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Anaerobic Digestion of Nssc Pulping Effluent

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ABSTRACT: Two UASB reactors R-I and R-II, each of 10liters capacity, made up of acryl material were operated parallel and continuously for a period of 220 days, using actual effluent of the NSSC pulp and paper mill, at mesophilic temperature and neutral pH. The main objective was to investigate the treatability performance of the reactors at varying OLR and HRT in terms of TOC and lignin removal. The reactor R-I was used by conventional procedure, whereas, for the reactor R-II methanol and activated carbon was added to supplement its efficiency. Corresponding to an optimum OLR of 4.5kg/m^3-day and HRT of 18hrs, the reactor R-II gives 69% TOC and 60% lignin removal. But, in the reactor R-I at same operating conditions, only 56% TOC and 51% lignin removal was noticed. However, the biogas yield in the reactor R-I was noticed relatively more than in the R-II. The biogas yield observed was 0.28m^3/kg-COD$_{rem}$ and 0.18m^3/kg-COD$_{rem}$ in the reactor R-I and R-II, respectively, with 56-58% methane content in both the reactors. The average VFAs concentration observed in the reactor R-I and R-II is 360mg/L and 230mg/L, respectively. The results of this study suggest that the use of methanol and activated carbon to enhance the treatability efficiency of the UASB reactor is a viable option for the treatment of NSSC pulping effluent.

Key words: NSSC pulping effluent, UASB reactor,TOC, Lignin

INTRODUCTION

In terms of fresh water extraction, the pulp and paper industry is a water intensive industry and stands third in number over all after the primary metals and the chemical industries. It is also considered to be the sixth major polluter releasing a jumble of gaseous, liquid and solid wastes (Ali *et al.*, 2001, Khallas *et al.*, 1994). In Pakistan, there are more than sixty pulps and paper mills in operation and majority of the mills do not have wastewater treatment facilities to treat their effluent and hence discharge the entire major effluent stream into environment thus poses a great threat to the entire ecosystem (Pak-EPA, 1999). The bleaching effluent of the paper industry contains high amount of toxic and persistent pollutants, which are created as a product of reaction among remaining lignin from the pulping step and chlorine and chlorine mix utilized in the bleaching practice (Pak-EPA, 1999, Savant *et al.,* 2005). Up to now, 500 diverse chlorinated organic compounds have been recognized in the bleaching waste of pulp and paper mills. In wastewater, these compounds are knowable jointly as “absorbable organic halides” (AOX). Quantity of these compounds is completely relative to utilization of chlorine in the bleaching practice of pulp and paper manufacturing (Yan *et al.*, 1994). Most of these compounds are hydrophobic, persistent, bio-accumulative and carcinogenic in nature and causes a varity of harms to human health (Savant *et al.,* 2005). A vast variety of methods have been considered for treating the industrial wastewater all around the world (Uemura *et al.*, 2012a; Uemura *et al.*, 2012b; Shafieiyoun *et al.*, 2012; Rahman and Al-Malack, 2012). Applying anaerobic technology, which is technically simple, fairly economical and uses very little energy, is considered to be one of the most encouraging choice for the treatment of different industrial effluents along with the pulp and paper industrial wastes (Lettinga et al. 1980, Bhatti 1995, Mtethiwa et al. 2008). The employing of anaerobic treatment in pulp and paper industry initiated in early seventies (Savant, 2005). The initial anaerobic digestion process launched was anaerobic lagoon. Contact reactor and upflow anaerobic sludge blanket reactor (UASB) technology was started nearly in eighties. Nowadays, above fifty complete anaerobic treatment systems, treating pulp and paper mills waste matter are working in the world (Bajpai, 2000). Anaerobic processes have turned into extensively recognized treatment systems after the information added in the functioning of numerous anaerobic systems in the world (Schellinkhout, 1993). Complete UASB reactors are functioning now in various parts of the world like Japan, India, Colombia, Brazil etc. for the handling of multiple type of wastes material (Driessen et al. 1994, Vieira et al. 1988; Chernicharo

