Experimental Study on Diffusion Bonding in Pure Magnesium

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Applicability of diffusion bonding was examined on pure magnesium rolled sheet with an initial grain size of 85 μm . The diffusion bonding tests were carried out in air at the pressure range from 2 to 20 MPa and at the temperature range from 573 to 673 K and for the times up to 72 h. Before diffusion bonding, the surfaces to be bonded were blasted to make rugged. The post-bonded mechanical properties were estimated by the compression lap shear test, and the microstructure after bonding were also observed by the microscopy in order to achieve the optimal process for diffusion bonding. The maximum of lap shear strength was 0.888 at a bonding pressure of 20 MPa and a bonding temperature of 673 K, with a bonding time of 1 hour. In high ratio of lap shear strengths, the bond line was not identified by optical microscopy, and surface fracture after compression lap shear test was also fully ductile failure. The present results revealed that there is a possibility to fabricate magnesium products using diffusion bonding technique.

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1. Introduction

Recently, the research activities of magnesium become higher in order to reduce the weight of components such as motor vehicles from the economical and ecological point of view. There are many potential opportunities for the use of magnesium in motor vehicle components. This is not only a result of magnesium's relatively low density, which can directly and substantially reduce vehicle weight, but is also a result of its good damping characteristics, dimensional stability, machinability and low casting cost. These attributes help magnesium to economically replace many zinc and aluminum die-castings, as well as cast iron and steel components.¹⁾

Despite these advantages, magnesium normally exhibits limited ductility because of hcp structure. Therefore, recent fabrications of magnesium-based materials were mainly performed by die casting with hot- or cold-chamber, or semi-solid processing of thixotropic molding. Recently, the combination of cast and wrought materials is also expected to fabricate structural components. For structural components in automobiles, the material should exhibit sufficient ductility as well as specific strength. Because wrought magnesium alloys generally have higher strength and ductility, there is a strong need to use wrought materials. In such a case, joining between cast and wrought materials will be indispensable technology.²⁻⁵⁾

Diffusion bonding is one of the joining techniques and it is a solid state joining process in which two clean metallic surfaces are brought into contact at elevated temperature, $< 0.7T_m$, under a low pressure, where T_m is the absolute melting points of materials. Diffusion bonding process also permits the production of high quality joints with a little need for post-weld machining.^{6,7)}

There are several advantages in using solid state bonding. Since the bonding is performed without a fusion process, the mechanical properties deteriorate very little in the welding

zone as a result of non-microstructural changes. And diffusion bonded materials have high dimensional stability even after bonding. It is also possible to bond dissimilar materials such as non-metallic materials, refractory materials, or composite materials.⁸⁻¹⁰⁾

It is important to consider the optimal conditions for diffusion bonding in order to achieve high quality joints: temperature, pressure, time and so on. It is also necessary to obtain an extremely close contact throughout the bonding interface. When the two bonded surfaces are brought into contact, diffusion bonding occurs primarily as a result of time-dependent plastic flow and diffusion of material into the interfacial voids created. Given sufficient time, the voids will disappear completely across the original surfaces.^{11,12)}

It has been reported that many materials can be successfully diffusion bonded *e.g.* titanium alloys, aluminum alloys and steels.¹²⁻²³⁾ For example, titanium alloys have come to be widely used, especially, in the aerospace industry.¹²⁻¹⁶⁾ Aluminum alloys and steels have already obtained the optimal conditions to produce high quality in diffusion bonding.¹⁷⁻²²⁾ The summary of diffusion bonding conditions between the above materials together with the fracture strengths is shown in Table 1. However, the possibility of diffusion bonding using magnesium-based materials has hardly been investigated.²⁴⁾ Therefore, in the present study, diffusion bonding tests for pure magnesium were carried out in the wide range of bonding temperature, pressure and time to investigate its optimal conditions.

2. Experimental Procedures

Pure magnesium was used in order to investigate the diffusion bonding conditions. The chemical composition of the material is given in Table 2. Pure magnesium of 99.95% purity was supplied by Osaka Fuji Corp. as a rolled sheet with a thickness of 2.3 mm. The microstructures of the specimens were examined by optical microscopy. The average grain size, d , was estimated using the equation, $d = 1.74L$; L is

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Table 1 Summary of the conditions for diffusion bonding between Al alloys and Ti alloys together with the fracture strengths of the bonds.

