positive instance (false negative) is more critical, such as in fraud detection or safety-critical systems, where failing to identify an issue can have severe consequences.

$$Recall = \frac{TP}{TP + FN}, \tag{8}$$

The F1 score, which is the harmonic mean of precision and recall, provides a better measure of the model's performance on imbalanced data by considering both false positives and false negatives

$$F1 = \frac{2(Precision \times Recall)}{Precision + Recall}. \tag{9}$$

In particular, the F1 score is preferred over accuracy, precision, and recall in this paper because the data set is imbalanced. Specifically, we have a large amount of non-defect data but only a limited amount of defect data in our experiments.

When it comes to regression problem, the mean squared error (MSE), mean absolute error (MAE) and coefficient of determination (R^2) are used to determine performance of the regression model and defined as below. Here we assume a data set with a total number of N data points. Here y_i and $\tilde{y}$ represent the true value and prediction value of the i-th data sample. MSE measures the average squared difference between the predicted and actual values. Lower MSE values indicate better model performance, as they imply smaller errors.

$$MSE = \frac{1}{N}\Sigma_{i=1}^{N}(y_i - \tilde{y})^2 \tag{10}$$

MAE measures the average absolute difference between the predicted and actual values. Like MSE, lower MAE values indicate better model performance by showing smaller absolute errors.

$$MAE = \frac{1}{N}\Sigma_{i=1}^{N} \mid y_i - \tilde{y} \mid \tag{11}$$

R^2 measures the proportion of the variance in the dependent variable that is predictable from the independent variables. An R^2 value closer to 1 indicates

a better fit of the model to the data.

$$R^2 = 1 - \frac{\Sigma_{i=1}^{N}(y_i - \tilde{y})^2}{\Sigma_{i=1}^{N}(y_i - \overline{y})^2} \tag{12}$$

Therefore, when both MSE and MAE are smaller and R^2 approaches 1, the model achieves a good prediction accuracy and performs well.

2.2 Electroplating Experimental Procedures

This section describes the experimental procedure of electrochemical process with a boric acid-free black nickel plating formulation given in Military Specification MIL-P-18317 [14].

The schematic diagram of a beaker set-up is shown in Fig. 3. In this work, black nickel coating was plated onto copper panel substrate. The initial pH of the black nickel plating bath was checked using a pH meter (Metrohm 826 pH mobile). The sample was the connected to a DC rectifier (Extech Quad Output DC Power Supply) and immersed "live" (current was turned on before immersing) into the plating bath. A Pt-Ti mesh served as the anode. The black nickel electroplating recipe used in this work in general follows the boric acid-free formulation given in Military Specification MIL-P-18317 and is shown in Table 2.

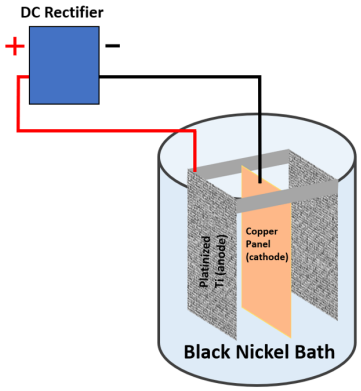

Fig. 3 Schematic diagram of a beaker set-up for electroplating experiment.

The authors used 2 different approaches to generate data for building the machine learning models from beaker electroplating experiments. First approach (experimental setting E_1) was to use the same chemical bath repeatedly while varying the plating bath's pH value, current density (CD), sample surface area (SA), and tracking accumulated ampere-hour (AAH) as the bath aged with repeated use. The second approach (experimental setting E_2) was to use fresh chemical baths with different zinc concentrations ($[Z_n]$), while

Table 2 Formulation of black nickel electroplating solution given in MIL-P-183 17.

Chemical	Amount (g/L)
Ammonium nickel sulfate hexahydrate ($(NH_4)_2SO_4.NiSO_4.6H_2O$)	60
Zinc sulfate heptahydrate ($ZnSO_4.7H_2O$)	7.5
Sodium thiocyanate (NaSCN)	15
pH adjustment (concentrated ammonia solution to increase pH value, sulfuric acid to decrease pH value)	Depends on the experiment

Table 3 Experimental data

Process Parameter (unit)	Range	Output
1. pH value (initial)	2.80 to 6.46	1. Defect label
2. Current density (ASD)	0.06 to 1.37	2. Colour label
3. Surface area (cm^2)	13.8 to 103.7	3. Coating mass (g)
4. Accumulated ampere-hour (Ah)	0 to 1.18	
5. Time (min)	20	
6. $[Z_n]$ (g/L)	0.17 to 1.995	
7. Agitation rate (RPM)	350	
8. Temperature	lab ambient condition	

varying the plating bath's pH value, current density, and sample surface area. There are a total of 60 data sets for E_1 setting (with AAH) and 120 data sets for E_2 setting (with $[Z_n]$). The process parameters range can be found in Table 3.

