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Biodegradation of Acetaldehyde wastewater in UASBR by mixed cultures of anaerobic bacteria

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Abstract
The wastewater generated from an acetaldehyde production unit consists of ethanol along with acetic acid and traces of acetaldehyde. These volatile organic compounds (VOCs) are hazardous pollutants and form acidic and toxic intermediates during the process of biodegradation. Present study reports commissioning of a UASB reactor to treat such toxic wastewater. The COD removal efficiency at laboratory and plant scale reactor were found to be higher than 75 %. The UASB reactor was able to withstand the fluctuations in VOCs loading in wastewater during plant start-ups and shutdown, without exhibiting any effect on treatment efficiency.

Keywords: Biodegradation, UASBR, acetaldehyde, ethanol, acetic acid, bioremediation

Introduction
Volatile organic compounds (VOCs) are hazardous pollutants emitted from paints, solvents, preservatives, automobile exhaust, industrial facilities, etc (Sano, et al., 2004; Jones, 1999; Wargocki et al., 2002; Wolkoff and Nielsen, 2001; Fanger, 2001; Racciatti et al., 2001). VOCs are recognized as causative agents of many conditions like sick building syndrome and cancer. In addition VOCs can contribute to ozone depletion and global warming.(Sano,et al., 2004).

Acetaldehyde a colourless, flammable liquid finding application in the manufacture of perfumes, flavors and as precursor for other industrial chemicals like acetic acid, ethyl acetate, Pentaerythritol, is produced by chemical oxidation of ethanol in presence of silver catalyst. The manufacturing process leads to generation of wastewater comprising of acetic acid, ethanol and traces of acetaldehyde. Acetaldehyde, acetic acid and ethanol are widely used VOCs which are toxic chemicals highly soluble in water (Lyman et al., 1982; Verschueren, 1983; Reimers et al., 2004) and therefore have severe impacts on aquatic environment and pose potential hazards to living beings (Nishijima, 2004). Therefore, remediation of VOCs from wastewaters is must before their discharge in to water bodies.

Ethanol during degradation forms toxic intermediates like acetaldehyde, acetic acid and ethyl acetate which lower down the pH inhibiting the treatment process (Devinny et al., 2012). Acetaldehyde on the other hand has negative impacts on aquatic environment mainly owing to its toxic nature (Morris, 1997; Xi et al., 2004; Guo & Ren., 2010). Only few studies in the literature address removal of acetaldehyde from contaminated water (Hebert, et al., 2010; Liviac, et al., 2010). Acetic acid is reported to be non-degradable compound (Park et al, 2007).The conventional techniques to remove VOCs is by adsorption on activated carbon, which however needs to be replaced and pose a problem of absorbent disposal (Sano et al., 2003). Many alternate methods are being investigated such as ozone oxidation method, catalytic combustion method, non thermal plasma method, etc (Futamura et al., 2002; Evans et al., 1993). These methods are either capital intensive or require heat source making them impractical for wastewaters having low concentration of VOCs.

Aerobic biodegradation of acetic acid in fluidized bed reactor has been reported for vinegar containing wastewater (Kumbha et al., 2014). Biological activated carbon filters were used for removal of VOCs in water (Bandosz, 2006; Arulneyam and Swaminathan, 2000; Chun et al., 2013). However, details regarding microorganisms employed in these studies are described by authors. (Nercessian et al., 2005; Wang et al., 2006).

Anaerobic digestion technology for organic industrial effluents was reported by Rajeshwari et al. (2000). Anaerobic treatment in UASB appears to be cost effective and environmentally sound approach. The main advantage is low installation and operation maintenance cost, high removal efficiencies and most important generation of bye-product methane which can be utilized as an energy source. The successful UASB start up for 10% acetic acid containing distillery effluent was reported (Wolmarans and de Villiers, 2001)

In this present study an Upflow Anaerobic Sludge blanket (UASB) reactor was commissioned to treat the VOCs containing wastewater generated from acetaldehyde production unit.

Materials and methods

Organism and Culture medium

Initial commissioning of UASB Reactor was carried out by using fresh cow dung 10 % mixed in fresh raw water. The mixed culture microorganisms surviving in UASBR were continuously acclimated to acetaldehyde wastewater as the carbon source with a nutrient solution consisting of (g/l) 0.01 $FeCl_3$; 0.8 K_2HPO_4; 0.05 $CaSO_4$ in deionized water plunged with Nitrogen gas to attain anaerobic conditions. Samples were drawn after 48 h and plated on Anaerobic Basal agar (HI MEDIA M1635) as well as on Anaerobic Agar (HIMEDIA M228) for identification of organisms in UASB reactor.

