



Short Communication

New catalytic system for oxidation of isopropyl alcohol with thin film catalysts

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ABSTRACT

This paper describes a new catalytic system developed to study catalysts deposited as thin films on a metallic support. This device uses the electromagnetic induction for heating the metallic support. Therefore, it enables homogeneous sample heating, up to $400\text{ °C}\cdot\text{min}^{-1}$ with a good regulation and a very low thermal inertia allowing fast cooling and accurate hold on temperature plateau. Catalysts SnO_2 , 0.3 and 1 wt.% Pt on SnO_2 were deposited on stainless steel by an electrophoretic technique and evaluated in the abatement of isopropyl alcohol in air, a common model for VOCs.

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1. Introduction

Volatile organic compounds (VOCs) are considered as the first source of indoor environmental pollutants both in living and working places [1]. There is more and more evidence for the relationship between the amount of such pollution and the occurrence of some types of cancers [2]. It has long been recognized that isopropyl alcohol (IPA) used in the industry as solvent/cleaner [3] is a toxic compound even at low level which can induce cancer in the upper respiratory tract [4]. Its total catalytic oxidation to CO_2 and H_2O is a very attractive abatement method but can be much energy-consuming. This is also the case of thermal incineration which, in addition, may generate undesirable byproducts [5]. Filtration/adsorption can be used but needs to process the used sorbent [6]. Photocatalytic [7], biological [8] and non-thermal plasma [9] techniques are energy-saving but sometimes suffer from low reaction rates and are not very efficient to deal with heavily contaminated atmospheres. Therefore, the combination of catalytic combustion with an efficient heating technique such as electromagnetic induction has been proposed very recently [10] and can be considered as a good candidate because it puts together an excellent temperature regulation, fast abatement of accidental indoor air contamination and is quite energy-saving.

In the recent literature, many papers deal with the use of microwaves or cold plasma to improve performance in catalytic systems.

Anyway, the use of electromagnetic induction as a heating device in a catalytic process is scarcely reported. Main applications are the synthesis of carbon nanotubes [11], heating materials in a flow reactor [12] and fast pyrolysis process [13]. A couple of papers describe the possible use of induction heating in exhaust treatment [14], biomass reforming [15], copper flow reactor heating [16] and fundamental studies on microreactors [17].

Metal oxide and metal oxide-supported precious metal catalysts are used for low-temperature combustion of VOCs [18–20] and more especially Pt on Al_2O_3 or SnO_2 [21–23]. The purpose of this study is to show the performance of an innovative system using the electromagnetic induction for a fast and accurate heating. This device is a good tool for evaluation and ranking of thin film catalysts deposited on a metallic support. For illustration purpose, we focused on IPA oxidation on Pt/ SnO_2 and SnO_2 solids deposited on a stainless steel support.

2. Experimental

2.1. Materials

The catalyst carriers were austenitic stainless steel (AISI 304L SS) tubes 6.35 mm in o.d. (from Swagelok) cut at 50 mm in length. Catalyst support was SnO_2 (from Sigma-Aldrich, 99.9%, specific surface area $6.8\text{ m}^2\cdot\text{g}^{-1}$) which is known as a fair oxidation catalyst able to repeated oxygen adsorption/desorption cycles [24] and shows a good thermal stability. The catalysts prepared were 1 wt.% Pt/ SnO_2 and 0.3 wt.% Pt/ SnO_2 . A classical wet impregnation method was chosen [23], typically for the 1 wt.% Pt/ SnO_2 , 9.9 g of SnO_2 were added to a solution containing 0.1 g H_2PtCl_6 , $6\text{H}_2\text{O}$ (from Alfa Aesar) in 4 ml of water. The so-obtained

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solids were dried at 100 °C for 24 h, and then calcined for 5 h at 450 °C in air.

2.2. Catalyst film deposition

Electrophoretic deposition (EPD) was chosen because (a) it is a quite simple technique which allows a good control of the solid mass deposited, (b) it induces limited modifications of the initial solid and (c) it generally provides deposits with a good adherence to the support [25] that is favorable for the present aims. EPD has been applied according to the procedure described by Aruna and Rajam [26]. Stainless steel tube was pickled in 5 vol.% HF–15 vol.% HNO₃ aqueous solution at room temperature during 5 min in order to remove residual oxides and contaminants without a significant modification of the surface composition of the alloy [27]. The solid powder was placed with I₂ in a mixture of ethanol and acetone (25/75 in volume). The sample was positioned as the cathode and a sheet of stainless steel (AISI 304, 0.01 mm thick from Goodfellow) acted as the anode. Duration and voltage were selected according to the desired deposit final thickness. After deposition, the samples were fired at 450 °C in air during 30 min (heating rate: 1 °C·min⁻¹).

2.3. Catalytic performance test

A schematic drawing of the experimental setup is shown in Fig. 1. O₂/N₂ gas mixtures at atmospheric pressure were controlled by two mass flowmeters (range: 10–100 N mL·min⁻¹) and a third flowmeter (range: 1–10 N mL·min⁻¹) fixed the helium flow rate passing through a saturator/condenser for the control (acting both on He flow and condenser temperature) of the IPA concentration in the final stream. The condenser was mainly placed in melting ice corresponding to an IPA vapor pressure of 8.9 Torr [28]. For the present study, total flow rate was 100 N mL·min⁻¹ and the reactive mixture was 1000 ppm IPA/20% O₂/9.9% He/70% N₂. Helium was selected as IPA carrier instead of N₂ because it is a useful marker for the IPA containing steam. The mixture can be directed toward a reactor of annular type [29], as shown in Fig. 1 with a Pyrex® glass tube (10 mm inner dia.) for the outer part whereas the inner part was the 304L SS tube (with or without catalyst deposit) obstructed at both the extremities in order to force the reactant flow into the gap (1.8 mm) between glass and catalyst film. The SS tube was kept centered on the Pyrex® tube axis in a perfectly aligned position by a ceramic tube (mullite, from SCERAM, France). The volume of the reactive sheath (gap between the inner wall of the Pyrex tube and the outer wall of the SS tube) was about 2.3 cm³.

