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Predictive modeling of boriding kinetics on armco iron using linear regression approach

ABSTRACT

Tools inevitably degrade over time, with lifetimesvarying based on material and usage. To mitigate this, thermochemical treatments such as boriding are employed. This process forms a boride layer with favorable tribological and mechanical properties. Generally, depending on the processing conditions, the layer may result in either a mono-phased Fe_2B or a bi-phased FeB/Fe_2B. This study examines a mono-phased Fe_2B layer formed by powder pack boriding of Armco Iron. The objective is to predict the thickness of this layer using a linear approach with minimal variables, and to compare the predictions with a more complex approach requiring additional parameters. Results show that both models yield similar predictive capabilities within the available interval, validating the linear model. However, the linear approach shows divergence when applied beyond the investigated interval, indicating limitations in its generalizability.

Keywords: *Boride, boriding, layer, linear, regression, thickness*

1. INTRODUCTION

Due to the importance of tribological aspects of materials in different fields, hardening processes became an unavoidable domain to research at every level giving rise to various processes that followed the industrial revolution. Every year, billions of dollars are lost in damages from components failures; thus, over the development and by doing research studies about surface treatments on tools and parts, their wear resistance increased and, in parallel, leading to the enhancement in their lifetime[1].

One of the hardening techniquesare nitriding, carburizing, chromium plating, thermal spraying, chemical vapor deposition (CVD) or physical vapor deposition (PVD) and others [2,3]. Our area of interest is boron diffusion, also known as boridingor boronizing, where significant andadvantageous mechanical properties are gained after diffusing boron to the surface of the material [4].

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Differentiation in the nomenclature of the process occurs based on the manner in which the thermochemical treatment is carried out, whether in solid, liquid, or gaseous media. For instance, there are variations such as electrochemical boriding (EB), gas boriding (GP), plasma gas boriding (PGB), paste boriding (PB), plasma paste boriding (PPB), powder pack boriding process (PPBP), and ultra-fast boriding (UFB) [5,6]. All of these applications expanded onto numerous domains; automotive, fittings, power plant, stamping, milling and crushing technologies, nuclear, textile, pump constructions, oil and gas industries, agricultural machinery, renewable energies, biomedical applications and many more [7-9]. In all these domains, different materials ranging from cast iron, cast steel and most steel grades to nickel-based alloys except silicon and aluminum were subjected to the process where each material had different complex intermetallic compounds created between the borides and the elements included in each material; like iron, chromium, nickel, vanadium, cobalt, molybdenum, tungsten or titanium [5,10].

Upon diffusing boron atoms at high temperatures, a boride layer is formed being either monophase Fe_2B or multiphase FeB/Fe_2B [11,12]. This layer has tribological properties [13] that manifest in the increased resistance to wear from abrasion [14], acids and alkalis [15], corrosion [16] and adhesion [17]. Depending on the materials used and the other aspects, the process may increase the material's hardness to more or less than 20 GPa [10].

From a cost point of view, modelling the kinetics of boride layers by characterizing the required variables to predict its thickness with the least experimentations needed became essential due to the expensiveness of processes. Thus, researchers developed different methods to predict the boride layer thickness. The integral method and the artificial neural network modelingare among the many techniques available [18-21].

This work investigates the kinetics of a monophaseFe_2B layer formed after boridingof Armco Iron by simulating its thickness using a linear regression method. The method is based on a multilinear regression approach done with the least variables possible, temperature and time. After getting the results, they are analogized with another method used in aprevious research, the integral method [18]. Consequently, it shall be seen that, if not taking other variables, such as concentrations, the diffusion coefficient, incubation time, or others, into account leads to unsatisfactory estimations. The study aims to establish whether a more straightforward linear approach can be comparable to a complex one that needs more variables in the kinetic studies of Fe_2B layers.

2. METHODS

2.1. Integral method

When the boriding is processed, boron atoms are diffused into the substrate until saturation which takes a period called incubation time t_0 [12]. After that, the formation of continuous and compact Fe_2B boride layer begins having a certain thickness u over a treatment time t.

Boron concentrations C_{Fe_2B} vary across the boride layer, as illustrated schematically in Figure 1. C_{ads}denotes the absorbed boron concentration at the material surface [22]. $C_0$is the solubility limit of boron within the material substrate, which can be neglected[23,24].

