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Predicting the Corrosion Rate of Al and Mg Alloys Coated by Plasma the Spraying Method with Machine Learning

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ABSTRACT: Developing technology has increased the need for materials that are more economical in terms of cost and more reliable in terms of strength, chemical and physical properties in all industrial areas. This has necessitated the development of new materials or the improvement of existing material properties. Surface coating methods are used to improve existing material properties. In this study, Al and Mg alloys, which are considered as an alternative to steel material in terms of being lightweight materials, were coated with Al_2O_3 and TiO_2 at different rates by plasma spraying method, and the corrosion behaviors of the coatings in different environments were predicted using machine learning methods. AA7075 and AZ91 non-metal materials were chosen as the substrate for the study. Different ratios of Al_2O_3 and TiO_2 ceramic materials were coated on the substrates. To determine the corrosion resistance of the coated samples, corrosion experiments were carried out in 3.5% NaCl and 0.3M H_2SO_4 environments. Using the experimental results, corrosion rate values were estimated using machine learning algorithms such as XGBoost, Random Forest (RF) and artificial neural networks (ANN) methods, depending on the substrate material, corrosive environment and coating rates. At the end of the study, corrosion rate values were estimated with low error rates and the best estimate was obtained with the XGBoost method (0.9968 R^2 value).

Keywords: Coating, AZ91,AA7075,Seramic Materials, Machine Learning

1. INTRODUCTION

Al is a metal with a density of 2.7 g/cm^3, good corrosion resistance and easy production. The strength/density ratio is approximately 3 times higher than structural steels. Due to its non-toxic structure, it is a material group suitable for use in sectors such as the beverage industry and medical

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pharmaceuticals. Al is a preferable engineering material due to its high electrical conductivity, thermal conductivity, magnetic and high reflection properties, as well as its low cost. Due to all these features, it is widely preferred in many areas, especially defense, space, automotive industry (Ashby,2004; Picas et al.,2005; Gibbons et al.,2006). Metal materials are used in the manufacturing of many machine elements such as construction, oil, heat exchangers and valves, and there are problems with resistance to corrosion and abrasion (Bolelli,2009; He et al.2007).

Mg began to take its place as the lightest material group among green metallic materials in the 21st century. It is a group of non-metallic materials used in the aviation and automotive industries due to their properties such as low density (1.73 g/cm^3), high specific strength and hardness, excellent castability and good magnetic shielding performance (Kojima et al., 2001; Mordike et al., 2001; Rotshtein et al.,2004). Although it has many superior properties, its low corrosion resistance has limited the use of Mg and its alloys (Shi et al., 2005, Song Gand StJohn,2004).

