Dynamic Accommodation of Internal Stress and Selection of Crystallographic Orientation Relationship in Pearlite

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To gain a deeper understanding of the dynamic accommodation mechanism of the internal stress in pearlite originating from the lattice misfit between ferrite and cementite phases, the lattice parameter ratios of cementite, $b_{\theta}/a_{\theta}$ and $c_{\theta}/a_{\theta}$, were locally analyzed in detail using the electron backscatter diffraction (EBSD) technique. The EBSD analysis revealed that the lattice parameter ratios of the cementite lamellae clearly differed from those of the spheroidized cementite particles, indicating that pearlite has a certain amount of internal stress as long as it maintains a lamellar structure. The internal stress in pearlite gradually decreased during the isothermal holding at 923 K after the pearlitic transformation due to the interfacial atomic diffusion of Fe atoms. However, a comparison with the theoretical values obtained from the Pitsch-Petch orientation relationship revealed that a large amount of internal stress had been already accommodated during the pearlitic transformation by the introduction of misfit dislocations and structural ledges on ferrite/cementite lamellar interfaces, i.e., the internal stress of pearlite is dynamically reduced by two different processes; built-in accommodation upon pearlitic transformation and additional time-dependent relaxation after pearlitic transformation. On the other hand, the analysis of the EBSD and neutron diffraction results yielded intriguingly different lattice parameters of cementite, indicating that various crystallographic orientation relationships between ferrite and cementite coexist in pearlite. Furthermore, the elastic strain energy analysis suggests that the invariant-line criterion on the ferrite/cementite interface plays an important role for the selection of orientation relationships in pearlite.

KEY WORDS: pearlite; misfit; internal stress; built-in accommodation; diffusional relaxation; cementite; electron backscatter diffraction.

1. Introduction

Pearlite (P) is a eutectoid transformation microstructure consisting of ferrite ($\alpha$) and cementite ($\theta$) in the Fe–C alloy, having a lamellar structure resulting from the cooperative growth of these phases during the eutectoid reaction. In addition to the lamellar structure, pearlite also has hierarchical substructures known as pearlite colonies, where the cementite lamellae are aligned in the same direction.1) A larger region composed of several colonies in which the ferrite matrix has an identical crystal orientation is defined as a pearlite block or nodule.2) Previous studies on the hierarchical substructures and their relationship with the mechanical properties of pearlitic steels reported that the strength and ductility are enhanced by the refinement of the interlamellar spacing3) and block size,2) respectively. In order to control the mechanical properties of pearlitic steels, it is essential to develop a thorough understanding of the pearlitic transformation mechanism including the formation of the hierarchical substructures. The pearlitic transformation is controlled by C diffusion as both ferrite and cementite precipitate simultaneously from austenite ($\gamma$) via C-partitioning between them. The Zener-Hillert model has been widely recognized to explain the C diffusion-controlled process based on the local equilibrium among the $\gamma/\alpha/\theta$ phases, especially because the pearlitic transformation occurs in the steady state condition. On the other hand, the cementite lamellae maintain a specific orientation relationship with respect to the ferrite
matrix in pearlite. A recent study\(^5\) reported that a large amount of internal stress is generated by lattice misfit between the ferrite and cementite phases when the \(\alpha/\theta\) lamellar interphase is coherent, and that the generated elastic strain energy is equivalent to the driving force for the pearlitic transformation. Furthermore, it was proposed that the misfit dislocations are dynamically introduced at the \(\alpha/\theta\) lamellar interphases on the pearlitic transformation that reduce the elastic strain energy, leading to an acceleration of the pearlitic transformation. Zhou and Shiflet\(^6\) investigated the pearlite growth mechanism by transmission electron microscopy and reported that a growth ledge on a \(P/\gamma\) growing interphase converts into a direction step (structural ledge) on the \(\alpha/\theta\) lamellar interface. Consequently, a lot of misfit dislocations are generated on the \(\alpha/\theta\) lamellar interphases. Considering that the reduction of elastic strain energy substantially increases the total driving force for the pearlitic transformation, it may be assumed that the pearlitic transformation is controlled not only by C diffusion, but also the dynamic accommodation of the internal stress. Therefore, the internal stress state of pearlite must be evaluated.

In this regard, we present a detailed evaluation of the internal stress state in pearlite based on the analysis of neutron diffraction and electron microscopy experiments.\(^5\) Besides, we attempt to simulate the transformation model taking into account the dynamic accommodation of the internal stress for a complete understanding of the pearlitic transformation. In this study, the measurement of the lattice parameter ratio using the electron back scatter diffraction (EBSD) technique was conducted for cementite lamellae distributed in the ferrite matrix to evaluate the variation of internal stress state in pearlite during isothermal holding and cementite spheroidization annealing after the pearlitic transformation. The results were compared with the average lattice parameters of cementite evaluated by neutron diffractometry, and the selection of the \(\alpha/\theta\) crystallographic orientation relationship in pearlite is discussed based on the analysis.