Analysis method

Acetaldehyde and acetic acid concentration in inlet and outlet wastewater were analyzed by method described by Indian Specification standard (IS 15356(2003) and IS 695(1986), respectively. The ethanol concentration was analyzed by high performance liquid chromatography (HPLC) using a Waters μBondapak C18 10μm 125 A Column (3.9 X 300mm); 100 % deionized water was used as mobile phase and refractometer as the detector. Biodegradation process indicators including chemical oxidation demand (COD), biomass, MLVSS, and pH in the inlet and outlet wastewater were analyzed by methods described in APHA (1976).

Experimental Equipment UASB Lab scale and plant scale

A schematic diagram of the UASB reactor system is illustrated in fig 1. For the reactor a rectangular plastic container of 5 L capacity was used. Feeding was done through bottom with help of feed pipe extended to bottom; overflow from the tank was collected from a point as shown in figure. A vent was supplied for escape of gas formed inside the reactor. On the same line UASB reactor of 200 M^3 capacity was designed to conduct experiments at plant scale, feeding was done from bottom of digester by help of open impeller pump, inside through a circular pipe with holes throughout for uniform distribution of feed. Overflow was allowed to flow in launder V notch at top of digester from where it was collected in a tank through pipe line. Four vents were provided for escape and collection of gases from headspace of the digester.

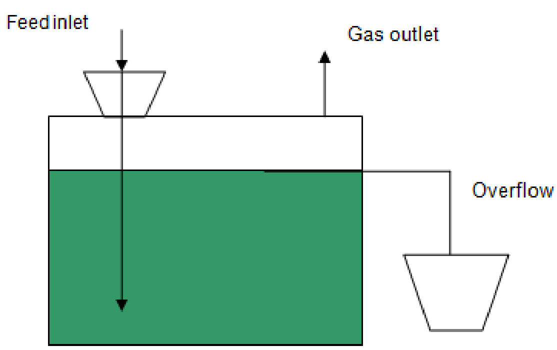

Figure 1: Schematic diagram of the laboratory scale UASB reactor showing inlet, outlet pipes along with point of venting for gases generated.

Commissioning and operation of UASB at lab and plant scale

Initial commissioning of the UASB was carried out using fresh cow dung in deionized water, solution of 20 % (w/v) cow dung was prepared and the UASB was loaded up to 20 % of total volume, the waste water emanating from acetaldehyde plant was neutralized to pH 6.5 to 6.8 with caustic solution and fed to the digester. Initially the feed rate was 3.5 g L^{-1} for 16 days, which was gradually increased to 6.9 g L^{-1} and subsequent increase in loading rate was done every 5th day until it reached to 55.0 g L^{-1} .The UASB reactor was found stable at this feed with a retention time of 19 d.

UASB reactor at plant scale was commissioned with 20 % (w/v) solution of cow dung in raw water up to 25 % of reactor volume; acetaldehyde wastewater was neutralized to pH 6.0 to 6.5 with caustic solution and fed in the reactor with initial COD loading rate of 20.0 kg per day. The loading rate was gradually increased up to 290.0 kg per day. Samples at inlet and outlet of UASB were collected regularly and parameters such as pH, Cell viability, COD were monitored.

Results

Profile of incoming wastewater from Acetaldehyde production unit

The analysis of wastewater emanating from acetaldehyde production plant, used in the present study is shown in Table 1 where acetic acid, ethanol, and acetaldehyde contribute mainly to COD of the effluent.

Table 1: Physicochemical analysis of wastewater of aldehyde production unit.

Waste water parameter	Range
pH	1.9 to 2.5
Acetic acid (ppm)	25000 to 75000
Ethanol (ppm)	900 to 1000
Acetaldehyde (ppm)	100 to 200
C.O.D (ppm)	25000 to 35000

*average values including values at plant stoppage and startups.

The acetic acid and ethanol concentration in the wastewater fluctuated mainly during plant start up and stoppage condition, the equipment washing led to higher values of COD in wastewater.

UASBR performance

Fig 2 shows process performance of UASB reactor at lab scale COD loading was successfully achieved up till 55 g L^{-1} in 50 days, the cells were viable and MLVSS was found to increase in the reactor from initial 5 % to 71 % and then was maintained at that level. High COD loading rates at 33rd day lead to reduction in cell viability but microorganisms rapidly acclimatized to the altered condition and viability was restored at an average of 70 % in the UASBR. Average reduction in COD was observed to be around 83 % and pH of the reactor was maintained at an average of about 7.65, while reactor temperature was maintained around 30 to 34 °C.

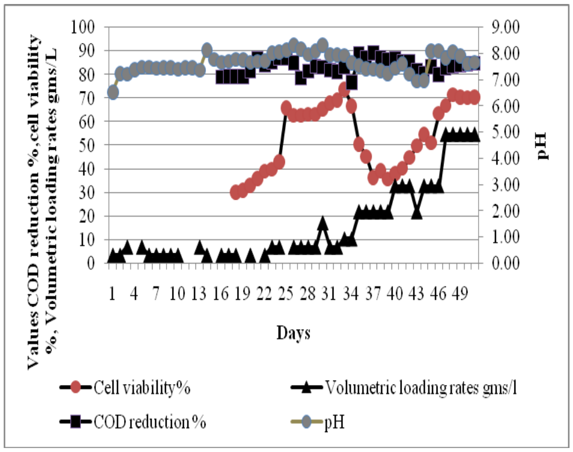

Figure 2: Profile UASB lab scale.

