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Effect of carbon content on microstructure of in-situ Al_2O_{3p}-TiC_p/Al composites①

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[Abstract] The in-situ Al_2O_{3p}-TiC_p/Al composite was fabricated by XD (exothermic dispersion) process in TiO_2-Al-C system, and the effect of carbon content on the microstructure of the fabricated composite and the reactive temperature characteristics had been studied. The results show that carbon content affects the microstructure and reactive temperature of Al_2O_{3p}-TiC_p/Al composite greatly. The quantity of TiC phases in the fabricated composites increases, while the Al_3Ti phases reduces with increasing carbon content. And when C/TiO_2 molecular ratio is equal to one, the Al_3Ti phase nearly disappears. High temperature metallurgical field caused by strong exothermic reaction influences the synthetic reaction greatly, and higher synthetic temperature was favorable to the synthesis of Al_2O_{3p}-TiC_p reinforce particles.

[Key words] XD technique; carbon content; in-situ Al matrix composites
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1 INTRODUCTION

XD technique is a patent of Martin Marietta Corporation[1]. Composites fabricated by this technique possess many advantages: 1) The synthesized reinforce particles are fine in size and equiaxed in shape[2,3]; 2) The interfacial compatibility between in-situ reinforce particles and matrix is super[4,5]. A series of aluminum composites reinforced with TiC_p, Al_2O_{3p} and TiB_2 had been successfully produced by XD process[6]. The in-situ Al_2O_{3p}-TiC_p/Al composites seem to have a great potential because of their high strength to mass ratio[7]. Previous researches in this field were mainly on the process[8,9] and kinetic mechanism of XD technique[10]. On the synthetic process of Al-Ti-C system, effects of heating parameters[11] (heating rate and hold temperature) and aluminum content[12] on the microstructure and properties of in-situ TiC/Al composites have been studied. Only few researches were reported on influences of TiO_2 and aluminum content on the microstructure in Al-TiO_2-C system, whose results showed that elemental proportion in system has a great influence on the microstructure and property of composite[13,14].

The purpose of this paper is to deal with the effect of the carbon content on the reactive temperature character of the system and the microstructure of the composites.

2 EXPERIMENTAL

The chemical formula to fabricate in-situ Al_2O_{3p}-TiC_p/Al composite by XD process in TiO_2-Al-C system can be given as

$$(13-3x)Al + 3TiO_2 + xC = 2\alpha-Al_2O_3 + xTiC + (3-x)Al_3Ti \quad (1)$$

Materials for experiment were pure aluminum powder (purity is 99.98%, and size 30~50 μm), TiO_2 powder (purity is 99%, size 5~10 μm, contain two types crystals of anatase and rutile) and carbon powder (purity is 99%, and size 30~95 μm).

The raw materials were mixed with the ratio of Al:TiO_2:C = 4:3:x and x express 0, 2/3 and 1 respectively. Then 60% diluent aluminum power was added as a diluent, the mixed powers were milled for 5 h (rotating speed was 200 r/min and the rate of ball and feed was 3:1) in a high energy miller. As milled powders were died to the preform (the diameter is 20 mm, thickness 10 mm and die press 190 MPa) and sintered in furnace in Argon atmosphere. The sample was heated up to 660 ℃ and hold for 40 min. Then the sample was heated up to 900 ℃ and hold for 30 min. At the same time, the temperature change in the sample was measured with PC temperature measure system. The microstructure of synthesized composites was observed by means of LOM and SEM, and phase constitute was analyzed with XRD.

3 RESULTS AND DISCUSSION

In TiO_2-Al-C system, following reactions take place under the condition of XD process:

$$4Al + 3TiO_2 = 2\alpha-Al_2O_3 + 3[Ti] \quad (2)$$

$$[Ti] + 3Al = Al_3Ti \quad (3)$$

$$[Ti] + C = TiC \quad (4)$$

$$4Al + 3C = Al_4C_3 \quad (5)$$

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Fig. 1 shows the relationships between free energy differences of forming Al_3Ti, TiC and Al_4C_3[6] and temperatures when diluent aluminum content is 60%. When temperature is over the critical point 1 685 K, reaction (4) has minimum ΔG, or TiC is more stable than the Al_3Ti. Al_4C_3 is not easy to form in the system.

