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Effect of Alloy Composition on Carburizing Performance of Steel

Olga K. Rowan and Richard D. Sisson Jr.

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This paper investigates the effect of alloy composition on the gas carburizing performance of AISI 1018, 4820, 5120, and 8620 steels. The mass-transfer coefficients and carbon diffusivities were calculated from experimental measurements using direct flux integration. Although steels with high concentration of austenite-stabilizing elements (Si, Ni) increased carbon diffusivity in austenite, they significantly reduced the kinetics of carbon transfer from the atmosphere to the steel surface and resulted in lower weight gain. Despite lowering the carbon diffusivities, steels alloyed with carbide-forming elements (Cr, Mo) significantly increased the mass-transfer coefficient in the atmosphere and enhanced the rate of carbon profile evolution. The experimentally determined carbon diffusivities were in good agreement with the carbon diffusivities obtained from the thermodynamic and kinetic databases in DICTRA. Overall, using the concentration dependent mass-transfer coefficient and carbon diffusivity in various alloy steels helped explain the experimentally observed variations in the carbon concentration profiles and the effective case depths. Recommendations are made to help achieve better case depth uniformity within a carburizing workload.

Keywords carbon activity, carbon diffusivity, gas carburizing, kinetics, mass-transfer coefficient, modeling

1. Introduction

Gas carburizing is an important heat treatment process used for surface hardening of automotive and aerospace steel components. Despite its worldwide application, the process faces certain challenges in the process control and case depth variability. Carburizing performance of steel is influenced by the furnace design, the process parameters (i.e., gas atmosphere composition, carburizing temperature, and time), and by the steel composition. Considerable research has been done to investigate the effect of these process parameters on the carburizing performance. In practice, however, even with a well-controlled process, some variation in the effective case depth and surface carbon concentration are observed that remain unresolved. Therefore, the goal of this work is to develop a better understanding of the effect of steel composition on the kinetics of carbon transfer during the process and on the overall carburizing performance of steel. Specifically, the objective is to qualitatively and quantitatively investigate the contribution of the major alloying elements on the mass-transfer

coefficient in the gas atmosphere and on the carbon diffusivity in austenite.

The effect of alloy composition on the rate of gas carburizing has been investigated by many researchers.^[1-11] Wada et al.^[1-3] studied the effect of alloy composition on carbon activity in austenite and developed thermodynamic models for several ternary Fe-C-X systems. Other researchers^[4-8] studied the effect of alloy composition on carbon mobility and carbon diffusivity in austenite. Most of these investigations were based on the analysis of diffusion couples. Application of such models to gas carburizing, introduces a certain level of uncertainty because of the assumption of a constant surface concentration. Therefore, the most common approach to account for the effect of steel composition involves adjusting the effective carbon potential in the gas atmosphere by an “alloying factor.”^[9-11] While this empirically developed correction factor yields acceptable results, it does not provide a clear relationship between the alloy composition and the coefficients of mass transfer from the atmosphere to the steel surface or the carbon diffusivity in austenite. Therefore, to explore the nature of their relationship, the calculations and data analysis in this work are based on the modified method of direct flux integration.^[12] This method enables calculation of the mass-transfer coefficient and the carbon diffusivity in austenite from a simple experimental setup and has previously been validated.^[12]

2. Thermodynamics of Mass Transfer During Gas Carburizing

The process of gas carburizing can be viewed as diffusion in a vapor-solid diffusion couple. Carbon transport

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Section I: Basic and Applied Research

during the process is governed by the gradient in chemical potential and is determined by the rate-limiting process, which kinetically becomes the controlling stage of carburizing. The maximum carburizing rate is obtained when carbon transfer from the gas atmosphere is equal to or greater than the carbon diffusion rate in the steel. In practice, however, the process is mixed controlled^[10,13] and is governed both by the mass-transfer coefficient and by the carbon diffusivity in steel. According to the thermodynamics of irreversible processes,^[14] the driving force for mass transfer during carburizing is the gradient in carbon chemical potential. The chemical potential is determined by the carburizing temperature and the thermodynamic carbon activity:

$$\mu_C = \mu_C^0 + RT \ln a_C \quad (\text{Eq 1})$$

where μ_C is the chemical potential of carbon, R is the universal gas constant, T is the process temperature in Kelvin, and a_C is the carbon activity in austenite.

