Bioprocess Biosyst Eng (2003) 26: 69–74
DOI 10.1007/s00449-003-0334-0

ORIGINAL PAPER

I. Vinage · P. Rudolf von Rohr

Biological waste gas treatment with a modified rotating biological contactor. I. Control of biofilm growth and long-term performance

Received: 28 May 2003 / Accepted: 4 July 2003 / Published online: 16 October 2003

Abstract In this work, we introduce a modified rotating biological contactor (RBC) system and demonstrate its feasibility by applying the newly devised process to the biological treatment of artificial waste gas. In the proposed system, the waste gas is introduced to the bioreactor in the spacings between the rotating discs through a hollow shaft, thus allowing for intimate gas–liquid contact. A 91-l modified RBC containing 20 biofilm support discs 40 cm in diameter was used in the experiments. Toluene was used as the model pollutant, and the system was operated under standard operating conditions for more than one year in order to investigate its long-term performance and assess its ability to control the growth of the biofilm. It was demonstrated that the proposed system allows to efficiently control the growth of the biofilm, thus overcoming the clogging problem inherent in most conventional methods for the biological treatment of waste gas. Moreover, the system was shown to exhibit stationary long-term performance for a period of more than one year, hence indicating its feasibility for industrial application.

Keywords Rotating biological contactor · Biofilm · Shear stress · Toluene

Introduction

Biological waste gas treatment is an attractive and environmentally friendly alternative to physico-chemical methods. However, the operational problems of conventional biological waste gas treatment facilities, namely the difficult control of operating parameters such as pH, temperature, humidity and nutrient supply in biofilters or the clogging of biotrickling filters due to unlimited biofilm growth, are well known and have not yet been successfully resolved. Moreover, biofilters are not suitable for the treatment of contaminants yielding acidic metabolites during the degradation process, as in the degradation of chlorinated hydrocarbons yielding hydrochloric acid for example, because the pH cannot be efficiently controlled in these systems. Furthermore, biofilter beds are often subject to compacting with time, or to clogging due to particulate matter. Sabo et al. [1] have proposed a rotor-biofilter, which showed no compacting and clogging problems. However, the regulation problem inherent in biofilters remains unsolved. Accurate control of process parameters is more easily achieved in biotrickling filters. However, their industrial application is hindered by the difficulty of operating biotrickling filters in a stable manner over prolonged periods of time. For example, it has been shown experimentally that unlimited biofilm growth may lead to an increase in the pressure drop and finally to complete clogging of the reactor [2, 3]. Clogging was usually observed at high inlet concentrations of compounds which are easily biodegraded. Several approaches to preventing clogging of biotrickling filters, either by removal of excess biomass or by limiting the biofilm growth, have been investigated on a laboratory scale. Reduction of the biomass accumulation rate can be achieved by a reduction of the microbial growth rate by nutrient limitation [2], by using nitrate instead of ammonium as the nitrogen source, or by maintaining growth-limiting concentrations of NaCl in the recycling liquid [4]. However, these methods have the common drawback that the specific microbial activity is reduced, hence reducing the pollutant elimination capacity. The use of protozoan predation has also been investigated [5]. Protozoan predation does not affect the activity of the biomass, but microbial growth is not efficiently controlled in this method. Hence, clogging is only delayed, but not prevented. Removal of excess biomass may be achieved

I. Vinage · P. Rudolf von Rohr (✉)
Institute of Process Engineering, ETH Zurich, Sonneggstrasse 3, 8092 Zurich, Switzerland
E-mail: vonrohr@ivuk.mavt.ethz.ch
Tel.: +41-1-632 2488
Fax: +41-1-631141

mechanically or chemically. Mechanical removal relies on biofilm detachment through high shear forces, and was achieved by backwashing [6]. This technique was proved to be effective, but required rather high liquid flow rates and an enlargement of the bed volume by approximately 40% so as to allow for expansion of the bed during fluidization if random packing material was used [5]. Moreover, the method produced large amounts of high biochemical oxygen demand (BOD) waste water, and most importantly, non-continuous operation of the system. Stirring the randomly packed material periodically in order to shear off excess biomass was shown to be effective on a laboratory scale, but may raise scale-up problems [7]. Chemical washing of the packed bed has been investigated by different research groups [8, 2]. Solutions of sodium hydroxide, sodium chloride, and hydrogen peroxide as well as mixtures thereof were used, resulting in partial or complete inhibition of the microbial activity and in the production of large quantities of waste water. Gai et al. [9] proposed a system consisting of rotating packing elements combined with special nozzles for liquid repartition intended to ensure uniform growth conditions for the biofilm. However, discontinuous operation was mandatory, as the system needed to be washed intermittently in order to prevent it from clogging.