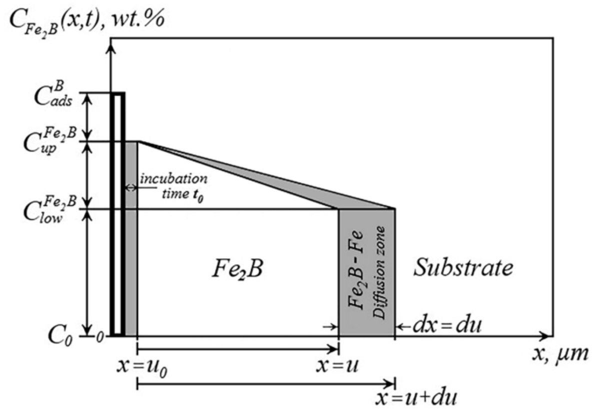

Figure 1. Boron concentration distribution across the Fe2B layer

To establish this method, certain assumptions, found in Figure 1, are used.

- Initial condition:

$$C_{Fe_2B}(t=0, x>0) = C_0$$

- Boundary conditions:

$$C_{Fe_2B}(x=0, t=t_0) = C_{up}^{Fe_2B} \quad \text{for} \quad C_{ads} > C_{low}^{Fe_2B}$$

$$C_{Fe_2B}(x=u, t=t) = C_{low}^{Fe_2B} \quad \text{for} \quad C_{ads} < C_{low}^{Fe_2B}$$

The Fick's second law given by Eq. (1) is a partial differential equation (PDE) that elucidates how diffusion causes the boron concentration to change with respect to time t knowing that the diffusion coefficient $D_B^{Fe_2B}$is dependent on the boriding temperature.

$$\frac{\partial C_{Fe_2B}}{\partial t} = D_B^{Fe_2B} \frac{\partial^2 C_{Fe_2B}}{\partial x^2} \qquad (1)$$

Integrating the second Fick's law between 0 and u(t), Eq. (2) is obtained.

$$\frac{u(t)^2}{2}\frac{da(t)}{dt} + a(t)u(t)\frac{du(t)}{dt} + \frac{u(t)^3}{3}\frac{db(t)}{dt} + b(t)u(t)^2\frac{du(t)}{dt} = 2D_B^{Fe_2B} b(t)u(t) \qquad (2)$$

Using Goodman's heat balance integral method (HBIM) [25], the expression of the boron concentration profile throughout the boride layer is formulatedby Eq. (3).

$$C_{Fe_2B}(x,t) = C_{low}^{Fe_2B} + a(t)(u(t)-x) + b(t)(u(t)-x)^2 \mathit{for} 0 \le x \le u \qquad (3)$$

Where the boron concentration inside the boride layer depends on three time-dependent variables$a(t)$, $b(t)$ and $u(t)$.

Via the second boundary condition, a first algebraic equation, Eq. (4), is deduced.

$$\left(C_{up}^{Fe_2B} - C_{low}^{Fe_2B}\right) = a(t)u(t) + b(t)u(t)^2 \quad (4)$$

Based on the principle of mass conservation at the (Fe_2B/substrate) interface,Eq. (5),is used to formulate another algebraic equation, Eq. (6).

$$W \frac{\partial x}{\partial t}\bigg|_{x=u} = -D_B^{Fe_2B} \frac{\partial C_{Fe_2B}}{\partial x}\bigg|_{x=u} \quad (5)$$

with

$$W = \frac{C_{up}^{Fe_2B} + C_{low}^{Fe_2B} - 2C_0}{2}$$

$$\left(C_{up}^{Fe_2B} + C_{low}^{Fe_2B}\right)b(t) = a(t)^2 \quad (6)$$

Accordingly, Eqs. (2), (4) and (6) depend on$a(t)$, $b(t)$ and $u(t)$, and form a system of differential algebraic equations (DAE) that can be solved either analytically [26-28] or numerically [29].

