



Spherical Carbon as a New High-Rate Anode for Sodium-ion Batteries



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ABSTRACT

Ambient temperature sodium-ion batteries are emerging as a new chemistry platform for energy storage technologies. Anodes that consist of carbon have been used for this application due to their highly stable and reversible Na cycling nature. In the search of new carbon materials with various microstructure morphologies, we have synthesized spherical carbon particles via an autogenic process. ²³Na MAS solid-state NMR characterization method as a function of the state of charge of carbon was used to determine that these spherical carbon anodes show pure, reversible Na intercalation into graphene regions: these materials are devoid of nanocavity or nanopore filling of Na. This feature and the spherical morphology create low reactivity behavior with the electrolyte which assists in the material's highly reversible (de)sodiation. Specific capacity is 40 mAhg⁻¹ at a high-rate of 10C (1.5 Ag⁻¹) thus demonstrating that the graphene element in hard carbons contributes largely to the rate capability.

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1. Introduction

The introduction of new high performance low cost batteries and their technological development is a key driver for the deployment of advanced large scale electrical energy storage systems. Na-ion batteries that operate at ambient temperature constitute a serious contender for this application if the energy density can be improved. The search for electrode materials that possess desirable properties such as low cost and low toxicity, long cycle life, high rate capability, high energy density that are easily synthesized will foster the rollout of an environmentally sustainable battery technology [1].

Most evident as anodes for Na-ion batteries are carbon materials. Carbonaceous materials such as petroleum cokes, [2–4] carbon black, [5] templated carbons, [6] pitch-based carbon-fibers, [7] and polymers (poly(para-phenylene)) [8,9] are known to insert Na. Mostly abundant and low cost, such anodes have been shown to possess a variety of electrochemical performances due to their

difference in morphologies, surface areas, pore size and percentage in the material. These anodes are much less reactive than dendrite forming sodium metal in non-aqueous systems as they operate at a slightly higher voltage than metallic reactive Na metal and do not plate Na.

Graphite, the most commonly used anode in lithium ion cells has a theoretical specific capacity of 372 mAh/g, but it does not intercalate sodium to any appreciable extent and is electrochemically irreversible [10]. In contrast, so-called hard-carbons which are materials that are synthesized at high-temperatures (~1000 °C, inert atmosphere) from carbon-based precursors such as sugars have been utilized largely in Na-ion systems with variable performance. Recently, carbon nanomaterials such as carbon nanofiber, [11] reduced graphene oxide, [12] and hollow carbon nanosphere [13] also have shown impressive performance. The great flexibility in carbon-source precursors leads to low cost, but other factors such as electrode packing density, rate capability and safety are very important for their use.

In this work, we have synthesized micron-sized carbon material that combines the structural similarities for Na-insertion in a conventionally synthesized hard carbon, but instead featuring a unique spherical shape [14] (hereafter called sphere carbon) possessing a characteristic smooth surface, with a solid and dense core. It is expected that a carbon sphere is preferred not only from electrode

