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Review Paper

Biological Oxidation for Treatment of VOCs –A Review

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Abstract

Presence of volatile organic compounds (VOCs) in our atmosphere is a major environmental concern. There are several methods available for the treatment of VOCs exists in the atmosphere. Biological oxidation methods which are adopted for controlling gaseous VOCs have been found as simple techniques to control contaminated gases present in air due to its simple operating conditions, normal design and low cost. This review includes an overview of the various biological oxidation methods which have been used for VOC removal from air. The methods discussed in this review also include details information of their configuration and design, operating mechanism, internal activities of microbial biodegradation process and requirement of future R&D in this area.

Keywords: VOCs, Biofilter, Biological Oxidation, Removal Efficiency, Elimination Capacity.

Introduction

The organic compounds present in gaseous form in air with a vapor pressure over 10.3 Pa at normal temperature (293.15 K) and pressure (101.325 kPa) are classified as volatile organic compounds (VOCs)[1] and considered as a major contributor to air pollution. Various VOCs released by industrial and manufacturing operations in to the air on a large scale, VOCs are significant environmental concern as some contribute to the ozone depletion potential, photochemical ozone creation potential, global warming potential, toxicity, and carcinogenicity and local nuisance. There are many volatile organic compounds which are organic chemicals and released in to the atmosphere by natural and man made sources. Many industries as well as manufacturing plants uses a wide range of organic solvents during chemical synthesis, formulation, extraction and product recovery[2].

There are three main industrial sectors which act as main source for VOC emissions. These include coating and surface treatment related industry which generates 18% of VOCs in atmosphere, chemical industry produce 22% of VOCs while the production of VOCs in power generation and fossil fuel burning sector is 41% [3]. In a survey it was observed that exhaust which produced from transportation play the major role in contribution of VOC generation. After the transportation, painting operations, gasoline vapor and liquefied petroleum gas (LPG) sectors come in to the contributor list.

According to recent studies it has observed that in developed countries such as Canada, the industries related to paints and solvents activities perform nearly 20% contribution of total man-made VOC emission[4]. Glues and lacquers release ketones in spite that agents which deals in carpets and cleaning release chlorinated compounds and as tricholoroethylene or tetrachloroethylene into the air. Methyl acrylate is also a kind of VOCs which is colorless, has a repellent odor and not only effect human health but also create severe environmental disorders. Humans can sense even extremely low exposure of an odorant. It is estimated that only 108 or 109 molecules of odorant vapor in the nose is enough to trigger detection[5]. Acetone, methyl ethyl ketone (MEK), methyl isobutyl ketone (MIBK) and methyl isopropyl ketone (MIPK) are widely used industrial chemicals. These ketone compounds were designed high-priority toxic chemicals[6].

Various compounds which are aromatic in nature such as benzene, toluene, and ethyl benzene found naturally in petroleum products, gasoline and other fuels which are produced from crude oil. In spite of that liquid fuels which are used in automobile industries releases significant amounts of aromatic compounds into the atmosphere due to the incomplete combustion.

A very small amount of exposure of aromatic compounds present in polluted air can cause many types of heath disorders like weakness, loss of appetite and memory confusion, tiredness, nausea and also loss of sight while high level exposure of aromatic compounds can cause dangerous effects on health such as dizziness, unconsciousness and at the last stage death. Now days some widely found VOCs are categories in two group Benzene, toluene, ethylbenzene, and xylene (BTEX) and MEK, toluene, n-butyl acetate and o-xylene (MTBX).

According to United States Environmental Protection Agency (USEPA) benzene has been classified as more carcinogenic and hazardous compound. Due to the exposure to benzene several short-term and long-term health effects can create like dizziness, eye irritation, asthma, cancer and even liver and kidney damages[7].

Five VOC species i.e. methyl ethyl ketone (M), methyl iso-butyl ketone (B), toluene (T), ethyl benzene (E) and o-xylene (X) account for 75–80% of the total VOC load in these emissions. These compounds also constitute major part of the VOC emission from ink, glue and rubber manufacturing units[8].