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2001, Wiegant et al. 2001; Mobarak-Qamsari *et al.*, 2012; Selvamurugan *et al.*, 2012; Adl *et al.*, 2012; Mahmoudkhani *et al.*, 2012). However, very little information are available regarding the use of UASB reactor specifically for the treatment of NSSC (Neutral Semi-Sulfide Chemical) pulping effluent, hence further investigation are needed to evaluate the treatment feasibility and potential of the UASB reactor for the subject wastes. Sharma in 1992 concluded that the black liquor, alone, cannot be easily anaerobically degraded due to the presence of recalcitrant material in it. Recently, it was also shown that the UASB reactor could remove 35% of black-liquor corresponding to an OLR of 2.75kg-COD/m^3-day, which could be further improved by 30%, if an easily biodegradable substance like methanol is added with the feed solution to the reactor (Arshad et el., 2010). It is also the use of an activated carbon with the seeded sludge enhance the treatability efficiency of the UASB reactor, while working on the chlorophenolic (Arshad *et al.,* 2011). Therefore, this study was designed to study the removal of lignin-COD in a single-stage UASB reactor using actual NSSC pulping effluent in the presence of methanol and activated carbon. The key objective of this study was to examine the performance of the reactor at different OLR (Organic Loading Rate) and HRT (Hydraulic Retention Time).

MATERIALS & METHODS

Two UASB reactors, namely R-I and R-II made up of acryl resin material, each of 10liters capacity were used in this study. Water jackets were provided to the reactors to maintain their constant mesophilic temperature. The reactors were also equipped with gas solid separators (GSS) and mixing devices of turbine shape of 1.5"x3.0" size (Arshad and Hashim, 2010). Both the reactors were run at same operating conditions. Actual NSSC pulping effluent obtained from the nearby pulp and paper mill was used as the carbon provider in the supply (influent) for both the reactors, R-I and R-II. For reactor R-II, methanol was added to the actual effluent in ratio of 1:10. Nitrogen and Phosphorous were added to them in the form of $(NH_4)_2SO_4$ and KH_2PO_4 in accordance with the C:N:P ratio of 350:5:1, $MgSO_4.7H_2O$ was added in concentration of 0.1g/L. Trace nutrient like $FeCl_3.6H_2O$, $CoCl_2.6H_2O$, $ZnSO_4$, $CuSO_4$ etc were also added in required proportion (Bhatti et al. 1996, Yaochatchaval et al. 2008). A seeded sludge was prepared by mixing fully granular sludge obtained from a laboratory scale UASB reactor and the digested acclimatized sludge of NSSC pulping effluent. The seeded sludge obtained had mixed-liquid volatile suspended solids (MLVSS) content of 62g/L and VSS (Volatile Suspended Solids) content of 56.80g/L. This sludge was then used for the start-up of both the reactors. But for R-II this sludge was further added with granular activated carbon of an effective size of 1.5-2.5mm, to strengthen its mass, at a concentration ratio of 3:2 (Arshad el al. 2011). Both the reactors were operated continuously for approximately 220 days. Actual wastewater sludge was used in the study obtained from the nearby local NSSC pulp and paper mill. The wastewater characteristics data of the sludge is shown in the given Table 1.0. Biogas was collected over a tap water saturated with NaCl (Bhatti et al. 1996), and were deliberated according to the available standard analytical procedure as shown in the Table 2.0 (AWWA 2005).

