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THE APPLICATION OF THE PROCESS MODELLING OF ANODIC WET-STRIPPING OF CrN MULTI-LAYER COATINGS FOR CHARACTERISTICS PREDICTION

Keywords

Multi-layer coatings, anodic wet-stripping, statistical modelling, artificial neural networks, prognostic model.

Abstract

The paper presents the results of experimental research on a process of anodic wet-stripping of CrN multi-layer coatings. The stripping rate was correlated with the coating structure and the current density of the stripping process. The experimental data was statistically analysed and regression models of stripping thickness were created as a function of stripping time. The obtained results indicated that the anodic wet-stripping process can be described by means of linear function only in the case of one-layer coatings. Moreover, the general neural network model was created as a complex model including both quantitative and qualitative variables characterising the wet-stripping process. The developed models enable the estimation of the character and time of the stripping process, depending on the coating thickness, structure and current parameters.

Introduction

CrN coatings, deposited using PVD methods belong to the wear resistant coatings group, which importance in industry increases especially for the durability improvement of the moulds and tools in technological processes [1, 2, 3]. CrN coatings have favourable tribological (high wear resistance and low friction), mechanical (high hardness, toughness) and chemical (corrosion resistance) properties; therefore, the scope of their utilisation is broadening. However, the increased corrosion resistance and good oxidation resistance ensuring high durability of the tools operating in a chemical aggressive environment [4, 5, 6, 7] also cause essential a problem in the situation when the coating has to be stripped from the substrate. In industrial practice, the operation of coating stripping is most often needed in the following cases:

- when after deposition the coating defectiveness are revealed such as: structure heterogeneity, non assumed roughness, adhesion or thickness [8],
- when, the coated tool suffered damage but can be regenerated and require recoating before further use in production processes [9].

Because of high quality properties that have to be achieved for elements' surfaces, which are covered by PVD coatings, the proper technology of stripping should not interfere into the substrate's surface structure and may cause only negligible damage to substrates. Analysis of literature [9–13] has shown that the efficient technology of CrN coating stripping from steel substrates can be developed by adaptation of the well known, and used in galvanic-technique, method of the anodic wet-stripping of the chromium coatings.

Due to the increase in the number of different kinds of multi-layer coatings that are broadly applied in engineering practice, the important problems need to be solved, namely, the identification of both the process parameters and the coating structure influence on the stripping rate. This paper presents the results of the experimental research and the process modelling of the coatings anodic wet-stripping conducted in order to evaluate the period of time needed for total coating removal.

Four different kinds of coatings were examined. The experiments of wet-stripping were conducted in the condition of different current parameter variables.

1. Method of CrN multi-layer coatings anodic wet-stripping

All CrN multi-layer coatings were deposited onto 4H13 or W300 steel discs of ϕ = 30 mm diameter and 12 mm thickness. Table 1 shows the different structure and thickness of the coatings deposited by vacuum arc evaporation. The photomicrographs of coatings ball shaped section are shown in Fig. 1.

In order to evaluate the current parameter influence on the anodic wet-stripping rate of the CrN multi-layer, the stripping experiments were conducted with different values of the current density: 4.3 A/dm^2, 6.0 A/dm^2, 7.3 A/dm^2.

For each stripping process, the evaluation of the dissolution rate was investigated by the coating thickness measurement. The specimens were removed from the stripping solution at regular time intervals and coating thickness was determined using ϕ = 25 mm diameter ball crater measurements. The microscopy observations of the coating topography changes were also conducted. The average coating thickness was calculated from three measurements taken at different specimen positions. The stripping was continued until the complete removal of the CrN coating, and the total time was determined.

