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Multilayer nitride coating performance optimized by an artificial neural network approach

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Abstract

One of the most important problems occurred in many industries is due to friction and wear process. Over the years, minimizing friction and controlling wear is one of the difficult tasks for the researchers. Both properties can be minimized by the application of adequate coating technology. Many coating deposition technologies have been employed to limit friction and wear but only few succeeded, those are directly affected by the nature of the material under investigation and process parameters. A suitable coating strategy varying from single layer to multilayer should be applied to the materials whose superficial properties such as low friction, improved wear resistance, and adhesion are the prime interest. Multilayer coatings possess high hardness, ductility and fracture strength compared to single layer coatings. The advantageous properties of these multilayers can be preciously tailored according to specific application. For this purpose Physical Vapour Deposition (PVD) coatings have been developed considerably due to increasing industrial demands. In the present research, friction and wear study of multilayer PVD-nitride coating deposited on tool steel by unbalanced reactive magnetron sputtering technique have been discussed. Later on an Artificial Neural Network approach was used to predict the tribological properties of multilayer nitride films. Bias voltage, total gas flow rate, lap, time, velocity and load were considered as controllable factors. The regression and performance curve analysis is used to assess the optimized outcome of deposited film properties such as friction and wear. The analyzed results shows that experimented and predicted values are in a good agreement.

Keywords: physical vapour deposition; nitride; artificial neural network; friction; wear.

1. Introduction

In a modern world, steel research have been considerably gaining importance due to its impact on day to day life and varying from biological replacements to micro industries to heavy machineries. More noble materials are identified and developed during past years either through alloying one or more than two materials. At a same time their mechanical, physical and chemical properties are triggered through newly developed coating technologies and are handful to strengthened the materials functional properties such as tribological optical, electrical and electronic. Coating tribology is constantly increasing and fulfils the demands of tool steels such as drilling, moulding, blanking etc. in an efficient way by improving their friction and wear properties [1]. Surfaces with nitride coatings on tool steels reflect high lattice energy and strong attraction towards metallic cations. These nitride coatings with addition of tungsten reduce tensile stresses in the coating zone and develop compressive stresses during PVD process. The type of coating like chemical vapour deposition (CVD) [2], physical vapour deposition (PVD) and plasma spray process [3] mainly based on the type of application required and materials chemical-physical properties.

Nowadays, in order to get the desired properties such as mechanical, physical and chemical, PVD coatings

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gaining valuable interest varying from monolayer to multilayer. These Multilayer coatings exhibit increased life with fracture resistance, adhesion, Young's modulus and improved wear resistance compared to monolayer analogues. Low friction and wear can be obtained by adding various monolayers in a thin and hard multilayer coating [4,5] with the help of PVD process. These multiple monolayer on the material surface changes the overall surface property and gives lower friction and wear values compared to initial one; which is depends on the relationship between material and corresponding nitride coating [6]. With the combined effect of these advantages it is important to evaluate its tribological properties.

Artificial neural network (ANN) is an appropriate tool to study complex processes and it obtain using only data generated by the model and an automatic training procedure. Recently, ANN have been applied to the complex problem in the field of engineering such as aerospace, manufacturing, electronics, measurement, plasma science and materials and metallurgy [7,8]. The most advantageous thing of this technique is non dependency of concerned material property [9]. ANN approach have been a focus of interest over the last decade and give powerful means for optimization, prediction, system identification, control, classification, and non-linear mappings. ANN had been shown to be a superior design method for non-linear model where the network itself is a non linear [10] system.

To the best of author's knowledge first time ever multilayer PVD tungsten nitride coatings were sputtered on chromium-molybdenum-vanadium tool steel to investigate its tribological properties. In the present research, multilayer tungsten nitride coatings were deposited through PVD-unbalanced reactive magnetron sputtering technique and their tribological performance characteristic were evaluated through Pin on Disk (POD) machine. Furthermore, parameters were optimized by ANN technique to predict the characteristics of experimented material. The approached ANN method is suitable for industrial point of view where we can predict the results for the future event. Sometimes, due to time constraint it is very hard to evaluate step by step properties of the materials, those are under investigation. To overcome from such difficulties ANN is employed to predict system behaviour at given input variables. Artificial Neural Networks have been applied to the problems those are too complex to be found such as regression analysis, prediction outcomes, and pattern recognition which makes it very flexible and powerful tool to solve real world problem with ease.

