Electrospinning β-SiC fibers from SiC nanoparticles dispersed in various polymer solutions as the electrospinning agents

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Abstract. Silicon carbide (SiC) fibers were synthesized by electrospinning method from SiC nanoparticles dispersed in polymer solutions, i.e., polyethylene glycol (PEG) and polyvinyl alcohol (PVA). The SiC nanoparticle used in this research was synthesized from sucrose and natural silica via a sonochemical method. The natural silica was extracted from local pyrophyllite by a sol-gel method. The characterization was performed via x-ray fluorescence (XRF), X-ray diffraction (XRD), scanning electron microscopy (SEM). The XRD characterization results showed that the sample possessed a β-SiC phase and formed a cubic-structured crystal with a lattice parameter of \(a = b = c = 4.3448\,\text{Å}\). The use of PEG and PVA in the electrospinning process resulted in fractal and fiber structured SiC, respectively.

1. Introduction

Various non-oxide ceramics are used for many industrial applications. Silicon carbide (SiC) serves as an exclusive example of such non-oxide ceramics due to its superior characteristics such as good mechanical property, high level of thermal and electrical conductivity, excellent chemical stability, high radiation resistance, and so forth [1]. Such distinctive properties have turned SiC to be a promising material to be applied as a nuclear, turbine, and furnace reactor, and a jet engine, or even for the needs of space exploration [2–4]. Moreover, fiber structured SiC can also be applied to a solar panel or as a photocatalyst in a hydrogen production via water splitting process [5,6].

Commonly, the synthesis of SiC fibers is conducted by chemical vapor deposition (CVD), template, arc-discharge, carbothermal reduction, and molten salt synthesis methods [7–9]. Several obstacles are found in such methods, which include the needs of a very high synthesis temperature, an expensive and rare precursor, and high and complex technology. Therefore, it is necessary to find an alternative that allows a low price and simple synthesis of SiC fibers.

This research aimed to synthesize silicon carbide fibers through a straightforward and inexpensive method. In order to achieve such purpose, the synthesis was performed using sucrose and silica extracted from pyrophyllite as precursors. Besides reducing the production cost, the utilization of silica extracted from pyrophyllite as a primary precursor is also improving the functional value of local resources in Indonesia.

2. Experimental
2.1. Silica extraction from pyrophyllite
The silica was extracted from pyrophyllite via a sol-gel method. The content of Si and other elements in the pyrophyllite are presented in table 1. Furthermore, 20 grams of pyrophyllite powders were put into a beaker, and then added with 500 ml 5M sodium hydroxide solution and stirred at 120 °C. Then, the mixture was filtered. The filtrate in the form of a sodium silicate was then titrated using a 5M hydrochloric acid solution until it reached pH 7. The silica gel formed in the titration process was rinsed using distilled water to be then drained.

Table 1. Element contents in pyrophyllite.

| No. | Element | Content of element (%) |
|-----|---------|------------------------|
| 1.  | Si      | 64.70                  |
| 2.  | Al      | 16.00                  |
| 3.  | Fe      | 8.95                   |
| 4.  | Ti      | 7.17                   |
| 5.  | Ca      | 1.32                   |
| 6.  | Other elements | 1.86               |

2.2. Synthesis of SiC nanoparticles
The SiC nanoparticles were synthesized using a sonochemical method. 3 grams silica and 9 grams sucrose were added to a 240 ml distilled water and ethanol mixture. Next, the mixture was stirred and sonicated at 70 °C. The result was then put into a furnace for the following preheating process for 1 hour at 180 °C. After that, the process continued by annealing at 950 °C in Argon ambiance until SiC nanoparticles were produced.

2.3. Synthesis of SiC fibers
0.1 grams SiC nanoparticles was added in to a 2 ml PEG solution (Mw ~6,000 Da) and a PVA solution (Mw ~186,000 Da). In order to ensure the even dispersion of the SiC nanoparticles, each of the solutions was stirred and sonicated for 1 hour. The sonicated solution was then put into a syringe with 25 gauge needle. After that, the electrospinning process was performed by using the parameters presented in table 2.

Table 2. Electrospinning process parameters.

| No.  | Parameter                        | Value  |
|------|----------------------------------|--------|
| 1.   | Tip to collector distance (TCD)  | 10 cm  |
| 2.   | Pump speed                       | 0.005 ml/min |
| 3.   | Electrospinning period           | 1 hour |
| 4.   | Applied voltage                  | 20 kV  |

2.4. Characterization
The percentages of the elements contained in pyrophyllite and natural silica were identified by an x-ray fluorescence (XRF, PANalytical) characterization. The SiC nanoparticles formed a crystalline phase that was identified via x-ray diffraction (XRD, Phillips X’Pert Pro) characterization. Morphology of the SiC fibers resulted from the electrospinning process was observed via a scanning electron microscopy (SEM, FEI INSPECT-S50) characterization.

