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# Ni–Pt sinter as a promising electrode for methanol electrocatalytic oxidation

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## ABSTRACT

Methanol is one of the chemical compounds utilized in fuel cells. The direct methanol fuel cell (DMFC) can be applied in many devices such as light electric vehicles and field equipment. Such a fuel cell is characterized by its high fuel energy density and low pollution. Despite many advantages of DMFCs, they are not commercially available, as the most efficient catalyst, which can be used in this process, has not been developed yet. Traditionally, it was platinum that was used in these fuel cells which is expensive and susceptible to CO poisoning. The solution to this is the use of bimetallic catalysts such as a Ni–Pt system. In this study, we used a sintered Ni–Pt electrode as the anode for the electrocatalytic oxidation of methanol. Based on our results, the sintered Ni–Pt electrodes exhibited much higher activity in the oxidation of methanol, when compared with some conventional anodes.

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## Introduction

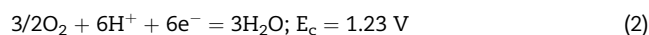
The electrocatalytic oxidation of methanol is a promising process for use in direct methanol fuel cells (DMFCs). DMFC uses methanol as a liquid fuel and it is a variant of the proton exchange membrane (PEM) fuel cell [1]. This fuel cell can be applied as an alternative for many energy devices and can be utilized in portable electronic devices, light electric vehicles, and field equipment. DMFCs are characterized by their high fuel energy density, low pollution, simplicity, rapid start-up as well as their ease of fuel recharging [2]. Additionally, DMFCs can work with high efficiency at temperatures below 100 °C. Moreover, the complexity and cost of the DMFCs are reduced as there is no need to use the fuel processor to convert the hydrocarbon

fuel into a consumable source of hydrogen. The DMFC has a maximum thermodynamic voltage of 1.18 V at 25 °C. The reactions which take place in the DMFC are as follows:

- Anode reaction:



- Cathode reaction:



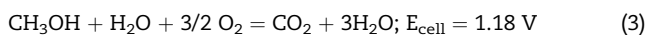
- Overall reaction:

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Despite the numerous advantages of DMFCs, they are not commercially applied. The choice of suitable electrocatalytic material is a major cause of limiting of this technology in the industry [1]. Therefore, the most important issue connected with the use of DMFCs is finding the effective and low-priced anode catalyst. Among the various types of electrocatalysts currently being investigated, the most popular is platinum. This metal exhibits excellent activity towards methanol oxidation [3,4]. However, the principal disadvantage of Pt and Pt-based catalysts is their propensity to poisoning by intermediate products of methanol oxidation, especially carbon monoxide. Further drawbacks of platinum catalysts are their high cost, slow kinetics, low stability, and low methanol-tolerance capacity [5,6]. For this reason, low Pt loadings, or Pt-X bimetallic catalysts are typically utilized. An alternative to platinum is palladium, due to its lower cost and better resistance to CO poisoning. Pd exhibits high activity in alkaline solutions during methanol oxidation. Nevertheless, the stability and activity of palladium are too low to justify their use in commercial applications [3,7]. Another option to obtain an active, low-cost catalyst, resistant to CO poisoning, and containing platinum is to combine Pt with other metals such as Ni [8–11], Co [12–14], Cu [15], Sn [16], Au [17], Ag [18], Ru [19] or Pd [20,21]. However, to create a reduced-cost and highly active catalyst for methanol oxidation, the best choice is to use a Ni–Pt electrode systems [22]. In Table 1 the comparison of bimetallic catalysts containing platinum or nickel are shown.

In this work, a sintered Ni–Pt electrode was employed as an anode for the electrocatalytic oxidation of methanol in an alkaline solution. The Ni–Pt catalyst has unique properties. Pt is the metal with the most favorable electrocatalytic properties in relation to the methanol oxidation reaction. Whereas, nickel significantly reduces the costs and the degree of catalyst poisoning [23–32]. The combination of these two metals is a beneficial alternative to the use of platinum. Other works concerning the combination of nickel and platinum confirm this statement. The unique structure of the produced catalyst also causes the catalyst to exhibit more favorable or even electrocatalytic properties in the methanol oxidation as it is already known in the literature of Ni–Pt catalysts. The effects of different scan rates during the cyclic voltammetry and the concentration of methanol on the process were investigated. The activity of the new Ni–Pt electrode was compared with that of Ni and Ti/Pt electrodes.

