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# ESTIMATION OF THE EFFECTIVE BORON-DIFFUSION COEFFICIENT IN THE $Fe_2B$ LAYERS GROWN ON GRAY CAST IRON

## DOLOČANJE EFEKTIVNEGA KOEFICIENTA DIFUZIJE BORA PRI RASTI PLASTI $Fe_2B$ NA SIVEM LITEM ŽELEZU

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The present work evaluates the effective diffusion coefficient of boron in the $Fe_2B$ layers grown on a substrate from gray cast iron. The boride layers were generated using powder-pack boriding in the temperature range of 1173–1273 K for (2, 4, 6 and 8) h of treatment. First, the diffusion coefficient of boron in $Fe_2B$ (free of chemical stresses) was obtained through solving the mass-balance equation at the ($Fe_2B$/substrate) interface using the experimental values of parabolic-growth constants taken from the literature.
Second, a simple equation was derived to evaluate the effective diffusion coefficient of boron in $Fe_2B$ by considering the effect of chemical stresses. The estimated values of boron activation energies were 154.8 kJ/mol and 164.8 kJ/mol with and without the presence of chemical stresses. Furthermore, the calculated values of effective diffusion coefficients were found to be sensitive to the change in the boriding temperature and to the increase in the upper boron concentration in $Fe_2B$.

Keywords: boriding, chemical stresses, kinetics, Fick's law, effective diffusion coefficient, incubation time

To delo ocenjuje efektivni koeficient difuzije bora v plasteh $Fe_2B$, zraslih na podlagi iz sivega litega železa. Boridne plasti so nastale med boriranjem v prahu v škatli, v temperaturnem območju 1173–1273 K in trajanju (2, 4, 6 in 8) h. Najprej je bil določen koeficient difuzije bora v $Fe_2B$ (brez kemijskih napetosti) z rešitvijo enačbe za masno ravnotežje ($Fe_2B$/podlaga) na stiku z uporabo eksperimentalnih vrednosti za konstanto parabolične rasti, dobljeno v literaturi.
Nato je bila izpeljana enostavna enačba za oceno efektivnega koeficienta difuzije bora v $Fe_2B$ z upoštevanjem kemijskih napetosti. Določeni sta bili vrednosti aktivacijske energije bora 154,8 kJ/mol in 164,8 kJ/mol s kemijskimi napetostmi in brez njih. Ugotovljeno je bilo tudi, da so izračunane vrednosti efektivnega koeficienta difuzije občutljive za spremembo temperature boriranja in za naraščanje koncentracije bora v $Fe_2B$.

Ključne besede: boriranje, kemijske napetosti, kinetika, Fickov zakon, efektivni koeficient difuzije, čas inkubacije

## 1 INTRODUCTION

Boriding is a well-known thermochemical treatment for creating boride layers with interesting properties by saturating the surface layer of a material with boron. It is used to improve the surface hardness, the frictional wear, the fatigue endurance and the corrosion resistance of ferrous and non-ferrous alloys. It can be performed between 1123 K and 1323 K to form iron borides on the material surface with the treatment times varying from 0.5 h to 10 h[1]. Different boriding methods exist in practice. However, the powder-pack boriding has the advantages of simplicity, flexibility with respect to the powder composition, minimum equipment and cost-effectiveness.[2–4]

The boriding temperature, the time duration, the boron potential of the medium and the chemical composition of the substrate are the key parameters controlling the morphology, the growth kinetics and the microstructural nature of boride layers.[5–7]

The present model considers the effect of chemical stresses when evaluating the effective diffusion coefficient of boron in the $Fe_2B$ layers grown on a gray-cast-iron substrate. In this context, some recent reference studies regarding the evaluation of the effective diffusion coefficient of boron in $Fe_2B$ were reported for borided AISI 1018 and AISI 4140 steels and borided Armco iron.[8–10]

Certain reference works[11,12] report on the chemical stresses resulting from the composition gradient similar to those caused by the thermal stresses in an isotropic medium. Diffusion-induced stresses (or chemical stresses) are then built up due to the composition inhomogeneity during mass transfer. Diffusion-induced stresses also change the mechanical properties of metal systems during mass transfer. Larché and Cahn[13–15] investigated the stresses arising from the inhomogeneities of materials. Consequently, chemical stresses enhance both the diffusion coefficient and the concentration.[16]

The objective of this work was to evaluate the diffusivity of boron in $Fe_2B$ (with and without the presence of chemical stresses) in the temperature range of 1173–1273 K by applying a kinetic model of a monolayer configuration of $Fe_2B$ grown on gray cast iron.

