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Simultaneous removal of soot and NOx over Ir-based catalysts in the presence of oxygen

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ABSTRACT

The potentiality of Ir-based catalysts for the simultaneous removal of soot and NOx in the presence of O_2 is investigated by using temperature programmed reaction (TPR) technique. The behaviors of the catalysts for the soot oxidation and the NOx reduction are also separately analyzed. The results show that soot plays an important role on the NOx reduction, the formed NO_2 promotes the soot oxidation, and soot and NOx can be removed simultaneously over Ir-based catalysts in the presence of O_2. The simultaneous removal paths of soot and NOx over Ir catalyst are proposed that the NO_2 formation by the oxidation of NO oxidizes the soot and the CO formation by the soot oxidation reduces NO. Furthermore, the adsorbed-dissociation of NO_2 is a path of the NOx reduction. Comparing with Pt catalyst, Ir catalyst is a more effective catalyst for the simultaneous removal of soot and NOx. The Ir contents and support materials have some effects on the catalytic activity and 1 wt.% Ir/ZSM-5 exhibits a high level activity for the simultaneous removal of soot and NOx.

1. Introduction

Nitrogen oxides (NOx) and soot emission from diesel exhaust have been causing severely environmental and healthy problems. NOx are major air pollutants to produce acid rain and photochemical smog. And soot, which contains hydrocarbons, can penetrate into lung tissue and leads to serious pulmonary edema and cancer. Since the reduction of both NOx and soot emission to the admitted level cannot be accomplished by engine modifications alone, after-treatment processes for the simultaneous reduction of their emission from diesel exhaust have attracted much attention in recent years. As a promising process, Yoshida et al. [1] proposed the catalytic process to remove soot and NOx simultaneously in the presence of O_2. Later, Teraoka et al. [2–4] studied the simultaneous removal of soot and NOx in oxygen-containing model exhaust gas and reported that perovskites and spinels were effective catalysts. However, the activity of these catalysts is low in the presence of water vapor and sulfur dioxide [5], and therefore it is difficult to apply these results to the case under the automotive exhaust gas condition.

During the past twenty years, many catalytic systems for single reduction of soot or NOx have been researched, and many efforts have been done to design catalysts in the presence of O_2, H_2O and SO_2 [6–13]. With respect to the reduction of soot emission, the noble metal of Pt catalyst [6–7] exhibits a high level of catalytic activity in the presence of O_2, H_2O and SO_2. With respect to the reduction of NOx emission, the noble metal catalysts [8–12] also seem to be more promising than non-noble metal catalysts [13]. In recent years, Ir is regarded as an effective catalyst for the NO reduction with CO. Ogura et al. [8] investigated the activity of supported iridium catalysts and found that NO can be successfully reduced to N_2 with CO over Ir/silicalite catalyst and that the catalytic activity is not influenced by coexisting SO_2. Wang et al. [9–10] reported that among Pt, Pd, Rh and Ir catalysts, Ir/ZSM-5 catalyst exhibits the highest activity for NO reduction by CO in the presence of excess O_2 and H_2O. Haneda et al. [11] discovered that Ir/SiO_2 shows marked catalytic activity for NO reduction with CO in the presence of O_2, H_2O and SO_2. The most significant feature of Ir catalyst is that the presence of SO_2, which normally poisons catalytic reactions, actually promotes NO reduction with CO in the presence of O_2. Because Ir catalyst has unique property for NOx reduction with CO in the presence of O_2, H_2O and SO_2, and soot is an important source to produce CO, it is necessary to investigate the simultaneous removal of soot and NOx from diesel exhaust over Ir catalyst. However, the noble metal of Ir catalyst has received little attention in the literatures on simultaneous removal of soot and NOx in the presence of O_2.

In this work, we investigate the simultaneous removal of soot and NOx over Ir-based catalysts in the presence of O_2, and discuss

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the effects of Ir contents and support materials on the catalytic activity.

2. Experimental

2.1. Catalyst preparation and characterization

Ir-based catalysts were prepared by iso-volumetric impregnation with aqueous solutions of $H_2IrCl_6{\cdot}6H_2O$ (Platinum Group Metals China) and the Ir content was 0.1 wt.%, 1 wt.% and 5 wt.%, respectively. The different content of Ir catalyst was prepared by given concentration solution. The support materials used were ZSM-5 (Nankai University, Si/Al = 50, 5–6 μm, 320 m^2/g), γ-Al_2O_3 (Dalian Luming Nanometer Material Co., Ltd, 200–500 nm, 160 m^2/g) and SiO_2 (Qingdao Haiyang Chemical Co., Ltd, 0.125–0.425 mm, 460 m^2/g). For comparison, Pt/γ-Al_2O_3 was also prepared by iso-volumetric impregnation of γ-Al_2O_3 with an aqueous solution of $H_2PtCl_6{\cdot}6H_2O$ (Platinum Group Metals China) and the Pt content was 1 wt.%. The impregnated catalyst precursors were dried at 110 °C for 10 h and finally calcined at 500 °C for 3 h in air. The powder catalysts were pressed at 10 MPa into pellets and crushed to granules of 0.250–0.425 mm for measuring the catalytic reaction.

