Synthesis of high-purity Ti_3SiC_2 and Ti_3AlC_2 by spark plasma sintering (SPS) technique

WEIBING ZHOU, BINGCHU MEI, JIAOQUN ZHU, XIAOLIN HONG
State Key Laboratory of Advanced Technology for Materials Synthesis and Processing, Wuhan University of Technology, Wuhan 430070, People's Republic of China
E-mail: jsyczwb@sina.com

Recently, Ti_3SiC_2 and Ti_3AlC_2 have received increasing attention because they show both metallic and ceramic properties simultaneously. Like metals, they are thermally and electrically conductive, easy to machine with conventional tools, and resistant to thermal shock; like ceramics, they have high strength, high melting point and thermal stability [1].

The fabrication of single-phase, bulk dense samples of Ti_3SiC_2 and Ti_3AlC_2, however, has proved to be very difficult. Recently, Barsoum *et al.* have fabricated high-purity Ti_3SiC_2 and Ti_3AlC_2 polycrystals by hot isostatically pressing (HIP) a mixture of Ti, graphite and SiC powders, and Ti, graphite and Al_4C_3, respectively [2, 3]. However, the process was very complex. More recently, our research revealed that appropriate addition of aluminum (or silicon) improved the synthesis of Ti_3SiC_2 (or Ti_3AlC_2), and polycrystalline bulk Ti_3SiC_2 and Ti_3AlC_2 materials with high purity could be fabricated by hot pressing (HP) [4, 5].

The objective of this work was to fabricate highpurity Ti_3SiC_2 and Ti_3AlC_2 using Spark Plasma Sintering (SPS) technique.

All of the work was conducted using powder mixtures of TiC (99.2% pure, 8.4 μm), Ti (99.0% pure, 10.6 μm), Si (99.5% pure, 9.5 μm), and Al (99.8% pure, 12.8 μm) (all from Institute of Non-Ferrous Metals, Beijing, China). In brief, the mixture with a designed composition was firstly mixed in ethanol for 24 hrs, then placed in a graphite die, 20 mm in diameter, and finally sintered in a spark plasma sintering system. The samples were heated at a rate of 80 °C/min until the requisite temperature was reached; the soaking times were 8 min, and the pressure was 30 MPa. The density of the sintered products was measured by the Archimedes' method. The sintered product was characterized by X-ray diffraction (XRD) using a rotating anode X-ray diffraction (Model D/MAX-RB, RIGAKU Corporation, Japan). The microstructures of the samples were investigated using scanning electron microscopy (SEM) (Model JSM-5610LV, Jeol Ltd., Japan).

Shown in Fig. 1 are the X-ray diffraction patterns of samples obtained from the mixture of raw materials of $2TiC + 1Ti + 1Si + 0.2Al$ (moles). When sintered at 1150 °C, the main phase was Ti_3SiC_2. However, the peak of TiC ($2\theta = 41.82°$), which was very weak in contrast with those of Ti_3SiC_2 was present. When the sintering temperature reached 1250 and 1300 °C, the product was pure Ti_3SiC_2; no phase but Ti_3SiC_2 was identified by X-ray diffraction.

Fig. 2 shows the X-ray diffraction patterns of samples obtained from the mixture of raw material ingredients of $2TiC + 1Ti + 1Al + 0.2Si$ (moles), i.e., 0.2 moles of Al were substituted by the same amount of Si. Weudged from the X-ray diffraction patterns, for sample sintered at 1150 °C, that the product reached a high purity, since very weak peaks of TiC ($2\theta = 41.82°$) and Al_3Ti ($2\theta = 47.3°$) were identified by X-ray diffraction. For samples sintered at both 1200 and 1250 °C, the products were pure Ti_3AlC_2; no phase but Ti_3AlC_2 was identified by X-ray diffraction. When the temperature reached 1300 °C, the peak of TiC reappeared, which indicates Ti_3AlC_2 has decomposed according to the reaction $Ti_3AlC_2 \rightarrow TiC + Al\uparrow$

