The Effects of Structure Orientation on the Growth of Fe₂B Boride by Multi-Phase-Field Simulation

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A morphological evolution of the growth of Fe₂B boride on steel substrate has been investigated using two dimensional (2D) multi-phase-field (MPF) simulations. In order to evaluate competitive growth between boride seeds during the coating process, variations on boride seed orientation have been implemented. In addition, in order to have anisotropy growth of boride, anisotropy of interfacial energy is considered on the evaluation of phase-field evolution. It was observed that boride seed with structure orientation of 90° shows a preferential growth as compared with the growth of boride seeds at other orientations. On the other hand, competitive growth between boride seeds at different crystal orientations can also be observed, where boride seeds approaching a preferential orientation angle grow faster and suppress the growth of boride seeds at the lower orientation angle. Both of these present observations agree with previous experimental observations that boride seeds tend to grow perpendicular to the substrate surface and the growth of boride seeds in this direction suppress growth in other directions. Additionally, it was observed that the preferential growth of boride is independent of the initial size of the boride seed. [doi:10.2320/matertrans.M2009227]

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1. Introduction

Boronizing has been recognized as one of the most conventional methods of surface hardening.¹⁾ Compared with other surface hardening processes, such as nitriding and carburizing, boronizing consistently outperforms these two processes. Compared with carburized or nitrided steels, the iron boride formed exhibits substantially higher hardness (1600–2000 HVs), whereas the first two products can reach a hardness of 650–900 HVs.²⁾ In particular, boronized steel exhibits excellent resistance to a variety of tribological wear mechanisms, as well as oxidation mechanism.^{3–5)} There are two types of boride products, FeB and Fe₂B boride. The first structure is considered to be harder than the latter structure, however it is more brittle. Therefore, normally having a single Fe₂B boride in the structure is more preferable than the presence of dual phase of boride. Regarding its outstanding characteristics, this process has been widely implemented in the broad area, and the most recent application of this process is in the biomedical field, as reported by Jelis *et al.*⁶⁾

It has been well understood that material properties are governed by their microstructural characteristics. They also depend on the growth behaviour during the transformation process. In the case of boride growth, it is considered that the morphology of boride structure significantly influences the adhesion bonding between boride layer and the substrate.^{7–10)} The interaction between the substrate and boride during the growth process is also considered to determine the kind of morphology of boride structure. It was reported that Fe₂B boride layers produced by the conventional boronizing process to have a strong (0 0 1) growth texture¹¹⁾ and grow perpendicular to the surface substrate.¹²⁾ The growth of boride along this direction is faster than in other directions, which is considered the preferential direction for boride

growth, and suppression of the growth of boride structure in other directions is considered to occur by the growth of boride in this direction.¹³⁾ Therefore in order to confirm this important phenomenon, systematic study on this morphological evolution is necessary.

Numerical study by phase-field method is expected to help our better understanding of the morphological growth of boride structure. This method has proven to be useful for investigating pattern formations during phase transformation, such as in the solidification process,^{14–17)} solid-state transformation,^{18–20)} coarsening and grain-growth,^{21–23)} and surface diffusion.^{24,25)} Compared with other numerical approaches, such as dynamic continuum simulation or Monte carlo method, phase-field simulation is thought to be easier since this method can simulate complicated morphological evolution without tracking the interface or surface, which normally cause mathematical difficulties. In the present study, multi-phase-field (MPF) method has been employed in order to perform the morphological evolution of single Fe₂B boride with the focus on the competitive growth between boride seeds with different crystal orientation. By using the MPF method, simulation on the growth of multiple grains can be done, and by setting artificially different orientation on each boride seed, we can expect competitive growth between each boride grain that has a different crystal orientation. In addition, anisotropy of interfacial energy is also introduced, thereby we can expect anisotropy growth of boride seed as was observed in experimental works.^{8,12)} To the author's knowledge, the present work is a volunteer for numerical works that evaluate the morphological evolution of Fe₂B boride. Previous numerical works on boride layer growth have focused on kinetic growth using the mass balance equation as the basic model.^{3,13)} Here, the kinetic growth of boride layer merely depends on the kinetic diffusion of boron without considering the interfacial properties of Fe₂B boride, such as interfacial energy, thickness and mobility, which are very important in determine the mobility of the growth of

