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Study of Nitrification and Denitrification in the High Ammonia and COD Load of Industrial Wastewater using an Ultracompacted Biofilm Reactor-Moving Bed System

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Abstract
Removing nitrogen, one of the most common and abundant pollutants of surface and ground water, is very important. For this purpose, biological nitrification and denitrification as the most economical method should be considered. The feasibility of high load COD (Chemical Oxygen Demand) (800-2000 mg/lit) and NH_4 (250-1000 mg/lit) industrial wastewater treatment, at different Hydraulic Retention Times (HRTs), was studied in 9-lit anaerobic-aerobic systems in the post-denitrification mode. The Ultracompacted Biofilm Reactor (UCBR) is a new system, with all the advantages of activated sludge and fluidized fixed bed processes, without the disadvantages of each system, so that the biofilm production takes place on the packings, moving along the height of the reactor. From the experiments carried out using this system, it can be said that higher ammonia removals take place at higher ammonia and lower organic loads. Denitrification increases at higher nitrification rates because of the effect increasing of NO_3^- entering to anaerobic reactor. Despite the fact that nitrifying bacteria are more sensitive than COD and NO_3^- for removing bacteria, after toxic shock by phenol as an organic source, the nitrification rate increases and COD removal decreases according to the damaging effect of phenol on COD-removing bacteria. Total COD removal during the study varied between 70-98%, this value changing between 50-90% for ammonia and 55-90% for nitrate.

Keywords: Industrial wast water, Denitrification, Nitrification, Ultracompaccted Biofilm Reactor(UCBR).

مطالعات یک بیورآکتور با بستر متراکم به منظور حذف نیترات در پسابهای صنعتی

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چکیده
حذف نیتروژن به‌عنوان یکی از معمولی‌ترین آلاینده‌های آب‌های سطحی و زیر زمینی حائز اهمیت است بدین ترتیب نیتریفیکاسیون و دی نیتریفیکاسیون بیولوژیکی اقتصادی‌ترین روش برای حذف آنها می‌باشد. در این تحقیق سعی گردیده تا با ایجاد شرایط بهینه، درصد حذف نیتروژن آمونیاکی و COD در پساب‌های صنعتی حاوی نیتروژن آمونیاکی بالا(۲۵۰-۱۰۰۰) و COD (۸۰۰-۲۰۰۰) در دو راکتور جداگانه هوازی و بی‌هوازی بصورت ناپیوسته در ۹ لیتر پساب مصنوعی بروش پس دی‌نیتریفیکاسیون مورد بررسی قرار گیرد. استفاده از سیستم ناپیوسته نیز به‌دلیل بررسی نحوه حذف نیتروژن در آن، عدم نیاز به بازگشت لجن، کیفیت مناسب بیولوژیکی پساب خروجی و ... می‌باشد. راکتورهای به‌کاررفته شامل دو راکتور هوازی(UCBR) و بی‌هوازی(MBS) می‌باشد که در اولی عمل اکسیژن‌رسانی با تزریق هوا از کف انجام شده و پس از انجام فرایند نیتریفیکاسیون، نمونه پساب به‌منظور انجام فرایند دی نیتریفیکاسیون وارد راکتور دوم می‌شود. (UCBR) یک سیستم جدید است که مزایای زیادی نسبت به لجن فعال و فرایند بسترهای متحرک و ثابت بدون معایب هر کدام از اینها دارد که لایه بیوفیلم آن بر روی بسترهای متحرک تشکیل می‌شود. بررسی انجام فرایند نیتریفیکاسیون در زمان ماندهای متفاوت(۲-۲۴ ساعت در هر دو راکتور) بهترین راندمان حذف آمونیاک و COD در راکتور هوازی (UCBR) را در زمان ماند ۲۰ ساعت و بترتیب برابر ۹۶ درصد و ۷۵ درصد و بهترین راندمان حذف نیترات و COD در راکتور بی‌هوازی(MBS) را در زمان ماند ۱۴ ساعت و به‌ترتیب برابر ۸۳/۲۳ درصد و ۸۳ درصد نشان می‌دهد.

کلیدواژه‌ها: پساب صنعتی، دی نیتریفیکاسیون، نیتریفیکاسیون، بیورآکتور با بستر متراکم.

