Effect of Eutectic Silicon Particle Morphology on the Fluidity of 4045 Aluminum Alloy Filler

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Abstract. The present work studies the effect of eutectic silicon particle morphology on the fluidity of 4045 aluminum alloy filler through dynamic spreading experiment, filling test and welding seam experiment. The experimental results indicate that the morphology of eutectic silicon particle has an excellent influence on the melting speed at the initial melting stage of brazing process: the alloy loaded with smaller eutectic silicon particles exhibits a faster melting speed and better fluidity. When the alloy completely melts along with those internal eutectic silicon particles in a variant morphology, particles sizes have limited impact on the fluidity. In addition, it is discovered that alloy introduced with smaller particles possesses a much lower surface activation energy; furthermore, the proportional relationship the ratio of length to diameter (S) and surface activation energy (Q) is verified in this article, and is consistent with the formula: $Q=2.35S+94.47$.

Introduction

Due to its low cost, good brazability and superior corrosion resistance, 4045 aluminum alloy filler has been widely used in the aluminum heat exchanger [1-2]. Al-Si binary alloy is a eutectic system with the eutectic composition at 12.6% wt.% Si. Silicon reduces the thermal expansion coefficient, increases corrosion and wear resistance, and improves casting and machining characteristics of the alloy. When the Al-Si alloy solidifies, the primary aluminum forms and grows in dendrites or silicon phase forms and grows in angular primary particles. When the eutectic point is reached, the eutectic Al-Si phases nucleate and grow until the end of solidification. At room temperature, hypoeutectic alloys consist of a soft and ductile primary aluminum phase and a hard and brittle eutectic silicon phase. Hypereutectic alloys usually contain coarse, angular primary silicon particles as well as a eutectic silicon phase [3]. Many researchers have studied the modification mechanism of aluminum alloy, and it exists two modification mechanisms: TPRE (twin-plane reentrant edge growth mechanism) [4] and interfacial edge mechanism [5].

There exist many eutectic silicon phases in the 4045 aluminum alloy, which presents different morphology with modification or not [6]. Previous investigations [7-9] have indicated that, after full modification, the flexural strength and ductility of the brazing joint can be enhanced as the morphology of the eutectic silicon transforms from thick sheet-like shape to a coral shape. But there is still few research about the effect of eutectic silicon particle morphology on the fluidity and wettability of filler metal. Many researches show that the quality of the brazed joint depends on the wettability and liquidity of the filler [10-11]. Wei Dai [12] investigated the Al-Si-Zn-Sr filler metals for brazing 6061 aluminum. The results clearly showed that tensile strength of the brazing joint increased from 120MPa to 132MPa as spreading area of the filler increased from 258mm$^2$ to 285mm$^2$. In the present study, the fluidity and wettability of the filler metal with different eutectic silicon particle morphology is discussed through spreading experiment, filling experiment, T-joint specimens typical fillet formation test. The result will contribute to control the morphology of eutectic silicon particle in industry and develop a new 4XXX aluminum alloy filler.
Experimental Procedures

Primary aluminum ingot (97wt. %Al) was melted by the SG2-12-10 crucible electrical resistance furnace at 760±5°C. Then metal Si (97wt. %Si) was added into the molten aluminum. When Si was totally melted, the resulting melt was refined and modified using diverse sodium salts (1# without Na, 2# with 0.6%Na, 3# with 0.9%Na). After the last casting, 4045 aluminum alloys with silicon particles in different morphology were acquired. Table 1 lists the measured compositions of the three alloys used in present work.

| Alloy | Si  | Fe  | Na   | impurity | Al   |
|-------|-----|-----|------|----------|------|
| 1#    | 10.6| 0.17| 0.00  | 0.02 max | Bal. |
| 2#    | 10.7| 0.15| 0.0049| 0.02 max | Bal. |
| 3#    | 10.7| 0.16| 0.0098| 0.02 max | Bal. |

The schematic diagram of dynamic spreading is shown in Figure 1. The dimension of 4045 filler was 4×4×4mm while the thickness of the 3003 baseplate was 1.5mm. The dynamic spreading experiment was performed in GTP110S dynamic vacuum furnace which can be controlled with tolerance among ±3°C assisted by K-type thermocouples. In the spreading experiment, the temperature of furnace was 575°C, 580°C, 590°C, respectively, with the heat rate 30°C/min. The spreading diameter of filler metal was measured in the real time when insulating and the spreading diameter is equal to the difference between the melting diameter and the initial diameter of filler metal.

