Biological waste air treatment in biofilters

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Recent studies in the area of biological waste air treatment in biofilters have addressed fundamental key issues such as microbial dynamics, microscopical characterization of the process culture and oxygen and nutrient limitations. The results from these studies have provided a deeper insight into the overall biofiltration process. In the coming years, such advances should allow for the design of better reactor controls and the improvement of pollutant removal in gas-phase bioreactors.

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Abbreviations

FAME fatty acid methyl esters
MEK methyl ethyl ketone
MIBK methyl isobutyl ketone
PLFA phospholipid fatty acids
VOC volatile organic compound

Introduction

Increasingly stringent environmental legislation is generating great interest in industry as to the effectiveness of biological waste air treatment techniques. Biological waste air treatment is relatively inexpensive compared with conventional techniques such as incineration or adsorption onto activated carbon. Also, biological treatment is environmentally friendly, treatment is performed at ambient temperatures, and it does not generate nitrogen oxides or secondary waste streams. Pollutants are generally converted to carbon dioxide under the action of growing or resting microorganisms. Biological waste air treatment techniques have become the method of choice in many instances for the control of low concentrations of odors, volatile organic compounds (VOCs), or hazardous air pollutants in large air streams.

In recent years, there has been significant maturation of biological waste air treatment research. This has resulted in a large number of papers concerning the performance and operation of the so-called biofilters, or biotrickling filters. Interestingly, the fundamental processes involved during the elimination of a pollutant in a gas-phase bioreactor are still very poorly understood. In this review, advances made in the past year are discussed and placed in an overall perspective for process understanding and process optimization. The discussion focuses on fundamental and microbiological aspects of pollutant elimination in biofilters. Although there have been some significant

contributions concerning the design and the operation of biofilters [1,2**] and concerning the marketability of biological waste air treatment techniques [3,4], these aspects, as well as recent advances in biotrickling filters and innovative gas-phase bioreactors, will not be discussed herein.

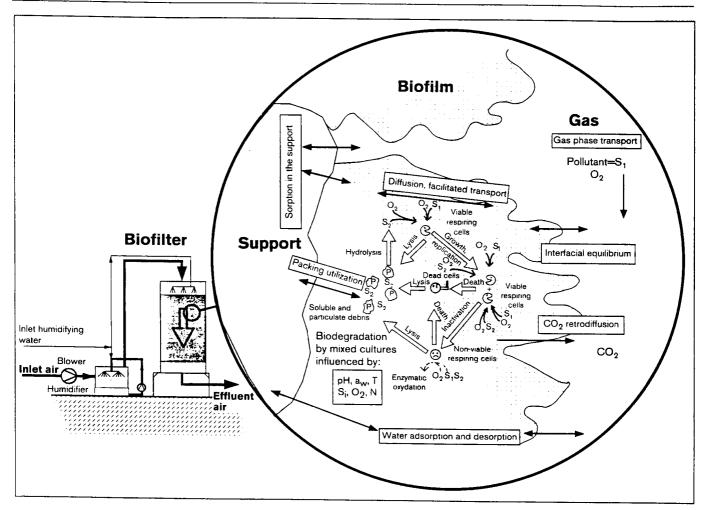
Biofiltration process

Biofilters are reactors in which a humid polluted air stream is passed through a porous packed bed (generally a peat or compost mixture) on which a mixed culture of pollutant-degrading organisms is naturally immobilized. As described in Figure 1, the elimination of a gaseous pollutant in a biofilter is the result of a complex combination of different physicochemical and biological phenomena. The following discussion seeks to identify the different steps likely to occur during biofiltration, so that reviewed papers can be placed in a general perspective for their contribution to the understanding of the process.

The pollutant vapors and oxygen are transported in humid air by forced convection. Interphase mass transfer occurs, and, provided that the biofilter bed particles are small, interfacial equilibrium is achieved so that gas-phase resistance can be neglected. In the biofilm, simultaneous diffusion and biodegradation of the pollutants occurs as a result of growing or resting microorganisms. Oxvgen is also subject to diffusional resistance. The sorption of all chemical species involved is possible either after diffusion through the biofilm as modeled before [5] or directly from the gas-solid contact occurring on bare portions of particles [6]. As biodegradation occurs in the biofilm, metabolite formation is possible [7-9]. If formed, metabolites will undergo the same simultaneous diffusion/biodegradation/sorption processes. Acidic metabolites will be neutralized by limestone or other pH buffer agents generally mixed with the support prior to the packing of the biofilter. The fact that metabolites are seldom observed suggests that they are degraded faster than the primary pollutant either by ancillary strains in direct contact with the primary pollutant degraders (consortia) or directly by the primary pollutant degraders themselves. The carbon dioxide resulting from the oxidation diffuses back and is further transferred to the gas phase. Some of it can also accumulate as carbonate.

