Conference Paper

Meso Carbon Micro Bead (MCMB) Based Graphitized Carbon for Negative Electrode in Lithium Ion Battery

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Abstract Lithium ion battery performance of graphitized Meso Carbon Micro Beads (MCMB) as an anode material was investigated in full cell battery system containing LiCoO₂ cathode, PE separator and LiPF₆ electrolyte. The commercial MCMB, which was fabricated by Linyi™, was sintered at 500°C for five hour to make graphitized MCMB. The microstructure of graphitized MCMB was characterized using XRD and SEM to show the crystalinity, crystal phase and morphology of the MCMB particle. The result indicated that the crystal phase of the sample was changed into graphitized carbon. The electrode was made using coating method. We used copper foil as the substrate for anode. The anode materials consist of graphitized MCMB (active material), Polyvinylidene fluoride/PVDF (binder) and acetylene black (additive material). Full cell battery was tested using charge-discharge and cyclic voltammetry (CV) methods. From the CV characterization, cyclic voltammograms of the cell show characteristic lithium intercalation through reduction-oxidation peak. Charge-discharge test showed the discharge and charge capacity of the cells. According charge discharge test, commercial MCMB was better that graphitized MCMB.

Keywords: Graphitized carbon, MCMB, anode material, lithium ion battery

1. Introduction

Lithium ion battery has been widely used as energy storage system for applications such as in the field of portable electronic, e.g. in mobile phone, notebook computer, gadget, camera [1] and also for electric vehicles and hybrid electric vehicles [2]. There are many parameters that should be optimized, including the energy density, safety, rate capability, cost, sustainability [2] and cycle life [3] to get better performance of the Li-ion batteries.

Carbon-based anode has been widely used as anode for commercial batteries such as mesocarbon microbead, artificial and natural graphite, carbon fiber or C-C composites material [4], carbon nanotube, and also graphene [5]. Natural graphite was the most promising material for lithium-ion battery because of its low cost and flat potential profile, high cumbic efficiency in proper electrolytes and relatively high reversible capacity. Synthetic graphite has many properties that are the same as those of natural graphite. Besides that, it has many unique merits such as high purity, variety of
structures suitable for smooth Li intercalation and diffusion. Graphitized MCMB is one of the benchmark synthetic graphite materials for lithium-ion battery [6].

Graphitized MCMB was better than MCMB before graphitization. Graphitized MCMB has many advantages e.g., high packing density that guarantees high-energy density, small surface area that decreases the irreversible capacity corresponding to electrolyte decomposition. Most of the surfaces of MCMB spheres are composed of edge-plane surfaces, thus Li\(^+\) intercalation becomes easier and the rate capacity increases and MCMB can be easily spread onto copper foil [6]. So, it was very interesting to analyze the synthetic graphite like graphitized MCMB to improve the performance of anode in the lithium ion battery.

2. Experimental

2.1. Preparation and characterizations of MCMB

The commercial MCMB was fabricated by LinyiGelon\(^{\text{TM}}\) with specific surface area \(\approx 1.5 \text{ m}^2/\text{g}\). This material was then sintered at 500\(^\circ\)C for five hours to make graphitized carbon. Commercial and graphitized MCMB were characterized by X-Ray Diffraction (XRD, Rigaku) using Cu \(\kappa\alpha\) (\(\lambda = 1.541862 \text{ Å}\)) to identify crystal phase of these material in the range 5\(^\circ\) - 90\(^\circ\) of two theta. The XRD technique could identify the graphitization of the material with crystal phase identification of these materials (commercial and graphitized MCMB). Scanning Electron Microscopy (SEM, SU3500, Hitachi) with tungsten as electron source was used to observe the morphology and to determine the particle size of these particles.

2.2. Preparation of the electrode and full cell battery

The full cell battery of commercial and graphitized MCMB were investigated in pouch cells. The positive electrode was prepared by commercial electrode LiCoO\(_2\) (MTI Corp., USA) with the thickness of active material was 100 \(\mu\text{m}\). The negative electrode were prepared using commercial and graphitized MCMB as the material active with the composition of slurry i.e. 85 wt% active material, 5 wt% acetylene black, and 10 wt% PVdF dispersed in DMAC as a solvent. Full cell battery was prepared by assembling positive electrode, negative electrode, and Celgard microporous polyethylene (PA grade, 25\(\mu\text{m}\) thickness) as separator with LiPF\(_6\) 1 M as electrolyte.

2.3. Battery testing

Cyclic voltammetry (CV, WonaTech WBCS3000) experiment was performed on the batteries with scanning rate 0.1 \(\mu\text{A/s}\) and potential range 2.5 – 4.5 volt. Charge discharge (CD, WonaTech WBCS3000) characteristic was recorded to determine the charge-discharge capacity at work voltage.
3. Result and Discussion

3.1. MCMB characterizations

XRD pattern (Fig. 1) showed the shifting of diffraction peak of commercial MCMB and graphitized MCMB. It indicated that there was a phase change in the crystal structure. By Rietveld method of interpretation, there are three phases in the commercial MCMB i.e. graphite-2H (48.8%), graphite-3R (39%), and carbon (12.6%), while for the graphitized MCMB has single phase, graphite-2H (100%). SEM image (in Fig. 2) show the morphology and size of the MCMB particles. Both of samples had a spherical structure with diameter range 5 – 20 μm.

3.2. Electrochemical and battery performance analysis

According to the cyclic voltammogram of the samples, it was typically the same. But, it was little bit different in the reduction peak (discharge voltage). The oxidation peak was 4.2 volt, while the reduction peak was 3.6 volt and 4 volt (for graphitized MCMB), while 3.5 volt and 4 volt for commercial MCMB. There are two reduction peaks in the cyclic voltammogram of the samples. It indicated that other material was performed as anode material in the cell, such as acetylene black.

Charge-discharge was performed at rate 0.1 C to measure the charge and discharge capacity of the samples. From the charge-discharge measurement, it shows that the commercial was better than the sample. In the first cycle, charging capacity of the
Figure 2: SEM images of (a) commercial MCMB and (b) graphitized MCMB sintered at 500 °C for five hours.

Figure 3: Cyclic voltammogram with scan rate 0.1mV/s of commercial MCMB and graphitized MCMB sintered at 500 °C for five hours.

commercial MCMB was 3.4 mAh, while the graphitized MCMB was 3.0 mAh. For discharge capacity, the commercial MCMB has 1.5 mAh, while the graphitized MCMB has 1.3 mAh. This effect was probably happened because of stability of MCMB structure, lithium intercalation process, and coating process.

4. Summary

Graphitized MCMB were prepared by sintering process at 500°C for five hours. Structure and morphology were evaluated by XRD and SEM, respectively. According to the XRD diffractogram, the diffraction peak showed the graphitized MCMB was produced. In the SEM image, the morphology of the particles was typically the same, spherical structure with diameter 5 – 20 μm. Reduction and oxidation peak in cyclic voltammogram
Figure 4: Comparison charge-discharge profile at C-rate 0.1C of (A) commercial MCMB and (B) graphitized MCMB sintered at 500 °C for five hours.

indicated the electrochemical reaction of the sample. The oxidation peak was 4.2 volt, while the reduction peak was 3.6 volt and 4 volt (for graphitized MCMB), while 3.5 volt and 4 volt for commercial MCMB. Charge-discharge profiles showed charge-discharge capacity of the commercial MCMB was 3.4 mAh and 1.5 mAh, while the graphitized MCMB was 3.0 mAh and 1.3 mAh, respectively. It indicated that the commercial MCMB was better than graphitized MCMB.

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References