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Influence of Mathematical – Kinetic Models for the Removal of Organics and Nutrients from Municipal Wastewater in Moving Bed Biofilm Reactor

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***Abstract* –** The moving bed biofilm reactor (MBBR) is a recent solution alternative to conventional wastewater treatment process. In this study, quantitative steady state mathematical kinetic models (Monod, modified Stover-Kinacannon and Michaelis Menten) were used which describes the removal of organics and nutrients in MBBR and its consequences for the principal parameters of the process like effluent quality and oxygen consumption. The model incorporates the mechanisms of diffusive mass transport and substrate utilization kinetics. The model solutions included the concentration profiles, the growth of suspended and attached biomasses and the effluent concentrations of chemical oxygen demand (COD), biochemical oxygen demand (BOD), total Kjeldahl nitrogen (TKN) and total phosphorus (TP). A lab-scale MBBR was set up to verify the three different kinetic model systems. The experimental results of effluent shows a good agreement with model prediction. Data analysis indicates that modified Stover-Kinacannon and Michaelis Menten's kinetic model can produce the best fit with the experimental results. The correlation between the experimental values of the process variables and the values predicted by the model was found excellent (> 90). The approaches of modeling and experiments presented in this study could be employed for the design of a full-scale MBBR process to remove organic matter and nutrients from wastewaters.

***Keywords* –** Moving bed biofilm reactor; Kinetics; Organic loading; Removal efficiency; Biofilms; Microbial growth

I. INTRODUCTION

In the course of the last few decades, it has become a need to improve the performance of existing municipal wastewater treatment plants. To achieve this, the development of more advanced treatment technologies is necessary in order to comply with the restrictive effluent limits and water quality guidelines that could be imposed in the coming future [1]. The organic matter content and the enrichment of nutrients in water bodies, like phosphorus and nitrogen are the major factors that influence the water quality [2]. Therefore, it is necessary to remove these contaminants from the wastewater in order to reduce the damage caused to the environment and public health [3].

Biological secondary treatment is the main process to remove the organic contaminants from the municipal wastewater [1]. Biological processes are classified into two different types: suspended biomass and attached biofilm processes. Biofilm is a complex surface-attached or associated with the interfaces of microbial communities formed in response to specific environmental conditions, such as nutrient and oxygen availability (Figure 1) [4], [5]. MBBR is an advanced treatment technology which provides the coexistence of both suspended and attached biofilm kinds of biomass, that could lead to the elimination of organic matter and nutrients present in municipal wastewater. Additionally, simultaneous nitrification and de-nitrification can also be successfully achieved since slow growing micro-organisms (nitrifiers) are retained by the biofilm growing over the biocarriers provided in the reactor [3], [6], [7]. Aerobic biodegradation rate is strongly affected by the DO concentration controlled by the aeration rate [8], [9].