Table 1. Structure and thickness of the coating used in experiments

Lp.	Coating type	Coating structure	Coating thickness
1	multiCrN-1	4M13 steel substrate - CrN/(Cr-CrN)x5	4.0 μm
2	multiCrN-2	4M13 steel substrate - CrN/(Ti-CrN)x5	5.0 μm
3	multiCrN-3	4M13 steel substrate - CrN/(TiN-CrN)x5	4.5 μm
4	NL-CrN	W300 steel with nitriding layer substrate - CrN	3.3 μm

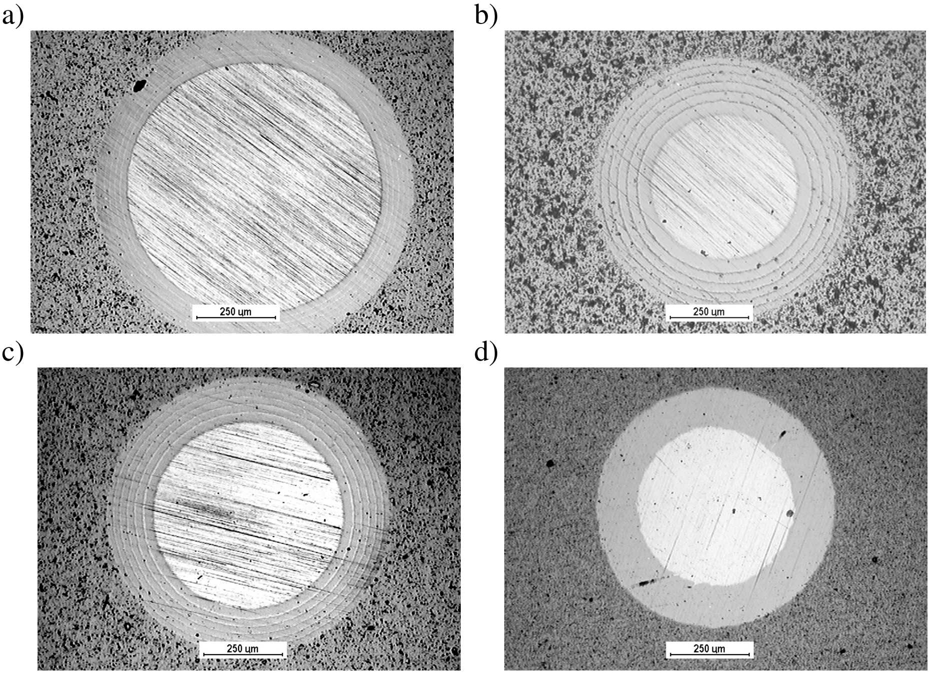

Fig. 1. Photomicrographs of coating ball shaped section: a) multiCrN1, b) multiCrN-2 c) multiCrN-3, d) NL-CrN

The experimental data (coating thickness as a stripping time function) were processed in statistical analysis [14]. Modelling research was conducted with the use of the non-linear module of StatisticaPL software version 6 [15]. As the measure (criterion) of model accuracy (on the significance level $\alpha = 0.05$) the coefficient R has been calculated:

$$R = \sqrt{\frac{\sum_{i=1}^{n} (\hat{y}_i - \bar{y})^2}{\sum_{i=1}^{n} (y_i - \bar{y})^2}} \tag{1}$$

where y_i – value of depended variable achieved from experiments,
$\hat{y}_i$ – theoretical value of depended variable calculated from regression function,
$\bar{y}$ – arithmetical average of y_i variables.

There was also applied modelling with the use of artificial neutral network method for the creation of the model of the coating thickness changes during anodic stripping as the function of time, process parameters and coating structure. In modelling the Neural Network module of Statistica PL software was used.

2. Modelling of the coatings stripping

The experimental results (presented in Fig. 2-4 as points) showed that the character of CrN coatings stripping differs in dependence on the coating structure. For all deposited coatings it is also noticeable that the stripping rate accelerates when current density increases.

In the beginning of statistical modelling a regression function type was chosen for describing relation between the stripping coating thickness and the time of the stripping period; then function parameters were calculated. Due to the importance in the technological applications of the time point in which the coating is totally removed from the substrate, in modelling, it was assumed that the experimentally established point ending the coating stripping should be especially considered.

