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Olga Karabelchtchkova

Richard D. Sisson Jr.

Worcester Polytechnic Institute, sisson@wpi.edu

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Carbon Diffusion in Steels: A Numerical Analysis Based on Direct Integration of the Flux

Olga Karabelchtchikova and Richard D. Sisson, Jr.

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In the early 1970s, Professor Dayananda developed a technique for the direct integration of fluxes from the concentration profiles in vapor-solid diffusion couples to determine diffusion coefficients and atomic mobilities. As part of a project to control and optimize the industrial carburization process in mild- and low-alloyed steels, a modified integration analysis was applied to determine the mass transfer coefficient in the gas boundary layer and carbon diffusivity in austenite. Because carbon flux and surface carbon content vary with time during single-stage carburizing even with a fixed carbon potential in the atmosphere, a mass balance at the gas-solid interface must serve as a boundary condition. This article discusses the numerical modeling of gas carburizing, and focuses on calculating the mass transfer and carbon diffusivity parameters using the simulated concentration profiles. This approach validates the proposed method by comparing the calculated parameters with those used in simulation. The results were compared with previous determinations and predictions reported in the literature.

Keywords carburization, computation, concentration profiles, diffusion modeling, diffusivity coefficient

1. Introduction

Carburization is one of the oldest heat treatments used for surface hardening. Nonetheless, it experiences certain challenges associated with the process performance and reliability. As part of the process control and optimization study of industrial gas carburizing, this article discusses modeling of the process and focuses on developing a method for calculating the coefficient of mass transfer at the gas boundary layer and the diffusion coefficient in steel during the process.

Carbon diffusivity is the main controlling parameter in the carburization heat treatment of steel, yet its value is difficult to measure. Often, the coefficient of carbon diffusion is determined from a model of a solid-solid diffusion couple.^[1-3] The application of such models to carburizing

invariably introduces a certain level of approximation and uncertainty due to a rough, though convenient, assumption of constant surface concentration at the interface with time. More accurate modeling of the gascarburizing process must account for mass transfer from the carburizing gas atmosphere to the steel surface, the surface reaction, and further carbon diffusion into the steel. Mass transfer in the gas atmosphere is the rate-limiting factor in the initial stages of carburizing,^[4,5] and carbon diffusion controls the process at longer times^[6,7]; more often, however, carburizing is considered to be controlled by a combination of both factors.^[8-11] If these coefficients could be calculated from the

Table of Symbols

J	flux of diffusing species
C	carbon concentration
M	atomic mobility
μ	chemical potential
x	space coordinate, depth from the interface
N_{Av}	avogadro number
T	time
K	reaction rate coefficient
a_C	carbon activity
C_p	carburizing potential in the gas atmosphere
C_s	surface carbon concentration
β	mass transfer coefficient
D	carbon diffusivity in austenite
N_1	stability criteria
N_2	biot number equivalent
m	mass
A	area

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Olga Karabelchtchikova and **Richard D. Sisson, Jr.**, Worcester Polytechnic Institute, Center for Heat Treating Excellence Materials Science and Engineering, 100 Institute Road, Worcester, MA 01609. Contact e-mail: olly@wpi.edu.

carbon concentration profile as a function of various process parameters, it would enable modeling and process control. This information could also be used for further process optimization.

The objective of this work was to develop a method for calculating the surface mass transfer and diffusion coefficients from carbon concentration profiles. The approach is based on the numerical modeling of the carburizing process. Carbon diffusivity and the mass transfer coefficient from the literature are used to simulate carbon concentration profiles, and a comparison with the calculated coefficients from these concentration profiles with the input concentration profile is then used for validation of the method. Once tested, this technique will be further applied to experimental data to determine coefficients for a range of steels of various composition and various process parameters.

2. Available Methods for Measuring Carbon Diffusivity

Carbon diffusivity in austenite was first measured by Smith^[12] using the steady-state method. The experimental setup included a steel tube that was carburized on the inside by natural gas decomposition and decarburized on the outside by wet hydrogen. The flux of carbon atoms was measured under steady-state conditions by determining the number of carbon atoms per second carried by the wet hydrogen. Measuring the flux and carbon concentration profile, the coefficients of carbon diffusivities for a range of carbon concentration were determined.