Material	Surface finish	Temperature, T/K	Pressure, P/MPa	Time, t/h	Strength, σ/MPa	Ref.
7475	grit blasted	788–823	2.0–20	2.0–12	40–150	17)
7475	ground	783	2.76	1.0	50–150	21), 22)
2004	grit blasted	733–753	1.0–5.0	4.0–6.0	55–105	20)
TiAl	chemical etching	1273	5.0–20	5.0–8.0	250–550	15)
SP700	ground	973–1223	3.5	1.0	150–580	16)

Table 2 Chemical compositions of pure magnesium used in the present investigation.

Mg	Zn	Si	Mn	Fe	Al	Pb	Ca
99.95	0.008	0.005	0.0037	0.003	0.003	0.001	0.001

(mass-%)

Fig. 1 Optical microstructure of as-received material.

the linear intercept size. The microstructure of as-received material is shown in Fig. 1. The average grain size was about 85 μm .

The specimens to be diffusion bonding were cut directly from the rolled sheet. The length is 30 mm and the width is 20 mm, respectively. Before diffusion bonding, the bonding surfaces were blasted using 20 μm diameter alumina grit for 60 s. The blasting pressure was about 0.2 MPa with separations of 300 mm between the specimen and the muzzle of a gun. This was the best condition, because the surfaces could form the most uniform roughness. The grit blasting treatment was done to enhance diffusion bonding quality as reported in the references.^{7,17)} After grit blasting treatment, the specimens were cleaned in ethanol using ultrasonic vibration cleaner to remove the alumina particle. The surface roughnesses of as-received and grit blasted surfaces are listed in Table 3. By using the grit blasting treatments, the surface became more rugged than that of as-received condition. It was found that the surface roughness was nearly equal to particle size used for grit blasting.

Diffusion bonding tests were carried out in air. The pressure range was from 2 to 20 MPa and at the temperature range from 573 to 673 K and for the times up to 72 h. The testing temperature range is equivalent to the absolute tempera-

Table 3 Roughness dimensions of as-received and grit blasting treated surfaces.

Surface	Rmax, (μm)	Rz, (μm)
As-received	10.2	0.35
Grit blasting	35.5	29.7

Rmax; The maximum roughness.

Rz; The average roughness.

ture ranges from 0.62 to 0.73 T_m , where T_m of magnesium is 924 K.²⁵⁾ The overall arrangement of the experimental setup for the production of the overlap joints is shown in Fig. 2(a). The overlap length is 1.5 t , where t is the thickness of the sheet. Temperature fluctuation during the diffusion bonding was less than 2.5 K.

Diffusion bonding quality was assessed by the microstructural observations and compressive lap shear tests. For observation by optical microscopy, the specimens after diffusion bonding were polished and then etched in 10%-acetic for 30 s. A sound bond was defined as a bond which did not show bond line.²³⁾ After the specimen was securely fixed in the jig, compression lap shear tests were carried out with an overlap of 1.5 t at a loading rate of 5 mm/min at room temperature. The schematic model of compression lap shear test is shown in Fig. 2(b). The absolute bond strength could not be used to evaluate the extent of bonding, because specimens having different bonding conditions would have different mechanical properties. For compression lap shear tests, a sound bond was defined as the bond which exhibited a lap shear strength ratio $\tau_b/\tau_p \geq 0.80$, where τ_b and τ_p are the compression lap shear strength of the bond and the compression shear strength of the parent metal, respectively.²³⁾ The fracture surfaces after the compression lap shear test were observed in SEM.

3. Results and Discussion

3.1 Mechanical test

The result of compression lap shear tests after diffusion bonding test at 673 K is listed in Table 4(a). It is apparent that the bonding strength depends on bonding pressure and time, *i.e.*, bonding time decreases with increasing pressure. The maximum ratio of lap shear strength was 0.888 when specimen was bonded at 20 MPa for 1 h. The summary of lap shear tests of diffusion bonding test at 623 K and 573 K is shown in Table 4(b). It is considered that the specimens attain a sound bond, τ_b/τ_p was 0.865, at 20 MPa for 1 h and 623 K. On the other hands, when the bonding temperature was lower than 573 K, the ratio of the lap shear strength was much lower than

Table 4 Summary of lap shear tests of diffusion bonding specimens at (a) 673 K and (b) 573 K–623 K.

