Latin American Applied Research

43:399-404 (2013)

REDUCTION OF ETHANOL EMISSIONS USING MANGANESE OXIDES SUPPORTED METALLIC MONOLITHS: A PILOT-SCALE PLANT STUDY.

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Abstract — Metallic monoliths were prepared by anodized aluminum-scale pilot plant, using sulfuric acid as electrolyte. Anodisation tests were conducted at different times and different temperatures of the electrolytic bath. Aluminum monolith anodized at 40°C for 30 min was selected for catalysts support. The monoliths were impregnated by two methods: i) impregnation in two stages using two Mn salts in an aqueous medium, and ii) impregnation using a KMnO₄ solution in acetone. The last monolith presented a higher and a more homogeneous deposit of manganese oxide layer. The catalytic activity was tested in the total oxidation reaction of ethanol in a pilot-scale reactor at 50 L min⁻¹, and ethanol concentrations of 900, 1800 and 3600 ppm. Monolith impregnated using acetone solution was more active that prepared in aqueous solution in all the experiments carried out.

Keywords — MnO_x; Pilot-scale plant; metallic monoliths; VOCs.

I. INTRODUCTION

Volatile organic compound (VOCs) emissions, produced from industrial processes, mobile sources, etc., are considered to be severe air pollutants. Catalytic oxidation is one of the most attractive routes for the elimination of VOC emissions. Noble metals (Pt, Pd) or transition metal based-oxides (Mn, Cu, Co, Fe, Ni) are commonly used as active phases (Alifanti *et al.*, 2005; Luo *et al.*, 2007; Peluso *et al.*, 2008a). Metal oxides have lower activity than noble metal catalysts but they are cheaper and have greater resistance to some poisons (Avila *et al.*, 2005). Among the transition metal oxides of interest for VOC oxidation, MnO_x-based materials exhibited great potential (Craciun *et al.*, 2003).

For environmental applications, high flow rates should be treated and a low-pressure drop is required. For that reason, the catalytic system most widely used for catalytic VOC oxidation is the monolithic reactor.

Metallic monoliths present higher mechanical resistance and thermal conductivity, thinner walls and lower pressure drop than those in ceramics. Additionally, metallic substrates are produced easily and in different and complicated forms adapted to a wide variety of uses (Twing and Webster, 1998).

Aluminum monoliths are generally produced by anodic oxidation, in which a porous alumina layer is obtained on the aluminum surface. The alumina texture can be controlled modifying anodisation parameters such as the current density and the temperature (Burgos et al., 2002). These authors deposited Pt over 6 cm³ volume Al₂O₃/Al monoliths to study the combustion of VOCs. Barrio et al. (2005) studied the impregnation of Al₂O₃/Al monoliths at lab scale with KMnO₄ using acetone and water as solvent. They found that pore sealing of alumina occurs in water solutions. However, this is prevented by the use of acetone.

A pilot-scale plant is used to test catalyst at operating conditions similar to the ones existing in to real gas turbine burners. Some works at pilot-scale plant are found using ceramic monoliths (Alvarez *et al.*, 2002; Requies *et al.*, 2008) but no metallic ones.

The aim of this work is to study the anodisation parameters to obtain metallic monoliths at higher dimensions than are usually found in lab scales, in order to prepare active catalysts for VOC oxidation with nonoble additives such as MnO_x . The prepared monoliths were tested in a pilot-scale plant using catalytic oxidation of ethanol as test reaction.

II. METHODS

A. Sample Preparation

Cylindrical monolith supports were prepared by anodisation of 120 cm x 5 cm aluminum foils of 100 µm thick. Each foil was first washed with water and degreased with acetone. Anodisation was carried out in a longitudinal glass tank with 2.55M sulphuric acid, which is placed in second glass tank contained water. All the experiments were carried out with a current density of 4 A dm⁻² and during 30 min. The electrolyte temperature used was 20, 30 and 40 °C. The aluminum sheet was fixed to the anode (working anode) between two aluminum foils connected to the cathode (counterelectrodes). The distance between electrodes was 5 cm. Agitation was provided by a pump which recirculates the electrolyte inside the bath. Temperature control of +/- 0.5°C was obtained with a refrigerating system and an electrical heater with an On-Off temperature controller. After anodising, aluminum foils were removed from the electrolytic bath, thoroughly washed with water to remove the acid, and then dried at 100 °C for 1h. Flat and corrugated foils were anodized separately and then both sheets were rolled together to form monoliths. Once the monoliths were prepared, they