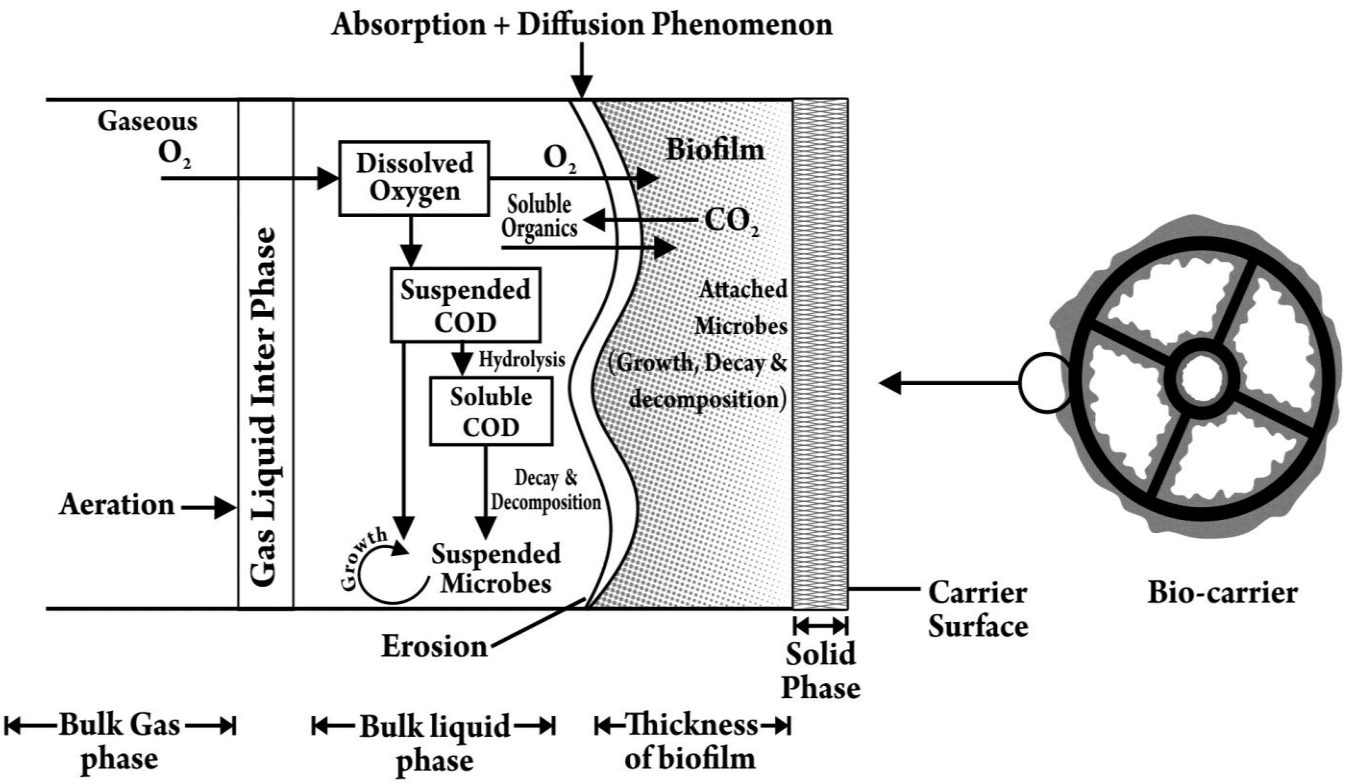

Figure 1 Schematic diagram of substrate penetration profile in a biofilm matrix

The MBBR process is relatively novel from the point of view of the kinetics and modeling. The modeling of MBBR systems remains very challenging to process engineers [10]. The kinetic parameters of both attached and suspended biomasses, modeling and dynamic simulation are an important tool for the design, operation and optimization of MBBR for wastewater treatment processes [11]. The kinetic models contribute to the understanding, determination and assessment of organic matter degradation in a biological reactor. Many studies have been published related to the performance evaluation, efficiency and application of MBBR process, but very little efforts have been made to describe the kinetic behavior and modeling of this biological reactor [12].

1.1 Kinetics of biofilm reactor

Biofilm kinetics can help to describe the substrate removal rate and the parameters which could affect the transport phenomenon in microbial films. Therefore, it is very useful to study and understand the mechanisms that control the process [7], [13]. To confront the various unique features of biofilm, numbers of models have been proposed in the literatures, such as first-order substrate removal model, Monod, Stover-Kincannon, Michaelis Menten's, Opaken and Grau second order model, to describe the overall kinetics of biofilm reactors [12], [14].

Substrate transport to cells in biofilms is essential to maintain a viable biofilm for wastewater treatment. Aggregation of cells creates significant gradients in substrate concentrations. Mass transport of bulk substrate from outside the biofilm to inside is driven by concentration differences. Bacteria on the inside of biofilms are often exposed to substrate concentrations substantially lower than that measured in the bulk liquid. Therefore, the rates of substrate utilization and cell growth are not uniform throughout the depth of a biofilm, but depend on the cell location within the film [15], [16]. Three concentration profiles that are possible in a biofilm are noted in Figure 2. They are (1) deep biofilm, in which the substrate concentration approaches zero at some point in the biofilm, (2) shallow biofilm where substrate concentration in biofilm ($S_{f)}$ remains above zero at all points in the film, and (3) fully penetrated biofilm, which occurs when the substrate concentration has negligible gradient [16].

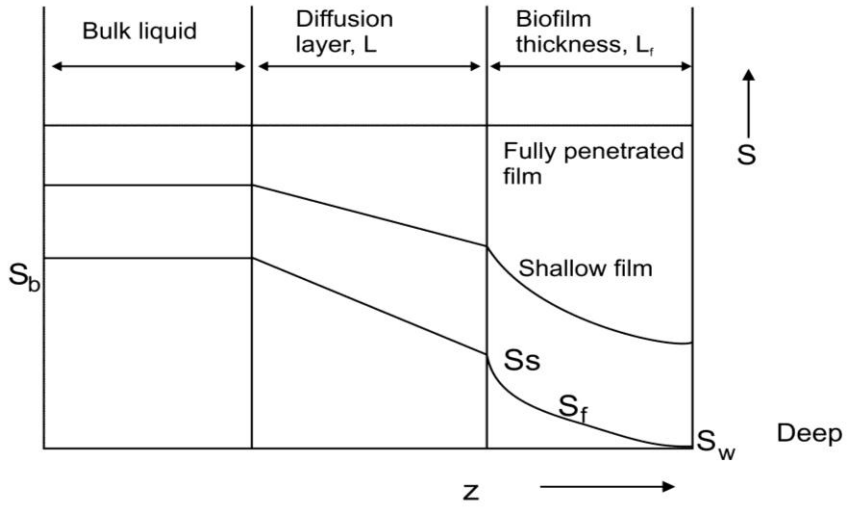

Figure 2 Substrate penetration profile in a biofilm structure

where S_b is effluent or bulk substrate concentration (as mg L^{-1}), S is concentration of rate-limiting substrate in the bulk liquid (as mg L^{-1}), Ss is readily biodegradable soluble substrate concentration (as mg L^{-1}), S_f is substrate concentration within the biofilm (as mg L^{-1}).

The substrate removal kinetics in biofilm applications is strongly dependent on the concentration of substrate in the wastewater being treated. This is illustrated in Figure 3, which shows the development of the kinetic description from a first (1') order expression at low concentrations to a zero (0') order expression at very high concentration. The transition from low to very high substrate concentration is described with a half (½') order expression [17], [18], [19].

