General paper

Solid State Diffusion Bonding of Superplastic Duplex Stainless Steel with Carbon Steel

Iswadi JAUHARI*, Hiroyuki OGIYAMA * and Hitoshi TSUKUDA*

* Mechanical Engineering Department, Ehime University, Matsuyama City
Bunkyocho 3 Ban790-8577 Ehime, Japan

Abstract: Solid state bonding between superplastic duplex stainless steel with carbon steel under low pressures is studied for the objectives to understand bonding behaviors and to determine mechanisms controlling bonding through the activation energy analysis. Experiments were carried out by combining several bonding parameters such as bonding temperatures, bonding pressures and surface roughness. Bond quality was evaluated by its tensile strength. Bonding strength increased with temperature, time and pressure but decreased with surface roughness. Sound bonds comparable to that of the parent metal were obtained at considerably short bonding time and low pressure. The best bonding condition obtained in this study was 1373 K temperature, 3 MPa pressure, $0.32 \mu\text{m } R_{\text{max}}$ and 220 s bonding time, producing the parent metal strength with around 1 % of deformation ratio. Changes in microstructures and hardness were observed across the bonding interface, due to diffusion of atoms, mainly C, from the carbon steel side to the duplex stainless steel side. A hard carburized layer formed at the DSS side and a soft decarburized layer at the carbon steel side. From the value of activation energy and experimental data, bonding was controlled mainly by two mechanisms, which were the superplastic deformation at the early stage and the diffusion of carbon at the second stage.

Key words: Superplastic duplex stainless steel, Carbon steel, Diffusion bonding, Activation energy

1. INTRODUCTION

($\alpha + \gamma$) duplex stainless steel (DSS) contains ferrite and austenite phases with approximately equal composition. The alloy has good resistance to corrosion cracking and corrosion fatigue. At a certain range of temperature and strain rate, DSS with fine microstructures is also known for showing a large degree of elongation called superplasticity [1]. It was reported that the superplasticity could provide to the potential for solid state bonding in which the high ductility of superplastic materials might be used to accelerate the contact process on the bonding interface [2]. A Previous work on solid state bonding of the superplastic DSS to the same and different ferrous alloys showed that bonds failed at the parent metal could be achieved in a relatively short bonding time [3]. However, the pressure used to obtain such bonds was considerably high at 19.6 MPa, which would lead to a large deformation of the bonding couple.

In the current situation, solid-state bondings between stainless steels to other ferrous steels such as carbon steels are performed mostly by pressure or friction welding. However, again the pressures used to obtain such bonds are considerably high, and for design

emphasizing on accuracy such as near net shape design, the method might not be suitable.

For this reason, in this work solid state bonding between the superplastic DSS with carbon steel under low pressures is studied with the objectives to understand the bonding behaviors and to determine mechanisms controlling bonding through the activation energy analysis.

Experiments were carried out by combining several bonding parameters such as bonding temperatures, bonding pressures and surface roughness.

2. EXPERIMENTAL PROCEDURE

2.1. Materials Preparation

The materials used in the present study were a duplex stainless steel (JIS SUS329J1) and a carbon steel (S45C). Table 1 shows the chemical composition for both steels. In order to obtain superplastic microstructure, the as-received DSS was initially solution-treated at 1573 K for 1 hour followed by water quenching. It was then cold-rolled to a plate through a reduction area of 75 %. The thermo-mechanically treated alloy showed a superplastic elongation of 1050 % at a temperature of 1223 K

Table 1. Chemical composition of a duplex stainless steel and a carbon steel used (wt.%).

	C	Si	Mn	P	S	Ni	Cr	Mo	N
SUS329J1	0.02	0.36	0.08	0.027	0.007	5.43	23.8	1.23	0.03
S45C	0.43	0.20	0.67	0.019	0.014	0.20	0.14	—	—

and tensile strain rate of $1 \times 10^{-2} \text{ s}^{-1}$ [4] (Fig. 1). Bonding specimens were cut from the cold rolled plate by wire cutting with the bond interface normal to the cold rolled direction. In order to obtain different surface roughness, the samples were ground by various grades of emery paper and some were buffed, producing mirror-like surface. The roughness of the surfaces R_{max} was measured perpendicular to the grinding direction.

2.2. Bonding Procedure

Bonding experiments were carried out in a vacuum hot-press furnace chamber schematically shown in Fig. 2. As shown in Fig. 3, after evacuating to a pressure of 100 Pa, the bonding specimen was heated to the required temperature at a rate of 1 K/s, and after homogenizing for 6 minutes, bonding pressure was applied and maintained for a certain bonding time. The pressure was released at the end of bonding and the specimen was air-cooled to room temperature.