Objectives

The objective of the current research project was to develop and characterize a new system where biofilm accumulation can be controlled without any additional intervention. This system should exhibit stationary long-term performance along with high degradation efficiency and minimum maintenance requirements. The proposed system is a modified form of the rotating biological contactor (RBC), which is widely used in the treatment of waste water. In this part of our work, we investigate the long-term performance of the modified RBC under standard operating conditions and assess its ability to efficiently control the growth of the biofilm. In this study, toluene was used as the model pollutant. Toluene is an organic solvent widely used in industry. It is generally considered as moderately water soluble and easily biodegradable.

RBC for waste water treatment

The RBC, which combines the advantages of low energy consumption and control of biofilm growth, is a well-known system for the treatment of municipal and industrial waste water [10]. In its simplest form, an RBC consists of a series of discs mounted on a horizontal shaft which is slowly rotated. The discs are typically made of polymer material such as PVC, polystyrene, or polyethylene, and partly immersed in a waste water stream. About 40% of the disc surface is immersed in the waste water, and micro-organisms grow on the discs to form a biofilm. The biofilm is submerged in the waste water flow and exposed to air alternately due to the rotation of the discs. During the periodic submersion, the biofilm is brought into contact with the organic pollutants solubilized in the waste water flow. While exposed to air, a film of waste water is carried along the biofilm, which absorbs oxygen from the air, thereby aerating the waste water. Hence, in this process, the micro-organisms are provided with a steady supply of both organic material and dissolved oxygen. Shear forces are exerted on the biofilm due to the rotation of the discs through the waste water. These shear forces strip the discs of excess biomass, and provide a sufficient degree of mixing to keep the sloughed biomass in suspension until it is carried out of the reactor tank with the bulk flow, thus eliminating the possibility of reactor clogging due to biomass accumulation. The rotation of the discs also humidifies the biofilm surface uniformly, and provides turbulence in the liquid phase . Moreover, the presence of a bulk liquid phase allows for accurate control of the main operating parameters such as pH, temperature and nutrient supply.

RBCs are often designed as a series of stages. The series arrangement results in a high degree of organic removal and denitrification, since micro-organisms that grow in a particular stage are adapted to the waste water of that particular stage. The residence time distribution approaches plug-flow in a multistage RBC, hence increasing the BOD removal.

RBC for waste gas treatment

It is worth noting that there is only limited experience with this type of reactor in waste gas treatment. In fact, a Canadian company (CMS Group Inc., Concord, ON) has applied conventional RBC technology to waste gas treatment. However, in their system, the polluted air is flowing parallel to the discs. Results about ammonia removal from waste gas with a conventional RBC have also been reported by Vis and Rinzema [11].

Preliminary experiments with a conventional four-stage RBC, kindly provided by CMS Group Inc., and using methylene chloride as model pollutant were carried out at our laboratories in 1997 [3, 12]. These measurements were continued over a period of 4 months. Degradation results obtained with this set-up showed that the system was very suitable for the degradation of VOCs from waste gas. Furthermore, the biofilm growth was successfully limited. However, it is worth noting at this point that degradation results were lower when compared with typical degradation data from biotric-kling filters. This is explained by considering the non-optimal contact between gas and biofilm in this system. In fact, the gas phase was introduced parallel to the discs above the water level. As a result of this experimental configuration, a significant part of the gas was never in contact with the liquid film on the discs.

Material and methods

Experimental set-up

In order to be degraded in any waste gas treatment system, the pollutant must be absorbed from the gas into the water phase. Hence, design considerations have explicitly addressed the optimization of gas–liquid mass transfer rates through intimate gas–liquid contact. In the novel, modified RBC system, the synthetic waste gas is introduced to the reactor through a hollow shaft, thus ensuring a uniform gas supply along the discs and optimal contact between gas and liquid phase. The principle of the modified RBC for waste gas treatment is shown in Fig. 1. It exploits the main advantages of waste water RBCs, i.e., control of biofilm growth ensuring stationary long-term performance, low maintenance requirements, and accurate control of process parameters.

