Effects of Carbon Content on the Diffusion Bonding of Iron and Steel to Titanium

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1) Welding

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A commercially pure titanium has been diffusion-bonded to iron and steels of various carbon contents: low carbon steel with 0.01 % C (ULC), mild steel with 0.19 % C (S20C) and spheroidal graphite cast iron with 3.75 % C (FCD). The tensile strength of joints bonded below 900°C was hardly influenced by the carbon content, and increased with the rise in bonding temperature. Above 900°C however, the strength of the ULC/Ti joint was almost independent of the bonding temperature, that of the S20C/Ti joint decreased with the rise in bonding temperature, and that of the FCD/Ti increased. The interlayer forming along the bond interface consisted only of TiC in the FCD/Ti joint and of TiFe and TiFe₂ in the ULC/Ti. Since Ti and Fe diffused over much longer distances in the ULC/Ti than in the FCD/Ti, the TiC layer may be considered to interfere with the interdiffusion of Ti and Fe, and consequently to suppress the formation of TiFe and TiFe₂ which decrease the joint strength more seriously than TiC. The interlayer in the S20C/Ti consisted of TiC as well as TiFe and TiFe₂. The coexisting carbide and intermetallic compounds in the interlayer seems to have more detrimental effect on the joint strength than those formed individually.

KEY WORDS: diffusion bonding; composite metal; titanium; carbon steel; cast iron; intermetallic compound; joint strength; carbide; diffusion.

1. Introduction

With the recent developments of mass production technology of clad steels, their application to industries has been extended very widely, ranging from chemical plants to dam gates.¹⁾ They are produced by various processes, such as hot rolling,²⁾ explosive bonding,³⁾ overlay welding,^{4,5)} enshrouding casting⁶⁾ and diffusion bonding.⁷⁾ The titanium clad steel, one of the most widely used clad steels, is produced by the hot rolling and explosive bonding processes.

Since brittle intermetallic compounds are formed in the binary system of iron and titanium, many investigations⁸⁻¹⁰⁾ have been reported of the effect of the intermetallic compound on the bond strength, including the application of the intermediate metal to preventing the formation of the intermetallic compound. For example, when steels of various carbon contents were bonded to a titanium by the hot rolling process without an intermediate metal, the shear strength of the joint scattered to a large extent, and so no systematic relation could be found between the strength and the bonding temperature.¹⁰⁾ On the other hand, Arata *et al.*¹¹⁾ reported that maximum joint strength could be obtained at bonding temperatures from 800 to 920°C in the diffusion bonding of a mild steel (JIS SS41) to a titanium, but Shimazaki *et al.*⁸⁾ reported that the joint strength increased with the rise in bonding temperature. In these studies, it was also suggested that the titanium carbide was formed at the bond interface through the reaction of

titanium with carbon in the steel and exerted detrimental effects on the joint strength.

The significant discrepancy in the joint strength as mentioned above should be considered in connection with the difference in the kind and/or morphology of the intermetallic compound formed at the bond interface. The main purpose of the present investigation is, therefore, to make clear the relation between the joint strength and the formation of the intermetallic compound in the diffusion bonding^{12,13)} of titanium to iron-base alloys of various carbon contents with particular reference to the formation of titanium carbide.

2. Experimental Procedures

Chemical compositions of the base metals used are shown in Table 1. A low carbon steel (ULC), mild steel (S20C) and ferritic spheroidal graphite cast iron (FCD) were used as the iron-base metals, and commercially pure Ti (JIS TB35C) as the titanium base metal. The base metal was cut in a lathe to rod specimens $\phi 14 \times 20$ mm for the diffusion bonding. One of the end faces of the specimen, the face to be bonded, was finished by grinding on #240 emery paper, and subsequently subjected to degreasing with acetone and to wire-brushing (stainless steel wire 0.3 mm diameter and 20 mm long) just before the bonding.

The diffusion bonding was carried out in a vacuum of the order of 10^{-4} Torr using an apparatus as schematically shown in Fig. 1. A couple of specimens

were set between the pressing axes of alumina rods, so that the brushing marks were crossed almost perpendicularly with each other. The bonding pressure, which was applied with a hydraulic press, was monitored with a small load cell. The set specimens were heated in a infrared image furnace at the heating rate of 1°C/s , and the temperature of the specimens was kept at a desired bonding temperature to an accuracy of $\pm 1^{\circ}\text{C}$. The temperature of the specimen was monitored with a CA thermocouple (0.3 mm thick) percussion-welded to the specimen within 2 mm from the bond interface. The bonding pressure and time were fixed at 0.15 kgf/mm² and 30 min, respectively.

