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Removal of nitrogen and phosphorus from municipal wastewater using intermittent cycle moving bed biofilm reactor (ICMBBR)

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ABSTRACT

In present study nitrogen and phosphorus removal from municipal wastewater using ICMBBR reactor, was investigated. ICMBBR reactor is an integration of intermittent cycle extended aeration system (ICEAS) and moving bed biofilm reactor (MBBR) which comprise benefits of both systems in term of their efficiency in nitrogen and phosphorus removal. A laboratory –scale Plexiglas reactor with volume of 4 Lit which was filled with kaldnes (a plastic media) as biofilm carrier with specific surface area of 400m^3/m^2 and 20-50% occupation of reactor volume was used in this study. The reactor was operated in an aerobic and anaerobic continues condition. Efficiency assessment of the reactor was obtained at HRT of 3-6 hr. The average removal efficiency of 68.1±0.12, 67.7±0.09, 67.5±0.12, 75.2±0.1, and 53.6±0.17 were achieved re spectively for TN, TP, TKN, NH_4-N, and N-org within 6 month experiment. the maximum removal efficiency was observed at aeration time of 4h, mixing time of 90min and kaldnes filling ratio of 50%. In such condition, removal efficiency of 91.8%, 88.95%, 92.8%, 93.3%, and 92% were achieved for TN, TP, TKN, NH_4-N, and N-org respectively. Based on the results, this study confirms that ICMBBR can be considered as an efficient method for removal of nitrogen and phosphorus from municipal wastewater.

Key words: ICMBBR, wastewater treatment, nutrient removal, kaldnes

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INTRODUCTION

Discharge of untreated (raw) wastewater which contains nutrients (nitrogen and phosphorus) can be hazardous for environment, due to its potential to cause autrophication, oxygen reduction, and also toxicity in water bodies. Thus it is needed to treat polluted water in term of nutrient removal before discharging in environment (1). Generally, chemical and biological techniques are applied for nutrient removal from wastewater, but chemical methods owing to being costly, producing high sludge content, changing pH, and reducing settling ability are not very popular (2,3). Biological systems because of possessing diverse benefits including, high efficiency, simplicity of application, cost effectiveness (economical), and being environmentally compatible and safe, have become more popular than other methods (4). Recently almost all of the wastewater treatment plants have been equipped with nitrogen and phosphorus removal systems. In such systems, biological processes which are consisted of sequential anaerobic, anoxic, and aerobic zones (for releasing and reabsorbing phosphorus) are used (5). Nitrification and denitrification are among the most accepted biological nutrient removal methods. In this method, biological oxidation accomplishes via ammoniac oxidative autotrophic bacteria and as a final point ammonia nitrogen is converted to nitrate. In the next stage, heterotrophic bacteria develop nitrification process in anoxic condition and alter nitrate to nitrogen gas which is released out of system (6). Activated sludge process is the most accepted aerobic method for biological wastewater treatment (7). Moving bed biofilm reactor (MBBR) and intermittent cycle extended aeration system (ICEAS) are among new techniques for biological wastewater treatment. The ICEAS is a form of activated sludge reactor with continues flow which consists of diverse phases, including mixing, aeration, settling, and discharge phase (8). The MBBR reactor is an advanced technique for wastewater treatment. The crucial point in this process is biofilm carrier which provides a suitable site for microorganism growth, due to its high specific surface (9). The ICMBBR reactor possess many advantages including no need to primary and secondary settling tank (land saving), reducing operation cost, high efficiency, operation simplicity, nitrogen and phosphorus high removal efficiency, and sludge bulking reduction. The advantages of MBBR reactor was grow of biomass (microorganism) on biofilm, no clogging and no need to return the sludge, in comparison to conventional activated sludge system (10-13). In this work to reach the desired characteristic in treated wastewater, a combination of ACEAS and AMBBR reactor was used which is called intermittent cycle moving bed biofilm reactor (ICMBBR) that acquires advantages of both systems. The MBBR reactor has been effectively applied for municipal and industrial wastewater such as paper mill, wood mill, dairy, refinery and slaughter industries (14). The SBR reactor with continuous flow have been approved of possessing high removal efficiency of BOD, COD, TKN, TSS, and mean removal efficiency for TN and TP (15). The MBBR reactor is proficient of high TN and TP removal (16). Up to our knowledge, nutrient removal via continues flow ICMBBR reactor is a new-fangled technology that have not been used for municipal wastewater up to now. This research aims at evaluating the ICMBBR reactor efficiency in nutrient removal from municipal wastewater and spreading the method in Iran. Therefore the authors came to a decision to present optimum model to appraise (survey) the process and viable factors affecting this process in order to achieve a high nutrient removal rate.

