ELEVENTH INTERNATIONAL CONFERENCE ON INTERGRANULAR AND INTERPHASE BOUNDARIES 2004

JOURNAL OF MATERIALS SCIENCE **40** (2005) 3191 – 3198

Diffusion of carbon and titanium in γ-iron in a magnetic field and a magnetic field gradient

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The diffusion coefficients of carbon and titanium in γ-iron were measured in a 6T magnetic field and in magnetic field gradients ranging from 30 to 45 T/m. We have found that the diffusion of carbon in γ-iron is retarded by application of a 6T magnetic field. In contrast with carbon diffusion, no noticeable effect of a magnetic field on the diffusivity of titanium in γ-iron is observed. On the other hand, the diffusion of carbon in γ-iron can be enhanced in a magnetic field gradient when carbon atoms move towards the direction with a higher magnetic field strength. The higher the magnetic field gradient strength becomes, the more the carbon diffusion is enhanced. Nevertheless, a magnetic field gradient causes a decrease in diffusivity of carbon in γ-iron when the opposite magnetic field gradient is applied.

1. Introduction

The relationship between microstructure and properties of materials has been well established. It is widely recognized that bulk properties of both structural and functional materials are often governed by microstructures like grain/interphase boundary, precipitate and dislocation. The design and control of microstructures is therefore a key issue in order to confer desirable properties and performance to engineering materials.

A strategy for microstructures control using a magnetic field has drawn a significant interest of researchers recently. Extensive studies have shown that an external magnetic field can affect many metallurgical phenomena such as recrystallization [1–5], grain growth (grain boundary migration) [6–11], phase transformation [12–16] and precipitation [17]. Although most changes in microstructure occur by diffusion, only a few reports have been made on the effect of a magnetic field on diffusion in solids so far. Youdelis *et al.*, who used a 3T magnetic field, reported that a magnetic field retarded diffusion of copper in aluminum [18]. On the other hand, Nakajima *et al.*, who used a 4T magnetic field, did not find any magnetic field effects on the diffusion of nickel in titanium [19]. Also, Pokoev *et al.* [20] and Poloev and Stepanov [21] have measured the diffusivity of nickel in α-iron and Fe-1.94 at%Si alloy in the magnetic fields ranging 0–0.7 T, respectively. They reported that the magnetic field exerted a non-monotonic influence on the diffusivity of nickel in α-iron and in Fe-Si alloy at ferromagnetic temperatures. A maximum increase in diffusivity occurred at a weak magnetic field strength (e.g. the maximum in diffusivity of nickel in α-iron was observed at 0.1T at 1003 K). However, the diffusivity was decreased with further increasing magnetic field strength and eventually it proved to be smaller in a magnetic field than without magnetic field. In addition, no noticeable influence of a magnetic field on diffusion was observed at paramagnetic temperatures.

Extensive studies on the magnetic field–applied materials development have simulated our interest in ferromagnetic materials, especially steel. During microstructural changes in steels, diffusion of carbon plays an important role. Accordingly, it is of particular

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importance to study the effect of a magnetic field on diffusion of carbon in iron. Unfortunately, to the author's knowledge, there is no report on diffusion of carbon in iron in a magnetic field.

In this article, particular attentions will be paid to effects of the magnetic field on the diffusion of carbon by the interstitial mechanism and that of titanium by the vacancy mechanism in iron. In addition to the diffusion in a uniform magnetic field, we also examine diffusion of carbon and titanium in iron in a magnetic field gradient.

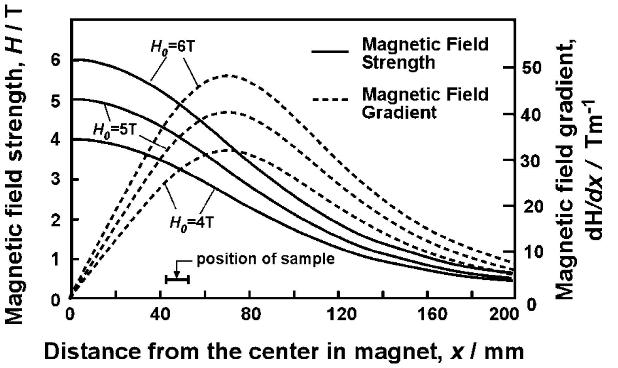

Figure 2 Variation of a magnetic field H and a magnetic field gradient $\mathrm{d}H/\mathrm{d}x$ with the position x in the magnet for different nominal magnetic fields H_0.

