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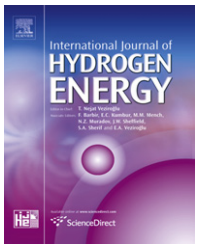

Combinatorial investigation of Pt–Ru–Sn alloys as an anode electrocatalysts for direct alcohol fuel cells

Young Hwan Chu [a,*], *Yong Gun Shul* [b]

[a] *Department of New Energy·Resource Engineering, College of Science & Engineering, Sangji University, 124, Sangjidae-gil, Wonju-si, Gangwon-Do 220-702, Republic of Korea*

[b] *Department of Chemical and Biomolecular Engineering, Yonsei University, 134, Shinchon-Dong, Seodaemun-Gu, Seoul 120-749, Republic of Korea*

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ABSTRACT

Low-temperature direct alcohol fuel cells fed with different kinds of alcohol (methanol, ethanol and 2-propanol) have been investigated by employing ternary electrocatalysts (Pt–Ru–Sn) as anode catalysts. Combinatorial chemistry has been applied to screen the 66-PtRuSn-anode arrays at the same time to reduce cost, time, and effort when we select the optimum composition of electrocatalysts for DAFCs (Direct Alcohol Fuel Cells). PtRuSn (80:20:0) showed the lowest onset potential for methanol electro-oxidation, PtRuSn (50:0:50) for ethanol, and PtRuSn (20:70:10) for 2-propanol in CV results respectively, and single cell performance test indicated that Ru is more suitable for direct methanol fuel cell system, Sn for direct ethanol fuel cell system, and 2-propanol could be applied as fuel with low platinum composition anode electrocatalyst. The single cell performance results and electrochemical results (CV) were well matched with the combinatorial electrochemical results. As a result, we could verify the availability of combinatorial chemistry by comparing the results of each extreme electrocatalysts compositions as follows: PtRuSn (80:20:0) for methanol, PtRuSn (50:0:50) for ethanol and PtRuSn (20:70:10) for 2-propanol.

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1. Introduction

Electric vehicles powered with polymer-electrolyte fuel cells using hydrogen as fuel are being tested for vehicular applications. However it is very inconvenient to store and transport hydrogen in vehicles. Therefore low-temperature polymer-electrolyte membrane fuel cells (PEMFCs) fueled directly with liquid fuels are one of the alternative potential candidate application for fuel cell vehicles, stationary application and portable power sources such as mobile phones, PDAs, and laptops [37]. Operation on liquid fuels without the external bulky fuel-reforming system could greatly simplify the fuel cell system, therefore would result in its rapid practical use. This advantage makes direct methanol fuel cells (DMFCs) the most promising alternative to lithium ion batteries for application in portable power sources. Present lithium ion batteries suffer from long recharge times, while DMFCs can be easily refueled in a short time [38]. At present DMFCs are being actively investigated and during the last a couple of decades significant progress has been achieved in this field [1–3].

Ethanol [3–5], 1-propanol [6], and 2-propanol [7,8] are also liquid fuels which are easily available. The use of alcohols with longer chains for applications in fuel cells can also be attractive considering the higher energy content of the fuel. Methanol, ethanol and 2-propanol have a very good energy density (We) such as 6.09, 8.00, and 8.58 kWh/kg respectively, close to that of

* *Corresponding author*. Tel.: +82 33 730 0456; fax: +82 33 730 0403.
E-mail address: yhchu@sangji.ac.kr (Y.H. Chu).

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hydrocarbons and gasoline (e.g. 10–11 kWh/kg) [39]. These fuels for Direct Alcohol Fuel Cells (DAFCs) are inexpensive and less toxic than methanol. Particularly, ethanol is an attractive alternative to methanol as a fuel for fuel cells since in some countries, such as Brazil, an already well-established infrastructure for the use of ethanol in automobile combustion engines exists so that an introduction of direct ethanol fuel cells for electricity generation purposes may be possible without requiring any modification of the infrastructure [9]. But the present development of direct alcohol fuel cells is not so satisfying as much as DMFCs. There are several obstacles for DAFCs to overcome before their commercialization. One of the main problems of DAFCs is the poor performance of electrocatalysts, especially anode catalysts at lower temperatures because of the anode surface poisoning by CO-like intermediates [10]. Making alloys with a second or a third metal is a convenient way to modify electrocatalytic properties of Pt in order to overcome poisoning effects. Notable enhancement of the electrocatalytic activity of platinum-based electrocatalyst has been observed by introducing a second metal: Pt–Ru, Pt–Sn, Pt–Pd, Pt–W [11].

