Vol. 34

2008

No. 4

MIROSŁAW SZKLARCZYK*, WALDEMAR ADAMIAK*, JOANNA SZYMBORSKA**

BIOFILTRATION OF BENZENE AND TOLUENE VAPOURS. INFLUENCE OF QUALITY OF FILTRATION BED ON PROCESS RESULTS

The results of biofiltration of the vapours of aromatic compounds occurring in air were examined. The experiments were carried out in a laboratory-bench scale on benzene and toluene. The first one represented an aromatic compound without any substituent, the second one was a benzene derivative with substituted methyl group. First, the effects of biofiltration were examined in the bed composed of acid peat inoculated with microorganisms of activated sludge. Further examinations were carried out on a modified bed consisting of deacidified peat inoculated with microorganisms adapted to in vitro decomposition of benzene. To improve structural properties of the bed, it was mixed with perlite, being a neutral component. The modifications considerably reduced adaptation period of the bed and increased biofiltration rate. To maintain high performance of the modified bed, it was necessary to increase the doses of mineral media.

1. INTRODUCTION

Biological purification of gases consists in decomposition of impurities adsorbed from waste gases by microorganisms living in sorbent. The prerequisite for the occurrence of the biodegradation process is sorption, i.e. the drifting of organic compounds into the environment of microorganisms (water, wet organic materials) as well as the susceptibility of the organic impurities to biological degradation. The most popular method for biological purification of gases is biofiltration. The heart of the filter, which has a critical effect on the efficiency of the process, is a bed of wet layer of organic material (peat or compost) covered with microorganisms.

Main features of the bed, which determine its quality, are:

• structure,

^{*} Institute of Environmental Protection Engineering, Wrocław University of Technology, Wybrzeże Wyspiańskiego 27, 50-370 Wrocław, Poland.

^{**} ALSTOM Poland.

- settlement (type and quantity of microorganisms),
- moisture content,
- concentration of biogenic compounds,
- reaction.

Organic solids are carriers for microorganisms, as well as for water and biogenic substances, like nitrogen, phosphorus and microelements.

- The results of biological filtration to a great extent depend upon [1]:
- susceptibility of vapours to biological degradation,
- dwell time of the flue gas in the filtration bed,
- concentration of impurities in flue gas.

The paper presents relations between the features of the filtration bed and the effectiveness of biological purification of air polluted with benzene and toluene.

2. MATERIALS AND METHODS

The experiments were carried out in the laboratory biofilter, shown in figure 1, with two beds made of acid peat. One bed was inoculated with activated sludge from laboratory culture, whereas the second one – with bacteria culture adapted for decomposition of benzene, obtained by the enrichment method. Later, the peat bed inoculated with the adapted bacteria was mixed with perlite in a ratio of 1:1 v/v. Single layer of the bed was 0.3 m thick, and its cross-section area amounted to $7.85 \cdot 10^{-3} \text{ m}^2$. Moisture content of the bed was maintained within the range of 30-50%. The experiments were carried out at room temperature. Relative humidity of air, contaminated with vapours of benzene or toluene, approximated to 100%. In both cases, wide ranges of the concentration of pollutants were examined. Stream of air amounted to $0.25 \text{ m}^3/\text{h}$, which corresponded to linear flow rate of 0.88 cm/s.

The effectiveness of biofiltration was evaluated by comparing the concentration of the pollutants in the inlet and in the outlet parts of the apparatus, as measured during several dozen days lasting continuous operation of the system. The concentrations of pollutants were measured by the gas chromatography method. The chromatograph of the GCHF 18.3 type, with flame ionization detector, was equipped with a column 1 m long and 5 mm in diameter, filled with chromosorb WAW 80/100 covered with 10% carbowax. The results of biofiltration were characterized by the following parameters:

• mass loading of the bed

$$O = \frac{c_p \cdot V}{V_z} \quad (\text{mg/(m^3 \cdot s))},$$

elimination capacity

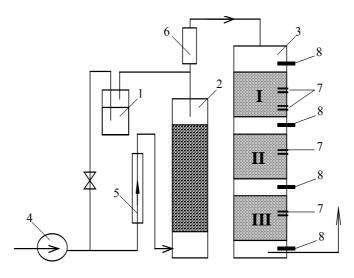
$$V = \frac{(c_p - c_k) \cdot V}{V_z} \quad (\text{mg/(m3·s)}),$$

removal efficiency

$$\eta = \frac{c_p - c_k}{c_p} \cdot 100\%,$$

where:

- c_p initial concentration, mg/m³,
- c_k final concentration, mg/m³,
- V_z volume of the bed used, m³,
- V_z volumetric flow rate of air, m³/s.



