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## Stability and Dynamic Behaviour of a Fluidised Bed Bioreactor Treating Phenolic Effluent

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**Abstract:** Stability aspects and dynamic behaviour of a draft-tube fluidised bed bioreactor treating phenolic effluent from a coal carbonisation plant have been investigated in the present study. Experiments were carried out to study the biodegradation of phenolic wastewater using the microorganism *Pseudomonas* sp., immobilised on solid support particles. The dynamics of the reactor system were monitored at various experimental conditions viz., feed (substrate) flow rates and phenol concentrations in the feed. The draft tube fluidised bed bioreactor was assumed to operate in a completely backmix mode and a model to predict the outlet phenol concentration for given inlet and equipment parameters was developed. The model equations were solved using 4<sup>th</sup> order Runge-Kutta technique. The predictions made by the dynamic model were compared with experimental findings and the agreement between the two was good. Stability of the reactor to input disturbances was examined and it was found to be stable.

**Keywords:** Biodegradation, Wastewater, Phenol, Fluidised bed bioreactor, Dynamic behaviour.

### Introduction

Phenol is one of the pollutants present in effluents from chemical process plants. Process industries, which are major sources of phenolic discharges, include petroleum refineries, coal carbonisation units, gas and coke plants and fibreglass units. Smelting and connected metallurgical operations, plastic industries, paint and varnish industries and textile units making use of organic dyes also contribute to phenolic liquid waste. Biodegradation of phenol in fluidised bed bioreactors (FBRs) has been reported because of their superior performance and some inherent advantages [1-7]. The superior performance of FBRs is due to very high concentration of immobilised cells on the solid particles, prevention of washout of the microbes, lack of clogging of the biomass, ease of separation of cells from

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product stream and elimination of limit on liquid flow rates due to decoupling of residence time of liquid phase and of microbial cells.

For a variety of reasons it is highly desirable to understand the transient behaviour of a FBR, especially for wastewater treatment. The reactor is in transient stage during start-up and shut down. Also, any disturbances in input conditions viz., flow rate and feed concentration, disturb the steady state of the system leading to a transient. Several workers have attempted to study this transient behaviour [8-14]. The dynamic behaviour of a phenol oxidising fixed film using FBR was analysed by Worden and Donaldson [15]. A pulse of phenol was added to perturb the system and the phenol concentration was monitored continuously until steady state was again achieved. The experimental dynamics were compared with a dynamic model based on diffusion and reaction with the biofilm, liquid mixing and biofilm growth. Tang et al. [16] studied the transient behaviour of a draft tube gas-liquid-solid fluidised bed biofilm reactor to a step increase in influent phenol concentration. Shahalam et al. [17] developed a theoretical model describing the process occurring in a FBR. It covered both the unsteady and steady state conditions of the process, taking into account the bulk liquid biomass activity. Tsuneda et al. [18] have studied the dynamic response of completely mixed three-phase fluidised bed biofilm reactor treating simulated domestic wastewater after a step change has been given in inlet concentration. It was found that the response curves showed second order characteristics.

There have not been any reports regarding the stability and dynamic behaviour of draft tube FBR treating phenolic effluents from actual plant units. An attempt has been made in the present work to address these aspects. The dynamics and stability of the reactor system were monitored at various experimental conditions viz., feed (substrate) flow rates and feed concentration of phenol.

## Experimental

### Sample

Samples of wastewater were collected from a low temperature coal carbonisation plant in Andhra Pradesh, India. Table 1 gives details of analysis of the plant's wastewater.

