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SILICON CARBIDE FORMATION FROM NATURAL WOODS

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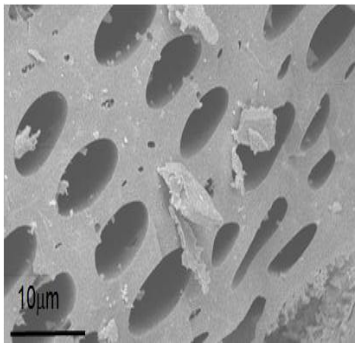
Graphical abstract



Abstract

Processing of cellular ceramics with anisotropic pore structure by using silicon infiltration into carbonized template was investigated. Biomorphic silicon carbide (bioSiC) was produced by using two different types of natural woods which are Kapur and Dark Red Meranti. Carbon template was produced from a pyrolysis process followed by an infiltration process of melting silicon to produce bioSiC. The samples were dried in an oven in order to remove the moisture of the samples. The pyrolysis was done in two stages at a temperature of 500°C followed by 850°C. This study was to investigate the effect of infiltration temperature in the formation of SiC composites. Two different infiltration temperatures of 1500°C and 1600°C were used with constant holding time of 1 hour. The characteristic of the biomorphic silicon carbide was analyzed using the TGA, FESEM and EDX analysis. A wide variety of microstructures, densities and porosities were found depending on the type of wood used. Instead of carbon, it was found that both woods also reacted with nitrogen gas, which reduced the formation of SiC. The density of samples was increased as the working temperature increased. Dark Red Meranti was found to be denser and exhibit higher porosity than Kapur due to the higher formation of SiC.

Keywords: Biomorphic silicon carbide, pyrolysis, infiltration, temperature, porosity

1.0 INTRODUCTION

Silicon carbides have good specific strength, thermal shock resistance and oxidation resistance at high temperature. Most of traditional manufacturing processes such as carbothermal reduction reaction, hot pressing, chemical vapor deposition and reaction bonding process require high processing cost in fabricate silicon carbide. However, biomorphic silicon carbide shows great attraction to utilize biomorphic approaches in order to replace conventional manufacturing process. It is a cost effective process as it requires less energy to manufacture due to the low fabrication temperatures [1]. Besides, a wide variety of microstructures that can be obtained due to varies morphology of the template biomaterials [2]. Biomorphic silicon carbide (bioSiC) utilizes a unique process of fabricate porous ceramics from wood. This process involves pyrolysis of natural wood precursors, followed by the infiltration of silicon to form silicon carbide (SiC), retaining the initial wood structure. There are many potential high-temperature applications for these porous ceramics, including heat exchangers, molten metal filters, catalyst supports, and heating elements [3, 4]. In this research, Carbonous preform of Kapur and Dark Red Meranti were used for infiltration process at different temperatures and holding times. This research is to investigate the microstructure, densities and porosities under two different temperatures of infiltration process.

2.0 EXPERIMENTAL

Dark Red Meranti and Kapur wood were chosen as precursors to be transformed into silicon carbide. The woods were cut into blocks (90mm x 20mm x 18 mm) and dried in oven furnace at 110°C for 24 hours to remove the moisture contained in the samples. In order to avoid crack happen to the samples, the pyrolysis process was done in two stages where the first stage was at 500°C of temperature with 1°C /min heating rate followed by second stage up to 850°C temperature with 2°C /min heating rate. Nitrogen gas was used as a flow gas. The weight loss and shrinkage rate in the samples were evaluated after the pyrolysis process.

After pyrolysis process, the samples were then infiltrated with the silicon powders in the vacuum furnace at two different temperatures of 1500°C and 1600°C with 5°C/min heating rate and 1 hour holding time.

The characterization of the wood decomposition behavior was performed with a thermogravimetric analyzer (TGA) while density was determined using Archimedes Principle. Pore size was determined by using mercury porosimeter. The microstructures of the carbon preform and ceramics were observed using Field Emission Scanning Electron Microscopy (FESEM) and Energy-Dispersive X-ray Spectroscopy (EDX) was used to analyze the structure and composition of the composites.

3.0 RESULTS AND DISCUSSION

3.1 Thermogravimetric Analysis (TGA).

Based on TGA analysis for both Kapur and Dark Red Meranti precursors, it is found that both samples give the similar pattern of weight loss as shown in Figure 1. Weight lost at 100°C was attributed for dehydration of the wood. The TGA curve clearly showed that both woods started to lose weight at temperature about 275°C to 440°C. During this temperature, process of decomposition of cellulose, hemicellulose and lignin of wood materials occurs simultaneously [5]. The weight loss is due to pyrolysis of the wood precursors and almost completed at about 650°C. It is then continued by breakdown of carbon chains in the structure and formation of graphite carbon structure at temperature above 600°C [5]. Wood samples experienced 60% weight loss compared to the original precursors, due to the thermal degradation process. Beside, a porous carbon template is then formed at the end of the process which is essential for the silicon infiltration stage.

