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Simple solution-combustion synthesis of Ni-NiO@C nanocomposites with highly electrocatalytic activity for methanol oxidation

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ABSTRACT

Transition metal and its oxide composite nanomaterials are attracting increasing research interest due to their superior properties and extensive applications in many fields. In this paper, Ni-NiO@C nanocomposites were successfully synthesized in one step via a simple solution-combustion route, employing $NiCl_2$ as the Ni source, oxygen in the atmosphere as the oxygen source, and ethanol as the solvent. The final product was characterized by powder X-ray diffraction (XRD), X-ray photoelectron spectroscopy (XPS), scanning electron microscopy (SEM), energy dispersive X-ray spectrometry (EDS), (high resolution) transmission electron microscopy (TEM/HRTEM), and Raman spectra. N_2 gas sorption-desorption experiments uncovered that the BET surface area of Ni-NiO@C nanocomposites reached 161.9 m^2 g^{-1}, far higher than 34.2 m^2 g^{-1} of Ni-NiO. The electrochemical measurement showed that the as-produced Ni-NiO@C nanocomposites presented better catalytic activity for the electro-oxidation of methanol than Ni-NiO and NiO, which provides a new catalyst selection for the electro-oxidation of methanol.

1. Introduction

Nowadays, energy shortage and environmental pollution are two major challenges to the human society. Therefore, the exploration of new clean energy resources and the related devices has drawn worldwide interest [1,2]. As one of new clean energy resources, direct methanol fuel cells (DMFCs) are paid much attention due to the high energy conversion efficiency, low pollution emission and simple operation device [3,4]. To date, many studies have uncovered that the electrode materials with high electrocatalytic activity act a vital role for enhancing the performance of a DMFC [5]. At present, several noble metals, such as Pt [6], Pd [7] Ru [8] and their alloys [9–13], are often used as electrocatalysts owing to their high electrocatalytic activity. However, the high costs and low utilization of these noble metals obstruct the commercialization of DMFCs [14]. To promote the commercialization of DMFCs, it is a key to find new catalysts with low cost and high electroactivity to replace noble metal catalysts.

Generally, transition metal oxides, **such as NiO** [15], **$Ni(OH)_2$** [16], Co_3O_4 [17] and MnO_x [18], bear high catalytic activity and can be potential candidate for electrode materials. Among them, NiO is considered to be one of the most promising electrode materials for the methanol oxidation due to its low cost and high electrocatalytic activity [19]. However, the low conductivity of NiO is a fatal imperfection for its application. It is well known that metallic Ni possesses good conductivity and catalytic activity. It is possible that Ni-NiO composites show better performance for methanol electrooxidation than single NiO [20].

In order to prepare Ni-NiO composites, two approaches have been developed: One is the oxidative route and another is the reductive one. Through the former metallic Ni is firstly prepared, and then is partially oxidized to form Ni-NiO composites. For example, Zhang and coworkers [21] firstly electrodepositedly obtained porous Ni film on the surface of the stainless steel from an electrolyte solution containing nickel chloride and ammonium chloride. Then, porous Ni film was annealed at different temperatures in air to form Ni-NiO microstructures with controlled oil adhesions. By the latter the nickel compound containing oxygen is firstly obtained, and subsequently is partly reduced at high temperature to form Ni-NiO composites. For instance, Huang *et al* [22] firstly synthesized $Ni_2(OH)_2CO_3$ precursor via a precipitation route, and then $Ni_2(OH)_2CO_3$ precursor was reduced by ethanol at 700 °C to form Ni-NiO nanocomposites. It was found that the as-prepared Ni-NiO nanocomposites could be used as an anode material for Li-ion batteries. Obviously, the above two routes need two steps at least, and high temperature. Can Ni-NiO nanocomposites, then, be obtained by a simple one-step route?