The structure of supported Ir particles was analyzed by TEM (JEOL, JEM-2010) at an acceleration voltage of 200 kV.

2.2. Catalytic activity measurements

Catalytic activity was measured with temperature programmed reaction (TPR) technique. Printex-U (Degussa AG; 25 nm, 100 m^2/g) was used as model soot. The catalyst granules (0.05 g) or catalyst/soot (10/1, w/w, 0.055 g) mixture held in a tubular quartz reactor (6 mm i.d.). The mixture was carefully mixed with spatula and kept "loose" contact between catalyst and soot, which was close to that found in practical cases. The reaction gas, containing NO (420 ppm) and O_2 (4.4 vol.%) diluted in He as the balance gas, was fed through the catalyst bed at a rate of 100 ml/min (SV = 35,000 h^{-1}). In some experiments, the components of reaction gas were changed but the concentration of each gas and the total flow rate were unchanged. The reaction temperature was monitored with a thermocouple directly inserted into the region of the catalyst bed and controlled by a PID-regulation system (Bchy, CKW-2200). The reactor temperature was raised at a rate of 4 °C/min from 100 °C to 750 °C. The concentrations of CO and CO_2 emitted from soot oxidation were measured with an on-line gas chromatograph (Shanghai Precision & Scientific Instrument Co., Ltd, GC-112A), equipped with a flame ionization detector (FID). The CO and CO_2 concentrations were determined after separating over a Porapak-Q column and converting to methane over a Ni catalyst at 360 °C. The concentrations of NO, NO_2 and NOx (=NO + NO_2) were continuously detected by Nitrogen Oxides Analyzer (Ecotech, EC-9841B).

