Synthesis of Si-MWCNT nanocomposite using triple DC thermal plasma jet system

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Abstract: The Si-MWCNT(Multi wall carbon nanotube) nanocomposite which is considered to be a promising substitute for graphite as anode material of Lithium-ion batteries was synthesized by thermal plasma. The mass ratio of MWCNT and micron-sized Si powder as starting material was varied, Si and MWCNT crystalline was observed with silicon carbide in the product by X-ray diffraction. It was revealed that the silicon carbide bonding is formed between MWCNT surface and attached silicon nanoparticle.

Keywords: Synthesis, Thermal plasma, Silicon, MWCNT, nanocomposite, Silicon carbide

1. Introduction

Many efforts to improve the performance of Li-ion batteries have been studied in accordance with the continuing progress of portable electronic devices and electric vehicles and the increasing market scale of energy storage system. Carbon has been used as an anode material for Li-ion batteries, but small capacity of graphite have limitation to meet the growing energy demand, and new anode materials are required.

Among them, silicon is one of the most potential material due to the large deposits and the higher capacity of lithium ions than Carbon material. However, it has a critical disadvantage that the volume of silicon is expanded and finally cracks occur when charging and discharging are repeated. In addition, due to the decomposition reaction of the electrolyte in the battery, an unstable solid electrolyte interphase (SEI) layer is repeatedly formed on the silicon surface, resulting in rapid capacity drop and low efficiency [1].

In order to overcome these disadvantages, many studies have been tried to control the silicon morphology into a hole or wire structure [2]. However, the process is complicated and difficult to succeed, so we suggest a solution to synthesize silicon nanoparticles attached in carbon nanotube.

The high tensile strength of MWCNT prevents the volume expansion of silicon. In addition, nano-sized silicon particles are effective to minimize their volume change rate. Thermal plasma provides advantages over other synthesis processes. Since thermal plasma process does not require pre- or post-treatment, it is more environmentally friendly than wet methods. The temperature of the thermal plasma is generally over 10,000 K and the velocity is generally over few hundreds m/s. Therefore, it might be able to heterogeneous nucleation of silicon on the surface of carbon nanotube.

In this study, we aimed to find the optimal synthesis condition and understand the formation process by varying the mass ratio of Si and MWCNT and input power.

2. Experimental setup

Thermal plasma system used for Si-MWCNT synthesis consists of a power supply, triple plasma torch, powder feeder, reactors and cyclone filter. The mixed powder of micron sized Si and MWCNT was injected into the triple plasma jet as raw material. The detail of experimental condition is presented in Table 1. Total input power was set as about 19 kW. The mass ratio of Si and MWCNT was controlled in Exp. 1, 2, and 3 conditions. The mixed powder was injected under 0.5 g/min with Ar carrier gas.

Table 1. Experimental condition for synthesis of Si-MWCNT nanocomposite.

Experimental No.	Exp. 1	Exp.2	Exp.3
Total input power (kW)	18.6	19.1	19.9
Feeding rate of starting material (g/min)	0.32	0.31	0.5
Plasma forming gas (L/min)	N_2 (6), Ar (4)		
Mass ratio of injected powder (Si : MWCNT)	2 : 1	5 : 1	1 : 2

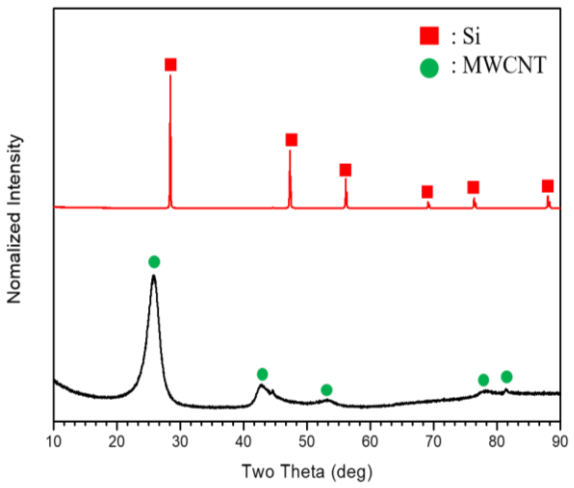

Fig. 1. XRD patterns of the raw materials

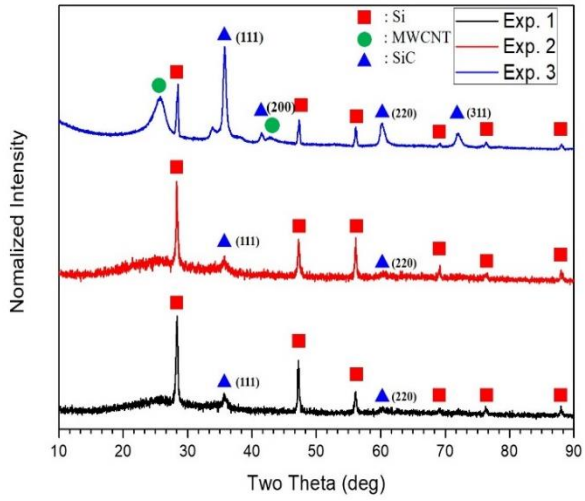

Fig. 2. XRD patterns of synthesized products in Exp. 1, 2, and 3.

