# Method for increasing β-SiC yield on solid state reaction of coal fly ash and activated carbon powder

*by* Sulardjaka Sulardjaka

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# Method for increasing β-SiC yield on solid state reaction of coal fly ash and activated carbon powder

**SULARDJAKA*, JAMASRI, M W WILDAN and KUSNANTO†**

Department of Mechanical and Industrial Engineering, Gadjah Mada University, Yogyakarta 55281, Indonesia

†Department of Engineering Physics, Gadjah Mada University, Yogyakarta 55281, Indonesia

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**Abstract.** **A novel process for increasing β-SiC yield on solid state reaction of coal fly ash and micro powder activated carbon powder has been proposed. β-SiC powder was synthesized at temperature 1300°C for 2 h under vacuum condition with 1 l/min argon flow. Cycling synthesis process has been developed for increasing β-SiC yield on solid state reaction of coal fly ash and activated carbon powder. Synthesized products were analyzed by XRD with Cu-Kα radiation, FTIR spectrometer and SEM fitted with EDAX. The results show that the amount of relative β-SiC is increased with the number of cycling synthesis.**

**Keywords.** **SiC; cycling synthesis; solid state reaction.**

## 1. Introduction

Fly ash, a waste product of coal combustion in thermal power plants, is produced in dry form in large quantity. Large quantities of fly ash are produced during the combustion of coal in the production of electricity. Coal combustion by product in USA and EU is estimated around 115 million tons per year (Querol *et al* 2002). Suralaya power plant in Indonesia which is fueled by coal, has generated about 0·2 million tons fly ash in 2005 (Sulardjaka *et al* 2009). In South Africa, where high content coal is used in power generation, 24 million ton fly ash were produced in 1997, and only 5% of this ash was reused (Woolard *et al* 2000). The current annual production of worldwide coal ash is estimated around 600 millions tons, with fly ash constituting about 500 millions tons (Hongjie *et al* 2001). Most of the fly ash production is still stored in a disposal site and only 20–30% is reused as a valuable resource (Joshi and Lothia 1997). The amount of the fly ash released by factories and thermal power plants has been increasing throughout the world, and the disposal of fly ash has become a serious environmental problem. The compositions of the fly ash varies depending on the source of coal and the combustion process. The main constituents are metal oxides, predominantly of silicon and aluminum in addition to other metal oxides. Utilization of fly ash in producing novel materials, waste management and recovery of metals are the new research areas that will expand the positive reuse of this abundant material, thereby helping to reduce the environmental and economical impacts of its disposal.

Silicon Carbide (SiC) has superior strength and corrosion resistance at elevated temperature (Ryu *et al* 2002). In industrial scale, SiC is produced by the Acheon process, that is carbothermal reduction of silica sand with green petroleum coke in temperature of around 2400°C (Hongjie *at al* 2001). The carbothermal reduction is relatively simple and time–cost effective technique; in this process, mixture of carbon and silica or silicon is heated in a reactor in an inert atmosphere (Shi *et al* 2006). In order to optimize the production of SiC particles, several authors have proposed methods which used fine starting materials and reaction mixtures prepared by sol–gel techniques (Russell-Floyd *et al* 1993; Qiang Jin and Yun Go 2003; Shi *et al* 2006). Synthesis of SiC requires a series of reaction. At reaction temperature below ~1400°C, most of the SiC is generated through reaction solid $SiO_2$ and solid carbon (Krstic 1992; Qian 2004). The reaction takes place at the contact point between solid carbon and solid $SiO_2$. Fabrication of silicon carbide at low temperature needs long time, because it is limited by the diffusion rate of $SiO_2$ and C (Krstic 1992).

The 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 produce silicon carbide. β-SiC powder had been successfully synthesized from fly ash and 8·21 μm carbon black (Hongjie *et al* 2001). Our previous work is also successful in synthesizing β-SiC form fly ash and 32 μm activated carbon at temperature 1300°C under argon flow (Sulardjaka *et al*

*Author for correspondence (s_djaka@yahoo.com)

**Table 1.** Composition of Suralaya coal fly ash.

| Compound | $SiO_2$ | $Al_2O_3$ | $Fe_2O_3$ | CaO | MgO | $Na_2O$ | MnO | $K_2O$ | Others |
|---|---|---|---|---|---|---|---|---|---|
| Weight | 45·51 | 30·35 | 8·71 | 5·49 | 2·75 | 1·45 | 0·1 | 0·64 | 5·0 |

