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Kinetic analysis of treatment of formaldehyde containing wastewater in UAFB reactor

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ABSTRACT

Anaerobic degradation of formaldehyde containing wastewater was studied in an upflow anaerobic fixed film (UAFB) reactor. The reactor was operated at 0.18–3.61 kg COD m^{-3} d^{-1} corresponding to formaldehyde concentration of 65–92 mg L^{-1}. COD and formaldehyde removal were found to be 92–24% and 99–41%, respectively. The efficiency of the reactor was investigated at different hydraulic retention time of 10–24 h. Kinetic models were applied to the data obtained from the studies in the anaerobic reactor. Second order and Stover–Kincannon models were best fitted to the data on UAFB reactor. The second order substrate removal rate ($K_{2(s)}$) was found to be 3.2 h^{-1}. U_{max} and K_B value constants for Stover–Kincannon models were found to be 3.4 g d^{-1} and 4.6 g d^{-1}, respectively.

1. Introduction

Many industrial activities utilize formaldehyde as a key chemical for the production of penta erythritol, ethylene glycol, synthetic resins, paper products, wood processing, paints, medicine and drugs [1]. Formaldehyde is also used as a disinfectant for killing bacteria, fungi, etc. [2]. USEPA has classified formaldehyde as a "Probable Human Carcinogen" [3]. The effluents arising from these applications may contain significant amount of formaldehyde [4]. Formaldehyde reacts with DNA, RNA and damage cells which cause death of the microorganisms [5] and was found to be mutagenic and carcinogenic when exposed to high concentration [6]. Formaldehyde can inhibit the growth of aerobic bacteria at lower concentrations [2]. As a disinfectant, formaldehyde solution (0.5%) destroys all species of microorganisms in a period of 6–12 h [7]. The process urea–formaldehyde–resin releases wastewater with high COD (50–200 g L^{-1}) and formaldehyde (2–4 g L^{-1}) concentration. Formaldehyde ranks top in the list of environmental impacts of among 45 chemical products [7].

Anaerobic biodegradation was found to be an alternative for the treatment of wastewater containing high organics. Moustafa et al. [3] reported that formaldehyde was removed from high strength organic wastewater in anaerobic granular activated carbon fluidized bed reactor. The reactor removed more than 95% of the dissolved organic carbon and 99.9% of formaldehyde of waste under continuous operation. Vidal et al. [8] studied the removal of formaldehyde in anaerobic sludge blanket reactor using glucose as a co substrate. Oliveira et al. [7] showed effective removal of formaldehyde and COD (99.9% and 92%, respectively) using horizontal anaerobic immobilized sludge bed (HAIB) reactor. Formaldehyde concentration of 26.2–11,586 mg L^{-1} were applied in the reactor, resulting in formaldehyde and COD removal efficiencies of 99.7% and 92%, respectively. In a multi upflow filter anaerobic reactor, Garrido et al. [9] obtained 100% formaldehyde removal by increasing the concentration of formaldehyde from 250 to 1000 mg L^{-1}.

Many other reactors including granular sludge blanket reactor [10], fluidized bed bioreactor [11], horizontal flow anaerobic immobilized sludge reactor [7] and expanded sludge blanket reactor [12] have been studied for the degradation of formaldehyde. In the present investigation, upflow anaerobic fixed film reactor (UAFB) was used for the degradation of formaldehyde containing wastewater. UAFB is known to operate at high solid retention time (SRT) and low hydraulic retention time (HRT). The purpose of the study was to evaluate the performance of UAFB having insulated beads as support material for biomass immobilization in the treatment of formaldehyde containing wastewater. Additionally, the process kinetics provides a useful technique for predicting the performance of the reactor in order to evaluate formaldehyde removal and to determine kinetic constants. Different mathematical models, including Monad model, second-order kinetic model, Stover–Kincannon model were applied to the reactor and the kinetic constants were determined.

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Nomenclature

ds/dt	substrate removal rate ($g\,L^1\,d^{-1}$)
k	the maximum rate of substrate removal ($L^{-1}\,d^{-1}$)
K_B	the saturation value constant ($g\,L^1\,d^{-1}$)
K_s	the half-velocity constant (and V is the clean-bed volume of the reactor
U_{max}	maximum utilization rate constant ($g\,L^{-1}\,d^{-1}$)
X	the microorganism concentration (VSS) in the UAFB ($g\,L^{-1}$)

Table 1
Characteristics of formaldehyde containing wastewater

Parameters	Concentration
pH	2.5–3.0
Total solids	1,000–1,210
Chloride	35–41
Chemical oxygen demand	10,976–11,840
Bio chemical Oxygen Demand	3,200–3,250
Formaldehyde	8,400–8,545
Methanol	2,800–2,950
Total organic carbon	3,461–3,523

All the parameters expressed in mg L^{-1}except pH.

