SiO$_2$/C Composite as a High Capacity Anode Material of LiNi$_{0.8}$Co$_{0.15}$Al$_{0.05}$O$_2$ Battery Derived from Coal Combustion Fly Ash

Arif Jumari $^{1,2}$, Cornelius Satria Yudha $^{1,2}$, Hendri Widiyandari $^{2,3}$, Annisa Puji Lestari $^{1,2}$, Rina Amelia Rosada $^{1,2}$, Sigit Puji Santosa $^4$ and Agus Purwanto $^{1,2,*}$

$^1$ Department of Chemical Engineering, Faculty of Engineering, Universitas Sebelas Maret, Jl. Ir. Sutami 36 A, Surakarta, Central Java 57126, Indonesia; arifjumari_ft@staff.uns.ac.id (A.J.); corneliussatriayudha@gmail.com (C.S.Y.); annisa.pl@student.uns.ac.id (A.P.L.); rosadaameliarina@gmail.com (R.A.R.)

$^2$ Centre of Excellence for Electrical Energy Storage Technology, Universitas Sebelas Maret, Jl. Slamet Riyadi, 435, Surakarta, Central Java 57146, Indonesia; hendrividiyandari@staff.uns.ac.id

$^3$ Department of Physics, Faculty of Mathematic and Natural Science, Universitas Sebelas Maret, Jl. Ir. Sutami 36 A, Surakarta, Central Java 57126, Indonesia

$^4$ Department of Mechanical Engineering, Institut Teknologi Bandung, Jl. Ganesha No.10, Bandung 40132, Indonesia

* Correspondence: aguspurwanto@staff.uns.ac.id

Received: 30 October 2020; Accepted: 25 November 2020; Published: 26 November 2020

Abstract: Abundantly available SiO$_2$ (silica) has great potential as an anode material for lithium-ion batteries because it is inexpensive and flexible. However, silicon oxide-based anode material preparation usually requires many complex steps. In this article, we report a facile method for preparing a SiO$_2$/C composite derived from coal combustion fly ash as an anode material for Li-ion batteries. SiO$_2$ was obtained by caustic extraction and HCl precipitation. Then, the SiO$_2$/C composite was successfully obtained by mechanical milling followed by heat treatment. The samples were characterized by X-ray diffraction (XRD), Fourier transform infrared spectroscopy (FTIR), and scanning electron microscopy (SEM). Electrochemical properties were tested using an 18650 cylindrical cell utilizing LiNi$_{0.8}$Co$_{0.15}$Al$_{0.05}$O$_2$ (NCA) as the counter electrode. Based on the obtained results, the physicochemical characteristics and electrochemical performance, it was determined that SiO$_2$/C composites were greatly affected by the temperature of heat treatment. The best result was obtained with the SiO$_2$ content of 10% w/w, heating temperature of 500 °C, initial specific discharge capacity of 586 mAh g$^{-1}$ at 0.1 C (1 C = 378 mAh g$^{-1}$), and reversible capacity of 87% after 20 cycles. These results confirmed that the obtained materials had good initial discharge capacity, cyclability, high performance, and exhibited great potential as an anode material for LIBs.