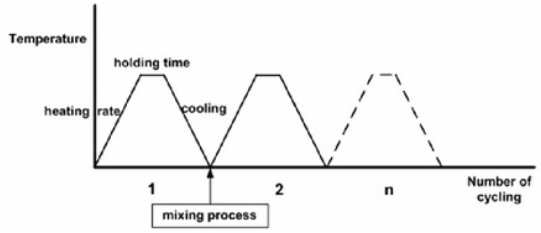


**Figure 1.** Schematic cycling synthesis.

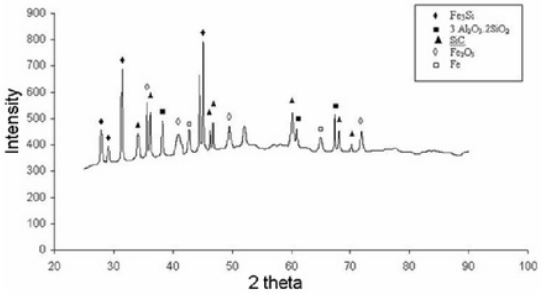


**Figure 2.** XRD pattern at reaction temperature 1300°C for 2 h.

2009). SiC yield from synthesis process can be increased by increasing temperature, use very fine starting material and longer holding time reaction. However, those methods need higher cost process. Therefore, this paper proposes a method for increasing SiC yield at low temperature synthesis process, shorter holding time and utilizing cheap materials for synthesis $\beta$-SiC powder.

## 2. Materials and experimental procedure

Our experiment used starting materials which ultilized fly ash and activated carbon powder. The fly ash was collected from Suralaya power plant, Indonesia. Composition of the coal fly ash was analyzed by atomic absorption spectroscopy (AAS) as shown in table 1.

Activated-carbon powder with particle size – 400 mesh (< 32 μm) was used as the carbon sources. The carbon source was made from granular activated-carbon that was ball milled and sieved with – 400 mesh sieving machine. Weight of the fly ash in starting precursor was determined based on the amount of $SiO_2$ in the fly ash. Molar ratio of $SiO_2$ and activated-carbon powders was 1 : 4. The fly ash and activated carbon powder were mixed using magnetic stirrer for 6 h in ethanol solution.

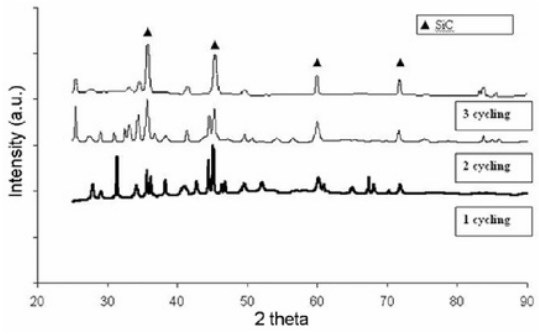


**Figure 3.** XRD pattern at reaction temperature 1300 for 2 h with cycling process.

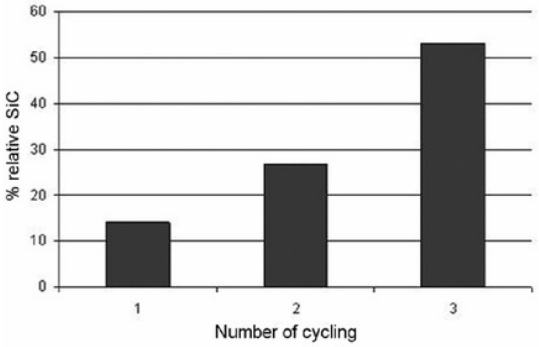


**Figure 4.** The effect of cycling on percentage of SiC yield.

Synthesis of $\beta$-SiC was conducted in the carbolite vacuum furnace at temperature of 1300°C, with a heating rate of 10°C/minute, and holding time process of 2 h, then followed cooling in the furnace by switching the furnace off. All synthesis reactions were carried out on vacuum condition under 1 l/min argon flow. After the synthesis process, the product was heated at temperature 850°C for 4 h in atmosphere condition to burn the excess carbon. Synthesized products were examined by X-ray powder diffraction (XRD) using CuK$\alpha$ radiation, Fourier transformation infrared spectrometer (FTIR) and SEM fitted with EDAX. Peaks obtain from XRD test were interpreted by PANalytical. The amount of $\beta$-SiC yield in each process was estimated by comparing the relative height of the $\beta$-SiC diffraction peaks with that of all diffraction peaks. Cycling synthesis process has been developed for increasing SiC yield at temperature synthesis