2. Experimental Procedure

A steel with near-eutectoid composition (0.83% C–1.46% Mn, in mass%) was used in this study. The C–Mn balance was adjusted to ensure that the pearlitic transformation is not controlled by the long-range Mn diffusion, while keeping sufficient quench hardenability. A hot-rolled sheet homogenized at 1 523 K for 36 ks was subjected to the heat treatment as shown in Fig. 1. A sample of volume 20 × 10 × 10 mm\(^3\) (length × width × thickness) was cut from the hot-rolled sheet and solution treated at 1 323 K for 1.8 ks, and then isothermally held at 923 K for 300 s and 600 s in a salt bath furnace to obtain specimens of the partially and fully transformed materials, respectively (lamellar pearlitic material). The incubation period for the pearlitic transformation\(^5\) was 100 s at 923 K under this heat treatment condition. For spheroidization of the lamellar cementite, the lamellar pearlitic material obtained by isothermal holding at 923 K for 600 s was annealed additionally at 923 K for 36 ks after cold rolling it at 40% thickness reduction (spheroidized pearlitic material). The microstructure was observed using an optical microscope and a scanning electron microscope (SEM developed by JEOL Ltd., JSM-7001F). The surface to be observed was automatically polished with colloidal silica and then etched with 5% nital etchant. The crystal orientation was analyzed using the EBSD technique. EBSD patterns were captured by a CCD camera attached to the SEM and analyzed using the OIM Data Collection ver. 7.1.0. The data thus obtained was displayed as an inverse pole figure (IPF) map with the OIM Analysis ver. 7.3.0. These softwares were developed by TSL solutions. The working distance, acceleration voltage, and step size for the EBSD analysis were set at 15.0 mm, 15.0 kV, and 0.01 μm, respectively. In addition, the microscopic lattice parameter ratio of cementite phase was evaluated by the EBSD technique to analyze the internal stress state in the pearlite. In the OIM Data Collection, crystal orientation is indexed by comparing an experimentally obtained EBSD pattern with a theoretical one calculated from the crystal structure and lattice parameters. The width of each band in the EBSD pattern corresponds to an interplane spacing of the diffraction plane, although its accurate measurement is impossible owing to the low spatial resolution of the CCD camera. Therefore, the crystal orientation is actually indexed from the angular relation between some of the diffraction bands in the EBSD analysis. The information of the angle and position of each EBSD band is transformed in two-dimensional coordinates using the Hough conversion, and then the angular deviation between the theoretical values and experimental measurements (fit value) is minimized in the coordinates. Therefore, similar to the analysis method used for cementite characterization as an orthorhombic crystal (Pnma), the ratios of the three lattice parameters, \(a_\theta\), \(b_\theta\), and \(c_\theta\) can be evaluated with the fit value as an indication instead of their independent values. With this advantage of the EBSD technique, several thousand EBSD data points obtained from a single cementite particle were reanalyzed by changing \(b\) and \(c\) arbitrarily while setting \(a_\theta = 0.5090\) nm in the OIM Data Collection to evaluate the average fit value. After investigating the change

\(^{1}\): The diameter of the largest nodule isolated within an untransformed austenite matrix was measured as a function of the isothermal holding time. Since the pearlitic transformation undergoes in the steady state, the time variation of the diameter should be constant. Therefore, the critical time estimated by extrapolating the constant time variation was regarded as the incubation period for the pearlitic transformation.\(^{5}\)

![Image](https://example.com/image1.png)

**Fig. 1.** Heat treatment route to obtain lamellar and spheroidized pearlite.
in the fit value as a function of $b_\theta$ and $c_\theta$, the $b_\theta/a_\theta$ and $c_\theta/a_\theta$ values were evaluated accurately from the minimum fit value. In the OIM Data Collection, $a_\theta = 0.5090$ nm, $b_\theta = 0.6748$ nm, and $c_\theta = 0.4523$ nm were set as the initial lattice parameters of cementite, and at least three EBSD bands were used for calculation in the Hough transformation. On the contrary, the macroscopic lattice parameters of the cementite phase were evaluated using in situ neutron diffractometry with the Engineering Materials Diffractometer (TAKUMI, BL19) operated at the Material and Life science Experimental Facility (MLF) at the Japan Proton Accelerator Research Complex (J-PARC). For the neutron diffraction measurements, an accelerator output energy of 150 kW, an incident slit area of $5 \times 5$ mm$^2$, and a receiving slit width of 5 mm were used. Austenitization of a specimen with a volume of 5 mm$^3$ was conducted by heating it from ambient temperature to 1 323 K at 10 K/s. After holding the specimen at 1 323 K for 900 s, its austenitized form thus obtained was cooled at 20 K/s and then isothermally held at 873 K for 10.8 ks. The diffraction peaks from the cementite phase were continuously measured during the isothermal holding and the corresponding lattice parameters were evaluated using the Z-Rietveld software developed by J-PARC with the orthorhombic cementite of space group Pnma and No. 62 used as reference.