2. Experimental procedures

We used a decarburization technique to observe carbon diffusion in iron. Although decarburization is usually carried out in a wet hydrogen atmosphere, we applied the reaction of carbon with titanium, which has a very low carbide formation energy, to decarbutization. The materials used were hypoeutectoid steel with 0.09 mass% C and commercially available pure titanium. The steel and titanium plates were explosively joined at The Shock Wave and Condensed Matter Research Center of Kumamoto university. Specimens were cut from the joined steel/titanium samples into the plate of 10 mm × 8 mm × 1.4 mm in dimensions. Decarburization annealing was carried out at temperatures ranging from 873 to 1323 K with a magnetic field or a magnetic field gradient in a vacuum of 1.0×10^{-3} Pa using a specially designed superconducting magnetic field heat treatment system ($H_{\mathrm{max}} = 6$T, $T_{\mathrm{max}} = 1773$ K) shown in Fig. 1. The system was composed of a helium free superconducting magnet (Sumitomo Heavy Machine Corporation) and a high-temperature furnace with molybdenum sheet heating element (Futek Furnace inc.). A specially designed carbon sample holder was used for decarburization annealing. Tungsten sheets were inserted between the sample and the carbon holder to prevent the sample from a contact with the carbon holder. The applied magnetic field strength was 6T and magnetic field gradients ranged from 30 to 45 T/m ($H(\mathrm{d}H/\mathrm{d}x) = 0.92 \times 10^2$ T^2/m –2.08×10^2 T^2/m). A magnetic field gradient was applied by placing the samples apart from a uniform magnetic field region. Fig. 2 shows the variation of magnetic field strength H and magnetic field gradient $\mathrm{d}H/\mathrm{d}x$ along the position from the center of the magnet for different nominal magnetic field strength H_0. The direction of the applied magnetic field was perpendicular to the titanium/steel interface, that is, parallel to the diffusion direction.

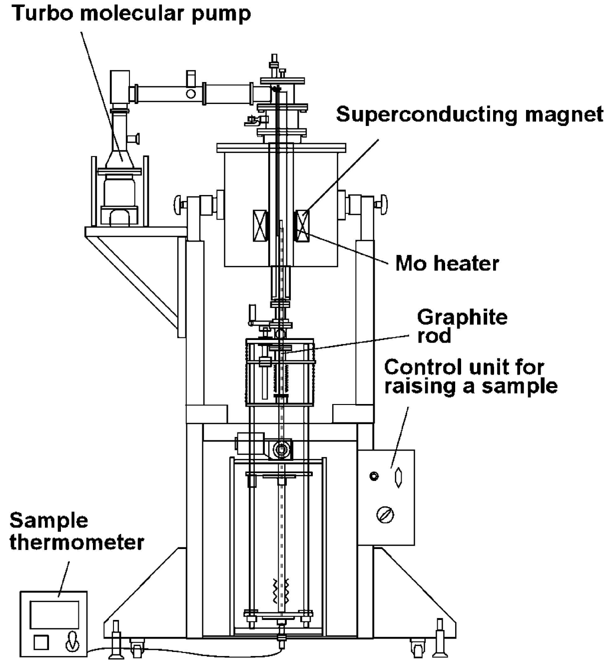

Figure 1 Schematic illustration of a superconducting magnetic field heat treatment system.

After decarburization annealing, the samples were mechanically polished with waterproof papers of #320–#2400, and buff-polished with alumina particles of 3, 1, 0.3 and 0.1 μm. Finally the samples were electrically polished in a mixture of acetic acid, hyperchloric acid and methanol with 9:1:1 by volume at a current density of 1.75 A/cm^2 to remove residual surface layer, and etched in a mixture of nitric acid and ethanol with 2:78 by volume for SEM observations.

The carbon concentration profiles were drawn by measuring the area fraction of pearlite phase as a function of the distance from the titanium/steel interface. During decarburization annealing, titanium atoms diffuse into the steel region, which is the opposite direction to carbon diffusion, and form titanium carbide particles. Then, we measured the penetration depth of titanium into the hypoeutectoid steel and evaluated diffusivity of titanium in iron as well.

3. Results and discussion

3.1. Decarburization annealing

Fig. 3 shows SEM micrographs for titanium/steel samples annealed at 873 K (a), 1123 K (b) in α-iron temperature region and 1223 K (c) in γ-iron temperature region for 10 h without a magnetic field. The arrow at the left-hand side of each micrograph indicates the titanium/steel interface. A pearlite phase can be seen as bright areas in these micrographs. The layer near the titanium/steel interface observed in Fig. 3c is the region where titanium atoms penetrated into the steel to form titanium carbide particles. These micrographs reveal that decarburization occurs inadequately at 873 and

