

## Catalysts for the Removal of CO in a Reformed Gas by Selective Oxidation – Influence of CO<sub>2</sub> and H<sub>2</sub>O on the CO Oxidation over Pt-Alloy Supported on Zeolite

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In polymer electrolyte fuel cells (PEFCs), a small amount of CO contained in a reformed gas is known to lower the performance of PEFC by poisoning Pt anode catalyst. Therefore, it is essential to develop a simple system, which can minimize the CO level up to ppm level.<sup>1-5</sup> We have shown that Pt-Ru or Pt-Fe supported on mordenite exhibit extremely high activity and selectivity for the CO oxidation.<sup>3-5</sup> In this study, we present this preferential oxidation of CO on Pt-Fe/mordenite at low temperature in details.

Pt-Fe (2:1; weight ratio) catalyst was supported on mordenite by a conventional ion-exchange method.<sup>1-5</sup> The loading amounts of Pt and Fe were 4wt% and 2wt%, respectively. Before usage, it was heat-treated in a reactor under an O<sub>2</sub> flow for 1 h and H<sub>2</sub> flow 1 h at 500°C. The catalytic oxidation of CO was carried out in a conventional flow reactor. The reactant mixture consisted of 1.0% CO, 0.5% O<sub>2</sub>, and H<sub>2</sub> balance or additional CO<sub>2</sub> and/or H<sub>2</sub>O. Flow rate condition (W/F = amounts of catalyst loaded to the flow rate of reactant gas) was 0.03 g s cm<sup>-3</sup>. On-line gas chromatograph with TCD detectors was used to measure reactant- and product-gas compositions. The lower limit of CO detection was 20 ppm in this work.

Figure 1 shows dependencies of CO conversion, O<sub>2</sub> conversion, and CO selectivity at the Pt-Fe/mordenite on reaction temperature. The CO conversion and the selectivity of 100% were achieved at the temperature from 200°C to 80°C. By the FTIR measurement on the Pt-Fe/mordenite, no CO adsorption was seen at the low temperature of 30°C. Since CO adsorption was lowered and weakened on Pt sites on the alloy surface, an enhanced adsorption of O<sub>2</sub> may occur on sites free from CO, resulting in the enhanced CO oxidation. At higher temperature than 200°C, the performances are lowered noticeably, probably due to a reverse shift reaction described below.

About 25% of CO<sub>2</sub> and 20% of H<sub>2</sub>O are usually contained in a reformed gas. So, the effects of CO<sub>2</sub> and H<sub>2</sub>O contents on the preferential oxidation of CO were examined. Figure 2 shows the effect of CO<sub>2</sub> concentration on the Pt-Fe/mordenite performances. At a low temperature such as 100°C or less, the influence was not observed at all, but at higher temperature the reverse shift reaction (CO<sub>2</sub> + H<sub>2</sub> = CO + H<sub>2</sub>O) tended to occur. The reverse shift reaction was enhanced as the CO<sub>2</sub> concentration increased, *e.g.*, at 200°C and 25% CO<sub>2</sub>, CO concentration became 1600 ppm. As shown in Fig. 3, however, the reverse shift reaction was noticeably suppressed by the presence of additional 20% H<sub>2</sub>O. The CO selectivity is dramatically improved without prevention of the CO conversion, *i.e.*, *ca.* 100% at 80°C to 200°C. The performance was stable during all of the experiments.

### References

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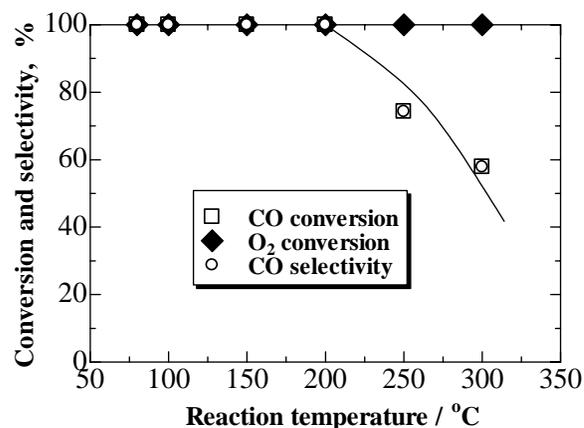


Fig. 1. Dependencies of CO conversion, O<sub>2</sub> conversion, selectivity on reaction temperature. CO 1.0%, O<sub>2</sub> 0.5%, H<sub>2</sub> balance, W/F=0.03 g s cm<sup>-3</sup>.

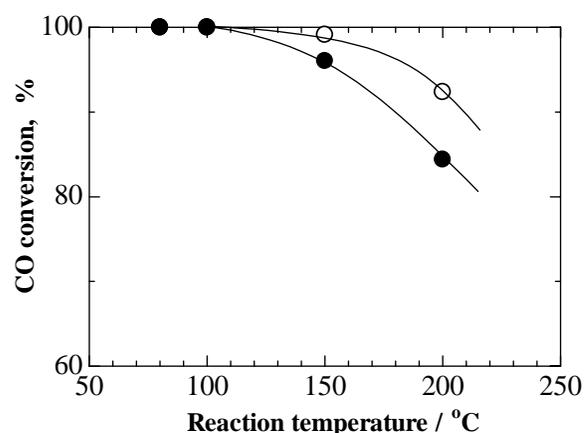


Fig. 2. Dependencies of CO conversion in the presence of CO<sub>2</sub> on reaction temperature. CO<sub>2</sub> 10% (○), CO<sub>2</sub> 25% (●), CO 1.0%, O<sub>2</sub> 0.5%, H<sub>2</sub> balance, W/F=0.03 g s cm<sup>-3</sup>.

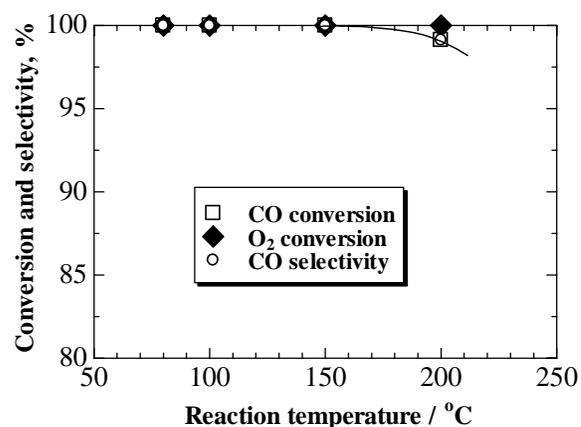


Fig. 3. Dependencies of CO conversion, O<sub>2</sub> conversion, selectivity on reaction temperature. The reaction conditions were same as those shown in Fig.1 except for containing CO<sub>2</sub> 25% and H<sub>2</sub>O 20%.