2. Materials and Methodology

High alloyed chromium-molybdenum-vanadium tool steel of hardness 52HRC with following (C: 0.39wt.%, Cr: 5.2wt.%, Mo: 1.4wt.%, Si: 1.0wt.% and V: 0.9wt.%) composition were employed as a testing substrate. An alumina ball (93% purity, Hardness: 66HRC) were used as a sliding counterpart. Sliding friction and wear test were conducted with pin on disk apparatus under constant load (5N) and fixed sliding speed (9.41cm/s) in which a vertical pin consists of alumina ball of dia 6 mm rotates against a prepared substrate.

Multilayer tungsten/tungsten nitride (W/W_2N) PVD coatings were deposited through commercial magnetron sputter unit. The specimens were mirror polished (1μm diamonds) with surface roughness value of <20nm before deposition and further ultrasonically cleaned in acetone for 5 min and dried it for approximately 20min in a vacuum dryer. The coating deposition parameters are as follows: coating temperature 300°C, the DC substrate bias voltage -75V, gas flow rate 60sccm and partial pressure of 0.093Pa respectively. Total six sub-layers of W/W_2N thin film were deposited on substrate with total coating thickness of 2.70 μm whereas, layer thickness of W/W_2N were 0.3 μm and 0.6μm respectively.

2.1. ANN training, optimization and problem formulations

The artificial neural network (ANN) is a non-linear data modelling, mapping [11] tool that has been found a wide practice in engineering sciences. ANN technique is versatile and used by many researchers for predicting the range of applications such as optical properties [12], tribological properties [13], manufacturing process control [14] and in-flight particle characteristics [3]. The network consists of input layer, hidden layer and an output layer (Fig. 1). It used to model complex relationship between a set of inputs and outputs without any prior assumption. Therefore it is considered as a non-linear statistical data modelling tool. Neuron is the basic unit in the network and they are connected to each other with weight factors [15,16]. Simulation of the values and outputting the final vector is done by feeding the data forward through each layer whereas, network training is performed by calculating the output node error and sending them back through the network to aid in weight training. During training process, in order to get desired input-output relationship a proper optimization of weight matrix is required which gives minimum error between predicted and actual output.

Multilayer perceptron (MLP) are used to understand data processed by individual neurons and their final output. Fig. 1a, only represents a two input MLP architecture, not a general one; whereas Fig. 1b represents the simple neuron model. In this architecture X_1, X_2,...X_n are input variables comprising the input layer. The first set of arrows, see Fig. 1a, represents the weights a_{01}, a_{02},...a_{0n} (input hidden neurons) connected to the middle hidden layer, consisting of many hidden neurons. These hidden neurons are not exposed to the open environment as input and output neurons. Each hidden neurons passes its weighted sum through a non linear transfer function (*f*) [17,18]. The hidden neurons output are fed to the second set of weights; b_{11}, b_{12},...b_n (output hidden neurons). By combining these neurons a single output (Z and Z') or a multiple output can be received. Furthermore, these neurons belong to input parameters and making them most powerful tool to predict system behavior.

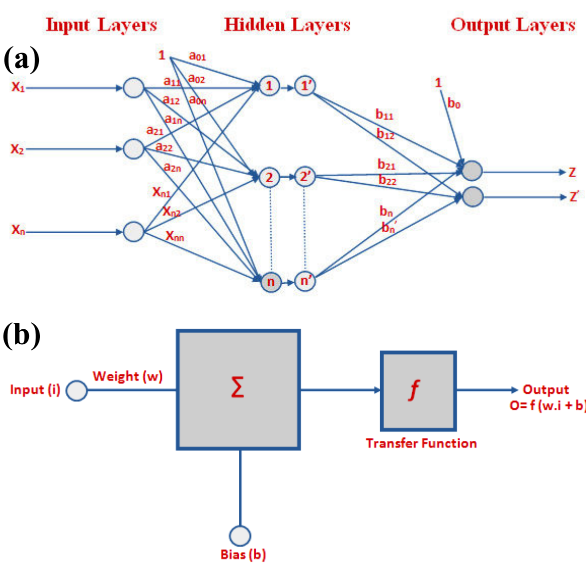

Fig. 1. Architecture of (a) multilayer perceptron network structure; and (b) a simple neuron model.

