The application of Brij 35 in biofiltration of the air polluted with toluene vapours

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Abstract. Removal of certain organic pollutants from the environment may be hindered due to their weak water solubility and high vapour pressure. In particular, these are factors that limit the application of biological methods in remediation since they have an influence on the bio-accessibility of the xenobiotics. For that reason, we carried out research on the use of surface-active agents that have impact on the increase in solubility of hydrophobic compounds. In this publication, we present the results of laboratory tests on the application of Brij 35 in purification of the air polluted with toluene vapours by the biofiltration method. Within the range of surfactant concentrations subjected to the research (200, 300, 400 mg/dm^3), we observed an improvement of the removal efficiency as compared to the control series (without the surfactant).

1 Introduction

Gases containing volatile organic compounds may be purified through the use of physical, chemical or biological methods. The choice is determined by such factors as: proprieties of the purified gases, concentration of the emitted pollutants, type of the emission source, and also the planned purification efficiency. Biological gas purification is based on biodegradation processes occurring naturally in the environment – the ability of certain heterotrophic microorganisms to the decomposition of organic matter as well as the ability of autotrophs to oxidise the non-organic compounds. Therefore, a prerequisite to the application of biological methods of gas purification is the vulnerability of pollutants to the biodegradation. The purification mechanism is based on two main processes: sorption and biodegradation.

The removal of hydrophobic pollutants is a very important issue in gas biofiltration. The majority of biofiltration models are restricted only to pollutants that are easily soluble in water and at low concentrations [1]. A decreasing efficiency of removal of certain groups of organic pollutants according with the sequence: alcohols > esters > ketones > aromatic hydrocarbons > aliphatic hydrocarbons, is directly correlated with the Henry's constant: the pollutants' water solubility decreases along with a possibility of their biodegradation in the biofilter bed [2]. For that reason, scientists did both: the initial purification of gases routed into the filter bed and the change of parameters of purification process and modification of

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the filter bed for the improvement of its sorption and degradation proprieties (e.g. the use of microfungi) [3-4]. Furthermore, they focus on interactions between the pollutants contained in the gases subjected to purification that could have significant, both positive and negative impact on the pollutants' bio-accessibility, and at the same time, on the efficiency of the gas purification process, especially in the case of removal of compounds with hydrophobic proprieties [5]. It was noticed that simultaneous removal of easily water-soluble compound along with a removal of a poorly-soluble compound may hinder the removal of the latter [6] or, on the contrary, facilitate the transport of the hydrophobic pollutants to aqueous phase and have a positive impact on efficiency of gas purification [2]. The impact of some chemicals on water solubility of the others was used in the environmental engineering, mainly in soil bioremediation [7].

By lowering the surface tension and micelles forming, the surface-active agents affect the improvement of the bio-accessibility of the organic compounds in two ways: by increasing the hydrocarbons' solubility and by affecting the hydrophobicity of the cell surface of microorganisms [8]. Surfactants can adsorb onto cells or interact with their external layers causing an increase of their hydrophobicity and at the same time, raising their affinity of the microbial cells to the poorly-soluble particles of organic compounds and facilitates their mutual contact [9]. Furthermore, the surface-active agents enable the emulsification of hydrophobic pollutants, increasing at the same time the easiness of their leaching from the soil. In the soil purification process, the proper choice of the surface-active agent (or their mixes) is very important, since they present a high acting selectivity and, by accelerating the biodegradation of specific pollutants, they can also obstruct the development of strains of microorganisms that are responsible for decomposition of the other compounds [10]. For example, the Tween 80 preparation may cause an increased activity of the *Sphingomonas sp.* strains, by obstructing the development of the *Mycobacterium sp.* [13]. The research also showed a high dependency of the biodegradation efficiency on the type and chemical structure of pollutants [11]. Therefore, the surface-active agents may have positive, neutral or negative impact on the biodegradation of pollutants. The main factors taken into consideration while choosing the surfactants are: Hydrophilic Lipophilic Balance (HLB), Critical Micelle Concentration (CMC), stability of created emulsion, surfactant's biodegradability and its toxicity towards the microorganisms [13].

