



Synthesis of high purity titanium silicon carbide from elemental powders using arc melting method



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ABSTRACT

The objective of this study is to investigate the formation of Ti_3SiC_2 from Ti/Si/C powders using the arc melting method. The results show that the sample sintered at 80 s produced a near single-phase of Ti_3SiC_2 (99.2 wt.%) with a relative density of 88.9%. These results were confirmed by phase determination using XRD analysis and were supported with micrographs from FESEM/EDX analyses. The relative density and porosity of all samples were dependent on the formation of macropores in bulk samples and micropores in TiC_x grains. The proposed reaction mechanisms for the synthesis of Ti_3SiC_2 by arc melting is that Ti_3SiC_2 might be formed from $TiC_x + Si$, $Ti_5Si_3C_x + C$, and $Ti_5Si_3C_x + TiC_x$ at early arcing time (≤ 10 s), while $TiC_x + TiSi_2$ take place at 15 s to 80 s. After 80 s, decomposition of Ti_3SiC_2 into TiC_x , $TiSi_2$ and C was observed.

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Introduction

Recently, chemists and researchers have paid considerable interests in the use of titanium silicon carbide (Ti_3SiC_2) for many applications ranging from chemical, physical, electrical, and mechanical properties [1–12] due to its unique characteristics of combining metal and ceramic performance [13]. Like metals, it has excellent electrical ($\sim 4.5 \times 10^6 \Omega^{-1} m^{-1}$) [6] and thermal ($\sim 37 W/m K$) [5] conductivities, relatively low hardness (~ 4 GPa) [7], high Young modulus (~ 325 GPa) [3], not susceptible to thermal shock and damage tolerance at high temperatures [5,12]. It also duplicates several ceramic properties such as good oxidation resistance of up to 1400 °C [4], relatively low density ($\sim 4.52 g/cm^3$) [7], high melting temperature up to 3000 °C [14]. Moreover, porous structure of this material was found to have better damping capacity and excellent dissipating factor of heat than its fully dense components [15]. More detailed descriptions of Ti_3SiC_2 properties have been given by Barsoum et al. [1–7]. A near single-phase Ti_3SiC_2 can be synthesized through several routes (thermal-induction, thermal-combustion, thermal-propagation, electro-discharge) using different starting reactants (Ti, Si, C, SiC, TiC) and sintering aids (Al, Ni, etc.). In most cases, the synthesis of pure Ti_3SiC_2 using thermal-induction route requires high sintering temperatures (1200–1700 °C) for long durations of sintering time (2–100 h), thereby leading to a very expensive process.

For this very reason, most researchers nowadays are focusing on thermal-combustion syntheses such as reactive sintering, thermal explosion (TE), self-propagating high-temperature synthesis (SHS), equipped with hot isostatic pressing (SHS-HIP) [14,16–25]. This is due to their capability of yielding high purity dense Ti_3SiC_2 in a very short time (0.1–100 s). Compared to conventional thermal-induction syntheses, thermal-combustion synthesis that provides more time saving with higher energy efficiency due to heat movement through a reactant is predominantly in the propagation mode, and utilizes extra exothermic reaction in the reactant. Apart from these synthesis routes, electro-discharge synthesis where heat is localized by the electro-discharge of current through the reactants has become a highly potential contender to replace the existing thermal-combustion route because it offers extra benefits such as lower cost and fewer equipment preparations. Unfortunately, the use of electro-discharge synthesis (e.g. arc melting, pulse discharge sintering (PDS), spark plasma sintering (SPS), plasma activated sintering (PAS)) [11,26–32] and elemental reactants without post-annealed products has shown very limited success and the content of the Ti_3SiC_2 produced has never exceeded 60 wt.% [17,24,26]. Moreover, to the best of our knowledge, there is very little available literature on the synthesis of pure Ti_3SiC_2 using arc melting technique [26] and the governing reaction mechanisms among particles when subjected to electro-discharge of current remain unresolved.