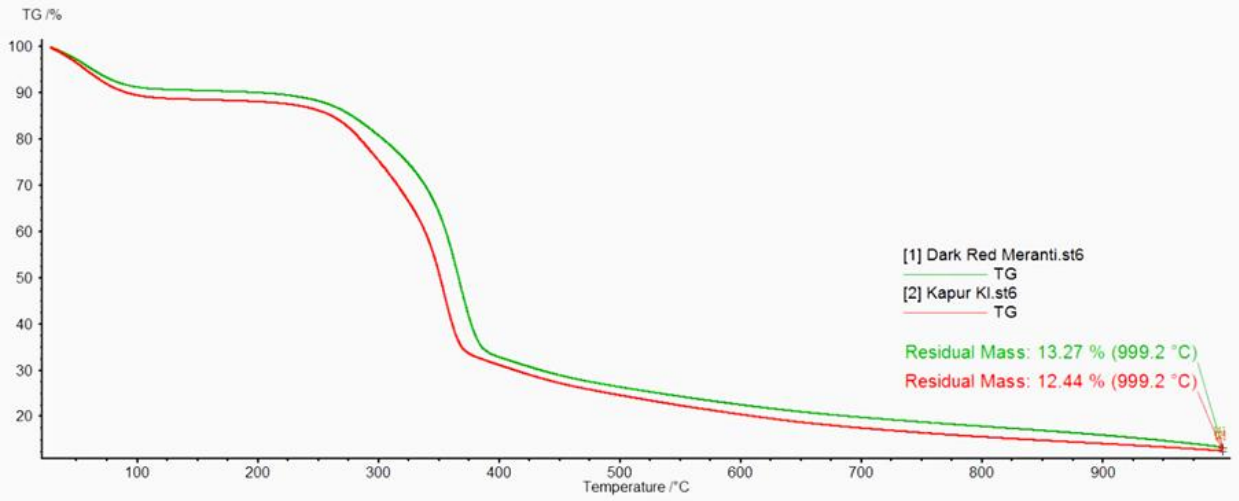


Figure 1 TGA analysis of Kapur and Dark Red Meranti samples

3.2 Density

Table 1 shows the density result for Kapur wood and Dark Red Meranti wood throughout the processes. The calculated values of density of both wood precursors are approximate to their theoretical values of 0.58-0.82 g/cm3 [6] and 0.415-0.885 g/cm3 [7], respectively. From the table above, it is found that density of the precursors decreases after undergo drying and pyrolysis process. This is due to water lost and decomposition of hemicellulose, cellulose and lignin of the wood samples during carbonization process [8]. The density of both samples increased after infiltrated with the silicon powders. Silicon powders melt and then diffused into carbon preform pore and cause the density increases. Higher infiltration temperature will produce denser SiC composites. However, the density of SiC composites was much lower than the theoretical value of SiC (3.21g/cm3) [9]. This means that the wood samples were not full reacted with the silicon during infiltration process.

Table 1 Density of wood samples

Sample	Density (g/cm³)				
	Original	After Drying process	After Pyrolysis process	After Infiltration process	
				1500°C	1600°C
Kapur	0.872	0.770	0.703	0.786	0.9
Dark Red Meranti	0.833	0.733	0.690	0.733	0.9

