DEVELOPMENT AND COMPARATIVE STUDY OF A SEMI-FLUIDIZED BED BIOREACTOR FOR TREATMENT OF WASTEWATER FROM PROCESS INDUSTRIES

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ABSTRACT

The harmful effects of various pollutants from chemical process industries with there sources of occurrence have been highlighted. A comparative analysis has been critically made to examine the suitability if immobilized culture semi-fluidized bed bioreactor with comparison to other conventional reactors. In this paper an attempt has been made to develop a semi-fluidized bed bioreactor for performance analysis. Among the immobilized cell bioreactors, no doubt the semi-fluidized bed bioreactor is a novel and efficient one, which can be adopted for the treatment of industrial wastewater containing phenolic compounds and other pollutants even at lower concentration. Experimental investigations will continue to characterize and evaluation of its performance for phenolic wastewater treatment. A proper choice of immobilized culture, careful consideration of various design parameters of semi-fluidized bed bioreactors will make treatment process cost effective in the long run.

Keywords: Pollution Control; Water Pollution; Wastewater; Semi-fluidized bed; Hazardous Wastes; Environmental Pollution; Waste Management

Introduction

Environmental pollution is an emerging threat and of great concern in today's context pertaining to its effect on the ecosystem. Water pollution is one of the greatest concerns now a day. In recent years, considerable attention has been paid to industrial wastes discharged to land and surface water. Industrial effluents often contain various toxic metals, harmful dissolved gases, and several organic and inorganic compounds. These may accumulate in soil in excessive quantities in long-term use, ultimately physiologically adverse effects on crop productivity.

The worldwide rise in population and the industrialization during the last few decades have resulted in ecological unbalance and degradation of the natural resources. One of the most essential natural resources, which have been the worst victim of population explosion and growing industrialization, is water.

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Huge quantity of wastewater generated from human settlement and industrial sectors accompany the disposal system either as municipal wastewater or industrial wastewater. This wastewater is enriched with varied pollutants and harmful both for human being and the aquatic flora and fauna, finds it way out into the nearly flowing or stationary water bodies and thus make natural sources of water seriously contaminated. The presence of some harmful pollutants in wastewater deteriorates the water quality considerably and has damaging effect on both aquatic life and human health [1].

It has been estimated that over 5 million chemical substances produced by industries have been identified and about 12000 of these are marketed which amount to around half of the total production. Due to discharge of toxic effluents long-term consequence of exposure can cause cancer, delayed nervous damage, malformation in urban children, mutagenic changes, neurological disorders etc. Various acid manufacturing industries discharge acidic effluent, which not only make the land infertile but make the water of the river acidic also. The high acidity causes stomach diseases and skin ailments in human beings [1,2].

Alkaline effluents cause infertility of the soil and destroy the flora and fauna of the vicinity. Contaminated water by pesticides, such as DDT, aldrin, dieldrin, heptachlor etc is harm full for aquatic life and human beings as well. Discharge of cyanide-contained wastewater to water mass may lead to death of fish and other aquatic life therein. Use of water containing fluoride can causes mental disorders and stomach ailments and can also reduces agricultural production [1-3]. Characteristics of wastewater from few process industries are shown in table-1.

So far public health is concerned, wastewater from chemical process industries is almost totally responsible due to presence of toxic and harmful chemicals, like phenol, heavy metals (Pb As, Cd, Cr, Zn etc) and pesticides therein [1-4].

The widely used mercury in industrial applications is highly toxic in its ionic form due to its potential for remobilization even from low soluble forms through microbial generation of methyl mercury and its ability to accumulate in nutrition chains [5].

Nitrate-nitrogen is a common pollutant in water. The main source is over fertilization in agriculture, human and animal waste disposal, and effluent after nitrifying treatment in fertilizer plant, explosive manufacturing plants, NO_x absorption in air-washing devices and recovery of nuclear fuels and cause severe health effects. United States Environmental Protection Agency (US EPA) and World Health Organization (WHO), have set limit levels of 10 and 1 ppm for nitrate as nitrogen (NO_3^--N) and nitrite as nitrogen (NO_2^--N) respectively to regulate to regulate nitrate concentration in drinking water [6,7].