The role of the packing is to support the biofilm. In addition, it serves as a reservoir for water, pollutants and nutrients by adsorption on its matrix and absorption in pore water. The packing material generally comprises a fair amount of organic residues that may be utilized as nutrients by the process culture, even if it is still not clear to what extent they are utilized. Little is known

Figure 1



The biofiltration principle, the fate of pollutants during the process, and the physiological states (inspired from [12]) of the process culture. a_{wi} water activity; N, nutrients (N, P, K, and trace elements); S_1 , primary substrates (pollutants); S_2 , secondary substrates.

about biofilter process cultures and how they are affected by operating conditions or their direct environment. In most instances, microorganism emissions [10] and nutrient leaching [11] can be neglected, such that biofilters can be considered as closed systems with respect to nutrient balance. Hence, a (pseudo) steady state must exist between the different physiological states as described in Figure 1. The 'cryptic' growth of microorganisms [12] is not the only means of recycling nutrients because higher organisms like protozoa are also present in biofilters. Even if they do not contribute directly to the pollutant elimination, they are certainly essential for nutrients cycles in the system. In the future, as fundamental understanding of the phenomena described in Figure 1 progresses, reactor performance and process control will be improved.

Microbial dynamics

A number of questions exsist as to the composition of the process culture. Classical microbiological techniques have revealed the presence of mixed populations of bacteria, yeast, fungi and higher organisms in biofilters. In most

cases, these techniques are limited to the quantification and characterization of culturable organisms, that is, a small proportion of the actual microflora. To overcome this limitation, two recent studies [13°,14°] have taken advantage of specific lipid markers to characterize in situ changes in the bacterial communities. De Castro et al. [13°] used the analysis of 50 fatty acid methyl esters (FAMEs) generally utilized for microbial identification and reduced the results to two principal components: to characterize different sources of biofilter inoculum and to quantify the changes occurring to the biofilter process culture over 90 days. The biofilters treated vapors of α-pinene, a common pollutant in the wood industry. The results demonstrated that the composition of the process culture differentiated rapidly from the inoculum, and that significant differences were observed between the inlet and the outlet side of the biofilter. This implies that the widespread assumption of homogeneous culture structure throughout the bed height is not valid. In a similar approach, phospholipid fatty acid (PLFA) fingerprints were used by Webster et al. [14°] to monitor various aspects

of the physiology and composition of the process culture. Several biofilters treating low concentrations of both VOCs and hydrogen sulfide were operated with and without pH control. Unexpectedly, the results show that changes in the microflora occur even after more than 500 days of operation, whereas it is usually assumed that a constant microflora is generally achieved within a few weeks. Whether the observed changes were related to operational factors such as moisture content or related to the aging of the biofilter could not be established. Interestingly, the long-term composition of the process culture was not influenced by the packing type (compost or granular activated carbon). This suggests that, in some cases, the long-term deactivation of biofilters is related to packing structural changes (channeling and compaction) rather than to process culture deactivation. This contradicts nutrient limitations discussed further in this review and emphasizes the fact that biofilters can be subject to various limitations depending on the application and the operating mode. In addition to providing fundamental insights, the use of markers such as FAMEs or PLFAs might, in the future, serve as high-tech diagnostic tools for troubleshooting biofilters exhibiting poor performance.

