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Carbon Nanomaterial from Tea leaves as an Anode in Lithium Secondary Batteries

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Abstract : Carbon nanomaterials have been synthesized by pyrolysis of plant based precursors (i.e. Tea leaves), without using any catalyst. These materials are characterized by SEM, Raman and XRD. Carbon nanomaterials are utilized as the anode in Li secondary batteries. The charging/discharging of the lithium batteries are studied. The carbon obtained shows the highest capacity of 63.31mAh/gm.

Keywords: Carbon nanotubes, Lithium battery carbon, Lithium intercalation.

Introduction

Graphite is the most commonly used anode for Li-ion secondary battery. Lithium secondary batteries are the only commercially available batteries which can provide voltage greater than 3 V per cell. Lithium metal anodes, in conjunction with an organic electrolyte, result in non-uniform formation of a passive film on the anode surface (Koch *et al.*, 1982; Hirai *et al.*, 1994), which causes dendrite growth of lithium metal. This problem is tackled by utilizing the electrochemical intercalation of lithium in carbon, which generates a considerable negative potential close to that of lithium, but this is less reactive and easily reversible. Longer charging-discharging cycles are one of the important requirements for making a commercially viable lithium ion battery. The structural characteristics of carbon are believed to be the major element that controls the performance of Li-ion batteries. The intercalation of Li-ion with carbon depends upon factors such as the preparative history, carbon precursor, etc. Recently, attention has been drawn to use disordered carbon materials (Herold, 1955), which may store Li via a mechanism which is completely different from that associated with graphite. Sharon *et al.,* 2002 and Kumar *et al*., 2000, studied the intercalation of lithium with camphor-based carbon Nano-beads and found the cell to be stable for 10-20 days. The reversible Li-ion intercalation capacity was found to be 45%-61% of that obtained with graphite (Sharon *et al.*, 2002). In this paper, we report the result of lithium intercalation by carbon nanomaterial obtained from the pyrolysis of tea leaves (*Ehretia microphylla*).

Experimental

Tea leaves (*Ehretia microphylla*) obtained from a local vendor is boiled in water for 1 hr to remove the artificial color and flavor added by manufacturing company. Solution is filtered and leaves are washed several times with water and finally with acetone and dried in oven for three hours at 100ºC. The dried precursor is placed in a quartz boat and inserted in a quartz tube which is kept in a furnace as shown in Fig.1, such that the precursor is placed in the high temp zone of the furnace. Nitrogen gas was flushed for 15 min to remove air from the tube. The furnace was heated at 850º C with a flow of nitrogen gas at a flow rate of 50 cc per minute and maintained for 3 hrs to complete the process of pyrolysis. Later the furnace is cooled to room temperature and carbon materials are taken out. The synthesized carbon material is powdered using a motor pestle. These carbon materials were weighed to get the % yield. These carbon materials are purified by soaking in 3M Nitric Acid for 3 hrs to remove any metal impurity present. Then these were washed several times with distilled water and finally with acetone and dried at 100ºC. These materials were then characterized with Scanning electron microscopy (FEI quanta 200 electron microscope), X-Ray diffraction (PHILIPS PW 1710 Differactometer) and Raman Spectra (JASCO, NRS-2100 Raman spectrometer).

Purified carbon sample are mixed with ethylene-propylene dimethyl-monomer (EPDM) in cyclohexane and then pressed into a stainless-steel grid on the stainless-steel plate as an anode. A constant dc current was applied across the anode and a Li-metal counter electrode to measure the extent of Li-intercalation capacity of the anode. Ethylene carbonate (EC): dimethyl carbonate DMQ (1:20 solution), dissolved in $LiPF_6$ (1 wt. %), was used as an electrolyte. The charge-discharge characteristics were measured between 0 and 1.5 V. The charging of battery is shown by curve "A" and that of discharging behavior is shown by curve "B" in Fig. 2A. The reversibility of the battery was studied by measuring the change in capacity versus the number of cycle under the constant current of 1mA (Fig. 2B)