Purifying and recycling wastewater is imperative in view of reduced availability and deteriorating water quality. Phenol along with other xenobiotic compounds is one of the most common contaminants present in effluents from chemical process industries. Even at lower concentration these compounds adversely affect aquatic as well as human life [1-4,8-13]. Also these compounds form complexes with metal ions discharged from other industries, which are carcinogenic in nature. It is water soluble and highly mobile. This imparts medicinal taste and odor even at much lower concentration of 2 μg/l and it is lethal to fish at concentrations of 5-25 mg/l [10]. The maximum permitted concentration level of phenol being 0.5-1 mg/l for industrial wastewater and 1μg/l for drinking water [15,17]. So it is highly essential to save the water resources and aquatic life by removing these compounds from wastewater before disposal.

The main sources of phenolic wastewater are coal chemical plants, oil refineries, petrochemical industries, fibre glass units, explosive manufacture, phenol-based polymerization process, pharmaceuticals, plastic, paints and varnish producing units, textile units making use of organic dyes, anticeptics, antirust products, biocides, photographic chemicals and smelting and related metallurgical operations, etc [2,8-10, 17,20].

Table 1: Characteristics of wastewater from process industries

Parameter / Source & amount range, mg/l	From steel industry	From petroleum refinery	From LT coal carbonization
PH	8.5-9.5	-	9.0
Total solids	175-1300	-	6720
Dissolved solids	125-800	-	5312
Suspended solids	50-500	200-400	1408
Oils and grease	-	2000-3000	-
Chlorides as Cl	-		Nil
HS and mercaptans	-	10-220	-
Nitrogen	800-1400	-	-
Sulphates / sulfides	110-220	09	802
Cyanides	10-50	-	4576
Thiocyanates	50-100	-	2840
Phenol	500-1000	1500-2000	10240
Total alkalinity	-	-	14670
Phenolphthalein alkalinity	-	-	Nil
Turbidity	-	-	-
BOD	160	100-300	11100ppm
COD	790-2450	-	20400ppm

Treatment of wastewater

The conventional methods of treatment of phenolic and nitrate-nitrogen wastewater are largely physical and chemical processes but these processes led to secondary effluent problems due to formation of toxic materials such as cyanates, chlorinated phenols, hydrocarbons, etc. These methods are mainly chlorination, ozonation, solvent extraction, incineration, chemical oxidation, membrane process, coagulation, flocculation, adsorption, ion exchange, reverse osmosis, electrolysis, etc [2,8,9,19].

In solvent extraction there is a danger of contamination of treated water by the solvent. The solvents used for phenol recovery are benzene, isopropyl ethyl and butyl acetate. In addition to the presence of solvent in treated water, the high cost of solvent is another disadvantage. In adsorption commonly activated carbon is used which is disposed by incineration. The process of incineration generates many new compounds such as dioxins and furans have very severe consequences on human health. Chemical oxidation requires a reactor, which operates at high temperature and high pressure, ultimately huge energy [2,30].

Biological treatment is attractive due to the potential to almost degrade phenol and other pollutants while producing innocuous end products, reduced capital and operating costs, maintain phenol concentrations below the toxic limit. However difficulty arise in such treatment due to the toxicity of phenol to the microbial population [30]. In the biological dentrification, in the water is converted into gaseous nitrogen (N_2) [7]. The biological degradation of phenol is accomplished through benzene ring cleavage using the enzyme present in the microorganism. The bacteria express differently when exposed to different initial phenol concentrations and other conditions [10]. The most efficient *Pseudomonas Putida* is capable of using phenol as the sole source of carbon and energy for cell growth and metabolism degrade phenol via meta-pathway. That is the benzene ring of phenol is dehydroxylated to form catechol derivative and the ring is then opened through meta-oxidation. The final products are molecules that can enter the tricarboxylic acid cycle [24].

