



Performance of continuous stirred-tank reactors connected in series as a photocatalytic reactor system



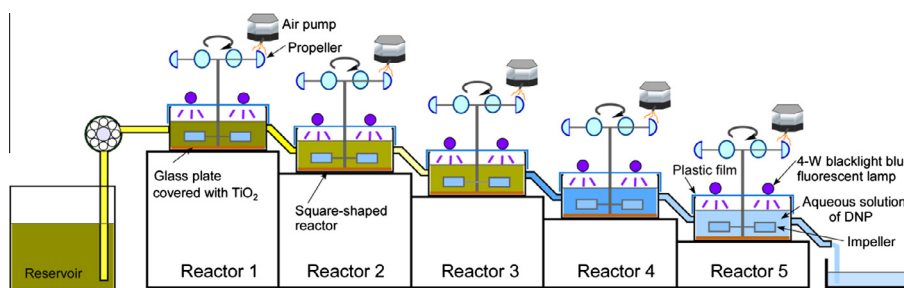
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HIGHLIGHTS

- Performance of five CSTRs in series as a photocatalytic reactor system is evaluated.
- Five CSTRs arranged here can decompose DNP in the absence of diffusional resistance.
- A five-CSTR system with sufficient mixing is close to that of a plug-flow reactor.
- Five-CSTR arrangement has practical potential as a photocatalytic reactor system.

GRAPHICAL ABSTRACT



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ABSTRACT

Film-diffusion resistance significantly reduces the decomposition of organic compounds over an immobilized photocatalyst in the vicinity. To overcome this problem, the performance of a photocatalytic system consisting of five continuous stirred-tank reactors (CSTRs) in series was investigated. Each reactor was a square-shaped container with a glass plate immobilized with a film of TiO_2 photocatalyst at the bottom. Two 4-W blacklight blue-fluorescent lamps fixed above the container provided UV light. A 10-ppm aqueous solution of 2,4-dinitrophenol (DNP) supplied continuously to the reactor was stirred by an impeller fixed to a shaft with a wind propeller at its upper end. The change in the DNP concentration in each reactor with increasing flow rate agreed with the result calculated by a mathematical model, constructed for the photocatalytic system, which included a mass-transfer coefficient expressed in terms of rotational speed. Hence, this model was used to simulate the performance of the reactor system. At sufficiently high rotational speeds, the conversion of DNP at the exit of the fifth CSTR was substantially higher than that at the exit of a single CSTR with the same liquid volume, and was close to the calculated value for a plug-flow reactor without film-diffusion resistance. At low rotational speeds, however, the DNP concentration remained remarkably high because of large film-diffusion resistance, demonstrating that the photocatalytic reaction requires sufficient liquid mixing to enhance the reaction rate. Thus, the system of five CSTRs in series is effective as a photocatalytic reactor system.

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1. Introduction

The operational mode of a chemical reactor can be divided into two main groups; batch-wise and continuous-flow modes [1,2].

The typical example for the former mode is the stirred-tank reactor and that for the latter mode is the continuous stirred-tank reactor (CSTR) and plug-flow reactor (PFR). Fixed-bed, fluidized-bed, membrane reactors, etc. are also used according to the property of reactants and products, the specificity of chemical reactions, operating conditions (temperatures, pressures, flow rates), etc. [3]. The CSTR and PFR are further used to define perfectly-mixed flow and plug

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flow, respectively. These ideal flows express both extremes of the liquid flows in chemical reactors, and the liquid flows in actual chemical reactors are included between the ideal extremes.

The continuous-flow mode is effective in treating a large amount of liquid solution [4,5]. Once operation of a reactor is started in this mode, the reactor can easily be controlled compared with that in the batch-wise mode. Thus, the continuous-flow mode has been utilized to operate many industrial reactors [6,7]. In the perfectly-mixed flow, the liquid solution is well mixed and the reactant concentration is uniform throughout. Hence, the reactant in the CSTR is diluted with the progress of the reaction, which leads to a reduction in the rate of reaction and, in turn, lowers the performance of the reactor. In the plug flow, on the other hand, there is no liquid mixing in the direction of flow. Hence, the reactant concentration in this reactor steadily decreases along the flow direction while taking a largest value at each position, which provides a theoretically highest performance for the reactor [1,2]. When many CSTRs are connected in series, the entire flow pattern approaches the plug flow; when the number of CSTRs is infinite, the performance of the reactor is identical to that of the PFR [1,8]. This theoretical fact is useful when one must inevitably adopt a CSTR system and wants to increase the performance of the system under a limited reactor volume as highly as possible. However, little practical application is available as to the multiple CSTRs.

Titanium dioxide (TiO₂) irradiated with UV light can decompose organic compounds in its vicinity [9–16]. In general, such a photocatalyst is used to decompose harmful substances in an environment. When a reactor containing a suspension of TiO₂ fine particles is operated in a continuous-flow mode, the particles easily flow out with the effluent. It is reasonable, therefore, to use TiO₂ by immobilizing it on a support [10,17–19]. However, the concentrations of harmful substances in the environment are generally less than or on the order of 1 ppm (parts per million). When such substances are treated using the immobilized TiO₂, the very low concentrations create a large film-diffusion resistance in the vicinity of TiO₂ in both gaseous reactions [20] and liquid reactions [21,22], leading to a marked decrease in the rate of photocatalytic decomposition.

