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Application of artificial neural networks to optimization the parameters of ion-plasma deposition of thermionic coatings

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A method for optimizing the parameters of the ion-plasma sputtering process of triple carbonates of alkaline-earth metals Ba, Sr, Ca is proposed. An artificial neural network (ANN) with a genetic algorithm is used to build a predictive model. The sputtering parameters are determined to obtain an emission current in the range of 70–120 μ A for thin-film thermionic microcathodes. The prediction results are compared with the actual emission data of the manufactured microcathodes and those calculated by other machine learning models. Based on the comparison, it was shown that the ANN allowed for finding a more accurate combination of coating deposition parameters to obtain efficient thermionic microcathodes.

Keywords: molecular sputter-deposited oxide microcathode (MSOC), ion-plasma deposition, machine learning, forecasting, artificial neural network, genetic algorithm.

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Introduction

Ion-plasma (magnetron) sputtering (IPS) technologies allow for control over the deposited film thickness and composition. A plasma medium is employed in these processes to eject target material for deposition onto a substrate to form a thin film. Complex physicochemical phenomena occurring during coating deposition require precise control of key process parameters, such as pressure, working gas flow rate, substrate temperature, and sputtering power. The study of these processes enables control over film stoichiometry and structure, which, in turn, affects their functional characteristics [1].

The authors of [2] developed a hybrid machine learning (ML) approach that combines respective classification and regression models to address challenges related to producing high-mobility amorphous In_{0.9}O_{0.1}:Sn films deposited via RF magnetron sputtering.

In [3], various characteristics of low-temperature plasma were reviewed along with

their complexity, which is driven by diverse gas-phase compositions, operating pressures, power densities, and spatial boundaries. Under these conditions, the direct application of machine learning methods is not always feasible. To overcome these difficulties, progress is required in plasma diagnostics and data acquisition systems. Processing large amounts of data is one of the key tasks to achieve this goal.

By simulating the transport of argon ions bombarding a Ti–Al composite, the authors of [4] obtained a series of representative energy distribution functions of incident particles. Subsequently, a multilayer artificial neural network (ANN) was trained and tested on this dataset of input and output distributions. Following the training phase, an error analysis was performed, and the model performance was compared for two sets of hyperparameters. The trained ANN demonstrates the ability to predict sputtered particle distributions for unknown incident ion energy distributions of arbitrary shape.

In [5], an efficient process design system was introduced to determine optimal operating conditions through a combination of deep learning and plasma simulation. To create the training model, a dataset was generated using a two-dimensional (2D) hybrid plasma equipment model code for an inductively coupled argon plasma system. The model was trained on the generated dataset to investigate the functional dependencies between operating conditions and subsequent plasma states.

The refractive indices of SiN_x films deposited on silicon substrates via magnetron sputtering were predicted in [6] using an ANN. Experimental wavelength and temperature values were utilized as inputs for ANN training, with the refractive index as the output parameter. The simulation and prediction results obtained using the model were compared with experimental data. The results demonstrate that the ANN approach is suitable for the simulation and prediction of the refractive index.

Soft computing techniques, including genetic algorithms (GAs) and ANNs, are widely utilized in addressing various nonlinear problems, owing to the superior performance of these models [7, 8]. ANNs enable the extraction of complex patterns from data.

The subject of this study is thin-film molecularly deposited oxide cathodes (MOCs) used in cyclotron protection devices (CPDs), which are deposited via ion-plasma sputtering (IPS) of cathode targets prepared from alkaline earth metal carbonates. They provide a current density of 1–2 A/cm² for 5,000–10,000 hours. The thickness of the deposited emission film on the micro-cathodes is $\approx 1 \mu\text{m}$ [1, 9].

The objective of this study is to construct and optimize a model to predict the target variable, micro-cathode current (I_{cat}), based on a set of deposition process parameters.

Materials and Methods

The deposition of coatings onto the micro-cathodes was carried out using an URM3.279.011 vacuum ion-plasma sputtering system manufactured in Russia. The geometric dimensions, operating principles, and key electrical parameters of the cathode-heater

assembly (CHA) and the working chamber of this system are identical to those described in [10,11]. The (Ba, Sr, Ca)CO₃ cathode targets for IPS were prepared by spraying from powdered AEM carbonates of the KTA-1-6-sp grade. The ratio of active AEM elements in the target was 47:45:8, and the thickness was 130–135 μm . The turbo-molecular pump provided a pumping speed of 500 L/s. The ultimate vacuum level was 1×10^{-6} Pa. After target carbonate decomposition, the DC voltage applied between the target and the substrate provided the energy required to ionize argon atoms and dissociate CO₂ gas, resulting in a plasma state. Driven by the applied electric field, argon ions bombard the target, causing material sputtering. As the sputtered atoms and molecules move toward the substrate holder, they react with the gas and deposit onto the micro-cathode cores, forming thin films of AEM oxides and carbonates. The sputtering process is regulated by adjusting the operating parameters of the system.

