Analysis of mixed and dual-layer LNT-SCR catalyst configurations for effective storage and utilization of NH_3

Shephali Singh[1] and Divesh Bhatia[1]*

1 Department of Chemical Engineering, Indian Institute of Technology Delhi, Hauz Khas, New Delhi -110016

**Corresponding author: dbhatia@chemical.iitd.ac.in*

Highlights

- More NH_3 gets stored on mixed LNT-SCR than dual layer configuration during regeneration.
- LNT catalyst is more active than SCR in mixed configuration during lean condtions.
- SCR catalyst is more active than LNT in dual layer configuration during lean conditions.
- Diffusional limitations are more significant in dual layer compared to mixed catalyst.

1. Introduction

Combined LNT–SCR is a potential technology for the control of NO_x emissions from diesel engines by operating under cyclic fuel-rich and fuel-lean conditions. In such systems, the NH_3 that is produced over the LNT catalyst during the fuel-rich conditions is stored in the SCR catalyst. The stored NH_3 is then utilized to reduce the NO_x during the fuel-lean conditions. Use of LNT-SCR catalyst systems has the potential to reduce the precious metal costs for the LNT functionality and result in removing the urea–dosing unit. However, there is no agreement between research groups on the most effective configuration of the LNT–SCR system. Corbos et al. [1] reported that the mixed catalyst resulted in a higher NO_x conversion than the sequential LNT-SCR catalyst when CO is used as a reductant. However, Liu et al. [2] reported that the mixed LNT-SCR catalysts resulted in a lower NO_x conversion compared to the dual layer configuration due to the oxidation of stored NH_3 over the LNT catalyst. In the current work, dual layer and mixed LNT–SCR catalyst configurations are analyzed using global kinetic models and the favorable operating conditions, specific to the individual configurations are suggested.

2. Methods

A two-phase model incorporating the effect of diffusional limitations is used in the present work. The reaction rate expressions and the kinetic parameters were taken from the global kinetic model developed by Shakya et al. [3]. During the lean phase of the cyclic operation, the feed consisted of 500 ppm NO, 5% O_2 and remaining N_2, whereas the rich phase consisted of 5000 ppm H_2 and remaining N_2. The performance of dual layer and mixed catalyst configurations for various operating parameters, including temperature, lean time, rich time, total cycle time, and washcoat loading of LNT and SCR is studied in the present work.

3. Results and discussion

The effect of temperature was studied for both the dual layer and mixed catalyst configurations at a fixed ratio of the LNT and SCR catalyst volumes. The results are shown in Figure 1 and it is observed that the amount of adsorbed NH_3 in the SCR catalyst at the beginning of lean phase is higher for the mixed configuration than the dual layer catalyst for all the temperatures analyzed. This is attributed to the close proximity of the LNT and SCR catalyst in the mixed configuration, which favors the storage of NH_3 formed over LNT. On the other hand, NH_3 that is produced in the LNT layer of the dual layer catalyst diffuses through the LNT layer towards the SCR layer and gets adsorbed on the SCR catalyst. Both the diffusional limitations as well as the reaction of NH_3 with the NO_x stored in the LNT layer reduces the amount of NH_3 available for storage in the SCR layer. However, in the mixed configuration, NH_3 is transported readily from the LNT to the SCR because of the contiguousness of the two catalysts, thus preventing its consumption with the NO_x stored in the LNT catalyst to form N_2. This is confirmed by the lower amount of N_2 formed during the rich phase for the case of mixed catalyst as compared to the dual layer catalyst. A higher amount of NH_3 stored in the SCR catalyst for the mixed configuration results in a higher NO_x reduction during the fuel-lean conditions as compared to the dual layer catalyst.