The training is based on the Levenberg–Marquardt back propagation algorithm [16] to tune the weight structure. This algorithm allows prediction of complex non-linear relationship between input-output parameters in less time. This method is actually a combination of the gradient descent method and the Gauss-Newton method and is designed to reach the second order training speed without computing the Hessian matrix. The Levenberg-Marquardt algorithm uses following approximation as shown in Eq. 1.

$$Y_{n+1} = Y_n-[J^TJ+\mu I]^{-1} J^Te \qquad (1)$$

Where, Y_n: old parameter value; Y_{n+1}: new parameter value; I: Identity matrix and μ becomes scalar. Term 'J' is the Jacobian matrix, 'T' is network target and 'e' is a vector of network errors; those are calculated through a back-propagation technique.

The back propagation procedure involves the errors output (actual node output errors) which are shared between the processing elements (input layer and hidden neurons) and the actual output of the network. In back propagation, method allows network to quick convergence on satisfactory local minima for errors which it is suited. In other words, the prediction of error is fed backward through the network in order to adjust those weights and minimize error. In order to obtain a high performance of a network, training is done by increasing the number of hidden layer. The maximum number of allowed epoch (iteration) for training cycle is limited to 1000 or early stopping to combat the problem of over-fitting. A large number of training data with cross validation and early stopping enriches the level of accuracy. In the present study data were divided into three subsets: 70%, 15%, 15% for training, validation, and testing purpose respectively. If the chosen dataset percentage increases further the total number of dataset for validation and testing purpose also increases, with this increment, enhanced value of regression coefficient can be achieved. For the present study, the coefficient of regression value is 0.999 on the chosen dataset of 70%, 15%, 15% respectively and this leads no further modification in the increased dataset percentage. The subsets have been chosen in such a way such that test set error and validation error reaches its minimum value at equal number of epochs (iterations). During the study numbers of hidden layers at initial stage were kept two and increases continuously till satisfying the network performance at its best.

The present work investigated the friction, wear resistance of multilayer tungsten nitride coating, and the potential of a neural network approach to correlate and predict their performances with testing conditions to obtain a deeper insight into their functionality while acting as a diffusion barrier. The friction and wear tests were performed with a pin on disk tribometer. Effect of various parameters like time, velocity, load, lap, bias voltage and gas flow rate is considered for the study purpose. The range of these parameters is presented in Table 1.

Table 1. Parameters used for ANN formulation.

S. No.	Significant Parameters	Parameters Range
1.	Time (s)	0-1000
2.	Velocity (cm/s)	9.41
3.	Load (N)	5
4.	Lap	1-5000
5.	Bias Voltage (V)	-75
6.	Gas Flow Rate (sccm)	60

ANN structure of the input-output parameters are dipicted in Fig. 2. These parameters are affecting the overall coating properties. As time progresses friction and wear of the tested materials changes due to formation of debris between the contacting material during continuous sliding. Other parameters such as lap, load and velocity also affects the friction and wear characteristics. Bias voltage and gas flow rate are coating deposition parameters which should be selected properly that they can not induce any tensile stresses with in the coating surface and material interfaces.

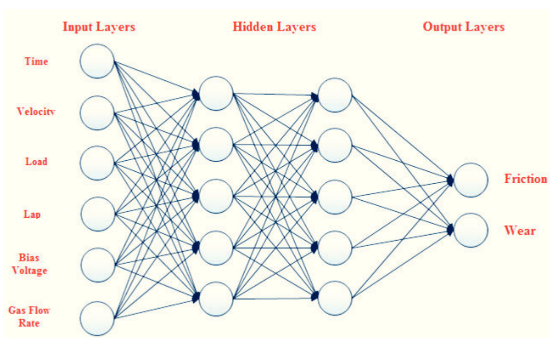

Fig. 2. ANN structure of input-output database.

