Synthesis the SiO Powders by a New Method

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Abstract. Lithium ion batteries with the advantages of the long cycle life, high energy density, high power density etc. have been widely used in mobile phones, notebook computers and other areas of the power. The SiO is one of the most promising materials of lithium ion battery cathode material because of the theoretical specific capacity more than graphite 10 times. While traditional method, the chemical vapor deposition method has been unable to meet the increasing demand for the SiO material. In this paper, the SiO powders have been synthesized by a new method which is named Crucible Rotating Up method. The technological process of this method has been introduced in this paper and the X-ray diffraction analysis and SEM-EDS have been performed for the test samples. The result shows that the high purity SiO gas has been generated in the Crucible Rotating Up Furnace.

1. Introduction
As the global coal, oil and other traditional energy increasingly scarce materials, people are trying to develop new energy to replace traditional fossil energy. As a kind of important chemical power supply, the lithium ion battery (LIB) has been widely utilized as an energy storage device in many electronic applications due to its favorable energy density and cycle performance. Especially, in recent years, with the rapidly growing market for the electric vehicle (EV), hybrid electric vehicle (HEV) as well as stationary battery storage systems, the need for LIBs is thus shifting to larger scale applications. LIBs will further replace the lead-acid battery and nickel cadmium battery because of the current impact on the environment[1-6]. In recent years, along with the continuous renewal of electronic devices, a new generation of high specific capacity lithium ion battery research and development has become increasingly urgent[7-10]. At present, the low-cost and high energy-density electrode materials are critical for high-energy LIBs, whereas the conventional anode materials are primarily graphitic anode materials with a limited capacity(370mAhg−1) [11-13]. Therefore, there is an urgent need to develop an alternative to the graphite material type lithium ion battery anode materials with high capacity. Among the promising anode electrodes, Si has been attracting much attention because of its abundance and highest gravimetric specific capacity of 4200 mAhg−1[14, 15]. However, Si suffers from dramatic pulverization and rapid capacity decay caused by the unavoidable large volume changes during the charge and discharge processes [16-18]. Silicon monoxide is one of the most attractive anode materials because of its relatively high theoretical capacity of 2400mAhg−1, which is greater than that of conventional graphite electrodes[3].The theoretical specific capacity of the SiO material is high and the cycle performance is poorer for volume effect, but the cycle stability of the graphite is good and...
the specific capacity is low. So the organic combination of the SiO material and the graphite can effectively improve the battery energy density[18-20]. In a word, with the increase demand of high-power power supply, the demand of the SiO material application in lithium ion battery anode will greatly improve.

While traditional method, the method of chemical vapor deposition have been unable to meet the increasing demand for the SiO material[21]. To solve the problem of the SiO production, this paper provides a new synthesis method for the nanoscale SiO powder materials. The particle size of the SiO powder material prepared by the new method is small, the process flow is simple and this method can be industrialized mass and continuous production.

2. Materials and Methods

2.1. Material and the Principle Preparation of the SiO Powders

The 6N pure polycrystalline silicon powders and the quartz rod were used as starting materials and the reaction is as following: Si + SiO2—2SiO. The gaseous SiO powder should be synthesized according to the reaction of Si and SiO2 at high temperature in the Crucible-Rotating-Up Furnace (CRUF) [22]. Fig.1 is the schematic diagram of SiO powders preparation device.

![Figure 1. The schematic diagram of SiO powders preparation device](image)

2.2. X-ray Diffraction Analysis

The X-ray diffraction analysis (XRD) was carried out in the 2θ range from 10° to 90° by using the Philips XPERT-MED X-ray diffractometer with a conventional CuKα radiation.

2.3. SEM and EDS Analysis

A scanning electron microscope (SEM) incorporated with energy dispersive spectrometer (EDS) was used to detect the powders of the samples. The analysis was performed on a ZEISS-SUPRA55 scanning electronic microscope with an Oxford-AztecX-Max80 energy X-ray spectrometer. The working voltage was 5.0 KV and the working distance is 6.5 mm.
3. Results and Discussion