Therefore, the objectives of this study are to synthesize pure Ti_3SiC_2 by the arc melting method using Ti/Si/C powders and to attain a better understanding of the reaction mechanisms that take place in the Ti–Si–C system when exposed to the arcing process where the heat produced is beyond Ti–Si eutectic temperature. Raw reactant materials containing a small amount of excess-Si (off-stoichiometry formulation) were used with the aim to restrict TiC formation by creating a bigger

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volume of melt Si-rich liquid phase during reaction. This is essential to maximize Ti_3SiC_2 crystallisation.

Experimental details

Powders of titanium (99.99% purity, average particle size (d_{50}) = 40 μm), silicon (99.9% purity, d_{50} = 2 μm) and graphite (99.9% purity, d_{50} = 2 μm) from Sigma-Aldrich were used as the raw materials. Based on our previous work [33], the molar ratio of Ti/Si/C was selected to be 3:1.2:2 (slightly off-stoichiometry formulation). This ratio has produced a near single-phase Ti_3SiC_2 . The mixture was dry-mixed using a heavy-duty high speed mixer (Fast Mill FM-2) with zirconia (ZrO_2) balls at 10:1 ratio for 1 h. It was then compacted into cylindrical pellet having a diameter of 13 mm and a height of 10 mm under uniaxial pressure at 150 MPa. The green compact density was 60% of the theoretical density. The cylindrical pellets were arced by an arc melting machine (UNI-ARC DC inverter TIG welder model 200C) equipped with non-consumable thoriated-tungsten electrode alloys (EWTh-2, diameter: 2.4 mm) at a fixed of arcing current (30 A) and voltage (30.54 V) for various schedules of arcing time (5–120 s), shielded by high-purity argon gas (99.99%) at 7 l/min in the enclosed copper crucible. The axial direction of the arc was set perpendicular to the cylindrical pellet surface, where the distance of the end-tip tungsten electrode to the cylindrical pellet surface was fixed at 5 mm. It is noted that one of electrode-end was connected to the anode terminal of arc melting machine (known as the direct electrode negative), resulted to ~30% of heat is concentrated on the other end of electrode (cathode terminal) and ~70% of heat is concentrated on the surface of compacted pellet. The actual temperature of the sample was recorded at a fixed point (approximately 5 mm distance from pellet surface, denoted as X) using an infrared thermometer (high-temperature handheld infrared thermometer model OmegaScope OS524E, 538–2482 °C) at selected duration of arcing time. Schematic diagram of arcing sample with the location X is illustrated in Fig. 1.

The arced samples were cut in a straight line from the top (at the centre point of circle) to the bottom of the surface using a diamond cutter before the surface was ground using abrasive SiC paper, from 100 grits to 2000 grits. It was then polished with 0.5 μm alumina (Al_2O_3) powder. Phase identification of the surface of arced sample (taken approximately at 5 mm distance from top polished sample) was analysed by an X-ray diffractometer (XRD model Bruker AXS D8 Advance) with $CuK\alpha$ radiation operating at 40 kV, 30 mA, at a scanning speed of $0.02^\circ s^{-1}$ and the $2\theta = 10-90^\circ$. The qualitative and quantitative analyses of these XRD profiles were performed by the calibrated standard automatic-mode method using Rietveld refinement of PANalytical X'Pert Highscore Plus software. The weighted residual profile (R_{wp}) and goodness of fit (GOF) of XRD profiles were recorded as indicators for refinement quality level. It is noted that the value of R_{wp} should be

below 20% while GOF value should be below 4 for good-quality refinement process. Many macropores were observed in the combustion samples, comparable in literatures [21,22]; therefore, microstructures were observed onto two sites (taken at the same location as observed by XRD analysis); dense-site (to observe phase texture) and pore-site (to observe phase growth) using a field emission scanning electron microscope (FESEM model Supra 35VP Zeiss) equipped with an energy-dispersive spectroscopy (EDS) system. The theoretical density of the samples was calculated from the crystal structure and chemical composition of phases as observed from the XRD profile's results. Finally, relative density and porosity of the arced samples were calculated according to the Archimedes principle.