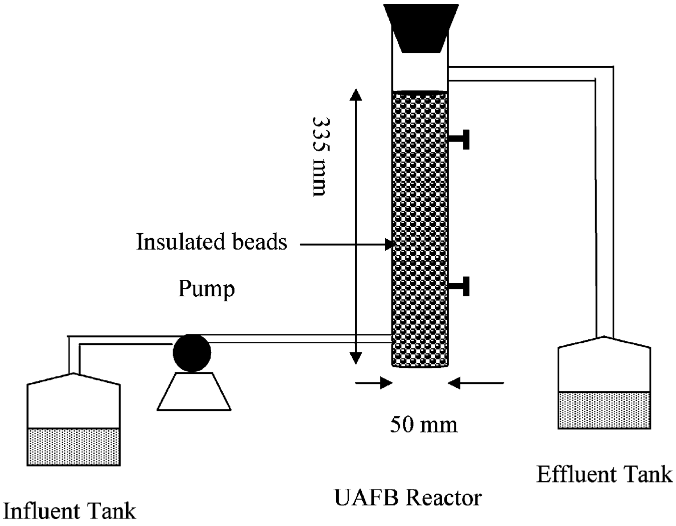

Fig. 1. Schematic diagram of UAFB reactor.

2. Materials and methods

2.1. Set up and operational details of experimental system

The UAFB reactor used in this study was made up of glass column of 50 mm diameter and 335 mm height packed with chemically inert insulated beads to a height of 255 mm which served as a supporting media and the set up was maintained at room temperature (30 °C). The empty bed volume and the void volume of the reactor were 760 and 360 mL, respectively. The reactor was wrapped with black paper to prevent the photo oxidation of formaldehyde. The reactor was fed from the bottom and the effluent was collected from the outlet provided at the top portion of the reactor. Fig. 1 represents the schematic of the reactor. In the beginning of the experiment, the reactor was fed with municipal sewage from a sewage treatment plant for the development of biofilm. The formation of active biofilm was indicated by high and consistent removal of COD. Acclimation of the reactor to the formaldehyde was carried out by gradually increasing the concentration of the substrate and reducing the sewage in the feed. COD concentration was gradually increased to 1000 mg L^{-1} of formaldehyde.

The formaldehyde containing wastewater used in this study was collected from paint industry situated in Tamilnadu. The characteristics of formaldehyde wastewater used in the study are given in Table 1. The wastewater was acidic in nature with COD of the wastewater ranged from 10,976 to 11,840 mg L^{-1}. The formaldehyde concentration was 8400–8545 mg L^{-1}. The wastewater dose not has nitrogen and phosphorus, and di-ammonium hydrogen phosphate (DAP) was added as nutrient. The dilute wastewater was fed along with nutrients after pH adjustment to 7.0, as biological biomass is active at this pH.

2.2. Analytical methods

The pH, volatile suspended solids (VSS), chemical oxygen demand (COD) and chloride were determined according to Standard Methods [13]. Low concentration of formaldehyde was determined by the colorimetric method using chromotrophic acid [14] and higher concentration by titremetric method using sodium sulphite [15].

3. Results and discussion

3.1. Effect of initial COD on formaldehyde and COD removal

The removal of organic substrate by heterogeneous microorganisms in UASB reactor can be determined on the basis of COD removal rate as a function of the substrate concentration. The reactor was operated with increase in COD concentration from 100 to 2000 mg L^{-1} at HRT of 14 h. All the results of COD and formaldehyde removal efficiencies at different organic loading rates ranging from 0.18 kg COD $m^{-3}\,d^{-1}$ and 3.6 kg COD $m^{-3}\,d^{-1}$ during the experimental studies are given in Table 2. The steady state removal of formaldehyde and COD with different influent COD concentrations is presented in Fig. 2. COD and formaldehyde removal efficiency decreased with increase in COD concentration and it was found to be 92–24% and 99–41%, respectively. The reactor was functioning efficiently up to influent COD of 1000 mg L^{-1}. Fig. 3 represents the COD loading removal rate as a function COD loading. The COD removal rate increases with increase in COD load-