The most common Bio-reactors are (1) Aerated lagoon, (2) Oxidation Ditch, (3) Activated sludge system, (4) Anaerobic digestion system, (5) Oxidation pond, (6) Trickling filters, (7) Rotating discs biological reactors, (8) Basket type bioreactors, (9) Hollow fiber membrane bioreactor, and (10) Fluidized bed bioreactors [1,2,4-24].

Comparison of Bioreactors

Treatment of industrial and/or domestic wastewaters requires a great deal of space when using systems based on activated sludge or aerated lagoons in which retention time is many days [13]. Wastewater having lower phenol concentration in the range 5-500 ppm is suitably treated in the bioreactors like Activated sludge, Aerated lagoons, trickling filter, oxidation ponds. The major constraints in using bioreactors with free cells for biodegradation of phenol include maintenance of proper cell concentration, removal of cell sludge, settling and sedimentation of sludge, sludge recycling etc [1-2,8-15].

A bioreactor integrated to a membrane module is referred as membrane bioreactors. The advantages with MBRs are that they offer long culture retention time and short hydraulic retention time and reduce number of the post treatment processes. The membrane has the objective of removal of particulate substances that replaces the gravitational clarifier to separate the biomass from the treated effluent and retainment of low-growth microbes in the reactor for high cell density operation [7,16,23]. The limitation of this rector is high membrane cost and large energy inputs for membrane operation [23].

In free-culture bioreactors, the microorganisms suffer from substrate inhibition, whereby growth (and consequently pollutant degradation) is inhibited at high pollutant concentrations [30].

Biological fixed films exhibit properties that make them preferable to suspended-cell systems for a wide variety of waste water treatment applications. These properties include high concentrations, enhanced cell retention due to cell immobilization and an increased resistance to the detriment effects of toxic shock loadings [15,17].

Rotating biological contactor gives very good phenol removal efficiency at moderate loading rate. It posses high surface area, provides vigorous contact for the biological growth with wastewater and efficiently aerates the wastewater [1,21].

Two-phase partitioning bioreactors (TPPBs) are characterized by a cell-containing aqueous phase and a second immiscible phase that contains toxic and/or hydrophobic substrates that portion to the cells at sub inhibitory levels in response to the metabolic demand of the organisms. This reactor is capable of degrading the highly toxic chemicals at very concentrations [22].

Hollow-fiber membrane bioreactor (HFMBR) with immobilized culture (biofilm) is an extractive membrane bioreactor, could completely degrade phenol up to 3000 mg/l with moderate hydraulic loading rate [24].

Trickling bed reactors posses a very good biomass concentration show high treatment efficiency at high hydraulic loading rates. But it has limitations like channeling, clogging and high-energy consumption [17].

Over the conventional type free-culture bio-reactors the immobilization cell bioreactors like CSTR, PFR, fluidized bed, air lift type, etc. has the following advantages like continuous reactor operation at any desired liquid throughput without risk of cell washout, protection of cells from toxic substrates, higher growth rate gives high concentration of cells in the reactor, easy cell-treated water separation, enhanced gas-liquid mass transfer rate, plug flow operation by maintaining the immobilized cells as a stationary phase [1,2,8-10,14,15,17,24]. The fluidized bed bioreactors are superior in performance due to immobilization of cells on solid particles reduce the time of treatment, volume of reactor is extremely small, lack of clogging of bio-mass and removal of phenol even at lower concentrations [1,2,4-6,9-19].

Cell Immobilization

Cells of mixed culture collected from soils containing pollutants or specific culture (pure) isolated from the pollutant containing soil are immobilized in/on solid matrix. The specific cultures such as *Pseudomonas Putida* (NICM, Sp, MTCC, Q5, DSM, KT etc) either psychotropic or mesophilic type, *T cutaneum* R57 used for biodegradation of phenol, Catechol, Azo dyes removal of ionic mercury etc., *Pseudomonas* spp. And Bacillus spp. used for denitrification, green sulfur bacteria for sulfide removal etc. are used for immobilization [2,4-12,21,31].

