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THE MICROSTRUCTURE, PHYSICAL AND MECHANICAL PROPERTIES OF SILICON CARBIDE WITH ALUMINA AND YTTRIA AS SINTERING ADDITIVES

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ABSTRACT

Silicon carbide has gain much attention in recent years as the best material for the application in harsh environment condition. This is due to their excellent properties such as good wear resistance with high hardness and strength at elevated temperature. In this study, the sintered specimen of silicon carbide with sintering additives of four different compositions were fabricated using powder metallurgy method. Physical properties were measured by means of bulk density and apparent porosity whereas the mechanical property was measured in term of hardness. From this study, it can be observed that the silicon carbide was sintered to about 0.93 of its relative density by using alumina and yttria as sintering additives. This finding was as a result of a eutectic liquid formed between alumina and yttria at sintering temperature of 1930°C. The specimen with 25 wt. % of sintering additives gave the maximum value of hardness of 2601 H_V.

Keywords: silicon carbide, ceramics, sintering, microstructure, mechanical properties.

INTRODUCTION

For decades, silicon carbide (SiC) has been known to have outstanding mechanical properties including high strength, stiffness, good resistance to wear and corrosion. The material is composed of one carbon atom and one silicon atom which is bonded together by a strong chemical bond in the crystal lattice. This results in a very hard and strong material. In addition, the existence of the strong covalent bond between the atoms produces a material that is extremely stable in various range of temperatures and chemical environments [1]. The chemical and physical properties of SiC have made it an exceptional material for the applications in high temperature environment [2]. These properties comprised of excellent corrosion resistance and high strength at elevated temperature [3-5].

However, as a ceramic material, SiC is brittle and its mechanical strength is considered to be less than that of other metal carbides. Aside from that, SiC is susceptible to chipping and fracture when introduced to large mechanical stress or shock [6]. Another disadvantage of SiC is that the material is difficult to be manufactured in any required shapes especially for component design for certain application [1]. Moreover, it is difficult to densify the SiC without the use of suitable additives and external pressure [4]. This is mainly due to the covalent nature of silicon-carbon (Si-C) bonding together with its low self-diffusion coefficient.

Although it is not an easy task to attain the densified SiC, addition of sintering additives has provided tremendous help in obtaining the aforementioned material with desired properties particularly the density. There are many types of sintering aids that can be used and each has their own advantages and disadvantages on the final specimen. The main reason for using such aids is to help in reducing the sintering time and temperature [7-11]. Among common sintering aids to obtain a dense silicon carbide are aluminium oxide (Al_2O_3), yttrium oxide (Y_2O_3) and boron oxide (B_2O_3).

As experimentally investigated by Omori and Takei [8], β-SiC could be densified by sintering at the temperature of 2100 ˚C using yttrium and aluminium hydroxide. The additives in the later stage reacted with each other and formed oxides, which subsequently facilitated densification process via liquid-phase sintering. The liquid phase which is formed between the reactants has lowered the sintering temperature sintering, and on the other hand, can help improving the microstructure of SiC during the sintering or heat treatment [9-12].

Besides utilizing the sintering aids, several factors have been acknowledged to influence the final product of the sintered SiC. One of them is the starting raw material where very fine SiC powders offers possibility for densification to occur at the lowest possible temperature. The grain type of silicon powders also affects to the refinement of final specimen's microstructures and in turn attainment of the desired mechanical properties [13-14]. Irrespective of substantial microstructural complication, there is a comprehensive agreement that the densification of liquid phase sintered structures proceeds fundamentally in three stages, as shown in Figure-1.

Sintering process usually takes about 10 to 60 minutes to be completed. However, it is found that sintering time did not provide any significant impact on the properties and microstructure [2]. Meanwhile, during hot pressing process, the atmosphere must be inert to SiC. Hence, the process cannot be conducted in any oxidizing atmospheres such as air since conversion into silica through oxidation process tends to occur in these kinds of environments. Subsequently, it will interfere with the sintering process and alter the properties of the specimen.

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Such examples of inert atmospheres include the using of argon, helium, and nitrogen gases [16-17].