**Table 1. Analysis of plant wastewater**

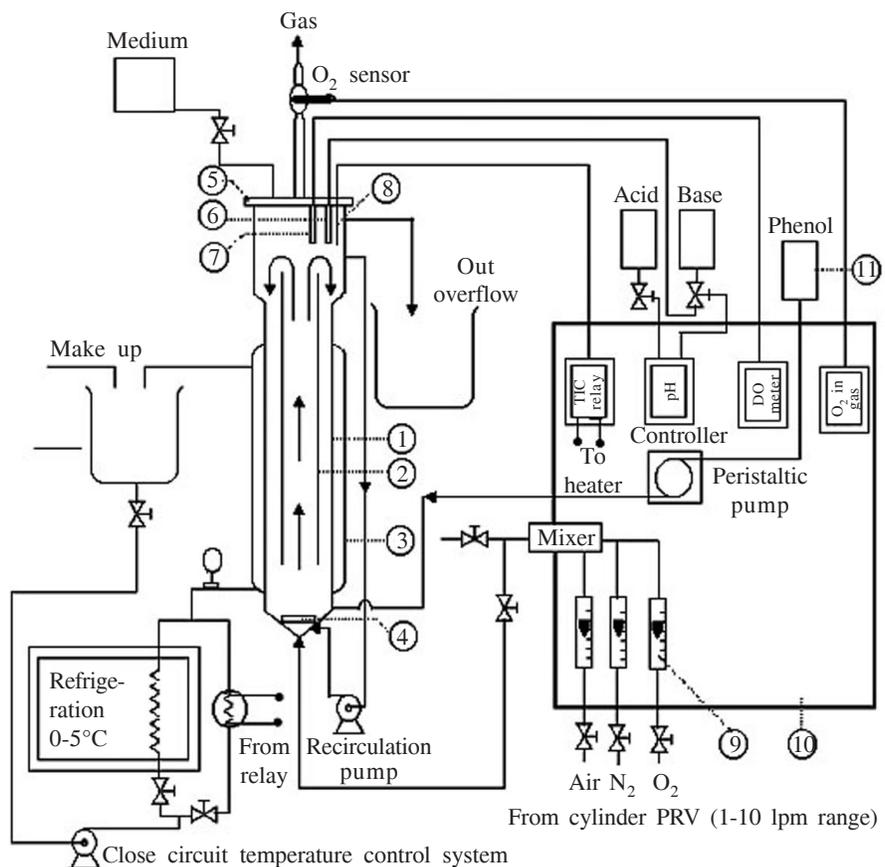
pH	9.0
Density	1.0288 gcm <sup>-3</sup>
Total solids	6,720 mg l <sup>-1</sup>
Dissolved solids	5,312 mg l <sup>-1</sup>
Suspended solids	1,408 mg l <sup>-1</sup>
Chlorides	Nil
Sulphates	0.802 g l <sup>-1</sup>
Phenol	10,240 mg l <sup>-1</sup>
Thiocyanates	2.84 g l <sup>-1</sup>
Total alkalinity	14,670 ppm
C.O.D.	20,400 ppm
B.O.D.	11,100 ppm

### Reactor Set-up

Figure 1 shows the schematic diagram of the draft tube FBR used in the present work. The various dimensions of the column and draft tube are shown in Fig. 2.

### Reactor and Draft Tube

The FBR and draft tube were made of glass. A sparger made of the same material was provided at the bottom of the reactor through which air could be sparged into the reactor. The total volume of the reactor was about  $2.67 \times 10^{-3} \text{ m}^3$  (2.67 l). The top of the glass



- 1: Reactor column
- 2: Draft tube
- 3: Jacket
- 4: Gas distributor
- 5: Top enclosure
- 6: pH electrode
- 7: DO electrode

- 8: Thermocouple
- 9: Rotameter
- 10: Panel mounting
- 11: Feed tank

- Reactor: Glass
- Vessel dia.: 80 mm
- Larger-end dia.: 100 mm
- Jacket dia.: 100 mm
- Length: 400 mm
- Draft tube dia.: 50 mm

Fig. 1. Schematic diagram of the experimental set-up.