Table 2
Performance of UAFB reactor during the experimental study (14 h HRT)

OLR (kg COD $m^{-3}\,d^{-1}$)	COD (mg L^{-1})		Removal (%)	HCHO (mg L^{-1})		Removal (%)
	Inf	Eff		Inf	Eff	
0.18	100	8	92	65	1	99
0.45	250	22	91	97	2	98
0.90	500	46	91	242	9	96
1.80	1000	117	88	408	40	90
2.71	1500	640	57	686	82	88
3.61	2000	1508	24	918	542	41

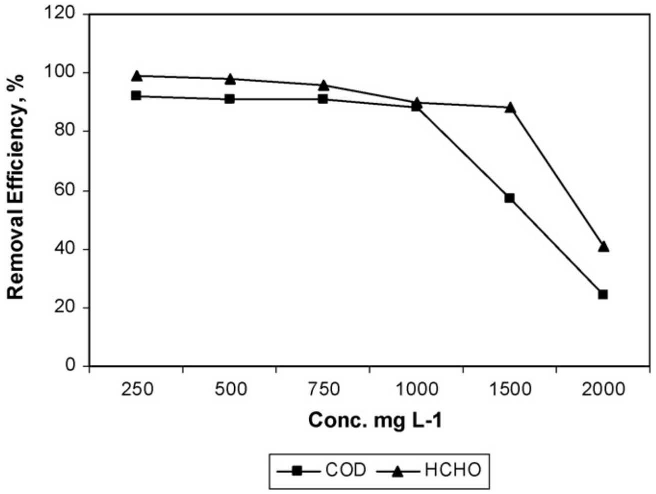

Fig. 2. COD and formaldehyde removal at different COD concentrations in UAFB reactor (HRT-14 h).

ing from 0.18 kg COD m^{-3} d^{-1} to 1.8 kg COD m^{-3} d^{-1} and decreases with further increase in COD loading from 2.7 kg COD m^{-3} d^{-1} to 3.6 kg COD m^{-3} d^{-1}. del Pozo et al. [16], while working on anaerobic slaughter house wastewater using fixed film reactor have shown that COD removal efficiencies ranging from 85% to 95% were achieved for organic loading rate of 8 kg COD m^{-3} d^{-1}, while the highest organic loading rates (35 kg COD m^{-3} d^{-1}) lead to decrease in efficiencies from 55% to 75%. Similarly the COD removal efficiencies were consistently over 96% for a loading from 15.8 g COD m^{-3} d^{-1} as reported by Shin et al. [17]. At higher loading rates over 18.47 g COD m^{-3} d^{-1}, the COD removal decreased due to sludge flotation and washed out in the UASB reactor [17]. The decrease in COD removal from 90% to 23% with the increase in organic loading was observed by Almendariz et al. [18]. The removal of formaldehyde by adsorption in the reactor was considered negligible although no experimental tests were concluded. The system was properly sealed to prevent loss due to evaporation.

3.2. Effect of hydraulic retention time on COD and formaldehyde removal

The reactor was continuously operated at different HRT (6–24 h) and at initial COD concentration of 500 mg L^{-1}. Fig. 4 depicts the percentage removal of formaldehyde and COD in the reactor with variation in HRT. Degradation of formaldehyde and COD decreases with decrease in HRT. When HRT was decreased from 24 to 6 h the formaldehyde and COD removal decreased from 99% to 83% and 91–31%, respectively. At 10 h HRT the COD and formaldehyde concentrations were 500 and 60 mg L^{-1} which was removed efficiently in UAFB with the treated effluent COD of 25 mg L^{-1}. The decrease in COD removal was observed for the decrease in HRT from 24 to 6 h, respectively. At 24 h HRT, COD remaining was very low (25 mg L^{-1}) indicating efficient functioning of reactor with optimal COD removal. Elmitwalli and Otterpohl [19] have operated UASB for the treatment of grey water at different HRT of 16, 10 and 6 h and observed decrease in the total COD removal from 64% to 52% with decrease in HRT. In the treatment of low concentration industrial chemicals mixture using UASB, reactor Castila et al. have shown the decrease in COD removal efficiency from 90% to 74% with decrease in HRT from 12 to 4 h [20]. As the COD loading increases from 0.5 to 0.90 kg COD m^{-3} d^{-1}, the COD loading removal rate also increases to some extent, but further increase in the loading shows the decrease in removal rate correspondingly as represented in Fig. 5.