As a result of these and other sources, over 300 VOCs have been detected indoors. With inadequate ventilation rates in buildings the frequently the conditions exist where VOCs may accumulate[9]. For the treatment of VOCs physical–chemical methods are being gradually replaced by biological techniques, since the latter can provide an efficient VOC abatement at lower operating costs and environmental impacts than their physical as well as chemical counterparts.

Biological treatment is an attractive approach for removing volatile contaminants present in gaseous waste streams. Biofilter is the most widely used biological air treatment process due to its attractive process for the elimination of volatile organic compounds from waste air streams[10]. For the complete degradation of VOCs to generate products such as carbon dioxide, water, and biomass several biological processes are available[11].

Gases which are released during incineration such as CO_2 as well as NO_2 have dangerous affects on environmental conditions. Various poisonous gases which are present on the surface of adsorbents may be responsible for climatic disordered in case of land-filling. Hence it is better to avoid these types of methods to remove contaminates[12]. On the other hand, biological waste air treatments are cost effective, eco-friendly and also simple to operate for the removal of VOCs.

Biological Oxidation Methods

Biofiltration: The biofilter system is a technique which includes the treatment process of organic, inorganic or synthetic media and receives liquid through a spray nozzle on the top of the reactor. Biofilters work as removal technique for various VOCs and also it is the first technique which was widely applied for biological treatment of waste gases present in the atmosphere.

For the purposes of industrial applications different bioreactor configurations have been proposed so far but biofilter is the most commonly used and successful reactor for industrial applications. This filtration technique has very low operating costs for its maintenance and also to operate it required low energy. Residual products are also absent in this system so there is no requirement of disposal or further treatment[13].

Biofiltration process involves many organic microorganisms that found in nature and immobilized in the form of a biofilm which acts as a filtration membrane on a porous medium such as compost, peat, synthetic substances, soil or their combination. This medium provides a hospitable environment in the form of moisture, temperature, oxygen, nutrients and pH to the microorganisms. When contaminated air stream passes through the filter-bed, contaminants are transferred to the biofilm which is developed on the packing materials.

Pollutants are metabolized in their primary components (such as carbon dioxide and water in the case of carbonaceous pollutants) and additional biomass and innocuous metabolic products with the help of microorganisms[14]. In biofiltration unit (Figure-1) contaminated air is passed through the supporting media and packing material is considered as the core of a biofilter, since its nature influences both removal performance of contaminants and operational costs.

Biotrickling filter: Among the various biological oxidation techniques, biotrikling filter (BTFs) are the most preferred technique due to their stable operation, high removal rates, low capital expenditure as well as better pH control. However, they are prone to excess biomass accumulation in the bed[15].

This technology used for gaseous treatment relies on odorant degrading microorganisms which are growing in a biofilm attached to an inert packing material. Biotrickling filter utilized a packing material which is chemically inert such as plastic supports, lava rock, polyurethane foam and pall rings that can be arranged either in a random or a structured manner[16].

The polluted air stream passes through the packed bed transferring the gaseous pollutants to the biofilm while an aqueous solution is trickled over the packing media to provide essential nutrients and water to the microorganism[17]. Hence, it is observed that for the treatment of complex combination of various organic and inorganic gaseous pollutants such as mixtures of methanol, a-pinene, and hydrogen sulfide (H_2S), biotrickling filter are considered as suitable bioreactors. The filtering material used in a BTF has to resist crushing and compaction as well as favor the development of the microflora.

For the best meet of these specifications BTF packing are made from inert materials. As the contact between the microorganisms and the pollutants occurs after the VOC diffusion in the liquid film, the liquid flow rate and the recycling rate are recognized to be critical parameters for BTF operation[18].

