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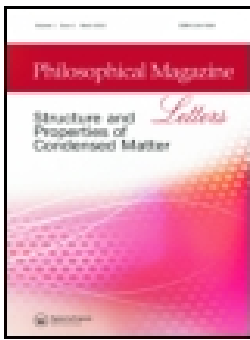
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# Predicting the cooperative effect of Mn–Si and Mn–Mo on the incomplete bainite formation in quaternary Fe–C alloys

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## ABSTRACT

Predicting the effect of alloying elements on the degree of incomplete austenite to bainite transformation in low carbon steels is of great industrial importance. This study introduces an extended Gibbs energy balance model which makes use of an additive approach to calculate the coupled effect of Mn, Si and Mo on the fraction of bainitic ferrite after the incomplete transformation in multicomponent steels. The model predicts significant effects of Mn and Mo and the negligible effect of Si levels on the fraction of bainitic ferrite. This is attributed to the high value of dissipation of Gibbs energy caused by interfacial diffusion of Mn and Mo and low values caused by Si diffusion. The model predictions for quaternary Fe–C–Mn–Si system are comparable with the experimentally measured values of bainite fraction. For the Fe–C–Mn–Mo system, the agreement is less accurate and the accuracy decreases with increasing Mo content, which is attributed a substantial carbide formation but interaction effects between Mn and Mo or a temperature dependent binding energy cannot be ruled out.

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Bainite; incomplete reaction; phase transformation; Gibbs energy balance; steel; modelling

## 1. Introduction

The effect of alloying elements on the incomplete transformation (ICT) phenomenon [1,2] during bainite formation in low carbon steels is of great technical importance. The simultaneous presence of alloying elements like Mn, Mo and Si in appropriate amounts enhances the mechanical properties achieved by bainitic microstructures obtained during quasi-isothermal annealing during

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industrial production; However, the addition of such alloying elements increases the degree of ICT and hence the fraction of untransformed austenite or unscheduled martensite formation. As the mechanical properties of the final microstructure of the alloy are strongly influenced by the bainite fraction, it is essential to predict the effect of alloying elements on the degree of ICT [3,4].

Two general theories have been proposed in the past to describe the mechanism of the bainite formation and consequently describe the effect of alloying elements on the ICT phenomenon in the bainite formation. The first theory assumes a non-diffusional and displacive growth mode for sheaves of bainitic ferrite [5–7]. According to this approach, the austenite to bainite transformation is stopped when the free energy of the supersaturated ferritic bainite equals the free energy of the untransformed austenite at the transformation temperature ( $T_0$  model). Therefore, this theory models the effect of alloying elements on the ICT by predicting their effect on the total free energy of the supersaturated ferritic bainite and the austenite. In a later extension of the model a strain energy term compensating for the local elastic stresses due the bainite formation was added in order to improve the agreement between the experimental data and the model predictions. This model modification is called the  $T'_0$  model. The second theory is based on a diffusion-controlled mechanism for the austenite to bainite transformation. In this theory, the effect of the alloying elements on the incomplete bainite formation is modelled by considering their effect on the migration velocity of the austenite/bainitic ferrite interface. Both schools have applied their models to predict the effect of alloying elements on the incomplete bainite formation and are able to describe the experimental observations to extent reasonable degree [8–10].

Recently, the Gibbs energy balance (GEB) model has been successfully introduced to predict the effect of alloying elements on the degree of the ICT in isothermal bainite transformation more accurately [11–13]. The GEB model, closest related to the diffusional theory, focusses on the solute drag effect of substitutional alloying elements at the migrating austenite/bainitic ferrite interface [14]. In this model, the velocity of migrating austenite/bainitic ferrite interface is calculated by matching the chemical driving force of the isothermal transformation as a function of the degree of transformation with the energy required to move the austenite-ferrite locally enriched by solute atoms trapped in the interface. The model assumes the carbon to partition to proceed with such a high speed that a mean field approximation can be applied. In the GEB model the bainite reaction will stop if the available energy is insufficient to drive the interface forward with a realistic velocity. For ternary, Fe-C-X alloys, it has been shown that the GEB model predictions of the degree of incomplete transformation as a function of the alloy composition and the transformation temperature are significantly better than those of the  $T'_0$  model with a constant value for the strain energy contribution [13].

