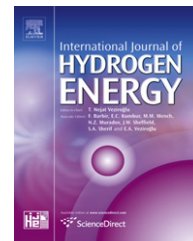


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Synthesis of ZSM-5 zeolite: Electrochemical behavior of carbon paste electrode modified with Ni (II)–zeolite and its application for electrocatalytic oxidation of methanol

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ABSTRACT

In this research, we reported a novel method for synthesis of ZSM-5 zeolite. The synthesized zeolite was characterized using X-ray diffraction, scanning electronic microscopy and FT-IR techniques. The modified carbon paste electrode was prepared by incorporation of Ni (II)–zeolite in the carbon paste matrix. The electrochemical oxidation of methanol was investigated at the surface of this modified electrode in alkaline solution using cyclic voltammetry and chronoamperometry methods. It was found that methanol was oxidized by NiOOH generated with further electrochemical oxidation of nickel hydroxide on the surface of this modified electrode during the anodic potential sweep. The effect of some parameters such as scan rate of potential, concentration of methanol, amount of Ni (II)–zeolite was investigated on the oxidation of methanol at this electrode. Also, the rate constant for the catalytic reaction (k) of methanol was obtained. Copyright © 2010, Hydrogen Energy Publications, LLC. Published by Elsevier Ltd. All rights reserved.

1. Introduction

Fuel cells are being considered as an important technology that can be used for various power applications. They are devices in which the energy of a chemical reaction is converted directly into electricity. In direct methanol fuel cells (DMFCs), methanol is used as a fuel and a great deal of interest exists in the development of materials with capability for the electrocatalytic oxidation of methanol [1]. Although some metals such as Pt- and Pt–Ru are very active in the anodic oxidation of methanol [2–4], they are too expensive for practical applications. Unfortunately, compounds such as methanol with large oxidation overpotential at ordinary carbon electrodes are not suitable analytes for these electrodes. One promising approach for minimizing overvoltage effects is in

carrying out the electrocatalytic process at chemically modified electrodes [5,6]. Zeolite-modified electrodes (ZMEs) from a sub-category of the so-called chemically modified electrodes (CME), were largely studied and promoted by Murray and co-workers [7–10].

The use of zeolites as supported electrodes represents one of the solutions overcome the problem of catalytic oxidation of methanol. Zeolites are crystalline microporous solids that contain many channel-networks proving molecular-sized cages and passageways for excellent steric control of reaction paths. For catalytic purposes, modification of zeolites is, therefore, carried out with some metals [11,12]. The pore geometry is considered as the main reason for the different activity of zeolite-based catalyst. Transition metal-containing zeolites were found to exhibit a high catalytic activity.

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Ni-containing zeolites [13], Ti-zeolites [14], Pt-and Pt-Ru-zeolites [15] have been used for catalytic purposes. This article reports a novel method for synthesizing ZSM-5 zeolite, studies of the electrochemical behavior of Nickel (II) incorporated in zeolite-modified carbon paste electrode and the application of the modified electrode for the electrochemical oxidation of methanol in 0.1 M NaOH solution.

2. Experimental

2.1. Apparatus and chemicals

The ZSM-5 zeolite was synthesized in our laboratory. Sodium aluminate was from Fison. Graphite powder, nickel chloride, potassium chloride were from Fluka. Silicic acid, tetrapropyl ammonium hydroxide (TPAOH), methanol, and sodium hydroxide used in this work were also purchased from Merck. All reagents were of analytical grade and used as received without further purification.

Electrochemical experiments were carried out using a potentiostat/galvanostat (sama 500-c Electrochemical Analysis system, sama, Iran). An Ag|AgCl|KCl (3 M) electrode as reference electrode, a platinum wire as the auxiliary electrode and Ni/ZMCPE as the working electrode were used.

The X-ray diffractograms of zeolitic sample was measured by using an X-ray diffractometer (XRD, GBC MMA Instrument, $\lambda = 1.5418 \text{ \AA}$ and 28 mA). FT-IR spectrum was recorded at room temperature using FT-IR spectrometer (Vector 22-Bruker), in the range of $450\text{--}2000 \text{ cm}^{-1}$. Scanning electron microscopy (SEM) was done on selected samples to determine the crystallite size and morphology using a JEOL JXA-840 SEM.

