Synthesis of Silicon Carbide Fiber as semiconductor substrate for Betavoltaic cell

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Abstract. Silicon carbide (SiC) has excellent properties to be utilized as a semiconductor substrate because this material with high thermal conductivity and has stable properties against mechanical and chemical and also resistant to radiation. In this research, SiC fiber synthesis of polycarbosilane by electrospinning method with N, N-dimethylformamide (DMF) and toluene solvent was used. This electrospinning method is excellent for making fibers with controlled diameters and less than 10 μm. Some parameters that affect the fiber results will be studied that is by varying the voltage and temperature in the pyrolysis process. SiC fibers obtained were characterized by Microscop optic, FTIR, XRD and SEM. The SiC fibers were made into composites that poly vinyl alcohol (PVA) used as matrix. The obtained composites were then tested for electrical conductivity and dielectric constant using LCR meter in order to show the ability as a semiconductor to be applied on betavoltaic cell. The Results obtained from FTIR, XRD and SEM-EDS shows SiC fibers have been successfully synthesized by this method. SiC fiber morphology is known of changes fiber diameter and fiber uniformity. The average fiber diameter as-received from electrospinning process were decrease with increasing the applied voltage during electrospining process. The average fiber diameter result from applied voltage for 10 kV were at 10.4 μm. The average diameter of fiber after pyrolisys with temperature variation at 1200, 1300 and 1400 °C has a tendency to become smaller with an increase of temperature in all applied voltage. The SiC fiber produced in all variations of voltage and pyrolysis temperature was a semiconductor material. The electrical conductivity value at frequencies 1 to 1000 Hz, the area of electrical conductivity values ranges from $8 \times 10^{-6}$ to $7 \times 10^{-6}$.

Keywords: SiC fibers, SiC composites, polycarbosilane, semiconductors, betavoltaic cell

1. Introduction
The need for long-lived power sources primarily to be placed in hard-to-reach areas and difficult access and also for long-term applications has led to the need for energy power sources from these radioisotopes into energy solutions that should be considered an alternative to next-generation battery technologies in the future. Lifetime of betavoltaic cell battery life is very long if we compared with other types of batteries that can live 10 to 100 years [1, 2].

The betavoltaic cell like a mini portable nuclear power plant in that the working principle of this cell is to convert a portion of the energy of beta particles (β) into electric power. The kinetic energy of the beta particles creates an electron pair. The magnitude of the voltage on the device is proportional to the band gap of the semiconductor and the distance between the positive and negative poles
produced. The working principle of this betavoltaic cell is similar to that of solar cells, with illustrations of converting beta radiation energy to electric energy as follows:

![Figure 1. Illustration of converting beta radiation energy into electrical energy][1]

The choice of Silicon carbide (SiC) as a semiconductor because SiC has excellent mechanical properties, so it can be used for many applications. Its hardness and wear resistance make SiC ideal for abrasive machining, such as grinding and sand blasting, it can also be used at high temperatures and resistant to radiation. SiC is a candidate for electronic devices such as high power switching, high-voltage light-emitting diodes and wafers.[3]

Electrospinning is a highly versatile method that, through processing of polymeric solutions or melts, produces fibres with diameters ranging from a few microns to tens of nanometers [4]. The use of polycarbosilan (PCS) as SiC-forming precursor polymer has been done by several researchers such as Sugimoto et al [7] and Yue, et al [5], where in processing PCS into SiC fibers consist of several stages of methods: synthesis, spinning, curing and pyrolysis. The mechanism of the SiC fiber formation reaction of PCS fibers occurs during the pyrolysis process, due to differences in chemical bonding energies on Si and C atoms in the PCS main chains as described in Fig. 2 [3], [5].

![Figure 2. Mechanism of chemical reaction of SiC fiber synthesis from PCS][5]

2. Experimental
Polycarbosilan is used as a precursor by dissolving a toluene solvent and N, N-dimethylformamide (DMF) with a composition at 30% (% DMF / toluene), during the electrospinning process a variation of voltage of 10,15,20 kV is applied. Then the fibers were characterized by microstructure and average diameter by optical microscope, functional group with FTIR and optical microscope. The electrospinning fiber is then cured at 200 °C, for 1 hour at a rate of 2-3 °C/min and then pyrolysis. In the process of pyrolysis conducted temperature variations are 1200, 1300, 1400, 1500 °C. Then characterization of functional group analysis with FTIR with range 400-1400 cm⁻¹, crystal structure and phase composition with XRD, fiber morphology with optical microscope and SEM. SiC as semiconductor is made in composite form, composite solution is made with polymer matrix of poly
vinyl alcohol (PVA). 10% PVA is stirred for 12 hours with stirrer at 80 °C. In Preparation of composites, SiC fibers are added in matrix solution. Then let stand until dry then composite heated up to 70 °C. Composite testing as a semiconductor is done by testing the electrical conductivity with LCR Meter.