EXPERIMENTAL SECTION

A laboratory-scale Plexiglas reactor with effective volume of 4 lit, with an internal diameter of 10 mm, wall thickness of 4mm, and height of 11mm was used in this experiment. The reactor was filled with kaldnes with specific biofilm surface area of $400m^3$/m2, (the kaldnes characteristic presented in table2) and added (occupied-filled) at different filling ratio (%20, %35, and % 50) of reactor volume. Organic and hydraulic loading has been done for every stage during the reactor operation. After the hydraulic test of reactor, the system was started up with the sludge seed obtained from (the activated sludge of Doroodfaraman town wastewater treatment plant) and was fed with municipal wastewater. After microbial acclimation, when the effluent COD concentration and turbidity controlled (maintained) below, 100m/l and 5 NTU respectively the reactor was operated with hydraulic retention time (HRT) of 3-6 hr. In this experiment the MLSS concentration was kept 4000 mg/l and Total suspended loading has been controlled (adjusted) according to kaldnes filling ratio in the reactor volume. The temperature was kept at value of 24 to 28C, and pH arranged between 6.8 to7.2, and DO concentration was always controlled between 3-5 mg/l during the experiment. The reactor equipped with peristaltic pump that provide continuous influent during the operation. Furthermore a circulator pump was used, supplying sufficient influent mixing and freely moving of carriers in the reactor. The blowers system, possessing 4 nozzles was placed in the reactor to provide the sufficient air for the system. All experiments have been done in Kermanshah University pilot laboratory followed by standard method book and each test 3 times has been repeated. The schematic diagram of ICMBBR was presented in fig1.

Table1.technical data of, air and circulator blower used in this study

	Circulator blower		**Air blower**
		model	ACO-5505
Power(w)	12	Power (watt)	6.5
Max head(cm)	80	Pressure(M pa)	0.25
Max Q(L/h)	600	Inlet flow (L/min)	5.5

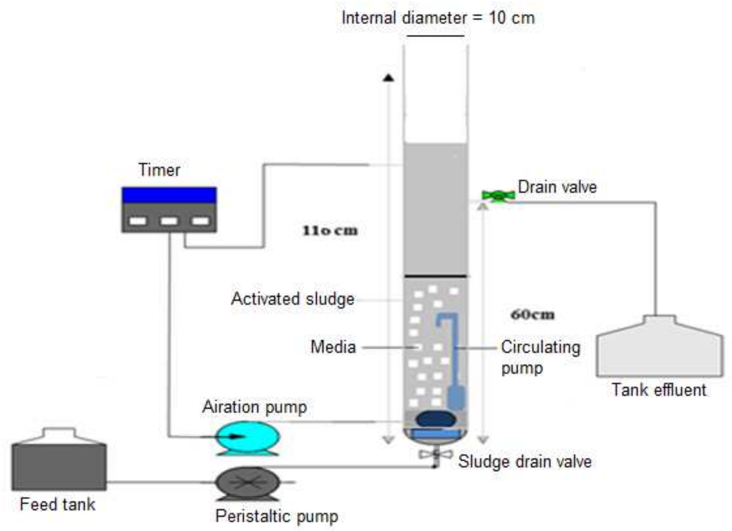

Fig1. The schematic diagram of ICMBBR (intermittent cycle moving bed biofilm reactor)

