

Electronic Supplementary Information

for

Membraneless hydrogen peroxide fuel cell using Prussian Blue as cathode material

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Synthesis of Prussian Blue (PB)

In order to prepare Prussian Blue, an amount of 0.5 M FeCl_3 from Sigma-Aldrich in 0.1 M HCl from Sigma-Aldrich was added dropwise to a solution of $\text{K}_3[\text{Fe}(\text{CN})_6]$ from Sigma-Aldrich in 0.1 M HCl at room temperature. An immediate formation of dark blue colloid was observed. The solution was stirred for 1 hour. The produced PB was collected by filtration through a PTFE filter with an average pore size of 0.22 μm from Millipore. The collected PB was washed with 0.1 M HCl until the filtrate become colorless. PB was removed from the filter and kept in an oven at 100 °C for 3 hours. To make PB supported on carbon, the required amount of Vulcan XC-72 was suspended in 0.1 M HCl and agitated in an ultrasound water bath. The required amount of $\text{K}_3[\text{Fe}(\text{CN})_6]$ in 0.1 M HCl and 0.5 M FeCl_3 from in 0.1 M HCl were added dropwise to the carbon slurry, respectively. The remaining procedure was the same as above.

Preparation of catalyst ink and the carbon paper-based cathode

Catalyst solution for modifying the glassy carbon and for spraying on carbon paper was made by adding 4 ml Isopropanol and 1.2 ml DI water from Millipore (Direct Q) to 24 mg of PB. The solution was agitated in an ultrasound water bath for 30 minutes. Subsequently, 100 μl of Nafion with a concentration of 5% was added to the solution and again agitated for 30 minutes. For cyclic voltammetry experiment, small portions of unsupported and carbon-supported PB solution (2 μl) were placed on the glassy carbon electrode with a diameter of 4 mm and dried under a light bulb for 30 minutes.

In order to make the electrodes, the catalyst solution was sprayed on the carbon paper using a painting gun, from Badger model 100 LG, connected to a regulator to reduce the N_2 gas pressure from a cylinder. The spraying process was done under the light of an incandescent light bulb to facilitate the evaporation of water and isopropanol from the carbon paper. To

make the catalyst layer with a uniform loading of 10 mg/cm^2 over the carbon paper, frequent spraying with subsequent drying under the light was repeated until the whole solution was sprayed on the carbon paper.

Electrochemical characterization

Electrodes were connected to an electric load system (PGSTAT 302 with GPES Manager as interface software) using alligator clips. A potential scanning rate of 10 mV/s with a potential step of 10 mV was set. The characterization was carried out at room temperature of $25 \text{ }^\circ\text{C}$. All experiments were carried out using $0.5 \text{ M H}_2\text{O}_2$ in a 0.1 M HCl solution as supporting electrolyte.

For all fabricated cells, the open-circuit potential (OCV) was monitored for 5 minutes before the I-V test. As shown in Figure S1, the fuel cell with nickel anode showed an OCV of 0.60 V with a slow degradation. But silver produced a stable OCV of 0.53 V .

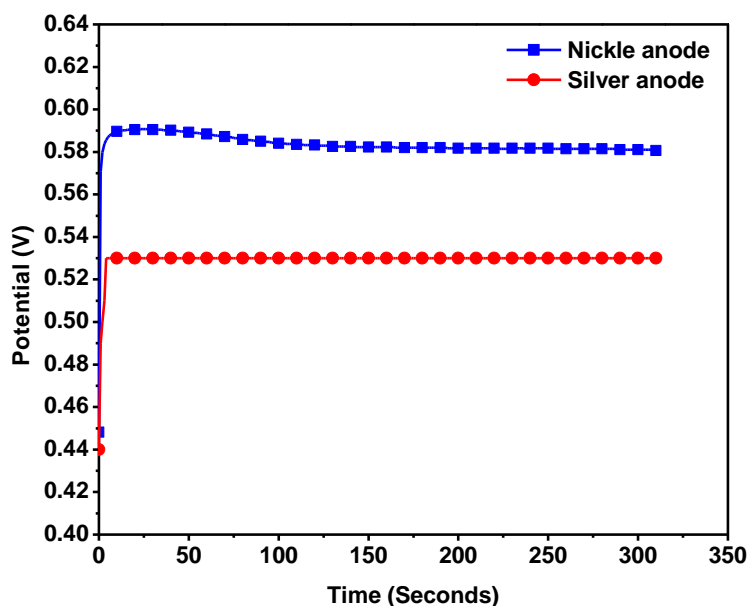


Figure S1. Open-circuit potential of the single-compartment H_2O_2 fuel cell with nickel and silver anodes and unsupported PB coated on carbon paper as cathode. Tests were done under acidic condition using 0.1 M HCl and $0.5 \text{ M H}_2\text{O}_2$.

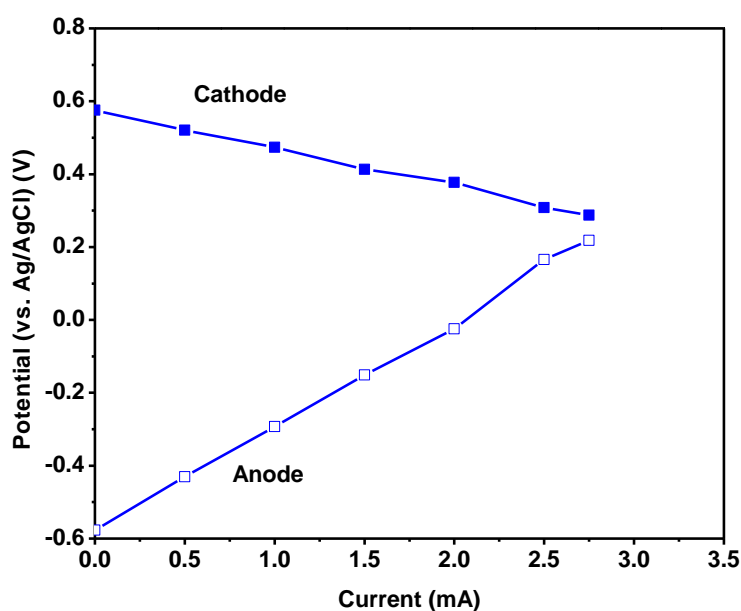


Figure S2. Steady-state anodic and cathodic polarization data for the flow-through fuel cell measured in situ at room temperature under chronoamperometry control, using a platinum electrode as a counter electrode and an external Ag/AgCl reference electrode.

Figure S2 shows individual anodic and cathodic polarization curves obtained versus an external Ag/AgCl reference electrode. This plot reveals that silver anode has a more contribution in limiting the overall fuel cell performance. This may be associated with the electrochemical kinetics and mass transport. Since no significant mass transport limitation was observed, the main contribution may be related to the electrochemical kinetics. In this case, there is a huge potential for further improvements on catalyst selection.