

Simultaneous COD and Ammonium Nitrogen Removal from a High-strength Wastewater in a Shaft-type Aerobic Hybrid Bioreactor

Debabrata Mazumder

Abstract — A hybrid model of activated sludge reactor has been developed in laboratory scale to explore the feasibility of carbon oxidation along with nitrogen removal through nitrification and denitrification in the same reactor. The reactor was operated initially under suspended growth only and then hybrid system containing 20 g/L of tyre tube beads. The reactor was operated continuously with the high strength composite synthetic wastewater containing COD and NH_4^+ -N in the range of (1000 – 3500) mg/L and (500 – 1750) mg/L respectively that follows COD : NH_4^+ -N equals to 2 : 1. About 80% removal of COD was achieved at a HRT of 8 hrs. even under purely suspended growth condition. At least 12 hrs. HRT was needed to achieve more than 90% NH_4^+ -N removal for an influent NH_4^+ -N of 1000 mg/L or more. Nitrite and nitrate formation was promoted with the addition of attached biomass by means of bio-carriers. The maximum extent of nitrite and nitrate formation was about 43.9% and 35.3% of influent NH_4^+ -N respectively.

Index Terms— ammonia nitrogen removal, COD removal, hybrid bioreactor, shaft-type.

I. INTRODUCTION

The aerobic hybrid bioreactor is recently being used to augment the additional treatment capacity of conventional activated sludge process. This system essentially comprises biomass both in suspended growth and attached phase, either under fixed or moving condition. Obviously, the introduction of attached biomass by means of carrier materials or fixed media enhances the total microorganism concentration in the reactor resulting in better removal of carbonaceous organic mater (expressed in terms of COD). Apart from heterotrophic carbon oxidation, a considerable amount of nitrification can also take place depending on the availability of dissolved oxygen (DO) concentration by the biomass attached with the media. With the progress of nitrification, DO level deteriorates reaching to a limiting value for aerobic oxidation, which may inculcate denitrification by the facultative denitrifiers present in the suspended growth stage. Furthermore, DO concentration gradually decreases towards the inner surface of the biofilm, creating an anoxic zone. Therefore, denitrification can take place at the inside of the

biofilm in presence of organic carbon derived from endogenous decay of the heterotrophs in addition to some amount of residual organic carbon.

In the year 1996, one of the two streams of the Annapolis, Maryland, USA step aeration activated sludge treatment plant of capacity 37,000 m³/d was modified by single-sludge anoxic-aerobic operation, and then fixed-film media were integrated into the aerobic zone to enhance nitrification [14]. The integrated *Ringlace* media increased the nitrification rate per unit volume to 225% of that observed in the control section, attaining a value of 1.75 kg NH_3 -N /d per linear meter at 15°C. The media also increased denitrification in the aerobic media section permitting a potential of 25% or more reduction in the volume of the anoxic zone. Nitrification-denitrification was studied by Jih *et. al.* (2001) in a single-sludge reactor system treating high-strength nitrogen wastewater [6]. The NH_4^+ -N removal efficiencies (98 - 82%) were observed to be higher than total nitrogen removal efficiencies (71 - 43%).

The hybrid activated sludge-biofilm system like Integrated Fixed Film Activated Sludge (IFAS) has recently become popular for enhanced nitrification and denitrification in aerobic zones because it is an alternative to increasing the capacity of treatment plant units for accomplishing year round nitrification and nitrogen removal [16]. Simultaneous nitrification and denitrification (SND) via the nitrite pathway has also become a promising method that can significantly reduce the energy and COD demand for nitrogen and phosphorus removal [18].

The South-Budapest Municipal Wastewater Treatment Plant (SBWWTP) based on the high-load activated sludge process (ASP) was upgraded into nutrient removal in 1998-1999 in Hungary [11]. The study conducted over 25 months revealed that besides the efficient pre-denitrification obtained in the AS basin, significant ammonium oxidation was observed in the aerated zone. In the same year, a series of bench-scale nitrification/denitrification tests were carried out with both suspended growth and hybrid bioreactors [15]. The hybrid reactor was filled with plastic (polyethylene) media to evaluate the effects of biofilm. The results from the reactor operated under a MCRT of 10 days demonstrated that the hybrid system showed greater stability in nitrification and higher denitrification efficiency than the suspended growth system.

