## A coupled phase-field and finite element method to simulate the elasto-

### plastic deformation induced cementite dissolution in pearlitic rail steels

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#### Abstract

A coupled phase-field and finite element method is originally proposed to investigate the cementite dissolution behavior in pearlitic steels subjected to cyclic deformation. The stress distribution and plastic strain accumulation are calculated by a rolling-sliding contact finite element model assisted with a plastic strain accumulation model. With the input parameters from the finite element model, an elasto-plastic phase-field model is then employed to simulate the real-time evolution of cementite volume fraction, microstructure morphology and carbon distribution for different rolling cycles and contact depths. Upon experimental validations, the proposed model predicts more accurate and realistic results than Sauvage's model. A three-stage dissolution kinetics is also revealed, which well explains an experimentally observed microstructure gradient along the depth direction. The proposed elasto-plastic phase-field model can be potentially extended to simulate cementite dissolution under various manufacturing or serving conditions, and even any stress-driven microstructure evolution containing cementite dissolution.

**Keywords:** phase-field; finite element; cyclic deformation; cementite dissolution; microstructure prediction

#### 1. Introduction

Rails are subjected to increasing mechanical wear and rolling contact fatigue (RCF) due to fast train speed [1] and massive axle loads [2] in modern railway systems. Therefore, there are surging interests to figure out the mechanism of formation of those material failures to improve the properties of existing rail steels and to prolong service life of rails. Microstructure change in the surface layer of rail steels is considered to be related to the degradation of mechanical properties and thus microcrack formation in rails [3][4]. For example, the formation of White Etching Layer (WEL), a hard and brittle layer with considerable dissolution of cementite and grain refinement, on rail surface is usually believed to be favorable locations of crack initiation and be detrimental to the rail lifetime [5][6]. However, the microstructure changes, including pearlite deformation, grain refinement and especially cementite dissolution, during rolling-sliding contact loading remains elusive. Therefore, there is currently a critical need to understand the fundamental mechanism of microstructure evolution of rail steels under rolling-sliding contact deformation.

As reviewed by Gavriljuk [7], there are generally two interpretations of cementite dissolution under severe loadings. Gridnev et al. [8] proposed that the cementite dissolution is due to the higher binding energy between carbon interstitials and dislocations as compared to the binding energy between carbon atoms and iron atoms in cementite. On the other hand, Languillaume et al. [9] thought that the increase in free energy caused by the geometrical thinning of cementite lamellae and the formation of slip steps during plastic deformation. Sauvage et al. [10] semiquantitatively calculated cementite dissolution rate in cold-drawn pearlitic wires based on the Gibbs-Thomson effect and a diffusion-controlled dissolution process. Unfortunately, the predicted dissolution rates appeared to be underestimated in comparison with the experimental measurements. Further, the work was limited to cold-drawn pearlitic wires in which the mechanical deformation is applied in the fashion of only one or a few cycles. Pearlitic rail steels, however, have to endure thousands of rolling-sliding contact deformation cycles during service. More recently, Nematollahi et al. [11] suggested that in addition to the free energy increase mechanism proposed by Languillaume, an elastic strain induced thermodynamic driving force is possible according to first principle calculation, which may account for the underestimation of previous simulation. Hence combing the elastic and plastic effect may get a more reasonable result.

In the present study, an elasto-plastic phase-field model, integrating an elastic strain and plastic strain induced free energy increase in cementite and ferrite, is originally proposed to understand the fundamental mechanism of mechanical deformation induced cementite dissolution. The simulated cementite morphology agrees well with the experimental results.

#### 2. Model description

In this study, a multiscale computational framework, in which an elasto-plastic phase-field model is coupled with the finite-element method, is proposed to model the cyclic mechanical deformation-driven cementite dissolution. The distributions of stress and strain as functions of rolling cycles and contact depths, at the macro-scale, are first calculated by a rolling-sliding contact finite element model. The accumulated strain is then fed into the phase-field model to simulate the real-time evolution of cementite volume fraction and microstructure morphology at the meso-scale.