BTFs perform more consistent and convenient operation as compared to traditional biofilters (BFs) and the reason behind this, control of overall pressure drop, maintain pH value, nutrient concentration, and enables better elimination rates for higher pollutant to be obtained for a broader range of pollutants.

Bioscrubbers: Bioscrubber unit consist two subunits namely an absorption unit and a bioreactor unit. Input gaseous contaminants can be transfer to the liquid phase with the help of absorption unit. Column may contain the packing material and gas as well as liquid phases flow counter currently, however in liquid phase further treatment is also required. Packing medium used in bioscrubber, is used to provide increased transfer surface between the VOC and the aqueous phase .After the gaseous treatment, solution of the waste may be recycled back.

Main advantages of bioscrubbers include an ability to control pH, nutrients, and separate operational methods for the proper growth of microorganism and their activity[19]. Bioscrubbers are rarely used for the treatment of VOCs due to volatile nature of VOCs and have less water solubility but the current up gradation and designing process show their increased interest in various applications.

Membrane Bioreactor: For the removals of hydrophobic pollutants membrane bioreactors (MBRs) are the best filtration technique since they provide a larger gas–liquid interface. In spite of that they also have excellent mass transfer properties. In this filtration process, the VOCs are transferred with the help of a membrane, where they are degraded with the help of biofilm.

Generally the supply of oxygen and VOC compounds to biomass can be done through the gas phase, whereas the growth factor necessary for microbial activity such as nutrients and water can be supply through the aqueous phase. Important advantages of MBRs over other biological reactors are – only selective pollutants can pass through it and its capability to degrade those VOCs which are poorly soluble in water[9].