Analytically, aparabolic equation is set as in Eq. (7), which can be rewritten as Eq. (8). These equations form the solution of the system with Eqs. (9) and (10).

$$u(t) = k\sqrt{t - t_0} \quad (7)$$

$$u(t) = k_1\sqrt{t} \quad (8)$$

where k the parabolic growth constant of the Fe_2B layer, and k_1 another parabolic growth constant.

$$a(t) = \frac{\alpha}{u(t)} \quad (9)$$

$$b(t) = \frac{\beta}{u(t)^2} \quad (10)$$

By substituting the solutions, Eqs. (9) and (10) into Eqs. (4) and (6), two algebraic equations are obtained, Eqs. (11) and (12).

$$\left(C_{up}^{Fe_2B} - C_{low}^{Fe_2B}\right) = \alpha + \beta \quad (11)$$

$$\left(C_{up}^{Fe_2B} + C_{low}^{Fe_2B}\right)\beta = \alpha^2 \quad (12)$$

After solving the algebraic system formed by Eqs. (11) and (12), the two unknown parameters α and β are determined as follows:

$$\alpha = \left(C_{up}^{Fe_2B} + C_{low}^{Fe_2B}\right)\left(-1 + \sqrt{1 + 4\left(\frac{C_{up}^{Fe_2B} - C_{low}^{Fe_2B}}{C_{up}^{Fe_2B} + C_{low}^{Fe_2B}}\right)}\right)\Big/2$$

$$\beta = \left(C_{up}^{Fe_2B} + C_{low}^{Fe_2B}\right)\left(2 + 4\left(\frac{C_{up}^{Fe_2B} - C_{low}^{Fe_2B}}{C_{up}^{Fe_2B} + C_{low}^{Fe_2B}}\right) - 2\sqrt{1 + 4\left(\frac{C_{up}^{Fe_2B} - C_{low}^{Fe_2B}}{C_{up}^{Fe_2B} + C_{low}^{Fe_2B}}\right)}\right)\Big/4$$

Using all that, and by substituting Eq. (7) or Eq. (8), along with Eqs. (9) and (10) into Eq. (2), and deriving with respect to time t,the diffusion coefficient is deduced, Eq. (13).

$$D_B^{Fe_2B} = \eta k^2 \quad (13)$$

with

$$\eta = \frac{1}{12} + \frac{1}{16}\frac{\left(C_{up}^{Fe_2B} + C_{low}^{Fe_2B}\right)}{\left(C_{up}^{Fe_2B} - C_{low}^{Fe_2B}\right)}\left(1 + \sqrt{1 + 4\left(\frac{C_{up}^{Fe_2B} - C_{low}^{Fe_2B}}{C_{up}^{Fe_2B} + C_{low}^{Fe_2B}}\right)}\right)$$

Lastly, Eq. (14) is attained, helping estimate the boride layer thickness after determining the diffusion coefficient $D_B^{Fe_2B}$ [18].

$$u_l(t) = \sqrt{\frac{D_B^{Fe_2B}}{\eta} t} \quad (14)$$

2.2. Linear method

The linear method used here is a statistical approach based on a multiple linear regression (MLR). It uses two or more independent variables to predict the outcome of a dependent variable. It is usually formulated by Eq. (15) [30].

$$y = \beta_0 + \beta_1 x_1 + \beta_2 x_2 + \cdots + \beta_p x_p + \varepsilon \quad (15)$$

Where

y the dependent variable,

$x_1 x_2 \ldots x_p$the independent variables,

$\beta_0 \beta_1 \beta_2 \ldots \beta_p$regression coefficients,

ε the model's error term (residual).

This method is based on certain assumptions [31,32]:

- Dependent and independent variables have a linear relationship.
- Weak correlation between independent variables.

- Fixed residual variance.
- Observations are independent.
- Normality multivariate.

In some cases, like this studied case, variables have nonlinear or highly correlated relationships, which does not satisfy the earlier assumptions. To overcomethis issue, an interaction term is injected between those variables [31].

Taking all that into account, the linear method's equation ends up as in Eq. (16).

$$u_L(T,t) = \beta_0 + \beta_1 T + \beta_2 t + \beta_3 Tt \qquad (16)$$

where u_L the dependent boride layer thickness variable,

$T$and t the independent variables, temperature and time,

$\beta_0\beta_1\beta_2\beta_3$the regression coefficients.