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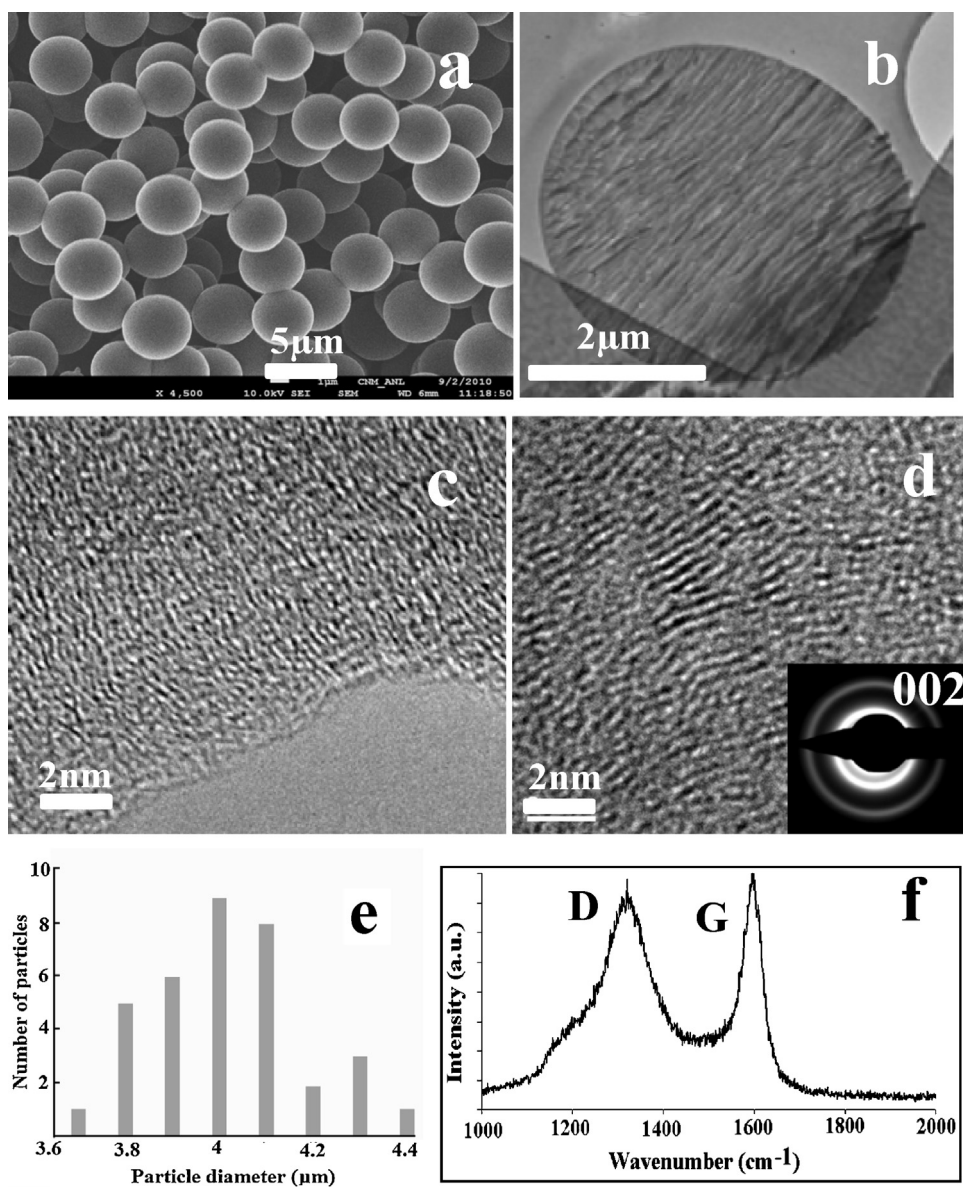


Fig. 1. Carbon spheres shown: a) by scanning electron micrograph, b) cross-section SEM, c) surface structure shown by TEM, d) inner structure by TEM, e) particle size distribution of spherical carbon determined from SEM Fig. 1a, and F) Raman spectrum results.

packing density (better than fluffy, typical hard carbon with random particles), but from the standpoint of homogeneous primary current distribution across the surface and the avoidance of SEI over-buildup and Na consumption at sharp edges and morphology irregularities.

2. Experimental

2.1. Synthesis

The synthesis of carbon sphere particles was carried out by the controlled thermal decomposition of mesitylene (C₉H₁₂) or used low density polyethylene bags in a specially designed, enclosed (but ventable), Haynes 230 alloy autoclave reactor under argon gas [14]. The reactor was heated to 700 °C with a heating rate of 20 °C/min; the reactor was held at 700 °C for 5 min, which generated an autogenic pressure of less than 3310 kPa. Thereafter, the reactor was cooled gradually to room temperature. The yield of dry, carbon spheres was approximately 45 wt. %. For the commercial hard

carbon, the Carbotron P material was obtained from Kureha Corp. (Japan)

2.2. Electrochemical test

The working electrode consisted of 85 wt% of sphere carbon, 8 wt% carbon black, and 7 wt% polyvinylidene difluoride binder laminated onto a copper foil current collector. Electrochemical evaluations of sphere carbon electrodes were undertaken using coin cells (size 2032) with a sodium metal foil counter electrode and an electrolyte consisting of 1 M NaClO₄ in propylene carbonate (PC) solvents. The cells were assembled in a helium-filled glove box. Cells were galvanostatically charged and discharged between 0.001 and 2.0 V vs. Na at room temperature.

2.3. Characterization

A Raman spectrum of sphere carbon was recorded at room temperature using a triple Jobin Yvon/Atago-Bussan 92 T-6400

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