The results of UASB performance at plant scale is shown in fig. 3. The reactor performance was monitored for 15 months and an average COD reduction of 80 % was achieved with COD loading rate of 288 kg per day.

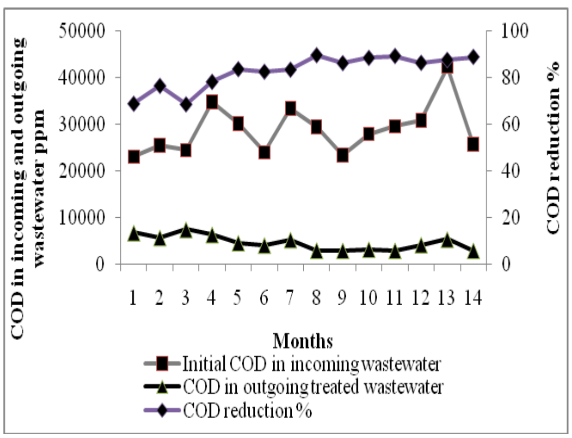

Figure 3: Performance of plant scale UASB reactor.

The analysis profile of incoming and outgoing effluent shows removal of acetic acid and ethanol in reactor, while aldehyde was detected in the samples which contributed to the COD in the treated effluent (table 2). Moreover it was observed that during plant start up and shutdown the higher content of ethanol and acetic acid was detected in the treated wastewater but the reactor performance became stable subsequently and was thus found to be stable to shock loadings.

Table 2: Inlet and treated wastewater quality from UASB reactor.

Parameter	Inlet	Outlet
pH	2.55	7.8
Acetic acid (ppm)	55000 to 75000	traces
Ethanol (ppm)	900 to 1000	traces
Acetaldehyde (ppm)	100 to 200	10 to 20
COD (ppm)	25000 to 35000	4000 to 5000

Plating of samples from UASB on anaerobic basal agar and anaerobic agar showed population of *Bacteroides sp.* and *Clostridium sp.* thriving luxuriantly on these plates (data not shown) indicating these groups of organisms are actively involved in removal of VOCs from acetaldehyde production wastewater.

Discussion

VOCs are hazardous pollutants emitted from production plants, scanty reports are available for biodegradation of wastewater from acetaldehyde production units, acetaldehyde in effluent sample are either adsorbed in carbon or degraded using Pt – loaded TiO_2 catalyst, 97% to 89 % degradation of acetaldehyde was reported when 100 ppm of acetaldehyde was introduced, however the degradation considerably decreased when acetaldehyde concentration was increased to 400 ppm (Sano et al., 2004). Biological activated carbon filters reduced the concentration of acetaldehyde in wastewaters from 0.8 ppm to 1.5 ppm to below 0.05 ppm but with considerable time and bacterial population which could utilize acetaldehyde or its metabolites as energy resource became prominent and other groups got inhibited (Chun-lei et al., 2013) suggesting acetaldehyde is toxic for other groups thriving on ethanol or acetic acid as carbon source. Bio filters for ethanol biodegradation have been reported by Arulneyam (2000) where 2000 ppm ethanol vapours were successfully degraded. Joseph et al. (2012) reported toxic and acidic intermediates like acetaldehyde, acetic acid and ethyl acetate generation on ethanol bio filters which inhibited the treatment process. Decomposition of 500 ppm acetic acid in wastewaters has been demonstrated by Park and Lee (2009) employing UV radiation on TIO_2-UV-H_2O_2 catalyst. Acetic acid present in distillery and wine effluents up to 7% (w/v) concentration has been successfully degraded in anaerobic fluidised bed reactor (Kumbha et al,. 2014). Also COD loading rate of 18 kg per day in effluent containing 10 % acetic acid was treated in UASB reactor without disturbance while plant start up and shut down (Wolmarans et al., 2001). On the similar lines an attempt was made to treat the wastewater of acetaldehyde plant which had acetic acid concentration of 7.5 % (w/v) along with ethanol and acetaldehyde; higher than 75 % reduction in COD was achieved with 19 days retention time, the reactor temperature was fairly constant between 34 and 36 °C. The reactor efficiently degraded acetic acid and ethanol while acetaldehyde was removed with 90 % efficiency.

Conclusion

UASB technology is well suited for the treatment of wastewaters containing VOCs particularly acetic acid, acetaldehyde and ethanol. Organic loading rates up to 290 Kg COD /d was applied to UASB reactor without decrease in performance of reactor.

Acknowledgement

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