A laboratory-scale fluidized-bed reactor with an external aeration loop was used for nitrification of high-strength ammonium wastewater (up to 500 mg NH_4 -N/L) [2]. The

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results demonstrated that the system was capable of handling ammonium removal rates up to 2.5 kg NH₄-N/m³.d, while removal efficiencies were as high as 98% and independent of the applied ammonium loading rates. Ammonium loading rates higher than 2.5 kg NH₄-N/m³.d resulted in reduced ammonium removal efficiency. The analysis also showed that almost complete ammonium removal occurred at DO concentrations as low as 0.3 - 0.5 mg/L. Activated sludge/biofilm hybrid process treating municipal wastewater was also studied in pilot plant trials [3]. A new type of suspended carrier, with large effective surface area, was employed in the process with the aim of enhancing nitrification. The nitrification rate obtained on the new carrier within the hybrid stage was in the range of 0.9 - 1.2 g NH₄-N/m².d corresponding to a volumetric rate of 19 - 23 g NH₄-N/m³.h. More than 80% of the total nitrification took place on the carrier and the remainder in the suspended solids.

The Kolkata Municipal Corporation is the municipal authority of metropolitan city Kolkata in India. It has a centenary-old drainage system, which appears to be insufficient to comply with the minimum environmental standards for disposal of wastewater. The existing drainage system was overviewed and pollution status was examined at various stretches by means of wastewater characterization [10]. The results of characterization implied that water pollution in some places, especially adjacent to the industries became severe. There are a number of wastewater treatment plants based on conventional technologies like activated sludge process or trickling filter in the city for treating domestic wastewater. With the development of various townships, a few low-cost treatment systems like aerated lagoon, anaerobic-aerobic pond are also under operation for treating community wastewater. There is a cluster of small and medium scale tanneries in the eastern part of the city that contributes wastewater to the municipal sewer. Moreover, a large number of small and primitive slaughterhouses are located in the area, discharging wastewater with high amount of organic carbon and nitrogen. Indeed, those two industrial wastewater sources may get diluted with the domestic one causing reduction in effective pollution load, it often creates problem to the existing sewage treatment plant (STP).

Since the domestic wastewater contains a marginal amount of organic carbon and little amount of nitrogen, the existing treatment plants are designed to cater such a pollution load only. In most cases, the treatment plants become failed to meet the effluent discharge standard in terms of COD (representing organic carbon) and TKN (representing total nitrogen) as laid down in Indian Standards [5]. A recent wastewater survey on East Calcutta Wetland also revealed this fact showing high value of COD and TKN. In view of that the above two industrial wastewater streams should be separately treated prior to disposal into public sewer. The tannery wastewater exhibits all the characteristics of strong wastewater with moderate organic carbon and inorganic nitrogen [13]. Furthermore, the slaughterhouse wastewater is a potential source of organic carbon and organic nitrogen that can also be used as a carbon source for denitrification [1].

To study the feasibility of individual/combined treatment of both those wastewater, one laboratory scale "Hybrid

Reactor" has been developed in form of a shaft type ASP system and operated with similar type of synthetic wastewater. The high-rate aerobic systems are applicable for a BOD₅ concentration as high as 2500 mg/L corresponding to (F/M) ratio of 1.5/day, HRT of 4 hrs. and MLVSS concentration of 10000 mg/L [12]. Therefore, the present study was conducted under that range of COD concentration with additional NH₄⁺-N concentration equals to 50% of COD. The present paper encompassed such a study with the synthetic wastewater bearing COD and NH₄⁺-N of (1000 - 3500) mg/L and (500 - 1750) mg/L, respectively.