## 2 MATHEMATICAL MODEL

A schematic non-linear concentration profile of boron in the $Fe_2B$ layer is depicted in **Figure 1** where the boron potential allows the formation of $Fe_2B$ on the surface of the material.

$C_{up}^{Fe_2B}$ denotes the upper boron concentration in $Fe_2B$, $C_{low}^{Fe_2B}$ (= 59.2 × 10[3] mol m[–3]) represents the lower boron concentration in $Fe_2B$ and $t_0(T)$ is the boride incubation time depending on the boriding temperature. $C_{ads}$ is defined as the effective boron concentration[17]. Distance *u* is the layer depth at the ($Fe_2B$/substrate) interface as a function of treatment time *t*. $C_0$ is the boron solubility in the matrix. The upper boron concentration in $Fe_2B$ is between $59.2 \times 10^3$ mol m$^{-3}$ and $60 \times 10^3$ mol m$^{-3}$ since this phase exhibits a narrow composition range as pointed out by Massalski[18].

When building a mathematical model of the problem, the following assumptions are made:

- The growth kinetics is controlled by the boron diffusion in the $Fe_2B$ layer
- The growth of the $Fe_2B$ boride layer is a consequence of the boron diffusion perpendicular to the sample surface
- The solid solution of boron in the matrix behaves ideally (i.e., the activity coefficient of boron in the solid solution is independent on the concentration)
- The $Fe_2B$ iron boride nucleates after a certain incubation time
- The boride layer is thin in comparison to the sample thickness
- Local equilibrium is held at the ($Fe_2B$/substrate) interface
- A planar morphology is assumed for the ($Fe_2B$/substrate) interface
- The volume change during the phase transformation is neglected

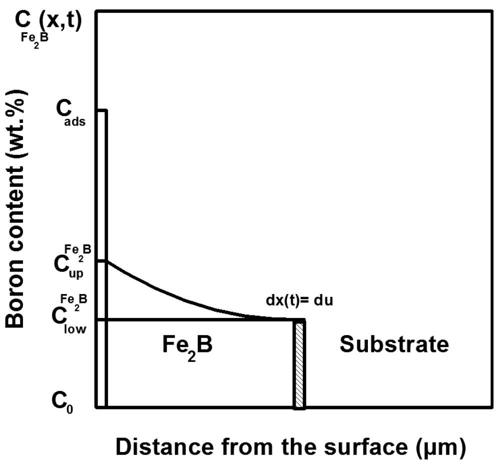


**Figure 1:** Schematic non-linear concentration profile of boron in the $Fe_2B$ layer

**Slika 1:** Shematični prikaz nelinearnega profila koncentracije bora skozi plast $Fe_2B$

- A uniform temperature is assumed throughout the sample
- No effect of the alloying elements on the boron diffusion is assumed
- The pressure effect on the boron effective diffusion coefficient in $Fe_2B$ is ignored

The initial condition of the diffusion problem is expressed with Equation (1):

$$C_{Fe_2B}(x,0) = C_0 \tag{1}$$

The boundary conditions of the diffusion problem are given with Equations (2) and (3):

$$C_{Fe_2B}\{x[t=t_0(T)]=0, t_0(T)\} = C_{up}^{Fe_2B} \quad \text{for } C_{ads} \succ 59.2 \times 10^3 \text{ mol m}^{-3} \tag{2}$$

$$C_{Fe_2B}\{x(t=t)=u, t\} = C_{low}^{Fe_2B} \quad \text{for } C_{ads} \prec 59.2 \times 10^3 \text{ mol m}^{-3} \tag{3}$$

for $0 \le x \le u$.

