The Relation between Deposited Weight and Quality of Coating in EPD Method Derived by Genetic programming

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ABSTRACT

In this work, the relation between deposited weight and the quality of electrophoretically deposited coating has been derived using genetic programming method. Although, the accumulated mass is thicken by time, its quality varies at different times of coating procedure. Three different suspensions i.e. Mullite, SiC and Mullite/SiC were stabilized in ethanol medium and the suspended particles were electrophoretically deposited on C-C composite at several different times. The results of SEM micrographs show that the quality of coating rises by time and after some time it starts to drop for all three suspensions. The results of Zeta potential of suspension after different times of coating that is derived by pH measurement, illustrate the same pattern. There is a maximum for zeta potential after 150 sec of deposition process. Accordingly, the quality of coating rises as a result of enhancement of Zeta potential in suspensions. Last but not least, there is a relation between deposition time and quality of coating which is mathematically modeled using genetic programming method. In this case, the root of multiplication of Z-w and w-t differential equations could show the optimum time of deposition process.
1. Introduction

1.1. Zeta potential

In colloid suspensions, electro-kinetic potential is described by Zeta potential. Zeta potential theoretically is the electric potential difference between interfacial double layer (DL) and bulk fluid away from the interface. It means that the zeta potential is the potential difference between the dispersion medium and the dispersed particle [1–7]. There is a slipping plane around the suspended particles. The location and electrical charge of slipping layer is two main factors affecting the amount of zeta potential. There are also other potentials in the double layer like Stern potential or electric surface potential. The main differences among these potentials are the different locations of charged layer [8–15]. Zeta potential is usually used for the study of the stability of colloidal dispersions. The magnitude of the zeta potential indicates the amount of electrostatic repulsion between suspended particles which is a key factor affecting the stability of suspensions. Higher attractive force while the repulsion is low due to the reduction of zeta potential is the reason for flocculation of suspension. It means that high zeta potential suspensions are stabilized while low zeta potential suspensions are flocculated [16–20]. Zeta potential of suspensions is practically measured by applying an electric field across the dispersion. Under a constant external electrical field, particles suspended in the dispersion will move with a velocity proportional to the magnitude of the zeta potential [21].

1.2. Modeling by genetic programming

Genetic programming (GP) is a method of artificial intelligence considered as a member of soft calculation method. Methods like genetic programming have been frequently used because of the nonlinear properties, inherent uncertainty and complexity existed in physical phenomena [22]. Genetic programming is an automatic technique of programming and solution of problem could be calculated via computer programs. Evolutionary algorithms (such as GP) that have been evaluated based on the Darwin’s theory, have the modeling ability of nonlinear processes [23]. GP can automatically choose the variables that are more effective than the others. It is the advantage of genetic programming in compare with other modeling methods. This advantage doesn’t exist in other modeling methods such as neural networks [24]. The main disadvantage of genetic programming that has bothered the scientists of Artificial intelligence is bloating of program. Bloating is the excessive growth of codes along the evolutionary process which causes the stagnation of process. Bloating control could be done via different methods. One of the newest of them that has been used in this investigation is ‘waiting room’ method [23]. For the initiation of work with GP, function set and terminal set must be determined. Function set is used in computer programs. Also terminal set includes of variables, constants and null-arity functions [25]. After recognition of function and terminal set, that are the building blocks of GP, parameters such as number of initial population and kind of genetic operators must be defined. The most important part of GP application is the recognition of different, in use parameters. These parameters are determined by experience or even trial and error methods. For an example, estimation of initial population and number of produced generation depend on the
characterization of computer system that is used [23–26]. Although GP has been used for prediction and optimization of processes in engineering fields, it is not conventionally used in materials science and coating processes. By the way, there are several investigations in the field of mechanical properties of materials [27–30].

In the present investigation, the relation between deposited weight and the quality of electrophoretically deposited coating will be discussed using genetic programming method. Accordingly, zeta potential changes were calculated and it is used for optimization by GP. The aim of this investigation is to find the optimum time of deposition for production of the most qualified surface.

2. Experimental and modeling procedure

2.1. Experimental procedure

Suspensions were prepared by adding 4 g SiC, Mullite or Mullite/SiC powder to 50 ml Ethanol and 20 ml/l triethylamine (TEA) as dispersant. For suitable dispersion of suspensions, magnetic stirrer (Alfa D-500, Iran) and ultrasonic bath (hielscher model UP 100 H, Ultrasound Technology, Germany) were utilized.

Surface preparation of C-C composites were done by grinding up to 800 grit using SiC papers, cleaning in acetone and ethanol using an ultrasonic bath, washing with distilled water and drying in air.

