

Design and analysis of bimetal supported zeolite catalysts for selective CO oxidation in reforming gas for fuel cells

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Introduction

The hydrogen used as fuel in polymer electrolyte fuel cell (PEFC) should be essentially free from CO to avoid poisoning the Pt-based anode catalyst⁽¹⁾. Selective oxidation of CO in hydrogen-rich reformer gas is an important reaction in the PEFC technology.

We found that Pt supported on mordenite showed higher conversion of CO to CO₂ than that of conventional Pt/Al₂O₃ catalyst used for a preferential oxidation of CO (PROX) in hydrogen-rich gas⁽²⁾. Recently, we found Pt-Fe/mordenite exhibited higher CO conversion and selectivity than Pt/mordenite catalyst.

In this report, we investigate an electronic state of Pt particles supported on mordenite before/after the preferential oxidation of CO (PROX) by in-situ XAFS.

Experimental

Pt-Fe/mordenite (donated as Pt-Fe/M) catalysts were prepared by ion-exchange of Pt(NH₃)₄Cl₂ and dried in vacuum evaporator.

Samples were oxidized and then reduced at 500°C with O₂ and H₂, respectively. After the sample was used for the PROX at 473K in 1%CO, 0.5%O₂, and H₂ balance gas for 1h, it was cooled down to room temperature in same gas.

In-situ EXAFS measurements of Pt LIII edge were carried out at room temperature in PF BL-9A. The data were collected in transmission mode.

Subtraction of the background and normalization of the spectra by using the program autobkg were employed to lead EXAFS function $\chi(k)$. For Fourier transformation of the $\chi(k)$ to R space, Gaussian Window was used. The phase shift and backscattering amplitude factor were corrected by using the calculated value of FEFF8.

Results and discussion

Figure 1 shows Pt LIII edge XANES spectra of ion-exchanged catalyst just after drying and those of some compounds as the reference. The edge position of the catalyst is near to that of Pt foil. Therefore, we concluded that Pt species in Pt-Fe/M was assigned to zero-charged Pt. We also performed EXAFS measurement on the same Pt-Fe/M catalyst. It was found that Pt is coordinated with four nitrogen atoms. So we conclude Pt species exist in mordenite cage as Pt(NH₃)₄.

Figure 2 shows Pt LIII edge XANES spectra on Pt-Fe/M catalysts after the PROX measurement and some reference compounds. The edge position of Pt-Fe/M is near to that of PtO₂. This indicated that Pt species in Pt-Fe/M catalyst have electron deficiency. We think that such a high activity of Pt-Fe/M catalysts might be ascribed to this anomalous oxidative Pt state.

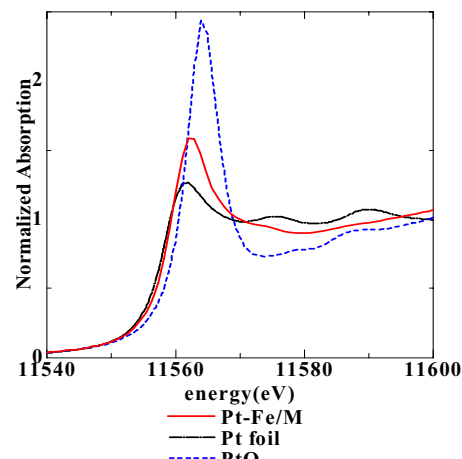


Fig.1 XANES spectra of Pt-Fe/M catalyst after dry

Table1 results of EXAFS analysis

Scattering atom	CN	R/ Å	$\sigma^2 / \text{Å}^2 \times 10^{-3}$	$\Delta E_0/\text{eV}$	R/%
N	3.9	2.04	2.6	6.6	1.7

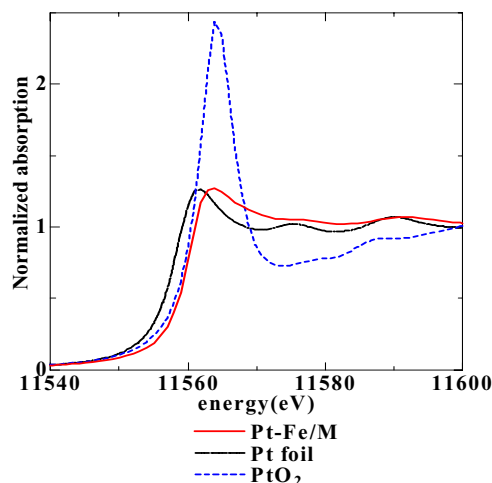


Fig.2 XANES spectra of Pt-Fe/M catalyst after treatment

References

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