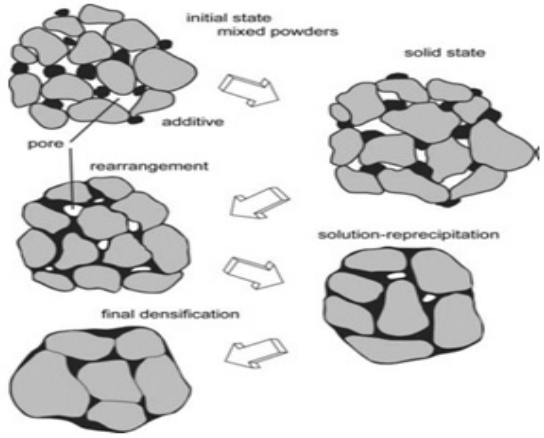
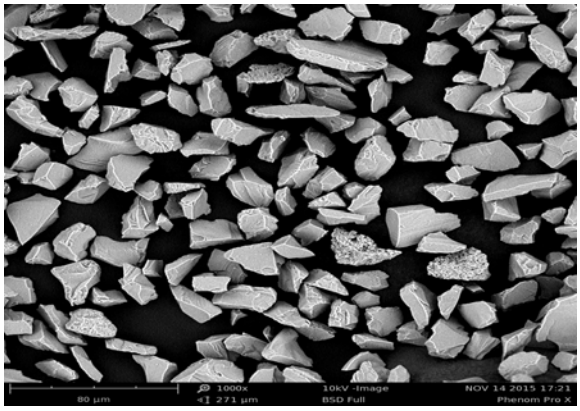


Figure-1. The theoretical steps to liquid phase sintering using binary powders [15].

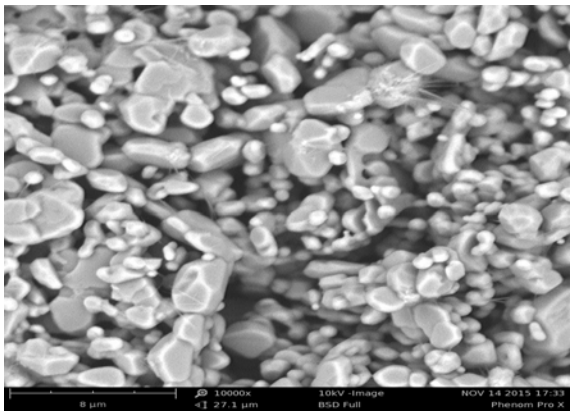
In this study, effect of sintering additives namely Al_2O_3 and Y_2O_3 with different composition on the properties of sintered SiC was investigated. In addition to the microstructure, physical and mechanical properties of the sintered specimen were then examined and discussed.

EXPERIMENTAL METHODOLOGY

The SiC specimens were produced from the homogenously blended of β-SiC, Al_2O_3 and Y_2O_3 powders. These powders were purchased from Merck Chemical Ltd where their morphology can be viewed in Figure-2 (a), (b) and (c) respectively.



(a)



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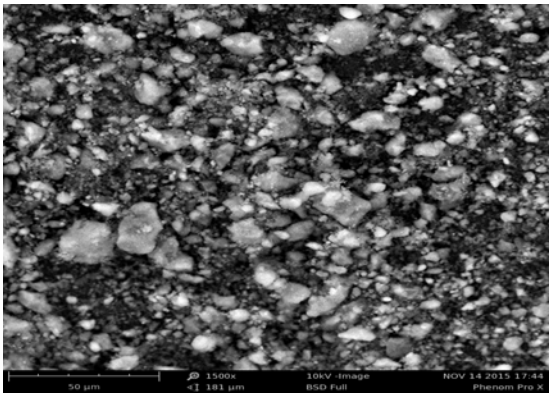

(c)

Figure-2. Powders used in the study: (a) SiC powder (b) Al_2O_3 powder (c) Y_2O_3 powder.

Four different composition of the specimens were prepared as presented in the Table 1. In this study, the sintering additives composition were fixed at 64.4 wt.% and 35.6 wt.% for the respective Al_2O_3 and Y_2O_3.