#### 2.1. Finite-element model

To improve the computation efficiency, a 2-D plane strain finite element model is employed to simulate the stress distribution in the rolling-sliding experiment. The penalty function methods are used to solve the contact and friction problem. Contact load and friction torque applied are 1500 N and 18 Nm respectively, from rolling-sliding experiments. The Young's modulus and the Poisson's ratio of the rail are set as 210 GPa and 0.3. The yield strength and tensile strength are 490 MPa and 900 MPa, respectively.

#### 2.2. Plastic strain accumulation model

A plastic strain accumulation model proposed by Kapoor and Franklin [12] is used. With the shear stress distribution and material hardening behavior, the plastic strain accumulation as a function of rolling cycles and contact depths can be calculated. The shear strain increases with rolling cycles is given by [12]:

$$\Delta \varepsilon_p^{[z]} = C \left( \frac{\tau_{zx(\max)}}{k_{eff}^{[z]}} - 1 \right) \tag{1}$$

where  $\tau_{zx(\max)}^{[z]}$  is the maximum shear stress at the depth of z,  $k_{eff}^{[z]}$  is the effective shear strength of material at the depth of z, and C has a constant value of 0.00237 [13].

The total accumulated shear strain is then calculated by:

$$\varepsilon_p^{[z]} = \sum_N \Delta \varepsilon_p^{[z]} \tag{2}$$

where N is the number of rolling cycles.

#### 2.3. Phase-field model

Ferrite and cementite are described by a field variable  $\phi$ , ranging from 0 in ferrite to 1 in cementite. The carbon distribution in the phases is described by the concentration field, *c*. Specifically,  $c^{\alpha}$  and  $c^{\text{Fe}_3\text{C}}$  represent the carbon composition (in atomic ratio) of the two phases, respectively. Because of its accuracy of modeling stoichiometric compounds with sharp curvature of free energy function, Kim-Kim-Suzuki (KKS) model [14] is employed. The total free energy  $F_{total}$  of the system is given by:

$$F_{total} = \int_{V} \left[ \frac{1}{\Omega_0} f_{local}(c, \phi, \varepsilon) + \frac{\kappa}{2} |\nabla \phi|^2 \right] dV$$
(3)

where  $\Omega_0$  is the molar volume of the system, and  $f_{local}(c,\phi,\varepsilon)$  is the local free energy, which is related to the free energy of ferrite,  $f^{\alpha}$ , and cementite,  $f^{\text{Fe}_3\text{C}}$ . The second gradient term partially accounts for the interfacial inhomogeneity between two phases and  $\kappa$  is a gradient coefficient.

The key novelty of the current phase-field model is to involve both the elastic strain and plastic strain contributions in  $f^{\alpha}$  and  $f^{\text{Fe}_3\text{C}}$ :

$$f^{\alpha} = f^{\alpha}_{chem}(c^{\alpha}) + f^{\alpha}_{el}(\mathcal{E}_{e}) + f^{\alpha}_{pl}(\mathcal{E}_{p})$$
(4)

$$f^{\text{Fe}_{3}\text{C}} = f_{chem}^{\text{Fe}_{3}\text{C}}(c^{\text{Fe}_{3}\text{C}}) + f_{el}^{\text{Fe}_{3}\text{C}}(\varepsilon_{e}) + f_{pl}^{\text{Fe}_{3}\text{C}}(\varepsilon_{p})$$
(5)

where  $f_{chem}^{\alpha}$  and  $f_{chem}^{\text{Fe}_3\text{C}}$  are the chemical free energy of ferrite and cementite, respectively. The chemical free energy of ferrite follows the work of Loginova [15], which is based on regular solution model. The elastic part,  $f_{el}^{\alpha}(\varepsilon_e)$  and  $f_{el}^{\text{Fe}_3\text{C}}(\varepsilon_e)$ , accounting for the lattice distortion of cementite and ferrite crystals, are obtained by fitting the total energy of bcc Fe and cementite as a function of elastic strain in Ref. [11] into a parabolic function.