3. Result and Discussion
The PCS fibers obtained from the electrospinning process were characterized with FTIR and the results showed in Figure 3. The assignment for structural bond in PCS already showed in three major structural bonds exist in PCS, they are: Si-C, Si-CH$_2$-Si and Si-CH$_3$ bonds. These Si-C, Si-CH$_3$ and Si-CH$_2$-Si bonds are important due to the relationship of oxygen control which is introduced in the subsequent curing process. The C –H and Si-H bonds are the side groups of PCS.

![Figure 3](image_url)

**Figure 3.** FTIR spectra of PCS fiber

The fiber diameter changes with different applied voltage and different pyrolysis temperature showed in Fig 4. The average fiber diameter as-received from electrospinning process were decrease with increasing the applied voltage during electrospinning process. The average fiber diameter result from applied voltage for 10 kV were at 10.4 μm comparable to other result at 9.5 ± 1.5 μm. [8] Higher voltage applied during electrospinning the fibers diameter were decreased. The smallest fibers diameter was at 5.862 μm with applied voltage at 17 kV.

The thermal curing was applied to the PCS fibers to achieve high order crosslinking and to increase the integrity of fiber. The oxygen atmosphere interacts with the fibers during curing. The fibers evaporate small amount H$_2$ gases and the sites that were abandoned by hydrogen would be occupied by oxygen beside crosslinking of Si-C or Si-Si occurs. [9]. The average diameter of fiber after pyrolys is with temperature variation at 1200, 1300 and 1400 °C were plotted in Fig 4, has a tendency to become smaller with an increase of temperature in all applied voltage. During pyrolysis there were a lot of H$_2$, CH$_4$ or O$_2$ gases that released. Higher temperature would increase the production rate of gases. The reactive of Si and C would interact during the pyrolys is in the inert atmosphere.
Figure 4. The average fiber diameter changes that processed with different applied voltage (a) 10 kV, (b) 15 kV and (c) 17 kV.

The XRD results in Fig. 5 shows that the crystalline SiC phase only formed at sample pyrolyzed at 1400 °C. Below this temperature there were only amorphous SiC that formed. As there showed a peak between 30-40° that indicating to β-SiC (111) has been formed, as well as a low peak between 60-70° indicating β-SiC (220). From the data also seen the increase of phase crystallinity with increasing temperature as seen from the greater its intensity and the sharp peak.
Figure 5. The difraction of SiC in variation temperature of pyrolysis

Figure 6 shows the SEM image results explain the morphology of the fibers, which are pipe-shaped fibers, have a good uniformity and can retain their shape by increasing pyrolysis temperatures. From the image is also visible fiber without fracture and showed a minimal pore on the surface.

Figure 6. SEM image results of SiC Fibers pyrolyzed at (a) 1200 °C, (b) 1300 °C and (c) 1400 °C.

Assuming the chemical structure was Si-O-C, the result that derived from EDS currently SiO_{1.911}C_{0.802}. The oxygen contents were increased caused by the oxidation during the curing process carried out in the air resulting the replacement of Si-CH\textsubscript{3} bond with Si-O-Si and Si-O-C, which causes the percentage of carbon to be reduced and the oxygen content increases in the fiber.

The composites were take placed for electrical characteristic using the LCR meter. The measurements with the LCR meter were done at frequencies ranged from 1 to 100,000 Hz. The interesting phenomena of electrical conductivity of composite was in the range 0 to 80 Hz as seen on Fig 7. Its showed constant value until 10 Hz and the composite processed with 10 kV and pyrolysis at 1200 °C showed electrical conductivity value decreasing almost 10 %. The electrical conductivity value at frequencies 1 to 1000 Hz, the area of electrical conductivity values ranges from 8×10\textsuperscript{-6} to 7×10\textsuperscript{-6}. The SiC fiber produced in all variations of voltage and pyrolysis temperature was a semiconductor material. Increase in voltage on the electrospinning process and the increase in temperature in the pyrolysis process, the electrical conductivity value tends to increase, seen in sample with a voltage of 17 kV and temperature 1400 °C has the highest electrical conductivity value with increasing frequency.
4. Summary
SiC fibers by electrospinning method have been successfully synthesized, as evidenced by the PCS functional groups resulting from the FTIR characterization, and also results of PCS fibers characterized by XRD, SEM and EDS after curing and pyrolysis. The results show that SiC crystalline phase has been formed and morphology of the fibers, which are pipe-shaped fibers, have a good uniformity and can retain their shape by increasing pyrolysis temperatures. From the image is also visible fiber has a minimal fracture and minimal pore. SiC fiber morphology is known of chances fiber diameter. The average fiber diameter as-received from electrospinning process were decrease with increasing the applied voltage during electrospining process. The average diameter of fiber after pyrosisys with temperature variation at 1200, 1300 and 1400 °C has a tendency to become smaller with an increase of temperature in all applied voltage. The electrical conductivity value at frequencies 1 to 1000 Hz, the area of electrical conductivity values ranges from $8 \times 10^{-6}$ to $7 \times 10^{-6}$. The SiC fiber produced in all variations of voltage and pyrolysis temperature was a semiconductor material. Increase in voltage on the electrospinning process and the increase in temperature in the pyrolysis process, the electrical conductivity value tends to increase.

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