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Influence of Fe and Sn on selectivity of nitrate reduction at Pd alloy electrodes

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PURPOSE OF THE ABSTRACT

Nitrate in groundwater, lakes, and rivers causes pollution problem, which has attracted a lot of attention in these days [1]. It is the source of eutrophication and is also pointed out as substance damaging human health. Several treatment methods have ever been examined and developed [2]. Electrocatalytic method is one of them and attracts interests because of the cost-effective and environmental-friendly approach. Although Cu-Pd electrocatalyst is one of the promising materials for nitrate reduction, it produces some amount of ammonium. It is required to enhance the selectivity toward nitrogen by improving electrode catalyst. Cu is considered the main factor of lowering the selectivity due to the high performance of conversion nitrate to ammonium. In this paper, we examined Fe-Pd and Sn-Pd electrocatalysts since Fe and Sn are the preferable materials for reducing nitrate [3]. Fe-Pd and Sn-Pd films were prepared by co-electrodeposition, because this method was cost-effective and interesting for the nitrate reduction. The purpose of our experiment is to find the electrocatalytic behavior of these electrodes in a nitrate solution and to improve the selectivity toward nitrogen.

The surface morphology, composition, and structure of Fe-Pd and Sn-Pd alloy films were investigated by SEM, EPMA and XRD. Nitrate electroreduction were performed in three-compartment cell to avoid aqueous contamination. The prepared alloy electrode was used as working electrode, and the compartment was filled with 10 mL solution of 0.01 M KNO3 and 1 M NaOH. The other compartments were filled with solution of 1 M NaOH. The counter electrode was Pt gauze and the reference electrode was Ag/AgCl. Electrolysis were performed at room temperature. After the electrolysis, the concentrations of nitrate, nitrite and ammonia in the cathode electrolyte were measured by HPIC (ICS-1100, Thermo Scientific) and UV-Vis spectroscopy (UV-2600, Shimadzu). The amount of produced nitrogen was calculated from the other N-species concentrations because of difficulty of directly measuring nitrogen.

The activity and the selectivity depended on the film composition, while the relation was not simple. While the results of activity was contrary to expectations, pure Pd film electrode did not reduce nitrate mostly in our other experiment. It means that small Fe contents in Fe-Pd alloy film significantly promoted nitrate reduction. The result that the activity decreases with Fe contents shows that there was the most appropriate composition in the alloy. It was the same in the selectivity. In order to investigate the effect of cathode potential, the electroreduction were performed with Fe20Pd80 electrode. The activity and the selectivity were dependent on electrolysis potential. At the potential of -1.0 V, nitrate was not reduced effectively, and nitrite remained a lot. At the potential of -1.1 and -1.2 V, nitrate and nitrite were almost reduced. The selectivity toward nitrogen was 61%, 89% and 95% at each potential. The activity and the selectivity increased at the more negative potential, whereas the current efficiency decreased due to H2 evolution. The reduction rate of nitrate for Pd60Sn40 film was 77%, while it was 25% for Pd30Sn70 film. The reduction products of nitrate are mainly nitrite, ammonia, and nitrogen. Although ammonia is an undesirable final product, its production rate was 13% for Pd60Sn40 film and 16% for Pd30Sn70 film. The reduction from the nitrate ion to nitrite ion was promoted by Sn at Pd-Sn alloy electrode, and in addition, the nitrite ion is reduced to the nitrogen at Pd sites. The selectivity to the nitrogen became a maximum at Pd60Sn40 alloy electrode in higher potential condition.

FIGURES

FIGURE 1

FIGURE 2

KEYWORDS

Nitrate reduction | Fe-Pd electrocatalysts | Sn-Pd electrocatalysts | Selectivity

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