2.3 Coating Characterization

The coated samples were characterized for coating defect, coating appearance (i.e. coating colour) and deposited coating mass. In particular, we focus on three outputs for coating characterization 1) defect, 2) coating colour, and 3) coating mass as indicated in Table 3. To identify coating defect, the black nickel coating was visually inspected after drying by the operator for any plating defects, such as localized discolourations, skip plating and burns. If no observable defect exists, the defect label of sampling is set to be 0, otherwise, it is set to be 1. To quantify the colour of the coating, a sphere spectrometer X-rite Ci60 was used. In particular, lightness L (with 0 being absolute black, 100 being absolute white) is used for classifying whether the coating colour meets the requirement. Based on results from a pilot study, we empirically set $L = 40$ as the threshold of colour [15]. If $L <= 40$, we label the colour to be 1, i.e. the colour meets the black requirement.

For the coating mass deposited $\triangle m$, it was calculated using the following Eq. 13

$$\triangle m = m_{\mathrm{f}} - m_{\mathrm{i}}, \tag{13}$$

where $\mathrm{m_f}$ represents the final mass and $\mathrm{m_i}$ the initial mass of the sample. The coating mass is an output parameter that can be experimentally measured

easily and can be used to estimate the average coating thickness if the plated substrate surface area and the density of the coating are known.

3 Results

3.1 Experimental Results of Black Nickel Plating

In the experiments, good quality black nickel coatings as well as defective black nickel coatings were produced under different bath and process conditions. Fig. 4 shows examples of good black nickel coating and defective black nickel plating. Good black nickel plating had uniform surface appearance with L value less than or equal to 40. There are cases where even if the coating is uniform in appearance without coating defect, it may be silvery (L >40) instead of black in appearance and thus not meet the criteria for a black nickel coating. Defects observed include non-uniform coatings exhibiting an uneven and powdery appearance. In certain cases, the formation of interference film on the surface gives rise to a "rainbow effect".

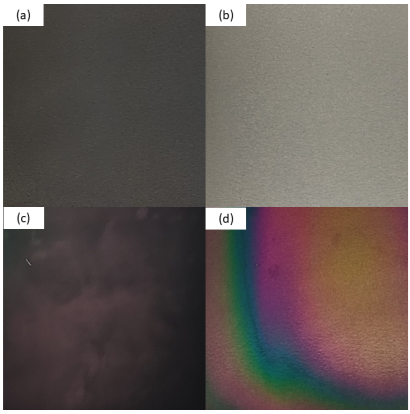

Fig. 4 Images of deposited black nickel plating for (a) uniform black coating (L = 40), (b) uniform silvery coating (L = 58), (c) defective coating with powdery surface, and (d) defective coating with rainbow effect.

The outputs are coating defect label (0 - no observable defect, 1 - observable defect), coating colour label (0 - non-black and 1 - black for colour classification model, with a threshold value of maximum L value of 40 considered as black) and coating mass value.

3.2 Machine Learning Modelling

Before building the machine learning model, we first pre-process the data set and remove the process parameters that were not actively varied such as coating time, temperature and bath agitation rate.

To build the machine learning models for classification problem, i.e. defect prediction and colour prediction, imbalance occurs because one class (e.g. defect class with label 1) in the dataset has significantly fewer instances than the other class (e.g. non-defect class with label 0). This results in a biased model performance, meaning that a machine learning model trained on an imbalanced dataset consistently favours the class with a higher number of instances (majority class) over the class with a smaller number of instances (minority class). Here we employ ADASYN (Adaptive Synthetic Sampling) library to solve this imbalance issue. ADASYN is an oversampling technique that focuses on generating synthetic examples for the minority class, making the distribution more balanced. Unlike simple oversampling methods that duplicate existing instances, ADASYN generates synthetic examples by considering the density distribution of the minority class. After applying the ADASYN method, 80% of data samples were randomly selected as training samples while 20% of the data were selected as testing data. The random selection with the ratio 80-20 was also applied when it comes to the development of machine learning model for regression problem, i.e. the prediction of coating mass.

For each of the machine learning models (coating defect model, coating colour model, and coating mass model), the authors decided to build 2 versions for comparison, with one version of machine learning model built under experimental setting E_1 using {pH, CD, SA, AAH} as input set while the other version of machine learning model is built under E_2 using {pH, CD, SA, $[Z_n]$} as input set. This allows a comparison of whether AAH or zinc concentration $[Z_n]$ is a better input parameter for model building.

To optimize the performance of each machine learning model, we utilize Grid Search as a technique for tuning parameters. This method helps identify the optimal combination of hyperparameter values, leading to the model's highest performance level. Following the grid search process, we save the model along with its associated hyperparameters that yield the highest average 5-fold F1 score. We denote $M_{def_{E_1}}$, $M_{clr_{E_1}}$, $M_{cm_{E_1}}$ as the machine learning models for defect, colour and coating mass predictions under experimental setting E_1, while $M_{def_{E_2}}$, $M_{clr_{E_2}}$, $M_{cm_{E_2}}$ as the machine learning models for defect, colour and coating mass predictions under experimental setting E_2.