3. Results and Discussion

3.1. Accommodation of Internal Stress in Pearlite during and after Transformation

3.1.1. Measurement of Lattice Parameter Ratio of Cementite by the EBSD Technique

Figure 2 shows the EBSD-analyzed phase and the IPF maps of the lamellar pearlitic material formed at 923 K for 600 s (a, b) and the spheroidized pearlitic material (c, d). RD and ND indicate the rolling and normal directions of the rolled face on cold rolling, respectively. The EBSD analysis for the cementite phase was performed with the initial lattice parameters and the corresponding results with a confidence index value greater than 0.1 are shown at the extreme right of Fig. 2. Although it is difficult to index the crystal orientation of cementite owing to its lower crystal symmetry and very fine lamellar structure, both ferrite and cementite could be identified in this figure and preferential etching of the ferrite matrix led to the increase in the cementite area fraction. The lamellar pearlitic material (a, b) exhibits a lamellar structure with a slightly waved interface and an identical lamellar arrangement. These results indicate that the observed area was in a pearlite colony. Furthermore, it is found that the crystal orientations of both the ferrite and cementite phases changed continuously in the colony. Previous reports on the simultaneous crystal rotation of both the phases suggest the existence of an internal stress in pearlite. On the contrary, in the spheroidized pearlitic material (c, d), it was found that the spheroidized cementite particles were distributed at the boundaries of the equiaxed and fine ferrite matrix. This reveals that the spheroidization of the cementite lamellae and the recrystallization of pearlitic ferrite had been completed. The crystal orientation appeared to be identical in both ferrite and cementite, suggesting the release of the internal stress. To identify the $\alpha/\theta$ orientation relationship in the lamellar pearlitic material, the crystal orientations of cementite and ferrite were analyzed at point A (white circle) and B (black circle), respectively. Figure 3 shows the pole figures for (a) (010)$_\alpha$ and (b) [100]$_\alpha$ of cementite, and (c) (215)$_\alpha$ and (d) [311]$_\alpha$ of ferrite. Upon comparing both the phases we find that (010)$_\alpha$ and [100]$_\alpha$ are nearly parallel to (215)$_\alpha$ and [311]$_\alpha$, respectively, as indicated by the dotted circles. In other words, the Pitsch-Petch orientation relationship ((010)$_\alpha$ // (215)$_\alpha$, [100]$_\alpha$ // [311]$_\alpha$) holds true between the lamellar $\alpha/\theta$. It has been previously reported that the pearlitic ferrite and cementite crystals cooperatively rotate with an identical rotational axis and angle. A similar trend confirmed in the lamellar pearlite revealed that the Pitsch-Petch orientation relationship had been maintained at the lamellar $\alpha/\theta$ interface within the colony. On the other hand, a specific orientation relationship was not identified between $\alpha/\theta$ in the spheroidized pearlitic

![Fig. 2.](image-url) (a), (c) Phase and (b), (d) IPF maps showing (a), (b) lamellar pearlite isothermally transformed at 923 K for 600 s and (c), (d) spheroidized pearlite annealed at 923 K for 36 ks after 40% cold rolling.)
material due to recrystallization of the ferrite matrix.

Figure 4 shows the dependence of the average fit value of cementite lamellae on the lattice parameters $b_0$ and $c_0$. The measurement was carried out with the same data as shown in Fig. 2(a, b), keeping $a_0 = 0.5090$ nm. The fit value varied as a continuous function of the lattice parameters, exhibiting a curved surface projecting downward. This resulted in the minimization of the fit value at $b_0 = 0.6988$ nm and $c_0 = 0.4623$ nm of the lamellar pearlitic material, while the spheroidized pearlitic material (Fig. 2(c, d)) yielded the minimum fit value at $b_0 = 0.6688$ nm and $c_0 = 0.4523$ nm. These results suggest that the cementite lattice parameter differs between the two materials, which may be caused by alloy partitioning between the ferrite and cementite phases during spheroidization. However, X-ray diffractometry and chemical analysis carried out with powder cementite obtained from the extraction residue of both the materials and chemical analysis carried out with powder cementite during spheroidization. However, X-ray diffractometry and chemical analysis carried out with powder cementite obtained from the extraction residue of both the materials confirmed that there is little difference in the lattice parameter and solute Mn concentration of cementite between the lamellar and spheroidized pearlitic materials. These results imply that the lattice parameters of cementite are different between lamellar and spheroidized pearlitic materials when cementite exists within the ferrite matrix that is caused by differences in the microstructure, such as cementite morphology and the $\alpha/\theta$ crystal orientation relationship.