## Experimental

For the preparation of Ni–Pt sinters a spherical nickel powder (Alfa Aesar) of 99.8% purity and a platinum powder (Goodfellow) of 99.99% purity were used. A detailed methodology of the Ni–Pt sinters preparation was described in the previous papers [33,34]. The Ni–Pt sinters surface area may be approximated by their geometric surface [34–36].

The electrochemical activity of the Ni–Pt sinters was compared with that of Ni and Ti/Pt electrodes. The Ti/Pt electrode was prepared by electrochemical deposition of Pt on

a Ti substrate. The Ni electrode was a nickel plate (99.99%) [37]. The structures of the Ni–Pt, Ni, and Ti/Pt electrodes surfaces were described in our previous paper [37].

Electrochemical measurements of Ni–Pt, Ni, and Ti/Pt electrodes were conducted in 1 M KOH (Chempur, Poland), in both the absence and presence of methanol (Chempur, Poland) at varying concentrations (0.0, 0.1 and 1.0 M). The apparatus included a standard electrolysis cell with the following three electrodes: a working electrode, platinum auxiliary electrode and Haber-Luggin capillary containing a reference electrode (saturated calomel electrode, SCE). The electrolysis cell was powered by a potentiostat (PARSTAT 4000, Ametek) equipped with Versa Studio software. The investigations included the following measurements:

- cyclic voltammetry from  $-0.8$  to  $0.2$  V vs. SCE at scan rates of  $10$ ,  $50$  and  $100$   $\text{mV s}^{-1}$ ,
- chronoamperometry at a constant potential of  $-0.3$  V vs. SCE for  $1$  h.

## Results and discussion

The cyclic voltammograms of the Ni–Pt electrode obtained in 1 M KOH solution are shown in Fig. 1a. A peak at  $-101$  mV (at  $10$   $\text{mV s}^{-1}$ ) is visible in the anodic region of the curves. This peak corresponds to the oxidation of Ni to  $\text{Ni}(\text{OH})_2$ . The onset potential of the nickel oxidation is  $-350$  mV. At negative potential in alkaline solution, the nickel electrode can be oxidized to  $\text{Ni}(\text{OH})_2$  [38]. This compound can occur in two crystallographic phases:  $\alpha$ - $\text{Ni}(\text{OH})_2$  and  $\beta$ - $\text{Ni}(\text{OH})_2$  [39,40]. As potential increases in time, the hydrous and less stable  $\alpha$ - $\text{Ni}(\text{OH})_2$  form is converted to the anhydrous and more stable  $\beta$ - $\text{Ni}(\text{OH})_2$ . When the potential is increased even further the  $\beta$ - $\text{Ni}(\text{OH})_2$  can be oxidized to  $\beta$ - $\text{NiOOH}$ , which then may be partially converted to  $\gamma$ - $\text{NiOOH}$  [41]. The second visible peak observed at  $-221$  mV (at  $10$   $\text{mV s}^{-1}$ ) in the cathodic region corresponds to the reduction reaction of  $\text{Ni}(\text{OH})_2$  to Ni. The third visible peak at  $-470$  mV, also in the cathodic region, corresponds to the desorption of hydroxyl groups on the catalyst surface and/or reduction of platinum oxide (PtO). The adsorption peak of the hydroxyl groups or formation of PtO is not visible as it is masked by the peak caused by the oxidation of nickel. These peaks relate to the presence of platinum on the electrode surface. This confirms the position of the peaks on the voltammetric curve for the Ti/Pt electrode (peaks at  $-46$  mV in the anodic region and  $-307$  mV in the cathodic region) [42,43]. The current density of the nickel oxidation peak and the current density in the vertex potential increase with the increasing scan rate. At the same time, the current density of the reduction peak of nickel hydroxide decreased. With the increase of the scan rate, the oxidation peak moved towards more positive potential values and the reduction peak was shifted towards the more negative potentials. Thus, the reactions involved were not reversible reactions. On the curve obtained for the Ni electrode, the same oxidation and reduction peaks ( $\text{Ni}/\text{Ni}^{2+}$ ), as for the Ni–Pt electrode, were present, however, the values of the current density peaks were much smaller, especially the current density of the reduction peak. In the 1 M KOH solution, the Ni–Pt electrode

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