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Low-temperature oxidation of volatile organic compounds by ozone on Mn-promoted W/Al_2O_3

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Catalyst preparation

A sample containing 8 wt.% W was prepared by incipient wetness impregnation ɣ-Al_2O_3 ("Sasol", S_{BET} = 76 m^2 g^{-1}) with an aqueous solution of $(NH_4)_6W_{12}O_{39}xH_2O$, (Aldrich). A 1% Mn/Al_2O_3 sample was prepared by impregnation with an aqueous solution of $Mn(NO_3)_2 \bullet 4H_2O$ (≥97%, Sigma Aldrich, USA). A 1%Mn8%W/Al_2O_3 sample was prepared by additional impregnation of a monolayer 8%W/Al_2O_3 with an aqueous solution of $Mn(NO_3)_2 \bullet 4H_2O$ (≥97%, Sigma Aldrich, USA). After impregnation, the samples were dried at room temperature and then calcined for 2 hours at 550°C.

Catalyst characterization

Temperature-programmed reduction (H_2-TPR). The catalysts were studied by temperature-programmed reduction by hydrogen on an USGA-101 semiautomatic flow setup (Unisit LLC, Russia) equipped with a catharometer calibrated against the reduction of NiO (99.99%, Sigma—Aldrich, USA). An U-shaped quartz reactor with a chromel—alumel thermocouple was loaded with a weighed sample of the catalyst (0.3 g). The sample was preliminarily purged with argon (99.999%, Linde Gas Rus PLC, Russia; flow rate 30 mL min^{-1}) on heating from room temperature to 150°C at a rate of 10°C min^{-1} and then allowed to stay at a specified temperature for 1 h. Then, the sample was cooled in Ar flow to 50°C and kept at a preset temperature in a 5%H_2/Ar stream (Linde Gas Rus PLC, Russia) (30 mL min^{-1}) until a stable baseline emerged (~20-30 min). The catalyst sample was reduced in a 5%H_2/Ar flow (30 mL min^{-1}) on heating to 650°C at a rate of 10°C min^{-1}. To remove water vapor formed during the reduction reaction, a trap cooled to a temperature of –20°C with a Peltier element was placed between the reactor and the catharometer.

Scanning electron microscopy (SEM-EDS) The surface morphology of catalysts was characterized using SU8000 (Hitachi, Tokyo, Japan) field-emission scanning electron microscope (FE-SEM). Before measurements the samples were mounted on a 25 mm aluminum specimen stub and fixed by conductive graphite adhesive tape. Qualitative and quantitative analysis of catalyst elements were carried out using energy dispersive X-ray spectroscopy (EDS) on Oxford Instruments X-max 80 EDS system with 15 kV accelerating voltage.

Catalytic tests

Catalytic performance of samples in ozone decomposition and ozone catalytic oxidation of n-C_4H_{10} (OZCO) was studied using fixed-bed flow quartz reactor (id=10mm) operating at atmospheric pressure (Fig S1). The composition of the feed gas contains 100 ppm n-C_4H_{10}, 1600 ppm O_3, 10 vol. % O_2, balanced with N_2. Ozone was obtained using a Medozone YOTA-60-01 ozone generator (Medozone, Russia). Total flow rate was fixed at 750 mL/min (GHSV = 150 000 h^{-1}). The temperature was ranged from RT to 350°C at a heating rate of 5°C/min. Ozone content at the inlet and outlet of the reactor was measured by ozone analyzer Medozone 254/5 (Medozone, Russia). Reaction products were analyzed by FTIR gas analyzer “Gasmet’’ (Temet Instruments DX-4000). The conversion of n-C_4H_{10}, was calculated using the follow equation:

$$X = \frac{C_{in} - C_{out}}{C_{in}} \times 100\%$$

where subscripts *in* and *out* are the inlet and outlet concentrations, respectively.