Alloying and surface coating methods are used to increase the corrosion and wear resistance of Al and Mg materials, which are candidates to replace metal materials because they have many superior properties. In order to improve the wear and corrosion resistance of light metal alloys, thermal spray surface coating methods such as atmospheric plasma spray (APS), high velocity oxygen fuel (HVOF) and wire arc are used (Ernst P and Fletcher K., 2011). In thermal spray coating methods, metals and ceramic materials as well as hard metals are widely used as coating materials. Atmospheric plasma spraying method is one of the methods used to make ceramic coatings due to its high melting temperatures (Toma et al.,2010). Al_2O_3 is a brittle ceramic material, and with the addition of TiO_2, the hardness value decreases and the toughness value increases (Šuopys et al., preprint; Basha et al.,2020). In addition, TiO_2, which has a low melting point, increases the coating performance by lowering the melting point when added to Al_2O_3. For this reason, instead of using only Al_2O_3 for surface coating, coatings obtained by adding TiO_2 into Al_2O_3 are used and studies have focused on this type of coatings. Ya-Li et al. (2007) in their study, applied Al_2O_3 ceramic coating process on Mg alloy using plasma coating and laser melting plasma spraying methods. They investigated the wear and corrosion resistance of the coatings. At the end of the study, the authors determined that there was a 3-fold increase in wear resistance compared to uncoated materials and a 1-fold increase compared to coatings using the plasma coating method. In addition, the authors determined that there was a 5-fold increase in corrosion resistance in uncoated samples and a 3-fold increase compared to the plasma coating method (Ya-li et al., 2007). In their study, Morks and Akimato (2008) examined the effect of nozzle diameter on the quality of Al_2O_3/TiO_2 coated materials by plasma spraying method. At the end of the study, a denser coating was obtained with a 7.5 mm nozzle diameter compared to 8 mm, Micro Vickers hardness values decreased as the nozzle diameter increased, and greater wear resistance was obtained with a lower nozzle diameter (Morks et al., 2008).In their study, Islak and Buytoz (2011) coated Al_2O_3/TiO_2 materials on AISI 304 steel. Authors who examined the coatings concluded that as the amount of Al_2O_3/TiO_2 increased, the porous structure decreased and the hardness increased four to five times compared to the substrate (Islak and Buytoz (2011). Jia et al. (2015) coated Al_2O_3/TiO_2 coating materials with different ratios of TiO_2 added on AA6061 aluminum material using the plasma spraying method and examined the corrosion resistance of the coatings. At the end of the study, the authors concluded that as the TiO_2 ratio increases, corrosion resistance increases and thermal insulation properties decrease (Jia et al. 2015). Basha et al. (2020) discussed the Al_2O_3-TiO_2 coating process using the atmospheric plasma method in their study. At the end of the study, they determined that the coatings obtained from agglomerated powders of nano-sized alumina-titania showed better wear resistance than the coatings of conventional powders (Basha et al. 2020). In their study, Bakhsheshi-Rod et al. (2020) applied nanostructured titania (TiO2, n-TO) and nanostructured alumina alumina-titania (Al_2O_3-13% TiO2; n-ATo) coatings on the AA6061 substrate. At the end of the study, they determined that n-TO coatings had superior hardness and higher wear resistance than uncoated and n-ATO coatings. In addition, electrochemical examinations of the coatings were made and it was determined that n-ATO coatings have 50% lower corrosion resistance due to their looser structure than n-TO coatings (Bakhsheshi-Rod et al. 2020). In their study, Harju et al. (2007) examined the effects of surface properties, surface stress and phase inhomogeneity of TiO_2, Al_2O_3based coatings (Harju et al. 2007). Toma et al. (2009) examined the corrosion behavior of materials coated with different ratios of Al2 Al_2O_3/TiO_2. At the end of the study, the authors determined that there was an increase in corrosion resistance with the addition of TiO_2 (Toma et al. (2009). Michalak et al. (2020) in their study, they discussed the determination of tribological properties of samples coated with 3, 13 and 40% TiO_2. In the study, they concluded that when the TiO_2 ratio increased, the pure Al_2O_3 phases decreased and the TiO_2 phases increased. They also determined that the best tribological performance was obtained in coatings with 13% TiO_2 added, where a decrease in hardness occurred as TiO_2 increased (Michalak et al. 2020).

When the literature studies were examined, it was determined that many studies were carried out to determine the behavior of different corrosive environments and different coatings. The most important problem experienced in experimental studies is the losses in terms of time and cost due to the large number of experiments. In order to eliminate this problem, analytical methods are used to determine the optimum parameters (Giard and Karlsson, 2021; Altinkok, and Koker,2004). In recent years, the data-driven machine learning approach has been used in optimizing the performance of materials and designing new materials (Xinming et al.,2023; Lei et al., 2022; Cheng et al.,2021; Dey et al.,2016). In their studies, the authors used machine learning (ML) and ANN methods to predict the mechanical properties of materials. Dey et al. discussed the prediction of mechanical properties of ageable wrought alloys using the ANN method (Dey et al.,2016). Again, Giard and Karlsson used the ANN method in their study to predict the mechanical properties of duplex stainless steel (Giard and Karlsson, 2021). El Rehim et al. aimed to determine the change in hardness of AZ91 magnesium alloy using the ANN method (El Rehim et al.,2020). In addition, different statistical methods have been used to develop prediction models using real corrosion data for corrosion rate prediction and to develop prediction models using real corrosion data. Some of the methods used in the studies are linear regression (LR) (Al-Fakih et al.,2016); It can be expressed as multivariate regression (Vel´azquez et al. 2009). However, when the basic assumptions in statistical methods are not fully met, there is difficulty in making accurate predictions. For this reason, methods such as machine learning have been used. For example, Liu et al. (2012) proposed a support vector machine (SVM) regression model with a radial basis function (RBF) kernel for oil boat line corrosion prediction in their study. They used the particle swarm optimization (PSO) algorithm in SVM regression (Liu et al.,2012). Chern-Tong and Aziz (2016) used the CMARGA model to create an optimal decision tree model using a genetic algorithm (Chern-Tong and Aziz 2016). In his study, developed machine learning approaches (component analysis (PCA) and reinforcement machine (GBM)) to predict corrosion defect depth growth in pipelines. Among these approaches, it was determined that the PCA-GBM model achieved 3.52-5.32 times more accuracy than the others (Ossoi, 2019). Ren et al. estimated the internal control rate of underground natural gas pipelines using backpropagation artificial neural networks.In the study, it was determined that the performance of the prediction made using the model was high (Ren et al.,2012).

