Study of Carbon-Nafion-Membrane Catalyst for Synthesis of Pure H_2O_2 Aqueous Solutions

Ichiro Yamanaka,[1]* Yu Tamada,[1] Daisuke Hiraki,[1] Hitoshi Ogihara[1]
[1] *Department of Chemical Science and Engineering, Tokyo Institute of Technology, Tokyo, 1528550, Japan*
*E-mail: yamanaka.i.aa@m.titech.ac.jp

Application of a hydrogen permeable membrane in chemical process has been expected because there is a great advantage for reduction of energy consumption. Previous application of hydrogen permeable membrane was limited for Pd based materials over 673 K.[1)] Most of all works were studied for H_2 production by steam forming at higher temperatures. A part of works described the hydrogen permeation at 298 K.[2)] They reported applications for the direct synthesis of H_2O_2 using O_2 and permeated H_2; however, it can not prevent a participation of pin halls and cracks of the Pd membrane by the hydrogen embrittlement. Purposes of this work are to develop a new type of hydrogen permeable membrane and to perform direct synthesis of pure H_2O_2 aqueous solutions from O_2 and H_2.

Hydrogen permeable membrane developed in this work consists three sheets, oxidation-catalyst sheet, proton and electron conductive sheet and reduction-catalyst sheet, as shown in Figure 1. This three-layered membrane was fabricated as below. The electron and proton conductive membrane was prepared by a casting method using a paste of carbon and Nafion materials. The paste was prepared from carbon fiber (VGCF) powder and 20wt%-Nafion solution by ultrasonic mixing. The paste was soaked in a flat-bottom Teflon-vessel and dried up at 60 ˚C. The black composite sheet was cut a round shape of 30 ø. Oxidation and reduction catalyst sheets (2.0 cm^2) were prepared from 50 wt% Pt/XC72, VGCF and PTFE powders. The composite sheet was sandwiched with Pt/XC72 sheets and these were connected by hot-pressing at 140˚C.

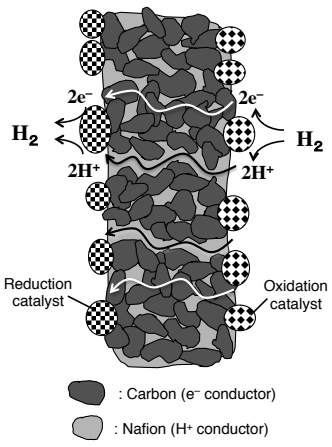

Fig. 1. Hydrogen permeable membrane.

Table 1 shows effects of various combinations of the three-layered membrane on permeation rates of H_2 and He from the oxidation-catalyst side to the reduction-catalyst side. To be note results in run 1 using the typical membrane, a high permeation rate of H_2 and a low permeation rate of He were observed. A ratio of the permeation rates of H_2 and He was 125. When XC72 without Pt was used for reduction-catalyst sheet (run 2), a very low permeation rate of H_2 as low as that of He was observed and the H_2/He ratio was unity. When XC72 alone was used for oxidation-catalyst sheet (run 3), a very similar result to the run 2 was obtained. When a pure H_2 of 1 atm was introduced to the oxidation catalyst side, a fast permeation rate of H_2 was observed (run 4).

Table 1. Permeation of a gas mixture of H_2 and He through the three-layered membrane.

Run	Red-Catal.	Ox-Catal., gases	Perm. rate / μmol min^{-1} cm^{-2}		H_2/He
			H_2	He	
1	Pt/XC72	Pt/XC72, H_2+He	175.1	1.4	125
2	XC72	Pt/XC72, H_2+He	1.4	1.4	1
3	Pt/XC72	XC72, H_2+He	1.5	1.5	1
4	Pt/XC72	Pt/XC72, H_2	250	no	–

A reduction catalyst sheet prepared from a catalyst ink of the pyrolyzed Co-TPP/VGCF and Nafion solutions was used for a direct synthesis of pure H_2O_2 aqueous solution from H_2 and O_2. Products were only H_2O_2 and water in the reduction-catalyst compartment and no H_2. The maxima of the concentration of 3.8 mol dm^{-3} and the selectivity of 42% were achieved.[3)]

REFERENCES

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