Laboratory-scale biofiltration system used: 1 – bubbler for saturation of air with pollutant, 2 – air damping column, 3 – biofilter, 4 – pump, 5 – rotameter, 6 – mixing chamber, 7 – pipe connectors used for filter bed moisture control, 8 – pipe connectors for gas sampling

To obtain inoculum with high metabolic activity in respect to benzene, an enrichment culture was established. The mineral medium, similar to that used by FOCHT [2], was somewhat modified and contained NH₄NO₃ (0.8 g), K₂HPO₄ (0.8 g), KH₂PO₄ (0.2 g), CaCl₂ · 7H₂O (0.1 g), MgSO₄ (0.1 g), FeCl₃ (10 mg) in 1000 cm³ of water. The medium (also in concentrated form) was used to supply a bed with biogenic components as well. Benzene, added to the medium, was the only source of carbon. The medium was inoculated with microorganisms that live in a great abundance in the compost-based bed. The inoculated sample was taken from the biofilter used for odour control in municipal composting plant. Under such conditions, mainly microorganisms that are able to assimilate benzene have survived. The culture was grown in glass conical flasks ($V = 250 \text{ cm}^3$) with a ground glass joint filled with 100 cm³ of mineral medium, benzene and inoculum. At first, benzene was added in small quantity (5–10 mm³), then this quantity was gradually increased up to 200 mm³. In the first period, every second day, and then every day, 10 cm³ of the liquid culture was transferred to a new medium and continuously shaked (100 rpm). Incubation was conducted at room temperature for 1.5 year.

3. RESULTS AND DISCUSSION

3.1. EXAMINATION OF BENZENE BIOFILTRATION

The first series of tests was carried out with filtration bed based on acid garden peat, pH 3.5–4. Because this peat is rather sterile (approx. 10^5 cfu/g d.m.), it was enriched with activated sludge, cultured under laboratory conditions, and finally the medium was supplemented with molasses. This allowed the population of microorganisms to be increased up to approx. $9 \cdot 10^6$ cfu/g d.m. In the first period, which lasted for approx. 1 month, elimination capacity did not exceed 3 mg/(m³·s) in spite of rather low loadings, i.e. 2–9 mg/(m³·s). Later, the elimination capacity gradually increased. After one month also a removal efficiency increased considerably – some over 70% of benzene was removed from the gas at the loadings approximated to 15 mg/(m³·s). The maximum elimination capacity approximated to 13 mg/(m³·s) at the loadings of 24 mg/(m³·s), but it was accompanied by a lower removal efficiency, which decreased to approx. 50%.

It should be stressed that a relatively long time is required for adaptation of filtration bed for benzene removal. The elimination capacity increased very slowly, probably due to a slow growth rate of the microorganisms capable of benzene degradation.

To increase the removal efficiency, the peat bed was inoculated with microorganisms sampled from the enrichment culture. In consequence, a final number of microorganisms approximated to 10^8 cfu/g d.m. However, this did not improve significantly the operation of the biofilter. Only deacidification of the bed with CaO considerably increased the efficiency of biofiltration. The efficiency of one layer of deacidified bed exceeded the efficiency of three layers of acid beds (table). Moreover, the bed deacidification resulted in an instantaneous increase in its elimination capacity and this improvement occurred within few hours after deacidification. This means that the adaptation rate of a modified bed significantly increased compared with not deacidified beds and inoculated with non-adapted microorganisms. It should be noted that according to bibliography the adaptation periods of the beds designed for biofilters used for the removal of benzene vapours from gases exceed two days – in spite of the presence of adapted inoculum and low loadings, which should promote further adaptation [3]–[5].

Table 1

Bed	Initial	Final	Gas	Load	Biofiltration
	concentration	concentration	purification	of the bed	rate
	C_p	c_k	η	0	V
	(mg/m^3)	(mg/m^3)	(%)	$(mg/(m^3 \cdot s))$	$(mg/(m^3 \cdot s))$
Acid, I layer	1200	860	28	35.4	10.0
Acid, II layer	860	450	48	25.4	12.1
Acid, III layer	450	220	51	13.3	6.8
Deacidified, I layer	1120	30	97	33.0	32.1
Deacidified,	1020	0	100	27.5	27.5
peat+perlite,	1560	0	100	42	42
I layer	2710	15	99.5	73.1	72.7

Comparison of results of benzene biofiltration through various beds. Acid peat inoculated with activated sludge, deacidified peat inoculated with culture obtained in vitro and mixture of deacidified peat and perlite inoculated with bacterial culture obtained in vitro

Further operation of the biofilter at high loadings (above 20 mg/($m^3 \cdot s$)) required an additional medium because of an intensive depletion of mineral salts (usually, biofilter beds operating at low loadings do not require extra media for their whole operation life). However, addition of standard mineral medium does not increase the effectiveness of the filter operation. Quite the opposite, a fluid medium supplied twice a day worsened the structure of the bed by closing up its voids, which reduced the effectivetiveness of the purification process.

