PM combustion of Ag-loaded perovskite-type oxides prepared from heteronuclear cyano-complex precursors

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Abstract: Two types of Ag-loaded perovskite-type oxides synthesized from a cyano complex precursor were examined for PM combustion activity. The positive effect of Ag loading on the catalytic activity for PM combustion was observed when Ag was loaded on a cyano complex precursor before calcination. Such a noticeable enhancement in activity was suggested to come from high dispersion of Ag loaded on perovskite-type oxide.

Keywords: Soot oxidation, Ag dispersion, Particulate matter.

1. Introduction

The removal of particulate matter (PM) has been attracted much attention as a new catalytic process for the environmental protect. Up to date, the silver-loaded ceria (Ag/CeO_2) has been reported to be the best catalyst for the removal of PM.[1] The perovskite type oxide has been also reported to be active for PM oxidation. Prasad et al. reported that $LaCoO_3$ partially substituted by Sr exhibited the high catalytic activity for PM oxidation.[2] Xiao et al. reported that $LaFeO_3$ with high surface area, which was prepared by using PMMA, is active for PM oxidation.[3] However, little is known about the catalytic activity of Ag-loaded perovskite-type oxide for PM oxidation. In the present study, the PM combustion was investigated for Ag-loaded perovskite-type oxides prepared by two methods.

2. Experimental

The heteronuclear cyano complex, $AB(CN)_6 \cdot nH_2O$ (A = La and Sm, B = Fe and Co), was prepared as a precursor of perovskite-type oxide.[4] Ag-loaded catalysts were prepared by the following two methods. (Method I) $AB(CN)_6 \cdot nH_2O$ was calcined at 873-973 K for 1 h in air to yield the corresponding perovskite-type oxide. Ag was loaded on perovskite-type oxide by impregnation with $AgNO_3$ solution. The catalyst was finally calcined at 873 K for 3 h. (Method II) $AB(CN)_6 \cdot nH_2O$ was stirred in $AgNO_3$ solution. In this case, Ag^+ ions were adsorbed on $AB(CN)_6 \cdot nH_2O$ surface probably due to electrostatic interaction between Ag^+ and CN^-. The filtrated samples were calcined at 873-973 K for 1 h in air. The amount of Ag loaded on perovskite-type oxide was determined by inductively coupled plasma (ICP).

Acetylene carbon black (ACB) was used as a carbon source. The catalyst was physically mixed with ACB in a mortar for 10 min (tight contact). The catalytic activity for carbon combustion was evaluated by a technique of the temperature-programmed reaction (TPR) as reported previously.[5] The value of T_{max} was defined as the temperature giving the maximum CO_2 concentration.

3. Results and discussion

Figure 1 shows TPR profiles of perovskite-type oxides prepared by Method I and Method II. TPR profiles of perovskite-type oxide without Ag loading are also shown. The TPR profiles of Ag loaded perovskite-type oxides prepared by Method I were almost similar to those of perovskite-type oxide without Ag loading. This suggests no or less positive effect of Ag addition on the catalytic activity for PM oxidation of perovskite-type oxide prepared by Method I. On the other hand, Method II was significantly effective for

enhancing the catalytic activity of PM oxidation. In particular, T_{max} of $Ag/LaFeO_3$ was more than 100 °C lower than that of $LaFeO_3$, being comparable to that of Ag/CeO_2. Similar effects were observed for $Ag/SmFeO_3$ and $Ag/SmCoO_3$.

FE-SEM measurements were conducted to elucidate Ag particles supported on $LaFeO_3$ (Fig. 2). The Ag particle sizes of $Ag/LaFeO_3$ prepared by Method I and II were 60-100 nm and *ca*. 10 nm, respectively. It was found that $Ag/LaFeO_3$ prepared by Method II provided higher dispersion state of Ag than $Ag/LaFeO_3$ prepared by Method I.

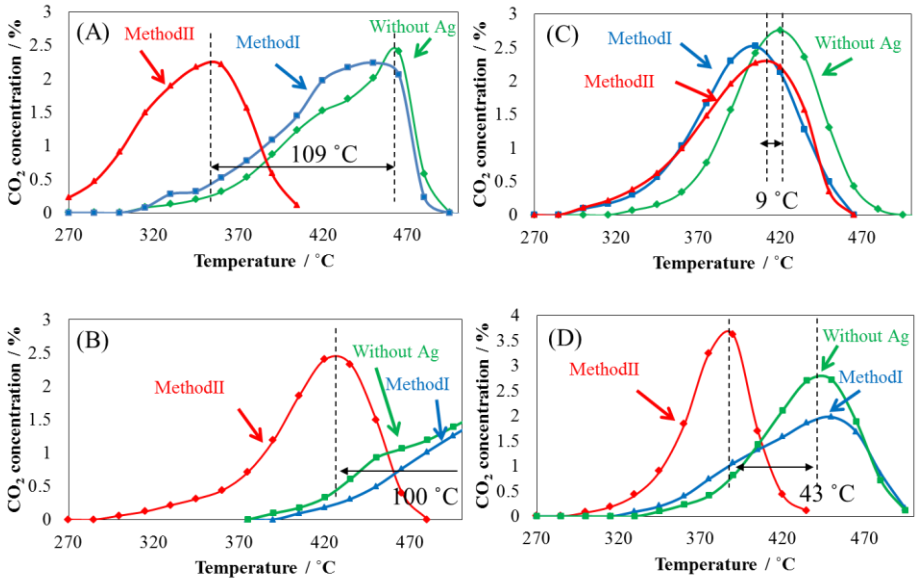

Figure 1. TPR profiles of Ag-loaded perovskite-type oxides prepared by Method I and Method II. The dotted line indicates TPR profile of perovskite-type oxide without Ag loading. (A) $Ag/LaFeO_3$, (B) $Ag/SmFeO_3$, (C) $Ag/LaCoO_3$, and (D) $Ag/SmCoO_3$.

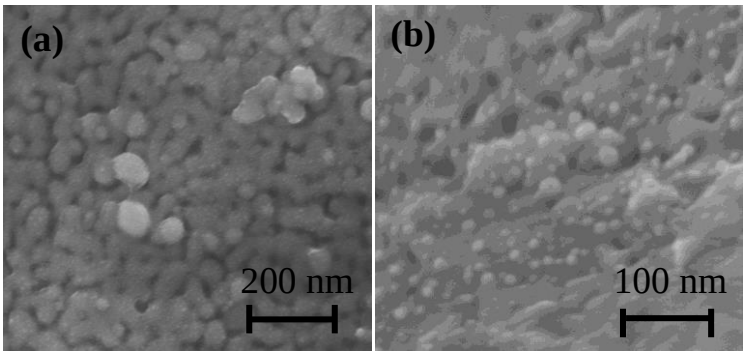

Figure 2. FE-SEM images of (a)$Ag/LaFeO_3$ prepared by Method I and (b)$Ag/LaFeO_3$ prepared by Method II.

4. Conclusions

The present study suggests that Ag-loaded perovskite-oxides prepared by Method II is one of the promising catalysts for PM combustion. $Ag/LaFeO_3$ prepared by Method II was the most active and its PM combustion activity was comparable to that of Ag/CeO_2. It was found that Ag particles (*ca*. 10 nm) of $Ag/LaFeO_3$ prepared by Method II were highly dispersed on the surface of perovskite-type oxide.

References

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