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Mathematical modeling of treatment of oily wastewater by biological method – A mini review

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With the increasing industrial and commercial activities, various toxic and inhibitory organic substances including oil and grease come out in wastewater generated. It is very challenging to remove oil and grease from wastewater as it falls under the category of hazardous waste. The parameters like COD, Total Petroleum Hydrocarbon (TPH), BOD etc. exhibit higher values due to presence of excessive organic matter in oily wastewater. Biological method is found to be effective for the removal of oil and grease from wastewater after pre-treatment. The knowledge of mathematical modeling and bio-kinetics is very important for design of biological system treating oily wastewater. It is also relevant for augmentation of numerous operational conditions in order to get good quality effluent. This review paper elucidates the source, characteristics and treatment approaches of oily wastewater, requirement of mathematical modeling together with model development and validation in case of biological process.

Keywords: Oily wastewater, biological treatment, mathematical modeling, model comparison, model validation.

Introduction

In the advent of growing population, urbanization and industrialization there is considerable generation of oily wastewater every year. Wastewater containing oil and grease is originated from kitchens of houses, restaurants, marine transport, garages, workshops, different industries such as food processing, cosmetic, metal finishing, petrochemicals and vegetable oil industries¹. The major parameters to be analyzed in oily wastewater include COD, oil and grease, BOD, TPH (Total Petroleum Hydrocarbon), TSS and dissolved solids. The treatment and disposal of this wastewater is a matter of concern as it is considered as a hazardous waste which detrimentally affects water, soil, air as well as human beings^{2,3}. Hence, suitable treatment techniques must be adopted on the basis of type and nature of oily wastewater.

There are different types of treatment methods adopted for oily wastewater such as physicochemical, biological and combined treatment methods based on the source and characteristics³. The cost of chemical and physical methods is high because of handling of chemical sludge and cost of different equipments as well as chemicals⁴. Hence, more preference should be given to biological methods for treatment

of oil and grease containing wastewater owing to its comparatively low cost and simplicity in operation.

Various biological treatment methods adopted so far for oily wastewater include activated sludge process (ASP), rotating biological contactor (RBC), upflow anaerobic sludge blanket (UASB), anaerobic baffled reactor (ABR), fixed bed reactor and sequencing batch reactor (SBR)^{3,5}. The biological treatment including activated sludge process, bio-filters, aerated pond and many more help in the removal of numerous organic contaminants in dissolved form including dissolved oil⁶. It was also reported by Primasari *et al.*⁷ that the utilization of facultative as well as anaerobic digestion helps in overcoming the pollution caused by the oily effluent.

However, in order to obtain effluent of superior quality by modifying operational conditions, utilization of appropriate treatment methods and mathematical models are extremely essential. Mathematical modelling is a process of representation, analysis and making predictions in order to provide insight into the real world phenomena by utilizing the concept of mathematics. It starts with defining a problem and continues with taking some assumptions, figuring out few key variables, analysis and assessment of model solution

and ends up with validation of model. Though a large number of mathematical models have already been implemented for biological treatment of wastewater, a very few of them have been developed for the treatment of oil and grease containing wastewater in biological reactor. The majority of the studies on mathematical modelling of biological treatment of oily wastewater have laid stress on degradation of several organic pollutants including oil and grease, total organic carbon (TOC), total petroleum hydrocarbon (TPH), COD, phenols and TSS^{6,8,9}.

It is important to estimate very precise as well as accurate model parameters from the experimental data. Model assessment involves two phases i.e. estimation of parameter and selection of model structure. Considering either aerobic or anaerobic type, various mathematical models have already been developed for treatment of oily wastewater^{6,8}. Setiadi *et al.*¹⁰ studied recycle impact on the performance of anaerobic baffled reactor for the treatment of palm oil mill effluents of P.T. Perkebunan XI (West Java) and focused on bio-kinetic parameters. Again, Faisal and Unno⁵ studied the treatment of wastewater coming out of palm oil mill (from Perkebunan Nusantara, Indonesia) by using one modified anaerobic baffled bioreactor (MABR) in steady state and also focused on bio-kinetic parameters. The biokinetic equations are essential for understanding substrate utilization rate, growth of biomass, mean cell residence time, food to microorganisms ratio and also utilized for design of various biological wastewater treatment systems¹¹. Another kinetic study

was performed by Nakhla *et al.*⁹ for aerobic biodegradation of pet food wastewater containing high amount of oil and grease in batch scale activated sludge reactor. Again, Santo *et al.*⁶ developed a mass balance model for treatment of petroleum refinery wastewater in activated sludge process.