The experimental investigations were carried out on a laboratory scale one-stage modified RBC. A process flow diagram of the experimental set-up is shown in Fig. 2.

The cylindrical reactor is made of polypropylene, and has an inner diameter of 0.5 m and a length of 0.465 m. It contains 20 polypropylene discs with a diameter of 0.4 m and a thickness of 5 mm. The spacing between the discs, which were roughened with a sand jet to facilitate good adhesion of the biofilm, was arbitrarily chosen to be 10 mm. The 20 discs are mounted on a hollow shaft made of stainless steel with outer and inner diameters of 42 mm and 36 mm, respectively. Twelve perforations with a diameter of 8 mm are placed along the circumference of the shaft, and 19 series of holes are drilled along the length of the shaft with a spacing of 15 mm between the centers of the holes to provide sufficient space to place the discs between two series of holes.

A sintered stainless steel tube (GKN Sinter Metals, Radevormwald, Germany) with inner and outer diameters of 24 and 30 mm, respectively, was placed inside the shaft to ensure a uniform supply of gas between the discs along the shaft. The rotation of the shaft is driven by a speed-regulated direct current electric motor (ITT, Angst + Pfister, Zurich, Switzerland), where transmission is accomplished through a toothed-belt, and speed is regulated by a speed controller (Typ DC 10-S, Angst + Pfister, Zurich, Switzerland). Three segmented discs were specially designed for the purpose of taking samples from different compartments of the reactor. These samples were then used for the measurement of biofilm thickness profiles and further biofilm analysis. The rotating discs in the reactor are immersed in the nutrients solution up to a radius of 0.0315 m, which corresponds to about 40% of the total disc surface. The nutrient solution is pumped through the system continuously at a constant flow rate.