Keywords: anode material; composite material; lithium-ion batteries; SiO$_2$/C

1. Introduction

In recent decades, energy shortage and environmental pollution have become serious issues along with the world population growth. Extensive research on renewable energy sources has been rapidly performed. Current renewable energy cannot be separated from energy storage systems. Lithium-ion batteries (LIBs) have become the main candidates for a wide range of applications owing to their fast charging, long lifespan, low cost, high energy density, and ability to be used repeatedly with little degradation in performance [1–5]. Currently, graphite is widely used as a common anode material in the industry owing to its advantage of long lifespan and low cost [6–8]. In addition to
graphite, Li$_4$Ti$_5$O$_{12}$ or LTO has also become a promising candidate because it has fast charging and long shelf life [9,10]. However, both of them exhibit low storage capacity (graphite = 372 mAh g$^{-1}$ and LTO = 160 mAh g$^{-1}$), which has become a huge obstacle to further application, especially for electric vehicles and electronic devices [1,11]. Various different anode materials with higher capacities have been studied as a candidate of the next generation of anode material. Among the various anode materials studied, Si is considered as the most promising anode material owing to its high theoretical capacity (4200 mAh g$^{-1}$), moderate operating voltage (0.1–0.5 V vs. Li/Li$^+$), and low stable plateau potential [12,13]. Unfortunately, Si anode materials experience some challenges such as low electrical conductivity, large volume expansion (>300%) during lithiation and delithiation, and an unstable solid electrolyte interphase (SEI), which leads to poor cycling performance [1,9]. Therefore, silicon suboxide or SiO$_x$ (x < 2) has been suggested as an alternative anode material. Even though SiO$_x$ has a smaller theoretical specific capacity (1961 mAh g$^{-1}$) than Si, it exhibits better cycling stability, smaller volume expansion, lower cost, and eco-friendliness. SiO$_x$ also has a drawback of poor conductivity and low Coulombic efficiency at the initial charge–discharge cycle. On the other hand, it is difficult to produce SiO$_x$ owing to the partial oxidation of silicon or semi-reduction of SiO$_2$, which require complex and convoluted synthesis steps. It is advantageous to use SiO$_2$ as an anode material owing to its high availability and because it can be easily extracted from biomass-based waste. Similar to SiO$_x$, SiO$_2$ has low conductivity, which needs to be improved before being applied in full batteries [14,15].

Many approaches have been tried to overcome these problems such as inserting Li into SiO$_2$ and modifying the size of SiO$_2$ [14]. However, one of the most effective ways is to use composite SiO$_2$ with carbon. The composite improves electrical conductivity and also reduces volume expansion during the charge–discharge cycling to maintain a stable electrochemical performance [16]. For example, it has been reported that the SiO$_2$/C composite showed an impressive reversible capacity of 635.7 mAh g$^{-1}$ at the discharge current of 100 mA g$^{-1}$ [17]. The 3D SiO$_2$@graphene aerogel composite shows a reversible capacity of 300 mAh g$^{-1}$ at the discharge current of 500 mA g$^{-1}$ [18]. However, the SiO$_2$-carbon (SiO$_2$/C) composite preparation for anode material usually requires many complex steps such as additional processes to modify the morphology, expensive silicon precursor, e.g., Tetraethyl orthosilicate (TEOS), pretreatments [18] and multiple high temperature and long heating process [17], which greatly hinders further improvement. Thus, these additional processes will directly affect the overall production cost of SiO$_2$/C anode material. The use of cheap raw materials, especially from waste, and a simple processing method reduce the production cost in a significant amount. Hence, it is desirable to design a facile method to fabricate SiO$_2$/C composites to realize high-performance anode materials for LIBs.

In this article, we reported a facile and inexpensive method for synthesizing the SiO$_2$/C composite from coal combustion-derived fly ash for active anode material by mechanically milling SiO$_2$/C and heat-treating it in an inert atmosphere. The use of silica from coal fly ash is a sustainable approach to recycle, reduce, and reuse industrial waste. SiO$_2$/C was directly used as the anode material for the LiNi$_{0.8}$Co$_{0.15}$Al$_{0.05}$O$_2$ battery. This approach has never been studied before.

2. Materials and Methods

2.1. Synthesize of the SiO$_2$/C Composite from Coal Fly Ash

Coal combustion-derived fly ash was provided by PT Semen Indonesia and was stored in an oven. The chemical composition of fly ash is shown in Table 1. A total of 50 g of coal fly was extracted using 2 L of 2-M NaOH (Merck, Darmstadt, Germany). The mixture was heated at 90 °C for 4 h under stirring at 300 rpm. The separation of filtrate and solid particles was done by gravity filtration followed by the removal of the filter cake. SiO$_2$ particles were obtained by adding 10 N HCl (Merck, Darmstadt, Germany) to the filtrate until pH 6–7. The resulted precipitates were filtered, washed using hot demineralized water three times, and dried under oven at 100 °C for 12 h. Then, the SiO$_2$/C composite was successfully obtained through mechanical milling of SiO$_2$/C with various SiO$_2$ composition using a ball mill for 2 h and heating at 500 °C for 30 min under argon flow. Variations in heat treatment
were applied to the optimized composite and performed at 400–600 °C under the same conditions. The overall process of SiO$_2$/C composite synthesis can be seen in Figure 1.