On the basis of these experiences, the researches currently study the use of surface-active agents in biological gas purification form the hydrophobic pollutants [14]. Due to a better biodegradability, the most often we use the anionic and non-ionic surfactants. Furthermore, the non-ionic surfactants do not ionise in water, that is why they can be safely and dosed at the filter bed along with the nutrient solution [15]. Ramirez et al. [15] studied the impact of dosage of different non-ionic substrates on the efficiency of methane biofiltration. Methane occurring in a natural environment is easy degradable by microorganisms occurring in soil and in water, and for this reason, it may be removed from gases by biological methods. However, a poor solubility of methane in water hinders its biodegradation in biofilters. The influence of a series of non-ionic surfactants (Brij 35, Brij 58, Brij 78, Tween 20, Tween 40, Tween 60) on the efficiency of biofiltration and on biomass growth was compared. The study was done in a separated filtration column for each surfactant, while keeping the constant surfactant concentration in the medium at the level of 0.5% and biofilter work in average initial methane loading rate equal to 68.5 g/m^{3}h. It was observed that along with the increase of the HLB value, the biomass production rate was decreasing, thus, the surface-active agent was acting as a typical detergent. The influence on the stability of the biofilter's work was observed, since the reduction of biomass excess prevented clogging_of the filter bed, improving at the same time its efficiency. For a biofilter without the surfactant, the elimination capacity amounted to

24 g/m^{3}h. With surfactants from the Brij group, elimination capacity oscillated within the range from 25.5 to 32.5 g/m^{3}h, whereas for Tweens, it amounted from 28.5 to 32.5 g/m^{3}h. The highest elimination capacity was observed for the Brij 58 and Tween 20 surfactants; however, the latter was highly toxic for the bed's microflora. The maximum elimination capacity amounted to 45 g/m^{3}h.

Surfactants from the Brij group were also tested for the toluene biofiltration. The authors [16, 17] regarding the example of two surface-active agents (Brij 30 and Brij 35), determined the influence of their concentration on the toluene solubility in water by determining the S/S_0 ratio (the ratio of toluene concentration in water with surfactant to the toluene concentration without surfactant). It was revealed that both the Brij 30 and Brij 35 had an impact on the improvement of toluene solubility in the aqueous phase. The best effects were obtained for the surfactant concentration range below the value of Critical Micelle Concentration (CMC). It was also observed that the toluene solubility in water increases along with an increase of the surface-active agent concentration; however, only to a certain critical value above which there is a decrease of a positive surfactant impact. The optimal Brij 30 concentration, in which the highest S/S_0, ratio was obtained, amounted to 35 mg/dm^3. In the case of Brij 35, within the whole range of its concentration that was subjected to research (i.e. from 0 to 540 mg/dm^3), there was an improvement in toluene solubility. The best effects were obtained for the surfactant concentrations below the CMC value. The obtained results might suggest that, along with an increase of toluene solubility, its bio-accessibility also increases, what can be used in order to reach a higher efficiency of biofiltration of polluted gases. However, the experiments revealed a decrease of the maximum biofiltration rate along with an increase of surfactant concentration, and an obstructing influence of detergent on the microorganisms growth. The research paper's does not contain results for the variant in which the surfactant concentration is higher than the critical micelle concentration (CMC). The results obtained by the authors may be explained not only by a negative influence of surfactants on the colonisation of the bed, but also by vulnerability of the microorganisms colonising the biofilter bed on the toxic influence of toluene in concentration equal to 200 ppm. Most likely, this bed was inoculated with an unadapted inoculum, that is to say, activated sludge from a sewage treatment plant [18]. Hwang and Tang [19], using higher toluene concentrations and much higher loads applied on the filter bed, stated high biofiltration rates (97 g/m^{3}h at maximum) on a bed inoculated with specially-adapted inocula.