The plastic part  $f_{pl}^{\text{Fe}_{3}C}(\varepsilon_{p})$  in cementite phase is then estimated using a linear approximation [10]:

$$f_{pl}^{\rm Fe_3C}(\varepsilon_p) = \gamma V_m \frac{1 + \ln(1 + \varepsilon_p)}{t}$$
(6)

where  $\gamma$  is the ferrite/cementite interfacial energy without plastic deformation,  $V_m$  is the molar volume of cementite and t is the lamellar thickness of cementite,  $\varepsilon_p$  is the plastic strain accumulated during the rolling-sliding contact deformation calculated from the plastic strain accumulation model.

The spatial and temporal evolutions of composition and order parameters are governed by the Cahn–Hilliard equation [16] and Allen–Cahn equation [17], respectively:

$$\frac{\partial c}{\partial t} = \nabla \cdot \left[ \frac{D(T)}{(f_{local})_{cc}} \nabla \frac{\partial f_{local}}{\partial c} \right] = \nabla \cdot \left[ M(T) \nabla \frac{\partial f_{local}}{\partial c} \right]$$
(7)

$$\frac{\partial \phi}{\partial t} = -L \frac{\delta F}{\delta \phi} = \frac{L}{\Omega_0} \left[ -\frac{\partial f_{local}}{\partial \phi} + \Omega_0 \kappa \nabla^2 \phi \right]$$
(8)

where D(T) and M(T) are chemical diffusivity and mobility, respectively,  $(f_{local})_{cc}$  is the second derivative of  $f_{local}$ , and L is the interface mobility coefficient.

Both kinetic equations are solved in Fourier space [18] using the FFT algorithm in reduced units, with nondimensionalizing factors  $E = 1.18 \times 10^{11}$  J/m<sup>3</sup> and  $l = 10^{-9}$  m. A 2-D phase-field model with a size of 600 × 600 grid, representing a physical domain of 600 nm × 600 nm, is employed. Unless otherwise specified, the interface thickness (2 $\lambda$ ) is 4 nm in the present simulation.

#### 3. Model validation

The proposed elasto-plastic phase-field model is first validated by comparing the cementite dissolution amount with Sauvage's prediction and the existing experimental results in colddrawn wires, and the results, i.e., the amount of dissolved cementite after cold drawing as a function of temperature are shown in Fig. 1. The parameters used in the validated case all follow those of Sauvage's work [10], in which the wire temperature is 200°C, the true logarithmic strain is 3.5 and the interlamellar spacing of pearlite is 90 nm. When the temperature decreases to 100°C, the dissolution amount from phase-field simulations, either with or without the elastic energy, agrees well with the experimentally measured amount, in contrast to Sauvage's model [10] which presents a clear (70%) underestimation of the dissolved cementite. As the ferrite/cementite interface thickness decreases to 1.3 nm, close to the grid size, the prediction of present phase-field model converges to Sauvage's model. Hence Sauvage's model is a special case of present model and the discrepancy between the above two models is primarily attributed to the difference in the description of the ferrite/cementite interface. In Sauvage's model, a sharp interface and a constant carbon concentration (25 at.%) in cementite were considered; whereas a diffuse interface proposed in the phase-field model presents a carbon concentration gradient at the cementite side of the ferrite/cementite interface, which is more consistent with the experimental results measured by 3DAP [19], as shown in the inserted picture in Fig. 1. The model with the elastic energy clearly outperforms the counterpart without the elastic contribution, when compared with the experimental measurements at the same strain level [20]. Consequently, we rationalize the necessity of the contribution by the elastic strain in the free energy, in addition to the plastic contribution. The phase-field model, integrating both elastic and plastic energy, is subsequently applied to the pearlitic steels subjected to the cyclic rolling-sliding contact deformation.

