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Effect of carbon sources on the formation of carbon-coated silicon nanoparticles by induction thermal plasma

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The development of electrical vehicles and portable electronics require higher energy density and better performance of Li-ion battery. For the anode material, silicon is promising because of its high charge storage capacity of 4200 mAh/g that is approximately 10 times higher than graphite which is used commercially. However, silicon undergoes a 400% volume expansion during de-/lithiation process and leads to serious electrode failure mechanisms, which has severely reduced its promise for practical applications.

Applying carbon coatings is believed a useful way to prevent electrode pulverization, and provides continuous electronic and ionic conduction pathways during cycling. Purpose of this study is to synthesize silicon nanoparticles with carbon coating by induction thermal plasmas, and investigate the formation mechanism.

The experimental setup mainly consists of three parts, RF torch, chamber and filter. The plasma was generated by the RF power supply of 4MHz at 10 kW. Silicon powders with a diameter around 5 µm were injected into torch at 400 g/min. Methane and ethylene were chosen as carbon source to investigate their effect on carbon coating. Particles were characterized by XRD, Raman and EDS.

Figure 1 shows the components with different carbon sources injection. SiC can be observed in both cases seems that some hydrocarbon gases were injected into zones where temperature is still higher than the melting point of silicon. Then carbon atoms reacted with melted silicon and unfavorable SiC was generated. On the other, there is no crystalline carbon can be observed from Fig. 1 suggests that carbon mainly exist as amorphous phase.

Electron diffraction analysis as shown in Figure 2 indicates the structure of synthesized nanoparticles. Here diffraction patterns of nanoparticles synthesized with CH_4 was used as an example, and a clear core-shell structure can be observed in both cases. According to the EDS results which are not shown in here, the core is crystalline silicon, while shell keeps amorphous phase and mainly consists of carbon and/or organic compound.

The Raman spectra from 1100 to 1700 cm^{-1} for particles are shown in Figure 3. Both sp^2 and sp^3 sites were confirmed, and hydrocarbon also exists. Therefore, the silicon core is coated with amorphous hydrogenated carbon by thermal plasma. Carbon coating is very useful to protect particles from oxidation, and C_2H_4 has more effective protection as the ratio of Si-O band is lower than the value in CH_4 case.

The formation mechanism of coating is explained on the basis of Raman spectra. Firstly, CH_4 and C_2H_4 molecules were injected into tail of plasma and decomposed. CH_4 decomposes into CH_3 + H or CH_2 + H_2 at about 880 K [1], while two C_2H_4 molecules break down into C_2H_3 + C_2H_5 at about 780 K [2]. Secondly, these radicals bond to the surface of the silicon particles. Finally, more and more radicals were bonded to the surface, and lead coating forms and thickness increases.

Silicon nanoparticles coated with amorphous hydrogenated carbon were successfully synthesized in induction thermal plasma. The performance of coating can be controlled with different carbon sources.

References

[1] C.J. Chen, et al., Canadian Journal of Chemistry, 54, 3175 (1976).
[2] M.L. Boyd, et al., Canadian Journal of Chemistry, 46, 2427 (1968)

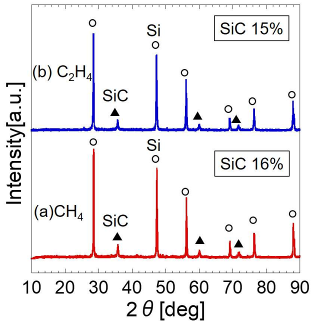

Fig. 1 Components and morphology of synthesized nanoparticles with (a) CH_4 and (b)C_2H_4. Inserts are the mole fractions of SiC in two cases.

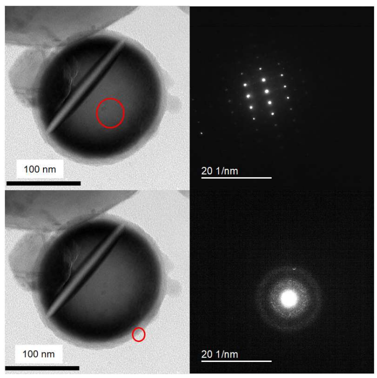

Fig. 2 Diffraction patterns of nanoparticles synthesized with CH_4. (red cycle is the focus of the electron beam)

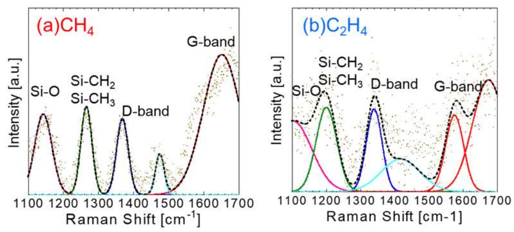

Fig. 3 Raman spectra of prepared Si nanoparticles with carbon source of (a) CH_4 and (b) C_2H_4