Table 1. XRF analysis of coal combustion-derived fly ash from PT Semen Indonesia.

| Component | O | Si | Fe | Ca | Al | Na | Mg | K | S | Ti | Cl | P | Mn | Trace Metals |
|-----------|---|----|----|----|----|----|----|----|----|----|----|----|----|-------------|
| wt%       | 36.6 | 20.4 | 18.1 | 9.4 | 4.0 | 3.5 | 3.1 | 1.2 | 1.1 | 0.7 | 0.5 | 0.4 | 0.3 | 0.7         |

Figure 1. Process flow diagram of SiO$_2$ and SiO$_2$/C composite synthesis.

2.2. Material Characterizations

The crystallographic structure of materials was studied by X-ray diffraction (XRD, Rigaku Miniflex 600 Benchtop XRD). The compositional analysis of materials was performed with Fourier transform infrared spectroscopy (FTIR, Shimadzu IRPrestige-21) and X-ray fluorescence (Bruker XRF Spectrometer, Germany). The morphologies of the materials were investigated by scanning electron microscopy (SEM, JSM-6510LA).

2.3. Electrochemical Measurements

Electrochemical measurements were performed using a cylindrical cell (18650) assembled in an argon-filled glove box. LiNi$_{0.8}$Co$_{0.15}$Al$_{0.05}$O$_2$/NCA (MTI, CA, USA) was used as the counter electrode; monolayer polypropylene film (Celgard 2400) was used as the separator, and 1-mol/L LiPF$_6$ dissolved in a mixture of ethylene carbonate and diethyl carbonate (EC/DEC = 1:1, v/v) was used as the electrolyte. The working electrode was prepared with active materials, acetylene black, carboxymethyl cellulose (CMC), and styrene–butadiene rubber (SBR) obtained from MTI, CA, USA with the weight ratio of 93:1:1:5 in deionized water. The mixture was stirred for 5 h at room temperature; then, it was coated onto both sides of copper foil using the doctor blade method. The copper foil was dried at 80 °C for 30 min in a vacuum oven. The load mass of active material was approximately 18 mg/cm$^2$. The overall assembly process of cylindrical Li-ion cells is described elsewhere [19,20].

The electrochemical performance of materials was studied by galvanostatic charge–discharge cycling tests using a battery testing system (Neware BTS 6000, Neware Electronic Co., Shenzen, China) at various current rates (1 C = 372 mAh g$^{-1}$) and room temperature (25 °C).

3. Results

3.1. SiO$_2$ Characterization

Figure 2 shows the as-prepared SiO$_2$ powder obtained from fly ash characterization by XRD (Figure 2a), FTIR (Figure 2b), SEM (Figure 2c), and XRF (Figure 2d). Based on the XRD pattern, the widening peak is detected at 15–35° of 2 theta; thus, it can be concluded that the samples exhibit amorphous properties. The impurity peaks (31° and 45°) of sodium chloride (NaCl) are detected in the
The presence of impurities can be caused by an unfinished cleaning process or salt entrapment in silica matrices. In addition to the impurity peak, all peaks from all samples are well indexed to ICSD Card no. 061662. Based on the FTIR spectra, the absorption peaks at 3400, 1630, and 950 cm$^{-1}$ are attributed to the stretch vibration of –OH from silicanol (Si–OH) or H$_2$O and Si–O from siloxane (Si–O–Si). The peak at 790 cm$^{-1}$ is attributed to symmetrical Si–O from Si–O–Si stretching vibration. Based on the XRD and FTIR result, SiO$_2$ is successfully obtained by caustic fusion and precipitation through following reactions [21].

\[
\begin{align*}
\text{SiO}_2(\text{s}) + 2\text{NaOH}(\text{aq}) & \rightarrow \text{Na}_2\text{SiO}_3(\text{aq}) + \text{H}_2\text{O}(\text{l}) \\
\text{Na}_2\text{SiO}_3(\text{aq}) + 2\text{HCl}(\text{aq}) & \rightarrow \text{H}_2\text{SiO}_3(\text{aq}) + 2\text{NaCl}(\text{aq}) \\
\text{H}_2\text{SiO}_3(\text{aq}) + \text{H}_2\text{O}(\text{aq}) & \rightarrow \text{Si}(_4\text{OH})(\text{s}) \\
\text{Si}(_4\text{OH})(\text{s}) & \rightarrow \text{SiO}_2(\text{s}) + 2\text{H}_2\text{O}(\text{g})
\end{align*}
\]

