

A study of the preparation conditions of Pt/SiO₂ catalysts for the total oxidation of naphthalene a model polyaromatic hydrocarbon

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Introduction

Polycyclic aromatic hydrocarbons (PAHs) are a specific class of Volatile Organic Compounds (VOCs) and have been cited as carcinogenic and environmentally hazardous substances. Their release into the atmosphere results predominantly from the incomplete combustion of many fuels, and originates from many domestic and industrial sources. The health and environmental issues associated with PAH emissions has led to stringent regional and international restrictions on the concentration levels permitted for discharge [1]. A number of control technologies are available, however, catalytic total oxidation is one of the most efficient and economic methods for VOC control and hence potentially it is also important for control of PAH emissions.

The number of studies investigating the catalytic oxidation of polycyclic aromatic compounds is limited in number. A number of studies have concentrated on Pt-based catalysts as platinum has been reported to be the most active metals for total oxidation [2]. In previous work we have investigated the influence of support for platinum catalysts and identified silica as being more effective when compared with alumina, ceria and tin oxide [3]. In this work, we investigate the effect of a variety of preparation variables, which were metal loading and calcination time, temperature and atmosphere, on the activity of Pt/SiO₂ catalysts for naphthalene total oxidation. Naphthalene is selected as a model compound as it is widely accepted as a suitable model for a typical PAH.

Materials and Methods

Catalysts were prepared using an impregnation technique at 80°C and high surface area silica was used as the support. The Pt loading was varied from 0.1 wt.% up to 5 wt.%. After drying, the calcination conditions were varied systematically and the influence of calcination temperature, time and atmosphere were investigated. Catalyst activity for naphthalene oxidation was evaluated using a laboratory micro reactor with on-line gas chromatography analysis. Reaction conditions employed a concentration of 100 vppm naphthalene in air with a GHSV of 45000 h⁻¹. Catalysts were characterized using a variety of techniques, including BET surface area measurement, pulsed CO chemisorption, scanning electron microscopy and X-ray photoelectron spectroscopy.

Results and Discussion

The metal loading greatly influenced the activity of the platinum catalysts. The most active catalyst contained 2.5% Pt, whilst increasing the loading to 5% decreased activity, as did reducing the loading to 1% and below (Figure 1). The platinum dispersion decreased as the metal loading was increased, and the dispersion for the most active 2.5% platinum catalyst was around 4%. As expected the average platinum particle size increased with increasing

loading, and data indicates that for best activity relatively large platinum particles (ca. 280 nm) were required, although increasing above this value did eventually result in a less active catalyst. The catalytic activity did not correlate directly with the number of surface platinum sites and turnover frequencies for the 2.5 and 5% platinum catalysts were considerably higher than more highly dispersed catalysts with lower metal loadings. Surface analysis by XPS showed that the more active catalysts had higher ratios of metallic to oxidised platinum and this seems to be an important factor influencing activity.

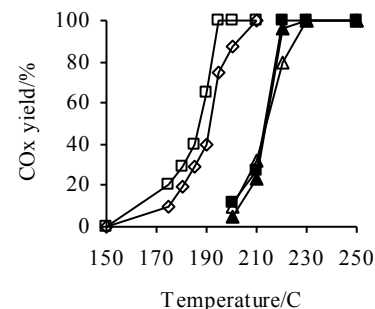


Figure 1. The effect of platinum loading on the catalytic oxidation of naphthalene to carbon dioxide (100 vppm Np in air, GHSV = 45000 h⁻¹): 0.1% Pt/SiO₂ (△), 0.5% Pt/SiO₂ (■), 1% Pt/SiO₂ (▲), 2.5% Pt/SiO₂ (□), 5% Pt/SiO₂ (◇).

The influence of calcination parameters on catalyst activity has also been systematically investigated. It was found that 2.5 wt% Pt calcined at 550°C for 12h in static air with a ramp rate of 5 °C min⁻¹ produced the most active catalyst, as 100 % yield to CO₂ was obtained below 200 °C. As indicated previously the surface ratio of metallic to oxidized platinum was an important factor controlling activity.

Significance

The influence of metal loading and calcination conditions have been investigated for a silica supported platinum catalyst for the total oxidation of the model PAH naphthalene, and the factors of metal dispersion, particle size and surface ratio of metallic/oxidised species have been identified as important factors.

References

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