Dhamwichukorn et al. [20] used a mix of non-ionic surfactants featuring a following composition: Triton X-100 (0.05%), Brij 35 (0.1%) and Brij 58 (0.05%) in purification at a higher temperature of gases polluted with α-Pinene and methanol. These are pollutants that are characteristic for waste gases from papermaking industry. These gases have a high temperature: that is why their biofiltration was conducted on a heated bed with a temperature of 55°C and inoculated with thermophilic microflora isolated from a compost waste. Adding a mix of non-ionic surfactants: Triton X-100, Brij 35 and Brij 58, significantly increased the α-Pinene's solubility in water, increasing the biofiltration rate and shortening the time of the retention of gases within the filter bed. The research was conducted in a low gas flow rate (18 dm^3/h) and with pollutants loads applied on the filter bed within the range of 1.5–2 g/m^{3}h. The biofilter with a filter bed moistened with surfactant mixture reached the maximum elimination capacity at the level of about 1.8 g/m^{3}h, and it was three times higher as compared to the control (for a biofilter without the surfactant dosage, the maximal elimination capacity amounted only to 0.6 g/m^{3}h).

2 Materials and methods

The in-house research on toluene biofiltration was conducted at a research installation in a laboratory scale (the conceptual scheme was presented on Fig. 1). The filtration column was constituted by a PVC tube with an internal diameter of 0.1 m, filled with three layers of a filtration material with a height of 0.3 m each. The filtration material used in research was a coconut fibre with perlite addition, enhanced with a mineral medium featuring the following composition: total nitrogen 23%, phosphorus pentoxide 9%, potassium oxide 10%, magnesium oxide 2%, copper 0.005%, iron 0.05%, manganese 0.03%, zinc 0.005%. The air was supplied firstly to the column for gas pre-conditioning with a height of 1 m, filed with a humid active carbon. Humidified air was routed into a mixer, where it was mixed with the air dosed from a gas wash bottle in a form toluene vapours. The gas stream was routed from the top of the filtration column through the next bed layers. The gas samples were taken from spigots situated on the inlet to the biofilter, and also after every three layers of the filter bed. The toluene content in the air was determined by the gas chromatography method. The surfactant (Brij 35) was being dosed on the filtration bed during its humidification.

The bio-filter's work, in the assumed flow rate equal to 0.75 m^3/h, was described with the following parameters:

- loading rate of the filter bed O_z, g/m^3h:

$$O_z = \frac{c_p \cdot \dot{V}_g}{V_z} \tag{1}$$

where:
C_p – inlet concentration, g/m^3;
V_g – flow rate, m^3/h;
V_z – filter bed's volume, m^3;

- biofiltration rate V_r, g/m^3h:

$$V_r = \frac{(c_p - c_k) \cdot \dot{V}_g}{V_z} \tag{2}$$

where:
C_k – outlet concentration, g/m^3;
C_p – inlet concentration, g/m^3;
V_g – flow rate, m^3/h;
V_z – filter bed's volume, m^3;

- removal efficiency η, %:

$$\eta = \frac{c_p - c_k}{c_p} \cdot 100\% \tag{3}$$

where:
C_p – inlet concentration, g/m^3;
C_k – outlet concentration, g/m^3.

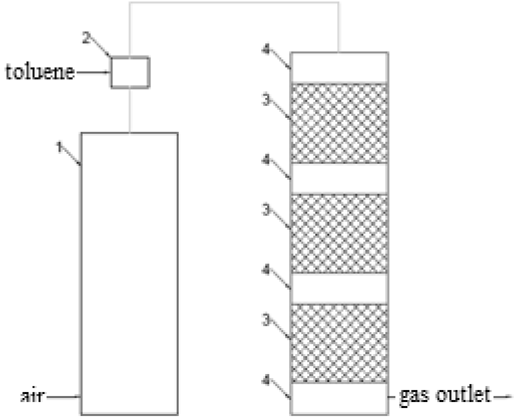

Fig. 1. Research installation scheme: 1 – column for gas pre-conditioning, 2 – mixer, 3 – filter bed, 4 – gas intake spigot.