When the studies are examined, the use of machine learning in the development of new materials for material selection, performance evaluation and lifetime prediction is increasing (Kılıç et al.,2020; Yan et al.,2020; Liu et al.,2022). Machine learning is a particularly suitable method for modeling the relationship between nonlinear material behavior and complex influencing factors (Tian et al.,2017; Shi et al., 2018). For this, Random Forest (RF) (Pei et al., 2020) ; Popular machine learning methods such as adaptive boosting (AD) (Schmidt et al.,2017) , light gradient boosting machine (LGB) (Behara et al; 2021). Gradient boosting (XGBoost) (Fan et al.,2018) and artificial neural network (ANN) (Kumari and Tiyyagura; 2018) are used.ANN model is a comprehensive database; It is suitable for conditions where the data set is incomplete and complex (Hongyu et al.,2023). If the data set is small, the RF model gives the best prediction result (Shi et al., 2018). The RF model is used in many areas such as image recognition (Yang et al.,20179) and corrosion [(Hou et al.,2018) because it is easily applicable, has a simple structure, is suitable for non-linear data, and is suitable for a small number of data.

In this study, ANN; RF and XGBoost regression methods were used to predict the corrosion behavior of AA7075 and AZ91 alloys coated with different ratios of Al_2O_3 and TiO2 in different corrosive environments. In addition, determining the most appropriate method by using more than one method is another aim of the study.

2. MATERIALS AND METHODS

2.1 Experimental Procedure

AA7075 Al alloy and AZ91 Mg alloy were selected as the substrate materials in the study. The chemical compositions of the selected materials are given in Table 1.

Table 1. Chemical properties of the substrate materials used in the study

AZ91	Alloy element	Al	Zn	Mn	Fe	Si	Cu
	%	8.8	0.61	0.18	0.02	0.02	0.005
AA7075	Alloy element	Zn	Mg	Cu	Fe	Si	Mn
	%	6.2	2	1.7	0.5	0.4	0.1

Al_2O_3 and TiO_2 ceramic materials were chosen as coating materials for the study. Al_2O_3-TiO_2 powders with an average size of 40-80 μm were used and these powders were mixed homogeneously. Before the coating process, surface preparation processes were applied to the substrates and the substrates were prepared for coating. For this purpose, the substrate materials are first sandblasted and then ultrasonically cleaned. Then, by applying Ni-Cr primer material to the substrates, the bonding ability of the coating is increased. Atmospheric plasma spraying method was used in the study. The plasma spray coating process parameters used in the study are given in Table 2. Coated samples are given in Figure 1.

Table 2. Process parameters used in the study

Coating Material	Spraying distance (cm)	Ampere (A)	Voltage (V)	Powder feed rate (g/saat)	Argon /Hidrogen (l/dak)
%40 TiO_2	6	530	55-58	1,38	40-42,5/59-63,72
%13 TiO_2	7	550	58-60	1,84	40,59-42,5/56,64
%3 TiO_2	8	570	60-62	1,61	40,59-43,42/61,36-64,66
%100 Al_2O_3	9	580	69	1,73	40-42,5/59-63,72

