Investigating the effect of reduction conditions on diesel oxidation reaction over Pt-based NSR catalyst

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Abstract: The effect of reduction conditions on the catalytic diesel oxidation reaction has been investigated. It is found that the CO oxidation is substantially improved after the reduction process at various time (1, 5, and 10 s) and temperature (150, 300, 450, and 600 °C). The shift of light-off temperature for CO conversion to low temperature after CO reduction at 600 °C for 10 s is achieved by 80 °C. This is a crucial factor for emission control for light-duty passenger cars in order to maintain and achieve the catalytic activity of CO oxidation at low temperature region.

Keywords: Pt-based NSR catalyst, DOC, CO oxidation

1. Introduction

Diesel and lean-burn engines provide better fuel economy and produce lower carbon dioxide (CO₂) emissions in comparison with gasoline engines. The removal of HC/CO/NOx requires the combination of after-treatment technologies. For many years, catalytic converters have been used to control the exhaust emissions from diesel vehicles. These have involved a variety of approaches including diesel oxidation catalysts (DOC), diesel particulate filters (DPF), lean NOx trap (LNT) or NOx storage reduction (NSR) catalysts and selective catalytic reduction (SCR) [1,2].

Currently, SCR technology has been implemented to reduce NOx using urea or ammonia, which is effective for heavy-duty vehicles. The control of NOx emission at low temperatures for small diesel vehicles is challenging because the catalytic activity is not sufficient for future low fuel consumption and exhaust emission. Under this situation, high HC/CO emission is inevitable at low temperatures and a NSR catalyst has been investigated as a potential oxidation catalyst in combined after-treatment systems [3].

In the present study, we have investigated the effect of reduction conditions on HC/CO oxidation activities over aged NSR catalyst. Understanding these effects is important since this will help to clarify the reactions which occur over a NSR catalyst and to meet more stringent regulations which require high HC/CO/NOx conversions at low temperatures.

2. Experimental

A NSR catalyst containing Pt, Rh, BaO, and CeO₂ was prepared by the impregnation of γ -Al₂O₃ with different aqueous nitrate solutions followed by drying and calcination at 550°C for 4 h to give a sample with the following loading of 2.3%, 0.2%, 14.7% and 12.7%, respectively. The resulting catalyst was aged under flowing air at 780 °C for 20 h. Catalyst testing was performed in a quartz tube (4 mm i.d) plug flow reactor. A typical gas composition of 1000 ppm CO, 900 ppm C₃H₆, 200 ppm NO, 4.5% vol H₂O, 12% O₂, 1% Kr and balance Ar was used at a flow rate of 200 ml/min. Various reduction gas feeds were used at four different temperature (150, 300, 450, and 600 °C) to investigate the influence on the CO oxidation. The analysis of the products and reactants was carried out with a Hiden HPR 20 mass spectrometer.

3. Results and discussion

Figure 1 shows the CO conversion profiles on aged NSR catalyst obtained after 1 s reduction by C_3H_6 with λ =0.9 at different temperatures. Considering the CO oxidation profile without the pre-reduction, the CO conversion starts at about 80°C and slowly rises to 100% conversion at about 240 °C, giving the light-off temperature for CO conversion at 183 °C. Similarly, after 1 s C_3H_6 reduction at 150 °C, the profile of CO conversion is not changed since C_3H_6 is not active at low temperatures. However, it is clear that there are significant changes in CO conversion when the catalyst is treated at high temperatures, resulting in the enhancement of catalytic activity. Figure 1 also shows that the light-off temperature for CO conversion is

decreased by 65% after reducing the catalyst with C_3H_6 at 450 and 600 °C. Moreover, a similar trend has been observed for which CO is used as a reductant (results not shown here). The only difference between the two reductants is that CO is able to reduce the NSR catalyst at low temperature. It is observed that the light-off temperature for CO oxidation is shifted by 20% to lower temperature after 1 s CO reduction at 150 °C.

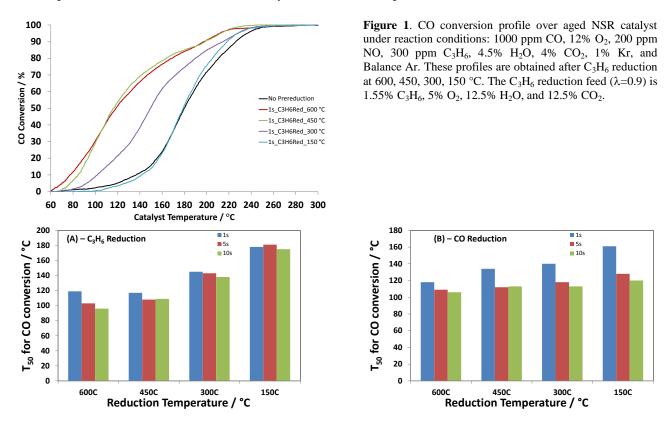


Figure 2. The light off temperature for CO conversion as a function of reduction temperatures over aged NSR catalyst under reaction conditions: 1000 ppm CO, 12% O₂, 200 ppm NO, 300 ppm C₃H₆, 4.5% H₂O, 1% Kr, 4% CO₂, and Balance Ar. The C₃H₆ reduction feed (A, λ =0.9) is 1.55% C₃H₆, 5% O₂, 12.5% H₂O, and 12.5% CO₂. The CO reduction feed (B, λ =0.9) is 7.5% CO, 2.1% O₂, 12.5% H₂O, and 11.4% CO₂.

Figure 2 shows the comparison of light-off temperature for CO conversion over aged NSR catalyst as a function of reduction time and temperature when using either C_3H_6 or CO as a reductant. It is clearly seen that the light-off temperature is shifted to low temperature after the reduction, especially at temperatures above 300 °C.

4. Conclusions

The present study shows that the catalytic CO oxidation can be substantially improved after the reduction process at various temperatures. The light-off temperature for CO conversion is significantly decreased after reducing the catalyst with either CO or C_3H_6 .

References

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