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Modelling the formation kinetics of Fe₂B layers with a diffusion model using Taylor expansion

ABSTRACT

This work aimed to model the growth kinetics of Fe₂B layers on the DIN 1.2738 steel by using a novel kinetic approach. The proposed model considered the transient diffusion regime of boron atoms through the surface of treated steel. The distribution of boron atoms across the Fe₂B layer was expressed as a Taylor expansion of second order. Afterwards, the boron activation energy in the Fe₂B layers was assessed as equal to 214.48 kJmol⁻¹ in the temperature range 1123-1223 K using the experimental results taken from the literature. Finally the present model has been validated experimentally by using additional boriding condition (1198 K for 4.5 h). The experimental Fe₂B layer thickness obtained at 1198 K for 4.5 h aligned with the predicted value provided by the model. Furthermore, the mass gain per unit area resulting from the formation of Fe₂B layer was also calculated as a function of processing parameters.

Keywords: Boriding / Iron boride / Kinetics / Diffusion model/ Boron activation energy

1. INTRODUCTION

The boronizing process is intended to surface harden the surface of treated ferrous or non-ferrous alloys. It leads to the improvement in tribological properties and resistance against attack in some acids and alkali media of treated materials [1]. The boriding treatment requires the process temperatures ranging from 800 to 1050°C for 0.5-10 h. For the boronizing process, the active boron can be supplied from different sources: (solid [2], liquid [3], plasma-paste [4] and gas [5]). The powder-pack boronizing is selected as one of the most attractive processes to generate the boride layers at the surface of a workpiece due to its easy handling and to its economic advantage [6]. For steels and Armco iron, the two kinds of iron borides could be formed. In practice, a single phase boride layer (Fe₂B) is more suited for practical use than the dual phase boride layer (FeB+ Fe₂B) due to the development of cracks located across the (FeB/Fe₂B) interface. Kinetically, many different kinetic approaches were proposed to investigate the kinetics of

boronized layers on Armco iron [7-9] and steels [10-18]. Some of these diffusion models considered the occurrence of boride incubation times during the formation of either dual-phase boride layer (FeB + Fe₂B) or the Fe₂B monolayer. For instance, Nait Abdellah et al. [8] examined the growth kinetics of Fe₂B layers on Armco iron using two different mathematical models. The first approach involved applying the mass balance equation at the (Fe₂B/substrate) interface, taking into account the boride incubation time. The second approach used dimensional analysis based on the Buckingham-Pi theorem as another powerful tool. In another study, Morales-Robles et al.[14] investigated the influence of the chemical potential of boron in the powders mixture being used three-fold to treat the ASTM A36 steel. A kinetic model using the Taylor expansion was capable of estimating the value of boron activation energy of 158.62 kJ mol⁻¹, unaffected by the lowering of boron chemical potential.

Raden Dadan et al.[17] suggested an approach using the phase-field method to simulate the growth kinetics of Fe₂B layers on Armco iron. In their study, the optimized parameter was the free energy driving force for the Fe₂B phase derived from the austenite phase, which enabled them to reproduce the experimental results. Campos et al. [18] analyzed the growth kinetics of Fe₂B layers on

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AISI 1045 steel using neural networks and least squares techniques, and varying the boron paste thickness from 2 to 5 mm. The results indicated that the mean error in Fe₂B layer thicknesses was 5.31% for the neural network model and 3.42% for the least squares method.

In the present work, a new kinetic approach [14] has been employed for the boriding kinetics of DIN 1.2738 steel to assess the values of boron diffusion coefficients in the Fe₂B layers in the temperature range 1123-1223 K.

In addition, this mathematical model has been verified experimentally by a direct comparison of the experimental Fe₂B layer thickness with the predicted value for the corresponding boriding condition (1198 K during 4.5 h). Lastly, the mass gain per unit area resulting from the formation of Fe₂B layer was evaluated as a function of time duration for the given boriding temperature.

2. THE DIFFUSION MODEL USING TAYLOR EXPANSION

This diffusion model [14] considers the growth of a single boride layer (Fe₂B) over a saturated substrate with boron atoms of DIN 1.2738 steel. Schematic boron concentration– profile through the Fe₂B layer is displayed in Figure 1.

