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Laboratory biotrickling filter for the removal of odorous volatile compounds from air

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ABSTRACT:

The paper presents the results of experimental investigations of the deodorization process of air contaminated with toluene vapors in a trickled-bed biofilter (i.e. biotrickling filter). A laboratory set-up for biofiltration process was proposed, based on a two-section bioreactor filled with Raschig ceramic rings. The biofilter bed was inoculated with microorganisms from the *Candida* species. Aqueous solution of mineral salts was used as the trickling liquid. The effectiveness of toluene removal from its mixture with air was investigated and compared using a trickling liquid without addition and with the addition of sodium dodecyl sulphate. It has been found that the addition of a surfactant increases the degree of toluene removal in the biofiltration process. It was pointed out that the proposed set-up can be successfully applied during laboratory investigations of the biofiltration process, ensuring system stability and reducing technical problems related to excessive biomass growth in the biofilter bed.

Laboratoryjny biofiltr strużkowy do oczyszczania powietrza z lotnych związków organicznych o charakterze odorowym

Słowa kluczowe: biofiltracja, biofiltr strużkowy, złoże zraszane, toluen, stopień usunięcia

STRESZCZENIE:

W pracy przedstawiono wyniki badań doświadczalnych procesu dezodoryzacji powietrza zanieczyszczonego parami toluenu w biofiltrze ze złożem zraszanym (tj. w biofiltrze strużkowym). Zaproponowano układ badawczy do procesu biofiltracji, oparty na bioreaktorze dwusekcyjnym, wypełnionym ceramicznymi pierścieniami Raschiga. Złoże biofiltra zaszczepiono drobnoustrojami z rodzaju *Candida*. Jako ciecz zraszającą stosowano wodny roztwór soli mineralnych. Zbadano i porównano skuteczność usuwania toluenu z mieszaniny z powietrzem, stosując ciecz zraszającą bez dodatku i z dodatkiem dodecylosiarczanu sodowego. Stwierdzono, że dodatek związku powierzchniowo czynnego powoduje wzrost stopnia usunięcia toluenu w procesie biofiltracji. Wskazano, że proponowane stanowisko może być z powodzeniem stosowane podczas laboratoryjnych badań procesu biofiltracji, zapewniając stabilność pracy układu oraz ograniczenie problemów technicznych związanych z nadmiernym wzrostem biomasy w złożu biofiltra.

1. INTRODUCTION

Progress in economic and industrial development results in increased emissions of pollutants to ambient air, especially from industrial facilities [1, 2]. Among many different chemical compounds, the odors (odorous compounds) play a significant role. The odors are volatile compounds sensed by animals and humans via olfactory receptors at very low concentrations and identified by brain as unpleasant sensations. Compounds characterized by an unpleasant odor include [3] inorganic (e.g. hydrogen sulphide, ammonia, sulphur dioxide, nitrogen oxide, hydrogen fluoride and hydrogen arsenide) and organic compounds (e.g. thiols, sulphides and disulphides, amines, carboxylic acids, aldehydes, ketones, aromatic hydrocarbons). Deodorization of gases polluted with odorous compounds may be realized either by the removal of malodorous pollutants or by transformation of malodorous pollutants into odorless chemical compounds or the compounds characterized by high odor threshold. Another deodorization method includes introduction of additives changing the character of odor or decreasing its intensity of hedonic tone (i.e. masking compounds). Selection of the most effective deodorization method is difficult and depends on many factors including emission intensity, total content of pollutants, odorous character of emitted gases. There are four main groups of techniques used to reduce odorants in the air: combustion, adsorption, absorption and biological methods.

The use of biological methods for air purification has been known for over 60 years. Devices called biofilters are used for this purpose, among others, for the removal of odorous compounds from gases, e.g. in food processing plants, sewage treatment plants and from landfill [4]. The process of biofiltration consists in the decomposition of contaminants by bacteria or other microorganisms that colonize the porous packing bed of the biofilter. The mechanism of the process involves the diffusion of pollutants from the gas phase to the biofilm, covering the surface of the packing elements. The compounds absorbed in the biofilm are being biodegraded and the cleaned gas leaves the biofilter.

