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Low temperature VOC catalytic total oxidation by O₃

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PURPOSE OF THE ABSTRACT

Introduction

Elimination of volatile organic compounds (VOCs) contaminants of indoor air needs the use of new technologies with a low energy cost since VOC concentrations are low and the air flow to be treated is high. Catalytic oxidation with oxygen needs a temperature higher than 200°C to be efficient, leading to an important energy consumption to treat large air flow. This drawback can be circumvented by using oxidative species able to perform total oxidation reactions at room temperature. Amongst the various possibilities we have chosen to use ozone as oxidative species. Ozone is largely used as oxidant in waste water treatment and is easily produced by flowing air or pure oxygen through a non-thermal plasma reactor [1, 2]. Manganese oxide based catalysts are well-known to be effective for the oxidation of organic compounds by ozone.

In order to optimize the role of the catalyst and the gas-solid contact we have prepared a mesoporous-microporous TiO₂ with controlled porosity and high specific surface area on which the catalytically active phase (5 wt% of manganese oxid variously doped by palladium) was supported. In the present work the catalytic oxidation of EtOH (chosen as VOC model molecule) by few ppm of ozone at room temperature on our Mn-Pd/TiO₂ catalyst has shown that such system can be used as efficient indoor air treatment.

0.5%Pd-5%Mn/TiO₂ being the most efficient catalyst to decompose EtOH into CO₂.

Materials and Methods

The catalytic activity tests were carried out in a conventional flow reactor with atmospheric pressure at 40°C. VOC were generated by a Bronkhorst system with a specific flow and concentration. Then, the ozone was generated by a craft device by flowing pure oxygen through a non-thermal plasma reactor. For wet conditions, water was introduced into the gas stream using a saturator-type vessel containing water. The amount of water was fixed by controlling the temperature of the saturator using a cryostat. Moisture was set at 10% relative humidity at 25°C; corresponding to 3130 ppm of water in the final gas stream.

Ozone was analyzed online by an Ozone analyzer (Environment Type A. O3 42 M) based on the UV photometric

method at 254 nm [3] and VOC were analyzed by Varian GC (for heavier products) and Varian micro-GC (for light products, CO and CO₂).

Results and Discussion

The study of the catalytic performances in the elimination of a model VOC (ethanol) by oxidation by ozone of catalytic formulations under identical conditions allow us to select the most promising formulations on the basis of the best selectivity in carbon oxides. A carbon balance close to 100% and a rational use of ozone were obtained. We chose to use a quantity of ozone slightly greater than the stoichiometry of the total oxidation of ethanol in CO₂ and water, ie a molar ratio of 7 instead of 6 (stoichiometry = 6 molecules of ozone per molecule of ethanol completely mineralized).

The best catalyst systems selected in powder form are either palladium-doped manganese oxide supported on TiO₂ or cerium oxide (Mn-Pd/TiO₂ and Mn-Pd/CeO₂); a slight advantage being obtained with the Pd-Mn/TiO₂ system. The results are shown in Table (1); best composition being 0.5%Pd5%Mn/TiO₂.

A more specific study on these systems by varying the catalyst mass (the residence time) has shown that an increase in the residence time, in addition to the increase in the conversion, makes possible the reduction of the by-products formation, such as acetaldehyde or CO.

In order to better approach the actual conditions of use of the catalysts, we have succeeded in depositing the active phase Pd-Mn/TiO₂ on cordierite monoliths, previously coated with alumina. The obtained results confirm the good catalytic efficiency of our active phase which allow to obtain a complete conversion of the model VOC used (ethanol) with a selectivity of more than 98% in CO₂, without formation of acetaldehyde and with almost total elimination of injected ozone (less than 10 ppb of residual ozone). These very promising results allow to consider the development of a VOC treatment process at room temperature and with a reasonable reactor size (V ? 56 L for a flow rate of 1000 m³.h⁻¹).

Significance

The quality of the air we breathe and especially the air of confined spaces (indoor air of homes, offices, workshops, vehicles ...) is a major public health concern, but also has an impact on the notion of well-being and comfort.

Many pollutants can be present in the indoor air either from the outside (polluted environment, proximity to busy roads ...) or the room itself because of the presence and human activities or materials present in this environment (degassing of synthetic polymers or paints, for example).

The main objective of this work is to improve the air quality in public transports.

FIGURES

Catalysts	TiO ₂	5%Mn/TiO ₂	0.1Pd-5Mn/TiO ₂	0.5Pd-5Mn/TiO ₂	1Pd-5Mn/TiO ₂
Conv. EtOH (%)	73	92	94	93	86
S. CO ₂ (%)	61	85	87	91	77
S. CO (%)	6	6	6	6	9
S. acetaldehyde (%)	33	9	7	3	14
Conv. O ₃ (%)	12	61	75	70	76

FIGURE 1

Table 1. Results of elimination of ethanol with ozone on powder catalysts
Elimination of EtOH by O₃

FIGURE 2

KEYWORDS

VOCs | EtOH | O₃ | Catalysts

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