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this structure. In the present work, phase-field model for the growth of boride phase is derived from Ginzburg-Landau free energy functional that considers the free energy value from thermo-chemical database and related physical parameters, such as interfacial energy and thickness of the corresponding material. Several important parameters such as the coefficient of free-energy driving force, diffusivity of boron, activation energy and diffusion flux of boron were taken from our previous work.²⁶⁾

2. Phase-field Model

In order to construct a multi-phase-field model for Fe₂B boride evolution, it is considered that a system contains N different phase field variables, $\phi_1, \phi_2, \phi_3, \dots, \phi_N$, where each phase-field is associated with the local density of a different phase or grain. The free energy functional of this system is given by the following equation,²⁷⁾

$$F = \int_V \left[\sum_{\alpha=1}^N \sum_{\beta=\alpha+1}^N \left(-\frac{a_{\alpha\beta}^2}{2} \nabla \phi_\alpha \cdot \nabla \phi_\beta + W_{\alpha\beta} \phi_\alpha \phi_\beta \right) + f_c \right] dV, \quad (1)$$

where, $a_{\alpha\beta}$ and $W_{\alpha\beta}$ are the gradient coefficient and the height of double well potential between α -th and β -th grains, on the other hand f_c is the free energy density in the grains. The phase field, ϕ_α , takes a value of 1 inside the α -th grain, 0 inside other grains, and $0 < \phi_\alpha < 1$ at grain boundary. Each of these phase-field variables are connected by the following constraint,

$$\sum_{\alpha=1}^N \phi_\alpha = 1. \quad (2)$$

We then define the step function, σ_α , with $\sigma_\alpha = 1$ if $0 < \phi < 1$ and $\sigma_\alpha = 0$ elsewhere. Using this step function, the number of locally present phase n is expressed as

Fig. 1 Boride and substrate system with the defined interface orientation of boride.

$$n = \sum_{\alpha=1}^N \sigma_\alpha(x, t). \quad (3)$$

Henceforth, N in the equations 1 and 2 will be substituted by n in the eq. (3).

In addition, in order to create anisotropy growth of Fe₂B boride, anisotropy is introduced by using anisotropy function for the gradient coefficient, a_{ij} . The gradient coefficient is expressed as a function of θ , where θ is the anti-clock wise interface orientation between x -axis and surface normal of boride as illustrated in the Fig. 1. The gradient coefficient that include the anisotropy function is then defined as

$$a_{ij}(\theta) = \frac{\bar{a}_{ij}}{1+u} [1 + u \cos\{\xi(\theta - \theta_0)\}], \quad (4)$$

where $\bar{a}_{ij}$ is constant related to grain boundary thickness and grain boundary energy, θ_0 is the crystal orientation of each grain/phase, u is the strength of anisotropy and ξ is the mode of anisotropy.

Taking into consideration the anisotropy of interfacial energy as $a_{ij} = a_{ij}(\theta)$, the evaluation of phase-field time evolution is obtained explicitly as

$$\frac{\partial \phi}{\partial t} = - \sum_{j=1}^n \frac{M_{ij}^\phi}{n} \left[\sum_{\substack{k=1 \\ (k \neq i \neq j)}}^n (W_{ik} - W_{jk}) \phi_k + \sum_{\substack{k=1 \\ (k \neq i \neq j)}}^n \left[\frac{\partial}{\partial x} \left\{ \frac{(a_{ik}^2 - a_{jk}^2)}{2} \frac{\partial \phi_k}{\partial x} \right\} + \frac{\partial}{\partial y} \left\{ \frac{(a_{ik}^2 - a_{jk}^2)}{2} \frac{\partial \phi_k}{\partial y} \right\} \right] \right. \\ \left. + \sum_{\substack{k=1 \\ (k \neq i \neq j)}}^n \left[\frac{\partial}{\partial x} \left\{ \left(a_{ik} \frac{\partial a_{ik}}{\partial \theta} \frac{\partial \theta}{\partial \phi_{i,x}} \nabla \phi_i \nabla \phi_k \right) - \left(a_{jk} \frac{\partial a_{jk}}{\partial \theta} \frac{\partial \theta}{\partial \phi_{j,x}} \nabla \phi_j \nabla \phi_k \right) \right\} + \right. \right. \\ \left. \left. \frac{\partial}{\partial y} \left\{ \left(a_{ik} \frac{\partial a_{ik}}{\partial \theta} \frac{\partial \theta}{\partial \phi_{i,y}} \nabla \phi_i \nabla \phi_k \right) - \left(a_{jk} \frac{\partial a_{jk}}{\partial \theta} \frac{\partial \theta}{\partial \phi_{j,y}} \nabla \phi_j \nabla \phi_k \right) \right\} \right] \right. \\ \left. + \frac{8}{\pi} \sqrt{\phi_i \phi_j} \Delta G_{ij} \right], \quad (5)$$