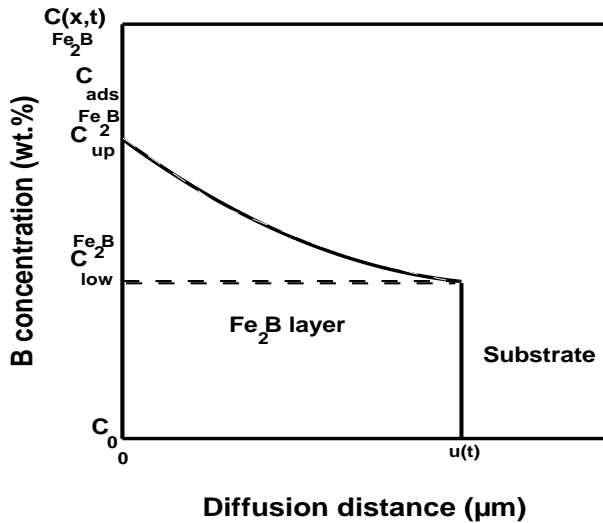


Figure 1. Schematic illustration of boron distribution across the Fe₂B layer

$$C_{Fe_2B}(x,t) = C_{Fe_2B}(u,t) + \left. \frac{\partial C_{Fe_2B}(x,t)}{\partial x} \right|_{x=u} (u(t) - x) + \frac{1}{2} \left. \frac{\partial^2 C_{Fe_2B}(x,t)}{\partial x^2} \right|_{x=u} (u(t) - x)^2 \quad \text{for } 0 \leq x \leq u \quad (6)$$

The mass balance equation at the (Fe₂B/substrate) interface is given by Equation (7) :

$$w \frac{du}{dt} = -D_{Fe_2B} \left. \frac{\partial C_{Fe_2B}(x,t)}{\partial x} \right|_{x=u} \quad (7)$$

$C_{up}^{Fe_2B}$ represents the upper limit of boron content in Fe₂B (=9 wt.%) while $C_{low}^{Fe_2B}$ is the lower limit of boron content in Fe₂B (=8.83 wt.%). The point $x = u(t)$ stands for the Fe₂B layer thickness or the position of (Fe₂B/substrate) interface. The term C_{ads} is the adsorbed boron concentration in the boride layer during the boriding treatment [19]. C_0 is the boron solubility in the matrix which is very low (≈ 0 wt.%) [20].

The initial and boundary conditions for the diffusion problem are given by:

Initial condition:

$$t = 0, x > 0, \\ \text{with } C_{Fe_2B}[x(t), t = 0] = C_0 \approx 0 \text{ wt. \%} \quad (1)$$

Boundary conditions:

$$C_{Fe_2B}(x = 0, t = 0) = C_{up}^{Fe_2B} \\ \text{for } C_{ads}^B > 8.83 \text{ wt. \%} \quad (2)$$

$$C_{Fe_2B}(x = u, t) = C_{low}^{Fe_2B} \\ \text{for } C_{ads}^B < 8.83 \text{ wt. \%} \quad (3)$$

The Fe₂B layer thickness is given by Equation (4) :

$$u = 2\varepsilon \sqrt{D_{Fe_2B} t} \quad (4)$$

where ε is a dimensionless constant to be determined and D_{Fe_2B} the value of diffusion coefficient in Fe₂B. The Second Fick's law that describes the evolution of boron concentration in Fe₂B as a function of diffusion distance x and time t is expressed by Equation (5) :

$$\frac{\partial C_{Fe_2B}(x,t)}{\partial t} = D_{Fe_2B} \frac{\partial^2 C_{Fe_2B}(x,t)}{\partial x^2} \quad (5)$$

The expression of boron-concentration profile through the Fe₂B layer is approximated using Equation (6) as a Taylor series of second order.

with

$$w = \left[\frac{(C_{up}^{Fe_2B} + C_{low}^{Fe_2B})}{2} - C_0 \right]$$

At the (Fe₂B/substrate) interface, the boron concentration is kept constant and the velocity of

the growing interface can be expressed by Equation (8):

$$\frac{du}{dt} = - \frac{D_{Fe_2B} \left. \frac{\partial^2 C_{Fe_2B}(x,t)}{\partial x^2} \right|_{x=u}}{\left. \frac{\partial C_{Fe_2B}(x,t)}{\partial x} \right|_{x=u}} \quad (8)$$

