

Process technology of biotechniques

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PROCESS TECHNOLOGY OF BIOTECHNIQUES)*

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Introduction.

As a result of the increasing concern about environmental pollution, in many countries a statutory control nowadays takes place on the emission of toxic compounds into environment. Stronger regulations are put into action and hence in many different branches of industry much interest exists in reliable, simple and cheap purification techniques for the elimination of undesirable contaminations present in waste gases. One promising technique is the biological treatment of contaminations. In the last few

decades the number of applications of biological treatment systems for off-gas purification has strongly increased. Although this development is quite recent, the principle of waste gas purification by contacting a contaminated gas flow with a suitable microbial population is much older. Already in 1923 the biological elimination of H₂S emissions from waste water treatment plants was discussed [1]. Afterwards, in 1934, probably one of the earliest patents in this field was applied

reference	Maurer (1979) [2]		Jäger & Jäger (1978) [3]	Lith (1990) [4]	
process	investment costs DM/(m ³ /h)	operational costs DM/1000m ³ (Application not specified)	total costs DM/1000m ³ (composting works) price level 1974	total costs DM/1000m ³ (VOC: 100-2000 mg/m ³)	Gas flow 10000 m ³ /h
Thermal incineration	12-14 ¹	1.4-1.7	9.10 (fuel costs only)	7 - 9	(50% energy recovery)
Catalytic incineration	14-16	1.3-1.5	-	6 - 8	(50% energy recovery)
Adsorbtion	5 -20	0.5-1.0	1.5 (incl. regeneration incineration)	14 - 18	(incl. steam by regeneration)
Absorbtion	8 - 10	0.8-1.0	4.20 (chlorine)	-	
Ozone oxidation	6 - 8	0.4-0.6	4.2	-	
Biofilter open	3-10	0.3-0.5	0.6	-	
closed				0.5 - 3	

Table I: Relative capital and operational costs for off-gas purification [2,3,4]; Capital costs are given per (m³ waste gas/h) (related to the gas flow); operational costs are given per 1000m³ of waste gas treated.

for, claiming a biological purification system concerning 'die Reinigung von luft- oder sauerstoff-haltigen Gasmischen die biologisch zerstörbare Riech und/oder Feststoffe enthalten...' (the purification of air- or oxygen containing gas mixtures, which contain biologically degradable odorous compounds or particles...) [5]. Reports on the actual application of the technique on a larger scale date back from the early fifties, when soil bed filters were mostly applied to purify odorous waste gases from municipal sewage treatment plants. Ever since, a lot of microbiological as well as process engineering research has been carried out on the development of biological elimination systems for the removal of volatile organic and inorganic compounds, as will be discussed below. Although at the outset biofiltration was mainly applied for odour abatement, it has nowadays become an important alternative to many physical- and chemical methods of waste gas purification, as the application of biofilters generally appears to be quite reliable and effective at relatively low cost. Table 1 gives a rough indication of the relative capital and operational costs, involved in the application of different waste gas purification techniques.

Microbial substrate degradation.

The application of a biological treatment system is primarily based upon the microbial degradability of the compounds present in the waste gas. However, the performance of a continuously operating bioreactor is the ultimate result of a complex interaction between the microbiological and physical phenomena, often denoted as the macro-kinetics of the process. The physical phenomena include the mass transfer between gas- and liquid phase, the mass transfer to the microorganisms, the average residence time of the

mobile phases etc. Some of the microbial phenomena, often denoted as the micro-kinetics, are e.g. the reaction rate of the degradation, the substrate or product inhibition and di-auxic phenomena.

The micro-kinetics of the degradation process are generally investigated and modelled for pure cultures of suspended microorganisms. However, in bioreactor systems for environmental purposes mainly heterogeneous mixed cultures of microorganisms are present rather than monocultures, which means that the application of the micro-kinetic results may be limited for bioreactor design purpose.

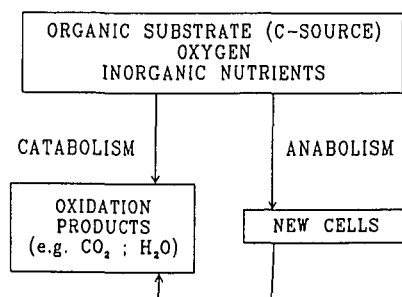
In many bioreactor systems the degradation also takes place within fixed biofilms, which means that additional mass transfer phenomena (i.e. mass transfer to the biofilm and internal diffusion) should be taken into account.

In the following, some aspects concerning the micro-kinetics will be reviewed. Thereafter essential differences of substrate degradations within heterogeneous biofilms will be discussed.

Micro-kinetics of suspended microorganisms.

The elimination of organic substrates by microorganisms results from the fact that those organisms can use the organic compounds as its sole energy- (catabolism) and carbon source (anabolism) (Fig. 1). Approximately 50% of the carbon of the organic substrate is involved in each reaction.

The degradability of a compound often reflects its origin: biogenic compounds are easily biodegradable, while anthropogenic (i.e. man-made) compounds sometimes possess such unnatural structures (xenobiotics) that biological degradation is difficult (recalcitrant compounds) or even impossible (persistent



ENDOGENEOUS RESPIRATION

Figure 1: Aerobic microbial substrate oxidation.

compounds). However, due to intensive microbiological research in recent years, much of progress has been made in isolating, selecting or constructing strains or mixed cultures of microorganisms (mainly bacteria), which can degrade some recalcitrant compounds to such an extent, that the application of a biological waste gas treatment system even in such cases offers real prospects by this time. This is illustrated by Table II, which lists the growth rate of some bacteria on xenobiotic chlorinated hydrocarbons that have been isolated from among others activated sludge, contaminated soils.

