

Complex Pt/Al₂O₃ materials for small catalytic systems

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Abstract

This paper reports on the preparation of catalytic materials composed of a porous metallic preform containing winding microchannels whose surface is coated by a complex catalytic film. Al₂O₃ is deposited inside the pores by means of wet impregnation in a first step, in order to increase the surface and to serve as an intermediate layer on which Pt nanoparticles are deposited in a second step. The latter deposition takes place by means of forced metalorganic chemical vapour infiltration (F-MOCVI). Pt(acac)₂, is used in appropriate low pressure operating conditions, to allow for a process with low thermal budget, compatible with the geometrical and physical characteristics of the porous substrate. The results are evaluated by means of BET, SIMS, SEM/EDX and FEG/SEM. The catalytic material is finally being tested following the oxidation of carbon monoxide, a model reaction with high industrial interest.

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1. Introduction

Catalysts have an enormous impact on the chemical industry and every-day applications, with the catalysis industry being the largest single economic activity today [1]. High catalytic activity induces either high productivity from relatively small reactors or mild operating conditions. Catalyst preparation is the secret to achieving the desired activity, selectivity and lifetime. The activity of solid catalysts is proportional to the active surface area per unit volume of catalyst. This consequently calls for small catalytic particles. The high reactivity of nanoparticles makes them good candidates for environmental catalysts for the treatment of automotive exhaust [2]. Preparation of large quantities of heterogeneous materials containing monodispersed nanoparticles is becoming one of the bottlenecks that hinder the development of industrial devices [3]. In most cases, the catalysts are being supported by a highly porous and thermally stable material, onto which the active species are being dispersed. The main purpose of

a support is to achieve an optimal dispersion of the catalytically active component(s) and to stabilize it/them against sintering. The most rapid procedure to prepare catalysts of such type is to start from commercially available preshaped support materials of the desired size, shape, porosity and mechanical properties. Applying the active element finely on the support, leads to the final material. Alumina is a widely used support body since it is a hard material with an excellent chemical stability [4]. Pt is an ideal catalytic active material. The use of metalorganic complexes as precursors of supported metal catalysts provides an important way to prepare highly dispersed and selective catalysts [5–9].

The present work deals with the preparation of a complex catalytic material for use in small catalytic systems. It consists of a commercially available metallic disk shaped preform of specific pore size and porosity. Alumina was deposited inside the pores via wet impregnation to increase the surface area and to serve as anchoring sites for the subsequent deposition of Pt particles. Platinum was deposited via forced metalorganic chemical vapour infiltration (F-MOCVI) in a hot-wall reactor, starting from platinum bis-acetylacetonate precursor, Pt(acac)₂. The obtained catalytic material is finally being tested following

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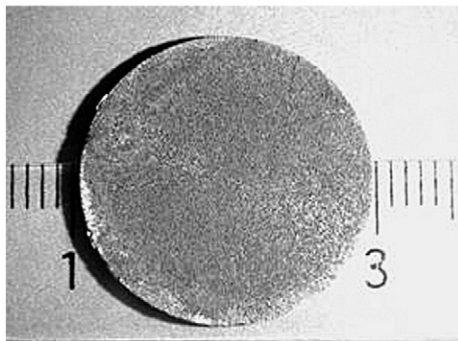


Fig. 1. Optical micrograph of the as-delivered porous preform.

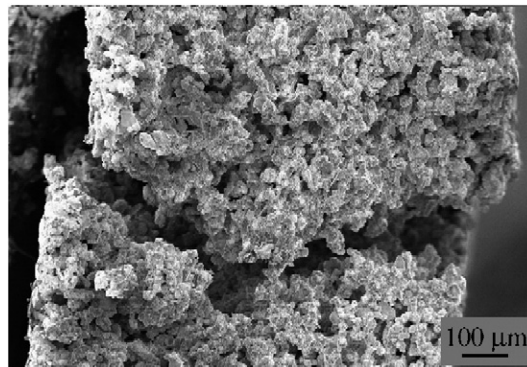


Fig. 3. SEM micrograph of the cross-section of stainless steel porous disk (100 μm pore size, 70% porosity) after wet impregnation of alumina film.

the oxidation of carbon monoxide, a model reaction with high industrial interest.