Therefore, the quantity $\left. \frac{\partial C_{Fe_2B}(x,t)}{\partial x} \right|_{x=u}$ can be deduced from Equation (7) by considering the derivation of Equation (4) with respect of the treatment time t :

$$\left. \frac{\partial C_{Fe_2B}(x,t)}{\partial x} \right|_{x=u} = - \frac{w}{D_{Fe_2B}} \left(\frac{du}{dt} \right) = - \frac{2w\varepsilon^2}{u(t)} \quad (9)$$

$$C_{Fe_2B}(x,t) = C_{low}^{Fe_2B} - \frac{w}{D_{Fe_2B}} \left(\frac{du}{dt} \right) (x-u) + \frac{w}{2D_{Fe_2B}^2} \left(\frac{du}{dt} \right)^2 (x-u)^2 \quad (11)$$

By substituting $x=0$ into Equation (11) for which $C_{Fe_2B}(x=0,t) = C_{up}^{Fe_2B}$, Equation (12) is derived as follows:

$$\frac{w}{D_{Fe_2B}} \left(u \frac{du}{dt} \right) \left[1 + \frac{1}{2D_{Fe_2B}} \left(u \frac{du}{dt} \right) \right] = (C_{up}^{Fe_2B} - C_{low}^{Fe_2B}) \quad (12)$$

By considering the time derivative of Equation (4), the following biquadratic Equation is obtained from Equation (12) as follows:

$$2w\varepsilon^2 + 2w\varepsilon^4 = (C_{up}^{Fe_2B} - C_{low}^{Fe_2B}) \quad (13)$$

By solving Equation (13) for the unknown ε , one finds the unique positive solution given by Equation (14):

$$\varepsilon = \sqrt{\frac{(-1 + \sqrt{1 + 2 \frac{(C_{up}^{Fe_2B} - C_{low}^{Fe_2B})}{w}})}{2}} \quad (14)$$

The value of boron diffusion coefficient in Fe₂B can be calculated from Equation (15) as:

$$D_{Fe_2B} = \left(\frac{k}{2\varepsilon} \right)^2 \quad (15)$$

The second derivative of $\left. \frac{\partial^2 C_{Fe_2B}(x,t)}{\partial x^2} \right|_{x=u}$ at

point $x=u$ can be readily obtained from Equation (8) with the consideration of Equation (9):

$$\left. \frac{\partial^2 C_{Fe_2B}(x,t)}{\partial x^2} \right|_{x=u} = + \frac{w}{D_{Fe_2B}} \left(\frac{du}{dt} \right)^2 = + \frac{4w\varepsilon^4}{u^2(t)} \quad (10)$$

Thus, the boron concentration profile through the Fe₂B layer is given by Equation (11) by considering the boundary condition provided by Equation (3):

The mass gain per unit area generated by the formation of Fe₂B layer at the material surface can be computed by using Equation (16).

$$G = \int_0^t \rho_{Fe} D_{Fe_2B} \left. \frac{\partial C}{\partial x} \right|_{x=0} dt = 2\rho_{Fe} w \varepsilon \sqrt{D_{Fe_2B} t} \quad (16)$$

where $\rho_{Fe} = 7.86 \text{ g cm}^{-3}$ which denotes the iron density with $w = 8.915 \text{ wt.}\%$

It is worth noting that the analytical solution of Equation (12) yields a parabolic relation without assuming it a priori, and it also confirms the parabolic growth law of Fe₂B layers as described experimentally by Equation (4).

3. SIMULATION RESULTS AND DISCUSSIONS

The proposed approach [14] has been employed to calculate the boron diffusivities in the Fe₂B layers in case of DIN 1.2738 steel. The kinetic data taken from the reference work [21] was used for this purpose. This thermochemical treatment was performed between 1123 and 1223 K during 3-9 h using a conventional furnace within inert atmosphere of high purity Argon gas. The specimens have been placed in a sealed stainless steel box containing the powders mixture with 72.5 wt.% H₃BO₃, 5 wt.% KBF₄ and 72.5 wt.% SiC. The chemical composition of DIN 1.2738 steel has 0.36 wt.% C, 0.26 wt.% Si, 1.44 wt.% Mn, 1.71 wt.% Cr, 0.23 wt.% Mo, 1.01 wt.% Ni and 0.10 wt.% Cu. It is specially employed for plastic injection die

moulds. To ensure a good accuracy of results, a great number of measurements should be considered in different locations of cross-sections of boronized layers to obtain reliable mean values of layers' thicknesses.