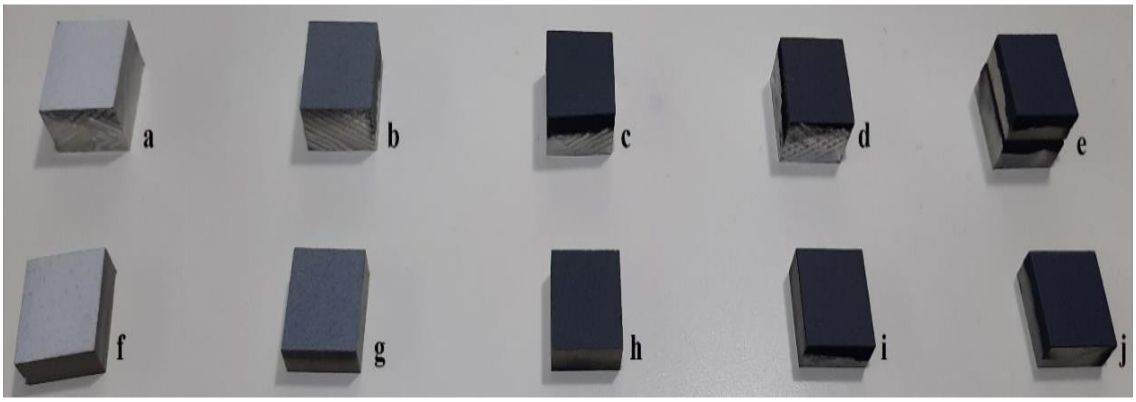

Figure 1. Coated samples by using Atmospheric plasma spraying method (a) AA7075 Saf Al_2O_3 (b) AA7075 %97 Al_2O_3 + %3 TiO_2 (c) AA7075 %87 Al_2O_3 + %13 TiO_2 (d) AA7075 %60 Al_2O_3 + %40 TiO_2 (e) AA7075 Saf TiO_2 (f) AZ91 Saf Al_2O_3 (g) AZ91 %97 Al_2O_3 + %3 TiO_2 (h) AZ91%87 Al_2O_3 + %13 TiO_2 (i) AZ91 %60 Al_2O_3 + %40 TiO_2 (j) AZ91 Saf TiO_2

Corrosion tests were carried out to determine the corrosion resistance of the coatings. Gamry Reference 600 potentiostat/galvonostat device was used for the experiments. Since the study aimed to determine the effects of different corrosive environments, 3.5% NaCl and 0.3 M H_2SO_4 corrosive liquids were used. Before corrosion experiments, the samples were cleaned ultrasonically for 15 minutes with acetone, 15 minutes with ethanol and double distilled water for 15 minutes, and dried in an oven at 50^oC for 1 hour. Experiments were repeated 3 times. Before the corrosion tests, the samples were kept in a corrosive environment for 1 hour to ensure that the system reached equilibrium. The general view of the electrodes and corrosion cell used for corrosion experiments is given in Figure 2. In the study, SEM examinations were also performed before and after corrosion.

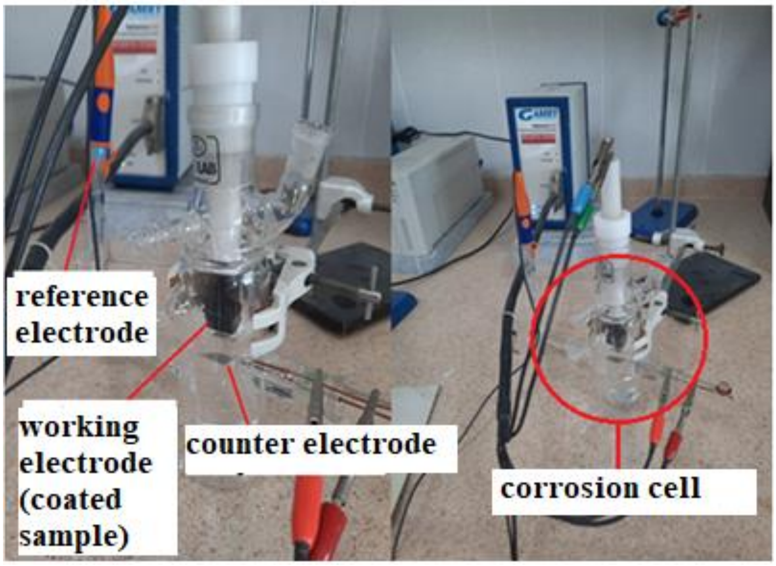

Figure 2. The general view of the electrodes and corrosion cell used for corrosion experiments