3.1. Synthesis the SiO Powders

The raw materials were weighed according to its stoichiometric composition after drying at 200°C for 3 hours. Then the cylindrical graphite crucible was be installed in the outer and the quartz crucible was be placed in the inner, as shown in the Fig.1. The polycrystalline silicon powders were loaded into the cylindrical quartz crucible and the quartz rod was fixed by the molybdenum clip. The diameter of the quartz rod is about 1/3 of the quartz crucible. Then the CRUF chamber was shut down and the vacuum pump was started. The program of temperature was executed with the rising at the rate of 200°C/h when the vacuum was 3 Pa. Then the argon gas switch was opened and the flow velocity of argon was 2L/min from the gas collection processing equipment. At the same time the end pump of SiO powder collection system was opened and the velocity of gas was set reasonably. The temperature was kept at 1450°C for several hours to ensure polycrystalline silicon powders was completely melted and to provide a stable temperature distribution. Then height of quartz rod was adjusted when the quartz rod and polycrystalline silicon melt contacted and the drop speed of quartz rod was maintained at 1-10mm/min, the rotation speed of quartz rod was 5-15r/min, the rotation speed of quartz crucible was 5-15r/min in the opposite direction with the quartz rod. The generated SiO gas flows along the drive direction of argon. Argon inflows from guide cylinder to the reaction interface of polycrystalline silicon melt and quartz rod. The mixed gas of argon and the SiO gas will be output in the molybdenum guide cylinder and enter to the water cooling deposition device and coolant filtering system. The end of the reaction process, the argon gas and the end pump operation have been maintained until the temperature reached the room temperature. Then the powder collection system was opened and the SiO powders were collected and encapsulated.

3.2. XRD Analysis

The X-ray diffraction analysis (XRD) was carried out for the samples in the 2θ range from 10° to 90° by using the Philips XPERT-MED X-ray diffractometer with a conventional CuKα radiation. The XRD patterns of SiO powders are presented in Fig.2. An amorphous diffraction peak is found in 20 to 30 degrees of the low diffraction angle region and the intensity of the amorphous diffraction peak attenuates gradually and becomes smooth, as shown in the Fig.2. It is obvious that the samples are the amorphous material.

![Figure 2. XRD patterns of the samples](image-url)
3.3. SEM and EDS Analysis
A scanning electron microscope (SEM) incorporated with energy dispersive spectrometer (EDS) was used to detect the powders of the samples. The analysis was performed on a ZEISS-SUPRA55 scanning electronic microscope with an Oxford-AztecX-Max80 energy X-ray spectrometer. The working voltage was 5.0 KV and the working distance is 6.5 mm. The SEM and EDS analysis for the samples, are shown in Fig. 3 and Fig. 4. Results of SEM and EDS analyses (Figs. 3, 4) show the surface morphology and composition. The O and Si elementals are distributed corresponding to the peak place 0.501keV and 1.755keV, respectively and the peaks of 0.261keV and 2.126 keV were ignored. Table 1 is the results from elemental microanalysis (SEM-EDS) of numbers. The results can be seen from Table 1 that the ratio of silicon atoms and oxygen atoms is about 0.628 between 0.5 and 1.0, and more close to 0.5. So it is concluded preliminarily that the ingredient of the samples is most of SiO$_2$ and a small amount of SiO powders. The reason is that the high purity of SiO powders are generated in the Crucible-Rotating-Up Furnace and the SiO gas enters the powder collection system along the molybdenum guide cylinder. The sealing of the powder collection system is not good maybe, mixed with O$_2$, thus under the condition of high temperature, the chemical reaction of SiO and O$_2$ generates the SiO$_2$. The results show that the SiO powders can be synthesized using the setting process parameters in the Crucible-Rotating-Up Furnace.

![Figure 3](image)

**Figure 3.** Micrograph of acquired Site of interest 1 showing embedded/ SEM micrographs specimens and spectrum acquisition targets (1–Spectrum 1)
Figure 4. Site of interest 1 spectra showing relative elemental abundance. See Fig. 3 for spectral locations.

Table 1. Results from elemental microanalysis (SEM-EDS) of numbers

| Element | Weight% | Atomic% |
|---------|---------|---------|
| O       | 47.56   | 61.42   |
| Si      | 52.44   | 38.58   |
| Total   | 100     | 100     |

4. Conclusions
In summary, the SiO powders have been synthesized by a new method in the Crucible-Rotating-Up Furnace (CRUF). The idea of the reaction device is derived from the crystal growth method of Czochralski and the method of Czochralski is referred to as pulling method in China. So the reaction device is named CRUF according to the characteristics of the device and the new method is named Crucible-Rotating-Up (CRU) method. The powder of SiO\(_x\) (x=1 or 2) can be produced by CRU method and this new method can be industrialized mass and continuous production and the process flow is simple.

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