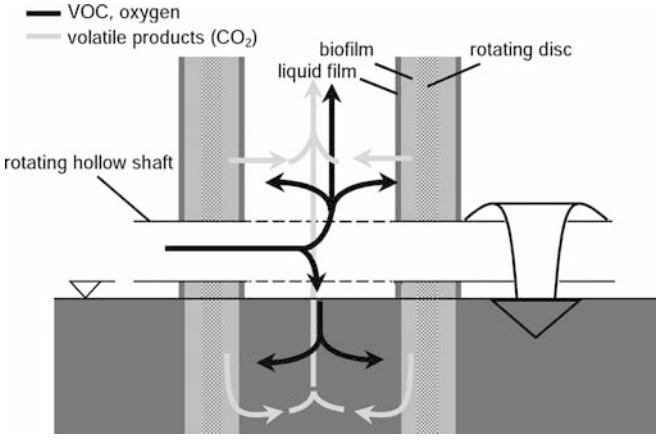

Fig. 1 Principle of the modified RBC for waste gas treatment

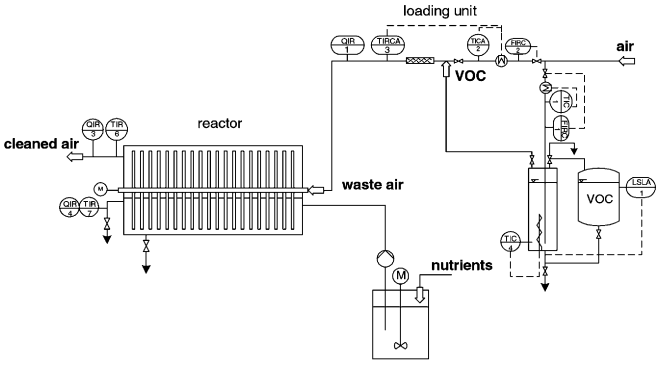

Fig. 2 Process flow diagram of the experimental set-up of a modified RBC

The operation principle of the loading unit can also be identified in Fig. 2. A stream of compressed air was passed through a microfilter (EAFM4000-F04, SMC Pneumatik, Weisslingen, Switzerland) in order to remove potential impurities. The air stream was then divided into two streams, i.e., the primary and secondary air streams, which were both controlled by individual mass flow controllers (Brooks, Venendaal, The Netherlands). The secondary stream was then bubbled into a temperature controlled column (76.1×600 mm) containing liquid toluene at 40°C. This column was connected to a larger tank (273×360 mm) containing about 10 l of liquid toluene, which was used to control the liquid level in the temperature controlled column. The temperature of the liquid phase in the toluene column was controlled by a heating cartridge (Elektrolux, Aarau, Switzerland) and a PID temperature controller (RR210, Elektrolux, Aarau, Switzerland). A level switch (KSR 701-L, Heinrich Kübler AG, Baar, Switzerland) was placed into the column and used to ensure that heating was only applied to the cartridge when it was completely covered with liquid toluene. Hence, the secondary air stream was loaded with toluene, and then diluted by remixing with the primary air stream in three consecutive static mixers (SMV-4 DN20, Sulzer Chemtech AG, Winterthur, Switzerland). The synthetic waste gas stream was brought to a temperature of about 30°C using a band heater and a PID temperature controller (MC 102, Thuba Engineering, Basel, Switzerland) prior to entering the RBC reactor. Tanks and piping were made of stainless steel and equipped with the necessary safety precautions.

The specifications of the modified RBC are again summarized in Table 1.

Bacteria and medium

The RBC was initially inoculated with a biomass suspension of *Pseudomonas putida* F1 and *Rhodococcus erythropolis* PWD1 cultivated in shake flasks. Fresh medium was continuously added to the reactor at a volumetric flow rate of approximately 5 l/day. The

Table 1 Specifications of the modified RBC

Number of discs	20
Disc diameter	0.4 m
Disc material	Polypropylene
Disc thickness	0.005 m
Discs spacing	0.01 m
Rotational speed	0–11 rpm
Disc immersion	40%
Shaft diameter	0.042 m
Reactor diameter	0.5 m
Reactor material	Polypropylene

liquid level in the reactor was adjusted by an overflow device. The nutrient solution was prepared with the following concentrations: 5.35 g/l NH_4Cl, 17.4 g/l K_2HPO_4, 0.764 g/l NTA in NaOH 0.6 M, 0.264 g/l $MgCl_2$, 0.284 g/l Na_2SO_4, 0.300 g/l $CaCl_2$, 0.012 g/l $(NH_4)6Mo_7O_{24}.4H_2O$, 2.46 ml/l HCl 37%, 8.16 mg/l ZnO, 1.08 g/l $FeCl_3$, 0.04 g/l $MnCl_2$, 3.4 mg/l $CuCl_2$, 9.52 mg/l $CoCl_2$, 1.24 mg/l H_3BO_3.

Results of 16S rDNA-sequencing showed that the microflora in the biofilm evolved to a mixed culture, which exhibits more robustness than a pure culture towards changes of the environmental conditions, such as, for example, higher loading. Some sequences matched the sequence of the initial strains very well. Other bacterial strains which can grow on toluene were also found and, according to the sequence, these might be *Nocardia* sp. and *Ochrobactrum* sp.

Gas phase analysis

The total concentrations of toluene, organic carbon, and carbon dioxide are registered on-line. Total organic carbon was measured using a flame ionization detector (FID VE7, J.U.M. Engineering, Karlsfeld, Germany), and the toluene concentration was determined on a gas chromatograph (Carlo Erba Strumentazione, Milan, Italy) equipped with a DB5 column (0.5 mm film, 0.32 mm ID, 30 m length). Helium was used as the carrier gas, and the sample was injected at a temperature of 100°C. The temperature of the column was maintained at 100°C, and detection occurred at 200°C with an FID detector. Continuous analysis of the gas phase was performed by using an eight-port external volume sample injector with a 500 ml loop (VICI, Houston, USA). The concentration of carbon dioxide was determined by IR spectroscopy (UNOR 610, Maihak, Hamburg, Germany).

Liquid phase analysis

The concentration of toluene in the liquid phase was determined off-line using HPLC (Seperation Module 2690, Waters AG, Rupperswil, Switzerland). The sample was collected in a glass vial with a PTFE-silicone sealing. Suspended cells were removed from the sample by centrifugation, and 10 µl of the solution were injected into the packed column (Nucleosil 100-C18, 3 mm-100 A°, 4.6×40 mm) using an auto-sampling device. Impurities were removed on a pre-column of the same type with a length of 10 mm. Methanol (0.7 ml/min) and water (0.3 ml/min) were used as eluents, and a UV detector (Photodiode Array Detector 996, Waters AG, Rupperswil, Switzerland) at 210 nm was used.