The Fick's second law of diffusion[19] relating to the change in the boron concentration throughout the $Fe_2B$ layer with time *t* and location *x*(*t*) is:

$$\frac{\partial C_{Fe_2B}(x,t)}{\partial t} = D_B^{Fe_2B} \frac{\partial^2 C_{Fe_2B}(x,t)}{\partial x^2} \tag{4}$$

where $D_B^{Fe_2B}$ is the diffusion coefficient of boron in $Fe_2B$ free of chemical stresses and $C_{Fe_2B}(x,t)$ is the boron concentration. The boron concentration along the boride layer, which is the solution of the Fick's second law (Equation (4)) in a semi-infinite medium, is consistent with **Figure 1**. Its expression is given with Equation (5):

$$C_{Fe_2B}(x,t) = C_{up}^{Fe_2B} + \left( \frac{C_{low}^{Fe_2B} - C_{up}^{Fe_2B}}{erf \dfrac{u}{2\sqrt{D_B^{Fe_2B} \cdot t}}} \right) \cdot erf \frac{x}{2\sqrt{D_B^{Fe_2B} \cdot t}} \tag{5}$$

for $0 \le x \le u$.

The continuity equation at the ($Fe_2B$/substrate) interface is expressed with Equation (6):

$$w_{Fe_2B} \left[ \frac{dx}{dt} \right]_{x=u} = -D_B^{Fe_2B} \left[ \frac{\partial C_{Fe_2B}}{\partial x} \right]_{x=u} \tag{6}$$

with $w_{Fe_2B} = \left[ 0.5 \times (C_{up}^{Fe_2B} + C_{low}^{Fe_2B}) - C_0 \right]$.

Boride layer thickness *u* follows the parabolic-growth law expressed with Equation (7), where *k* represents the parabolic-growth constant at the ($Fe_2B$/substrate) interface:

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

where $[t - t_0(T)]$ is considered as the effective growth time of the $Fe_2B$ layer[9,10,20–22]. According to Brakman et al.[23], boride incubation time $t_0(T)$ is decreased as the temperature goes up. The $\beta(T)$ parameter, which takes

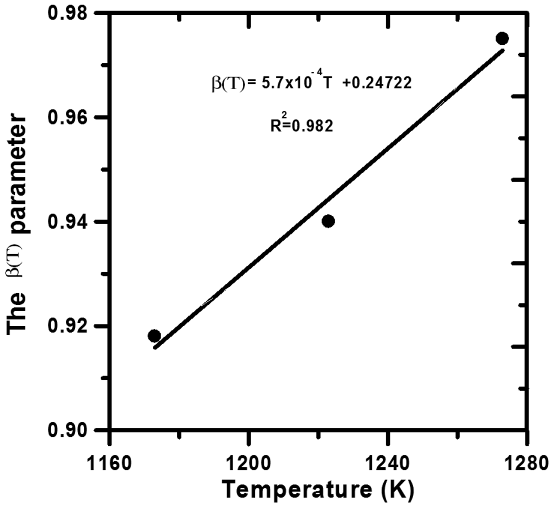


**Figure 2:** Temperature dependence of the $\beta(T)$ parameter
**Slika 2:** Temperaturna odvisnost parametra $\beta(T)$

into account the effect of the boride incubation time, is given with Equation (8):

$$\beta(T)=k\sqrt{1-\frac{t_0(T)}{t}} \tag{8}$$

The $\beta(T)$ parameter can be approached with a linear relationship (Equation (9)), illustrated also in **Figure 2** (with a correlation factor = 0.982):

$$\beta(T)=(5\cdot 7\times 10^{-4}\,T+0.24722) \tag{9}$$

By derivation of Equation (5) with respect to distance $x(t)$ and Equation (7) with respect to time $t$, Equation (6) can be rewritten[24] as follows:

$$\left[0.5(C_{\rm up}^{\rm Fe_2B}+C_{\rm low}^{\rm Fe_2B})-C_0\right]k=$$
$$=2\sqrt{\frac{D_{\rm B}^{\rm Fe_2B}}{\pi}}\cdot\frac{(C_{\rm low}^{\rm Fe_2B}-C_{\rm up}^{\rm Fe_2B})}{erf\,\dfrac{k\beta(T)}{2\sqrt{D_{\rm B}^{\rm Fe_2B}}}}\cdot\exp\left(-\frac{\beta^2(T)k^2}{4D_{\rm B}^{\rm Fe_2B}}\right)\cdot\beta(T) \tag{10}$$

