Novel Heterogeneous Contacting System – Microfibrous Entrapped Catalysts for Low Temperature Carbon Monoxide (CO) Oxidation

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Introduction

Low temperature CO oxidation is characterized by slow surface reaction kinetics and strong CO self-poisoning, which compete with one another. This tradeoff becomes especially problematic at lower temperatures due to the higher activation energy for the surface reaction compared to diffusion. In the case of larger catalyst particles and higher CO concentrations, the increased thermal mass of the large particle and its lower effectiveness factor limit the opportunity for, and potential benefits of, ignition. Conventional packed beds of small particles (ca. < 200 micron) currently pose technological problems due to intrabed flow maldistributions, high pressure drops, and the inability to immobilize/stabilize thin beds toward upsets in orientation and vibration. Monolithic reactors provide low pressure drop for immobilized catalyst washcoats but suffer from low volumetric catalyst loading, high external resistances, and relatively high levels of inert thermal mass.

To circumvent a number of the above noted problems and tradeoffs, a new class of micro-structured materials consisting of small sorbent/catalyst particulates entrapped by metal, ceramic, or polymer microfibers (MFES/MFEC)[1,2] has been developed and optimized for low temperature CO oxidation, among other applications. These materials immobilize active particulates (D_P: 10-200µm) by entrapping them within a network of sinter-locked fibers (1.5-12µm dia). The immobilization of small particles from 15 to 40 vol% loading results in higher heterogeneous contacting efficiency, reduced pressure drop, and lower effective thermal mass. Fibers are shown to strongly reduce both microscale interparticle peaking velocities and intrabed flow maldistributions normally associated with small particle intrabed clustering[3]. In comparison with conventional packed beds, MFES provide significant increases in volumetric reactivity[4] and radial dispersion at reduced driving forces normally used for promoting heterogeneous contacting (e.g., reduced pressure drop, temperature, catalyst loading, etc.).

Materials and methods

A promoted noble metal catalyst supported on silica has been developed for low temperature CO oxidation, wherein the promoter can be either tin oxide or cerium oxide. The catalysts were prepared by incipient wetness impregnation. The CO oxidation activity of the different catalysts was determined using a tubular quartz microreactor operated in both integral and differential modes of operation. The CO concentration was detected by means of an electrochemical sensor. MFEC structures were prepared using standard wet lay paper making techniques. Characterization of the catalysts was carried out using CO chemisorption, N_2 physisorption, O_2/H_2 chemisorption, and X-ray diffraction (XRD).

Results and discussion

During this investigation, catalysts developed by the authors for low temperature CO oxidation were prepared and compared in microreactor evaluations as a function of particle size and bed type (viz., packed bed versus microfibrous entrapment). The effective CO oxidation activity of the catalysts increased with decreasing particle size as shown in Figure 1. This showed a clear transport limitation for larger particles (η<0.95, even for D_P=100µm). The comparison of CO oxidation activity for packed beds and MFEC structures employing the same particle size and catalyst loading is shown in Figure 2. MFES catalysts outperformed packed beds at equivalent particle sizes and catalyst loadings demonstrating enhanced contacting efficiency at reduced driving forces. From Figure 2, it can also be seen that the packed bed experienced increased CO self-poisoning compared to the MFEC structure. The better performance of MFEC can be attributed to decreased flow maldistributions and enhanced ignition prospects, especially at higher CO concentrations.

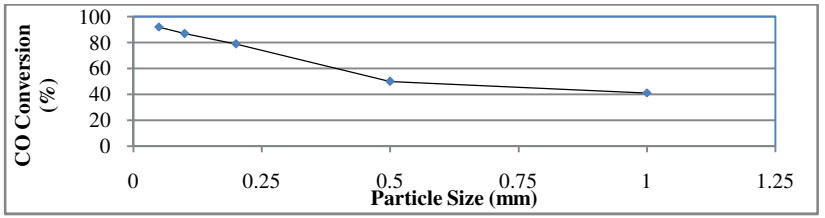

Figure 1: Test Conditions: CO: **750 ppm**; temp: 25°C; humidity: 90% RH; catalyst bed thickness: 8mm; catalyst: 3%Pt-25%SnO_2 on Silica; face velocity: 45 cm/sec [**Packed Bed**]

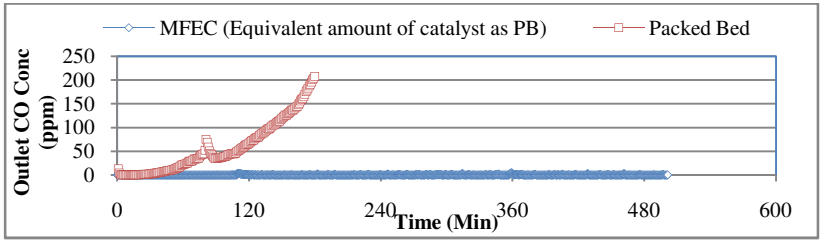

Figure 2: Test Conditions: CO: **2500 ppm**; temp: 25°C; humidity: 90% RH; catalyst: 1.0g; D_P: 200 µm; face velocity: 10 cm/sec; catalyst: 3%Pt-25%SnO_2 on Silica; MFEC: 30 vol% loading

Significance

Microfibrous entrapped catalysts provide relatively high heterogeneous contacting efficiency and high volumetric CO oxidation rates at low temperature and low pressure drop. MFEC are useful for performance limiting CO oxidation applications where the driving forces for heterogeneous contacting must be minimized such as in air purification and cathode air protection of PEM fuel cells.

References

1. B.J. Tatarchuk, *et al* US Patent 5080963
2. B.J. Tatarchuk, *et al* US Patent 5102745
3. R.R. Kalluri, *et al*, *Separation and Purification Technology* 62 (2008) 304-316
4. B.K. Chang, *et al*, *J. Mater. Eng. Perform*. 15 (2006) 195-202