Biofilm thickness measurement

Biofilm thickness profiles along the radius of a disc were measured using a laser distance sensor based on the triangulation principle (LDS1-010, Raytec System AG, Chur, Switzerland). The method is rather similar to the method described by Okkerse et al. [13]. It is worth noting that the technique adapted is non-destructive and precise, and allows biofilm profiles to be measured over lengths of up to 14 cm in our particular application. The measurement principle of the laser triangulation method is shown in Fig. 3. A modulated laser beam generated by a semi-conductor laser diode is projected on the biofilm surface through a system of lenses. Another system of lenses is used to record the reflection of the laser beam on the biofilm surface by a high-resolution position sensitive detector. The position of the beam of light on the receptor is a function of the distance between the laser and the biofilm surface. The laser diode has a wavelength of 780 nm, a focusing diameter of 50 µm, an accuracy of 25 µm, and a measuring range of 10 mm. The distance between the laser and the surface under examination has to be between 50 and 60 mm, and the maximum sampling frequency is 500 kHz.

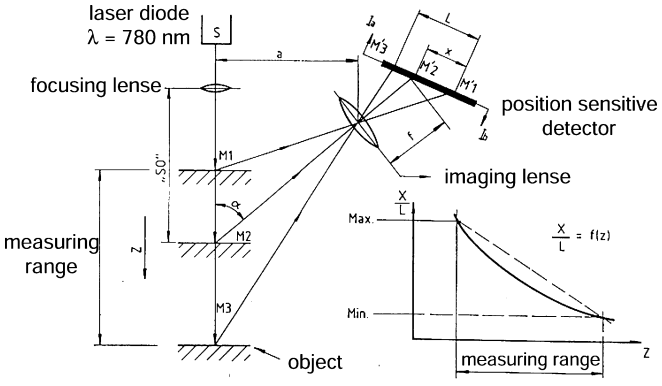

Fig. 3 Principle of the biofilm thickness measurements using a laser diode sensor utilizing the triangulation principle

As mentioned above, three discs were segmented into eight parts which can be taken out of the reactor individually. The segments and the measurement system were designed such as to leave the biofilm on both sides of the disc intact. The laser sensor is fixed on a linear-track guide equipped with a step motor (LF5, Heeb Elektro AG, Küsnacht, Switzerland). It is moved along the disc at a given velocity, and measures the biofilm thickness profile along the disc radius. In addition to the movement along the track guide, the sensor can be moved perpendicularly to the sample using a micropositioning table that can be adjusted manually to bring the sample into the laser measuring range. With this non-destructive method, the sample must be removed from the reactor for approximately 5 min only, while three consecutive measurements are carried out.

Experimental results

The performance of the modified RBC was studied during more than one year of continuous operations. The standard operating conditions were chosen as follows: gas flow rate, 0.96 m^3/h; toluene inlet concentration, 3.1 g/m^3; rotational speed of the discs, 6 rpm; and nutrients flow rate, 0.21 l/h. These standard operating conditions were always resumed after the experiments in other conditions were completed in order to return to the same regime of operation. The experiments at non-standard operating conditions were typically finished within one day, and were only initiated after the reactor performance had returned to steady state under standard operating conditions.

Long-term performance at standard conditions

Stationary long-term performance is required for any waste gas treatment system used on an industrial scale. This issue was investigated for the modified RBC system, and the results are summarized in Fig. 4, where load and elimination capacity at the standard conditions summarized above are shown as a function of time. It is worth noting that steady-state operation at optimal standard conditions was achieved approximately 100 days after inoculation of the reactor.

The system was operated at a relatively high toluene inlet concentration of 3.1 g/m^3 corresponding to an inlet

load of about 53 g toluene per cubic meter reactor and hour. This was chosen to show that the system is not subject to clogging even at high inlet loadings, as biofilm growth increases with increasing substrate loading [14].

It is also obvious from Fig. 4 that the elimination capacity remains approximately constant during the entire investigation. This result clearly indicates that the proposed reactor system is capable of providing the required stationary long-term performance.