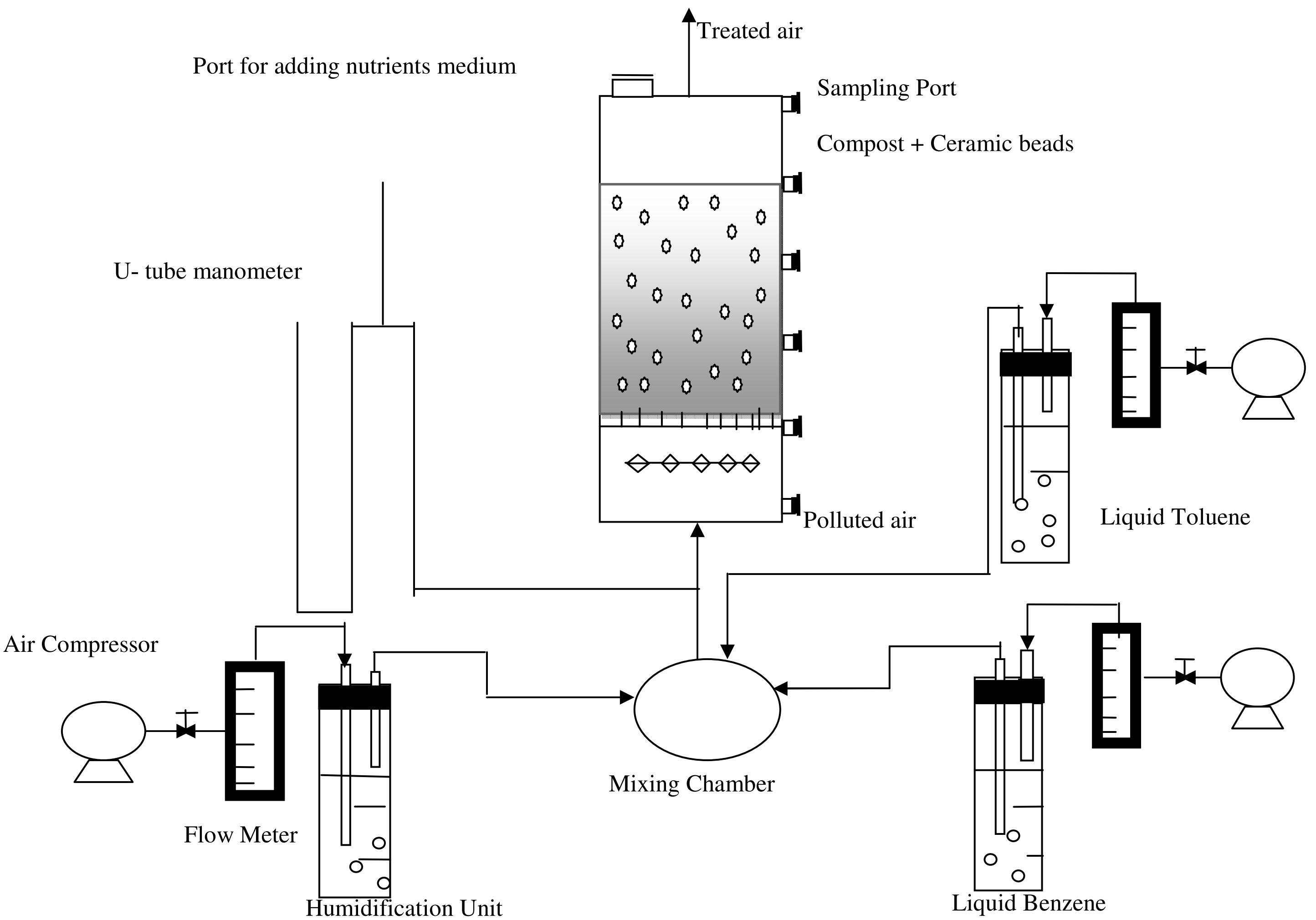

Figure- 1
Schematic Diagram of biofiltration unit[31]

For the biodegradation of pollutants microorganism can be grown with the help of membrane. The performance of MBRs is determined by the type of membrane configuration plate and frame, spiral wounded, tubular, capillary or hollow fiber modules and membrane material like polydimethylsiloxane (PDMS), polypropylene (PP), polyethylene (PE), polyvinylidene difluoride (PVDF), etc.[20]. Summary of different biological oxidation methods for removal of VOCs are listed in Table-1.

Suspended bioreactor: Some problems can become severe in several experimental studies due to packed-bed bioreactors are subjected to high VOC loading rates over an extended period of operation. On the other hand suspended-growth bioreactors have been tested to provide reliable performance[21].

In suspended bioreactor the pollutants present in gaseous form are passing through aqueous phase which contain microorganisms suspended through out the phase. With the help of this method a biomass can be produced and also for the removal of this biomass a process of gravity sedimentation is used. After this process, the recycling of the obtained settled biomass can be performing to get a desired concentration of mixed-liquor suspended-solids in the bioreactor.

Therefore, microorganism present in the biofilm play an important role for degradation of VOCs in a biofilter, whereas in case of suspended cell bioreactors degradation of VOCs occurs because of those microorganisms which are suspended in an agitated mixture.

Rotating biological contactor: Earlier this type of filtration method was used with fixed carriers and mainly applied for waste water treatment but currently it has applications for removing the various pollutants from air. For the treatment of polluted gases, these techniques can be utilized with tangentially flow of gases along the discs of carriers which are known as support medium.

This support medium further connected to a bar which is horizontal. This bar help to make disc rotated at the desired rpm.

For the better connectivity of gaseous and liquid phase the half of disc is submerged in the nutrient solution and half exposed to the air. The rotation of the discs creates a tangential force on the biomass and this biomass help to prevent the clogging of the media.

The unique advantage of rotating biological contactor (RBC) is continuously removal of biomass due to which it has tendency to operate for a long duration without any interference. However, for the treatment of gaseous contaminants rotating biological contactors play a very limited role[22]. Characteristics, Advantages and disadvantages of various biological oxidation methods are summarizing in Table-2.