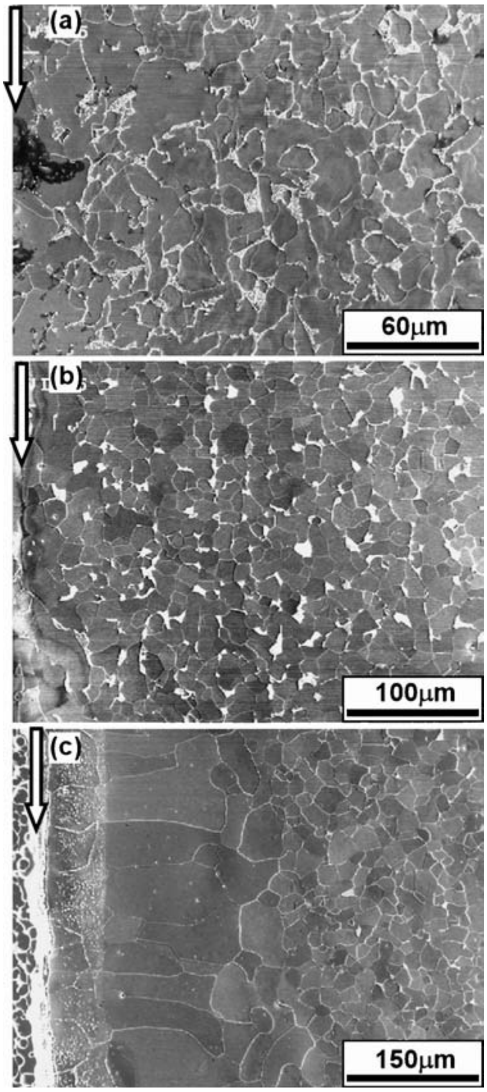

Figure 3 SEM micrographs for Ti / Steel samples annealed at (a) 873 K, (b) 1123 K and (c) 1223 K for 10 h without magnetic field. The arrows show the Ti/Steel interface.

1123 K in the α-iron temperature region, while sufficient decarburization occurs at 1223 K in the γ-iron temperature region. The diffusivity of carbon is approximately two order magnitude higher in α-iron than in α-titanium [22, 23]. Hence, a dense titanium carbide layer is probably formed at the titanium/steel interface due to accumulation of carbon atoms at the interface. Because of extremely low diffusivity of carbon in titanium carbide [24], the titanium carbide layer at the interface will act as a barrier against the diffusion of carbon at 873 and 1123 K. Therefore, the decarburization does not proceed in the α-iron temperature regime.

On the other hand, the diffusivities of carbon are almost the same both in γ-iron and in β-titanium at 1223 K [23, 25]. Thereby, such the dense titanium carbide layer acting as the barrier against the carbon diffusion dose not form at the interface, resulting in occurrence of decarburization. Hence, decarburization annealing in a magnetic field was conducted at temperatures above 1223 K.

3.2. Diffusion of carbon in γ-iron in a magnetic field

Fig. 4 presents SEM micrographs showing changes in microstructure along the direction from the titanium/steel interface to the steel side after decarburization annealing at 1273 K for 10 h without a magnetic field (a) and with a 6T magnetic field (b). The direction of the applied magnetic field is parallel to the direction of carbon diffusion as shown by a bold arrow in the micrograph. Carbon concentration profiles after the decarburization annealing were drawn by measuring the pearlite area fraction at different positions along the direction perpendicular to the titanium/steel interface, because the area fraction of pearlite phase to ferrite phase reflects carbon concentration at a given area. Fig. 5 shows the area fraction of pearlite phase as a function of the distance from the titanium/steel interface after decarburization annealing at different temperatures for 10h with and without a 6T magnetic field: (a)1223, (b) 1273 and (c) 1323 K. The area fraction in Fig. 5 was normalized by the original fraction of pearlite phase in each hypoeutectoid steel sample. It is found that the fraction of pearlite phase is somewhat larger in the samples decarburized with a magnetic field than in the samples without a magnetic field. This finding

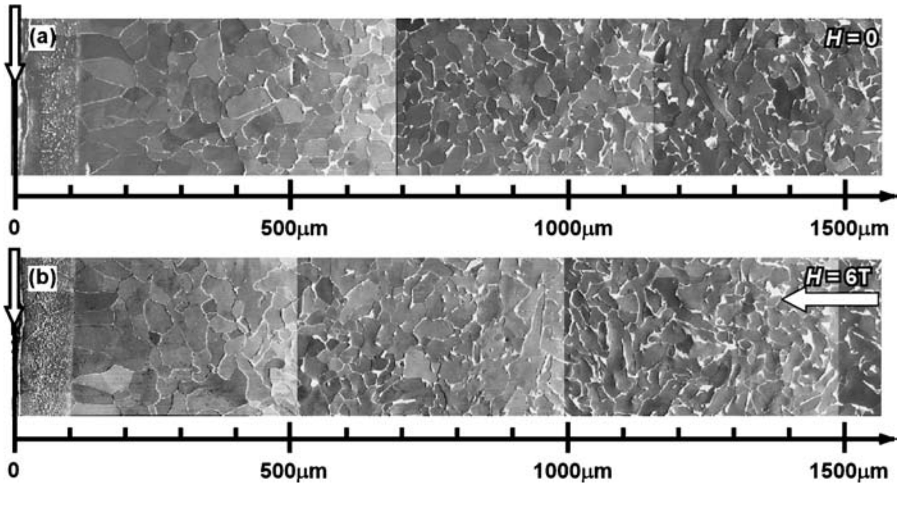

Figure 4 SEM micrographs showing the changes in microstructure due to decarburization along the direction from the Ti/Steel interface to steel interior. The decarburization annealing was carried out at 1273 K for 10 h (a) without and (b) with a 6T magnetic field.