The present study introduces an extended GEB model. The extended GEB model makes use of a simple additive approach in the calculation of the Gibbs energy dissipation caused by interfacial diffusion of substitutional alloying elements in order to predict the cooperative effect of alloying elements on the bainitic ferrite fraction ( $f_\alpha$ ) in multicomponent steels and a function of the isothermal transformation temperature. The assumption of a simple additivity rule was prompted by the observation that the length of the stagnant stage during partially cyclic transformations, which is also related to the trapping of the interface by the local enrichment of the substitutional alloying element was found to depend on the linear sum of the concentrations of the alloying elements [15]. The predictions of  $f_\alpha$  versus the critical concentrations of alloying elements in Fe–C–Mn–Si and Fe–C–Mn–Mo quaternary steels are presented, discussed and analysed and compared to experimental data reported in the literature as well as predictions by the  $T_0$  and the  $T'_0$  model.

## 2. Model

The GEB model as developed by Chen et al. [16] is based on two fundamental components. The first component is the chemical driving force of the phase transformation, which is taken to be the Gibbs energy change for the transformation. The chemical driving force due to compositional and thermodynamic differences between the austenite and bainitic ferrite phases can be generally calculated using

$$\Delta G_m^{\text{chem}} = \sum_i^n x_i^0 (\mu_i^{\gamma/\alpha}(x_i^{\gamma/\alpha}) - \mu_i^{\alpha/\gamma}(x_i^{\alpha/\gamma})), \quad (1)$$

where  $\Delta G_m^{\text{chem}}$  is the chemical driving force per mole atom,  $i$  is the element in the alloy,  $n$  is the total number of elements in the alloy,  $x_i^0$  is the composition of material transferred over the interface,  $\mu_i^{\alpha/\gamma}$  and  $\mu_i^{\gamma/\alpha}$  are the chemical potential of the element  $i$  ferrite and austenite, and  $x_i^{\alpha/\gamma}$  is the mole fraction of element  $i$  in ferritic side of interface and  $x_i^{\gamma/\alpha}$  is the mole fraction at austenitic sides of interface [17]. For substitutional alloying elements,  $x_i^{\alpha/\gamma}$  and  $x_i^{\gamma/\alpha}$  are chosen according to the Negligible Partitioning Local Equilibrium (NPLE) thermodynamic model, but for carbon, being a fast diffusing interstitial alloying element,  $x_C^{\alpha/\gamma}$  is assumed to be equal to the equilibrium concentrations of carbon in ferrite.  $x_C^{\gamma/\alpha}$  is calculated from the Zener–Hillert equation [18]:

$$x_C^{\gamma/\alpha} = x_C^\gamma + \sqrt{\frac{vx_C^\gamma}{25 \times 10^5 D_C^\gamma T}}, \quad (2)$$

where  $x_C^\gamma$  is the average carbon concentration in austenite,  $v$  is the velocity of the migrating austenite/ferritic bainite interface,  $D_C^\gamma$  is the diffusion coefficient of carbon in austenite and  $T$  is the temperature. The equilibrium carbon

concentration in bainitic ferrite compared to carbon content of austenite is assumed to be negligible. As the carbon diffusion in austenite is much faster than of the substitutional alloying elements, the carbon content in the remaining austenite can be calculated using the mean-field approximation leading to

$$x_C^\gamma = \frac{x_0^\gamma}{1 - f_\alpha}, \quad (3)$$

where  $f_\alpha$  is the fraction of bainitic ferrite and  $x_0^\gamma$  is the initial carbon concentration in austenite. Combining Equations (2) and (3),  $x_C^{\gamma/\alpha}$  can be calculated as a function  $f_\alpha$  and migration rate of austenite/bainitic ferrite interface.