In order to determine a total number of microorganisms and determine the number of yeasts and moulds, the surface culture method was applied respectively on the nutrient agar and Sabouraud agar dextr. 2%. The results of colonisation of the filter bed were indicated in cfu/g of d.m. (colony-forming units per one gram of dry matter of the filter bed).

3 Results and discussion

The concentrations of aqueous solution of Brij 35 preparation that served to moisturise the filter bed amounted to 200, 300 and 400 mg/dm^3 successively. The solutions were being dosed respectively on the 32nd, 46th and 60th day of conducting the biofiltration process. The results of the research on the influence of different concentrations of the Brij 35 preparation above the CMC with different loads applied on the filter bed on the removal efficiency of air polluted with the toluene vapours were presented in Figure 2. For each measurement series, the toluene loads applied on the filter bed were being gradually increased.

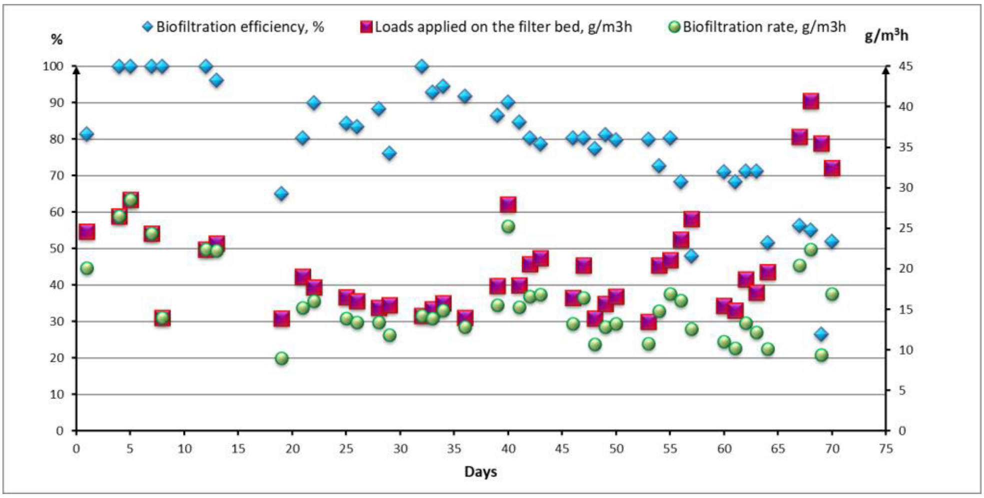

Fig. 2. Dependency of the rate and efficiency of biofiltration on the toluene loads applied on the filter bed for a series in which the Brij 35 was being dosed.