To prevent further worsening of the bed, peat was mixed with fine-grained perlite in a volume ratio of 1:1. Perlite is durable and allows better bed aeration. Addition of perlite practically solved the problem, and a frequent sprinkling of the bed with extra medium concentrate (concentration of 20 g/dm³) proceeded without obstructions and considerably improved the biofilter operation. At the inlet concentration of benzene ranging from 360 mg/m³ to 2905 mg/m³ (loading range of 9.7–78.4 mg/(m³·s)), the removal efficiency exceeded 95%, whereas the maximum elimination capacity amounted to 73 mg/(m³·s) (table 1). This value is by one order higher than that for acid peat beds inoculated with activated sludge, and also higher than that reported by other researchers who examined the biofiltration of benzene [6]–[8].

3.2. BIOFILTRATION OF TOLUENE

The first series of experiments with the biofiltration of toluene has been carried out on the bed made of acid peat inoculated with activated sludge. The properties of the bed and operation conditions were as described above for biofiltration of benzene. The experiments were carried out for 61 days. An initial concentration of toluene was changed within the range from 136 mg/m³ to 670 mg/m³. Hence, the maximum loading of the bed amounted to 19.8 mg/(m³·s). The bed adaptation period finished after 9 days. At the concentration of toluene slightly exceeding 200 mg/m³ and three layers of the filtration bed (0.3 m each) the air at the outlet was almost clean. After an increase of the initial concentration of toluene above 400 mg/m³, the removal efficiency dropped by several dozen per cent.

There was also examined the possibility of utilizing the modified and deacidified peat mixed with perlite, formerly inoculated with bacteria adapted in vitro for the decomposition of benzene (0.3-m thick single layer), for biofiltration of toluene. The bed for several weeks effectively purified the air polluted with benzene. The concentration of toluene in the inlet air varied from 300 mg/m³ to 1440 mg/m³. After a 3-day adaptation time, the bed removed 100% toluene, provided that its concentration did not exceed 1180 mg/m³ (loadings up to 32 mg/(m³·s)). The maximum elimination capacity amounted to 34 mg/(m³·s) under the load of 39 mg/(m³·s), whereas the critical loadings of the bed approximated to 32 mg/(m³·s) (table 2).

It has been found that the peat and perlite mixture, inoculated with benzenedecomposing bacteria, allowed much more efficient removal of toluene compared

Table 2

Comparison of results of biofiltration of toluene through various beds. Acid peat inoculated with activated sludge and mixture of deacidified peat and perlite inoculated with bacterial culture obtained in vitro

Bed	Initial concentration c_p (mg/m ³)	Final concentration c_k (mg/m ³)	Gas purification η (%)	Load of the bed O (mg/(m ³ ·s))	Biofiltration rate V (mg/(m ³ ·s))
Acid peat, I layer	400	260	35	11.8	4.1
Acid peat, II layer	260	133	49	7.7	3.8
Acid peat, III layer	133	0	100	3.9	3.9
	300	0	100	8.1	8.1
Deacidified	425	0	100	11.5	11.5
peat + perlite,	825	0	100	22.3	22.3
I layer	1180	0	100	32.0	32.0
	1440	125	87.2	39.0	34.0

to that obtained with the conventional bed made of acid peat inoculated with activated sludge. The adaptation period was reduced from 9 to 3 days, and 100% purification was obtained under ten times higher loadings (3.9 mg/(m³·s) and 32 mg/(m³·s), respectively). Also the parameters of the operation of biofilters used for the removal of toluene, reported by other researchers, are considerably lower in comparison with those for the peat and perlite mixture described here (there are reported critical loads of 4–15.3 mg/(m³ · s)) [9]–[11]. It is evident that the bacterial cultures adapted in vitro and used for inoculation of the bed have high metabolic effectiveness.

4. CONCLUSIONS

Considering the results presented above, the following conclusions may be drawn:

1. Peat is the prerequisite for obtaining a full biological activity of microorganisms inoculated in the bed.

2. The drawback of peat, as the base for the filtration beds, is its low stability and loss of structural features under conditions of high moisture.

3. Perlite is a usefull additive of biofiltration beds. Its mixture with peat is characterized by a stable structure.

4. To obtain an efficient inoculum, microorganisms should be slowly adapted for the decomposition of air pollutants. It should be carried out in vitro.

