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TREATMENT OF LANDFILL LEACHATE BY MEMBRANE BIOREACTOR AND ELECTRO FENTON PROCESS

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ABSTRACT

Land filling is one of the least expensive methods for disposal of municipal solid waste (MSW). Hence about 90% of MSW is disposed in the open dumps and landfill unscientifically, creating problem to public health and the environment. The leachate generated from the municipal landfill contains organic and inorganic pollutants, which make it unsuitable for discharge in natural bodies without any prior treatment. Advanced oxidation process (AOPs) are promising methods to treat effectively the recalcitrant substances present in landfill leachate. Among APOs fenton process is considered the most promising treatment for remediation of highly contaminated water. In this study electro fenton pretreated landfill leachate was treated by membrane bioreactor process and then post treated by electro fenton process. The pretreated values of parameters were taken from previous studies which has BOD/COD ratio 0.39. Membrane bioreactors have proven quite effective in removing organic and inorganic contaminants as well as biological entities from waste water. In MBR the membrane arrangement consist of hollow fiber membrane module having pore size of 0.1µm. After MBR process, the effluent quality did not meet the general Standards for discharge of environmental pollutants. But after post treatment all the parameters have met the relevant Indian standards for discharge as irrigation water and all the parameter except BOD and sulphide have met the relevant Indian standards for discharge in to inland surface water.

KEYWORDS: Municipal solid waste; Membrane Bioreactor process; Electro fenton process; Response Surface Methodology

INTRODUCTION

Sanitary Landfill is considered to be the most common way of disposing urban solid waste. An important problem associated with sanitary landfills is the production of leachate. Leachate is generally formed when rain water percolates through dumped waste and takes up the organic and inorganic products from both physical extraction and hydrolytic and fermentation process. Generally, leachate contains high concentrations of soluble organic matter and inorganic ions. Due to its high strength nature, direct discharge of leachate in the environment is not recommended.

Landfill leachate is the liquid produced by natural humidity and water present in the residue of organic matter, the result of the biological degradation of organic matter present and by water infiltration in the covering and inner layers of landfill cells, supplementing dissolved or suspended material originating from the residue mass. The chemical and microbiological composition of landfill leachate is complex and variable, since apart from being depended upon features of residual deposit, it is influenced by environmental conditions, the operational manner of the landfill and by the dynamics of the decomposition process that occurs inside the cells. Landfill leachate is generally a dark colored liquid, with a strong smell, which carries a high organic load. One of its characteristic features is an aqueous solution in which four group of pollutant are present: dissolved organic matter (volatile fatty acid and more refractory organic matter such as humic substance), macro inorganic compounds, heavy metals and xenobiotic organic compounds originating from chemical and domestic residue present at low concentrations (aromatic hydrocarbons, phenols, pesticides, etc.) [1].

Landfill effluents (leachate) need to be treated on site to meet the standards for its discharge into the server or its direct disposal into surface water. In the world the problem of leachate treatment has been existed for some time now, but a universal solution has not been found. Under emerging cleaner production technologies treatment methods for leachate and sludge need to be developed. Application of advanced oxidation methods are considered to enhance existing treatment technologies. Advanced oxidation is the use of radicals especially hydroxide radical to enhance oxidation in a treatment process. Hydroxide radical is one of the most powerful radicals used to treat water and wastewater. The solid liquid separation that is conventionally carried out in gravity based clarifier is replaced by membrane filtration in MBR systems thus combining the strength of biological treatment process and efficiency of membrane filtration. This and several other advantages have made the MBR system ideally suited for treatment of strong industrial wastewater and reclamation of water.

Advanced Oxidation Process

Advanced oxidation process are frequently used to oxidize complex organic constituents found in wastewater, which are difficult to be degraded biologically in to simpler end products. In this process, free hydroxyl radical is used as a strong oxidant to destroy the complex organic compounds. AOP process has become an alternative efficient process for mineralization of recalcitrant organics in landfill leachate. The main purpose of AOP is to enhance chemical oxidation efficiency by generating more hydroxyl radicals. This process includes non-photochemical methods generating hydroxyl radicals without light energy: ozonation (O_3) at elevated pH > 8.5; O_3/H_2O_2; O_3/catalyst; Fenton process (H_2O_2/Fe^{2+}), and photochemical methods: O_3/UV; H_2O_2/UV; O_3/H_2O_2/UV; photo-Fenton; photo catalysis (UV/TiO_2). Advanced Oxidation Processes (AOP) has been proposed in recent years as an effective alternative for mineralization of recalcitrant organics in landfill leachate. Among these processes, Fenton process seems to be the best compromise because the process is technologically simple, there is no mass transfer limitation and both iron and hydrogen peroxide are cheap and non toxic [2].