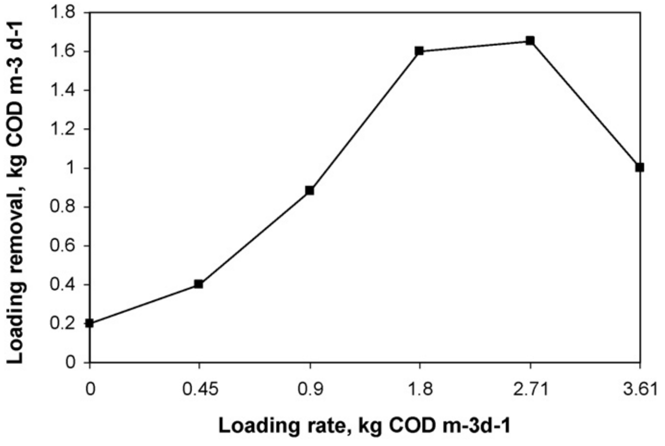

Fig. 3. COD removal as a function of COD loading rate (HRT-14 h).

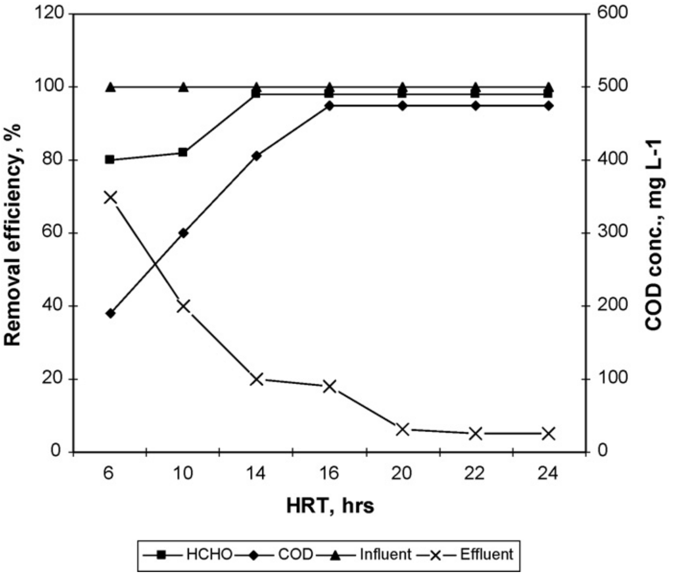

Fig. 4. COD and formaldehyde removal at different HRTs in UAFB reactor (COD conc. 500 mg L^{-1}).

3.3. Second-order kinetic application to the UAFB

There are several models such as Monod model, Second order kinetics model, Stover–Kincannon model, etc., which have been

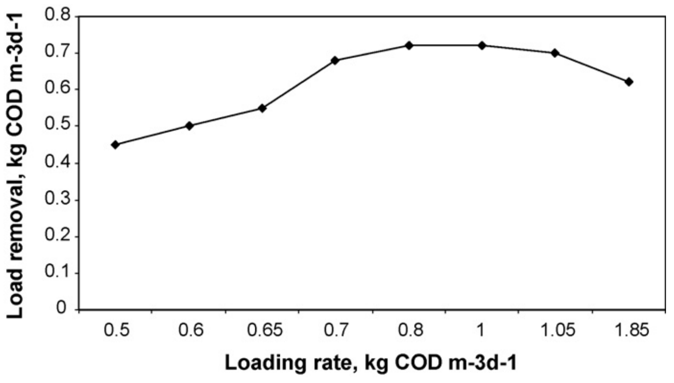

Fig. 5. Rate of COD loading removal as a function of COD loading rate.