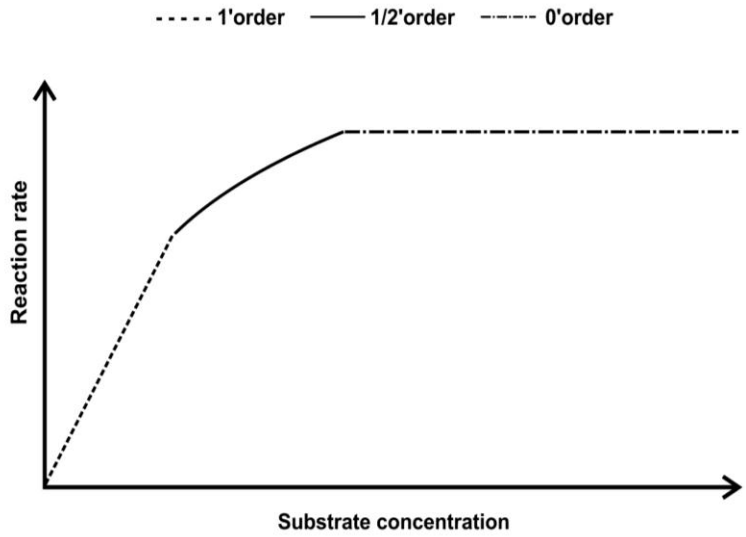

Figure 3 The kinetic description with reaction rate as a function of the substrate concentration

The above graph also depicts that the substrate removal rate is limited by the substrate concentration only at low concentrations where a small change in concentration gives a proportional change in the degradation. At high substrate concentrations, the rate is limited by the diffusion of substrate into the biofilm. Thus, as the concentration increases, the kinetics begins to shift from being concentration dependent to being diffusion dependent and eventually the kinetics becomes independent of the substrate concentration, this is described by half (1/2') order kinetics. At very high substrate concentration, the enzymatic efficiency restrains the removal rate—zero (0') order dependence [7], [18], [19].

Performance of any wastewater treatment process is typically evaluated by using analytical measurements such as BOD, COD, TKN and TP. These measurements provide information on the macroscopic performance of bioreactor, but provide limited quantitative information on cellular biological activity [20]. The principal aim was to study different kinetic-mathematical models that could describe the behavior of the bioreactor, kinetic parameters which characterize the biomass, cellular growth and availability of limiting substrate. The purpose of this objective was to find the best suitable model which could closely follow the experimental results and could describe the overall kinetics of the MBBR system treating municipal wastewater. The second objective of this investigation was to quantify organic matter and nutrient removal capacities so that the approaches of modeling and experiments could be employed for designing a full-scale MBBR process.

II. MATERIAL AND METHODS

2.1 Experimental setup

In order to study the various factors influencing the reactor kinetics, a laboratory scale rectangular reactor (length 60 cm; width 35.5 cm and height 14 cm) with a liquid volume of 30 L was made by using acrylic sheet. The effective working volume (total liquid volume excluding volume of the plastic media) was 20 L. The reactor was filled with commercially available high density polyethylene (HDPE) biofilm carriers (density: < 0.97 g cm^{-3}; diameter: 22 mm; height: 20 mm; thickness: 1 mm; specific gravity: 0.90 – 0.95 g cm^{-3} and specific surface area: 450 m^2 m^{-3}). These suspended carriers were used in the reactor to enlarge the surface area for better microorganism growth and accommodation. An aeration pump was used to supply air and for keeping the sludge in suspension. The schematic of the experimental setup used is shown in Figure 4 and the actual photograph of the bioreactor and biocarriers is shown in Figure 5.

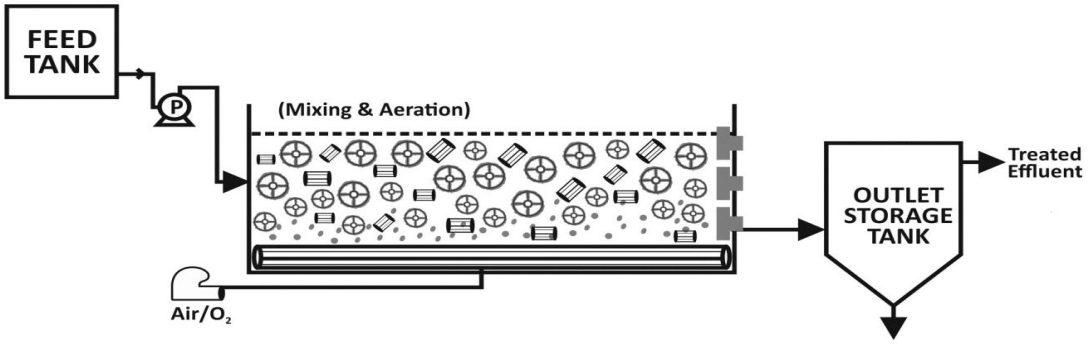

Figure 4 Schematic of the experimental setup

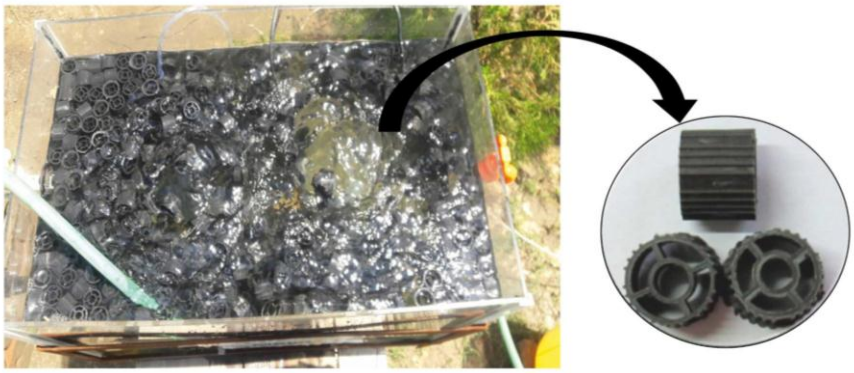

Figure 5 Real photograph of moving bed biofilm reactor and polyethylene biocarriers used for the study