#### 4. Application in rolling-sliding condition

The shear stress distribution of contact zone is shown in Fig. 2(a). It can be seen that the stress pattern is only existed in surface region of about 3 mm in thickness, so the use of a circular segment with height of 6mm in the rolling-sliding contact finite model would not affect the stress calculation of contact zone. An enlarge picture of the contact zone is displayed in Fig. 2(b). It can be observed that the maximum shear stress is located at the top surface with a value of 543 MPa. The shear stress which is large than the shear strength of material is within 200  $\mu$ m region, so the maximum shear stress distribution at the corresponding depths is plotted in Fig. 2(c). The maximum shear stress decreased linearly to 400 MPa with the depth increasing to 50  $\mu$ m, and then the maximum shear stress stayed unchanged in the next 25  $\mu$ m, resulting from the position indicating by the dot circle in the inserted figure. After that, the maximum shear stress gradually declines to 300 MPa at depth of 200  $\mu$ m.



Fig. 1. Cementite dissolution behavior simulated by present phase-field model with ferrite/cementite interface thickness of 4 nm, by Sauvage's model [10] and of experiment results [19] in cold-drawn wires; and cementite dissolution behavior with different ferrite/cementite interface thicknesses (2 nm, 1.8 nm, 1.6 nm, 1.4 nm and 1.3 nm). Carbon concentration across ferrite/cementite interface based on sharp interface assumption [10], present diffusion interface assumption and experiment measurement [20] in inserted picture.



## Fig. 2. (a) Shear stress distribution of sample under rolling-sliding loading; (b) magnified view of the vicinity of contact region; (c) maximum shear stress distribution with depth.

The accumulated strain as functions of contact depths and rolling cycles is shown in Fig. 3. In Fig. 3(a), it can be seen that at 2000 cycles, the accumulated strain at the top surface is 2.5, and it gradually decreases with the increase of depth, to almost 0 at the depth of 200  $\mu$ m. With the increase of rolling cycles, the accumulated strain increased rapidly at the top surface reaching the critical value 11 at 14000 cycles. However, the strain accumulation speed is much slower at inner part as shown by the slope of the curves in Fig. 3(b). This is because that the maximum shear stress at inner part is not much larger than the yield strength of material and strain hardening even narrows the difference. After 14000 cycles, the accumulated strains reach their maximum values at all depths, i.e. maximum shear stress reaches an equilibrium with the

effective yield strength. The maximum shear strains at depth of 10  $\mu$ m, 20  $\mu$ m, 30  $\mu$ m, 40  $\mu$ m, 60  $\mu$ m and 100  $\mu$ m are 9.18, 7.13, 4.98, 3.21, 2.65 and 2.00 respectively.



# Fig. 3. Accumulated plastic shear strain as a function of (a) contact depth and of (b) rolling cycles at the contact depth of 0 µm,10 µm, 20 µm, 30 µm, 40 µm, 60 µm, and 100 µm.

The evolution of cementite morphology and carbon concentration is shown in Fig. 4. At the contact surface, i.e. at the depth of 0  $\mu$ m, cementite shows a fastest dissolution rate and the thickness of cementite decreases to 2.7 nm and starts to show breakage or discontinuity at 34700 rolling cycles. After that cementite length witnesses a rapid shrinkage and only four small pieces of cementite left at 45000 cycles. After 60000 cycles, no cementite left in the phase field,  $\phi$ , profile and carbon concentration shows a redistribution.

Although cementite is still almost fully dissolved at 10  $\mu$ m below the contact surface after 90000 cycles, the dissolution speed is a little slower than that of the contact surface. The breakage of cementite appears at 50000 cycles with an average cementite thickness of 4.0 nm. At 51500 cycles, cementite lamellae are broken into small particles and the boundaries between cementite and ferrite starts to disappear.

At depth of 20  $\mu$ m, there is still 3.6% volume fraction of cementite left after 120000 cycles, which may because that the driving force is not large enough to induce a full dissolution or the rolling cycles is not enough for the system to reach another quasi-equilibrium. At depth of 30  $\mu$ m, however, only decrease in the thickness cementite lamellae is observed in the morphology profile, without any breakage showing up.

