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EFFECT OF PROCESS PARAMETERS ON THE PRODUCTION OF Ti_3SiC_2 VIA MODIFIED SHS THROUGH ARC MELTING

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Abstract—Ti_3SiC_2 was synthesized by modified-SHS system through arc melting. Elemental powder of titanium, silicon, and graphite were weighed according to the stoichiometric ratio of Ti:Si:C = 3:1:2. The powders were mixed and milled by high speed mill for 1 hour. 120 MPa of pressure is used for the compaction. The samples were sent for arc melting process and the effect of arc melting duration (60s, 75s and 90s) was studied. The welding current was varied (70A, 100A, and 130A) to get the best parameters to produce Ti_3SiC_2 in high composition. The samples were sent for XRD analysis and SEM observation. For arc melting, the best composition of Ti_3SiC_2 was produced with 100A welding current for 90s instead of impurity TiC formation. Hence, Ti_3SiC_2 can be produced via modified-SHS through arc melting with mixing duration of 1 hour using high speed mill, and arc melting duration of 90s with weld current of 100A.

Keywords: Ti_3SiC_2 ,SHS, arc melting

I. INTRODUCTION

THE Ti_3SiC_2 was firstly synthesized via chemical reaction in 1967, and then by chemical vapor deposition in 1987. Titanium silicon carbide (Ti_3SiC_2) has the crystal structure of which is comprised of hexagonal nets of Si atoms separated by three nearly closed-packed Ti layers that accommodate C atoms in the octahedral sites between them. Its room-temperature electrical and thermal conductivities are excellent, being about $4.5 \times 10^{-6}\ \Omega^{-1}\ m^{-1}$ and 37 W/mK, respectively. It is relatively soft (the Vikers hardness of which is about 4 GPa), damage tolerant and resistant to thermal shock, and easily machinable. Furthermore, its density is relatively low (4.5 g/cm^3) and it is stable up to at least 1700 °C in inert atmospheres and vacuum [1].

Conventional production method often involves various pretreatments, for example, hot uniaxial pressing, hot isostatic pressing and chemical vapor deposition. These processes feature the use of strong external heating to accelerate processing and hence, are regarded as energy intensive technologies [2].

Combustion synthesis, or self-propagating high temperature synthesis (SHS) provides an attractive practical alternative to the conventional methods of producing advanced materials, such as ceramics, ceramic composites and intermetallic compounds, since SHS offers advantages with respect to process economics and process simplicity. The underlying basis of SHS relies on the ability of highly exothermic reactions to be self-sustaining and, therefore, energetically efficient [3, 4].

However so far there is no published report of production of titanium silicon carbide via modified SHS through arc melting using a TIG arc welding machine.

II. EXPERIMENTAL PROCEDURE

The raw materials used in this research are titanium, silicon, and graphite. All of the raw materials are in the form of powders. The elemental powders are prepared based on the stoichiometric ratio of Ti : Si : C to 3:1:2. High speed mill is used for the mixing and milling process. The grinding media used in this study are zirconia balls. This is due to the higher hardness of zirconia compared to titanium. The ball-to-mass ratio used is 10:1. After mixing and milling process, the mixed powder will be compacted at 120 MPa into pellets. A mold with 13mm diameter is used, together with a hydraulic press. The compacted mixture in pellet form will be taken for combustion process. In this process the TIG arc welding machine is used. Argon gas is flowed to the chamber of crucible to provide inert atmosphere that does not react during the

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combustion process. Water supply also will be flowed around the crucible to help in cooling the chamber and surrounding. The electrode of the TIG arc welding machine is made from tungsten. When the combustion process is done the combusted pellet will be taken out and allowed to cool down before it is taken for characterizations. During the combustion process, two parameters were varied. The first parameter in this research is the combustion time. The pellets will be combusted for 60, 75 and 90 seconds in the TIG arc welding machine. The next parameter in this research is the welding current. The pellets will be combusted at current of 70A, 100A and 130A. The characterizations used for this research is X-ray diffraction analysis (XRD).