The dataset was generated by depositing coatings onto MOCs and then measuring the emission current of the micro-cathodes inside the vacuum devices. This dataset was obtained from processes conducted under various conditions with varying coating deposition parameters. The target variable I_{cat} was separated from the features. The dataset was split into training (80%) and testing (20%) sets. All features were scaled using the training set and then the transformation was applied to the testing set. The initial dataset contained 253 rows and 13 columns.

The study consisted of the following stages: data collection and processing, model development and prediction using ANNs, and optimization via GAs. Columns with completely missing values were removed. The missing data was checked and processed. Twelve deposition parameters were selected: discharge voltage U_{dis} , V; discharge current I_{dis} , mA; target heating voltage during sputtering U_{nm} , V; target heating current during sputtering I_{nm} , A; reflector voltage U_{otr} , V; reflector current I_{otr} , A; solenoid current I_{sol} , A; sputtering pressure P , Pa; substrate temperature T_{sub} , °C; target carbonate decomposition time t_{dec} , min; maximum target decomposition current I_{dec} , A; and sputtering time t_{sup} , min. The GA was adapted to find the optimal combination of parameter settings. The objective

function of this study was to maximize the cathode emission current I_{cat} . The result obtained using the GA was verified with the ANN and compared against experimental data as well as results from other machine learning (ML) methods, such as support vector regression (SVR), random forest (RF), gradient boosting (GB), and XGBoost (extreme gradient boosting) [12–14, 15]. The GA is based on the evolutionary principle and involves four main stages. These include creating a population of solutions, evaluating the fitness of each solution, selecting individuals, and forming a new population based on three genetic operations: reproduction, crossover, and mutation [7, 8]. Reproduction occurs when each individual is duplicated based on its fitness values [16]. The mutation selected in this study replaces the cathode current with a uniformly distributed random value within the specified lower and upper limits of 60–120 μA , corresponding to the low-temperature range (480–490 $^{\circ}\text{C}$) of the MOC emission characteristics [1, 11]. This low-temperature region was chosen because it characterizes and determines the emission activity and quality of the micro-cathode across the entire emission curve. The objective function of the study was to maximize the emission current of the sputtered cathodes in a specific low-temperature range of underheating characteristics. The model inputs were the sputtering parameters, while the output was the micro-cathode current. The neural network architecture was defined using the Keras library. The baseline model consisted of four fully connected layers with LeakyReLU activation functions (with a parameter of 0.01) and batch normalization layers. L2 regularization was applied to control overfitting and improve the generalization capability of the model. The input layer consisted of 12 neurons. The hidden layers had 128, 128, 64, and 32 neurons, respectively. The output layer contained a single neuron with a

linear activation function for the regression task. The model was trained for 100 epochs with early stopping. The model was compiled with the Adam optimizer (learning rate of 0.001), the MSE (mean squared error) loss function, and evaluation metrics including MAE (mean absolute error) and R^2 . The metrics of the baseline model with four hidden layers on the test data were as follows: MSE: 431.9; MAE: 15.5; R^2 score: 0.47. The R^2 score represents the percentage of variability in the dependent variable that is predictable from the independent variables used in the model. The target metric for optimization was R^2 with a maximization direction. A total of 30 trials were conducted, where each model configuration was trained twice to enhance the stability of the evaluation.

Training the optimized model using the selected hyperparameters demonstrated a significant performance improvement. The model was trained for 250 epochs. The Keras library along with the `keras_tuner` package and the RandomSearch algorithm were used. Different combinations of hyperparameters were randomly selected. The algorithm trained the model on the training data for each selected hyperparameter combination. Based on the optimized hyperparameters, an ANN model was constructed with the following architecture and training configuration: four hidden layers. The first layer had 480 neurons with the 'relu' activation function, the second layer had 160 neurons with 'tanh', the third layer had 96 neurons with the 'leaky_relu' activation function, and the fourth layer had 128 neurons with 'relu'.

After training the optimized ANN, its performance metrics on the testing set changed as follows: MSE: 229.68; MAE: 12.17; and R^2 : 0.72. The tuning process significantly improved the predictive performance of the model. The learning curves of the optimized model are presented in Fig. 1 (a, b).