3. Results and Discussion

Friction and wear results of W/W_2N coated substarte were shown in Fig. 3a, while slide against alumina ball at room temperature. After several attempt of sliding the obtained average friction cefficient was 0.22. The friction coefficient first increased rapidly with increase in time/running cycle due to sudden rise in pressure or applied load which shears the protective layer from the substrate surfaces. As the time progresses, due to sliding, surface film starts forming a protective layer and tries to stabilize the friction coefficient values. These protective layers act as a diffusion barrier and further suppress the formation of transferred layer. Generally, this state arises due to physiochemical process takes place between substrate and alumina balls. At the same time rise in temperature gradually decreases friction coefficient to its minimum value because of the stable and protective oxides films developed during the process which really benefits the surfaces from wear out. Wear rate were calculated by measuring the wear track dia of a substrate at several locations after sliding test and average value has been considered for the calculations. The calculated wear rate value for multilayer nitride coating was very less and optimum for any tribological application. Multilayer coating protects the surface from wear out because it remained intact at higher loads whereas thin coatings might be worn through quickly with minimum effect.

Scanning Electron Microscopy (SEM) microstructure of wear surface after sliding test were depicted in Fig. 3b and shows that after running in period W/W_2N has clear microstructure. With the addition of nitride its physical properties are enhanced but the disadvantage of these coatings are their high oxidation rates after 500°C. Wear track of W/W_2N coatings shows rare material displacement to the outer surface of the steel substrate. W/W_2N coating showed typical columnar morphology (Fig. 3c) developed in the films by surface diffusion process and influenced by geometrical shadowing effects. The Energy Dispersive X-ray Spectroscopy (EDS), Fig. 3d, analysis mainly revealed constituent material for W/W_2N multilayer coating. The EDS analysis showed a few iron content, which means that material transfer takes place between substrate and ball which further extending wear rate. Abrasion and oxidation were the primary wear mechanisms. Wear rate generally depends on these transferred materials during the sliding process due to occurrence of materials oxidation which further fractured and turn into debris.

In order to predict the tribological properties of nitride coatings as a function of six different parameters, the ANN modeling was done in MATLAB version R2012a using inbuilt Neural Network Toolbox. In the present study, ANN predictive model is developed using 41 data sets. The input processing parameters for ANN with experimental results were listed in Table 2. Experiments were carried out several times to match the repeatability of obtained results and at last average data have been taken in to account for the relative error calculation. The network was trained with an input-output database and ten hidden neurons (preliminary stage) in order to predict the friction and wear behavior of multilayer coating.

To achieve the highest accuracy of the network number of neurons was increased further in step wise ranging from ten to twenty.