Firstly, all processes were modelled as linear models, and for all experiments those models appeared to be statistically significant. However, only in the case of the NlCrN coating, the high R coefficient was achieved ($R \in [0.988 – 0.998]$ this proved a very good linear function adjustment to the experimental results (Fig. 2; Table 2). Because coefficients R of regression linear models for all other multi-layer coatings stripping were not so high, namely: 0.945 to 0.978:

for multiCrN1, 0.842÷0.901 for multiCrN2 and 0.883÷0.917 for multiCrN3, the regression non-linear models were developed for those coatings. Among non-linear models (parabolic, hyperbolic and exponential) only exponential models, $y = e^{ax+b}$, were statistically significant for all processes of the CrNx coating stripping.

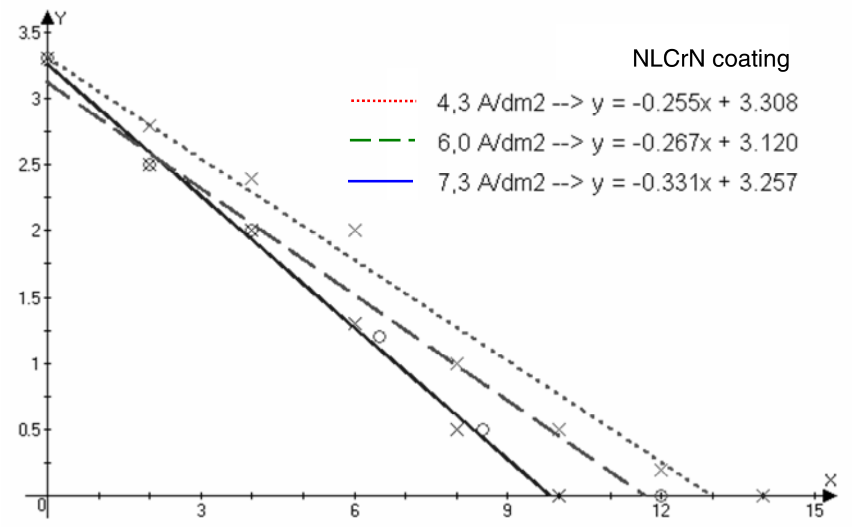

Fig. 2. Experiment results and regression function adjusted to NLCrN coating stripping for different values of the current parameters

Table 2. Statistical parameters of the regression linear models

Coating	Current density	Parameter a	Stand. error of a	Parameter b	Stand. error of b	R
NLCrN Y=ax+b	4.3	-0.255	0.016	3.308	0.137	0.988
	6.0	-0.267	0.014	3.120	0.101	0.994
	7.3	-0.331	0.010	3.257	0.064	0.998

The regression exponential model $y = e^{ax+b}$ was modified as $y = e^{ax+b} - C_0$ in order to achieve "0" point when a coating is totally removed. C_0 value was calculated from equation: $C_0 = y(x_0)$; where x_0 is the time of total coating removing period, determined from the experiment. The statistical parameters of the regression models: $y = e^{ax+b} - C_0$ have been shown in Table. 3 and model graphs in Fig. 3-5.

High values of R coefficients, statistical significance and low standard errors of all estimated parameters proved very good exponential models fitting the experiment results.

Table 3. Statistical parameters of regression exponential models

Coating	Current density	Parameter a	Stand. error of a	Parameter b	Stand. error of b	C_0	R
MultiCrN1 $Y = e^{ax + b} - C_0$	4.3	-0.140	0.012	1.558	0.047	1.059	0.991
	6.0	-0.184	0.012	1.539	0.037	0.504	0.993
	7.3	-0.235	0.012	1.528	0.029	0.339	0.996
MultiCrN2 $Y = e^{ax + b} - C_0$	4.3	-0.116	0.008	1.742	0.044	0.485	0.984
	6.0	-0.154	0.014	1.709	0.052	0.106	0.984
	7.3	-0.163	0.020	1.643	0.020	0.106	0.998
MultiCrN3 $Y = e^{ax + b} – C_0$	4.3	-0.053	0.003	1.563	0.033	0.213	0.984
	6.0	-0.067	0.003	1.483	0.026	0.092	0.993
	7.3	-0.089	0.004	1.496	0.025	0.062	0.996