Biofilter Operation

Evaluation Parameter: Henry's law is used to describe the partition between air and water, which is given by the equation.

$$C_{gi} = H_c \times Cl_i$$

where C_{gi} is the concentration of pollutant *i* in the gas phase, H_c is Henry's coefficient and C_i is the concentration of *i* in the liquid phase. Using a non-dimensional Henrys' coefficient, substances with values over 0.01 are considered volatile, and the higher the value, the less soluble the substrate is in water[23].

Empty bed residence time: Empty bed residence time (EBRT) is a relative measure of gas resistance time within the biofilter medium. It over estimates the actual treatment time. The medium occupies a substantial fraction of the biofiter, reducing the volume within which air flows and shortening the contact time. It is defined as empty bed filter time volume divided by the air flow rate.

$$EBRT = \frac{V}{Q}$$

Removal efficiency: Removal efficiency (RE) is the fraction of the contaminants which are removed by the biofilter. It is mainly controlled by the mass transfer rate of the substrate in the biofilm and in the gas boundary layer, which in turn is controlled by the residence time in biofilter.

$$RE = \left(1 - \frac{C_{go}}{C_{gi}}\right) \times 100$$

Loading Rate: The loading rate (LR) is the mass of contaminated air applied to biofilter per unit medium volume per unit time. Due to the flow remains constant through the bed, the loading rate along the length of the bed will reduce as contaminants are removed. Overall inlet loading rate in biofilter is defined as:

$$LR = C_{gi}\left(\frac{Q}{V}\right)$$

Elimination capacity: Elimination capacity (EC) of biofilter is defined as the mass of contaminants degraded per unit mass of filter material per unit time, and is always equal to or less than the load. Elimination capacity can be expressed as:

$$EC = \frac{(C_{gi} - C_{go})Q}{V}$$

Where: C_{goi} is inlet concentration of pollutant in the biofilter (g m^{-3}), C_{go} is outlet concentration of pollutant in the biofilter (g m^{-3}), LR is inlet loading rate (g m^{-3} h^{-1}), EC is elimination capacity (g m^{-3} h^{-1}), V is the biofilter bed volume (m^3), Q is the volumetric gas flow rate (m^3 h^{-1})[24].

Biofilm: Biofilm is the key element of the biofilteration unit, and work for the biodegradation of the pollutants. 'Biofilm' is the mass of organisms and need surface of solid support to grow; it converts the pollutants to harmless products and carries out the catabolic activity (Figure-2).

The type of pollutant, its flow rate through the biofilter, the material used in bedding, and the design and configuration of the treatment system being used are the various factors which decide the thickness of biofilm. The bulk and substrate phenomenon is used to supply organics or nutrients to the microorganism in the biofilm. The time for development of bioflm may be few days or months depending on the concentration of microorganism.

Biofiltration system consists following three main biological processes. i. Attachment of microorganism, ii. Growth of microorganism and iii. Decay and detachment of microorganism.

The thickness of biofilm vary according to several factors like type of pollutants, its rate of flow through the biofilter, the material used for bedding and the design and configuration of the treatment system being used.

Packing Materials: The important part of the biofiltration process is biofilter bed. It provides the support for microbial growth so called the heart of biofiltration process[25].

The important desirable characteristics of the biofilter bed include the following points: i. It provides high specific surface area for development of a microbial biofilm and gas-biofilm mass transferm, ii. For the homogeneous distribution of gases it has high porosity. iii. A good water holding capacity to avoid bed drying, iv. Intrinsic nutrients are available, and in spite of that peat, soil, compost, and wood chips are the most important basic materials in biofilter beds.

The required desirable criteria are satisfied by these materials. Each of these materials has their own advantages and disadvantage.

Air flow rate: The degradation of VOCs depends on the two important parameters that are flow rate and concentration. The flow rate and residence time are inversely proportional to each other that mean it the value of flow rates become high, the value of residence time will be low and this type of combination can not perform complete biodegradation. Besides it increased value of flow rate leads to strip of water in biofilter bed, due to this biofilter become desiccate.