The second component of the GEB model is the dissipation of Gibbs energy due to interfacial diffusion of the substitutional alloying elements. As in the approach by Purdy and Bréchet [19] a triangular potential well inside the interface is assumed and the solute profile is calculated as a function of interface velocity. Based on the diffusion equation and the potential well and the solute profile inside the interface with a width of  $2\delta$  at a given velocity, the dissipation of Gibbs energy due to diffusion inside interface can be calculated from

$$\Delta G_i^{\text{diff}} = - \int_{-\delta}^{+\delta} (x_i - x_i^0) \left( \frac{dE}{dz} \right)_i dz, \quad (4)$$

where  $\Delta G_i^{\text{diff}}$  is the dissipation of Gibbs energy due to diffusion of the substitutional alloying element of  $i$  inside the interface,  $x_i$  is the concentration of the element  $i$  at point  $z$  inside the interface,  $x_i^0$  are the nominal concentration of the element  $i$  and  $dE/dz$  is the gradient of the potential energy inside the interface. According to [19],  $(dE/dz)_i = ((E_0 - |\Delta E|) - \mu_i^{\alpha/\gamma})/\delta$  in the ferritic side of the interface and  $(dE/dz)_i = \mu_i^{\gamma/\alpha} - (E_0 - |\Delta E|)/\delta$  in the austenitic side of the interface, where  $\Delta E = (\mu_i^{\gamma/\alpha} - \mu_i^{\alpha/\gamma})/2$  and  $E_0$  is the binding energy for atoms of alloy  $i$  inside the interface [17,19,20].

In the present model, the total dissipation of Gibbs energy caused by interfacial diffusion is calculated separately for each of the substitutional alloying elements present, and the total dissipation,  $\Delta G_{\text{total}}^{\text{diss}}$ , is the summation of all contributions

$$\Delta G_{\text{total}}^{\text{diss}} = \sum_i^m \Delta G_i^{\text{diff}}, \quad (5)$$

where  $m$  is the number of substitutional alloying elements.

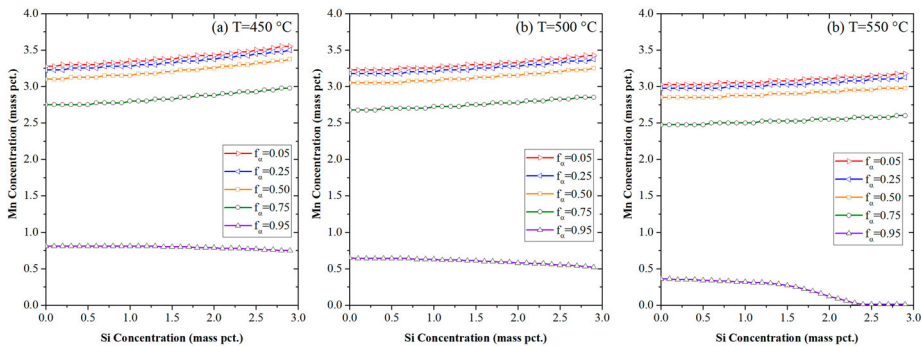
As in the original GEB model, for a given composition and transformation temperature, in the simulations  $f_\alpha$  is gradually increased and the interface velocity is calculated in a recursive loop establishing a balance between the components of the model described above. Once the interface velocity is calculated to be smaller than  $0.1 \mu\text{m/s}$ , the condition for the stagnant stage in

bainite transformation is assumed to have been reached and the ferrite fraction at that state is taken to be the bainitic (stasis) fraction.

### 3. Results

In this section, the results of the GEB model in predicting the total fraction of isothermally formed bainitic ferrite in Fe–C–Mn–Si and Fe–C–Mn–Mo quaternary steels are presented. For both systems, the thickness of the interface,  $2\delta$ , is assumed to be 0.5 nm and independent of the transformation temperature and the degree of enrichment. Similar to the assumptions in previous studies [12,13], the binding energies for the substitutional alloying elements Mn, Si and Mo are taken as 9.9, 12.3 and 30.3 kJ/mol, respectively. The interfacial diffusion coefficient of these elements are again assumed to be the geometric average of the diffusion coefficients in austenite, in ferrite, and in the ferrite grain boundary [21]. Solute interaction terms or interactions between Carbon and the substitutional alloying elements are not considered.

