Synthesis of silicon carbide from coal fly ash and activated carbon powder

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ABSTRACT

Micrometer silicon carbide powder has been synthesized from coal fly ash and activated carbon powder. Calcinated Suralaya fly ash used as SiO₂ resources on carbothermal reduction process with activated carbon powder. Particle size of activated carbon powder is < 32 mm. Synthesis process has been carried out on temperature 1200 °C and and 1300 °C on vacuum atmosphere under argon flow. Holding time for all synthesis is 2 hour. Synthesis products were analyzed by XRD with Cu-K α radiation, FT – IR spectrophotometer and SEM fitted with EDAX. SiC powder with particle size about 5 mm were successfully synthesized at temperature 1300 °C.

Key word: Silicon carbide, coal fly ash, carbon powder.

INTRODUCTION

Fly ash is a byproduct of the burning process at coal-fired power plant. Fly ash is obtained by electrostatic or mechanical precipitation from the flue gases of furnaces fired with pulverized coal. In USA and Europe coal combustion by product is estimated around 115 million tons per year¹. Suralaya power plant, once of the Indonesia’s power plant that used coal, generated about 180,039.1 tons fly ash in 2005. The employed fly ash as a valuable resource in China is only 20 – 30 %, in Europe is about 60 % and in USA is about 30 %²,³. Most of fly ash production is still stored in disposal site. So, the storage of fly ash has become a serious environmental pollution problem.

Silicon Carbide (SiC) is a kind of covalent material. SiC has excellent mechanical strength and high hardness, excellent resistance to oxidation and corrosion, low coefficient of thermal expansion, high heat-transfer capability, excellent thermal stabilities and chemical inertness²,³. Silicon Carbide can be produced by chemical vapor deposition (CVD) and carbothermal reduction. Chemical vapor deposition (CVD) is a chemical process used to produce high-purity, high-performance solid materials. In a typical CVD process, the substrate is exposed to one or more volatile precursors, which react and/or decompose on the substrate surface to produce the desired deposit. Frequently, volatile by-products are also produced, which are removed by gas flow through the reaction chamber⁴. The carbothermal reduction is relative simple and time-cost effective technique, in this process : mixture of carbon and silica or silicon is heated in a reactor in an inert atmosphere⁵. Reaction between carbon and silica to form SiC is as follows⁶,⁷:
\[ \text{SiO}_2(s) + 3C(s) \rightarrow \text{SiC}(s) + 2\text{CO}(g) \]  
...(1)

Reaction (1) proceeds from two stages 8:

\[ \text{SiO}_2(s) + C(s) \leftrightarrow \text{SiO}(g) + \text{CO}(g) \]  
...(2)

\[ \text{SiO}(g) + 2C(s) \leftrightarrow \text{SiC}(s) + \text{CO}(g) \]  
...(3)

Major component of fly ash is 40 % wt SiO$_2$. It is potential SiO$_2$ resources on carbothermal reduction process. Quartz (SiO$_2$) and SiO can be reacted with activated carbon powder in carbothermal reduction process to form silicon carbide. SiC powder had been successfully synthesized from fly ash and 8.21 mm carbon black$^2$. On this process, the use of very fine carbon sources caused high cost process because starting material is expensive. Developing process with cheap starting material is focused on this research. This paper presents the synthesized process of silicon carbide uses cheap carbon source.

**MATERIAL AND METHODS**

The starting materials were calcinated fly ash and activated carbon powder. The fly ash used in this experiment was collected from Suralaya power plant. Composition of coal fly ash was tested by atomic absorption spectroscopy, the composition is shown in Table 1. Calcinations process has been done in carbolite furnace at temperature 850 °C for 4 h in atmosphere condition. XRD pattern of calcinated fly ash and FTIR analysis are shown in Figure 1 and Figure 2. Activated carbon powder with particle size-400 mesh (<32mm) was used as carbon resources. The carbon source is made form granular activated carbon then ball mill and sieved with-400 mesh sieving machine. This process resulted-400 (<32mm) activated carbon powder. Weigh of fly ash in starting precursor was determined based amount of SiO$_2$ in fly ash. Molar ratio SiO$_2$ and activated carbon powders was 1 : 4. Fly ash and activated carbon powder were mixed using magnetic stirrer for 6 h in ethanol solution. Before synthesis process, the mixture was dried.