Joints obtained were machined to tensile test specimens in the form of bars with the gauge length of 15 mm and the gauge diameter of 7 mm. The tensile test was carried out at the crosshead speed of 1.0 mm/min at room temperature.

The intermetallic compound formed at the bond interface was identified by analyses of X-ray diffraction pattern from fractured surfaces of joints using Cu-K α radiation. Since all joints obtained were fractured in the close vicinity of or at the bond interface with rather flat fractured surfaces, the X-ray diffraction pattern may be consisted to indicate correctly the intermetallic compound formed at the bond interface.

Table 1. Chemical composition of base metals used. (wt%)

	C	Si	Mn	P	S	Mg
ULC	0.01	0.26	0.19	0.011	0.008	—
S20C	0.19	0.07	0.72	0.026	0.026	—
FCD	3.75	3.00	0.13	0.038	0.010	0.042

	Fe	N	O	H
Ti	0.12	0.0065	0.126	0.0035

Fig. 1. Schematic drawing of diffusion bonding apparatus.

3. Experimental Results

3.1. Diffusion Bonding of FCD to Ti

Microstructures of FCD/Ti joints bonded at 800 and 1 000°C are shown in Figs. 2(a) and 2(b), respectively. No interlayer could be found at bonding temperatures T_B below 800°C by the optical microstructure, while a interlayer 4–5 μm thick was observed at T_B of 1 000°C. X-ray diffraction patterns from fractured surfaces after the tensile test are shown in Fig. 3. As shown in Fig. 3, on the fractured surface of the FCD side, no diffraction line from intermetallic compounds of Fe–Ti binary system could be found, but those from TiC were observed in addition to those from α -Fe base metals. On the Ti side, more intensive diffraction lines from TiC were observed than those on the FCD side. The fact that the diffraction line from TiC was observed on both sides suggests that the joint fractured in the TiC layer and/or at the

Fig. 3. X-ray diffraction patterns from fractured surfaces of a FCD/Ti joint bonded at 1 000°C.

(a) Bonding temperature $T_B = 800^{\circ}\text{C}$
(b) $T_B = 1\,000^{\circ}\text{C}$

Fig. 2. Microstructures of FCD/Ti joints.

interfaces between TiC and base metals. Since the intensity of the diffraction lines from TiC was higher on the Ti side, the bond strength of the Ti/TiC interface was probably higher than that of the FCD/Ti interface.

In order to investigate the dependence of the thickness of TiC layer on the bonding temperature, the intensity of the diffraction line from TiC {111} plane is plotted against the bonding temperature in Fig. 4. The intensity of the diffraction line increased with the rise in bonding temperature, suggesting that the TiC layer became thicker with the rise in bonding temperature.

Distributions of Fe, Ti and C in a FCD/Ti joint bonded at 1 000°C were analyzed by EPMA as shown in Fig. 5. The distribution curve of carbon had a peak at the bond interface, suggesting that TiC was formed in the interlayer as shown in Fig. 2(b). The amounts of Ti diffusing into FCD and of Fe diffusing into Ti were much smaller than those in a ULC/Ti

joint bonded under the same conditions (see Fig. 7). These results suggest that the interdiffusion of Ti and Fe during the diffusion bonding was interfered by the presence of the TiC layer. The effects of bonding temperature on the tensile strength of FCD/Ti joints are shown in Fig. 6. Though all the joints tested were fractured in the close vicinity of or at the bond interface, their strength increased with the rise in bonding temperature. Thus, the increase in the thickness of the TiC layer did not decrease the joint strength at bonding temperatures below 1 000°C. However, the increase in the joint strength tends to be saturated at bonding temperatures above 900°C. This suggests that the tensile strength of FCD/Ti joints was influenced by the growth of the TiC layer and that it would be rather difficult to obtain bond strength not less than the strength of the base metal by increasing the bonding temperature.

Though the FCD used contained 3.0 % Si, it was not observed that Si had any significant effects on the formations of TiC or intermetallic compounds of Ti-Si system.