SEM images (Figure 2c) confirm that there are two types of particles, i.e., primary and secondary. The secondary particle has a diameter of ~7.5 µm, which consists of clustering sub-micron-sized primary particles and small adherents on the surface. These adherents or aggregates enhanced the overall surface area of silica, which resulted in good ionic transfer during Li insertion. The XRF result shows that mostly silicon was obtained, while there are also small sodium impurities (6%), which is confirmed by the XRD result. The NaCl impurities is still detected due to the entrapment of NaCl during the silica precipitation process and exist even after multiple washing steps.
The as-obtained SiO$_2$ powder was directly applied as anode in a NCA/SiO$_2$ battery. Figure 3 shows the electrochemical performance of the cell. The theoretical capacity of SiO$_2$ is adapted from a study by Liu et al. (1965 mAh/g), while the theoretical capacity of NCA is estimated at 200 mAh/g. In the final cell assembly, the SiO$_2$ anode was selected as the limiting reactant; therefore, the specific capacity was referred to as the specific capacity of SiO$_2$. Figure 2a shows that the specific charge and discharge capacity of SiO$_2$ is approximately 1396 mAh/g and 1188 mAh/g, respectively. The discharge capacity was maintained for 10 cycles. Even though SiO$_2$ has a considerably lower specific capacity than its theoretical capacity, the capacity was stable at 1/10 C. At increasing rate, as shown in Figure 2b, SiO$_2$ exhibits an inferior rate performance and high level of irreversibility. This can be caused by the formation of electrochemically inactive lithiated SiO$_2$ at high rate (>0.5 C). Previous studies claimed that the reaction mechanism between Li-ion and SiO$_2$ was not been completely understood yet; however, most studies showed a significant irreversible capacity loss during initial cycling [16]. This phenomenon directly indicates the importance of compositing SiO$_2$ with a more stable and highly conductive material such as artificial graphite or Mesocarbon microbeads (MCMB) [17,22].

![Figure 3](image1)

**Figure 3.** (a) Charge discharge curve and (b) ratability of SiO$_2$ as the LiNi$_{0.8}$Co$_{0.15}$Al$_{0.05}$O$_2$ (NCA) battery anode.

### 3.2. Effect of SiO$_2$ Composition in the SiO$_2$/C Composite on Its Structure and Coulombic Capacity

As-prepared SiO$_2$ was mechanically milled with MCMB and heated at 500 °C for 30 min under flowing argon. Figure 4 shows the XRD analysis of the SiO$_2$/C composite with various SiO$_2$ wt%. Because as-obtained SiO$_2$ is in an amorphous form, the diffraction peaks of graphite are dominant in all samples, and the SiO$_2$ peak is not detected; however, there is an increase in the peak width, specifically at 2 theta of 27°, which may be caused by an increase in the amount of amorphous silica (SiO$_2$).

![Figure 4](image2)

**Figure 4.** Electrochemical performance of SiO$_2$ in the NCA battery.
The initial charge/discharge curves of the samples at different SiO\(_2\) content at 1/10 C are shown in Figure 5a. It is easily observed that the specific capacity of the SiO\(_2\)/C composite depends on the SiO\(_2\) content in the composite. It is observed that the first capacity of the SiO\(_2\)/C composite considerably increased with an increase in the SiO\(_2\) content. However, when the content of SiO\(_2\) was greater than 10\%, the capacity was insignificantly increased.

![Graphs showing charge/discharge curves](image)

**Figure 5.** (a) Initial charge/discharge curve of the samples at different SiO\(_2\) content at 0.05 C; (b) magnified charge–discharge curve of the samples; (c) cycling performance of the samples at different SiO\(_2\) content; (d) rate and cycling performance of the SiO\(_2\)/C 10\% composite.

Table 2 shows that all samples have greater capacity than batteries that contain carbon materials (340 mAh g\(^{-1}\)) (0.1 V vs. Li/Li\(^+\)) [1]. Increased capacity occurs because SiO\(_2\) (1961 mAh g\(^{-1}\)) acts as an active material in composites and is responsible for increasing battery capacity. The increased capacity also indicates that SiO\(_2\) used in the composite exhibits electrochemical activity [16–18]. Based on previous reports, the small amount of NaCl within silica matrices can potentially enhance the electrochemical performance of LIBs. Borong Wu et.al. claimed that 1\% NaCl in electrolyte of Li-Graphite cell can improve the SEI formation on graphite surface which enhance the cycling performance. Study by Ikhsanudin et al. had stated that NaCl can improve the capacity of NCA cathode. Since the specific discharge capacity listed in Table 2 exceeds the capacity of pure graphite, we can conclude that the effect of a small amount of NaCl is still insignificant compared to the effect of silica composite on the graphite anode. However, the effect of NaCl additive on NCA/Graphite cell can be furtherly studied in future research [23,24].
Table 2. Initial specific capacity of the SiO$_2$/C composite.