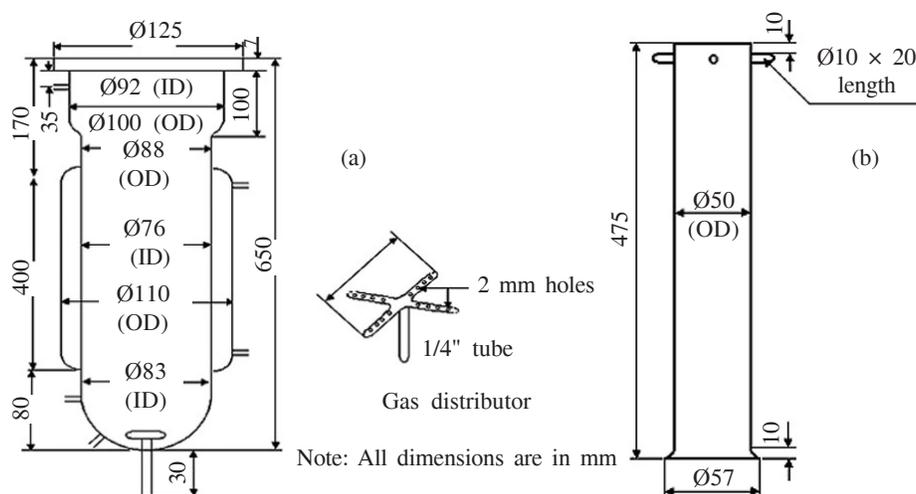


Fig. 2. (a) Reactor with jacket and (b) draft tube.

reactor was closed with a plate through which all the probes and sensors were inserted into the reactor. An overflow line was provided near the top so that the reaction medium flowed out of the reactor in continuous operation.

Plastic beads with a density of  $1,005 \text{ kg/m}^3$  were used for immobilisation of the microorganism. The average diameter of the beads was 3.895 mm. Two peristaltic pumps, one each for media and feed into the reactor, were provided. The flow rate of these pumps can be set at the required value using a flow controller. The capacity of the pumps was  $0.11 \times 10^{-7}$  to  $9.7 \times 10^{-7} \text{ m}^3/\text{s}$  (40-3,500 ml/h). The reactor was provided with a glass jacket to maintain the temperature of the reactor system. Depending on the temperature set for the reactor operation, the controller activated either the heating or refrigeration circuit. Separate tanks made of stainless steel were provided for supplying the feed, medium, acid and base solutions for pH control.

#### Reactor Instrumentation

A pH meter and a controller were provided to maintain the pH of the system. The pH was maintained by addition of acid or base from the tanks provided at the top. The oxygen required for the degradation of phenol by the microorganism was supplied in the form of air from a compressor. The oxygen content in the reaction medium was measured using a DO meter. The flow rate of air was measured using a rotameter with a range of  $0.167 \times 10^{-4}$  to  $1.67 \times 10^{-4} \text{ m}^3/\text{s}$  (1-10 lpm).

#### Microbial Culture

A strain of the microorganism *Pseudomonas* sp. reported to be capable of utilising phenol as the sole carbon and energy source was obtained from Regional Research Laboratory, Jammu, India.

### **Culture Preparation**

The culture was maintained by periodic subculture on nutrient agar and stored in a refrigerator. The reaction medium was prepared from this strain by growing the bacteria on  $2.6 \times 10^{-3} \text{ m}^3$  (2.6 l) of  $0.05 \text{ kg/m}^3$  (50 ppm) phenol solution containing growth medium [2, 6]. Before inoculation of the organism, phenol solution was sterilised in autoclave at a gauge pressure of  $1.034 \times 10^5 \text{ N/m}^2$  (15 psi) for 20 min. This was done to selectively grow the *Pseudomonas* species.

### **Growth Medium**

The growth medium was prepared using tap water. Sterile conditions were not maintained during continuous operation of the reactor, in order to simulate treatment of actual plant wastewater as the latter would contain different contaminants.

