Oxidation and decomposition of NH₃ over combustion synthesized Al₂O₃ and CeO₂ supported Pt, Pd and Ag catalysts

Parthasarathi Bera & M S Hegde

Solid State and Structural Chemistry Unit, Indian Institute of Science, Bangalore 560 012, India

Received 9 April 2002

The catalytic oxidation and decomposition of NH₃ have been carried out over combustion synthesized Al₂O₃ and CeO₂ supported Pt, Pd and Ag catalysts using temperature programmed reaction (TPR) technique in a packed bed tubular reactor. Metals are ionically dispersed over CeO₂ and fine metal particles are found on Al₂O₃. NH₃ oxidation occurs over 1% Pt/Al₂O₃, 1% Pb/Al₂O₃ and 1% Ag/Al₂O₃ at 175, 270 and 350 °C respectively producing N₂, NO, N₂O and H₂O, whereas 1% Pt/CeO₂, 1% Pb/CeO₂ and 1% Ag/CeO₂ give N₂ along with NO, N₂O and H₂O at 200, 225 and 250 °C respectively. N₂ predominates over other nitrogen-containing products during the reaction on all catalysts. At less O₂ concentration, N₂ and H₂O are the only products obtained during NH₃ oxidation. NH₃ decomposition over all the catalysts occurs above 450 °C.

Oxidation of NH₃ has been studied for a long time over various catalysts as either a primary reaction or as a side reaction. It is well established that NH₃ oxidation plays an important role in the selective catalytic reduction (SCR) of NO by NH₃ in the presence of O₂. But here oxidation of NH₃ to NO or N₂ is an unwanted side reaction. The selectivity of SCR cannot be understood without investigating the role played by oxidation of NH₃. Past and present investigations on NH₃ oxidation over noble metals and various oxides, zeolites have shown that either NO or N₂ is the selective oxidation product over most of the catalysts. In a few cases, N₂O has been found as a trace product but at higher temperatures it disappears. Ozkan et al. and Ramis et al. have studied the role of NH₃ oxidation in SCR of NO over vanadia, CuO-TiO₂ and MgO-Fe₂O₃ based catalysts. Supported Mo, V and Cu catalysts are active and selective to N₂ for NH₃ oxidation. Gang et al. have shown that CuAl₂O₄ like phase is more active than a CuO phase for NH₃ oxidation whereas [Cu–O–Cu] like species or small copper oxygen aggregates in copper zeolites are the active centers at low temperature reaction. Yahiro et al. have reported NH₃ oxidation over SiO₂-pillared oxy-compound where N₂, N₂O and H₂O are the main products.

Recently, Burch and Southward have established internal selective catalytic reduction (iSCR) through NH₃ oxidation during combustion of gasified biomass under O₂-limited environment. They have shown the selective oxidation of NH₃ to N₂ in biomass derived gases by a coupled NH₃ oxidation to NO and subsequent reduction of NO by CO or H₂ over 2% Rh/Al₂O₃, 2% Rh/10% CeO₂-Al₂O₃, 1% Pt/20% CuO/Al₂O₃, 1% Pt/20% BaO/Al₂O₃ and Al₂O₃ supported precious metal catalysts. Ayyoob and Hegde have shown that high concentration of dissociatively chemisorbed oxygen on potassium covered Pt surface favors the formation of NO from NH₃ in contrast to N₂ over pure Pt at lower oxygen concentration. Bradley et al. and Kim et al. have reported that NO is the major product on pure Pt metal at high oxygen coverage (θ₃ > 0.2 ML) and N₂ is formed at lower oxygen coverage (θ₃ < 0.2 ML). In the present study, Al₂O₃ and CeO₂ supported Pt, Pd and Ag catalysts have been synthesized by a combustion method. Here Pt, Pd and Ag metals are ionically dispersed over CeO₂ whereas fine nano size metal particles have been formed over α-Al₂O₃ support. These catalysts are examined for NH₃ oxidation using temperature programmed reaction (TPR) technique in a packed bed tubular reactor. We report herein a comparative study among Pt, Pd and Ag catalysis over Al₂O₃ and CeO₂ supports.