Introduction

Biological nitrification and denitrification processes are the most important wastewater treatment processes because of the abundance of nitrogen pollutant compounds in water and wastewater and also because of the growing trend in population and the increasing number of industrial plants and agricultural fields, especially in developing countries.

It is generally believed, on a relative basis, that ammonia and nitrite oxidation is carried out mainly by autotrophs of the types Nitrosomonase sp. And Nitrobacter sp.

A few features of the autotrophic Nitrifying bacteria, Nitrosomonase and Nitrobacter are summarized in Table 1. In denitrification, nitrite reduction to N2 is carried out by heterotrops of the Psudomonase.

One potential biofilm process, which may be compact, is the one based on submerged biological filters. There are many reports concerning the possibility of using biofilm processes for treating wastewater (MBBr, 2000; Carrera *et al.*, 2003; ong *et al.*, 2003; chen *et al.*, 1995; Halling- sarenson and Jorgensen, 1993) but the disadvantage of some biological filters is the possibility of clogging of the biofilm media (MBBr, 2000; carrera *et al.*, 2003, ong *et al.*, 2003; chen *et al.*, 1995; Rusten *et al.*, 1994; Rusten *et al.*, 1996).

The biofilm process in the Ultracompacted Biofilm Reactor (UCBR) has a high specific surface, but none of the clogging (Al- Ghusain, 1994). In this reactor, the biofilm grows on carriers circulating inside the tank. The carriers are shaped to maximize growth, by protecting the biofilm from abrasion (Van loosdrecht *et al.*, 1995; carrera *et al.*, 2003).

The First and best study on (UCBR) process was developed by Ong, Lee, Hu and Ng at the National University of Singapore on January 2003 (Al-Ghusain, 1994). The basic idea behind the (UCBR) in this research was to have a batch operating, with a non-cloggable biofilm reactor with no need for backwashing, low-loss and a high specific biofilm surface area(Al- Ghusain, 1994). This reactor is becoming increasingly popular and is now being used in many plants around the world for various treatment purposes (BOD/COD removal, nitrification and denitrification) in both municipal and industrial wastewater (yang and zhang, 1995).

This paper examines the results obtained from two pilot anaerobic-aerobic (UCBR)-(MBS) plants in their application to both organic carbon and nitrogen removal.

Table 1- Characteristics of Nitrosomonase and Nitrobacter

	Nitrosomonase	Nitrobacter
Morphology	Rod-shaped	Rod-shaped
Cell size	1*10-6 by 1.5*10-6 (m)	0.5 *10-6 by 1*10-6 (m)
Gram Test	Negative	Negative
Mobile	May or may not be	May or may not be
Autotroph	Obligate	Facultative
Dissolved Oxygen Requirement	Strict Aerobic	Strict Aerobic
Optimum Temperature	5-35 (0 c)	5-35 (0 c)
Optimum p H	7.8-9.2	8.2-9.2
Estimated Generation Time	8-36 (hr)	12-59 (hr)
Free-Energy Efficiency	11-27	34

Materials and Methods

The technical and operating data as well as a simplified figure and flowsheet of the pilot plant are shown in Table 2, Figure 1 and Figure 2 respectively. The pilot plant was operated in the post-denitrification mode, with two reactors in use. The first reactor was always aerobic and the second one was anaerobic. The process was based on the biofilm principle and the biomass grew on small elements that move along with the wastewater in reactor.

The movement was typically produced by coarse bubble aeration in aerobic and mechanical mixing in an anaerobic reactor.The biofilm carrier elements were made of 0.9 specific gravity of polyethylene, about 13 mm long and 13.5 mm in diameter. The aerobic reactor was filled to 80% volume and the Anaerobic reactor was filled to 60%volume, providing a specific surface area equal to 192.5 m^2/m^3. As shown in Figures 1 and 2 , two anaerobic- aerobic reactors were operated in post-denitrification mode to study the feasibility of treating high ammonia and COD load wastewater without spending extra expense to add an external carbon source and to provide high C/N ratio for the denitrification process.