Microorganism	Substrate	Growth rate [h ⁻¹]
<i>Hyphomicrobium</i>	methylchloride	0.09 [6]
<i>Pseudomonas DM1</i>	dichloromethane	0.11 [7]
<i>Methylobact. DM11</i>	dichloromethane	0.17 [8]
<i>Xanthobacter GJ10</i>	1,2-dichlorethane	0.12 [9]
<i>Mycobacterium L1</i>	vinylchloride	0.05 [10]
<i>Pseudomonas AD1</i>	epichlorohydrine	0.20 [11]
<i>Pseudom. WR1306</i>	chlorobenzene	0.79 [12]
<i>Pseudom. GJ60</i>	1,2-dichlorobenz.	0.33 [13]
<i>Pseudomonas</i>	1,3-dichlorobenz.	0.07 [14]
<i>Alcaligenes A175</i>	1,4-dichlorobenz.	0.13 [15]

Table II: The aerobic degradation of some xenobiotic chlorinated hydrocarbons by pure cultures of microorganisms.

It is surprising to conclude from the data presented in Table II, that the growth rate of suited strains on many xenobiotic compounds is of the same order of magnitude as encountered in the degradation of many biogenic substrates. This stresses the importance of thorough isolation and

adaptation procedures.

The degradation of substrates and the subsequent growth of microorganisms is generally described by the Monod equation. The value of the Monod-constant K_s for organic substrates generally amounts to 1-10 g/m³, while for oxygen it is about 0.1 g/m³ [16]. However, the concentrations of substrate and oxygen in the liquid phase in bioreactor systems often exceed the value of K_s . The degradation can hence be described as a zeroth order process.

Apart from the substrate availability, the growth rate also depends upon the physiological conditions e.g. the temperature, pH and inorganic nutrients. Fig. 2 shows the influence of the temperature on the maximum growth rate of *Hyphomicrobium GJ21* growing on dichloromethane [17]. An optimum of around 30 °C can be observed, whereas at higher temperatures the growth rate

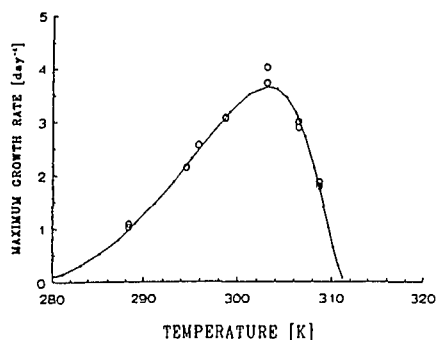


Fig. 2: The influence of the temperature on the maximum growth rate of *Hyphomicrobium GJ21*. For the range of 15 °C to 25 °C an activation energy of 76 kJ/mol was calculated.

quickly decreases. Within the temperature range of practical interest (15°C - 25°C) this influence can be described by an activation energy of 76 kJ/mol, according to the

Arrhenius equation. In general the temperature influence on the growth rate of many mesophylic bacteria is similar to the one shown in Fig. 2.

Most microorganisms are able to grow over a pH range of about 4 pH units, but the growth rate generally has a pH optimum around 6.5-7.5. Above 8.5 and below 5 the growth rate may become very low.

Substrate degradation in biofilms.

Opposed to most micro-kinetic experiments performed on microbial suspensions in laboratory, biological purification systems are often based upon fixed film degradation processes. The microorganisms are immobilized inside the pores or on the surface of a carrier material. The spontaneous formation of aggregates sometimes occurs as well. Fig. 3. shows a microscopic view on an immobilized biofilm of *Hyphomicrobium GJ21* growing on dichloromethane. The immobilization of microorganisms has the advantage that biomass concentrations, hence the volumetric reaction rates, can be considerably increased. However, the kinetics of immobilized cells may substantially differ from that of freely suspended cells.

The most important feature of a biofilm is the existence of concentration gradients of substrates and products. These gradients result from the internal mass transport by diffusion and the substrate depletion by reaction. The reaction rate therefore may vary throughout the biofilm and also serious diffusion limitations can occur. This may result in starvation and decay of cells in deeper parts of the biofilm, which may eventually result in detachment.

If Monod kinetics are applied to describe the biological reaction rate, it is obvious that the substrate gradient will also considerably affect the overall reaction rate. This is illustrated in Fig. 4, in which the effectiveness of a homogeneous, flat

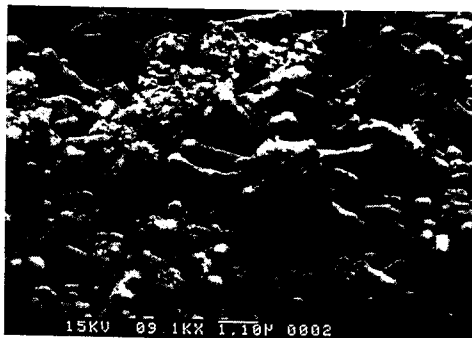


Fig. 3: *Hyphomicrobium GJ21* for the degradation of dichloromethane; immobilized on a glass ring, taken from a continuously operating biological trickling filter (magnification 9100 times).

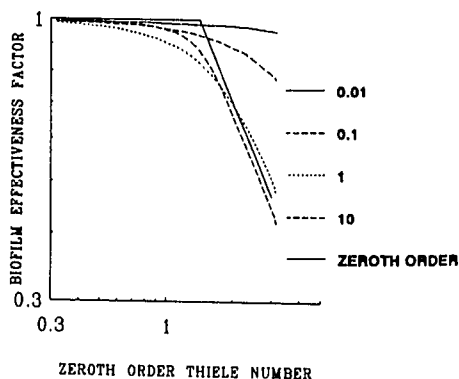


Fig. 4: The effectiveness of a flat biofilm plotted versus the zeroth order Thiele number for different values of α .

biofilm is plotted versus the zeroth order Thiele number for different ratios of the biofilm surface concentration and the Monod-constant. The Thiele number reflects the ratio of the maximum rate of degradation and the maximum rate of diffusion in the biofilm. For low values of the Thiele number the biofilm efficiency approaches unity, whereas at higher Thiele numbers the efficiency decreases. This effect increases at lower values of α .

