

# CHARACTERISTICS OF DISORDERED CARBON MATERIALS AS NEGATIVE ELECTRODES FOR LITHIUM SECONDARY BATTERIES

Y. Takahashi, H. Sakamoto, J. Ohishi, K. Kanno Y. Miki,  
T. Higashiizumi, M. Yoshimura and K. Shibahara

Mitsubishi Gas Chemical Company, Inc.  
Corporate Research Laboratory  
22 Wadai, Tsukuba-shi, Ibaraki, 300-42 Japan

## INTRODUCTION

Lithium rechargeable batteries have attracted much attention because their high energy density enables the use of smaller and lighter batteries in consumer electronics. These batteries have recently been commercialized and their range of application is rapidly expanding. To insure that the use of these rechargeable batteries is safe, carbonaceous materials which can store and release lithium ions are used in the negative electrodes. For these batteries to have energy density sufficient to meet the needs of today's applications, an improvement in capacity over present commercially available levels is essential. In this study, we present novel disordered carbon<sup>1,2)</sup> which has rechargeable capacity 30% larger than the theoretical capacity of  $C_6Li$  and explain this material's characteristic.

## EXPERIMENTAL

The disordered carbons was obtained by heat treatment of precursor synthesized from organic compounds at around 1000 °C. Working electrodes were prepared by the following procedure: Powdered carbonaceous material of mean particle size 12~15  $\mu m$  was mixed with 4 wt% of PTFE as a binder and made into 60 ~ 70  $\mu m$  thick sheets, which were then pressed onto a nickel mesh. The electrochemical properties of these electrodes were measured in a three-electrode cell containing a counter electrode and a reference electrode of lithium metal. The electrolyte used was 1M  $LiClO_4$  dissolved in a 50/50 mixture of PC and DME.

The cells were charged at a constant current of 2 mA/cm<sup>2</sup> against a fixed potential of 1mV. Discharge was conducted at 1 mA/cm<sup>2</sup> to a cutoff at 1.2 V.

## RESULTS AND DISCUSSION

The data listed in Table 1 are the physical properties of the carbons obtained by the synthetic method described above. The broad X-ray diffraction peak and the relatively low density of the material indicate that this is typical disordered carbon.

Figure 1 and 2 show the discharge curves and charge/discharge capacities by different charge times ranging from 0.4 to 40 hours. When charged up to 1 hour, it shows the discharge curve similar to those of coke materials. But when charge time was extended to over 1 hour, the increase in the charge capacity took the form of an extension of the voltage plateau appearing between 0.05-1 V. The full charge capacity (~ 1.2V) observed was 485 mAh/g and the discharge capacity accounted for by the region up to 0.2 V was 322 mAh/g accounting for 66.4% of the full discharge capacity.

These results show that this material has a discharge capacity 30% larger than the theoretical capacity of  $C_6Li$ . Consequently, we examined the charged material using <sup>7</sup>Li-NMR (Figure 3). When the material was charged up to 650 mAh/g, the NMR spectrum of the material showed a single peak at -90 ppm (vs LiCl). When it was charged up to 700 mAh/g, a different and very small peak appeared at -270 ppm. After charging up to 800 mAh/g, the spectrum clearly showed the two different peaks at -270 and -95 ppm. From these results, we concluded that charging the material to less than 650 mAh/g results in no deposition of lithium metal, and that the larger shift than is observed for graphite, though the reason is not yet clear, it must have some relevance to the larger than a theoretical capacity and long plateau region observed for this material.

## REFERENCES

1. Y.Takahashi et al., U.S.Pat. 5,326,658.
2. Y.Takahashi et al., Abstracts of the 35th Battery Symposium in Japan, 39(1994).

Table 1 - Physical properties of the disordered carbon material

d <sub>002</sub> (nm)	0.379
L <sub>c</sub> (nm)	1.15
Density (g/cm <sup>3</sup> )	1.54
Surface area (m <sup>2</sup> /g)	0.78

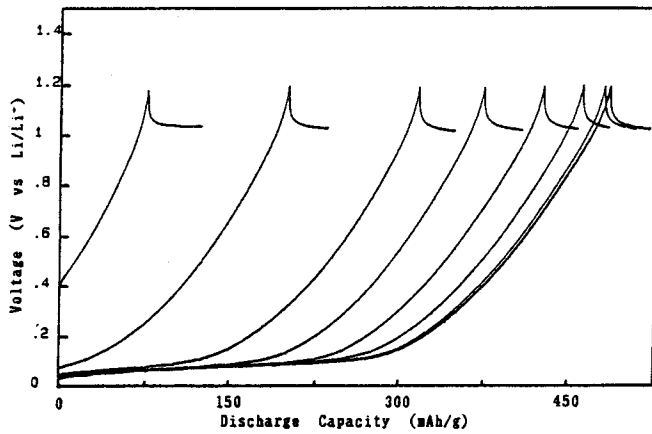


Figure 1 - Discharge curves for different charge times. Discharge current density; 150mAh/g (1mA/cm<sup>2</sup>).

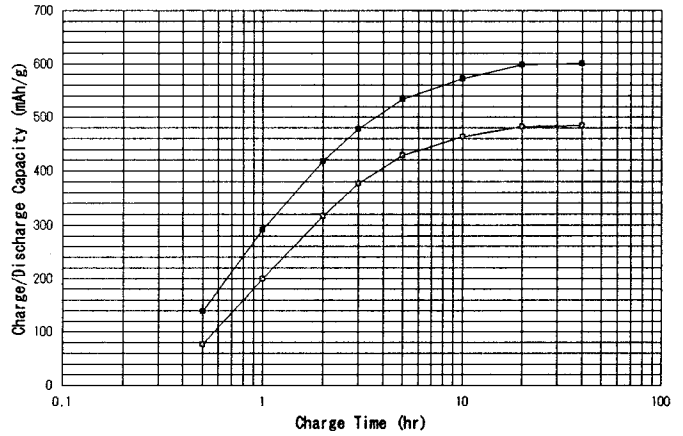


Figure 2 - Initial charge (■) and discharge (□) capacity.

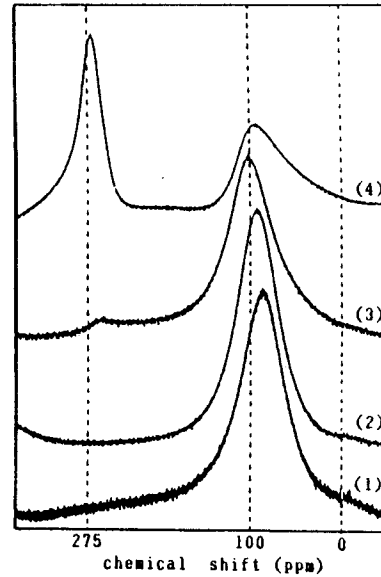


Figure 3 - <sup>7</sup>Li-NMR spectra of disordered carbon after charging  
Charged capacity; (1)600mAh/g (2)650mAh/g  
(3)700mAh/g (4)800mAh/g