

## SPRAYING OXIDATION AND DEPOSITION

PROCESS OF Al/Al<sub>2</sub>O<sub>3</sub> COMPOSITES<sup>①</sup>

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**ABSTRACT** Al/Al<sub>2</sub>O<sub>3</sub> and Al-5Si/Al<sub>2</sub>O<sub>3</sub> composites were produced by spraying oxidation and deposition process. The alumina was formed directly through the oxidation reaction of the molten aluminum during spraying atomization and was dispersed in the Al matrix after deposition and solidification. The structure of the composites was examined and the mechanical properties were determined. Also the effects of process parameters were investigated.

**Key words** Al matrix composites spraying oxidation and deposition structure and property

## 1 INTRODUCTION

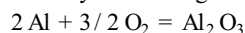
The research and development of metal matrix composites has been one of the most active areas in materials science and technology for past decades. Significant progress has been achieved in this field and some continuous fiber reinforced metal matrix composites have been successfully used in the aerospace and automobile industries<sup>[1-9]</sup>.

The continuous fiber reinforced metal matrix composites, compared with the conventional engineering materials, provide much more excellent properties, but the complicate processing technology and high cost limit their application in the industries. In recent years, more and more interests have been paid to the particle or whisker reinforced metal matrix composites because these kinds of composites can be produced by powder metallurgy or casting process methods<sup>[10-12]</sup>. However, there are considerable problems involved in the introduction of the reinforcement phases into the matrix metal, such as the dispersion of particles which are not wetted by the liquid metal, the segregation during solidification, or the interface reaction between the reinforcement phases and the molten metals.

The Spraying Oxidation and Deposition (SOD) process was developed to produce Al/Al<sub>2</sub>O<sub>3</sub> composites<sup>[13,14]</sup>. The molten aluminum was atomized with an oxidizing gas and the alumina was formed directly through the oxidation reaction of the molten aluminum during spraying atomization and was dispersed in the Al matrix after deposition and solidification. In this method, the problems involved in the introduction of reinforcement phases to the matrix metal could be overcome.

## 2 EXPERIMENTAL PROCEDURE

Fig. 1 is the illustration of the SOD process. Under oxidizing atmosphere, Al will be oxidized immediately according to following reaction:



This reaction is spontaneously towards forming Al<sub>2</sub>O<sub>3</sub> thermodynamically and the oxidation reaction rate is directly related to the temperature of Al<sup>[15]</sup>. The oxidation rate increases with temperature increasing. As the temperature close to the melting point of Al, the oxidation rate rapidly reaches the maximum within very short time. Once a complete Al<sub>2</sub>O<sub>3</sub> layer at the Al surfaces forms, the oxidation process is con-

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trolled by the diffusion process and the rate is reduced greatly.