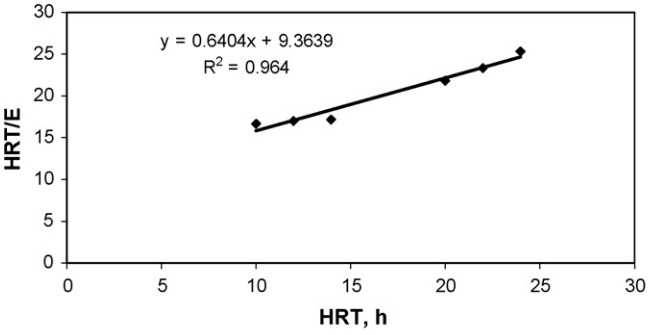

Fig. 6. Second-order kinetic model application for UAFB reactor.

used to describe the overall kinetics of biological reactor. The second-order model was applied to experimental results for UAFB treating formaldehyde containing waste waster. The general second-order kinetic model equation was represented as follows [21].

$$\frac{-\mathrm{d}s}{\mathrm{d}t} = K_{2(s)}X\left(\frac{S}{S_0}\right)^2 \tag{1}$$

After integration and linearilized, the equation was represented as

$$\frac{S_0\theta}{S_0 - S} = \theta + \frac{S_0}{K_{2(s)}X_0} \tag{2}$$

If the second term of the right part of this equation was accepted as a constant, equation given below will be obtained as.

$$\frac{S_0\theta}{S_0 - S} = a + b\theta \tag{3}$$

where $a = S_0/(K_{2(s)}\ X)$ and b is a constant greater then unity. $(S_0 - S)/S_0$ expresses the substrate removal efficiency and in symbolized as E. Therefore, the last equation can be written as follows:

$$\frac{\theta}{E} = a + b\theta \tag{4}$$

where S and S_0 are the effluent and influent substrate concentration ($\mathrm{mg\,L^{-1}}$); X, the average biomass concentration in the reactor ($\mathrm{mg\,VSS\,L^{-1}}$); θ, hydraulic retention time (h), and $K_{2(s)}$ is the second-order substrate removal rate constant ($\mathrm{d^{-1}}$).

Data used for a second-order kinetic model were given in Table 2 and (a) and (b) values were obtained using Fig. 6. for UAFB reactor. From the figure (a) and (b) values were found to be 0.64 and 9.36, respectively, with correlation coefficient of 0.96. Second-order substrate removal rate constants (K_2), which were calculated, were given in Table 3.

Table 3
Data for second-order kinetics model for UAFB

HRT (h)	S_0 ($\mathrm{mg\,L^{-1}}$)	S ($\mathrm{mg\,L^{-1}}$)	E (%)	θ/E	$K_{2(s)}$ ($\mathrm{h^{-1}}$)[a]
10	500	200	60	16.7	3.2
12	500	150	71.2	16.9	3.2
14	500	84	83.2	16.8	3.2
16	520	65	87.5	18.3	3.3
20	500	43	91.4	21.9	3.2
22	520	30	94	23.4	3.3
24	500	25	95	25.3	3.2
					3.2

The average biomass concentration in the reactor was 247 $\mathrm{mg\,VSS\,L^{-1}}$.
[a] Avg.

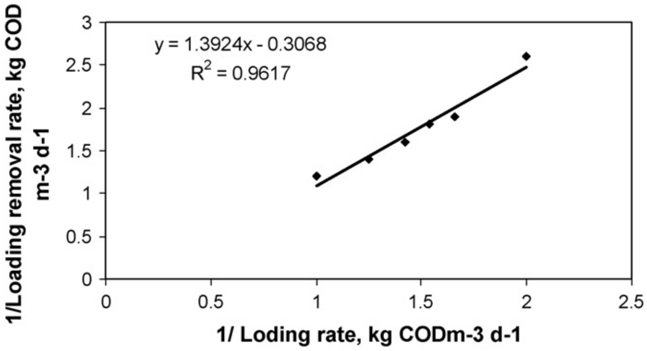

Fig. 7. Stover–Kincannon model application for UAFB reactor.

3.4. Modified Stover–Kincannon model for UAFB reactor

Monod type kinetic analysis based on COD loading and for organic substance removal in continuously operated anaerobic reactors has been developed [7,22,23]. Stover–Kincannon model proposed for rotating biological contractor (RBC) system [24]. However, for an anaerobic filter system the volume (V) of anaerobic filter is used instead of surface area. Previous studies by Yu et al. applied to the up flow anaerobic filter for soybean waste water treatment [25] and Sandhya and Swaminathan [21] to the textile wastewater treatment in hybrid column UAFB reactor have shown that fixed biomass on the media contribute a significant and stable removal efficiencies.