RESULTS & DISCUSSION

Both the reactors were started-up simultaneously, operating parallel at constant mesophilic temperature, since it was previously reported that a number of potential problems are associated with higher thermophilic temperature like higher residual volatile fatty acids (VFAs), and at lower temperature the biodegradation process is relatively slow (Buhr et al. 1977, Bryant 1979, Grin *et al.,* 1985, Henze *et al.,* 1983, Kennedy, 1982, Stronach et al. 1986, Switzenbaum et al. 1980, Zinder, 1998). The average temperature hence recorded during this study in both the reactors was 29°C. The pH is also among the most important operating parameters for the anaerobic digestion. As shown earlier the microbial activity is at optimum at neutral pH (Bryant 1979, Bhatti *et al.,* 1995, Lettinga *et al.,* 1984), therefore, during this study the pH in both the reactors were kept around neutral by using an external buffer solution in the form of 0.03M $NaHCO_3$. Though at the start of study no external buffers solution was added as in both the reactors the pH was observed to be beyond 7.4, but after a couple of days a drastic drop in the reactors pH was noticed, thereafter the buffer solution was started added with the feed solution to both the reactors that helps to maintain the neutral pH in both the reactors. The organic loading rate (OLR) was gradually increased, starting from 0.2kg/m^3-day, in order to avoid the volumetric shock to the reactors. The maximum OLR observed was 8.6kg/m^3-day. Likewise, the hydraulic retention time (HRT) was too slowly lowered down from 42hrs to 10hrs, to avoid the wash out of sludge from the reactors. HRT and OLR are the significant design parameters that decide the capital cost, and set up the engineering and financial viability of a particular technology. The connection among HRT and OLR with respect to TOC and lignin elimination competency of the reactors is illustrated in Fig 1-4 and 5-8, respectively.

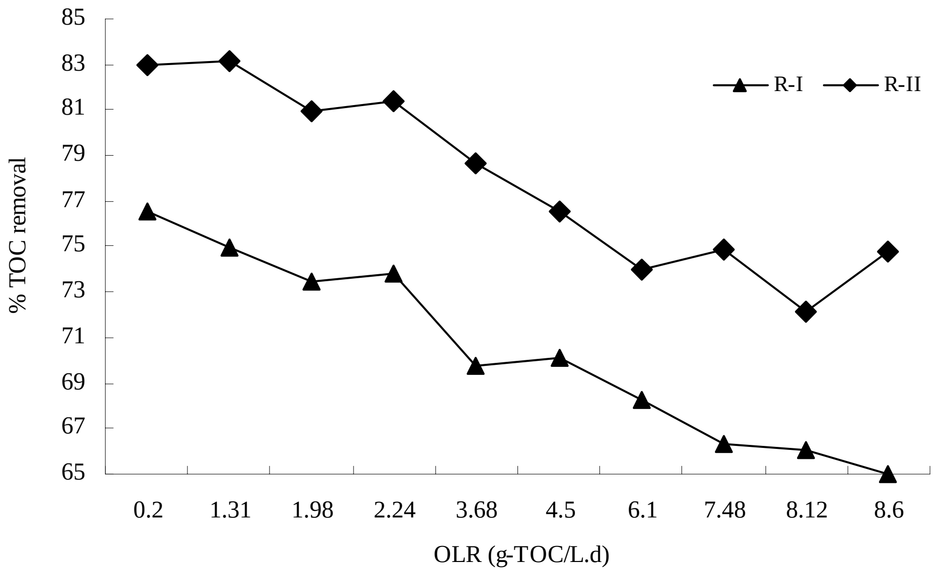
These results shown are derived at stable operating conditions corresponding to each HRT. The higher HRT, as usual proves better treatability performance in terms of TOC and lignin removal, in either reactors. Like indicated, both the TOC and lignin removal is more than 60% corresponding to an OLR of less than 8.5kg/m^3-day in both the reactors. But if the same amount of TOC removal is desire to achieve at lower HRT of 10-18hrs, then the OLR must be kept as

Table 1. Wastewater Characteristics of the Paper Mills Effluent

Parameters	Concentration	Parameters	Concentration
pH	8.6	Chemical Oxygen Demand, COD(mg/L)	1835
Color (units)	1745	Total Suspended Solids, TSS(mg/L)	940
Lignin (mg/L)	498	Total Dissolved Solids, TDS(mg/L)	1585

Table 2. Parameters - frequency and analytical procedure

Parameter	Sampling port	Frequency	Methodology
pH	Reactor	Daily	Electrometric method
Temperature	Reactor	Daily	--
TOC	Inlet and outlet	Twice a week	Multi N/C 3100 Analytik Jena AG (Differential method)
Lignin	Inlet and outlet	Twice a week	Tyrosine method (DR/2010 Spectrophotometer)
VFAs	Outlet	Twice a week	Distillation method
Methane	Biogas	Once a week	Gas chromatographic method