From Fig. 4 it can be concluded that already at

$\alpha=1$ the biofilm effectiveness factor can be approximated by a zeroth order behaviour.

The diffusion of oxygen in aerobic biofilms is known to have even more pronounced effects upon the overall activity of a biofilm. Typical oxygen penetration depths amount to 100-200 μm , while the whole biofilm may be as thick as several mm (Fig.5) [16,17,18]

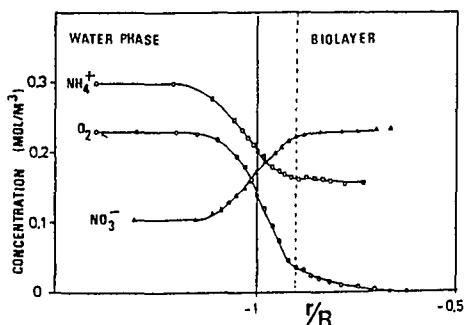


Fig. 5: Concentration gradients of ammonium (\square), oxygen (\circ) and nitrate (Δ) in a nitrifying biofilm. The dotted line indicates a penetration depth of 100 μm . Adapted from [17].

Diffusion limitation aspects also hold for inorganic nutrients. The minimal nutrient requirement during the growth of microorganisms may follow from the stoichiometry of the elemental composition of biomass. However, this stoichiometrical composition is not sufficient to calculate the minimum concentrations required. In a fixed film process, diffusion limitations may further increase the minimal requirements.

Inhibiting products which must be transported outwards will also influence the local activity in the biofilm. Fig 6. shows the existence of a pH gradient which has been determined in a dichloromethane degrading biofilm, in which HCl is produced. Due to the influence of the pH on the reaction rate, the overall biofilm effectivity will decrease.

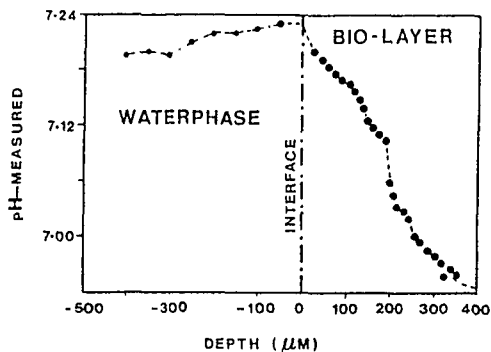


Fig. 6: A pH gradient in a biofilm of *Hyphomicrobium GJ21* degrading dichloromethane. (Measurements by Beer de D., University of Amsterdam, 1990).

Sofar only gradients of diffusing compounds in biofilms of monocultures have been discussed. As bioreactors for environmental purposes generally are continuously operated under non-septic conditions, a heterogeneous population of microorganisms may develop in the system.

Heterogeneous population may develop for two reasons. First, in the field of waste gas purification the carbon source generally comprises a mixture of different substrates. Different microorganisms may be required for an efficient removal of all those components. Secondly, a heterogeneous population can also develop as biogenic material is available in the system, due to the lyses of microorganisms e.g. in deeper parts of the biofilms. Also the presence of predators like protozoa and metazoa (e.g. nematodes, ciliates, worms), is known [16].

Thus the biofilm may no longer be homogeneous, which means that porosity, density, activity etc. may vary throughout the film.

As micro-kinetics are often investigated for pure cultures of bacteria only, the application

of those results may also be limited.

When modelling the macro-kinetics of the environmental bioprocesses much simplifications are necessary, due to the complex nature of the characteristics of immobilized non-septic biofilms, as described above.

Biofilters.

Three groups of biological waste gas purification systems are known, which can be distinguished (Table III) by the behaviour of the liquid phase (which is either continuously moving or stationary present in the contact apparatus) and of the microorganisms (which are either freely dispersed in the aqueous phase or immobilized on a carrier material).

Microbial flora	Aqueous phase	
	Mobile	Stationary
Dispersed	Bioscrubber	-
Immobilized	Trickling filter	Biofilter

Table III: Distinctions between different biological waste gas purification systems.

Biofilters reflect the easier mode of operation as compared to other biological purification systems. In a biofilter (Fig. 7) the waste gas is forced to rise through a simple structured packed bed of materials, in which a suitable microbial population develops in course of time, or by inoculation of the material with suitable microbial strains. It is generally assumed, that the constituent particles of the packing material are surrounded by a wet biolayer.

The volatile compounds and oxygen present in the waste gas are transferred from the gas phase into this biofilm, where the microbial degradation takes place. The packing material normally consists of small particles ($d < 10\text{mm}$), hence a high specific area ($300\text{-}1000\text{ m}^2/\text{m}^3$) and an excellent

mass transfer are established.

As inorganic nutrients (phosphate, nitrogen, sulfur etc.) must also be supplied by the carrier material, mostly natural materials (like compost, peat etc.) are applied. Extra addition of nutrients have sometimes shown to increase the conversion rate of a biofilter. Don [19] reported that the removal efficiency of a

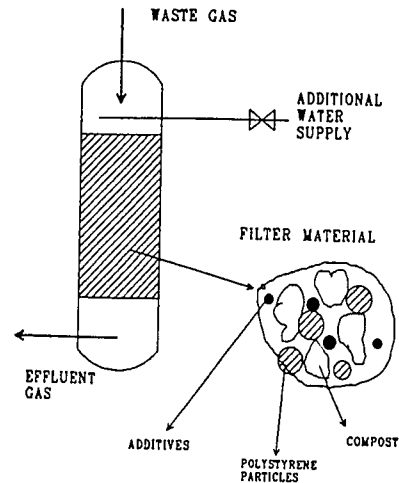


Fig. 7: Experimental set-up of a biofilter system. The filter material consists of e.g. compost, mixed with additives.

toluene eliminating biofilter could be increased from 50% to 95% at an inlet concentration of 100 mg/m^3 and a gas velocity of 100 m/h , by the addition of inorganic nutrients to the filter material.