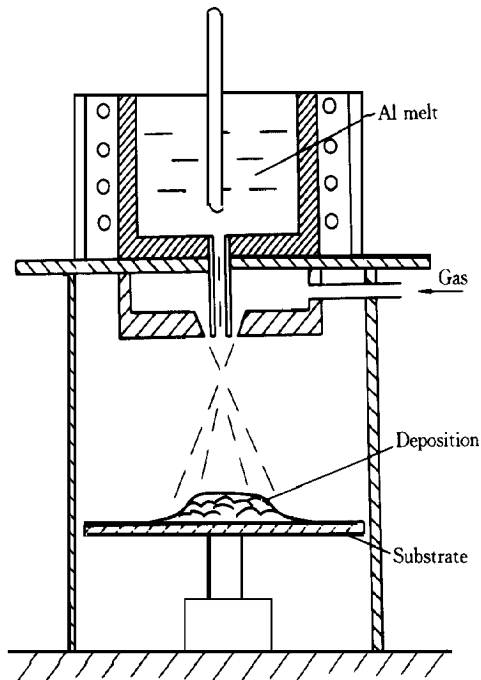


Fig.1 Illustration of Spraying Oxidation and Deposition (SOD) process

Another important factor influencing the oxidation degree of Al is the surface area of Al exposed to the oxidizing medium. With larger surface area of exposed Al, more Al<sub>2</sub>O<sub>3</sub> will be produced. In the SOD process Al melt is atomized by high pressure oxidizing gas into a lot of liquid droplets, producing much greater surface areas exposed to the oxidizing medium. Al<sub>2</sub>O<sub>3</sub> is formed at the surfaces of these Al droplets and the mixture of Al and Al<sub>2</sub>O<sub>3</sub> is deposited together on the substrate. The Al<sub>2</sub>O<sub>3</sub> layer at the surfaces of the Al droplets may be broken into a number of smaller fragments, which are dispersed in the rapidly solidified Al matrix. The whole process can be finished within very short time and the process can be performed continuously. After the melting temperature, the oxidizing medium, or the atomization process is adjusted, and the oxidation degree of Al melt can be controlled.

The materials used in this experiment were pure foundry Al metal and Al-5Si alloy. The ma-

trix alloys were melted in an electrical resistance furnace and the melting temperature was within 780 ~ 800 °C. After the flooding inclusions being removed, the melt was poured into the atomization chamber and was atomized into droplets by a gas mixture of O<sub>2</sub> and N<sub>2</sub>. The oxygen content in the gas mixture was adjusted by changing the partial pressure of O<sub>2</sub> and N<sub>2</sub>, the total gas pressure kept unchanged. The variables investigated in this study include the total gas pressure, the oxygen content in the gas mixture, and the pouring temperature.

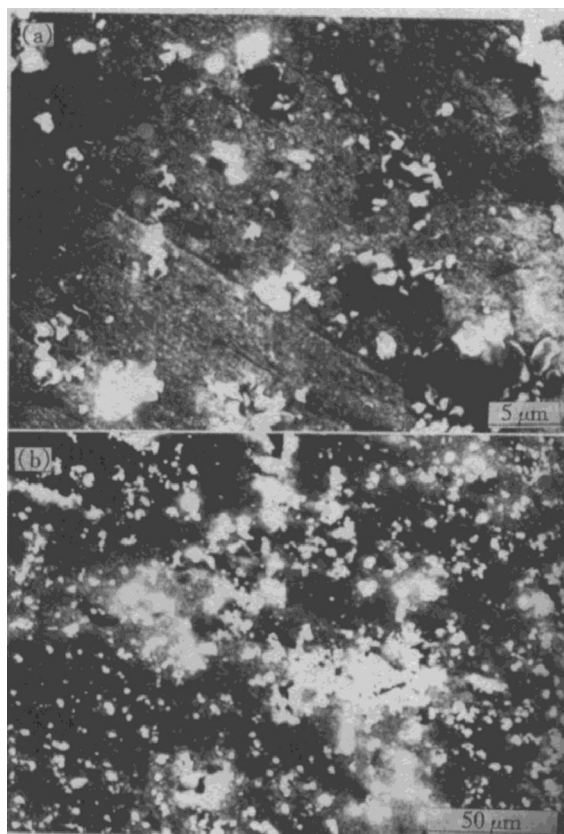
Microstructure examination and mechanical property tests were performed on the samples of SOD processed Al/ Al<sub>2</sub>O<sub>3</sub> and Al-Si5/ Al<sub>2</sub>O<sub>3</sub> composites. Flat sheet specimens with gauge section of 6 mm × 3 mm were used to determine the tensile properties. For the purpose of comparing, samples of same matrix metals were also produced by sand casting method and the mechanical properties were tested.