5. A suitable modification of the bed and the application of active inoculum enabled:

a) a considerable reduction of the time (to few hours) required for adaptation of the bed for removal of benzene (in references there are reported periods from several to ten or fifteen days),

b) very high elimination capacity of benzene under high loadings of the bed; critical loadings exceeded 70 mg/($m^3 \cdot s$), whereas for conventional beds they range from 1.5 to 13 mg/($m^3 \cdot s$)).

6. In the biofilter operating under high loadings, a considerable concentration of biogenic salts is undispensable.

7. The bed inoculated with benzene-decomposing bacteria proved to be suitable for the removal of toluene as well. It was found that the critical value of the loading is relatively high (approx. $32 \text{ mg/m}^3 \cdot \text{s}$) and considerably exceeds the values reported by other authors (4–15.3 mg/m³ · s).

REFERENCES

 VDI-Richtlinien 3477 – Biologische Abgas-/Abluftreinigung Biofilter, Kommision Reinhaltung der Luft im VDI und DIN, Beuth Verlag, 1991.

- [2] FOCHT D., Growth kinetics of Pseudomonas alcaligenes C-0 relative to inoculation and 3chlorobenzoate metabolism in soil, Appl. Environ. Microbiol., 1987, Vol. 53, 8, 1846–1849.
- [3] SENE L., CONVERTI A., FELIPE M., ZILLI M., Sugarcane bagasse as alternative packing material for biofiltration of benzene polluted gaseous streams: a preliminary study, Bioresource Technology, 2002, Vol. 83, 2, 153–157.
- [4] ZILLI M., DAFFONCHIO D., DI FELICE R., GIORDANI M., CONVERTI A., Treatment of benzenecontaminated airstreams in laboratory-scale biofilters packed with raw and sieved sugarcane bagasse and with peat, Biodegradation, 2004, Vol. 15, 87–96.
- [5] CHOI S.-CH., OH Y.-S., Simultaneous removal of benzene, toluene and xylenes mixture by a constructed microbial consortium during biofiltration, Biotechnology Letters, 2002, Vol. 24, 1269– 1275.
- [6] SHAREEFDEEN Z., BALTZIS B., OH Y.S., BARTHA R., [in:] Advances in Bioprocess Engineering, Galindo E., Ramirez O.T. (Eds.), Kluwer Academic, Dordrecht, the Netherlands, 1994, 397–404.
- [7] JOHNSON C., DESHUSSES M., Quantitative structure-activity relationships for VOC biodegradation in biofilters, Proceedings of the Fourth International in Situ and on-Site Bioreclamation Symposium, Vol. 5, Battelle Press, Columbus, 1997.
- [8] EITNER D., *Biofilter in der praxis* [in:] *Biologische Abluftreinigung*, Expert Verlag Ehningen bei Boeblingen, 55, Germany, 1990.
- [9] SEED L.P., CORSI R.L., [in:] Proceeding of the 89th Annual Meeting and Exhibition Air & Waste Management Association, paper 96 – WP87A.06; Air & Waste Management Association, Pittsburgh, PA, 1996, 15 pp.
- [10] GRIBBINS M., LOEHR R., Effect of medium nitrogen concentration on biofilter performance, J. Air Waste Manage. Assoc., 1998, 48 (3), 475.
- [11] DESHUSSES M., JOHNSON C., Development and validation of a simple protocol to rapidly determine the performance of biofilters for VOC treatment, Environ. Sci. Technol., 2000, 34, 461–467.

BIOFILTRACJA PAR BENZENU I TOLUENU. WPŁYW JAKOŚCI ZŁOŻA FILTRACYJNEGO NA EFEKTY PROCESU

Badano efekty biofiltracji par związków aromatycznych zawartych w powietrzu. Doświadczenia przeprowadzono w skali laboratoryjnej. Do badań wybrano benzen jako związek aromatyczny bez podstawnika w pierścieniu oraz toluen jako pochodną benzenu z podstawioną grupą metylową. W pierwszej kolejności badano efekty biofiltracji na złożu uzyskanym przez zaszczepienie torfu kwaśnego mikroorganizmami osadu czynnego. Dalsze badania wykonano na złożu zmodyfikowanym, uzyskanym przez zaszczepienie torfu odkwaszonego mikroorganizmami przystosowanymi do rozkładu benzenu w hodowli in vitro. Ponadto modyfikacja złoża polegała na dodaniu do torfu czynnika neutralnego – perlitu, który poprawił właściwości strukturalne złoża. Dzięki zastosowaniu złoża zmodyfikowanego uzyskano znaczne skrócenie czasu adaptacji złoża oraz zwiększenie szybkości biofiltracji. Utrzymanie korzystnych efektów pracy złoża zmodyfikowanego wymagało dostarczania zwiększonych dawek pożywek mineralnych.