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Effects of different Sn contents on formation of Ti_2SnC by self-propagating high-temperature synthesis method in Ti–Sn–C and Ti–Sn–C–TiC systems

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Effect of different Sn contents on combustion synthesis of Ti_2SnC was studied using elemental Ti, Sn, C and TiC powders as raw materials in the Ti–Sn–C and Ti–Sn–C–TiC system, in which the molar ratio of Ti/C was set as 2:1. The reaction mechanism for the formation of Ti_2SnC was also investigated. The results showed that the amount of Ti_2SnC in combustion products firstly increased with increasing of Sn content (0.6 to 0.8 mol), and then decreased with further increasing of Sn content (1.0 to 1.2 mol). Upon addition of 15 % TiC instead of Ti and C, the optimum addition of Sn decreased to 0.7 mol and a higher purity of Ti_2SnC was obtained. The Ti_2SnC powders were characterized by scanning electron microscopy (SEM) and X-ray diffraction (XRD).

Keywords: *Ti_2SnC; self propagating high-temperature synthesis; TiC; Sn; reaction mechanism*

1. Introduction

Ti_2SnC is one of the layered ternary carbides, which was first identified by Jeitschko et al. in 1963 [1]. They proposed that Ti_2SnC has a hexagonal symmetry with a space group of $P6_3/mmc$ and lattice parameters of a = 0.3168 nm and c = 1.363 nm. The crystal structure of Ti_2SnC is shown in Fig. 1. It has a unique combination of both metallic and ceramic properties: high thermal and electrical conductivity, excellent thermal shock resistance, easy machinability, low hardness and self-lubricity, etc. [2–5]. Despite the remarkable advantages, it is difficult to synthesize pure phase products. At present, there are several processing routes, such as hot pressing (HP) [6], hot isostatic pressing (HIP) [7], pressureless sintering [8] and self propagating high-temperature synthesis (SHS) [9–11]. The SHS method has many attractive advantages, such as high purity of products, low processing cost, energy and time efficiency, non-polluting traits, etc. [10].

The main purpose of this paper is to investigate the effect of Sn content on the phase composition and microstructure of synthesized Ti_2SnC and establish the reaction mechanism of the compound formation in the Ti–Sn–C and Ti–Sn–C–TiC system.

2. Experimental procedures

Commercial Ti (average particle size: 500 mesh, 99.7 purity), Sn (average particle size: 200 mesh, 99.6 purity), carbon black (average particle size: 330 mesh, 99.5 purity) and self-made TiC (average particle size: 200 mesh) powders were used as the starting materials. The powders of Ti, Sn, C and TiC were mixed at a stoichiometric ratio of Ti_2SnC with ethanol by wet ball milling for 8 h at 20 rpm. After ball milling and drying, the mixed powders were cold pressed to form green compacts with the theoretical density of 50 %, a diameter of about 30 mm and a height of about

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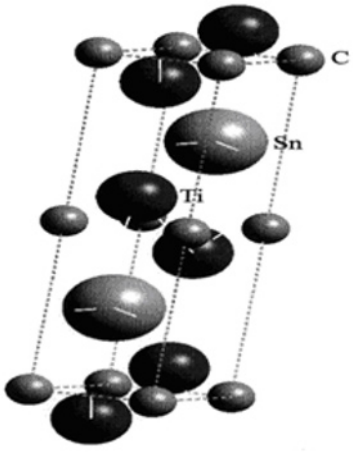

Fig. 1. Crystal structure of Ti_2SnC.

45 mm. The reactants were ignited with a tungsten wire ring under argon atmosphere protection. The combustion reaction temperature was tested with a W/3%Re-W/25%Re thermocouple and recorded by a computer data acquisition system, which was connected with the thermocouple. The X-ray diffraction was used to analyze the phase composition of the products. Scanning electron microscopy was used to observe the microstructure morphology. The relative quantity of Ti_2SnC was represented by F value (the ratio of diffraction intensities of non-overlapping Ti_2SnC_2 (002, $2\theta = 12.97°$) and TiC (111, $2\theta = 35.9°$).

