

Decomposition of ozone on Ag/SiO₂ catalyst for abatement of waste gases emissions

A. Naydenov^{a,*}, P. Konova^a, Pen. Nikolov^a, F. Klingstedt^b, N. Kumar^b,
D. Kovacheva^a, P. Stefanov^a, R. Stoyanova^a, D. Mehandjiev^a

^a*Institute of General and Inorganic Chemistry, Bulgarian Academy of Sciences, "Acad. G. Bonchev" Street, Building 11, 1113 Sofia, Bulgaria*

^b*Laboratory of Industrial Chemistry, Process Chemistry Centre, Åbo Akademi University, Biskopsgatan 8, 20500 Åbo/Turku, Finland*

Received 25 July 2007; received in revised form 5 November 2007; accepted 8 November 2007

Available online 27 December 2007

Abstract

A silica-supported Ag system made by the incipient wetness impregnation method was investigated in the reaction of heterogeneous catalytic decomposition of ozone. It was established that the catalytic ozone decomposition on Ag/SiO₂ proceeded in the temperature interval –40 °C to 25 °C as a first order reaction with activation energy of 65 kJ/mol (pre-exponential factor $5.0 \times 10^{14} \text{ s}^{-1}$). Based on the results from the instrumental methods (SEM, XRD, XPS, EPR, TPD) it can be concluded that in presence of ozone the silver is oxidized to a complicated mixture of Ag₂O₃ and AgO. Due to the high activity and stability of the Ag/SiO₂ catalyst, it is promising for neutralization of waste gases containing ozone. © 2007 Elsevier B.V. All rights reserved.

Keywords: Ozone decomposition; Ag/SiO₂; Waste gases; Silver oxide; Silica

1. Introduction

Ozone is known as a strong oxidizing agent for waste and drinking water treatment, sterilization and deodorization [1–4]. The exit gases from such processes contain residual ozone and due to the fact that the ozone itself is very toxic [1], an additional air pollution problem must be solved. The most effective method for neutralization of waste gases containing ozone is the heterogeneous catalytic decomposition [2–4]. The disadvantage of this method is the deactivation of the catalysts due to presence of nitrogen oxides formed during ozone synthesis [5], the presence of water vapour and carbon dioxide [6,7]. Therefore, it is of importance to develop a new reliable and active catalyst for ozone decomposition. The behavior of silver catalysts in the reaction of ozone decomposition was a subject of several investigations [8–11]. Ag-containing catalysts show highest activity when compared with the oxides of Co, Ni, Fe, Mn, Ce, Cu, Pb, Bi, Sn, Mo, V and Si [8]. During the catalytic decomposition of ozone, a highly reactive oxygen

species is produced and it is able to oxidize completely the toxic compounds at room temperatures [12–14].

The aim of the present paper is to investigate the kinetics and the mechanism of ozone decomposition on silver-modified SiO₂ with a view to its further application to the neutralization of waste gases containing residual ozone and possible use for decontamination of polluted gases from toxic volatile organic compounds (VOCs) by applying of ozone-catalytic method [12].

2. Experimental

The modification of SiO₂ (delivered commercially from Merck company) with Ag was carried out by the incipient wetness impregnation method. The reaction proceeded at ambient temperature for 24 h in a rotator evaporator device using an aqueous solution of silver nitrate. After impregnation, the sample was dried at 110 °C and calcined at 500 °C. The specific surface area of the supported catalyst was determined by low-temperature nitrogen adsorption using Sorptomatic 1900 (Carlo Erba Instruments). The BET equation was used for calculation of the surface area. The morphology of the samples was investigated by a scanning electron microscope (Cambridge Leica 360). The metal content of the sample was

* Corresponding author. Tel.: +359 2 979 39 09; fax: +359 2 870 50 24.

E-mail address: naydenov@svr.igic.bas.bg (A. Naydenov).

determined by inductively coupled plasma atomic emission spectrometry (ICP-AES) with a Jobin Yvon 38 (France). Phase composition and particle sizes of the samples were studied by X-ray powder diffraction. XRD spectra were recorded at room temperature using a Bruker D8 Advance powder diffractometer with Cu K α radiation and SolX detector. Data were collected in the 2θ range from 10° to 80° with a step 0.04° and counting time of 2 s/step. The XPS studies were performed in a VG Escalab II electron spectrometer using AlK α radiation with energy of 1486.6 eV. The residual gas pressure in the analysis chamber was 10^{-7} Pa. Binding energy values (BE ± 0.2 eV) were referenced to the C 1s line at 285.0 eV. Oxygen TPD – spectra were recorded by using a fast response Teledyne (3000 P) O $_2$ – gas analyser in argon (99.999%) flow (9.0 l/h), the catalyst sample being 0.2 g. The EPR spectra were recorded as a first derivative of the absorption signal of an ERS-220/Q (ex-GDR) spectrometer within the temperature range of -195 to 130 °C. The g -factors were determined with respect to a Mn $^{2+}$ /ZnS standard. The catalytic activity of the sample in the reaction of ozone decomposition was investigated in a plug flow reactor system permitting isothermal operation. The reactor was made of glass and had an inner diameter of 4.1 mm. The catalyst particle sizes were varying: 0.1–0.2 mm and 0.5–0.6 mm. The volume rate was $120,000$ h $^{-1}$. Ozone was synthesized in a flow of oxygen (99.7%) dried with silica gel using an ozone generator with silent discharge and coaxial electrodes. The inlet concentration of ozone was varying from 5 to 15 g/m 3 (0.1 – 0.3 mol/m 3). Ozone concentration was measured by an Ozomat GM (Anseros, Germany) ozone analyzer with a resolution of ± 0.1 g/m 3 . The reaction temperature was -35 to 25 °C and was maintained with an accuracy of ± 0.2 °C.