3.1.2. Change in the Lattice Parameter Ratio of Cementite on Isothermal Holding after the Pearlitic Transformation

Figure 5 shows the rate of change in lattice parameter ratios of cementite, $R_{b/a}$ and $R_{c/a}$, in lamellar and spheroidized pearlitic materials. In addition to the isothermal holding at 923 K for 600 s, the holding time was shortened to obtain another lamellar pearlitic material where the pearlitic transformation had not yet completed. For the lamellar pearlitic material, EBSD analysis was carried out at the central part of an isolated pearlite nodule in order to evaluate the cementite that was formed at an early stage of the pearlitic transformation. Furthermore, the theoretical values, $R_{b/a}^*$ and $R_{c/a}^*$ were calculated by the theory of elasticity under the Pitsch-Petch orientation relationship (O.R.). Here, $R_{b/a}$ and $R_{c/a}$ are the rate of change in $b_0/a_0$ and $c_0/a_0$ respectively, based on the lattice parameters of cementite extracted from the spheroidized pearlitic material ($a_0^* = 0.5074$ nm, $b_0^* = 0.6742$ nm, $c_0^* = 0.4520$ nm) and defined by the following equations.

$$R_{b/a} = \frac{b_0}{a_0} - \frac{b_0^*}{a_0^*} \frac{a_0}{a_0^*}$$

and

$$R_{c/a} = \frac{c_0}{a_0} - \frac{c_0^*}{a_0^*} \frac{a_0}{a_0^*} \hspace{1cm} (1)$$

The first noticeable characteristic is that the cementite in the spheroidized pearlitic material has almost the same lattice parameters as the cementite relieved from the ferrite matrix due to comparable $(R_{b/a}, R_{c/a}) = (0, 0)$. On the contrary, $R_{b/a}$ and $R_{c/a}$ values of the lamellar pearlitic materials are clearly in the first quadrant and increase upon decreasing the holding time. It should be noted here that the theoretical $R_{b/a}^*$ and $R_{c/a}^*$ values under the Pitsch-Petch O.R. were predicted by extrapolating the measured data. This suggests that the difference in the cementite lattice parameter ratio between the lamellar and spheroidized pearlitic materials could be attributed to the internal stress generated by the lattice misfit between $\alpha/\theta$. This internal stress decreases during the isothermal holding and spheroidized annealing after the pearlitic transformation. This implies that the internal stress in pearlite is continuously decreased by the transition of $\alpha/\theta$ interface coherency from coherent to semi-coherent and incoherent interfaces.

3.1.3. Relaxation of the Elastic Strain on the Pearlitic Transformation and Subsequent Holding

$R_{b/a}$ and $R_{c/a}$ in lamellar pearlitic materials shown in Fig. 5
are plotted in Fig. 6 as a function of the holding time after solution treatment. Additionally, the theoretical values of \( R_{\alpha \theta}^* \) and \( R_{\alpha \theta}^{**} \) under the Pitsch-Petch O.R. were plotted for \( t = 100 \) s, which corresponds to the start time for the pearlitic transformation. \( R_{\alpha \theta}^* \) and \( R_{\alpha \theta}^{**} \) decrease monotonically as the holding time increases, which reveals that the internal stress generated by the misfit strain between \( \alpha/\theta \) continuously reduced during the isothermal holding after the pearlitic transformation. Onaka and Kato\(^{14}\) investigated the high temperature relaxation mechanism of the elastic strain in the spherical shaped second phase \( e_{ij}^{*} \) and proposed that the relaxation behavior of \( e_{ij} \) should be described according to the attenuation function with the holding time, \( t \), as follows:

\[
e_{ij} = e_{ij}^* \exp \left( -\frac{t}{\tau} \right) \quad \text{......(2)}
\]

where \( e_{ij}^* \) is the initial elastic strain generated in the second phase precipitate with a spherical shape, and \( \tau \) is the relaxation time necessary for obtaining the relation \( e_{ij} = 1/e \times e_{ij}^* \) (e: the base of natural logarithm). Furthermore, they conducted a micromechanical analysis of the elastic strain relaxation mechanism by the interface diffusion between the matrix and second phases and established a relation with the extent of accommodation may depend on their densities. From this point of view, it is essential to develop a complete understanding of the dynamic accommodation mechanism of the internal stress on the pearlitic transformation by carrying out further investigation on the temperature dependence and its correlation with the thermal activation process.