Acclimatization of microorganisms is done by increasing the pollutant concentration (say of phenol) gradually during culture preparation. The acclimated culture is used for the immobilization in/on the solid matrix [8].

Immobilization of cells means that the cells have been confined or localized so that it can be reused continuously. These exhibit totally different hydrodynamic characteristics than surrounding environment [24]. Living cells produce enzymes (biological catalysts) to catalyze cellular reactions vital to the organism. The microorganisms are normally immobilized on natural and synthetic supports [10]. Various types of solid matrices like polyacrylamide gel, Ca alginate, porous glass, plastic beds, activated carbon, sand, charcoal, diatomaceous earth, cement balls made of coal ash, cellulose, polymeric materials, polymeric ions, chitosan, lignins, chitins, coal, collagens etc. have been used for immobilization of whole cells [29]. In the recent years, the immobilization of biocatalysts with polyvalent salts of alginic acids has received much attention because of low cost of alginate and the mild conditions of immobilization [8].

Techniques of immobilization are broadly classified into four categories namely covalent bonding, cross-linking (chemical methods), entrapment and adsorption (physical methods). Covalent binding most extensively used technique, where cells or enzymes are covalently linked to the support through the groups in them or through the functional groups in the support material. In the cross-linking technique, the cells are immobilized through chemical cross-linking using homo as well as hetero-bifunctional cross-linking agents. Adsorption is the simplest of all techniques and does not alter the activity of the bound cells. Adsorption involves adhesion or condensation of the cells to the surface of a carrier. The diving force causing immobilization is the combined hydrophobic interactions, hydrogen bonding and salt bridge formation between the adsorbent and cells. Entrapment within gels or fiber is a convenient method for reactions involving low molecular weight substrates and mainly used for immobilization of whole cells. This method is nothing but the polymerization of the unsaturated monomers in the presence of cells results in the entrapment of the cells within the interstitial spaces of the gel.

Fluidized bed bioreactor for wastewater treatment

This reactor had been successfully applied in the treatment of several kinds of wastewater such as ammonia-nitrogen containing wastewater, photographic processing wastewater, phenolic waste water, coke oven wastewater, and other domestic and industrial wastes. Also used successfully for the reductive biotransformation of mercuric ions to elemental mercury present in the effluents from industrial amalgam process, combustors and power stations [1,2,4-6,9,11-15,18,19].

A fluidized bed bioreactor (FBB) is capable of achieving treatment in low retention time because of the high biomass concentration. FBB offers distinct mechanical advantages, which allow small and high surface area media to be used for biomass growth [1,2,9,13-15].

Fluidization overcomes operating problems such as bed clogging and the high-pressure drop, which would occur if small and high surface area media were employed in packed-bed operation. Rather than clog with new biomass growth, the fluidized bed simply expands. Thus for a comparable treatment efficiency, the required bioreactor volume is greatly reduced. A further advantage is the possible elimination of the secondary clarifier, although this must be weighed against the medium-biomass separator [13,15,25].

The superior performance of the FBB stems from the very high biomass concentration (up to 30-40 kg/m^3) and its ability to produce less amount of excess sludge compared to activated sludge process. The limit on the operating liquid flow rates imposed by the microbial maximum specific growth rate, as encountered in the continuous stirred tank bioreactor, is eliminated due to the decoupling of the residence time of the liquid phase and the growth of the microbial cells. The use of biomass support allows the partial replenishment of the fluidized bed without interrupting the operation in order to maintain high microbial activity [25].

An FBB has attracted considerable interest as an alternative to the conventional suspended growth and fixed-film process in wastewater treatment application due to its high efficiency performance. Once fluidized, each particle provides a large surface area for biofilm formation and growth. The support media eventually become covered with biofilm and the vast available growth surface afforded by the media results in a biomass concentration approximately an order of magnitude greater than that maintained in a suspended growth system [1,2,9,13-15,25].

