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The use of surfactant from the Tween group in toluene biofiltration

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Abstract: Due to the lower energy consumption and waste production compared to traditional methods, the environmental bioremediation methods based on natural processes have been gradually becoming more prevalent in environmental engineering. Biological methods are used in waste management, wastewater treatment, gas treatment or soil remediation. For the low solubility of some pollutants and lower bioavailability, the use of biological methods may be hampered. This problem might be mitigated with the use of surfactants. This paper presents the results of studies regarding the effect of dosing a selected surfactant from the Tween group (Tween 20) on the efficiency of toluene elimination from the air by biofiltration. The obtained maximum biofiltration rate was 21.2 g/m^3/h and 19.8 g/m^3/h for the control bed and for the bed to which the Tween 20 solution was dosed, respectively. The effect of Tween was neutral (the effectiveness of toluene removal was insignificantly comparable to the effectiveness of the control series), it did not affect the effectiveness or limited the development of the biofilter microflora.

Introduction

Biological methods of environmental remediation are based on biodegradation processes naturally occurring in the environment. In order to successfully perform the process of biofiltration, pollutants are to be susceptible to biological degradation. In the biofilter the pollutants are absorbed in the water moistening the bed, and thus are conveyed into the biofilm where the biodegradation of the eliminated substance occurs. The process of microbiological degradation depends on the type, chemical structures of compounds, physio-chemical properties and their concentration. Low solubility of some pollutants in water may limit their biological elimination. The improved efficiency of hydrophobic compounds elimination by biofiltration can be obtained by using consortia of microscopic fungi demonstrating the decomposition property of organic compounds (Gospodarek et al. 2019). One of the main advantages of this method is the high resistance of fungi to environmental conditions, such as low pH, which gives the possibility of hydrocarbon degradation in a wide range of process conditions (Rene et al. 2012).

Volatile organic compounds present in purified gases can react with one another, undergo transformations, change their properties and affect the bioavailability and rate of biodegradation. In the case of the biofilter process these interactions can have a significant, both positive and negative, impact on the bioavailability of pollutants, and consecutively – the effectiveness of the gas purification process, especially when removing compounds with hydrophobic properties. Ikemoto et al. (Ikemoto et al. 2006) studied the effect of the presence of hydrophilic compounds in the gas on the biodegradation of hydrophobic compounds. As expected and shown – the presence of hydrophilic impurities in the treated gases significantly inhibits the elimination of hydrophobic components. Reducing the concentration of the hydrophilic component in the mixture results in reducing its negative impact on the elimination of the hydrophobic component. Balasubramanian et al. (Balasubramanian et al. 2012) determined that high ethanol concentration has a negative effect on the degradation of toluene and benzene, while its presence can cause an increase in the elimination rate of dichlorobenzene. Hassan and Sorial (Aly Hassan and Sorial 2010) conducted biofiltration studies on n-hexane in the presence of benzene. N-hexane is relatively soluble in benzene, and thus, when mixed with benzene, its solubility in water is even improved. Studies have shown that the presence of benzene vapors has a significant positive effect on the biodegradation of n-hexane, but the presence of hexane does not prove to sustain such the effect on benzene elimination.

Similar increasing solubility of hydrophobic impurities and their bioavailability can be achieved using surfactants (Miller et al. 2018). Surfactants, by reducing surface tension, improve the solubility of organic compounds and increase their bioavailability (Maier et al. 2000). In addition, surfactants allow for the emulsification of hydrophobic impurities while improving leaching from the soil. However, surfactants show high selectivity and by increasing the biodegradation rate of a given pollutant they can simultaneously block the development of strains of microorganisms responsible for the decomposition of other compounds. For example, Tween 80 may cause increased activity of the *Sphingomonas* strains by inhibiting the development of *Mycobacterium* (Błaszczyk 2009).