### 3 RESULTS AND DISCUSSION

Fig.2 shows the typical microstructure of Al/ Al<sub>2</sub>O<sub>3</sub> composites produced by SOD process. The X-ray diffraction and EDAX analyses revealed the presence of certain amount of Al<sub>2</sub>O<sub>3</sub> in the composites. The results obtained from X-ray diffraction are listed in table 1. The 2θ values were measured from the diffraction peaks in the

Table 1 Results of X-ray diffraction of Al/ Al<sub>2</sub>O<sub>3</sub> composites

| 2θ (°) | d( calculated)<br>/ Å | d( data)<br>/ Å | Material                       |
|--------|-----------------------|-----------------|--------------------------------|
| 38.3   | 2.3482                | 2.33            | Al                             |
| 39.4   | 2.2869                | 2.28            | Al <sub>2</sub> O <sub>3</sub> |
| 44.7   | 2.0257                | 2.02            | Al                             |
| 45.6   | 1.9893                | 1.98            | Al <sub>2</sub> O <sub>3</sub> |
| 65.1   | 1.4317                | 1.43            | Al                             |
| 66.3   | 1.4159                | 1.41            | Al <sub>2</sub> O <sub>3</sub> |
| 78.3   | 1.2201                | 1.22            | Al                             |
| 83.4   | 1.1590                | 1.15            | Al <sub>2</sub> O <sub>3</sub> |



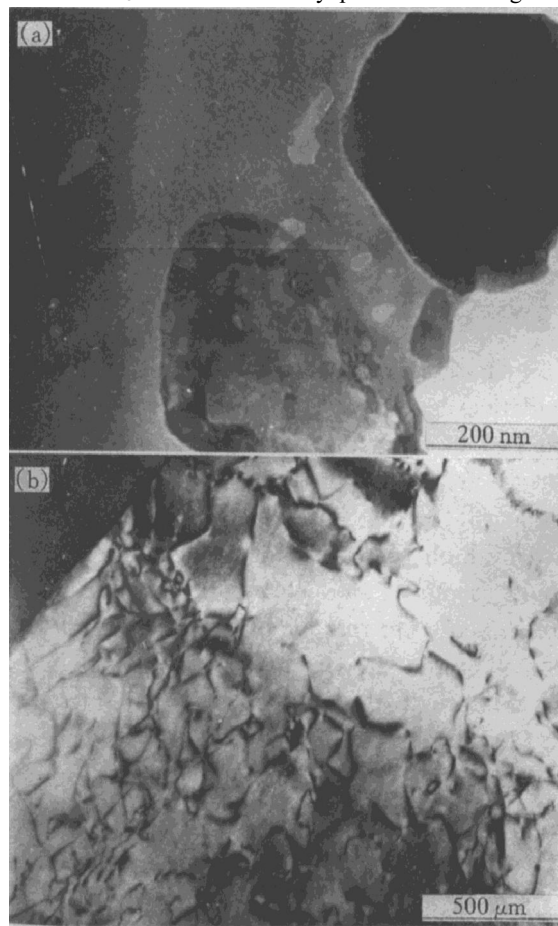
**Fig.2** Typical microstructures of Al/ Al<sub>2</sub>O<sub>3</sub> composites

recorded diffraction curves. The corresponding  $d$  values were calculated by Bragg Law. These  $d$  values were compared with the diffraction data in the 1980 diffraction cards and to be identified as Al and Al<sub>2</sub>O<sub>3</sub> materials, respectively. In the view of a relatively high concentrations of O<sub>2</sub> used in the gas mixture, it is most likely that the oxidation of Al is the dominant reaction between the gas and the Al melt droplets and Al<sub>2</sub>O<sub>3</sub> is the predominant oxidation products.

SEM observations indicated that the Al<sub>2</sub>O<sub>3</sub> particles appeared as irregular forms and were randomly dispersed in the Al matrix. The particle sizes ranged from a few of microns to nanometers. Usually the larger particles consist of a number of smaller particles clustered together, which is possibly formed during the deposition process of larger size metal droplets. Direct experimental evidence providing support to the phenomenon of clustering is not available now.

However, extensive fragmentations of the outer solid layer of the larger size metal droplets are present during the deposition impingement. Regarding the deformation and spreading out of metal droplets at the deposition impingement, the liquid metal surrounds the fragments and solidifies rapidly, causing the fragments clustered together. In addition, a small amount of micro porosity was present in the as-deposited composites, which is related to the spraying and deposition processes and may be reduced effectively by optimizing the processing parameters.