3.2. Diffusion Bonding of ULC to Ti

Microstructures of ULC/Ti joints bonded at 800, 900 and 1 000°C are shown in Figs. 7(a), 7(b) and 7(c), respectively. No intermetallic compound layer could be recognized at the bonding temperature of 800°C. As shown in Figs. 7(b) and 7(c), however, an interlayer and remarkable change in the microstructure of Ti adjacent to the bond interface were observed at the bonding temperatures of 900 and 1 000°C, i.e., above $\alpha \rightleftharpoons \beta$ transformation temperature of Ti. That is, in Fig. 7(c), the dark thin layer just next to the bond interface indicated by the arrow 'C' is regarded as the interlayer consisting of intermetallic compounds. The white non-etched region indicated by 'D' is the β -Ti phase formed by the diffusion of Fe into Ti, as can be seen from the results of X-ray diffraction analysis and EPMA described later (see Figs. 8 and 9). This β -Ti region was much harder than the Ti base metal; i.e., the Vicker's hard-

Fig. 4. Effect of bonding temperature on the peak height of {111}-TiC diffraction line from fractured surfaces of FCD/Ti joints.

Fig. 5. EPMA line-scanning profiles across the bonding interface of a FCD/Ti joint bonded at 1 000°C.

Fig. 6. Tensile strength of FCD/Ti joints as a function of bonding temperature.

Fig. 7. Microstructures of ULC/Ti joints.

ness number of the β -Ti region, which decreased with the increase in the distance from the bond interface, ranged from 463 to 365 as compared to that of the Ti base metal ~ 170 . Region 'E', showing Widmanstätten's structure, was formed as a result of $\alpha \rightarrow \beta$ transformation during the cooling process after the bonding. As shown in Figs. 7(b) and 7(c), the width of these phases increased with the bonding temperature. This is consistent with the result reported by Shimazaki *et al.*⁸⁾ that the width of white layer increased with the bonding temperature.

X-ray diffraction patterns from fractured surfaces of a joint bonded at 1000°C are shown in Fig. 8. On the fractured surface of the ULC side, diffraction lines from TiFe_2 , TiFe , α -Ti and β -Ti were observed in addition to those from α -Fe, but those from TiC could not be detected. On the fractured surface of Ti side, strong diffraction lines from TiFe and rather weak diffraction lines from TiFe_2 were observed in addition to those from α -Ti and β -Ti. These results indicate that the interlayer shown in Fig. 7(c) consisted of TiFe and TiFe_2 . Since diffraction lines from TiFe , TiFe_2 and β -Ti could be observed on the fractured surfaces of both ULC side and Ti side, the crack was probably propagated through the intermetallic compound layer and partially the β -Ti region. This suggests that the strength of this joint is controlled by the strength of the interlayer consisting of TiFe and TiFe_2 and that of the β -Ti region.

In order to assign the phases identified by the X-ray diffraction analysis to the microstructures shown in Fig. 7(c), the distribution of Fe, Ti and C was investigated with EPM analysis. As shown in Fig. 9, considerable amounts of Fe diffused into Ti base metal, while no detectable change in C content could be observed. Region 'D', where a significant increase in Fe content due to the diffusion was observed, can be assigned to β -Ti, considering that Fe is an element stabilizing β -Ti. The amount of Ti diffusing into ULC was rather small, though a small increase in Ti content was observed in region 'B'. Since α -Fe containing 2–7% Ti is in equilibrium with TiFe_2 at 1000°C,¹⁴⁾ region 'B' in Fig. 7(c) may be regarded as the α -Fe. The interlayer 'C' probably consisted of TiFe and TiFe_2 , though it was too thin to detect with EPMA.

The relation between tensile strength of the joint and bonding temperature is shown in Fig. 10. The tensile strength increased with the rise in bonding temperature up to about 900°C, and became almost

Fig. 8. X-ray diffraction patterns from fractured surfaces of a ULC/Ti joint bonded at 1000°C.