Figure 1(a–c) displays the calculated bainite fractions at which the stasis occurs as a function of the Mn and Si concentration for three isothermal transformation temperatures  $T = 450, 500$  and  $550^\circ\text{C}$  in Fe-0.1C-xMn-ySi system, where  $0 < x < 3$  mass% and  $0 < y < 4$  mass%. In each plot, the iso-bainite fraction contours are plotted for the following bainitic stasis volume fractions  $f_\alpha = 0.05, 0.25, 0.50, 0.75$  and  $0.95$ . The contours were determined on the basis of linear interpolations over the results of a large set of simulations. The curve of  $f_\alpha = 0.05$  specifies the concentrations above which bainite is predicted not to form. On the other side, the curve of  $f_\alpha = 0.95$  marks the domain below which the bainite formation is predicted to run close to the final thermodynamic fraction, i.e. bainitic stasis not to occur. The space between both curves is split up by the iso-bainite fraction contours. The results show that more or less irrespective of the transformation temperature, the fraction of isothermally formed bainitic



**Figure 1.** The effect of Si and Mn addition on the volume fraction of bainitic ferrite calculated by GEB model at (a)  $T = 450^\circ\text{C}$ , (b)  $T = 500^\circ\text{C}$  and (c)  $T = 550^\circ\text{C}$  in Fe-0.1C-xSi-yMn system ( $0 < x < 3$  mass%,  $0 < y < 4$  mass%).

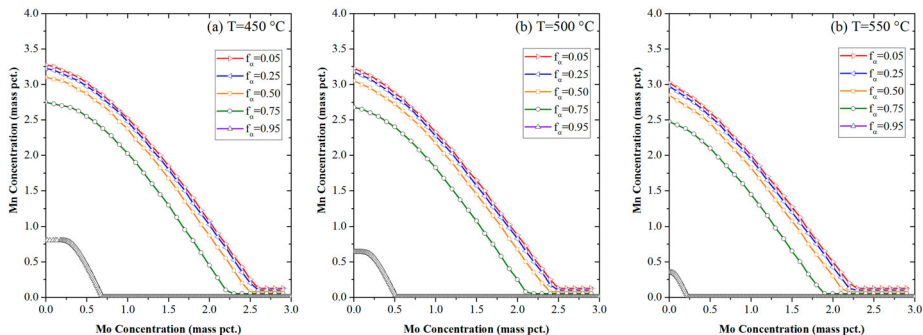
ferrite depends primarily on the Mn concentration, while the effect of Si content seems to be negligible. This is in agreement with previous experimental findings on Mn and Si quaternary alloys and steels that the ICT phenomenon critically depends on the composition of the alloy, in particular the Mn concentration [22]. The critical Mn concentration above which no bainite is predicted to increase from 3.0 to 3.25 mass% when the transformation temperature is lowered from 550°C to 450°C. For the  $f_{\alpha} = 0.05$  contour, the critical Mn concentration increases slightly with the Si concentration, while the opposite trend is observed for the  $f_{\alpha} = 0.95$  contour.

The calculated effect of the combinations of specific Mo and Mn levels on the total bainitic ferrite fractions for temperatures  $T = 450, 500$  and  $550^{\circ}\text{C}$  in Fe-0.1C-xMo-yMn system, where  $0 < x < 3$  mass% and  $0 < y < 4$  mass%, is shown in Figure 2(a-c). It is immediately obvious that the dependence on elemental concentration is very different from that in the Si-Mn system. The model predicts a major effect of the Mo concentration on the degree of ICT. The critical Mn concentration for bainite formation drops from 3.25 mass% at 0 mass% Mo to about 0.1% for 2.5 mass% Mo. A lowering of the transformation temperature shifts the iso-bainite contours to lower Mn and Mo concentration levels.

