Identification of predominant interfacial contact mechanisms in solid-state bonding of superplastic high carbon steel

Zhonghao Heng¹, Masakatsu Maeda² and Yasuo Takahashi²

¹1. Graduate School of Engineering, Osaka University, 2-1 Yamadaoka, Suita, Osaka 565-0871, Japan
²2. Joining and Welding Research Institute, Osaka University, Japan

E-mail: takasy@jwri.osaka-u.ac.jp

Abstract. Interfacial contact mechanisms and the superplastic behaviour of fine-grained high carbon steel during solid-state bonding were investigated. According to the results, typical superplastic deformation occurred at the bonding interface under conditions of superplasticity-causing temperature (\(T = 1003\)–\(1053\)K), bonding pressure (\(P = 34\)–\(44.3\)MPa), and bonding area ratio (\(S = 30\)–\(50\)%). The stress exponent, \(n\), was about 2.25 while the activation energy, \(Q\), was about 189 kJmol\(^{-1}\), both indicating and confirming superplastic solid-state bonding. Predominant bonding mechanism controlled by superplastic deformation was experimentally identified.

1. Introduction

Superplastic materials exhibit high deformability and strong diffusion properties under appropriate conditions of temperature and pressure [1]. One of the important properties of fine-grained superplastic materials is that they can often be readily bonded in the solid state, and this is particularly true for metals with poor weldability [2]. For instance, concurrent superplastic forming and diffusion bonding (SPF/DB) has been recognized as a viable and highly economical manufacturing technology in the aviation industry.

Current researches on solid-state bonding of superplastic materials primarily focus on the optimization of diffusion bonding parameters [3-5]. Although there is no doubt that superplastic flow can facilitate the early stage of bonding process, the predominant bonding mechanisms, including the effect of superplasticity on enhancement of interfacial contact, are not fully understood yet. The role of superplastic deformation therefore should be clarified to maximize the benefits of its application.

Previous experimental researches on conventional solid-state bonding have elucidated fundamental mechanisms of void shrinkage at the bond interfaces [6-8]. In the present study, the predominant bonding mechanism of interfacial contact process was investigated on the basis of the model proposed by Takahashi [6]. The effect of superplastic flow during solid-state bonding was also studied. This research is of great value in clarifying the predominant bonding mechanisms during different bonding stages and in optimizing the bonding parameters of superplastic materials.

2. Identification method

A new method is required to identify the predominant interfacial contact mechanisms in superplastic materials. Referring to an existing theoretical model [6], Figure 1(a) schematically illustrates the solid-
state bonding test of superplastic and non-superplastic metals. The bonding surface of the non-superplastic hard metal is perfectly flat while that of superplastic metal is machined to concentric circular ridges (interval of $2L_0$). The voids shrinkage $V_s (= V_2 - V_1)$ in the period of $t_s (= t_2 - t_1)$ is shown in Figure 1(b). The geometrical parameters $X, w,$ and $h$ in Figure 1 are, respectively, half of the bonded length of a unit, half the void width, and the void height. The subscript $0$ indicates initial value of the respective parameters.

![Figure 1](image.png)

**Figure 1.** Schematic illustrations for superplastic solid-state bonding. (a) voids at the bonding interface. (b) interfacial contact process.

For solid-state bonding, the interface self-diffusion and volume self-diffusion are predominant when the bonding pressure, $P$, is low enough. On the other hand, high-temperature deformation becomes more important as $P$ increases [9]. In regard to the bonding conditions used in the present study, the core discussion is focused on the interfacial deformation, that is, interfacial contact controlled by diffusion has been excluded. As the interfacial contact processes occur under conditions of high vacuum and high temperature, effect of surface oxide film were ignored. Interfacial deformation mainly occurs on the side of the superplastic metal under identical bonding conditions, that is, the plastic deformation of the non-superplastic metal need not be considered. Initial contact is made through instantaneous plastic deformation in the early stage of bonding process, in general, when the bonding ratio is less than 20%. In the present study, the set value $X_1$ is greater than the critical value $X_c$ given by the instantaneous plastic flow, thus, the contribution of instantaneous plastic deformation to the interfacial contact can be ruled out.

Referring to the identification model of void shrinkage due to creep proposed by Takahashi [6], the relation between bonded area growth $\Delta X$ and $t_s$ is expressed as

$$\Delta X = \frac{A_c}{kT} \left( \frac{P}{G} \right)^n \exp \left( -\frac{Q}{RT} \right) M \{ F(L, X, n) \} t_s, \tag{1}$$

where $A_c$ is a constant when the bonding temperature $T$ is kept constant, $P$ is the bonding pressure, $G$ is the shear modulus, $n$ is the stress exponent, $F(L, X, n)$ is a geometrical function of $L, X,$ and $n, Q$ is the activation energy for creep. $\Delta V$ is roughly proportional to $\Delta X$. If $\Delta X$ is kept constant, $M \{ F(L, X, n) \}$ also becomes constant, and the following relations are expressed as

$$t_s \propto \left( \frac{1}{P} \right)^n \tag{2}$$

when $P$ is constant, and

$$\frac{T}{t_s} G^{n-1} \propto \exp \left( -\frac{Q}{RT} \right) \tag{3}$$
when $T$ is designated properly and kept constant. So, the predominant mechanism during solid-state bonding can be identified by the gradients of $\log t_s$ vs $\log P$ and $\log (T/t_s)$ vs $(1/T)$ plots, that is, the gradients are the stress exponent, $n$, and the activation energy, $Q$, respectively. The $n$ value is about 4–6 when power law creep is predominant, whereas the $n$ value is 2–3 and the $Q$ value is close to the interface self-diffusion activation energy $Q_b$ when the superplastic flow is predominant during solid-state bonding [2, 6].