Results and discussion

Measurement of arcing temperature

A time-temperature plot of location X as recorded by the infrared thermometer is shown in Fig. 2. Once the welder machine was switched on, the semi arc flame is clearly seen on the sample. The propagation of heat through a sample can be seen on the changes of colour appearance, where the bright yellow (flame front) moved downward along the reactant (black). To support heat propagation, the recorded temperature of location X is increased with an increase of time, up to 15 s. This finding is in agreement with the previous reports made by Yeh et al. [21,22], where the flame front of heat propagation moved downward through a reactant within a few seconds of combustion time. After arcing time was prolonged from 15 to 120 s, the arcing temperature decreased gradually to 1904 °C, due to small radiation and convection heat transfer to its surroundings. Therefore, it is safe to suggest that the actual temperature of arcing process is ~1900 °C. This value is in the range of combustion temperature used to synthesis Ti_3SiC_2 as reported by Lis et al. [19]. After welder machine switched off at the longest duration of 120 s, the arcing temperature was dropped significantly from ~1900 to 112 °C, within 80 s.

Physical appearance

The physical appearance of pellet and arced sample is shown in Fig. 3. It is shown that the size and dimension of arced samples are quite similar with compacted pellet. However, there are significantly slight of expansion in pellet thickness (axial direction), and shrinkage in pellet diameter (radial direction). Observation on the surface of arced sample indicates that the reactant is completely in molten (approximately 0.2 to 1 mm depth), followed with the formation of laminate layers to the bottom. Many macropores were observed at the interfaced of these lamella layers, due to the explosion of air/gas pockets

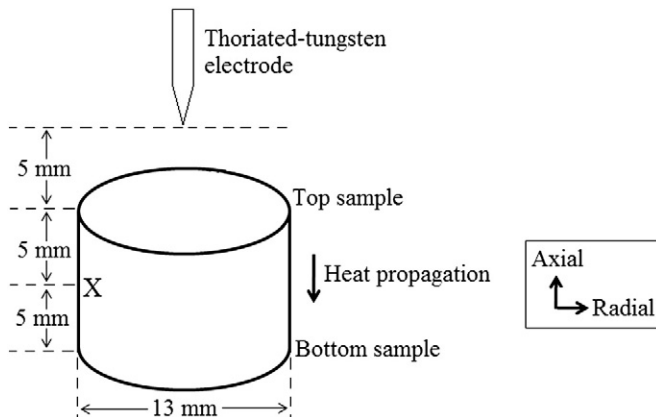


Fig. 1. Schematic diagram of arcing sample.

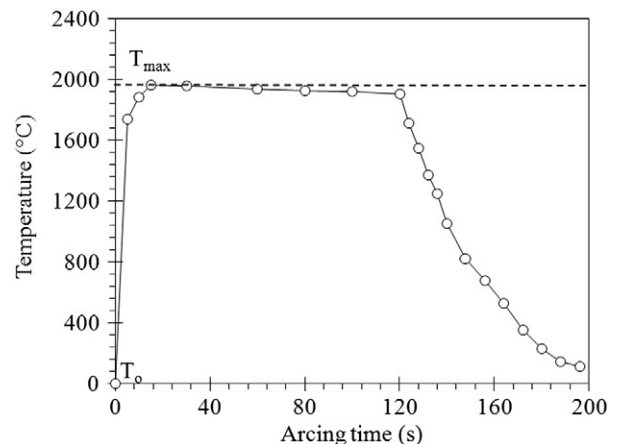


Fig. 2. Arcing temperature profiles of location X as a function of arcing time.

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