2.2 Wastewater characteristics

The reactor was continuously fed with synthetic municipal wastewater with the help of peristaltic pump (flow rate: 15 mL min^{-1}) and control valve in the middle ports of the reactor. The synthetic wastewater used throughout the experiments contained CH_4N_2O: 91.74 mg L^{-1}, NH_4Cl: 12.75 mg L^{-1}, $C_2H_3NaO_2$: 79.37 mg L^{-1}, peptone: 17.41 mg L^{-1}, $MgHPO_4.3H_2O$: 29.02 mg L^{-1}, KH_2PO_4: 23.40 mg L^{-1}, $FeSO_4.7H_2O$: 5.80 mg L^{-1}, starch: 122.0 mg L^{-1}, glucose: 120 mg L^{-1}, yeast: 52.24 mg L^{-1} and trace elements solution was composed of $Cr(NO_3)_3.9H_2O$: 0.770 mg L^{-1}; $CuCl_2.2H_2O$: 0.536 mg L^{-1}; $MnSO_4.H_2O$: 0.108 mg L^{-1}; $NiSO_4.6H_2O$: 0.336 mg L^{-1}; $PbCl_2$: 0.100 mg L^{-1} and $ZnCl_2$: 0.208 mg L^{-1} [21].

2.3 Initiation of the reactor under optimal conditions

The reactor was filled with an optimized biocarrier filling ratio of 30%. To achieve even distribution of oxygen in the reactors, the residence time as well as the contact area to volume ratio was increased by fine air bubbles. To initiate biological growth and development of the biofilm, the reactor was inoculated with mixed culture of biomass. In order to acclimate the wastewater and microbial growth, the COD:N:P ratio was maintained as 100:14:2. Intermittent air was introduced into the reactor with an air pump at the bottom of the reactor with a flow rate of 0.21 m^3 h^{-1}. The dissolved oxygen (DO) concentration was normally kept above 4 mg L^{-1} throughout the experiment [3], [9]. The pH varied from 6.8 to 8.0. This study was performed under ambient conditions.

2.4 Analytical methods

Before starting the experiment, reactor was acclimatized with biomass for almost 15-20 days. Then the experimental study was carried out for 15 days. The influent and effluent DO, COD, BOD, TKN and TP concentrations were measured in accordance with the standard methods for the examination of water and wastewater [22]. DO, temperature and pH values were measured with a DO meter, digital thermometer and pH meter, respectively. All experiments were conducted until the system reached steady state in terms of effluent concentration.

III. RESULTS AND DISCUSSION

The substrate removal kinetics in biofilm applications is strongly dependent on the concentration of substrate in the wastewater being treated ([7], [19], [23]. Among the most widely used models, Monod, advanced Stover-Kincannon, and Michaelis Menten's kinetic models were selected and evaluated for considering organic load and nutrient removal in a MBBR. The results and interpretation of these models are described below:

3.1 Monod model

Monod's growth model was proposed as an empirical model to describe microbial growth kinetics [15], [24], [25], [26]. The model defines the relationship between the growth rate and the concentration of the limiting nutrient:

$$\frac{dS}{dt} = \frac{Q}{V}(S_i - S_e) = \frac{U_{max}(\frac{QS_i}{V})}{K_B + (\frac{QS_i}{V})} \quad (1)$$

where, *dS/dt* is the specific substrate utilization rate (g L^{-1}.day^{-1}), S, S_i and S_e are the concentrations of volatile suspended solid (VSS) in the feed, reactor and reactor effluent (g VSS L^{-1}), Q is flow rate (L day^{-1}), V is reactor volume (L), K_B is effective half saturation coefficient (g L^{-1}.day) and U_{max} is maximum utilization rate constant (g L^{-1}.day).

The half saturation coefficient (K_B) in the Monod equation is the concentration that gives one-half the maximum specific growth rate. For suspended growth and simple substrates, where mass transport is often disregarded, K_B values are usually low [15]. The important features of the model are that the growth rate is zero when there is no substrate and tends to an upper limit when the substrate is in great excess.

Under steady state conditions, the rate of change in substrate concentration (*dS/dt = 0 and -dS/dt =0*) is negligible and HRT is defined as the volume of the reactor divided by the flow rate of the influent, then following equation could be obtained by rearranging Equation (1) in to Equation (2):

$$\frac{S_i - S_e}{HRT} = K_B S \quad (2)$$

The value of the first-order kinetic constant can be obtained by plotting $(S_i - S_e)/HRT$ versus *S*, according to Equation (2). The value of K_B is obtained from the slope of the straight line. From Figure 6, K_B value can be estimated as 19.48, 16.66, 2.51 and 0.28 per day with a correlation coefficient of 0.527, 0.531, 0.646 and 0.659 per day for the corresponding concentrations of COD, BOD, TKN and TP, respectively. The high decay coefficient values might be due to substantial decay of cells that occur as a result of endogenous respiration. It also heavily depends on substrate utilization rate [27], [28], [29]. So, the higher yield coefficient values obtained in the study could be due to relatively larger proportion of biodegradable organic matter, while synthesized into new cells. The low value of the coefficient (R^2) indicates that first order kinetics cannot be applied with good precision.

