



Letter

Effect of C/Ti ratio on self-propagating high-temperature synthesis reaction of Sn–Ti–C system for fabricating Ti₂SnC ternary compounds

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ARTICLE INFO

Article history:

Received 31 March 2011

Received in revised form 10 June 2011

Accepted 14 June 2011

Available online 25 June 2011

Keywords:

Intermetallics

Laser processing

Microstructure

X-ray diffraction

ABSTRACT

Ti₂SnC ternary compound was successfully synthesized utilizing laser ignited self-propagating high-temperature synthesis (SHS) of Sn–Ti–C system with the different C/Ti molar ratio. When C/Ti ratio is 0.7, Ti₆Sn₅ as the main phase appears, and a small amount of TiC is also found, most of the Ti₆Sn₅ phases synthesized exhibit the polygon-shaped coarse appearance with an obviously sintered morphology, and the distinct transgranular and intergranular microcracks can be observed. When C/Ti ratio increases over the range from 0.8 to 1.0, the relative content of Ti₂SnC increase and the plate-like shape Ti₂SnC appears. Furthermore, the sintered density increases firstly and then decreases with the increasing of C/Ti ratio.

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1. Introduction

Titanium tin carbide (Ti₂SnC), because of its low hardness, high electrical conductivity, self-lubricity, machinability, damage tolerant capability, resistance to corrosion, etc., is one of the most interesting materials in the family of the layered ternary compounds [1–4]. There are only several synthesis methods such as hot isostatic pressing (HIP), hot pressing (HP) and pressureless sintering adopted to fabricate Ti₂SnC from the powder mixtures of Ti, Sn and C. Barsoum and coworkers [1,2] prepared polycrystalline Ti₂SnC by HIP of Ti, Sn and C powders in the temperature range of 1250–1325 °C for 4–10 h under 50–70 MPa. Zhou et al. [3] fabricated the bulk Ti₂SnC by HP of Ti, Sn and C at 1250 °C for 2 h under 30 MPa. Dong et al. [4] obtained Ti₂SnC powder by pressureless sintering of Ti, Sn and graphite at 1150 °C for 2 h at Ar atmosphere. Vincent et al. [5] synthesized Ti₂SnC powder by pressureless sintering of Ti, Sn and graphite at 1200 °C for 1–6 h at Ar atmosphere. Li et al. [6,7] employed the pressureless sintering technique to fabricate high-purity Ti₂SnC powders from the Ti–Sn–C and Ti–Sn–TiC powder mixtures at 1200 °C for 15–60 min in vacuum. In general, the synthesized Ti₂SnC from most of the previous studies was accompanied by small amounts of TiC, Ti₆Sn₅, and Sn. All these methods not only needed high cost, but also the purity of the synthesized Ti₂SnC powder was low. From an industrial point of view, it is nec-

essary to develop a new method to fabricate Ti₂SnC in relatively short time.

Self-propagating high-temperature synthesis (SHS), developed by Merzhanov and Borovinskaya [8] in the late 1960s, is a new kind of method to produce materials. It has been extensively employed for the production of ceramic [9,10], ceramic matrix composites [11], and so on. It has many attractive advantages, such as high purity of products, low processing cost, and energy and time efficiency, no high-temperature furnace process, non-polluting traits, etc. Applying this method, Yeh et al. [12] employed Self-propagating high-temperature synthesis technique to synthesize Ti₂SnC powders from the Ti–Sn–C and Ti–Sn–TiC powder mixtures.

The main purpose of the present study is to investigate the effect of the C/Ti atomic ratio on the phase composition, microstructure and densification of the synthesized Ti₂SnC by the self-propagating high-temperature synthesis reaction of Ti–Sn–C system.

2. Experimental procedures

Commercial powders of Sn (99.0% purity, 38 μm), titanium (99.5% purity, 15 μm) and graphite (99.9% purity, 38 μm) were used for experiments. Reactant powder mixtures with 30 wt.% Sn and C/Ti atomic ratios ranging from 0.6 to 1 were prepared. The powders were mixed in a ball mill for 3 h and then pressed at 8 MPa pressure to obtain densities of 70% theoretical density. Microstructures and phases of the synthesized compacts were investigated by the scanning electron microscopy (SEM) (Model JSM-5310, Japan) equipped with energy-dispersive spectrometer (EDS) (Model Link-ISIS, Britain) and X-ray diffraction (XRD) (Model D/Max 2500PC Rigaku, Japan).

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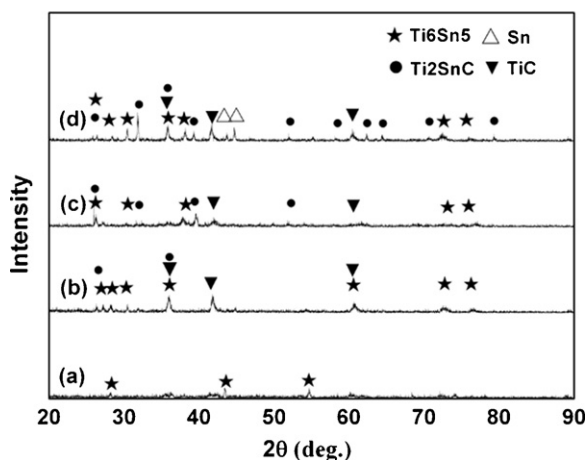


Fig. 1. The XRD patterns of the SHS reaction products synthesized by green compacts with 30 wt.% Sn mixed with different C/Ti (molar ratio), (a) 0.7, (b) 0.8, (c) 0.9 and (d) 1.0.