3.3 Performance of Machine Learning Models

Table 4 shows the performance of the machine learning models for coating defect prediction with its corresponding inputs which achieved the best 5-fold F1 sore among all the combinations of inputs. Table 5 shows the performance of the machine learning models for coating colour prediction with its corresponding inputs which achieved the best 5-fold F1 sore among all the combinations of inputs. For coating mass prediction which was dealt with a regression approach, we use R^2, MAE and MSE as the performance measurement. Table

Table 4 Machine learning model performance for defect prediction

Best 5-fold F1 score	$M_{def_{E_1}}$ (inputs of $M_{def_{E_1}}$)	$M_{def_{E_2}}$ (inputs of $M_{def_{E_2}}$)
AdaBoost classifier	0.9875 (pH, CD, SA, AAH)	0.8433 (pH, CD, $[Z_n]$)
Support vector classifier	0.9467 (CD, SA, AAH)	0.8238 (pH, CD, $[Z_n]$)
Random forest classifier	0.9733 (pH, CD, SA, AAH)	0.8538 (pH, CD, $[Z_n]$)
Multi-layer perceptron classifier	0.8683 (pH, SA, AAH)	0.7753 (pH, CD, SA, $[Z_n]$)
Logistic regression	0.8675 (pH, CD, SA, AAH)	0.7753 (pH, CD, SA, $[Z_n]$)

Table 5 Machine learning model performance for colour prediction

Best 5-fold F1 score	$M_{clr_{E_1}}$ (inputs of $M_{clr_{E_1}}$)	$M_{clr_{E_2}}$ (inputs of $M_{clr_{E_2}}$)
AdaBoost classifier	0.8956 (CD, SA, AAH)	0.9500 (pH, CD, SA, $[Z_n]$)
Support vector classifier	0.8824 (pH, CD, SA, AAH)	0.8778 (pH, CD, $[Z_n]$)
Random forest classifier	0.9099 (CD, SA, AAH)	0.9429 (pH, CD, SA, $[Z_n]$)
Multi-layer perceptron classifier	0.8516 (pH, CD, SA, AAH)	0.7960 (CD, SA, $[Z_n]$)
Logistic regression	0.7758 (pH, CD, AAH)	0.7963 (CD, SA, $[Z_n]$)

6 and Table 7 show the performance of the machine learning models for coating mass prediction with its corresponding inputs which achieves the best R^2 along all the combination of the inputs.

3.3.1 Coating Defect Model

Five AI models (AdaBoost, SVC, RF, MLP, Logistic Regression) were applied in this study to predict defects. In general, defect prediction achieves a high F1 score using machine learning techniques for both E_1 and E_2. Table 4 shows that under E_1, the AdaBoost classifier achieves the best performance with an F1 score of 0.9875, using input parameters AAH, SA, pH, and CD. Under E_2,

Table 6 Machine learning model performance for coating mass prediction under E_1

M_{cmE_1}	R^2	MAE	MSE
AdaBoost Regressor (pH, CD, SA)	0.9501	0.0045	0.00001
Random Forest Regressor (pH, CD, SA)	0.9561	0.0039	0.00001
Multi-Layer Perceptron Regressor (pH, CD, SA, AAH)	0.8979	0.0060	0.0001
Linear Regression (pH, CD, SA)	0.5398	0.0001	0.007

Table 7 Machine learning model performance for coating mass prediction under E_2

M_{cmE_2}	R^2	MAE	MSE
AdaBoost regressor (CD, SA, $[Z_n]$)	0.9725	0.0113	0.0002
Random forest regressor (CD, SA, $[Z_n]$)	0.9634	0.0113	0.0003
Multi-layer perceptron regressor (pH, CD, SA)	0.9489	0.0113	0.0003
Linear regression (pH, CD, SA)	0.9352	0.0002	0.0092

the RF classifier achieves the best performance with an F1 score of 0.8538, using input parameters $[Z_n]$, pH, and CD.

It is observed that AI models under E_1 outperform the same type of AI models under E_2. For example, the best support vector classifier under E_1 achieves an F1 score of 0.9467, while the best support vector classifier under E_2 achieves an F1 score of 0.8238. Similarly, this trend is seen for RF classifiers, MLP, and logistic regression models.

Therefore, it can be seen that that AAH parameter is more important than the zinc concentration parameter for defect prediction. The AAH parameter encompasses other aspects such as concentration of other solution constituents like nickel and thiocyanate, hence this shows that the other constituents in the bath are also important in determining whether there will be any coating defect formed.

AdaBoost and random forest classifiers demonstrate superior performance compared to MLP and logistic regression in this study due to their ability to handle complex, nonlinear relationships and interactions among features. AdaBoost combines multiple weak classifiers to form a strong classifier, improving performance, particularly in cases with imbalanced data. Random forest,

with its ensemble of decision trees, enhances robustness and reduces overfitting, making it more effective in capturing the underlying patterns in the data. In contrast, MLP and logistic regression may struggle with capturing these complexities and interactions, leading to lower performance in defect prediction tasks.

3.3.2 Coating Colour Model

Regarding the machine learning model for color prediction, all AI models performed with high F1-scores. Table 5 shows that the AdaBoost classifier under E_2 achieved the best performance, with a 5-fold F1 score of 0.9500 using input parameters of pH, CD, SA, and $[Z_n]$. Similar to defect prediction, AdaBoost and random forest demonstrate superior performance over MLP and logistic regression in predicting whether the coating colour will be sufficiently black in both experimental settings E_1 and E_2. This superiority can be attributed to their use of ensemble learning techniques, which aggregate predictions from multiple individual models to form a final prediction. Ensemble methods like AdaBoost and random forest are more robust to noisy data compared to single models such as logistic regression or MLP. Moreover, it can be observed that $[Z_n]$ plays an important role in colour prediction by comparing the results from E_1 and E_2. For example, the F1 score of AdaBoost under E_2, which includes $[Z_n]$ as one of the inputs, is higher than AdaBoost under E_1. For coating colour, the concentration of zinc is more crucial than other bath constituents in determining whether the coating will be sufficiently black. This finding is consistent with earlier reports that the ratio of deposited zinc to nickel atoms in the coating needs to be sufficiently high for a good black nickel coating [15–17]. Additionally, it is noted that if the black nickel plating is not thick enough, it can end up looking bluish instead of black, highlighting the importance of having a sufficient coating mass to achieve the desired black surface appearance.

