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Bio-treatment of landfill leachate having low Carbon–Nitrogen ratio in a bio-film reactor packed with granular activated carbon under control of oxygen gas concentration

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ABSTRACT

Micro-organisms attached to granular activated carbon (GAC) in a fixed bed reactor were applied to treat synthetic leachate with different ratios of carbon to nitrogen. The leachate treatment reactor used in this study was run in batch operation under control of temperature, circulating flow rate, and oxygen concentration in pore space. The efficiency of a new device was estimated by treating artificial leachate having low levels of C/N ratios (0 to 5) at a temperature of 30°C, flow rate of 40 ml/min and oxygen gas concentration ranging from 1%, 5%, 10%, 15% to 21%. It was found that this device is adequate for the elimination of both dissolved organic carbon and ammoniacal-nitrogen. Even with the absence of external carbon supply, the denitrification process occurred. Partial bio-mass decay is considered the main source that supplies carbon to denitrifying bacteria. The average time needed for the elimination of dissolved organic compound (DOC) and NH_4^+ — N was 1 and 2 d, respectively.

Keywords: Leachate; Nitrogen; Oxygen; Activated carbon; Bio-film; C/N ratio

1. Introduction

Integrated waste management was initiated to carefully handle the different wastes. Incineration, composting, recycling, reuse and landfilling are the principal methods used for waste treatment. Sanitary landfill stands alone as the only waste disposal method that can deal with all materials in the solid waste streams. Other option such as biological or thermal treatments produces waste residues, and that subsequently need to be landfilled. Landfill sites are well known to be a long term source of pollution. In fact, long term emission controls as well as reclamation of completed landfill are the main problems in the field of sanitary landfill operation. The time required for leachate, landfill gas and landfilled waste to become environmentally stabilized and harmless was also estimated to fall within several decades to century.

Landfill leachate is a high strength wastewater characterized by high levels of organic constituents, ammonia and heavy metals. A typical young leachate may have a chemical oxygen demand (COD) 36 times higher than raw sewage, whereas a mature leachate may be equal in COD to raw sewage but containing much more recalcitrant organic constituents than domestic sewage [1].

The aftercare of landfill leachate is increasing the charge of landfill treatment. The current study is looking

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to create a compact unit to treat leachate, having moderate low Carbon–Nitrogen ratio, with a reduction of charge.

1.1. Method of carbon and nitrogen removal: general survey

An extensive review of landfill techniques and leachate treatment was well documented [2]. Leachate quality can be used as an indicator of stabilization within a landfill [2]. This will help for the assessment of the leachate management and its long-term cost. The causes of decreasing Carbon–Nitrogen ratio of landfill leachate with time have been studied [3,4]. The organic carbon contained in old leachate is attributed to substances having high molecular weight and is less amenable to microbial degradation [5,6]. Physical-chemical treatment of stabilized landfill leachate, using mainly coagulation/precipitation and adsorption on powdered activated carbon in conjunction with air stripping for ammonia removal was performed [7].

Simultaneous carbon and nitrogen removal processes were investigated in new down flow hanging sponge cubes (DHS) bed reactor where sponge was used as an attachment site. The DHS was used mainly as a post treatment process of an up flow anaerobic sludge blanket (UASB). Anaerobic pre-treatment process receiving a municipal sewage have an average ratio of C/N = 4.5. The DHS process performed 71% of carbon biodegradation and 78% of ammonia conversion to nitrate. Accumulated nitrate reached 30 mg/l accompanied with a decrease with the pH value [8]. Also, the simultaneous organic carbon removal and nitrification from waste water was carried out by a single stage bio-film process with membrane oxygenation [9].

Full anaerobic treatment of old landfill leachate is proved to be ineffective because most of the polluting substances are non biodegradable [10]. There are some researches of nitrogen removal using biofilm reactor. In this regard, nitrogen loss in an aerobic bio-film was attributed to a combination of nitrification and aerobic denitrification with nitrite as electron acceptor at low dissolved oxygen concentrations [11]. The influence of dissolved oxygen on nitrification in a circulating bed reactor using a synthetic solution containing inorganic carbon and organic nitrogen have been investigated [12]. The study showed that either the ammonia or the oxygen concentration could be limiting factors for the nitrification rate.