The thickness of cementite lamellae as a function of rolling cycles and contact depths is displayed in Fig. 5. The thickness is the average value calculated from the volume fraction of cementite, so it is underestimated after the emergence of cementite breakage. Because the trend of cementite dissolution is more important than the exact value in present study and the thickness of cementite is already very small after breakage, using the above average thickness is acceptable. It can be seen that in Fig. 5, before a huge drop to 1.6 nm during 30000 to 35000 cycles, the thickness of cementite lamellae decreases dramatically to 6.1 nm in the first 30000 cycles at the depth of 0  $\mu$ m. The cementite dissolution behavior at depth of 10  $\mu$ m is similar to that of the contact surface except that the huge drop comes up at 45000 cycles. Cementite lamellae both witness a complete dissolution in those two depths; however, at the depth of 20  $\mu$ m, the dissolution speed is much slower and gradually decreases to 5.3 nm at 120000 cycles. For depths at more than 40  $\mu$ m, the thickness of cementite lamellae shows obvious decrease from 18nm to 14.8 nm at depth 40  $\mu$ m, to 15.3 at depth 60  $\mu$ m and to 15.8 nm at depth 100  $\mu$ m in the first 50000 cycles and then stays almost constant.



Fig. 4. Microstructure morphology (yellow) and carbon distribution (white-black) of pearlite after different rolling cycles and at different contact depths.



Fig. 5. Cementite lamellae thickness evolved with rolling cycles.

As revealed in Fig. 6(1), cementite lamellae show a three stages dissolution behavior. In stage I, cementite displays a linear dissolution rate due to the increase of free energy as indicated by line a–b, d–e and g–h. At the transition point between stage I and II, discontinuity or breakage starts to appear in cementite lamellae. The increase of interface area where the cementite breaks can significantly enhance the dissolution kinetics in stage II. In addition, the curvature at the surface of the breakage part is much larger than the plain interface between the prior ferrite and cementite lamellae, which can lead to a fast shortening of cementite lamellae due to the Gibbs-Thomson effect. Consequently, a steep dissolution behavior is observed in stage II. At the end of stage II, the interfaces between ferrite and cementite lamellae become vague and only some small particles located at prior cementite lamellae are left. In stage III, spheroidization of cementite and diffusion of carbon atoms are achieved.

It should be noted that the cementite dissolution behavior is strongly dependent on the depth. More importantly, due to the fast dissolution kinetics in stage II, the "transition zone" for different kinetic stages in the sample after certain rolling cycles is quite sharp. As illustrated in Fig. 6(2), after 120000 cycles, the volume fractions of cementite at depths of less than 17.5  $\mu$ m are all well below 1.0%, showing an almost full dissolution behavior. However, at the depth of 20  $\mu$ m, 3.6% cementite is present with an average lamellae thickness of 5.3 nm, indicating a sharp transition zone of less than 2.5  $\mu$ m. It is worth noting that sharp boundaries between the WEL with complete cementite dissolution and the layer below are frequently observed experimentally under rail surfaces after service and are often used to support the heat induced cementite dissolution and WEL formation mechanism. In our simulation assuming the mechanical deformation-driven cementite dissolution mechanism, however, if the full dissolution regions are considered as the WEL, the sharp boundaries can also be realized due to the transition of the cementite dissolution kinetics.



Fig. 6. (1) Cementite dissolution kinetics; and (2) cementite dissolution behavior across the transition zone, i.e. at the depth of 10  $\mu$ m, 12.5  $\mu$ m, 15  $\mu$ m, 17.5  $\mu$ m and 20  $\mu$ m.

#### 5. Conclusions

An elasto-plastic phase-field model is proposed to simulate mechanical deformation-driven cementite dissolution behavior of pearlitic rail steels under cyclic deformation. The proposed elasto-plastic phase-field model can well predict the evolution of cementite morphology and carbon concentration of pearlitic steel. A three stages dissolution kinetics of cementite lamellae is determined from the phase-field simulations. Because of the kinetics transition, a great microstructure gradient is predicted along the contact depth, which well explains an experimentally observed sharp microstructure transition along the depth direction. The proposed elasto-plastic phase-field model can be potentially extended to simulate cementite dissolution under various manufacturing or serving conditions, and even any stress-driven microstructure evolution containing cementite dissolution.

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