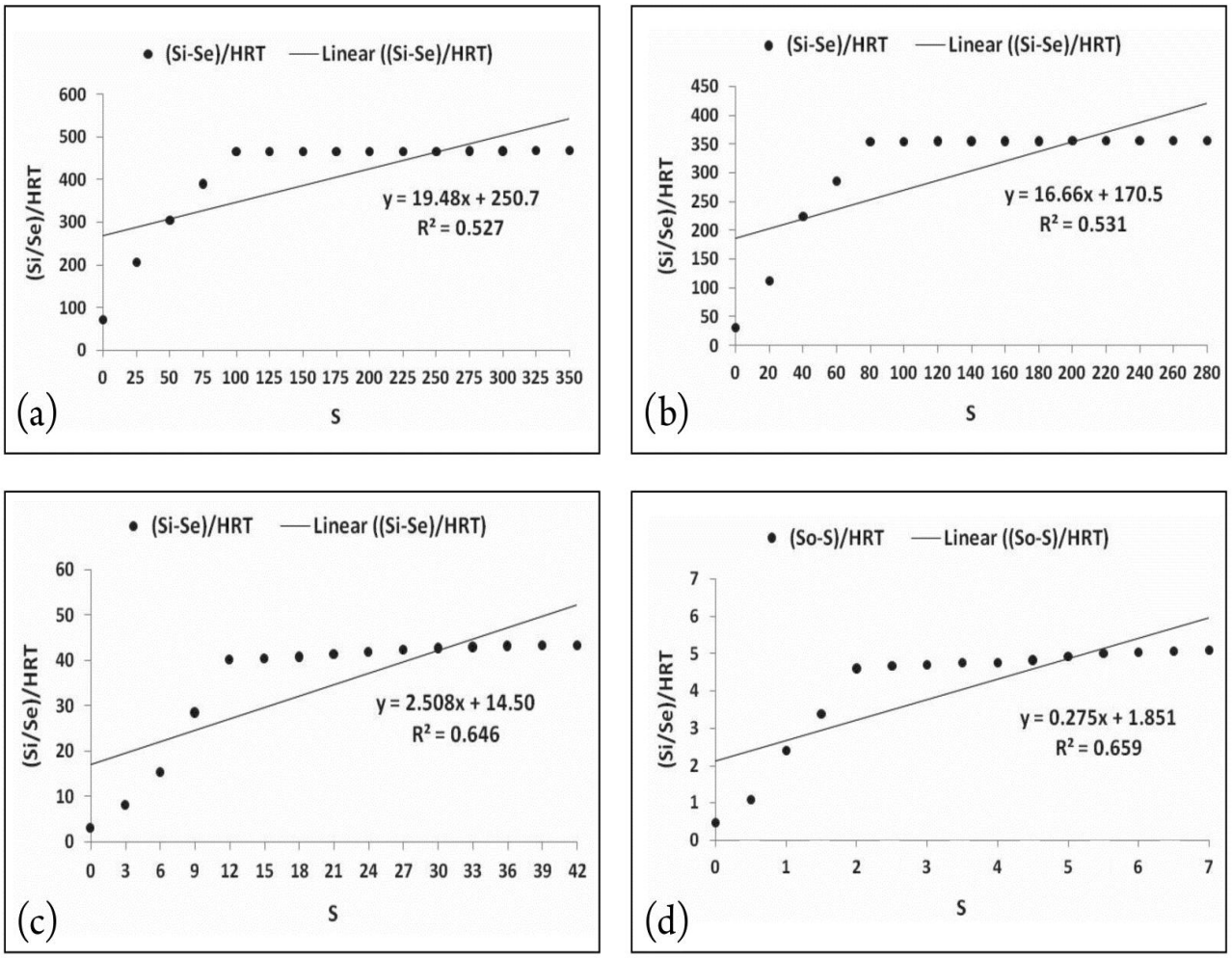

Figure 6 Determination of maximum specific growth rate and correlation between Monod model and experimental data for (a) COD, (b) BOD, (c) TKN and (d) TP removal from MBBR

3.2 Modified Stover-Kincannon model

In this kinetic model, the substrate utilization rate is expressed as a function of the organic loading rate (OLR) which is considered as the most important parameter influencing the behavior of the MBBR operating under steady-state conditions [10], [12]. Because of the difficulties in measuring the active surface area of the biocarriers, which supports the biofilm growth, the reactor's effective volume is used for this model [13], [30]. This modified model can be expressed as follows:

$$\frac{ds}{dt} = \frac{U_{max}\ (\frac{QS_i}{V})}{K_B + (\frac{QS_i}{V})} \tag{3}$$

Here *dS/dt* , the rate of substrate utilization is defined as:

$$\frac{ds}{dt} = \frac{Q}{V}(S_i - S_e) \tag{4}$$

where, *dS/dt* is the specific substrate utilization rate (g VSS L^{-1}.day^{-1}), U_{max} is maximum utilization rate constant (g L^{-1}.day), Q is flow rate (L day^{-1}), V is reactor volume (L), K_B is saturation constant (g L^{-1}.day), S_i is influent COD concentration and S_e is effluent COD concentration (g VSS L^{-1}). Linearization of the above Equation (3) and (4) gives the following relationship:

$$\left(\frac{dS}{dt}\right)^{-1} = \frac{V}{Q(S_0 - S)} = \frac{K_B}{U_{max}}\left(\frac{V}{QS_0}\right) + \frac{1}{U_{max}} \tag{5}$$

The plot of $V/[Q(S_i - S_e)]$, inverse of the removal rate, versus $V/(QS_i)$, inverse of the total loading rate, results in a straight line. A least squares linear regressions were applied. Maximum utilization rate (U_{max}) and saturation constant, (K_B), were calculated from the intercept and slope of the line, respectively. The substrate balance for the reactor can be written as follows:

$$QS_i = QS_e + V\frac{dS}{dt} \quad (6)$$

Substituting of Equation (5) into (4) gives

$$QS_i = QS_e + \frac{U_{max}\left(\frac{QS_i}{V}\right)}{K_B+\left(\frac{QS_i}{V}\right)} \times V \quad (7)$$

This expression can then be solved for either the effluent substrate concentration (Equation (8)) or the required volume of the reactor (Equation (9)) by substituting kinetic constants U_{max} and K_B.