One way to reduce the cost, time, and effort, when develop new electrocatalysts, is applying the combinatorial electrochemistry to this research field. The first report of combinatorial electrochemical screening was that of Mallouk and coworkers [12]. Their method was based on an optical response of a fluorescent dye due to changes in the local pH by the electrochemical reaction of interest. In their first report, the method was applied to the screening of catalysts for methanol oxidation. Subsequently, the method has been applied to the discovery of electrocatalysts for oxygen reduction and water oxidation [13,14] and catalysts for amperometric glucose sensors [15]. The method has the advantage of the simplicity of a single working electrode and the electrochemical control and measurement using a single potentiostat and current follower.

The use of an array of independently addressable catalyst electrodes, rather than a single electrode on which an array of catalysts is deposited, enables the direct current measurement at each electrode and obviates the requirement of an indicator. Such arrays of electrodes may be screened rapidly in series either using moveable counter and working electrodes that are brought into electrochemical contact with each of the individual working electrodes in turn [16,17] or using a single electrochemical cell containing all of the working electrodes in the same electrolyte solution and fixed counter and reference electrodes [18,19]. The moveable electrode method has been applied to the screening of a 144-element array of commercial electrocatalyst powders in Nafion for methanol oxidation [16] and to the screening of photoelectrochemical materials [17]. This approach, although economical, requiring only one potentiostat and current follower, is inherently slow as a result of the need to move the counter and reference electrodes. The serial examination of each electrode in the array or library in the same electrochemical cell using one potentiostat and current follower has been applied to the study of organosulfur monolayers adsorbed on gold electrodes [20,21]. Improved multiplexing in the screening of electrochemical libraries has been reported by Hintsche et al. [22]. They developed special ASIC switches that enabled the application of a potential to all the working electrodes of the library via bias lines, whereas the currents from individual working electrodes were read in series. Such a pseudoparallel method is more economical than very large multipotentiostats while retaining much of the multiplexing advantage of such systems.

In this study, we have applied the combinatorial chemistry to fuel cell anode electrocatalysts research to reduce cost, time, and effort when we select the optimum metal composition of electrocatalysts for DAFCs. Platinum is commonly used metal electrocatalyst for organic liquid fuel cells. However, platinum single metal itself does not show quiet good performance. Therefore, many research groups have applied Pt–Ru binary electrocatalyst and Pt–Sn binary electrocatalyst for enhancement of methanol and ethanol electrooxidation activity respectively [6,33,36]. As a result, we combined the two binary electrocatalysts system as one ternary electrocatalyst system for various alcohols electrooxidation reaction. Pt–Ru–Sn metal alloys system was applied as the anode electrocatalysts for the direct alcohol (methanol, ethanol and 2-propanol) fuel cells. The optimum compositions of electrocatalyst for the each alcohol were chosen by combinatorial chemistry and carbon supported electrocatalysts were prepared by impregnation method. All carbon supported Pt–Ru–Sn/C anode catalysts were characterized by X-ray diffraction (XRD). The electrochemical properties of the electrodes which contain different compositions of Pt, Ru and Sn, were tested by cyclic voltammetry, impedance spectroscopy and a direct alcohol fuel cell performance test. We have investigated the availability of combinatorial chemistry on electrocatalysts development by comparing the combinatorial chemistry results with the results of electrochemical analysis method such as single cell performance and cyclic voltammetry.

2. Experimental

2.1. *Combinatorial chemistry*

Fig. 1 shows the experimental scheme of combinatorial screening method to select the active catalysts for each alcohol (methanol, ethanol, 2-propanol) electro-oxidation reaction. Electrode arrays were prepared by dispersing 0.5 M aqueous solutions of three metal salts (H_2PtCl_6, $RuCl_3$, Aldrich, and $SnCl_2$, Junsei) onto a Teflon-coated Toray carbon sheet, using manually prepared solution mixtures. The completed array was reduced by 0.5 M sodium borohydride, and the reduced array was thoroughly washed with nanopure water. Fig. 2 shows the combinatorial electrode array map of Pt–Ru–Sn ternary electrocatalysts for alcohols electrooxidation.

The electrolyte solution was prepared with 100 μM proton indicator (Quinine), 6 M of reactant alcohol, and the diluted H_2SO_4 which was added to adjust the solution pH to 7. The potential-step experiments were carried out using a potentiostat/galvanostat controlled by an IBM PC. Each composition was used as a working electrode. Cyclic voltammetry experiments were performed between 0.0 V and 0.8 V RHE at a sweep rate of 1 mV/s. The most active working electrode was selected by the brightest spot of the array obtained by fluorescence emission excited by UV of 254 nm [23].

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