Biofilm architecture and microbial density

The study of biofilm architecture is a difficult task. In biofilters, the ill-defined nature of the support (generally compost) and the absence of thick biofilms makes it even more difficult. Therefore, most investigations on biofilms in gas-phase bioreactors use a biotrickling filter configuration, allowing an inert support to be used and thicker biofilms to be sampled. The results of biotrickling filter studies can probably be used to understand biofilter biofilms, although differences must exist because of the absence of shear or liquid convection in biofilters. Two remarkable studies are worthy of mention [15.,16.]. Moller et al. [15.] described the distribution of Pseudomonas putida in the biofilm of a toluene-degrading biotrickling filter, using, amongst other techniques, scanning confocal laser microscopy, 16S rRNA probes, and various staining techniques. Interestingly, P. putida constituted only 4% of the total biofilm population, but was responsible for about 65% of the degradation of the toluene vapors. Further, a comparison of the rRNA content of P. putida in the biofilm and growing under optimum conditions in suspension indicated that toluene degradation activity by P. putida was substantially lower in the biofilm than in suspension. This can probably be attributed to various stresses on the immobilized culture, but it also suggests that the full potential of biofilters and biotrickling filter has not vet been fully explored. Finally, the microscopic observation of fully hydrated biofilm sections revealed important heterogeneities, with large channels extending from the gas-liquid interface of the biofilm to the substratum. Such channels clearly increase pollutant and oxygen availability. In a previous study with submerged biofilms [17], de Beer et al. evaluated that the supply of oxygen through

such voids and channels was roughly 50% of the total oxygen transfer. This might explain why primary pollutant degraders are found throughout the biofilm, and not only at the gas-liquid interface. Further, it raises the question of the validity of traditional flat surface biofilm modeling. Another study reports highly heterogeneous biofilms in gas phase bioreactors [16]. Detailed microscopic observation of relatively thick biofilms (2-5 mm) in biotrickling filters revealed that the biofilm comprised three regions, the relative thicknesses of which were approximately constant. The external film (5% of total thickness) had a high bacterial and hyphal tip density. It also comprised a number of pocket-like structures free of cells. These structures could correspond to the channels described by Moller et al. [15.]. The intermediate region (20% of total thickness) was characterized by reduced numbers of bacteria, an increased number of hyphae and an absence of pocket-like structures. Also, a number of nematodes were detected. Finally, the basal region (75%) of total thickness) composed of tightly compacted hyphae against the substratum exhibited little staining with haemato-eosin, indicating the absence of cytoplasmic material, that is, dead or starved biomass. The fact that large amounts of presumably heterotrophic fungi, that is, those requiring an organic carbon source for growth, were detected in a reactor dedicated to hydrogen sulfide and carbon disulfide elimination (i.e. in the absence of organic carbon) raises a number of questions concerning the ecology of the process culture and the fate of carbon during waste air biotreatment.

Because of experimental difficulties encountered during direct microbiological investigations of the process culture, a number of studies focus on the determination of indirect or bulk microbial parameters such as protein content or respiration rates. One of the most interesting recent contributions is by Cox et al. [18**] who report protein content, crude lipid (a storage material in the cells) and organic matter content in a styrene-degrading biofilter containing the yeast Exophiala jeanselmei. The results demonstrated an approximately homogeneous distribution of those compounds over the height of the biofilter. This supports previous work [19-22] on biofilter modeling, where the biofilm was assumed to be homogeneous throughout the height of the reactor without much justification. Further, the microscopic observation of biofilms showed that the average thickness of the biofilm increased from 80 µm on day 35 to 280 µm on day 300, but that no improvement in pollutant elimination occurred. During this time, the total protein amount in the biofilter remained approximately constant, suggesting that the mass of active microorganisms also remained constant. Hence, it demonstrates that most of the biofilm was made of inactive cells. In general, whether this inactivity is due to the limited availability of nutrients, diffusion limitation of oxygen in the biofilm, a poor turnover of dead biomass or another phenomenon, such as the presence of a toxic compound, requires evaluation on a case-by-case basis.

Oxygen limitation

In the case of Cox et al. [18.] discussed in the previous section, experiments with air enriched with oxygen improved the performance of the biofilter and demonstrated that oxygen was indeed limiting. In similar experiments, Deshusses et al. [23] found that no significant improvement of the simultaneous removal of mixtures of methyl ethyl ketone (MEK) and methyl isobutyl ketone (MIBK) was observed when the oxygen content in air was increased. In this case, the fact that significant cross-inhibition of MEK and MIBK biodegradation occurred suggests that kinetic effects were more important than diffusion effects. This was further demonstrated in transient experiments where pulses of either compound were injected into biofilters, and both cross- and self-inhibitions were observed. Clearly, these two examples demonstrate that oxygen limitation is case specific. As a rule of thumb, it is most likely to occur in high-performance biofilters or when thick biofilms exist. Other studies suggest that fortuitous anaerobic microenvironment conditions exist in biofilters [24.25], as indicated by the biodegradation of traces of perchloroethylene (PCE), a compound recalcitrant to aerobic degradation. A step further was accomplished by du Plessis et al. [26] who promoted anaerobic conditions in a biofilter so that nitric oxide could be denitrified. To establish an anaerobic biofilm, toluene-contaminated air was enriched with acetylene. The removal rate of nitric oxide remained low, and process optimization will be necessary before the effective treatment of combustion gases can be achieved. These latter examples are the exception rather than the rule. In most applications, biofilter control will seek to avoid anaerobic conditions because of the formation of odorous compounds under anaerobic conditions and the limitations resulting in pollutant elimination.