Table-1. Composition of specimen.

Specimen	Composition (wt.%)	
	SiC	Sintering Additives (Al_2O_3-Y_2O_3)
1	95	5
2	90	10
3	85	15
4	75	25

The homogenously blended powders were compacted into pellets with diameter of 20 mm and thickness of 10 mm using hand press machine at pressure of 200 MPa. The specimens were then sintered at 1930°C for 60 min in Argon gas atmosphere.

The grinded and polished specimens were etched using Murakami's Reagent. The revealed microstructure of the specimens was observed and captured by PHENOM Pro X scanning electron microscope. The elemental

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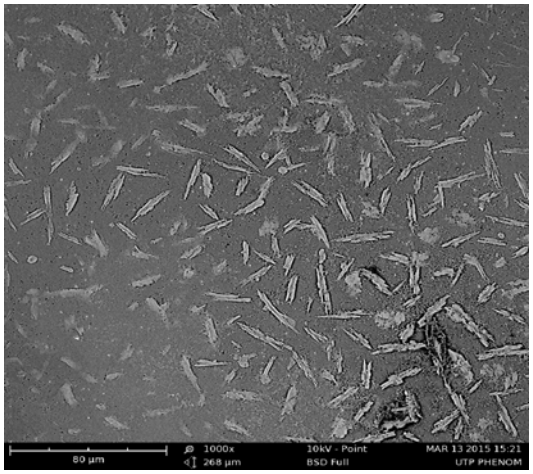
compositions of the microstructure were determined using Energy Dispersive X-Ray (EDX) spectroscopy.

Archimedes method was utilised in order to measure the density of the specimens. Meanwhile in terms of relative density (RD), it was estimated based on the mixture theory.

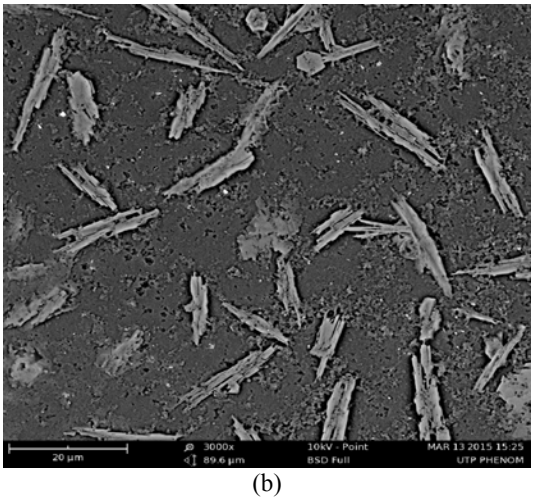
As for the hardness of the specimens, micro Vickers hardness test (ASTM C1327) had been employed. It utilized 1 kgf of test load and the indentation on the polished surface was carried out for 15 s of dwell time.

RESULTS AND DISCUSSION

Figure-3 shows the electron micrograph of sintered SiC-25 wt. % (Al_2O_3-Y_2O_3) which consists of flaky shape particles that is homogenously distributed in the microstructure. The composition of flaky shape particles (point 1 of Figure-4) which was characterized by EDX analysis indicates the presence of Si, Al, Y and O element. Figure-5 shows the energy dispersive spectrum of the particles and its chemical composition is tabulated in Table-2.



(a)



(b)

Figure-3. Electron micrographs of sintered SiC-25 wt. % (Al_2O_3-Y_2O_3) at (a) 1000x (b) 3000x.

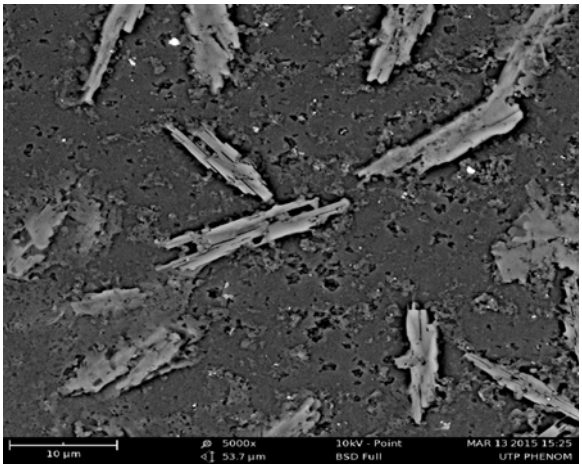


Figure-4. Electron micrographs of sintered SiC-25 wt. % (Al_2O_3-Y_2O_3) at x5000.