3. Results and discussion

3.1. Effect of amount of Sn on the phase evolution and combustion temperature of obtained product

Fig. 2 shows the XRD patterns for the samples with the mole ratio of Ti:C = 2:1, in which the addition content of Sn has been changed of 0.6 mol, 0.7 mol, 0.8 mol, 1.0 mol and 1.2 mol. Fig. 2a indicates that the Ti_2SnC dominates over TiC in the final product. Besides, TiC is identified as a secondary phase. Fig. 2 (b – c) reveals that Ti_2SnC is still the predominating phase but the diffraction peak of TiC decreases with the increasing of Sn content. When the content of Sn increases to 1.0 mol, some unreacted Sn is also detected, indicating that the amount of Sn in the starting materials is in excess. So we determine the optimum raw materials mole ratio as Ti:Sn:C = 2:0.8:1.

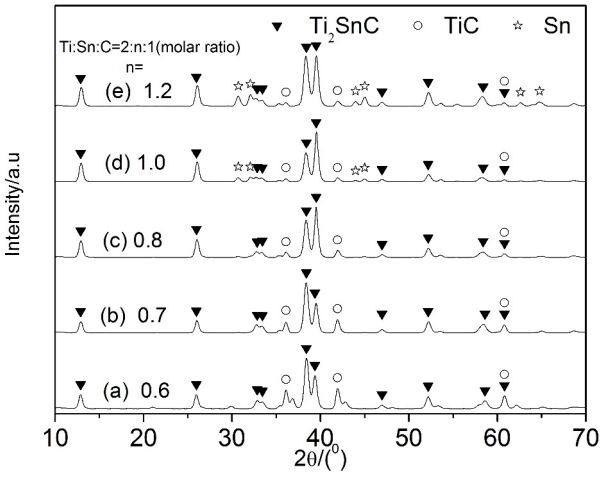

Fig. 2. XRD patterns of products synthesized with different amounts of Sn additions at Ti:C mole ratio of 2:1.

Fig. 3 shows the profiles of solid state combustion of the investigated powders compacts with different amounts of Sn. As shown in Fig. 3, the abrupt rise in temperature signifies rapid arrival of the combustion front and the peak value corresponds to the combustion front temperature. After the passage of the combustion wave, an appreciable decrease in temperature is a consequence of heat losses to the surroundings. The flame-front temperature achieves 2192.75 °C, 2187.44 °C and 2174.22 °C for Sn contents changing of 0.6, 0.7 and 0.8 mol, respectively.

Fig. 4 provides the relationship between the maximum reaction temperature and Sn content. The plot shows that the maximum reaction temperature is substantially affected by Sn addition content and decreases approximately linearly with increasing Sn content.

3.2. Characterization of self made TiC powders

XRD pattern in Fig. 5 shows the phase composition of the self made TiC. It can be seen that a high purity TiC is obtained, and no other impurity phase is detected.

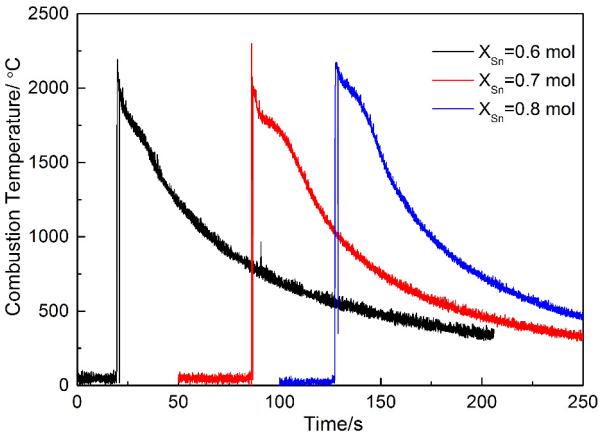

Fig. 3. Effect of Sn content on combustion temperature of Ti–Sn–C powder compacts.

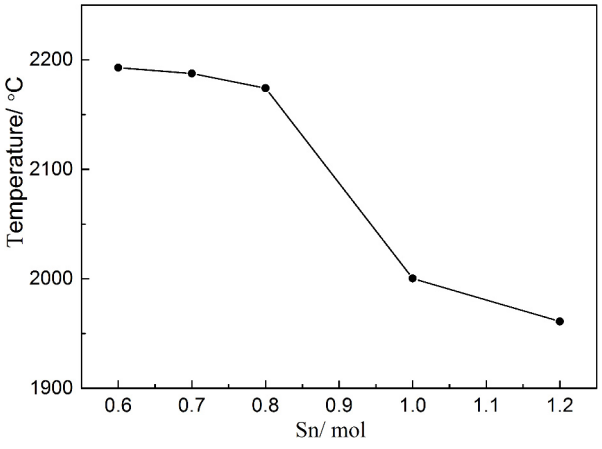

Fig. 4. The relationship between the maximum reaction temperature and Sn addition.