In environmental engineering, surfactants are used in soil remediation processes. One method of soil purification is washing. In this procedure the liquid with absorbed pollutants is separated by passing the washing solution through the soil layer. Due to the low solubility of many organic compounds in the wash water, surfactant solutions are used. Impurity particles are solubilized in micelles, which increases their mobility and facilitates leaching them from the soil (Rakowska et al. 2012). The results of researches on the removal of polycyclic aromatic hydrocarbons (PAHs) from soil show that the use of surfactants as rinsing agents significantly improves the cleaning effects. Among the surfactants tested, the best effects were obtained with beta-cyclodextrin and rhamnolipids (Wojnowska-Baryła 2011). Another method of removing pollutants, especially petroleum substances, from soil is dispersing them in water. Surfactants act as dispersants here. This method reduces the interfacial tension of the oil-water system by introducing a surfactant into it, which causes oil particles to disperse and aids its further biodegradation (Radwan et al. 2012). The studies (Kim et al. 2001) of the influence of Brij 30, Tween 80 and Triton X-100 surfactants on the dispersion of polycyclic aromatic hydrocarbons (naphthalene and phenanthrene) in the aqueous phase showed that the PAH solubility was proportional to the concentration of surfactants in the range concentrations above the CMC (critical micelle concentration) value. Below this value, no increase in PAH solubility was observed.

Due to their better biodegradability, anionic and non-ionic surfactants are most commonly used in biological purification of gases from poorly water-soluble compounds. In addition, non-ionic surfactants do not ionize in water, hence, they can be safely dosed to the filter bed along with the nutrient solution (Ramirez et al. 2012). Surfactants from this group demonstrate good surface-active properties; they also show low sensitivity to changes in the pH of the environment and to the electrolytes present in the solution. Surfactants from the Tween group are compounds from the group of polyoxyethylene derivatives of sorbitan esters (Przondo 2007), that are produced in the process of condensing sorbitan fatty acid esters with ethylene oxide in the presence of catalysts. They are produced from readily available natural raw materials, such as fat and sugar, so they can be widely used as additives in the food and pharmaceutical industries (Salager 2002). They are also exploited in cosmetics (Tadros 2005). Tweens belong to non-ionic surfactants. They show no irritating properties and prove to be low in toxicity, so they can be intended for internal use. They are used as wetting agents, stabilizers and emulsifiers. They are hydrophilic and water-soluble or can disperse in it, depending on the degree of their ethoxylation (Przondo 2007).

Ramirez et al. (Ramirez et al. 2012) conducted a study on the effect of dosing various non-ionic surfactants on the effectiveness of methane biofiltration. Conducted studies compared the impact of the number of non-ionic surfactants (Brij 35, Brij 58, Brij 78, Tween 20, Tween 40, Tween 60) on the effectiveness of biofiltration as well as on the biomass growth. The tests were carried out in separate filtration columns for each surfactant while sustaining the constant concentration of surfactant in the medium at the level of 0.5% and biofilter operation under the average bed load rate equal to 68.5 $g/m^3/h$. It was observed that as the HLB (the hydrophilic-lipophilic balance) value increased, the biomass production rate decreased, so the surfactant acted like a typical detergent. It also had an impact on the stability of the biofilter, because the reduction of excess biomass prevented the filter bed from clogging, and thus improved its efficiency. For the biofilter without the addition of surfactant, the biofiltration rate was 24 $g/m^3/h$. With the addition of surfactants from the Brij group, the rate was within the range from 25.5 $g/m^3/h$ to 32.5 $g/m^3/h$, while the ones from the Tween group ranged between 28.5 $g/m^3/h$ and 32.5 $g/m^3/h$. The highest rate of biofiltration was observed for surfactants Brij 58 and Tween 20, however the latter had a highly toxic effect on the bed microflora. The maximum biofiltration rate was 45 $g/m^3/h$. Tween 20 was used in combination with Zn (II) in the elimination of ethylbenzene from gases (Wang et al. 2014). The experiment was carried out on two filtration columns (K1 and K2), of which only one (K2) was fed with Tween 20 and Zn (II). For the control column (K1) with the increase of the bed load of ethylbenzene from 64.8 $g/m^3/h$ to 189.0 $g/m^3/h$, the biofiltration efficiency decreased from 74% to 54%, while for K2 from 94% to 69%. At the same time, a significant reduction was observed in the degree of biomass accumulation in the filter bed of K2 column. However, other researchers (Park et al. 2008) designed a loop reactor with a surfactant solution loop. The researchers raised the problem of the need for preliminary gas treatment to ensure stable operation of the biofilter. The use of surfactant was aimed at reducing the concentration of volatile organic compounds, especially those of hydrophobic nature. Toluene, which is poorly soluble in water, was chosen as the model impurity. In order to select the most effective surfactant concerning the improved solubility of toluene, the following non-ionic surfactants were compared: LA5, LA7, Span 20, Tween 20 and Tween 80. The solubility of toluene in the presence of surfactant was measured as the concentration of toluene in the water phase over 30 minutes at 298 K. On this basis, Tween 81 was selected for further biofiltration studies in the loop reactor as potentially the most effective for increasing the bioavailability of the model pollutant. In the study the authors obtained 90% efficiency in toluene elimination also with increasing bed loads. They showed that the use of surfactant allowed to ensure stable operation of the biofilter even in the case of sudden and unexpected changes in VOC concentration.