Micro-organism attached activated carbon fluidized bed (MAACFB) was used to treat leachate from old landfill having an average C/N of 0.7 [12]. MAACFB process could remove 60 and 70% of refractory organic and nitrogen, respectively. In situ nitrogen removal using leachate re-circulation to provide separate aerobic and anaerobic zones for ammonia nitrogen transformations to nitrate and nitrogen was reported [13]. Three simulated optional stages of methanogenesis, nitrification and denitrification showed that nitrogen conversion and removal depend on the operational stage and 95% nitrogen conversion to nitrate was achieved by this method, while the usual combined reactor operation with single pass leaching yielded 30 to 52% nitrification and 16–25% denitrification per cycle [13]. Hence, the attenuation of leachate ammonia nitrogen concentration to acceptable discharge levels could be achieved. The suspended carrier biofilm process (SCBP) or moving bed process is an attached growth system on plastic carrier elements which are kept in suspension and movement. It has been reported that such reactor exhibits a high rate of leachate nitrification at low temperatures (5°C). It was reported that the use of such reactor could cause the highest nitrification, and denitrification rate resulting in almost complete removal of the inorganic nitrogen (90%) [14].

Biological processes under aerobic, anaerobic and/or combined conditions are considered the most cost-effective methods to treat landfill leachate [15,16]. It is recognized that leachate with high BOD/COD ratio are easily bio treated [17].

Several researches have proved that use of combined anaerobic and aerobic systems for the removal of COD and ammonia from landfill leachate is quite efficient [18–20]. In fact, both reductive and oxidative biotransformations might occur concomitantly to complete mineralization of highly substituted compounds under micro-aeration [18]. Under oxygen-limited conditions, simultaneous aerobic and anaerobic (SAA) occur as a result of dissolved oxygen concentration gradients arising from diffusion limitation [21]. The activated sludge process are widely used for carbon and nitrogen removal from municipal and industrial wastewater, while it remains limited for treating landfill leachates. Landfill leachates are characterized by high levels of ammonia and require high hydraulic residence times. The effectiveness of biological nitrogen removal for treating a mature municipal landfill leachate, reached low ammonia removal efficiency (i.e., 20%) even given a long hydraulic residence time (20 d) [22]. Treatment of landfill leachate by attached-growth biomass systems was used and many experiences were reported by several researchers. Due to problems of sludge bulking or inadequate separability in conventional aerobic systems, a number of innovative aerobic processes, called attached-growth biomass systems, using bio film, have been developed [23]. These systems present the advantage of low loss of active biomass. Also nitrification is less affected by low temperatures [24] than in suspended-growth systems. Trickling filters are also among techniques that consist of attached biomass. It has been investigated for the biological nitrogen lowering from municipal landfill leachate. Trickling filters remain an interesting and attractive option for nitrification due to low-cost filter media [25]. Above 90% nitrification of leachate was achieved in laboratory and on-site pilot aerobic crushed brick filters with loading rates between 100 and 130 mgNH_4^+—N l^{-1} d^{-1} at 25°C and 50 mgNH_4^+—N L^{-1} d^{-1} even at temperatures as low as 5–10°C, respectively [25]. In the last decade, maximum ammonia rejections in a trickling filter were of 97 and 75%, respectively [26,27].

Previous experiments using pure culture treating synthetic wastewater offer reproducible results [28,29], they may not reflect outcomes at more realistic treatment scales. Under realistic operating conditions, a synergistic and antagonistic [30] effects depending of the wastewater type, the environmental operating conditions and the age composition of the exposed microbial population.

