Soot Oxidation with Ir-based Catalysts in the Presence of O_2 and NO

ZHU Rong-shu, GUO Ming-xin, CI Xin-bo, OUYANG Feng
(Environmental Science and Engineering Research Center, Shenzhen Graduate School, Harbin Institute of Technology, Shenzhen 518055, China)

Abstract: The soot oxidation with Ir-based catalysts was investigated using simulated diesel exhaust gases. By comparing with Pt/SiO_2, Ir/SiO_2 showed similarly catalytic activity and high CO selectivity. The effect of Ir content on the catalytic activity and CO selectivity was examined. With the increasing of Ir content in the range examined (from zero to 5%), the catalytic activity increased and the CO selectivity decreased. Furthermore, the effect of support oxides was obtained: the catalytic activity was Ir/ZSM-5 > Ir/SiO_2 > Ir/γ-Al_2O_3 and the CO selectivity was Ir/ZSM-5 < Ir/γ-Al_2O_3 < Ir/SiO_2.

Key words: soot oxidation; Ir-based catalysts; NO
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1 Introduction

Soot emitted from diesel engines causes a serious problem to human health and environment. The use of a catalyst is the most preferential way to promote soot oxidation at exhaust gas temperature. During the past decade, many catalytic systems have been researched[1-3]. Among them, the Pt-based catalysts exhibit a high level of catalytic activity. However, this system that obviously does not remove NO is blamed for the increase in the NO_2 concentration in the exhausts.

In our research, we are interested in Ir-based catalysts for soot oxidation, since they show high catalytic activity for NO reduction with CO in the presence of excess oxygen[4-5]. It is well known that soot oxidation is an important source to produce CO. If we can control soot to oxidize to CO and then use this CO to reduce NO to N_2, we will realize to simultaneously remove the soot and NO. Thus, the aim of this work is to investigate the catalytic activity and CO selectivity of Ir-based catalysts for soot oxidation using simulated exhaust gases, including O_2 and NO.

2 Experimental

The supported catalysts were prepared by incipient wetness impregnation method with the solution of $H_2IrCl_6 \cdot 6H_2O$ and $H_2PtCl_6 \cdot 6H_2O$. Catalytic activity for soot oxidation was measured with temperature programmed reaction (TPR) system. The catalytic activity was evaluated at the initial temperature (T_i: the temperature when the CO+CO_2 concentration reached 100 ppm). The CO selectivity (S_{CO}: the ratio of CO to the sum of CO and CO_2 formed during the reaction) was also calculated.

Tab. 1 T_i and S_{CO} for catalysts

Catalyst	T_i (℃)	S_{CO} (%)
1% Pt/SiO_2	330	0
SiO_2	480	49
0.02% Ir/SiO_2	461	9
0.1% Ir/SiO_2	421	7
1% Ir/SiO_2	329	6
5% Ir/SiO_2	315	3
ZSM-5	471	42
0.02% Ir/ZSM-5	402	5
0.1% Ir/ZSM-5	377	4
1% Ir/ZSM-5	316	4
5% Ir/ZSM-5	302	1
γ-Al_2O_3	480	37
0.02% Ir/γ-Al_2O_3	462	9
0.1% Ir/γ-Al_2O_3	445	5
1% Ir/γ-Al_2O_3	340	4
5% Ir/γ-Al_2O_3	321	2

Catalyst weight = 0.05 g; soot weight = 0.005 g; Feed gas composition = 420 ppm NO + 4.4% O_2 in He; flow rate = 100 ml/min.

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作者简介: 朱荣淑 (1977年生), 女, 博士后; 通讯联系人: 欧阳峰; E-mail: ouyangfk@hit.edu.cn

3 Results and discussion

Table 1 showed the T_i and S_{CO} of a series of catalysts for soot oxidation. Compared the T_i and S_{CO} of 1% of Ir/SiO_2 with that of 1% of Pt/SiO_2, we could see that the T_i of Ir/SiO_2 was near to that of Pt/SiO_2 but the S_{CO} of Ir/SiO_2 was higher than that of Pt/SiO_2, which was in agreement with that reported by Wang et al[4].

From the data on Ir/SiO_2 that contained different Ir metal contents, we could see that from blank SiO_2 to 5% of Ir/SiO_2, the corresponding T_i was lowered and S_{CO} was decreased. On the samples supported with γ-Al_2O_3 or ZSM-5, the trend was agreement with that of Ir/SiO_2. This phenomena was similar to that reported by Xue et al[6]. In our work, although the correlation had not been established between the Ir content and the particle size, it seemed that the oxidation of soot and the selectivity to CO depended on the size of particles.

By comparing the T_i and S_{CO} in the same Ir content, the effect of support oxides was obtained. The data in the table1, show that the order of T_i was ZSM-5 < SiO_2 < γ-Al_2O_3 and the order of S_{CO} was ZSM-5 < γ-Al_2O_3 < SiO_2. Xue et al[6] examined the effect of the support oxides on the oxidation of NO to NO_2 and found the catalytic activity order was Pt/SiO_2 > Pt/γ-Al_2O_3. This result was similar to our results in the T_i of soot oxidation. The order of S_{CO} differed from the order of T_i was attributed to the interaction between γ-Al_2O_3 and NO_2 and the adsorption of NO_2 promoted the selectivity to CO_2.

4 Conclusion

The soot oxidation with Ir-based catalysts was investigated using simulated diesel exhaust gases. Ir-based catalysts showed high catalytic activity for oxidation of soot and high CO selectivity in the presence of O_2 and NO. With the increasing of Ir content in the range examined (from zero to 5%), the catalytic activity increased and the CO selectivity decreased. Furthermore, support oxides had a little effect on the soot oxidation; the order of catalytic activity was $Ir/ZSM\text{-}5 > Ir/SiO_2 > Ir/\gamma\text{-}Al_2O_3$ and the order of selectivity to CO was $Ir/ZSM\text{-}5 < Ir/\gamma\text{-}Al_2O_3 < Ir/SiO_2$.

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在 O_2和 NO存在下 Ir催化氧化黑烟的研究

朱荣淑, 郭明新, 次新波, 欧阳峰

(哈尔滨工业大学深圳研究生院环境科学与工程研究中心, 广东 深圳 518055)

摘 要: 模拟柴油机尾气中 O_2和 NO的氛围, 研究了以 Ir为基础的催化剂对黑烟的催化氧化。与 Pt/SiO_2比较, Ir/SiO_2对黑烟氧化有其相似的催化活性, 但有更高的 CO选择性。通过测定 Ir含量对催化活性和 CO选择性的影响, 发现在研究的范围内 (0到 5%), 随着 Ir含量的增加, 催化活性增加, CO选择性降低。此外, 测定了氧化物载体对催化活性及 CO选择性的影响, 得到了催化活性顺序为 $Ir/ZSM\text{-}5 > Ir/SiO_2 > Ir/\gamma\text{-}Al_2O_3$, CO选择性顺序为 $Ir/ZSM\text{-}5 < Ir/\gamma\text{-}Al_2O_3 < Ir/SiO_2$。

关键词: 黑烟氧化; Ir催化剂; NO

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