where i, j and k are the local grain numbers from 1 to n at a numerical grid point, and subscript $(\cdot)_{,x}$ and $(\cdot)_{,y}$ refer to derivation to x and y -axis. Here, the last term on the right hand side is the phenomenological thermodynamic driving force, where the magnitude of the driving force, ΔG_{ij} , is given by the optimum free energy approach from our previous works.²⁶⁾ The free energy driving force is defined as $\Delta G(X_B, T) = G_m^{\text{Fe}_2\text{B}}(X_B, T) - G_m^\gamma(X_B, T)$. $G_m^{\text{Fe}_2\text{B}}$ and G_m^γ represent molar free energy of Fe₂B and austenite, whereas X_B and T represent molar fraction and temperature respectively. Here, we use the molar fraction, (X_B) , term for the concentration value of boron, and it will be used for all

calculations involving boron concentration in the present works. In addition, as in our previous works, $G_m^{\text{Fe}_2\text{B}}$ and G_m^γ are approached with the assumption that free energy has a parabolic relationship with concentration and their minimum free energy is similar with the free energy provided by the thermo-chemical database (SGTE database). The free energy of austenite and Fe₂B boride in the present works are then approached with the following equations²⁶⁾

$$G_m^\gamma = G_{\text{AV}} + 3.2 \times 10^6 (0.1 - X_B)^2, \quad (6)$$

$$G_m^{\text{Fe}_2\text{B}} = 3.8 \times 10^6 (X_B - 0.33)^2 + G^{\text{Fe}_2\text{B}}, \quad (7)$$

where ${}^oG^{\text{Fe}_2\text{B}}$ and G_{AV} are the reference energy terms for Fe_2B and austenite, and are obtained from the optimization of SGTE database value by Van Rompaey *et al.*²⁸⁾

In addition, W_{ij} , $\bar{a}_{ij}$ and M_{ij}^ϕ are energy barrier height, reference value of gradient coefficient and mobility of phase-field respectively, and are related to the material parameters as follow²⁹⁾

$$W_{ij} = \frac{4\gamma_{ij}}{\delta}, \quad \bar{a}_{ij} = \frac{2}{\pi} \sqrt{2\delta\gamma_{ij}}, \quad M_{ij}^\phi = \frac{\pi^2}{8\delta} M_{ij}, \quad (8)$$

where δ is the grain boundary thickness, and γ_{ij} and M_{ij} are the grain boundary energy and mobility between i -th and j -th grains.

On the other hand, in order to provide the diffusion evolution of boron atoms during the growth of Fe_2B boride from austenite phase, we employ diffusion equation in a multi-phase system proposed by Tiaden *et al.*³⁰⁾ The total boron concentration is defined as a linear function of the local boron concentration, b_i^Λ , weighted by the phase-field variables, ϕ_i . The local boron concentration is boron concentration in each grain, where it may refers to boron concentration of Fe_2B or Austenite. Initial boron concentration in Fe_2B is set at 0.33 molar fraction of boron, which is a stoichiometric compound of Fe_2B . On the other hand, initial boron concentration in austenite phase is set at 0.0035 molar fraction of boron. The total boron concentration is written as,

$$b = \sum_{i=1}^N \phi_i b_i^\Lambda, \quad (9)$$

where the subscripts i and Λ denote the number of grains ($i = 1, \dots, N$) and the phase ($\Lambda = \text{Fe}_2\text{B}$ or γ) respectively. Hereafter, since we consider 2 phases conditions, Fe_2B and austenite phases, the term of bulk phase corresponds to both of these phases. On the other hand boron concentration value here refers to molar fraction of boron (X_B), as was previously described.