The objective of the present review article is to investigate (a) nature and general characteristics of oil and grease containing wastewater, (b) scope of biological treatment for oily wastewater, (c) necessity of mathematical modeling especially for biological treatment of oily wastewater and (d) development and validation of various models obtained so far for biological treatment of such wastewater.

Characteristic features of oily wastewater

As oily wastewater contains huge amount of oil, water, other organic matter and sludge, the discharge of such wastewater is amenable to environmental pollution. Hence, it is essential to understand the characteristics of oily wastewater to prevent natural water bodies, aquatic life, air and soil from its adverse effect. The characteristics of oil bearing wastewater coming out of different sources reported by various authors have been presented in Table 1.

Treatability of oil and grease containing wastewater

Oil content from the wastewater can be reduced by utilizing different physical, chemical as well as biological methods. El-Naas *et al.*²⁰ found 63% COD removal from petroleum refinery wastewater in electrocoagulation process.

Table 1. Characteristics of various oily wastewater

Sources	Important characteristic parameters								Ref.
	pH	COD (mg/L)	BOD (mg/L)	TKN (mg/L)	Oil and grease (mg/L)	TS (mg/L)	TSS (mg/L)	NH ₄ ⁺ -N (mg/L)	
Palm oil mill effluents of P.T. Perkebunan XI (West Java)	3.3–4.6	15103–65100	8200–354000	12–126	–	16580–94106	1330–50700	2.5–5	10
Palm oil mill of Perkebunan Nusantara, Indonesia	4.8	16000	8700	179	410	–	–	–	5
Pet food wastewater (raw wastewater)	–	18850	8820	–	13500	–	14470	–	9
Palm oil mill effluent	3.8–4.4	42500–55700	23000–26000	500–700	4900–5700	–	16500–19500	–	14
Oil refinery, wastewater collected after primary treatment	6.7	373	165	26.1	291	–	461	–	6
Petrochemical wastewater	13	5260–12820	–	–	–	210–1026	530–4146	–	18
Metal working fluid from Automotive spare parts industry	8.8–9.05	63000–90000	6000–7000	–	700	–	2700–3400	–	19

Again, Gursoy-Haksevenler and Arslan-Alaton²¹ studied acid cracking as well as filtration technique for removal of suspended organic matter as well as oil and grease from palm oil mill wastewater. They reported removal of suspended organic matter and oil and grease as 96% and 95% respectively. Various other physical and chemical methods used for treatment of oil bearing wastewater include flotation, coagulation, ultrafiltration, reverse osmosis and many more³.

Setiadi *et al.*¹⁰ studied the performance of anaerobic baffled reactor for the treatment of palm oil mill effluents of P. T. Perkebunan XI (West Java). The removal efficiency was found to be (64.52–82.88)% for oil and grease, (41.66–86.04)% for BOD (total) and (59.6–80.5)% for soluble COD. Shariati *et al.*²² studied the removal of pollutants after treatment of synthetic petroleum wastewater by membrane sequencing batch reactor (MSBR). The aromatic as well as aliphatic hydrocarbon removal was found to be more than 97% irrespective of HRT. Again, the removal of (12.9–54.8)% oil and grease and (85.1–97.1)% of COD was obtained during treatment of oily wastewater from a food industry in aerobic biological process using stirred tank reactor. It was further observed that the percentage of removal was more with higher rate of aeration⁷.