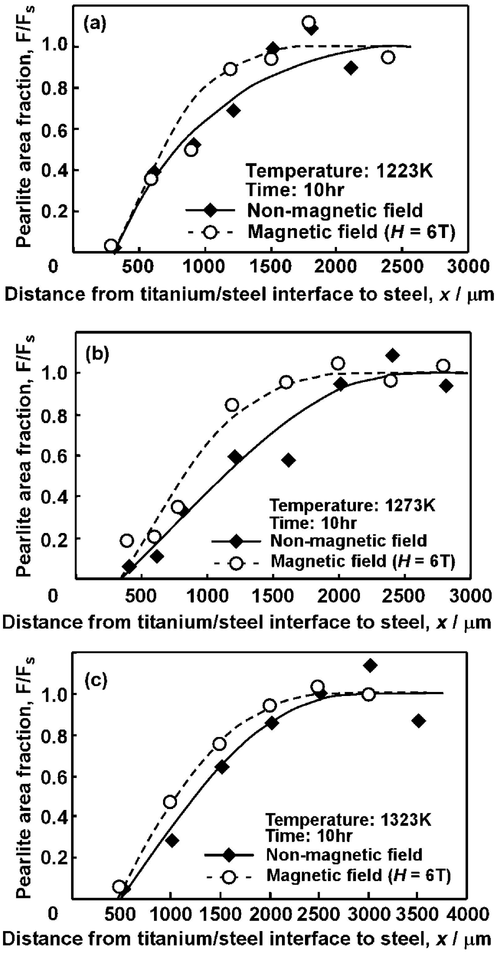

Figure 5 The reduced pearlite area fraction profiles in samples decarburized at (a) 1223 K, (b) 1273 K and (c) 1323 K for 10 h with and without a magnetic field. The fractions were normalized by the original area fraction in the hypoeutectoid steel sample.

suggests that a magnetic field retards carbon diffusion in γ-iron.

Diffusion coefficients of carbon in γ-iron were determined from the normalized pearlite area fraction carves shown in Fig. 5. An analytical expression for the carbon concentration profile due to decarburization can be given by the Equation 1 by solving Fick's second law using the boundary conditions: $c(x, t) = 0$ at $x = 0, t > 0$ and $c(x, t) = c_0$ at $x > 0, t = 0$ where c_0 is the original carbon concentration .

$$c(x, t) = c_0 erf\left(\frac{x}{2\sqrt{Dt}}\right) \qquad (1)$$

where "*erf*" stands for the error function and D diffusion coefficient. Since $erf(0.5) \approx 0.5$, the diffusion coefficient of carbon at a given temperature can be obtained by measuring the distance where the normalized pearlite fraction is 0.5.

Fig. 6 shows the Arrhenius plot for the measured diffusion coefficients of carbon in γ-iron with a 6T magnetic field and without a magnetic field. For comparison, the reported data [25], which was obtained without a magnetic field, is shown by a broken line in Fig. 6. Of particular importance is the finding that the diffusion coefficient of carbon in γ-iron reduced to approximately half by the application of a 6T magnetic field.

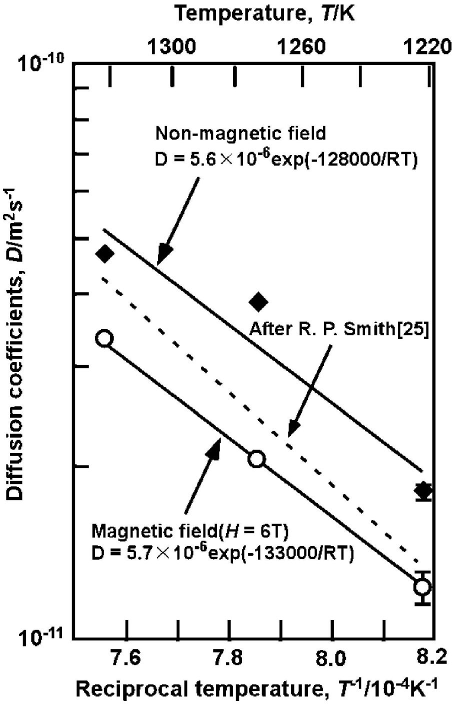

Figure 6 Arrhenius plots for diffusion coefficients of carbon in γ-iron with and without a magnetic field.