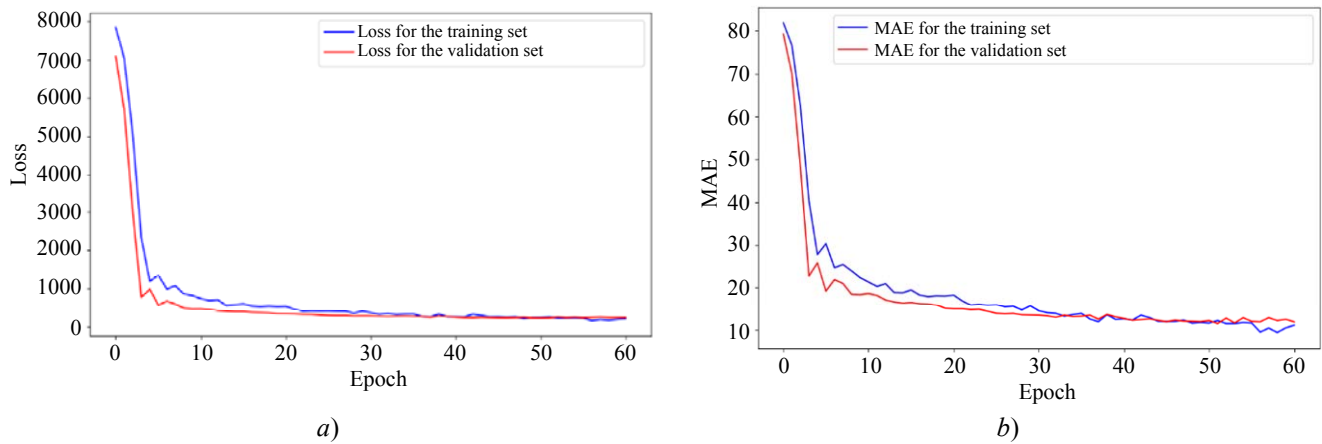


Fig. 1. Model training history on the training and validation sets: a) loss, b) MAE

Results and Discussion

The process data related to target carbonate heating and decomposition, coating deposition in the IPS system, and emission measurements were used in this study. Values recorded at other stages this re only indirectly related to the emission current, such as vacuum pumping or assembly of the sputtered cathodes, were not considered. The addition of these stages would have required the creation of a much larger dataset, thereby significantly increasing the complexity of the problem.

The training history curves (Fig. 1a, b) show that both training and validation errors decreased over the epochs, demonstrating that the model learned successfully. Two key metrics are evaluated: loss (Loss) and mean absolute error (MAE). For each metric, two lines are displayed: the blue line shows the metric values on the training set (the data the model learned from), while the red line represents the metric values on the validation set (data excluded from the training process to evaluate the model's generalization capability). The loss curve illustrates the evolution of the model error over the training epochs. As observes, the validation loss does not significantly diverge from the training loss, indicating that no overfitting occurs and the model performs well on new data, thus maintaining its generalization capability. The MAE metric reflects the mean absolute deviation of predicted values from the actual values, where a lower MAE metric reflects that the model's predictions are closer to the actual values on average. Similar to the loss, both MAE lines also decrease and then stabilize. The low MAE values

on the validation set confirm that the model delivers accurate predictions. The similarity between the training and validation MAE indicates the absence of significant overfitting.

A feature importance analysis was conducted based on the averaged absolute connection weights of the first layer of the optimized model. The feature importance of the optimized ANN model was calculated and sorted in ascending order. The resulting vertical bar chart (Fig. 2a) illustrates the numerical importance of each feature.

According to the analysis results (Fig. 2a), the most important features (with the largest average absolute weights in the first layer) are the deposition parameters: I_{dec} , U_{dis} , I_{nm} , I_{dis} , while the least important are I_{otr} , U_{nm} , P . The parameter I_{dec} has the highest importance. Randomly varying this parameter significantly degrades the model's prediction quality. The subsequent thermionic emission capability and gas ionization for cathode target sputtering depend on the correctness of the vacuum thermal decomposition of the cathode target carbonates prior to the actual coating deposition onto the micro-cathode cores. This decomposition process, in turn, depends on the temperature of the AEM carbonates, the duration of this process step, and the residual gas pressure in the chamber. During thermal target decomposition, critical physicochemical processes occur both within its bulk and on its surface, forming an oxide layer of the appropriate composition from which the sputtering onto the micro-cathodes takes place. U_{dis} determines the accelerating voltage required for cathode target sputtering. Simultaneously, I_{nm} along with U_{nm}

determine the heating power essential for the heating and the diffusion of active substance

atoms to the target surface during coating deposition.