2.2. Synthesis of ZSM-5 zeolite

Silicic acid and sodium aluminate were used as Si and Al sources, respectively, (TPAOH) was used as structure directing agent. The sodium source was NaOH pellets. Seeding gel was prepared by mixing 36 ml of water, 0.5 g sodium hydroxide, 0.04 g TPAOH and appropriate amount of silicic acid. The resulting mixture was aged at $100 \text{ }^\circ\text{C}$ for 16 h. In the second step, 0.35 g sodium hydroxide and 0.462 g sodium aluminate were dissolved in water and mixed thoroughly. Then 5.202 g silicic acid was added and the synthesized gel was shaken at ambient temperature. After that 2.10 g of seeding gel was added and shaken. The gel was transferred to PTFE-lined stainless steel autoclave and placed in an oven at $180 \text{ }^\circ\text{C}$ for 40 h. The zeolite was calcinated in an electrical furnace at $550 \text{ }^\circ\text{C}$ for 6 h to remove of organic template.

2.3. Characterization of synthesized zeolite

XRD pattern of the synthesized sample is illustrated in Fig. 1 with comparing the main peaks at $2\theta = 7.9, 8.9, 23.2$ and 24.5 with the reference sample [16–18], crystallization of almost pure ZSM-5 phase will be ascertained.

FT-IR spectrum of the synthesized sample is shown in Fig. 2. The bands around $790, 1080\text{--}1200 \text{ cm}^{-1}$ are characteristic of SiO_4 tetrahedron units. The absorption bands at 1219 and 542 cm^{-1} provide information on the difference between

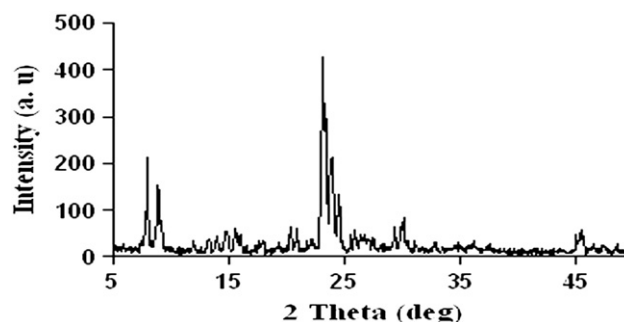


Fig. 1 – XRD patterns of synthesized zeolite.

ZSM-5 zeolite and other type of zeolites. The external asymmetric stretching vibration near 1219 cm^{-1} is due to the presence of structures containing four chains of five-member rings of ZSM-5 structure. The band near 790 cm^{-1} is assigned to the symmetric stretching of external linkages and the one near 542 cm^{-1} is attributed to a structure-sensitive vibration caused by the double five-member rings of the external linkages. The absorption band near 450 cm^{-1} is due to the T–O bending vibrations of SiO_4 and AlO_4 internal tetrahedral. The presence of absorption bands around 542 and 450 cm^{-1} are characteristic of the ZSM-5 crystalline structure [19,20].

The SEM image of crystalline phase is a useful approach that one can determine the size and morphology of obtained crystals. SEM micrograph of the synthesized zeolite sample is illustrated in Fig. 3, which the formation of cubical crystallites with average particle size of $\sim 1 \mu\text{m}$ can be seen.

2.4. Electrode preparation

0.5 g of ZSM-5 zeolite was lightly ground and immersed to 5 ml 0.1 M NiCl_2 solution for 3 h. Then the solid was dried in oven at 347 K for 8 h. The zeolite exchanged washed with deionized water to remove surface-adsorbed species. The 30% (W/W) zeolite loading Ni^{2+} was made by mixing the given quantity of zeolite loading Ni^{2+} in diethyl ether and hand mixing with 70 times its weight of graphite powder. The solvent was evaporated by stirring a mixture and paraffin was blended by hand mixing and the resulting paste was inserted in the bottom of a glass tube (with internal radius 1.5 mm). At least, modified electrode was immersed in 0.1 M Ni solution for 5 min.

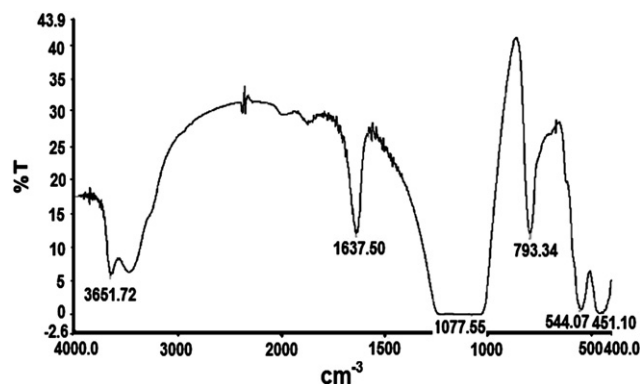


Fig. 2 – FT-IR spectra of synthesized ZSM-5 zeolite.

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