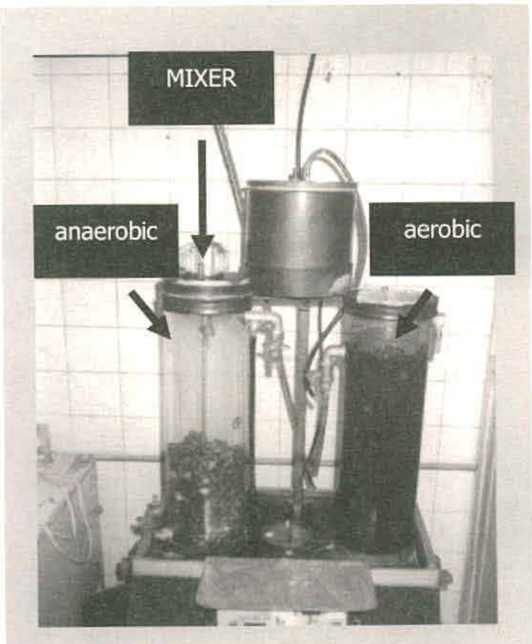

Figure 1- The two anaerobic- aerobic reactors

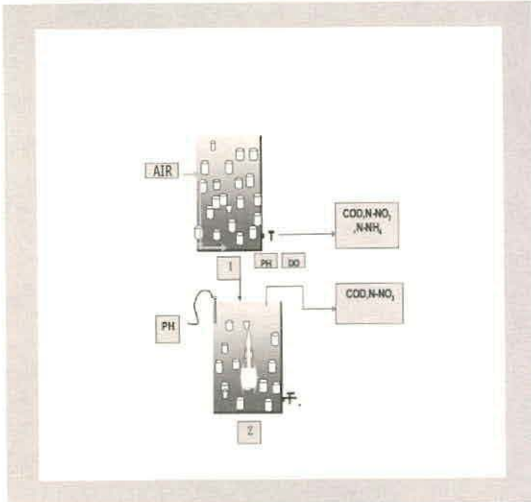

Figure 2- Flowsheet of the two reactors

This configuration also helps to reduce influent COD to the denitrification reactor .

The technical and operating data of the pilot plant are given in Table 2.

Table 2- Technical and Operating data for UCBR and MBS

Technical Dates	Feed Tank	UCBR	MBS
Hight	62 cm	55 cm	55cm
Diameter	33 cm	15 cm	15 cm
Volume	50 lit	9 lit	9/1 lit
Shaft Height	....	-----	48 cm
Impeller Diameter	....	10 cm	10 cm
Filling Ratio	....	80%	60%
Electromotor	....	-----	*DC.5 Amp.,40 Volt*
Aeration	....	*Coarse Bubble*	-----

A water lock was located above denitrification reactor and any biogas exiting from anaerobic reactor passes through a water column and exits from a water lock to prevent air entering the anaerobic reactor.

These experiments were carried out to study the effect of HRT, COD load and NH_4^+ load on nitrification and denitrification rate in 2-20 HRTs, two COD loads in each HRT and variable ammonia concentrations for each COD load. The process was tested in a pilot plant for the treatment of a high ammonia and COD load. The composition of the synthetic wastewater is: (NH_4-N: 250-1000 (mg/lit), COD: 800-1500 (mg/lit),). NH_4HCO_3 and NH_4CL were used as the ammonia sources and phosphorous was provided by adding KH_2PO_4, NH_4HCO_3 and KH2PO_4 which were chosen as buffer compounds to control the pH of process. Micronutrients such as Cu,Cl,Mg,Na,and Fe were added to the system as $CuSO_4$ (2 mg/lit), $MgSO_4$ (3 mg/lit), $FeCl_3$ (0.4 mg/lit), and NaCl (0.7 mg/lit).

This wastewater consisted of: 764.2mg/l ammonium chloride,1029.4mg/l sodium acetate, 1200mg/l sodium bicarbonate,28.1mg/l di-potassium hydrogen phosphate and 1mg/l of trace element solution. Each liter of trace element solution contained 10g calcium chloride, 8g ferric chloride, 5g magnesium sulphate, 2g cobalt chloride, 2g thiamine-HCL, 1g sodium silicate, 550mg aluminum sulphate, 50mg manganese chloride, 1mg ammonium molybdate, 1mg copper sulphate, 1mg zinc sulphate and1mg boric acid (Table 3).