There are three main groups of apparatus designed for biofiltration i.e. bioscrubbers, biofilters and biotrickling filters (Figure 1) [5-8]. In bioscrubbers, the components from the gas phase penetrate into the counter-currently flowing liquid enriched with activated sludge. The liquid circulates in a closed system and it is periodically regenerated and aerated. In the case of a conventional biofilter, the contaminated gas is humidified in a separate chamber and then flows to a bioreactor packed with a bed made from natural materials. Microorganisms that are capable of degrading odorants develop in the biofilter. In a biotrickling filter (BTF), absorption and decomposition of pollutants take place in one apparatus, the packing of which is trickled with liquid enriched with nutrients for microorganisms. Beside the fact that the apparatus for biotrick-



Figure 1 Schematic diagrams of the most popular apparatus for biofiltration: a – conventional biofilter, b – bioscrubber, c – biotrickling filter

ling filtration is relatively complex as compared to conventional biofilters or bioscrubbers, it has a number of advantages over typical biofilters, including: greater process stability, pH and temperature regulation of the trickling liquid, lower flow resistance and reduced space requirements [7]. The mentioned features are conducive to the ongoing development of this group of bioreactors in biofiltration processes [9, 10]. Additionally, ease of process control together with possibility of modification of trickling liquid composition makes biotrickling filters an advantageous bioreactor configuration for the removal of hydrophobic compounds. A brief comparison of abovementioned bioreactor types is presented in Table 1 [5, 6].

Among the research areas in the field of biotrickling filtration, the removal of hydrophobic volatile organic compounds is an urgent topic [5, 11]. This is because hydrophilic compounds are easily biodegraded in biological systems, while it is not true for hydrophobic compounds, i.e. due to low affinity as well as high diffusion resistance in aqueous phase. Toluene, a popular organic solvent, characterized with Henry's constant of 0.0015 mol m⁻³ Pa⁻¹, is a representative hydrophobic volatile organic compound (VOC) [5]. Toluene is not easily removed from air in biological systems. As toluene is present in waste gases emitted from various industrial facilities (e.g. from municipal landfill sites), introducing new solutions to improve the efficiency of its removal is of importance. The selected research on the removal of toluene in biotrickling filters is given in Table 2.

The aim of the investigations is to verify the usefulness as well as to test the performance of a laboratory biotrickling filter designed in the Department of Process Engineering and Chemical Technology, Chemical Faculty, Gdańsk University of Technology. The investigations were performed in a biotrickling filter packed with ceramic elements and toluene was selected as a target compound to be removed from its mixture with air. Moreover, the effect of trickling liquid modification with a surface active substance on the toluene removal efficiency is evaluated.

Type of bioreactor	Advantages	Disadvantages
Conventional biofilter	Low investment and operational costs; ease of operation and maintenance; high deodorization efficiency	Large surface area requirements; limited durability of natural packing materials; process difficult to control
Bioscrubber	Low pressure drop; low space requirements; operational stability	Effective for hydrophilic compounds only; complex operation and maintenance; secondary pollution generation
Biotrickling filter	Low operating costs; ease of process control; high durability of a packing material	Complex construction; problems with biomass overgrowth; generation of waste trickling liquid streams

Table 1 Advantages and disadvantages of biological reactors for gas deodorization

Table 2 Selected research or	toluene biot	rickling filtration
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Reference	[12]	[13]	[14]
Packing material	Polyurethane foam	Kaldnes K1 rings	Bundle of plastic tubes glued together
Trickling liquid	Sulphate-free mineral salt medium	Mineral salt medium	Water
BTF height, m	1.00	0.60	1.00
BTF diameter, m	0.08	0.07	1.95
Empty bed residence time, s	50	180	-
Removal efficiency, %	70	50	95

2. MATERIALS AND METHODS

2.1 Experimental set-up

Investigations were carried out in a two-section biotrickling filter made of plexi glass, constructed according to the general scheme presented in Figure 2. A BTF was packed with inert ceramic Raschig rings. The filter packing was inoculated with *Candida sp.* environmental isolates, purchased from the Department of Molecular Biotechnology and Microbiology, Faculty of Chemistry, Gdańsk University of Technology. The inoculation



Figure 2 Schematic diagram of a laboratory biotrickling filter; P – manometer

was performed by circulating a nutrient solution, containing Candida sp., for 7 days prior to the start-up of toluene biofiltration. A mixture of air and toluene ($C_{toluene} = 4 \text{ ppm v/v}$) was supplied from the gas mixture generator to the bottom of the biotrickling filter, according to the methodology previously described [15]. From the BTF top, a liquid mineral salt medium (MSM; aqueous solution of Na, HPO, × 2H, O, KH, PO, NaCl, NH, Cl) was trickled over the filter packing. When stated, sodium dodecyl sulphate (SDS) was added to the trickling liquid (initial concentration of SDS was 36 mg dm⁻³). The trickling liquid was circulating in a closed system with periodical regeneration (once per 5 working days). Gas samples were taken once a day from both inlet and outlet streams. The concentration of toluene was determined using Varian CP3800 gas chromatograph equipped with flame ionization detector. A HP-5MS column (30 m \times 0.250 mm ID \times 0.25 μ m) and nitrogen as carrier gas were used (0.7 mL/min). Oven temperature was set at 160°C. The calibration was performed using an external standard method. Details of the experimental conditions are given in Table 3, while the photograph of a laboratory BTF is shown in Figure 3.