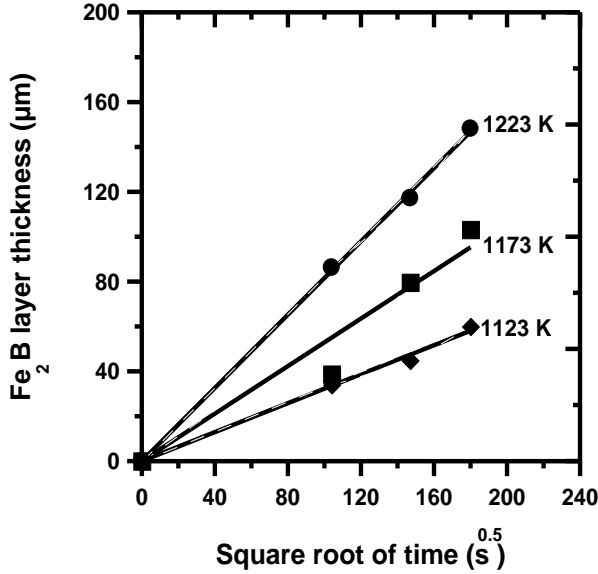


Figure 2. Change in the layer thickness of Fe₂B with the treatment time at different temperatures

Figure 2 allows describing the evolution of Fe₂B layer thickness versus the processing time at increasing temperatures. The experimental parabolic growth constants listed in Table 1 correspond to the slopes of plotted straight lines shown in the Figure 2.

Table 1. Experimental parabolic growth constants [21] at the concerned interface

T(K)	<i>k</i> (µm·s ^{-0.5})
1123	0.3226
1173	0.5612
1223	0.8158

Table 2 shows the calculated values of boron diffusion coefficients in the Fe₂B layers in the temperature range 1123-1223 K by using Equation (15) along with the values of ε parameter unaffected by the processing temperature.

Table 2. Calculated boron diffusion coefficients in Fe₂B and the values of ε parameter

T(K)	ε parameter calculated from Equation (14)	<i>D</i> _{Fe₂B} (m ² ·s ⁻¹) calculated from Equation (15)
1123	0.0971	2.75×10 ⁻¹²
1173	0.0971	8.33×10 ⁻¹²

1223	0.0971	17.61×10 ⁻¹²
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Figure 3 presents the variation of natural logarithm of calculated boron diffusivities in the Fe₂B layers with the change in boriding temperature. The boron activation energy can be inferred as the slope of the corresponding straight line displayed in Figure 3.

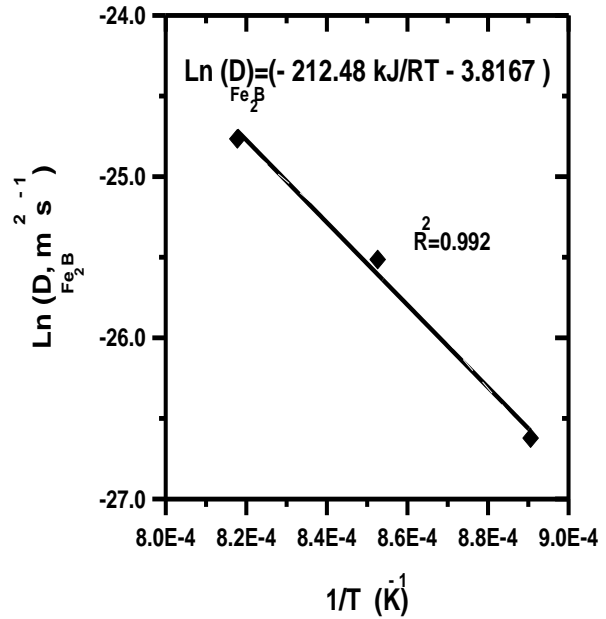


Figure 3. Evolution of natural logarithm of diffusion coefficient of boron in Fe₂B with the boriding temperature

The expression of the temperature dependence of boron diffusion coefficient in Fe₂B, valid between 1123 and 1223 K, is given by Equation (17):

$$D_{Fe_2B} = 2.20 \times 10^{-2} \exp\left(\frac{-212.48 kJ mol^{-1}}{RT}\right) \quad (17)$$

With

$$R = 8.314 J mol^{-1} K^{-1}$$

The estimated value of boron activation energy in Fe₂B (=212.48 kJ mol⁻¹) in case of DIN 1.2738 steel is an necessary amount of energy to make the boriding process possible and to activate the solid state diffusion of boron atoms [16] along the favoured crystallographic pathway [001].