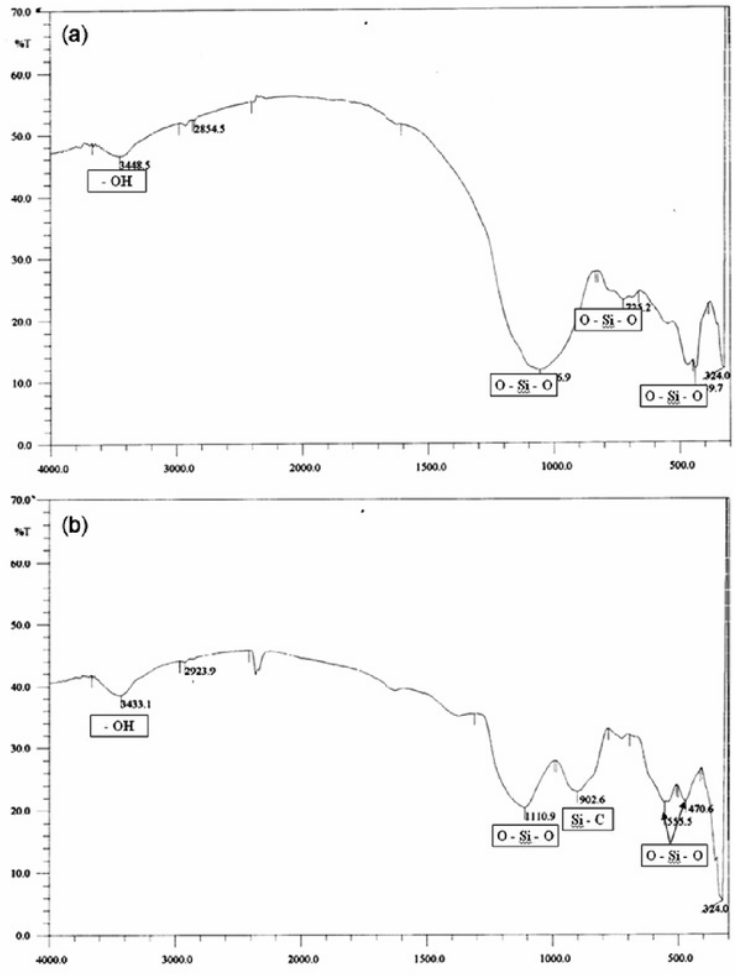


**Figure 5.** FTIR spectrum of fly ash (**a**) and synthesis product (**b**).

1300°C. The objective of the cycling process is to make new surface contact between $SiO_2$ and C. The cycling process is resynthesis of the product from previous synthesis process, the schematic of cycling synthesis is shown on figure 1. After one cycling synthesis process, the synthesized product was added with 1 mol activated carbon powder then cruised and mixed on jar mill with steel ball for 2 h.

## 3. Results and discussion

Figure 2 shows the XRD pattern synthesized product at temperature 1300°C from which $\beta$-SiC was formed by present major peaks at 35·6°, 41·3°, 60·1° and 72·1°.

Figure 3 shows the XRD pattern of the powders produced at 1300°C with the cycling process. It shows that peaks at $2\theta = 35{\cdot}6°$, 41·3°, 60·1° and 72·1° is higher with the increasing of the cycling process. This increase affects the relative amount of $\beta$-SiC as depicted in figure 4. Figures 3 and 4 show that SiC yield increases with the cycling process. Under the experimental condition of the present work, the reaction between C and $SiO_2$ for producing SiC is shown in reaction (1) (Vix-Gutherl and Ehrburger 1997). At low temperature reaction (below 1400°C) most of SiC formation is generated through reaction (1). The reaction consists of a solid–solid, solid–liquid types reaction between C and $SiO_2$. At this reaction, formation of $\beta$-SiC is formed by diffusion of $SiO_2$ and C which takes place at the contact points between solid C and $SiO_2$ (Krstic 1992; Russell-Floyd 1993). The cycling synthesis process requires mixing process between one and another previous synthesis process and produces new contact point between $SiO_2$ and C and promote the diffusion rate.