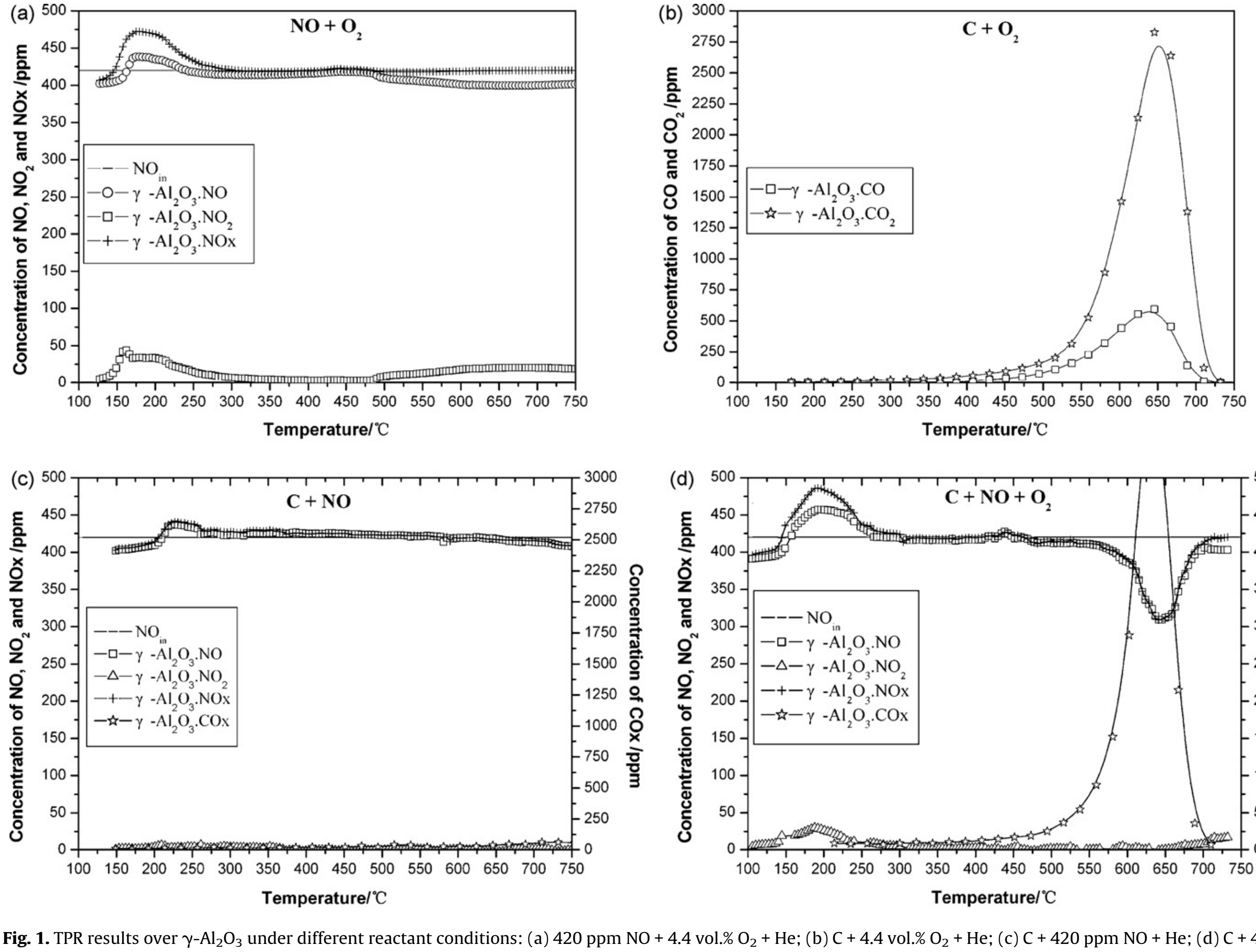

Fig. 1. TPR results over γ-Al_2O_3 under different reactant conditions: (a) 420 ppm NO + 4.4 vol.% O_2 + He; (b) C + 4.4 vol.% O_2 + He; (c) C + 420 ppm NO + He; (d) C + 420 ppm NO + 4.4 vol.% O_2 + He. Catalyst weight = 0.05 g, soot weight = 0.005 g, flow rate: 100 ml/min.

The catalytic activity was evaluated by temperature of soot oxidation and total amount of NOx reduction throughout a TPR run. The temperature of soot oxidation was estimated by the value of T_{10}, T_{50} and T_{90}, which were defined as the temperatures by which 10%, 50% and 90% of soot, respectively, were oxidized during the TPR procedure. The total amount of NOx reduction throughout the TPR run ($\upsilon_{\mathrm{deNOx}}$) was obtained by integrating NOx reduction versus temperature (time) curve. The selectivity to CO (S_{CO}) was also calculated by the ratio of CO to the sum of CO and CO_2 formed during the TPR.

3. Results and discussion

3.1. Simultaneous removal paths of soot and NOx

Fig. 1(a) shows TPR results of NO, NO_2 and NOx over bare γ-Al_2O_3 under 420 ppm NO + 4.4 vol.% O_2 + He. NO_2 starts to appear at 490 °C but the concentrations are very low even at high temperature. In the same temperature region as that of NO_2 increase, the concentrations of NO decrease and the concentrations of NOx are invariant. These results indicate that conversion only between NO and NO_2 occurs for the nitrogen oxide species (Eq. (1)). The low conversions of NO into NO_2 are limited by the thermodynamic equilibrium among NO, O_2 and NO_2. For the concentrations of NO, NO_2 and NOx increasing in the temperature range 100–250 °C, as discussed elsewhere [14], they are associated to their adsorption at lower temperature.

$$NO + O_2 \rightarrow NO_2 \quad (1)$$

Fig. 1(b) shows TPR results of CO and CO_2 over γ-Al_2O_3–soot mixture under 4.4 vol.% O_2 + He. CO and CO_2 start to appear around 500 °C (T_{10} = 543 °C). This is a typically non-catalyzed oxidation of carbon using O_2 (Eq. (2)).

$$C + O_2 \rightarrow CO_2\ (\text{or CO}) \quad (2)$$

Fig. 1(c) shows TPR results of NO, NO_2, NOx and COx (COx = CO + CO_2) over γ-Al_2O_3–soot mixture under 420 ppm NO + He. COx are not produced and NOx are not reduced over the whole temperature range examined, suggesting that the reaction of "NO + C" cannot occur when the temperature is lower than 750 °C in the absence of O_2.

Fig. 1(d) shows TPR results of NO, NO_2, NOx and COx over γ-Al_2O_3–soot mixture under 420 ppm NO + 4.4 vol.% O_2 + He. COx start to appear around 490 °C (T_{10} = 534 °C), almost at the same temperature as in Fig. 1(b), indicating that the soot oxidation mainly results from the oxidation by O_2 (Eq. (2)) and is little affected by the existence of NO. Contrasted with Fig. 1(a), it is obvious that the NOx reduction occurs in Fig. 1(d) and appears in the same temperature region of the soot oxidation, suggesting the NOx reduction is related to the soot oxidation. From the above discussion, the reaction of "NO + C" cannot occur in the temperature region of the NOx