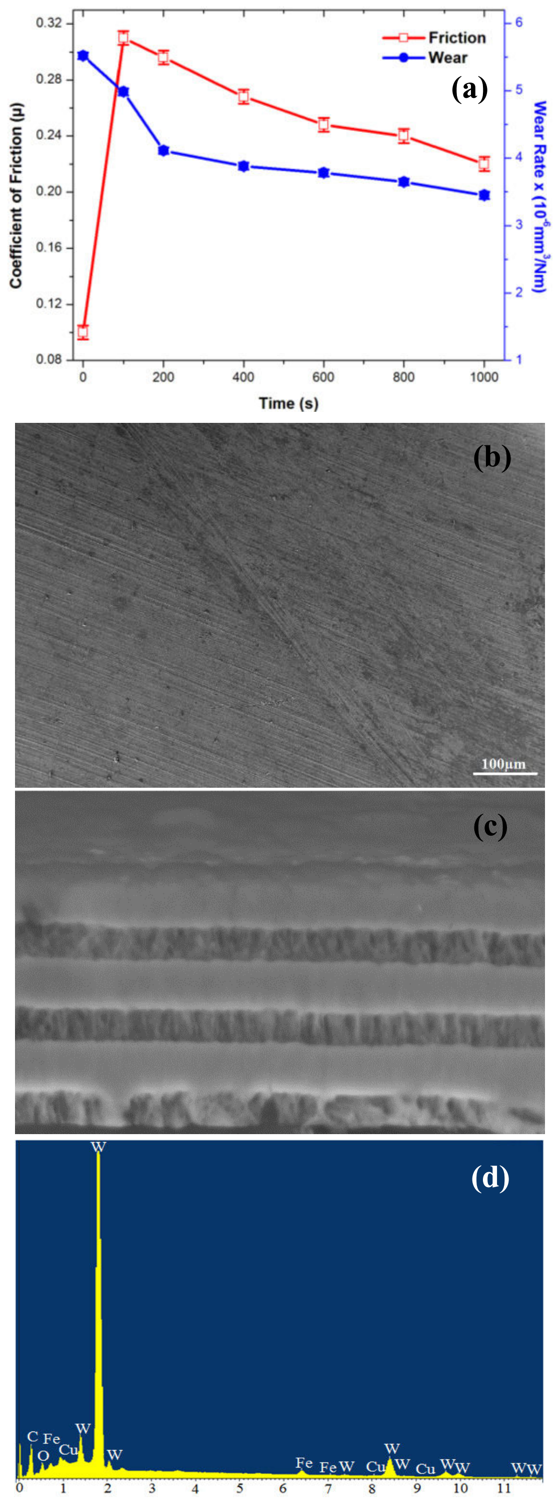

Fig. 3. (a) Average friction and wear performance of nitride coating; (b) SEM images after sliding test; (c) cross-section SEM micrograph of coated surface before sliding test; and (d) EDS spectra of chemical composition present in the multilayer nitride structure.

Highest number of neurons gives most accurate desired value and less relative error compared to lower ones.

All the ANN predicted result was latterly compared with the actual experimented values and with their respective error percentage. Relative error was measured using absolute error (experimented – ANN predicted) divided by the original experimented value.

The friction and wear behaviour of steel substrate showed how well the predicted values follow the experimental one and it has been seen that both experimented and predicted values are close to each other with minimum number of relative error which is an acceptable result for any ANN network. Wear phenomena is quite different on the surfaces and it basically based on the nature of the material, physical-chemical process between the ball and substrate. Although the friction performances of substrate material were measured lower but due to the intrinsic interaction and surface reaction of two materials differs the wear characteristic. As in case of tested W/W_2N coating friction value was little higher but achieved wear value was lower. The lowest average relative error for friction and wear were 0.90%, 0.87% respectively. All the relative errors to the multilayer coating measurement implying that the ANN method adopted within this database is acceptable.

The overall ANN predicted values showed similar trend towards the experimented values with respect to their input processing parameter. For the above analysis, correlation coefficients were $R^2 = 0.99976$ and 0.99998 for friction, wear analysis respectively which is shown in Fig. 4a,b. Levenberg-Marquardt algorithm [16] was successfully deployed for validation, training process and optimization with improving the process input-output relationship. The network was trained several times to get the optimum solution with different number of hidden layers. The trained ANN network was successful in predicting the average friction and wear behaviour with minimum error of less than 10% with sufficient accuracy and such results were considered as satisfactory. Some of the ANN predicted values shows slight scatter plot in comparison to other plots. The main reason behind this may be due to the fact that input information database are not sufficient enough for the network to correlate between time and their friction or wear characteristic. Gholamreza Khalaj [19] has also predicted the layer thickness of pre-nitrided steels by an ANN technique. The observed R^2 values are very close to the experimented ones which signify its true function of the trained network. ANN approach was an excellent in terms of simplifying the complex industrial processes, leading to save time and overall involved cost. In another work by Khalaj and Pouraliakbar [20] showed an alternative approach to consider process parameters for predicting the desired property of coatings. The predicted result agrees with the suitability of proposed model where duplex treated ceramic coating values were very close to the experimental ones.