Measurements of carbon diffusivity using diffusion-annealed couples were studied by various researchers.^[1-3,13,14] In their analyses, the coefficients of carbon diffusion were calculated from the concentration profiles using the Boltzmann-Matano method.^[15] The driving force for diffusion is the concentration gradient between the components of the diffusion couples and/or the differences in carbon activities due to the effect of alloying. While this approach to determining the coefficient of carbon diffusion in steel yields a good approximation of the diffusivity coefficient, it assumes a time-invariant carbon content at the interface of the two solids. When applied to carburizing, this assumption implies that there is no resistance barrier to carbon transfer in the atmosphere and that diffusion in the steel is rate limiting. As a result, we are often not able to explain the effect of variations in furnace parameters, such as temperature, atmosphere characteristics, and/or material-related parameters.

Dayananda^[16] developed a method of direct flux integration, which allowed calculation of the intrinsic diffusivities in solid-solid and solid-vapor diffusion couples. Assuming negligible interactions between fluxes at the lattice fixed frame of reference, the intrinsic flux of species within the solid is defined as:

$$J = -C \cdot M \cdot \frac{\partial \mu}{\partial x} \cdot \frac{1}{N_{Av}} \quad (\text{Eq 1})$$

where C and M , respectively, are carbon concentration and atomic mobility of the component, $\partial \mu / \partial x$ is the gradient in chemical potential, and N_{Av} is the Avogadro number. While the above equation is valid for all sections of the diffusion couple, the limitation to its usefulness, as noted by Dayananda,^[16] is that the instantaneous intrinsic flux cannot be measured directly. To compensate for this limitation, a continuity equation was used that allowed an estimation of the cumulative intrinsic flux of atoms diffusing past the marker plane with time by integrating the corresponding area under the concentration profile:

$$\int_{x_0}^{x_\infty} C(x,t) dx = \frac{1}{N_{Av}} \int_0^t -C \cdot M \cdot \frac{\partial \mu}{\partial x} dt \quad (\text{Eq 2})$$

where x_0 is the initial location of the interface between the two components of the diffusion couple, x_∞ is the depth beyond which no concentration gradient exists, and t is the diffusion time. Based on the assumption of constant surface concentration, application of the Boltzmann parameter and Fick's law of diffusion yielded:

$$D = C \cdot M \cdot \frac{\partial \mu}{\partial C} \cdot \frac{1}{N_{Av}} \quad (\text{Eq 3})$$

The method of direct flux integration developed by Dayananda and colleagues^[17-21] is extensively used in the analysis of solid-solid and vapor-solid diffusion couples. Considering the carburizing process as diffusion in a vapor-solid diffusion couple, the goal of this article is to develop a modified method for direct flux integration that would account for the surface boundary condition. As such, with slight modifications, the proposed method would allow one to calculate not only concentration-dependent carbon diffusivity but the mass transfer coefficient as well.

3. Kinetics of Carbon Transfer in Carburizing

The process of the gas carburizing of steel can be viewed as diffusion in a vapor-solid diffusion couple, as shown in Fig. 1.^[22] Carbon transfer from the atmosphere to the solid is determined by the rate-limiting process, which kinetically becomes the controlling stage of carburizing. The maximum carburizing rate is obtained when the carbon transfer from the atmosphere is equal or greater than the carbon diffusion rate in the solid state. Such a diffusion-controlled process has no deficiency of carbon supplied to the interface for its further transport into the solid. In this case, the assumption of constant surface carbon content can be justified. In practice, however, the carbon transfer from the atmosphere to the steel surface is often reported to be the rate-limiting factor,^[4,5] especially at the start of the carburizing process. After this initial stage, the process becomes mixed controlled,^[8-11] and should be modeled correspondingly.

Carburizing can be modeled using a parabolic partial differential equation (PDE) for carbon diffusion in steel and