### 3.2. Difference between Microscopic and Macroscopic Internal Stresses in Pearlite

#### 3.2.1. Effect of Orientation Relationship between Ferrite and Cementite on the Internal Stress in Pearlite

In the previous section, the microscopic evaluation of the lattice parameter ratio of cementite clarified that the internal stress induced by the lattice misfit between \( \alpha/\theta \) is dynamically accommodated by two different mechanisms.

\[ R_{\alpha \theta} \]

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**Fig. 5.** Distribution of \( R_{\alpha \theta} \) and \( R_{\alpha \theta}^{**} \) of lamellar and spheroidized cementite. The theoretical values under the Pitsch-Petch orientation relationship (O.R.) is also plotted as a solid mark. **Fig. 6.** Changes in \( R_{\alpha \theta} \) and \( R_{\alpha \theta}^{**} \) in lamellar cementite as a function of isothermal holding time at 923 K. The theoretical values under the Pitsch-Petch O.R. is also plotted as solid marks.
In addition, we conducted the macroscopic evaluation of the lattice parameter of pearlitic cementite with the same specimen using neutron diffractometry. In Fig. 7, the time dependence of cementite lattice parameters, $a_0^\alpha$, $b_0^\alpha$, and $c_0^\alpha$, is plotted for isothermal holding at 873 K after completion of the pearlitic transformation. Comparison of $R_{b/a}$ and $R_{c/a}$ calculated from the results presented in Fig. 7 with those presented in Fig. 5 revealed that they differed depending on the evaluation method used, i.e., the microscopic analysis by EBSD and the macroscopic one by neutron diffractometry.

The macroscopic lattice parameter ratios were evaluated to be ($R_{b/a}$, $R_{c/a}$)$_{Neutron}$ = (0.00297, −0.000969) with the cementite lattice parameters, $a_0^\beta$, $b_0^\beta$, and $c_0^\beta$, as references as mentioned above as the pearlitic cementite had $a_0 = 0.5116$ nm, $b_0 = 0.6818$ nm, and $c_0 = 0.4553$ nm just after the pearlitic transformation at 873 K, while, they were ($R_{b/a}$, $R_{c/a}$)$_{EBSD}$ = (0.03175, 0.01296) in the EBSD analysis (see Fig. 5), which are much higher than ($R_{b/a}$, $R_{c/a}$)$_{Neutron}$. The EBSD analysis is a scanning electron microscope-based microstructural, crystallographic characterization technique, wherein some fraction of the incident electrons entering the sample may backscatter. Since the penetration depth of electrons is generally lesser than 100 nm in metallic materials, attention should be paid to a loss of restriction due to the free surface. However, the difference in the lattice parameter ratio cannot be explained by the free-surface effect since ($R_{b/a}$, $R_{c/a}$)$_{EBSD}$ is larger than ($R_{b/a}$, $R_{c/a}$)$_{Neutron}$. In addition, there is a small difference between the thermal expansion coefficient of ferrite and cementite, and thus, the effect of thermal stress shall not be attributed to the difference in the measured temperature. Indeed, the full width at half maximum of the pearlitic ferrite measured by X-ray diffractometry showed insignificant changes before and after cooling from the annealing temperature. On the other hand, attention should also be paid to the size difference of the evaluated area. EBSD analysis can evaluate the lattice parameter ratio of cementite at the microscopic scale, while the average lattice parameter of a large number of cementite particles can be determined by neutron diffractometry as it is based on the macroscopic diffraction phenomenon in a bulk sample. Considering that the lattice parameter dependence of cementite can be attributed to the lattice misfit between α-Fe and that it is strongly affected by O.R. between them, the difference between ($R_{b/a}$, $R_{c/a}$)$_{EBSD}$ and ($R_{b/a}$, $R_{c/a}$)$_{Neutron}$ suggests the coexistence of various O.R.s, i.e., although the Pitsch-Petch O.R. was identified in the lamellar pearlitic material as shown in Fig. 2, another O.R. may be satisfied in a different area. In fact, for a different selected area as shown in Fig. 8, it was confirmed that the Isaichev O.R. ((0 1 1)$_{\alpha}$/ (1 1 2)$_{\gamma}$ [0 T 1]$_{\alpha}$ // [1 T 0]$_{\gamma}$) was appropriate between the lamellar ferrite and cementite in the same sample as shown in Fig. 2. We shall now discuss the effect of the coexistence of O.R.s on the internal stress of pearlite.