Materials and Methods

Synthesis and characterization

Pt, Pd and Ag metals supported on Al₂O₃ and CeO₂ have been synthesized by combustion technique. The details of the preparation have been described elsewhere. The combustion mixture for the preparation of 1% Pd/CeO₂ contained...
(NH₄)₂Ce(NO₃)₆, PdCl₂ and C₂H₆N₄O₂ (oxalyl-dihydrazide) in the molar ratio 0.99 : 0.01 : 2.33. In a typical preparation of 1% Pd/CeO₂, 10 g of (NH₄)₂Ce(NO₃)₆ (E. Merck India Ltd., 99%), 0.033 g of PdCl₂ (Glaxo India Ltd., 99%) and 5.175 g of C₂H₆N₄O₂ were dissolved in the minimum volume of water in a borosilicate dish of 130 cm³ capacity. The dish containing the redox mixture was introduced into a muffle furnace maintained at 350 °C. Initially the solution boiled with frothing and foaming and underwent dehydration. At the point of complete dehydration, the surface ignited, burning with a flame (~1000 °C) and yielding a voluminous solid product within 5 min. Similarly alumina and ceria supported Pt and Ag catalysts are prepared by the same method. These catalysts have been characterized by XRD, TEM and XPS techniques.

XRD patterns of the catalysts were recorded on a JEOL JDX-8P diffractometer using CuKα radiation with Ni filter and scan rate of 2° min⁻¹. TEM of the catalysts was carried out in a JEOL JEM-200CX transmission electron microscope operated at 200 kV.

XPS of the catalysts were recorded on an ESCA-3 Mark II spectrometer (VG Scientific Ltd., England) using AlKα radiation (1486.6 eV). Binding energies calibrated with respect to C(1s) at 285 eV were accurate within ± 0.2 eV. For the XPS analysis the powder samples were made into pellets of 8 mm diameter and kept in an ultra high vacuum (UHV) chamber at 10⁻⁹ Torr housing the analyzer. Prior to mounting the sample into the analyzing chamber it was kept in the preparation chamber at 10⁻⁹ Torr for 5 h in order to desorb any volatile species present on the catalyst.

**Temperature programmed reaction (TPR)**

The catalytic reactions were carried out in a temperature programmed reaction system equipped with a quadrupole mass spectrometer QXK300 (VG Scientific Ltd., England) for product analysis. The details of the TPR experiments have been reported earlier. Typically 0.1–0.2 g of the catalyst was loaded in a quartz tube reactor of 20 cm length and 6 mm diameter. The gases were passed over the catalyst at a flow rate of 25 μmol s⁻¹ and it was varied from 10–40 μmol s⁻¹. Accordingly, space velocity was in the range of 5 × 10⁵ to 2 × 10⁴ mol g⁻¹ s⁻¹. O₂ to NH₃ volume ratio was 1.5:1 in the excess O₂ condition whereas it was 0.7:1 in O₂ lean condition. Concentrations of reactants and products are obtained by dividing the relative intensities of molecular ions from the mass spectrometric analysis with their respective ionization cross-sections. A series of gas mixtures (e.g. NH₃ + O₂, NO + N₂) of different volume ratios were analyzed in the mass spectrometer and intensities of molecular ions were confirmed to be proportional to the respective ionization cross-sections. Errors in concentrations reported in our results are within 5%. NH₃ and O₂ gases were obtained from Bhoruka Gases Ltd., Bangalore. Their purities were better than 99% as analyzed by the quadrupole mass spectrometer.

**Results**

**Structural studies**

XRD patterns of 1% M/Al₂O₃ (M = Pt, Pd and Ag) show the broad peaks of Pt, Pd and Ag metals along with a-Al₂O₃ peaks. Metal particle sizes are in the range of 6–20 nm as calculated from Debye-Scherrer method. On the other hand, the diffraction lines agree well with CeO₂ in fluorite structure and there are no reflections corresponding to the metal in 1% M/CeO₂ patterns. Crystallite sizes of CeO₂ calculated from the Debye-Scherrer method are in the range of 15–30 nm. In all the cases, peaks corresponding to these metal oxides are not seen. Typical XRD patterns of Al₂O₃ and CeO₂ supported catalysts are shown in Fig. 1.

TEM studies show that the crystallites of CeO₂ are in the range of 15–25 nm. Very few Pt particles of 3–5 nm size are seen on CeO₂ crystallite surface in 1% Pt/CeO₂ catalyst. If all the platinum in 1%...
Pt/CeO$_2$ have been formed as Pt metal particles of 3–5 nm size, at least 10 times more number of Pt particles should have been observed in the TEM images. Therefore, over 90% of Pt in 1% Pt/CeO$_2$ are dispersed in ionic form. However, corresponding Pd and Ag particles are absent on 1% Pd/CeO$_2$ and 1% Ag/CeO$_2$ catalysts. In contrast, dispersed metal particles of 7–20 nm have been obtained on α-Al$_2$O$_3$ support. Analysis of electron diffraction patterns does show rings due to Pt, Pd and Ag metals in M/Al$_2$O$_3$.