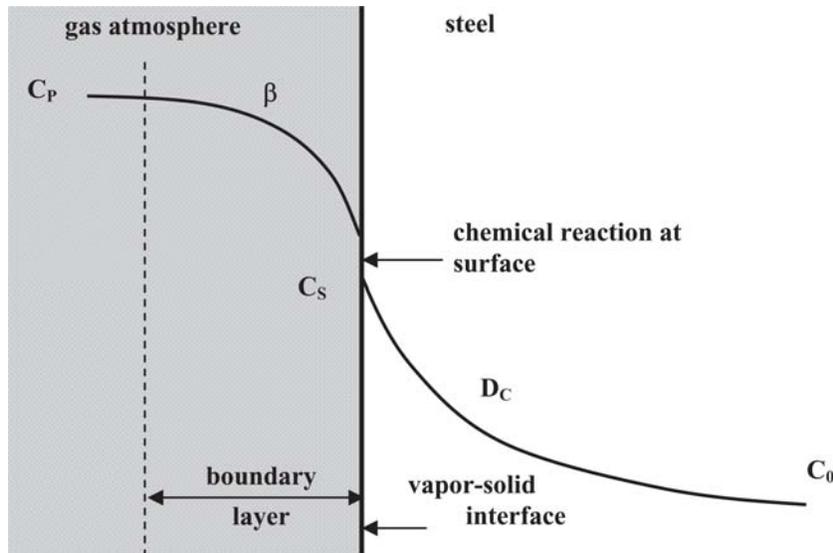


Fig. 1 Schematic representation of carbon transport in carburizing^[22]

a set of boundary conditions accounting for the mass transfer coefficient:

$$\frac{\partial C}{\partial t} = \frac{\partial}{\partial x} \left(D \frac{\partial C}{\partial x} \right) + u \cdot \frac{D}{r + ux} \cdot \frac{\partial C}{\partial x} \quad (\text{Eq 4})$$

where $u = -1$ for a convex surface, $u = 0$ for a plane surface, and $u = 1$ for a concave surface, D is the coefficient of carbon diffusion in steel, x is the distance from the surface, and r is the radius in the case of a convex or a concave surface.

The boundary condition is specified by assuming a mass balance at the steel surface:

$$\beta(C_p - C_s) = -D \frac{\partial C}{\partial x} \quad (\text{Eq 5})$$

where $\partial C/\partial x$ is the carbon concentration gradient at the surface and β is the mass transfer coefficient (in centimeters per second), C_p is the carbon potential in the gas phase, and C_s is the carbon concentration in the solid. The mass transfer coefficient, as defined in Eq 5 accounts for all of the phenomena at the phase boundary between gas atmosphere and steel.^[4] Therefore, the two primary parameters governing carburizing are the mass transfer coefficient (β) and carbon diffusivity (D) in austenite.

4. Numerical Approach to the Parameters Calculation

4.1 Carbon Profile Simulation

Since the analytical solution to carbon diffusion in steel (Eq 4) with the flux balance boundary conditions (Eq 5) is not available for concentration-dependent diffusivity, the method proposed in this article is based on a numerical analysis. A computer program was written in the MATLAB,

which transformed the governing PDE with its corresponding boundary conditions into a set of finite difference equations. Initially, carbon concentration profiles were generated with the mass transfer coefficient (β) and carbon diffusivity (D) from the literature. Then these concentration profiles were analyzed to determine the β and D coefficients. As such, this approach served two purposes: preliminary computer experiments tested the capability of the technique using numerically simulated data; and calculated values of the parameters were validated by comparing them against the parameters used for the concentration profiles generation.

The Dusinger numerical method^[23] was used in this study as it enables one to relate boundary conditions to the rate of carbon transfer at the gas boundary layer and across the steel surface. Concentration profiles were computed using an iterative method for generating the case into a solid of semiinfinite geometry, initially at uniform concentration. The method is second-order accurate and provides a stable convergent solution. Assuming a simple plane geometry and one-dimensional diffusion, the following expression transforms the continuum of Eq 4 into the finite difference expression:

$$C_i^{t+\Delta t} = \frac{D\Delta t}{(\Delta x)^2} \left[C_{i-1}^t + C_i^t \left(\frac{(\Delta x)^2}{D\Delta t} - 2 \right) + C_{i+1}^t \right] \quad (\text{Eq 6})$$

To account for the mass transfer at the surface, the carbon concentration at the boundary nodes was calculated as:

$$C_{\text{surf}}^{t+1} = \frac{1}{N_1} \cdot [2N_2 \cdot C_p + [N_1 - (2N_2 + 2)]C_{\text{surf}}^t + 2C_{x_1}^t] \quad (\text{Eq 7})$$

where

$$N_1 = \frac{D \cdot \Delta t}{(\Delta x)^2} \quad \text{and} \quad N_2 = \frac{\beta}{D} \cdot \Delta x \quad (\text{Eq 8})$$