Zhou and Shiflet pointed out the possibility of the following four O.R.s in pearlite according to the lattice correspondence of Fe atoms on a habit plane between α/θ:

(a) Bagaryatsky O.R.$^{19}$ (0 1 0)$_{\alpha}$/ (1 1 2)$_{\gamma}$ [001]$_{\alpha}$  // [1 T]$_{\gamma}$/ [011]$_{\gamma}$

(b) Isaichev O.R.$^{20}$ (0 1 1)$_{\alpha}$/ (1 1 2)$_{\gamma}$ [0 T1]$_{\alpha}$ // [1 T0]$_{\gamma}$/ [0 11]$_{\gamma}$

(c) Pitsch-Petch O.R.$^{21,22}$ (0 1 0)$_{\alpha}$/ (2 2 5)$_{\gamma}$ [0 0 1]$_{\alpha}$ // [3 1 1]$_{\gamma}$/ [1 0 0]$_{\gamma}$ 2.6° // [3 1 1]$_{\gamma}$/ [1 0 0]$_{\gamma}$ and

(d) Unknown O.R. (0 1 1)$_{\alpha}$/ (2 2 5)$_{\gamma}$ [0 0 1]$_{\alpha}$ 2.6° // [3 1 1]$_{\gamma}$/ [1 0 0]$_{\gamma}$ 2.6° // [1 3 1]$_{\gamma}$

Except for the fourth O.R., all the others have been identified in previous studies. In these O.R., each lattice misfit could be geometrically evaluated as two principal strains, $\varepsilon_{11}'$ and $\varepsilon_{22}'$, with the two-dimensional lattice correspondence of the Fe atoms between the ferrite and cementite on their respective habit planes. Furthermore, the elastic strain energy can be calculated with $\varepsilon_{11}^*$ and $\varepsilon_{22}^*$ by applying the Eshelby inclusion theory to pearlite with cementite lamellae.$^{23}$ Furthermore, if the ferrite and cementite phases keep perfect coherence, then recalibration of the stress state of cementite in the coordinate system of the cementite crystal could help estimate the theoretical lattice parameters, $a_0^\alpha$, $b_0^\alpha$, and $c_0^\alpha$ of cementite. Based on the above calculations, $\varepsilon_{11}^*$, $\varepsilon_{22}^*$, $a_0^\alpha$, $b_0^\alpha$, $c_0^\alpha$, and $R_{b/a}^\alpha$, $R_{c/a}^\alpha$ are listed in Table 1. Here the lattice parameters of cementite, $a_0^\beta$, $b_0^\beta$, and $c_0^\beta$, and of ferrite, $a_0^\alpha = 0.2867$ nm, were used as reference for calculating $R_{b/a}^\alpha$ and $R_{c/a}^\alpha$, respectively, wherein both the phases were regarded as isolated elastic bodies. The experimental results obtained by the EBSD analysis and neutron diffractometry have also been listed in the table as the linear expansion coefficients along each axis of the cementite crystal do not differ noticeably at the measured temperatures.$^{15}$ The values $a_0^\alpha$, $b_0^\alpha$, and $c_0^\alpha$ inherently depend on the O.R., leading to a difference of the plus/minus sign and the absolute values of $R_{b/a}^\alpha$ and $R_{c/a}^\alpha$. This calculation indicates that the values of $R_{b/a}^\alpha$ and $R_{c/a}^\alpha$ become smaller on average due not only to the Pitsch-Petch O.R. but also if the Isaichev O.R. exists in the same sample. From this perspective, the difference in the cementite lattice parameter ratio based on the evaluation method can be semi-quantitatively explained by the coexistence of various O.R.s.

### 3.2.2. Examination of Variant Selection Mechanism in Pearlite

As mentioned above, some O.R.s have been confirmed for α/θ interface in pearlite. In addition, it was reported that the Pitsch-Petch O.R. is frequently observed in eutectoid steels, while the Isaichev and Bagaryat'sky O.R.s tend to be formed in proeutectoid and hypoeutectoid steels, respect-
Therefore, it is assumed that the nucleation sites associated with proeutectoid ferrite and cementite precipitation have a large influence on the O.R. in pearlite. However, this hypothesis depends on the observations from transmission electron microscopy measurements wherein limited areas are observed with high magnification. However, as indicated in Figs. 2 and 8, the Pitsch-Petch and Isaichev O.R.s were observed in the same sample, proving the coexistence of various O.R.s in pearlite. We will discuss the variant selection rule of the above four O.R.s further in the manuscript.