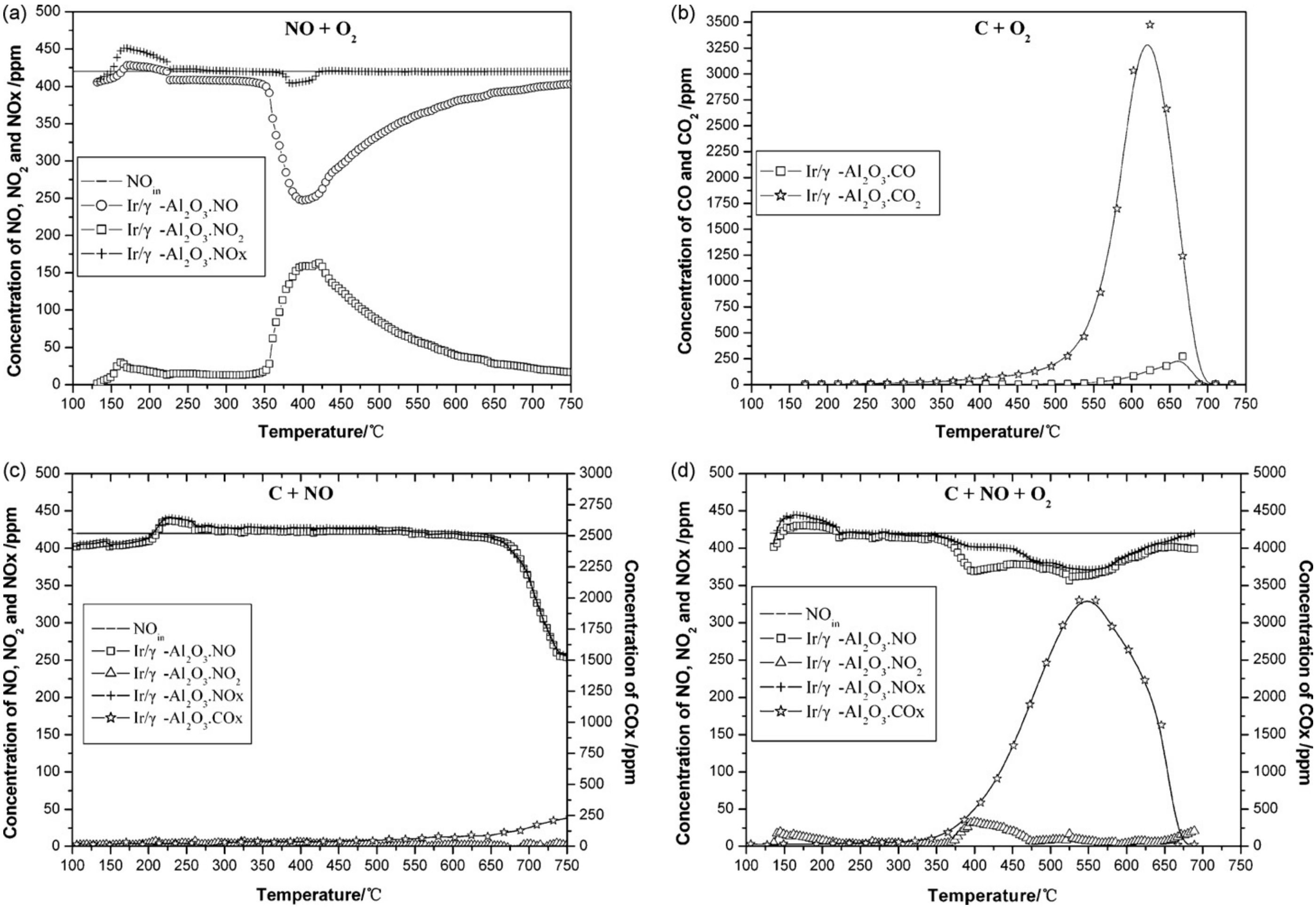

Fig. 2. TPR results over Ir/γ-Al_2O_3 under different reactant conditions: (a) 420 ppm NO + 4.4 vol.% O_2 + He; (b) C + 4.4 vol.% O_2 + He; (c) C + 420 ppm NO + He; (d) C + 420 ppm NO + 4.4 vol.% O_2 + He. Catalyst weight = 0.05 g, soot weight = 0.005 g, flow rate: 100 ml/min.

reduction. So, the NOx reduction is mainly attributed to the reaction of NO with carbon–oxygen (CO) (Eq. (3)) by the soot oxidation, which is similar to the result reported by Yamashita and Tomita [15]. The increase of NOx concentration at higher temperature than 650 °C is due to the exhaustion of the soot.

$$NO + CO \rightarrow CO_2 + N_2\,(\text{or}\,N_2O) \qquad (3)$$

Fig. 2(a) shows TPR results of an experiment similar to Fig. 1(a) but over 1 wt.% Ir/γ-Al_2O_3. NO_2 starts to appear at 352 °C, reaches a maximum at 390 °C and then decreases with temperature increase. Simultaneously, the concentrations of NOx decrease around 350 °C, reach a maximum reduction at 390 °C. Contrasted with Fig. 1(a), it is obvious that noble metal of Ir promotes not only the oxidation of NO (Eq. (1)) at lower temperature but also the reduction of NOx. For the NOx reduction over Ir catalyst, Wang et al. [10] had investigated the TPD of NO after co-adsorption with O_2 over 0.1% Ir/ZSM-5 and found that NO can dissociate to N_2 and N_2O around 395 °C. Although our instruments cannot detect the signals of N_2 and N_2O, a TPR experiment over 0.1% Ir/ZSM-5 has been carried under 420 ppm NO + 4.4 vol.% O_2 + He (results shown in Fig. 3). Obviously, the temperature at the NOx reduction is same as that at the N_2 and N_2O appearance reported by Wang et al. [10], suggesting that the NOx reduction mainly forms N_2 and N_2O. Furthermore, the NOx reduction correlates to the conversion of NO to NO_2 in Fig. 2(a) and one possible explanation of this phenomenon is that NO_2 is adsorbed dissociatively on the catalyst surface to form adsorbed species (Eq. (4)) [16,17]. In order to explain these findings, the following paths are proposed (Eq. (4) and (5)) [10,16,17].