3.3.3 Coating Mass Model

Unlike classification problems, where performance is typically measured using metrics like accuracy, precision, and recall, regression problems are evaluated using metrics such as MAE, MSE, and R^2. Lower values of MAE and MSE indicate better performance of a regression model, while R^2 provides insight into the model's explanatory power, with values closer to 1 indicating better performance.

In the context of predicting coating mass under experimental setting E_1, the RF regressor demonstrates the best performance metrics, achieving an R^2 value of 0.9561, along with the lowest MAE (0.0039) and MSE (close to 0) using input parameters pH, CD, and SA. Conversely under experimental setting E_2, the AdaBoost regressor exhibits superior performance, with an R^2 of 0.9725, and the lowest MAE (0.0113) and MSE (0.0002) utilizing input parameters CD, SA, and $[Z_n]$.

Figure 5 illustrates the comparison between predicted and actual mass values in the testing set using the AdaBoost regressor ($R^2 = 0.9725$). The close alignment of the predicted values with the true coating mass values highlights the model's effectiveness in accurate prediction.

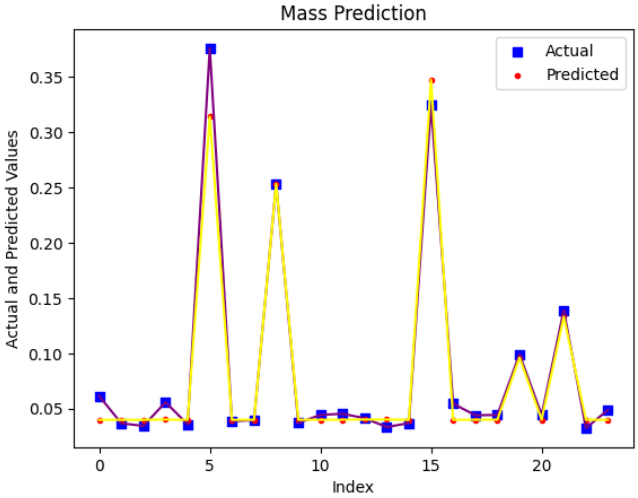

Fig. 5 Mass Model Prediction on Testing Set

We further examined the correlation coefficients for coating mass prediction, with a focus on identifying the most important parameters. The bar charts in Figure 6 and Figure 7 illustrate that current density (CD) and surface area (SA) are highly correlated with coating mass, which aligns with expectations. Specifically, a higher current density results in more coating deposited per unit area over a given time, while a larger surface area results in a greater overall coating mass. This is because a coating on a larger sample is heavier compared to the same thickness on a smaller sample due to increased coating coverage. The correlation coefficients under both experimental settings (E_1 and E_2) indicate that CD and SA are more important than either AAH or zinc concentration for predicting coating mass.

3.4 Process Optimization Using Machine Learning Models

This section studies how to find the optimal values for process parameters by using the machine learning models developed in the previous section. Further experiments were conducted to validate the prediction results.

3.4.1 Process Optimization Algorithm

This section describes the process optimization algorithm. Here we leverage the machine learning models under E_2, because the process parameter [Zn] is

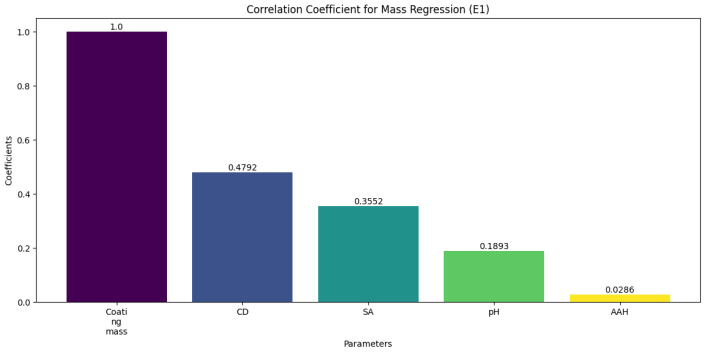

Fig. 6 Correlation coefficient of Coating Mass under E_1

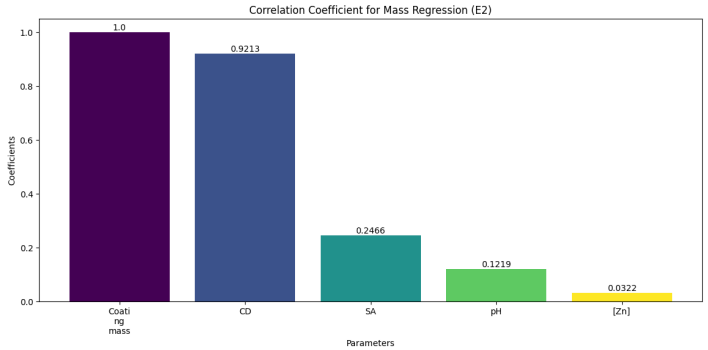

Fig. 7 Correlation coefficient of Coating Mass under E_2

more controllable in lab (i.e. more easily varied at will for validation purpose) compared with AAH in E_1. Moreover, based on the results given in section 3.3, in general ML models built using $[Z_n]$ performed better than those built using AAH as input parameter.