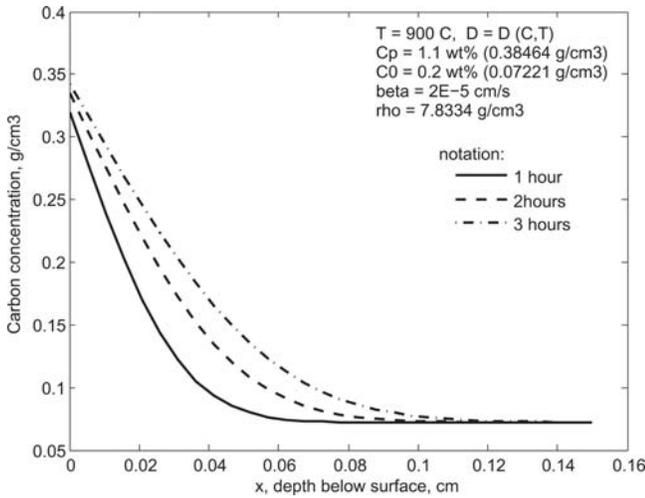


Fig. 2 Numerically calculated carbon concentration profiles after 2 h of carburizing at $T = 900\text{ }^{\circ}\text{C}$ and $C_p = 1.1\text{ wt.}\%$

The two stability criteria were assured to be fulfilled simultaneously: $N_1 > 2$, and $N_1 > 2 N_2 + 2$, where N_2 is the equivalent Biot number, which relates the mass transfer resistance within the steel and at the steel surface. The maximum stable time increment from the previously determined Δx -grid space interval, D , and β values were calculated as:

$$\Delta t < \frac{(\Delta x)^2}{2\beta \cdot \Delta x + 2D} \quad (\text{Eq 9})$$

The input diffusivity values for carbon profiles simulation were calculated from the equations for carbon diffusion in austenite that have been reported in the literature.^[10-12,24-33] These equations were subdivided into two categories: the equations that depend on temperature only; and those that consider carbon content in the steel as well. To account for the variation of diffusivity with concentration, the values were recalculated for instantaneous carbon concentration levels along every space and time increment. Each of the two sets of diffusivity equations was used to calculate the mean values, which served as input for carbon diffusivity in the MATLAB code execution.

It was assumed that no volume change takes place in the crystal lattice of austenite during carburizing, which is a valid assumption for interstitial diffusion processes.^[29] The results of the numerically simulated carbon profiles are given in Fig. 2, and the adequacy of the prediction was tested by comparing the simulation results with the available analytical solution for the flux balance boundary condition and constant carbon diffusivity.^[34]

$$\frac{C(x,t) - C_0}{C_p - C_0} = \text{erfc}\left(\frac{x}{2\sqrt{Dt}}\right) - \exp\left(\frac{\beta x + \beta^2 t}{D}\right) \cdot \text{erfc}\left(\frac{x + 2\beta t}{2\sqrt{Dt}}\right) \quad (\text{Eq 10})$$

Given the sufficiently small spatial increment used for the calculation, the numerical solution for any time greater than 0.25 h accurately reproduced the analytical solution in Eq 10. This comparison validated the accuracy of the numerical calculation and gave confidence to further use the code for modeling carburizing using concentration-dependent diffusivities.

4.2 Calculation of the Mass Transfer Coefficient

From the flux balance condition at the steel interface and the continuity equation of the mass accumulation within the solid, the rate at which the total mass of the solid changes per unit cross section area is:

$$\int_{x_\infty}^{x_0} C(x,t) dx = \int_{t_0}^{t_f} J dt = \frac{\Delta m}{A} \quad (\text{Eq 11})$$

where m is the mass and A is the surface area of the work-piece.

The total quantity of the species diffusing through the surface is found by integrating the concentration profile over the depth of the carburized layer. Further differentiation of the total weight gain by the steel over the carburizing time yields the following expression for the total flux of carbon atoms through the vapor-solid interface:

$$J^t = \frac{\partial(\Delta m/A)}{\partial t} = \beta(C_p - C_s^t) \quad (\text{Eq 12})$$

Assuming a time-dependent nature for the process, the mass transfer coefficient can be found as:

$$\beta^t = \frac{\frac{\partial}{\partial t} \int_{x_\infty}^{x_0} C(x,t) dx}{(C_p - C_s^t)} = \frac{(\Delta m/A)|_{t_0 \rightarrow t}}{t(C_p - C_s^t)} \quad (\text{Eq 13})$$

If weight gain is expressed in grams per square centimeter, time in seconds, and carbon concentration in grams per cubic centimeter, the calculated mass transfer coefficient is expressed in centimeters per second.