From the conventional soil- and compost filters, which were used in the early 1950s, a high pressure drop (Fig 8) and a non-homogeneous structure were known to exist [19,20]. In order to reduce energy consumption, the height of such a filterbed amounted to $0.5\text{-}1.0\text{ m}$, while the initial gas loads applied amounted to $5\text{-}10\text{ m}^3/\text{m}^2\text{ h}$ [19]. Fairly long residence times (up to several minutes) were needed in order to achieve a high removal

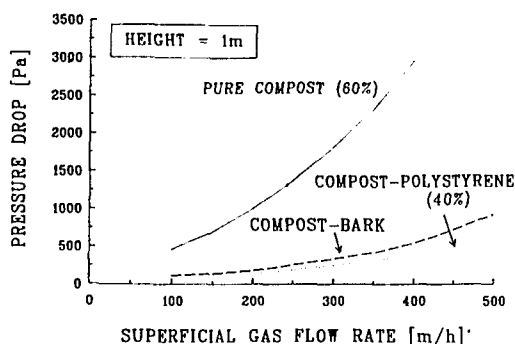


Fig. 8: The pressure drop of a filterbed versus the superficial gas load; the percentages at each curve indicate the water content (bed height = 1m).

efficiency.

This initiated intensive research projects for the development of better filter materials. Nowadays mainly compost and peat are applied, or mixtures of these materials with wooden chips, heather, bark, polystyrene or lava particles [21,22]. The latter materials are added to in order to create a stable filterbed structure in course of time and to decrease the pressure drop ($< 500 \text{ Pa/m}$ at superficial gas loads up to $100\text{-}500 \text{ m}^3/\text{m}^2 \text{ h}$) (Fig. 8). Due to the optimized conditions in present biofilter systems provided with sufficiently active packing materials, the superficial gas contact time is in the range

of 10-30s. Microorganisms generally applied in biofilter systems are mesophylic. The degradation should hence take place within the temperature range of $15\text{-}40 \text{ }^\circ\text{C}$. Furthermore, it has been reported that the optimal water content of the packing material should be in the range of 40%-60% w/w. In order to preserve the microbial activity it should at least exceed 40% [20,23].

Water will evaporate from the filter bed, unless the inlet gas flow is completely saturated. This process will be enhanced by the heat generated by the microbial substrate degradation. In a pretreatment section therefore the inlet gas may

advantageously be humidified. As 100% saturation can hardly be achieved, an additional (periodical) water supply to the filter at the top of the filterbed may sometimes be necessary. As the dry-out process generally starts at the gas inlet side, in those cases the filter may advantageously be operated downflow.

The water balance control of a biofilter is also connected to the lay-out of the filter.

Many different designs and forms have been presented in literature [21,24] and can be divided into 'open-' and 'closed' biofilters.

Open filters are generally subjected to changing weather conditions, thus to a strongly changing temperature and humidity in time and place. This may result in setting and shrinking of the packing material and an increasing formation of a non-homogeneous structure (e.g. channeling) etc.

Since a number of years the interest in closed biofilters is increasing, although these completely housed and insulated systems generally require somewhat higher capital costs. The biofilter performance can be better controlled and ageing phenomena of the packing material prevented, hence overdesign can be minimized. In this way the microbial activity can be exploited for long periods of time (3-5 years) with low operational cost and maintenance [4,25].

Intensive experimental investigations on laboratory, semi-technical as well as full-scale have been carried out to determine the macrokinetics of the filtration process and the values of the rate parameters for many volatile compounds discharged by industry.

For biofilter systems experimental results may be summarized as follows [22,26,27]:

- the macro-kinetics of the elimination proces-

ses in a biological filter bed can be described by an absorption process in a wet biolayer, surrounding the constituent particles, accompanied by a simultaneous biological degradation reaction;

- the elimination of nearly all of the compounds investigated, like alcohols, ketones, esters, aromatics etc., in a biological filter bed follows zeroth order reaction kinetics down to very low concentration levels. This has been confirmed by batch investigations of the degradation process in aqueous solutions of the compounds concerned;
- at low gas phase concentration levels or low water solubility of the compounds concerned, the elimination rate of the filter bed may shift towards a diffusion controlled regime;
- due to the predominantly zeroth order character of the elimination process, the degree of removal of any biodegradable compound may be close to 100% at finite residence times of the gas phase in the filter bed.

The general path of the elimination capacity EC of a filter bed with height H as a function of the inlet gas phase concentration C_p at constant gas flow rate ω is shown in Fig. 9. EC is defined as:

$$EC = \frac{\omega}{H} \cdot (C_p - C_p^*)$$

Three operational regimes may generally be distinguished:

- i) Below point A in the graph the organic load $(\omega/H) \cdot C_p$ to the filter is so low, that practically 100% conversion of the compound is achieved;
- ii) if the gas phase is higher than C_p^* (Fig. 9) the load is so high that the maximum elimination capacity EC_{max} of the filter is reached. The system is in the so-called reaction limited regime [26], the activity of the biolayer surrounding the packing particles is fully utilized and no reaction free zone exists (see also curve 1, Fig 10).

iii) if anywhere in the filter bed the concentration C_t equals C_p^* two zones can be distinguished in the filter bed. If the filter is operated downflow the upper part of the bed is still in the reaction

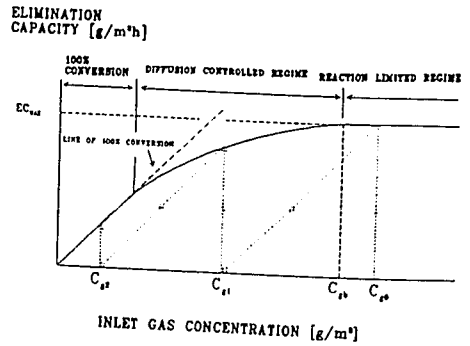


Fig. 9: Schematic path of the elimination capacity of a biofilter as a function of the gas inlet concentration, at constant gas flow rate ω .

