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# Decolorization of azo dye in a FBR reactor using immobilized bacteria

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#### Abstract

Decolorization of azo dye (RED RBN) was carried out experimentally in a liquid–solid fluidized bed reactor (FBR) using polyvinyl alcohol (PVA)-immobilized cell beads as support carriers. The effects of various operating conditions such as bed expansion, cell bead number density, initial dye concentration, hydraulic retention time (HRT), and diameters of immobilized cell beads on the decolorization of azo dye were demonstrated experimentally. It was found that azo dye degradation time reaching initially a steady state decreased with an increase in bed expansion, cell bead number density as well as HRT. The mean cell residence time ( $\theta_C$ ) in FBR using PVA-immobilized cell beads increased insignificantly from 1014.1 to 1014.9 days as the HRT increased from 3 to 24 h, and thus the impact of convectional reduced  $\theta_C$  could be minimized by using the same polymer as support carriers. In addition, a mathematical model was used to describe the simultaneous diffusion and reaction of azo dye in the FBR. The internal mass-transfer resistance, rather than the film diffusion resistance, played an important role in azo dye utilization in FBR when film modulus ( $m_f$ ) was smaller than 1. The model was employed to analyze and predict both the  $k_L$  value and the operational efficiency of FBR (which was running in a well-mixed state) in azo dye biodegradation using immobilized cell beads during a steady-state operation. The simulated results corresponded satisfactorily with the experimental data in the azo dye concentration ranging up to 2200 mg L<sup>-1</sup>.

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### 1. Introduction

Color removal of industrial effluents has been a major concern in wastewater treatment, especially for wastewater that originates from textile and dyestuff plants with a continuous discharge of a great quantity of remaining dyes to the environment. Biological processes are frequently applied to decolorization of textile and dyestuff wastewater due to cost effectiveness. However, conventional activated sludge processes cannot achieve a satisfactory efficiency in color removal because the azo dyes are difficult to degrade aerobically [1,2]. Moreover, the solid–liquid separation in the settling tank occasionally leads to difficulties in process operation. To resolve such problems, the feasibility of applying immobilized cell technology to wastewater treatment has attracted increasing attention [3–6]. Moreover, selecting a suitable reactor is very crucial in improving the economy and efficiency of immobilized cell process. Previous studies have indicated that the PVA gel beads at a PVA concentration of 7% possessed not only good chemical resistance, but also great decolorization [7]. The performance was so obvious even when using PVA-immobilized cells in an aerated or stirred system, because the inhibitory effect of oxygen was markedly depressed in the immobilized cell system. The result indicates that the PVA-immobilized mixed cultures system may be operated in many different types of reactors (e.g. air-lift reactor, continuous stirred-tank reactor, fixed bed reactor, and fluidized bed reactor) for treating the textile effluents. For practical application in biological wastewater treatment, the chosen reactors are expected to be able to process continuously with less area requirements. As a result, the FBR has emerged as an alternative to other biological treatment systems or as a complementary treatment method. Consequently, the process combining FBR with immobilized-cell technology was used to achieve the decolorization of azo dye in this study.

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There have been many studies on the mathematical modeling of biological FBRs with immobilized enzymes or cells in porous particles [8–11]. Most of these models took into account the diffusion across the biofilm, but neglected the hydrodynamic characteristics of particles in the FBR. Although design equations were offered, the applications of the models were restricted to laboratory-scale reactors or a particular biological system. Moreover, for PVA-immobilized cell beads, the biofilm thickness could be negligible [12]. Therefore, the experimental data and models in the literature were not sufficient to offer design equations for FBRs using immobilized cell beads systems. Furthermore, for practical application, the predication of the general behavior of the liquid-solid FBRs is very important for process kinetics that takes into account the hydrodynamic characteristics of particles and the diffusional limitations. The design models that cover above-mentioned phenomena for FBRs with immobilized cell beads systems have not been found in the available literature.

This work is an extension of our previous research done in the laboratory using PVA gel beads as the support of biomass in FBR. Our previous study has examined the intrinsic kinetics of immobilized cell beads and hydrodynamic characteristics of the immobilized cell beads in FBR [12,13]. The objectives of this work are to study the operational performances (such as bed expansion, hydraulic retention time, beads number density, initial dye concentration, and recirculation ratio) of the FBRs combined with immobilized cell beads system on decolorization of azo dyes. Moreover, a general model was developed for the design of FBRs with immobilized cells in the gel beads. The hydrodynamic characteristics of particles and the internal diffusional limitations were taken into account. The validity of the proposed model was also verified by comparing the predicted results with the experimental data obtained from a FBR using PVA-immobilized cell beads. The model is useful for predicting the effluent dye concentration in respond to any specific operation condition under a steadystate with well-mixed conditions. In addition, the model is also applicable to the design and simulation of FBR operated with an immobilized cell beads system.