Fig.1. Undeformed (top) and 1050% superplastically deformed (bottom) specimens.

Fig.2. Schematic diagram of a diffusion bonding apparatus.

2.3. Bonding evaluation

The bond quality was evaluated by its tensile strength. The tensile test specimens were machined from the joints. Tensile tests were performed at room temperature at a strain rate of $3 \times 10^{-4} \text{ s}^{-1}$. Microhardness values were measured across the bonding interface. The microstructures and fracture surfaces were examined using an optical and charge-coupled device (CCD) camera.

3. RESULTS AND DISCUSSION

3.1. Effect of Bonding Parameters on Bonding Strength

In this study, it was identified that the tensile strength of the carbon steel experienced the thermal history at all bonding conditions was lower than that of DSS and did not vary with the temperatures. Thus, the absolute strength was used instead of the specific strength (i.e. a bonding strength/a parent metal strength, experiencing the same thermal history) in evaluating the bonding strength.

Figure 4 shows the relation between the bonding strength and time for three different temperatures. As can be seen from the figure, the bonding strength increases with an increase in temperature and time. It is understood that increasing temperature reduces the yield stress of the materials and activates the creep deformation and the diffusion of atoms. These would accelerate the bonding process and finally lead to an increase in the bonding strength.

The bonding strength also increases with increasing pressure, as shown Fig. 5. Besides what was mentioned earlier, increasing pressure helps to accelerate the bonding especially through the plastic deformation of surface asperities. However, as the pressure increased, the parent metals, particularly the DSS would easily be deformed as it has the superplastic characteristics. Figure 6 shows the relation between the deformation ratio of DSS parent metal and the pressuring time for three

Fig.3. Illustration of experimental procedure: temperature change with time.

different pressures. The deformation ratio, D_r , is defined by

$$D_r = [(l_s - l_f) / l_s] \times 100 \%, \quad (1)$$

where l_s and l_f are respectively the material length before and after pressured. From the Fig., 9 MPa pressure shows a considerably large deformation ratio compared to 3 and 6 MPa pressures even from the start of pressuring due to the superplastic deformation. Earlier works show that for the superplastic DSS deformed at ~ 1373 K and initial strain rate of $\sim 10^{-4} \text{ s}^{-1}$, a peak stress of approximately 9 MPa was obtained [1,5].

From this result, although increasing the pressure could improve the bonding strength, it would consequently lead to a large deformation in the bonding material and this is regarded as undesirable from a practical point of view.

The relation between the bonding strength and the surface roughness is shown in Fig. 7. At R_{max} of 0.02 and $0.32 \mu\text{m}$, the bonding strength comparable to that of the parent metal was obtained, but macroscopically failed at the bonding interface (this would be discussed in more detail later). As the surface roughness increased, the bonding strength decreased. The results agree to the fact obtained in earlier work [6] that increasing the surface roughness would increase the volume fraction of voids in

bonding interface and finally decreasing the bonding strength. It also shows that in a low-pressure condition such as in this study, the surface roughness has a significant effect in the bonding process. This is because bonds that were conducted at a higher pressure showed almost no surface roughness effect [3].

Figure 8 shows the hardness distribution across the bonding interface for two different types of bonds, the superplastic DSS to the carbon steel and the as-received DSS to the carbon steel. For both bonds, the bonding conditions were exactly the same except for their surface

Fig.6. Relationship between the deformation ratio and the pressuring time at different pressures.

Fig.4. Relationship between the bonding strength and time at different temperatures.

Fig.7. Relationship between the bonding strength and the surface roughness.

Fig.5. Relationship between the bonding strength and pressure.

Fig.8. Hardness distribution near the bonding interface for the superplastic DSS/S45C and nonsuperplastic DSS/S45C couples.

roughness ($0.32 \mu\text{m } R_{\text{max}}$ for the former and $1.5 \mu\text{m } R_{\text{max}}$ for the latter). The bonding condition for the former bond is the optimum condition obtained in this study where the bond with the strength comparable to that of the parent metal was obtained at relatively shorter time than the one mentioned earlier (see Fig. 7). The latter bond shows the bonding strength of 344 MPa. From Fig. 8, a change in the hardness distribution can be seen across the bonding interface. The hardness increased sharply at the DSS side while decreased at the carbon steel side. These are mainly due to the diffusion of C atoms from the carbon steel side to DSS because the carbon steel has a higher content of carbon, although it is also expected that Cr and Ni which are the main alloying elements of DSS also would diffuse to the carbon steel side [7]. As a result, a hard carburized layer formed at the DSS side and a soft decarburized layer at the carbon steel side. It is interesting to note that for both bonds, despite the large difference obtained in the bonding strength, the hardness distributions across the bonding interfaces showed almost the similar result.