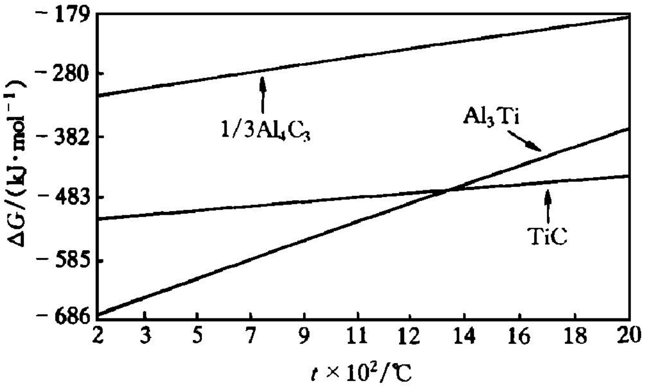

Fig. 1 Free energy difference ΔG of Al_3Ti, $1/3Al_4C_3$ and TiC at different temperatures

Because reactions (2) and (4) are the strong exothermic reaction, once the reaction starts, the exothermic reaction makes the temperature of system rise up quickly. As reaction (4) is stronger exothermic than reaction (3), the increase of carbon content should make the maximum temperature in the system rise up. Fig. 2 shows the relationship between temperature and time measured during the reaction. The maximum temperature of carbon free system rises to 1 240 ℃, which is much higher than the temperature of furnace. When the ratio of carbon and TiO_2 reaches to 1, the exothermic reaction is so strong that NiCr-NiSi thermocouple is melt and broken immediately, shown as Fig. 2(b).

From above experiment, increasing the carbon content makes the reaction speed up, which results in the formation of high temperature in the system. In fact, within the reactive layer, the temperature should rise much higher than the temperature measured in samples. From thermodynamic, higher temperature is favorable to the formation of TiC particles. So when the carbon content is enough, considering of thermodynamic factors, there will be no Al_3Ti in the synthesized composites.

The result of the X-ray diffraction further proves the thermodynamic analysis. Figs. 3(a) and (b) show the results of X-ray diffraction with the ratio of C and TiO_2 being 2/3 and 1 respectively. It shows that the main phases of composites are aluminum matrix, α-Al_2O_3 and Al_3Ti for carbon free system. When the ratio of C and TiO_2 reaches to 1, the main phases are aluminum matrix, α-Al_2O_3 and TiC, no Al_3Ti and other phases are detected. It is clear that Al_3Ti has been restrained by increasing the carbon content.

Figs. 4(a), (b) and (c) show the microstructures of the composite when the molecular ratio of C and TiO_2 is 0, 2/3 and 1 respectively. It can be seen that the white wattle phases of Al_3Ti decrease with the increase of carbon content, and the fine reinforcing particles TiC_p and Al_2O_3 increase correspondingly. When the C and TiO_2 is 1, Al_3Ti nearly disappears. Fig. 4(c) shows that the composites contains mainly aluminum alloy matrix, α-Al_2O_3 and TiC particles distributed uniformly in the matrix.

Figs. 5(a) and (b) are the SEM images of the composites as the ratio of C and TiO_2 is 0 and 1 respectively. It can be seen from Fig. 5(b) that the reinforcing particles α-Al_2O_3 and TiC distributed in the matrix uniformly are very fine, and the average size is 1~ 5 μm.

According to the above experiment and analysis, to manufacture the high quality composite in which the fine α-Al_2O_3 and TiC particles distributed uniformly, the reactive temperature factor is very important. The increase of carbon content results in the formation of higher temperature in metallurgical reaction layer, which is favorable to formation of TiC and α-Al_2O_3 and not favorable to the formation of Al_3Ti from the thermodynamics.