Most of the available models for carbon activity reported in the literature have been developed for ternary Fe-C-X systems.^[1-3] Such models are based on the characteristic distribution of carbon atoms in the matrix of alloyed austenite and the localized forces of their interactions. The presence of Si and Ni in steel increases the carbon activity and the coefficient of carbon diffusion in austenite.^[9,13,14] Nevertheless, the presence of these alloying elements in steel impedes the carburizing process.^[13] In comparison, Cr and Mo decrease the carbon diffusivity in austenite,^[4,15] though these elements accelerate the overall carburizing performance of steel.^[13] These phenomena become more convoluted as the composition of alloy steels grows increasingly complex. Since carburizing of alloy steels helps attain necessary steel hardenability, it is important to understand the effect of alloying elements on the carburizing response of medium- and high-alloy steels to help ensure repeatable and well-controlled results. Several thermodynamic databases, including Thermo-Calc,^[16] have been developed to provide these data and analyses in a user-friendly format.

Gas carburizing is modeled using Fick's Second Law with a flux balance boundary at the gas/steel interface (i.e., carbon flux from the atmosphere to the steel surface equals the flux of carbon atoms diffusing in steel) assuming no soot accumulation occurs^[9]:

$$\sum_i^n \frac{k_i}{a_{\text{surf}_i}} (a_C^{\text{gas}} - a_C^{\text{surf}}) = -D \frac{da_C^{\text{surf}}}{dx} \quad (\text{Eq 2})$$

where k_i is the rate coefficient of the atmosphere chemical reactions; a_C^{surf} and a_C^{gas} are the carbon activity at the steel surface and gas atmosphere, respectively; D is the carbon diffusivity in austenite; and x is the depth below the steel surface. The summation sign (\sum) indicates that several chemical carburizing reactions can take place simultaneously. For carbon concentration profiles with less than 1 wt.% C, the mass-transfer coefficient is often expressed as the ratio between the rate coefficient for the chemical reactions and the carbon activity at the steel surface.^[17,18]

$$\left(\beta = \sum_i^n \frac{k_i}{a_{\text{surf}_i}} \right)$$

The mass-transfer coefficient (β) has been reported to be a complex function of the atmosphere gas composition, carburizing potential, and temperature.^[18-21] To the authors' best knowledge, there has been little work published to relate quantitatively the effect of alloy composition to the rate of carbon transfer in the gas atmosphere and across the gas/steel interface. Using a concentration dependent β would allow modeling the carbon concentration profiles to help explain the observed variations in the effective case depth and the carbon concentration evolution in various alloy steels.

The carbon diffusivity in austenite (D) is another critical parameter that is influenced by the carburizing temperature and steel composition.^[22] For low-alloy steels, this influence may be negligible, while for medium- and high-alloy steels the effect of alloying elements may be significant and should be taken into consideration. Understanding the effect of alloying on the carburizing performance requires knowledge of thermodynamic data including the activity coefficient and carbon mobility in the face-centered cubic (fcc) lattice of alloyed austenite. Such experimental data are scarce and are not always readily available in the published literature. Therefore, the mass-transfer coefficient and carbon diffusivity in this work were determined using the method of direct flux integration, which allows calculation of both kinetic parameters from a simple experiment.

3. Experimental Protocol

Four steel grades with the same bulk carbon concentration were selected for this work. These included plain carbon steel (AISI 1018) and three medium-alloyed steels (AISI 4820, 5120, 8620), with the chemical composition given in Table 1. The steel grades were selected to provide various combinations of (low-high) concentration of the major alloying elements (Ni, Mo, Cr, and Si). Carburizing AISI 1018 was intended to serve as the baseline for evaluating the effect of steel composition on carburizing performance.

The AISI 5120 and AISI 8620 steel bars were received in the hot rolled condition, while AISI 4820 and AISI 1018 were supplied in the annealed and cold-finished condition.

