Journal of Metallurgical and Materials Engineering, Vol.30, No.1, 2019.

Fabrication of TiB_2-TiC Composite through Microwave Assisted Self-propagating High-Temperature Synthesis Method

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1- Introduction

TiB_2-TiC composite has excellent properties such as high melting point, excellent abrasion resistance, high hardness, good thermal stability and high corrosion resistance. Hence, this composite has been used in various applications such as abrasion resistant parts, cutting tools, forming molds, lightweight parts, jet engines and heat exchangers.

TiB_2-TiC composite is usually produced by powder metallurgy (PM) method. This method requires high-temperatures and long heating times and therefore is costly. Recently, use of microwave assisted self-propagating high-temperature synthesis (MASHS) has been proposed for the production of TiB_2-TiC composite. Some of the benefits of this method are: high production rates, simplicity, low energy consumption and product purity.

It has been shown that the TiB_2-TiC composite can be synthesized from a Ti-B_4C mixture and in a microwave oven. Increasing the mixing time of the reagents eliminated the formation of the unwanted products. It also prevented the synthesis of the intermediate compounds in the final powder and shortened the synthesis time. The main drawback of this route was the high-price of the reagents.

In this research, the fabrication of TiB_2-TiC composite via MASHS method and by use of low-cost raw materials (i.e. oxides compounds) has been studied. Synthesis was performed in a domestic microwave oven. Moreover, thermodynamic calculations were employed in order to predict the type of possible reactions in the system.

2- Experimental

In this study, an inexpensive route was proposed for the fabrication of TiB_2-TiC composite powder. Raw materials (i.e. TiO_2, B_2O_3, Mg and C) were mixed by hand and pressed into cylindrical samples (10 mm diameter and 4 mm height). Microwave heating was performed using a domestic microwave oven (SAMSUNG: GE2370G, 850 watts output power). During the microwave heating, the ignition and sparking of the samples were considered as the signs for the occurrence of a combustion reaction.

Phase compositions of the heated samples were determined using an XRD device. Moreover, an SEM (equipped with EDX detector) machine was employed for the observation of the microstructure and morphology of the samples.

In order to obtain a pure TiB_2-TiC powder, the byproduct of the reactions (i.e. MgO phase) was leached out using 20 vol. % HCl solution. Temperature and time of the acid leaching process were 50° C and 2 hours, respectively.

In all parts of this research, ΔG°, ΔH° and T_{ad} (i.e. adiabatic temperature) values of the probable reactions were calculated and used, in order to get an insight into the system and predict the type of the reactions.

3- Results and Discussion

Thermodynamic calculations: The overall reaction in the TiO_2-B_2O_3-Mg-C system can be written as follows:

$$2TiO_2 + B_2O_3 + C + 7Mg = TiB_2 + TiC + 7MgO \quad (1)$$

In fact, reaction (1) is the combination of the reduction reactions (reactions (2) and (3)), the reaction between the reduced elements (reaction (4)) and the reaction in the Ti-C system (reaction (5)).

$$TiO_2 + 2Mg = Ti + 2MgO \quad (2)$$

$$B_2O_3 + 3Mg = 2B + 3MgO \quad (3)$$

$$Ti + 2B = TiB_2 \quad (4)$$

$$Ti + C = TiC \quad (5)$$

Thermodynamic results showed that reactions (2) to (5) have negative ΔG° values. This indicates the tendency for the reactions progress. In addition, these reactions are very exothermic (i.e. $\Delta H^\circ < 0$). Therefore, it is expected that the temperature of the system will be increased during the reactions. Our calculations showed that the adiabatic temperature (T_{ad}) of all reactions are above 1800 K. Thus, according to the Marzhanov's criterion, the type of reactions (2) to (5) will be SHS.

Further investigations revealed that the chemical reactions in the TiO_2-B_2O_3-Mg-C mixture most probably initiate with reaction (3). Subsequently, the heat released from reaction (3) activates reactions (2), (4) and (5) which forms the TiB_2-TiC-MgO composite (reaction (1)). ΔG°, ΔH° and T_{ad} curves of the reaction (1) are presented in Fig. 1.