**XPS studies**

The XPS of Pt(4f) core level region in 1%Pt/Al$_2$O$_3$ and 1% Pt/CeO$_2$ are given in Fig. 2. As binding energies of Pt(4f) and Al(2p) core levels are in closer region, a broad spectrum is obtained in the case of 1% Pt/Al$_2$O$_3$ [Fig. 2 (a)]. This spectrum can be deconvoluted into different doublets. Accordingly, Pt(4f$_{7/2,5/2}$) peaks at 71.0, 74.2, 71.9, 75.1 and 74.3, 77.5 eV could be attributed to Pt metal, Pt$^{2+}$ and Pt$^{4+}$ respectively [Fig. 2 (b)]. Here Pt is found to be dispersed mostly in +2 (72%) and +4 (21%) oxidation states on CeO$_2$ crystallites with only 7 % Pt present as Pt metal particles. This observation agrees well with the TEM studies. However, Pd and Ag are present in +2 and +1 oxidation states in 1% Pd/CeO$_2$ and 1% Ag/CeO$_2$ respectively. The respective surface concentrations of Pt, Pd and Ag have been estimated by the relation

$$X_M = \frac{I_M \sigma_M \lambda_M D_M(Ce)}{I_{Ce} \sigma_{Ce} \lambda_{Ce} D_{Ce}(M)} \quad \cdots (1)$$

where $X$, $I$, $\sigma$, $\lambda$ and $D_E$ are the surface concentration, intensity, photoionization cross section, mean escape depth and geometric factor respectively. Integrated intensities of metal core level and Ce(3d) peaks have been taken into account to estimate the concentration. Photoionization cross sections and mean escape depths have been obtained from the literature$^{34,35}$. Accordingly, surface concentrations of as-prepared Pt, Pd and Ag in CeO$_2$ are 15, 5 and 12 %.

![Fig. 2—XPS of Pt(4f) core level region in (a) 1% Pt/Al$_2$O$_3$ and (b) 1% Pt/CeO$_2$. In 1% Pt/Al$_2$O$_3$, Al(2p) of α-Al$_2$O$_3$ is also shown.](image)

![Fig. 3—TPR profiles of NH$_3$ + O$_2$ reaction over (a) 1% Pt/Al$_2$O$_3$ and (b) 1% Pt/CeO$_2$.](image)
respectively against 1% taken in the preparation. Similarly, surface concentrations of as-prepared Pt, Pd and Ag in Al₂O₃ support are 0.7, 0.8 and 0.7% respectively which are close to the bulk concentrations of the respective metals in Al₂O₃. Thus, higher dispersion of metals is achieved over CeO₂ support.

TPR studies

The catalytic properties of these materials have been investigated for the oxidation and decomposition of NH₃. Oxidation of NH₃ by O₂ occurs at 175 and 170 °C over 1% Pt/Al₂O₃ and 1% Pt/CeO₂ respectively giving N₂, N₂O and H₂O as the reaction products. Typical TPR profiles of NH₃ + O₂ reaction on 1% Pt/CeO₂ and 1% Pt/Al₂O₃ are shown in Fig. 3. In Pt/Al₂O₃, N₂ is formed selectively as the main product at lower temperature region, whereas NO concentration increases in the temperature range of 250-450 °C in Pt/Al₂O₃. At a higher temperature (>450 °C) NO intensity decreases and accordingly, increase in N₂ intensity is observed. In the case of Pt/CeO₂, NO concentration decreases above 300 °C and N₂ concentration increases. In both the cases N₂O disappears at higher temperature. On Pt/Al₂O₃ the reaction can be written as follows

\[ 6\text{NH}_3 + 6\text{O}_2 \rightarrow \text{N}_2 + 2\text{NO} + \text{N}_2\text{O} + 9\text{H}_2\text{O} <450^\circ\text{C} \]  
\[ 4\text{NH}_3 + 3\text{O}_2 \rightarrow 2\text{N}_2 + 6\text{H}_2\text{O} >450^\circ\text{C} \]  

NH₃ oxidation over 1% Pd/Al₂O₃ starts at 270 °C and N₂, NO, N₂O and H₂O are the reaction products. Here NO intensity increases and N₂O disappears at higher temperatures (>400 °C). In 1% Pd/CeO₂, NH₃ oxidation starts at 250 °C giving N₂ as the main nitrogen-containing product. Above 350 °C NO concentration increases, N₂ intensity decreases and N₂O decomposes. But at a higher temperature N₂ intensity increases.