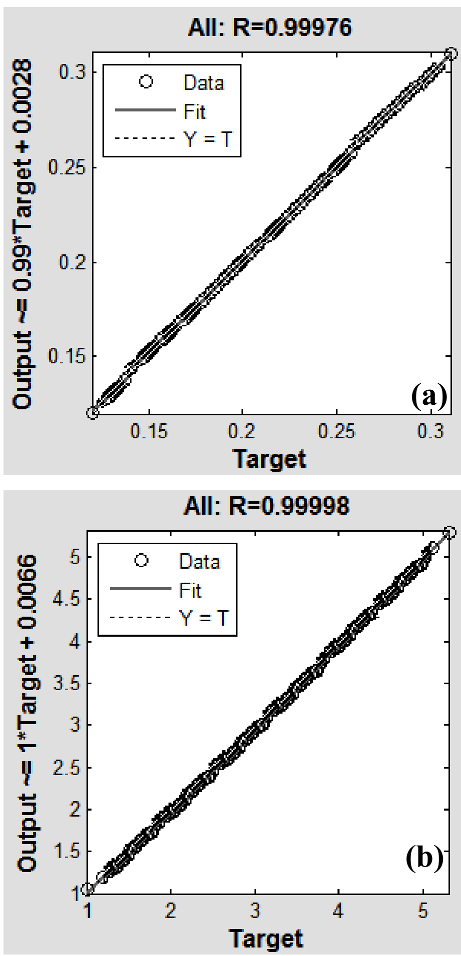

Fig. 4. Overall predictive performances of ANN model (a) friction; and (b) wear.

A plot of the training errors, validation errors, and test errors for overall friction coefficient and wear rate is shown in Fig. 5a,b respectively. The obtained results were reasonable because of the following reasons: the final mean square error (MSE) was small; the test and validations set error had similar characteristics; there is no significant over fitting occurred by iterations (at the point of best validation performance). In order to carry out training for the following ANN architecture, the process variables were assigned intermediate values, the confirmation of test and the measured responses were presented in Table 3. The predicted ANNs friction coefficient and wear rate values were compared with the actual ones and a good agreement was observed.

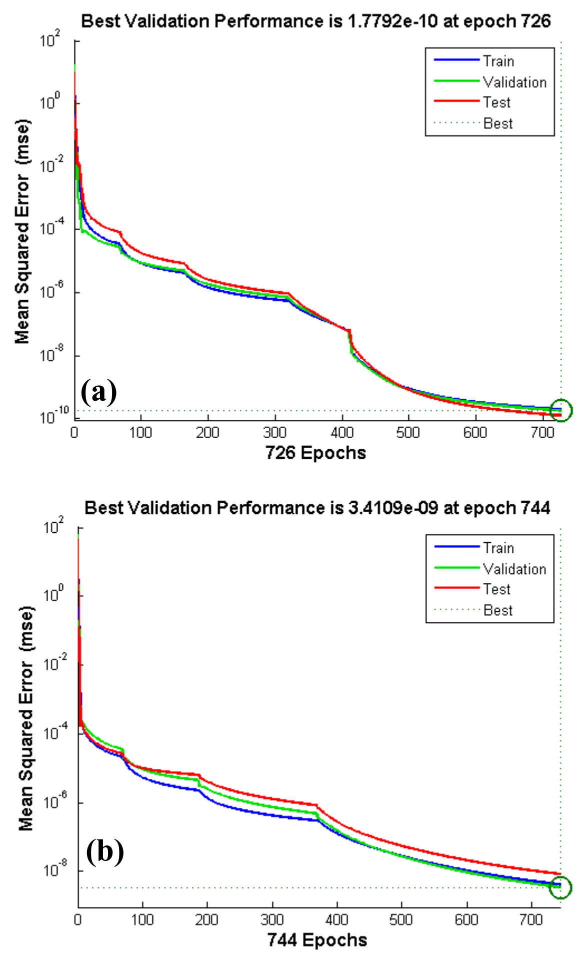

Fig. 5. Overall ANN (a) friction and (b) wear performance of multilayer coating at room temperature.

Table 2. Input processing parameters for ANN architecture and experimented output results corresponds to input parameters.