$$S_e = S_i - \frac{U_{max}S_i}{K_B+\left(\frac{QS_i}{V}\right)} \quad (8)$$

$$V = \frac{QS_i}{\left[\frac{U_{max}S_i}{S_i-S_e}\right]-K_B}\text{B} \quad (9)$$

The modified Stover–Kincannon model was applied to experimental results of the continuously operated bioreactor system for organic and nutrient load removal. Measuring the slope and intercept of lines in Figure 7, the K_B and U_{max} values were determined and so were calculated as 11.20 and 10.13 g L^{-1} day^{-1} for COD, 8.12 and 9.45 g L^{-1} day^{-1} for BOD, 4.25 and 5.34 g L^{-1} day^{-1} for TKN and 1.53 and 2.76 g L^{-1} day^{-1} for TP, respectively. Therefore, the regression lines had a R^2 of 0.953, 0.961, 0.933 and 0.970 for COD, BOD, TKN and TP, where R is degree of regression. The effluent concentrations of COD, BOD, TKN and TP can then be predicted by using Equations (10-13). The saturation value indicates the substrate removed by microorganisms and the maximum utilization rate shows the maximum substrate removed by aerobic organisms versus time.

$$S_e = S_i - \frac{10.13\ S_i}{11.20+\left(\frac{QS_i}{V}\right)} \quad (10)$$

$$S_e = S_i - \frac{9.45\ S_i}{8.12+\left(\frac{QS_i}{V}\right)} \quad (11)$$

$$S_e = S_i - \frac{5.34\ S_i}{4.25+\left(\frac{QS_i}{V}\right)} \quad (12)$$

$$S_e = S_i - \frac{2.76\ S_i}{1.53+\left(\frac{QS_i}{V}\right)} \quad (13)$$

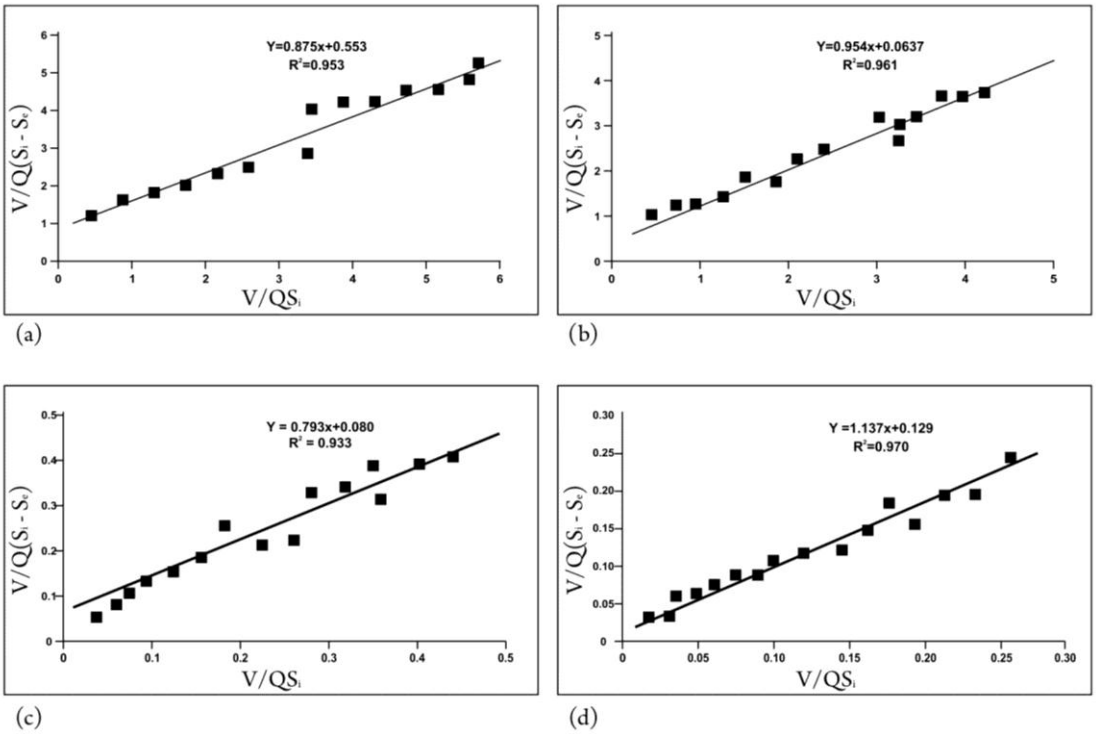

Figure 7 Determination of maximum specific growth rate (U_{max}) and saturation constant (K_B) for the modified Stover–Kincannon model for (a) COD, (b) BOD, (c) TKN and (d) TP removal

In an aerobic MBBR system, Hosseiny and Borghei [13] also conducted the similar work and observed K_B and U_{max} values as 9.45 and 8.3 g L^{-1} day^{-1}, respectively. Their findings indicated that Stover-Kincannon model is more applicable model for describing the kinetics of organic removal in MBBR. Similarly, by using the modified Stover–Kincannon model, Sarariah and Chakraborty [31], also reported maximum K_B and U_{max} values as 13.39 and 10.537 g L^{-1} day^{-1}.

The substrate removal rates, TKN and TP loading rates were calculated at the studied RRT and initial concentrations. The variation of substrate removal rates of COD, BOD, TKN and TP with effluent concentration is shown in Figure 8.

