



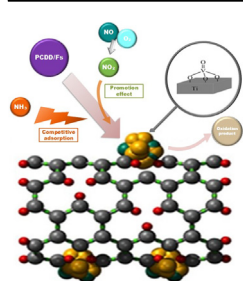
## Short communication

Catalytic decomposition of gaseous PCDD/Fs over  $V_2O_5/TiO_2$ -CNTs catalyst: Effect of NO and  $NH_3$  additionQiulin Wang<sup>a, b</sup>, Pao Chang Hung<sup>c</sup>, Shengyong Lu<sup>b, \*</sup>, Moo Been Chang<sup>c</sup><sup>a</sup> School of Energy and Power Engineering, University of Shanghai for Science and Technology, Shanghai 200093, China<sup>b</sup> State Key Laboratory of Clean Energy Utilization, Institute for Thermal Power Engineering, Zhejiang University, Hangzhou 310027, China<sup>c</sup> Graduate Institute of Environmental Engineering, National Central University, No. 300, Jungdad Road, Chungli 320, Taiwan

## HIGHLIGHTS

- Catalytic decomposition of gaseous PCDD/Fs over  $V_2O_5/TiO_2$ -CNTs composite catalysts.
- The removal efficiency of PCDD/Fs reaches 99.9% at 150 °C while adsorption is the main reason.
- $NH_3$  restricts the adsorption of PCDD/Fs on catalyst, hence, reduces the removal efficiency.
- The positive effect of NO is due to the oxidation of NO to  $NO_2$  that speeds up the catalyst reoxidation.
- The activation energies for OCDD/F via catalysis are evidently reduced in the addition of NO and  $NH_3$ .

## GRAPHICAL ABSTRACT



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## ABSTRACT

There is a strong need for a control technology that simultaneously achieving the abatement of PCDD/Fs (polychlorinated dibenzo-p-dioxins and polychlorinated dibenzofurans) and nitrogen oxides ( $NO_x$ ) emissions in waste incineration industry.  $TiO_2$  and carbon nanotubes (CNTs) were used as composite carriers to support vanadium oxide as an innovative catalyst to simultaneously control PCDD/Fs and  $NO_x$  emissions. The removal efficiencies (RE) of PCDD/Fs by  $V_2O_5/TiO_2$ -CNTs catalyst under a space velocity (SV) of 20,000  $h^{-1}$  reaches 99.9% at 150 °C and adsorption is supposed to be the main mechanism at this temperature. The influence of  $NO-NH_3$  reaction on PCDD/Fs catalytic reaction is investigated. The kinetics analysis exhibits that the addition of NO and  $NH_3$  reduces the activation energies for OCDD (octachlorodibenzo-p-dioxin) and OCDF (octachlorodibenzofuran) decomposition to 3.6 kJ/mol and 5.4 kJ/mol respectively.

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## 1. Introduction

The active temperatures of conventional selective catalytic reduction (SCR) catalyst (e.g.  $V_2O_5/TiO_2$ ) for PCDD/Fs destruction (200–300 °C) (Chang et al., 2009; Ji et al., 2013, 2014; Yang et al.,

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2008) and deNO<sub>x</sub> via NH<sub>3</sub>-SCR (300–400 °C) (Roy et al., 2009; Skalska et al., 2010) are different. To accomplish the combining catalytic removal of PCDD/Fs and NO<sub>x</sub> generated during waste incineration, it is essential to develop inexpensive novel catalysts with high destruction efficiency at lower temperature (<200 °C) (Dvořák et al., 2010; Liu et al., 2015).

Carbon nanotubes (CNTs) has been proved to be the excellent sorbent for PCDD/Fs (Long and Yang, 2001a) and NO<sub>x</sub> (Long and Yang, 2001b). Very recent studies have claimed that introducing CNTs to alternative SCR catalyst can effectively enhance the low-temperature activity of several composite catalysts and facilitate the elimination of chlorobenzene (Fan et al., 2011; Nie et al., 2011) and NO<sub>x</sub> (Li et al., 2011). This study investigates the catalytic performance of home-made V<sub>2</sub>O<sub>5</sub>/TiO<sub>2</sub>-CNTs catalyst on gas-phase PCDD/Fs treatment for the first time. Besides, the effects of NO and NH<sub>3</sub> on PCDD/Fs catalytic destruction and adsorption are also studied. Moreover, reaction rate constant and apparent activation energy of OCDD and OCDF decomposition on V<sub>2</sub>O<sub>5</sub>/TiO<sub>2</sub>-CNTs catalyst are calculated before and after NO and NH<sub>3</sub> addition.

## 2. Material and methods

### 2.1. Catalyst preparation and characterization

Purchased multi-walled carbon nanotubes (MWCNTs, purity > 95%, BOYU GAOKE Co., Beijing) with outer-diameter of 20–30 nm, a length of 10–30 μm and specific area of 154 m<sup>2</sup>/g were used without further treatment. Previous study has investigated the thermogravimetric analysis of CNTs and found that the oxidation of CNTs occurs at 650 °C (Misra et al., 2006). The stability of CNTs within the temperature of 150–320 °C can be guaranteed as a consequence.