Table2.characteristic of kaldnes (the plastic media)

Value	Characteristic	Value	Characteristic
274	The carrier number within %20 reactor volume occupied	600	Total specific surface (m^2/m^3)
422	The carrier number within %20 reactor volume occupied	20,35,50	Filling ratio
		10	(mm)height
648	The carrier number within %20 reactor volume occupied	10	(mm)diameter
		0.9	(g/cm)density

RESULTS AND DISCUSSION

In biological nutrient removal techniques, precise pH monitoring (control) must be met. In the ICMBBR reactor that was used, some processes including nitrification, denitrification, phosphorus releasing, and reabsorbing occurred in which for any of them a specific pH range was needed. Based on researches pH is a key factor, especially in anaerobic zone (1). The influent pH was about 7.6 that due to high alkalinity, there is no need to add extra alkalinity to set pH. During the stable operation phase of the ICMBBR reactor, the mean ph value was reported 7.4 (at the beginning of mixing period in anaerobic condition) in consequence of denitrification and alkalinity production. But at the end of mixing period without aeration (in anaerobic condition) the ph value descends to 6.67 as a result of volatile fatty acid (VFA) creation, like acetate that are absorbed via polyphosphate collector microorganism and are stored as PHB. In anaerobic condition, pH reduction is an indicator for denitrification speed lessening. But this reduction can be modified through wastewater aeration and CO_2 removal. That is why pH has arrived at 7.28 in aerobic condition. The reactor effluent pH has fallen slightly (pH7.06) in comparison to aerobic condition. In ICMBBR reactor, in settling phase (30 min) after aeration, pH will drop down due to relative anaerobic phase developing which this reduction will be improved through high pH of continues influent. Generally author can claim that, through the reaction pH had been in a suitable range for nitrification, denitrification, phosphorus releasing and reabsorbing processes(Fig.2). In this system, during nitrification process (in aerobic condition) ammonia converts to nitrate and then nitrite, but through denitrification course (in anaerobic condition) at first, nitrate changes to nitrite, then to nitric oxide, nitrous oxide, and finally to nitrogen gas. In nitrification process, carbon source is CO_2 (inorganic carbon), hence by influent alkalinity consumption, pH drop a little down in comparison to influent. Ammonium and nitrite ions are as electron receptor, and nitrate and nitrite ions as final products. Results, declare that raw (untreated) wastewater, contain a low concentration of nitrite and nitrate while in effluent, nitrite concentration has increased and nitrate concentration has fallen down (Fig.3,4). Generally, a stable nitrate effluent has been observed.