A practical problem, which occurs in the operation of an FBB, is the excessive growth of biomass on support media. This can lead to the channeling of bioparticles in fluidized beds since biomass loading can increase to such extent that the bioparticles began to be carried over from a bioreactor. The problem of over expansion of fluidized bed due to biomass growth has generally been solved by the removal of heavily biomass-laden particles from bioreactor, followed by the addition of biomass-free particles. However this solution complicates operation of a bioreactor and introduces the need for additional equipment external to the bioreactor, such as a vibrating screen or an incinerator [13-15,25,26].

One way of achieving the constant biomass loading in an FBB is the regulation of mass of cells grown on surface media so that a steady state is reached where the rate of biomass growth is equal to the rate of biomass attrition. Livingston and Chase have demonstrated that a practically steady biomass loading can be achieved in a draft tube fluidized bed bioreactor where shear forces, occurring between the particle and the liquid, slough off excess biomass from support particles [15,25,26].

Another way is the application of a light (matrix particle density smaller than that of liquid) biomass support in a conventional FBB. Sokol and Halfani have reported that steady-state biomass loading was achieved in a three phase (gas-liquid-solid) fluidized bed bioreactor (TPFBB) with KMT particles (made of polypropylene) for over a 9- month operation. Rusten et al. have demonstrated practically constant biomass loading was attained in a bioreactor with a biomass support made of polyethylene [15,25,27].

Conventional FBB are operated in two different ways. In a bioreactor with a heavy (matrix particle density larger than that of liquid) biomass support (e.g. silica sand, coal), fluidization is commonly conducted with an upward co current flow of gas and liquid through a bed of particles. Under fluidization conditions, the bed is fluidized with an upward flow of a liquid counter to the net gravitational force of the particles. Once fluidized, each particle provides a large surface area for biofilm formation and growth. The support media eventually become covered with biofilm and the vast available growth surface afforded by the media results in a biomass concentration approximately an order of magnitude greater than that maintained in a suspended growth system [13,15,25].

In a bioreactor with light support media, fluidization can be conducted either by an upward flow of gas and a liquid through a bed or by a downward flow of liquid and countercurrent upward flow of gas. The countercurrent flow of the gas and the liquid eliminates one of the chief disadvantages of the conventional (co current flow) three phase fluidized bed, namely that of elutriation of particles due to the passage of bubbles through the bed [13,15,25].

The use of biomass support allows the partial replenishment of the fluidized bed without interrupting the operation in order to maintain high microbial activity. The limit on the operating liquid flow rates imposed by the microbial maximum specific growth rate, as encountered in the continuous stirred-tank bioreactor, is eliminated due to the decoupling of the residence time of the liquid phase and of the growth microbial cells. As a result, loading rates that can be applied in FBBs is greater than those used in the suspended biomass growth systems. Shieh and Keenan have reported that for FBBS a volumetric loading rate of 9.8 x 10^{-4} kg BOD_5/m^3 s can be applied to produce effluent values of 0.02 kg BOD_5/m^3 and 0.03 kg suspended solids/m^3. This value is fairly high than the design value of approximately 1.3 x 10^{-4} kg BOD_5/m^3 s for conventional air activated sludge processes [15,28].

The degradation of phenolic type liquors, derived from coal processes, in a continuous stirred-tank bioreactor (CSTB), packed-bed bioreactor (PBB) and FBB shown in [15]. The degradation rates of 0.087, 0.053 and 0.012 kg phenol/m^3 were achieved in the FBB, PBB and CSTB respectively. The effluent concentrations produced by three bioreactors are shown in Table 2.

Table-3 shows a comparison of the FBB with other reactors in terms of specific surface area and biomass concentration.The nutrients for microbial growth are transported first from bulk phase to the surface of the biofilm, and then transported to the inner regions of the biofilm via diffusion. The limiting mass transport rate controls the performance of the biofilm reactor [12,15,32].