Table 1 Chemical composition of steels

AISI steel designation	Composition, wt.%							
	C	Mn	P	S	Si	Ni	Cr	Mo
1018	0.2	0.8	0.01	0.029	0.26	0.1	0.07	0.03
4820	0.2	0.6	0.007	0.02	0.28	3.28	0.12	0.26
5120	0.2	0.79	0.007	0.01	0.23	0.05	0.77	0.02
8620	0.19	0.87	0.013	0.031	0.19	0.42	0.57	0.21

Microstructural analysis revealed a mixture of ferrite and pearlite uniformly distributed in the transverse direction and having a banded structure in the longitudinal direction, parallel to the direction of rolling. All steel bars were normalized for 4 h at 900 °C, which minimized the differences in prior rolling conditions and reduced the grain size from 6.5 to 8 (ASTM E 112).

The normalized bars were machined into disks 3.1 cm in diameter and 1 cm in thickness. The samples were carburized at 925 °C for 1 and 2 h in an integral quench furnace at an industrial research facility. The endothermic atmosphere was produced by blending the endothermic carrier gas with natural gas enrichment, and the carbon potential in the furnace was controlled at 1.1 wt.% C using an oxygen probe and infrared (IR) analyzers. The weight gain measurements upon carburizing were collected on a laboratory scale sensitive to 0.1 mg. Surface carbon concentration and carbon concentration profiles were measured by spectral analysis using LECO optical emission spectrometer (OES) (St. Joseph, ME) with an accuracy of ± 0.01 wt.% C. A layer of the material of exact known depth was sequentially removed from the surface and analyzed for chemical composition. In order to measure the carbon concentration profiles, this procedure was repeated until a zero carbon gradient (for three consecutive measurements) was reached, which indicated the bulk carbon concentration.

For computation analysis, experimentally measured carbon concentration profiles were interpolated to form smooth curves using a cubic spline interpolation algorithm executed in MATLAB.^[23] Cubic spline interpolation method was based on fitting a series of unique cubic polynomials between the data points ensuring that the curve obtained be continuous and appears smooth. The interpolated carbon concentration profiles were then used to determine the slope and the integrated area under the curve, required for carbon diffusivity calculations, as will be discussed in the following sections.

4. Results and Discussion

Figure 1 shows the experimentally measured carbon concentration profiles in the carburized parts after 1 and 2 h. Although all parts were carburized in the same basket under the same carburizing conditions, the laboratory analysis revealed distinct differences among these steels in the carbon concentration profiles and the total weight gain (ΔM). It was observed that plain carbon steel exhibited the maximum carbon uptake. Depending on the level of alloying and on the nature of carbon and alloying elements atomic interactions, the carbon concentrations profiles in the alloy steels were lower than in the plain carbon steel.

If the effective case depth is defined as the depth to 0.4 wt.% C, the measured case depth variation was found to be ± 0.03 mm after 1 h carburizing and ± 0.06 mm after 2 h carburizing. From Fig. 1(a), the primary differences in the carbon concentration profiles after 1 h were observed at near surface layer up to a depth of 0.4 mm. This observation was attributed to the rate of carbon transfer from the atmosphere