Dimensions of a single section of BTF	internal diameter: 0.08 m; height: 0.35 m	
Gas flow rate	800 ml min ⁻¹	
EBRT	210 s	
Trickling liquid flow rate	0.001 dm³ s ⁻¹	
Trickling frequency	1 minute every 30 minutes	
Packing elements	Ceramic Raschig rings (6 × 1.5 mm); porosity: 0.52	

Table 3 Selected experimental parameters



Figure 3 Laboratory biotrickling filter with developed biofilm

The efficiency of toluene removal was evaluated using removal efficiency factor:

$$RE = ((C_{in} - C_{out})/C_{in}) \times 100\%$$
(1)

where C_{in} and C_{out} stand for inlet and outlet concentrations of toluene.

Empty bed residence time was calculated as follows:

$$EBRT = V / Q$$
 (2)

where V is volume of the filter bed and Q is the volumetric gas flow rate.

3. RESULTS AND DISCUSSION

The performance of laboratory bioreactors for air deodorization is typically evaluated on the basis of changes in removal efficiency of target compound with process time. Figure 4 presents such results for two investigated systems used for the removal of toluene from air [16, 17]. In one system, mixture of air and toluene was treated in a biotrickling filter using mineral salt medium as trickling liquid (MSM). During the first days of biofiltration process (up to five days from the process start-up), the removal of toluene is fluctuating and is rather law (about 30%). Then, the removal of toluene slightly increases, reaching about 50% after 10 days from the process startup. The mentioned fluctuations as well as low removal efficiency may be attributed to accommodation period for microbes inhabiting the biofilter bed as well as the development of a biofilm at the elements of filter packing. Because toluene is a representative of hydrophobic VOC, its affinity towards MSM aqueous phase is rather low and thus the mass transfer from gaseous to liquid phase is limited. This is why the removal efficiency is about 50% when steady-state conditions are considered for the investigated time of the process (from 10th to 20th day of the process).



Figure 4 Changes of removal efficiency with biofiltration time; MSM – mineral salt medium; SDS – sodium dodecyl sulphate

It is known that modifying the composition of a trickling liquid with a surface active substance may enhance the removal of hydrophobic VOCs during biofiltration [11]. Thus, the removal efficiency of toluene obtained in the second examined system (MSM + SDS) is higher than in the one formerly discussed (MSM) (Fig. 4). During the first twenty days of biotrickling filtration, RE increases steadily up to about 75% and the increase in RE values coincides with the development of a biofilm at the elements of a filter bed. Then, until the end of investigations (fixed time of the process i.e. 40 days of biofiltration), the removal efficiency is kept constant (RE = 75-80%), representing the steady-state conditions. Obtained results for both investigated systems are in accordance with available literature data. Based on data presented in Table 2, it may be stated that toluene is removed for air streams with an average removal efficiency of about 70%, when water with mineral salts is used as a trickling liquid. Results of investigations presented in Figure 4 shows that values of RE are about 50% and 80%, when MSM or MSM supplemented with SDS is used as a trickling liquid, respectively. Thus, the application of a surface active substance enhances the removal of hydrophobic VOCs e.g. toluene in a biotrickling filter. Similar results were obtained by Cheng et al. [18] for the removal of hexane from air.

It is worth noting that beside the removal efficiency, ease of maintenance as well as the resulting process stability are important parameters describing the laboratory set-up for biotrickling filtration. Considering its operation for the investigated time period, the constructed biofilter setup has proven to be useful for both short- and mid-term investigations. Biofilter packing material (i.e. ceramic Raschig rings) after the process completion may easily be removed from the filter column and, after cleaning and sterilizing, may be used in the next process. During the investigations, no problems with the bed clogging were detected (pressure drop in the range 60-80 mm $H_{2}O$). It is a result of at least two specific features of the investigated system: trickling frequency was selected accurately so as to limit the biomass overgrowth and the two-section biofilter configuration reduced the possibility of a bed clogging when compared to similar volume of single-stage packed bed. Above given features of the proposed laboratory biotrickling filter makes it useful both for conducting research as well as didactic purposes.

4. CONCLUSIONS

The proposed laboratory biotrickling filter enabled efficient removal of toluene from its mixture with air. Removal efficiency reached about 75-80% after 20 days from the process start-up and remained constant until the end of the investigations (40 days), when trickling liquid (MSM) was supplemented with surface active substance (SDS). The proposed construction pattern of BTF revealed its suitability for short as well as medium time-length experiments. Additionally, the durability of the packing and the stability of the formed biofilm at the surface of packing elements were confirmed with no detected problems of clogging for the investigated time period of the experiment. Presented results reveal that modification of a trickling liquid composition affects the removal efficiency of the biofiltration system and a biotrickling filter may be tuned so as to effectively treat air streams containing hydrophobic volatile organic compounds. Future development of the proposed laboratory set-up should include more precise process control (pH and temperature of trickling liquid) as well as online monitoring of the process efficiency with the use of an electronic nose [5].

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