Fig. 9. EPMA line-scanning profiles across the bonding interface of a ULC/Ti joint bonded at 1000°C. Symbols A–E correspond to those in Fig. 7(c).

constant above 900°C. Since TiFe and TiFe_2 were observed on the fractured surfaces of both Ti side and ULC side, it can be accounted for by the growth of these intermetallic compound layers that the tensile strength of joint did not increased with the rise in bonding temperature above 900°C. This means that the tensile strength of joint bonded at temperatures

above 900°C is controlled mainly by the strength of the intermetallic compound layers. The layers of TiFe and TiFe₂ seems to have more detrimental effect on the joint strength than TiC, since the tensile strength of the FCD/Ti joint in which only TiC was formed increased with bonding temperature even above 900°C as shown in Fig. 6. The increase in the joint strength with bonding temperature below 900°C can be accounted for by the decrease in the unbonded area,¹⁵⁾ since SEM observation of the fractured surface of joints showed that unbonded area characterized by the wire-brushing mark decreased with the rise in bonding temperature.

3.3. Diffusion Bonding of S20C to Ti

Microstructures of S20C/Ti joints bonded at 880 and 1 000°C are shown in Figs. 11(a) and 11(b), respectively. At the bonding temperature of 880°C, a discontinuous dark layer, which probably consisted of TiC, was observed at the bond interface. A decarburized band characterized by the disappearance of pearlite structure was also observed in the S20C adjacent to the bond interface. At the bonding temperature of 1 000°C, the decarburized band became wider, and a remarkable difference in the width of reaction or diffusion zone was observed over the whole bond interface. That is, the bond zone consisted of two characteristic places; the place where regions similar to 'B' and 'D' in the ULC/Ti joint were formed and the place where Ti adjacent to the bond interface showed Widmanstätten's structure and region 'B' was not observed.

Fig. 10. Tensile strength of ULC/Ti joints as a function of bonding temperature.

(a) $T_B = 880^\circ\text{C}$
(b) $T_B = 1\,000^\circ\text{C}$

Fig. 11. Microstructures of S20C/Ti joints.

X-ray diffraction patterns from the fractured surfaces of a joint bonded at 1 000°C are shown in Fig. 12. As shown in Fig. 12, diffraction lines from TiC, TiFe₂ and TiFe were observed on the S20C side, and diffraction lines from TiC and TiFe on the Ti side. These results, combined with those of the observation of microstructure, suggest that TiFe and TiFe₂ were formed at the place having region 'B' and 'D' similar to those of the ULC/Ti joint and TiC was formed at the place without region 'B' or 'D'. In fact, the distributions of Fe, Ti and C analyzed with EPMA were almost the same as those of FCD/Ti joint shown in Fig. 5 at the place where TiC was formed, and almost the same as those of the ULC/Ti joint shown in Fig. 9 at the place where intermetallic compounds TiFe and TiFe₂ were formed. That is, at the place where the TiC was formed, the interdiffusion of Ti and Fe was much suppressed as compared with the place where TiFe and TiFe₂ were formed. This result also supports that the TiC layer acts as a barrier against the interdiffusion of Ti and Fe. At bonding temperatures below 900°C, X-ray diffraction lines observed were only those from TiC except for α -Fe and α -Ti.

The relation between tensile strength of joints and bonding temperature is shown in Fig. 13. The tensile

Fig. 12. X-ray diffraction patterns from fractured surfaces of a S20C/Ti joint bonded at 1 000°C.

Fig. 13. Tensile strength of S20C/Ti joints as a function of bonding temperature.

strength of joints increased with temperature below 870°C similarly to that of FCD/Ti joints, but above 870°C decreased with the rise in bonding temperature. That is, the tensile strength of the S20C/Ti joint decreased with the rise in bonding temperature when the interlayer of TiC and the interlayer of TiFe and TiFe₂ coexisted at the bond interface. This fact suggests that the TiFe and TiFe₂ interlayer coexisting with the TiC interlayer had more detrimental effect on the joint strength than that in the ULC/Ti joint.

4. Discussion

As described above, the microstructure of the diffusion-bonded joint of iron-base alloys to titanium is influenced strongly by the carbon content; i.e., only the TiC layer was formed in the FCD/Ti joint, the intermetallic compounds layer of TiFe and TiFe₂ was formed in the ULC/Ti joint, and both the TiC layer and the intermetallic compound layer were formed in the S20C/Ti joint. In order to discuss the formation of such compounds, the standard free energy changes due to the formation of TiC, TiFe and TiFe₂ are shown in Fig. 14.^{16,17)} The standard free energy change due to the formation of TiC was lowest of all the compounds of TiFe, TiFe₂ and TiC. Therefore, TiC is preferentially formed at the bond interface of the FCD/Ti joint, in which the amount of C enough to form TiC is supplied from the FCD base metal. As mentioned in Sec. 3.1, the TiC layer acted as a barrier against the interdiffusion of Ti and Fe across the bond interface, and made it difficult to form the intermetallic compounds of Ti-Fe binary system. For this reason, the intermetallic compound layer of Ti-Fe binary system or β -Ti layer was not observed in the FCD/Ti joint.