Own research was conducted to determine the effect of Tween 20 dosage on the removal efficiency of toluene in a classic biofilter.

Materials and methods

Own research on biofiltration of selected pollutant was performed on a laboratory scale. The conceptual scheme is depicted in Figure 1. The filtration column was a PVC pipe with an inner diameter of 0.1 m, consisting of three parts, each of them 0.5 m high. The column was filled with three layers of filter material, each 0.3 m high (total height of the biofilter bed was 0.9 m). The installation contained also a pre-conditioning column, 1.0 m high, filled with moistened activated carbon. A constant gas stream was established at a rate of 750 dm^3/h. Toluene was chosen as a model compound for the kinetics of the biofiltration process of volatile organic compounds. A stream of gas passed through the scrubber containing toluene in a liquid form was being added to the mixer, in which it blended with the flowing air. Gas samples for analysis were taken from four ports located at the inlet to the biofilter and after each of the three layers of the bed.

The filtration material consisted of coconut fibre with the addition of coarse pearlite at the volume ratio of 1:1. In order to feed the biofilter bed with biogenic salts, the mineral medium of fertilizer granules containing: (23%), P_2O_5 (9%), K_2O (10%), MgO (2%), Cu (0,005%), Fe (0,05%), Mn (0,03%), Zn (0,005%) was added. The filter bed was regularly moistened (water was introduced from the top of the biofilter) in order to sustain its humidity at about 60%. The study used the indigenous microflora inert for the filter material.

The concentrations of the aqueous solution of Tween 20, which were used to moisten the bed, were respectively: 200, 300 and 400 mg/dm^3. The solutions (2 dm^3 each time) were dosed after the bed adaptation period, on the 32nd, 46th and 60th day of the biofiltration process, respectively (during the adaptation period, the bed was moistened only with water). For each surfactant concentration, the experiment was carried out for 14 days, during which the toluene concentration at the inlet to the biofilter was gradually increased. At the same time, a toluene biofiltration control series was carried out in other column with identical parameters, in which the bed was moistened with water without the addition of surfactant.

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

Results and discussion

Figure 2 presents the influence of different concentrations of Tween 20 above CMC at different bed loads on the effectiveness of biofiltration of air contaminated with toluene vapors. The toluene load on the bed was gradually increased for each measurement series.

For increasing values of bed loading with toluene substantial decreases in biofiltration efficiency have been observed. The results (only results after the bed adaptation period have been included – after starting surfactant dosing) were compared to the control series (the biofiltration of toluene without the addition of surfactant – bed moistened with water) (Figure 3). The obtained maximum biofiltration rates were 21.2 $g/m^3/h$ and 19.8 $g/m^3/h$ for the control bed and for the bed to which Tween 20 solution was dosed, respectively. During the Tween 20 dosing trial no improvement in toluene biofiltration was observed. Similar results were obtained during the experiment performed with beta-cyclodextrin. The maximum biofiltration rate was 20 $g/m^3/h$ (Sówka et al. 2016). For comparison, biofiltration of toluene using other surfactants resulted in the following biofiltration rates: 32.3 $g/m^3/h$ (Betaine used) (Miller et al. 2019), 26.2 $g/m^3/h$ (Triton X-100 used) (Sówka et al. 2016) and 25.3 $g/m^3/h$ (Brij 35 used) (Miller et al. 2018). All these surfactants were dosed analogously at concentrations above CMC. Similar results were obtained by Wu-Chung i Hui-Zheng (Wu-Chung & Hui-Zheng, 2009) in biofiltration of toluene using Brij 35. The biofiltration rate was from 17.4 to 26.1 $g/m^3/h$ (at a range of surfactant concentration from 0 to 540 mg/dm^3).