2.2 Machine Learning Methods

In this study, corrosion rate values of different non-metal materials were tried to be determined by using different coating rates and different corrosive media. For this purpose, the study tried to estimate corrosion rate values using XGBoost regression, Artificial Neural Network (ANN) and random Forest regression methods. Artificial neural networks (ANN) are expressed as an artificial intelligence method in which the concept of learning is modeled in computer systems (Han et al.2023). Artificial neural networks are widely used in many fields such as financial affairs and computer image processing. In addition, artificial neural networks are frequently used in regression and classification processes (Aslan, 2022). In addition to ANN, regression methods are also used to solve prediction problems. XGBoost algorithm is one of the decision tree-based ensemble learning-based machine learning algorithms that uses the gradient boosting framework (Morde, 2023; Kurt et al., 2020) . In this algorithm, it is used in structured or tabular data sets in classification and regression problem (Brownlee,2020). This method has advantages such as calculation speed and regularization, such as focusing on the performance of the model (Verma et al.,2018). Random Forest algorithm is among the commonly used methods among community classification algorithms. This algorithm is a type of classifier in tree structure and consists of multiple trees. When predicting the class of new data, predictions are made from each of the multiple trees created in the training phase. The predictions made are called votes, and the class with the most votes is determined as the predicted value of the new data (Bilgin et al.,2018; Breiman, and Cutler; 2020).

In forecasting techniques, it is important to evaluate the performance of the technique. The standard deviation of the forecast errors was determined as the root mean square error (RMSE) by Chai and Draxler (Chai and Draxler, 2014). A low RMSE value is a desirable situation. How close the regression line is to the actual data is indicated by R2 (Cameron and Windmeijer,1997). The important thing here is that the R2 value must be between 0-1 and close to 1.

3. RESULTS AND DISCUSSION

3.1 Experimental Results

In the study, AA7075 aluminum alloy and AZ91 magnesium alloy were coated with different ratios of Al_2O_3 and TiO_2 ceramic materials using the atmospheric plasma method. The corrosion resistance of the coatings made after the coating process was determined. SEM examinations of the coated samples were carried out before and after the corrosion tests. The absence of errors during coating can be expressed as an indication that the coating regime between the substrate and coating material is regular. SEM images of the samples before corrosion tests are given in Figure 3. When the Figure 3 is examined, it is determined that the amount of pores occurring in TiO_2-doped coatings on both AA7075 substrate and AZ91 substrate is higher than Al_2O_3 coatings. This is due to the presence of pores formed during the nucleation of TiO_2.

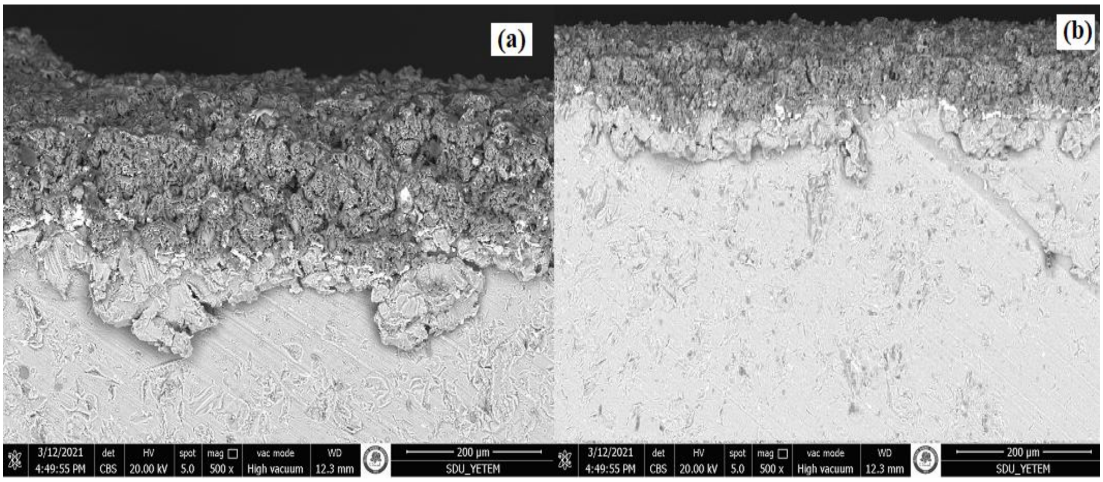

Figure 3. SEM images of the samples before corrosion tests (a) %40 TiO_2 and %60 Al_2O_3 coated AA7075 substrate (b) %40 TiO_2 and %60 Al_2O_3 coated AZ91 substrate