Eq. (16) is scripted into a matrixial form as in Eq. (17), for computing purposes.

$$U = X\beta \qquad (17)$$

where: $U = [u_1 u_2 \ \dots \ u_n]'$

$$X = \begin{bmatrix} 1 & T_1 t_1 T_1 t_1 \\ 1 & T_2 t_2 T_2 t_2 \\ & \vdots \\ 1 & T_n t_n T_n t_n \end{bmatrix}$$

$$\beta = [\beta_0 \beta_1 \beta_2 \beta_3]'$$

while$n$ represents the number of data sets.

The resolution of that matrixial system via a computing platform or mathematically [33] gives the regression coefficients β_0, β_1, β_2 and β_3, characterizing the equation (16) that estimates the final boride layer thickness u_L.

3. BORIDING EXPERIMENT

The boriding treatment [34] was carried on samples of Armco iron which has insignificant amounts of alloying elements, 0.02 carbon, 0.2 manganese, 0.015 phosphorus, 0.015 sulfur, 0.007 nitrogen, and 0.06 copper.

The samples were cubically sectioned with one centimeter on each side, polished, ultrasonically cleaned, and dried. Then, they were embedded in a stainless steel AISI304L cylindrical container where they are englobed by a Durboridpowder containing boron carbide, silicon carbide, and potassium fluoroborate. For each treatment, the container was placed in a furnace under a pure argon atmosphere at temperatures of 850, 900; 950, and 1000 $°C$, each with diverse treatment times, 2, 4, 6, and 8 h. After which, it is left to slowly cool down to room temperature.

After boriding, the treated samples were cross-sectioned using a cutting precision machine and the Fe_2Blayers with a saw-tooth morphology were observed by optical microscopy[34].

These observed Fe_2B boride layers' thicknesses were defined as the average value of the long boride teeth and estimated with fifty measurements collected in different sections. The experimental data [34] presented in Figure 2 was used to model the kinetics of Fe_2B layers using thelinear regression approach.

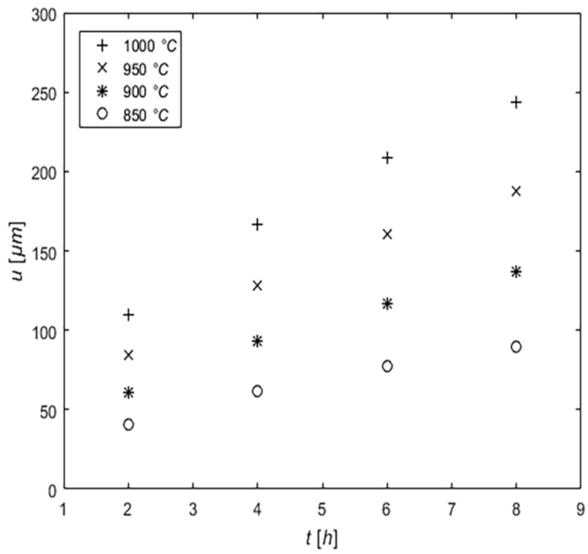

Figure 2.Variation of Fe_2B layer thickness with the treatment time for increasing temperatures

3. RESULTS AND DISCUSSIONS

The Fe_2B layer thicknesses are predicted using Eq. (16), which was attained using the multiple linear regressionu_L with the coefficients given in Table 1.

Table 1.Coefficients of the multiple linear regression approach

Coefficients	β_0	β_1	β_2	β_3
Values	-228.4195	0.2985	-71.6751	$9.3737\ 10^{-2}$

These linear regression approach'spredictions are plotted in Figure 3parallelly with a previous research's results that used the integral method referred as $u_I$obtained from Eq. (14) [18].