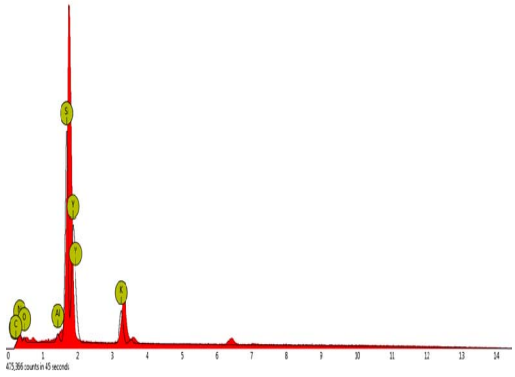


Figure-5. EDX spectrum of flaky shape particle (Point 1).

Table-2. Elemental compositions of flaky shape particle.

Element	Weight %
Y	55.5
Si	22.0
Al	9.2
O	13.3
Total	100

In Table-3, the linear shrinkage of all specimens which have been sintered at 1930 ˚C is presented. The results indicate that the shrinkage for all specimen is within 18.2-19.0 %. It also disclosed an increasing pattern with the increase of additives composition in the mixture. As the content of sintering additives is increased, the formation of liquid phase between Al_2O_3 and Y_2O_3 at sintering temperature of 1930 °C will vacate the SiC grains. This phenomenon leads to the increase of the shrinkage of the specimen.

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Table-3. Variation of linear shrinkage (%) of SiC with different sintering additive compositions.

Specimen	Linear Shrinkage (%)
SiC-5 wt. % (Al_2O_3-Y_2O_3)	18.2
SiC-10 wt. % (Al_2O_3-Y_2O_3)	18.8
SiC-15 wt. % (Al_2O_3-Y_2O_3)	18.9
SiC-25 wt. % (Al_2O_3-Y_2O_3)	19.0

As can been anticipated from Table-4, specimen 4 with the highest sintering additives composition has the highest density with a value of 3.19 gcm^{-3} with 0.93 of its RD. Meanwhile, specimen 1 with the lowest sintering additives content yields a density value of 2.95 gcm^{-3} with 0.90 of its RD. From the given data in Table-3 and Table-4, the sintered density of the specimen is correlated with that of its linear shrinkage behaviour. However, this finding is contradicted with the work of She and Ueno [7]. The densities of all measured specimens are slightly less than 3.21 gcm^{-3} which is the theoretical density of SiC. All RD calculated values are higher than 0.9 of their RD, suggesting that all samples have achieved high densification progression.

Table-4. Variation of density and its RD of SiC with different sintering additives composition.

Specimen	Density (gcm^{-3})	RD
1	3.01	0.90
2	3.05	0.91
3	3.08	0.92
4	3.19	0.93

The density and porosity of the SiC with different sintering additives composition are plotted as depicted by Figure-6. The density of the specimens decreases from 3.01 gcm^{-3} to 3.19 gcm^{-3} when the sintering additives composition of the specimen is increased from 5 to 25%. Meanwhile the value of the porosity of the specimen decreases from 9.70% to 8.25% as the sintering additives composition increase from 5% to 25%. It can be deduced that densities of the specimens are closely related to their porosities.

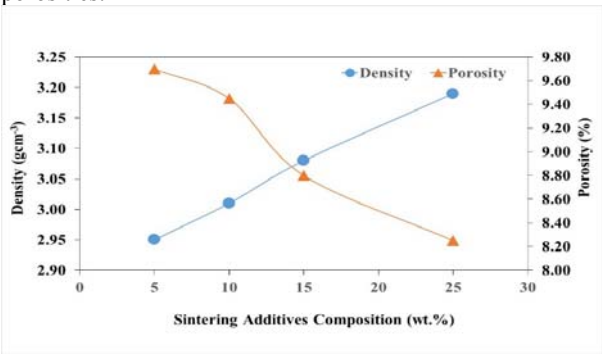

Figure-6. Variation of density and porosity of sintered SiC with different sintering additives composition.