## 2. Model

#### 2.1. Pore diffusion associated with film diffusion

For most kinetic studies of immobilized cell systems, external diffusion is often negligible due to the efficient mixing of the solution in a stirred batch reactor [13]. However, it remains unclear whether the external mass transport effects have been eliminated in a FBR even if the recirculation ratios are kept larger. The film resistance in cell particles is specifically not negligible in a column reactor. Immobilized cell beads are considered as porous spheres where cells are uniformly distributed on the surface of and inside the beads. Furthermore, the following assumptions are made: (1) The enzymatic reactions with three azo-degrading-bacteria are the same and they are all mono-substrate; (2) The effective diffusivity  $(D_e)$  of azo dye decreases with an increase in cell density as discussed in our previous study [12], but the  $D_{\rm e}$ value of RED RBN inside the immobilized cell beads is kept constant at a steady state because the cell concentration is also kept constant; (3) The ratio of the amount of biomass in the solid phase to that in the bulk liquid phase  $(m_P/m_L)$  is greater than 97%. In other words, no color removal is due to suspended growth, rendering the influence of free cells in the bulk liquid negligible; (4) The dye diffusion through the stagnant liquid film surrounding the gel bead and inside the immobilized cell beads could be modeled by Fick's law; (5) The rate of reaction could be represented by Michaelis-Menten kinetics and the rate of reaction is unaffected by the presence of reaction products; (6) At the solid-liquid interface, there is no accumulation of substrate or products. With the above assumptions and under steady-state conditions with simultaneous reactions in the immobilized cell beads, the mass balance for pore diffusion yields is:

$$D_{\rm e}\left(\frac{\partial^2 C_{\rm p}}{\partial r^2} + \frac{2}{r}\frac{\partial C_{\rm p}}{\partial r}\right) = \frac{\phi_{\rm cv}V_{\rm m}'C_{\rm p}}{K_{\rm m}' + C_{\rm p}} \tag{1}$$

## $\phi_{\rm cv} = \phi_{\rm cw} \rho_{\rm p}$

On the other hand, under steady-state conditions, the mass balance for film diffusion is:

$$k_{\rm L}S_{\rm V}(C_{\rm L} - C_{\rm P}) = \frac{V'_{\rm m}C_{\rm P}}{K'_{\rm m} + C_{\rm P}}$$
(2)

For combined effects of film and pore diffusion, the boundary conditions are:

$$\frac{\partial C_{\rm P}}{\partial r} = 0 \quad \text{at} \quad r = 0 \tag{3}$$

$$D_{\rm e} \left(\frac{{\rm d}C_{\rm P}}{{\rm d}r}\right)_{r=R} = k_{\rm L}(C_{\rm L} - C_{\rm PR}) \quad \text{at} \quad r=R \tag{4}$$

$$C_{\rm P} = C_{\rm PR} = K_{\rm P}C_{\rm L} \quad \text{at} \quad r = R \tag{5}$$

where  $C_{PR}$  and  $C_L$  are the surface concentration and the bulk concentration, respectively.  $V'_{\rm m}$  and  $K'_{\rm m}$  are the intrinsic catalytic constant and the Michaelis-Menten constant, respectively, for reaction within the immobilized cell beads.  $k_{\rm L}$  and  $S_{\rm v}$  are the external mass-transfer coefficient and the total surface area per unit volume of reaction solution, respectively.  $\psi_{\rm cv}$  is the dry-cell weight per unit volume of immobilized cell beads, i.e. the density of cells in the immobilized cell beads.  $\psi_{cw}$  is the dry-cell weight per unit weight of immobilized cell beads.  $\rho_p$  is the density of immobilized-cell beads. A convenient way to describe the influence of diffusional resistance on reactor performance is to use the concept of effectiveness factors. Here,  $\eta_D$  is the effectiveness factor of immobilized cell beads, and is defined as the ratio of actual reaction rate to the reaction rate without diffusion limation (for internal mass transfer controlling reaction only):

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