The diffusion equation for total boron concentration is expressed by the sum of diffusion fluxes of boron atoms in the individual grains as

$$\frac{\partial b}{\partial t} = \nabla \cdot \left(\sum_{i=1}^N \phi_i D_b^\Lambda \nabla b_i^\Lambda \right), \quad (10)$$

where D_b^Λ is denoted as the diffusion coefficient of boron in the bulk Λ and is given by $D_b^\Lambda = D_b^{o\Lambda} \exp(-Q_\Lambda/RT)$, with $D_b^{o\Lambda}$ is the pre-exponential factor of diffusivity in the bulk phase, Q_Λ is the activation energy in the bulk Λ phase, and R and T are the gas constant and temperature respectively.

By having eqs. (5) and (10), we provide a system of coupled differential equations to calculate the evolution of phase-field variable and boron concentration with location and time which can describe the kinetic growth from austenite to Fe_2B phase.

3. Numerical Algorithm to Solve MPF Equations

The usage of eq. (5) in the present study indicates that we do not have to solve eq. (5) on the grid point with $n = 1$ and it is sufficient to save the values of not N but n phase fields. In addition, as is explained in the Ref. 29), the following algorithm is applied to solve eq. (5) very efficiently,

(1) Solve eq. (5) for n phase fields.

(2) During (1), if ϕ_i at previous time step is zero and the calculated increment $\Delta\phi_i$ has a minus value, i -th phase field is put away from the phase field group on the grid point. And, steps (1) and (2) are repeated until all phase fields that satisfy the above condition are removed.

(3) The phase fields are rearranged in the order of largest to smallest. Phase fields from the largest until n_ϕ -th are recorded. Here, n_ϕ is a predefined maximum number of the recorded phase field.

(4) The phase fields are replaced by $\phi_i^* = \phi_i / \sum_{j=1}^{n_\phi} \phi_j$ so as to satisfy $\sum_{i=1}^{n_\phi} \phi_i^* = 1$.

(5) If a phase field is not saved at a grid point of (l, m) and its value at grid points of $(l \pm 1, m \pm 1)$, or the nearest four neighbors, is not zero, the phase field is added to the phase field group on the grid (l, m) .

The above algorithm is taken from Kim *et al.*³¹⁾ and the iteration in steps (1) and (2) is added by Takaki *et al.*²⁹⁾ in order to achieve higher accuracy.

4. Numerical Conditions

The two-dimensional simulation is performed for the growth of Fe_2B boride from austenite phase. The computational domain size is $400\Delta x \times 200\Delta x$, where Δx is the grid size, which is set at $0.05 \mu\text{m}$. Initially 20 seeds of Fe_2B boride with different orientations are nucleated at the bottom side of the computational domain, as shown in the Fig. 2. Here, the bottom side is set as the substrate surface, where boron atoms diffuse and form the boride layer. Based on this description, the Neumann boundary condition is applied at the top and bottom of computational domain, while the gradient value is derived from the flux (J) of boron entering the surface, $J = -D\partial b/\partial y$, where D is diffusivity of boron in Fe_2B , $1.19 \times 10^{-13} \text{m}^2/\text{s}$,²⁶⁾ and $\partial b/\partial y$ is the average of surface concentration gradient. On the other hand, periodic boundary conditions are applied at the both left and right side of computational domain, representing that the computational domain used here is a small part of the boride-austenite steel substrate system.

In the Fig. 2, each boride seed is given a number from 1 to 20. Each boride seed number refers to a different orientation value given in the Table 1. By applying different orientation values of boride, it is expected to observe competitive growth between these boride seeds. Here, the austenite substrate orientation is set at crystal orientation of 90° .

Fig. 2 Initial Fe_2B borides colony with its number distribution of crystal orientation (the grey scale shows distribution of phase-field).