The above mentioned methods for nitrogen removal showed good performance, but it required external carbon sources. Therefore, a new device is proposed in this study. The concept of our new design will have four prominent advantages:

i. it does not require external carbon sources such as methanol, during the nitrification-denitrification reaction,
ii. excess sludge neither formed nor withdrawal is needed,
iii. the new prototype can be placed within the boundary of old landfill site as automated and self system for the local treatment of leachate, and
iv. It is believed to be a cost-benefit method for treatment of long-term leachate.

In the device pre-treatment as well as post-treatment of leachate is totally avoided. Thus, in the present work, a new device consists of biomass attached to granular activated carbon bed with leachate circulating with control of aeration. Optimal operational conditions such as flow rate, oxygen gas concentration and C/N ratio were determined.

2. Materials and methods

2.1. Materials and dimensions of reactors

The bed-reactor system used in this study is shown in Fig. 1. Both reactors made of acrylic, have identical dimensions (height: 60 cm, diameter: 7 cm, total volume: 2 l and effective bed volume: 1 l). Granular activated carbon (500 g) having an average diameter of 4 mm was soaked into deionised distilled water. Small balls made of plastic mesh bag were formed to contain the granule

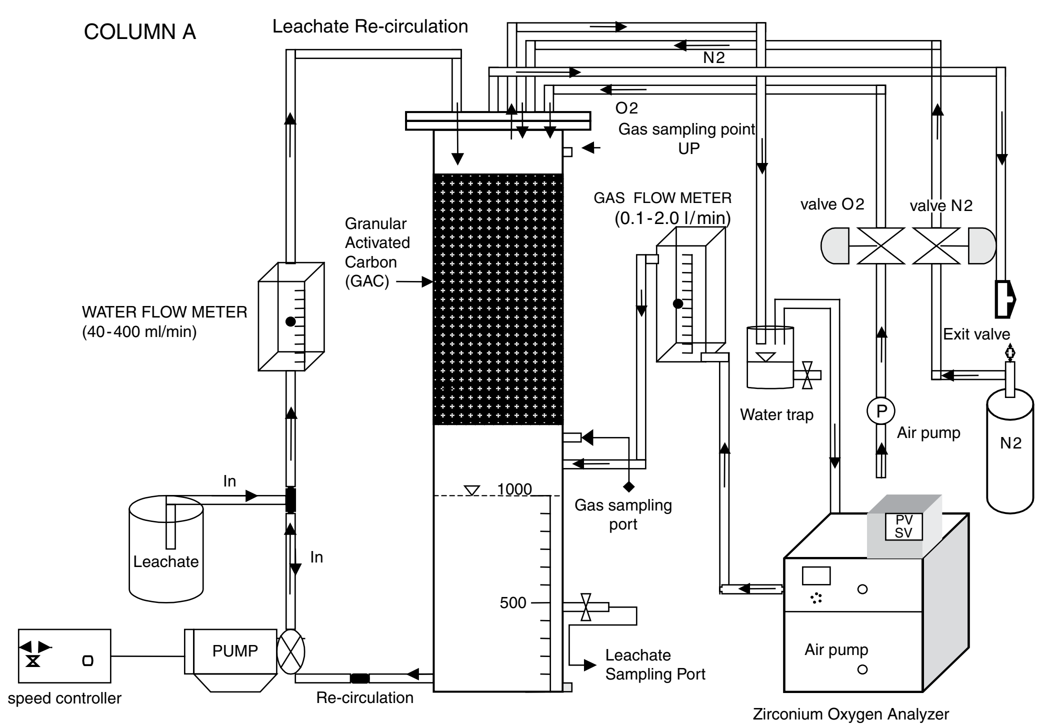

Fig. 1. Schematic configuration of fixed bio-filter with control of oxygen gas concentration.

of activated carbon. The balls were then packed into reactor, and micro-organisms were attached on the GAC.

Leachate was re-circulated with a constant flow rate. In column A, oxygen was controlled with a zirconium oxygen analyzer. Column B was open only from the top during the experiment without oxygen controller. Both column reactors A and B were operated under the same conditions, such as mass of granular activated carbon, inoculation procedure, controlled temperature and flow rate. The initial concentration of leachate was also identical. For homogeneous gas concentration the gas in reactor was circulated continuously at a rate of 1 l/min.