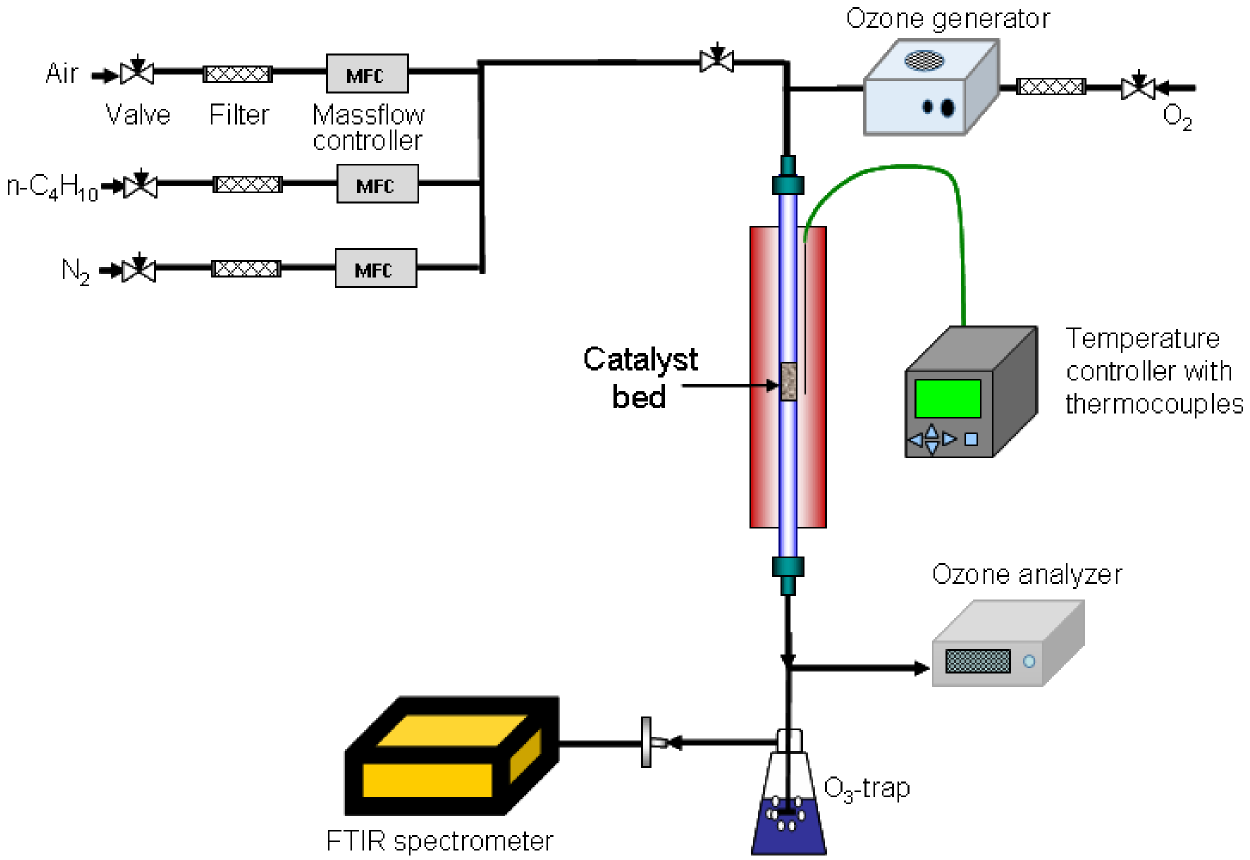

Figure S1. Experimental setup

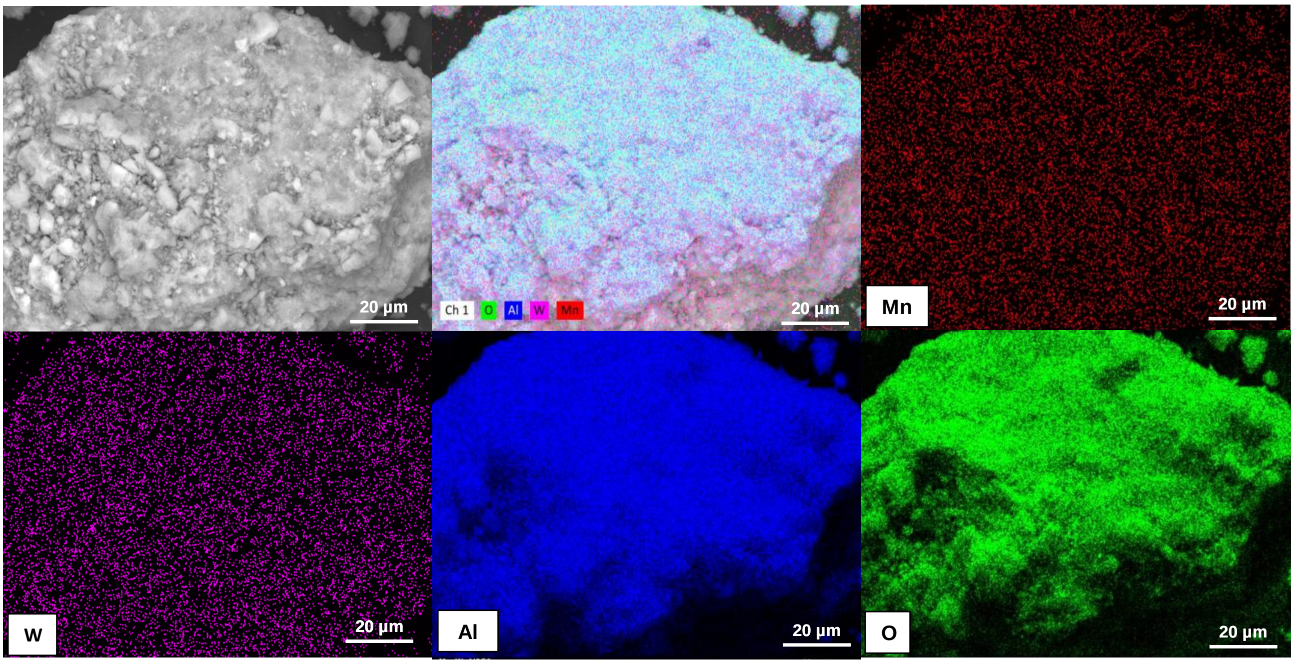

Figure S2. Surface morphology and EDS of 1%Mn8%W/Al_2O_3