An electrophoretic cell is containing a 150 ml beaker, C-C composite electrode as electrodes with surface area of 10 × 10 mm, and a fixer for fixing the location of electrodes with 1 cm distance from each other. During electrophoretic deposition, constant voltages of 60 V were applied by a power supply (Mastech, DC power supply HY30001E, 9225). Electric current during deposition process were measured by means of Escort, 3146A Dual Display Multi meter. The samples were dried in desiccators for 24 h, after deposition process to be ready for weight measurements, and SEM micrographs. During the deposition process pH of suspensions were measured using pH meter, Hanna Instruments, USA.

2.2. Modeling procedure

Weight and Zeta potential vs. time curves of SiC and Mullite suspensions were set as input and output for GP Toolbox of MATLAB® software. Obtained model was tested by the data of SiC/Mullite suspension. Rmse and R-square tests were used for the evaluation of fitness of the calculated model. As a conclusion, suitable equation of controlled and optimum coating was obtained.

It must be noticed that the computer program was adjusted as it is seen in table 1 while both training and test stages. Furthermore, modeling was done using a computer system, Pentum 4, RAM 4 GB and CPU2.66 GHz.
Table 1
GP adjusted value for computer program while training and test.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Population Size</td>
<td>30</td>
</tr>
<tr>
<td>Mutation Rate</td>
<td>0.3</td>
</tr>
<tr>
<td>Crossover Rate</td>
<td>1</td>
</tr>
<tr>
<td>Maximum Initial Tree Size</td>
<td>15</td>
</tr>
<tr>
<td>Maximum Tree Size</td>
<td>45</td>
</tr>
</tbody>
</table>

3. Results and discussion

Fig. 1 illustrates weight vs. time curves for SiC, Mullite and SiC/Mullite suspensions. As it is illustrated, gradient of curves decreases gradually.

![Fig. 1. Weight vs. time of coating for various suspensions.](image)

Fig. 2 shows SEM micrographs of coated samples in different times of deposition process for Mullite suspension. As it is illustrated, the quality of coating differs in different times of coating procedure. It is noticeable that SEM micrographs of SiC and SiC/Mullite coatings are similar to those shown in fig. 2. The quality of coatings is related to the amount of Zeta potential, directly. The higher stability of suspension will be a reason for more quality of coating. Accordingly, the pH of suspensions was measured during deposition process and upon the variation of pH, Zeta potential of suspensions were calculated as their values shown in fig. 3.

Fig. 3 depicts Zeta potential vs. time for SiC, Mullite and SiC/Mullite suspensions. As it is illustrated, the maximal point with the highest value of Zeta potential in all three curves shows the highest stability of suspension. The other points have gotten lower Zeta potential which is the lower stability of suspensions.
Fig. 2. SEM micrographs of SiC coating on c-c composite in different times of coating.

Fig. 3. Zeta Potential vs. time of coating for various suspensions.
According to fig. 1, coating weight increases exponentially with time of deposition. Equation of curves illustrated in fig. 2 was fitted and determined as

\[ w = w_M(1 - e^{-kt}) \]  

(1)

Where \( w \) is the coated weight at time \( t \), \( w_M \) is the maximum weight deposable on surface and \( k \) is a constant that probably depends on the voltage of electrophoretic deposition process, composition of suspension and so on.

Fig. 1 and Eq. (1) are similar to charging curve and equation of a reservoir in a RC circuit. Charging of a reservoir vs. time determines as

\[ v = v_T(1 - e^{-\frac{1}{RC}t}) \]  

(2)

By comparison between parameters in Eq. (1) and (2), \( k \) corresponds to \( 1/RC \). \( R \) and \( C \) are defined by

\[ R = \frac{\rho L}{A} \]  

(3)

and

\[ C = \frac{\varepsilon A}{L} \]  

(4)

Where \( \rho \) is the specific resistivity, \( L \) is the distance between electrodes, \( A \) is the cross section of anode and \( \varepsilon \) is the permeability of suspension.

If behavior of electrophoretic deposition is considered as a RC circuit, electrophoretic cell could have characterizations of resistor and reservoir, simultaneously. As a result \( k \) could be defined by

\[ k = \frac{1}{RC} = \frac{1}{\rho \varepsilon} \]  

(5)

Both \( \rho \) and \( \varepsilon \) are the inherent parameters of suspension [26]. Therefore, \( k \) constant depends on the composition and temperature of suspension as same as the voltage of electrophoretic deposition process. The deposition process finally could be defined by,

\[ w = w_M(1 - e^{-\frac{t}{\rho \varepsilon}}) \]  

(6)

It was explained earlier in section (2.2) that weight and Zeta potential vs. time curves of SiC and Mullite suspensions were set as input and output for GP Toolbox of MATLAB® software. Among the suggested models (equations), the one which possessed the following characterizations, was chosen as the best model. Best fitness and lack of complication in model. Relation between \( Z(t) \) and \( W(t) \) that has been modeled via genetic programming is defined by,

\[ Z(t) = \text{atan}\left( -\frac{C_1}{\cos(\sqrt{W(t)})} \right) + \sin\left( \sqrt{W(t)} \right) + \left( \sqrt{C_2W(t)} \right)^{0.3333} \]  

(5)

Where \( Z(t) \) is the Zeta potential of suspension at time \( t \), \( W(t) \) is the weight of powder deposited on the sample at time \( t \) and \( C_1 \) as same as \( C_2 \) are the constants that were calculated as,
\[
\begin{align*}
C_1 &= 0.1899 \\
C_2 &= 2.950501
\end{align*}
\] (6)

These two constants could be related to the inherent characterization of suspension. By the way, nature of modeling is black box and discussion about these constants is not possible. For the reorganization of these constants, more experimental data, and white box modeling would be required.