2.2. Synthetic leachate

Artificial leachate made of glucose, ammonium chloride, and peptone was used. The composition of the artificial leachate is given in Table 1. Artificial leachate was merely chosen to facilitate the laboratory test because it is easy to make and we ensure whatever desired replicate without any change of its composition. We used artificial leachate to determine the optimal conditions necessary for the treatment of leachate having different C/N ratios.

2.3. Experimental procedure

Reactor was inoculated with 500 ml of digested sewage sludge with rich bio-mass of high removal ability of organic carbon and nitrogen. The process was operated as batch reactor at 30°C. At designated concentration, the mixture of glucose, peptone and ammonium chloride, etc. was charged.

The concentrations of dissolved organic carbon (DOC), NH_4—N, NO_2—N, and NO_3-N were analyzed at predetermined time intervals. The experiment ended when DOC and NH_4—N decreased sufficiently. After seeding phase, effects of flow rate, oxygen gas concentration and C/N ratio were investigated for the treatment of artificial leachate.

2.4. Analytical methods

Dissolved Organic Carbon (DOC), NH_4—N, NO_2—N, NO_3—N, gas composition, pH, temperature were determined according to the Japanese Standard Methods [31]. All parameters determined for filtrated samples. Dissolved constituents were defined as those that passed through a 0.45 μm membrane filter. DOC was determined by Shimadzu TOC-500 analyzer. NH_4—N, NO_2—N, and NO_3—N were determined by ion exchange chromatography. The pH was determined by a pH meter. Temperature was read directly from thermometer attached to each column. The process values of oxygen were read directly from the screen of oxygen analyzer (GE Panametrics CGA 351 Oxygen Analyzer, US) and also measured by a gas chromatography (Gas chromatography with a thermal conductivity detector (Hitachi Type 164, column type: WG-100, flow rate of He: 33 ml/min; detector temperature: 50°C).

3. Results and discussions

3.1. Effect of flow rate

Relationship between flow rate, carbon and nitrogen removal were investigated. In this experiment C/N, and oxygen gas concentrations were kept constant to 5 and 15%, respectively. Setting values of flow rate were varied at 60, 100, 140 ml/min. The total volume of loaded

Table 1
Example of artificial leachate composition when C/N is 5, TOC = 1000 mg/l and NH_4—N = 200 mg/l

Substrate use in the composition of synthetic leachate	Substrate name	Weight of chemical (g)	Volume of distilled water (l)
	Glucose	1.25	0.5
	Peptone	*	
	NH_4Cl	0.38	
A**	K_2HPO_4	21.75	1
	KH_2PO_4	8.50	
	$Na_2HP \cdot 12H_2O$	44.60	
	NH_4CL	1.70	
B**	$MgSO_4 \cdot 7H_2O$	22.50	1
C**	$CaCl_2$	27.50	1
D**	$FeCl_3 \cdot 6H_2O$	0.25	1

* Peptone was added only to increase the biomass population when biomass activity is decreased.
** 1 ml of each solution of A, B, C and D was added to 0.5 l of leachate.

leachate was one litre. The summarized influences of flow rate on carbon and nitrogen removal are shown in Figs. 2 and 3, respectively. The results show that by increasing the flow rate, an increase of the removal rates of DOC and NH_4^+—N were observed. However, when flow rate increased, a remarkable fraction of sludge was observed on the bottom of the reactor. To avoid such conditions, the water flow rate was kept to minimum (40 ml/min). Even, at flow rate of 60 ml/min the percentage of carbon and nitrogen elimination could reach 95% and 90% within 2 d, respectively.