Equations of the modified Stover–Kincannon model were as follows:

$$\frac{\mathrm{d}s}{\mathrm{d}t} = \frac{Q}{V}(S_i - S_\mathrm{e}) \tag{5}$$

where as $\mathrm{d}s/\mathrm{d}t$ was defined in two way as follows:

$$\frac{\mathrm{d}s}{\mathrm{d}t} = \frac{U_{\max}(QS_i/V)}{K_\mathrm{B} + (QS_i/V)} \tag{6}$$

$$\frac{\mathrm{d}s}{\mathrm{d}t} = \frac{KXS_\mathrm{e}}{K_\mathrm{s} + S_\mathrm{e}}$$

$$\left[\frac{\mathrm{d}s}{\mathrm{d}t}\right]^{-1} = \frac{V}{Q(S_i - S_\mathrm{e})} = \frac{K_\mathrm{B}}{U_{\max}}\frac{V}{QSi} + \frac{1}{U_{\max}}$$

where $\mathrm{d}s/\mathrm{d}t$, substrate removal rate ($\mathrm{g\,L^{-1}\,d^{-1}}$) $U_{\max}$ maximum utilization rate constant ($\mathrm{g\,L^{-1}\,d^{-1}}$); K_B the saturation value constant ($\mathrm{g\,L^{1}\,d^{-1}}$); k the maximum rate of substrate removal ($\mathrm{L^{-1}\,d^{-1}}$); X the microorganism concentration (VSS) in the UAFB ($\mathrm{g\,L^{-1}}$); K_s the half-velocity constant (and V is the clean-bed volume of the reactor (L). Eq. (6) is a Monod model, while Eq. (5) results from a simple modification of Stover–Kincannon model. If $(\mathrm{d}s/\mathrm{d}t)^{-1}$ was taken as $V/[Q(S_i - S_\mathrm{e})]$, which was the inverse of the loading removal rate and this was plotted against the inverse of the total loading rate $V/(QS_i)$, a straight line portion of intercept $1/U_{\max}$ and a slope of $K_\mathrm{B}/U_{\max}$ results. From Fig. 7 ($K_\mathrm{B}/U_{\max}$) and $1/U_{\max}$ were 1.40 and 0.30, respectively with the high correlation of 0.96. The maximum removal rate constant ($U_{\max}$) was 3.4 $\mathrm{g\,L^{-1}\,d^{-1}}$ and the saturation value constant (K_B) was 4.6 $\mathrm{g\,L^{-1}\,d^{-1}}$ for the UAFB reactor. The $U_{\max}$ and K_B values obtained in this study were lower than values found by Yu et al. [25] and Buyukkamaci and Filibeli [26]. The possible reasons for the differences may be variation in reactor configuration, wastewater characteristics and microorganisms used in the study. Yu et al. [25] obtained value higher, which are $U_{\max} = 83.3\ \mathrm{g\,L^{-1}\,d^{-1}}$ and $K_\mathrm{B} = 85.5\ \mathrm{g\,L^{-1}\,d^{-1}}$. The values obtained in present studies are comparable with Oliveira et al. [7] for the formaldehyde degradation using an anaerobic packed bed-bioreactor and in the biodegradation of resorcinol, catechol and hydroquinone in anaerobic up flow fixed film fixed bed reactor [27].

The performance relationship expressed by Eq. (6), provide good estimates of the performance of UAFB treating formaldehyde/methanol wastewater. Stover–Kincannon [24] have shown that the relationship developed from the laboratory scale experiments could be used for all media. The results from the present study on UAFB are in agreement with Stover–Kincannon. This again approves to support the applicability of the concepts and methodology used in Stover–Kincannon model.

4. Conclusion

The result obtained in this study of formaldehyde containing wastewater in an UAFB reactor led to the following conclusions.

The UAFB reactor was suitable for the treatment of formaldehyde containing wastewater. Efficient formaldehyde and COD degradations were achieved by applying HRT 14 h for influent COD concentrations ranging from 100 to 1000 $\mathrm{mg\,L^{-1}}$ and further increase in COD up to 2000 $\mathrm{mg\,L^{-1}}$ leads to decrease in removal efficiency due to its toxic effect.

Biokinetics model such as second order kinetics, Stover–Kincannon model were applied for the UAFB reactor. The second-order substrate removal rate constant ($K_{2(\mathrm{s})}$) was 3.2 $\mathrm{h^{-1}}$ for UAFB reactor. Modified Stover–Kincannon model to UAFB reactor, maximum removal rate constant ($U_{\max}$) and saturation value constant (K_{B}) were 3.4 and 4.6 $\mathrm{g\,L^{-1}\,d^{-1}}$, respectively.

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