

The kinetic of the catalytic decomposition of methyl isobutyl ketone over a Pt/ γ -Al₂O₃ catalyst

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Abstract

Environmental catalysis also can refer to catalytic technologies for reducing emission of environmentally unacceptable compounds. Catalytic decomposition also is one of the cost-effective technologies to solve the troublesome volatile organic compounds. This study treated methyl isobutyl ketone (MIBK) by a commercial catalyst, Pt/ γ -Al₂O₃, in an isothermal fixed bed differential reactor. The effects of O₂ and MIBK content in carrier gas on the catalysis's reaction rate are also observed. Three kinetic models, i.e. the Mars and van Krevelen model, Langmuir–Hinshelwood model and power-rate law were applied to best fit the experimental results. The results indicate that the kinetic behavior of MIBK oxidation with catalysis can be accounted for by using the rate expression of the Mars and van Krevelen model and Langmuir–Hinshelwood model. Kinetic parameters are also determined on the basis of the differential reactor data. The experimental results are compared with those of the model predicted.

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1. Introduction

Volatile organic compounds (VOCs) are defined as the organic compounds that have high vapor pressure and are easily vaporized at ambient temperature and pressure. Most hydrocarbons, including nitrogenous, chlorinated, and sulfonated organics, are determined to be VOCs. These compounds are usually found in the industries that manufacture or utilize organic solvents, e.g., petrochemical, pulp, or coating industries.

In addition to causing harmful effects on human organs, VOCs may also react with NO_x in the atmosphere to form even more toxic photochemical smog and ozone (de Nevers, 2000). To remove these puzzling substances, a number of technologies have been developed. Among them, catalytic incineration has been received lately because it is a final disposal and an energy saving process (Van der Vaart et al., 1991a,b). Catalytic incineration is more selective and, as it requires less heating, is more cost effective than the other technologies, such as direct combustion (Choi and Yi, 2000), when the VOCs concentration is lower than 10000 ppm (Lahousse et al., 1998).

Several kinetic models for exploring the mechanism of catalytic incineration of VOCs on the catalyst have been raised and investigated. These models include the

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power-rate law (Lou and Lee, 1997), Mars and van Krevelen model (Hsu and Teng, 2001) and Langmuir–Hinshelwood model (Chu and Horng, 1998; Tseng and Chu, 2001). Power-rate law is a simple expression used to fit the experimental data. There is, although, no theoretical basis to this model, it can be described the total reaction rate occurring on the catalyst. The Mars and van Krevelen model and Langmuir–Hinshelwood model are more precise alternatives. The Mars and van Krevelen model depicts reaction of reactants on the catalyst surface mainly via an oxidation–reduction mechanism; most catalysts with oxidizing activity can be described by this redox mechanism. On the other hand, the Langmuir–Hinshelwood model depicts reactions in which the adsorbates can only adsorb chemically onto specific sites of the catalyst, each site can only adsorb one molecular (or atom), and the adsorption is limited to a monolayer.

Methyl isobutyl ketone, MIBK, is used as an excellent solvent for the paint industry and an important reagent in dewaxing mineral oils. It is also a hazardous material, which is on the control list of Taiwan Environmental Protection Agency.

This study was carried out by catalytic incineration of MIBK over a Pt/ γ -Al₂O₃ isothermal fixed-bed differential reactor. Three kinetic models, i.e. the Mars and van Krevelen model, Langmuir–Hinshelwood model and power-rate law were applied to best fit the experimental results. The effects of the various operating conditions and kinetic behavior of MIBK oxidation with catalysis are also determined. The present work can provide valuable information for designing and treating a VOC system.

2. Experimental

The catalytic decomposition of this study was done in a bench scale fixed bed reactor system. The system can be divided into three parts: an effluent gas simulation system, a catalytic decomposition system, and a combustion gas analyzing system. The effluent gas simulation system was composed of an air compressor (SWAN, 186.5 Nt m s⁻¹), a N₂ cylinder (99.9%, San Fu), three mass flowmeters (Teledyne Hasting-Raydist, HFC-202), two plug-flow mixers (Omega, FMX7106), a water bath (Deng Yng, 253 to 353 K), and two VOC vapor generators (Pyrex). Flow rates of dilution N₂, purge N₂, and dilution air were controlled by three mass flowmeters, to prepare the desired concentrations of MIBK and O₂. The simulated gas was then preheated by an electrical heating tape before going through the catalytic reactor. The materials of piping, valves, regulators, or fittings used were either SS-316 or polytetra-fluoroethylene.

The catalytic incineration system was composed of a custom-made SS-316 tube reactor and an electrical heater. The length, internal diameter, and outer dia-

meter of the reactor were 45 cm, 1.5 cm, and 2.0 cm, respectively. A 200-mesh SS-316 sieve was set in the reactor, 24 cm below the top of the tube, to support the catalyst. The weight of catalyst packing was 0.60 g (thickness 0.75 cm and bulk density 0.40 g cm⁻³). A thin layer of glass fiber and a layer of glass bead with a 2-mm diameter covered the catalyst packing to uniformly distribute the gas. Two K type thermocouples were inserted into the reactor to the positions on the top and bottom of the catalyst packing, respectively, to measure and control (up position) and measure (down position) the inlet and outlet temperature.

The gas analyzing system was composed of a gas chromatograph (GC) (Shimadzu, GC-14B) and three combustion gas analyzers: O₂ analyzer (Signal Model magneto dynamic type), CO analyzer (Signal Series 2000), and CO₂ analyzer (Signal Series 2000). The GC column was a 30-m long capillary column with diameter of 0.53-mm (J&W Scientific #115-3432) and connected to flow ionization detector (FID). An icebox impingement condenser (Pyrex) and two particle filters (Balston, 95S6 and 45 G) were installed between the sampling port and three combustion gas analyzers. This arrangement was designed to prevent the analyzers from being damaged by condensed water and particles. All mass flowmeters and rotameters were calibrated by a bubble meter (Humonic digital flowmeter 650) or a dry gas meter (Shinagawa DK-SCF-T) at their proper ranges. MIBK standard was a product of Chem Service Inc. (99%). The catalysts used were commercial products of NIKKI NS-10 Pt/ γ -Al₂O₃. Liquid MIBK used for the catalytic incineration system was a product of Merck Chemical Inc. (purity > 99.5%). Standard gases included zero gas (N₂, 99.9995%, San Fu) and O₂ span gas (21%, San Fu).

A series tests were performed earlier on conversion of methanol over the same Pt catalyst with three-size ranges (Chu and Lee, 1998). The results showed that the conversion differences were not significant. Therefore, the catalyst being ground to 50–100 mesh (0.03–0.015 cm) was chosen to carry out the experiments for this study in order to reduce both the interfacial mass transfer limitation and intraparticle mass transfer resistance. The catalyst's deactivation is not obvious during the tests. All the experiments were performed at temperatures ranging from 373–513 K and a gas hourly space velocity (GHSV) of 260 000 h⁻¹ (Table 1).

The procedures consisted of experiments with an integral reaction and a differential reaction. The integral reaction experiments were carried out to investigate the experimental conversions of MIBK. The differential reaction experiments were performed to investigate the effects of oxygen and MIBK content in carrier gas on the reaction rate. Moreover, various kinetics models

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