Table 1 Related number of boride seeds from Fig. 2 and their orientation angle.

Boride seeds No	Boride seeds orientation (degree)	Boride seeds No	Boride seeds orientation (degree)	Boride seeds No	Boride seeds orientation (degree)	Boride seeds No	Boride seeds orientation (degree)
1	24.88	6	57.26	11	75.97	16	0.16
2	79.30	7	1.32	12	70.70	17	19.40
3	20.69	8	46.70	13	89.99	18	44.81
4	38.33	9	83.82	14	52.85	19	85.10
5	69.67	10	9.45	15	9.16	20	51.83

On the other hand, other parameter's conditions are as follows, the interface thickness, $\delta = 10\Delta x$; interface energy, $\gamma = 1 \text{ J/m}^2$; and time increment, $\Delta t = 0.0656 \text{ s}$. In addition grain boundary mobility (M), is defined as $M = M_0 \exp(-Q/RT)$, where M_0 is pre-exponential factor and is set at $5 \times 10^{-5} \text{ m}^2/\text{Ns}$, Q is activation energy and is set at $2.56 \times 10^6 \text{ J/mol}$,²⁶⁾ R is gas constant at $8.314 \text{ m}^3 \cdot \text{Pa} \cdot \text{K}^{-1} \cdot \text{mol}^{-1}$ and T is temperature at 1323 K . The value of pre-exponential factor for diffusivity in both austenite and Fe₂B boride, as well as their activation energy for the calculation of eq. (10), were taken from our previous works²⁶⁾ based on the reference values provided by Keddari.³²⁾ These parameters were set at $4.4 \times 10^{-8} \text{ m}^2/\text{s}$ and $1.13 \times 10^{-6} \text{ m}^2/\text{s}$ for pre-exponential diffusivity in boride and austenite, and 1.51×10^5 and $8.15 \times 10^4 \text{ J/mol}$ for activation energy in boride and austenite. Anisotropy strength, u , is set at 0.3 whereas the anisotropy mode, ξ , is set at 2, representing two-fold symmetry. Experimentally, it was observed that, due to the tendency of Fe₂B crystals to grow along a direction of minimum resistance perpendicular to the external surface,⁸⁾ boride structures prefer to have morphology in the form of a columnar shape with 'toothing' characteristics on the tips.⁷⁻¹⁰⁾ As this type of structure has two-fold symmetry, the present simulation work also uses two-fold symmetry for the calculation of eq. (4), in order to obtain the appropriate approach for simulating this type of shape structure.

5. Numerical Results

In order to understand boride seed characteristics that grows at different crystal orientations, Fig. 3 shows the growth of single seed of boride, which having different crystal orientation. Here, substrate orientation is set at 90 degrees and computational domain at $100\Delta x \times 100\Delta x$. We can observe from this figure that in general the tip interface shows a thicker interface as compared with the side interface. The variations of the interface width can be explained with the facts that the interface width varies with orientation and is proportional to the anisotropy function.³³⁾ On the other hand, boride seed with a crystal orientation of 90 degrees grows perpendicular to the substrate surface and shows faster growth at this direction compared with other seeds. In addition, as the crystal orientation approaches 90 degrees faster growth is observed. According to these facts, we can expect that boride seeds at a crystal orientation of 90 degrees will provide better directions for the growth of boride. This condition also agrees with previous experimental facts that boride grains tend to grow perpendicular to the substrate, which is considered giving the minimum resistance for crystal growth.¹⁶⁾

Figure 4 shows the growth of multiple-seeds of boride with the initial condition as is described in the Fig. 2 and Table 1. We can observe that competitive growth of boride seeds with different crystal orientations occur. Boride seeds with higher orientation and approaching 90 degrees or perpendicular to the substrate grow higher and suppress the growth of its neighbors, which having a lower orientation angle. As is the case of boride seeds number 1, 2, 3, 4 and 5, which have an orientation angle of 24.88, 79.30, 20.69, 38.33 and 69.67 degree respectively. It can be observed, that boride seeds number 2 and 5 grow fast compared with their neighbor and suppress the growth of boride grain with lower orientation angles. This condition agrees with previous results for the growth of single boride seed, that boride seeds with orientation angles approaching 90 degrees grow fast

Fig. 3 The growth of single boride seeds with different crystal orientation (the grey scale shows distribution of phase-field).