The filling experiment was carried out according to GB/T11364-1989 <test method of spread ability and clearance filling ability for filler metal>, as shown in Figure 2.

Figure 3 shows the schematic of T-joint specimens used for fillet formation measurements according to JB/T6966-1993 <assessment method of brazing seam appearance quality>. The filling experiment and T-joint test were carried out in a vacuum brazing furnace at 580°C, with heat rate of 30°C/min. Samples were held in furnace for 20s and 300s.

The microstructures of the filler and the brazing joint were observed using Zeiss Axio Imager A2 metalloscope and the ration of length to diameter of the eutectic silicon particle was measured by the Nano Measurer software.
Results

Microstructure of 4045 Aluminum with Different Sodium Modification

The microstructures of modified 4045 aluminum alloys containing variant additional sodium are shown in Figure 4. With unmodified one as control, from which it can be observed that silicon exists in two types of phases: primary silicon and eutectic silicon particles. The content of primary silicon almost remains at the same level after modification using sodium salt solutions, which is consistent with previous researches, nevertheless, which does affect the size of eutectic silicon particles. For unmodified 4045 aluminum alloy, eutectic silicon particles are long rod-like stripes, whereas for modified alloys with sodium enriched, the particles are greatly refined: the size of particles becomes much smaller, disrupting long stripes into pieces and even spots, with the average ration of length to diameter of eutectic Si particle as ca. 36.85 and 16.65, for alloy with 0.0049wt% and 0.0098wt% sodium, respectively, compared to 51.87 for unmodified alloy.

Figure 4. Microstructure of 4045 aluminum alloy modified by different Na additions: (a) 0%; (b) 0.0049%; (c) 0.0098%.

Spreading Experiment

Figure 5 shows the effect of holding time on the spreading diameter of 1#, 2#, 3# 4045 aluminum alloy and Figure 6 shows the photo of the three alloys at initial melting stage and final melting stage at 580°C. As illustrated in Figure 6, the filler with different eutectic silicon particle morphology shows different melting behavior at the initial melting stage. The completely spreading time decreases as the ration of length to diameter of eutectic Si particle decreases, as shown in Table 2. It can also be seen from Figure 6 that the eutectic silicon particle morphology has little influence on the complete spreading diameter after completely melting.
Figure 5. The results of dynamic spreading experiment: the effect of brazing time on spreading diameter.

Figure 6. The photos of filling experiment at 580℃, (a)(d) 1# alloy; (b)(e) 2# alloy; (c)(f) 3# alloy.

Table 2. The melting time of completely spreading.

| 4045 aluminum alloy | melting time/s |
|---------------------|----------------|
|                     | 848.2K         | 853.2K         | 863.2K         |
| 1#                  | 380            | 300            | 220            |
| 2#                  | 300            | 240            | 190            |
| 3#                  | 220            | 190            | 170            |

Filling Experiment

Table 3 lists the filling height at the initial melting stage and complete melting stage of 1#, 2#, 3# alloy at 580℃. It can be concluded that the alloy with smaller eutectic silicon morphology shows larger filling height at the initial melting stage. However, after completely melting, the filling height is almost the same, which is about 43mm.
Table 3. The filling height of alloys at brazing temperature 580°C.

| Alloy | the filling height/mm 20s | 300s |
|-------|--------------------------|------|
| 1#    | 21.5                     | 43.4 |
| 2#    | 38.0                     | 42.8 |
| 3#    | 41.8                     | 43.1 |

T-joint Specimens Typical Fillet Formation Test

Figure 7 shows the cross sections of the fillets of 1#, 2# and 3# filler at initial and final melting period. As illustrated in Figure 8 (a), (c) and (e), at the initial stage, alloy 1# do not melt sufficiently, which leads to an imperfect spreading, yielding the smallest value of \( L_v \), \( L_h \), and \( L_v/L_h \). The value of \( L_v \), \( L_h \), and \( L_v/L_h \) becomes larger as the eutectic silicon particles get more refines. As illustrated in Figure 8 (b), (d) and (f), at final stage, the fillet of the three alloy is almost the same.

Table 4 summarizes the height \( L_h \) and width \( L_v \) of the fillet at different temperature for the three alloys. It is noticed that the alloy with smaller eutectic silicon particle shows larger ratio of \( L_v/L_h \) while holding 20s. However, with extended heating to 250s, the value of \( L_v/L_h \) tends to be stable regardless of size of particles.