| Content of SiO$_2$ (wt%) | Initial Specific Capacity Based on the Anode (mAh g$^{-1}$) | Columbic Efficiency (%) |
|-------------------------|---------------------------------|------------------------|
| 0                       | 340                             | 85                     |
| 1                       | 356                             | 83                     |
| 3                       | 365                             | 80                     |
| 5                       | 423                             | 78                     |
| 10                      | 541                             | 76                     |
| 30                      | 552                             | 72                     |

However, an increase in the SiO$_2$ content in the anode material will also result in a decrease in battery stability (cyclability). Figure 5c shows that the reversible capacity of SiO$_2$/C 30% remains at 51% after 15 cycles. This is mostly due to the unstable solid electrolyte interphase (SEI) on the surface of Si [25], which causes lithium to be trapped in the active surface of Si and results in the rapid loss of irreversible capacity and low initial Coulombic efficiency (CE) [16–18]. These reactions that occur in the process can be summarized as follows:

$$2\text{SiO}_2 + 4\text{Li}^+ + 4e^- \rightarrow \text{Li}_4\text{SiO}_4 + \text{Si}$$ \hspace{1cm} (5)

$$\text{SiO}_2 + 2\text{Li}^+ + 2e^- \rightarrow \text{Li}_2\text{O} + \text{Si}$$ \hspace{1cm} (6)

$$\text{Si} + x\text{Li} + xe^- \rightarrow \text{Li}_4\text{Si}$$ \hspace{1cm} (7)

Reactions (2) and (3) result in the generation of irreversible capacity, which leads to capacity loss. In addition, Li$_4$SiO$_4$ compounds are reported to be electrochemically inactivated. However, Li$_4$SiO$_4$ and Li$_2$O act as buffer components and accommodate volume expansion that occurs in Li$_x$Si during lithiation and delithiation. This occurs because Li$_4$SiO$_4$ (2.39 g cm$^{-3}$) and Li$_2$O (2.02 g cm$^{-3}$) are denser materials than Li$_x$Si. The presence of oxide compounds shows a significant decrease in the volume expansion compared to when the material is homogeneous Li$_x$Si [16,26,27].

The rate performance of the SiO$_2$/C 10% composite was further examined, and the results are shown in Figure 5d; the samples tested at the discharge current of 0.1, 0.5, and 1 C provided the best performance. It is clearly observed that the sample still has a high specific capacity of approximately 395 mAh g$^{-1}$ at the current rate of 1 C. Figure 5c also shows that the reversible capacity slowly increased from 541 mAh g$^{-1}$ to 601 mAh g$^{-1}$ after 3 cycles and was stable after the first 15 cycles. The latest studies explain that an increase in reversible capacity is due to SiO$_2$, which is not fully active at the beginning of cycling and will be active in the next cycling [9,10,24]. Figure 5d shows that the sample has high stability. It was possible to obtain the reversible capacity at 87% after 31 cycles at 1 C and CE was always above 98%. These results confirm that the SiO$_2$/C composite with 10 wt% SiO$_2$ exhibited the best performance owing to its excellent specific capacity, good cyclability, and high performance, and showed considerable potential as an anode material for LIBs.