Chunshuang *et al.*¹⁵ studied the performance of UASB reactor for the treatment of high saline wastewater obtained from heavy oil production process. The removal of COD was found to be 65.08% at HRT of more than 24 h under influent COD of (350–640) mg/L and the removal of oil was 74.33% at 112–205 mg/L initial oil concentration. This indicated efficient treatment of heavy oil produced wastewater in UASB reactor. Similarly numerous other biological methods used for the treatment of oil containing wastewater include biological aerated filter, SBR, CSTR, MBR, anaerobic ponds, anaerobic digesters, fluidized bed reactors, ASP and many more^{3,4,10}.

Though physicochemical processes remove free oil, colloidal and suspended solids, but cannot remove dissolved or emulsified oil which is possible to be removed by biological treatment²². Again, Kumar *et al.*²³ reported that physicochemical methods of treatment are energy intensive with poor recovery efficiency and disposal problem. The cost of these methods is also very high which include the cost of sludge disposal, chemicals and different equipments⁴.

The advantages of biological treatment towards oil and

grease removal have been reported by different researchers. It was mentioned by Jafarinejad²⁴ and Santo *et al.*⁶ that numerous dissolved organic contaminants including dissolved oil can be removed by biological treatment like ASP, bio-filters, aerated pond etc. from petroleum refinery wastewater. Again, biological treatment was reported to be extensively used treatment method to remove dissolved oil from oil refineries wastewater^{22,25}. The advantages and limitations of different biological treatment towards oil and grease removal are mentioned in Table 2.

The oily wastewater was treated successfully by many researchers utilizing numerous laboratory and pilot scale biological processes. Some of the laboratory scale studies include treatment of POME by ABR¹⁰, POME by MABR⁵, pet food wastewater by batch ASP reactor⁹, POME by UASFF, heavy oil production wastewater by UASB¹⁵, petroleum refinery wastewater by ASP⁶ and oil refinery wastewater by anaerobic digester¹⁷. Some of the pilot scale treatment include treatment of fish-canning wastewater by anaerobic biological process²⁶, oil field produced water by skimming and ASP²⁷, oil refinery wastewater by fixed film bio-reactor²⁸ and heavy crude oil wastewater by fixed film bioreactor²⁹. Some of the studies focused on both laboratory scale as well as pilot scale biological process for the treatment of oily wastewater¹³.

Mathematical models of biological treatment of oily wastewater

The mathematical models studied by different researchers on biological treatment of oily wastewater have mainly focused on COD as performance parameter. Setiadi *et al.*¹⁰ focused on the impact of recycle for the treatment of palm oil mill effluent in ABR using COD as the performance parameter and studied various bio-kinetic parameters. Again, Faisal and Unno⁵ considered influent COD as performance parameter for the treatment of palm oil mill wastewater under steady state condition in a modified anaerobic baffled bioreactor (MABR). Furthermore, Al-Zuhair¹² presented a kinetic model which was similar to Tsai and Chang enzymatic hydrolysis model using batch reactor to estimate kinetic parameters. Accurate representation of experimental data by that kinetic model was also observed. A study on the Monod model of zero and first order type for the treatment of high oil and grease by utilizing batch activated sludge treatment was

Table 2. Advantages and disadvantages of biological treatment towards oil and grease removal

Sl. No.	Biological treatment methods	Advantages	Disadvantages	Ref.
1.	Anaerobic ponds	Greater percentage of BOD reduction	More retention time Necessity of greater digestion volume Bad odour Problems in collection of methane gas	10
2.	Anaerobic tanks/digesters	Greater percentage of BOD reduction	More retention time Necessity of greater digestion volume	10
3.	Fluidized bed reactor	Scope of high biomass retention	Maintenance of biofilm thickness is difficult More energy requirement for fluidization of bio-particle	10
4.	Anaerobic baffled reactor (ABR)	Simple and economical in construction Stability against shock loading Greater volumetric rates	Substrate-biomass contact may not be adequate Higher energy for pumping to overcome headloss.	10
5.	SBR	Less space requirement Sequence time flexibility	It is difficult to reduce suspended solids to almost zero Necessitates requirements for frequent start and stop operation	18 22
		No need of clarifier and sludge return pump Better removal efficiency than ASP	Needs higher pressure drop	30
6.	MSBR (Membrane sequencing batch reactor)	Separates suspended solids better than SBR Almost complete hydrocarbon removal	Membrane fouling Increase in the size of sludge particle Enhances apparent viscosity	22
7.	MBR	–	Membrane fouling	22

performed by Liu *et al.*¹⁵ considering soluble COD (sCOD) as performance parameter.