Temperature and pH were measured in each bioreactor every working day, immediately before sampling. The influent wastewater and the content of the UCBR and MBS at the end of aerobic and anaerobic reactors were sampled everyday. The samples were analysed immediately after sampling to obtain the parameters shown in Table 4 and 5. The parameters were measured according to the Standard Methods (1992) (Halling- sorensen and Jorgensen, 1993).

Results

Batch Operation

The experiment was aimed at studying the behavior of the MBBR for COD removal and also simultaneous nitrification and denitrification during the aerobic and anaerobic stages. The batch operstion was used as a start-up for the growth of biofilm on packing. After this period, the biofilm appeared on the packing elements and UCBR appeared to be ready for batch operation. Characteristics of the initial aerobic and anaerobic wastewater are given in Table 6.

Table 3- Analysis of the trace elements

	ammonium chloride	sodium acetate	sodium bicarbonate	di-potassium hydrogen phosphate	Trace elements											
					1											
mg/l	764.2	1029.4	1200	28.1	calcium chloride	ferric chloride	magnesium sulphate	cobalt chloride	thiamine-HCL	sodium silicate	aluminum sulphate	manganese chloride	ammonium molybdate	copper sulphate	zinc sulphate	boric acid
					10g	8g	5g	2g	2g	1g	550mg	50mg	1mg	1mg	1mg	1mg

Table 4- Removal rate of the ammonia in UCBR with Vari-Inf-Ammonia

AEROBIC- COD=800mg/l									
R emoval %	Conc (mg/l)	R emoval %	Conc (mg/l)	R emoval %	Conc (mg/l)	R emoval %	Conc (mg/l)	R time	NO
0.0	400	0.0	350	0.0	300	0.0	250	0	1
22.0	312	11.1	311	23.3	230	14.4	214	2	2
36.0	256	23.4	268	35.0	195	30.4	185	4	3
58.8	165	55.4	156	55.0	135	38.4	154	6	4
53.8	185	53.7	162	66.7	100	36.0	160	8	5
51.5	194	70.9	102	78.3	65	51.6	121	10	6
62.0	152	74.0	91	82.0	54	64.4	89	12	7
76.5	94	86.3	48	84.7	46	68.8	78	14	8
76.3	95	90.0	35	89.3	32	70.4	74	16	9
85.0	60	94.0	21	94.0	18	75.6	61	18	10
97.5	10	93.4	23	95.0	15	96.0	10	20	11
96.5	14	97.7	8	96.0	12	98.4	4	24	12
0.0	700	0.0	650	0.0	520	0.0	450	0	1
27.1	510	15.7	548	27.5	377	22.2	350	2	2
28.6	500	36.8	411	47.3	274	28.7	321	4	3
39.9	421	30.6	451	61.7	199	37.8	280	6	4
44.6	388	50.6	321	72.1	145	48.4	232	8	5
63.6	255	62.9	241	79.8	105	44.2	251	10	6
70.0	210	71.5	185	85.4	76	58.0	189	12	7
73.6	185	73.2	174	89.4	55	63.3	165	14	8
79.7	142	81.1	123	91.9	42	68.9	140	16	9
90.7	65	84.9	98	94.2	30	75.1	112	18	10
97.0	21	86.9	85	96.0	21	96.0	18	20	11
98.0	14	93.7	41	95.2	25	95.3	21	24	12

AEROBIC - COD=800mg/l									
R emoval %	Conc (mg/l)	R emoval %	Conc (mg/l)	R emoval %	Conc (mg/l)	R emoval %	Conc (mg/l)	R time	NO
0.0	1000	0.0	900	0.0	800	0.0	750	0	1
14.5	855	19.7	723	9.9	721	3.9	721	2	2
28.8	712	33.3	600	12.5	700	13.2	651	4	3
64.5	355	54.3	411	33.4	533	29.1	532	6	4
71.5	285	75.3	222	48.6	411	40.8	444	8	5
75.8	242	71.8	254	62.3	302	57.1	322	10	6
80.0	200	80.7	174	59.8	322	53.1	352	12	7
89.6	104	87.7	111	64.4	285	63.3	275	14	8
91.5	85	86.3	123	73.8	210	71.3	215	16	9
94.0	60	95.1	44	86.1	111	78.0	165	18	10
96.0	40	92.7	66	95.9	33	91.7	62	20	11
95.4	46	98.7	12	93.9	49	96.9	23	24	12