#### 4. Discussion

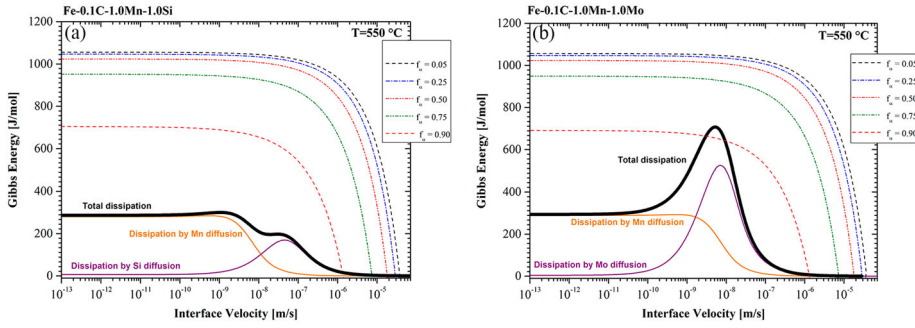
The present model on the cooperative effect of substitutional alloying elements on the degree of incomplete bainite phase transformation showed significant effects of Mn and Mo and the negligible effect of Si addition on the degree of ICT. In this section, first the predictions of the GEB model for each system are discussed, followed by a more detailed analysis of model performance.

Figure 3(a) shows the calculated dissipation of Gibbs energy caused by Mn and Si diffusion as a function of the interface velocity (in solid lines) and the calculated chemical driving force for the phase transformation for different bainitic



**Figure 2.** The effect of Mo and Mn addition on the volume fraction of bainitic ferrite calculated by GEB model at (a)  $T = 450^{\circ}\text{C}$ , (b)  $T = 500^{\circ}\text{C}$  and (c)  $T = 550^{\circ}\text{C}$  in Fe-0.1C-xMo-yMn system ( $0 < x < 3$  mass%,  $0 < y < 4$  mass%).



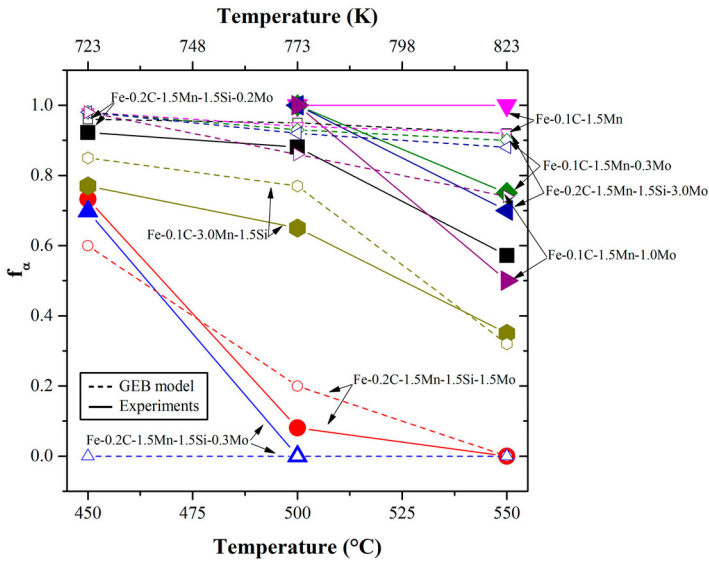


**Figure 3.** The calculated dissipation of Gibbs energy (solid lines) and chemical driving force (dashed lines) for isothermal bainite transformation in (a) Fe-0.1C-1.0Mn-1.0Si and (b) Fe-0.1C-1.0Mn-1.0Mo systems at  $T = 550^\circ\text{C}$ .

ferrite fractions (in dashed lines) for the case of isothermal transformations in Fe-0.1C-1.0Mn-1.0Si quaternary system at  $T = 550^\circ\text{C}$  at several stages of bainite formation. It is evident that the contribution of Si diffusion in the total dissipation of energy is negligible as the dissipation of energy by interfacial diffusion of Mn is much higher and the critical conditions for reaching the critical velocity, taken to represent the bainitic stasis, are barely affected by the Si level and for this combination of composition and temperature the transformation is predicted to run more or less to completion.