In our previous reports, a mild solution-combustion method was

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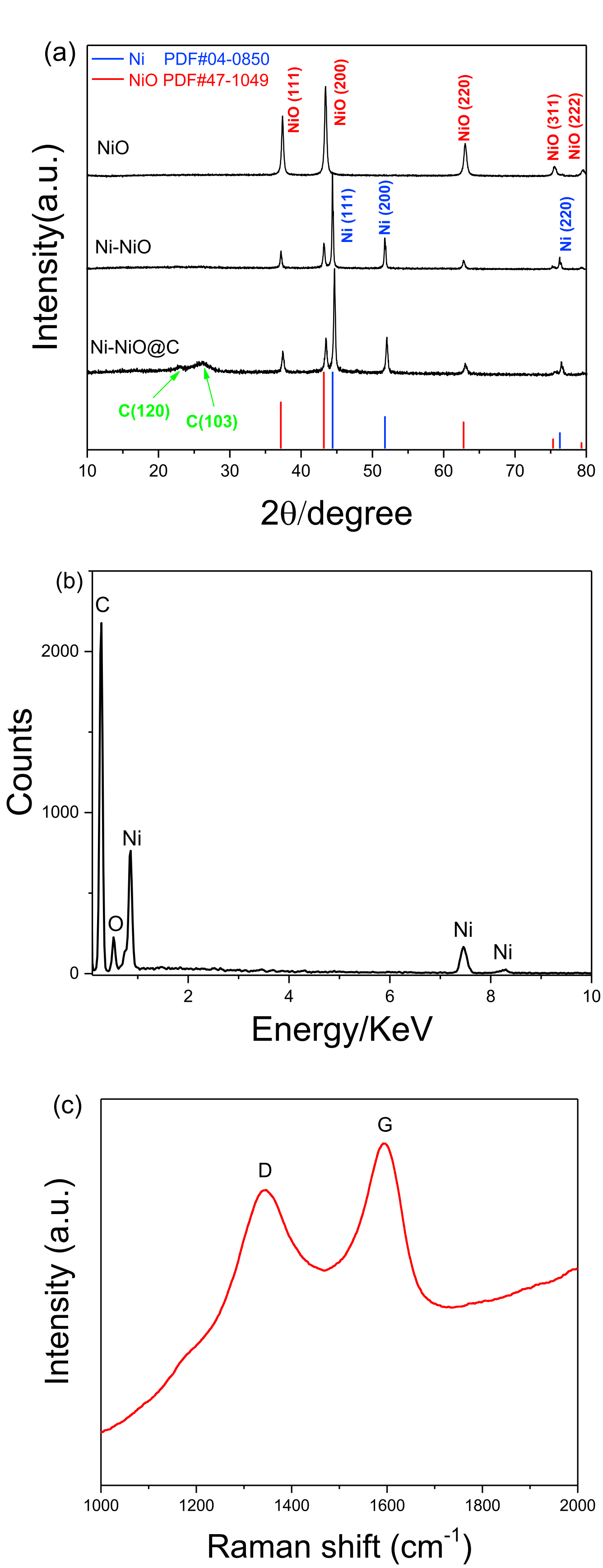

Fig. 1. (a) XRD patterns of the as-prepared Ni-NiO@C, Ni-NiO and NiO, (b) EDS analysis and (c) Raman spectrum of the as-prepared Ni-NiO@C.

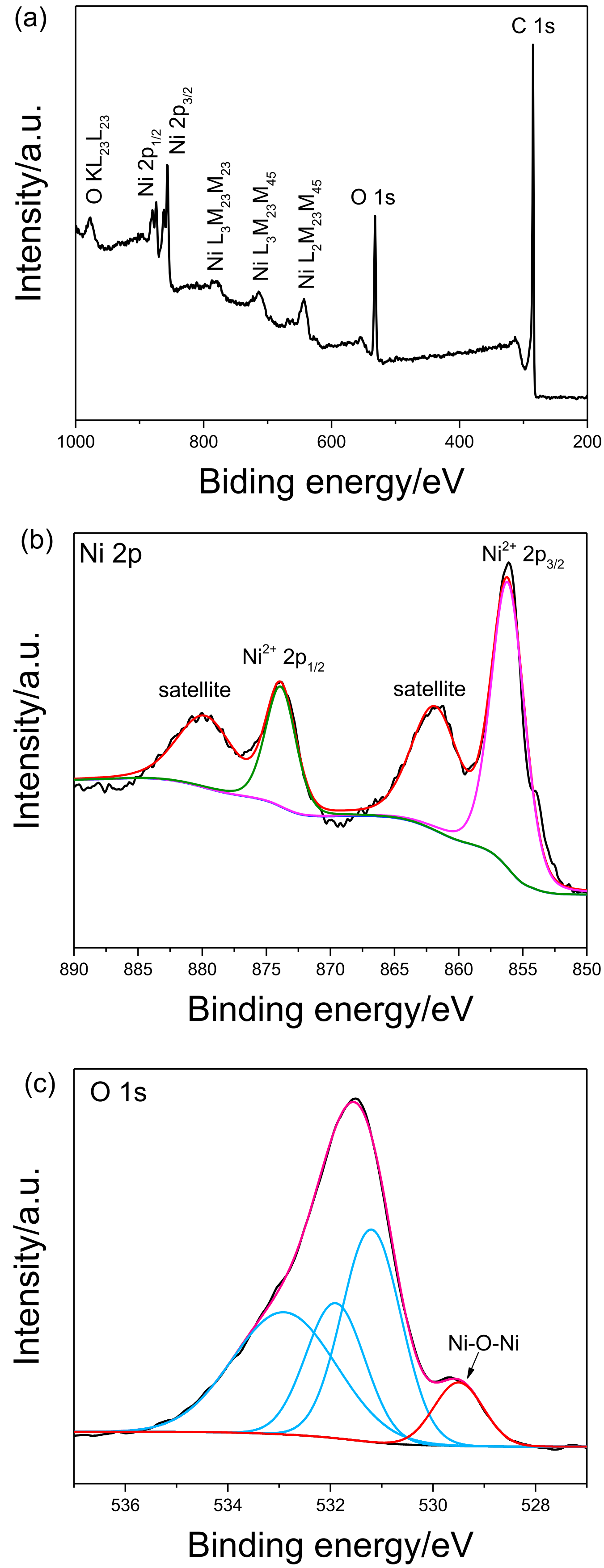

Fig. 2. XPS spectra of Ni-NiO@C nanocomposites: (a) Survey spectrum, (b) Ni 2p and (c) O 1s.