AEROBIC- COD=1500mg/l									
Removal %	Conc (mg/l)	Removal %	Conc (mg/l)	Removal %	Conc (mg/l)	Removal %	Conc (mg/l)	R time	NO
0.0	400	0.0	350	0.0	300	0.0	250	0	1
10.0	360	8.0	322	23.3	230	12.0	220	2	2
17.5	330	18.6	285	30.0	210	25.0	195	4	3
61.3	155	40.0	210	51.7	145	38.4	154	6	4
53.8	185	27.1	255	66.0	102	32.0	170	8	5
49.5	202	65.4	121	71.7	85	46.4	134	10	6
58.8	165	71.4	100	78.3	65	64.0	90	12	7
75.0	100	86.3	48	78.0	66	68.8	78	14	8
74.3	103	84.6	54	85.0	45	68.4	79	16	9
78.8	85	90.6	33	86.3	41	74.0	65	18	10
93.8	25	91.4	30	92.0	24	91.6	21	20	11
92.5	30	90.0	35	90.7	28	98.0	5	24	12

Removal %	Conc (mg/l)	Removal %	Conc (mg/l)	Removal %	Conc (mg/l)	Removal %	Conc (mg/l)	R time	NO
0.0	700	0.0	650	0.0	520	0.0	450	0	1
5.7	660	2.2	636	8.3	477	2.2	440	2	2
35.6	451	25.4	485	38.1	322	11.6	398	4	3
39.9	421	24.9	488	59.4	211	21.1	355	6	4
43.0	399	43.8	365	73.1	140	28.7	321	8	5
63.6	255	56.2	285	68.3	165	40.4	268	10	6
67.9	225	67.5	211	83.1	88	55.1	202	12	7
70.0	210	69.2	200	85.2	77	57.8	190	14	8
76.4	165	76.0	156	87.5	65	57.3	192	16	9
89.3	75	83.2	109	92.7	38	63.1	166	18	10
95.3	33	84.8	99	95.6	23	81.1	85	20	11
96.9	22	90.0	65	96.2	20	85.6	65	24	12

AEROBIC- COD=1500mg/l

Removal %	Conc (mg/l)	Removal %	Conc (mg/l)	Removal %	Conc (mg/l)	Removal %	Conc (mg/l)	R time	NO
0.0	1000	0.0	900	0.0	800	0.0	750	0	1
1.0	990	5.0	855	2.9	777	3.9	721	2	2
23.5	765	22.3	699	12.5	700	13.2	651	4	3
64.5	355	42.1	521	18.1	655	26.0	555	6	4
45.6	544	64.2	322	47.4	421	40.8	444	8	5
67.8	322	70.6	265	61.3	310	51.3	365	10	6
73.5	265	79.4	185	59.8	322	51.7	362	12	7
83.5	165	70.6	265	64.4	285	60.1	299	14	8
89.9	101	85.0	135	75.0	200	61.6	288	16	9
92.3	77	92.7	66	80.6	155	72.0	210	18	10
95.5	45	92.7	66	95.0	40	92.0	60	20	11
92.2	78	92.7	66	95.3	38	90.7	70	24	12