Fenton oxidation has been extensively studied in recent years and analyses indicate fenton process to be one of the most cost effective alternatives for this application. In fen ton process, aqueous hydrogen peroxide and Fe^{2+} (ferrous ions) in acidic conditions generate hydroxyl radicals that are strongly oxidative.

Electro fenton process is the use of electrically assisted fenton reactions. It includes electro chemical reactions for the in situ generation of the reagents used for the fenton reaction. The generated reagents depend on solution conditions, cell potential and nature of electrodes. Ferrous ions may be produced by oxidative dissolution of anodes such as iron metal or by reduction of ferric ions at an inert cathode such as platinum. Moreover H_2O_2 may be produced by dioxygen reduction at the cathode [3].

At anode:

$$Fe\ (s) \rightarrow Fe^{2+}\ (aq) + 2e^- \qquad (1)$$

$$Fe^{2+}\ (aq) + 2\ OH^-(aq) \rightarrow Fe\ (OH)_2 \qquad (2)$$

At cathode:

$$H_2O\ (l) + 2e^- \rightarrow H_2\ (g) + 2\ OH^-\ (aq) \qquad (3)$$

Overall:

$$Fe\ (s) + 2\ H_2O\ (l) \rightarrow Fe\ (OH)_2 + H_2\ (g) \qquad (4)$$

Membrane bioreactor process

Membrane bioreactor concept is a combination of conventional biological wastewater treatment and membrane filtration. The concept is technically similar to that of traditional wastewater treatment plant except for the separation of activated sludge and treated wastewater. In an MBR installation this separation is not done by sedimentation in a secondary clarification tank, but by membrane filtration. The entire biomass is confined within the system, providing both exact control of the residence time for the microorganisms in the reactor (solid retention time) and the disinfection of the effluent. A membrane is a material that one type of substance can pass more readily than others. It is manufactured in order to achieve the reasonable mechanical strength and can maintain a high throughput of a desired permeate with a high degree of selectivity. The optimum geometry or configuration may have these characteristics. MBR systems can be classified in to two major categories according to the location of the membrane component. The first category is normally referred to as the external membrane MBR configuration and the second one is submerged MBR configurations. In external membrane MBR, the membrane component is placed in a separate vessel, outside the bioreactor basin. In this system the mixed liquor from the reactor is pumped in to the external membrane module. In submerged type, membrane component is immersed inside the bioreactor basin. Here the effluent is sucked out of the membrane module with the help of a permeate pump and the suspended solids fall back in to basin [4].

MATERIALS AND METHODS

Preparation of synthetic wastewater

Synthetic wastewater was prepared based on the physicochemical parameters obtained for the pretreated (electro fenton) leachate sample from previous studies. The composition of synthetic sample was pretreated by trial and error method so that reasonable match with the pretreated sample could be attained. Pretreated sample was treated by MBR process. After that a post treatment was done to remove the pollutants from the synthetic sample. The characteristics of synthetic sample are given in Table 2.

Table 1. Composition of synthetic sample

Chemicals required	Quantity in g/l
Ammonium chloride	0.8
Sodium sulphide	0.3
Sodium chloride	0.8
Calcium carbonate	0.095
Dipotassium hydrogen ortho phosphate	0.2
Ferrous sulphate	0.06
Magnesium sulphate	0.06
D- Glucose	7

Table 2. Characteristics of synthetic sample

Parameter	Unit	Concentration of parameters of pretreated synthetic wastewater
BOD	mg/l	1552.5
COD	mg/l	4032
Ammonium nitrogen	mg/l	1136
Phosphate	mg/l	25.5
Sulphate	mg/l	1155
Sulphide	mg/l	32.2
Chloride	mg/l	2881
BOD/COD	-	0.39

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Experimental Set up of MBR

An acrylic container of 12L capacity was used as the bioreactor. The reactor size was 20cm x 15cm x 40cm. the reactor was supplied with oxygen by aerator. Aeration rate of 5litre/min was provided. The membrane arrangement consist of hollow fiber membrane module having pore size of 0.1 µm. pump of capacity 0.7Lpm and DC adapter connected in series. This was provided for enhancing treatment efficiency.