Biofilm thickness

Clogging of the reactor needs to be avoided in order to guarantee the long-term stability of the waste gas treatment system. Clogging is most efficiently prevented if the thickness of the biofilm on the discs can be kept constant over a long period of time.

In fact, the performance of the modified RBC system was studied experimentally for more than one year, and the problem of clogging was not observed during this time. An example of the biofilm inside the reactor is shown in Fig. 5. The picture was taken 11 months after inoculation, and it can readily be observed that the biofilm is rather thin even after almost one year of continuous operation. These results clearly indicate the robustness of the process, and proves that clogging is efficiently avoided in the modified RBC system.

Profiles of the biofilm thickness along the radius of the three segmented discs described above were measured at regular intervals using a laser triangulation method which was discussed in detail in Sect. 2.5. An example is shown in Fig. 6, where the biofilm thickness profile 133 days after the start-up of the reactor is shown. In fact, the disc segment for which the measurement was carried out is shown in Fig. 7, and the

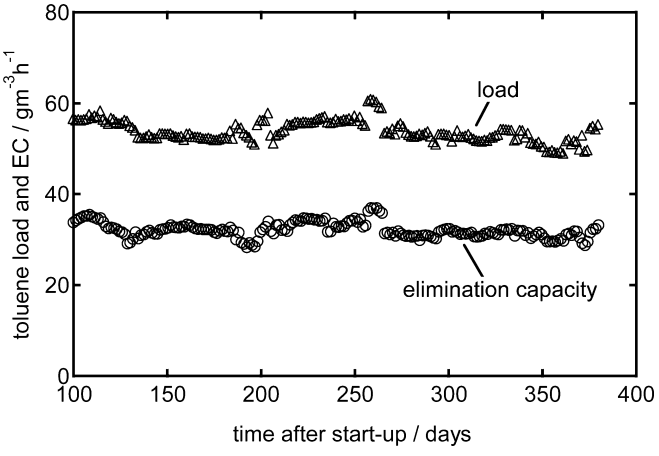

Fig. 4 Toluene load (*circles*) and elimination capacity (*triangles*) vs time after start-up for the standard operating conditions: gas flow rate = 0.96 g/m^{3}h, toluene inlet concentration = 3.1 g/m^3, disc rotational speed = 6 rpm

Fig. 5 Biofilm grown on the discs 11 months after inoculation

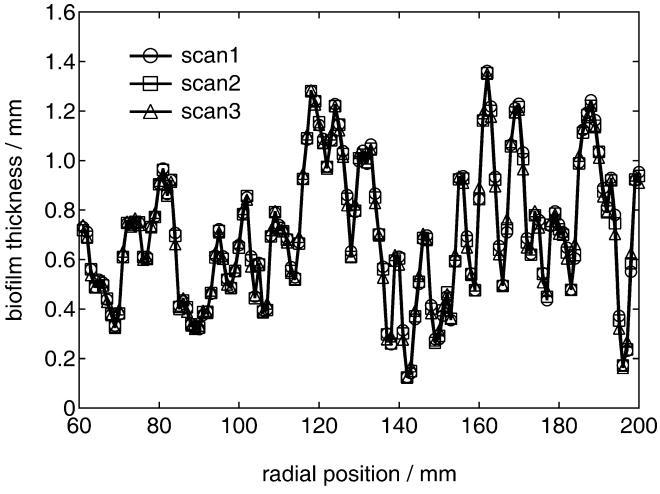

Fig. 6 Biofilm thickness measurements 133 days after start-up. The thickness profile was measured along the *dashed line* in Fig. 7