From the dynamics, the increasing temperature can raise the diffusion speed of [Ti] and other elements in the melt aluminum and also improve the

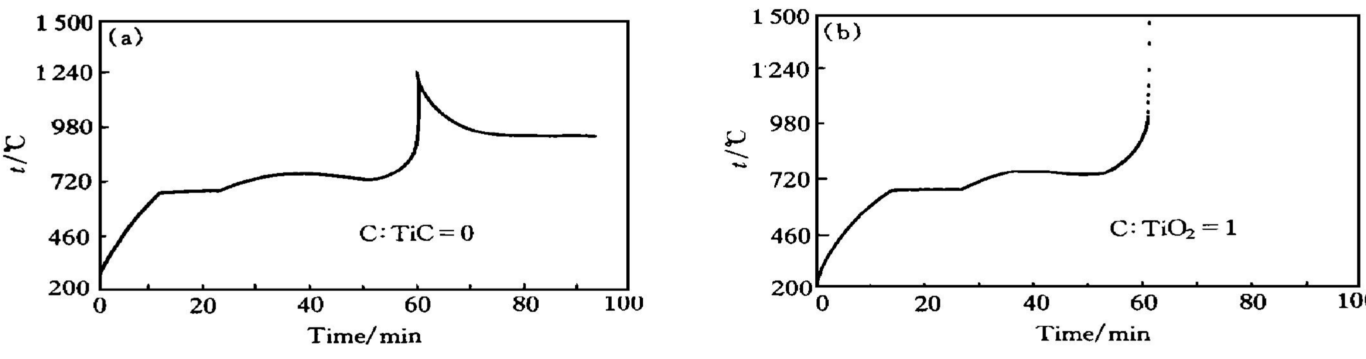

Fig. 2 Relationship between temperature and time in different carbon content systems
(a) —C: TiO_2= 0; (b) C: TiO_2= 1

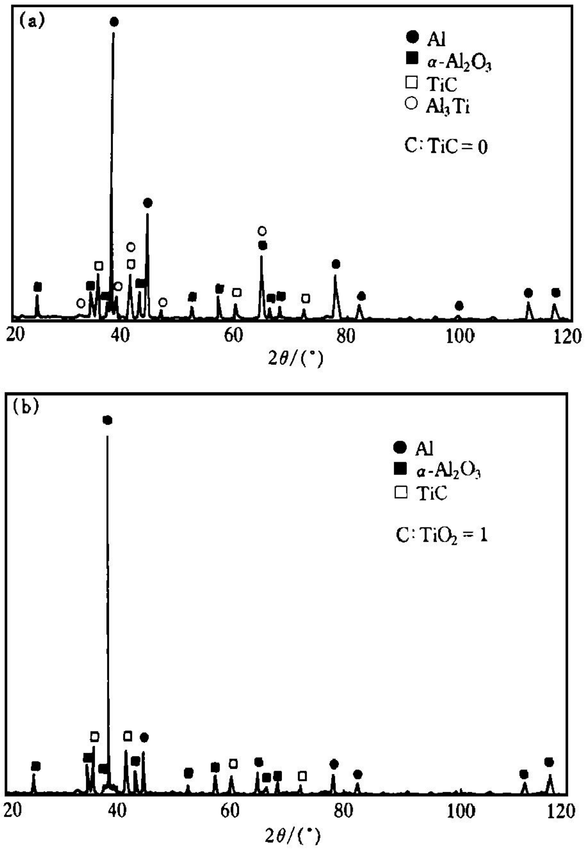

Fig. 3 X-ray diffraction patterns of composites of different ratios of C and TiO_2