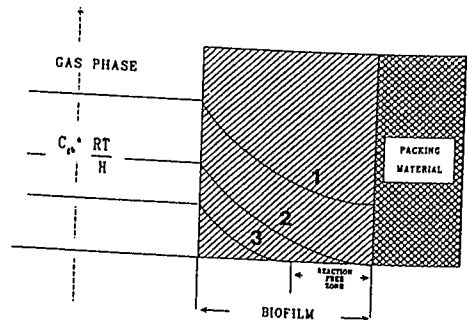


Fig. 10: Biophysical substrate penetration model according to zeroth order kinetics.

limited regime, the lower part however in the diffusion controlled regime. The transition between both regimes occurs at the critical gas phase concentration C_p^* shown in Fig 10. Below this critical concentration (as curve 3 in Fig. 10 shows) a reaction-free zone exists in the biolayer due to the zeroth order character of the micro-kinetics of the elimination process. It

can be shown that in a multi-stage filter, each stage having a height H , the final exit concentration may be found according to the construction shown in Fig. 9. Starting with a concentration C_p in the waste gas the exit concentration from the first stage amounts to C_{p1} , being the inlet concentration for the second stage. The exit concentration of this stage amounts to C_{p2} , etc. In this way the number of stages (or in other words the total height of the filter bed) may be calculated to achieve a desired degree of conversion.

Biofilters are nowadays applied in many branches of industry. Table III lists a number of full-scale applications for the removal of many different mixtures of volatile organic and inorganic substrates. From this Table it will be clear that degrees of conversion of over 90% can normally be reached. The specific costs of biofiltration

amount are generally of the order of magnitude of Hfl 0.5 - 3,- per 1000 m³ of waste gas treated (see Table I).

Biological trickling filters.

In the biological purification of waste gases problems may arise if acid metabolites are produced during the biological degradation. If this process takes place to such an extent that the pH buffering capacity of the filter material is effective for only a relatively short period of operation, the presence of a flowing liquid phase in the system is required for the continuous neutralization of the acids produced, as well as for the drainage of neutralization products from the system. This situation is encountered in the degradation of halogenated hydrocarbons, ammonia, hydrogen sulfide etc.

Application	Gas Flow [m ³ /h]	Elimination of	Number of filter stages	Total superficial Residence time [s]	Efficiency [%]
Gelatine production [28]	35000	odour / n.s.	0.6-1	12-21	70-93
Cocoa & Choc. processing [4]	10000	odour / n.s.	2	22	99
Fishmeal factory [29]	40000	odour / 230 mgC/m ³	1	20	50-90
Tobacco ind. [4]	30000	odour/ NH ₃ (1.5 mg/m ³) nicotine (3.5 mg/m ³)	2	14	95
Waste water treatment [4]	10000	odours / H ₂ S (10 mg/m ³) acetone (8 mg/m ³)	2	29	90-95
Flavour & Fragrance ind. [4]	25400	odours (10 ² o.u./m ³)	2	22	98
Paint prod. [4]	11700	org. solvents (1800 mg/m ³)	2	38	90
Pharmaceutical plant [4]	75000	org. solvents (aromatics, aliphatics, chlorinated compounds)	3	108	80
Photo film production [4]	140000	org. solv. (400 mg/m ³)	2	30	75
Food proc. ind. [4]	9000	odour from oil (10 ⁵ o.u./m ³)	2	20	93
Ceramics prod. [30]	30000	ethanol	1	8	98
Metal Foundry [31]	40000	benzene (9 mg/m ³)	1	30	80

Table IV: Examples of full-scale biofilter applications; * n.s. = no specification; o.u. = odour units; Height of each filter stage = 1m.

For example the degradation of dichloromethane yields hydrochloric acid:

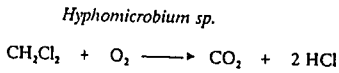


Fig. 11 shows the inhibiting influence of the pH upon the microbial activity of the microorganism *Hyphomicrobium sp. GJ10* [32].

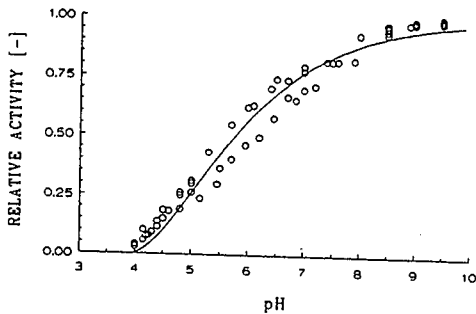


Fig. 11: The relative activity of *Hyphomicrobium GJ21* from a trickling filter versus the pH in the liquid phase.

The problems of acidification and neutralization can easily be solved by the application of a biological trickling filter (Fig. 12). In this system a water phase is continuously recirculated over a packed bed of a carrier material, on which biofilm is immobilized. The contaminants in the waste gas are absorbed in the liquid phase and transferred to the biolayer. Simultaneously, the acids produced are removed from the filter bed, while the pH value of the liquid is controlled by adding an alkaline solution (e.g. NaOH). As the neutralization product NaCl also inhibits the biological activity, as shown in Fig. 13 a continuous refreshment of the liquid phase takes place, thus keeping the NaCl-concentration below inhibiting levels (< 200 mM). The gas flow is forced to rise through the bed co- or counter-

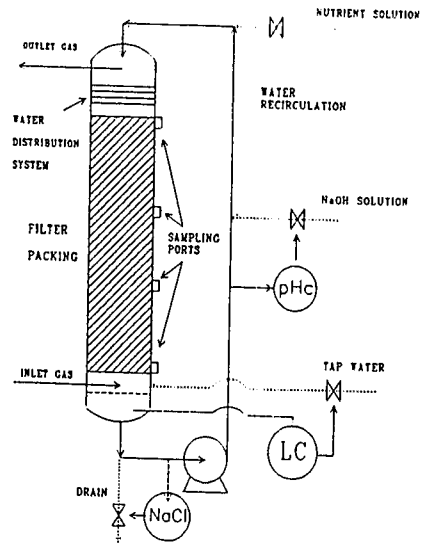


Fig. 12: Experimental set-up of a biological trickling filter system. The packing material consist of conventional packing elements.