3. Experimental

The material used in the present study was a high carbon steel, SK105. The chemical components and critical points ($Ac_1$: eutectoid transformation temperature, $Acm$: temperature of proeutectoid cementite precipitation) are shown in Table 1.

The original steel with its coarse pearlite structure (grain size > 10μm) did not exhibit superplasticity. The superplastic steel with fine-tempered martensite (grain size: 5~8μm) was obtained after an ultra fine heat treatment: circular quenching from 1053K thrice, followed by tempering at 473K [10]. Microstructures of both steels are shown in Figure 2.

| Steel  | C  | Mn  | Si  | Cr  | Ni  | $Ac_1$ | $Ac_3$ | $Acm$ |
|--------|----|-----|-----|-----|-----|--------|--------|-------|
| SK105  | 0.92 | 0.64 | 0.28 | 0.18 | 0.09 | 1003   | 1073   |       |

**Figure 2.** Microstructure of SK105. (a) pearlite in original steel. (b) tempered martensite in superplastic steel.

**Figure 3.** Bonding surface of superplastic steel.
The bonding surface of superplastic steel with concentric circular ridges was lathe-machined. As shown in Figure 3, the height was measured to be about 150 μm, and half the distance \( L_0 \) between the ridges was about 80μm. Bonding specimens were cylindrical in shaped of dimensions Φ10mm×12mm.

The bonding tests of the superplastic and original steel were carried out under several conditions for estimating the time \( t_s \) required to attain a certain increment \( \Delta S (= \Delta X/L_0) \) in percentage bonded area. The vacuum was 10\(^{-4}\) Pa and the bonding temperature was in the range of SK105 superplastic temperatures (1003~1053K) [11]. The range of bonding pressure was 25~60MPa. The pressure was applied in the form of a uniaxial compressive stress. The specimens were heated using a molybdenum heater and cooled after the bonding pressure was removed.

Choosing an appropriate \( \Delta S \) is necessary for identifying the predominant mechanisms in the middle stage of solid-state bonding. The change in \( \Delta S \) during cooling was ignored. The estimation of bonded area was inaccurate if the set value of \( \Delta S \) was less than 10%. Therefore, \( \Delta S = 20\% \) was adopted in the present study. Also, the bonded area percentage \( S_1 \) was set at 30% to rule out the influence of instantaneous plastic deformation. The bonded area increment was estimated by the scanning electron microscope (SEM) observation of the fractured surface after bonding. It was difficult to get the time \( t_s \) directly, therefore, \( t_s \) was estimated by interpolating the data of several bonding tests. Three repetitions of bonding tests under each condition were carried out and the mean value obtained were used to ensure the accuracy of \( \Delta S \) estimation.

4. Results and discussion

The joints of superplastic and original steel were broken in liquid nitrogen, so, the interfacial deformation that occurred during breaking was ignored. Figure 4 shows the bonding process and surface fractures at the bonded interface in different stages. The shrinkage of voids is relatively uniform. The rugged and bright areas are the bonded zones; the dark areas are the void surfaces. When the bonding time is 0 s, the initial state, \( S_0 \), is 0%. \( S_1 = 20\% \) is set as the stage 1, for a bonding time of \( t_1 \). When the bonding time \( t_2 = t_1 + t_s \), the bonded area ratio increment \( \Delta S \) is set to 20% at this stage.

The bonding pressure dependence of \( t_s \) at 1023K is shown in Figure 5. From equation (2), the stress exponent \( n \) value is calculated to be approximately 5.74 at low bonding pressure (\( P < 30\text{MPa} \)), therefore, the power law creep is the predominant mechanism. This result is also consistent with the value of \( n=6 \) provided by Takahashi [6]. The \( n \) value decreases gradually as the pressure increases, as a result of a change of the predominant mechanism of interfacial contact. The \( n \) value is about 2.3 for the bonding pressure \( P = 37\text{~}43\text{MPa} \). The strain rate sensitivity index, \( m \) (the reciprocal of \( n \) value), is

![Figure 4. SEM observations of fractured surfaces in different stages.](image)
about 0.43. This suggests that superplastic deformation predominantly contributes to the interfacial contact process. In other words, superplastic flow-based deformation occurs at the bonding interface at this stage. The \( n \) value increases with the further increase in bonding pressure and is larger than 3 when \( P > 43 \text{MPa} \), illustrating the change in the predominant bonding mechanism. At such pressure, the creep mechanism becomes increasingly influential and the effect of superplastic flow is weakened significantly. The predominant mechanism thus in effect comes back to exhibit power law creep.

