Journal of Power Sources 287 (2015) 276–282

Contents lists available at ScienceDirect

Journal of Power Sources

journal homepage: www.elsevier.com/locate/jpowsour

Surface natures of conductive carbon materials and their contributions to charge/discharge performance of cathodes for lithium ion batteries

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HIGHLIGHTS

- Surface natures of five commercial carbon materials were understood.
- Carbon with large specific surface area leads to low coulombic efficiency.
- Carbon with layered structure leads to low charge/discharge capacity of cathode.
- Negative effects become more significant as high voltage cathode material is used.

ARTICLE INFO

Article history:
Received 29 November 2014
Received in revised form
17 March 2015
Accepted 14 April 2015
Available online

Keywords:
Carbon materials
Electronic conductor
Surface nature
Charge/discharge performance
Cathode
Lithium ion battery

ABSTRACT

The surface natures of five carbon materials, acetylene black, Super P, ECP600JD, KS-6 and CNTs, are compared in terms of morphology, specific surface area and activity towards electrolyte decomposition and anion insertion, and their contributions as conductive additives to cathode performance of lithium ion batteries are understood. With the characterizations from scanning electron microscopy, Brunauer–Emmett–Teller analysis and cyclic voltammetry, it's demonstrated that: (1) the morphology is granular for acetylene black, Super P and ECP600JD, flake-like for KS-6 and wire-like for CNTs; (2) ECP600JD exhibits the largest specific surface area but KS-6 has the smallest one; (3) the activity is the same for all the samples towards the electrolyte decomposition but different from each other towards anion insertion. Charge/discharge tests of $LiMn_2O_4$ and $LiNi_{0.5}Mn_{1.5}O_4$ cathodes indicate that the surface natures of carbon materials play an important role in charge/discharge performance of cathodes for lithium ion batteries. ECP600JD with smallest particle size provides the largest site for electrolyte decomposition leading to the lowest coulombic efficiency, while KS-6 with a layered structure exhibits the highest activity towards anion insertion leading to the lowest charge and discharge capacity of cathode materials. These negative effects become more significant when high voltage cathode materials are used.

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1. Introduction

Lithium ion batteries are believed to be promising power sources for hybrid electric vehicles (HEVs) and electric vehicles (EVs) [1–4]. Currently commercial cathode materials, including $LiFePO_4$, $LiCoO_2$, $LiMn_2O_4$, provide lithium ion batteries with the working voltage of less than 4.2 V [5,6]. For the large-scale applications in HEVs and EVs, high energy density lithium ion batteries are required. Several high voltage cathode materials have been developed, including $LiNi_{0.5}Mn_{1.5}O_4$ [7,8], $LiM_{2x}Ni_{0.5-x}Mn_{1.5-x}O_4$ (M = Cr, Co, Al) [9–11] and $LiCoPO_4$ [12]. However, the voltage limitation of the state-of-art electrolytes hampers the practical application of the high voltage cathode materials. Many efforts have been made to understand the interaction between cathode materials and electrolyte in high voltage [13–17]. However, seldom attention has been paid to the contribution of conductive carbon materials, which are necessary as cathode additives because the cathode materials are poorly electronic conductors.

Based on the recorded oxidation current in linear sweep voltammetry, Zheng et al. concluded that electrolyte decomposed more easily on the carbon with larger specific surface area [18]. Large specific surface area provides more reaction sites for electrolyte decomposition and thus causes larger oxidation current, but this does not mean that the carbon with larger specific surface has