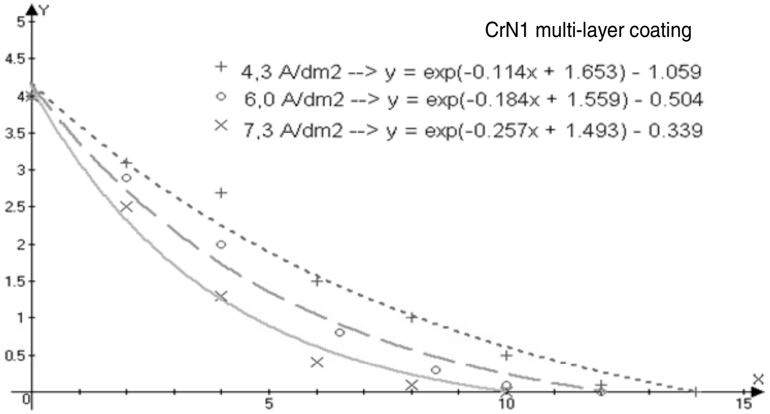

Fig. 3. Experiment results and regression exponential model: $y = e^{ax+b} - C_0$ for multiCrN1 coating stripping as different values of current parameters

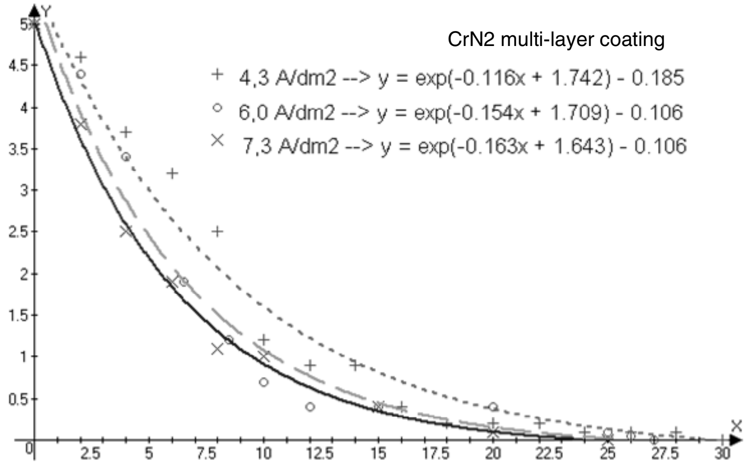

Fig. 4. Experiment results and regression exponential model: $y = e^{ax+b} - C_0$ for multiCrN2 coating stripping as different values of current parameters

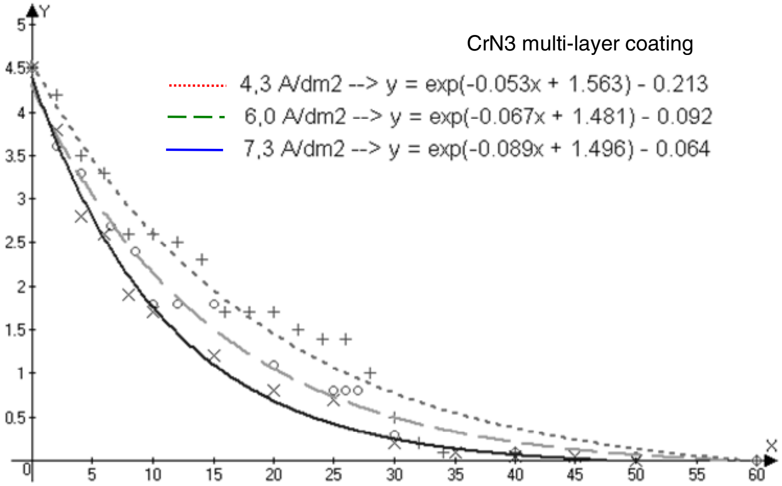

Fig. 5. Experiment results and regression exponential model: $y = e^{ax+b} - C_0$ for multiCrN3 coating stripping as different values of current parameters

