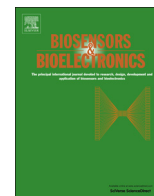




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Electrocatalysis and electroanalysis of nickel, its oxides, hydroxides and oxyhydroxides toward small molecules



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ABSTRACT

The electrocatalysis toward small molecules, especially small organic compounds, is of importance in a variety of areas. Nickel based materials such as nickel, its oxides, hydroxides as well as oxyhydroxides exhibit excellent electrocatalysis performances toward many small molecules, which are widely used for fuel cells, energy storage, organic synthesis, wastewater treatment, and electrochemical sensors for pharmaceutical, medical, food or environmental analysis. Their electrocatalytic mechanisms are proposed from three aspects such as Ni(OH)₂/NiOOH mediated electrolysis, direct electrocatalysis of Ni(OH)₂ or NiOOH. Under exposure to air or aqueous solution, two distinct layers form on the Ni surface with a Ni hydroxide layer at the air–oxide interface and an oxide layer between the metal substrate and the outer hydroxide layer. The transformation from nickel or its oxides to hydroxides or oxyhydroxides could be further speeded up in the strong alkaline solution under the cyclic scanning at relatively high positive potential. The redox transition between Ni(OH)₂ and NiOOH is also contributed to the electrocatalytic oxidation of Ni and its oxides toward small molecules in alkaline media. In addition, nickel based materials or nanomaterials, their preparations and applications are also overviewed here.

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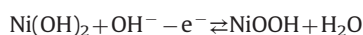
1. Introduction

Electrocatalysis toward small molecules, especially organic small compounds, is of importance in a variety of areas including fuel cells, energy storage, organic synthesis, wastewater treatment, and electrochemical sensors for medical, food or environmental analysis (Hutton et al., 2010; Wang et al., 2011).

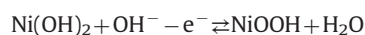
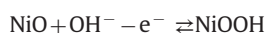
Electrocatalysis toward small molecules for various materials or substrates has been the extensively studied focus where a large number of metals (Au, Ni, Pt, Ru, Co, In, Ir, Cu etc.) and their oxides or hydroxides have been developed as electrochemical catalysts (Vedharathinam and Botte, 2012).

Compared to other metals, Ni with higher natural abundance is economically feasible, suitable for numerous applications in batch. Also, it has low toxicity. In 1966, Nesterov and Korovin pioneeringly studied the anodic oxidation of hydrazine on nickel in alkaline solution (Nesterov and Korovin 1966). In 1970s, Fleischmann et al. reported the electrochemical oxidation of some organic compounds at a Ni anode in alkaline solution (Fleischmann et al., 1971, 1972a, 1972b). After then, many Ni based materials such as Ni²⁺ (Raouf et al., 2009); Ni (Danaee et al., 2010); NiO (Sattarahmady et al., 2010), Ni(OH)₂ (Wang et al., 2011) and NiOOH (Hutton et al., 2010) had been reported, exhibiting excellent electrocatalytic activity toward the oxidation of a wide range of small compounds, such as glucose, glycine, methanol, ethanol, cyclohexanol, insulin, ammonia, acetylcholine etc. in alkaline media (Jafarian et al., 2003; Vedharathinam and Botte, 2012).

However, there have been various or even divergent expressions about their electrocatalytic mechanism. Usually, the Ni(OH)₂/NiOOH shift reaction is used to describe the redox couple mediated electrocatalysis for each of Ni, NiO and Ni(OH)₂ (Fleischmann et al., 1971):



Sometimes, the authors simply used Ni²⁺/Ni³⁺ rather than Ni(OH)₂/NiOOH to explain such a mechanism (Danaee et al., 2010). In the study of Liu et al. and Shamsipur et al., a pair of redox peaks and electrochemical response to glucose at Ni nanoparticle or NiO/carbon nanotubes modified electrodes in NaOH solution were attributed to the shift of Ni(II)/Ni(III) redox couple (Liu et al., 2009a, 2009b; Shamsipur et al., 2010). Mahshid et al. contributed the electrochemical signals of the Ni(II)/Ni(III) peaks to the following two possible electrochemical reactions (Mahshid et al., 2011):



Cheek and Grady reported their research with the title of “Redox behavior of the nickel oxide electrode system: quartz crystal microbalance studies” (Cheek and O’Grady, 1997). However, what the authors really studied was Ni(OH)₂ deposited onto the platinum films by cathodic reduction of nickel sulfate.

Completely different from the Ni(II)/Ni(III) or Ni(OH)₂/NiOOH mediated electrocatalytic mechanism, such a couple of redox peaks at NiO/carbon nanotubes electrode was even attributed to the redox couple of Ni(II)/Ni(0) and the electrocatalytic signal of glucose came from the shift of Ni(II)/Ni(0) (Zhang et al., 2010).

In addition, not all electrocatalysis toward organic small compounds is based on the redox couple mediated electrocatalytic mechanism. Sometimes, the direct electrocatalysis of some organic small compounds occurred at Ni(OH)₂ or NiOOH (Hutton et al., 2010; Jia et al., 2011).

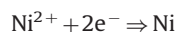
In this review, we comprehensively summarize the recent progress in the electrocatalysis and electroanalysis of nickel, its oxides, hydroxides and oxyhydroxides toward small molecules, especially organic small compounds. Their electrocatalytic mechanisms are well discussed. Their preparations and applications are described as well. Most of the relevant literatures appeared in recent decade. One of the earliest literatures could be tracked to 1966.

2. Preparation of nickel, its oxides and hydroxides

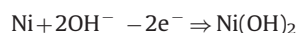
Ni, its oxides, hydroxides and oxyhydroxides can be prepared in a large number of ways, including electrochemical growth, chemical precipitation or physical techniques, etc.

2.1. Electrochemical deposition

Electrochemical approaches are often employed to deposit Ni, Ni(OH)₂ or NiOOH. Usually, nickel is cathodically electrodeposited from a nickel salt containing solution:



Electrochemical preparation of Ni(OH)₂ was achieved by applying a suitable anodic potential on Ni or Ni deposited electrodes in alkali solution (Hutton, Vidotti et al., 2010):



In some cases, it is also possible to form mixtures of Ni(OH)₂ and metallic Ni due to both of the above-mentioned processes

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