The activation energies are evaluated to be approximately 130 kJ/mol irrespective of whether a magnetic field is applied within the experimental error. Here a question arises: how does a magnetic field affect the activation energy and/or the frequency factor? It seems that from Fig. 6 the magnetic field could not affect the activation energy but could affect the frequency factor for carbon diffusion in γ-iron. Also, Tsurekawa *et al.*, who carried out sintering of pure iron compacts in a magnetic field, reported that the activation energies for both densification and grain growth do not change by application of a magnetic field even though a magnetic field can enhance the densification and grain growth during sintering [26]. However, further examination should be required to confirm the effect.

3.3. Diffusion of carbon in γ-iron in magnetic field gradient

Fig. 7 shows the reduced pearlite area fraction profiles after decarburization annealing at 1223 K for 5 h in

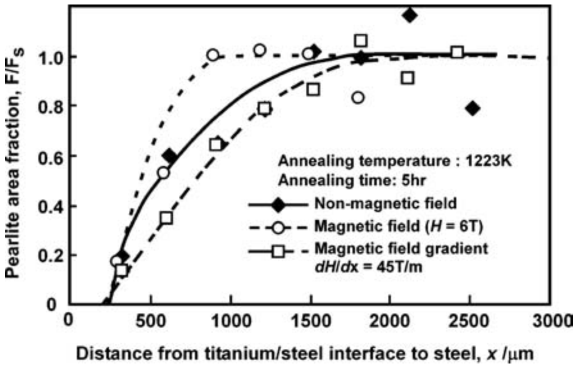

Figure 7 The reduced pearlite area fraction profiles in samples decarburized at 1223 K for 5 h without a magnetic field, with a 6T magnetic field and with a 45 T/m magnetic field gradient.

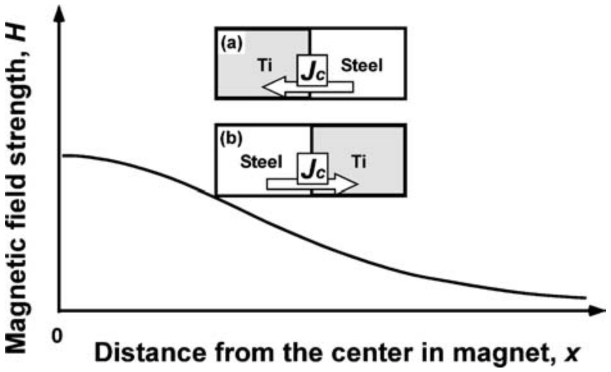

Figure 8 Definition of "negative" and "positive" magnetic field gradient: (a) negative and (b) positive. J stands for a flux of carbon atoms produced by a concentration gradient.

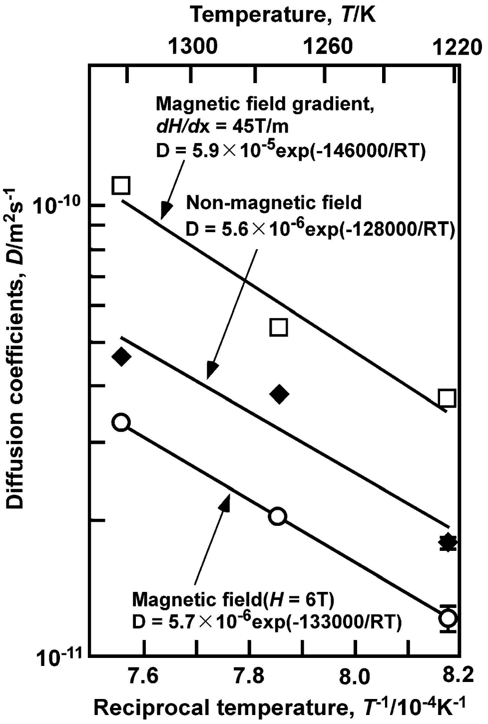

Figure 9 Temperature dependence of diffusion coefficients of carbon in γ-iron in a 45 T/m magnetic field gradient. For comparison, the data shown in Fig. 6 are also plotted in this figure.

a magnetic field gradient of 45 T/m ($H(\mathrm{d}H/\mathrm{d}x) = 2.08 \times 10^2$ T^2/m) as a function of the distance from the titanium/steel interface. For comparison, the profiles obtained from the samples decarburized with a 6T magnetic field and without a magnetic field are also shown in Fig. 7. Here, we define the sign of a magnetic field gradient as schematically shown in Fig. 8. When a flux of carbon atoms produced by a concentration gradient occurs towards negative direction, the magnetic field gradient is defined as "negative". It is found that the area fraction of pearlite phase is decreased by application of the negative magnetic field gradient at a given position in contrast to the effect of a uniform magnetic field on decarburization. In addition, the distance at which the reduced pearlite area fraction approaches a unity seems to be longer when the magnetic field gradient is applied.