Time (s)	Velocity (cm/s)	Load (N)	Laps	Bias Voltage (V)	Gas Flow Rate (sccm)	Experimented Output (Friction)	Predicted Output (Fiction)	Experimented output: Wear ((Wear rate x ($10^{-6}mm^3/Nm$))	Predicted Output: Wear ((Wear rate x ($10^{-6}mm^3/Nm$))
0	9.41	5	1	-75	60	0.100	0.097	5.520	5.517
25	9.41	5	125	-75	60	0.115	0.113	5.333	5.329
50	9.41	5	250	-75	60	0.266	0.262	5.105	5.101
75	9.41	5	375	-75	60	0.312	0.311	5.000	4.999
100	9.41	5	500	-75	60	0.310	0.307	4.983	4.982
125	9.41	5	625	-75	60	0.305	0.302	4.745	4.744
150	9.41	5	750	-75	60	0.298	0.299	4.655	4.654
175	9.41	5	875	-75	60	0.297	0.296	4.213	4.211
200	9.41	5	1000	-75	60	0.296	0.295	4.111	4.106
225	9.41	5	1125	-75	60	0.291	0.288	4.003	4.001
250	9.41	5	1250	-75	60	0.288	0.285	3.999	3.998
275	9.41	5	1375	-75	60	0.277	0.274	3.997	3.996
300	9.41	5	1500	-75	60	0.275	0.274	3.990	3.988
325	9.41	5	1625	-75	60	0.273	0.272	3.886	3.886
350	9.41	5	1750	-75	60	0.271	0.269	3.883	3.882
375	9.41	5	1875	-75	60	0.269	0.268	3.882	3.880
400	9.41	5	2000	-75	60	0.268	0.266	3.881	3.879
425	9.41	5	2125	-75	60	0.267	0.265	3.804	3.802
450	9.41	5	2250	-75	60	0.263	0.261	3.800	3.799
475	9.41	5	2375	-75	60	0.260	0.258	3.795	3.793
500	9.41	5	2500	-75	60	0.253	0.252	3.792	3.792
525	9.41	5	2625	-75	60	0.251	0.248	3.796	3.792
550	9.41	5	2750	-75	60	0.250	0.247	3.791	3.788
575	9.41	5	2875	-75	60	0.249	0.244	3.787	3.785
600	9.41	5	3000	-75	60	0.248	0.246	3.780	3.779
625	9.41	5	3125	-75	60	0.247	0.242	3.776	3.775
650	9.41	5	3250	-75	60	0.246	0.242	3.770	3.699
675	9.41	5	3375	-75	60	0.245	0.241	3.699	3.695
700	9.41	5	3500	-75	60	0.244	0.242	3.693	3.691
725	9.41	5	3625	-75	60	0.243	0.240	3.687	3.686
750	9.41	5	3750	-75	60	0.242	0.241	3.681	3.679
775	9.41	5	3875	-75	60	0.241	0.239	3.652	3.651
800	9.41	5	4000	-75	60	0.240	0.238	3.646	3.643
825	9.41	5	4125	-75	60	0.239	0.237	3.600	3.558
850	9.41	5	4250	-75	60	0.238	0.236	3.591	3.589
875	9.41	5	4375	-75	60	0.237	0.235	3.584	3.583
900	9.41	5	4500	-75	60	0.231	0.228	3.566	3.563
925	9.41	5	4625	-75	60	0.230	0.229	3.558	3.556
950	9.41	5	4750	-75	60	0.228	0.226	3.540	3.539
975	9.41	5	4875	-75	60	0.223	0.220	3.473	3.470
1000	9.41	5	5000	-75	60	0.220	0.218	3.450	3.420

The effect of input process parameters on the average friction and wear behaviour were depicted in Table 3.

Table 3. Average experimented and predicted values for tool steel.

Coating Material		Average Friction (μ)	Average Wear (mm^3/Nm)10^{-6}
	Experimented	0.220	3.450
W/W_2N	ANN Predicted	0.218	3.420
	% Error	0.900	0.870

4. Conclusion

In this study, the influence of multilayer tungsten nitride coating on tool steel was investigated. It was shown that multilayer structure influences initial material transfer and the coefficient of friction both at its best. Test results revealed that multilayer structure reduces friction and wear significantly. An artificial neural network (ANN) approach was succesfully applied for the multilayer coating. The network was trained and optimized to predict friction and wear behaviour with non-linear data relationship. The trained network was able to corelate the effect of processing input-output parameters and provide the optimzed result with respect to the desired coating properties. All the predicted results are found in a good agreement with the experimeantal database.

Acknowledgements

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