3.3. Effect of Heat Treatment on the SiO$_2$/C Composite

Figure 6a shows the FTIR spectra of SiO$_2$ in the composite after heat treatment at 400, 500, and 600 °C. The FTIR spectra of SiO$_2$/C show similar results at various heat treatment temperatures. The peaks at 3432, 3434, and 3433 cm$^{-1}$ are assigned to the –OH stretching in SiO$_2$/C heat-treated at 400, 500, and 600 °C, respectively. Furthermore, the peak at approximately 1600 cm$^{-1}$ indicates the presence of an –OH bending band. This hydroxide group indicates the presence of water molecules [15]. It is observed that an increase in the heat treatment temperature will decrease the water content in SiO$_2$/C (Figure 6b). The presence of SiO$_2$ is detected as the Si–O–Si stretching vibration of the sharp peak at approximately 1100 cm$^{-1}$ [28,29]. SiO$_2$/C heat-treated at 600 °C shows smaller transmittance than SiO$_2$/C heat-treated at 400 and 500 °C, specifically for the peak at approximately 1100 cm$^{-1}$. On the
other hand, SiO$_2$/C heat-treated at 600 °C has better transmittance than SiO$_2$/C heat-treated at 400 and 500 °C for the peak at approximately 400 cm$^{-1}$. It can be concluded that heat treatment affects the SiO$_2$ surface chemistry. The FTIR spectra of graphite also show peaks at approximately 3400 and 1600 cm$^{-1}$, which indicates the presence of water molecules in graphite, as previously described.

![FTIR spectrum of composite SiO$_2$/C 10% heated at 400, 500, and 600 °C and (b) Magnified zone at hydroxyl peak.](image)

**Figure 6.** (a) FTIR spectrum of composite SiO$_2$/C 10% heated at 400, 500, and 600 °C and (b) Magnified zone at hydroxyl peak.

Figure 7 shows the XRD patterns of SiO$_2$/C at various heat treatment temperatures. Based on the figure, all samples exhibit a graphite structure (JCPDS PDF 75-2078) owing to low content and crystallinity of silica compared to that of artificial graphite. The crystallite size of the samples, which is determined by the Scherer equation [19,30], is affected by the heat treatment temperature; it increased along with an increase in temperature, although heat treatment was performed for only 30 min.

![XRD pattern of composite SiO$_2$/C heated at 400, 500, and 600 °C.](image)

**Figure 7.** XRD pattern of composite SiO$_2$/C heated at 400, 500, and 600 °C.
The SEM characterization of artificial graphite and SiO$_2$/C composite heated at various temperatures is shown in Figure 8. Most graphite particles have a spherical shape with ~27-µm size. Flakes of carbon were observed between spherical particles. The abovementioned figure shows that SiO$_2$/C has rough surface morphology, owing to the presence of 10% of SiO$_2$ aggregates. SiO$_2$ secondary particles break into micron-sized primary particles during the ball milling process. The average primary particle size of SiO$_2$ on the graphite surface heated at 400, 500, and 600 °C is 344, 399, and 450 nm, respectively. Moreover, with an increase in temperature, the average particle size of SiO$_2$/C increases, owing to the agglomeration of particles [9,31].

![Figure 8](image_url)

**Figure 8.** SEM images (500× and 10,000× magnification) of graphite (a,b) and 10% SiO$_2$/C composite with heat treatment at (c,d) 400 °C, (e,f) 500 °C, and (g,h) 600 °C.

Table 3 shows that all samples have greater specific capacity than batteries using carbon materials. Compared to graphite, all samples have higher initial discharge capacity with an increase in temperature. Thus far, it can be concluded that with an addition of SiO$_2$ and increase in the heat treatment temperature, all of our samples exhibit high initial capacity; this improvement is achieved using an easy and simple method.
Table 3. Initial specific capacity of the SiO$_2$/C composite depending on the heat treatment temperature.

| Heat Treatment Temperature ($^\circ$C) | Initial Specific Capacity (mAh g$^{-1}$) | Columbic Efficiency (%) |
|----------------------------------------|------------------------------------------|-------------------------|
| 400                                    | 506                                      | 74                      |
| 500                                    | 586                                      | 76                      |
| 600                                    | 705                                      | 82                      |

The initial charge/discharge curve of the samples at different heat treatment temperature of SiO$_2$/C is shown in Figure 9a. Based on the cyclability and rate ability of the cell, 500 $^\circ$C is the optimum temperature to obtain the SiO$_2$/C composite. It is predicted that at higher temperature, the SiO$_2$/C composite is agglomerated into larger sizes, which lowers the diffusion kinetics of Li-ion during the charge–discharge process. At low temperature, the composite process did not completely occur.