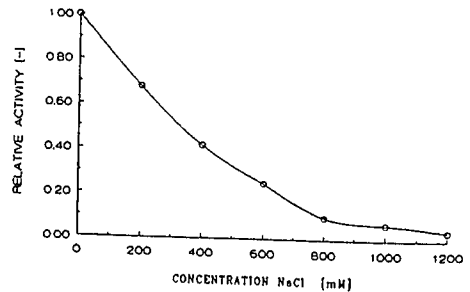


Fig. 13: The influence of the NaCl concentration on the degradation rate of dichloromethane by *Hyphomicrobium GJ21*.

currently to the liquid phase. The packed bed consists of packing elements (e.g. Pall-rings, Novalox-saddles or a structured packing of corrugated sheets) made of inert materials (e.g. glass, plastics, ceramics). On the surface of

these carriers the biofilm is present, which generally develops naturally in course of time, after a trickling filter system is inoculated. A macroscopic view of a biofilm grown on a packing element in a trickling filter is shown in Fig. 14 (see Fig. 4 for a microscopic view).

Other phenomena depending on the liquid flow rate should also be taken into account, as they may strongly determine the efficiency of the trickling filter [32].

In the first place the liquid flow rate strongly determines the degree of wetting of the packing material. Within the range of the liquid flow rate mentioned, the wetted area in a packed bed of dumped elements ($a_w \approx 100 - 350$; element size $> \frac{1}{2}$ ") is normally less than 40% of the specific area available [33]. Somewhat higher degrees of wetting may be found for structured packings. The total rate of mass transfer as well as the conversion rate of the system is proportional to the total biofilm area and hence proportional to the wetted area.

Secondly, the production of acid results in the existence of an axial pH gradient, as the liquid is generally neutralized only before it enters the column. A pH drop from pH=8 to pH=4 has for example been observed during the degradation of dichloromethane in a 2.7 m biological trickling filter at a low liquid flow rate (2 m/h) and a high inlet gas concentration (8 g/m³). This pH-gradient, which also depends on the concentration of dissolved buffering components (phosphates, carbonates, ammonium etc.), will result in a decreased elimination performance due to the pH-dependency of the biological reaction rate (Fig. 11). In Fig. 15 the trickling filter performance for the removal of dichloromethane is shown at different liquid flow rates. The influence of the above mentioned phenomena is clearly shown.



Fig. 14: A polypropylene-novalox saddle, fully grown with a dichloromethane degrading biofilm.

From the phenomena mentioned above, a high superficial liquid flow seems preferable. However, high liquid flow rates enhance the energy costs of the process and increase undesired sloughing of attached biomass.

The optimal value of the liquid flow rate should therefore be found by experiments.

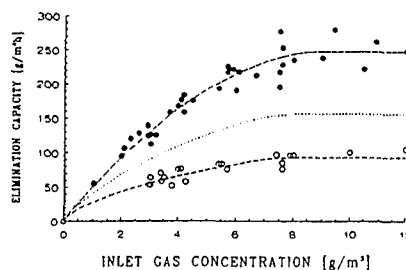


Fig. 15: The influence of the liquid flow rate on the elimination capacity in a biological trickling filter; $u_L = 1.8$ m/h (O, dotted line); $u_L = 3.6$ m/h (O, solid line); $u_L = 7.2$ m/h (O, dashed line).

Finally, some remarks should be made about the risk of clogging of the filter bed. This phenomenon is well-known from waste water treatment plants, where low rate trickling filters are applied, using dumped packings with

characteristic sizes of 40-75 mm [34].

If the average organic load exceeds certain limits (generally 0.08-1 kg/m³ d at hydraulic loads of 0.04-1.6 m/h), serious clogging may result from the extensive growth of biomass and the retention of suspended solids from the waste water.

At own experiments on the elimination of 1,2-dichloroethane and toluene from waste gases in a trickling filter, clogging was also observed. Within a few weeks, a packed bed of 1/2" ceramic novalox saddles was fully grown with biomass. However, for the elimination of dichloromethane such a packed bed proved to be stable for considerably longer periods of time (years). This different behaviour was suspected to be caused by the morphology of biofilms formed by the different microbial strain applied. In the case of toluene and 1,2-dichloroethane the formation of a network of long films and filaments between the packing elements was observed. However, the biofilms of *Hyphomicrobium* in the degradation of dichloromethane were characterized by an aggregation of small flocs.

It may thus be concluded that also the morphology of a biofilm may strongly determine the successful application of the trickling filter.

The number of full-scale applications of a biological trickling filter for the purification of waste gases is very limited as far as the authors know. However, from laboratory scale investigations [32] it may be concluded that this technique is very promising and offers real prospects for a practical application.

Bioscrubbers.

As indicated in Table III, the common aspect of trickling filters and bioscrubbers is the mobility of the liquid phase. However, in a biotrickling filter the biomass is immobilized on a carrier material, while in a bioscrubber (Fig. 16) the biomass is

freely suspended in the liquid phase.

A bioscrubber normally consists of a scrubber section, in which the mass transfer between gas- and liquid phase takes place. This section may be a packed bed, similar to a trickling filter, but also different designs have been presented [21,35,36].