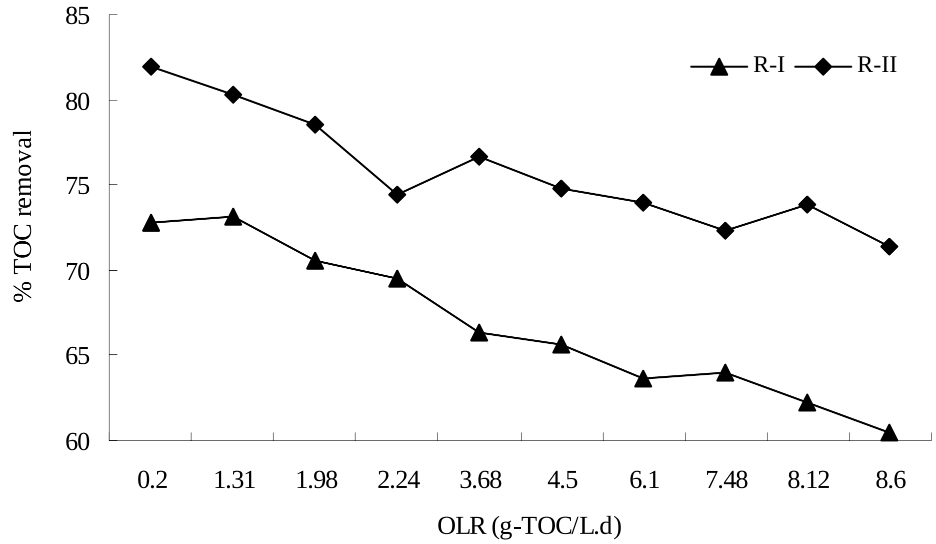


Fig. 2. Relationship b/w OLR and TOC removal at HRT of 30hrs

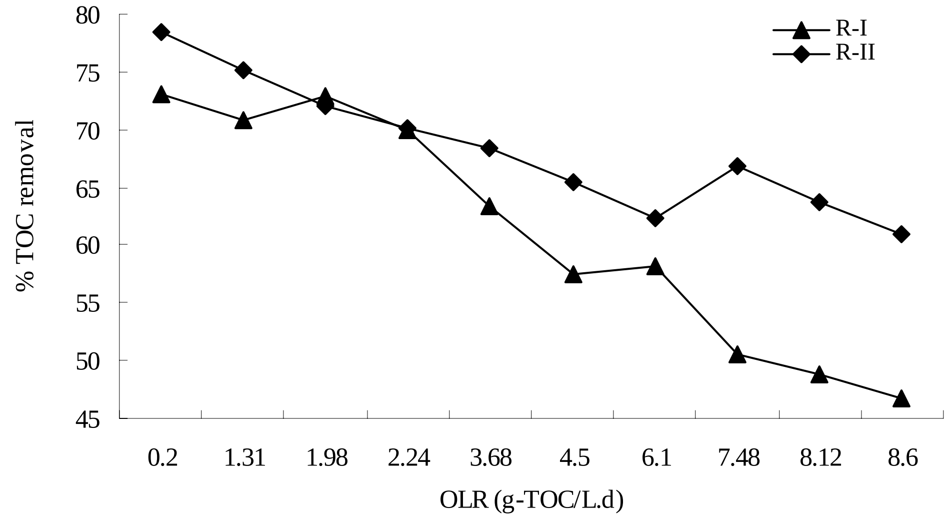


Fig. 3. Relationship b/w OLR and TOC removal at HRT of 18 hrs

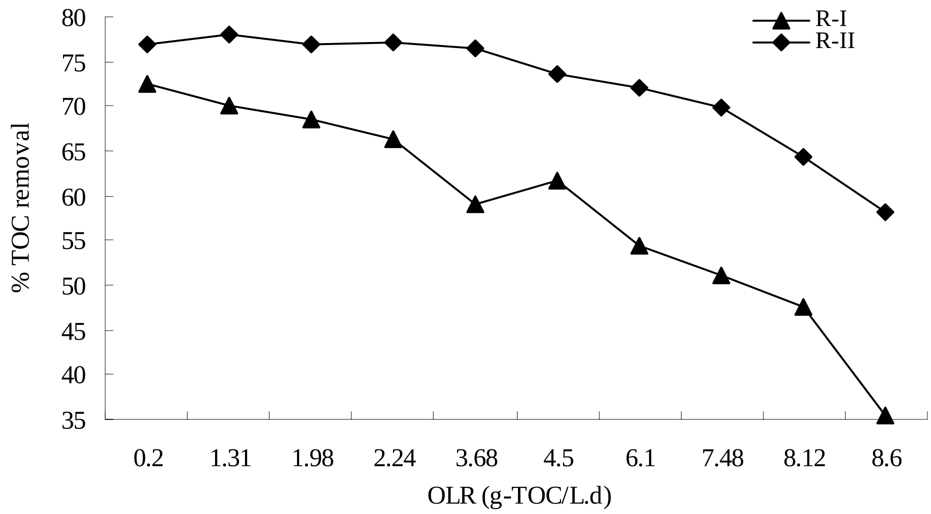


Fig. 4. Relationship b/w OLR and TOC removal at HRT of 10 hrs

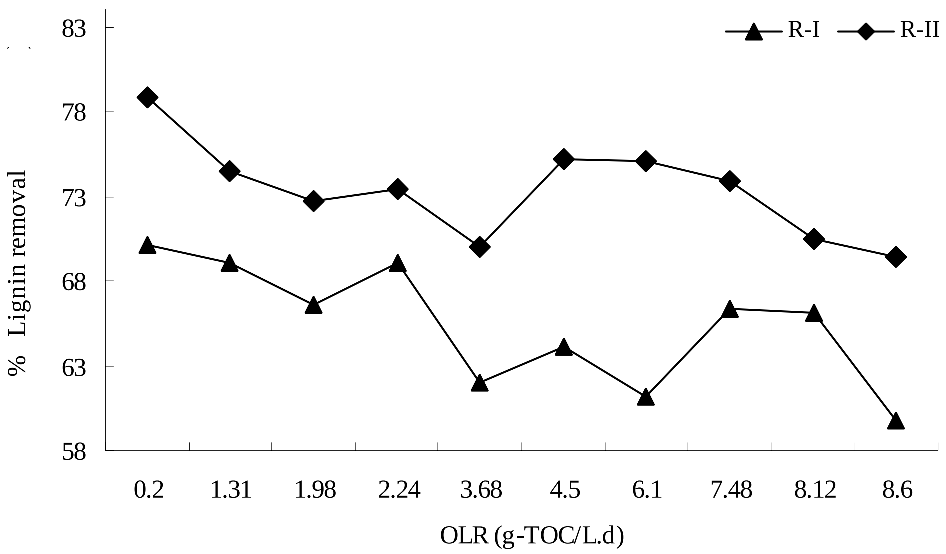