Figure 1: Appearance of sample before and after arc melting

Figure 2: Appearance of sample that was cut

I. RESULTS AND DISCUSSION

Figure 1 shows the appearances of the samples after the process of modified-SHS through arc melting. The shapes had been deformed significantly from the original shape after pressing process which is in compacted pellet form. During the arc melting process, it could be seen that there was presence of liquid phase. When heat was applied, the pellet would at first be seen at red colour due to heat and then liquid phase could be seen. In Ti–Si binary system there are two eutectic reactions for the Si–$TiSi_2$ and Ti-Ti_5Si_3 compositions both at the temperature of 1330°C [5]. Therefore it can be deduced from the appearance that the synthesis process must had exceeded 1330°C.

Figure 2 shows the appearances of example of sample that were cut. . From the figure it can be seen that the sample is very porous after arc melting process. Porous products from self- propagating high-temperature synthesis are one the major difficulties of SHS [6].

From figure 3, for the sample that is combusted for 60 seconds, it is majorly consisted of TiC and $TiSi_2$. There is only a little trace of Ti_3SiC_2. This means that Ti_3SiC_2 is just starting to form. Li et al. [7], in their study, found that impurities like $TiSi_2$ will occur in samples that are sintered at low temperature or/and at short time. The reaction to yield Ti_3SiC_2 is said to be mass-diffusion and time-consuming process. $TiSi_2$ might be a phase separating out from the eutectic Si-abundant liquid phase. Sun et al. [8] found that amount of $TiSi_2$ will increase with the increasing of amount of Si in the starting material. TiC is expected to be one of the impurities. Sun et al. [8] also mentioned that Ti_5Si_3, TiC, and silicon are prerequisite reactants for the formation of Ti_3SiC_2 through the following reaction

$$Ti_5Si_3 + TiC + Si \longrightarrow Ti_3SiC_2 \quad (1)$$

As the combustion time increase to 75 seconds and 90 seconds, the relative intensities of the peaks for Ti_3SiC_2 increases. The longer time allow the formation of Ti_3SiC_2.

The samples were combusted at three different weld current. The currents are 70 Ampere, 100 Ampere, and 130 Ampere. Weld current is chosen to be varied because as the current changes, the temperature given by the electrode changes. As weld current is increased, the temperatures dramatically increased [9]. Therefore in this research, varying the current is a mean of varying the temperature during the arc melting process. The combustion time of 90 seconds is used as it gives the best result as previously discussed. From figure 4, Ti_3SiC_2 is present in all samples but the intensity increases when weld current is increased from 70A to 100A, but the intensity drops when the weld current is increased to 130A. In the study of Zou et al. [5], they compared the results of the samples sintered at two temperatures of 1300 °C and 1350 °C and it was found that no single-phase Ti_3SiC_2 was obtained when sintered at 1300 °C even the sintering time was prolonged from 20 min to 120 minutes. In contrast, single-phase Ti_3SiC_2 was available when the sintering temperature was increased for 50 °C to 1350 °C in this narrow temperature range, with a short sintering time of 20 minutes. The mentioned temperature of 1350 °C is very important as in Ti–Si binary system there are two eutectic reactions for the Si–$TiSi_2$ and Ti–Ti_5Si_3 compositions both at the temperature of 1330 °C. When the weld current increases from 100A to 130A, the intensities of the peaks for Ti_3SiC_2 decrease. This may due to the increase of temperature and hence the decomposition of Ti_3SiC_2. Gao et al. [10] reported that Ti_3SiC_2 decomposed into Si and TiC in most furnace atmospheres at temperatures above 1350˚C. The partial pressure of Si in the furnace atmosphere was crucial for maintaining the stability of Ti_3SiC_2. Besides, with increase in temperature, there could be high possibilities of losses of silicon through vaporization. As stated by Sato et al. [11] and Pampuch [12], the evaporative losses of Si not only prevent the formation of Ti_3SiC_2 but increase the impurities like TiC. Common impurities like TiC and Ti_5Si_3 is present in samples 70A and 130A. However in sample 100A there is only TiC. Hence with the composition of only Ti_3SiC_2 and TiC in the sample 100A, it can be said that by using TIG arc welding machine, 100A is best to be used for SHS through arc melting of Ti_3SiC_2.