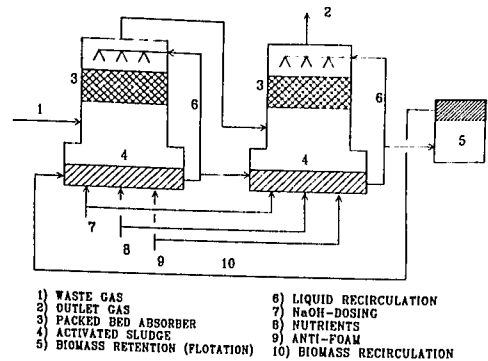


Fig. 16: The experimental set-up of a bioscrubber.

Regeneration of the liquid phase takes place in a regeneration tank by the suspended microorganisms. An additional oxygen supply in this compartment may be necessary, if the concentration of the substrates is so high, that the liquid itself does not contain enough oxygen for a complete degradation.

After the regeneration, the water is returned to the scrubber section. In order to increase the efficiency, biomass is also often recirculated to the scrubber compartment, where the mass transfer rate is increased by the simultaneous biological reaction. For example, the application of biomass suspensions containing 0,2 - 1 kgTSS/m³ (TSS= Total Suspended Solids) are reported for a venturi-scrubber [36],

while concentrations up to 60 kgTSS/m³ have been used in a packed bed absorber [37]. The latter application was tested on laboratory scale only. Clogging of the packed bed scrubber may be achieved by the application of a high liquid velocity (> 20 m/h) or specific packing elements [38].

In a bioscrubber a superficial gas flow rate of 0.5 - 1 m/s is normally applied. Degrees of conversion of over 90% can be reached in a bioscrubber for compounds with a relatively low value of the Henry-coefficient (< ca. 50 [Pa m³/mol]). At much higher values of the Henry-coefficient the required liquid flow rate, hence the energy consumption for liquid recirculation becomes too high [39].

This problem can partly be solved by allowing the biological degradation to take place in the scrubber section, either by suspended biomass, or by immobilized biomass as done in a trickling filter.

Also another solution for this problem has recently been presented. This concerns the addition of a high boiling organic solvent to the liquid phase [39,40]. For a successful operation, the organic solvent must have a very low water solvability, a very low Henry-coefficient of the compound to be absorbed and a high boiling point, thus a low partial pressure at operating conditions. Besides, it must neither be toxic for the microorganisms nor biodegradable. Organic fluids applied for this purpose mostly are silicone fluids or phthalates. A high mass transfer rate can be realized in the scrubber compartment by the high absorption capacity of the solvent. In this way both the reactor and the energy consumption can be reduced. The substrate concentration in the organic phase may be 100 to 1000 times higher than those in the aqueous phase. Thus in the regeneration compartment the compounds, which

have mainly been absorbed in the organic phase, are transferred to the aqueous phase where the microbial degradation takes place. Due to the buffering capacity of the organic phase, high substrate concentrations in the aqueous phase, hence toxic effects, can be avoided [40].

For bioscrubbers a small number of full scale applications are known, hence a comparison with e.g. trickling filter is still quite difficult.

Environmental aspects of biofiltration:

The emission of microorganisms.

As mentioned earlier, nowadays a clear trend exists to apply biofiltration on a much broader scale in different branches of industry and the interest in this technique is rapidly increasing. This necessitates the evaluation of possible risks of working with biologically active materials, and the consequent establishment of guidelines. Particularly in the food processing, the pharmaceutical- and the fermentation industry, some fear exists for an increased contamination of raw materials and products with undesirable microorganisms during handling processes, due to a significantly high microbial emission from a nearby installed biofilter.

Until today little is known on the subject of contamination of biologically treated air by microorganisms. Data dealing with the concentration of micro-organisms in an effluent gas from a biological system are very limited. However, in recent years a few investigations have been carried out on the subject [41,42] determining the number of microorganisms in effluent gases from full-scale as well as laboratory scaled biological waste gas purification systems. From the evaluation of the

results a number of important conclusions have been drawn.

In general it has been found that the number of microbial germs (mainly bacteria and fungi to a smaller extent) in the effluent gas of different full-scale biofilters varies from 10^3 to 10^4 m^{-3} , which is of the same order of magnitude as the numbers encountered in indoor air and slightly higher than encountered in open air.

On the basis of experimental investigations it was also found that a biofilter considerably reduces the concentration of microorganisms of waste gases contaminated with a high number of germs. Moreover, from laboratory experiments it followed, that the effect of the gas velocity on the discharge process of microorganisms from biofilters is due to a simultaneous process of capture (Fig. 17) and emission (Fig. 18).

A model, including these phenomena, has been developed, which is able to describe the experimental data rather well.

It can be concluded that the risks of any microbial contamination are generally not increased by the installation of a biological waste gas purification system on the location concerned.

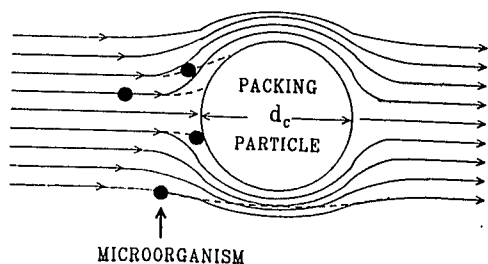


Fig. 17: Capture of microorganisms in a biofilter is mainly caused by inertial deposition.

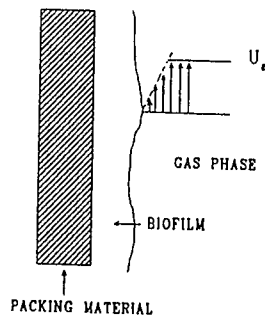


Fig. 18: The biolayer surrounding a compost particle is thought to be an exhaustible source of microorganisms.

Summary and conclusions.

From the above-presented overview on biological waste gas purification systems, it will be clear that as far as biofilters are concerned, this system can be considered as a state-of-art technology. It nowadays is an important alternative to many other physical- and chemical methods of waste gas purification, being safely to operate at relatively low costs. Concerning the bioscrubbers and the biological trickling filters, these systems do provide good prospects for the near future in the area where the biofilter application is limited. In specific cases, e.g. the removal of dichloromethane, the biological trickling filter has proved to operate successfully, while good results have also been reached for different compounds using a bioscrubber.