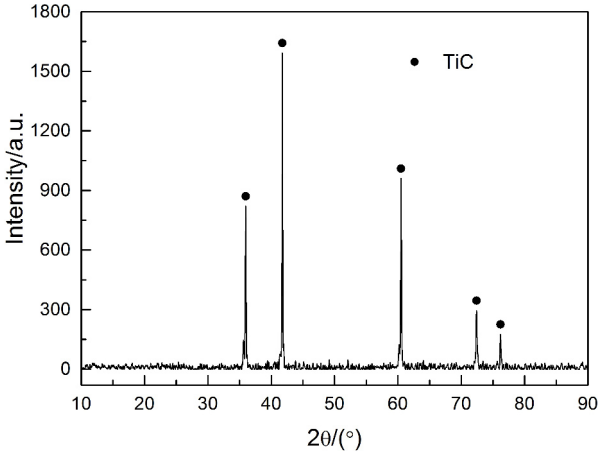

Fig. 5. XRD pattern of self made TiC powder.

Fig. 6 shows the SEM micrograph of the self made TiC powder. Based on the XRD information, it can be presumed that the faceted grains are composed of TiC. The single phase TiC has been prepared by combustion synthesis method.

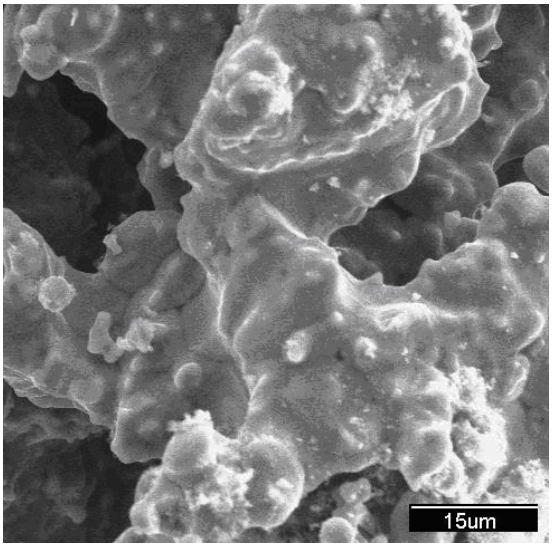

Fig. 6. SEM micrograph of self made TiC.

3.3. The effect of amount of Sn on phase evolution and combustion temperature of the product containing 15 % TiC addition

Fig. 7 shows the XRD patterns of the final products with the mole ratio of Ti:C = 2:1, in which instead of Ti and C 15 % TiC was added. Sn contents were changed from 0.5 mol to 1.3 mol. It can be seen that the amount of Sn has a great effect on the properties of Ti_2SnC. It should be noted that the Ti_2SnC is the predominating phase. Ti_5Sn_3, TiC and Sn are always accompanied by Ti_2SnC except for the case when the content of Sn is 0.7 mol. Moreover, Ti_2SnC and TiC are both predominating phases when the Sn addition is 0.5 mol. When the addition of Sn increases to 0.9 mol, TiC also becomes dominating and the peaks corresponding to Sn appear in the XRD patterns. So, the optimum addition of Sn is 0.7 mol. Compared with Fig. 2, we can see that a higher purity Ti_2SnC is obtained and Ti_5Sn_3 and TiC both show very week peaks after addition of 15 % TiC, indicating that the Ti_5Sn_3 phase almost reacted with TiC to form

Ti_2SnC. However, the amount of TiC in the initial composition was excessive. In the future research, we will adjust the composition of TiC to obtain a higher purity Ti_2SnC.

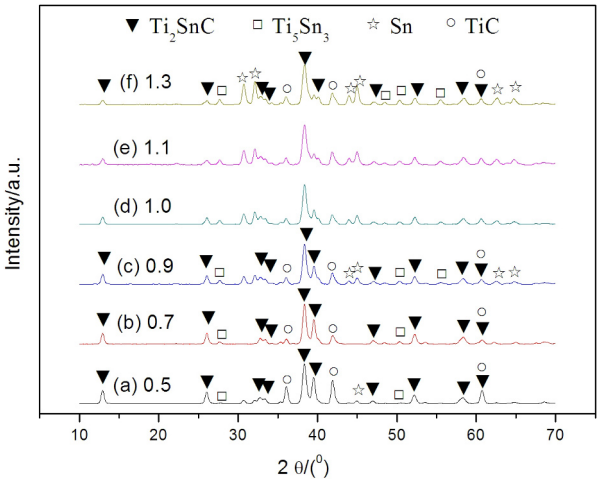

Fig. 7. XRD patterns of a sample made of Ti and C with a mole ratio of 2:1 with 15 wt.% TiC and different contents of Sn.