Since the decomposition ability of UV-irradiated TiO₂ is not so high enough to put to practical use at once, it is reasonable to ensure a reaction field where TiO₂ can substantially express its inherent catalytic activity because of being free from the film-diffusion resistance. However, the removal of this resistance in water treatment is not easy compared with that in air treatment. Although this problem may be solved by mixing a reaction mixture at a high speed, a high energy consumption problem arises as a result. For achievement of the practical use of photocatalytic reaction processes, we thus consider that the fundamental knowledge of chemical engineering must be used to design a reactor that is free from the film-diffusion resistance and has a higher performance.

To increase the conversion of a reactant in a PFR, the reaction mixture must be allowed to flow very slowly. Unfortunately, this increases the film-diffusion resistance, which may significantly decrease the rate of photocatalytic reaction. To solve this problem, we have previously investigated the performance of a continuous-flow recirculation reactor system [23]. In this arrangement, a reactant solution introduced from the reactor entrance was recirculated at a high speed between a mixed-flow container and an annular-flow photocatalytic reactor, and a part of the reaction mixture was allowed to flow out of the exit. We found that as the rate of photocatalytic decomposition increases with an increase in the rate of flow of a reaction mixture through the photocatalytic reactor, the reactor performance could be enhanced. Although the ability of this reactor system to eliminate the problem of film-diffusion resistance is beneficial for photocatalytic processes, it has the

disadvantage of large energy consumption, since the reaction mixture must constantly be recirculated at a very high speed.

An alternative to the photocatalytic reactor is the CSTR, in which the film-diffusion resistance can be removed if the reaction mixture is well mixed. However, the performance of the CSTR is theoretically inferior to that of the PFR, because the rate of reaction is lowered due to the fluid mixing [1,8]. According to the reactor theory stated above, this problem might be solved by use of multiple CSTRs in series. Unfortunately, experimental studies to verify whether this theoretical prediction is correct in an actual reactor system are highly limited.

The purpose of the present study is to determine a continuous-flow reactor system that is appropriate for a photocatalytic reaction using immobilized TiO₂ both theoretically and experimentally. To achieve this purpose, we first select a CSTR that is easy to remove the film-diffusion resistance and then consider to compensate for the resulting decrease in the rate of reaction by using five CSTRs in series. DNP (2,4-dinitrophenol) is used as a reactant. A kinetic analysis of experimental data is carried out to construct a mathematical model for the reactor system. The mathematical model is used to examine an effect of mixing speed on the rate of photocatalytic reaction by computer simulation. Finally, the usefulness of the CSTRs is discussed.

2. Theory

2.1. Kinetic expression for the photocatalytic decomposition of DNP

The DNP molecules are negatively charged [24], while the TiO₂ immobilized on a glass surface is positively charged, because an aqueous solution of DNP at 10 g·m⁻³ is acidic (pH ~ 4.5) [25]. Therefore, the photocatalytic decomposition of DNP on the TiO₂ surface is strongly affected by an electrostatic interaction. The diffusion flux, N_A [g·m⁻²·cat·min⁻¹], toward the photocatalyst surface can be expressed as [26–28].

$$N_A = k_L(C_b - C_i e^{\lambda}) \quad (1)$$

where k_L is the mass-transfer coefficient [m³·liq·m⁻²·cat·min⁻¹], λ is the dimensionless electrostatic potential, and C is the DNP concentration [g·m⁻³·liq] with the subscripts 'b' and 'i' denoting the concentrations of DNP in the bulk liquid and at the TiO₂ surface, respectively. In the absence of the electrostatic interaction, the rate of the DNP decomposition reaction, r [g·m⁻³·liq·min⁻¹], is given by

$$r = -\frac{dC_i}{dt} = \frac{kK_H C_i}{(1 + K_H C_i) \left(1 + \frac{C_b}{K_{ip}} \frac{C_{b0} - C_b}{C_{b0} - C_b}\right)} \quad (2)$$

where k is the rate constant [g·m⁻³·liq·min⁻¹], C_{b0} is the initial concentration of DNP [g·m⁻³·liq], K_H is the adsorption equilibrium constant [m³·liq·g⁻¹], K_{ip} is the product inhibition constant [g·m⁻³·liq], and t is the time [min]. During the reaction, λ changes as follows.

$$\lambda = -\frac{\kappa_1 \kappa_2 C_{b0} \left(\frac{C_b}{C_{b0}}\right)^{\kappa_4}}{\exp\left(-\kappa_3 C_{b0} \left(\frac{C_b}{C_{b0}}\right)^{\kappa_4}\right) + \kappa_2 C_{b0} \left(\frac{C_b}{C_{b0}}\right)^{\kappa_4}} \quad (3)$$

In a previous study [29], we determined the following values for the parameters; $k = 0.20$ g·m⁻³·min⁻¹, $K_H = 0.146$ g⁻¹·m³, $K_{ip} = 25.0$ g·m⁻³, $\kappa_1 = 2.44$, $\kappa_2 = 0.0378$, $\kappa_3 = 1.60$, and $\kappa_4 = 0.519$. The value of k_L in Eq. (1) changes in proportion to the degree of liquid mixing. If a reaction mixture is well mixed, the stagnant film near the TiO₂ surface is disturbed, so that DNP molecules in the bulk can reach the TiO₂ surface rapidly. The present study expresses k_L as

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