Figure 3(b) illustrates the calculated dissipation (Solid lines) and driving force (dashed lines) in Fe-0.1C-1.0Mn-1.0Mo (all in mass%) quaternary system at  $T = 550^\circ\text{C}$ . Contrary to Si, interfacial diffusion of Mo atoms generates huge dissipation of Gibbs energy and modifies the shape of the total dissipation curve and consequently shifts the intersection between the total dissipation curve and chemical driving force curves from  $v \approx 9 \mu\text{m/s}$  for  $f_\alpha = 0.75$  to  $v \approx 3 \text{ nm/s}$  for  $f_\alpha = 0.9$ . This sudden shift in migration rate of interface suggests that addition of Mo should be more effective than that of Mn in creating the transformation stasis. The predicted effect is supported by experimental observations of transformation stasis in Mo containing low-alloy low carbon steels. Furuhashi et al. [23] reported complete bainitic transformations for transformations above  $T = 550^\circ\text{C}$  for the Fe-0.15C-1.5Mn system but incomplete transformation for the alloy co-alloyed with 0.5 wt% Mo.

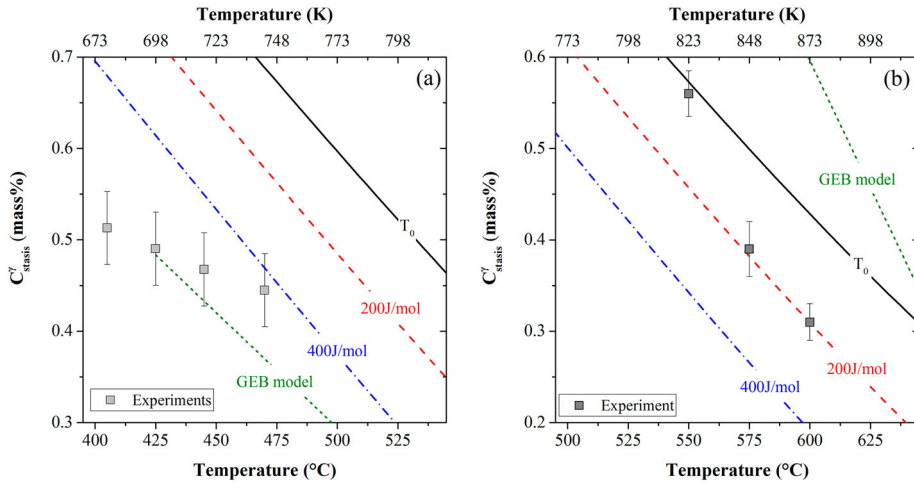
In order to evaluate the performance of the GEB model for the multicomponent alloys, its predictions of the volume fraction of bainitic ferrite ( $f_\alpha$ ) are compared with experimentally reported values. Figure 4 shows the comparison between prediction of the GEB model and experimental reports in Fe-0.1C-3.0Mn-1.5Si, Fe-0.1C-1.5Mn, Fe-0.1C-1.5Mn-0.3Mo, Fe-0.1C-1.5Mn-1Mo and Fe-0.2C-1.5Mn-1.5Si-xMo ( $x = 0.2, 1.5$  and 3 mass%) alloys taken from [3,12,24]. In general, the agreement between both data sets is rather good for the quaternary Fe-C-Mn-Si system. However, in Fe-C-Mn-Mo and Fe-C-Mn-Si-Mo system there are discrepancies between predictions of the current



**Figure 4.** Comparison between predicted values of fraction of bainitic ferrite ( $f_{\alpha}$ ) with the GEB model (shown with hollow symbols/dashed lines) and experimental reports (shown with solid symbols/solid lines) in Fe-0.1C-3.0Mn-1.5Si, Fe-0.1C-1.5Mn, Fe-0.1C-1.5Mn-0.3Mo, Fe-0.1C-1.5Mn-1Mo alloys Fe-0.2C-1.5Mn-1.5Si-xMo ( $x = 0.2, 1.5, 3.0$  mass%) [3,12,24].