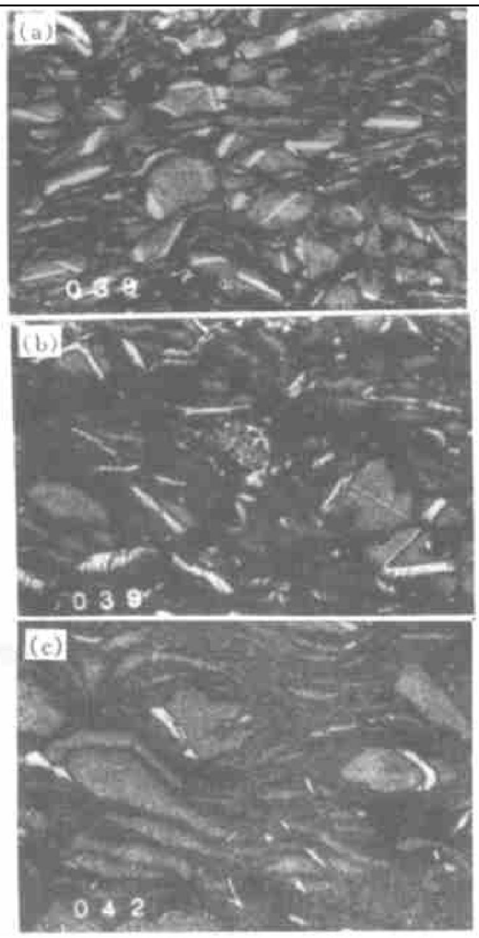

Fig. 4 Microstructures of composites in different ratios of C and TiO_2
(a) —C∶TiO_2= 0; (b) —C∶TiO_2= 2/3; (c) —C∶TiO_2= 1

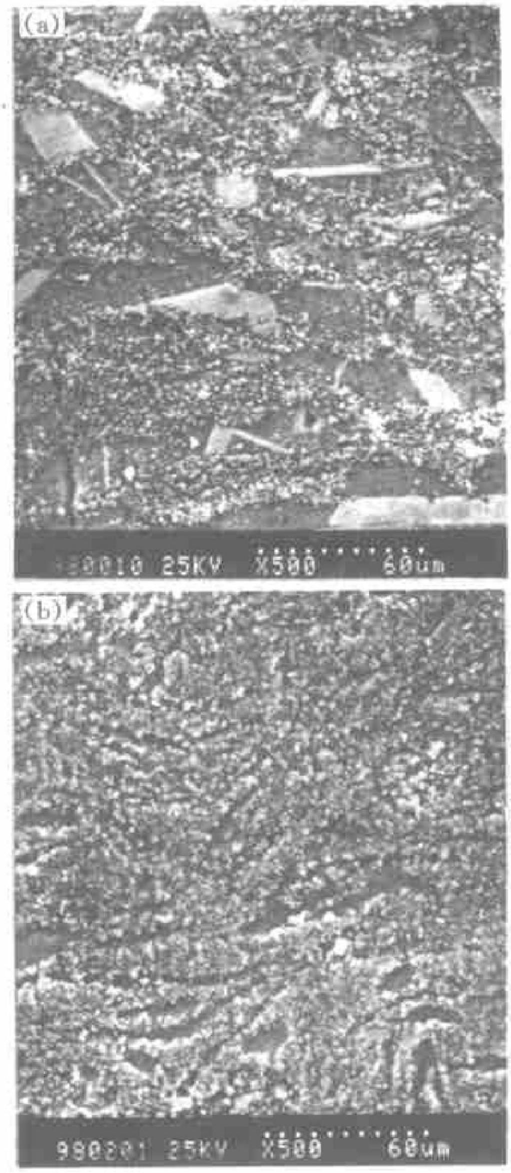

Fig. 5 SEM images of composite of different ratios of C and TiO_2
(a) —C∶TiO_2= 0; (b) —C∶TiO_2= 1

wettablity between the reinforced particles and melt aluminum, which is favorable for [Ti] and C to react as reaction (4) completely.

4 CONCLUSIONS

1) In TiO_2-Al-C system, the strong exothermic reaction results in the formation of high temperature in the reactive layer. The higher temperature in system is more favorable to the formation of TiC and α-Al_2O_3.

2) With the increase of carbon content, the maximum temperature in system, and the reinforcing particles TiC_p and α-Al_2O_3 increase, while unwanted phase Al_3Ti decrease.

3) When the ratio of C and TiO_2 reaches to 1, the composite mainly consist of α-Al_2O_3 and TiC particles distributed uniformly in the aluminum matrix, and the harmful phase Al_3Ti disappears.

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