Table 3 compares the listed values of boron activation energies in case of boronized Armco iron and some borided steels [7-13, 21-26] with the calculated value of boron activation energy from the present model. It is obvious that the values of activation energies in these materials are

depending on the chemical compositions of their substrates except for Armco iron, on the calculation method of boron activation energy and on the selected processing parameters.

Table 3. Reported literature results on boron activation energies in Armco iron and steels

Material	Solid boronizing	Activation energy (kJ mol ⁻¹)	Range of temperatures (K)	Approach employed	Ref.
Armco iron	Powder	(Fe ₂ B) 157.0	1223-1323	Diffusion model	[7]
Armco iron	Powder	(Fe ₂ B) 157.94	1123-1273	Mean diffusion coefficient method	[9]
AISI 1045 steel	Powder	180.0 (Fe ₂ B)	1123-1273	Diffusion model	[10]
ASTM A 36 steel	Powder	161.65 (Fe ₂ B)	1123-1273	Alternative diffusion model	[11]
AISI 1045 steel	Powder	154 ± 7 (FeB) and 141 ± 9 (Fe ₂ B)	1173-1273	Bilayer model	[12]
SAE 1020 steel	Powder	183.15 (Fe ₂ B)	1123-1223	Parabolic growth law	[13]
AISI 1050 steel	Powder	162.93 (FeB+ Fe ₂ B)	1123-1223	Parabolic growth law	[22]
16MnCr6 Steel	Powder	243.6 (FeB+ Fe ₂ B)	1123-1223	Parabolic growth law	[23]
ASP 2012 steel	Powder	314.716 (FeB+ Fe ₂ B)	1123-1223	Parabolic growth law	[24]
Hardox steel	Powder	157.99 (FeB+ Fe ₂ B)	1123-1223	Parabolic growth law	[25]
AISI M2	Powder	201.57 (Fe ₂ B)	1173-1373	Parabolic growth law	[26]
DIN 1.2738 steel	Powder	204.07 (Fe ₂ B)	1123-1223	Parabolic growth law	[21]
DIN 1.2738 steel	Powder	212.48 (Fe ₂ B)	1123-1223	Diffusion model using Taylor expansion	This work

For indication, Calik et al. [23] employed the solid boriding to treat the 16MnCr5 steel between 1123 and 1223 K. The XRD studies revealed the presence of dominant iron borides (FeB and Fe₂B) as well as minor precipitates (MnB and Cr₂B). The boron activation energy was assessed from the classical parabolic relationship as 243.6 kJmol⁻¹, higher than the value found in this work due to the difference in chemical composition of the treated steel.

In another study, Kayali et al. [24] boronized the powder metallurgy (PM) tool steel denominated ASP2012 in the temperature range 1123-1223 K. The XRD analyses corroborated the presence of dual boride layers (FeB+ Fe₂B) enriched by metal borides as precipitates like CrB, Mo₂B and W₂B. The value of boron activation energy in ASP 2012 steel was calculated as 314.716 kJ mol⁻¹. Such a value of activation energy was deemed very high ascribed to the high contents of alloying elements in the ASP@2012 steel.

In Table 4 are grouped the experimental Fe₂B layer thickness obtained at 1198 K for 4.5 h together with the computed layer thickness provided by Equation (12) for a maximum boron concentration in Fe₂B of 9 wt.%. The numerical solution of Equation (12) was obtained by help of

the Interactive Thermodynamic software (version 3.2) freely available by taking as initial condition $u_0=0.10 \mu\text{m}$. Equation (4) yields also the same simulation result as Equation (12) for $\varepsilon = 0.0971$. It is noticed that the experimental Fe₂B layer thickness [21] aligns with the calculation results from the present kinetic approach.