While TiC has a simple crystal structure of NaCl (A15) type,¹⁹⁾ TiFe₂ and TiFe have rather complicated ones; TiFe₂ is MgZn₂ (C14) and TiFe is BCC (A2) or CsCl (B2). Especially, TiFe₂ is known as the Laves phase, which is very hard and brittle.¹⁹⁾ In view of the crystal structures and mechanical prop-

Fig. 14. Standard free energy change, ΔG° of TiC, TiFe₂ and TiFe as a function of temperature.

erties of these phases, it seems likely that TiC is not so brittle as TiFe or TiFe₂. For this reason, the tensile strength of the FCD/Ti joint, in which only the TiC layer was formed, was higher than that of the ULC/Ti joint and the S20C/Ti joint.

It is said that the interlayer consisting of a brittle phase has detrimental effects on the mechanical properties of the joint when its thickness exceeds a critical value.¹⁹⁾ For instance, the strength of a joint between iron and titanium decreases abruptly if the thickness of the layer exceeds 3–5 μm ,¹⁹⁾ and the thickness for joints between iron and titanium is 4–6 μm .¹⁹⁾ The strength of the steel wire/aluminium composite decreases rapidly with the increase in the thickness of layer more than 6 μm .²⁰⁾ In the present investigation, the strength of FCD/Ti joints was increased with bonding temperature and the thickness of the TiC layer in these joints, increasing with bonding temperature, was less than 3 μm . Therefore, the critical thickness of TiC layer in FCD/Ti joints may be regarded as more than 3 μm .

As shown in Figs. 7, 10, 11 and 13, remarkable changes in both tensile strength and microstructure of ULC/Ti and S20C/Ti joints were observed at the bonding temperature of about 900°C. This bonding temperature is approximately equal to the temperature T_{tr} of $\alpha \rightleftharpoons \beta$ transformation of Ti (883°C), suggesting that the transformation has a great influence on the formation and growth of intermetallic compounds at the bond interface. That is, for ULC/Ti and S20C/Ti joints, the thickness of TiFe and TiFe₂ layers was increased rapidly with bonding temperature above T_{tr} , while they could not be observed below T_{tr} . Owing to this increase in the thickness of TiFe and TiFe₂ layers, the strength of ULC/Ti joints was not increased with the rise in bonding temperature above T_{tr} , and that of S20C/Ti joints was de-

creased.

5. Conclusions

Spheroidal graphite cast iron (FCD), low carbon steel with very low carbon content (ULC) and mild steel (S20C) have been diffusion-bonded to titanium, and the effects of carbon content on the microstructure and tensile strength of joint have been systematically investigated. Results obtained are summarized as follows:

(1) In the FCD/Ti joint, an interlayer of TiC was formed at the bond interface. The intermetallic compounds TiFe or TiFe₂, however, were not formed, since TiC was thermodynamically more stable than the intermetallic compounds and the interlayer of TiC acted as a barrier against the interdiffusion of Fe and Ti across the bond interface. The joint strength increased with bonding temperature; *i.e.*, no decrease in the joint strength due to the growth of the TiC interlayer was observed.

(2) In the ULC/Ti joint, an intermetallic compound layer consisting of TiFe₂ and TiFe was formed at the bond interface, while TiC could not be found. The bond strength increased with bonding temperature up to 900°C, but became almost constant above 900°C. This result suggests that the growth of the interlayer consisting of TiFe and TiFe₂ suppressed the increase in the joint strength at bonding temperatures above 900°C.

(3) At the bond interface of the S20C/Ti joint, a discontinuous interlayer of TiC was formed at bonding temperatures below 900°C. At bonding temperatures above 900°C, the bond interface consisted of the places where the interlayer of TiC was formed and where an interlayer of TiFe and TiFe₂ was formed. The tensile strength of the S20C/Ti joint increased with bonding temperature below 900°C, but above 900°C decreased with the rise in bonding temperature. These results suggest that the TiFe and TiFe₂ interlayer coexisting with the TiC interlayer had more detrimental effects on the bond strength than those

formed individually.

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