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High-Rate Synthesis of Si/C Nanoparticles using Pulse-Modulated Induction Thermal Plasmas with Intermittent Feeding of Feedstock

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1. Introduction

Silicon (Si) nanoparticle (NP) is anticipated as a negative electrode material for the next generation lithium ion battery (LIB). Silicon has a 10-times higher theoretical discharge capacity of 4200 mAh/g than graphite used as the present negative electrode material. However, there are mainly two issues in use of Si as the negative electrode in LIB. One is capacity deterioration due to a drastic change in Si volume during repetitive charge-discharge in batteries. Another is relatively lower electrical conductivity of Si. To overcome the former issue, Si NPs can be adopted instead of Si bulk. To improve the latter of electrical conductivity, Si/C nanocomposites are being studied, which have carbon (C) on the surface of a Si NP. Concerning such Si/C NPs, it is desired to develop a high rate production method.

In this report, we tried high rate synthesis of Si/C NPs using the PMITP+TCFF method that we have developed [1,2]. The PMITP is the pulse-modulated induction thermal plasma, and TCFF means the time controlled feeding of feedstock. Microsized Si feedstock powder was supplied intermittently to the Ar-H_2 PMITP, while CH_4 gas was injected as a carbon source from the downstream port with Ar quenching gas (QG). Synthesized particles were analyzed by FE-SEM,TEM and Raman spectrometry.

2. Experimental setup

Fig. 1 shows the experimental apparatus for Si/C NP synthesis. It consists of an induction plasma torch, a vertical reaction chamber, a downstream horizontal chamber and a filter with a vacuum pump. The coil is connected with an rf inverter power supply. The experimental condition was as follows: The time average input power from the power supply to the PMITP was set to 25 kW. The rf coil current was amplitude-modulated into a rectangular waveform with 80%SCL-80%DF as depicted on the left in Fig.1, where SCL indicates the degree of modulation, and DF denotes the duty ratio of the modulated coil current [2]. The pressure in the chamber was kept constant at 300 torr. Argon gas was supplied as sheath gas at a flow rate 90 slpm and H_2 at 1 slpm from the top of the torch. A mixed gas of Ar and CH_4 was supplied as QG at 50 slpm. The flow composition was set to 98%Ar and 2%CH_4. This QG was introduced from Port: B shown in Fig. 1. The raw material Si powder was supplied at 3 g/min with Ar carrier gas of 4 slpm. The feedstock Si powder was intermittently supplied into the torch with an electromagnetic valve. That opening/closing time were set to 8/7 ms, respectively. Synthesized NPs were collected in the filter.

3. Experimental result

Most of synthesized particles collected in the filter were found as NPs. Their mean diameter was evaluated as 86 nm from FE-SEM images. Fig. 2 shows a TEM image and the Raman spectra of the synthesized NPs. From the TEM image, Si NPs were found to be encapsulated by some layers. On the other hand, the Raman spectra for the NPs shows G-band and D-band of carbon. From the above results, the layers on the Si NPs are inferred as graphene. As a consequence, we could obtain Si/C NPs using the PMITP+TCFF method with a production rate of 111 g/h. Introduction of CH_4 from the reaction chamber downstream of the torch may involve decomposition of CH_4 to generate C atomic species in plasma, which may cover Si cluster surface after nucleation.

4. References

[1] Y.Tanaka, et al., *J.Phys.Conf.Ser.* **406**, 012001 (2012).
[2] Y.Tanaka, et al., *Thin Solid Films* **519**, 7100-7105(2011).

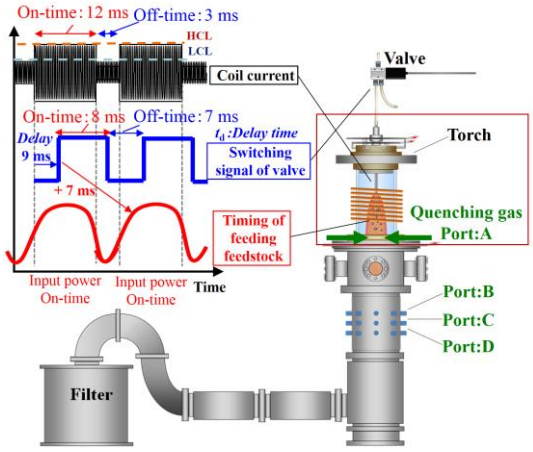

Fig. 1: Schematic of a nanoparticle synthesis system with PMITP+TCFF method.

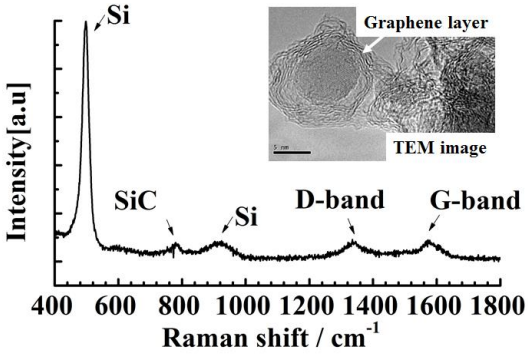

Fig. 2 : Raman spectrum and TEM image of synthesized nanoparticles. QG was injected from Port:B.