In spite of very good adjustment, developed regression statistical models are useless when the current parameters differ from those used in experiments. So, the other modelling method, namely artificial neural network [16] was used for developing a model of coating stripping. Such a method enables, among others, inclusion of both: qualitative (coating structure) and quantitative (thickness, time, density) data into one model. The neural network model for coating stripping was developed as Multi-Layer Perceptron (MLP) with four neurones of logistic activation function in a hidden layer (Fig. 6). Schemes of the neural network model fitting the experiment data are shown in Fig. 7.

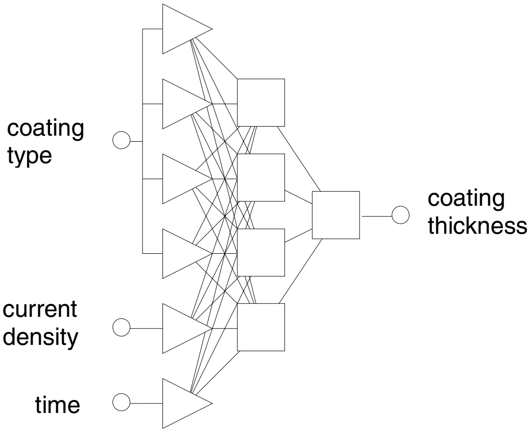

Fig. 6. Coatings stripping neural model of the MLP type

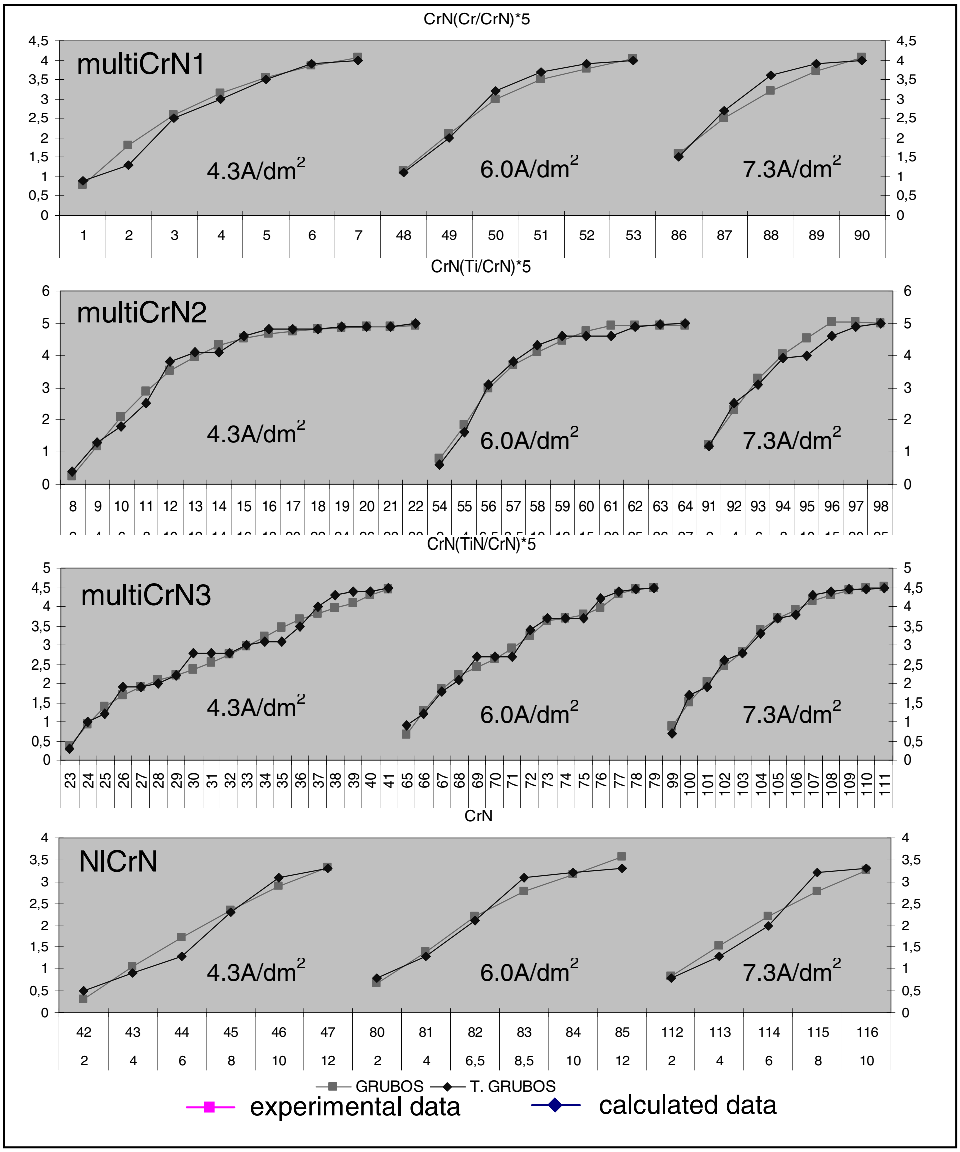

Fig. 7. Neural model fitting the experimental data for different current density both the data vector numbers and below the time period [in min] of stripping are given on the horizontal axe

As a result of an analysis of the inputs sensitivity, on the basis of which evaluation of the variables importance for model quality is carried out, the most important is the coating structure variable. The developed model investigation proved its good adjustment to experimental data; both training and verification correlation coefficients as well as testing data have high values as follows: 0.993; 0.991; 0.981, that show model ability of generalisation.

As it can be seen from Fig. 7 the model meets the assumption of precise representation of the final point of the coating removal.

Conclusions