Table 5- Removal rate of nitrate in the MBS with Vari-influent-Nitrate

ANAEROBIC- COD=800mg/l									
Removal %	Conce (mg/l)	Removal %	Conce (mg/l)	Removal %	Conc (mg/l)	Removal %	Conc (mg/l)	R Time	NO
0.0	600	0.0	507	0.0	400	0.0	250	0	1
25.0	450	19.7	407	25.0	300	20.0	200	2	2
29.8	421	35.7	326	37.5	250	44.0	162	4	3
53.3	280	48.3	262	59.5	162	51.6	121	6	4
57.5	255	58.6	210	59.5	162	55.2	112	8	5
66.7	200	66.7	169	72.3	111	59.2	102	10	6
81.7	110	73.4	135	73.0	108	77.6	56	12	7
83.3	100	83.2	85	82.5	70	83.2	42	14	8
76.7	140	83.2	85	82.3	71	82.8	43	16	9
81.3	112	86.2	70	80.0	80	84.4	39	18	10
ANAEROBIC- COD=1500mg/l									
Removal %	Conc (mg/l)	Removal %	Conc (mg/l)	Removal %	Conc (mg/l)	Removal %	Conc (mg/l)	R Time	NO
0.0	600	0.0	507	0.0	400	0.0	250	0	1
18.7	488	18.7	412	19.5	322	2.0	245	2	2
22.0	468	27.8	366	22.5	310	20.8	199	4	3
50.0	300	48.3	262	58.8	165	38.0	155	6	4
55.8	265	49.9	254	59.5	162	51.6	121	8	5
65.0	210	60.9	198	68.8	125	42.0	145	10	6
81.5	111	75.3	125	72.5	110	73.6	66	12	7
81.3	112	84.2	80	80.5	78	77.6	56	14	8
80.0	120	83.2	85	77.8	89	73.2	67	16	9
72.5	165	85.4	74	82.5	70	82.0	45	18	10

Table 6- Characteristics of initial wastewater in both systems

Parameter	Aerobic UCBR	Anaerobic MBS
COD(mg/lit)	800	200
NH_4^+(mg/lit)	520	22
NO_3^-(mg/lit)	350	475
PH	6/6-7/9	6/1-7/7

For a period of 160 days, the pilot plant was operated and the experimental results for different HRTs , COD and nitrogen-loading rates are shown in Figure 3 to Figure 6.

· Because of the high concentration of nitrates in the effluent, the Hydraulic Retention Time (HRT) was increased to 24hr.