A biofilter requires an airflow rate of 0.01 cfm. For the better performance of biofilter the flow rates of VOCs should be low while EBRT should be greater. In most of the successfully operated biofilters the EBRT varies from 15 s to several minutes. According to many researchers the longer EBRT can be achieved at lower flow rate and this give rise to better VOC removal efficiencies.

Table-1
Summary of different biological oxidation methods for removal of VOCs[9]

Methods	Pollutants Applied	RE (%)	Remark
Biofilter	Mixture of hydrophilic and hydrophobic VOCs	90-95	Presence of hydrophilic compound, suppressed the growth of hydrophobic degrading microbial community, thereby reducing the removal of hydrophobic compound
Biortickiling filte	VOCs	60	Increase in EBCT is needed to degrade the hydrophobic VOCs, which increases the performance of a reactor
Bioscrubber	Mixture of VOCs	80-85	Doubling of inlet load involved a decrease in the removal efficiency of the VOC.
Membrane Bioreactor	Benzene	98-99	Higher surface area of latex tube, increase the removal of benzene around 99.8% as compared to conventional biofilter
Suspended Bioreactor	Toluene and butylacete	~100	Recirculation of gaseous effluent improved the performance of the reactor
Rotating Biological Contactor	Diethyl ether	43-99	With increase in VOC loading rate, the removal efficiency is dropped

Table-2
Characteristics, Advantages and disadvantages of various biological oxidation methods

Biological Methods	**Characteristics**	**Advantages**	**Disadvantages**
Biofilter	a. Immobilized microorganism b. Immobile water Phase	a. Low operating and capital cost. b. High gas liquid surface area	a. Cogging of the medium b. Large area required
Biortickiling filte	a. Immobilized microorganism b. Nutrient solution is continuously recirculated	a. Better retention for slow growing microorganism b. Low pressure drop	a. More complex to construct and operate. b. Disposal of excess sludge
Bioscrubber	a. Suspended biomass which consist of two units i.e. scrubber and bioreactor	a. No clogging problem b. Better control of nutrients and pH	a. Slowest growing microorganism being washed out b. Disposal of excess sludge
Membrane Bioreactor	a. Microorganism are available in two forms-fixed film cultures and suspended growth cultures	a. No gas phase clogging b. Suitable for low water solubility compounds	a. High operation and capital cost b. Clogging of the liquid channel due to excess biomass
Suspended Bioreactor	a. Suspended biomass which consist of single units	a. Absence of plugging and easier biomass and nutrient control b. Drying of medium does not occurs	a. High operating and capital cost. b. High mass loading rate decreases and performance of reactor
Rotating Biological Contactor	a. Attached and suspended growth of biomass b. Both aerobic and anaerobic degradation occurs	a. Provide higher oxygen transfer, better mixing and high surface area b. Lower operating cost c. Low sludge volume index d. Enable better contact of the air with biofilm for efficient biodegradation	a. Not suitable for highly concentrated pollutants b. Difficult to scale up c. Slow process start up