We first define the following concept. Let $X_p = [a_p, b_p]$, $X_{cd} = [a_{cd}, b_{cd}]$, $X_Z = [a_z, b_z]$ and $X_s = [a_s, b_s]$ represent the range of initial pH, current density, concentration of $[Z_n]$ and surface area. Let Δ_p, Δ_{cd}, Δ_z and Δ_s represent the step size of initial pH, current density, $[Z_n]$ and surface area. The process range and step size here vary based on the precision of the equipment. Let T_{des} and η represent the desired coating mass and the tolerance parameter for coating mass, where $T_{des} > 0$ and $\eta > 0$. Let f_{def}, f_{clr} and f_{cm} represent the function of the best machine learning model for the prediction of defect, colour and coating mass respectively. Therefore, the outputs of f_{def}, f_{clr} and f_{cm} are the prediction results of defect, colour and coating mass. Take defect model as an example, for any $(x_p, x_{cd}, x_z, x_s) \in X_p \times X_{cd} \times X_z, \times X_s$, the output $f_{def}(x_p, x_{cd}, x_z, x_s) = 1$ represents the prediction of defect is 1. That is, machine learning model predicts that a defect will occur if the process

parameters of pH, current density, $[Z_n]$ and surface area are set to be $\{x_p, x_z, x_s, x_z\}$.

The pseudocode of process optimization algorithm to guarantee the product with good quality (no defect), desired appearance (black colour) as well as achieving the desired coating mass is given in Algorithm 1 and shown as a flowchart in Fig. 8. In particular, the steps of the algorithm are shown as below.

1. Initialize the algorithm by giving the initial range of pH, current density, $[Z_n]$ and surface area i.e. X_p, X_{cd}, X_z and X_s, the related step size Δ_p, Δ_{cd}, Δ_z, Δ_s, the desired coating mass T_{des} and the tolerance parameter η.
2. For any parameter with the initial range, do the searching to find out the set of process parameters which satisfy the following conditions, (1) no defect $f_{def}(x_p, x_{cd}, x_z, x_s) = 0$, and (2) desired colour, i.e. $f_{clr}(x_p, x_{cd}, x_z, x_s) = 1$. Therefore, by leveraging conditions (1) and (2), process parameters obtained in X_{opt_q} guarantee good coating quality and appearance under beaker experimental setting.
3. From above set of process parameters which guarantees no defects and desired colour, find out the ones within the tolerance range of the desired coating mass. Therefore, the obtained set X_{opt} achieves good coating quality, desired appearance as well as the right coating mass.

Algorithm 1 Calculate the Optimized Process Parameter Set X_{opt}

Require: X_p, X_{cd}, X_z, X_s,Δ_p,Δ_{cd}, Δ_z, Δ_s, L_{upp}, T_{des} and η.
Ensure: X_{opt_q} and X_{opt} ;

$X_{opt_q} = \Phi$ and $X_{opt} = \Phi$
for x_p in range(a_p, b_p, Δ_p) **do**
  for x_{cd} in range$(a_{cd}, b_{cd}, \Delta_{cd})$ **do**
    for x_z in range(a_z, b_z, Δ_z) **do**
      for x_s in range(a_s, b_s, Δ_s) **do**
        if $(f_{def}(x_p, x_{cd}, x_z, x_s) == 0) \wedge (f_{clr}(x_p, x_{cd}, x_z, x_s) == 1)$ **then**
          $X_{opt_q} = X_{opt_q} \cup \{(x_p, x_{cd}, x_{ah}, x_z, x_s)\}$
        end if
      end for
    end for
  end for
end for
for $(x_p, x_{cd}, x_z, x_s) \in X_{opt_q}$ **do**
  if $T_{des} - \eta \leq f_{cm}(x_p, x_{cd}, x_z, x_s) \leq T_{des} + \eta$ **then**
    $X_{opt} = X_{opt} \cup \{(x_p, x_{cd}, x_z, x_s)\}$
  end if
end for