Fig.3 shows the TEM photographs of the Al/ Al<sub>2</sub>O<sub>3</sub> composites. It is considered that in the SOD processed composites there are two types of possible interface structures between Al matrix and Al<sub>2</sub>O<sub>3</sub> which are likely produced during the



**Fig.3** TEM photographs of Al/ Al<sub>2</sub>O<sub>3</sub> composites

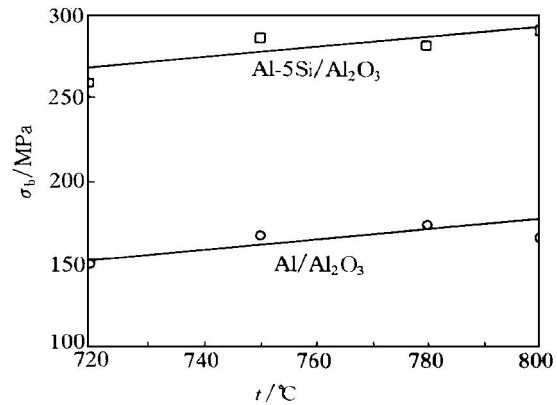
SOD process. One is the continuous structure from the surface Al<sub>2</sub>O<sub>3</sub> layer to the inside unoxidized Al alloys. Another is formed during the deposition impingement, that is, the broken Al<sub>2</sub>O<sub>3</sub> fragments surrounded by the unoxidized and rapidly solidified Al. There is no obvious transition layer observed at the Al/Al<sub>2</sub>O<sub>3</sub> interfaces as shown in Fig. 3. It appears that at temperatures over the melting point of Al, the fresh surfaces of Al<sub>2</sub>O<sub>3</sub> and Al melt are clean and active. A good connection between Al<sub>2</sub>O<sub>3</sub> and Al melt can be formed.

Tensile properties and hardness was measured and the results are listed in table 2. The total pressure was 1.2 MPa, oxygen content in the gas mixture was 15 %, and the pouring temperature was 750 °C. Compared with the matrix metals, the composites samples produced by SOD process provide much higher tensile strength and hardness. The increase in tensile strength is 73 MPa for the Al/Al<sub>2</sub>O<sub>3</sub> composites samples and 105 MPa for the Al-5Si/Al<sub>2</sub>O<sub>3</sub> composites samples. It is possible that the strengthening effects of Al<sub>2</sub>O<sub>3</sub> composites were mainly contributed from the dispersion of Al<sub>2</sub>O<sub>3</sub> in the matrix. For the Al-5Si/Al<sub>2</sub>O<sub>3</sub> composites, additional strengthening may be obtained from the refinement of the matrix alloy, as well as the dispersion of Al<sub>2</sub>O<sub>3</sub>.

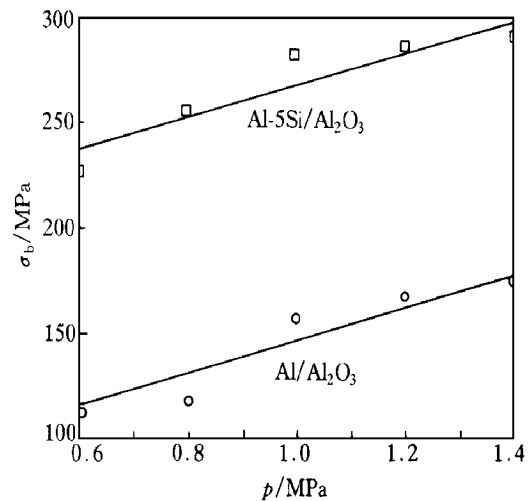
**Table 2** Tensile properties and hardness