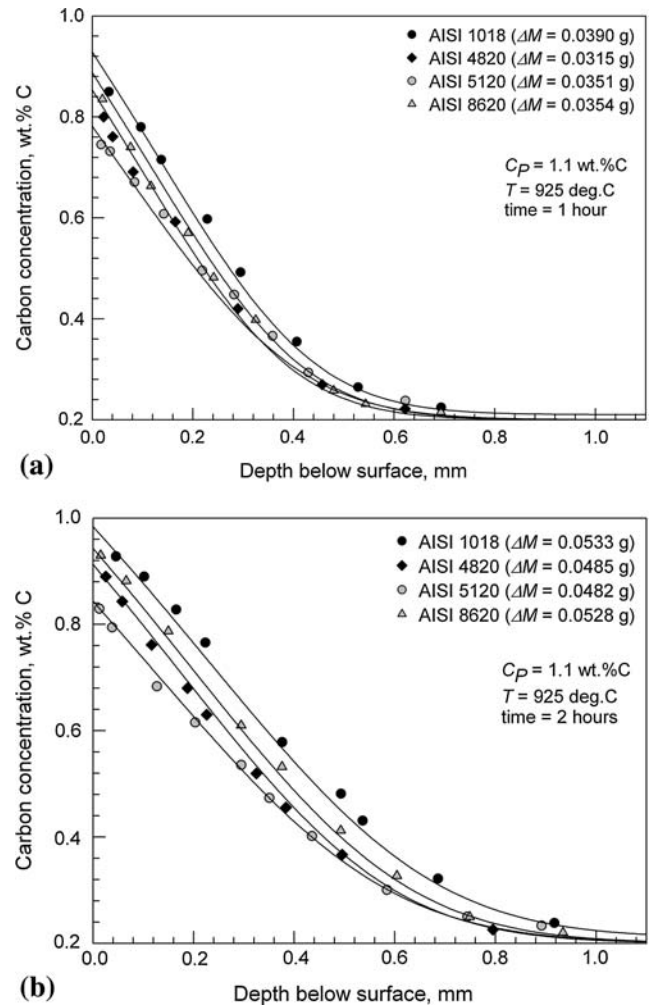


Fig. 1 Carbon concentration profiles after 1 and 2 h carburizing (data points are the reported average of three measurements)

to the steel surface, which is kinetically the rate-limiting process at the initial stage of carburizing.^[18] As time proceeds, the process becomes mixed controlled,^[10,13] where both the mass-transfer coefficient and the carbon diffusivity contribute to the carbon profile evolution (Fig. 1b). During this stage, the instantaneous carbon flux across the gas/steel interface is determined by the balance between the carbon flux from the gas atmosphere to the steel surface and the rate of carbon diffusion in steel. Therefore, the overall observed differences in the carburizing performance of various alloy steels are the result of the effect of alloying elements on the carbon activity at the steel surface (affecting the rate of carbon transfer across the gas/steel interface) and the carbon diffusivity in austenite.

By definition, carbon potential (C_p) in the atmosphere is defined as the amount of carbon that is in equilibrium with the surface carbon concentration in unalloyed austenite. For the given carburizing temperature and C_p of 1.1 wt.%, the thermodynamic carbon activity in austenite and the corresponding activity coefficients for various alloy steels were calculated using Thermo-Calc^[16] and are given in Table 2.

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Of the three alloy steels, AISI 5120 steel (with high-Cr and zero-Ni concentrations) provides the highest equilibrium carbon concentration and, therefore, it exhibits the strongest tendency to reduce the carbon activity from its unalloyed counterpart, that is, AISI 1018. AISI 8620 steel with a more balanced combination of the austenite-stabilizing and carbide-forming elements also increases the equilibrium carbon concentration from that of the plain carbon steels but to a smaller degree. The opposite effect is observed in case of AISI 4820 steel, the chemical composition of which significantly reduces the equilibrium concentration of carbon in austenite and increases the thermodynamic carbon activity in steel. These data are used further throughout the paper to help explain the effect of steel composition on the kinetic carburizing parameters.

4.1 Mass-Transfer Coefficient

From the flux balance condition at the steel interface and the continuity equation of mass accumulation within steel, the total amount of diffused carbon atoms per unit area can be estimated from the area under the carbon concentration profile and the integrated carbon flux:

$$\int_0^{x_\infty} C(x, t) dx = \int_{t_0}^{t_f} J_C dt = \frac{\Delta M}{A} \quad (\text{Eq 3})$$

where x_∞ is the depth beyond which no concentration gradient exists, J_C is the carbon flux, t_0 and t_f are the initial and final carburizing time, $\Delta M/A$ is the weight gain per unit

area of the carburized part. Further differentiation of the weight gain over carburizing time yields the following expression for the carbon flux at a particular time t through the gas/steel interface:

$$J_C^t = \frac{\partial}{\partial t} \left(\frac{\Delta M}{A} \right) = \beta^t (C_P - C_S^t) \quad (\text{Eq 4})$$