3. Results and Discussion
3.1. Element contents in natural silica

Elements contained in natural silica extracted from pyrophyllite were identified via XRF. The XRF characterization results are presented in table 3. According to the XRF results, it is known that the extracted silica contains 95.4 % Si element, which indicated an improvement on the silica purity after the extraction process compared with the initial pyrophyllite that only contained 64.7 % Si element (table 3).

| No. | Element | Content of element (%) |
|-----|---------|------------------------|
| 1.  | Si      | 95.40                  |
| 2.  | Ca      | 1.30                   |
| 3.  | Ti      | 0.12                   |
| 4.  | Fe      | 0.43                   |
| 5.  | Other elements | 2.75                 |

3.2. Crystalline phase, size, and structure of SiC nanoparticles

The diffraction pattern of the SiC nanoparticles is displayed in figure 1. All of the diffraction peaks formed were in good agreement with β-SiC diffraction peaks based on ICSD No. 01-072-1708, without any other phase. The results of the Rietveld analysis identified that the SiC nanoparticles are cubic-structured with a lattice parameter of a = b = c = 4.3448 Å. Mostly, the SiC nanoparticles have a size of 18.1 nm as measured via a Scherrer equation.

3.3. Morphology of SiC nanoparticles

The SiC nanoparticle morphology is shown in figure 2. It can be seen that the SiC nanoparticles tend to agglomerate and form a bigger sized aggregate. It indicates an occurrence of agglomeration in the sample. The average size of the SiC nanoparticles was 48.291 nm.

3.4. Morphology of SiC fibers

Figure 3 displays the morphology of the SiC synthesized via electrospinning method using the PEG solution. The use of PEG produced fractal or irregular-structured SiC. It was due to the overly low molecular weight (Mw ~ 6.000 Da) of the PEG used in the process. The molecular weight of a polymer represents the length of such polymer chain. A low molecular weight of a polymer will have a short
chain that is breakable during the electrospinning process [10]. The synthesis of the SiC fibers by using PEG with a higher molecular weight (Mw ~300,000 Da) is proven to be able to produce SiC fibers in diameters ranging from 200-800 nm [11].

Figure 3. Morphology of SiC fibers synthesized by electrospinning method using a PEG solution

Figure 4 shows the SEM characterization results of the SiC synthesized via electrospinning method using a PVA solution. The utilization of PVA in the electrospinning process produced beaded SiC fibers, which indicates that the PVA concentration was still too low. A polymer solution with a low concentration will be mostly formed by the solvent molecule with less polymer molecule. A high ratio of the solvent molecule to polymer molecule causes the solvent surface area to dominate the polymer solution during the electrospinning process. The high surface tension of the solvent will cause it to maintain its spherical form along the polymer chain leading to the production of the beaded fibers [10].

Different with PEG, the use of PVA in an electrospinning process can result in fiber-structured SiC (figure 4). It is because PVA has a higher molecular weight than PEG. The high molecular weight of PVA indicates that it has a longer polymer chain, which tends to form an entanglement. In an electrospinning process, when the solution was injected from the needle to the collector, the polymer chain of the solution would stretch because of an electric field. An entanglement of a polymer chain would prevent it to break during such stretching process so that it could result in a continuous production of fibers [10].

Figure 4. Morphology of SiC fibers synthesized by electrospinning method using a PVA solution of 10% w/v.

Figure 5. Morphology of SiC fibers synthesized by electrospinning method using a PVA solution of 12.5% w/v.
The growth of the beaded fibers can be avoided by increasing the molecular number of PVA such as by increasing the concentration of the PVA. Figure 5 shows the morphology of the β-SiC that produced by electrospinning technique for the concentration of PVA 12.5% w/v. The mean diameter of the β-SiC fibers is 228 nm. The balance comparison of PVA with solvent yang tended to reduce the surface tension of solvent and the solvent distributed homogeneously along with the fibers [10]. Therefore, it produced the fibers with a homogeneous diameter as presented in figure 5.

4. Conclusion
In this research, β-SiC fibers have been successfully synthesized using an electrospinning method from SiC nanoparticles dispersed in a polymer solution. The produced SiC is in the form of a cubic-structured β-SiC phase with a crystal size of 18.1 nm. The use of PEG in an electrospinning process led to the production of fractal-structured β-SiC while the utilization of PVA resulted in β-SiC fibers. Hence, the β-SiC fibers were well-synthesized by an electrospinning method using PVA as the electrospinning agent.

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