Fig. 4 The growth of multiple-seed of boride with different crystal orientation (the grey scale shows distribution of phase-field).

compared with lower orientation angle. Real Fe_2B boride growth as observed in the experimental works,^{12,34)} structures might grow at a different crystal plane. And it was observed, Fe_2B boride that grow on the (001) crystal plane, which is perpendicular to the substrate, grow faster than the other crystal planes and suppress the growth of Fe_2B boride on other crystal planes. Therefore the present crystal orientation refers to the growth of Fe_2B boride seeds at different crystal planes in the real material, and by using this approach we can capture competitive growth of boride seeds with different crystal orientations, which is observed in the experimental observation.

Figure 5 shows the time slice for the growth of multiple-seeds of boride, while Fig. 5(a) shows variation of initial boride seed size. Figure 5(b) uses homogeneous initial size of boride similar with the condition in the Fig. 4. Here, we consider that different sizes of boride seeds might have different surface curvature and thus have a chance to influence interfacial directions. Therefore, we should try to evaluate whether variations of initial size of boride seeds have significant influences on their growth, which have different crystal orientation. To perform this evaluation, the distribution orientation of boride seeds for Fig. 5(a) and (b) is similar to the previous condition in the Fig. 4, and in the case of Fig. 5(b), the initial seed size is also similar to Fig. 4. In Fig. 5(a), several boride seeds with higher orientation angles are set at smaller sizes compared with its neighbor, i.e. in the case of boride seeds number 2 and 19 which have smaller sizes compared with its neighbor. It can be observed in the Fig. 5(a), that the bigger seeds with lower orientation angles can ‘compete’ against the smaller but higher orientation boride seeds only at the initial stage, i.e. in the case of boride seeds number 2 and its neighbors up to 65.6 seconds. After this time, boride seed 2 starts to grow higher and suppress the growth of its neighbors. On the other hand in the Fig. 5(b), we can observe that boride seed number 2 starts to surpass the growth of its neighbor earlier than in Fig. 5(a). In addition, looking at the final boride layer produced by the two cases, we can only observe small differences in morphologies between the two cases. By having these facts, we may consider that the condition of initial boride seeds size might affect kinetic growth only at the early stages, whereas the later stage shows similar result in both cases, and the growth is dependent on the crystal orientation of each boride seed. Therefore, in general we can conclude conditions for initial

Fig. 5 The growth of multiple-seeds of boride with non-homogeneous size of initial seeds of boride (a), and homogeneous size of initial seeds of boride (b) (in both cases, the distribution orientation is similar and the grey scale shows distribution of phase-field).

boride seeds size do not significantly influence preferential growth, where the bigger seeds at lower orientation angles can not suppress the growth of smaller boride seeds with higher orientation angles.

Unfortunately, compared with experimental observation of Fe_2B boride that shows sharp structure of this phase,¹²⁾ the present works has not been able to capture a sharp boride

structure. Therefore, it will be our homework in the future to simulate sharper structures of boride, while maintaining boride growth as it is observed in experimental works.

6. Conclusions

Multi-phase-field simulations for the study of the growth of Fe₂B boride on the steel substrate have been well performed with the main focus on the effect of structure orientation on the morphological growth of Fe₂B boride. In order to obtain an appropriate growth of this structure, a consideration on the anisotropy dependence of interfacial energy is taken into account in the phase-field evolution model and variations of boride seeds with different orientations have also been taken into consideration. It is observed that a single Fe₂B boride, which grows perpendicular to the substrate, shows preferential growth and is in a good agreement with previous experimental observations. In addition, competitive growth between Fe₂B seed at different orientation angles can be observed, where boride seeds with orientation angles approaching preferential direction grow higher and suppress their neighbors with lower orientation angles. On the other hand, it was also observed that preferential growth direction for boride seeds is independent of the initial sizes. It is necessary for the future works to simulate a sharper structure of boride in order to simulate the more appropriate morphology of Fe₂B boride layers.

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