3. Results and discussion

The metal content was measured to be 4.85 wt.% and the surface area (BET), 342 m 2 /g. The pore size distribution curve can be decomposed to four different peaks with maxima at 1.6, 2.3, 2.9 and 3.7 nm, i.e. the main part of the pores are in a narrow range from 1.5 to 5.5 nm. The scanning electron micrograph (SEM) of the Ag-SiO $_2$ catalyst reveals a uniform particle size distribution of Ag over the silica support, the average particle size of Ag clusters ranging from 0.5 to 1 μ m.

Fig. 1, curve a, presents the temperature-conversion dependence of 5 wt.% Ag/SiO $_2$ catalyst in the temperature interval -35 to 25 °C, measured after the reaching of steady state conditions. As can be seen, the conversion is 50% at -13 °C and reaches almost 100% at room temperature. The lowering of the conversion observed with the higher particle diameter (Fig. 1, curve b, $d_p = 0.5$ – 0.6 mm) reveals that there is a significant increase of the effect of the internal diffusion.

A reactor model was used to obtain the values of the integrated reaction rates and, taking into account the diffusion effects, to establish the kinetics parameters [15]. On the basis of the prediction for the effectiveness factor and fitting with the experimental data, the following values for the kinetic parameters have been obtained in the temperature interval from -35 to 25 °C: $E_a = 65$ kJ/mol; $k_0 = 5.0 \times 10^{14}$ s $^{-1}$.

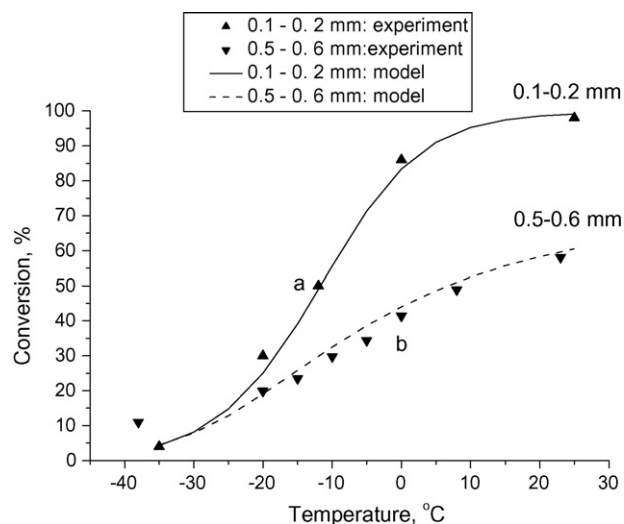


Fig. 1. Conversion–temperature dependences for the reaction of ozone decomposition over an 5 wt.% Ag/SiO $_2$ catalyst, $C_{O_3, \text{inlet}} = 0.25$ mol/m 3 (12 g/m 3), $GHSV = 120,000$ h $^{-1}$.

The X-ray powder diffraction patterns of fresh, ozonated and subjected to TPD samples of 5 wt.% Ag/SiO $_2$ are presented in Fig. 2. As can be seen, in the fresh sample silver is present as a metal phase (Ag 0). The major Ag diffraction peaks correspond

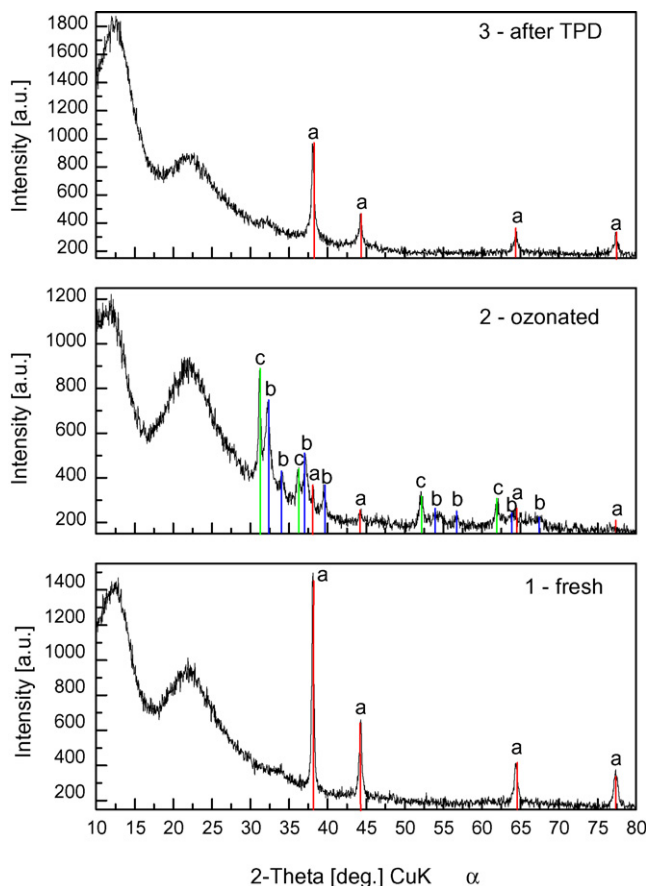


Fig. 2. X-ray powder diffraction patterns of fresh, ozonated and after TPD samples of 5 wt.% Ag-SiO $_2$: (a) metallic silver, (b) monoclinic AgO, (c) cubic Ag $_2$ O $_3$.

Download English Version:

<https://daneshyari.com/en/article/57557>

Download Persian Version:

<https://daneshyari.com/article/57557>

[Daneshyari.com](https://daneshyari.com)