In addition, Nakhla *et al.*⁹ presented Monod's model, Haldane's inhibition model, modified Monod's model, zero order and first order model while treating pet food wastewater. Initial substrate concentration (as sCOD) has been considered as an input parameter for model development. Zinatizadeh *et al.*¹⁴ used simplified Monod's model in order to explain kinetics of the treatment of palm oil mill effluent in upflow anaerobic sludge fixed film bioreactor (UASFF) considering COD as performance parameter. Apart from that, two models viz. back propagation neural network (BPNN) and linear regression techniques were successfully applied for treatment of heavy oil production wastewater in UASB reactor considering oil and COD as performance parameter¹⁵. Meanwhile, Santo *et al.*⁶ focused on improvement of the removal efficiency of a lab scale activated sludge reactor for biodegradable parameters including TOC, COD and TSS.

Apart from that, Mehryar *et al.*¹⁷ considered statistical model, mathematical as well as Artificial Neural Network

(ANN) model highlighting kinetic parameters, cumulative biomethane production (BMP) and cumulative biogas production (BGP) from oil refinery wastewater in anaerobic co-digestion treatment with COD as performance parameter. Brief overview of these models with performance parameters relevant to mathematical modeling and the model outcomes are presented in Table 3.

Utility of mathematical model for the removal of oil and grease

Till date a very few mathematical models have been developed for biological reactor treating oil and grease bearing wastewater. In most of the previous studies, kinetic models were mainly used to describe the biomass growth and substrate removal kinetics. These models generally vary from simple 0th order, 1st order or 2nd order kinetic models as per Monod's/Haldane's approach. However, application of these kinetic models is mostly system specific. Therefore, it becomes difficult to apply these kinetic models under varying environmental conditions. Hence, it is prerequisite to establish a correlation between all fundamental reactions oc-

Table 3. Brief overview on various mathematical models for removal of oil and grease

Types of model and process	Wastewater type and performance parameters	Important findings	Advantages	Limitations	Ref.
Kinetic model for substrate utilization and methane production suggested by Bathakur <i>et al.</i> ³² (Anaerobic baffled reactor)	Palm oil mill wastewater and total COD	Value of refractory coefficient (R) = 0.664 Kinetic parameter $A = 0.011$ Half saturation constant $K_S = 11.56$ g/L $\mu_m = 2.42$ /day	Good matching between model result and experimental data for COD as substrate. Model is suitable for complex substrate	Performance is expressed in terms of COD removal, not in terms of removal of oil and grease Same kinetic parameters are used for different recirculation ratios	10
Kinetic model for substrate utilization and methane production using Monod kinetic equation (Steady state MABR)	Palm oil mill wastewater and total COD	At influent COD (S_0) of 16 g/L, bio-kinetic parameters are $K_S = 0.313$ g COD/L, Kinetic parameter (A) = 0.329 and $\mu_m = 0.304$ /day	Experimental data were well represented by model equation	Performance is expressed in terms of COD removal, not in terms of removal of oil and grease Model gives good result only in specific situation	5
Hydrolysis model related to lipase catalyzed hydrolysis	Palm oil (Tributylin)	Enzyme activity was 15.7% more at interface than bulk	Rate of hydrolysis could be predicted in a batch reactor to determine optimal condition	Model is limited to low concentration of enzyme	12
Monod model of zero and first order in batch activated sludge process	Pet food processing wastewater and soluble COD	Value of $k = 0.168$ mg COD/mg VSS day	Kinetic data were fitted better in zero order kinetic model	The study mainly focused on removal of sCOD rather than oil and grease	13
Monod's kinetic model, Haldane's inhibition model and modified Monod's model as well as zero and first order model in activated sludge process	Pet food wastewater and soluble COD	Kinetic constants for Haldane's model were: $K_S = 17833$ – 23477 mg/L, $k = 1.28$ – 5.35 g COD/g VSS.d, $K_I = 48168$ mg/L and $Y = 0.13$ – 0.41 mg VSS/mg COD Kinetic constants for modified Monod's model were $K_S = 5580$ – 5600 mg COD/L, $k = 1$ – 1.3 g COD/g VSS.d, $K_H = 0.21$ – 0.66 d ⁻¹ and $Y = 0.08$ – 0.85 mg VSS/mg COD	Biomass and substrate data fitted better in Haldane's model than Monod's model for DAF pre-treated wastewater Kinetic data of raw wastewater fitted better in modified hydrolysis Monod's model	Slow biodegradation rate for DAF pretreated wastewater During simulation of substrate and biomass concentration, average percentage error (APE) was 12–154% for Monod model, 3–33% for Haldane model APE of zero order and first order substrate coefficient range 28–221%	9
Simplified Monod's model in UASFF under steady state	Palm oil mill wastewater and COD	The value of K linearly varies with VSS content at various influent COD At influent COD (S_0) of 34.75 g/L, bio-kinetic parameters are $K_S = 0.982$ g COD/L, $A = 0.738$ and $\mu_m = 0.207$ /day	Palm oil mill wastewater treatment using UASFF reactor can be fitted to kinetic equation	Reactor performance was not expressed in terms of oil and grease No information is available on process design	14