To evaluate the diffusion coefficient of boron in $Fe_2B$ $D_{\rm B}^{\rm Fe_2B}$ (free of chemical stresses), it is necessary to use the Newton-Raphson routine[25] to solve the problem.

For this purpose, a computer program was written in Matlab (version 6.5) to find the roots of Equation (10). By ignoring the pressure effect on diffusion, the effective diffusion coefficient of boron in $Fe_2B$ (see reference[10] for more details) can be evaluated from Equation (11) as follows:

$$D_{\rm B}^{\rm eff}=D_{\rm B}^{\rm Fe_2B}\left[1+\frac{2C_{\rm Fe_2B}(x,t)\overline{V}^2E}{9RT(1-v)}\right] \tag{11}$$

where $D_{\rm B}^{\rm eff}$ and $D_{\rm B}^{\rm Fe_2B}$ are the diffusion coefficients of boron in $Fe_2B$ with and without the chemical stresses, respectively. The origin of Equation (11) can be found elsewhere in[13–15].

Since the solid solution of boron in the matrix is ideal, the second term in Equation (11) is positive regardless of $\overline{V}$. Partial molar volume $\overline{V}$ is taken to be equal to $(1.01\times10^{-5}\ {\rm m^3\,mol^{-1}})$.[26]

$E$ = 290 GPa and $v$ = 0.3 are the Young's modulus and Poisson's ratio of the $Fe_2B$ layer, respectively[27,28]. Taking the mean value of the boron concentration throughout the $Fe_2B$ layer[10], i.e., $C_{\rm Fe_2B}(x,t)=\left[0.5\times(C_{\rm up}^{\rm Fe_2B}+C_{\rm low}^{\rm Fe_2B})\right]$, Equation (11) yields:

$$D_{\rm B}^{\rm eff}=D_{\rm B}^{\rm Fe_2B}\left[1+\frac{\overline{V}^2(C_{\rm up}^{\rm Fe_2B}+C_{\rm low}^{\rm Fe_2B})E}{9RT(1-v)}\right] \tag{12}$$

## 3 RESULTS AND DISCUSSION

To evaluate the diffusion coefficient of boron in $Fe_2B$ using Equation (10), the experimental results found in the reference work[29] in terms of the parabolic growth constants at the ($Fe_2B$/ substrate) interface and boride incubation times were used. The chemical composition (in mass fractions) of the gray cast iron (class 30, ASTM A48) to be borided is the following: (3.44–3.45 % C, 1.7–1.77 % Si, 0.5–0.6 % Mn, 0.2 % Cr, 0.45–0.5 % Cu). The boriding process was performed on the gray cast iron under an argon atmosphere in a conventional furnace at three temperatures (1173, 1223 and 1273) K and for variable times (2, 4, 6 and 8) h. The boriding medium is composed of boron carbide ($B_4C$) as the boron source and $KBF_4$ as the activator. Fifty measurements were taken on different cross-sections of the borided samples of the gray cast iron to estimate the thickness of the $Fe_2B$ layer. As the input data, the model uses the following parameters: the time, the temperature, the upper and lower boron concentrations in the $Fe_2B$ iron boride and the experimental values of the parabolic