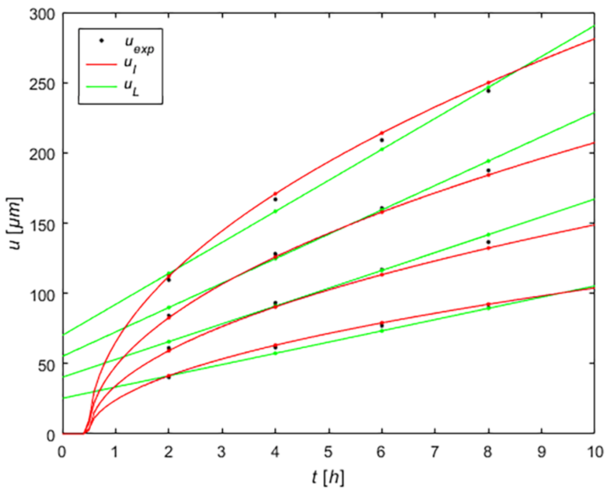

Figure 3.Predicted thicknesses of the Fe_2B boride layer by Integral and Linear methods

At first glance, it is observed from Figure 3 that both methods succeed in estimating the Fe_2B layer thickness.The integral method u_I took time and temperature into considerationalong withthe incubation time $t_0$and other variables, whereas the linear one u_L only had time and temperature as the sole variables.

The catchwith the linear method u_L is that before $2h$, as seen in Figure 3, there is a layer thickness with no treatment time, which is not possible. Thus, it can be said that the linear model cannot be used before 2 h and may result in bad predictions.

On the other hand, it can be said that the predictions add up well to the experimental data sets, but as given at each data point in Table 2 for comparisons, the numerical values seem somewhat dissimilar. To further establish each method's accuracy, a residual analysis is followed.

Table 2.Predicted thickness values of Fe_2B layers using the Integral and Linear methods

	$T[°C]$	t [h]			
		2	4	6	8
u_I [μm]	850	41,40	63,09	79,04	92,28
	900	59,18	90,36	113,25	132,24
	950	82,65	125,97	157,81	184,24
	1000	112,12	170,89	214,09	249,93
u_L [μm]	850	41,34	57,34	73,35	89,35
	900	65,64	91,02	116,40	141,77
	950	89,94	124,69	159,44	194,19
	1000	114,24	158,37	202,49	246,62

4. RESIDUAL ANALYSIS

Both modelsare analyzed to determine their accuracy by means of error calculations, i.e., residuals.

Figure 4 gives a bar chart of the absolute residuals obtained in each method, where it is observed that the integral model u_I had only two predictions with residuals exceeding 5 μm, while the linear method had five, which can be problematic.

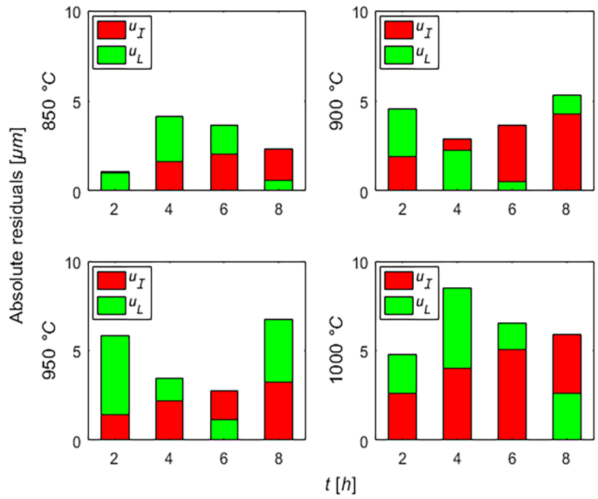

Figure 4.Absolute prediction residuals on both integral and linear methods

To further see if, even with this excess, the linear method can still be of use, the mean absolute error (MAE), Eq. (18), is introduced in each studied point (Figure 5), The mean absolute error obtained in the used method, the linear model u_L was 3.84, while it was 2.94 on the integral one u_I, which is better by 30%, but with it being marginal, the linear model compares quite accurately to it prediction-wise.

$$MAE = \frac{1}{n}\sum_{i=1}^{n}|u_i - \bar{u}| \quad (18)$$

Where

u_i denotes the predicted value,

$\bar{u}$ the actual value and

n the number of predictions.

Using the range of the mean absolute error (MAE) at each point, Figure 5 shows that 6 out of 16 data points were outside the estimated mean absolute error for the integral method. As for the linear method, there were 8. Thus, that is the price to pay precision-wise due to the use of only two variables to model, but it can be reconfirmed that the linear method was comparable in its predictions of the boride layer thickness to the integral method. Similar remarks were also observed in another paper [35] containing the same linear method compared to a method different than the integral method, and on a different substrate, XC38 steel.It was also concluded that the linear method has tolerable predictions [35].