Bio sludge collected from a secondary sedimentation tank was used as inoculums for the bioreactor system after acclimatizing with leachate for 1 month. The external membrane bioreactor consists of bioreactor and membrane filtration. The reactor was operated in batch mode which was filled with 1L sludge and 3L synthetic wastewater in order to maintain the MLSS concentration in the range 20g/l. after the reaction time, mixed liquor was allowed to settle for 30 minutes. The supernatant from the bioreactor was passed through the hollow fiber membrane module using the pump. The treated effluent was collected and analyzed. This process was repeated for different HRTs and pH maintained as neutral. The optimum HRT was obtained by analyzing the COD removal obtained in each day. The experimental set up of MBR is shown in Fig. 1

Fig. 1. Experimental Set up of MBR

Experimental Set up of Electro Fenton process

1000 ml borosil glass beaker was used for the electro fenton process. 800 ml of MBR treated effluent was used for the batch studies. Experiments were done at room temperature. Cast iron plates (12cm x 6.5cm x 0.1cm) with maximum surface area that will be fit to the 1L borosil glass beaker were used as electrodes. The electrodes were vertically positioned in the beaker. The distance between two electrodes was fixed as 2.5cm. The cast iron electrodes acted as the source of Fe^{2+} ions. A direct power supply was used to provide the desired current. The electric circuit consists of a regulated DC power supply connected to the reactor. The anode and cathode were connected to the positive and negative outlets of a DC power supply. The reaction mixture was continuously stirred by magnetic stirrer. The various parameters affecting the treatment are H_2O_2 concentration, current density and reaction time. pH of the sample was kept as 2.5 according to previous studies. The optimum condition for EF process was found by RSM using Minitab Software version 6.1. After the required reaction time the treated sample was allowed to settle for 2 hours and the supernatant was used for analyzing the parameters

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Fig. 2. Experimental Set up of Electro Fenton process

RESULTS AND DISCUSSION

Treatment of synthetic wastewater by MBR process

The synthetic wastewater was subjected to activated sludge process al neutral pH in the bioreactor. The optimum HRT was obtained by analyzing the COD removal efficiency in each day. COD of the effluent on each day is shown in Fig. 3.

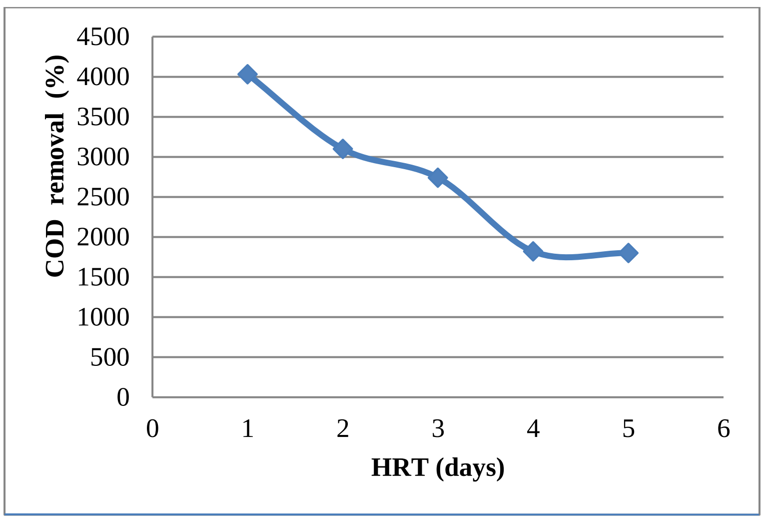

Fig. 3. COD removal at different HRTs

From the figure it can be seen that COD reduction reached a steady state on the 4th day. So the optimum HRT obtained as 4 days. So the synthetic sample was treated by MBR with an optimum HRT of 4 days. The removal of various parameters by MBR process was studied. Percentage removal of parameters after MBR process is given in Table 3.