In the study, SEM examinations were also carried out after the corrosion test.SEM examinations of AA7075 substrate after the corrosion tests in 3.5%NaCl and 0,3M H_2SO_4 enviroments in Figure 4; SEM examinations of AZ91 substrate after the corrosion tests in 3.5%NaCl and 0,3M H_2SO_4 enviroments are given in Figure 5. Al added into Mg eliminates the destructive effect of aggressive Cl- ions with its oxide film on the surface. For this reason, with the coating of Al_2O_3 on AZ91 material, the corrosion resistance (1,044 mpy) of AZ91 material increased and the situation is supported in SEM images. The addition of TiO_2 into Al_2O_3 caused the formation of cracks and pits in the samples, and these formations are observed in SEM images. When SEM images of corrosion experiments performed in 0.3M H_2SO_4 environment were examined, a decrease in corrosion resistance occurred with the increase in TiO_2 ratio, similar to 3.5%NaCl environment. After the corrosion tests of the coatings made on AA7075 substrate in 3.5% NaCl solution, the highest corrosion resistance value was obtained in 100% Al_2O_3 (3,486 mpy) coated samples due to the absence of defects such as porosity, and this is supported by SEM examinations. In 0.3M H_2SO_4 solution, the highest corrosion resistance was obtained in 100%TiO_2 coated samples (). This is due to the fact that TiO_2, which has a low melting temperature, melts and disperses into Al_2O_3, increasing the effectiveness of the coating (Jia et al.,2015). This situation is supported by SEM images.