| ID No. | Material                              | Condition    | $\sigma_b$<br>/ MPa | $\delta$<br>/ % | HB  |
|--------|---------------------------------------|--------------|---------------------|-----------------|-----|
| I-1    | Pure Al                               | As-cast      | 92.0                | 22.0            | 31  |
| I-2    | Pure Al                               | As-cast      | 99.1                | 19.0            | 28  |
| I-3    | Al/Al <sub>2</sub> O <sub>3</sub>     | As-deposited | 162.8               | 18.0            | 47  |
| I-4    | Al/Al <sub>2</sub> O <sub>3</sub>     | As-deposited | 174.0               | 12.0            | 44  |
| II-1   | Al-5Si                                | As-cast      | 184.3               | 2.4             | 112 |
| II-2   | Al-5Si                                | As-cast      | 167.0               | 2.8             | 105 |
| II-3   | Al-5Si/Al <sub>2</sub> O <sub>3</sub> | As-deposited | 278.0               | 5.8             | 139 |
| II-4   | Al-5Si/Al <sub>2</sub> O <sub>3</sub> | As-deposited | 293.6               | 5.5             | 145 |

The effects of the pouring temperature, total gas pressure, and oxygen content in the gas mixture on the strength of the composites were also investigated. The results are summarized in Fig. 4, Fig. 5 and Fig. 6. It appears that the tensile strength of the composites increased with



**Fig. 4** Effects of pouring temperature on tensile strength of composites



**Fig. 5** Effects of total gas pressure on tensile strength of composites

pouring temperature increasing within 720 ~ 800 °C and with the total gas pressure increasing within 0.6 ~ 1.4 MPa, respectively. When the oxygen content in the gas mixture changed from 5 % to 20 %, the tensile strength of the composites increased. With further increase of oxygen content, the tensile strength decreased. The processing parameters significantly influence the properties of the composites since the microstructure of the composites is closely related to the atomization, the oxidation, and the deposition processes. It is considered that the oxidation product Al<sub>2</sub>O<sub>3</sub>, as well as the refinement of the

matrix alloy, is the main contribution to the strengthening of the composites. However, the micro porosity in the composites negatively influences the properties. Hence, a better understanding of the process optimization is necessary in order to realize the full potential of SOD process for the manufacturing of oxide-dispersion-strengthened metal matrix composites.

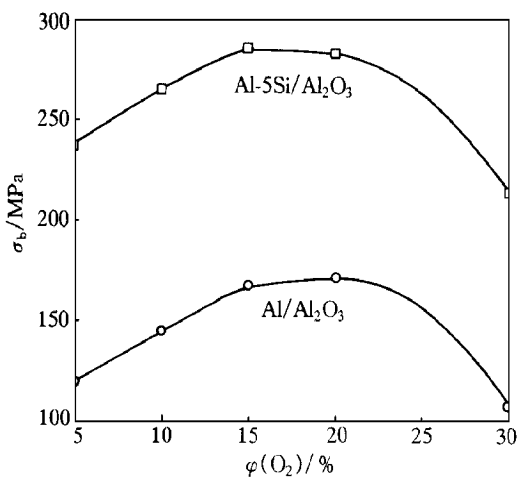


Fig. 6 Effects of oxygen content in gas mixture on tensile strength of composites

#### 4 CONCLUSIONS

(1) In the SOD process, Al or Al-5Si alloy melts are atomized and oxidized simultaneously by using a gas mixture of O<sub>2</sub> and N<sub>2</sub>. Al<sub>2</sub>O<sub>3</sub> forms on the surfaces of the droplets and after deposition and solidification, Al/Al<sub>2</sub>O<sub>3</sub> or Al-5Si/Al<sub>2</sub>O<sub>3</sub> composites are obtained.

(2) The Al<sub>2</sub>O<sub>3</sub> particles in the composites appear as irregular shapes, distribute randomly in the metal matrix. Their sizes are in the range of micron to nanometer. A good connection forms between the Al<sub>2</sub>O<sub>3</sub> and metal matrix.

(3) Compared with the matrix metals, the composites provide much higher tensile strength and hardness. Under the conditions of this investigation, the tensile strength of the composites increase with the total gas pressure and the

pouring temperature increasing. The tensile strength of the composites increase initially with oxygen content in the gas mixture increasing but decrease when the oxygen content is over 20%. The Al<sub>2</sub>O<sub>3</sub> forms in the composites exhibits significantly strengthening ability.

(4) It is suggested that with better understanding and optimization of SOD process, high quality composites with high strength, excellent wear resistance and heat resistance can be produced.

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