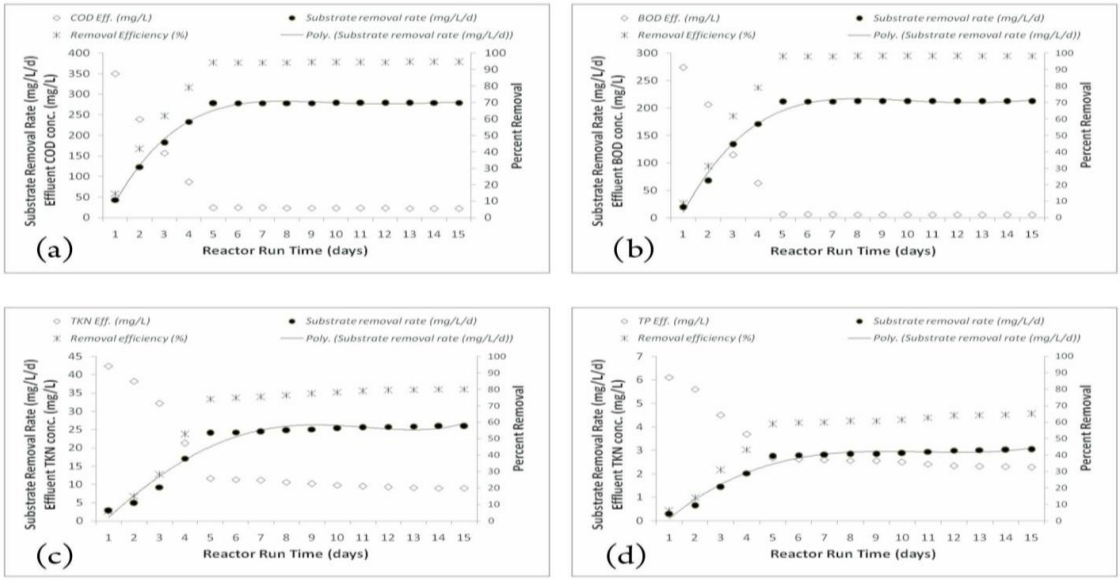

Figure 8 Modified Stover-Kincannon model plot for (a) COD, (b) BOD, (c) TKN and (d) TP removal from MBBR

3.3 Michaelis Menten's kinetic model

The kinetics of the organic matter and nutrient removal was also determined by fitting the data through the Michaelis Menten's kinetic model by using the following zero order (14) and first order (15) equations:

$$C = C_o - kt \quad (14)$$

$$C = C_o e^{-kt} \quad (15)$$

where, C is substrate concentration at time 't' (in mg L^{-1}), C_o is initial substrate concentration (in mg L^{-1}), k is rate constant and t is time (in days).

The organic load (mainly COD and BOD) grossly indicate the strength of sewage compositions. Hence, the effect of carrier filling rate on the bioreactor performance in terms of COD, BOD, TKN and TP removal was studied for the duration of 15 days using an optimized biocarrier filling ratio of 30%. Maximum organic load removal rate was observed at this filling ratio with respect to the kinetics as mixing intensity of carriers was found best which accelerated the renewal of surface or gas-liquid interphase, resulting in more oxygen to get absorbed and dissolved in the wastewater. The DO was observed to be in the range of 4.00 – 6.00 mg L^{-1} in the reactor. The high-density population of bacteria requires high oxygen demand to achieve high-rate biodegradation productivity within the system [7]. But, the increased RRT (or HRT) formed a little thick layer of biofilm around the carrier surface, which resulted in less penetration of oxygen and organic matter inside the deeper layer of biofilm [9], [32]. The various effluent parameters observed and the monitored values are mentioned in Table 1. This table shows that the maximum COD, BOD, TKN and TP reduction values were observed to be 94 ± 0.5%, 97 ± 1.3%, 74 ± 0.2% and 58 ± 1.2%, respectively. Whereas, the mean COD, BOD, TKN and TP values leaving the biological reactor were 22.0, 5.0, 9.0 and 2.3 mg L^{-1}, respectively.

