Silver catalysts for NO\textsubscript{x} storage and reduction using hydrogen

David W. J. McClymont,\textsuperscript{a,b} Stan T. Kolaczkowski,\textsuperscript{c} Kieran C. Molloy,\textsuperscript{c} Serpl Awdry\textsuperscript{b}\textsuperscript{a}\textsuperscript{a}\textsuperscript{a\textcopyright}Doctoral Training Centre, Centre for Sustainable Chemical Technologies, \textsuperscript{b}Department of Chemical Engineering, \textsuperscript{c}Department of Chemistry, University of Bath, BA2 7AY, UK.

e-mail: D.W.J.McClymont@bath.ac.uk; URL:http://www.bath.ac.uk/csct

Introduction

Legislation is in place to control emissions from various pollutant processes i.e. the Waste Incineration Directive (WID) regulates activities that involve burning or gasification of waste (Figure 1). Technologies have been developed which react a reductant with NO\textsubscript{x}, emissions forming harmless N\textsubscript{2} and H\textsubscript{2}O. Development of a material and process to treat NO\textsubscript{x} emissions using H\textsubscript{2} is the aim of this project. Utilising H\textsubscript{2} already present in the system (Figure 1) could provide a reductant which does not have to be specially manufactured (in contrast to e.g. NH\textsubscript{4}H\textsubscript{2}PO\textsubscript{4}, and hence would be a cleaner approach. H\textsubscript{2} can also be used in NO\textsubscript{x} storage and reduction (NSR) processes where NO\textsubscript{x} species are first ‘trapped’ and subsequently reduced through alternate lean and rich–burn cycles (Figure 2).

Theory

Pt/Ba/\textsubscript{Al}\textsubscript{2}O\textsubscript{3} is considered the ‘standard’ NSR catalyst and has been extensively studied for this process since original publication by Takahashi et al. (1996). As such, the chemical processes involved during NSR cycles are well understood. NSR catalysts generally consist of a noble metal and ‘storage component’ (alkaline earth metal) supported on alumina. Operating through alternate lean and rich conditions, the NO\textsubscript{x} is initially ‘stored’ on the catalyst surface during lean conditions (Figure 2), in the form of nitrates and nitrites. Subsequent introduction of a reductant, in this case H\textsubscript{2}, reduces the stored species to form N\textsubscript{2}. Although silver catalysts have previously been explored for related deno\textsubscript{x} applications, primarily in Selective Catalytic Reduction (SCR) approaches (e.g. Burch et al. (2004)), their performance in NSR reactions has not been reported

Experimental

Catalysts

Catalysts prepared using impregnation techniques and supported on honeycomb monoliths (Figures 3 & 4).

Table 1: Catalyst Compositions

<table>
<thead>
<tr>
<th>Loading (wt%)</th>
<th>Silver</th>
<th>Storage Component</th>
<th>Barium</th>
<th>Potassium</th>
</tr>
</thead>
<tbody>
<tr>
<td>5</td>
<td>14</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>5</td>
<td>-</td>
<td>38</td>
<td>-</td>
<td>-</td>
</tr>
</tbody>
</table>

Results

Figure 6: H\textsubscript{2}-NSR Lean and Rich cycle over Ag/Ba/\textsubscript{Al}\textsubscript{2}O\textsubscript{3} catalyst at 400 °C

Both catalysts appear to demonstrate some potential in their ability to ‘store’ and subsequently reduce NO (Figures 6 and 7). Ag/K offers a greater storage capacity (which may be related to storage component loading) and less formation of side products. Ag/Ba demonstrates a greater ability to reduce the stored NO\textsubscript{x} with minimal NO\textsubscript{x} desorption seen in the rich phase. Both catalysts demonstrate different affinities for NO (Figure 7), with Ag/K demonstrating reaction of surface species with NO, even in the absence of O\textsubscript{2}; formation of N\textsubscript{2}O (+1200 ppm) and NO\textsubscript{2} at higher temperatures.

Conclusions

Complete catalyst characterization including Temperature-programmed Surface Reaction (TPSR) studies investigate possible impact of ‘KK leaves’ \textsubscript{Al}O\textsubscript{3} support structure (Figure 9); freeze drying process produces thin leaves of \textsubscript{Al}O\textsubscript{3} (thickness 0.2-0.8 \textmu m), first reported by Kolaczkowski et al. (2006). The thin layer structure may offer less resistance to diffusion, possibly impacting the quantity of catalyst required

References


Burch, R. et al. (2000). Exceptional Activity for NO\textsubscript{x} Reduction at Low Temperatures Using Combinations of Hydrogen and Higher Hydrocarbons on Ag/Al\textsubscript{2}O\textsubscript{3} Catalysts. Topics in Catalysis, Vol. 30/31, No. 1-4, pp. 19-25.


Acknowledgements

The authors gratefully acknowledge the EPSRC (grant number EP/I008692/1), boundless support of our colleagues in the Chemistry and Chemical Engineering departments and the use of facilities in the University of Bath’s Centre for Sustainable Chemical Technologies. All authors have contributed equally to the research work presented in this paper.