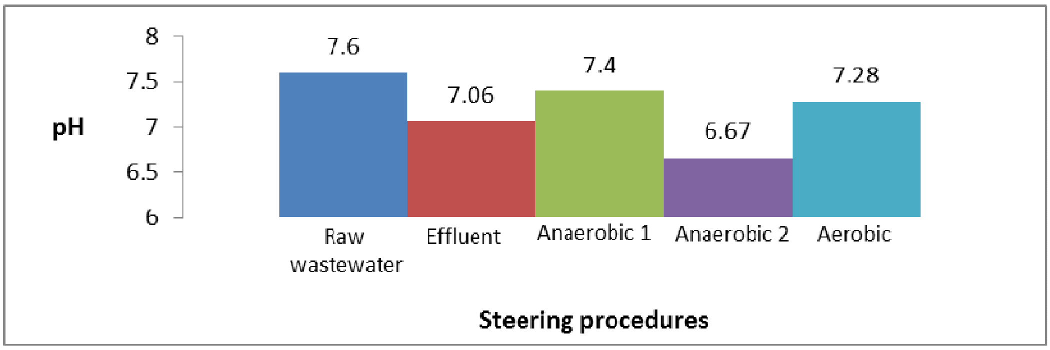

Fig. 2. Mean values of pH in the reactor during the reaction ICMBBR

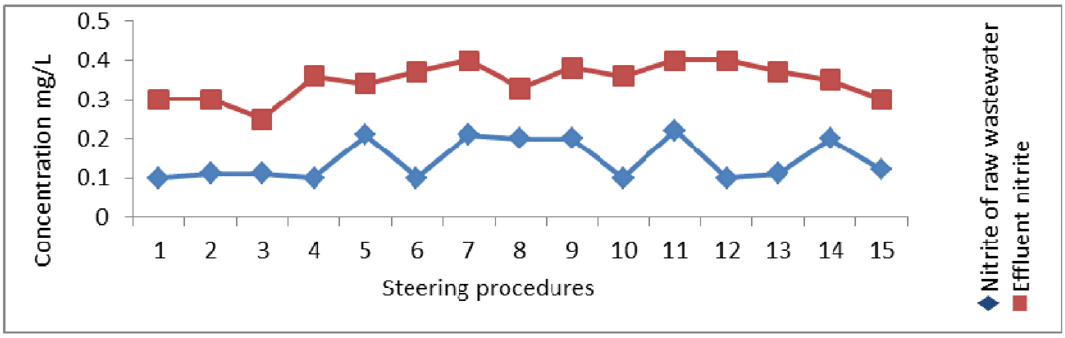

Fig.3. Nitrite Concentration in raw wastewater and effluent at various stages of Steering System ICMBBR

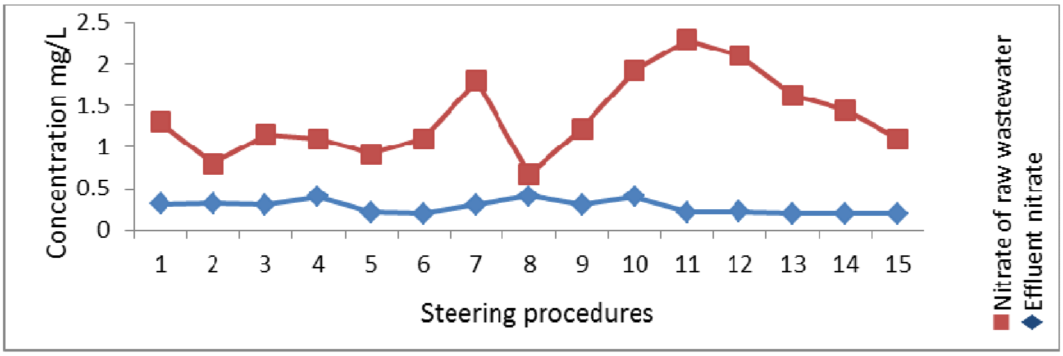

Fig.4. Nitrate concentrations in Raw wastewater and effluent at various stages of Steering System ICMBBR

Accepted theory toward enhanced biological phosphorus removal is that, sequential anaerobic-aerobic periods pose to competitive capability in substrate consumption and also microbial growth. In biological phosphorus removal, influent phosphorus is absorbed via cellular biomass and subsequently is removed from system as excessive sludge (17). In this work, biological phosphorus and nitrogen removal were integrated via incorporating anaerobic and aerobic zones. In anaerobic zone, orthophosphate concentration in mixed liquor has increased. High orthophosphate concentrate, imply phosphorus desorption via bacteria. Under anaerobic condition, intracellular orthophosphate concentration increases through stored material (substance) oxidation and subsequently energy generation. In aerobic phase, phosphorus absorbent organisms (PAO_s) metabolize intracellular poly hydroxyl alkaloids (PHA_s) and use produced energy for assimilation and also glycogen synthesize. A part of energy obtained from PHA_s oxidation is used for polyphosphate bonds formation and intracellular storage. Therefore, in this way most of orthophosphate can be removed from wastewater. Anoxic condition is the same as aerobic condition and orthophosphate is removed from wastewater. In anoxic environments, bacteria use nitrate as electron receptor instead of oxygen. These kinds of organisms are known as denitrifire phosphorus absorbent organisms ($DNPAO_s$) which can absorb orthophosphate by consuming nitrate under anoxic condition. Efficiency orthophosphate absorption in anoxic condition is inferior in comparison with aerobic environments. This result is in agreement with other studies (Fig.5) (18,19). In spite of phosphorus variation in raw wastewater and also anaerobic phase for diverse runs, there is a steady phosphorus effluent in all runs which demonstrate stable phosphorus removal efficiency.