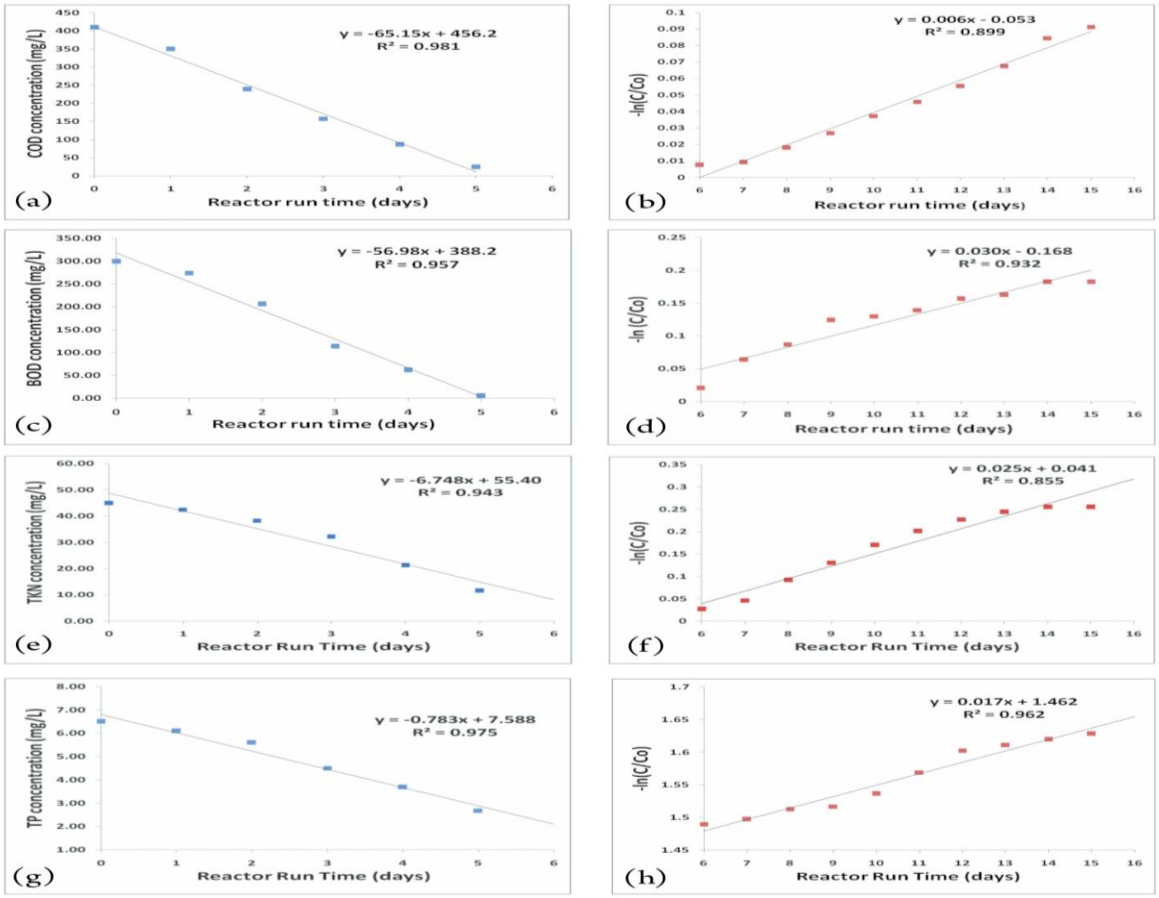

Figure 9 Zero order kinetics for (a) COD (c) BOD (e) TKN (g) TP removal and first order kinetics for (b) COD (d) BOD (f) TKN (h) TP removal by using Michaelis Menten's kinetic model

Figure 9 shows the trend of COD, BOD, TKN and TP removal rate in a bioreactor for the duration of 15 days. Rate of zero order varies from high substrate concentration of 410.00 to 24.10, 300.00 to 6.00, 45.00 to 11.62 and 6.50 to 2.68 mg L^{-1} for COD, BOD, TKN and TP removal, respectively, whereas the first order reaction rate varies from 23.92 to 22.00, 5.88 to 5.00, 11.31 to 9.00 and 2.62 to 2.28 mg L^{-1} for COD, BOD, TKN and TP removal, respectively. It can be observed that initially at high concentration, the rate of substrate reduction follows the zero order kinetics and then first order at low substrate concentration. With an increase in the residence time (reactor run time), extremely complex substrate controls the reaction rate, but after some time, the steady state is reached. Because at high organic content, microorganisms present in the wastewater are saturated with the substrate that made the rate to be almost constant (i.e. zero order). As the organic concentration decreased, only few available sites of the microbes were covered and that made the rate of reaction to be proportional to the substrate concentration (i.e. first order). This substrate reduction behavior is described by Michaelis-Menten kinetics [33], [34] with value of R^2 = 0.981 (zero order) and 0.899 (first order) for COD removal, 0.957 (zero order) and 0.932 (first order) for BOD removal, 0.943 (zero order) and 0.855 (first order) for TKN removal, 0.975 (zero order) and 0.962 (first order) for TP removal, respectively. The rate constants (k) for zero order were 77.18, 58.80, 6.68 and 0.76 mg L^{-1}.day and for first order regions were observed to be 0.008, 0.016, 0.023 and 0.014 d^{-1} for the COD, BOD, TKN and TP removal, respectively. Summary of organic matter and nutrient removal kinetics is given in Table 1.

Table 1 Summary of Michaelis-Menten kinetic reaction rates for the organic matter (COD, BOD) and nutrient (TKN, TP) removal from the municipal wastewater

Parameter	Order	
	Zero	**First**
COD Removal	$410 - 77.18t$	$23.92\ e^{-0.008t}$
BOD Removal	$300 - 58.80t$	$5.88\ e^{-0.016t}$
TKN Removal	$45 - 6.68t$	$11.31\ e^{-0.023t}$
TP Removal	$6.50 - 0.76t$	$2.62\ e^{-0.014t}$

Conclusion

An assessment of lab-scale moving bed biofilm reactor with the optimized filling ratio (30%) of commercially available biofilm plastic carriers as a medium was done for the simultaneous removal of organic matter (COD and BOD) and nutrients (TKN and TP) in a single aerobic lab-scale reactor. Under steady state conditions, three different kinetic –mathematical models such as Monod, modified Stover-Kincannon and Michaelis Menten's were applied to compare and predict the performance of the MBBR in terms of removal efficiency. The reactor was capable of achieving high removal efficiency of organic matter and nutrients. The maximum removal efficiencies of COD, BOD, TKN and TP in a specific area and volumetric loadings were observed to be 94 ± 0.5%, 97 ± 1.3%, 74 ± 0.2% and 58 ± 1.2%, respectively. It was also concluded that both modified Stover-Kincannon and Michaelis Menten's kinetic models with correlation coefficients of greater than 90 were found to be more suitable than the other applied model i.e., Monod Model, for predicting the performance of MBBR together with significant kinetic coefficients. The kinetic parameters have an increasing dependence with respect to the reactor run time. The approaches of model and experiments developed in this study could be applied in the design of a full-scale MBBR treatment process to remove carbon, nitrogen and phosphorus simultaneously in the existing small-sized or medium-sized wastewater treatment plants where land availability is limited.

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