II. EXPERIMENTAL

A. Experimental Setup

The experimental hybrid reactor was of overall volume 16 litres out of which 1.4 litres was used as the bio-carrier settler. A circular secondary clarifier of volume 5.7 L was also fabricated to facilitate sludge separation. The biomass recycling from the secondary clarifier to the hybrid reactor was made by means of a peristaltic pump. The feed was given using a centrifugal pump along with a bypass flow recovery system. Air was supplied by means of at least 6 nos. of small air pumps (commonly known as "Aqua Pump" and widely used for aeration in Aquariums), which ensured airflow of (7 - 8) L/min. Square shaped tyre tube beads of 5 mm average dimension were used as the carrier particles in the present study. The tyre tube beads possessed specific gravity of 1.14 and specific surface area of about 140 m²/m³. These particles were introduced to the main reactor with a mass required to make a particle concentration of 20 g per litre volume of the reactor. The required quantity of particle was directly poured from the top opening and was kept in suspension throughout the operation. These are also allowed to settle in an inclined tube of diameter 50 mm, connected to the main reactor, which prevents their entry into the secondary clarifier. After any reactor operation is over, the carrier particles were taken out from the bottom hopper along with the liquid and then separated. The schematic diagram of "Hybrid Reactor" system is shown in Figure 1. The reactor was operated firstly under purely suspended growth and then hybrid condition i.e. in presence of 20 g/L of tyre tube beads.

The reactor was started-up with the acclimated nitrifying biomass developed earlier by shake-flux method. The reactor was also provided with about 50 g/L of shredded tyre tube beads to facilitate the growth of sufficient biomass in the attached phase. This batch system was fed with 200 - 500 ml of synthetic wastewater prepared (Table 1) everyday and aerated with 4 nos. of air pumps. After 28 days, a suspended biomass (MLVSS) of about 2700 mg/L and attached biomass of 32.9 mg/g of tyre tube beads were achieved. During the start-up period undesirable fluctuations of pH was adjusted with buffering solution.

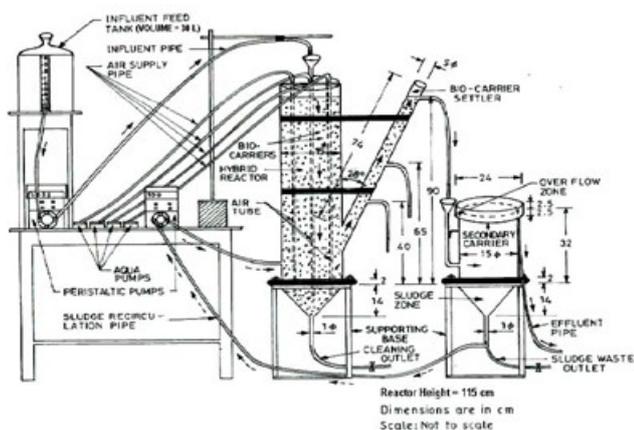


Figure 1. Schematic arrangement of hybrid beactor system

B. Feed Solution

Two types of synthetic wastewater were used – one containing predominantly organic carbon (Type I) and the other one composed of inorganic ammonia nitrogen as well as inorganic carbon (Type II). Synthetic wastewater of Type I was prepared for providing mainly biodegradable organic carbon (biodegradable COD). Dextrose was used in preparing the synthetic wastewater as a simple substrate, which is highly biodegradable. It was also selected in the reason that 1 g/L of dextrose solution is expected to produce a COD concentration of about 1000 mg/L. Synthetic wastewater (Type II) was used to feed the reactor with different concentrations of ammonia nitrogen ($\text{NH}_4^+\text{-N}$). The pH of Type I and Type II feed solution was estimated as 7.2 and 9.3 respectively. Initially, the activated sludge was acclimated with synthetic wastewater of Type II to produce a bulk amount of nitrifying biomass (autotrophic bacteria). Then varying (as per the input $\text{NH}_4^+\text{-N}$ concentration) fraction of the nitrifying biomass was added to the original activated sludge (predominantly containing heterotrophic bacteria) to conduct the study. Composite synthetic wastewater (feed solution) with desired COD and $\text{NH}_4^+\text{-N}$ was used in this study, which was prepared by mixing Type I and Type II synthetic wastewater accordingly. Concentrated synthetic wastewater (Type I and Type II) was diluted with tap water to obtain the desired concentration of COD and $\text{NH}_4^+\text{-N}$ during continuous study. The composition of the Type-I and Type-II synthetic wastewater is presented in Table 1 and Table 2 respectively.