Assuming a time-dependent nature of the process, the rate of carbon transfer at the gas/steel interface can be characterized by the instantaneous mass-transfer coefficient:

$$\beta^t = \frac{1}{(C_P - C_S^t)} \cdot \frac{\partial}{\partial t} \int_{x_\infty}^{x_0} C(x, t) dx \quad (\text{Eq 5})$$

or

$$\beta^t = \frac{1}{t(C_P - C_S^t)} \cdot \left(\frac{\Delta M}{A} \right) \Big|_{t_0 \rightarrow t} \quad (\text{Eq 6})$$

where t_0 is the initial time of carburizing, t is carburizing time of interest, and $(\Delta M/A)_{t_0 \rightarrow t}$ is the acquired weight gain in the carburized sample per unit area at any given time t .

Table 3 shows the measured weight gain and the surface carbon concentration in parts after 1 and 2 h carburizing. Carbon flux and the mass-transfer coefficient were calculated from the experimental data according to Eq 4 and 6. Although the mass-transfer coefficient (β) was shown to change slightly over time as the surface carbon concentration builds up,^[24] it is commonly assumed^[19,25,26] that $\beta = \text{const}$ for any given set of carburizing conditions (temperature, carbon potential, alloy). Therefore, the calculated values $\beta^{t=1\text{h}}$ and $\beta^{t=2\text{h}}$ were averaged and reported in Table 3.

While the parts were subjected to the same carburizing conditions, the calculated mass-transfer coefficients ranged from 8.91×10^{-6} to 1.82×10^{-5} cm/s depending on the steel composition. AISI 4820 steel exhibited the slowest kinetics of the mass transfer from the gas atmosphere to the steel surface and revealed the least weight gain after carburizing. As follows from Table 2, austenite-stabilizing

Table 2 Thermodynamic characteristics of various steels calculated at 925 °C and $C_P = 1.1$ wt. %

Parameters	AISI 1018	AISI 4820	AISI 5120	AISI 8620
a_C	0.803	0.803	0.803	0.803
Equilibrium C concentration, wt. %	1.029	0.964	1.043	1.039
γ , activity coefficient	15.85	16.94	15.63	15.67

Table 3 Calculation of the mass-transfer coefficient

Steel	Carburizing time, h	Experimentally measured			Calculated	
		Weight gain, mg	C_S (a), wt. %	C_S , g/cm ³	J_C , g/cm ² s	β , cm/s
1018	1	39 ± 0.55	0.88	0.309	1.47×10^{-6}	$(1.81 \pm 0.052) \times 10^{-5}$
	2	53.3 ± 0.46	0.95	0.334	8.98×10^{-7}	$(1.81 \pm 0.052) \times 10^{-5}$
4820	1	31.5 ± 0.45	0.75	0.264	1.19×10^{-6}	$(8.90 \pm 0.038) \times 10^{-6}$
	2	48.5 ± 0.35	0.82	0.289	8.17×10^{-7}	$(8.90 \pm 0.038) \times 10^{-6}$
5120	1	35.1 ± 0.48	0.83	0.291	1.32×10^{-6}	$(1.34 \pm 0.045) \times 10^{-5}$
	2	48.2 ± 0.43	0.92	0.323	8.12×10^{-7}	$(1.34 \pm 0.045) \times 10^{-5}$
8620	1	35.4 ± 0.51	0.85	0.299	1.33×10^{-6}	$(1.51 \pm 0.057) \times 10^{-5}$
	2	52.8 ± 0.48	0.92	0.323	8.9×10^{-7}	$(1.51 \pm 0.057) \times 10^{-5}$

(a) Experimentally measured surface carbon concentrations. Reported values represent the average of three measurements with ±0.01 wt. % measurement accuracy

elements (Ni and Si) reduce the equilibrium carbon concentration and increase the carbon activity (a_C^{surf}) at the steel surface. This decreases the mass-transfer coefficient and the corresponding carbon flux [$J_C \propto (a_C^{gas} - a_C^{surf})$] entering the steel surface. In comparison, carbide-stabilizing alloying elements (Cr, Mo) decrease the carbon activity and correspondingly increase the total carbon flux across the gas/steel interface. As a result, the calculated mass-transfer coefficients for AISI 5120 and AISI 8620 were greater than that of AISI 4820 and revealed larger weight gain and higher surface carbon concentration upon carburizing.