Fig. 9 shows the temperature dependence of diffusivities of carbon in γ-iron in a magnetic field gradient of 45 T/m. For comparison, the diffusivity in the absence of a magnetic field and in a 6T magnetic field are shown in the figure. It is found that carbon diffusion is enhanced by about twice in a 45 T/m negative magnetic field gradient, while a uniform magnetic field retards carbon diffusion. No noticeable influence of the magnetic field gradient on the activation energy for carbon diffusion in γ-iron is observed within an experimental error as in the case of a uniform magnetic field.

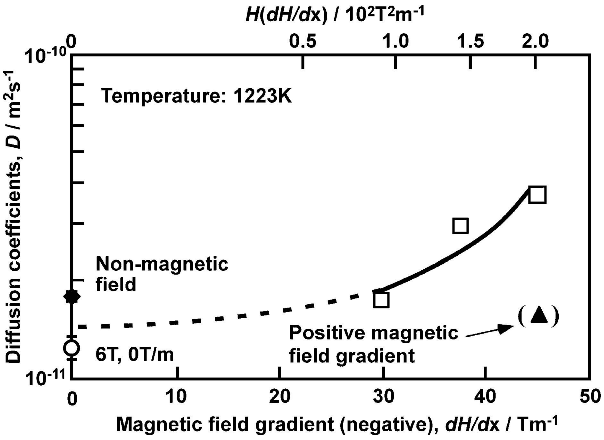

Figure 10 The dependence of magnetic field gradient strength on diffusivity of carbon in γ-iron at 1223 K.

The diffusion coefficients of carbon in γ-iron in different magnetic field gradients are shown in Fig. 10. Evident from Fig. 10 is the finding that the diffusivity of carbon in γ-iron increases with increasing the magnetic field gradient strength. It should be noted that the datum point indicated by a solid triangle in Fig. 10 was obtained when a 45 T/m "positive" magnetic field gradient was applied. It is interesting to observe that the positive magnetic field gradient is prone to reduce the diffusivity of carbon in γ-iron. From these results, this is probably because a magnetic field gradient produces a flux of carbon atoms in addition to that produced by a concentration gradient. The positive magnetic field gradient will give rise to a flux of carbon atoms against the flux due to a concentration gradient, resulting in a decrease in the diffusivity of carbon.

The measured diffusion coefficients of carbon in γ-iron in a uniform magnetic field and in magnetic field gradients are summarized in Table I.

3.4. Diffusion of titanium in γ-iron in a magnetic field

Diffusion of titanium in γ-iron occurs by a vacancy mechanism, while carbon atoms diffuse by an interstitial mechanism. Thus, a magnetic field may affect a different influence on diffusion between titanium and carbon in γ-iron. Fig. 11 shows the Arrhenius plot of diffusion coefficients of titanium in γ-iron in the absence of a magnetic field, in a 6T magnetic field and in a 45 T/m magnetic field gradient. In contrast to the findings of the magnetic field effects on carbon diffusion, both the magnetic field and the magnetic field gradient cannot exert a pronounced influence on the diffusivity of titanium in γ-iron. This finding is consistent with

TABLE I Diffusion coefficients of carbon in γ-iron in an magnetic field and in magnetic field gradients.

Temperature T/K	Annealing time t/h	Magetic field H/T	Magnetic field gradient $\mathrm{d}H/\mathrm{d}x$/ Tm^{-1}	 $H(\mathrm{d}H/\mathrm{d}x)/10^2\ \mathrm{T^2m^{-1}}$	Diffusivity, $D/\mathrm{m^2s^{-1}}$
1223	5	0	—	—	1.8×10^{-11}
		6	—	—	1.1×10^{-11}
		—	30(n)	0.92(n)	1.8×10^{-11}
		—	37(n)	1.44(n)	3.0×10^{-11}
		—	45(n)	2.08(n)	3.7×10^{-11}
		—	45(p)	2.08(p)	1.6×10^{-11}
	10	0	—	—	1.8×10^{-11}
		6	—	—	1.3×10^{-11}
1273	5	—	45(n)	2.08 (n)	5.4×10^{-11}
		0	—	—	3.8×10^{-11}
	10	6	—	—	2.0×10^{-11}
1323	5	—	45(n)	2.08(n)	1.1×10^{-10}
	10	0	—	—	4.6×10^{-11}
		6	—	—	3.3×10^{-11}