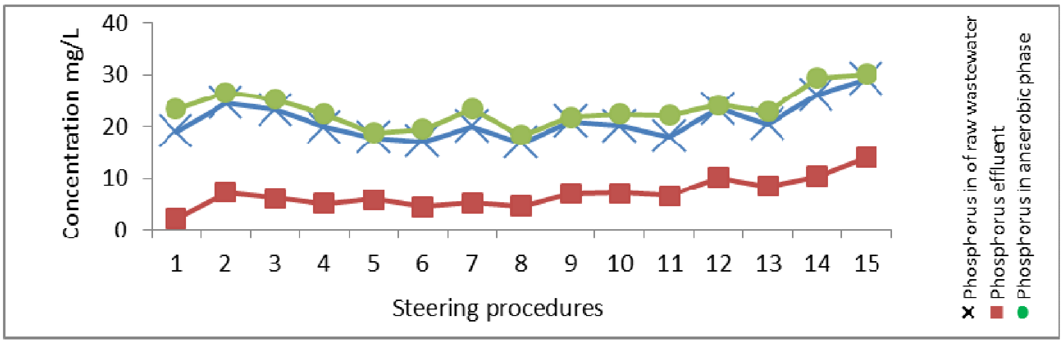

Fig. 5. changes total phosphorus inputs, outputs and phase anaerobic system Steering at various stages of ICMBBR

Findings imply that, nitrogen compounds removal efficiency increases through kaldnes filling ratio (MLSS improvement) and aeration time enhancement. In this study the mixing period without aeration (30-90min), didn't show any significant effect on results and just a slight difference was observed. So improving the mixing time without aeration from 30min to 90min, didn't affect nitrogen compounds removal efficiency (Fig.5). Nevertheless, by enhancing kaldnes filling ratio and mixing time (without aeration), phosphorus removal efficiency increases (Fig.6).