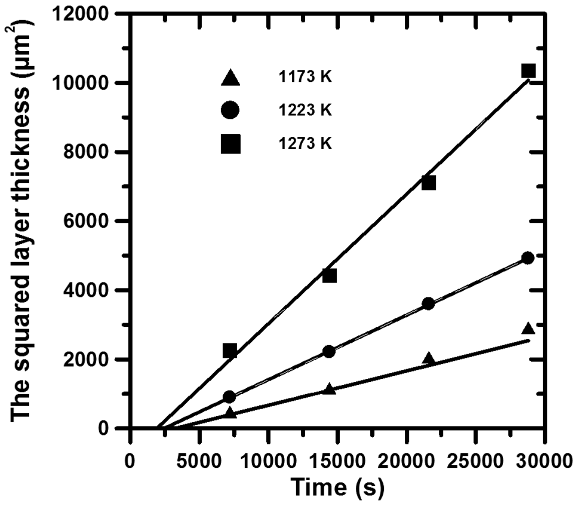


**Figure 3:** Evolution of the squared value of the $Fe_2B$ layer thickness versus the boriding time at different temperatures
**Slika 3:** Razvoj kvadratne vrednosti debeline plasti $Fe_2B$ v odvisnosti od časa boriranja pri različnih temperaturah

growth constants at the ($Fe_2B$/substrate) interface. The effective diffusion coefficient of boron in $Fe_2B$ was estimated on the basis of Equation (12). **Figure 3** shows the squared value of the experimental boride-layer thickness as a function of the boriding time, according to Equation (13):

$$u^2 = k^2 \left[t - t_0(T)\right] \qquad (13)$$

The squared values of parabolic-growth constants ($k^2$) are obtained from the slopes of the straight curves using the least-square method. The intercept with the abscissa determines the boride incubation time at each temperature.

**Table 1** gives the experimental values of parabolic-growth constants $k$ in the temperature range of 1173–1273 K, along with the corresponding boride incubation times deduced from **Figure 3**.

**Table 1:** Experimental values of parabolic-growth constants at the $Fe_2B$/substrate interface with the corresponding boride incubation times
**Tabela 1:** Eksperimentalne vrednosti konstant parabolične rasti na stiku $Fe_2B$/podlaga z ustreznimi inkubacijskimi časi boriranja

| $T$/K | Experimental parabolic-growth constant $k$/µm $s^{-0.5}$ | Incubation boride time (s) |
|---|---|---|
| 1173 | 0.3150 | 3200 |
| 1223 | 0.4321 | 2428 |
| 1273 | 0.6122 | 1902 |

**Table 2** lists the simulated values of $D_B^{Fe_2B}$ and $D_B^{eff}$ obtained at each boriding temperature using Equations (10) and (12), for the upper value of the boron content in $Fe_2B$ [10] being equal to 59.80 × $10^3$ mol $m^{-3}$.

**Table 2:** Computed values of the boron-diffusion coefficients for $Fe_2B$ and the effective diffusion coefficients of boron in $Fe_2B$, for the upper boron content in the $Fe_2B$ phase equal to 59.80 × $10^3$ mol $m^{-3}$
**Tabela 2:** Izračunane vrednosti koeficientov difuzije bora v $Fe_2B$ in efektivni koeficienti difuzije bora v $Fe_2B$ za zgornjo vsebnost bora v $Fe_2B$ fazi 59,80 × $10^3$ mol $m^{-3}$

| $T$/K | $D_B^{Fe_2B}$/($m^2$ $s^{-1}$) using Eq.(10) | $D_B^{eff}$/($m^2$ $s^{-1}$) using Eq.(12) |
|---|---|---|
| 1173 | 6.31 × $10^{-12}$ | 3.67 × $10^{-10}$ |
| 1223 | 11.881 × $10^{-12}$ | 6.64 × $10^{-10}$ |
| 1273 | 23.86 × $10^{-12}$ | 12.82 × $10^{-10}$ |

When incorporating the effect of chemical stresses on the boron diffusion, the effective diffusion coefficient of boron in $Fe_2B$ is increased with the boriding temperature. So, the dependence between the diffusivity of boron in $Fe_2B$ and the boriding temperature can be expressed with the Arrhenius equation. The temperature dependence of the diffusivity of boron in $Fe_2B$ is then depicted in **Figure 4**. The activation energy of boron (with and without the presence of chemical stresses) can be easily obtained from **Figure 4**.