In the literature, a higher NO_x conversion for the mixed catalyst has been attributed to the reaction of NO_x with the NH_3 stored over the SCR catalyst during the lean phase [1]. Contrary to this, our study suggested that for the mixed configuration, the NH_3 consumption during the lean phase occurs in the LNT in addition to that in the SCR. Figure 2 shows the comparison of the moles of NH_3 consumed for NO_x reduction during the lean phase in the LNT and SCR for both the dual layer and mixed catalyst at various temperatures. At low temperatures, the NH_3 consumption over the mixed catalyst is predicted to occur primarily in the SCR. The consumption of NH_3 in the SCR shows a maximum with temperature. The decrease at higher temperatures is because of an increase in the consumption of NH_3 through NO_x in the LNT during the regeneration phase. This also suggests that a lesser amount of NO_x will be available for storage in the LNT during the lean phase for the mixed catalyst as compared to the dual layer catalyst. Thus, the mixed catalyst will require lesser NO_x storage sites than the dual layer catalyst in order to achieve the same NO_x conversion. It is also observed from Figure 2 that for the dual layer configuration, NH_3 consumption during the lean phase occurred primarily in the SCR at all the temperatures. Thus, the difference in the NO_x reduction performance of the two catalyst configurations is dependent on the generation, storage and consumption of NH_3 during the rich phase and its efficient utilization during the lean phase of the cyclic operation.

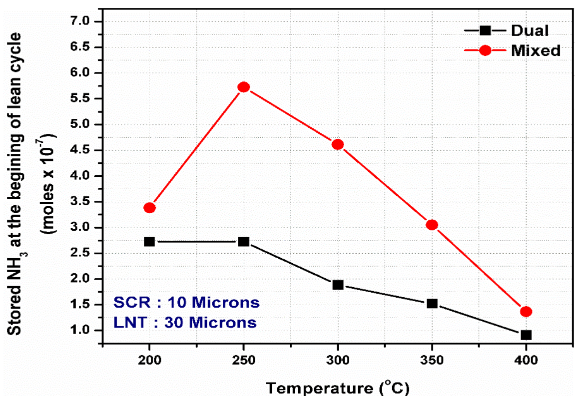

Figure 1. Comparison of the moles of NH_3 adsorbed in the SCR catalyst at the beginning of lean phase for the dual layer and mixed catalysts for various temperatures (Ratio of SCR and LNT catalyst volume = 1:3)

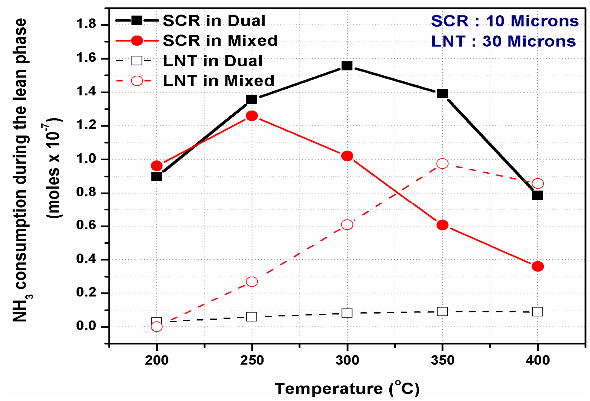

Figure 2. Comparison of NH_3 consumption in LNT and SCR during the lean phase for the dual layer and mixed catalysts at various temperatures (Ratio of SCR and LNT catalyst volume = 1:3)

4. Conclusions

It is shown that consumption of NH_3 in LNT during the lean phase plays a crucial role in the performance of the mixed catalyst, especially at high temperatures. Additionally, the close proximity of LNT and SCR catalysts in the mixed configuration results in a higher adsorption of NH_3 in the SCR during the rich phase as compared to the dual layer catalyst. The storage of NH_3 in the SCR for the dual layer catalyst is limited by the diffusional resistance and its reaction in the LNT. For the dual layer catalyst, the consumption of NH_3 during the lean phase occurs primarily in the SCR at all the temperatures. For the mixed configuration, the NH_3 consumption during the lean phase occurs primarily in the SCR at low temperatures and in the LNT at higher temperatures.

The relative contribution of the two catalysts towards NO_x reduction as well as the production and utilization of NH_3 were analyzed for various operating parameters such as the washcoat loading of LNT and SCR, lean time, rich time, overall cycle time and the concentration of reductant. These results will be presented in the complete manuscript.

References

[1] E. C. Corbos, M. Haneda, X. Courtois, P. Marecot, D. Duprez, H. Hamada, Catal. Commun. 10 (2008), 137-141.

[2] Y. Liu, M. P. Harold, D. Luss, Appl. Catal. B Environ. 121–122 (2012), 239–251.

[3] B. M. Shakya, M. P. Harold, V. Balakotaiah, Chem. Eng. J., 237 (2014), 109-122.

Keywords

LNT-SCR; dual layer; mixed catalyst; NH_3 slip.