Table 4. Experimental value of Fe₂B layer thicknesses obtained at 1198 K during 4.5 compared to the calculated value by the present approach

Boronizing conditions	Experimental Fe ₂ B layer thickness (μm) [21]	Predicted Fe ₂ B layer using the numerical solution of Equation (12)
1198 K for 4.5 h	83.66	85.54

Figure 4 shows the time dependency of mass gain per unit area related to the formation of Fe₂B layer at increasing temperatures. The G values follow a parabolic trend with treatment time for the given processing temperature, because the lattice-scale displacement of boron atoms diffusing through the steel surface is a diffusion-controlled

process. The G values calculated at 1223 K are approximately 2.5 times higher than those obtained at 1123 K. This indicates that the processing temperature has a significant influence on the G value.

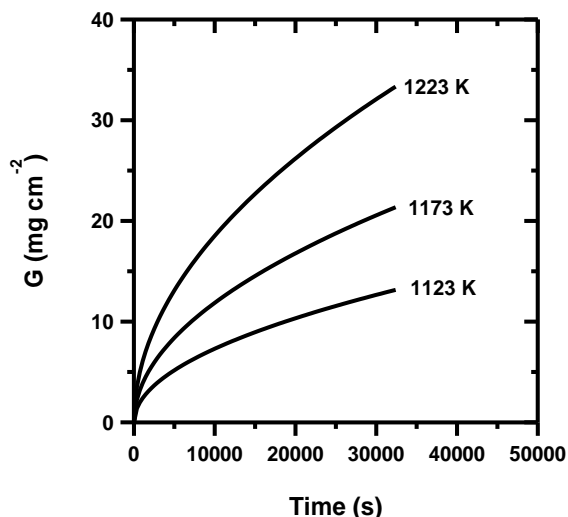


Figure 4. Change in the mass gain per unit of area associated with the formation of Fe₂B with the time duration at increasing temperatures

4. CONCLUSIONS

In the current work, the non linear diffusion model using Taylor expansion has been suggested to address the simulation of the growth kinetics of Fe₂B layers on DIN 1.2738 steel between 1123 and 1223 K. Afterwards, the value of the activation energy was estimated as 212.48 kJ mol⁻¹ for DIN 1.2738 steel and has been compared to the results from the literature. Finally, the present approach has been verified by a direct comparison of experimental layer thickness of Fe₂B (formed at 1198 K for 4.5 h) with the model result. The experimental result was in line with the value of layer thickness given by the diffusion model using Taylor expansion.

The mass gain per unit area was determined versus the processing parameters. It varied sensitively with the processing temperature due to the activation of diffusion phenomenon of boron atoms. For future researches, the non linear model based on Taylor expansion can be expanded to address multi-phase systems involving the diffusion of interstitial elements to simulate the kinetics of the corresponding thermochemical processes.

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IZVOD

MODELIRANJE KINETIKE OBRAZOVANJA Fe₂B SLOJEVA SA DIFUZIJSKIM MODELOM KORIŠĆENJEM TEJLOROVE EKSPANZIJE

Ovaj rad je imao za cilj da modelira kinetiku rasta Fe₂B slojeva na DIN 1.2738 čeliku korišćenjem novog kinetičkog pristupa. Predloženi model razmatra prelazni režim difuzije atoma bora kroz površinu obrađenog čelika. Raspodela atoma bora u sloju Fe₂B izražena je kao Tejlorova ekspanzija drugog reda.

Potom je energija aktivacije bora u slojevima Fe₂B procenjena na 214,48 kJmol⁻¹ u temperaturnom opsegu 1123-1223 K korišćenjem eksperimentalnih rezultata preuzetih iz literature . Konačno, sadašnji model je eksperimentalno validiran korišćenjem dodatnih uslova boriranja (1198 K tokom 4,5 h). Eksperimentalna debljina sloja Fe₂B dobijena na 1198 K tokom 4,5 h usklađena je sa predviđenom vrednošću u koju daje model . Štaviše, povećanje mase po jedinici površine kao rezultat formiranja sloja Fe₂B takođe je izračunato kao funkcija parametara obrade.

Ključne reči: Boriding, borid gvožđa, kinetika, difuzioni model, energija aktivacije bora

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