As a result, the diffusion coefficient of boron in $Fe_2B$ in the temperature range of 1173–1273 K is given with:

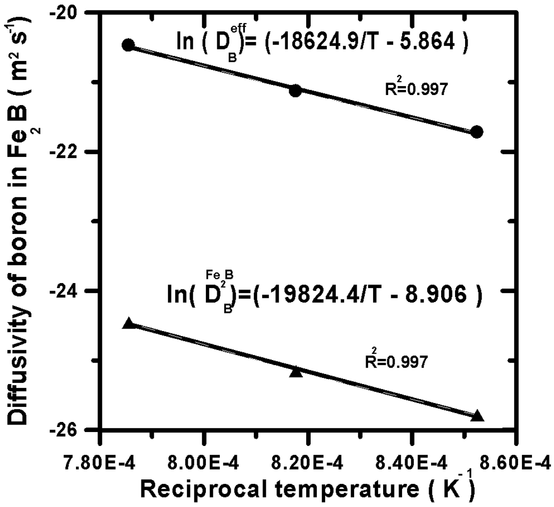


**Figure 4:** Temperature dependence of the diffusivity of boron in $Fe_2B$
**Slika 4:** Temperaturna odvisnost difuzivnosti bora v $Fe_2B$

$$D_B^{eff} = 1.35 \times 10^{-4} \exp\frac{-164.8 \text{kJ/mol}}{RT} \text{ m}^2/\text{s} \qquad (14)$$

The effective diffusion coefficient of boron in $Fe_2B$ is also determined as:

$$D_B^{eff} = 2.8 \times 10^{-3} \exp\frac{-154.8 \text{kJ/mol}}{RT} \text{ m}^2/\text{s} \qquad (15)$$

where $R$ is the universal gas constant (8.314 J/mol K) and $T$ represents the absolute temperature. The boron activation energy obtained in this work, in the absence of chemical stresses, was compared with the values found in the literature[29–34].

**Table 3** shows a comparison of the boron activation energies (in the absence of chemical stresses) obtained from different borided materials. The reported values in **Table 3** differ from each other depending on different factors such as the chemical composition of the substrate, the boriding method and the kinetic approach used to estimate the boron activation energy.

**Table 3:** Comparison of the boron activation energies (in the absence of chemical stresses) for borided ferrous alloys
**Tabela 3:** Primerjava aktivacijskih energij bora (pri odsotnosti kemijskih napetosti) za primere boriranja zlitin železa

| Material | Boron activation energy (kJ $mol^{-1}$) | Reference |
|---|---|---|
| Armco iron | 151 | 30 |
| Armco iron | 157 | 31 |
| AISI H13 | 186.2 | 32 |
| AISI 1045 | 169.6 | 33 |
| Gray cast iron | 177.4 | 29 |
| Gray cast iron | 175 | 34 |
| Gray cast iron | 164.8 | Present work |

It is seen that the calculated value of the boron activation energy (154.8 kJ $mol^{-1}$) under chemical stresses is lower than the one for 164.8 kJ $mol^{-1}$ due to an enhancement of the boron diffusion. The temperature dependence of the computed values of the effective

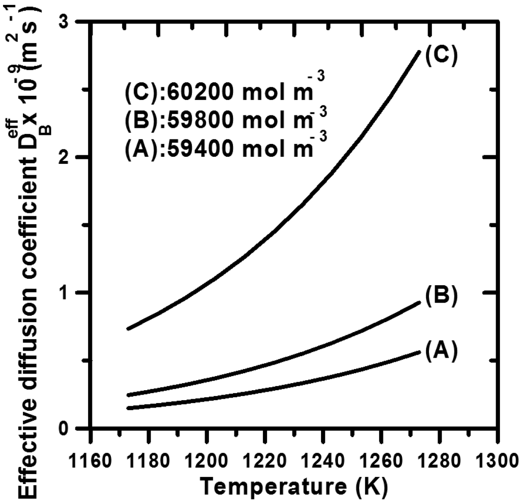


**Figure 5:** Temperature dependence of the computed values of the effective diffusion coefficient of boron in $Fe_2B$ for the increasing values of $C_{up}^{Fe_2B}$

**Slika 5:** Temperaturna odvisnost izračunanih vrednosti efektivnega koeficienta difuzije bora v $Fe_2B$ pri naraščajočih vrednostih $C_{up}^{Fe_2B}$

diffusion coefficient of boron in $Fe_2B$ for the increasing values of the upper boron contents in $Fe_2B$ is shown in **Figure 5**.