An example of such a calculation of the mass transfer coefficient is shown in Fig. 3. As follows from Eq 16, the data needed for the calculation include: total weight gain obtained by integrating total flux over carburizing time; and time evolution of the surface carbon concentration. The input value for β of $2 \times 10^{-5}\text{ cm/s}$ was used for the generation of the concentration profiles, while the calculated value of $2.046 \times 10^{-5}\text{ cm/s}$ was obtained by the application of the above-described method. The predictability was found to be dependent on the selection of the spatial grid size parameter. The predicted values corresponding to the initial time of carburizing were affected by the numerical error arising from the finite difference approximation; therefore, such an initial transient part should not be used.

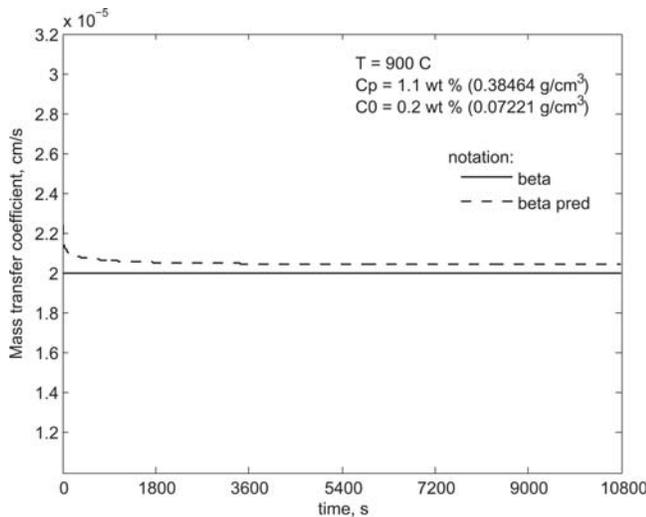


Fig. 3 Comparison of the calculated values of the mass transfer coefficient with the values used in the carbon concentration profile simulation

According to Rimmer et al.,^[35] the mass transfer coefficient in “technical” carburizing atmospheres, consisting of endogas and natural gas enrichment, may range from 1.3×10^{-5} to 2.7×10^{-5} cm/s. The result of the β calculation for a range of input parameters is shown in Fig. 4. The observed R^2 of the parameters correlation suggests that the accuracy of the calculation is independent of the parameters magnitude and gives a relative error of only 2.56% as seen from the slope of the fitted relationship.

4.3 Calculation of the Coefficient of Carbon Diffusion

As in the β calculation, the weight gain of the diffusing species in steel during carburizing was found by integrating the concentration profile over the distance at which the gradient exists:

$$\frac{d}{dt} \int_{x_\infty}^{x_0} C(x,t) dx - J(x_0) = 0 \quad (\text{Eq 14})$$

Assuming an isotropic media, the flux of the diffusing substances through a unit area is proportional to the concentration gradient measured normal to the section:

$$J(x_0) = -D(x_0) \cdot \frac{dC}{dx}(x_0,t) \quad (\text{Eq 15})$$

By equating the above two equations, the following expression for the diffusion coefficient from the concentration profiles can be derived:

$$D(x_0) = -\left(\frac{dC(x_0,t)}{dx}\right)^{-1} \cdot \frac{d}{dt} \int_{x_\infty}^{x_0} C(x,t) dx \quad (\text{Eq 16})$$

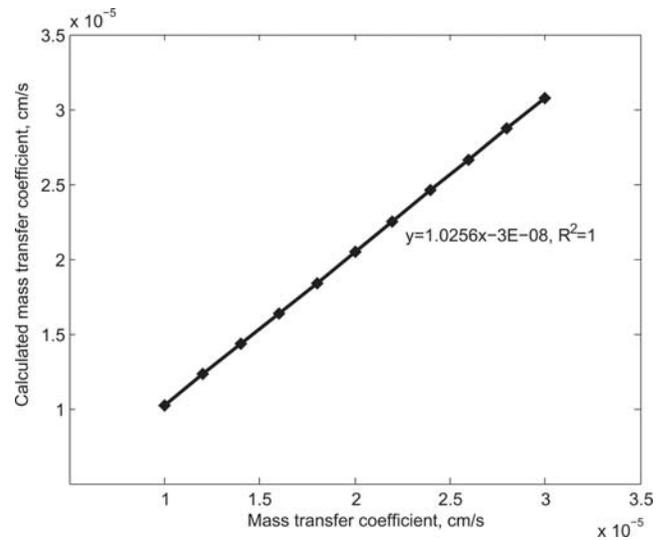


Fig. 4 Correlation of the mass transfer coefficient used in simulation and the corresponding calculated values for a range of input parameters