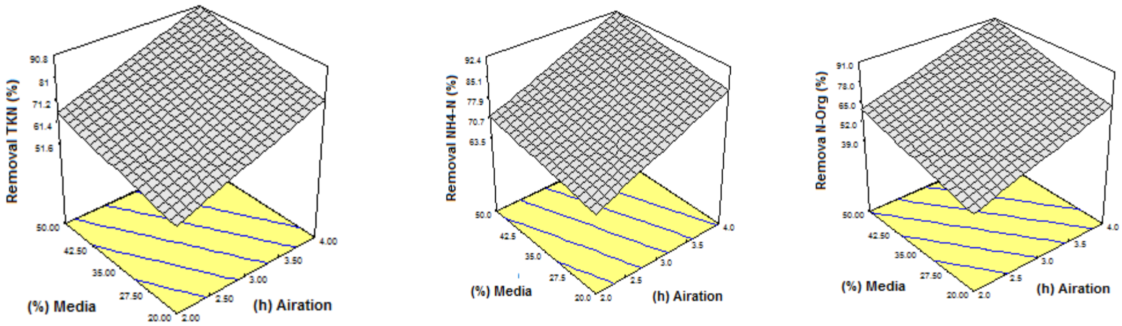

Fig.6. Three dimensional graph of TN and TP removal within mixing of 90 min

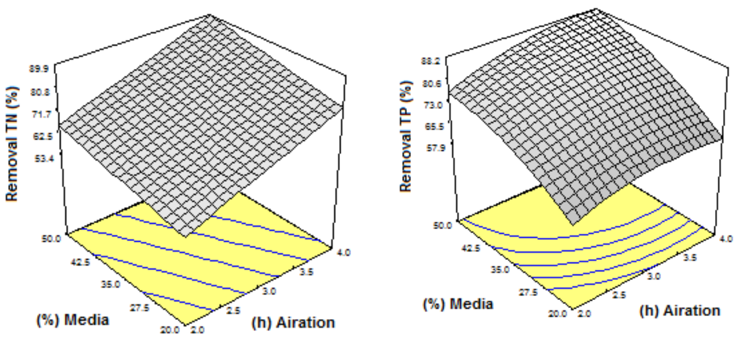

Fig.6. Three dimensional graph of TN and TP removal within mixing of 90 min

The maximum removal efficiency for TN, TP, TKN, NH_4-N, and N-Org were achieved 91.8, 88.95, 92.8, 93.3, and 92 whereas least values were 45.23, 52.2, 58.2, 45.2, and 26 respectively.

TN removal:
The results report a maximum influent TN concentration of 81.42mg/l, against effluent TN value of 44.59mg/l, indicating 45.23% removal efficiency. This removal occurred within aeration time of 2h, mixing period of 30min, and HRT of 3h. The lowest value for TN in influent and effluent were 49.26mg/l and 12.84mg/l respectively, demonstrating 73.93% removal efficiency. This removal occurred within aeration time of 3h, mixing period of 60min, and HRT of 4.5h (table 3).

TKN removal:
The highest concentrations of TKN in the ICMBBR reactor influent and effluent were 80.2mg/l and 44.09mg/l respectively which represent 45.1% removal efficiency (within aeration time of 2h, mixing period of 30min, and HRT of 3h). Based on findings, the lowest TKN value imported to the system was 48.4mg/l, against the effluent TKN value of 12.1, reflecting 75% removal efficiency within aeration time of 3h, mixing period of 60min, and HRT of 4.5h.

NH_4-N removal:
The results state a maximum influent NH_4-N concentration of 51.47mg/l, against effluent NH_4-N value of 18.02mg/l, indicating 65% removal efficiency. This removal occurred within aeration time of 2h, mixing period of 90min, and HRT of 4h. The lowest value for NH_4-N in influent and effluent were 31.78mg/l and 8.39mg/l respectively, demonstrating 73.6% removal efficiency. This removal occurred within aeration time of 3h, mixing period of 60min, and HRT of 4.5h (table 3).

N-Org removal:
The highest concentrations of N-Org in the ICMBBR reactor influent and effluent were 29.31mg/l and 21.69mg/l respectively which represent 26% removal efficiency (within aeration time of 2h, mixing period of 90min, and HRT of 4h). Based on findings, the lowest N-Org value imported to the system was 15.39mg/l, against the effluent N-Org value of 6.05mg/l, reflecting 60.7% removal efficiency within aeration time of 4h, mixing period of 60min, and HRT of 5.5h (table 3).

TP removal:
The results state a maximum influent TP concentration of 29.1mg/l, against effluent TP value of 13.92mg/l, indicating 52.2% removal efficiency. This removal occurred within aeration time of 2h, mixing period of 30min, and HRT of 3h. The lowest value for TP in influent and effluent were 16.85mg/l and 4.52mg/l respectively, demonstrating 73.2% removal efficiency. This removal occurred within aeration time of 3h, mixing period of 60min, and HRT of 4.5h (table 3).