4.2 Carbon Diffusivity in Austenite

As with the mass-transfer calculation, the weight gain of carbon atoms diffusing into the steel across any arbitrary plane parallel to the gas/steel interface can be found by integrating the concentration profile over the distance at which the gradient exists:

$$\frac{d}{dt} \int_{C'}^{C_0} x dC - J_C(x') = 0 \quad (\text{Eq 7})$$

where C' is the carbon concentration at the given depth (x') and C_0 is the bulk carbon concentration. 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:

$$J_C(x') = -D(x') \cdot \frac{dC}{dx}(x', t) \quad (\text{Eq 8})$$

By equating Eq 7 and 8, the following expression for calculating carbon diffusivity from the concentration profiles can be derived^[12]:

$$D(x') = - \left(\frac{dC(x', t)}{dx} \right)^{-1} \cdot \frac{d}{dt} \int_{C'}^{C_0} x dC \quad (\text{Eq 9})$$

Overall, calculation of the carbon diffusivity involves the product of two components: (a) negative inverse of the slope at any position x' on the carbon concentration profile and (b) integrated area under the concentration profile differentiated with respect to carburizing time. Figure 2 presents the carbon diffusivities calculated from the experimental carbon concentration profiles (shown in Fig. 1). The calculated data were compared with the carbon diffusivities to those calculated from the thermodynamic and kinetic databases in DICTRA.^[27] A good agreement was observed between the sets of data.

Figure 3 compares the carbon diffusivity in austenite for various alloy steels. It was observed that despite the lowest mass-transfer coefficient associated with AISI 4820, the presence of strong austenite-stabilizers (Ni, Si) increases the carbon diffusivity in austenite. This observation was attributed to weaker bonding energy and negative atomic interactions between the austenite-stabilizing elements and carbon atoms. Carbide-forming elements (Mo, Cr) induce positive atomic interactions and tend to attract interstitially diffusing carbon atoms. Such deviations from randomness impede the long-range diffusion of carbon atoms in the austenite matrix and therefore decrease the effective carbon diffusivity. Since the effect of carbide-forming elements is

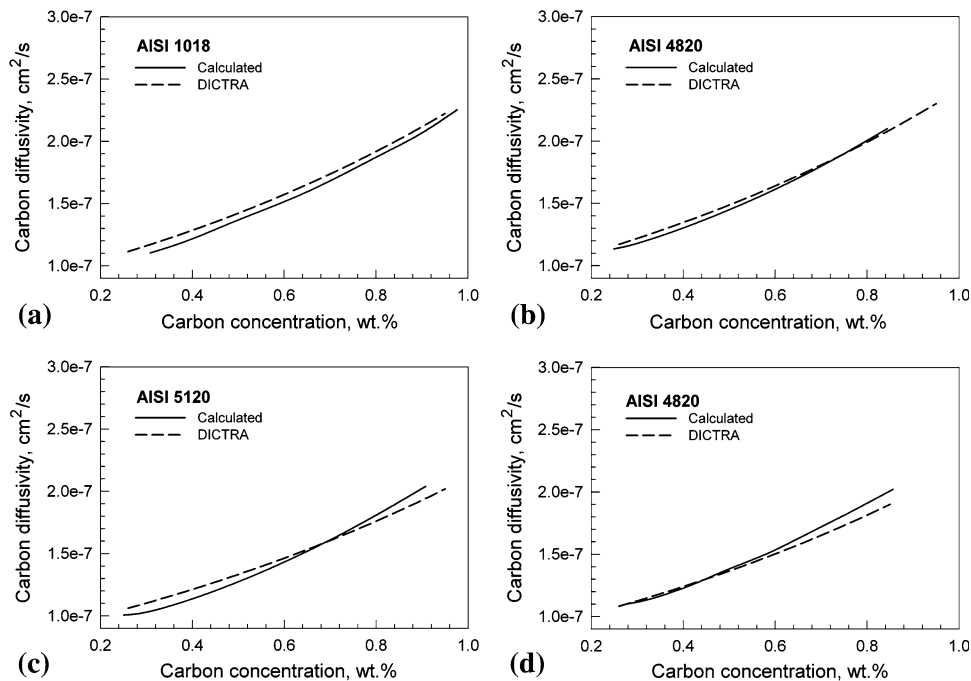


Fig. 2 Comparison of the calculated carbon diffusivities with those from DICTRA. (a) AISI 1018. (b) AISI 4820. (c) AISI 5120. (d) AISI 4820