$$NO_2\,(g) \rightarrow NO\,(ad) + O\,(ad) \qquad (4)$$

$$NO\,(ad) \rightarrow N_2\,(\text{or}\,N_2O) + O_2 \qquad (5)$$

Fig. 2(b) shows TPR results of an experiment similar to Fig. 1(b) but over 1 wt.% Ir/γ-Al_2O_3. CO_2 starts to appear around 500 °C (T_{10} = 545 °C), at the same temperature as in Fig. 1(b). This result indicates that Ir has little effect on ignition temperature of soot oxidation. Contrasted with Fig. 1(b), it is obvious that the concentrations of CO decrease and the concentrations of CO_2 increase, suggesting that Ir promotes CO to oxidize into CO_2 (Eq. (6)).

$$CO + O_2 \rightarrow CO_2 \qquad (6)$$

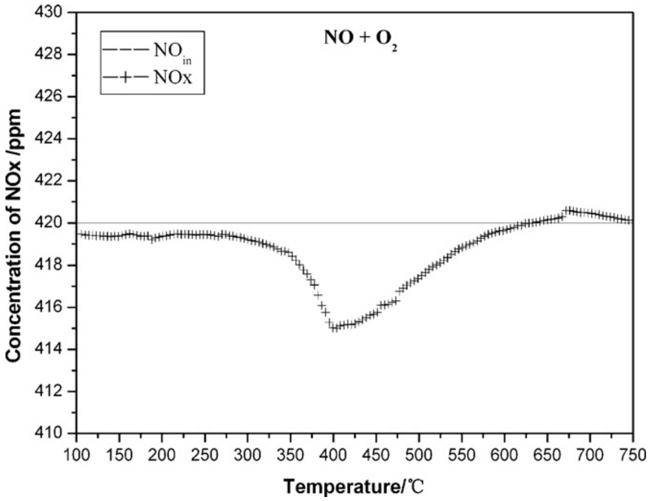

Fig. 3. TPR result of NOx over 0.1% Ir/ZSM-5. Catalyst weight = 0.05 g, reactant conditions: 420 ppm NO + 4.4 vol.% O_2 + He, flow rate: 100 ml/min.

Fig. 2(c) shows TPR results of an experiment similar to Fig. 1(c) but over 1 wt.% Ir/γ-Al_2O_3. COx appears around 650 °C. Simultaneously, the NOx reduction occurs. Contrasted with Fig. 1(c), it is obvious that Ir promotes the reaction of “NO + C”. However, the reaction of “NO + C” cannot takes place when the temperature is lower than 650 °C in the absence of O_2. The similar phenomenon was reported in previous investigation [3].

Fig. 2(d) shows TPR results of an experiment similar to Fig. 2(a) but using 1 wt.% Ir/γ-Al_2O_3 mixed with soot. Contrasted with Fig. 2(a), NO_2 starts to appear at almost the same temperature, but the concentrations of NO_2 decrease and the concentrations of NO increase between 350 °C and 650 °C distinctly. Simultaneously, at the same temperature region, COx appears (T_{10} = 442 °C). And contrasted with Fig. 2(b), it is obvious that T_{10} is much lower in Fig. 2(d). These results indicate that soot is oxidized by NO_2 to CO and CO_2, and then NO_2 goes back to NO (Eq. (7)). Furthermore, in Fig. 2(d), the NOx reduction appears not only at the same temperature region as in Fig. 2(a) but also at the same temperature region of the soot oxidation. The former correlates to the adsorbed-dissociation of NO_2 (Eq. (4) and (5)) and the latter is mainly related to the soot oxidation. Considering that the soot oxidation by O_2 normally starts at higher temperature than 500 °C and by NO normally starts at higher temperature than 650 °C, the NOx reduction is mainly attributed to a direct reaction of NO_2 with soot (Eq. (8)) when the temperature is lower than 500 °C and also includes Eq. (3) when the temperature is higher than 500 °C. The increase of NOx concentrations at higher temperature than 550 °C is also due to the exhaustion of soot.

$$NO_2 + C \rightarrow CO_2\,(\text{or}\,CO) + NO \qquad (7)$$

$$NO_2 + C \rightarrow CO_2 + N_2\,(\text{or}\,N_2O) \qquad (8)$$

From the above analysis, we can conclude that soot and NOx can be removed simultaneously in the presence of O_2. The soot plays an important role on the NOx reduction (Eqs. (3) and (8)) and the formed NO_2 promotes the soot oxidation (Eqs. (7) and (8)). In view of the interim process about Eq. (8), an equation can be written as Eq. (9) [17]. So, the simultaneous removal paths over Ir catalyst are proposed: the NO_2 formation by the oxidation of NO oxidizes the soot, and the CO formation by the soot oxidation reduces NO.