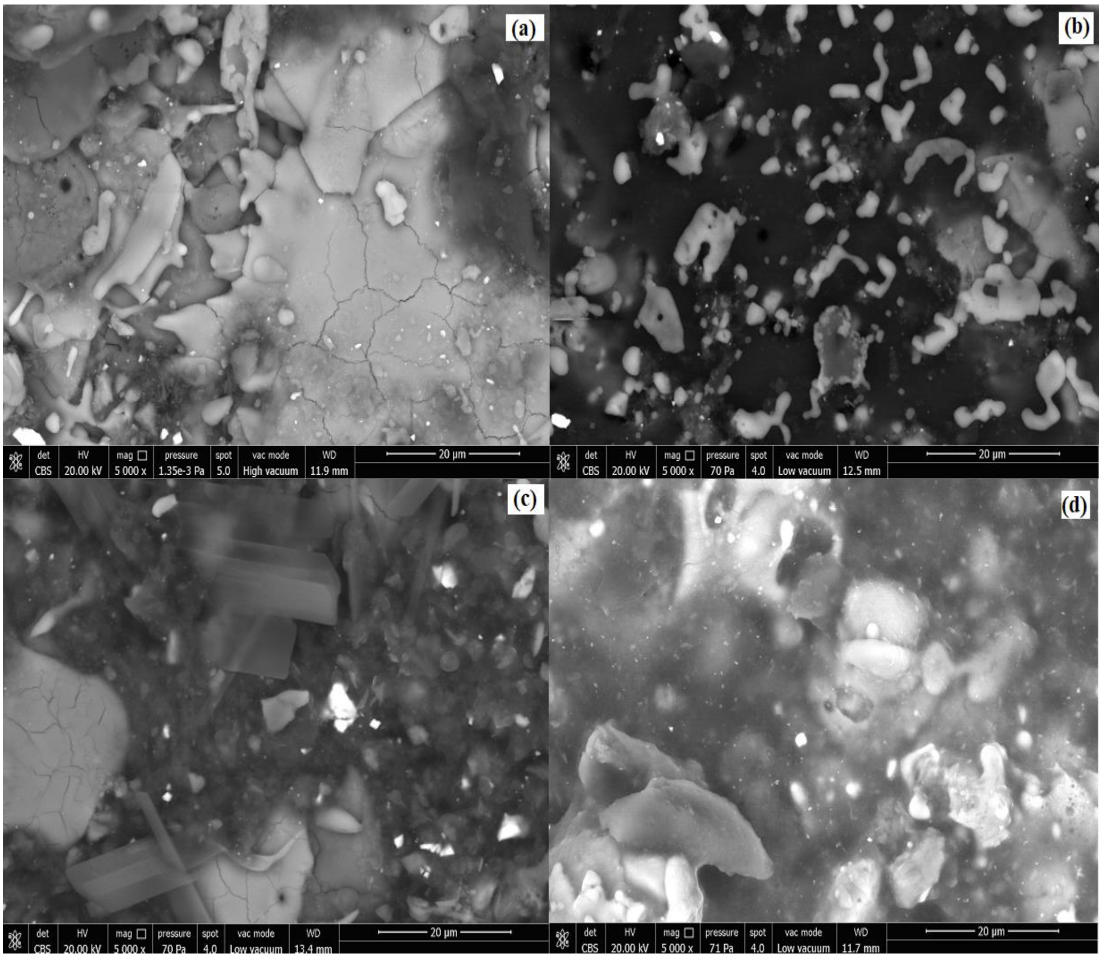

Figure 4. SEM examinations of AA7075 substrate after the corrosion tests in (a) %40 TiO2 coated samples in %3,5 NaCl enviroment (b) %100 Al_2O_3 coated samples in %3,5 NaCl enviroment (c) %40 TiO2 coated samples in 0,3M H_2SO_4 enviroment (d) %100 Al_2O_3 coated samples in 0,3M H_2SO_4 enviroment

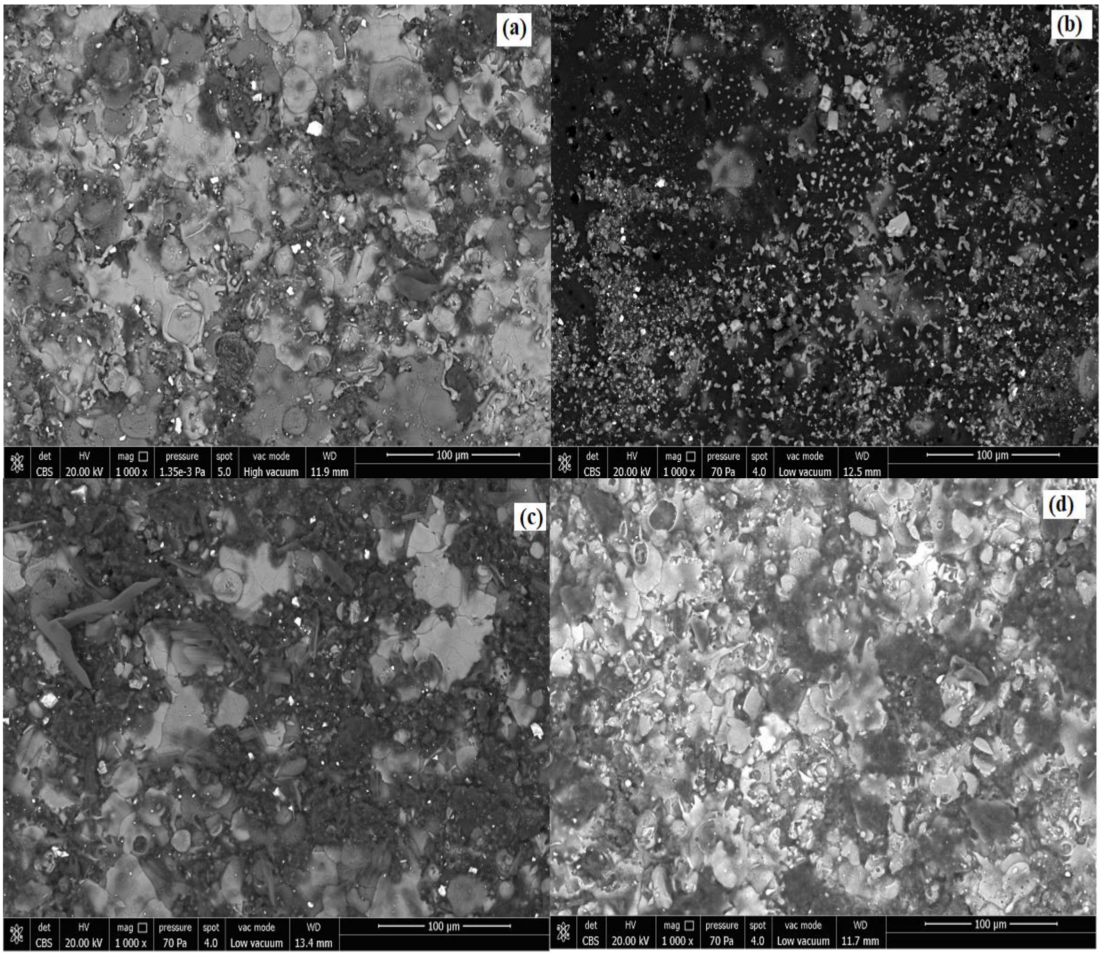

Figure 5. SEM examinations of AZ91 substrate after the corrosion tests in (a) %40 TiO2 coated samples in %3,5 NaCl enviroment (b) %100 Al_2O_3 coated samples in %3,5 NaCl enviroment (c) %40 TiO2 coated samples in 0,3M H_2SO_4 enviroment (d) %100 Al_2O_3 coated samples in 0,3M H_2SO_4 enviroment