2. Experimental procedure

The substrates consist of 1 mm thick and 19 mm diameter porous disks shown in Fig. 1. They are commercialized by SOLEA SAS. Different copper and 316 stainless steel porous disks, whose pore size and porosity vary from 20 μm to 100 μm and 50% to 70% respectively, were used. Copper disks were used as substrate for the secondary ion mass spectrometry (SIMS) analysis because such analysis is difficult on the complex surface of the stainless steel porous substrates. Wherever the medium pore size is mentioned, it should be taken into consideration that micropores of significantly smaller size coexist.

2.1. Alumina deposition

In the first deposition step, the porous disks are immersed under vacuum in a 1 M boehmite (Disperal P2, Sasol, GmbH) dispersion solution with addition of 10% alcohol. They are left to dry in air overnight and subsequently heated at 450 °C for 4 h. The latter step allows the transformation of boehmite into alumina [10]. The weight of the deposited alumina was calculated by weight difference of the samples before and after the process. The overall surface of each porous disk and the efficiency of the alumina coating method were evaluated by Brunauer, Emmett and Teller (BET) measurements with a Micromeritics ASAP2010M apparatus. At this stage, the catalytic activity of alumina-coated disks was measured considering the reaction of CO oxidation and was found to be close to zero. Taking into account (i) the non-negligible catalytic activity of as received Cu disks; (ii) the fact that pressure

drop through the substrate during CVI remains unchanged and (iii) cross-section SEM observations, it is concluded that the substrate had been completely covered without significant pores blocking.

2.2. Platinum deposition

In the second deposition step, a hot-wall CVD reactor, depicted in Fig. 2 was used for the deposition of Pt particles inside the pores of the preforms. Pt(acac)₂ (Acros Organics), a solid, air-stable and cost effective precursor was heated at ca. 155 °C under N₂ flow and 50 Torr in the low temperature section of the reactor. The formed vapours were driven through a quartz tube at the end of which the porous disk was glued with a Ceramobond 596 paste-like glue. In this way the gases passed through the open porosity where Pt deposition was obtained by heating the disk at 390 °C. Flat samples of the same material as the porous disk (stainless steel or copper) were positioned shortly before and after the porous disk. Visual observation and X-ray photoelectron spectrometry (XPS) analyses on the flat samples were used as indication for the deposition of Pt inside the pores of the substrate. After the deposition step, the reactor was left to cool under N₂ flow; then the disk was torn off the quartz tube. Because alumina paste had inevitably infiltrated radially some of the pores, a weight measurement to establish the weight of the deposited Pt was not used. Instead, Pt concentration was measured by Inductively Coupled Plasma–Atomic Emission Spectrometry (ICP–AES).

The evaluation of the catalytic activity of the disk was realized through the CO oxidation reaction with 1% CO and 0.66% O₂ in Ar (total flow rate 110 cm³/min). A detailed

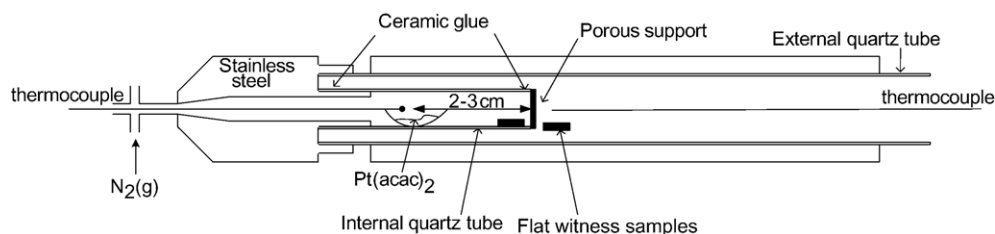


Fig. 2. Schematic representation of experimental set up for the F-MOCVI of Pt.

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