Statistical analysis of experimentally obtained data that characterise the changes of the multi-layer coatings thickness in the time of the stripping period has enabled the identification of the coating stripping regression model type and its parameters. Research results shown that coating stripping rate depends mainly on the coating structure and, in the investigated current range (4.3÷7.3 A/dm^2), increases along with a current density increase.

The stripping rate can be described by a linear model only in the case of CrN coating, which is the layer of NL CrN composite; other multi layers: multiCrN1, multiCrN2, multiCrN3 coatings stripping are well described by exponential regression. The exponential character in those cases of stripping can be explained by differences in the anodic stripping rate of the component layers of the coating that is determined by both process parameters and chemical composition of the coating layers.

Developed statistical models are very well adjusted to experimental data and can be used for prediction of the time period of the coating removal in a dependence on the coating thickness, and can replace the experimental try and errors method.

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Reviewer:
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Zastosowanie modelowania do prognozowania przebiegu anodowego rozpuszczania złożonych powłok CrN

Słowa kluczowe

Złożone powłoki CrN, rozpuszczanie anodowe, modelowanie statystyczne, sztuczne sieci neuronowe, model prognostyczny.

Streszczenie

W artykule przedstawiono wyniki badań eksperymentalnych anodowego procesu rozpuszczania złożonych powłok CrN. Uzyskane wyniki poddano analizie statystycznej, w rezultacie której wyznaczono modele regresyjne przebiegu procesu rozpuszczania w funkcji czasu. Ponadto wykorzystując sztuczne sieci neuronowe opracowano kompleksowy model procesu rozpuszczania anodowego. Opracowane modele umożliwiają oszacowanie przebiegu i czasu rozpuszczania w zależności od grubości powłoki oraz zastosowanych parametrów prądowych.