Bonding condition		Lap shear strength,			$\tau_b/\tau_p(\%)$
Pressure, P/MPa	Time, t/h	τ/MPa			
2	12	60.2	57.0	52.7	0.529
	24	78.1	77.3	68.4	0.697
	72	79.1	75.6	73.5	0.711
3	3	NA			NA
	6	75.9	68.4	67.2	0.659
	12	85.9	85.4	77.4	0.775
	24	97.0	87.4	85.2	0.840
5	0.25	NA			NA
	0.5	NA			NA
	1	79.4	60.5	58.6	0.537
	3	59.7	58.1	52.2	0.530
	6	79.3	75.2	68.4	0.694
	12	95.3	94.7	90.5	0.874
10	0.25	NA			NA
	0.50	68.4	66.7	65.0	0.571
	1.00	92.9	91.9	70.5	0.692
	3.00	92.3	87.9	81.6	0.816
20	0.25	63.1	61.4	59.6	0.525
	0.50	74.3	69.9	65.5	0.598
	1.00	109.4	106.8	95.7	0.888

NA; The strength was not determined, because of the specimens peeled away before compression lap shear tests.

Bonding condition			Lap shear strength,			τ_b/τ_p (%)
Temperature, T/K	Pressure, P/MPa	Time, t/h	τ/MPa			
573	5.0	0.25	NA			NA
		0.50	NA			NA
		1.00	NA			NA
	10	0.25	NA			NA
		0.50	NA			NA
		1.00	NA			NA
	20	0.25	NA			NA
		0.50	65.9	64.2	62.4	0.548
		1.00	76.3	74.8	58.7	0.565
623	5.0	0.25	NA			NA
		0.50	NA			NA
		1.00	NA			NA
	10	0.25	NA			NA
		0.50	72.4	71.6	70.8	0.612
		1.00	81.8	71.2	69.6	0.622
	20	0.25	67.2	65.9	64.7	0.563
		0.50	80.2	77.3	74.4	0.661
		1.00	108.9	104.8	101.8	0.865

Fig. 2 Experimental setup for (a) the production of overlap joints and (b) the compression lap shear tests.

unity. At these conditions, the specimens peeled away before measurement of the compression lap shear tests, because of weak bonding. It would be required more bonding times and pressures to attain high quality in diffusion bonding.

3.2 Microstructural observation

For microstructural observations, the optical microstructures near the bond are shown in Fig. 3. The white arrows in Fig. 3 show the original surface. The diffusion bonding test conditions were at bonding pressure of 20 MPa for 1 h at 673 K (a) and at 573 K (b), respectively. The bond line is not seen for high lap ratio of shear strength (≥ 0.80) in Fig. 3(a), but visible bond line is observed for low lap ratio of shear strength in Fig. 3(b). It is suggested that low bonding strength is associated with the visible bonding line. In order to obtain high ratio of lap shear strengths, the bond line should disappear from cross section.

SEM micrographs of the fracture surfaces after the compression lap shear tests are shown in Fig. 4. SEM micrograph of the fracture surface on specimens with high ratio of lap shear strengths (≥ 0.80) is shown in Fig. 4(a). The specimen was bonded at 673 K and 20 MPa for 1 h. The failure mode becomes very ductile with islands of cusps. It seems that it is

Fig. 3 Cross-sections through bonded specimens with 20 MPa for 1 h, at (a) 673 K and (b) 523 K. The allows show the bond lines.

difficult to peel away because of sufficiently joining together. The fracture surfaces of the other specimens with high lap shear strength were also fully ductile. In the case of low ratio of lap share strengths (< 0.75), SEM micrograph is shown in Fig. 4(b). The bonding condition was 673 K and 5 MPa for 0.25 h. Several flat areas, indicative of the traces of the original surface before diffusion bonding, are observed on the fracture surface as observed in Fig. 4(b). Similar fracture surfaces were mainly observed below the bonding temperature of 573 K. It is apparent that the trace of the original surface before diffusion bonding resulted from insufficient diffusion bonding. It noted that more bonding time or bonding pressure are required in order to enhance the bonding quality.