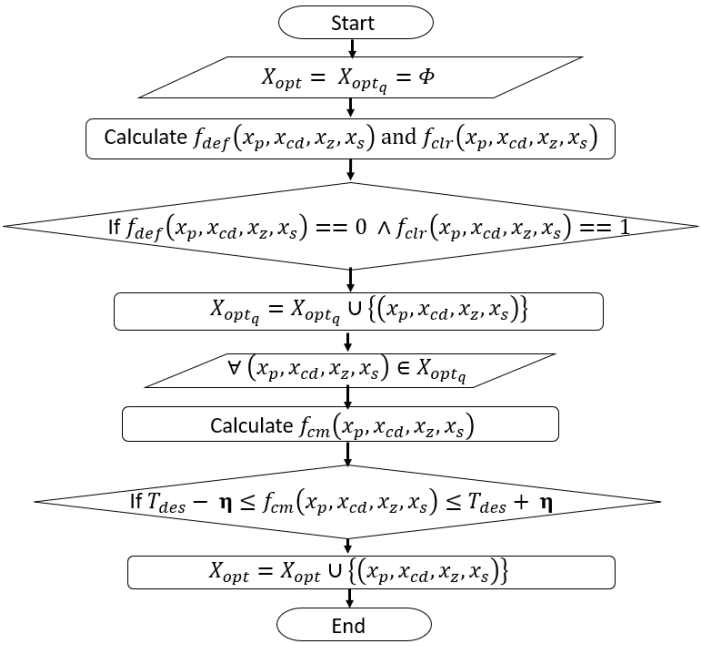

Fig. 8 Flow Chart of Optimization Algorithm

3.4.2 Illustration Example

Here is an example to illustrate how to obtain the optimized process parameter set by using the proposed algorithm. Specifically, we find out the minimal and maximal value for each parameter. Ranges for Initial pH, current density, $[Z_n]$ and surface area are $[2.8, 6.46]$, $[0.06, 1.37]$, $[0.17, 1.995]$ and $[51.7, 96]$. In order to find the optimal parameters, we generate a new data set to simulate a bunch of process parameter setting. The step size Δ_p, Δ_{cd}, Δ_z and Δ_s are set as 0.5, 0.05, 0.2, and 4, respectively. By mixing four parameters, we obtain total $25,920$ combinations. Then, we predict defect, colour label and coating mass for each simulated cases. The algorithm sorts out the process parameter set which achieves good coating quality, i.e., $f_{def} = 0$ and desired colour, e.g., $f_{clr} = 1$, and with the coating mass within the tolerance range.

For validation purposes, we chose a target coating mass of 0.04 g (a coating mass corresponding to an industrially practical thickness), then picked 4 experimental conditions spanning a typical range of zinc concentrations. The predictions and actual results of the validation experiments are shown in Table 8. The test results agreed well with the machine learning prediction results, with the defect and colour label predictions being correct (Figure 9) while the predicted coating mass values were close to 10% tolerance (Figure 10).

Table 8 Predictions and actual results of validation experiments.

					Predicted			Actual		
No.	SA	pH	CD	[Zn]	defect label	colour label	coating mass	defect label	colour label	coating mass
1.	94.7	6	0.15	1.534	0	1	0.03995	0	1	0.0447
2.	71.5	5.3	0.22	1.4	0	1	0.040	0	1	0.0366
3.	68.2	5.14	0.218	1.085	0	1	0.040	0	1	0.0371
4.	80.3	4.86	0.215	0.77	1	0	0.040	1	0	0.0429

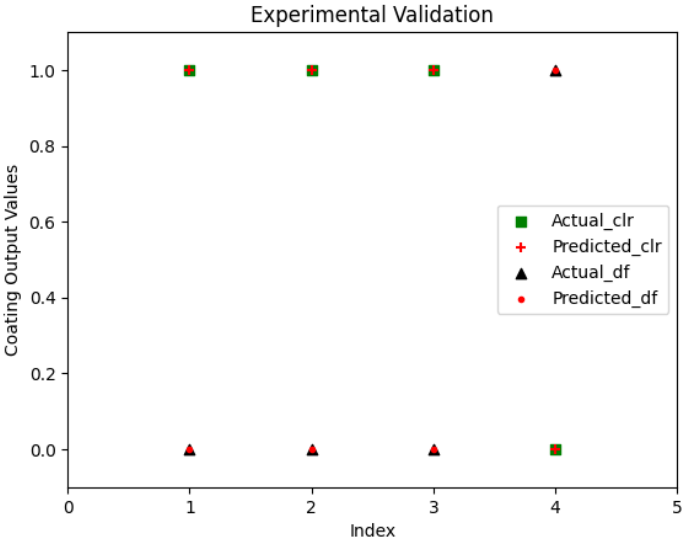

Fig. 9 Experimental Validation for Defect and Colour Labels

4 Discussion

For coating defect prediction, the best-performing model is the AdaBoost classifier under experimental setting E_1 using all four input parameters: pH, CD, SA, and AAH. Among the various machine learning approaches, the AdaBoost and RF classifiers achieved the highest F1 scores for both experimental settings E_1 and E_2. This superior performance is due to the ability of AdaBoost and RF classifiers to handle complex, nonlinear relationships and interactions among features. AdaBoost combines multiple weak classifiers to form a strong classifier, enhancing performance, particularly in cases with imbalanced data. The RF classifier, with its ensemble of decision trees, increases robustness and reduces overfitting, effectively capturing underlying patterns in the data. In contrast, MLP and logistic regression may struggle to capture these complexities and interactions, resulting in lower performance in defect prediction tasks. It can be seen that the F1 score of the various machine learning approaches is higher for experimental setting E_1 than for experimental setting E_2. This