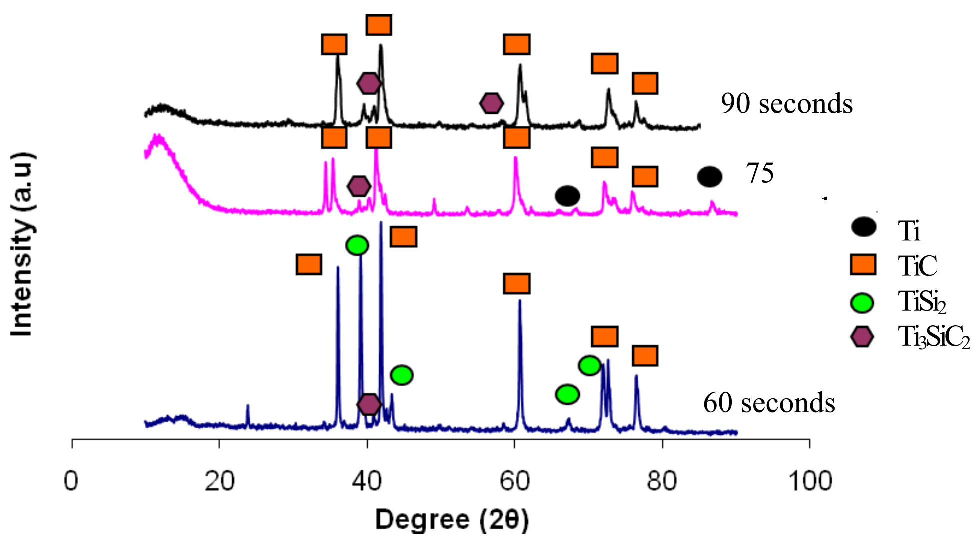

Figure 3: Comparison of XRD pattern for samples with combustion time of 60 seconds, 75 seconds, and 90 seconds

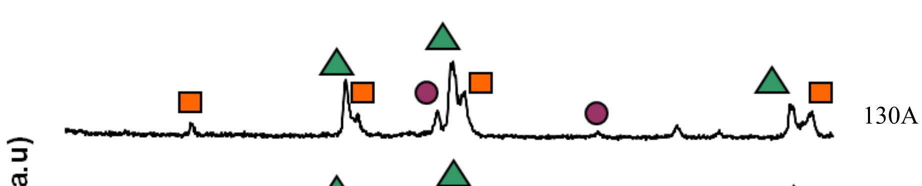

Figure 4: Comparison of XRD patter for samples arc melted with weld current of 70A, 100A and 130A

I. CONCLUSION

Ti_3SiC_2 can be synthesized in a very short time through arc melting method. The function of arc melting was to provide ignition temperature for SHS system to synthesis Ti_3SiC_2. However, it is not known to which extent that arc melting resembles the SHS system. Even though Ti_3SiC_2 was able to be synthesized via modified-SHS through arc melting in a very short time, it was found that there impurities such as Ti_5Si_3 and TiC always existed together with the product. The samples after arc melting always have high porosity. In this study, various process parameters were investigated to yield the best result for the synthesis of Ti_3SiC_2. In conclusion, high composition of Ti_3SiC_2 can be produced via modified-SHS through arc melting with mixing duration of 1 hour using high speed mill, and arc melting duration of 90s with weld current of 100A.

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