Fig. 5. Relationship b/w OLR and Lignin removal at HRT of 42 hrs

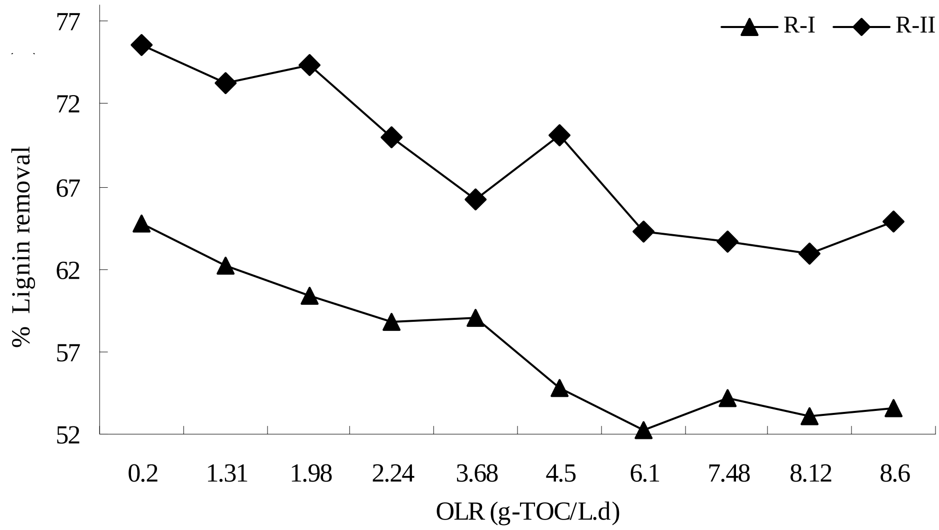


Fig. 6. Relationship b/w OLR and Lignin removal at HRT of 30hrs

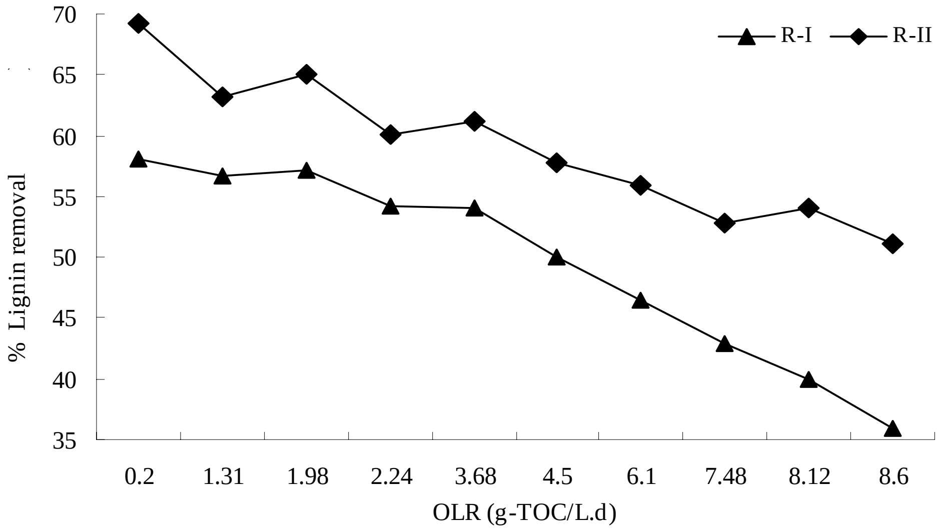


Fig. 7. Relationship b/w OLR and Lignin removal at HRT of 18 hrs

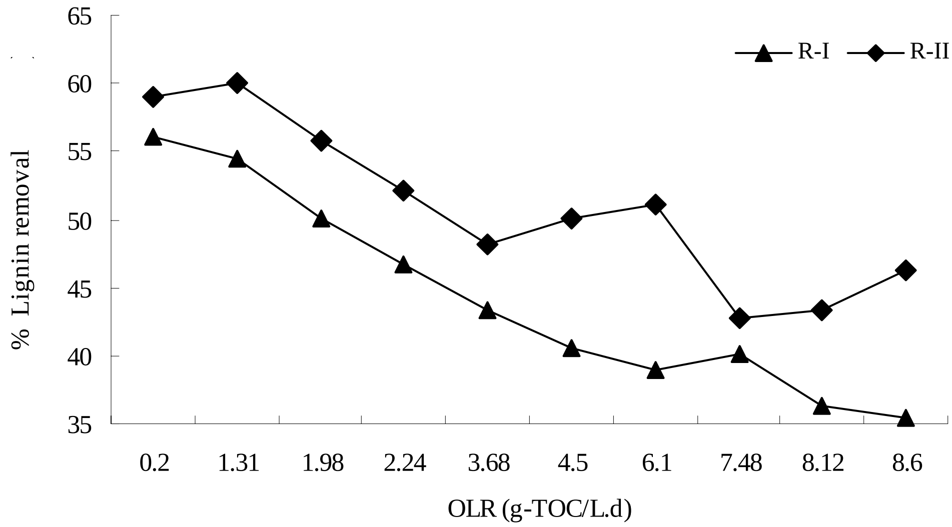


Fig. 8. Reltionship b/w OLR and Lignin removal at HRT of 10 hrs

low as 3.5kg/m^3-day. Further for the lignin reduction, especially corresponding to HRT of 10hrs, the OLR needs to be further reduced to 1.32kg/m^3-day. Although, the higher HRT gives better treatability, but makes the practical design of the reactor uneconomical.