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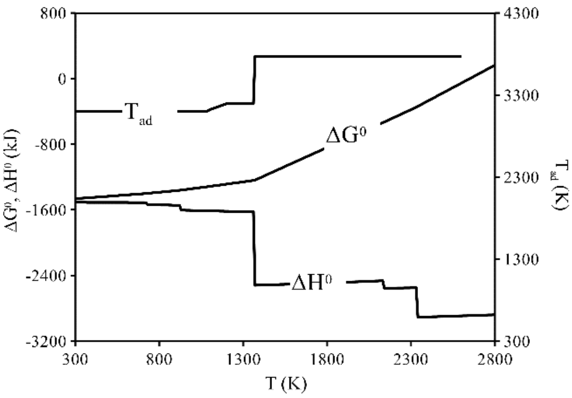

Fig. 1 ΔG°, ΔH° and T_{ad} curves of the formation reaction of TiB_2-TiC-MgO composite powder in the TiO_2-B_2O_3-Mg-C mixture (reaction (1))

Experimental results:

TiO_2-B_2O_3-Mg system: Experimental observations showed that an SHS reaction occurs during the microwave heating of TiO_2-B_2O_3-Mg mixture. XRD results showed that the product contains some amounts of unwanted phases (such as $Mg_3B_2O_6$). In order to eliminate the unwanted phases, Mg amount in the system was increased. It was found that a pure TiB_2-MgO composite powder can be obtained using TiO_2:B_2O_3:7Mg mixture.

TiO_2-Mg-C system: The results of this system revealed that the TiC-MgO composite powder (free from unwanted phases) can easily be synthesized from a TiO_2:2Mg:C mixture and via an SHS reaction activated by microwave heating.

TiO_2-B_2O_3-Mg-C system: Regarding the optimum amounts of Mg for the TiO_2-B_2O_3-Mg and TiO_2-Mg-C mixtures, TiB_2-TiC-MgO composite can be synthesized using a 2TiO_2:B_2O_3:9Mg:C mixture. In agreement with the thermodynamic predictions, microwave heating of the 2TiO_2:B_2O_3:9Mg:C mixture brought about the occurrence of an SHS reaction in the system. As Fig. 2A shows, the product of the SHS reaction was merely consisted of TiB_2, TiC and MgO phases.

The product was treated by a diluted HCl solution, which could dissolve the MgO phase and produce a pure TiB_2-TiC composite. The XRD result is shown in Fig. 2B and confirms the successful removal of MgO from the product.

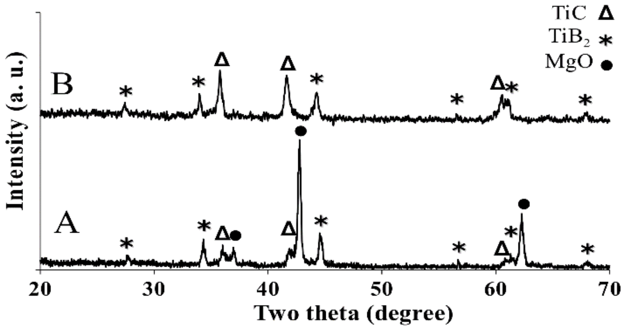

Fig. 2 XRD patterns of 2TiO_2:B_2O_3:9Mg:C mixture A) after microwave heating and B) after acid leaching

SEM investigations of the synthesized TiB_2-TiC-MgO powder showed the formation of large and dense agglomerates which was the sign of system high-temperatures. MgO (which forms a melt at high-temperatures) acted as an adhesive and connected the TiB_2-TiC particles.

It was found that the acid leaching of the MgO phase decreases the particles size of the product and yields a product with 100-150 nm average particle size. Fig. 3 represents the SEM image of the obtained TiB_2-TiC powder.

4- Conclusions

- Thermodynamic predictions and experimental results confirmed the occurrence of combustion reactions in the TiO_2-B_2O_3-Mg-C system as well as its sub-systems.

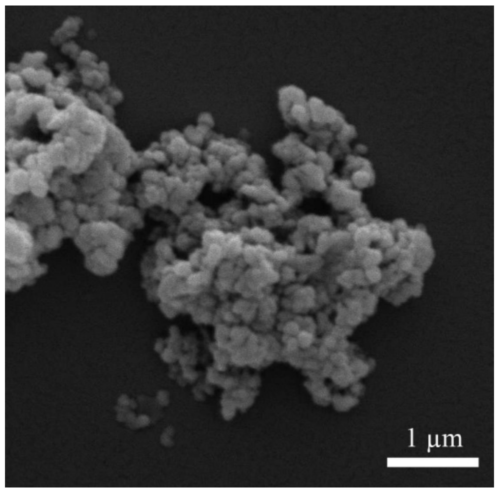

Fig. 3 SEM image of the synthesized TiB_2-TiC powder

- TiB_2-TiC-MgO composite was obtained by microwave heating of a 2TiO_2:B_2O_3:9Mg:C mixture.
- Acid leaching was found effective for the purification of the synthesized powder.
- The average particle size of the synthesized TiB_2-TiC powder was 100-150 nm.