3.3 Comparison by prediction analysis

Since there are many factors, *i.e.* temperature, pressure, time and so on, to obtain high quality in diffusion bonding, a number of theoretical models have been developed to achieve the optimal values for those factors.^{26–32} All existing models of diffusion bonding take into account the interfacial void shrinkage. However, these models differed from the origin. Some void shrinkage models were developed from powder sintering models and others were derived from void growth models under tensile creep, treating void shrinkage as negative void growth. The void growth models were generally classified into three categories: plasticity controlled growth,²⁹ diffusion controlled growth^{30,31} and surface con-

Fig. 4 Fracture surfaces of diffusion bonded joints at 673 K (a) 20 MPa, 1 h and (b) 5 MPa, 0.25 h. The allows show the flat areas.

trolled growth.³² The void growth models were noticed in this discussion.

Hancock developed a void growth model by plastic flow of the surrounding material.²⁹ This had originally been produced solely by bulk deformation. The growth rate was given by

$$\frac{dr}{dt} = r\dot{\epsilon} \quad (1)$$

where $\dot{\epsilon}$ is the local strain rate which can be determined from the power law creep equation, t is the time and r is instantaneous cavity radius. Chen and Argon had proposed a simple model to describe the rate of growth of voids during creep based on the Beere and Speight modification of the Hull and Rimmer analysis.^{30,31} They had been derived from the growth rate of the interfacial voids by diffusion. His model defined as

$$\frac{dV}{dt} = \alpha \left(\frac{8\pi\Omega\delta D_{gb}}{kT} \right) \left(\sigma - \frac{2\gamma}{r} \right) \quad (2)$$

where V is the cavity volume, Ω is the atomic volume, δ is the grain boundary width, D_{gb} is the coefficient for grain boundary diffusion, k is Boltzman's constant, T is the absolute temperature, σ is the applied stress, γ is the surface energy and α

Fig. 5 Comparison between the experimental bonding times and the bonding times by prediction analysis.

is the cavity size-spacing parameter defined as

$$\alpha = \frac{1}{4 \ln(B + r/r) - [3 - (r/r + B)^2][1 - (r/r + B)^2]} \quad (3)$$

where B was diffusion distance given as

$$B = \left(\frac{D_{gb} \delta \Omega \sigma}{kT\dot{\epsilon}} \right)^{1/3} \quad (4)$$

The experimental bonding times to obtain high value in lap shear strength were compared with those of prediction analysis by their models.^{29–31)} The comparison between experimental bonding time and that predicted in different diffusion bonding theories is shown in Fig. 5. The symbols of ○, △ and × in Fig. 5 show $\tau_b/\tau_p \geq 0.80$, $0.80 > \tau_b/\tau_p \geq 0.60$ and $\tau_b/\tau_p < 0.60$, respectively. The experimental results of the bonding times were roughly agreed with Chen-Argon model and Beere-Speight model less than 10 MPa of bonding pressure. When the bonding pressure was more than 10 MPa, the bonding times of the experimental results was shorter than that of theoretical analysis. The higher bonding pressure was, the larger errors became. Their models used to calculate the bonding time ignored plastic flow. It is assumed that the void growth was generally plasticity controlled in high stress regions. It is interred that large bonding pressure was easy to deform the surfaces to be jointed consisted of triangular section. It will be needed to constitute suitable model involved plastic flow.

4. Summary

Diffusion bonding was investigated in pure magnesium, using rolled sheet having equiaxed grain sizes with an average size of 85 μm. The following results were obtained.

(1) From the microstructural observations and compressive lap shear test, the optimal diffusion bonding condition was considered to be 20 MPa–1 h, 10 MPa–3 h, 5 MPa–12 h and 3 MPa–24 h at 673 K, respectively. The bond line could not be observed and fracture surfaces were very ductile, because diffusion bonding condition was suitable.

(2) The experimental results of the bonding times were roughly agreed with Chen-Argon model and Beere-Speight model under low bonding pressure. However, since their models did not include plasticity, the bonding times of theo-

retical analysis were overestimated in high bonding pressure.

(3) The present results revealed that there is a possibility to fabricate magnesium products using diffusion bonding technique.

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