Table3. Overview of TN, TP, TKN, NH_4-N, and N-Org removal rate presented within (base on –corresponding on) influential experiment variable

	A. mixing	B. Aeration	C. kaldnes Filling ratio	TKN removal rate percent	NH_4-N removal rate percent	N-Org Removal rate percent	TN Removal rate percent	TP removal rate percent
1	90	4	50	92.8	93.3	92	91.8	88.95
2	30	4	50	82.7	86.7	75	82.08	70.6
3	60	3	50	76.1	80.5	66.9	75.68	73.5
4	90	2	50	63	67.7	53.7	62.25	74.7
5	30	2	50	59.4	65.5	47.7	59.15	67
6	60	4	35	78.2	87.5	60.7	77.13	73.8
7	90	3	35	74.6	80.4	63.7	73.9	74.5
8	60	3	35	64.3	73.6	50.84	73.93	73.2
9	30	3	35	60.7	71.3	43.1	60.24	67
10	60	2	35	56.2	67.6	39	56.05	64.5
11	90	4	20	75.1	81.9	52.7	73.1	63.65
12	30	4	20	72.6	79.2	59.2	71.93	56.9
13	60	3	20	57.4	69.8	37.7	66.23	59.3
14	90	2	20	52.9	65	35.5	53	60.4
15	30	2	20	45.2	58.2	26	45.23	52.2

The result obtained removal efficiency based on design expert software:

In This study to analyze findings, Design Expert software has been (was) used. Based on this model, it was reported that, the aeration, high total solids loading (kaldnes filling ratio), and mixing can be consider as the most significant factors affecting nitrogen removal respectively. Whilst the total solids loading (kaldnes filling ratio), mixing, and aeration, respectively affected phosphorus removal. Findings demonstrate that according to accounts, nitrogen compounds removal was in agreement with Linear equation. Total phosphorus removal was followed by Quadratic second degree equation. It should be mentioned that resulted equations (presented in table 4) were identified based on subsequently omission of meaningless variable and finding the mutual coherence between them. Test analysis was accomplished using ANOVA test and SPSS statistic software (11) version. Statistical analysis results for nitrogen compounds and total phosphorus removal imply a meaningful correlation ($P<0.05$). Linear regression coefficient for TKN, NH4-N, N-Org, TN, and TP were reported 0.967, 0.964, 0.914, 0.967, and 0.967 respectively. Furthermore, the validity and reliability for all analysis (15.87-34.8), was remarkably greater than 4, this meant that an appropriate rate for test validated (validity assessment). Additionally, the standard deviations ranged from 2.14 to 3.99. Generally, the result obtained SPSS software and ANOVA statistic test, implies a significant (positive) correlation between nitrogen compounds average removal efficiency, aeration time, and kaldnes filling ratio ($p<0.05$), while no significant correlation with mixing period without aeration was observed ($P>0.05$). based on statistically analysis, variation of total phosphorus removal with mixing time without aeration, kaldnes filling ratio, and aeration time show a positive correlation ($p<0.05$). According to linear regression and standard deviation resulted from applied model (used in this study), aeration time factor has shown more significant effect than media filling ratio (total solids loading) and mixing time in nitrogen compound removal. It is evident the kaldnes filling ratio has depicted noticeable effect for removal rate in comparison to mixing, and aeration time (table5).

Table.4. The results of analysis of variance (ANOVA) for the response of the study program Design Expert (A: Mixer, B: Airation, C: Media)

Removal (%)	Significant equations for the desired response	Model	R^2	Adeq Precision	Std. Dev	P-value
TKN	+67.45 + 3.78 A + 12.47 B + 7.08 C	Linear	0.967	34.8	2.6	0.0001>
NH4	+75.19 + 2.74 A + 10.46 B + 3.96 C	Linear	0.964	31.08	2.14	0.0001>
N-Org	+53.58 + 4.66 A + 13.77 B + 12.42 C	Linear	0.914	21.3	5.61	0.0001>
TN	+64.48 + 3.33 A + 11.81 B + 7.94 C	Linear	0.967	34.57	2.59	0.0001>
TP	$+71.28 + 6.38 A + 3.04 B + 6.84 C – 5.38C^2 + 3.78 AC$	Quadratic	0.976	15.78	3.24	0.0318>

Table.5. Determine the significant variables in the regression model and determine the removal efficiency of nitrogen and phosphorus compounds