Throughout the study, it was observed that the use of methanol and activated carbon obviously increases the treatability performance of the UASB reactor. As illustrated in the figs, the reactor R-II gives higher efficiency than reactor R-I both in terms of TOC and lignin removal, corresponding to same operating conditions of OLR and HRT. For the optimum and practical design of UASB R-II at HRT of 18hrs and OLR of 4.5kg/m^3-day, the TOC and lignin removal of 69% and 60% could be achieved, respectively. While, the reactor R-I could give only 56% and 51% TOC and lignin removal, respectively. Comparison of this study with the previously work down using same type of substrate, as illustrated in the Table 3.0, show that the use of an activated carbon to enhance the treatability performance of granular sludge is a workable option in the presence of methanol.

Fig. 9.0 illustrates the biogas yield during the course of study period; this data has been extracted at relative higher treatability performance of the reactors, corresponding to various HRTs.

The results indicate that the higher HRT favors the biogas yields, as the lower HRT promotes the wash out of sludge from the reactor, thereby reduces the biogas production potential of the reactor. As shown corresponding to HRT of 18-30hrs, the biogas yield in the reactor R-I and R-II was noticed to be 0.28m^3/kg-COD_{rem} and 0.18m^3/kg-COD_{rem}, whereas corresponding to lower HRT of less than 10hrs, the biogas yield was reduced down to 0.2m^3/kg-COD_{rem} and 0.15m^3/kg-COD_{rem} in the reactor R-I and R-II, respectively. Refer to Table 3.0; the biogas yield observed during this study seems to be comparatively better, while using the same type of substrate at same operating conditions. But throughout the study it was noticed the reactor R-I is relatively more efficient in terms of biogas production, which might be due to the reason of denser granular sludge particles in reactor R-II on account of the activated carbon particles, which effects the smooth mixing of substrate and the biomass, thus the biogas production in the reactor R-II was observed lower than that of reactor R-I. Though the biogas production during anaerobic degradation at STP (Standard Temperature and Pressure) is theoretical equal to 0.35L/g-COD_{rem} (Bhatt et al. 1996) that indicates the production of lower biogas during this study in both the reactors, i.e. R-I and R-II, which might be due

Table 3. Comparison of similar work down

Substrate	Operating conditions	Treatment efficiency	Biogas production	Reference
NSSC pulping effluent	2.75 kg/m^3-day	35% COD	0.17 m^3/kg-COD_{rem}	Arshad et al 2009
NSSC pulping effluent (with methanol)	2.10 kg/m^3-day	57% TOC 55% COD 31% Lignin	0.34 L-CH_4/g-COD_{rem}	Arshad et al 2010
NSSC pulping effluent (with methanol + activated carbon)	4.5 kg/m^3-day	69% TOC 60% Lignin	0.29 m^3/kg-COD_{rem}	This study