The V<sub>2</sub>O<sub>5</sub>/TiO<sub>2</sub>-CNTs catalyst was prepared using sol-gel method (Wang et al., 2016) and was then characterized using N<sub>2</sub>-physorption (automated Brunauer-Emmet-Teller (BET)) and pore

analyzer (TRISTAR 3020, Mike Instrument Co., USA): BET specific surface area 140 m<sup>2</sup>/g, total pore volume 0.25 cm<sup>3</sup>/g, and average pore diameter 5.9 nm. The X-ray diffraction (Philips Model XD-98) analysis indicates that the TiO<sub>2</sub> phase in V<sub>2</sub>O<sub>5</sub>/TiO<sub>2</sub>-CNTs catalyst is anatase type, and VO<sub>x</sub> is either in amorphous or microcrystalline form. The surface morphology of the CNTs-containing catalyst is demonstrated in Fig. 1. Our previous study (Lu et al., 2014) has revealed that VO<sub>x</sub> connects to TiO<sub>2</sub> establishing V=O and V–O–Ti bonds which are closely related with the catalyst activity of the supported vanadia catalyst. In V<sub>2</sub>O<sub>5</sub>/TiO<sub>2</sub>-CNTs catalyst, the formed VO<sub>x</sub>/TiO<sub>2</sub> particles, independent VO<sub>x</sub> or TiO<sub>2</sub> particles are preferred to insert into the surface vacancies of CNTs (Rodriguez-Manzo et al., 2010; Song and Jiang, 2012), which enables the well dispersion of active components due to the larger surface area of CNTs.

### 2.2. Reactivity measurement and analysis

The catalyst test system mainly relies on a stable dioxin generator (Yang et al., 2008), a catalytic reactor and a tail gas adsorption part (Fig. 2). The inlet concentration of PCDD/Fs in gas phase (PCDD/Fs<sub>inlet</sub>) is detected as 12.2 ng TEQ/Nm<sup>3</sup> (mean value of seven runs, with a standard deviation of 0.3). A gas mixture of 11 vol% O<sub>2</sub> and N<sub>2</sub> (balance) was used as carrier gas and the gas hourly space velocity is fixed at 20,000 h<sup>-1</sup>. Before sampling, the test system maintains parameter stability for 60 min in advance. Then the effluent vapor-phase PCDD/Fs of the reactor was continuously collected by XAD-2 resin for the second 60 min (PCDD/Fs<sub>outlet</sub>). After each experimental run, the glass tube and the catalyst were then rinsed and Soxhlet extracted with toluene for the determination of residual PCDD/Fs on the surface of the catalyst and reactor (PCDD/Fs<sub>on-catalyst</sub>). PCDD/Fs pretreatment and analysis procedure were referred to that described by Wang et al. (2009). In our experiments, the surrogate recoveries range from 83% to 110% and meet the quality control of USEPA Method 23 (all surrogate recoveries shall be between 70% and 130%).

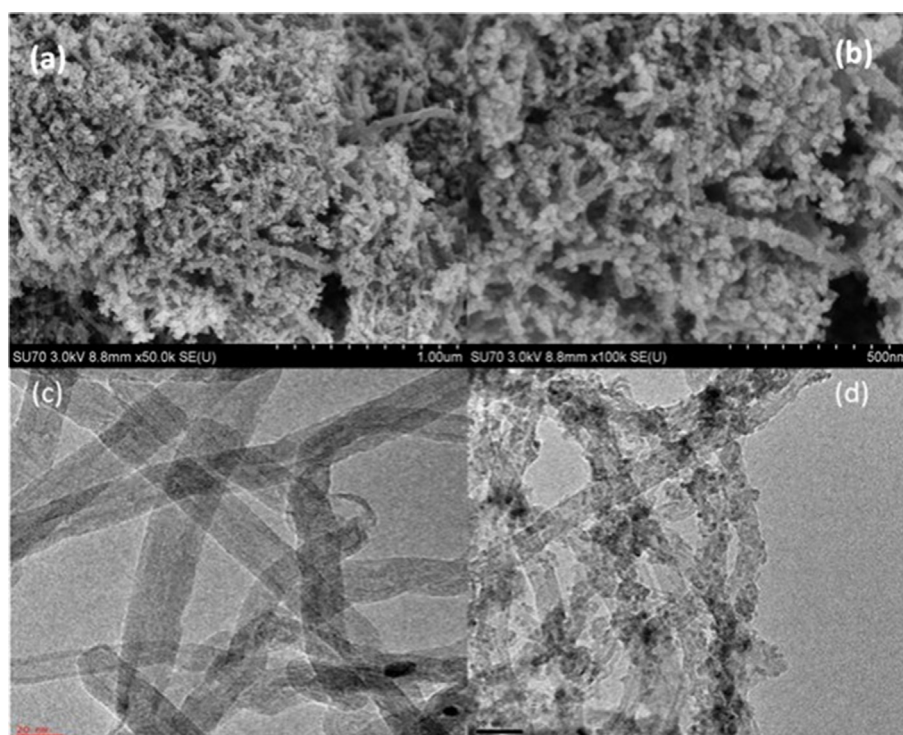


Fig. 1. SEM images of (a) (b) V<sub>2</sub>O<sub>5</sub>/TiO<sub>2</sub>-CNTs catalyst; TEM images of (c) pure CNTs and (d) V<sub>2</sub>O<sub>5</sub>/TiO<sub>2</sub>-CNTs catalyst.

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