$$NO_2 + C \rightarrow CO + NO \rightarrow CO_2 + N_2\,(\text{or}\,N_2O) \qquad (9)$$

3.2. Activity of Ir-based catalysts

Table 1 summarizes the results of T_{10}, T_{50}, T_{90}, S_{CO} and υ_{deNOx} over three types of catalysts with different Ir contents. Because the

Table 1
Activity of catalysts for simultaneous removal of soot and NOx

Catalyst	T_{10} (°C)	T_{50} (°C)	T_{90} (°C)	S_{CO} (%)	υ_{deNOx} ($\times10^{-5}$ mol)
1%Pt/γ-Al_2O_3	435	524	580	2	3.09
γ-Al_2O_3	534	625	662	39	3.07
0.1% Ir/γ-Al_2O_3	495	578	624	8	3.33
1% Ir/γ-Al_2O_3	443	543	624	6	3.45
5% Ir/γ-Al_2O_3	397	495	556	3	2.86
ZSM-5	490	614	666	41	2.38
0.1%Ir/ZSM-5	409	506	598	6	2.88
1%Ir/ZSM-5	390	490	554	2	3.37
5%Ir/ZSM-5	374	480	546	2	2.94
SiO_2	533	627	669	47	1.50
0.1%Ir/SiO_2	500	602	646	16	1.87
1%Ir/SiO_2	464	595	648	8	1.89
5%Ir/SiO_2	405	557	633	6	3.24

Catalyst weight = 0.05 g, soot weight = 0.005 g, reactant conditions: C + 420 ppm NO + 4.4 vol.% O_2 + He, flow rate: 100 ml/min.

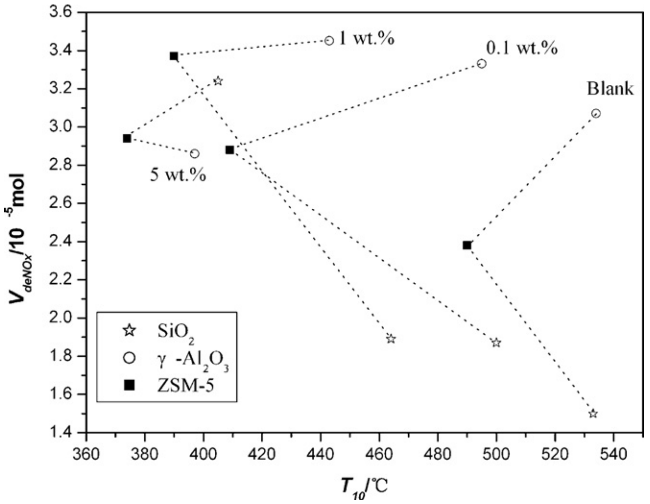

Fig. 4. Relation between T_{10} and $\upsilon_{\mathrm{deNOx}}$ over various Ir catalysts. Catalyst weight = 0.05 g, soot weight = 0.005 g, reactant conditions: C + 420 ppm NO + 4.4 vol.% O_2 + He, flow rate: 100 ml/min.

removal of soot and NOx at lower temperature is more important than the whole TPR profile in practical application, we especially focus on the results of T_{10} and $\upsilon_{\mathrm{deNOx}}$. Naturally a catalyst with high activity should be with low T_{10} and large $\upsilon_{\mathrm{deNOx}}$. Fig. 4 shows the relation between T_{10} and $\upsilon_{\mathrm{deNOx}}$ over various Ir catalysts. It is seen that the Ir contents and support materials have some effects on the catalytic activity and 1 wt.% Ir/ZSM-5 exhibits a high level activity for the simultaneous removal of soot and NOx.

3.2.1. *Effect of Ir contents*

Over all the Ir/SiO_2 catalysts, with an increase of Ir content, T_{10}, T_{50}, T_{90} and S_{CO} decrease and $\upsilon_{\mathrm{deNOx}}$ increases. The Ir/γ-Al_2O_3 catalysts also show that T_{10}, T_{50}, T_{90} and S_{CO} decrease with the increase of Ir content, whereas $\upsilon_{\mathrm{deNOx}}$ firstly increases and then decreases. The catalytic activity order over Ir/ZSM-5 shows the same trend as that over Ir/γ-Al_2O_3. These results indicate that a higher content of Ir metal is more beneficial for the oxidation of soot, while an optimal content of Ir metal exists for the reduction of NOx.

From Section 3.1, the catalytic activity for the soot oxidation is attributed to NO_2 formation by the oxidation of NO. In our previous work [12], we examined the effect of the Ir contents on the conversion of NO to NO_2 and found the activity order was 0.1 wt.% < 1 wt.% < 5 wt.%. This result is similar to our result of the soot oxidation. This similarity suggests that, for an Ir catalyst, the oxidation activity for soot is closely related to oxidation activity for NO.