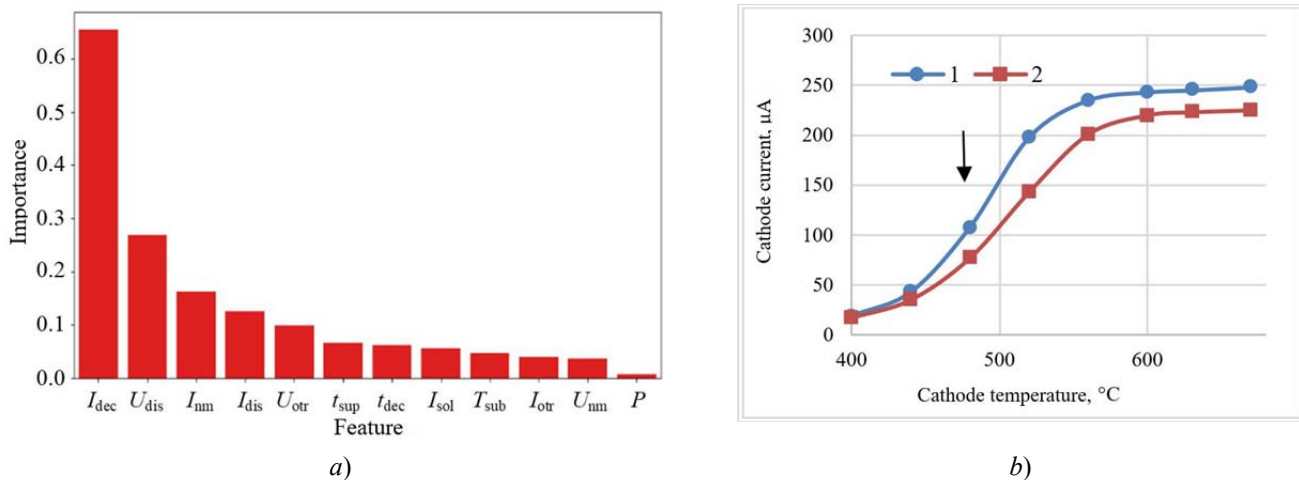


Fig. 2. Feature importance chart for the optimized model (a). Emission characteristics of two experimental MOCs obtained after thin-film deposition using GA (b)

As a result of the GA application, a set of input features (the best solution) was found that, when fed into the optimized ANN, determined the predicted value $I_{cat} = 90 \mu A$. This value corresponds to the center of the target range and maximizes the fitness function (Table 1).

The study also yielded the dependencies of the predicted cathode current on the sputtering process parameters. The predicted MOC current increases with rising accelerating discharge voltage, target carbonate decomposition, and discharge currents. Conversely, a decrease in the working gas pressure from 0.13 to 0.11 Pa results in an increase in the predicted I_{cat} . The results obtained are consistent with earlier studies reported in experimental works [1, 9–11].

After conducting the experimental sputtering onto the micro-cathodes using process parameters close to the GA-optimized values from the table, the I_{cat} values were measured for

two MOCs integrated into vacuum device mock-ups. The obtained underheating characteristics of the micro-cathodes are shown in Fig. 2b. In the low-temperature region of the emission curves (480–490 $^{\circ}C$), the measured I_{cat} values were 108 μA (No. 1) and 77 μA (No. 2), which fall within the target objective function range.

Other machine learning models (RF, GB, SVR, and XGBoost) were trained and evaluated for performance, with the R^2 metric used as the evaluation criterion. The comparison results for the models were as follows: ANN: 0.72; RF: 0.61; GB: 0.62; XGBoost: 0.42; and SVR: 0.25. The prediction accuracy of the ANN is higher than that of the other traditional ML methods. While the ANN, GB, and RF models proved to be the most suitable for predicting I_{cat} based on the current dataset, but the ANN outperformed all other models, demonstrating the highest predictive capability.

Table

Process Parameter Optimization Using GA

U_{dis} , V	I_{dis} , mA	U_{nm} , V	I_{nm} , A	U_{otr} , V	I_{otr} , A	I_{sol} , A	P , Pa	T_{sup} , $^{\circ}C$	t_{dec} , min	I_{dec} , A	t_{sup} , min	I_{cat} , μA
821	226	23	9.5	24	36	0.38	0.12	228	86	9.7	177	90

Conclusion

In conclusion, an ANN model architecture was developed and trained to predict the subsequent thermionic emission current of micro-cathodes based on experimental data from the vacuum ion-plasma sputtering of emission coatings. A method for optimizing the IPS process parameters was proposed for the fabrication of efficient thin-film emitters. Using a

genetic algorithm, the best set of sputtering parameters was found, ensuring a micro-cathode current in the range of 60–120 μA within the low-temperature region of the emission characteristics. The ANN showed acceptable prediction accuracy on the test data with an R^2 score of 0.72, surpassing traditional regression ML models. These simulation results can be used for scaling up sputtering processes in industry.

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