![Figure 7](image)

Figure 7. Cross sections of fillets in T-joint specimens at different holding time.

Table 4. The statistics size of T-joint fillet (brazing temperature: 580°C).

| Alloy | \( L_v/\mu m \) 20s | 300s | \( L_h/\mu m \) 20s | 300s | \( L_v/ L_h \) 20s | 300s |
|-------|-------------------|------|-------------------|------|-------------------|------|
| 1#    | 2003              | 9001 | 6838              | 31423| 0.29              | 0.29 |
| 2#    | 3829              | 9078 | 10993             | 30125| 0.35              | 0.30 |
| 3#    | 4537              | 8996 | 12128             | 30347| 0.37              | 0.30 |

Analysis and Discussion

The Effect of Silicon Particle Morphology on the Melting Process

All the three different brazing experimental indicate that the morphology of eutectic silicon particle has a remarkable effect on the melting speed at the initial melting stage. Given that eutectic silicon particle dissolves into aluminum matrix via diffusion during the melting process, the alloy with a smaller silicon particle has a much faster melting speed, because of the larger contact area between
silicon and aluminum matrix. At the initial stage, the smaller the silicon particles are in an alloy, the more readily they dissolve into the alloy when they are melting, resulting in an improved fluidity and lower viscosity of 4045 aluminum alloy, which reduces surface tension. It turns out that the 4045 aluminum alloy loaded with smaller silicon particles exhibits an optimized spreading behavior. When the eutectic silicon particles are completely melt, the performance of spreading can no longer be enhanced because of the uniform silicon distribution in the aluminum matrix.

**Spreading Kinetics**

The spreading process of filler metal technically relates to the element diffusion as well as thermal activation. The following equation can be used to fit the surface activation energy of the melting process [13].

\[
\frac{1}{t} = K \exp \left( -\frac{Q}{RT} \right)
\]  

(1)

where \( t \) is the total melting time in s, \( Q \) is the surface activation energy in kJ/mol, \( R \) is the gas constant 8.314J/(mol·K), \( T \) is the thermodynamic temperature in K, and \( K \) is a constant.

Figure 8 shows the relationship between the logarithm of \( t \) and the reciprocal of temperature \( T \). Through linear fitting, the surface activation energy \( Q \) for alloy 1#, 2#, 3# can be obtaining to be 217.34kJ/mol, 179.46kJ/mol, 134.31kJ/mol, respectively. The result means the filler with smaller silicon particles need lower surface activation energy for melting.

![Figure 8. The relationship between the logarithm of t and the reciprocal of temperature T.](image)

Figure 9 shows the relationship between surface activation energy and the ratio of length to diameter of eutectic silicon particle. It meets the formula as follow.

\[
Q = 2.35S + 94.47
\]  

(3)

where \( Q \) is the surface activation energy in kJ/mol and \( S \) is the ratio of length to diameter of eutectic silicon particle.

![Figure 9. The relationship between surface activation energy and the ratio of length to diameter of eutectic silicon particle.](image)
Conclusion

(1) The 4045 aluminum alloy with smaller eutectic silicon particle has better wettability and fluidity at the initial melting stage. For brazing at 580°C/20s, when the ratio of length to diameter of eutectic silicon particle is about 16.65, the spreading diameter, filling height and \( L_s / L_h \) of the filler is about 2.2mm, 41.8mm and 0.37, respectively. When the ratio of length to diameter is about 51.87, the spreading diameter, filling height and \( L_s / L_h \) of the filler reduce to 1.3mm, 21.5mm and 0.29, respectively.

(2) After completely melting, the eutectic silicon particle morphology has little influence on the fluidity of the filler. For brazing at 580°C/300s, the spreading diameter, filling height and \( L_s / L_h \) of three different filler are all about 4mm, 43.0mm and 0.30, respectively.

(3) The smaller the eutectic particle morphology is, the less melting time the filler needs. When the ratio of length to diameter of the eutectic silicon particle is about 51.87, 36.85 and 16.65, the corresponding surface activation energy is 217.34kJ/mol, 179.46kJ/mol, 134.31kJ/mol, respectively. Furthermore, the relationship surface activation energy \( (Q) \) and the ration of length to diameter \( (S) \) of eutectic silicon particle meets the formula: \( Q = 2.35S + 94.47 \).

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