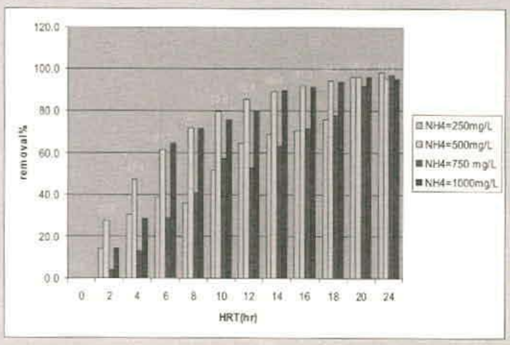

Figure 3- Ammonia removal at different HRTs in the aerated tank

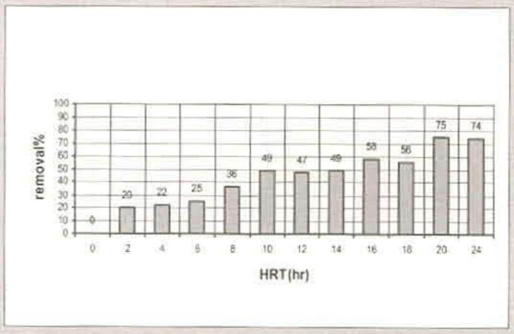

Figure 4- COD removal at different HRTs in the aerobic tank

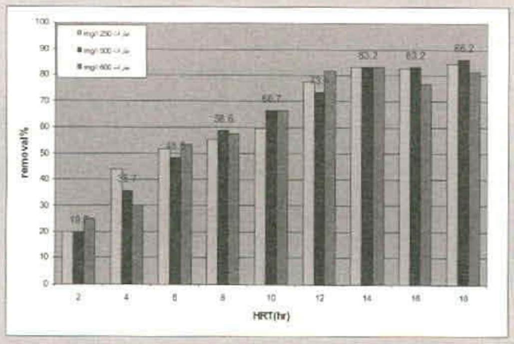

Figure 5- Nitrate removal at different HRTs in the anaerobic tank

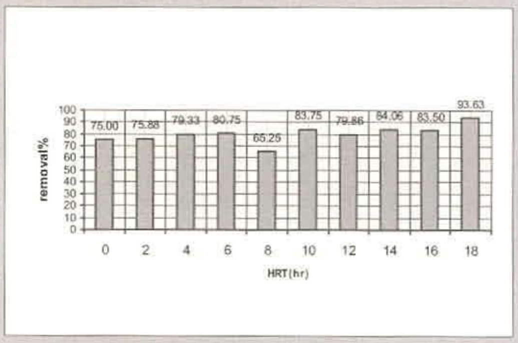

Figure 6- COD removal at different HRTs in the anaerobic tank

Discussion

As shown in Figures 3 to 6, the nitrification and denitrification rate increases when HRT increases. It can be concluded that the competitive inhibition effect at high COD loads influenced the nitrifier bacteria, which compete with carbonaceous bacteria at high COD loading rates. At higher ammonia loads it is easier for nitrifiers to compete with the other microorganisms, to consume the dissolved oxygen in system.

The nitrification rate has a dual effect on COD removal. On the one hand, COD removal increases when a high nitrification rate occurs because of the higher activity of the nitrifiers. On the other hand, when the nitrification rate increases, more nitrate enters the anaerobic reactor and, as a result, more denitrification and subsequently more COD removal occurs. The effect of nitrate concentration on denitrification rate is shown in Figure 5.

An another important result obtained is that the influence of nitrate concentration is more important than the C/N ratio which has been regarded as one of the most important factors on the denitrification rate.(see Figure 7 to Figure 9)

The other results show the low sensivity of the UCBR to HRT and the insignificant effect of HRT change on COD removal and the denitrification and nitrification process, showing the high stability of the UCBR.

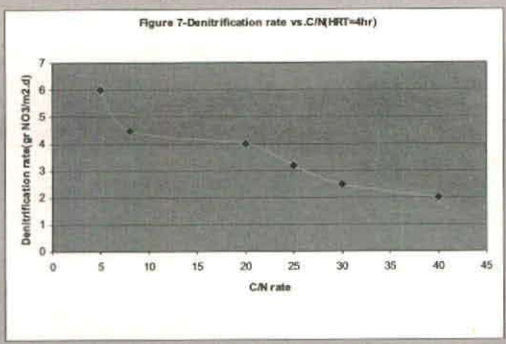

Figure 7- Denitrification rate vs.c/n(HRT=4hr)

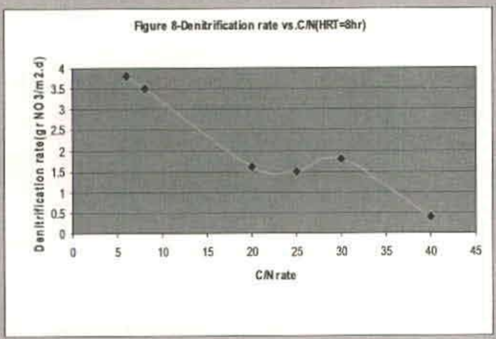

Figure 8- Denitrification rate vs.c/n(HRT=8hr)

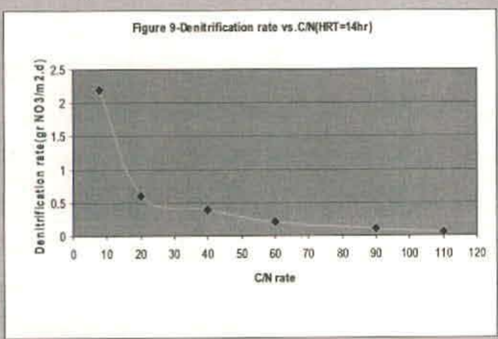

Figure 9- Denitrification rate vs.c/n(HRT=14hr)

Conclusions

From the different tests in pilot-scale plants, the following experiences have been gained with UCBR-MBS:

1) The reactor has demonstrated its capability for he nitrification, denitrification and organic removal process for a broad range of ammonia and COD.

2) The major advantages of UCBR as compared to other systems are its simplicity in operation, low space requirement, stability, reliability, good settlability, low head loss, no bulking and lack of bachwash requirement.

3) The percentage of COD removal did not fall below 75% and was most of the time more than 85%.

4) The percentage of ammonia removal was mostly more than 95% at 20 hr and the nitrate removal percentage above 80% at 14 hr.

5) The percentage of COD removal was more than 75% at 20 hr in aerobic tank and above 80% at 14 hr in an anaerobic tank.

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