Table-3 (contd.)

Two models of back propagation neural network (BPNN) and linear regression technique in UASB reactor	Heavy oil production wastewater and COD as well as oil and grease	Average error in removal of oil and COD were 0.84% and -0.65% respectively	BPNN model is feasible for the prediction of pollutant removal by UASB The models properly fit the data and simulate oil and COD removal	Model is very complex and cumbersome	15
Mechanistic model (pseudo-first order kinetic model and activated sludge reactor model)	Petroleum refinery wastewater and soluble COD	4% and 2% higher removal of TOC and COD respectively in case of recirculation of biomass Value of rate constant (k) as 0.059 and 0.055 mg^{-1} VSSL/day for without and with sludge recycle respectively	Describes reactor performance as well as kinetic model	Removal efficiency in terms of COD, not in terms of oil and grease	6
Statistical model, mathematical model and ANN algorithm	Oil refinery wastewater and soluble COD	Gompertz mathematical model satisfies experimental BMP and BPNN algorithm satisfies BGP data	Found to be very useful for optimization purpose	Removal efficiency is in terms of COD Model is very complex and cumbersome No information on process design	17

curing in bioreactors towards development of a simplified model for rational and accurate prediction of oil and grease concentration.

The utility of mathematical model treating oil and grease containing wastewater lies with optimization and improvement of the overall performance. It has been observed that the models used for treating oil and greases containing wastewater in ASP, SBR, USAB etc. mainly concentrated on determination the substrate removal rate under suspended growth condition. However, there are some mathematical models in case of biofilm reactors also. But application of these biofilm models in oil and grease bearing wastewater is very rare.

Moreover, no standard engineering guideline is available so far on the development of process design for treatment of oil and grease containing wastewater in biological system. The use of process design of a system is required to compute its volume, physical dimension and other operational requirements for any targeted effluent substrate concentration. There are various physical or statistical models developed so far on oil and grease containing wastewater. Therefore it is very important to develop a comprehensive mathematical model for oil and grease containing wastewater.

Drawbacks of available models

Accurate prediction of substrate removal and biomass growth are two critical issues for successful mathematical modeling in case of oily wastewater treatment. In most cases, biodegradation rate of a single component present in oily wastewater can be explained by the traditional kinetic expression. However, development of mathematical model for biodegradation of mixed substrates is quite limited when compared to biodegradation of single substrate. Setiadi *et al.*¹⁰ studied the performance of anaerobic baffled reactor with recycle, treating palm oil mill wastewater. Monod kinetic model with an additional refractory coefficient for degradation of mixed substrate present in palm oil mill wastewater was used. Kinetic coefficients of developed model equations were determined by the method of least square. The experimental data fitted with model equation shows good precision.