If the \( n \) value lies between the values associated with these two fundamental mechanisms, a combined effect of the two mechanisms can be expected to occur. By extending the characteristic curves of the different mechanisms, the intersection point is obtained. This point can be considered as a transition point of these two fundamental mechanisms. Thus, the bonding pressure range can be divided into three regions: \( P = 34 \sim 44.3 \text{MPa} \) when the superplastic flow is predominant; \( P > 44.3 \text{MPa} \) or \( P < 34 \text{MPa} \) when the power law creep is predominant. The predominant mechanisms during the bonding process can be identified thus.

This result, \( P = 34 \sim 44.3 \text{MPa} \), also correlates well to the superplastic flow stress range of SK105 (35\sim45\text{MPa}) obtained from uniaxial compression tests [10]. It should be noted that the pressure applied to the bonding interface is larger than that of the base metal. However the compression deformation at the bonding interface is different from that of uniaxial compression, as the interface tangential friction can have an impact on the interfacial contact process. This agreement of pressure is for reference only. The contribution of stress distribution to interfacial deformation needs further analysis.

Figure 6 shows the temperature dependence of \( T/t_s \) under 43MPa. The \( Q \) value, measured by the log \( (T/t_s) \) vs \( (1/T) \) plot, is about 190 kJmol\(^{-1}\) at \( T = 1003\sim1023\text{K} \) and 200 kJmol\(^{-1}\) at \( T = 1023\sim1053\text{K} \). The \( Q \) value increases slightly with increasing \( T \), and that is due to the change in superplasticity at different temperatures. Fine-grained SK105 steel exhibits superplastic behavior when \( T \) is in the proximity of the critical temperature \( A_{C1} \). Its microstructure is comprised of austenite, \( \gamma \), and cementite phases when \( T \) is slightly higher than \( A_{C1} \). This dual-phase fine-grained steel shows superplasticity well. With increasing \( T \), more carbides are dissolved, and the limiting effect of second phases (fine carbide) on grain growth is weakened, resulting in concomitantly reduced stability of grain size. Consequently, superplasticity relatively weakens while the \( Q \) value increases slightly.

The \( Q \) value during solid-state bonding (\( P = 43 \text{MPa}, T = 1003\sim1053\text{K} \)) is between the activation

![Figure 5. Bonding pressure dependence of \( t_s \).](image)
energy of $\gamma$-Fe interfacial self-diffusion ($Q_b = 159$ kJmol$^{-1}$) and that of volume self diffusion ($Q_v = 270$ kJmol$^{-1}$) [12]. Additionally, its value is comparatively closer to $Q_b$. This is reliable evidence of the predominance of superplastic deformation, a grain boundary phenomenon, as the interfacial contact process. The most commonly considered mechanisms for superplastic flow involve grain boundary sliding (GBS) and an accommodation process accompanying GBS [3, 13, 14]. Therefore, the activation energy for superplastic deformation should be equal to that of grain boundary diffusion. This fact is well represented by the results of the present study.

**Figure 6.** Temperature dependence of $T/t_s$.

5. Conclusions

It was found that superplastic deformation takes a dominant role in the interfacial contact process from a bonding area ratio of 30% to 50%, under the conditions of bonding temperature $T = 1003$–$1053$K and bonding pressure $P = 34$–$44.3$MPa. The predominant mechanism controlled by superplastic flow can be identified using the value of $n$ and $Q$. Superplastic deformation and its dominant effect are confirmed experimentally. Superplastic bonding was produced under conditions of $T = 1003$–$1053$K and $P = 34$–$44.3$MPa.

References

[1] Langdon TG 1991 *ICSAM-1991*. 3
[2] Nieh TG 1997 *Superplasticity in metals and ceramics* (New York: Cambridge University Press)
[3] Maehara Y, Langdon TG 1990 *Mater. Sci. Eng.* **128** 1
[4] Huang Y, Ridley N, Humphreys FJ and Cui JZ 1999 *Mater. Sci. Eng.* **266** 295
[5] Somekawa H, Hosokawa H, Watanabe H and Higashi K 2003 *Mater. Sci. Eng.* **339** 328
[6] Takahashi Y, Inoue K and Nishiguchi K 1993 *Acta metall.* **41** 3077
[7] Takahashi Y and Inoue K 1992 *Mater. Sci. Tech.* **8** 953
[8] Hill A and Wallach R 1989 *Acta metal.* **37** 2425
[9] Takahashi Y 2003 *Ceramic trans.* **138** 29
[10] Zhang KK, Yang YL, Liu SZ, Han CX and Xu D 2004 *Acta metall sinica.* **16** 538
[11] Zhang KK, Wang YL, Shi HX, Yu H and Liu S 2009 *Mater. Sci. Eng.* **499** 97
[12] James DW and Leak GM 1965 *Phil. Mag.* **12** 491
[13] Gifkins RC 1976 *Metall Trans.* **7** 1225
[14] Langdon TG 1970 *Phil. Mag.* **78** 689