GEB model and experimental data, which rise with increasing Mo content. In principle the value  $f_{\alpha}$  predicted by the GEB model could have been corrected by adjusting the values of binding energy ( $E_0$ ) for Mn or Mo as a function of the transformation temperature or the elemental concentration [12]; however, we could not find solid physical arguments which type of dependence to select. Instead, we may have a critical look at the general validity of the assumptions made in the GEB model.

In the GEB model, the carbide formation during bainite transformation is taken to be zero and according to Equation (3), it is assumed that during migration of the interface all of the carbon atoms are rejected to the austenite region making a flat profile. The accuracy of this assumptions can be investigated by comparing the carbon content in austenite at the transformation stasis stage ( $C_{\text{stasis}}^{\gamma}$ ) predicted by the model and the experimentally measured data. Figure 5(a,b) compares values of  $C_{\text{stasis}}^{\gamma}$  a function of temperature, experimentally measured in Fe-0.43C-3.0Mn-2.12Si [5] and Fe-0.1C-1.5Mn-0.3Mo (all in mass%) [3] quaternary alloys and calculated by the GEB model,  $T_0$  model and  $T'_0$  model predictions. In the  $T'_0$  model, additional deformation energies of 200 and 400 J/mol are assumed to take into account the shear deformation [25]. For Fe-0.43C-3.0Mn-2.12Si alloy, Figure 5(a), the experimental values of  $C_{\text{stasis}}^{\gamma}$  are close to the GEB model predictions but below the  $T'_0$  model assuming a strain energy of 400 J/mol. The high Si content in this system is known to suppress carbide formation [26,27], hence neglecting the



**Figure 5.** Carbon concentration in austenite in the stasis stage calculated by the GEB model,  $T_0$  and  $T'_0$  models and the experimentally measured values in (a) Fe-0.43C-3Mn-2.12Si [5] and (b) Fe-0.1C-1.5Mn-0.3Mo (mass%) [3] quaternary systems.

carbide formation in the GEB model is an accurate assumption for this system. In Fe-0.1C-1.5Mn-0.3Mo alloy, Figure 5(b), the experimental values of  $C^{\gamma}_{\text{stasis}}$  are much lower than the predictions by the GEB model but correspond better to the predictions of the  $T_0$  and  $T'_0$  model (assuming a strain energy of 200 J/mol) predictions. Neglecting the carbide formation the GEB model overestimates the carbon content in untransformed austenite, which has a major effect on the bainite formation in the presence of Mo [3].

In addition to ignoring carbide formation [3], or in general the interaction between different alloying elements [24,28,29], the assumed mean-field approximation of the carbon profile in austenite can limit the accuracy of the model predictions of the onset of transformation stasis [30]. Furthermore, the assumed perfect NPLE thermodynamic condition for calculation of the drag of substitutional elements during the whole transformation, may not be applicable under all conditions during the bainite formation. In general, bainite formation proceeds with transitions in partitioning mode of alloying elements at existing interfaces while concurrent nucleation of new bainitic plates and initiation of new interfaces takes place [11]. So, while the current model takes our understanding of the effect of alloying elements on bainite formation a little further, the need for more accurate models making fewer simplifying assumptions on the solute transport remains.

## 5. Conclusions

In this study, the original GEB model was extended in order to predict the coupled effect of Mn, Si and Mo on the fraction of bainitic ferrite after the

incomplete transformation in multicomponent steels alloys and the following conclusions were made:

Addition of Si has a minor effect on the degree of incomplete bainite transformation via immobilising migration of interfaces while addition of Mn and Mo significantly reduces the fraction of isothermally formed bainite and these effects are qualitatively well captured in the extended GEB model.

The predicted minor effect of Si addition and major effect of Mn and Mo addition on the degree of incomplete bainite transformation are attributed to the high value of dissipation of Gibbs energy caused by interfacial diffusion of Mn and Mo and low values caused by Si diffusion.

Considering possible interaction between alloying elements, especially carbide formation in presence of Mo, can improve model prediction of the onset of transformation stasis, but also the option to make the binding energy for each element a function of other parameters, such as co-solutes and concentrations levels is to be explored.

### Disclosure statement

No potential conflict of interest was reported by the authors.

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