### **Biomass**

To find the biomass concentration in the reaction medium, 25 ml of reactor medium was taken in every experimental run and filtered through  $0.7 \mu\text{m}$  filter paper to separate the biomass produced. The filter paper was dried at  $105^\circ\text{C}$  and weighed again after drying to obtain the weight of the biomass produced. The amount of biomass divided by the sample volume gives the biomass concentration in the reactor.

### **Determination of Gas Phase Volume Fraction**

In all the runs air at 2 lpm was used as source of oxygen. At the same time, it helped maintain the fluidisation of bioparticles (corresponding to a linear velocity of  $0.017 \text{ m/s}$  based on draft tube diameter). To find the gas phase volume in the reactor, air supply to the reactor was suddenly shut off when the reactor was operating in steady state. As a result, the level of reaction medium in the reactor fell by an amount equal to the volume of the gas phase present in the reactor when it was in steady state operation. This volume was determined by measuring the volume of liquid required to fill the reactor to the previous level. This measured volume was divided by the total reactor volume to obtain the gas phase volume fraction in the reactor.

### **Determination of Volume Fraction of Solid Particles**

In all the experimental runs the volume of solid particles was maintained at 10% of the total reactor volume. This volume was arrived at by taking a measuring cylinder and filling it with the solid particles and water. The difference of the total volume and volume of water is the volume of solid particles.

The analysis (Table 1) indicates that the phenol content of plant wastewater was very high (i.e. 10,240 ppm). This was diluted to bring down the concentration to 100 ppm for preparing the reaction medium. After 24 h of incubation, 2.6 l of reaction medium was added to the reactor (FBR). The reactor was run in batch mode for further 36 h for immobilisation of biomass onto the beads. Just before the reactor was started, the phenol concentration in the reactor was brought to feed concentration level by adding additional phenol to the reactor. The reactor was started with feed flow rate of  $470 \text{ ml/h}$  (corresponding to a dilution rate of  $0.201 \text{ h}^{-1}$ ). The experimental conditions maintained were:  $\text{pH} = 7.0$  and

temperature = 30°C. The same conditions were maintained at other flow rates of the wastewater. Phenol concentration in the outlet was monitored at regular intervals using the standard method of analysis [19].

The effect of feed flow rate on biodegradation was studied at different feed flow rates:  $1.3 \times 10^{-7}$ ,  $1.41 \times 10^{-7}$ ,  $1.66 \times 10^{-7}$ , and  $1.77 \times 10^{-7}$  m<sup>3</sup>/s (470, 510, 600 and 640 ml/h, respectively). The inlet phenol concentration can affect the reactor performance. The transient behaviour of the reactor was studied at four inlet phenol concentrations of 0.09, 0.15, 0.199 and 0.25 kg/m<sup>3</sup> (90, 150, 199 and 250 ppm, respectively).

### Dynamic Model

The FBR is assumed as a continuous stirred tank bioreactor, which is considered as an ideal reactor. It is based on the assumption that the reactor contents are well mixed. The RTD studies justify the assumption. Tracer studies carried out indicate backmix conditions. The results of one such study (Fig. 3) clearly show that the FBR behaves in backmix mode. Therefore, the concentrations of various components of the outlet stream are assumed to be the concentrations of these components in the reactor. A similar work, in which dynamic model has been applied to a biofilm process, has been reported in literature [18]. The assumptions made to develop the mathematical model are as follows:

1. The growth kinetics is assumed to follow substrate-inhibited kinetics with respect to phenol. The expression for the specific growth rate as a function of substrate concentration is

$$\mu = \frac{S\mu_{\max}}{(K_s + S + S^2 / K_i)}$$

2. The particle remains totally inert during all experiments and does not adsorb phenol.

The material balance over FBR for phenol and biomass results in the following equations:

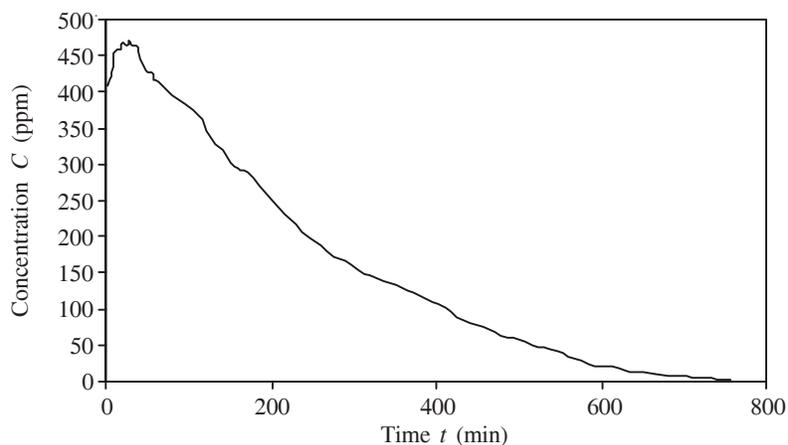


Fig. 3. Tracer concentration vs. time curve (feed flow rate = 396 ml/h, air flow rate = 2 lpm).

$$\frac{dS}{dt} = \frac{FS_o}{V} - \frac{FS}{V} - \frac{\mu_{\max}/Y_{x/s}}{K_s + S + S^2/K_i} XS \quad (1)$$

$$\frac{dX}{dt} = -\frac{FX}{V} + \frac{\mu_{\max}}{K_s + S + S^2/K_i} XS \quad (2)$$

$$V = (1 - \varepsilon_g - \varepsilon_p) V_R = \text{Active volume of the reactor}$$

### Solution Procedure

The mathematical model consists of mutually coupled differential Eqs. (1) and (2). The values of the model parameters  $\mu_{\max}$ ,  $K_s$ ,  $K_i$  and  $Y_{x/s}$  obtained previously [6] are given in Table 2. The values of other variables in Eqs. (1) and (2),  $F$ ,  $S_o$ , and  $V$  were obtained from experiments. The equations have been solved for  $S$  and  $X$  as function of time  $t$  using 4<sup>th</sup> order Runge-Kutta method.

**Table 2. Values of the model parameters**

Parameter	Value	Unit
$\mu_{\max}$	$1.436 \times 10^{-4}$	$s^{-1}$
$K_s$	$21.92 \times 10^{-3}$	$kg/m^3$
$K_i$	$522 \times 10^{-3}$	$kg/m^3$
$Y_{x/s}$	0.6	$kg/m^3$

### Stability of the Reactor to Infinitesimal Perturbations

The necessary and sufficient condition for asymptotic stability of a system to infinitesimal perturbations is that all the eigen values of  $\mathbf{A} = \nabla \mathbf{f}(\mathbf{x})$  have negative real parts (based on Liapunov function [20]). The  $\mathbf{f}(\mathbf{x})$  is an array of differential equations. The matrix  $\mathbf{A} = \nabla \mathbf{f}(\mathbf{x})$  has elements

$$A_{11} = \frac{\partial f_1}{\partial x_1}; A_{12} = \frac{\partial f_1}{\partial x_2}; A_{21} = \frac{\partial f_2}{\partial x_1}; A_{22} = \frac{\partial f_2}{\partial x_2}$$

In the study,  $x_1$  and  $x_2$  correspond to  $S$  and  $X$ , respectively, while  $f_1$  and  $f_2$  correspond to  $dS/dt$  and  $dX/dt$ , respectively. Eigen values  $\lambda_1$  and  $\lambda_2$  are the solutions of the characteristic equation

$$\lambda^2 - (A_{11} + A_{22})\lambda + (A_{11} A_{22} - A_{12} A_{21}) = 0$$

The necessary and sufficient conditions that the roots have negative real parts are

$$A_{11} A_{22} - A_{12} A_{21} > 0 \text{ and } A_{11} + A_{22} < 0 \quad (3)$$

In the present case

$$A_{11} = -F/V; A_{12} = -\mu/Y_{x/s} = -\frac{c\mu_{\max}S}{Y_{x/s}(S + K_s + S^2/K_i)}$$