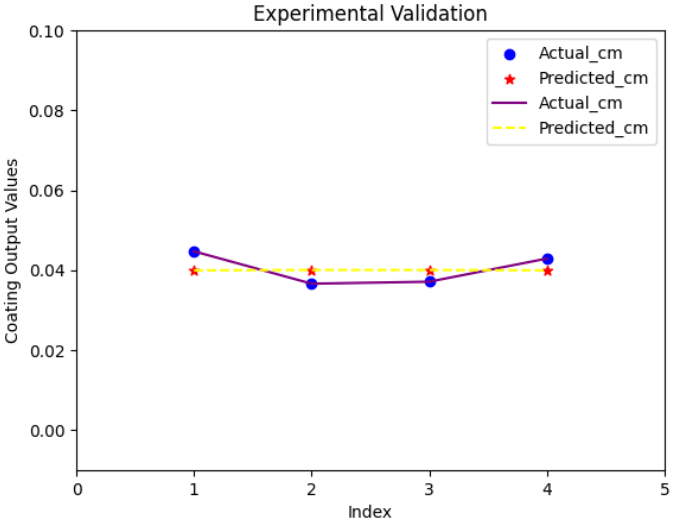

Fig. 10 Experimental Validation for Coating Mass

indicates that for defect prediction, the AAH is a more reliable input parameter compared to zinc concentration. This may be because black nickel plating is an alloy electroplating, which is more complex than typical single metal electroplating. The AAH would encompass various other aspects such as the nickel concentration and thiocyanate concentration, rather than just the zinc concentration alone.

For coating colour prediction, AdaBoost under experimental setting E_2 using all four input parameters (pH, CD, SA, and $[Z_n]$) performed best, achieving an F1 score of 0.95. Similar to coating defect prediction, AdaBoost and random forest demonstrate superior performance over MLP and logistic regression in predicting whether the coating colour will be sufficiently black in both experimental settings E_1 and E_2. This superiority can be attributed to their ability to capture nonlinear behaviour and their use of ensemble learning techniques, which aggregate predictions from multiple individual models to form a final prediction. Ensemble methods like AdaBoost and random forest are more robust to noisy data compared to single models such as logistic regression or MLP. Moreover, colour classification model under experimental setting E_2 performed better than those under experimental setting E_1 for three out of five of the machine learning approaches. This indicates that for coating colour, the zinc concentration is indeed the most important chemical species in the plating bath and that tracking this input is more important than tracking the AAH (which encompasses more than just the zinc concentration). This is consistent with domain expert understanding that zinc co-deposition with nickel is important to achieve a black coating appearance that results from a coating structure that is hypothesized to be mostly nickel sulfide and zinc plus free nickel [18]. Hydrogen evolution as a side reaction occurs during the deposition of nickel and zinc or zinc-nickel on the substrate. This results in a localized

increase in the pH value near the substrate surface, which then impedes the deposition of nickel and zinc due to complexation by ammonium ions. Reduction of thiocyanate then increases, which results in the deposition of nickel sulfide into the coating.

For coating mass prediction, the AdaBoost regressor exhibits superior performance under experimental setting E_2, with an R^2 of 0.9725, and the lowest MAE (0.0113) and MSE (0.0002), utilizing input parameters CD, SA, and $[Z_n]$. Other machine learning models under E_2, like the random forest regressor, multi-layer perceptron regressor (MLPR), and linear regression, also perform well with R^2 values greater than 0.95. These metrics suggest a close linear relationship between process parameters like CD, SA, and pH, and the output: coating mass prediction in E_2. However, for E_1, the RF regressor demonstrates the best performance metrics, achieving an R^2 value of 0.9561, along with the lowest MAE (0.0039) and MSE (close to 0) using input parameters pH, CD, and SA. Other machine learning models like the random forest regressor and MLPR also perform well, but the performance of linear regression is significantly lower, with an R^2 of only 0.5398. This discrepancy may be attributed to the limitations of linear regression in capturing complex, nonlinear relationships between input variables and coating mass. Moreover, all four regressor machine learning models under experimental setting E_2 performed better than their counterparts under E_1, implying that zinc concentration is more important than AAH, consistent with domain expert understanding. Under experimental setting E_1, the best models (AdaBoost and RF) do not include AAH as an input parameter, indicating that AAH is not as critical for predicting coating mass."

In general, AdaBoost classifier/regressor and Random Forest classifier/regressor perform better than MLP classifier/regressor and other linear models. This is due to the nonlinear and complex nature of the models required for coating defect, colour, and mass prediction. In particular, the AdaBoost approach outperformed Random Forest, which aligns with the general understanding that AdaBoost typically achieves higher performance with imbalanced datasets, as is the case with our data. However, it is also noted that AdaBoost is more sensitive to overfitting compared to Random Forest.

In the past, electroplaters are typically restricted to using simple statistical analysis and Hull Cell type experiments for process optimization. Process optimization with such conventional approaches are highly knowledge-driven, dependent on the expertise and experience of the electroplater. New insights from machine learning models, such as the critical factors and their importance ranking, are able to guide electroplaters for process optimization, and potentially how to adapt process parameters to changing plating bath conditions such as AAH increasing or zinc concentration decreasing with use.

The advantages of the machine learning approach are that machine learning is able to identify nonlinear patterns much better than domain experts, the predictions for coating mass is more convenient and can also be obtained much faster once the model is developed and optimized, with comparable or

better accuracy than predictions from domain experts. Using machine learning approach, the user can be data-driven instead of knowledge-driven, which can be advantageous for processes with highly nonlinear behaviour, such as the black nickel plating process studied in this work. This is also advantageous when there is a lack of local experts for the process, as the developed machine learning model can fill in the role of the domain expert and improve its performance via model refinement as more data is accumulated.