From Section 3.1, the catalytic activity for the NOx reduction is mainly attributed to the reaction between CO and NO. Although S_{CO} decreases with the increase of Ir contents, a temperature widow of the soot oxidation widens (not shown, but is clear by comparing Fig. 2(c) with Fig. 1(c)). The wide temperature widow of the soot oxidation improves the use of CO to reduce NO. Hence, $\upsilon_{\mathrm{deNOx}}$ increases with the increase of Ir contents. However, $\upsilon_{\mathrm{deNOx}}$ decreases over Ir/γ-Al_2O_3 and Ir/ZSM-5 when the Ir content is higher than 1 wt.%, which is due to the fact that 5 wt.% Ir catalysts have high oxidizability for the reaction of Eq. (6) and the concentration of CO is too low. Considering this reason, we do not discuss 5 wt.% Ir catalysts in the rest of this paper.

3.2.2. *Effect of support materials*

Over all bare support materials, the catalytic activity order is ZSM-5 > γ-Al_2O_3 > SiO_2 for the soot oxidation and γ-Al_2O_3 > ZSM-5 > SiO_2 for the NOx reduction. When the Ir content is 0.1 wt.% and 1 wt.%, respectively, the catalytic activity order is same as that over bare support materials. These results indicate ZSM-5 is most beneficial for the soot oxidation, while γ-Al_2O_3 is most beneficial for the NOx reduction.

For the soot oxidation, the catalytic activity is attributed to NO_2. In our previous work [12], we also examined the effect of the support materials on the conversion of NO to NO_2 and found the activity order was ZSM-5 > γ-Al_2O_3 > SiO_2. This order is similar to our result of the soot oxidation. This result indicates that, for a support material, the oxidation activity for soot is also closely related to the oxidation activity for NO.

For the NOx reduction, the catalytic activity is mainly attributed to the reaction between CO and NO. Haneda et al. [11] investigated the effect of support material on the NO reduction with CO and found the activity order was Ir/γ-Al_2O_3 > Ir/SiO_2 in the absence of SO_2, which is similar to our result. In their experiment, they discovered that the Ir particles on γ-Al_2O_3 consist of agglomerates of small particles with a size of 2–3 nm and the Ir particles are dispersed with a size of 10–20 nm over the surface of SiO_2, suggesting that the Ir particles with smaller particle size were more active for the NOx reduction with CO [18]. In our experiment, TEM observation was also performed for 1 wt.% Ir/γ-Al_2O_3, 1 wt.% Ir/ZSM-5 and 1 wt.% Ir/SiO_2 to examine the structure of supported Ir particles (shown in Fig. 5). The Ir particles on γ-Al_2O_3 clearly consist of agglomerates, which is similar to the result reported by Haneda et al. [11]. Accordingly, although we did not use high magnification to observe the agglomerates, they are likely to consist of small particles with a size of 2–3 nm. Over the surface of ZSM-5 support, the Ir particles are well dispersed with a size of 1–3 nm. In contrast, the structure of Ir particles deposited on SiO_2 seems to be different from the structure of those on γ-Al_2O_3 and ZSM-5, and are like to some “marking”. The size of “marking” is

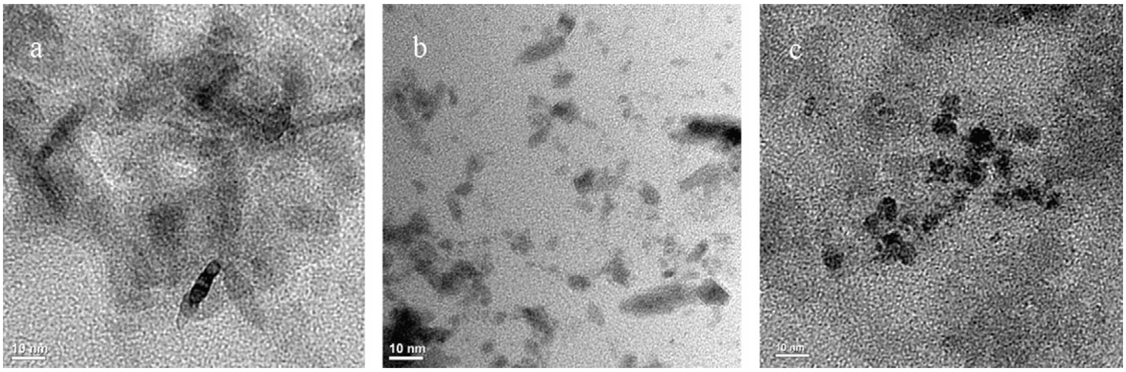

Fig. 5. TEM images of (a) 1 wt.% Ir/γ-Al_2O_3, (b) 1 wt.% Ir/ZSM-5 and (c) 1 wt.% Ir/SiO_2.