TABLE 1 : COMPOSITION OF SYNTHETIC WASTEWATER –TYPE I

Item	Concentration (mg/L)
$\text{C}_6\text{H}_{12}\text{O}_6$ (Dextrose)	10000
KH_2PO_4	877
$\text{MnCl}_2, 4\text{H}_2\text{O}$	11
$\text{CaCl}_2, 2\text{H}_2\text{O}$	30.8
$\text{MgSO}_4, 7\text{H}_2\text{O}$	56.5
FeCl_3	4.50

Note : pH of Type I Feed Solution = 7.2

TABLE 2 : COMPOSITION OF SYNTHETIC WASTEWATER (TYPE-II)

Item	Concentration (mg/L)
NH_4HCO_3	56420
NaHCO_3	58810
KH_2PO_4	4390
$\text{MnCl}_2, 4\text{H}_2\text{O}$	11
$\text{CaCl}_2, 2\text{H}_2\text{O}$	30.8
$\text{MgSO}_4, 7\text{H}_2\text{O}$	56.5
FeCl_3	4.50
MoCl_2	2.25

NH_4HCO_3	56420
NaHCO_3	58810
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$\text{CaCl}_2, 2\text{H}_2\text{O}$	30.8
$\text{MgSO}_4, 7\text{H}_2\text{O}$	56.5
FeCl_3	4.50
MoCl_2	2.25

Note : pH of Type II Feed Solution = 9.3

C. Experimental Procedure

Continuous study was conducted with composite synthetic wastewater with the COD : $\text{NH}_4^+\text{-N}$ equals to 2 : 1 to study the extent of COD reduction, $\text{NH}_4^+\text{-N}$ oxidation and denitrification. In this study, COD and $\text{NH}_4^+\text{-N}$ were adjusted in the range of 1000 – 3500 mg/L and 500 - 1750 mg/L respectively. The hydraulic retention time (HRT), solid retention time (SRT), suspended biomass (MLVSS) and attached biomass were considered in the range of (6 - 28) hrs., (51–93) hrs., (3052–3095) mg/L and (40.1– 44.8) mg/g of bio-carrier, respectively. The continuous study was undertaken firstly under purely suspended growth condition and then with hybrid system consisting of 20 g/L of bio-carrier. The thickened sludge concentration in the secondary clarifier remained about 10000 mg/L and based on it the recirculation ratio was adjusted to maintain the desired biomass level in the main reactor. Adequate aeration was done so that the DO concentration always remained in the range of 1.0 - 2.0 mg/L or more. The DO in the reactor suspension was continuously measured by an on-line DO meter made by ORION, USA. For each set of study, the process was allowed to run continuously for a number of times until a quasi-steady state was attained.

D. Analytical Procedure

Several parameters were measured including pH, DO concentration at the initial and final stage, Influent and effluent COD (soluble and biodegradable) and $\text{NH}_4^+\text{-N}$ (soluble) concentration and suspended as well as attached biomass concentration. In addition, initial and final Alkalinity, influent and effluent Nitrite (NO_2^-) as well as influent and effluent Nitrate (NO_3^-) concentration were also measured. All the parameters excepting attached biomass were measured in accordance with methods described in Standard Methods [17]. The attached biomass was estimated on the basis of protein content in the cells as per Lowry's method [9], which has been modified by Herbert *et. al.*[4].

III. RESULTS AND DISCUSSION

A. General Observation

The continuous study under COD : $\text{NH}_4^+\text{-N}$ = 2 : 1 indicated that a sufficiently higher HRT is required to accomplish the oxidation of $\text{NH}_4^+\text{-N}$ (equal to the 50% of COD) in addition to the COD. The COD removal was observed to be more than the $\text{NH}_4^+\text{-N}$ removal in all the cases indicating higher fraction of heterotrophs in the total biomass. The change in pH was not substantial (a maximum of about 2.0) and the final pH went equal or below the favorable pH for nitrification i.e. 7.2 in some cases. The DO level was more than 2 mg/L for COD and $\text{NH}_4^+\text{-N}$ concentration up to 1500 and 750 mg/L respectively. But, it varied in the range of

1.0 - 1.4 mg/L for higher COD and NH_4^+ -N loading. The NO_2^- -N build-up took place at a faster rate with the increase of hydraulic retention time. The NO_3^- -N concentration started to build up at an HRT of 8 hrs and onward. Although the NO_3^- -N is not significant for a lower HRT, it developed progressively with the increase in HRT.

