Ortho-dichlorobenzene oxidation over Pd/Co-sulfated zirconia washcoated minimonoliths

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Introduction

Catalytic oxidation is considered one of the best methods for the control of polychlorinated dibenzodioxin and dibenzofuran (PCDD/F) emissions from stationary sources [1,2]. This is a viable and economic approach due to its low destruction temperature and its excellent selectivity toward the formation of harmless products [2]. Due to the high toxicity of the original compounds, 1,2-dichlorobenzene (o-DCB) is considered a suitable model compound to study the catalyzed deep oxidation of PCDD/Fs [3]. Pd/Co sulfated zirconia powder catalysts were found to be promising for o-DCB oxidation under dry and wet conditions [3]. However, for industrial applications monolithic catalysts have several advantages compared with powder catalysts [4,5]. In this contribution we assessed the behavior of Pd/Co-ZS washcoated cordierite honeycomb minimonoliths for the oxidation of o-DCB under dry and wet conditions between 100 - 450 °C.

Materials and Methods

Pd/Co-SZ samples in powder form were synthesized following our previously reported procedure [3, 6]. Pd loading was approximately 0.2 wt-%. Slurries were prepared by mixing powder Pd/Co-SZ samples with the appropriate amount of water, water to catalyst powder ratio of 2.3, and 5 wt- % binder [4,5]. Under these conditions the slurries contained 25% solids and the pH was 2.3. Before coating, cordierite minimonoliths (10x10x12 mm, 36 channels) were treated in a nitric acid solution for 4 h and washed with water until neutral. Afterwards, minimonoliths were introduced into acetone for 2 h, dried in a microwave oven at 100 °C, and finally calcined under static conditions at 600 °C for 2 h. Washcoated samples were prepared by dipping minimonoliths into the slurry for 10 s and excess slurry softly blown off with compressed air. Washcoated minimonoliths were dried in a microwave oven and then weighed. These steps were repeated until a 15% washcoat loading was obtained. Resistance (adhesion, abrasion) and catalytic tests were carried out with fresh and used washcoated minimonoliths. Oxidation reactions were performed in a pyrex fixed-bed reactor under atmospheric pressure in the temperature range 100 - 450 °C. The feed consisted of about 500 ppmv o-DCB obtained by passage of a N_2 stream through an o-DCB saturator maintained at room temperature, 8% O_2 and balance N_2. Several runs were also performed in the presence of approximately 4.0–5.0% water vapor in the feed. The gas space velocity was approximately 60000 cm^3/g h. All samples were heated at 500 ºC during 4 h in flowing helium and then allowed to cool at room temperature before the reaction. Durability tests of Pd/Co-SZ washcoated minimonoliths were carried out using dry and wet feeds at 450 °C for 4h.

Results and Discussion

Resistance tests show that Pd/Co-SZ minimonolith washcoats have good adhesion properties (weight loss around 5 wt %.). Highly acidic conditions favor adequate slurry viscosity and could lead to an efficient coating process for honeycomb structures. The characteristic acidity of sulfated zirconia leads us to obtain a pH < 3 and additional acid addition was not required. Similar conversions and selectivities are obtained for the oxidation of o-DCB over both powder and Pd/Co-SZ washcoated minimonoliths (Figure 1). Conversions are noticeable at temperatures over 300 ºC. As shown in Figure 2 o-DCB conversion on Pd/Co-SZ washcoathed minimonoliths was similar for both dry and wet feeds even after four hours of reaction at 450 °C.

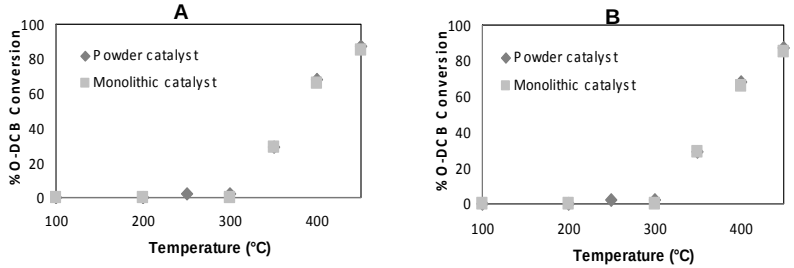

Figure 1 Effect of temperature on the oxidation of o-DCB over 0.2% Pd/Co-SZ for both powder and monoliths, under dry (A) and (B) wet feed. Space velocity 60000 $cm^3 g^{-1} h^{-1}$.

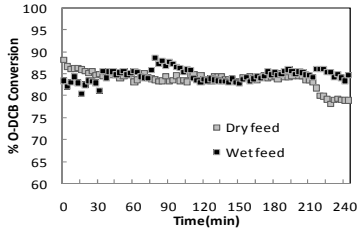

Figure 2 Durability tests for o-DCB oxidation at 450ºC over 0.2% Pd/Co-SZ minimonolith using dry and wet feeds. Space velocity 60000 $cm^3 g^{-1} h^{-1}$.

Significance

We developed a procedure to prepare washcoated Pd/Co-SZ ceramic minimonoliths with good adhesion properties. Besides, they exhibit o-DCB conversions similar to those in powder form even under wet conditions. The acidity of Pd/Co-SZ itself is adequate for obtaining an optimum viscosity of Pd/Co-SZ slurries.

References

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