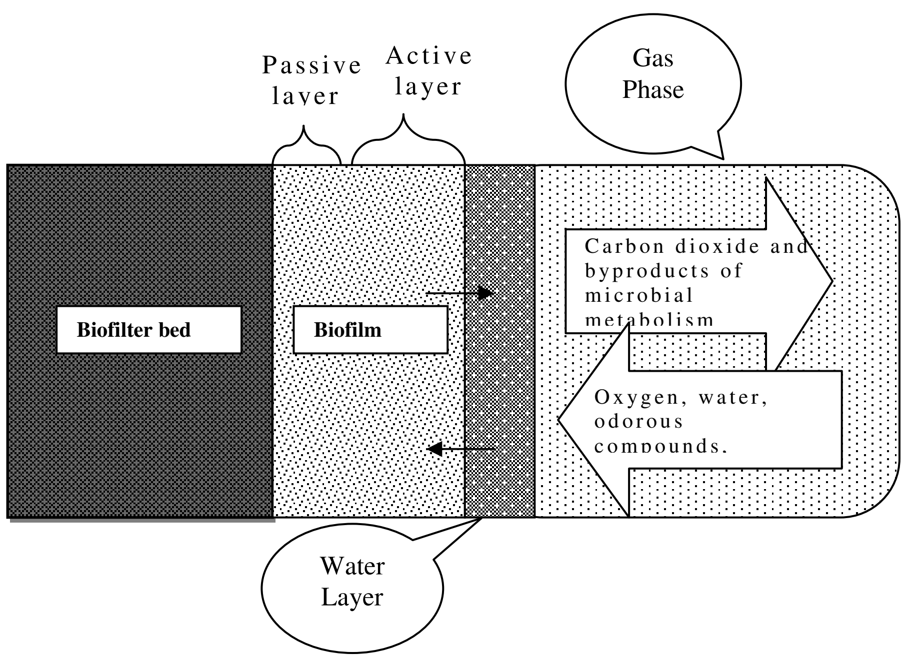

Figure-2
Schematic of a microbial layer on biofilter bed[30]

Pressure Drop (ΔP): Development in biofilm can increase in pressure drop. This will reduce the packing material pore size often increase the pressure drop. Hence huge amount of energy is required to pump the gas effluent into the biofilter. Several factors like flow rate, filter bed characteristics, moisture content, biomass density and wood chips influencing pressure drop across the filter bed. In the biofilter bed biomass accumulation can increase the pressure drop. Among the organic filter beds soil has least air permeable which give rise to high pressure drops. Small particles are present in filter beds that offer more resistance to gas flow and provide high surface area for microbial growth as well as increase the pressure drop. Pressure range from 2 to 8 hPa [25]. Care should be taken to minimize the biofilm which should reflect in pressure drop.

Isolation and characterization of microorganism: In any biological degradation technique the importance of isolating and characterizing of microbial culture is same as removal efficiency. The isolation process starts with the collection of sample then serially dilute and then culture by using spread-plate technique. After this process VOC tolerant strains are screened by culturing the isolate in a selective medium by providing VOC as the source of carbon .Using this approach, we can isolate the strain and further investigation can be performed by two methods phenotypically as well as genotypically. Conventional biochemical tests with reference to Bergey's manual can identify the genera during the phenotypic investigation while genotypic method is more reliable and accurate than phenotypic identification system. During genotypic identification technique such as 16S rRNA sequencing, gene is amplified with primers by the process called polymerase chain reaction (PCR) and then sequenced[27]. A basic local alignment search tool (BLAST) is used to compare the sequence obtained from PCR product with the reference sequence, and GenBank is used to obtain closely related sequences. Further alignment of these sequences can be performed by using CLUSTAL W and phylogenetic tree is predicted using 'njplotWIN95' software tool, which helps to identify the unknown bacteria. This method was used by Sei et al., 2013 for identification of bacteria to degrade 1, 4-dioxane. Moreover, mass spectrometry is another technique which may be used as an alternative technology for identification of strains reason is that, the mass of each nucleotide is different from the others and this difference is detectable by mass spectrometry.

pH: pH of medium in biofilter is very important, which effects the biodegradation process. Fungi are able to grow in acid or neutral pH (2-7). But ideal pH for biofilter process is from 7 - 8. The pH in biofilter varies due to production of acid metabolites this will drop in pH .Care should be taken to maintain the pH [28].VOCs that contain hetero-atoms (S, O, N) are converted into acidic products, which tend to reduce the pH, affect microorganisms and cause corrosion problems in downstream conduits. The bed irrigation method with nutrient solutions that contain pH buffers also used to control pH for example Ca $(OH)_2$, NaOH ,$NaHCO_3$ and urea etc .