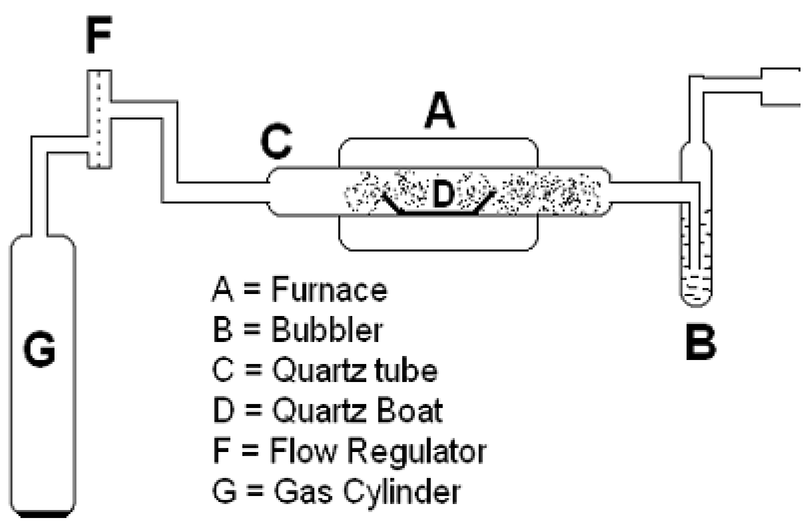
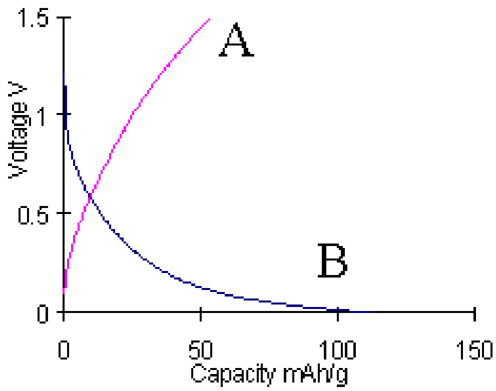


Fig.1 : Schematic diagram showing the apparatus for the preparation of carbon nanomaterials by pyrolysis of natural precursors



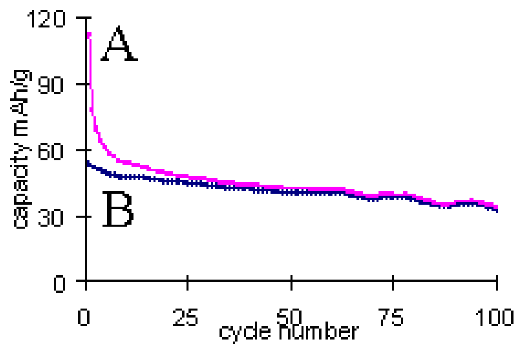


Fig.2 : Charge-Discharge Curve and Capacity of Li ion cell: A represents charging curve and B represents discharging curve

Results and Discussion

Scanning Electron Micrograph of the carbon materials obtained by the pyrolysis is shown in Fig. 2A & 2B. The Fig 2A which is taken at low resolution shows the bid formation with porous structure of carbon. From the Fig it can be concluded that the beads are having same spherical structure which is attached to the pores of the porous structure of the carbon. To analyze the bead formation the SEM image 2b was taken which confirms the bead formation having different boundaries.

A Raman spectrum taken is shown in Fig. 3. The Raman spectra show two different intense peaks at 1340 cm^{-1} and 1580 cm^{-1} which confirms occurring of D-band and G-band, respectively. These