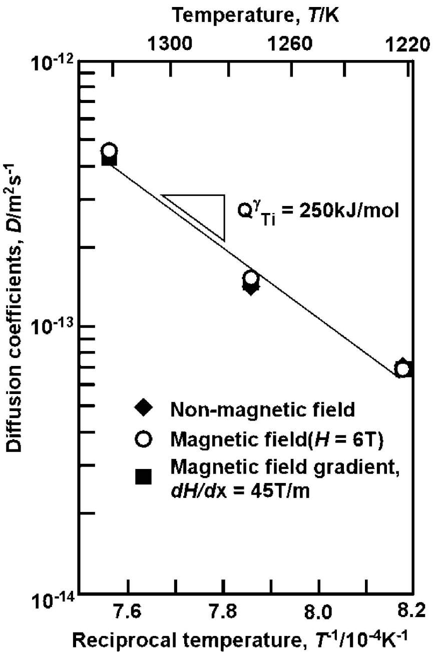

Figure 11 Arrhenius plots of diffusion coefficients of titanium in γ-iron without a magnetic field, with a 6T magnetic field and with a 45 T/m magnetic field gradient.

the result obtained by Nakajima *et al.* [19], who examined nickel diffusion by the vacancy mechanism in titanium with a magnetic field of 4T. The activation energy 250 kJ/mol obtained from this work is well consistent in the previously reported value [27] , irrespective of whether a magnetic field or a magnetic field gradient is applied. The measured diffusion coefficients of titanium in γ-iron are summarized in Table II.

3.5. Origin of magnetic field effects on diffusion

3.5.1. *Effect of a uniform magnetic field*

The carbon diffusion in γ-iron was found to be retarded in a uniform magnetic field. It has been recognized that the Arrhenius plots for the self-diffusion and carbon diffusion coefficients in iron shows an anomaly below the Curie temperature in ferromagnetic state: the plots of $\ln D$ vs. $1/T$ deviate downwards from the extrapolated liner Arrhenius relationship owing to the magnetic spin ordering [28, 29]. In a higher magnetic field, magnetic field-induced magnetization occurs even in the paramagnetic state [14]. Therefore, the observed retardation of carbon diffusion in γ-iron in a magnetic field would be explained by magnetic field induced magnetization.

In contrast to the effect of a magnetic field on carbon diffusion in γ-iron, there is no significant influence of a magnetic field on the titanium diffusion by the vacancy mechanism in γ-iron. Budke *et al.* [29] have pointed out that the carbon diffusion by interstitial mechanism in iron responds more strongly to the stiffening of the lattice due to the magnetic ordering than the vacancy mediated self-diffusion. This is because a stiffening of the lattice leads to an increase the activation barrier for diffusion jumps. Actually, Köster [30] has shown the anomaly in the elastic modulus below the Curie temperature in ferromagnetic materials like iron and nickel: the plot of elastic modulus vs. temperature deviates upwards from the extrapolated linear relationship between them above the Curie temperature. In case of the diffusion by the vacancy mechanism, a vacancy in the neighborhood of the jumping atom gives rise to a certain relaxation of its surrounding. The interstitially diffusing atoms do not take advantage of such a relaxation. In addition, we should take account of the influence of a magnetic field on the vacancy formation energy. It is considered that the vacancy formation energy should be increased in a magnetic field. This is because the formation of a vacancy may cause a decrease in magnetization of iron, and then an increase in the magnetic free energy per unit volume in a magnetic field. However, the volume of a vacancy is so small that the contribution of the applied magnetic field to the increase in the vacancy formation energy will be negligible. As was mentioned above, an external magnetic field involves a magnetic field-induced magnetization even above the Curie temperature. Therefore, Budke's assumption is a possible explanation for the observed discrepancy in the effects of a magnetic field on diffusion of carbon and titanium in γ-iron.

TABLE II Diffusion coefficients of titanium in γ-iron in a magnetic field and in magnetic field gradients

			Magnetic field gradient		
Temperature *T/K*	Annealing time *t/h*	Magnetic field *H/T*	$\mathrm{d}H/\mathrm{d}x/\ \mathrm{Tm}^{-1}$	$H(\mathrm{d}H/\mathrm{d}x)/10^2\ \mathrm{T}^2\mathrm{m}^{-1}$	Diffusivity, $D/\mathrm{m}^2\mathrm{s}^{-1}$
1223	5	—	45	2.08	6.9×10^{-14}
	10	0	—	—	7.0×10^{-14}
		6	—	—	6.8×10^{-14}
1273	5	–	45	2.08	1.5×10^{-13}
	10	0	—	—	1.5×10^{-13}
		6	—	—	1.6×10^{-13}
1323	5	—	45	2.08	4.3×10^{-13}
	10	0	—	—	4.6×10^{-13}
		6	—	—	4.7×10^{-13}