However, much research is still needed in order to understand and optimize the processes from a microbiological- as well as from an engineering point of view.

LIST OF SYMBOLS

- α : ratio of the concentration at the biofilm interface and the Monod constant
- a_w : specific wetted area [m^2 / m^3]
- C_{g0} : inlet gas phase concentration [g/m^3]
- C_{g1} : concentration at interface biofilm - gas phase [g/m^3]
- δ : biofilm thickness [m]
- D : effective diffusion coefficient in biofilm [m^2 / s]
- E_{act} : activation energy for the biological reaction [kJ/mol]
- EC: elimination capacity, or the amount of substrate degraded per unit of reactor volume and time [$g/m^3 \cdot h$]
- H : height of a filter bed [m]
- K_o : volumetric reaction rate in biofilm [$g/m^3 \cdot reactor \cdot h$]
- K_s : Monod constant [g/m^3]
- μ_{max} : maximum growth rate [day^{-1}]
- T : absolute temperature [K]
- u_g : superficial gas velocity [m/h]
- u_l : superficial liquid velocity [m/h]
- $\phi_o = \delta \cdot \left[\frac{K_o}{D \cdot C_{g1}} \right]^{1/2} =$ Zeroth order Thiele number
- ω : gas load = u_g

REFERENCES

- [1] Bach H., Gesundheits-Ingenieur, 46 38 (1923) 370-376.
- [2] Maurer P.G., BMFT Forschungsbericht T (1979) 79-114.
- [3] Jäger B. and Jäger J., Müll Abfall 5 (1978) 48.
- [4] Lith C., Clairtech b.v. Utrecht, The Netherlands, pers. communications.
- [5] Prüss M., Blunk H., German patent nr 710954 (1941) Appl. 1934.
- [6] Hartmans S. et al., J.Gen.Microbiol. 132 (1986) 1139-1142.
- [7] Brunner W. et al., Appl. Environ. Microb. 40 (1980) 950-958.
- [8] Scholtz R. et al., J. Bacteriol. 170 (1988) 5698-5704.
- [9] Janssen D. et al., Appl. Environ. Microb. 163 (1985) 635-639.
- [10] Hartmans S. et al., Biotechnol. Lett. (1985) 383-388.
- [11] Wijngaard v.d. A. et al., J. Gen. Microb. 135 (1989) 2199-2208.
- [12] Reineke W. et al., Appl. Environ. Microbiol. 47 (1984) 395-402.
- [13] Oldenhuis R. et al., Appl. Microbiol. Biotechnol. 30 (1989) 211-217.
- [14] Bont J. de et al., Appl. Environ. Microbiol. 52 (1986) 677-680.
- [15] Schraa G. et al., Appl. Environ. Microbiol. 52 (1986) 1374-1381.
- [16] Cooney L., in Biotechnology, Rehm H. Reed G. eds. VCH-Verlagsgesellschaft Weinheim vol 1, chap. 2 (1981)
- [17] Beer D. de, PhD Thesis Univ. of Amsterdam, Amsterdam The Netherlands (1990)
- [18] Harris N.P. et al, Water Reserch 10 (1976) 935-943
- [19] Don J., VDI-Berichte 561 (1986) 63-73
- [20] Ehtner E., VDI-Berichte 735 (1989) 191-214.
- [21] VDI-Berichte 735, VDI-Colloquium Biologische Gasreinigung, Köln 23-24 May (1989)
- [22] Ottengraf S.P.P., Oever A. vd., Biotechnol.&Bioeng. XIX (1986) 1411-1417.
- [23] Bardtke D., Proc. Int. Meet. Biol. Treatm. Ind. Waste Gases, Heidelberg 24-26 March (1987)
- [24] DECHEMA, Proc. Int. Meet. Biol. Treatm. Ind. Waste Gases, Heidelberg 24-26 March (1987)
- [25] Oude Luttikhuis H., VDI-Berichte 735 (1989) 341-348
- [26] Ottengraf S.P.P., in Biotechnology, Rehm H. Reed G. eds. VCH Verlagsgesellschaft Weinheim vol 8, chap. 12 (1981)
- [27] Ottengraf S.P.P. et al., Bioproc. eng. 1 (1986) 61-69
- [28] Hereth H., Proc. Int. Meet. Biol. Treatm. Ind. Waste Gases, DECHEMA, Heidelberg March 24-26 (1987).
- [29] Ljebe H., VDI-Berichte 735 (1989) 215-231
- [30] Koch W., VDI-Berichte 735 (1989) 349-355
- [31] Maier G., VDI-Berichte 735 (1989) 285-292
- [32] Diks R.M.M., Ottengraf S.P.P., part I-II Bioproc. Eng. 3 (1991), 390-399
- [33] Perry R.H. et al., Chem. Eng. Handbook, 6th ed. (1984) McGraw-Hill.
- [34] Bishop P., Kluener N., in Biotechnology, Rehm H. Reed G. eds. VCH-Verlagsgesellschaft Weinheim vol 8, chap. 3 (1981)
- [35] Kohler H., Fortschritt-Berichte VDI-Zeitschr. Reihe 15, 22 (1982).
- [36] Schmidt F., European patent nr 0-133-222, (1986).
- [37] Beck-Gasche B., Fortschritt-Berichte VDI-Zeitschr. Reihe 15, 68 (1989).
- [38] Schlippert E., VDI-Berichte 735 (1989) 77-88.
- [39] Schlippert E., VDI-Berichte 735 (1989) 161-177.
- [40] Lebeault J.M., Abstr. Forum Appl. Biotechnol. Gent September (1990) 20.
- [41] Ottengraf S.P.P., Konings J.H.G., 1991 To be published.
- [42] Klages S. et al., Thesis Univ. Stuttgart March 1987.

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