3.2. Effect of oxygen gas concentration

Oxygen plays a very important role in the aerobic degradation of both carbon and nitrogen. A complete aerobic degradation can degrade all the organic carbon. Yet, due to the formation of nitrate and nitrite from ammonium, if denitrification does not take place, the pH decrease (3 to 4) which impacts the life of bio-mass as well as the removal efficiency. Thus, oxygen gas concentration in pores of GAC bed becomes a main target to optimize the carbon and nitrogen removals without harming the biomass efficiency. Accordingly, we varied the levels of oxygen gas concentration for column A while column B was kept open from its top part. The oxygen gas concentrations for column A were set at 15, 10, 5 and 1% in decreasing order. C/N was fixed at 5 and temperature was fixed at 30°C. The summarized effect of decreasing and increasing O_2 gas concentration on DOC and NH_4^+—N removal rate are shown in Fig. 4. Fig. 4 shows that the rate of DOC removal increases with O_2 gas concentration. However, the effect on NH_4-N removal is not obvious. However, it seems that O_2 gas concentration of 5% to 10% is the best for simultaneous nitrification and denitrification. The curves of degradation of dissolved organic carbon (DOC) are much sharply than those of ammonia-nitrogen. Owing to competition between micro-organisms species, the oxidation of carbon occurred prior to the bio-oxidation of nitrogen. In addition, it is generally believed that nitrifiers are slow growing organisms in bio-film [5]. We observed that during the bio-oxidation of carbon, the pH decrease due to the formation of volatile fatty acids, e.g., carboxylic acid. Usually, the presence of nitrate in

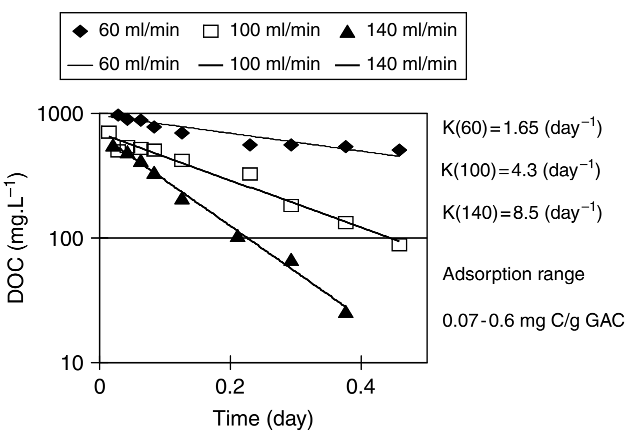

Fig. 2. Effect of flow rate on the removal of DOC in column A where *K* is reaction rate constant (t^{-1}), adsorption is given in mg/g solid.

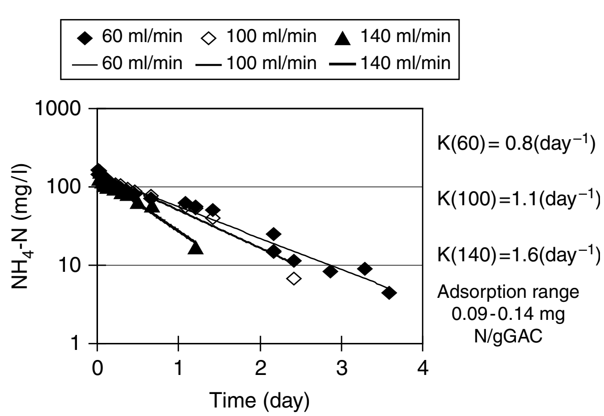

Fig. 3. Effect of flow rate on the removal of NH_4—N in column A where *K* is reaction rate constant (t^{-1}), adsorption is given in mg/g solid.