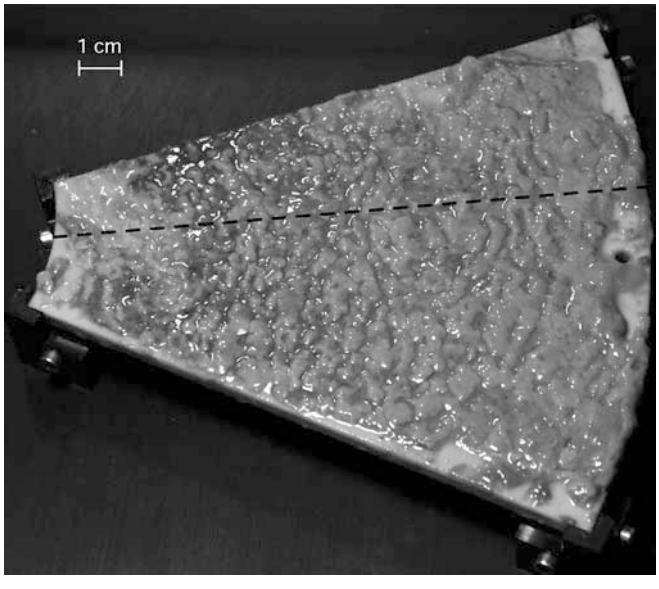

Fig. 7 Biofilm growing on disc 133 days after inoculation

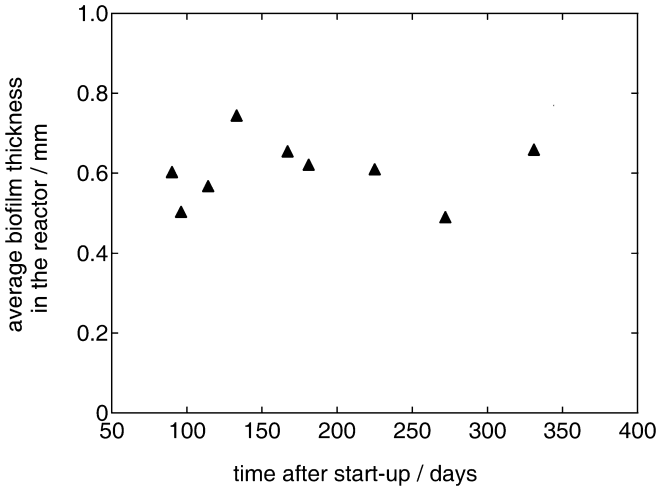

Fig. 8 Average biofilm thickness in the modified RBC vs time after start-up

thickness profile was measured along the dotted line in the same figure.

Three successive scans of the biofilm thickness were carried out, and the very high reproducibility is readily observed in Fig. 6. Furthermore, it can be concluded from the thickness profile that the biofilm comprises a base film and a surface film which consists of protrusions on the surface of the base film. In fact, these two compartments of the biofilm may also be observed in Fig. 7, where the base film appears dark, and the surface appears lighter.

In Fig. 6, the maximum biofilm thickness never exceeds 1.4 mm. Since the spacing between two discs is 10 mm, this result clearly shows that the problem of clogging is successfully prevented in the proposed system. An average biofilm of about 700 μm was calculated for the example shown in the figure. The mean biofilm thickness of the entire reactor was calculated by averaging the profiles obtained for the eight segments on the three segmented discs, i.e., 24 biofilm thickness profiles. The evolution of the mean biofilm thickness with time after start-up is shown in Fig. 8. It can be observed that it remains nearly constant at around 600 μm, hence indicating that the biofilm growth is efficiently controlled in this system.

Conclusions

In this work, we have introduced a novel reactor system for the biological treatment of waste gas, i.e., the modified RBC. In the proposed system, the synthetic waste gas is introduced to the reactor in the spacings between the rotating discs through a hollow shaft, thus allowing for intimate gas–liquid contact. Intimate contact of the liquid and gas phases is mandatory in waste gas treatment, as the pollutant has to be absorbed in the liquid phase prior to the biological degradation reaction occurring in the biofilm. The experimental characterization of the modified RBC was carried out in a 91-l laboratory scale set-up containing 20 biofilm support discs with a diameter of 40 cm, and toluene was chosen as the model pollutant. The unit was operated continuously over a period of more than one year in order to assess its long-term performance under standard operating conditions. Stationary long-term performance along with minimum maintenance requirements are needed for any industrial application of a waste gas treatment system, and the use of conventional systems such as biofilters or biotrickling filters is often hindered by the difficulty in controlling the process parameters accurately, and the risk of clogging due to unlimited biofilm growth. The newly devised system is able to overcome this limitation and efficiently control biofilm growth. In fact, this was successfully demonstrated by operating the unit for more than one year under standard operating conditions, and at a rather high inlet concentration of toluene. Measurements of the biofilm thickness revealed that the average thickness remained nearly constant at about 600 μm during the experimental investigation.

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