F value (a relative quantity of Ti_2SnC in the product) after addition of different Sn amounts into the Ti–Sn–C 15 % TiC system is shown in Fig. 8. It is found that the relative quantity of Ti_2SnC increased firstly with increasing of Sn content up to the maximum at the Sn content of 0.7 mol. While the content of Sn increased further, the F values began to decrease indicating that the addition of more Sn was not beneficial for the production of Ti_2SnC. So, when the ratio of the Ti:C = 2:1, the content of Sn is 0.7 mol, the relative quantity of synthesized Ti_2SnC is the largest.

Fig. 9 shows measured temperature profiles for 15 % TiC-added samples with different Sn contents. As shown in Fig. 9, the flame-front temperatures vary from 1304.82 °C to 1909.24 °C with the content of Sn ranging from 1.3 mol to 0.7 mol. Maybe the Sn has a similar dilution effect as the TiC on solid state combustion of the Ti–Sn–C–TiC powder compacts.

The SEM micrographs of the final products with different Sn contents are shown in Fig. 10. Large amounts of small, coarse, round-shaped granules of TiC [12, 13] and little plate-like

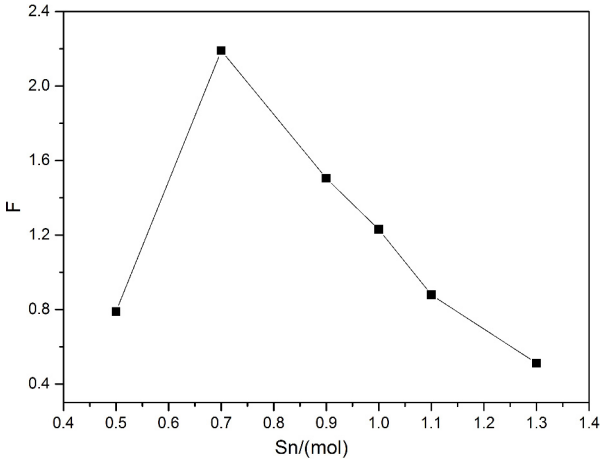

Fig. 8. F value (relative quantity of Ti_2SnC) in the product versus Sn content in the Ti–Sn–C 15 % TiC system.

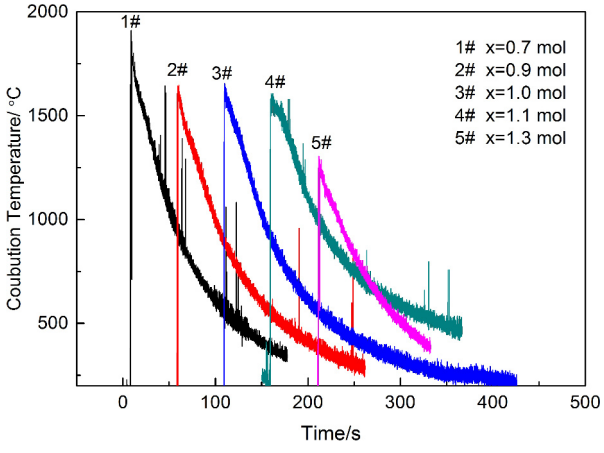

Fig. 9. Effect of Sn content on combustion temperature of Ti–Sn–C 15 % TiC powder compacts.

Ti_2SnC grains [8, 9] are observed (Fig. 10a). When the addition of Sn is 0.7 mol, a lot of well-developed plate-like Ti_2SnC structures evidently increases and the round-shaped TiC granules disappear (Fig. 10b), which is in good agreement with the results from the XRD patterns. Fig. 10c reveals that the Ti_2SnC grains are not well developed, and some rod shaped Ti_2SnC grains can be also observed in Fig. 10d. At the same time, some TiC particles are also found. The crystal structure morphology is determined by the grain growth rate in different planes reported by Hartman and Perdok [14]. Based on the study by Li et al. [15], the growth