B. Processing of Results

The results obtained from steady state operation of continuous study have been processed to determine the organic loading rate (COD or NH_4^+ -N), NO_2^- -N and NO_3^- -N formation. The performance of the reactor is expressed in terms of COD and NH_4^+ -N removal efficiency. Therefore, variation of COD or NH_4^+ -N removal efficiency is expressed in terms of COD or NH_4^+ -N loading rate. Typical performance curves for purely suspended growth and hybrid system containing 20 g/L of bio-carrier are shown in Figure 2 through Figure 5.

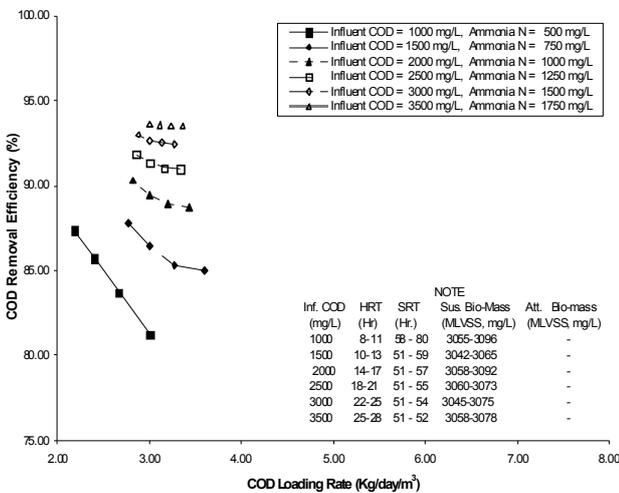


Figure 2 : COD Removal Efficiency Vs. COD Loading rate during continuous study with no bio-carrier

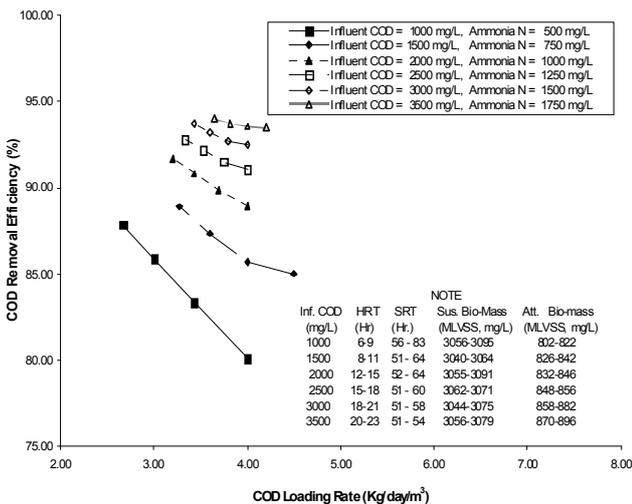


Figure 3 : COD Removal Efficiency Vs. COD Loading rate during continuous study with 20 g/L of bio-carrier

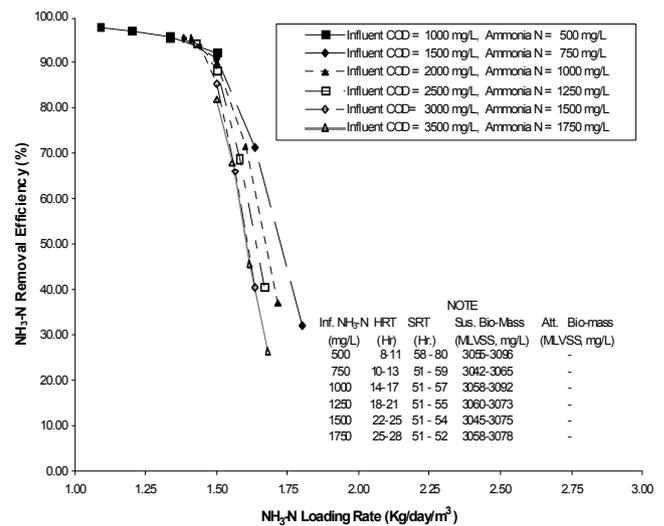


Figure 4: $\text{NH}_3\text{-N}$ Removal Efficiency Vs. $\text{NH}_3\text{-N}$ Loading rate during continuous study with no bio-carrier