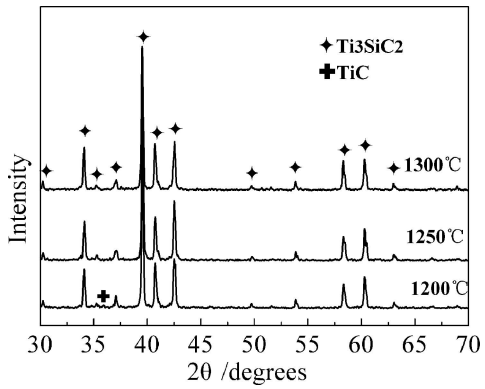


Figure 1 XRD patterns of samples from the starting raw materials made of $2TiC + 1Ti + 1Si + 0.2Al$ (moles).

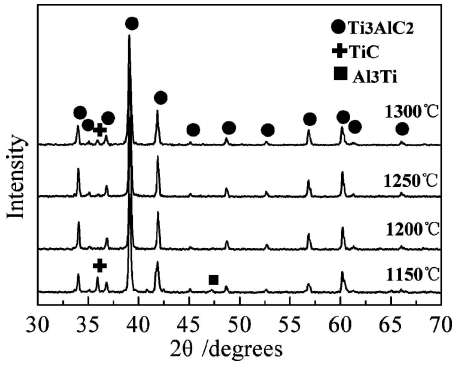
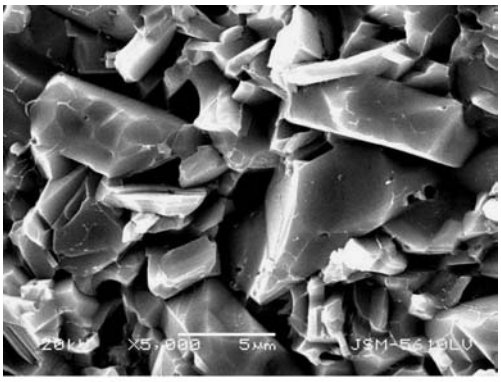
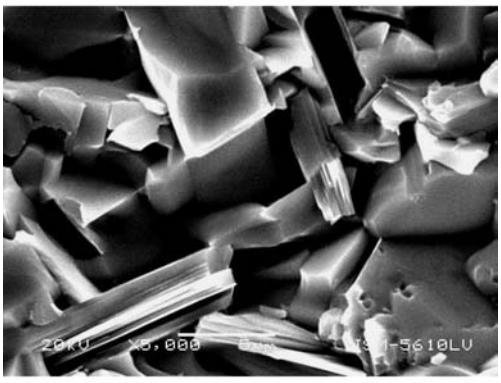


Figure 2 XRD patterns of samples made from the starting raw materials $2TiC + 1Ti + 1Al + 0.2Si$ (moles).



(a)



(b)

Figure 3 SEM micrographs of the fracture surfaces of: (a) Ti_3SiC_2 and (b) Ti_3AlC_2 synthesized at 1250 °C.

Fig. 3 shows the scanning electron micrographs of the fracture surfaces of Ti_3SiC_2 and Ti_3AlC_2 materials synthesized at 1250 °C. The grains of both Ti_3SiC_2 and Ti_3AlC_2 were plate-like. Ti_3SiC_2 grains are 2–10 μm in the elongated dimension, while Ti_3AlC_2 grains are 5–20 μm.

The densities of Ti_3SiC_2 and Ti_3AlC_2 materials prepared at 1250 °C were measured to be 4.47 and 4.21 g/cm^3, i.e., 98.7 and 99.1% of their theory densities, respectively.

It is concluded that polycrystalline bulk Ti_3SiC_2 and Ti_3AlC_2 materials with high purity and density can be fabricated by spark plasma sintering.

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Received 17 February
and accepted 10 August 2004