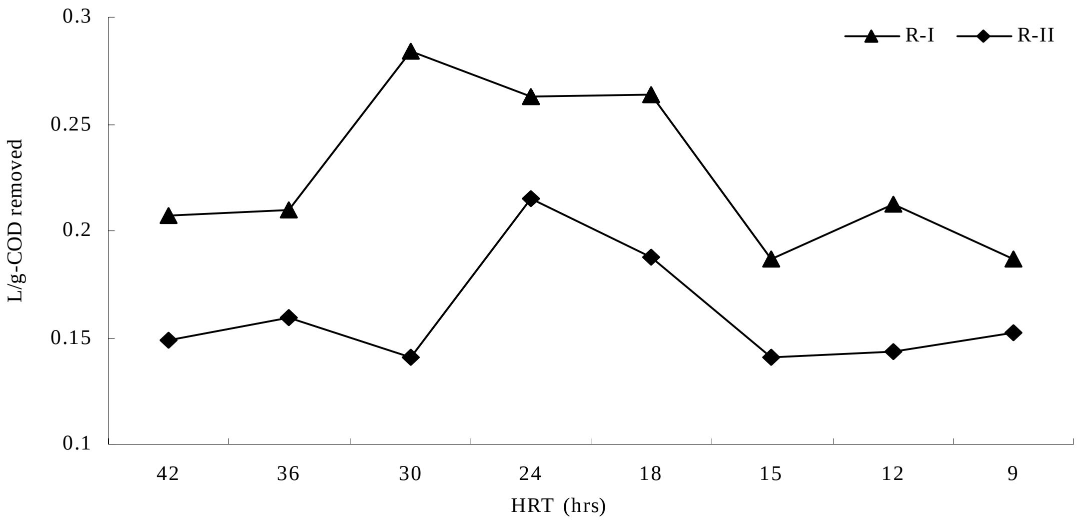


Fig. 9. Relationship b/w HRT and Biogas Production

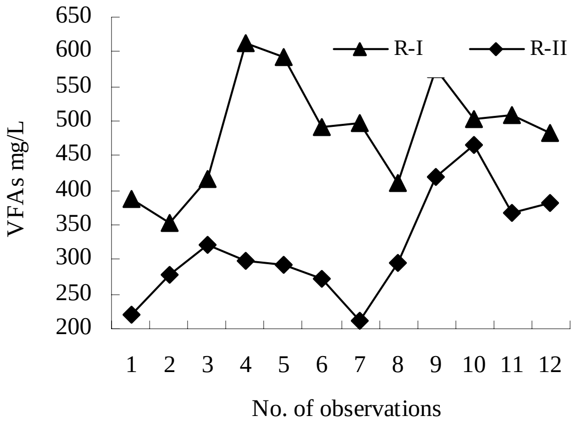


Fig. 10. Concentration of Effluent VFAs during the study period

to the reason of operating theses reactors at lower organic loading rates. Or it might be due to the present of some minor concentration of recalcitrant material in the substrate used, as previously identified in the previous studied (Sharama, 1992). The average methane composition observed in both the reactor during this study was about 56-58%, and the rest was noticed to be carbon dioxide.

As reported earlier, the concentration of effluent VFAs is inversely proportional to the treatability performance of the reactor (Mahadevaswamy, 2004) and the same reality has been confirmed during this study. The maximum concentration was observed as 610mg/L and 465mg/L in reactor R-I and R-II, respectively, corresponding to lower TOC removal of less than 56% in both the reactors. The effluent VFAs data obtained during optimum operating conditions, like HRT of 18hrs and OLR of 4.5kg/m^3-day, is illustrate in the Fig. 10. The inhibitory effect of VFAs on the anaerobic digestion also depends upon the buffering capacity within the reactor (Andrews 1969, Pohland 1969), as the average effluent VFAs concentration observed in the reactor R-I and R-II is 360mg/L and 230mg/L, respectively, indicates the low negative impact of VFAs on the treatability performance of the reactors, and shows effectives of using the external buffer solution used in this study.

CONCLUSION

From the results of this study, the following conclusions can be drawn:

1.The NSSC pulping effluent contains minor concentration of recalcitrant material that affects its treatability under anaerobic conditions.

2.The use of methanol and activated carbon in a UASB reactor enables it to improve its TOC and lignin removal efficiency by 19% and 15%, respectively.

3.At optimum operating conditions, the UASB reactor with methanol and activated carbon probably remove 69% TOC and 60% lignin. Whereas, the reactor without methanol and activated carbon can only remove 56% TOC and 51% lignin.

4.The biogas yield of reactor R-I is comparatively more than that of the reactor R-II, i.e., 0.28m^3/kg-COD_{rem} from R-I and 0.18m^3/kg-COD_{rem} from R-II, with 56-58% methane content in both the cases.

5.The average VFAs concentration observed in the reactor R-I and R-II is 360mg/L and 230mg/L, respectively.

The results of this study suggest that the use of methanol and activated carbon to enhance the treatability efficiency of the UASB reactor is highly feasible method for the treatment of NSSC pulping effluent. However, the exhaust period and regeneration methodology for the anaerobically used activated carbon requires further studies. Moreover, the bio-kinetic equation need to be developed, based on a long term studies of the same substrate.

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