Therefore, it is important to avoid evaluating the bonding strength directly from the interface's hardness.

The other thing that should be taken into account is the existence of the weak decarburized layer. Figure 9 shows the cross sections and fracture surfaces of the failed bonding specimens (superplastic DSS/carbon steel bonds) after tensile tests for the weak bond and the bond having the parent metal strength. For the weak bond, the cross section was straight while the fracture surface was completely flat and smooth. These are indicative of poor bonding, failed at the bonding interface. For the bond with the parent metal strength, the surface irregularities along the cross section, suggest that the failure's path did not completely pass through the bonding interface. As for the fracture surface, it was totally different from that of the weak bond one, which showed a typical brittle fracture surface throughout the area and also the ductile features at the edges of the surface with no sign of poor bond.

Figure 10 shows the microstructures in the vicinity of the bonding interfaces for the superplastic DSS to the

(a) Weak bond failed at 483 MPa (bonding condition: 1373K, 220 s, $1.5 \mu\text{m } R_{\text{max}}$, 3 MPa).

(b) Bond having the parent metal strength (bonding condition: 1373 K, 600 s, $0.32 \mu\text{m } R_{\text{max}}$, 3 MPa).

Fig.9. Tensile fracture surface of superplastic DSS/S45C couples.

(a) Bonding interface of the superplastic DSS/S45C couple ($0.32 \mu\text{m } R_{\text{max}}$).

(b) Bonding interface of as-received DSS/S45C couple ($1.5 \mu\text{m } R_{\text{max}}$).

Fig.10. Microstructures near the bonding interfaces; Arrows indicate the bonding interface (bonding condition: 1373 K, 220 s, 3 MPa).

carbon steel and the as-received DSS to the carbon steel bonds. From the figure, the decarburized carbon steels and carburized DSSs can be seen. The measured widths for carburized and decarburized layers were almost similar for both bonds, which were in good agreement with the hardness test results.

From these results, it can be understood that for bonds having the parent metal strength, failures actually did not occur at the bonding interface, but took place at the weak decarburized layers near the interface. However, since the size of the layer was extremely small, macroscopically, it seems that the failure occurred at the interface.

3.2. Bonding Mechanism

From Fig. 4, two obvious stages of bonding are observed based on the slopes of the curves. The first stages show a great increase in strength with time, but the rate decrease markedly in the second stage. Generally, the diffusion bonding process starts with the plastic deformation of surfaces asperities and followed mainly by the diffusion of atoms. Thus, from these results, it can be concluded that the first stage is due to the plastic deformation of surface asperities while the second stage is controlled by the diffusion of atoms. As stated in the previous work [6], the activation energy of bonding process can be determined by analyzing the increment rate data of bonding strength k , or in another word the slopes of the curves in Fig. 1, as a function of reciprocal absolute temperature ($1/T$). This is shown in Fig. 11 as an Arrhenius plot, where the activation energy is obtained from the slope of the plot. The rate constant k is defined by

$$k = d\sigma/dt, \quad (2)$$

where σ and t are the bonding strength and time, respectively. The calculated activation value was 248 kJ/mol for the first stage. However this method cannot be applied for determining the activation energy for the second stage because the rate constants do not show clear increment with the increasing of temperature. The reason for this will be further discussed later.

Generally the activation energy values for the superplastic deformation for a wide range of materials show them to be comparable with that for grain boundary diffusion although there are a few exceptions, where it is comparable with that for lattice diffusion [8]. In this study, although it was not possible to compare directly the activation energy obtained with that of lattice and grain boundary diffusion for DSS used, it is found that the activation energy is close to that of lattice diffusion of most main alloying elements of 18Cr-Ni stainless steel (240~280kJ/mol) [9]. From these results, it appears that the bonding process of the first stage is related to the superplastic deformation from their activation energy values. The similarity in the activation energy values suggests that the processes are controlled by the same mechanism. Thus, it can be concluded that the superplastic deformation process controls the early stage of the bonding process.