3. Results and discussion

3.1. XRD analysis

Fig. 1 shows the XRD patterns of the SHS synthesized products by green compacts with 30 wt.% Sn mixed with different C/Ti (molar ratio), (a) 0.7, (b) 0.8, (c) 0.9 and (d) 1.0, respectively. From Fig. 1, it can be observed that Ti_6Sn_5 as the main phase appears and a small amount of TiC are also found when C/Ti ratio is 0.7. The above results illustrate that reactions between Ti–Sn and Ti–C occur. When C/Ti ratio increases over the range from 0.8 to 1.0, besides Ti_6Sn_5 and TiC phases, the Ti_2SnC phases are detected, furthermore, the relative content of Ti_2SnC phases increase and Ti_6Sn_5 phases decrease with the increase of C/Ti ratio, respectively. This result illustrates that reaction between Ti_6Sn_5 and TiC occurs. It is interesting to note that the relative content of TiC phases decrease firstly and then increase with the increase of C/Ti ratio, Sn is detected when C/Ti ratio is 1.0. This result illustrates that Ti_2SnC phases begin to decompose. In a word, in the Ti–Sn–C system, Ti_6Sn_5 and TiC phases are firstly formed between Ti–Sn and Ti–C, and then Ti_6Sn_5 phases reacts with TiC to form Ti_2SnC , at last Ti_2SnC phases begin to decompose partly. Thus, the following four reactions are possible to occur during the SHS reaction:



3.2. Microstructure analysis

To gain a better insight into the microstructures, the typical microstructures of the fracture surface of the synthesized products by 30 wt.% Sn mixed with different C/Ti (molar ratio), (a) 0.7, (b) 0.8, (c) 0.9 and (d) 1.0, respectively are shown in Fig. 2(a)–(d). When C/Ti ratio is 0.7, most of the Ti_6Sn_5 phases synthesized exhibit the polygon-shaped coarse appearance with an obviously sintered morphology, and the distinct transgranular and intergranular microcracks were observed (Fig. 2(a)), which is in good agreement with Ref. [13]. The formation of microcracks may account for the great coefficient of thermal expansion (CTE) anisotropy of Ti_6Sn_5 and the high thermal residual stresses in the products, which has a harmful effect on the mechanical property of the material especially when the grains are coarse.

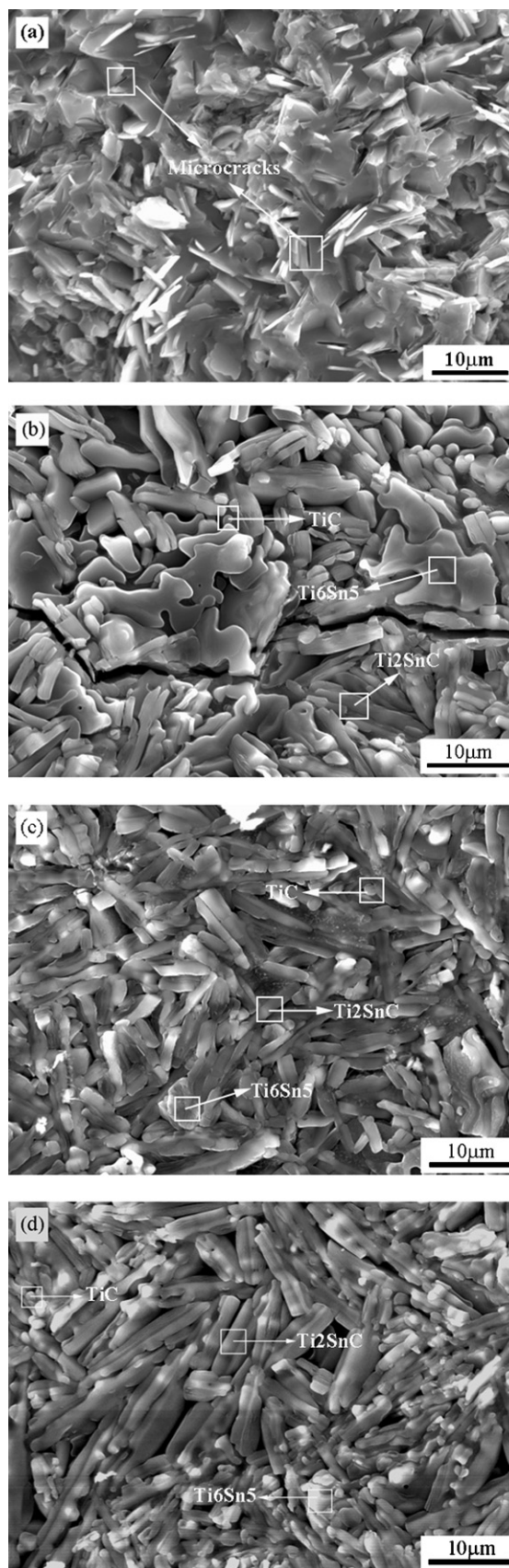


Fig. 2. The typical microstructures of the fracture surface of the products synthesized by 30 wt.% Sn mixed with different C/Ti (molar ratio), (a) 0.7, (b) 0.8, (c) 0.9 and (d) 1.0.