3.2 Dataset and Implementation Details

In this study, boosting (XGboost), tree-baggaging (randomforest) and neural network (ANN) structures were used. 80% of the 24 data was used for training and 20% for testing.It consists of corrosion rate values depending on the different corrosive environment and coating material for the study. Accordingly, the values of the substrate (I1), corrosive environment (I2) and coating material (I3) were taken as input, and the corrosion rate value (Q1) was taken as output. Python was used for this study.

3.3 Test and Evaluation

The important point in machine learning methods is to determine the features in the data set very well. In the study, firstly, training and test datasets were created. The developed model can then be used to predict the properties of the studied coating ratios. In this way, determination of optimum coating rates can be achieved quickly. In this study, 5 real test data and corrosion rate values predicted by machine learning models were compared (Table 3). When the results were examined, the best results were obtained as XG Boost Regression, Random Forest Regression and ANN, respectively.

Table 3. Test and predicted corrosion rate values

Experimental Results				XG Boost Regression	Random Forest	ANN
Substrate	Coating material	Corrosive enviroment	Real Output	Predict	Predict	Predict
AZ91	Uncoated	NaCl	826,9522	834,400	734,0888	833,8445
AA7075	%40 TiO_2	NaCl	89,2100	100,6060	123,0795	68,0182
AZ91	%3 TiO_2	NaCl	75,5805	75,7700	43,7842	1,7842
AZ91	%40 TiO_2	H_2SO_4	130,2622	97,6800	169,9654	3,4454
AZ91	%100 Al_2O_3	NaCl	45,9537	59,500	34,2997	1,1855

In addition, R^2, RMJE and MAE values were calculated for all 3 methods and are given in Table 4.

Table 4. Algorithm performances according to estimation results

Algoritma	R^2	RMJE	MAE
XG Boost Regression	0,9968	0,0203	0,0156
Random Forest Regression	0,9614	0,0709	0,0605
ANN	0,9584	0,0736	0,0023

When Table 4 is examined, the best results were obtained in the XG Boost Regression algorithm with 0.9968 R^2 value, 0.0203 RMJE and 0.0156 MAE values. This algorithm is followed by Random Forest and ANN.

When the results were examined, machine learning algorithms to obtain the corrosion rate value depending on different corrosive environment, substrate and coating material ratios gave results close to the real test data. Accordingly, time and cost savings can be achieved by using machine learning methods.

4. CONCLUSION

In the study, AA7075 aluminum alloy and AZ91 magnesium alloy were coated with different ratios of Al_2O_3 and TiO_2 ceramic materials using the atmospheric plasma method. It was observed that the amount of pores occurring in TiO2-doped coatings on both AA7075 substrate and AZ91 substrate is higher than Al_2O_3 coatings after coating process. coating of Al_2O_3 on AZ91 material, the corrosion resistance of AZ91 material increased both in %3,5NaCl and 0,3M H_2SO_4 enviroment. the coatings made on AA7075 substrate in 3.5% NaCl solution, the highest corrosion resistance value was obtained in 100% Al_2O_3 coated samples. In 0.3M H_2SO_4 solution, the highest corrosion resistance was obtained in 100%TiO_2 coated samples.

In this study, it is aimed to predict corrosion rate values using machine learning algorithms depending on different substrate material, corrosive environment and coating material ratios. At the end of the study, it was determined that corrosion rate values could be estimated with low error rates within the acceptable range. Thus, it has been concluded that estimating each time without experimenting can provide significant savings in terms of time and cost.

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6. CONFLICT OF INTEREST

The authors reported no conflict of interest.

7. AUTHOR CONTRIBUTION

Hüseyin ÖZKAVAK and Recai Fatih TUNAY determining the concept and design process of the research and research management, data collection and analysis, data analysis and interpretation of results'.

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