In the case of 1% Ag/Al₂O₃, N₂ starts at 350 °C during NH₃ oxidation. Above 400 °C NO and trace amount of N₂O are observed along with N₂. In the case of 1% Ag/CeO₂, N₂ and a very less amount of N₂O are obtained at 275 °C. Above 400 °C NO comes up as a product along with N₂ and accordingly the intensity of N₂ decreases. N₂O decomposition occurs in both cases at higher temperatures.

In NH₃ oxidation under excess oxygen condition, N₂, NO, N₂O and H₂O are the products from the reaction over all the catalysts. Complete NH₃ conversion occurs at higher temperatures in all the cases. NH₃ conversion and N₂ selectivity at a particular temperature during NH₃ oxidation over all the catalysts are given in Table 1. The selectivity to N₂ formation (Sₙ₂) is calculated as follows:

\[ S_{\text{N}_2} = \frac{100 \times \text{[Amount of N}_2\text{ formed]}}{\text{[Amount of NH}_3\text{ consumed]}} \]

\[ = \frac{100 \times \text{[Amount of N}_2\text{ formed]}}{\text{[Amount of N}_2\text{ formed] + [Amount of NO formed] + [Amount of N}_2\text{O formed]}} \]

Table 1—NH₃ conversion and N₂ selectivities during NH₃ oxidation over Al₂O₃ and CeO₂ supported catalysts.

<table>
<thead>
<tr>
<th>Catalyst</th>
<th>Temp. (°C)</th>
<th>NH₃ conversion (%)</th>
<th>N₂ selectivity (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pt/Al₂O₃</td>
<td>450</td>
<td>93</td>
<td>68</td>
</tr>
<tr>
<td></td>
<td>500</td>
<td>100</td>
<td>89</td>
</tr>
<tr>
<td>Pd/Al₂O₃</td>
<td>350</td>
<td>86</td>
<td>74</td>
</tr>
<tr>
<td></td>
<td>400</td>
<td>93</td>
<td>82</td>
</tr>
<tr>
<td>Ag/Al₂O₃</td>
<td>400</td>
<td>49</td>
<td>92</td>
</tr>
<tr>
<td></td>
<td>450</td>
<td>77</td>
<td>80</td>
</tr>
<tr>
<td>Pt/CeO₂</td>
<td>450</td>
<td>100</td>
<td>71</td>
</tr>
<tr>
<td></td>
<td>500</td>
<td>100</td>
<td>77</td>
</tr>
<tr>
<td>Pd/CeO₂</td>
<td>300</td>
<td>94</td>
<td>73</td>
</tr>
<tr>
<td></td>
<td>350</td>
<td>100</td>
<td>70</td>
</tr>
<tr>
<td>Ag/CeO₂</td>
<td>350</td>
<td>71</td>
<td>91</td>
</tr>
<tr>
<td></td>
<td>400</td>
<td>81</td>
<td>95</td>
</tr>
</tbody>
</table>

Fig. 4—Rates of N₂, NO and N₂O formation over (a) 1% Pt/Al₂O₃ and (b) 1% Pt/CeO₂ in NH₃ + O₂ reaction.
During NH₃ oxidation over all the catalysts N₂ predominates and selectivity to N₂ is higher in relation to other nitrogen-containing products. Rates of formation of all nitrogen-containing products such as N₂, NO and N₂O over Pt/Al₂O₃ and Pt/CeO₂ catalysts are given in Figs. 4 (a) and (b) respectively. It is important to note that N₂ formation rates are higher than NO and N₂O formation rates. Above 500 °C selectivity to N₂ is nearly 100% for NH₃ oxidation over Pt/Al₂O₃. In Figs. 5 (a) and (b), N₂, NO and N₂O formation rates are given for Pd/Al₂O₃ and Pd/CeO₂ respectively. Below 400 °C, NO formation rates are much lower than N₂ over Pd/Al₂O₃, but above 400 °C, NO formation rates increase. Rates of NO formation increase over 1% Pt/CeO₂ above 300 °C. Similarly, rates of N₂, NO and N₂O formation for Ag/Al₂O₃ are given in Fig. 6. Rates of N₂ formation are higher at higher temperatures over both Pt/Al₂O₃ and Pt/CeO₂ catalysts. Accordingly, NO formation rates are lower over these catalysts in the higher temperature region. On the other hand, N₂ formation rates decrease over Pd/Al₂O₃ and Ag/Al₂O₃ catalysts and the rates of NO formation increase in the case of Pd/Al₂O₃, Pt/CeO₂ and Ag/CeO₂ catalysts. But NO formation rates do not exceed N₂ formation rates in all the cases in the entire range of temperature. N₂ formation rates are much higher in comparison with NO and N₂O over all the catalysts.