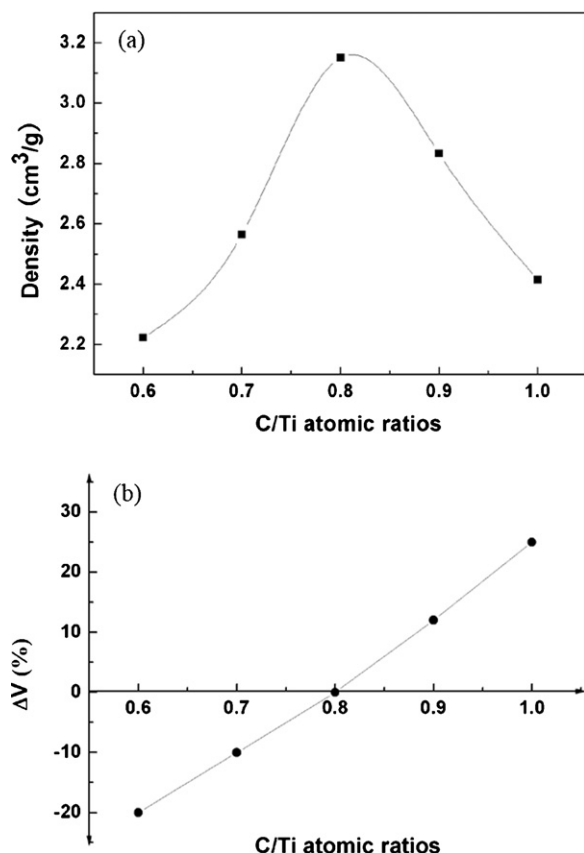


Fig. 3. (a) The sintered density with different C/Ti ratio; (b) effect of plot of V on different C/Ti ratio. ($V = (C + Ti)$ content (vol.%) – product porosity (%)).

When C/Ti ratio is 0.8, it can be observed that the plate-like shape Ti_2SnC appears with an average size of about $10\ \mu m$, furthermore, the amount of microcracks decrease and the size of microcracks increase (Fig. 2(b)). But when C/Ti ratio further increases over the range of 0.9–1.0, the amount of the plate-like shape Ti_2SnC increase, but the morphology and size of the Ti_2SnC grains do not change so much, at the same time, the amount of TiC increase and that of Ti_6Sn_5 decrease (Fig. 2(c) and (d)). When C/Ti ratio is 1.0, Ti_2SnC becomes the predominating phase (Fig. 2(d)), which is in good agreement with the results from the XRD patterns (Fig. 1). The reason of the increase of Ti_2SnC amount is due to the increase of C/Ti ratio accelerating the occurrence of the reaction (3). The reason of the increase of TiC amount is due to the part decomposability of Ti_2SnC (reaction (4)). Furthermore, it can be observed that the microcracks gradually disappear with the increase of C/Ti ratio (Fig. 2(c) and (d)).

3.3. Density analysis

The sintered density with different C/Ti ratio is given in Fig. 3. From Fig. 3, it can be seen that the sintered density increases before 0.8 (C/Ti ratio) and then decreases with further increasing C/Ti ratio. The maximum value of the sintered density is up to $3.15\ g/cm^3$ when C/Ti ratio is 0.8. The reason may be as follows: let us assume Ti_6Sn_5 is a filling phase, unconcerning the effect of sintering, the increase of C/Ti ratio induces that more filling phase will fill in porosity, which increases density of products, porosity is occupied completely just as C/Ti ratio is 0.8 (as shown in Fig. 3(b)), therefore, density of products will emerge the maximum value when C/Ti ratio is 0.8. The reason for the decrease in the sintered density for C/Ti ratio higher than 0.8 may be as follows: when C/Ti ratio is over 0.8, lots of Ti_6Sn_5 are consumed by the reaction (3), which makes the content of Ti_6Sn_5 in the Ti–Sn–C decrease.

4. Conclusions

Ti_2SnC ternary compounds have been successfully prepared by LISHS reaction of Ti, Sn and C powders. The results show that C/Ti ratio has a significant effect on phase composition and the density of the products. When C/Ti ratio increases, the amount of Ti_2SnC synthesized increase and the density of the products increases firstly and then decreases, a maximum value of density appears when C/Ti ratio is 0.8. Furthermore, it can be seen that the microstructural morphology of the products are dependent on C/Ti ratio.

Acknowledgements

The author would like to acknowledge the Youth Science Foundation of Shanxi (No. 2010021022–2), Natural Science Foundation of China (No. 50975264) and Ministry of Education, New Century Excellent Talents support project for the financial support.

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