It has been reported in literatures that the conversion of complex substrate into simple compounds takes place by means of hydrolysis leading to cell growth and formation of end product. A study was conducted by Zuhair *et al.*¹² regarding treatment of palm oil mill wastewater using lipase enzyme to determine the hydrolysis rate constant. A mechanistic model was also developed based on Monod kinetic

model for calculating the specific hydrolysis rate for various substrates present in oil mill effluent.

Treatability study and development of kinetic model were carried out by Liu *et al.*¹³ in a batch reactor fed with pre-treated and raw oil and grease bearing wastewater. Oil and grease biodegradation was explained by the first order and 0th order reaction. It was found from the experimental study that the 0th order rate constant was more significant compared to 1st order rate constant. These results indicate that rate of degradation of oil and grease depends on active biomass present in a system. At higher substrate concentration (as oil and grease), the rate of biodegradation is more accurately measured by the Haldane inhibition model⁹. However, Zahed *et al.*³¹ reported that the first order kinetic model could be explained more accurately for mixed microbial population treating the raw oil in presence of dispersant. It has also found that presence of dispersant improved the rate of biodegradation significantly.

Santo *et al.*⁶ proposed a mass balance equation based on CSTR model with biomass recirculation to calculate the stoichiometry coefficients, oxygen uptake rate and other kinetic rates for petroleum refinery wastewater in bench scale ASP reactor. Pseudo-first order kinetic expression was assumed for describing biodegradation kinetics of petroleum hydrocarbons present in the effluent. The model showed good agreement with respect to experimental observations. Yamaguchi *et al.*⁸ proposed a mechanistic model for Algal-RBC system fed with petroleum wastewater to examine its performance efficiency. However, that biofilm model was found difficult and cumbersome towards application. No process design approach has been developed so far in this regard.

Future prospect of mathematical modeling of oily wastewater

Out of very few literatures available for the modeling of biological treatment of wastewater containing oil and grease, major ones have considered suspended growth reactor such as ASP, ABR, MABR, SBR, MBER, UASB etc. as mentioned in Table 2. As very few studies are available on mathematical modeling of fixed film bioreactor for removal of oily substance from wastewater, experimental studies with these reactors are extremely essential. Most of the literatures focused on kinetic model especially Monod's model^{9-11,13}, a few on mechanistic model⁶ and statistical model^{15,17}.

In a simplified mathematical model Zinatizadeh *et al.*¹⁴ incorporated three fundamental steps of hydrolysis followed by substrate diffusion or transportation, which ended up with utilization. This is because, a major fraction of total COD present in oily wastewater include the dissolved and suspended hydrolysable substrate. Usually hydrolysis is a first order model for substrate as hydrolytic enzymes may not proportional to active biomass and linear nature of \log_e (particulate COD) with respect to time. In this process, complex substrates are converted to simplified hydrolyzed products which are used for cell growth¹⁰. Again, Faisal and Unno⁵ also mentioned that consideration of hydrolysis process is very much essential in developing a model in order to digest complex organics into soluble products which can be utilized for biomass growth. The model should be developed on the basis of characteristics of oily substances present in wastewater. Moreover, the model should consider hydrolytic degradation, biological uptake of substrate as well as microbial growth, incorporating their relevant kinetics in a systematic and rational way.

Concluding remarks

Discharge of wastewater containing oil and grease cause environmental hazard on account of slowly biodegradable COD, TDS, toxic/inhibitory substances, heavy metals and colour. Physicochemical methods are found to be inappropriate and also not techno-economically viable for this kind of waste streams. Pre-treatment of oil and grease bearing wastewater makes it fully compatible for biological treatment. However, biological treatment using specially acclimated microorganism can be practiced without any pre-treatment step in case of low concentration. In order to get satisfactory result, selection of suitable biological method and appropriate mathematical modeling are very essential. Various mathematical models have already been implemented for the prediction of effluent quality as well as for validation in aerobic and anaerobic processes for oil and grease containing wastewater. Although, numerous mathematical models are available for describing treatment of oily wastewater, very few of them are based on both kinetics and reactor mass balance. Hence, a simplified and comprehensive mathematical model needs to be developed incorporating all the relevant kinetic and process parameters. In that case, process design of the biological reactor for the treatment of oil containing wastewater would be more accurate.

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