NH₃ oxidation at low O₂ concentration was also carried out over Pt/CeO₂ and Pd/CeO₂ catalysts. When NH₃ and O₂ gases in 1:0.7 ratio were passed over Pt/CeO₂ catalyst, N₂ and H₂O are the only products over the entire range of temperatures. A sharp increase in N₂ and H₂O intensities is observed at 250 °C. Accordingly, O₂ is fully utilized in the reaction. Other nitrogen-containing products like NO and N₂O are absent during the reaction. Here excess NH₃ remains in the products indicating 'NH₃ slip'. The NH₃ + O₂ reaction can be written as follows:

\[ 6\text{NH}_3 + 3\text{O}_2 \rightarrow 2\text{N}_2 + 6\text{H}_2\text{O} + 2\text{NH}_3 \] ...(4)

Similarly, N₂ and H₂O are seen as the NH₃ + O₂ reaction products over 1% Pd/CeO₂ catalyst in O₂-lean condition. TPR profiles of NH₃ + O₂ reaction over 1% Pt/CeO₂ and 1% Pd/CeO₂ in O₂-lean condition are shown in Fig. 7.

In order to understand the role of lattice oxygen in NH₃ oxidation, NH₃ was passed over all the catalysts and N₂ and H₂ are obtained as the products, not N₂ and H₂O. Complete NH₃ decomposition occurs at 700–750 °C over Pt/CeO₂ catalyst. Over Pd/CeO₂ catalyst, H₂ and N₂ are obtained at 650 °C whereas over Pd/Al₂O₃ the decomposition occurs at 500 °C. The rates of N₂ and H₂ formation in NH₃ decomposition over Pt/CeO₂ are shown in Fig. 8. The concentration ratio of N₂ to H₂ of 1:3 is maintained during NH₃ decomposition and rate of formation of H₂ is three times higher than that of N₂ in entire range of temperature. Thus, absence of H₂O formation indicates that lattice oxygen from CeO₂ as well as Al₂O₃ is not utilized during the reaction. Therefore, in M/ CeO₂ and M/Al₂O₃ as catalysts, it is the feed oxygen that is utilized in the oxidation of NH₃.
BERA et al.: DECOMPOSITION OF NH₃ OVER Pt, Pd & Ag CATALYSTS

Discussion

NH₃ oxidation is of great importance regarding SCR of NO and HNO₃ synthesis. The possible reactions involved in NH₃ oxidation are as follows:

4NH₃ + 3O₂ → 2N₂ + 6H₂O  ...(5)
4NH₃ + 5O₂ → 4NO + 6H₂O  ...(6)
2NH₃ + 2O₂ → N₂O + 3H₂O  ...(7)
4NH₃ + 7O₂ → 4NO₂ + 6H₂O  ...(8)

Although the decomposition of NH₃ and NO are also possible reactions that could take place during NH₃ oxidation, they become significant only at a temperature above 500 °C. The reaction between NH₃ and O₂ produces some N₂O at low temperatures, but above 400 °C the only nitrogen-containing species observed are N₂ and NO. In fact, NO₂ was not observed during the reaction in any of the above experiments.

It is widely proposed that NH₃ oxidation is a multi-step process involving different intermediate species. Bodenstein, Andraßow and Zawadzki have advocated the formations of hydroxylamine, nitroxyl and imide respectively during the first step of the reaction. Fogel et al. and Nutt and Kapur have observed NO as the product of first reaction step which occurs on the catalyst surface. They suggested that N₂ production at low temperatures can be interpreted more simply as a bimolecular reaction between NO and NH₃ to produce N₂. However, at high temperatures the unimolecular decompositions of NH₃ and NO also produce N₂. Recently, Carley et al. have shown a number of distinct reaction pathways during NH₃ oxidation in the formation of either chemisorbed imide or nitrogen adatoms.