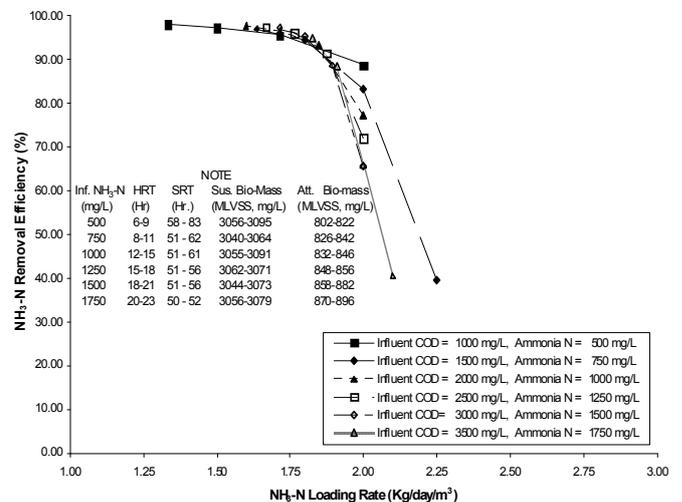


Figure 5: $\text{NH}_3\text{-N}$ Removal Efficiency Vs. $\text{NH}_3\text{-N}$ Loading rate during continuous study with 20 g/L of bio-carrier

Typical profiles of nitrite formation are plotted against the HRT for purely suspended growth and hybrid system containing 20 g/L of bio-carrier in Figure 6 and Figure 7.

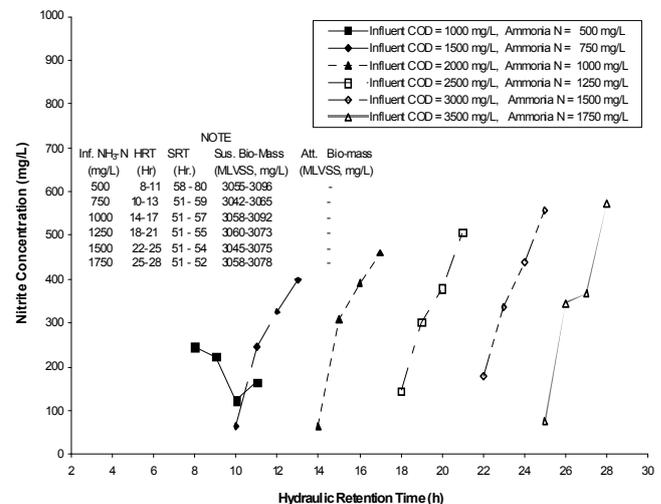


Figure 6 : $\text{NO}_2\text{-N}$ Formation Vs. Hydraulic Retention Time during continuous study with no bio-carrier

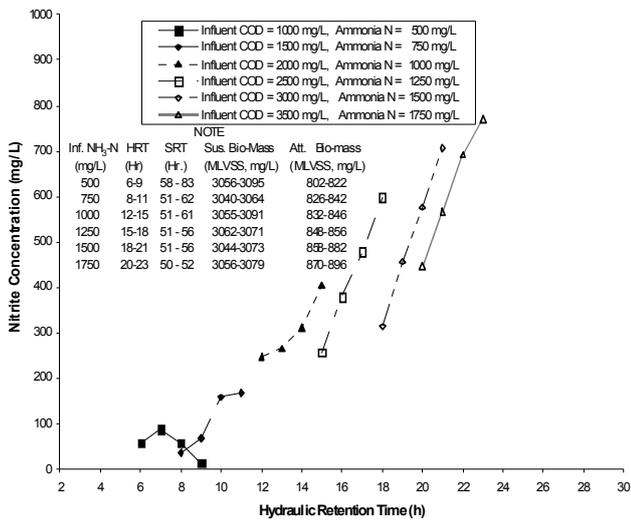


Figure 7 : NO₂-N Formation Vs. Hydraulic Retention Time during continuous study with 20 g/L of bio-carrier

On the other hand typical profiles of nitrate formation are shown against the HRT for purely suspended growth and hybrid system containing 20 g/L of bio-carrier in Figure 8 and Figure 9 respectively.