Figure 9. (a) Initial charge/discharge curve of the SiO$_2$/C composite at 0.1 C; (b) cycling performance of the SiO$_2$/C composite; (c) rate performance of the SiO$_2$/C composite at various temperatures.
The rate performance of the SiO$_2$/C composite depending on heat treatment variation is shown in Figure 9c. The samples were tested at the charge and discharge current of 0.1, 0.5, and 1 C. It is clearly observed that the samples treated at various temperatures have a high specific capacity of approximately 410 mAh g$^{-1}$ at the current rate of 1 C. Figure 9c also shows that the reversible capacity increases from 581 mAh g$^{-1}$ to 595 mAh g$^{-1}$ after two cycles at the heat treatment temperature of 500 °C. The latest research explains that an increase in reversible capacity owing to SiO$_2$ is not fully active at the beginning of cycling and will be active during next cycling [14,16,17]. The obtained results confirm that the SiO$_2$/C composite heat-treated at 500 °C produces the best results, owing to its excellent specific capacity, good cyclability, and high performance as anodes for LIBs [18,32,33]. Based on Table 4, the overall result is considered good since the starting material is far more inexpensive and the process is simple compared to previous reports.

### Table 4. Comparative study of SiO$_2$ based anode electrochemical performance.

| Precursors     | Product               | Methods       | Voltage (V) | Initial Specific Capacity (mAh/g) | Retention Capacity | Rate Performances Ref. |
|----------------|-----------------------|---------------|-------------|-----------------------------------|--------------------|------------------------|
| TEOS           | SiO$_2$/Graphene       | Hydrothermal  | 0–3 V       | 453 (half cell)                  | ~100% (500 cycles) | 103 mAh/g (5 A/g)       | [18]                   |
| TEOS           | SiO$_2$/C Hollow sphere| Precipitation | 0–3 V       | 400 (half cell)                  | 421% (160 cycles)  | -                      | [17]                   |
| TEOS           | SiO$_2$/C nanorods     | Precipitation | 0–3 V       | 498 (half cell)                  | 95% (100 cycles)   | 345 mAh/g (1 A/g)       | [33]                   |
| SiO$_2$ nanoparticle | Lithiated SiO$_2$   | Heat treatment| 0–3 V       | 1859 (half cell)                 | 70% (50 cycles)    | 100 mAh/g (~2 A/g)      | [14]                   |
| Coal derived  | SiO$_2$/Graphite       | Milling-Heat Treatment | 2.7–4.2 V | 541 (full-cell)                  | 87% (20 cycles)    | 410 mAh/g (572 mAh/g)   | This Work              |

4. Conclusions

In this study, a high-performance SiO$_2$/C composite was obtained by extracting SiO$_2$ from coal fly ash, mechanical milling of SiO$_2$-C at various SiO$_2$ content, and heating SiO$_2$-C composites at 400–600 °C. SiO$_2$/C composites exhibit specific gravity that is higher than that of carbon materials, which confirms that SiO$_2$ from fly ash acts as an active material and exhibits electrochemical activity. SiO$_2$/C composites with 10% of SiO$_2$ and heat treatment temperature of 500 °C shows the best result with the initial specific capacity of 586 mAh g$^{-1}$ at 0.1 C and the reversible capacity of 87% after 20 cycles. This method is advantageous for mass production owing to its low cost. Furthermore, the obtained SiO$_2$-C composites exhibit excellent reversible capacity, high initial specific capacity, good cyclability, and high performance of the anode. Based on these results, SiO$_2$ from coal fly ash is a great candidate for the anode material for LIBs.

**Author Contributions:** Conceptualization, H.W., S.P.S. and A.J.; data curation, A.P.L., R.A.R.; formal analysis, H.W., C.S.Y.; investigation, A.J., A.P. and H.W.; methodology, A.J., A.P.L., R.A.R.; project administration, A.P.; supervision, A.P.; validation, S.P.S., A.J. and A.P.; visualization, C.S.Y.; writing—original draft, A.J. and C.S.Y.; writing—review and editing, A.P. All authors have read and agreed to the published version of the manuscript.

**Funding:** This research is financially supported by the Indonesian Ministry of Education and Culture through World Class Research scheme with contract No. 112/UN27.21/HK/2020. This research is also partially funded by the Indonesian Ministry of Research and Technology/National Agency for Research and Innovation, and Indonesian Ministry of Education and Culture under World Class University Program managed by Institut Teknologi Bandung (contract No: 1534/C5/KB.07.02/2019).

**Acknowledgments:** We acknowledge PT. Semen Indonesia for providing the fly ash.

**Conflicts of Interest:** The authors declare no conflict of interest.
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