

Titania-supported Pt and Pt–Pd nanoparticle catalysts for the oxidation of sulfur dioxide

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Abstract

Several types of titania (anatase) were used as supports for pure platinum and Pt–Pd bimetallic alloy catalysts. The preparation methods, normal wet impregnation technique and flame aerosol synthesis, obtained metal loadings of 2% by weight. The prepared catalysts were tested for SO₂ oxidation activity at atmospheric pressure in the temperature range 250–600 °C. The SO₂ to SO₃ conversion efficiency of the Pt–Pd alloy was significantly higher than that of the individual metals. The effects of the preparation method and the titania type used on the properties and activity of the resulting catalyst are discussed.

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1. Introduction

The catalytic oxidation of sulfur oxides followed by absorption are key processes in industrial waste treatment, affecting air pollution worldwide. Measures from the EU and EPA to decrease the release of sulfur oxides with the aim of achieving a cleaner environment have resulted in increased research efforts for developing more efficient catalysts and catalytic units [1]. Platinum-based catalysts are widely used in conventional exhaust gas cleanup and have proven stable and tolerant to the typical poisons of the combustion engine exhaust. Although much progress has been made on the well-known vanadium oxide systems used in industry [2–4], it is obvious that systems based on the platinum group metals may be more suitable for higher activity at lower temperatures. Until World War II, platinum dominated as catalyst in the sulfuric acid production industries, oxidizing sulfur dioxide (the key step in sulfuric acid

production and flue gas desulfurization). This type of catalyst supported on oxide carriers has advantages over other catalysts used for the same catalytic process, because they can be regenerated in situ [5–10]. Synthesis of metal alloys is a common practice to improve the catalytic characteristics of monometallic catalysts [11–15]. Palladium, a platinum group metal, is thought to have relevant chemical properties and catalytic activity. The bimetallic alloy of the two metals has been tested as a catalyst for the SO₂ oxidation reaction, and the best conditions for improved catalytic performance have been investigated [16,17]. Such alloys have been tested in electrochemical fuel cells and in the exhaust gas treatment in car catalysts [18].

The use of noble metal-based catalysts is limited by their scarcity and high cost. Hence, improving their catalytic performance and decreasing their cost is important. The active phase in the final product should be highly dispersed and consist of nanoparticles. Achieving this depends on a number of factors, including the surface characteristics of the support, the nature of the metal precursors, the solvent, the rate of precursor salt formation, and more. The most commonly used support for platinum catalysts is silica, although tests with titania have shown that titania is a better support, because it is cheap, has

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better surface and electronic properties, is more acidic (very important for the specific reaction), and is resistant to steam and poisons in the gas mixture [19,20]. Although platinum-based catalysts supported on titania have been studied previously, improving their catalytic activity remains an important field for exploration. Catalytic properties depend on thermal treatment, doping, synthesis route, and surface characteristics of the carrier.

In the present work, we have investigated the effect of surface and morphological characteristics of several types of titania (anatase) as supports for platinum-based catalysts. We used two different preparation methods. The first method, which is based on impregnation, used acetylacetonate salts of the active metals as precursors and DMSO as the solvent. The activity of the bimetallic catalyst samples for the SO_2 oxidation reaction was compared with that of the respective pure metals. As an alternative method, the Pt/TiO₂ catalyst was prepared by a one-step flame synthesis method [21,22]. The following samples were investigated and compared for catalytic activity: wet impregnation of two commercially available titania samples, wet impregnation of flame-produced titania, and one-step synthesis of the supported noble metal in the flame with the same operating conditions as for pure flame-produced titania.

2. Experimental

2.1. Chemicals and gases

Two of the titania (anatase) samples used for catalyst preparation were commercial products (Alfa Aesar titanium (IV) oxide, anatase 99.9% and Millennium Co. TIONA-G5, anatase). Using the aerosol synthesis method, we prepared two titania (anatase) samples (T1 and T2, with specific surface areas of 57 and 98 m²/g, respectively). For the formation of titania in flame, the Ti-isopropoxide complex was used to ensure high vapor pressure in the saturator.