In addition to the stiffening due to magnetic ordering, the influence of magnetostriction on diffusion should be taken into consideration. Recently, Mironov *et al.* [31] have pointed out that the effect of magnetic field strength on nickel diffusion in α-iron [20], which was mentioned in the introduction, qualitatively agree with that on magnetostriction constant in iron. However, their suggestion is not to be simply acceptable because Honda reported that the magnetostriction in iron is always an increase when the temperature exceeds 573 K [32]. Nevertheless, the magnetostriction still draws our attention as a possible origin of the observed magnetic field effect on diffusion. In general, the magnetostriction as well as the magnetization approaches zero near the Curie point with increasing temperature. In an external high magnetic field, however, a forced magnetostriction is expected to occur even above the Curie temperature. If the forced magnetostriction yields a lattice contraction, the activation barrier to carbon diffusion by the interstitial mechanism could be increased. Unfortunately, little experimental information is presently available about the details of the forced magnetostriction particularly in γ-iron temperature region. Thus, we cannot provide a conclusive evidence for the origin of the observed magnetic field effect on diffusion.

3.5.2. Effect of a magnetic field gradient

We found that carbon diffusion in γ-iron is enhanced by a "negative" magnetic field gradient but is retarded by a "positive" magnetic field gradient. Let us consider the effect of the magnetic free energy in γ-iron matrix on carbon diffusion. The "negative" magnetic field gradient involves a "positive" magnetic free energy gradient in γ-iron matrix because of a positive value of susceptibility for γ-iron at annealing temperature: that is, the magnetic free energy of γ-iron decreases with approaching to the titanium/steel interface in the "negative" magnetic field gradient. For the paramagnetic state like γ-iron, the magnetic free energy is given for unit volume by,

$$U = -\frac{1}{2}\mu_0 \chi H^2, \tag{2}$$

where μ_0 is the magnetic permeability of vacuum, χ a susceptibility. Here, the demagnetizing term was omitted for simplification. From the Equation 2, the magnetic free energy would increase with increasing the carbon concentration in iron because the susceptibility χ of iron decreases with increase in carbon concentration [33]. It is reasonable to consider that the magnetic free energy gradient in γ-iron matrix would produce a flux of carbon atoms towards a lower magnetic free energy regime to achieve a homogeneous magnetic free energy in γ-iron matrix. Assuming that a magnetic potential gradient in γ-iron matrix as well as a concentration gradient can produce a flux of carbon atoms, the equation of a net flux of carbon atoms is given by the following equation.

$$J = -D\left(\frac{\mathrm{d}C}{\mathrm{d}x} + \frac{C}{kT}\frac{\mathrm{d}U}{\mathrm{d}x}\right), \tag{3}$$

where J is the flux of atoms, U magnetic free energy, and k and T usual meanings. From Equations 2 and 3, the following relation can be obtained:

$$J = -D\left(\frac{\mathrm{d}C}{\mathrm{d}x} + \frac{C\mu_0}{kT}\left(-\chi H\frac{\mathrm{d}H}{\mathrm{d}x}\right)\right). \tag{4}$$

Therefore, when a "negative" magnetic field gradient, $(\mathrm{d}H/\mathrm{d}x) < 0$, is applied, the total flux of carbon atoms would be increased because of a positive sign of susceptibility for γ-iron. This is a possible explanation for the enhancement of carbon diffusion in γ-iron by application of the "negative" magnetic field gradient.

4. Conclusions

Effects of a uniform magnetic field and a magnetic field gradient on carbon diffusion by interstitial mechanism and titanium by the vacancy mechanism in γ-iron were studied. The chief results obtained are as follows.

(1) The diffusivity of carbon in γ-iron is lowered in a 6T magnetic field, while no noticeable influence of a magnetic field on diffusion of titanium in γ-iron is observed. The stiffening of lattice due to field-induced magnetic ordering is responsible for a increase in activation barrier for jumping carbon atoms.

(2) A "negative" magnetic field gradient can enhance carbon diffusion in γ-iron. The higher the negative magnetic field gradient, the more diffusion of carbon is enhanced. On the other hand, a "positive" magnetic field causes a decrease in diffusivity of carbon in γ-iron. The magnetic free energy gradient in γ-iron matrix due to the magnetic field gradient would produce a flux of carbon atoms to achieve a homogeneous magnetic free energy in γ-iron matrix.

Acknowledgements

The authors would like to express their hearty thanks Mr. T. Tsurudome (Sumitomo Heavy Industries Ltd.) for supplying the magnetic field gradient data shown in Fig. 2, Mr. T. Matsuzaki for his help in the experimental work and Dr. K. Kawahara for useful discussion. This work was supported by the Grant-in-Aid for Basic Research (A) (Grant No.15201015) from the Ministry of Education, Science, Sports and Culture of Japan and ISIJ (The Iron and Steel Institute of Japan) Research Promotion Grant. Their supports are very much appreciated.

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Received 29 July 2004
and accepted 31 January 2005