	Regression model	Standardized coefficients			P-value - ANOVA		
		A: Mixer	B: Airation	C: Media	A: Mixer	B: Airation	C: Media
TKN	TKN = 0.052 + 0.001 A + 0.125 B + 0.005 C	0.248	0.818	0.464	0.111	0.000	0.001
NH4	NH4 = 0.288 + 0.001 A + 0.105 B + 0.003 C	0.233	0.888	0.363	0.202	0.000	0.033
N-Org	N-Org = -26.47 + 0.156 A + 13.77 B + 0.828 C	0.233	0.688	0.62	0.236	0.000	0.000
TN	TN = 3.84 + 0.111 A + 11.838 B + 0.529 C	0.233	0.764	0.532	0.325	0.000	0.001
TP	TP = 0.312 + 0.002 A + 0.03 B + 0.005 C	0.544	0.259	0.582	0.000	0.002	0.000

Graphic optimization (Multi-layer map)

A multi-layer map, optimized graphically was used (employ) in this experiment; therefore focus on the optimum zone for organic matter and turbidity removal rate that were defined criteria(table6). Aeration and mixing time and when 50% of bioreactor was occupied by media kaldnes was shown by graphical optimization.(table6).the crucial experimental variables influence on the removal efficiency percent(the dark zone) and the removal rate when kaldnes filling ratio was 50%(yellow zone) was demonstrated (table6). In optimum zone a point (punctuation) was chosen for assessing the validity and reliability of the model was presented in. (table.7). Accordingly, the bioreactor was operated for compare the real (calculated) and predicted value of removal rate. Consequently, DES software through (standard derivation) was applied to optimum zone for each removal rate, finally the current model value of actually calculated to predicted value.

Table.6. Optimization for criterion studied Response

Response	Value	Unit
Removal TKN	80<	**%**
Removal NH_4	80<	**%**
Removal N-Org	80<	**%**
Removal TN	80<	**%**
Removal TP	80<	**%**

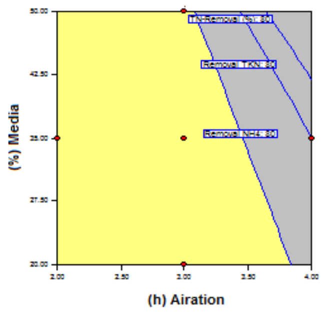

Fig.7. Overlaps Platt for optimal region

Table.7. The validation experiments for the optimal

Laboratory conditions	**Period experiments**	**Response**				
		Removal TP(%)	**Removal TN** (%)	**Removal N-Org**(%)	**Removal NH_4**(%)	**Removal TKN**(%)
Aeration = 3h Mixer = 60 min Media = 35%	Laboratories amounts	74	63.2	50.84	73	64.8
	Reply Model CI= %95	71.3	64.5	53.6	75	67.4
	Std. Dev	1.45	0.67	1.45	0.55	0.67

CONCLUSION

Findings imply a remarkable performance for ICMBBR reactor in municipal wastewater pollutants removal. The most import factors in nitrogen compound removal, using current system, were aeration time, total solids high loading (media filling ratio), and mixing time without aeration respectively. It was reported that the total solids loading, mixing time without aeration, and aeration time were the most significant factors in total phosphorus removal. The maximum nutrient removal efficiency was achieved within aeration time of 4h, mixing time without aeration of 90min, HRT of 6h, and media filling ratio of 50%. While The minimum nutrient removal efficiency was attained within aeration time of 2h, mixing time without aeration of 30min, HRT of 3h, and media filling ratio of 20%. Therefore HRT enhancements can result in removal high efficiency. The maximum removal efficiency for TKN, NH_4-N, N-Org, TN, and TP was reported 92.8, 93.3, 92, 91.8, and 88.95 whereas the least removal efficiency was achieved 45.5, 58.2, 26, 45.23, and 52.2 respectively. Finally it can be concluded that under suitable design and operation, the ICMBBR reactor can be consider as a promising technology for municipal wastewater treatment with respect to nutrient removal.

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