The advantage of using machine learning for process optimization in improving the process control of nonlinear process such as black nickel plating via a machine learning approach is pertinent for achieving high productivity, by ensuring the quality of the black nickel coating can be achieved right first time (i.e. no defects, right colour, right coating thickness/mass), in order to avoid unnecessary rework.

5 Conclusions

This paper studied the process optimization of black nickel electroplating process using a machine learning approach. To achieve a greener manufacturing, a boric acid-free process was investigated, which is a more challenging process to control due to the lack of pH buffer. Three machine learning models were developed for predicting coating defect, coating colour and coating mass respectively. This paper identified AdaBoost classifier as the best machine learning model to predict coating defect (with F1 score of 0.9875), coating colour (with F1 score of 0.95) and AdaBoost regressor (with R^2=0.9561, MAE=0.0039, MSE =0.00001) as the best machine learning model to predict coating mass. By using the above three models, we can simulate varied process parameter settings and predict whether the proposed parameters can achieve a black nickel plating with the desired surface appearance. A corresponding process optimization algorithm was further developed, that enables the selection of the process parameters which guarantee a good coating satisfying no defect, desired black colour and desired coating mass simultaneously. This will be helpful for industry to improve product quality as well as productivity.
Research Contribution
Compared with the existing literature, the research contributes of the paper laid in the following aspects: 1. Given the sensitivity of this electroplating process due to the lack of pH buffer, we developed three machine learning models for predicting coating defects, colour, and mass. To address the issue of the highly imbalanced dataset for defect and colour prediction, we employed adaptive synthetic sampling to generate synthetic examples for the minority class. This enabled us to develop effective machine learning models for both defect and colour prediction. 2. Unlike existing studies that focus solely on coating mass prediction, we developed machine learning models not only for coating mass prediction but also for defect and colour prediction, which are critical for ensuring coating quality. 3. We proposed a novel optimization algorithm based

on machine learning predictions for process tuning. This optimization process achieves both high coating quality and productivity, which is essential for industrial applications that require both efficient production and high-quality coatings. 4. Since the process optimization is based on machine learning models rather than domain expertise, tuning process parameters can be faster when conditions change compared to conventional process optimization methods.

Industrial Contribution

1. The results demonstrate how AI/ML can be effectively applied to industrial processes such as black nickel plating, which is relatively complex due to the presence of two main cations in the electroplating bath. 2. AI/ML enables more controlled plating operations, leading to consistent and high-quality outcomes. It serves as an enabler, helping beginners achieve expert-level skills by providing insights and guidance. The ability of an AI/ML-enabled machine to work autonomously can significantly increase productivity, allowing for operations such as overnight shifts with minimal human supervision. 3. AI/ML provides a better understanding of critical factors in the electroplating process, helping to extract meaningful signals from noise. Just-in-time rectification leads to less waste, as issues can be addressed promptly based on AI/ML predictions. 4. Easier tuning of parameters is possible when desired outputs change (e.g., different L value thresholds, different coating weights), without the need to start the analysis from scratch. Using small beaker experiments to vary parameters for data collection and model building is more feasible and less disruptive than altering large industry-scale plating tanks during actual operations. The ideal approach involves building the model largely on data from small-scale experiments and then fine-tuning and validating it using actual shopfloor data, ensuring its applicability and accuracy in real-world scenarios.

Limitations and Future Work

This study encountered several limitations in using a machine learning approach. Firstly, the predictions are significantly more accurate for interpolation than for extrapolation, especially when dealing with nonlinear behaviors such as those observed in black nickel plating. Secondly, there is a need for relatively large quantities of high-quality and standardized data. Researchers cannot mix data from different experimental settings, as building models with non-standardized data of various types is challenging.

Significance of Findings in the Electroplating Sector Context

AI/ML can be successfully applied to an electroplating process if a process with the right level of complexity is chosen. If the process is too simple, AI/ML may not be needed and a simpler analysis could suffice. If the process is too complex, data collection becomes more tedious, and the developed models require a higher level of complexity. Not all data can be easily and frequently collected. For example, critical organic compounds in the bath, such as levellers and brighteners, may be present in small quantities that are hard to measure accurately and differentiate between various species. This problem can be mitigated with advanced sensors capable of gathering data on these critical species quickly, accurately, and affordably.

There are still a few challenges to be solved for commercializing of this machine learning approach for black nickel electroplating to take place. Key challenges include the need to validate the model using actual shopfloor data, which is essential because the larger tank size may introduce new factors not present in small-scale (e.g., beaker) experiments. Additionally, it is crucial to have sensors in place to collect this shopfloor data. Finally, the developed models must be tuned using the shopfloor data to ensure their effectiveness and reliability in a real-world industrial setting.

Future work can explore the application of this approach to other types of electrochemical processes. Additionally, embedding the developed machine learning models into a system for industrial deployment could contribute to the development of smart auto-plating lines.

6 Statement

6.1 Competing Interests

The authors declare no competing interests

6.2 Funding Information

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6.3 Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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