The densification behaviour of the SiC specimens is owing to the formation of liquid phase between Al_2O_3 and Y_2O_3 at their eutectic composition. It is postulated that the solution precipitation process leads to the development of liquid phase sintering process for the SiC specimens with different sintering additives compositions [7]. The formation of pores as shown in Figure-3 (b) that create the presence of porosity in the specimen is caused by the presence of gaseous species such as SiO, Al_2O, YO and CO that formed during sintering process according to following reactions [7-9]:

$$SiC(s) + Al_2O_3(s) \rightarrow SiO(g) + Al_2O(g) + CO(g) \quad (1)$$

$$SiC(s) + 2Y_2O_3(g) \rightarrow SiO(g) + 4YO(g) \quad (2)$$

Hardness test that was conducted on the specimens had resulted in values ranging from 2517 H_V to 2601 H_V as given in Table-5. Therefore, regardless of the different sintering additives content, they produced quite similar hardness. Specimen 4 (SiC-25 wt. % (Al_2O_3-Y_2O_3)) which possess the highest content of additives is the hardest one with hardness of 2601 H_V. Overall, the measured values of all specimens are lower than the published hardness of commercial SiC which is 2804 H_V [1]. The reason behind this discrepancy of hardness can be attributed to the formation of porosity in the sintered specimens.

Table-5. Variation of vickers hardness of SiC with different sintering additive compositions.

Specimen	Vickers hardness (H_V)
SiC-5 wt. % (Al_2O_3-Y_2O_3)	2517
SiC-10 wt. % (Al_2O_3-Y_2O_3)	2539
SiC-15 wt. % (Al_2O_3-Y_2O_3)	2556
SiC-25 wt. % (Al_2O_3-Y_2O_3)	2601

Figure-7 indicates that the specimen with the lowest density has the lowest hardness and vice versa. Thus, the composition of sintering additives used has an influence on the overall hardness and density of the specimen. This can be explained by the fact that a denser specimen is usually more difficult to be indented and hence, indicating a higher hardness.

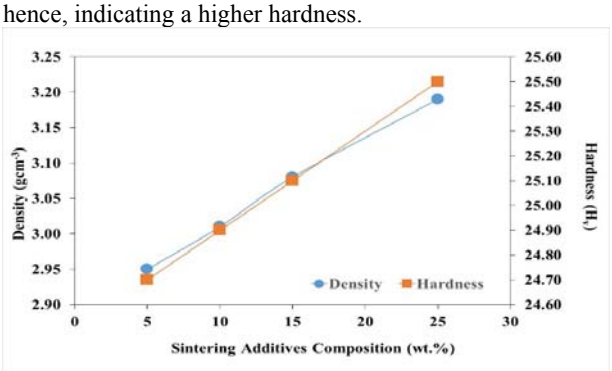

Figure-7. Variation of density and hardness of sintered SiC with different sintering additives composition.

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As the sintering process involves both SiC in solid and Al_2O_3 and Y_2O_3 in liquid phases, the sintered SiC with different additives compositions might have enhanced inter particle bonding resulting in increase of hardness and density values. In this system, the liquid phase from both Al_2O_3 and Y_2O_3 may enhance the mass transport mechanism. The solubility of SiC in Al_2O_3 and Y_2O_3 may develop the formation of intermediate compounds that leads to inter diffusion process. As a result of this occurrences, there is an increase in SiC-Al_2O_3-Y_2O_3 packing density that cause the reduction in pore volume within the specimen.

CONCLUSIONS

SiC with addition of sintering additives were successfully densified through solid-state sintering at the temperature of 1930 ˚C. From the results, it can observe that the specimen with the highest additives content exhibits the highest density and hardness value as compared to others. It can also be concluded that higher additives content could facilitate the better densification rate during sintering process and thus lead to higher physical and mechanical properties value of the produced specimen.

ACKNOWLEDGEMENTS

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