With the presence of chemical stress, the diffusion of boron atoms is accelerated with an increase in the boriding temperature since the diffusion process is a thermally activated phenomenon. At a given value of the upper boron content in $Fe_2B$, the computed values of $D_B^{eff}$ are affected by the change in the boriding temperature.

The variation in the calculated values of the effective diffusion coefficient of boron in $Fe_2B$ as a function of the upper boron content in the same phase ($C_{up}^{Fe_2B}$) is displayed in **Figure 6** for different boriding temperatures. The effective diffusion coefficient of boron in $Fe_2B$ is decreased with an increase in the upper boron content in $Fe_2B$. It can be explained with the saturation of the material surface with the active boron atoms for longer boriding times. It is concluded that the calculated values of the effective diffusion coefficients of boron in $Fe_2B$ vary notably with the upper boron content in $Fe_2B$ at a fixed boriding temperature.

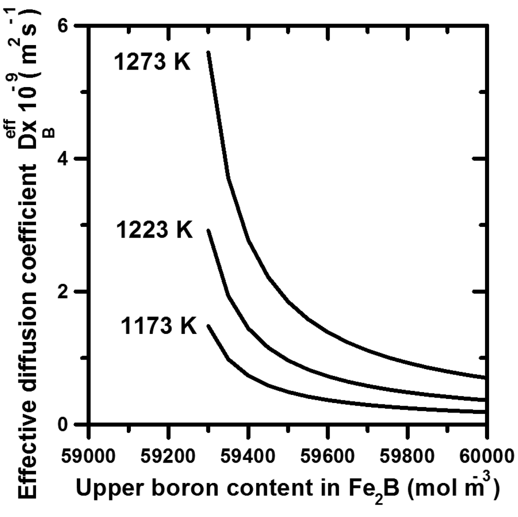


**Figure 6:** Variation in the calculated values of effective diffusion coefficients of boron in $Fe_2B$ as a function of the upper boron content in the same phase for different boriding temperatures

**Slika 6:** Spreminjanje izračunanih vrednosti efektivnega koeficienta difuzije bora v $Fe_2B$ v odvisnosti od zgornje vsebnosti bora v isti fazi pri različnih temperaturah boriranja

## 4 CONCLUSION

In the present work, the diffusion coefficient of boron in the $Fe_2B$ layers grown on gray cast iron was estimated through the mass-balance equation at the ($Fe_2B$/substrate) interface under certain assumptions. The model included the effect of boride incubation times by forming the $Fe_2B$ layers. Afterwards, the boron effective diffusion coefficient in $Fe_2B$ was evaluated by applying a simple equation based on the elasticity theory.

A lower value of the boron activation energy under chemical stresses was obtained (154.8 kJ $mol^{-1}$) for an upper boron content in $Fe_2B$ equal to 59.80 × $10^3$ mol $m^{-3}$. This result is a consequence of the chemical stresses enhancing the boron diffusion through the $Fe_2B$ layers.

In addition, it is also shown that the calculated values of the boron effective diffusion coefficient in $Fe_2B$ vary notably with the boriding temperature. The values of the boron effective diffusion coefficients in $Fe_2B$ are found to be decreased with an increase in the upper boron content in $Fe_2B$ at a given boriding temperature.

The model can be reformulated to be applied to a bilayer configuration (FeB + $Fe_2B$) as an extension of the present work to estimate the boron diffusion coefficients for the FeB and $Fe_2B$ layers in borided ferrous alloys, incorporating the effect of chemical stresses.

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