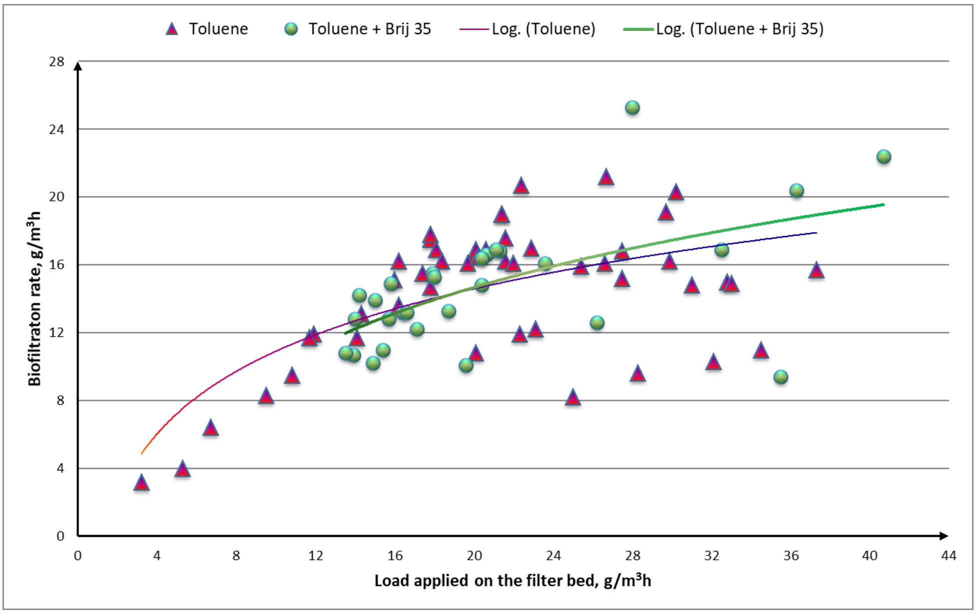

Fig. 3. Dependency of toluene biofiltration rate on the loads applied on filter bed enhanced by the Brij 35 as compared to the control series.

For increasing values of toluene loads applied on the filter bed, significant decreases in the efficiency of air purification were observed. The obtained results were compared to a control series (toluene biofiltration without the surfactant addition – filter bed moisturised with water). The maximum biofiltration rate amounted to 21.2 and 25.3 g/m^{3}h respectively for the control filter beds and for the filter beds on which the Brij 35 preparation solution was dosed (Figure 3).

The results of quantitative research on colonisation of filter bed were presented in Table 1. An increase in the total number of microorganisms as compared to the control filter bed was observed, which allows to conclude that within the studied Brij 35 concentration range, there is no toxic influence on the bacteria of the filter bed. However, a decrease in the number of microfungi was observed.

Table 1. Results of microbiological research.

Sample	Bacteria, cfu/g of d.m.	Fungi, cfu/g of d.m.
Output filter bed	31 x 10^4	43 x 10^4
Filter after the adaptation period	98 x 10^8	85 x 10^7
Control filter bed (66th day)	50 x 10^6	33 x 10^6
Filter bed after Brij 35 dosage	40 x 10^7	14 x 10^5

Wu-Chung and Hui-Zheng [16] obtained results, by using this surface-active agent also in biofiltration of the air polluted with toluene vapours, oscillating within the range from 17.41 to 26.12 g/m^{3}h (with the surfactant concentration range from 0 to 540 mg/dm^3). Therefore, the value of the maximal biofiltration rate, obtained on the basis of the in-house research, equal to 25.3 g/m^{3}h, is close to the upper value of results obtained by the above-mentioned authors. However, Wu-Chung and Hui-Zheng [16] observed a decrease in the maximal biofiltration rate along with an increase in the surfactant concentration, as well as an obstructing influence on the microorganisms' growth.

4 Conclusions

Surfactants, as agents affecting the increse in solubility of hydrophobic pollutants, and, at the same time, their bioaccessibility, have a large potential in terms of application in biological methods of environmental remediation, also including the biological purification of waste gases. The results of the research on toluene biofiltration show that the use of Triton X-100 in a concentration higher than the CMC value enables the increase of toluene removal efficiency, and, at the same time, gases purification with higher polluant loads applied on the filter bed. Within the studied surfactants' concentration range (for 200, 300, 400 mg/dm^3), the results indicated a positive influence of the Brij 35 on the biofiltration rate of the air polluated with toluene vapours. The surface active agents may also have a negative influence on the biodegradation of pollutants through toxic impact on the microorganisms colonising the filter bed. For this reason, the choice of the appropriate surfactant and its concentration is of crutial importance in order to optimise benefits resulting from its impact. Studies did not reveal toxic impact of the Brij 35 on quantitative colonisation of the filter bed.

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