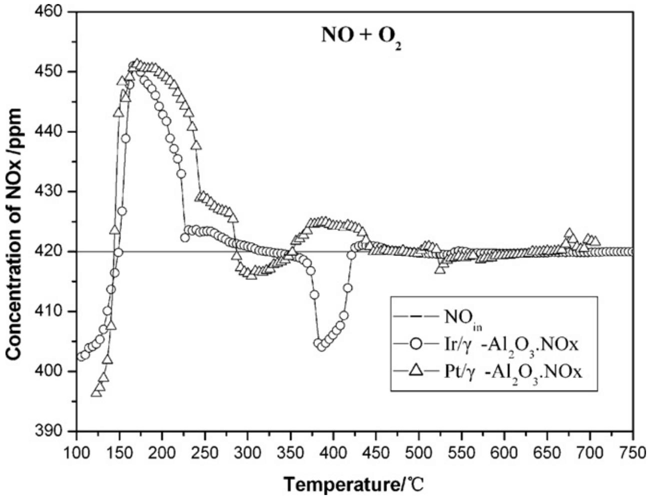

Fig. 6. TPR results of NOx over Ir/γ-Al_2O_3 and Pt/γ-Al_2O_3. Catalyst weight = 0.05 g, reactant conditions: 420 ppm NO + 4.4 vol.% O_2 + He, flow rate: 100 ml/min.

larger than 5 nm. These results indicate that the activities are sensitive to the Ir particle size. Furthermore, it is noteworthy that the catalytic activity of Ir/ZSM-5 for the NOx reduction is lower than that of Ir/γ-Al_2O_3, which maybe correlate to the high oxidizability of Ir/ZSM-5.

3.3. Comparison between Ir/γ-Al_2O_3 and Pt/γ-Al_2O_3

Table 1 also lists the catalytic activities of 1% Pt/γ-Al_2O_3 during TPRs over catalyst with soot under 420 ppm NO + 4.4 vol.% O_2 + He. On 1 wt.% of Pt/γ-Al_2O_3, T_{10} is 435 °C, which is similar to the result reported by Castoldi et al. [19]. Comparing the catalytic activities of Ir/γ-Al_2O_3 and Pt/γ-Al_2O_3, it is clear that T_{10} over Ir/γ-Al_2O_3 is very close to that over Pt/γ-Al_2O_3 and $\upsilon_{\mathrm{deNOx}}$ over Ir/γ-Al_2O_3 is higher than that over Pt/γ-Al_2O_3, whereas $\upsilon_{\mathrm{deNOx}}$ over Pt/γ-Al_2O_3 is same as that over bare γ-Al_2O_3. These results indicate that Ir/γ-Al_2O_3 catalyst is more beneficial for the simultaneous removal of soot and NOx than Pt/γ-Al_2O_3 catalyst.

From Section 3.1, the removal paths of NOx mainly include the reduction with CO (Eqs. (3) and (9)) and adsorbed-dissociation of NO_2 (Eqs. (4) and (5)). Wang et al. [9–11] reported that Ir exhibits a higher activity than Pt for NO reduction with CO in the presence of excess oxygen. This result is similar to our result, suggesting that Ir/γ-Al_2O_3 promotes NOx reduction with CO by the soot oxidation. Furthermore, Fig. 6 shows the results of NOx reduction over 1 wt.% Ir/γ-Al_2O_3 and 1 wt.% Pt/γ-Al_2O_3 under 420 ppm NO + 4.4 vol.% O_2 + He. Over Ir/γ-Al_2O_3, the NOx reduction correlates to the adsorbed-dissociation of NO_2 (Eqs. (4) and (5), discussed in Section 3.1). Whereas over Pt/γ-Al_2O_3, NOx is adsorbed between 270 °C and 350 °C and then desorbed between 350 °C and 440 °C, and the NOx reduction almost does not take place. This result indicates NOx is more easily dissociated over Ir/γ-Al_2O_3 than that over Pt/γ-Al_2O_3. So, Ir/γ-Al_2O_3 is a more effective catalyst for the simultaneous removal of soot and NOx.

4. Conclusions

The potentiality of Ir-based catalysts has been investigated for the simultaneous removal of soot and NOx in the presence of O_2. Soot and NOx can be removed simultaneously over Ir-based catalysts in the presence of O_2. The simultaneous removal paths are proposed that the NO_2 formation by the oxidation of NO oxidizes the soot and the CO formation by the soot oxidation reduces NO. Furthermore, the adsorbed-dissociation of NO_2 also promotes the NOx reduction. The comparison between Ir/γ-Al_2O_3 and Pt/γ-Al_2O_3 reveals that Ir catalyst is a more effective catalyst for the simultaneous removal of soot and NOx. The Ir contents and support materials have some effects on the catalytic activity and 1 wt.% Ir/ZSM-5 exhibits a high level activity for the simultaneous removal of soot and NOx.

Acknowledgments

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