Table 2: Typical assays of feed and effluent compositions for the CSTB, PBB and FBB

Constituent	CSTB			PBB			FBB		
	Concentration mg/l		Fractional conversion	Concentration mg/l		Fractional conversion	Concentration mg/l		Fractional conversion
	Feed	Product		Feed	Product		Feed	Product	
Phenol	800	0.5	0.99	800	1.0	0.99	990	<1	0.99
Thiocyanate	195	1.0	0.99	250	84	0.66	-	-	-
Cyanide	0.4	0.3	0.25	<1	<1	-	-	-	-
Sulphate	30	290	-	41	62	-	-	-	-
Chloride	115	20	0.76	<10	<10	-	-	-	-
Phosphate	125	115	0.08	250	245	0.12	125	115	0.09
Nitrate	554	1019	-	380	1221	-	16	13	0.19
Ammonium-Nitrogen	213	298	-	164	247	-	820	750	0.09
Total carbon	640	96	0.85	1780	496	0.71	750	<1	0.99

Table 3: Comparision of FBB with competing bioreactors in municipal applications [21,24, 32].

Parameter	Trickling filetr (PBB)	Rotating biological contactor	HFMBR	FBB
Specifiec suface area per bioreactor volume (m^2/m^3)	12-30	40-50	8-10	800-1200
Biomass concentration (kg/m^3)	Upto 170	Upto 6	Upto 22	30-40

From literature it is seen that the external resistance can be neglected in the case of a high fluidization flow rate [12].

In a three-phase fluidized bed bioreactor it is found reaction rate follows first-order kinetics with respect to oxygen and zero-order one with respect to phenol [33]. For chemical and bio-chemical process, where mass transfer is the rate-limiting step, it is important to know the gas hold-up as this is related directly to mass transfer [34].

The gas hold up at high pressures is always larger than that at low pressures, regardless of the liquid velocity and particle size in three-phase fluidization [35].

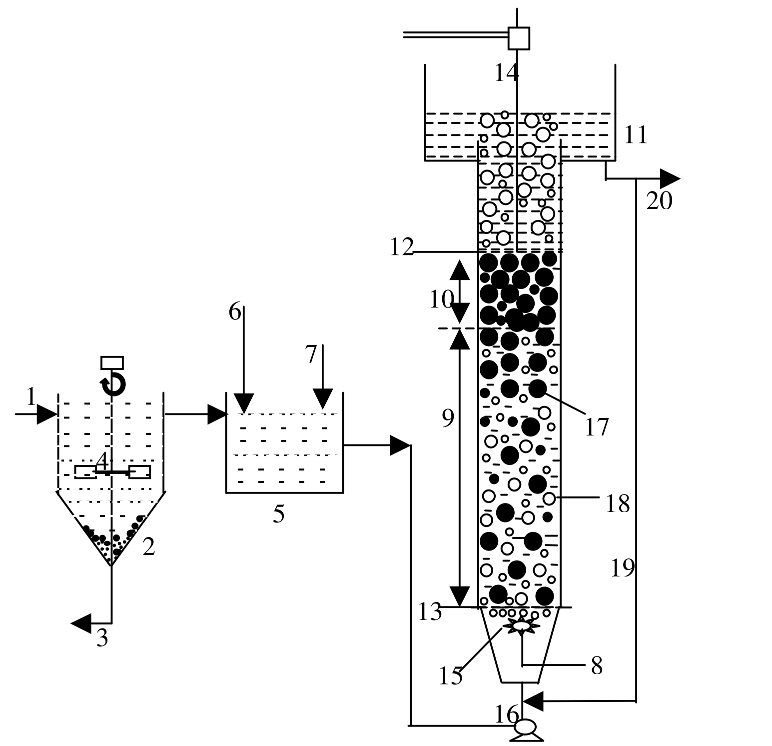
Semi-fluidized bed bioreactor for wastewater treatment

In this type of bioreactor, simultaneous formation of packed bed and fluidized bed is achieved by the prevention of free expansion of a fluidized bed with introduction of an adjustable top screen, which allows the fluid to pass through. The bottom portion of the bed will be fluidized condition while the top portion of the bed will be a packed bed.

