Low Temperature Carbon Monoxide (CO) Oxidation using Microfibrous Entrapped Pt-Ceria/Silica Catalyst

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Introduction

CO is harmful to humans as well as the environment, and catalytic oxidation of CO to CO$_2$ is an efficient way of removing CO. CO oxidation at low temperature still poses a major challenge$^1$, due to CO self-poisoning of the catalyst at ambient temperature. Although, supported gold catalysts are widely used for the low temperature CO oxidation, they lose their activity in the presence of moisture. A catalyst with active noble metal platinum and ceria as promoter, both supported in a nano-dispersed form on a high surface area silica support has been developed. The catalyst has a significant activity for CO oxidation even in the presence of moisture. The optimization of various catalyst preparation process parameters was carried out experimentally. The surface characterization was also carried out using CO chemisorption, N$_2$ physisorption, and powder X-ray diffraction (XRD). The characterization results were correlated with the activity of the catalysts.

The catalyst showed a significant increase in activity with decreasing particle size ($\eta<0.95$, even at 100µm), indicating a clear transport limitation. Considering the difficulties associated with packed bed of small particles, such as high pressure drop, bed channeling, and packing issues, a new class of micro-structured materials consisting of microfibrous entrapped sorbents/catalysts (MFES/MFEC)$^{2,3}$ has been developed at Auburn University. These MFEC have shown to increase radial dispersion and reduce flow maldistributions$^4$. When this newly developed Pt-CeO$_2$/SiO$_2$ catalyst was entrapped in a microfibrous matrix of metal fibers, this MFEC demonstrated a significant improvement in CO conversion compared to a conventional packed bed configuration while maintaining a lower pressure drop.

Materials and methods

Catalysts were prepared by successive incipient wetness impregnation method using aqueous cerium nitrate solution as a ceria precursor and diammine platinum (II) nitrite in ammonium hydroxide as a platinum precursor. The CO oxidation activity of catalysts was studied using a quartz tube reactor, wherein the CO concentration was detected by using an electrochemical sensor. The MFEC were prepared using wet lay paper making process.

Results and discussions

The Pt-Ceria/Silica catalyst was found to have a greater activity for low temperature CO oxidation in comparison with other catalysts as shown in Table 1. During this study, it was also found that MFEC when used in CO oxidation helped minimize the hot and cold spots in the reactor bed due to better inter-phase heat transfer rate. This in turn improved the activity of the catalyst and reduced the decay rate of the catalyst, which was particularly visible at high CO concentration wherein the CO inhibition kinetics takes over. Figure 1 shows the CO oxidation performance of MFEC at higher CO concentration at room temperature in humid air.

<table>
<thead>
<tr>
<th>Catalyst</th>
<th>Conversion (%)</th>
<th>Catalyst</th>
<th>Conversion (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Carulite</td>
<td>1%</td>
<td>Moleculete</td>
<td>1%</td>
</tr>
<tr>
<td>5%Pt-Alumina</td>
<td>23%</td>
<td>5%Pt-Alumina</td>
<td>60%</td>
</tr>
<tr>
<td>5%Pt-5%Co-Alumina</td>
<td>31%</td>
<td>5%Pt-5%Co-Silica</td>
<td>26%</td>
</tr>
<tr>
<td>1%Au/TiO$_2$</td>
<td>75%</td>
<td>3%Pt-12%CeO$_2$/SiO$_2$</td>
<td>99%</td>
</tr>
</tbody>
</table>

Test conditions: CO: 250 ppm; temp: 25°C; humidity: 50% RH; MFEC bed thickness: 2.5mm; particle size: 200 µm; face velocity: 45cm/sec

The activity of this catalyst can be attributed to the presence of ceria which acts as an oxygen reservoir. The proposed mechanism for CO oxidation is a redox mechanism which involves transfer of lattice oxygen from ceria to the adsorbed CO molecule on platinum. The resulting lattice oxygen vacancy is then immediately filled by oxygen from air.

Significance

This microfibrous entrapped Pt-CeO$_2$/SiO$_2$ catalyst demonstrated a great potential for CO removal applications involving moisture. This MFEC can be used for air purification, as a cathode air filter for PEM fuel cells, also in emergency escape products for CO abatement.

References

2. B.J. Tatarchuk et al, US patent 5080963