Furthermore, it is found that the activation energy for the first stage is much lower compared to that of plastic flow of 18Cr-Ni stainless steel and high alloy steel (~370 kJ/mol) [10,11]. A comparison with that of the first stage bonding process for the superplastic DSS with similar alloy [12] indicates that the activation energy has increased slightly from 204 kJ/mol to 248 kJ/mol, which is due to the fact that the superplastic DSS was bonded with the non-superplastic carbon steel.

Figure 12 shows the relationship between the bonding strength and time for the superplastic DSS to the carbon steel and the as-received DSS to the carbon steel bonds. It is clear from the figure that a large difference in the strength between the two bonds is observed at the early stage of bonding (220 s). However, with increasing bonding time, the bonding strength increment rates were very much alike.

From these results, it is suggested that by the using of superplastic material, the bonding was accelerated in the early stage but in the following stage it seems that the superplastic material influence was negligible. In addition, from the results of Fig. 4 and Fig. 12, it is suggested that in the second stage, bondings were

Fig.11. Arrhenius plot of the rate constant for the 1st stage bonding (superplastic DSS/ S45C couple).

Fig.12. Relationship between the bonding strength and time for the superplastic DSS/S45C and nonsuperplastic DSS/ S45C couples.

controlled mainly by the diffusions of C. This is because the activation energy values of carbon diffused in BCC and FCC iron are 84 kJ/mol and 142 kJ/mol respectively [13], which are relatively low compared to that for the diffusion of most main alloying elements of 18Cr-Ni stainless steel mentioned earlier. Low activation energy means that the diffusion rates do not vary so much with the variation of temperatures. This is probably the reason why the rate constants k for the second stage do not show clear increment with the increasing of temperature.

Finally, from the activation energy value and the experimental data obtained, it can be concluded that the superplastic deformation process controls the early stage of the bonding process, while in the second stage it is mainly controlled by the diffusion of C.

4. CONCLUSIONS

In this study superplastic DSS was used for the solid state diffusion bonding with the carbon steel. The bonding behaviors and mechanisms controlling bonding were studied. As a result, the following conclusions were obtained.

- (1) By using the superplastic DSS, bonds having the parent metal strength could be obtained easily and bondings were strongly influenced by the temperature, pressure, time and surface roughness.
- (2) The best bonding condition obtained in this study was 1373 K temperature, 3 MPa pressure, $0.32 \mu\text{m } R_{\text{max}}$ and 220 s bonding time, producing the parent metal strength with around 1 % of deformation ratio.
- (3) The mechanisms controlling bonding can be divided into two stages analyzing from the strength increment

rate data.

- (4) From the activation energy value and experimental data, it can be concluded that the first stage was controlled by the superplastic deformation while the second stage mainly by the diffusion of C.

REFERENCES

1. Y. Maehara and Y. Ohmori, *Met. Trans. A*, **18** (1987) 663.
2. Y. Komizo, Y. Maehara, and T. G. Langdon, *Mater. Sci. Tech.*, **4** (1988) 669.
3. Y. Komizo and Y. Maehara, *Trans J. Weld. Soc.*, **19** (1988) 83 (in Japanese).
4. H. Ogiyama, T. Shiraishi, T. Tsukuda and I. Jauhari, *J. Soc. Mater. Sci. Jpn.*, **50** (2001) 949 (in Japanese).
5. J. A. Jaminez, G. Frommeyer, M. Carsi and O. A. Ruano, *Mater. Sci. Eng.*, **A307** (2001) 134.
6. O. Ohashi and T. Hashimoto, *Trans J. Weld. Soc.*, **6** (1976) 71 (in Japanese).
7. I. Gutierrez, J. J. Urcola, J. M. Bilbao and L. M. Villar, *Mater. Sci. Technol.*, **7** (1991) 761.
8. K. A. Padmanabhan and G. J. Davies, *Superplasticity, Materials Research and Engineering*, Springer-Verlag, New York, **2** (1980) 72.
9. R. V. Patil, G. P. Tiwari and B. D. Sharma, *Met. Sci.*, **14** (1980) 525.
10. I. Tamura, C. Ouchi, T. Tanaka and H. Sekine, *Thermomechanical processing of high strength and low alloy steels*, London, Butterworth (1980).
11. D. J. Towel and T. Gladman, *Met. Sci.*, **13** (1979) 246.
12. I. Jauhari, H. Ogiyama and T. Tsukuda, *J. Jpn. Soc. Str. Frac. Mater.*, (2002) (submitted) (in Japanese).
13. J. P. Schaffer, A. Saxena, S. D. Antolovich, T. H. Sanders Jr, S. B. Warner, *The Science and Design of Engineering Materials*, McGraw-Hill, Singapore (1999).