$$A_{21} = 0, A_{22} = -F/V + \mu$$

### Results and Discussion

The results of the study on effect of flow rate on phenol concentration from the reactor are shown in Figs. 4 and 5. The lines indicate the theoretical predictions by the model, i.e. solving Eqs. (1) and (2) for outlet phenol concentration at different times. Experimental

data is represented by symbols. There is reasonably good agreement between the theoretical predictions and experimental results. In Fig. 4 the model predictions are higher than the experimental results. This can be attributed to the relatively simple model used in the study. At low flow rates the system reached steady state faster than at higher flow rates. This trend was observed both in model predictions and experimental findings. As the mean residence time increased the time taken for the system to reach steady state decreased. The assumption that FBR operates in backmix mode is justified.

The steady state outlet phenol concentration decreases as feed flow rate to the bioreactor decreased indicating the influence of mean residence time of phenol. In FBRs the biodegradation rate is decoupled from mean residence time to some extent due to the presence of biomass on the solid particles. But the findings of the present work indicated that residence time also plays a role in the rate of biodegradation. This is due to the fact that residence time of phenol is dependent on feed flow rate even though the residence time of immobilised biomass is decoupled from that of feed flow rate. Figure 5 shows the

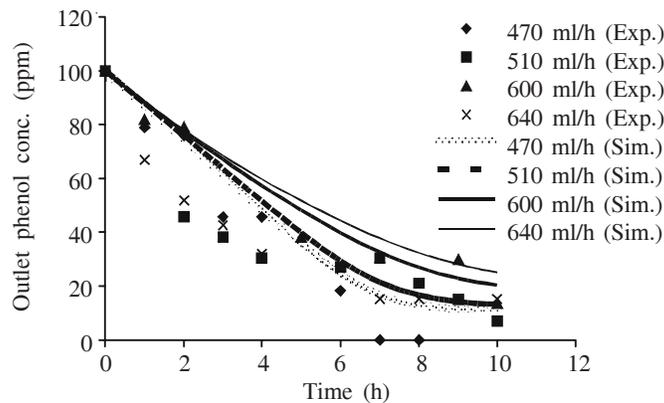


Fig. 4. Effect of feed flow rate on outlet phenol concentration (feed concentration of phenol = 100 ppm).

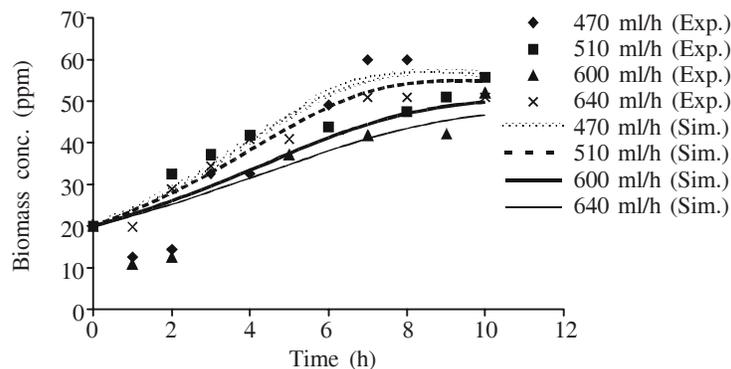


Fig. 5. Effect of feed flow rate on biomass concentration (feed concentration of phenol = 100 ppm).

dynamic behaviour of the reactor with respect to biomass concentration at various flow rates. The lines indicate simulated results given by Eqs. (1) and (2), and symbols correspond to experimental values. Initial concentration of biomass was maintained at 20 ppm in all cases. Figure 5 also shows that biomass concentration reached steady state faster at lower flow rates. Moreover, steady state value of biomass was higher at lower flow rates.