Figure 9 shows the theoretical ferrite (001) pole figure on the cementite standard stereographic projection under the (a) Bagaryatsky, (b) Isaichev, (c) Pitsch-Petch, and (d) Unknown O.R.s with respect to cementite. Here \( \langle 100 \rangle_\theta \) was fixed parallel to the normal direction of the projection since it is simultaneously on both \((010)_\theta\) and \((011)_\theta\) that correspond to the habit plane of (a, c) and (b, d), respectively. There are 2, 4, 8, and 16 ferrite variants under (a), (b), (c), and (d), respectively. Comparing the sets of (a)–(b) and (c)–(d), the orientations of the ferrite variants are found close to each other. This means that each O.R. can be converted without an obvious changing the crystal orientation but by the habit plane (lamellar interface) alone, suggesting the high variability of the O.R. between \( \alpha/\theta \). Adachi et al. investigated the three-dimensional morphology of lamellar cementite using the serial sectioning technique and reported that some cementite lamellae twist gradually within a pearlite colony. Their report suggests that the habit plane between \( \alpha/\theta \) is not identical but changeable on the pearlitic transformation. Therefore, we will consider the selection rule of the above four O.R.s in terms of the elastic strain energy. Figure 10 represents the elastic strain energy per mol \( <E_0^*> \) generated under each O.R. which is calculated by Eq. (4), with the principal strains \( \varepsilon_{11}^* \) and \( \varepsilon_{22}^* \) listed in Table 1.

\[
< E_0^* > = V_m f (1 - f) \left( \frac{\mu}{1 - \nu} \right) \left[ (\varepsilon_{11}^*)^2 + (\varepsilon_{22}^*)^2 + 2 \nu \varepsilon_{11}^* \varepsilon_{22}^* \right] \ldots (4)
\]

where \( V_m \) and \( f \) denote the molar volume and the volume fraction of the cementite, respectively, and \( f = 0.12 \) was

![Fig. 8. (a) Phase and (b) IPF maps of lamellar pearlite isothermally transformed at 923 K for 600 s. Sets of Pole figures of (c), (d) cementite and (e), (f) ferrite indicated by (A) white and (B) black points in (b), respectively, showing the Isaichev O.R. between the lamellar ferrite and cementite.](image-url)

**Table 1.** Theoretical principal strains on habit plane and lattice parameters in a lamellar cementite under four different orientation relationships between ferrite and cementite. Additionally, the experimental data obtained by EBSD and neutron diffraction methods are listed.