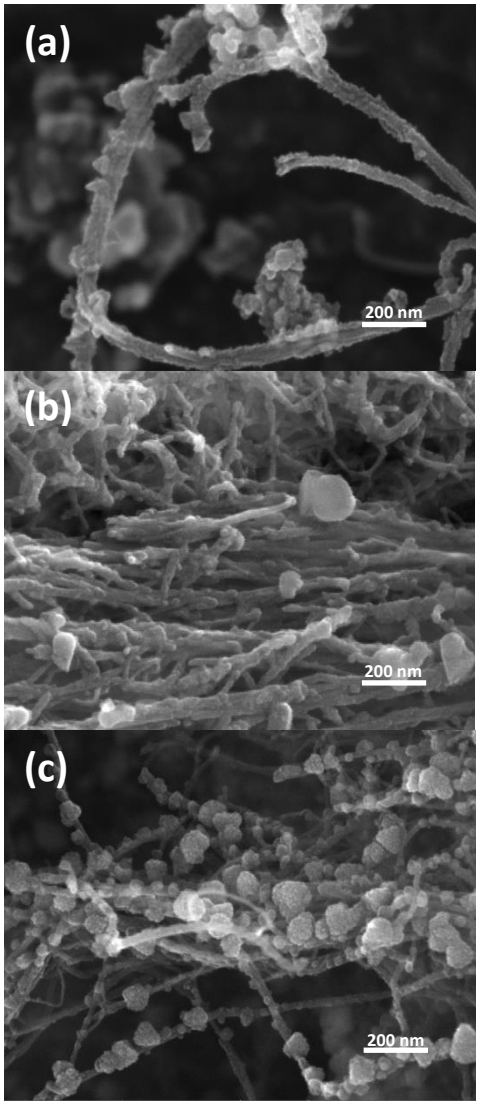

Fig. 3. FE-SEM images of synthesized Si-MWCNT nanocomposite in (a) Exp. 1, (b) Exp. 2, and (c) Exp. 3.

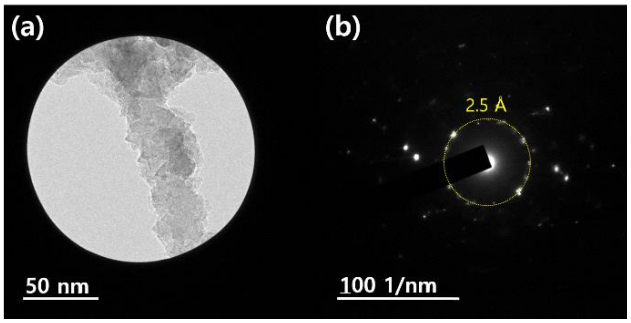

Fig. 4. (a) FE-TEM image and (b) SAED pattern of synthesized Si-MWCNT nanocomposite in Exp. 3.

The produced powder was collected at the inner wall of the reactor. It was analyzed by X-ray diffraction (XRD, Empyrean, PANaltocal), field emission scanning electron microscope (FE-SEM, MIRA3, TESCAN), and field emission transmission electron microscope (FE-TEM, JEM-2100F, JEOL).

3. Results and discussion

Fig. 1 shows the XRD patterns of the raw materials, Silicon and MWCNT. And the XRD patterns of synthesized product are presented in Fig. 2. In all products, Si and MWCNT peaks were observed, in addition, silicon carbide peaks were detected. Although the peak intensity of silicon carbide is weak in Exp. 1 and 2 (Si-rich condition), it is increased and become main crystalline peaks in Exp. 3 (MWCNT-rich condition). Fig. 3 shows the FE-SEM images of synthesized Si-MWCNT nanocomposite in all experimental conditions. It is observed that the amount of attached silicon nanoparticles is much higher in Fig. 3 (c). However, the size of the silicon nanoparticles is irregular, it is estimate that because of erratic vaporization and quenching process.

Fig. 4 shows the FE-TEM image and SAED pattern of the enlarged nanocomposite area. The d-spacing of spots in the diffraction pattern is 2.5 Å as SiC. It describes that the silicon nanoparticles were attached on the surface of MWCNT by SiC bonding. It means the silicon nanoparticles were not attached physically on the surface and it could be keep the structure without desorption during expansion by charging and discharging cycle.

Summary and future plan

Si-MWCNT was synthesized by triple DC thermal plasma jet system. The mass ratio of silicon and MWCNT was controlled. When the MWCNT ratio was high in Exp.3 condition, the peak intensity of silicon carbide was strongest in the XRD pattern and nanocomposite structure as silicon nanoparticles attached MWCNT was observed very well in SEM analysis. However, only nanoparticles and MWCNT beside Si-MWCNT were observed in all experiments. And some MWCNT was sublimated by injection into high temperature of the merged triple plasm jet. In order to improve the synthetic process, two powder feeding system will be applied in the future.

In the conference presentation, we will present for the optimal conditions and formation process for synthesis of Si-MWCNT nanocomposite.

References

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