Nutrient requirements

The effectiveness of nutrient additions to biofilters remains a highly debated question. Clearly, a continuous supply of nutrients is not desirable because rapid clogging of the biofilter bed by excessive biomass growth would occur, but an intermittent supply might prove useful to replenish exhausted biofilters or to temporarily increase biofilter performance. In this respect, a study on the effect of gaseous ammonia additions to biofilters reported by Auria et al. [27] merit comment. 48h after the first ammonia addition, toluene removal in the biofilter increased from approximately 10 g of toluene degraded per cubic meter biofilter per hour (g m⁻³ h⁻¹) to 80 g m⁻³ h⁻¹ (100% removal). The increase in performance correlated with increases in carbon dioxide production and in reactor temperature. After four days, the removal started to decrease slowly. It stabilized at about 30 g m⁻³ h⁻¹ 15 days after the ammonia injection. When a second injection of ammonia was given, the effect was much less pronounced, and only up to $40 \,\mathrm{g}\,\mathrm{m}^{-3}\,\mathrm{h}^{-1}$ of toluene was eliminated. Drving conditions and heterogeneities in the biofilter packing material were possible explanations discussed by

the authors as to why the second ammonia injection was not as effective as the first one. This was demonstrated by comparing toluene uptake in gaseous or liquid microcosms loaded biofilter packing material. Further evidence of a case where nitrogen was limiting pollutant elimination in a compost-based biofilter was presented by Morgenroth et al. [28]. When suboptimum hexane elimination (~10 g m⁻³ h⁻¹) was observed, several measures were used in an attempt to improve performance. This included mixing of the biofilter media, drying and mixing the media, and the addition of concentrated solutions of potassium nitrate to the biofilter bed. In the latter cases, a low concentration of nitrate improved performance for only four days, whereas the addition of a high concentration of nitrate resulted in the complete removal of hexane (21 g m⁻³ h⁻¹) for over two months. Because nitrogen limitation most probably depends on the availability of nitrogen from the packing, the authors suggest that determining potentially mineralizable nitrogen (PMN) in compost used for biofiltration, using ammonium-nitrogen production under waterlogged conditions. For sustained operation, composts with high PMN will be preferred because they allow for the slow release of assimilable nitrogen. As a whole, this study emphasizes that further research is needed to understand the nitrogen cycle in biofilters. In the future, when both the nitrogen and the carbon cycles are better defined, further research will be needed to understand the cycles for potassium and phosphate and to assess the role of trace elements in biofilters.

Conclusions

Biofilter technology was utilized in the field well before there was a basic understanding of its fundamental principles. This has resulted in several cases of unsuccessful or suboptimum operation of large-scale biofilters. Today, with recent advances in the understanding of the fundamental principles underlying biofiltration, promise exists for better reactor design with optimal operating conditions. Even so, a number of fundamental questions remain unanswered or require further clarification. These include, but are not limited to, the quantification of biomass turnover, the definition of biodegradation kinetic relationships and factors influencing these relationships, the question of the complex ecology of biofilter microflora, and the determination of the availability and cycles of pollutant, oxygen and essential nutrients. All of these factors have been shown to significantly influence both the performance and the long-term stability of biofilters, and thus require further investigation. In particular, quantitative studies are necessary. This should be made easier with the expanding use of modern tools of biotechnology. Finally, the largest problem to overcome will be the translation of recent and future basic advances into real process improvements; however, it is the only way for biofiltration technology to mature from the actual mysterious black box reactor to a well-engineered process. based on solid science rather than on trial and error.

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Webster TS, Devinny JS, Torres EM, Basrai SS: Microbial ecosystems in compost and granular activated carbon biofilters. Biotechnol Bioeng 1997, 53:296–303.

Granular activated carbon and yard waste compost biofilters were used to treat air contaminated with low concentrations of VOCs and hydrogen sulfide. Various pH control strategies were implemented and their effects on the process culture were monitored using PFLA markers. The results showed that pH significantly affected the stress of the process culture and

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