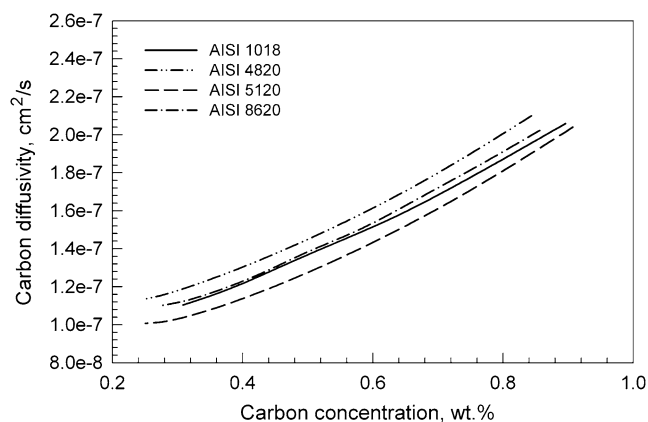


Fig. 3 Comparison of the coefficients of carbon diffusion for various alloyed steels

offset by the addition of austenite-stabilizing elements, the calculated carbon diffusivities in AISI 8620 and 1020 steels were found to be between those of AISI 4820 and 5120.

Overall, the implication of this work is that medium- and high-alloy steels with high-Cr and low-Ni concentrations significantly reduce the carbon diffusivity in steel and, therefore, require longer carburizing time to achieve the desired case depth. Carburizing such steel parts together with high-Ni steel components will inevitably lead to case depth variations within the same workload. Therefore, it is recommended that such steel components be carburized in a separate load and the carburizing time should be adjusted accordingly to achieve the desired case depth.

The importance of understanding and quantifying the effect of chemical composition of alloy steel on their carburizing performance should not be underestimated. For a given carburizing temperature and gas carburizing atmosphere, the mass-transfer coefficient and the carbon diffusivity in austenite vary with the steel composition. This implies that various alloy steels may require different carburizing times to achieve a desired case depth. Incorporating the concentration-dependent β and D in the available carburizing models can help achieve better case depth uniformity.

5. Conclusions

This paper investigated the effect of alloy composition on the kinetics of mass transfer during gas carburizing and on the overall carburizing performance of various alloy steels. The principal conclusions of this work are:

- Gas carburizing performance of various alloy steel is strongly affected by the alloy composition and should be taken into account when carburizing medium- and high-alloy steels to ensure repeatable and well-controlled results.
- AISI 4820 steel with high concentration of austenite-stabilizing elements (Ni, Si) exhibited the slowest kinetics of the mass transfer from the gas atmosphere

to the steel surface. Although the austenite-stabilizing elements increased the carbon diffusivity in austenite, the rate of carburizing was limited by the flux entering the steel surface, which significantly lowered the final carbon concentration profile compared to that of plain carbon steel with the same bulk carbon concentration.

- While carbide-forming elements (Cr, Mo) in AISI 5120 and 8620 lowered the carbon diffusivity in austenite, they increased the rate of carbon transfer from the atmosphere to the steel surface and accelerated the rate of carburizing.
- Carbon diffusivities calculated from the experimental data using the method of direct flux integration were compared to the carbon diffusivities obtained from the thermodynamic and kinetic databases in DICTRA and showed good agreement.
- Understanding and quantifying the contribution of alloy composition on the mass-transfer coefficient and carbon diffusivity in austenite helps explain the observed variations in the carbon concentration profiles of various alloy steels. Most importantly, it is recommended that the carburizing process time is adjusted by the steel composition to achieve the desired carburizing results with better case depth uniformity.

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