Based on the proposed method, the carbon diffusivity calculation requires at least two different concentration profiles for time differentiation of the corresponding weight gain. The diffusivity calculation involves the product of the two components: the negative inverse of the slope at any position of the concentration profile; and differentiated with respect to time integrated area under the corresponding section of the profile. The calculated carbon diffusivity, as opposed to the actual diffusivity values, used for the concentration profiles generation, are shown in Fig. 5.

As in the case of calculating the mass transfer coefficient, the predicted values of carbon diffusivities have some error due to the numerical approximation. This error is observed at depths where the carbon gradient asymptotically approaches zero. The corresponding rate of the weight gain change (term 2 in Eq 16) becomes negligible, and its further multiplication by the inverse of the slope causes an erroneous result. As such, it follows that this method can successfully be applied to the range of concentration profiles with concentration gradients greater than zero.

While the prediction of the diffusivities independent of carbon concentration is very accurate (Fig. 5), the technique applied to determining concentration-dependent coefficients of diffusion has some prediction error at the near-surface layer (Fig. 6). This difference between the input diffusivity and the calculated values arises from the estimation of the finite difference at the surface and consecutive recalculation of the carbon diffusion coefficient corresponding to the instantaneous carbon content at every depth of the profile.

5. Conclusions

In this article, the carburizing process was modeled using a finite difference method. The model prediction was validated by comparing the generated concentration profiles

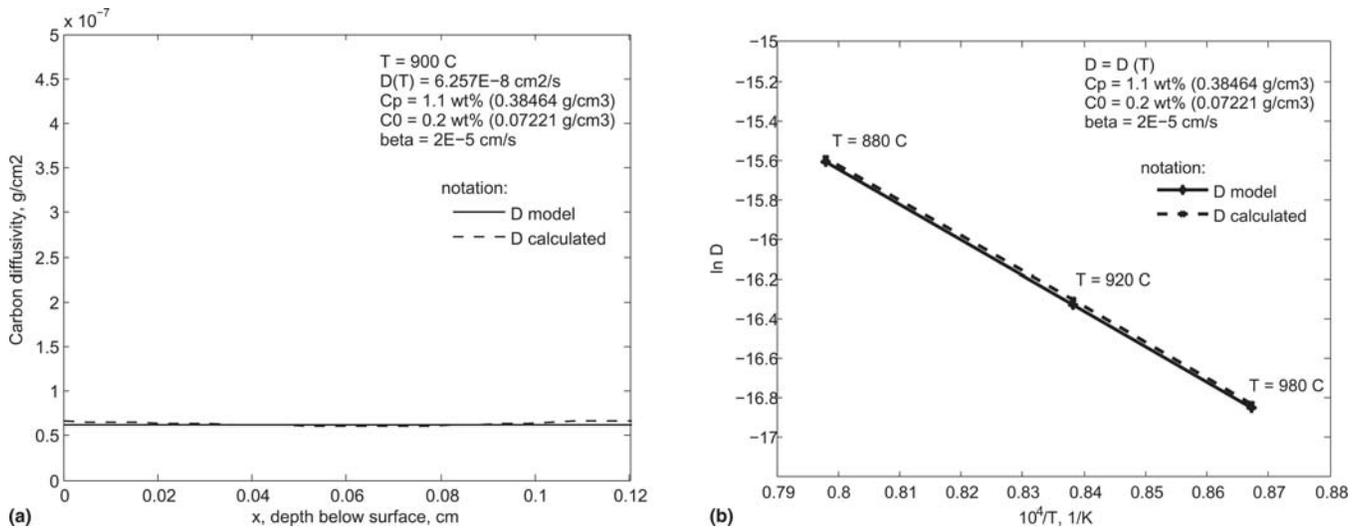


Fig. 5 Comparison of the predicted values of the carbon diffusion coefficient $D = D(T)$ with the modeled values used in the concentration profile simulation: (a) as a function of distance; and (b) Arrhenius type plot

