Preferential oxidation of CO in reformate on Pt-Fe/MOR catalyst

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1. Introduction

Polymer electrolyte fuel cells (PEFCs) are expected as residential power sources. But, a conventional Pt anode catalyst used for PEFCs operating with reformed gases seriously poisoned by small amount of CO. Preferential CO oxidation (PROX) is one of the best candidates to solve this problem. We have investigated Pt catalysts supported on zeolite and found mordenite is the best support for PROX catalysts.

In this presentation, we will report the PROX performances of Pt, Fe, Pt-Fe catalysts supported on mordenite. Moreover, we tested about the optimum composition of Pt and Fe. Finally, we will compare the PROX activities with that of Pt/Al_2O_3 catalyst which is conventionally used as PROX catalyst.

2.Experimental

Pt, Fe, Pt-Fe catalysts supported on mordenite (a reference catalyst support supplied by the Catalysis Society of Japan) were prepared by a conventional ion-exchange method (1-3). They denoted as Pt/M, Fe/M, and Pt-Fe/M, respectively. The PROX activity test was carried out in a conventional flow reactor with pyrex tube (with 6mm inner-diameter). Two mass flow controllers were used to make reaction mixture consisted of 1.0% CO, 0.5% O2, and H2 balance. On-line gas-chromatograph with a TCD detector (Hitachi Ltd. GC 263-30) was used to measure inlet- and outlet-gas composition. Catalyst activity was evaluated by CO and O2 conversion and CO selectivity. Before activity test, the supported catalyst was reduced at 300℃.

3. Results and discussion

PROX activity of 4wt%Pt, 2wt%Fe, 4wt%Pt-2wt%Fe/M catalysts were shown in Fig.1. In Pt/M catalyst, we could not detect an apparent PROX activity from 100 to 200℃. Also, Fe/M catalyst has no PROX activity from 100 to 300℃. But, Pt-Fe/M catalyst has PROX activity from 100 to 300℃. We think when both Pt and Fe was supported on mordenite, the PROX activity was appeared by the synergetic cooperation between both components even below 150℃, presumably in the binary alloy phase.

Next, we tested PROX activity of Pt-Fe/MOR catalyst as a function of Fe loading. The result is shown in Fig.2. Here, Pt loading was fixed at 4wt%. The PROX activity was influenced by Fe loading amounts. We found when Fe loading is 0.5wt%, Pt-Fe/MOR catalyst has the highest PROX activity.

Dependence of PROX activity upon the operating temperature was tested on the catalyst of 4wt%Pt-2wt%Fe/M. As seen in Fig.3, this catalyst showed extremely high activity at 200℃. In particularly, we note that CO conversion and selectivity was beyond 90% at 50℃ in the stoichiometric mixture (λ =1).

4.Reference

(1) M. Watanabe, H. Uchida, Chem. Lett. (1995) 21
(2) H. Igarashi, H. Uchida, and M. Watanabe, Chem. Lett. (2000) 1262
(3) H. Igarashi, H. Uchida, and M. Watanabe, Stud. Surf. Catal. 132 (2001)953

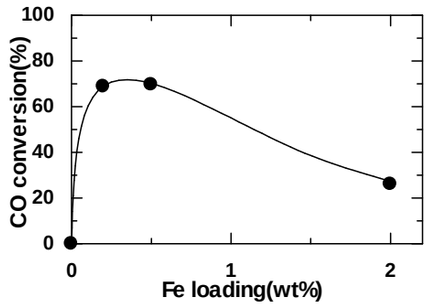

Fig.2 Effect of Fe loading on PROX for Pt-Fe/MOR catalyst

gas mixture:CO 1.0%,O_2 0.5%,H_2 balance
amount of catalyst:0.05g
W/F=0.03g・s・cm^{-3}
reaction temperature:100 ℃
pretreatment temperature:300 ℃
Pt loading :4wt%

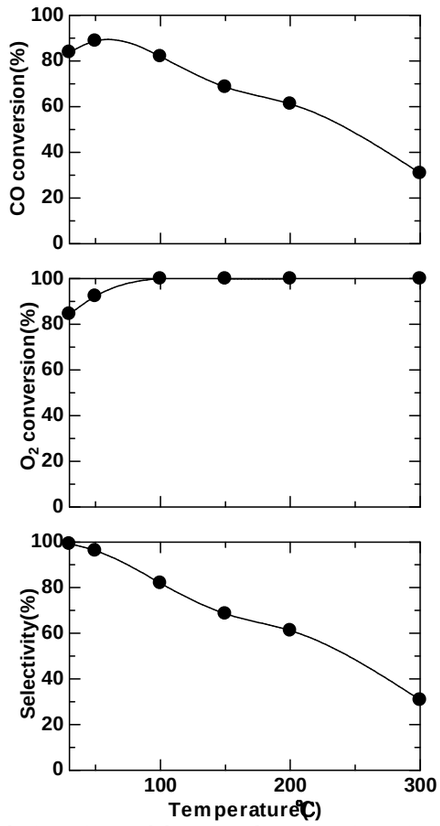

Fig.3 PROX activity test of 4wt%Pt-0.5wt%Fe/MOR

amount of catalyst:0.05g
gas mixture:1%CO,0,5% O_2 ,H_2 balance
W/F=0.03g・s・cm^{-3}