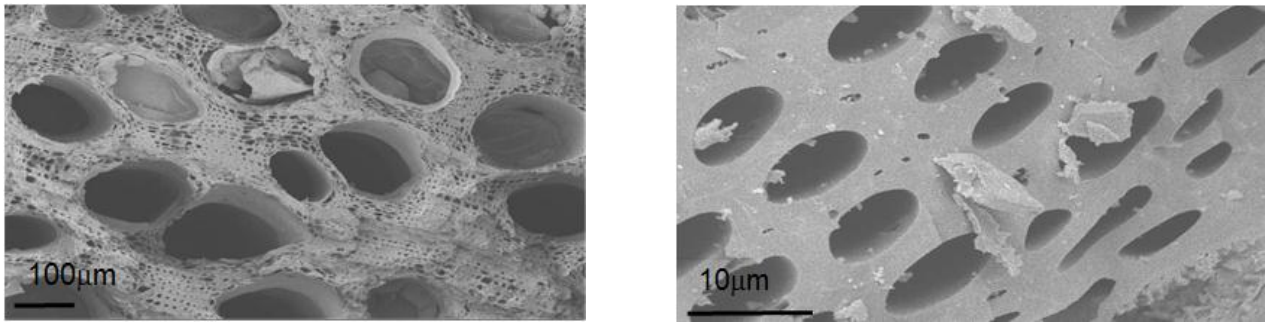
3.3 Porosity Analysis

Based on porosity analysis of both wood precursors, it is clearly shown that pyrolysis process enhanced the porosity of the wood precursors. Higher porosity will give higher chance of SiC formation in infiltration process. From Table 2, Dark Red Meranti wood consisted of higher porosity with 43.3190 % after pyrolysis process than Kapur wood, which is 36.3928 %. Porosity of wood cannot be measured and controlled easily, anyway charring process helped to develop porosity [10]. During infiltration process, silicon will infiltrate through the pores and react with the carbon of pyrolyzed wood structure. Past research showed variation of vessel distribution and pore distribution obtained mimicking those their carbon precursors with no pores smaller than 1µm [3]. Upon conversion to silicon carbide, all the small pores, below 1µm will be eliminated due to the volume expansion of almost 58% [3]. Besides, these porous structure of carbon template will give advantage in wide application of SiC ceramic that require open and highly oriented porous structure, especially in catalytic support, filtration and separation application [1,5,12].

Table 2 Porosities of Kapur and Dark Red Meranti before and after pyrolysis process.

Samples	Porosity of Samples (%)	
	Before Pyrolysis	After Pyrolysis
Kapur	32.6761	36.3928
Dark Red Meranti	39.5964	43.3190

3.4 FESEM-EDX Analysis



(a) (b)

Figure 2 FESEM analysis of (a) Kapur wood and (b) Dark Red Meranti wood after pyrolysis

Figure 2 shows the microstructure of kapur wood and Dark Meranti wood after pyrolysis process. High anisotropic porosity with original cellular structure of the wood is obtained after pyrolysis process takes place [11]. Due to charring process, the pore structure of the woods can be clearly seen. This is because the original wood precursors did not revealed any pore structure under microstructure examination. The similar effect also has been reported by other researchers [12]. Dark Red Meranti shows the pores ten times smaller than Kapur wood. Furthermore, more pores were found in Dard Red Meranti Wood. Pores will play an important role to Si infiltration later.

Composition of carbon template after pyrolysis process were tabulated in Table 3 which Kapur shows higher carbon content than Dark Red Meranti sample.

Table 3 Chemical composition contents in Kapur and Dark Red Meranti wood after pyrolysis

Element	Kapur		Dark Red Meranti	
	Weight %	Atomic %	Weight %	Atomic %
CK	94.03	95.45	91.03	93.82
OK	5.97	4.55	8.07	6.18
Total	100		100	

Table 4 Chemical composition in Kapur and Dark Red Meranti after infiltration process at 1500°C

Element	Kapur		Dark Red Meranti	
	Weight %	Atomic %	Weight %	Atomic %
N	31.95	52.45	33.21	39.68
Si	68.05	47.55	41.07	24.48
C	-	-	25.72	35.84
Total	100		100	

Table 4 shows the chemical composition content in Kapur wood and Dark Red Meranti wood after infiltration process under 15000C temperature. It is found that the Kapur wood remained high silicon content in the sample but no extra carbon. Silicon did not transformed into silicon carbide but the sample reacted with flow nitrogen and transform into Silicon Nitrite. On the other hand, the analysis shows that the Dark Red Meranti sample contained silicon carbide. Silicon reacted with carbon preform and formed silicon carbide in the Dark Red Meranti, at the same time silicon also reacted with nitrogen gas. However, limited porosity in Kapur structure only enables the silicon to react with nitrogen and remained in excessive silicon powders. Thus, high silicon element content was found in the Kapur.

Table 5 shows the chemical composition content in Kapur wood sample and Dark Red Meranti wood sample after infiltration process under 1600°C temperature. The analysis shows similar results in 1500°C. In Kapur sample, silicon not transformed into silicon carbide but the sample reacted with nitrogen and transform into Silicon Nitrite. For Dark Red Meranti sample, the analysis showed that the Dark Red Meranti sample have silicon carbide formation although the wood react with nitrogen at the same time.

Table 5 Chemical composition in Kapur and Dark Red Meranti after infiltration process (1600°C)

Element	Kapur		Dark Red Meranti	
	Weight %	Atomic %	Weight %	Atomic %
CK	-	-	22.19	31.36
NK	35.49	52.45	35.59	43.13
SiK	64.51	47.55	42.21	25.51
Total	100		100	

4.0 CONCLUSION

Low densities of biomorphic silicon carbide were obtained from the natural woods, Kapur and Dark red Meranti. This was due to the silicon powders that did not fully react with carbonous preform during the infiltration process. From 1500°C to 1600°C infiltration temperatures, Kapur wood was not transformed into biomorphic silicon carbide but transformed into silicon nitrite. This was because the silicon preferred to react with nitrogen gas instead of carbon. It was found that the most suitable wood sample to be transformed into biomorphic silicon carbide was the Dark Red Meranti wood due to their high density, high porosity, low pore diameter and more silicon carbide formation after the infiltration process. From the results obtained it was found that the most suitable temperature for SiC transformation was at 1600°C.

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