

Effects of reaction temperature and water vapor content on the heterogeneous photocatalytic oxidation of ethylene

Xianzhi Fu, Louis A. Clark, Walter A. Zeltner, Marc A. Anderson

Water Chemistry Program, University of Wisconsin-Madison, 660 N. Park St., Madison, WI 53706, USA

Received 7 June 1995; accepted 1 November 1995

Abstract

The photocatalytic degradation of ethylene in airstreams has been studied over the temperature range 30–110 °C using a packed bed reactor containing sol-gel-derived TiO₂ or platinumized TiO₂ particulates. Results of this study indicate that the reactivity of ethylene is greatly enhanced at increased temperatures and that the fraction of ethylene that reacts is stoichiometrically oxidized to CO₂ under all operating conditions. The effect of raising the temperature has been ascribed to decreasing adsorption of water for both types of catalysts, as well as an increase in conventional heterogeneous catalytic reactions occurring on the Pt/TiO₂ catalyst. In addition, platinumizing the TiO₂ photocatalyst and increasing the content of water vapor in the gaseous feed streams both decrease the rate of photocatalytic oxidation of ethylene. The activation energies for the photocatalytic and heterogeneous catalytic oxidation of ethylene were determined to be 13.9–16.0 kJ mol⁻¹ and 82.8 kJ mol⁻¹ respectively.

Keywords: Photocatalysis; Photocatalytic oxidation; Ethylene; Water vapour; Titania; Heterogeneous catalysis

1. Introduction

Purification of groundwater, soils and air polluted by volatile organic compounds (VOCs) is a task that is becoming increasingly important in environmental protection. One promising approach for remediating these systems is to employ semiconductor-mediated photocatalytic oxidation to obtain the complete mineralization (oxidation) of organic contaminants [1–7]. However, most heterogeneous photocatalysis studies have employed aqueous solutions at ambient temperature. Several of these studies have focused on the temperature dependence of the heterogeneous photocatalytic process [8–18]. In general, liquid phase photocatalytic reaction rates were improved by increasing the reaction temperature for temperatures in the range 20–60 °C. Arrhenius behavior has been reported in some cases with activation energies between 10 and 20 kJ mol⁻¹ [9,11,14–16]. These activation energies agree well with those found for OH radical reactions [19].

Studies involving temperature effects on gas phase photocatalytic reactions are far fewer [10,13,18]. The maximum rate for the photocatalytic oxidation of 1-butanol was obtained at 107 °C before it fell off at higher temperatures [13], while the complete mineralization of trichloroethylene (TCE) was obtained by increasing temperature from room temperature to 64 °C and by operating at *W/F* (mass of

catalyst divided by molar flow rate of contaminant) values greater than 6×10^7 g s mol⁻¹ [10]. However, reports also indicate that increasing the reaction temperature decreases the rate of photocatalytic oxidation of propene [18]. It thus appears that the precise role of reaction temperature in photocatalytic reactions requires further study.

Energy utilization in most photocatalytic oxidation processes is inefficient (especially when high power lamps are employed as UV light sources) because all lamps transform electrical energy into heat as well as light. According to the specifications for a 40 W Phillips fluorescent lamp emitting light at a peak of 350 nm, only 21% of the input electrical energy is utilized to produce UV light while 79% of that energy is transformed into heat. The efficiencies with which other types of commercially available UV light sources (e.g. mercury and incandescent) convert electrical energy to light are even lower than for fluorescent lamps. Note that most studies of photocatalytic reactions are conducted at ambient temperature by cooling the reaction apparatus to remove the thermal energy emitted by the lamp. This thermal energy is therefore being wasted rather than being utilized to help drive the oxidation process.

The major aim of the present study is to elucidate further the role of reaction temperature in the gas–solid photocatalytic oxidation of organic compounds. However, determina-