In a fluidized bed the reactor is operated at a liquid or gas velocity fairly less than the washout velocity of the cells. But in semi-fluidized bed higher velocity of fluid is possible which will lessen the external mass transfer resistance. As a top packed bed is formed in such a bioreactor, the reactor pressure drop is high that means it is operated under high-pressure condition. Hence the gas hold-up in the fluidizing section of the column will be more this will enhance the mass transfer rate.

If the semi fluidized bed can be used as bioreactor it will overcome the disadvantages of fluidized bed, namely back mixing, attrition and erosion of immobilized solids, reduction of concentration of culture by elutriation, instability due to fluctuation in flow rate of waste water, avoid agglomeration and also overcomes the drawbacks of packed bed such as particle segregation, non-uniformity in temperature and channeling. As the top restraining plate is adjustable slugging by bacterial growth can be prevented. Improved mass transfer in semi-fluidized bed at the cost of higher-pressure drop is compensated by lower operation cost through efficient use of oxygen. The top packed bed portion complements the fluidized bed portion by acting as a polishing section, so that the level of contaminants low compared to fluidized bed bioreactor.

The comparison of performance of different bioreactors with respect to phenol degradation in wastewater is shown in table-4 [9] and a typical semi-fluidized bed bioreactor is shown in figure-1.



1-Phenolic wastewater
2-Primery clarifier
3-Excess sludge
4-Agitator
5-Diluton tank
6-Fresh water
7-Nutrient
8-Air
9-Fluidized bed section
10-Packed bed section
11-Disengagement section
12-Top screen
13-Bottom grid
14–Clam holder to freely adjust the top screen level
15-Air sparger
16-Pump
17-Immobilized culture on solid matrix
18-Air bubbles
19-Recycle
20-Treated effluent

Figure 1: Sechematic diagram of a semi-fluidized bed bioreactor

Table 4: Comparison of performance of bioreactors with respect to phenol degradation of wastewater.

Performance data at max. Phenol degradation				
Condition of feed/ effluent	CSTR bioreactor	Packed bed bioreactor	Fluidized bed bioreactor	Semi-fluidized bed bioreactor
500 gm/lit of phenol	1.0 gm of phenol/ day/m^3 bioreactor	4.7 gm of phenol/ day/m^3 bioreactor	8.5 gm of phenol/ day/m^3 bioreactor	9.1 gm of phenol/ day/m^3 bioreactor
Treated effluent	0.25-1.0 mg/lit	0.21-1.0 mg/lit	0.01-0.5 mg/lit	0.008-0.45 mg/lit

The parameters, which govern the performance of a semi-fluidized bioreactor, are:

(i) Properties of particle; size, shape and density.
(ii) Properties of fluid; density, viscosity and velocity.
(iii) Dimensions of the column and its configuration.
(iv) Initial static bed height, height of top restraint and ratio of top packed bed
(v) To fluidized bed

Conclusion

In this paper the generation and treatment procedure for industrial wastewater generated from chemical process industries have been discussed. A comparative analysis has been critically made to examine the suitability if immobilized culture semi-fluidized bed bioreactor with comparison to other conventional reactors. In this paper an attempt has been made to develop a semi-fluidized bed bioreactor for performance analysis. Among the immobilized cell bioreactors, no doubt the semi-fluidized bed bioreactor is a novel and efficient one, which can be adopted for the treatment of industrial wastewater containing phenolic compounds and other pollutants even at lower concentration. Experimental investigations will continue to characterize and evaluation of its performance for phenolic wastewater treatment. A proper choice of immobilized culture, careful consideration of various design parameters of semi-fluidized bed bioreactors will make treatment process cost effective in the long run.

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