Preferential oxidation of CO over Pt-Sn/AC catalyst: Adsorption, performance and DRIFTS studies

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Introduction

The high activity of Pt-SnO_x system in CO oxidation has been ascribed to a synergistic bifunctional mechanism in which Pt provides the adsorption sites for CO, while oxygen adsorbs dissociatively on SnO_x **[1]**. In a series of papers, firstly several Pt-Sn/AC catalysts were prepared, characterized in a detailed fashion **[2,3]** and tested for their CO oxidation activities **[3]**. The results clearly showed that the CO oxidation performance of the catalyst strongly depends on Pt:Sn loading ratio, preparation procedure and AC pretreatment. The results hinted that the active phase on Pt-Sn catalyst prepared on nitric acid oxidized activated carbon (AC-N) is the Pt_3Sn alloy. Pt_3Sn is formed with the help of the carboxylic acid rich surface chemistry of the support and/or through the interaction between the metallic Pt phase and the SnO_x interface, and have resulted in enhanced CO oxidation ability **[3].**

In this work, PROX over Pt-Sn/AC-N and Pt/AC-N systems has been studied by using FTIR-DRIFTS-MS system under operando conditions with a feed including CO, H_2, O_2 and He. Adsorption properties of CO and CO_2 have been studied at reaction temperatures. In the performance tests on-line-real-time PROX activity profiles of an AC supported system for various feed conditions including the fully realistic feed were obtained. in order to understand the effect of reaction parameters on the PROX activity and selectivity.

Experimental

Commercial activated carbon was washed with 2 N HCl, then subjected to HNO_3 oxidation treatment. 1 wt% Pt-0.25 wt% Sn/AC-N catalyst was prepared by sequential impregnation. IR measurements were obtained in a FTIR Spectrometer (Bruker, Vertex V70) using a DRIFTS reaction cell. CO and CO_2 adsorption isotherms were obtained for fresh AC-N, Pt/AC-N and Pt-Sn/AC-N samples by using Intelligent Gravimetric Analyzer (Hiden Isochema). During the performance tests, the total flow rate was kept constant at 100 ml/min and for each freshly reduced sample, the reaction was conducted at 135˚C, 125˚C, 115˚C and 110 ˚C, in a Decreasing Temperature Progression fashion.

Results/Discussion

The overall evaluation of the performance test results at 110 ^oC (Figure 1) shows that Pt-Sn/AC-N is a very promising PROX catalyst to be used in fuel processors; it reached ca. 90% conversion level with a realistic feed having O_2:CO=1.25. It is clear from both performance tests and DRIFTS/adsorption studies that Pt_3Sn alloy formation on surface oxygen group rich AC-N support has led to augmented CO adsorption and faster surface reaction involving intermediate hydroxyl groups which bring along increase in CO conversion and selectivity as the temperature decreases within the range 110-135 ^oC. Besides the high CO oxidation activity and the fact that CO adsorption does not lead to CO poisoning, weak CO_2 adsorption strength of the catalyst limits CO_2 inhibition during PROX reaction.

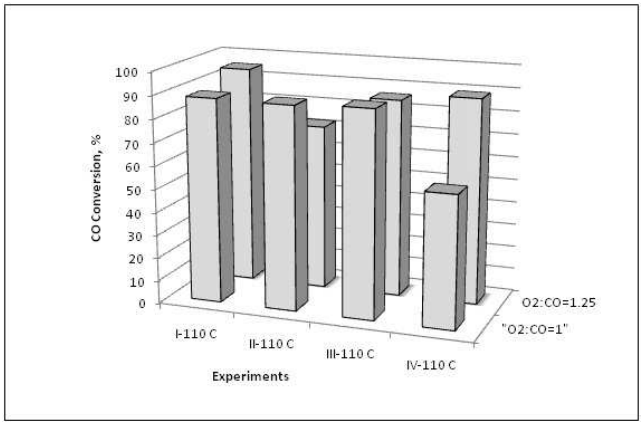

Figure 1. An overall comparison of PROX activities of the Pt-Sn/AC-N sample on CO conversion basis for different reaction conditions

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References.

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