| Orientation relationship | Temperature | \( \varepsilon_{11}^*/\% \) | \( \varepsilon_{22}^*/\% \) | \( a\theta^*/\text{nm} \) | \( b\theta^*/\text{nm} \) | \( c\theta^*/\text{nm} \) | \( R_{b/a}^*/\% \) | \( R_{c/a}^*/\% \) |
|-------------------------|-------------|--------------------------|--------------------------|--------------------------|--------------------------|--------------------------|--------------------------|--------------------------|
| Bagaryatsky             | Room temp. (293 K) | 2.179 × 10^{-2} | 1.148 × 10^{-1} | 0.4963 | 0.7137 | 0.4001 | 8.226 | -9.503 |
| Isaichev                | Room temp. (293 K) | 9.686 × 10^{-4} | 2.179 × 10^{-2} | 0.4963 | 0.6758 | 0.4531 | 2.479 | 2.899 |
| Pitsch-Petch           | -6.174 × 10^{-2} | 8.664 × 10^{-2} | 0.4714 | 0.6814 | 0.4728 | 8.786 | 12.59 |
| Unknown                | -1.534 × 10^{-1} | 7.84 × 10^{-2} | 0.4784 | 0.73503 | 0.46226 | 17.239 | 9.774 |
| Method | Temperature | \( \varepsilon_{11}^*/\% \) | \( \varepsilon_{22}^*/\% \) | \( a\theta^*/\text{nm} \) | \( b\theta^*/\text{nm} \) | \( c\theta^*/\text{nm} \) | \( R_{b/a}^*/\% \) | \( R_{c/a}^*/\% \) |
| EBSD                   | Room temp. | N/A | N/A | (0.5090) | (0.6978) | (0.4593) | 3.175 | 1.296 |
| Neutron diffraction    | 873 K       | N/A | N/A | 0.5118 | 0.6818 | 0.4553 | 0.5307 | 0.1534 |
used in the calculation for 0.8 mass% C steel. Since $\varepsilon_{11}^*$ and $\varepsilon_{22}^*$ are sensitive to the lattice parameters of both ferrite and cementite, $(a_\theta^0, b_\theta^0, c_\theta^0$, and $a_\alpha^0$) their parameters were changed at the rate of $-3\%$ to $3\%$, and indicated as $R_C$ and $R_F$, respectively. The three different lattice parameters of cementite were changed at the same rate for convenience of explanation. Furthermore, considering the invariant-line criterion on a habit plane, the result is marked in red if the corresponding criterion ($\varepsilon_{11}^*\varepsilon_{22}^* < 0$) is satisfied, otherwise the result is marked in black ($\varepsilon_{11}^*\varepsilon_{22}^* > 0$). $< E_0^* >$ was widely varied as a function of the lattice parameters of the both phases; the maximum value being several times higher than the minimum one in each O.R. Based on this result, the O.R. yielding the minimum value of $< E_0^* >$ at a given $R_C$ and $R_F$ was chosen and the preferential O.R. map is illustrated in Fig. 11. If the invariant-line criterion is not considered (a), $< E_0^* >$ is minimized under the Isaichev O.R in majority of the map. However, by considering the criterion as a necessity for the selection of O.R. between $\alpha/\theta$, the map (b) reveals the possibility of all O.R.s confirmed in the experiments, including the Pitsch-Petch O.R. as the preferred one. It is especially interesting that the preferential O.R. is shifted from the Pitsch-Petch to Isaichev O.R. in map (b), when ferrite or cementite changes its lattice parameter by $0.6\%$. Although the lattice parameters of ferrite and cementite are changed by a solid solution of the alloy elements, such large change in their values may hardly occur if we take into account the amount of alloy elements in conventional steels and its partitioning coefficient between $\alpha/\theta$. On the other hand, as listed in Table 1, considering that the lattice parameters change significantly by the internal stress, they may be deviated locally due to the internal stress leading to the O.R. transition on the pearlitic transformation. However, the necessity of the invariant-line criterion remains unclear in this case. The formation of the invariant-plane and -line have been discussed in the Phenomenological Theory of Martensite Crystallography and two-dimensional phase interface and they are geometrically

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**Fig. 9.** Cementite standard stereographic projections showing ferrite variants under four different orientation relationships. (a) Bagaryatsky O.R., (b) Isaichev O.R., (c) Pitsch-Petch O.R., and (d) Unknown O.R.

**Fig. 10.** Change in theoretical strain energy per mole in pearlite under four different orientation relationships as functions of ferrite and cementite lattice parameter ratios.
determined by the crystal structure and lattice parameters of the two phases, their lattice correspondence and the habit plane. Therefore, the formation of the invariant-line is not directly related with the elastic strain energy, although the chemical bonding energy between the interfacial atoms may have some influence. Thus, we present a fundamental approach for a deeper understanding of the selection and transition of O.R.s in pearlite in terms of the elastic strain energy considering the invariant-line criterion.

Based on the above results, in addition to the dynamic relaxation resulting from the introduction of structural ledges and misfit dislocations on the interface, it can be concluded from a macroscopic viewpoint that the internal stress generated by the lattice misfit between ferrite and cementite, which is caused by the chemical bonding energy, is minimized and the anisotropy is reduced by the effective internal stress relaxation resulting from the introduction of structural ledges and misfit dislocations on the interface, it can be

Fig. 11. Map of the preferential orientation relationship in pearlite (a) with and (b) without lattice invariant formation as functions of ferrite and cementite lattice parameter ratios.

4. Conclusions

In order to clarify the internal stress state in pearlite, the lattice parameter ratio of cementite distributed in pearlite was locally evaluated using electron backscatter diffraction (EBSD) method. The obtained results are as follows:

(1) The lattice parameter ratio clearly differs between lamellar and spheroidized cementite, which is caused by the internal stress generated by the lattice misfit between ferrite and cementite in pearlite.

(2) The major part of the internal stress is reduced by the structural ledges and misfit dislocations introduced at lamellar interfaces on the pearlitic transformation (built-in accommodation). In addition, residual internal stress is relaxed by the diffusion of Fe atoms at the interface when pearlite is annealed after the completion of transformation (additional time-dependent diffusional relaxation).

(3) The rate of lattice parameter ratio microscopically evaluated by the EBSD method is considerably different from the macroscopic value identified by neutron diffraction. This suggests the coexistence of certain orientational relationships between ferrite and cementite.

(4) The transition of orientational relationships in pearlite was satisfied when their selection takes place with a formation of the invariant-line as a requirement.

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