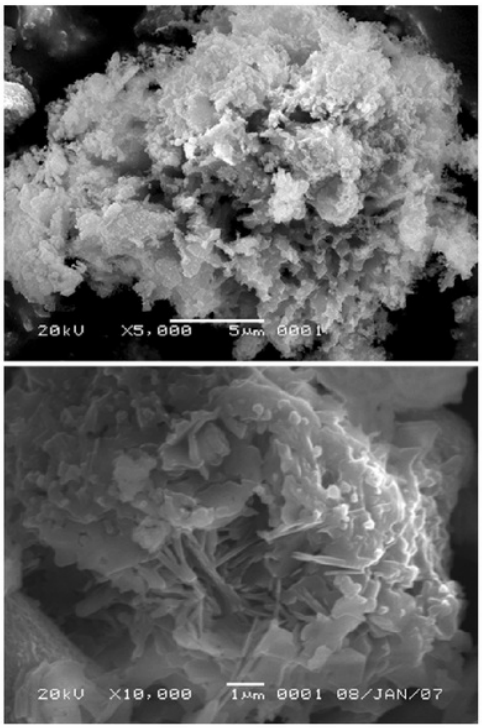


**Figure 6.** The SiC morphology.

In this research, there are possibility of two mechanisms that produce $\beta$-SiC. First, $\beta$-SiC is produced through mechanism diffusion process that follows (1), and second, $\beta$-SiC resulted through reaction (3) that involves catalyst of $Fe_2O_3$. $Fe_2O_3$ acts as catalyst in resulting intermetallic phase ($Fe_3Si$) as an intermediate phase as presented in figure 2. The presence of $Fe_3Si$ in this process makes possibility to obtain $\beta$-SiC at temperature reaction under 1500°C. In uncatalyzed reaction, silicon monoxide reacts with carbon to produce SiC as shown in reaction (2). In a catalyzed reaction, the possible explanations of the role of the catalyst $Fe_2O_3$ is (Narciso-Romero and Rodriguez-Reinoso 1996; Hongjie *et al* 2001): $Fe_2O_3$ produces a local decrease of CO produced because the metal oxide favours a disproportionate CO to give the $CO_2$, the $CO_2$ can react again with C to produce CO, the consequence of previous reaction is the formation of Fe as an intermediate stage. $Fe_2O_3$ is reduced by CO to produce Fe. In this work, Fe produced from this reaction is presented in XRD patterns as depicted in figure 2. Then, the intermediate phase $Fe_3Si$ is formed, which, at the reaction temperature (around 1300°C) $Fe_3Si$ is in liquid state, where C dissolved to form a supersaturated solid solution from which the solid phase SiC is removed (3).

$$SiO_2(s.l.) + 3C(s) \rightarrow SiC(s) + 2CO(g), \quad (1)$$

$$SiO(g) + 2C(s) \leftrightarrow SiC(s) + CO(g), \quad (2)$$

$$Fe(Si^m, C^n) \leftrightarrow SiC + Fe(Si^{m1}, C^{n1}), \quad (3)$$

where $m_1 < m$ and $n_1 < n$.

$\beta$-SiC powder from this synthesis process is also shown in FTIR spectrum as presented in figure 5(b). This figure shows the absorption peak around 902 $cm^{-1}$ which is caused by bond of Si–C. Reducing peaks of $SiO_2$ phase in XRD pattern (figure 4) and the last peak FTIR spectrum around 725 $cm^{-1}$ (figure 5(a)) indicate that $SiO_2$ reacts with another component to produce other phases such as: SiC or $Fe_3Si$. Figure 6 shows the morphology of $\beta$-SiC product. This process produces porous $\beta$-SiC micro powder.

## 4. Conclusions

$\beta$-SiC was successfully synthesized at temperature 1300°C for 2 h with starting materials fly ash and 32 μm activated carbon powder. There are two reaction mechanisms to produce $\beta$-SiC, that are solid reaction $SiO_2$ and C and segregation of $SiO_2$ and C from supersaturated solution of iron. The cycling synthesis process makes new contact point between silicon and carbon and promote the diffusion rate. In this research, $\beta$-SiC was successfully produced and increased by the cycling synthesis.

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