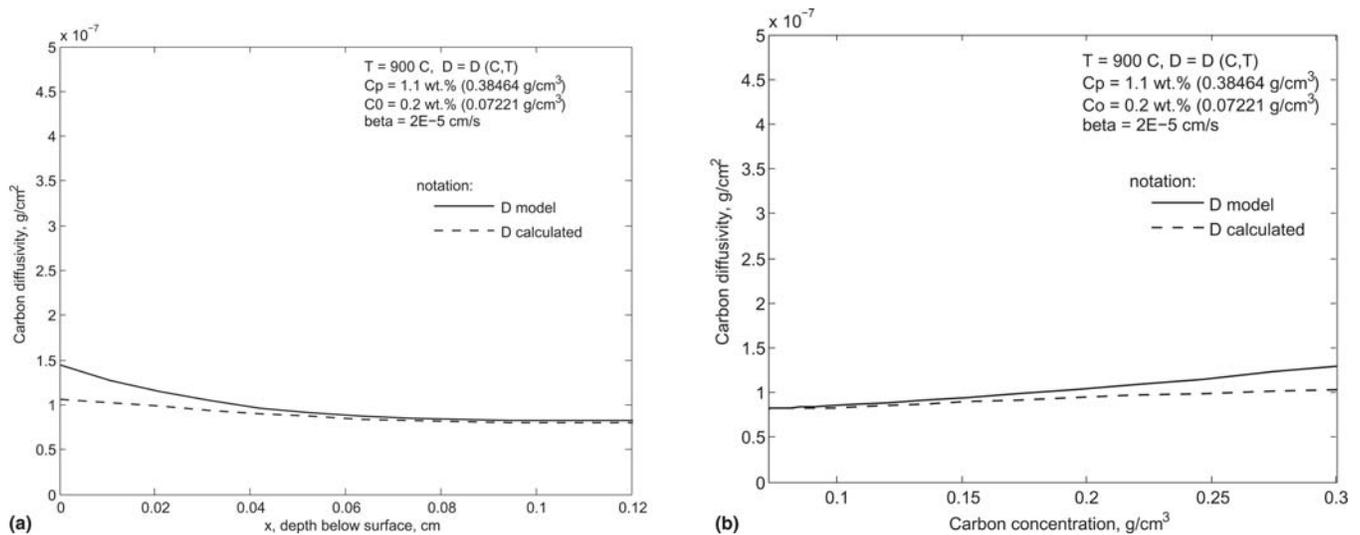


Fig. 6 Comparison of the predicted values of the carbon diffusion coefficient $D = D(C,T)$ with the modeled values used in the carbon concentration profile simulation: (a) as a function of depth; and (b) as a function of carbon content

with the analytical solution for constant diffusivity. The ongoing work uses these numerical simulations as a new method to calculate the mass transfer coefficient and concentration-dependent carbon diffusivity in austenite from the experimental carbon concentration profile. The adequacy of the method was validated by comparing the calculated coefficients against the models used in the generation of the concentration profiles.

The proposed method revealed good predictability and can be applied to determine the mass transfer coefficient in any vapor-solid diffusion system and any size of the steel part with no restriction on Biot number. Successful application of the method requires the available data on surface carbon concentration evolution with time and the carbon concentration profile. To the best of our knowledge, no

time-dependent mass transfer models were available in the published literature; therefore, at this moment it was not possible to test the prediction power to the variable mass transfer coefficient, although it is clear that the proposed method may also be used for time-dependent parameter calculations.

Similar to the mass transfer coefficient, the calculation of carbon diffusivity from the concentration profiles was capable of predicting the values, which agreed quite well with those from the input diffusivity models. The calculated values were slightly larger than the input values due to finite difference approximation.

Overall, since the proposed method for the mass transfer parameters calculation involves measurement of the concentration gradient and areas under the concentration pro-

Section I: Basic and Applied Research

files, it is possible that such a calculation using experimental data will have some level of uncertainty associated with it. Nonetheless, successful implementation of this technique gives a method for further analyses and will be validated using the experimental data. As such, the calculated mass transfer and carbon diffusivity values can then be related to the process parameters and materials characteristics, and may further be used for the control and optimization of the carburizing process.

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