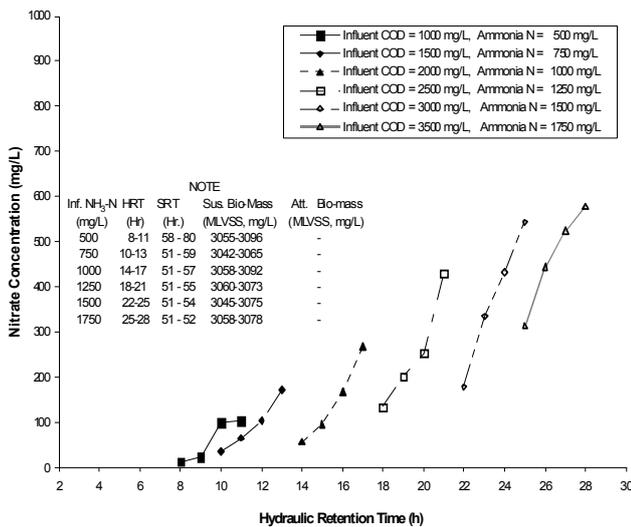


Figure 8 : NO₃-N Formation Vs. Hydraulic Retention Time during continuous study with no bio-carrier

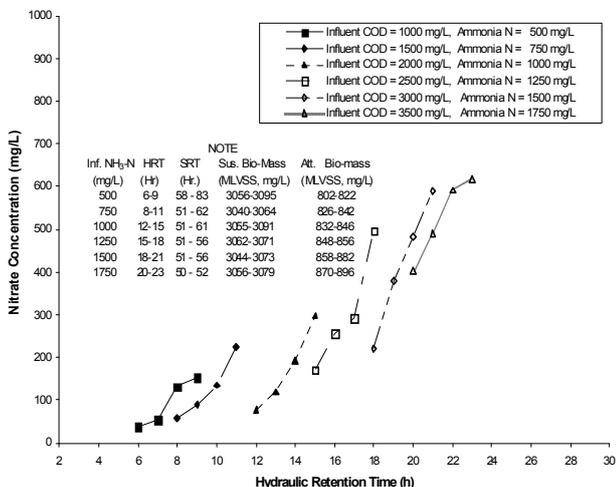


Figure 9 : NO₃-N Formation Vs. Hydraulic Retention Time during continuous study with 20 g/L of bio-carrier

C. COD Removal Performance

The identical nature of Figure 2 and Figure 3 revealed that the hybrid system performed in the same way as a purely suspended growth process. However, the hybrid system can withstand higher COD loading for the same removal efficiency in a suspended growth system. The COD loading rate was increased from (2.18–3.60) kg/day/m³ in purely suspended growth to (2.67–4.50) kg/day/m³ in the hybrid system resulting in a nominal change of COD removal from (81.2–93.6)% to (80.1–94.1)%. Therefore, the COD removal is in a good agreement with the results obtained by Junxin *et. al.*[7]. The COD removal efficiency decreased with the loading rate almost linearly for COD and NH₄⁺-N concentration of 1000 and 500 mg/L respectively. The effect of COD loading rate on the removal efficiency gradually decreased with the increase in influent COD concentration. It implied practically no effect on COD removal due to enhanced COD loading when the reactor comprised of additional attached biomass.

D. NH₄⁺-N Removal Performance

The NH₄⁺-N removal efficiency decreased very slowly with NH₄⁺-N loading rate up to a limiting point and then it decreased very sharply. For purely suspended growth condition, there was practically no effect of NH₄⁺-N loading rate on NH₄⁺-N removal efficiency up to a value of 1.5 kg/day/m³ (Figure 4). However, a drastic effect was observed beyond this loading rate for all the influent COD and NH₄⁺-N concentrations. When the bio-carrier concentration was increased to 20 g/L, NH₄⁺-N removal was increased in most cases. At the same time, the effect of NH₄⁺-N loading rate was observed to be nominal up to a loading rate of 2.00 kg/day/m³ (Figure 5). Beyond this loading rate the effect was prominent for influent NH₄⁺-N concentration of 750 and 1750 mg/L. The maximum NH₄⁺-N removal efficiency for the experimental runs, varied from 39.4 to 98.1%, which corroborates the observations made by Jih *et. al.*[6].