Moisture Content: In case of biofilter operation moisture contents play an important role. The moisture is a necessary parameter for microorganisms to carry out their normal metabolic activity. According to the requirement of filtering medium, for optimal operation of the biological filter the moisture content should be within 30–60% by weight and care should be taken to avoid the drying of filter bed. Besides it the supply of moisture on supplemental basis may also be required because bio-oxidation is an exothermic process which can leads to dryness within the bed. Moisture level in the biofilter is often maintained through the process of pre- humidification of the inlet gas stream. Localize dry spots can be caused due to drying of the packing material. Because of this dryness, activity of microorganism can be reduced and gas distribution can also be non uniform. Relative humidity should be maintained 90 – 95% for pollutant entering the biofilter.

Temperature: Temperature is one of the important parameter which affect performance of the biofilter. Only up to an appropriate range of temperature microorganisms can survive and effective. Though the reaction rate increase when the temperature rise, it should be not over the limit of those microorganisms. If the temperature is over the range, cell components such as enzyme and membrane can be destroyed. In the biofilter, several species of microorganism are developed. If the temperature changes rapidly, some species might not active except for those which can tolerate. Most of the biofilters are operate in the warm temperature because if the gas temperature changes rapidly, the thermophilic and phychrophillic could be inhibited. The range of optimum temperature is between 20 and 30ºC [28]. But in the case of mesothermic microorganism, temperature ranges are between 30 and 40ºC. In Some cases biodegradation process can be enhanced by increase in temperature. But care should be taken to control temperature below 40ºC; otherwise the decay of microorganism takes place.

Nutrients: For various microbial activities carbon and energy source are the main source to introduce pollutants into biofiltration. Other nutrients like hydrogen and oxygen are found in the air, in the growth medium, and sometimes in the VOCs. Other macronutrients (N, P, K, and S) and micronutrients (vitamins, metals) are partially fulfilled by the filtering materials used in the biofilters. Nutrients which are necessary for microbial growth are supplied in three ways either in the solid form or directly inserted into the filter bed, or as aqueous solutions. Wu et al. (1999) reviewed the most common nutrient solutions used in biofilters. These include KH_2PO_4,, KNO_3, $(NH_4)_2SO_4$, NH_4Cl, NH_4HCO_3, $CaCl_2$, $MgSO_4$, $MnSO_4$, $FeSO_4$, Na_2MoO_4, and vitamins (B1, etc.)[29].

Oxygen requirement: Oxygen is the most likely to affect high performance of biofilters or when thick biofilms exist. As the operation mode of biofilter is aerobic they require oxygen to perform. This oxygen is generally supplied to biofilter along with the pollutant stream. For each mole of VOCs in the gas form minimum quantity of oxygen in an aerobic biofilter should be hundred mol. In spite of that an additional requirement of O_2 for the biofilter is necessary which can be fulfilling with the help of a blower prior to humidification process. Oxygen is considered as a cut-off factor in contaminated air. Performance of the biofilter can be improved if the quantity of oxygen is larger in polluted air. According to a study the condition of micro-anaerobic help in the treatment of VOCs of organic nature in the biofilter[29].

Conclusion

If Biofilter systems utilized with their full capacity, may have negligible affect on the environment conditions because they utilized biodegradable materials. Most of them can be used second time, required low energy to operate and the obtained unwanted byproduct during filtration are usually very low and does not affect on environment. Besides these benefits the biggest advantage of biological oxidation method is that they do not compromise on the removal efficiency. These are the reasons due to which, filtration methods containing organic materials and biological methods in their operations are having more importance as well as do not create impact on ecosystem. The desired progress in understanding the fundamentals of the bioprocess can be achieved by the moderation of design in reactor so that a more innovative, logistic and focused approach in bioreactor design can be employed as well as its performance can be enhanced. For the better treatment of VOCs compounds from polluted air there is a necessity of continuous innovation and improvements in the arrangement of bioreactor.

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