In the present study, metals are ionically dispersed on CeO₂ support, whereas nano size metal particles are homogeneously dispersed on α-Al₂O₃ matrix. In 1% Pt/CeO₂, Pt is in +2 and +4 states along with small amount of Pt particles whereas Pd and Ag are in +2 and +1 states in 1% Pd/CeO₂ and 1% Ag/CeO₂ respectively. Oxidation of NH₃ over Pt/CeO₂ catalyst gives N₂, NO and N₂O as the reaction products. Above 300 °C N₂O decomposes to give N₂ and O₂.

In 1% Pd/Al₂O₃ and in 270–370 °C region, N₂ concentration increases and at higher temperatures (> 400 °C) it decreases. It is worthy to mention our earlier results of NH₃ oxidation over Cu/CeO₂ catalyst. NH₃ oxidation over 5% Cu/CeO₂ gives N₂ as the main product at lower temperatures (< 400 °C).
But NO is the main nitrogen-containing species at higher temperatures (> 400 °C) along with less content of N₂. Therefore, SCR of NO with NH₃ in the presence of O2 over Cu/CeO₂ works at 150–350 °C region. Above 350 °C NO predominates over N₂. On the other hand, in the entire range of temperature N₂ is the main product of NH₃ oxidation over CeO₂ and Al₂O₃ based Pt, Pd and Ag catalysts. In the case of less O₂ concentration, N₂ is the sole nitrogen-containing product. Formation of NO and N₂O requires more O₂. Due to unavailability of sufficient O₂ in the O₂-lean condition, NO and N₂O are not observed during the reaction in the entire range of temperature. These observations are in complete agreement with BHK model. However, NH₃ oxidation over Al₂O₃ and CeO₂ supported Pt, Pd and Ag catalysts shows that N₂ is the major product during the reaction compared to NO, N₂O and NO₂. Relatively low temperature of NH₃ oxidation over Pd/CeO₂ and Ag/CeO₂ can be due to higher dispersion of metals on CeO₂ support compared to Al₂O₃. However, a similar increase in the catalytic activity is not prominent in the case of Pt/CeO₂ in relation to Pt/Al₂O₃. Pt/Al₂O₃ and Pt/CeO₂ catalysts show relatively low temperature NH₃ oxidation compared to supported Pd and Ag catalysts. Rates of N₂ formation are higher in the case of Pt/Al₂O₃ and Pt/CeO₂ catalysts compared to Pd/Al₂O₃, Pd/CeO₂, Ag/Al₂O₃ and Ag/CeO₂ catalysts.

NH₃ decomposition over these catalysts predicts the role of lattice oxygen in NH₃ oxidation. There is no loosely bound O₂ in the lattice which can react with NH₃. As NH₃ decomposition to N₂ and H₂ occurs at a much higher temperature over these catalysts compared to NH₃ oxidation, formation of N₂ as a product during NH₃ oxidation is only due to oxidation. The unavailability of labile oxygen in the catalysts suggests that the reaction is unlikely to follow Mars-Van Krevelen pathway. On the other hand, lower reaction temperature in the presence of O₂ in a catalytic reaction indicates that the catalyst surface does provide suitable active sites for both NH₃ and O₂ adsorption to adopt a Langmuir-Hinshelwood pathway.

Conclusion

The important findings of the study are as follows: (i) combustion synthesized alumina and ceria supported Pt, Pd and Ag catalysts are all active in NH₃ oxidation at low temperature; (ii) ionic as well as metal sites are the active centers for NH₃ oxidation; (iii) Pt/CeO₂ and Pd/Al₂O₃ show low temperature NH₃ oxidation among all the catalysts; (iv) in all cases N₂ predominates over other nitrogen-containing products; (v) N₂ and H₂O are the only products under O₂-lean condition; and (vi) lattice oxygen is not utilized during the oxidation, only feed oxygen reacts with NH₃ to produce nitrogen-containing species.

Acknowledgement

PB is thankful to the CSIR, Government of India, for the award of a research fellowship. We thank Professor K S Gandhi, Department of Chemical Engineering, Indian Institute of Science, for useful discussions. Financial support from the Department of Science and Technology (DST), Government of India is gratefully acknowledged.

References

BERA et al.: DECOMPOSITION OF NH₃ OVER Pt, Pd & Ag CATALYSTS