Synthesis SiC has been conducted in the carbolite vacuum furnace an temperature : 1200 °C, 1300 °C and 1400 °C for 2 h and cooled naturally. All synthesis reactions were carried out on vacuum condition under argon flow, the schematic process is shown in Figure 3. After the synthesis process, the product was heated at temperature 850 for 4 h in atmosphere condition to burn the excess carbon. Synthesis products were examined by X-ray powder diffraction (XRD) using Cu $K_α$ radiation, Fourier transformation infrared spectrophotometer (FT-IR) and SEM fitted with EDAX. Peaks from XRD test were interpreted by PANalytical.

**RESULT AND DISCUSSION**

According to ASTM standard fly ash collected from Suralaya power plant is classified as type C. Figure 1 and Figure 2 show XRD patterns and FTIR spectrums of calcinated fly ash. XRD pattern show that calcinated fly ash consist of quartz (SiO$_2$) and hematite (Fe$_3$O$_4$) as the major component. Amount of SiO$_2$ on calcinated fly ash is about 45 %. The absorption peaks around 440 cm$^{-1}$, 725 cm$^{-1}$ and 1059 cm$^{-1}$ are characteristic absorption of SiO$_2$. Phases from synthesis process at temperature 1200 °C show in Figure 4. XRD pattern shows the low peak at 2q = 35.6 °, 41.3 °, 60.1 ° and 72.1 ° indicates that only small SiC powders were produced at this reaction temperature. The major phase that present at this temperature reaction is gupetite (Fe$_3$Si) and low quartz (SiO$_2$).

The XRD pattern of the powders produced at 1300 is shown in Figure 5, it shows that peaks at 2q = 35.6 °, 41.3 °, 60.1 ° and 72.1 ° is higher than in Figure 4. Based on the JCPDS data that shown in Figure 6, the peaks of the powders diffraction angles show that major component of this product

| No. | Component | % wt |
|-----|-----------|------|
| 1.  | SiO$_2$   | 45.52|
| 2.  | Al$_2$O$_3$ | 30.3 |
| 3.  | Fe$_3$O$_4$ | 8.72 |
| 4.  | CaO       | 5.5  |
| 5.  | MgO       | 2.75 |
| 6.  | others    | 7.21 |
Fig. 1: XRD pattern of calcinated fly ash

Fig. 2: FTIR spectrum of calcinated fly ash

Fig. 3: Schematic process synthesis SiC
Fig. 4: XRD pattern at reaction temperature 1200 for 2 h

Fig. 5: XRD pattern at reaction temperature 1300 for 2 h

Fig. 6: XRD pattern data of SiC and Fe$_3$Si
reaction are: b-SiC and Fe$_3$Si. Fe$_2$O$_3$ in this reaction acts as catalyst resulting in intermetallic phase (Fe$_3$Si) as an intermediate phase. According to thermodynamics, the standard free energy is endothermic until 1500 °C. Component Fe$_2$O$_3$ in fly ash can act as catalyst and forms the intermediate phase Fe$_3$Si which makes possible to obtain SiC at temperature reaction under 1500 °C. In uncatalysed reaction, silicon monoxide reacts with carbon to produce SiC as shown in reaction (3). At low temperature reaction (below 1400 °C) most of SiC formed is generated through reaction (4) which takes place at the contact points between solid carbon and SiO$_2$.

In a catalyzed reaction (reaction 5,6), silicon monoxide reacts with CO in the surface of the catalyst to yield CO$_2$, carbon and silicon, which are dissolved in the liquid metal. SiC is produced by segregation of silicon and carbon from supersaturated solution of iron [10].

\[
SiO_2 + 3C \rightarrow SiC(s) + 2CO(g) \quad ... (4)
\]

\[
SiO + 3CO + Fe(Si, C) \leftrightarrow 2CO_2 + Fe(Si^{SS}, C^{SS}) ... (5)
\]

\[
Fe(Si^{SS}, C^{SS}) \leftrightarrow SiC + Fe(Si^{S}, C^{S}) \quad ... (6)
\]
SiC powder from this synthesis process is also shown for FT – IR spectrum. Figure 7 shows the absorption peak around 902 cm⁻¹ that is caused by bond of Si – C. Reducing peaks of SiO₂ phase in XRD pattern and the lost peak FT-IR spectra around 725 cm⁻¹ indicated that SiO₂ reacted with another component to form another phases such as: SiC or Fe₃Si. The morphology of SiC product is shown in Figure 8. This process produces SiC powder with particle size about 5 mm.

**CONCLUSION**

1. β-SiC was successfully synthesized at temperature 1300 °C for 2 h with starting material fly ash and activated carbon powder.
2. Fe₂O₃ act as catalyst resulted the intermediate phase Fe₃Si and make possible to synthesized SiC on temperature under 1500 °C.
3. This process produces SiC powder with particle size about 5 mm.

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