Table 3. Percentage removal of pollutants after MBR

Parameter	Unit	Conc. of parameter before MBR	Conc. of parameter after MBR	% Removal
BOD	mg/l	1552.5	326	79
COD	mg/l	4032	1008	75
Ammonium nitrogen	mg/l	1136	295.4	74

Phosphate	mg/l	25.5	BDL	100
Sulphate	mg/l	1155	473	59
Sulphide	mg/l	32.2	10.8	66
Chloride	mg/l	2881	1267.6	56

The membrane can capture most of the suspended solids in the reactor because of its fine pore size. Therefore, non-biodegradable organic compounds are removed through filtration of particulates and discharge sludge. Aeration is also one of the main factors that affect the biochemical process of BOD and COD removals.

Treatment of effluent by Electro Fenton process
The effluent from MBR process was treated by EF process at optimum conditions. pH of the sample was kept as 2.5. H_2O_2 dosage, current density and reaction time were selected as factors in the Box- Behnken statistical design. The optimum condition obtained from the Minitab is H_2O_2 dosage = 60%, current density = 87.3 A/m^2 and reaction time = 38 min.

Table 4. Evaluation of the performance of EF process (post treatment) in synthetic wastewater

Parameter	Unit	Conc. of parameters before EF	Conc. of parameters after EF	% Removal
BOD	mg/l	326	84.8	74
COD	mg/l	1008	231.8	77
Ammonium nitrogen	mg/l	295.4	35.4	88
Sulphate	mg/l	473	165.6	65
Sulphide	mg/l	10.8	3.6	66
Chloride	mg/l	215.5	215.5	83

EF has very good efficiency during initial stage of electrolysis when easily oxidizable products were rapidly destroyed by hydroxyl radicals. Table 5 summarizes the result of all treatment done using synthetic wastewater. The table shows the performance of membrane bioreactor and electro fenton process based on relevant Indian Standards.

These standards are inserted by the Govt. of India by Rule 2(d) of the Environment (Protection) Second Amendment Rules, 1993 notified vide G.S.R. 422(E) dated 19.05.1993, published in the Gazette No. 174 dated 19.05.1993.

Table 5. Evaluation of performance of MBR followed by EF process in synthetic wastewater based on relevant Indian Standards.

Parameter	**Final effluent characteristics (mg/l)**	**General standards for discharge of environmental pollutants Part A: Effluents [Schedule VI] as per The Environment (Protection) Rules, 1986, Govt. of India**	
		Into surface water	**To land for irrigation**
BOD	84.8	<30	<100
COD	231.8	<250	-----
Ammonia Nitrogen	35.4	<50	-----
Phosphate	BDL	<5	-----
Sulphate	165.6	<1000	<1000
Sulphide	3.6	<2	-----
Chloride	215.5	<1000	<600

The table shows that final treated effluents have not met all the relevant Indian Standards. The value of sulphide is not within the limit and also the BOD has not met the standards of discharge in to surface water. So the effluent could not be discharged in to surface water. But the effluent after both the treatment has met relevant Indian Standards for discharge as irrigation water. So the effluent can be discharged as irrigation water.

CONCLUSION

In this study pretreated landfill leachate was treated by MBR and Electro Fenton process in order to meet the effluent discharge standards. The sample was treated by MBR with an optimum HRT of 4 days and at neutral pH in batch mode. In case of EF process, the optimization of various parameters was done by RSM using Minitab software, version 6.1. the optimum condition obtained from the Minitab were H_2O_2 dosage = 60%, current density = 87.3 A/m^2 and reaction time = 38 min. This optimum condition was used for further study. After MBR process the percentage removal obtained for BOD, COD, Ammonium nitrogen, Phosphate, Sulphate, Sulphide and Chloride were 79, 75, 74, 100, 59, 66 and 56% respectively. After EF followed by MBR process the percentage removal obtained for BOD, COD, Ammonium nitrogen, Phosphate, Sulphate, Sulphide and Chloride were 74, 77, 88, 65, 66 and 83% respectively. From the result it can be seen that percentage removal of pollutants increased after post treatment. But final treated effluent has not met all the relevant Indian Standards. All the parameters except BOD and Sulphide met the relevant Indian Standards for discharge in to surface water. But for discharging as irrigation water all the parameters have met the relevant Indian Standards. So the final treated effluent can be used as irrigation water.

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