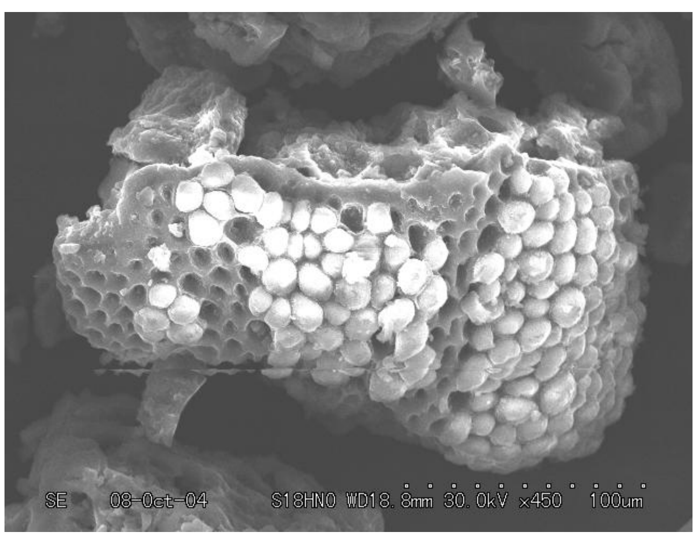


Fig.3 : SEM micrographs of carbon nanomaterials obtained from Tea leaves

Fig.3 : SEM micrographs of carbon nanomaterials obtained from Tea leaves

suggest the synthesized carbon material has both sp^2 and sp^3 hybridized carbon. From the Fig. 4 it has been noticed that the intensity of the both the D band and G band is around 0.775 (arb. units), which sugest the sp^2 and sp^3 ratio is of the same order.

X-Ray Diffraction studies were done by PHILIPS PW 1710 Differactometer with Cu Ka radiation. This is shown in Fig. 5. The XRD of carbon nanomaterials shows broad peak below 26 two theta which suggests the presence of amorphous carbon, also there is

Fig.4 : Raman spectra of carbon nanomaterials obtained from Tea leaves

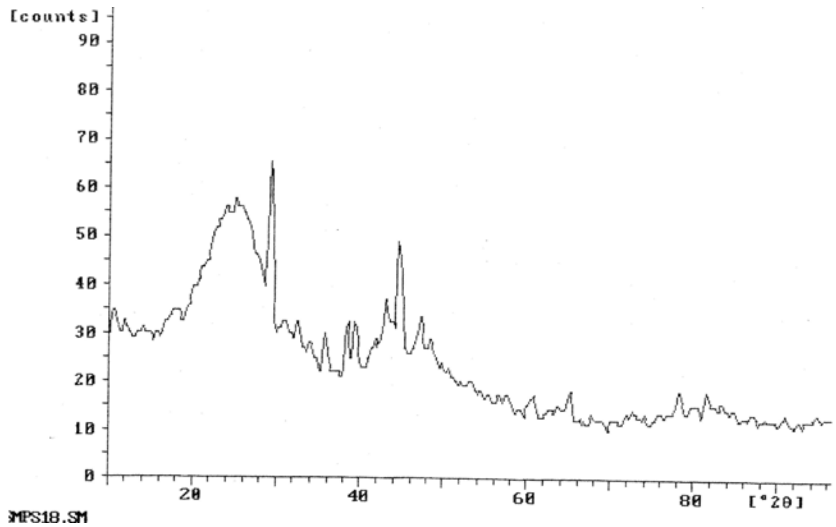


Fig.5 : XRD of carbon nanomaterials obtained from Tea leaves

one sharpe peak at two theta 42.9 which gives the value of interlayer spacing of 2.10 A° for $\alpha = 1$.

Conclusions

Carbon nanomaterials obtained from tea leaves (*Ehretia microphylla*) are examined for their application in lithium battery. Results of charging/discharging up to 1.5 V and number of reversible cycle are shown in Fig. 3. In the first cycle of discharging trend of carbon seems to show lower value of capacity (~ 100mAh/g) Carbon showed slow decrease in capacitance with the number of cycles, though its capacitance at 100th cycles was 63.31 mAh/gm. Efficiency of charging and discharging was found to be more than 96%.

Thus, Carbon nano material obtained from Tea leaves exhibit relatively high reversibility in charging-discharging profiles and good cycle stability.

References

Hirai T., Yoshimatsu I. and Yamaki J. (1994) : *J. Electrochem. Soc.*, **141**, 2300.

Herold A. (1955) : *Bull. Soc. Chim. France*, **197**, 999

Koch V.R., Goldman J.L., Mattos C.J. and Mulvaney M. (1982) : *J. Electrochem. Soc*. 129.

Kumar M., Kichambare P.D., Sharon M. and Avery N.R. (2000) : Krista J. Black. *Materials Chemistry and Physics* **66**, 83-89.

Sharon M., Hsu W.K., Kroto H.W., Walton D.R.M., Kawahara A., Ishihara T. and Takita Y. (2002) : *J. of Power Sources*, **104**, 148-153.