Figures 6 and 7 show the variation of outlet phenol and biomass concentrations from the reactor with time under transient conditions at different inlet phenol concentrations. Four feed concentrations of 0.09, 0.15, 0.199 and 0.25 kg/m<sup>3</sup> (90, 150, 199 and 250 mg/l, respectively) were used in the study. At higher concentrations the microorganism did not survive. The system reached steady state faster at lower inlet concentration than at higher

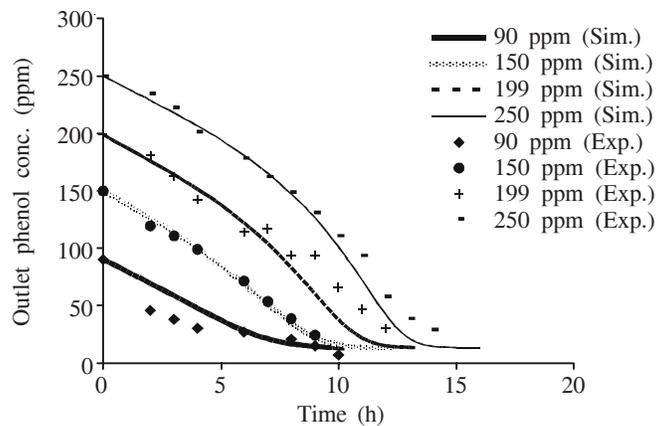


Fig. 6. Effect of feed concentration on outlet phenol concentration (flow rate = 510 ml/h).

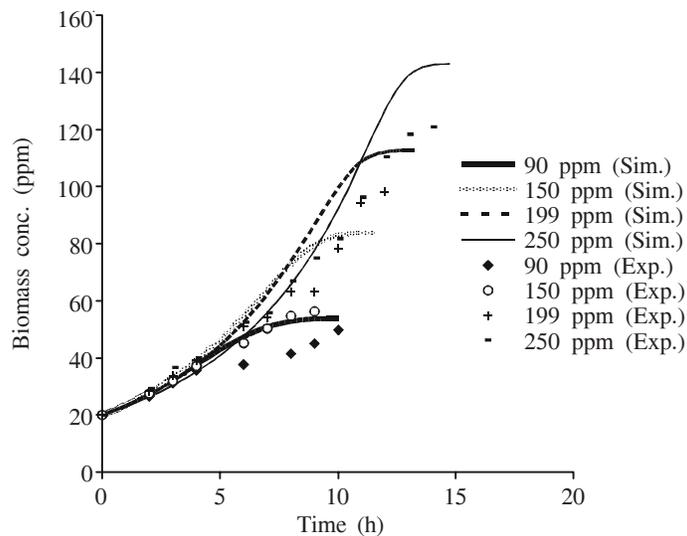


Fig. 7. Effect of feed concentration on biomass concentration (flow rate = 510 ml/h).

inlet concentration (Fig. 6). In all the four cases, final outlet concentration was the same. Figure 7 shows the transient behaviour of biomass when a step input was given to the system. The initial biomass concentrations were the same in both the cases. The steady state value of the biomass concentration was higher at higher inlet concentration because with steady state outlet phenol concentration being the same at all feed concentrations, the amount of biomass has to be higher at higher inlet phenol concentration. Also, at higher inlet phenol concentration the system took longer time to reach the steady state (Fig. 7) because with initial concentrations being the same in both the cases it would take longer for steady state to be reached for the case of higher phenol concentration in feed. There was good agreement between model predictions and experimental results (Fig. 6). In Fig. 7 the values of experimental results are smaller than the model predictions.

A similar work reported in literature is that of Tsuneda et al. [18] who considered absorption of substrate into the biofilm in the dynamic model. In their work, cement balls were used as support for the biofilm. The experiments were carried out using simulated domestic wastewater. The time required to reach steady state for TOC (total organic carbon) was about 100 h. This is considerably more than the steady state times in the present work. This is due to the fact that lower concentrations of phenol were used in this study.