E. Development of Nitrite Concentration

Nitrite concentration started to develop even at a HRT of 8 hrs. under purely suspended growth condition, which was further decreased to 6 hrs. in presence of 20 g/L of bio-carrier. It clearly indicated the real potential of attached biomass towards nitrification. The rate of nitrite formation increased sharply with the increase in HRT for a specific influent COD and NH₄⁺-N concentration. However, the profile of nitrite build-up for different influent COD and NH₄⁺-N concentration appears to be similar in nature (Figure 6). When, 20 g/L of bio-carrier was introduced in the reactor, the rate of nitrite formation became faster, which might be due to inability of further oxidation into nitrate (Figure 7). The extent of nitrite formation enhanced with the increase of COD as well as NH₄⁺-N concentration obviously due to dissolved oxygen shortage, which might have caused first stage denitrification (conversion of NO₃-N to NO₂-N). A distinct phenomenon was observed for a COD and NH₄⁺-N concentration of 1000 and 500 mg/L, where nitrite concentration fall both under suspended growth and hybrid system. This is also to observe that the effect of HRT on Nitrite formation drastically reduced due to addition of 20

g/L of bio-carrier in hybrid system. The maximum extent of nitrite conversion was 32.7% and 43.9% for purely suspended growth and hybrid system containing 20g/L of bio-carrier respectively, which is also commensurate with the results observed by Kual and Verstraete [8].

F. Building-up of Nitrate Concentration

Similar to nitrite ($\text{NO}_2\text{-N}$), nitrate ($\text{NO}_3\text{-N}$) also started to build-up even under purely suspended growth system. However, the extent of nitrate formation was low, which progressively increased with the increase in HRT. The maximum nitrate concentration was observed to be about 580 mg/L, which is 33% of the influent $\text{NH}_4^+\text{-N}$ concentration (Figure 8). When 20 g/L of bio-carrier was introduced, the maximum nitrate formation was further enhanced to 35.3%, although HRT was decreased to 23 hrs. (Figure 9). Both the results on nitrate formation are in agreement with the observations made by Kual and Verstraete [8]. For purely suspended growth condition, nitrate formation increased with a slow rate in case of influent $\text{NH}_4^+\text{-N}$ concentrations of 500 and 750 mg/l. However, the rate increased for influent $\text{NH}_4^+\text{-N}$ concentration up to 1500 mg/L and then decreased for influent $\text{NH}_4^+\text{-N}$ concentration of 1750 mg/L (Figure 8 and Figure 9). It revealed that nitrate concentration is highly dependant on HRT for the $\text{NH}_4^+\text{-N}$ concentrations above 750 mg/L.

IV. CONCLUSIONS

In case of combined carbon oxidation-nitrification with COD and $\text{NH}_4^+\text{-N}$ concentration ranging between (1000 – 3500) mg/L and (500 – 1750) mg/L respectively, about 80% removal of COD can be achieved at a HRT of 8 hrs. even under purely suspended growth condition. At least 12 hrs. HRT is needed to achieve more than 90% $\text{NH}_4^+\text{-N}$ removal for an influent $\text{NH}_4^+\text{-N}$ of 1000 mg/L or more. The present reactor is able to nitrify at a HRT of 8 hrs. even in absence of bio-carriers under the above condition, but an HRT of 6 hrs. is sufficient in case of hybrid condition. Both nitrite and nitrate formation get promoted with the addition of attached biomass in turn bio-carriers. The maximum extent of nitrite and nitrate formation was about 43.9% and 35.3% of influent $\text{NH}_4^+\text{-N}$ in case of COD and $\text{NH}_4^+\text{-N}$ concentration of (1000 – 3500) mg/L and (500 – 1750) mg/L respectively. The denitrification could also take place in the present reactor, which has been supported by the rapid increase in nitrite concentration associated with reduction in DO concentration.

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