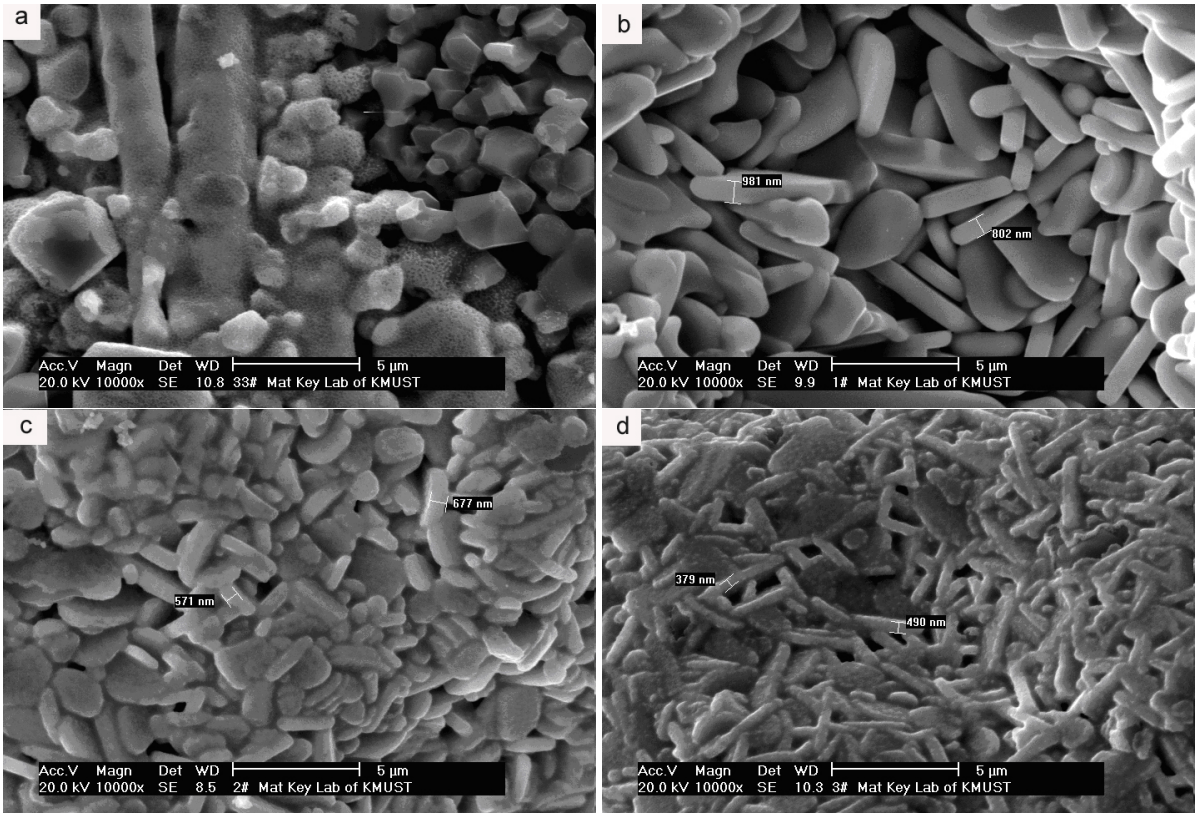

Fig. 10. SEM micrographs of the fracture surfaces of the samples prepared with different Sn amounts (a) 0.5 mol, (b) 0.7 mol, (c) 0.9 mol, (d) 1.1 mol.

environment with sufficient liquid phase is one of the reasons that promotes the development of Ti_2SnC rod-like microstructure. Therefore, it is believed in this study that the increase of Sn amount results in formation of more molten compound in the synthesis, which favors the growth of Ti_2SnC in the forms of rods instead of platelets.

4. Conclusions

The ternary carbide Ti_2SnC was prepared by self-propagating high-temperature synthesis method and the reaction mechanism of the formation of Ti_2SnC has been studied. The results showed that the addition of Sn had an effect on the purity of the final products and the amount of Ti_2SnC in combustion products. It firstly increased with increasing of Sn content (0.6 to 0.8 mol), and then decreased with further increasing of Sn content (1.0 to 1.2 mol). When 15 % TiC was added instead of some amount of Ti and C, the optimum addition of Sn decreased to 0.7 mol and a higher purity of Ti_2SnC was obtained. In the system of Ti–Sn–C–TiC, Ti_2SnC was formed from a reaction between Ti_5Sn_3 and TiC.

Acknowledgements

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