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http://dx.doi.org/10.1016/j.jpowsour.2015.04.077
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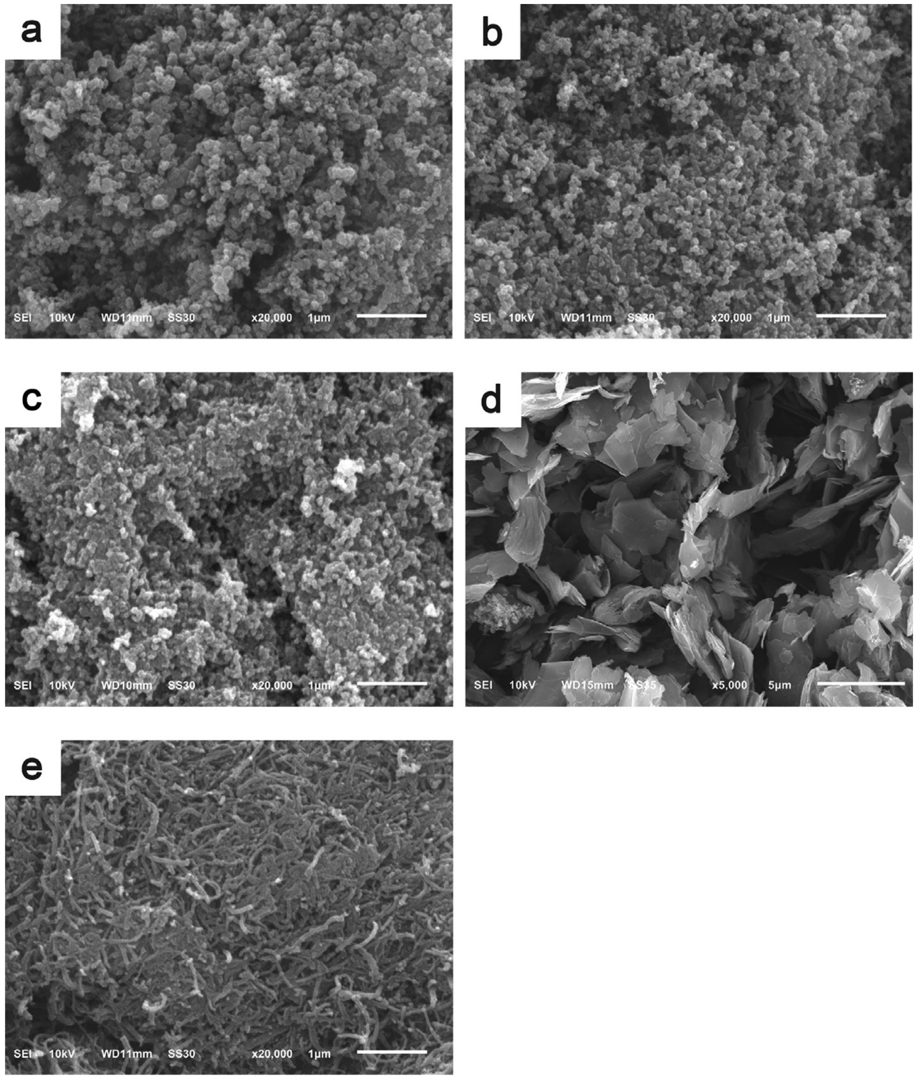

Fig. 1. SEM images of different conductive carbon materials: (a) Acetylene Black, (b) Super P, (c) ECP600JD, (d) KS-6, and (e) CNTs.

higher activity towards electrolyte oxidation because the oxidation current might become smaller when normalized with specific surface area and additionally the recorded current might be attributed to charge current for double layer capacitance. Up to date, knowledge has not been achieved on the activity of various carbon materials towards electrolyte oxidation, which is important to battery performance. It has been reported that conductive carbon materials possess activity for anion (PF_6^-) insertion at high potentials [19–21], but effect of this activity on the battery performance remains unknown.

To apply conductive carbon materials better and to help develop better carbon materials for lithium ion batteries, the surface natures of five commercial conductive carbon materials, acetylene black [22], Super P [23], Ketjen black ECP600JD [24], conductive graphite KS-6 [25] and carbon nanotubes (CNTs) [26,27], were understood systematically in terms of morphology, specific surface area and activity towards electrolyte decomposition and anion insertion. The contributions of these natures to cathode performances of lithium ion batteries were evaluated with $LiMn_2O_4$ and $LiNi_{0.5}Mn_{1.5}O_4$ [28–31].

2. Experimental

2.1. Electrochemical measurements

Cyclic voltammetry was used to determine the activity of conductive carbon materials towards electrolyte decomposition and anion insertion. Five commercial conductive carbon materials,

Table 1
Surface natures of carbon materials and their effects on charge/discharge performance of cathodes.

Samples	Acetylene black	Super P	ECP600JD	KS-6	CNT
Morphology	Granular	Granular	Granular	Flake	Wire-like
Surface area (m^2 g^{-1})	54.7	56.9	522.4	22.3	86.7
Onset potential for electrolyte decomposition (V, vs. Li/Li^+)	3.75	3.75	3.75	3.75	3.75
Coulombic efficiency of $LiMn_2O_4$ at the first cycle (%)	93	94	80	90	91
Coulombic efficiency of $LiNi_{0.5}Mn_{1.5}O_4$ at the first cycle (%)	81	81	49	50	74

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