The acetylacetonate salts of platinum and palladium (Pt(acac)₂ and Pd(acac)₂; Aldrich) were used as synthesis precursors for impregnation. Dimethyl-sulfoxide (DMSO), extra pure for chromatographic purposes (Aldrich) was used as a solvent. A catalyst sample, Pt/TiO₂, was prepared using the flame aerosol method by gas-phase decomposition of the Ti- and Pt-precursors [23].

Commercial gases SO_2 (>99.99%), O_2 (99.8% + 0.2% N_2 and Ar), and N_2 (<40 ppm O_2 + H_2O) were used for the catalytic activity tests. All gases were dried through P_2O_5 columns before entering the synthesis gas cylinder flask.

2.2. Catalyst preparation

2.2.1. Wet impregnation

In all cases, the loading of the active phase was 2 wt%, as confirmed by TEM/EDAX and by chemical analysis using atomic absorption spectroscopy (AAS). Several titania anatase samples were chosen as support materials and were impregnated with Pt(acac)₂ solutions using the wet impregnation technique and DMSO as solvent. For the preparation of the bimetal-

lic alloy samples, titania was coimpregnated in a solution with both the platinum and the palladium acetylacetonate salts. The desired content of the second metal was 20% on the final alloy composition. The suspension was stirred vigorously and maintained at 90 °C for 3 days to achieve equilibration, then the solvent was evaporated in a rotary evaporator, and the solid material was dried overnight at 110 °C. The sample was then reduced and calcinated at 420 °C, with the temperature slowly increased from room temperature to 420 °C over 6 h, then maintained at 420 °C for 3 h and finally decreased slowly to room temperature over 8 h.

2.2.2. Flame synthesis of pure TiO₂ support and Pt/TiO₂ catalyst

The setup for the aerosol synthesis of the pure titania (T1 and T2-TiO₂ preparations) and the catalyst sample of platinum supported on titania (T3-TiO₂) is shown in Fig. 1. The premixed flame with a central precursor jet is the same as that used by Hansen et al. [24]. Premixed methane and air react in a flat zone just above the arrestor plate. The arrestor is circular, 40 mm in diameter, and 5 mm thick and gives an even distribution of the methane/air mixture. The jet outlet in the center of the arrestor has a 1.27-mm inner diameter. The jet consists of nitrogen mixed with vapors of the precursors, i.e., Ti-isopropoxide, when only the titania was prepared, and Ti-isopropoxide along with Pt(acac)₂ in the case of direct, one-step catalyst preparation. The saturator for the solid Pt(acac)₂ in the

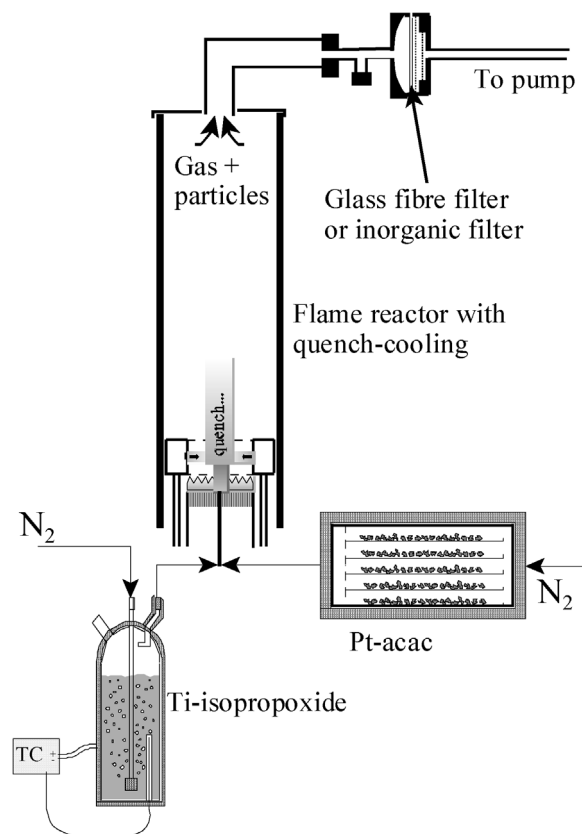


Fig. 1. Experimental setup for flame-aerosol synthesis of TiO₂ and Pt-doped TiO₂ particles (reproduced from Ref. [23]).

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