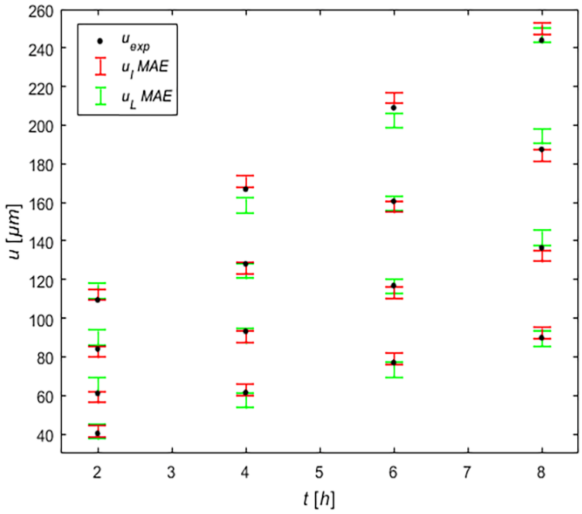

Figure 5.MAE at each predicted point for the integral and linear methods

Table 3.Prediction of the boride layer thickness at 980 $°C$ for 5 $h$with both methods and their absolute errors

980 °C for 5 h	u_{exp} [μm]	
	175 ± 12	
	u_I	u_L
	179,79	165,09
	Absoluteerror	
	4,79	9,91

At last, to affirm if the model works on other points in the studied interval and not out of it as concluded, a separate experiment that was carried out at a temperature of 980 $°C$ for 5 h [18] is examined to help test the accuracy of both models on a point not taken into account when the models were established. The results are given in Table 3, where the integral method was off by 4.79 μm while the linear method, by 9,91μm, which is more than the other for the same reasons seen previously, the use of less variables within the model. However, comparing with the experimental given error range ($\pm$12 μm), both methods predict within that estimated error range. Eventually, it can be said that the multilinear model is valid when predicting Fe_2B boride layer thicknesses.

5. CONCLUSIONS

A statistical approach has been used to develop a linear-based regression model, giving satisfactory estimation for the Fe_2B layer thickness on Armco iron with the possibleleast variables, time and temperature, from theexperimental data.

The linear regression employedin the approach gave predictions comparable to those of the integral method. However, the estimation from the linear regression is viable only if taken inside the scope of the data sets' studied interval. Otherwise, the prediction may diverge.

If the intent is simplicity, linear regression is preferable due to it having only two variables from which a model can be established, contrary to the integral method having many more variables and constants required.

The linear regression approach can be useful in predictive modeling of boriding kinetics concerning boride layer thicknesses.Specifically, multiple linear regression can further be utilized in studies of dual-phase boride layers or in other substrates.

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IZVOD

PREDIKTIVNO MODELIRANJE BORONIZACIJE KINETIKE U ARMCO UZORCIMA GVOŽĐA KORIŠTENJEM PRISTUPA LINEARNE REGRESIJE

Alati neizbježno degradiraju tokom vremena, s vijekom trajanja koji varira u zavisnosti od materijala i načina upotrebe. Kako bi se to ublažilo, koriste se termokemijski tretmani poput boronizacije. Ovim procesom formira se boridni sloj s povoljnim tribološkim i mehaničkim svojstvima. U zavisnosti od uslova obrade, sloj može biti mono-fazni Fe_2B ili bi-fazni FeB/Fe_2B. Ova studija ispituje mono-fazni Fe_2B sloj formiran boronizacijom Armco željeza metodom pakovanja u prah. Cilj je bio predvidjeti debljinu ovog sloja korištenjem linearnog pristupa s minimalnim brojem varijabli i uporediti te predikcije sa složenijim modelom koji zahtijeva dodatne parametre. Rezultati pokazuju da oba modela daju slične predikcije unutar dostupnog intervala, čime se potvrđuje tačnost linearnog modela. Međutim, linearni pristup pokazuje odstupanja kada se primijeni izvan istraživanog intervala, što ukazuje na ograničenja njegove generalizacije.
Ključne riječi*: Borid, boronizacija, sloj, linearno, regresija, debljina*

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