Electrochimica Acta 154 (2015) 94–101

Contents lists available at ScienceDirect

Electrochimica Acta

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

Synthesis of Carbon-coated Nanoplate α-Na_2MoO_4 and its Electrochemical Lithiation Process as Anode Material for Lithium-ion Batteries

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ARTICLE INFO

Article history:
Received 9 October 2014
Received in revised form 12 December 2014
Accepted 13 December 2014
Available online 16 December 2014

Keywords:
Lithium-ion batteries
Anode
α-Na_2MoO_4
Carbon-coating
Sol–gel method
Nanoplates

ABSTRACT

The carbon-coated α-Na_2MoO_4 nanoplate sample was fabricated via a facile sol–gel method involving the subsequent annealing under a reducing atmosphere to decompose the organic carbon source. X-ray diffraction with Rietveld refinement, high-resolution transmission electron microscopy (HRTEM) and X-ray photoelectron spectroscopy (XPS) results show that single-phase α-Na_2MoO_4 can be obtained even under the presence of carbon and reducing atmosphere. When evaluated as an anode material for lithium-ion batteries, the carbon-coated α-Na_2MoO_4 nanoplate electrode displays a discharge and recharge capacity of 806 $mAh\,g^{-1}$ and 409 $mAh\,g^{-1}$ respectively in the first cycle, while a reversible discharge–charge capacity of 350 $mAh\,g^{-1}$ can be retained after 30 cycles at 30 $mAh\,g^{-1}$. A capacity of ~320 $mAh\,g^{-1}$ at 30 $mAh\,g^{-1}$ can still recover after 50 cycles even following the discharge/charge process with the high current density of 480 $mAh\,g^{-1}$. Meanwhile, carbon-free and carbon-coated α-Na_2MoO_4 powders fabricated via a solid state reaction were also prepared for comparison. Furthermore, the structure change of α-Na_2MoO_4 and its Li storage mechanism upon lithiation and delithiation process are studied by ex-situ XRD and TEM in below.

1. Introduction

Lithium-ion batteries (LIBs) have been intensively utilized in portable electronics and show significant promise for the applications in hybrid electric vehicles (HEVs), electric vehicles (EVs), and smart grids. Graphite is used as the dominant anode in commercial LIBs, but its specific capacity is limited to 1 mol Li per C_6 (372 $mAh\,g^{-1}$). Besides, the Li intercalation into graphite appears at a low potential (<0.1 V) which is close to that of the Li-plating, which easily gives rise to the short circuit of batteries due to the formation of Li dendrites [1]. Therefore, extensive efforts have been made to search for the graphite alternatives in the hope of finding novel materials with both high capacities and slightly more positive intercalation voltage (vs. Li^+/Li) compared to graphite anodes. Transition metal oxides have long been known as competitive anode materials for advanced lithium-ion batteries due to their high reversible capacities and higher potential relative to Li^+/Li than graphite [2–9]. Specially, molybdenum oxide MoO_3 [10–12] and a series of molybdate such as $MMoO_4$(M = Mn, Cu, Zn, Ni, Fe, Ca) [13–15] have been reported about their electrochemical intercalation of lithium as possible anode materials for Li-ion batteries. Since Mo can exist in various oxidation states ranging from +6 to 0 during the electrochemical process, they display large capacity at the beginning of the electrochemical process. However, the large capacity could not be sustained on long-term cycling for their conventional bulk electrode and poor electronic conductivity. It has been widely acknowledged that nanosized materials provide higher electrochemical performance both in cathode or anode by means of decreasing lithium-ion diffusion distances and increasing the contact area between the electrode and the electrolyte [2,7,10,16]. Therefore, nanosized the Mo-based compounds with special morphology is an effective way to fully release their higher capacities and keeps an excellent electrochemical performance. Meanwhile, coating on the material surface is also a way to improve the electrochemical performance [17]. Especially to improve the electronic conductivity, carbon coating on the surface of materials has been a general method, therefore,