Corresponding *ghsv* at a total flow rate of 100 N mL·min⁻¹ was about $2.6 \times 10^3 \text{ h}^{-1}$ if we refer to the volume of the reactive sheath or about $1.4 \times 10^6 \text{ h}^{-1}$ if we refer to a standard catalyst deposit (i.e. 31 mg of SnO₂). A capillary K-type thermocouple (0.125 mm in dia. from Goodfellow) was guided into the ceramic tube and spot-welded onto the surface of the SS tube. This allows measuring the steel temperature accurately with a very low inertia. Gas analysis was performed by MS analysis using a Hiden HPR-20 QIC system. A switching valve allowed analyzing the composition of the gas mixtures at either the inlet or the outlet of the reactor. The heating device was a 400 kHz “Power Cube 32” from CEIA, interfaced to a “Power-C-V3” control system and fitted with a tailor made 5-turn induction coil outside the reactor [30]. Temperature regulation was achieved by a PID system (Eurotherm model 3504) reading sample temperature by means of the K-type thermocouple spot-welded on the stainless steel tube. The principal advantage of this technique is to heat only the stainless steel support.

3. Results and discussion

In order to evaluate the performance of the system in catalytic abatement of IPA in air, we compared the IPA conversion vs. temperature curves for the SS tube alone and then covered with SnO₂, 0.3% Pt/SnO₂ and 1% Pt/SnO₂. The MS allowed studying the evolutions of the molar fractions of He (m/e = 4), H₂O (m/e = 18), O₂ (m/e = 32), acetone (m/e = 43), CO₂ (m/e = 44) and IPA (m/e = 45) either at the inlet or at the outlet of the reactor. As a typical example of operating mode, Fig. 2 is relative to 0.3% Pt/SnO₂ on SS and shows the evolutions of the concentrations in IPA, acetone, CO₂ and the temperature of the SS tube vs. time on stream. Part A shows the corresponding signals in the absence of gas stream (background), Part B corresponds to the gas stream analyzed at the reactor inlet. Part C is recorded at the reactor outlet whereas in part D and E, the temperature was increased at 20 °C·min⁻¹. In part D a fraction of IPA desorbed from the catalyst and conversion to acetone started, then IPA complete oxidation proceeds (Part E). As widely reported in the literature, at lower temperatures IPA is first converted to acetone with almost 100% selectivity [31]. This is in agreement with the fact that acetone is, at least in its adsorbed form, an intermediate in IPA total oxidation [32]. For temperatures higher than 170–180 °C, acetone production rapidly falls to almost zero and only CO₂ and H₂O can be detected. A maximal conversion is reached at 275 °C and maintained. After 10 min at 400 °C, temperature is allowed to decrease freely and thus IPA concentration increases again to recover its initial value. Acetone was detected as the

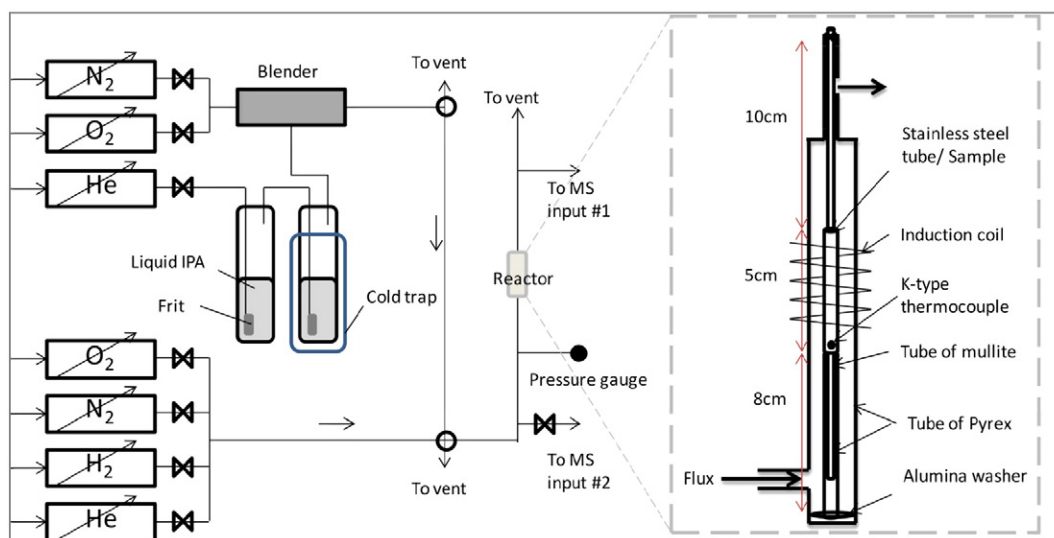


Fig. 1. Schematic drawing of the experimental setup.

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