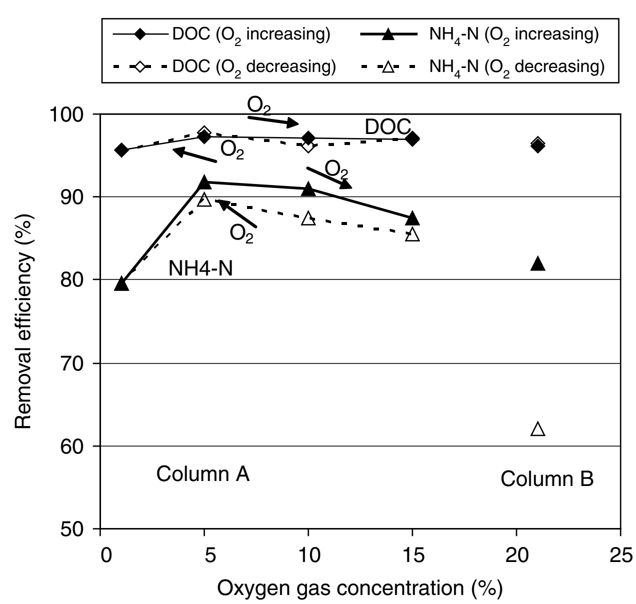

Fig. 4. Effect of oxygen gas concentration removal of DOC and NH_4—N.

leachate decreases also the pH. Before the production of nitrate pH of leachate decreased to 5 or below and this required increasing the pH of leachate. The pH of leachate were firstly adjusted by the addition of NaOH (5% w/v) and due to the close positions of detection peaks of sodium and ammonium in liquid chromatography, sodium hydroxide was substituted by potassium hydroxide.

3.3. Effect of C/N ratio

Leachate from old landfills usually contains the high concentration of ammonia, compared with carbon which is easily transformed to gas (CO_2, CH_4). Hence, leachate from completed and aged landfills exhibits a significantly low ratio of carbon to nitrogen. It was also reported that the ratio of BOD to COD usually bellow 0.1 which indicates a low biodegradability of organic carbon [32].

In this study, we focused to improve the performance of biological treatment process applied for the treatment of artificial leachate to remove organic carbon and ammonium nitrogen. From the section 3.2, the oxygen gas concentration level was set to 10%. The chosen values of C/N for this experimental runs were set to 5, 1, 0.5, 0.3, 0.1 and 0 in decreasing manner. The summarized effects of C/N ratio on DOC and NH_4—N removal are shown in Fig. 5. The relationship between C/N ratio and removal of total nitrogen at nearly steady state are shown in Fig. 5. The rate of NH_4—N removal decreases with decreasing C/N ratio. When C/N was 5, most of NH_4—N and total nitrogen were removed. Yet, the removal of both NH_4—N and total nitrogen remained high until C/N was 0.5. Compared to column A, column B is more efficient.

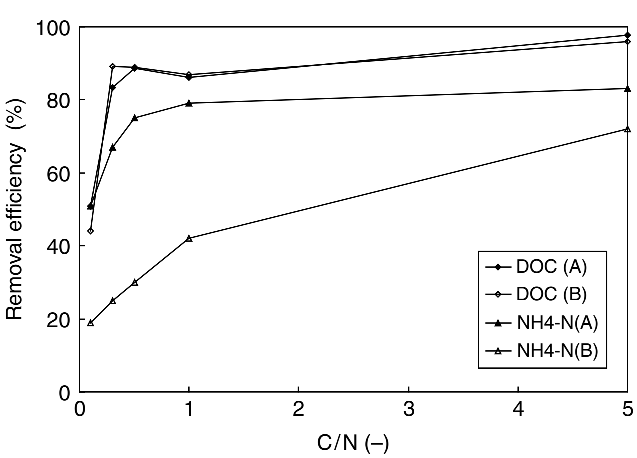

Fig. 5. Effect of C/N ratio on the efficiency of new device.

4. Conclusions

A new type of bio-film reactor with control of oxygen gas concentration in gas space is proposed to treat leachate having low carbon-nitrogen ratio. The optimal operating conditions when using artificial leachate were:

i. Control of oxygen between 5–10% in the reactor is effective to control biomass autolysis,
ii. Without external carbon sources, high nitrogen removal is achieved for artificial leachate of low C/N ratio 1,
iii. Within 1 to 2 d, 200 mg/l of ammonia nitrogen decreased to (20 mg/l) and nitrate nitrogen remained below 10 mg/l.

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