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http://dx.doi.org/10.1016/j.electacta.2014.12.082
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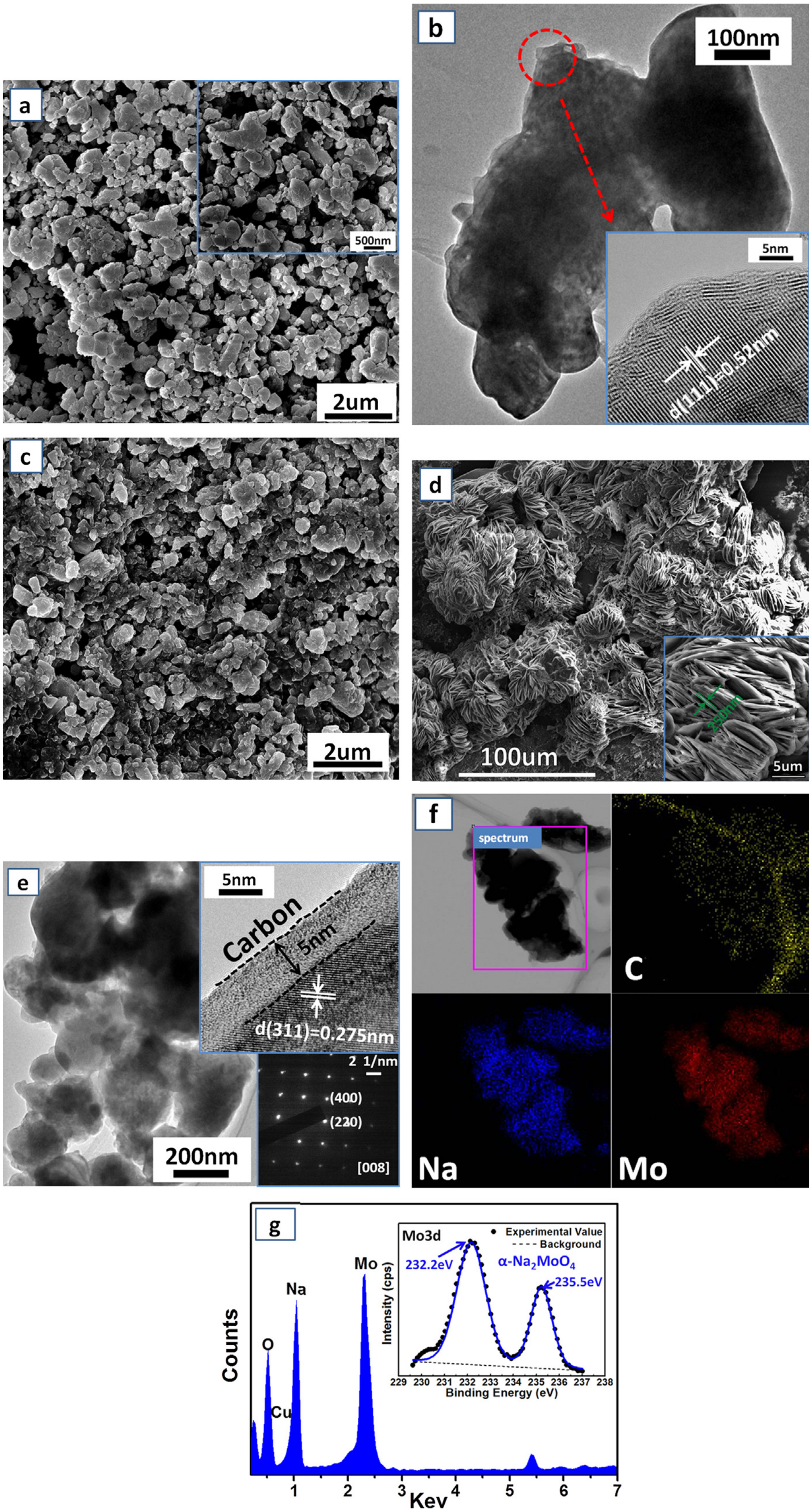

Fig. 1. SEM (a) and TEM (b) of the carbon-free powders; SEM (c) of the carbon-coated powders; SEM (d) and TEM (e) of the carbon-coated nanoplates; (f) and (g) EDS analysis of the fragments obtained from the nanoplates; The XPS of Mo3d in (g) as an inset.