The results on dynamic simulation show that the reactor (FBR) attains steady state after a certain time depending on the flow rates and concentration. Thereafter, the reactor continues to operate in steady state condition despite any input disturbances. If the input disturbances viz., in feed flow rate or feed concentration, result in a transient that takes the system to a new steady state, then the system can be called 'stable'. If the disturbance is amplified instead of being attenuated, the system becomes unstable and would never reach steady state. The FBR system in the present study was analysed for stability. The inequalities shown (Eq. (3)) are the necessary and sufficient conditions for steady state to be stable to infinitesimal perturbations. The last two rows of Table 3 show that the conditions are met, thus, indicating the stability of the reactor to infinitesimal perturbations.

**Table 3. Results of stability calculations**

Variable	Run 1	Run 2	Run 3	Run 4
Feed flow rate $F$ (l/h)	0.470	0.510	0.600	0.640
Feed flow rate $F$ ( $\text{m}^3/\text{h} \times 10^7$ )	1.3	1.41	1.66	1.77
Gas hold-up ( $\text{m}^3 \times 10^3$ )	0.065	0.065	0.065	0.065
Effective reactor volume* $V$ ( $\text{m}^3 \times 10^3$ )	2.338	2.338	2.338	2.338
Steady state phenol conc. $S$ (ppm) for calculation of $\mu$	11.15	13.0	20.84	25.69
$\mu$ ( $\text{h}^{-1}$ )	0.152	0.168	0.217	0.239
$A_{11}$	- 0.201	- 0.218	- 0.257	- 0.274
$A_{12}$	- 0.255	- 0.282	- 0.364	- 0.401
$A_{21}$	0	0	0	0
$A_{22}$	- 0.049	- 0.05	- 0.04	- 0.035
$A_{11} A_{22} - A_{12} A_{21}$	> 0	> 0	> 0	> 0
$A_{11} + A_{22}$	< 0	< 0	< 0	< 0

\* $V$  = Total reactor volume - Gas hold-up (65 ml) - Volume of solids (267 ml)

## Conclusion

Phenolic wastewater from a large-scale plant has been degraded using the microorganism *Pseudomonas* sp. The study of dynamic behaviour of the draft tube fluidised bed bioreactor indicates that the system reaches steady state faster at lower flow rates. Also, the system reaches steady state faster at lower inlet phenol concentrations than at higher inlet phenol concentrations. The fluidised bed bioreactor treating phenolic effluent was found to be a stable one.

## Nomenclature

$A_{11}, A_{22}, A_{12}, A_{21}$	Parameters in necessary and sufficient condition for stability
$c$	Constant accounting for the term $C/(K_o + C)$
$C$	Oxygen concentration, $\text{kg/m}^3$
$\mathbf{f}(\mathbf{x})$	Vector function (for differential equations)
$F$	Feed flow rate, $\text{m}^3/\text{s}$
$K_1$	Inhibition constant, $\text{kg/m}^3$
$K_o$	Monod (or saturation) constant for oxygen, $\text{kg/m}^3$
$K_s$	Monod (or saturation) constant for substrate, $\text{kg/m}^3$
$r_s$	Rate of substrate consumption for the bacterial reaction, $\text{kg/s}$
$r_x$	The rate of biomass production = $\mu X$ , $\text{kg/s}$
$S_o$	Inlet phenol concentration, $\text{kg/m}^3$
$S$	Phenol concentration in reactor, $\text{kg/m}^3$
$V$	Active volume of the reactor contents, $\text{m}^3$
$V_R$	Reactor volume, $\text{m}^3$
$X$	Biomass concentration, $\text{kg/m}^3$
$Y_{x/s}$	Yield coefficient, $\text{kg/kg}$

## Greek Symbols

$\epsilon_g$	Volume fraction of gas phase
$\epsilon_p$	Volume fraction of particle phase
$\lambda_1, \lambda_2$	Eigen values
$\mu$	Specific growth rate, $\text{h}^{-1}$
$\mu_{\text{max}}$	Maximum specific growth rate, $\text{h}^{-1}$

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