Fig. 4 shows the output of training process of model. In fig. 4-A the dispersion of simulated $Z$ toward the observed $Z$ has been depicted and in fig. 4-B bar chart of observed and simulated $Z$-t have been compared.

Fig. 5 shows the output of model’s testing procedure that its input was set by the data of fig. 1-B. Scatter plot of $Z$ and bar chart of $Z$-t have been illustrated by fig. 5-A and fig. 5-B respectively.

Fig. 4 and 5 illustrate that the observed data have been adapted with the simulated data; hence in this case, calculated model is completely suitable. Fig. 6 illustrates parse tree of model depicted by genetic programming software.

![Fig. 4. Output of training procedure of model.](image-url)
Fig. 5. Output of testing procedure of model.

Fig. 6. Tree schematic of model calculated by genetic programming.
For the evaluation of goodness of fitting, Rmse and R-square as two fitness measures were used. Evaluation criterions were measured by,

\[
Rmse = \left[ \frac{1}{n} \sum_{i=1}^{n} ((R_{rs} - R_{r0})_i)^2 \right]^{1/2}
\]

and

\[
R - Square = 1 - \frac{\sum (R_{rs} - R_{r0})^2}{\sum (R_{r0} - \bar{R}_{r0})^2}
\]

Among the 48 introduced models by genetic programming, the one which had the least difference between R-square values of training and test processes was chosen as the best model. Fitness of model in training and test stages has been shown using the evaluated criterions in Table 2.

**Table 2**
The results of model evaluation.

<table>
<thead>
<tr>
<th>Level</th>
<th>Rmse</th>
<th>R-square</th>
</tr>
</thead>
<tbody>
<tr>
<td>Train</td>
<td>0.14</td>
<td>0.97</td>
</tr>
<tr>
<td>Test</td>
<td>0.59</td>
<td>0.80</td>
</tr>
</tbody>
</table>

The method used in this investigation (GP) is a simulation Method. Minimization of error and maximization of R-square are the targets of simulation process. Therefore simulation seems to could be determined as an optimization process. Upon this sight of view and to be sure that the calculated model is a global optimum so it isn’t local, nonlinear regression was used. For this reason, \(C_1\) and \(C_2\) constants were calculated again via nonlinear regression and these two (constants calculated by GP and nonlinear regression) were compared using R-square. If model with GP constants have better fitness than nonlinear regression constants then GP will be the global optimum [21]. Hence, \(C_1\) and \(C_2\) constants were calculated via nonlinear regression, using MATLAB® software and via ‘nlinfit’ command. These could be illustrated by,

\[
\begin{cases}
C_1 = 0.1414 \\
C_2 = 2.9490
\end{cases}
\]

Table 3 compares the R-square value for Models calculated by GP and nonlinear regression.

**Table 3**
The R-square value for GP and Non Linear Regression Models.

<table>
<thead>
<tr>
<th>GP Model</th>
<th>Train</th>
<th>Test</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.975</td>
<td></td>
<td>0.816</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Non Linear Regression Model</th>
</tr>
</thead>
<tbody>
<tr>
<td>Train</td>
</tr>
<tr>
<td>0.976</td>
</tr>
</tbody>
</table>
Upon the results illustrated in table 3, it is clear that calculated model in GP is a global optimum and there isn’t any model better than the one determined in this investigation.

By multiplying eq. (4) and eq. (5), there will be an equation for curves depicted in fig. 3. Therefore predictive control of coating procedure could be achieved without doing any experimental procedure. Optimum time for calculation of maximum Z can be defined by,

\[
\frac{d(Z)}{d(W)} \times \frac{d(W)}{d(t)} = 0
\]

(10)

4. Conclusion

- The results of SEM micrographs show that the quality of coating rises by time and after some time it starts to drop for all three suspensions.

- The results of Zeta potential of suspension after different times of coating illustrate the same pattern with the quality of coating. It means that the quality of coating rises as a result of enhancement of Zeta potential in suspensions.

- There is a relation between deposition time and quality of coating which is mathematically modeled using genetic programming method.

- Genetic programming is a suitable modeling tool. Correlation coefficient of test procedure upper than 0.8 is a reason for this claim.

References


[23] Koza JR. Genetic programming: A paradigm for genetically breeding populations of computer programs to solve problems. Stanford University, Department of Computer Science Stanford, CA; 1990.