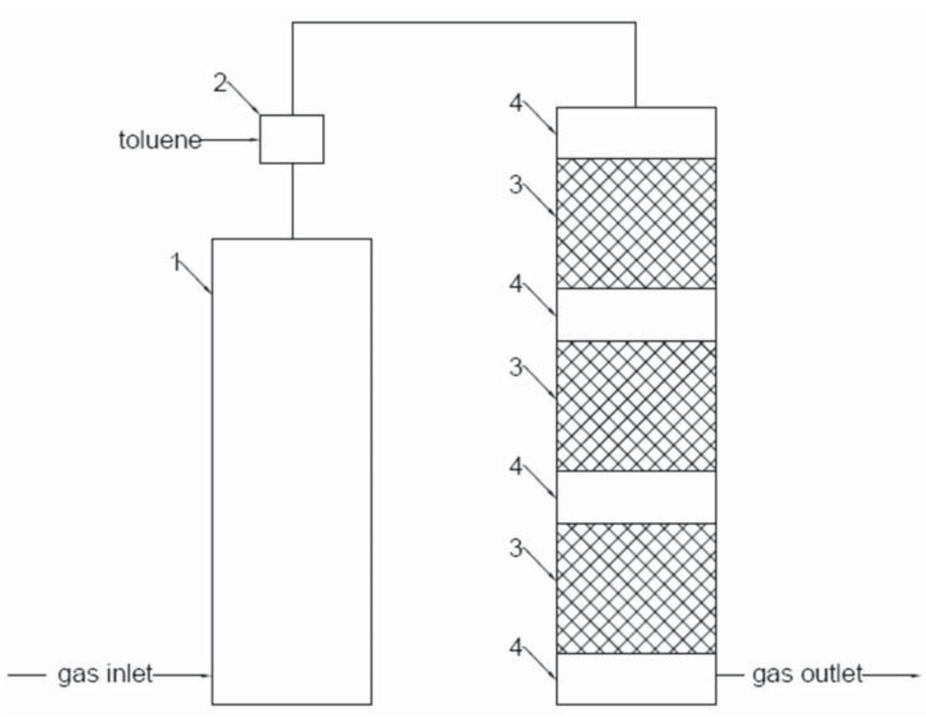

Fig. 1. Research installation scheme: 1 – column for gas pre-conditioning, 2 – mixer, 3 – filter bed, 4 – ports (Miller et al. 2019)

The biofiltration process is determined by many factors and leads to the large variety of the results obtained. According to the authors, the standard biofiltration rate in the uninoculated bed ranges from 14 to 55 g/m³/h (Szklarczyk et al. 2008). For example, Wieczorek (Wieczorek 2001) reached the maximum biofiltration rate of toluene with p-xylene at 40 g/m³/h. But research results published by other authors are very divergent. Park and Jung (Park & Jung, 2006) obtained a biofiltration rate of only 2.2 g/m³/h for a ceramic bed, while others (Singh et al. 2010) even 873 g/m³/h for biofilters filled with organic material in which the bed was inoculated. Therefore, the results can be compared for experiments carried out under similar conditions. These results show directions of actions aimed at optimizing the operation of biofilters on an industrial scale.

No toxic effect of surfactant on the biofilter microflora was revealed as the result of the quantitative settlement of the bed by microorganisms. A slight increase in the total number of microorganisms was observed from 50 × 10⁶ CFU/g d.m. (the control bed) up to 98 × 10⁶ CFU/g d.m. (the bed after dosing Tween). Alike is the result for fungi – respectively from 33 × 10⁶ CFU/g d.m. up to 48 × 10⁶ CFU/g d.m. (d.m. – dry mass).

Conclusion

Reducing interfacial tension and forming micelles, surfactants can improve the bioavailability of hydrophobic organic compounds. In the tested concentration range of Tween 20 no positive effect on the biofiltration of toluene has been demonstrated. The effect of Tween was neutral, it did not affect the effectiveness (the maximum biofiltration rate was 21.2 g/m³/h) or limited the development of the biofilter

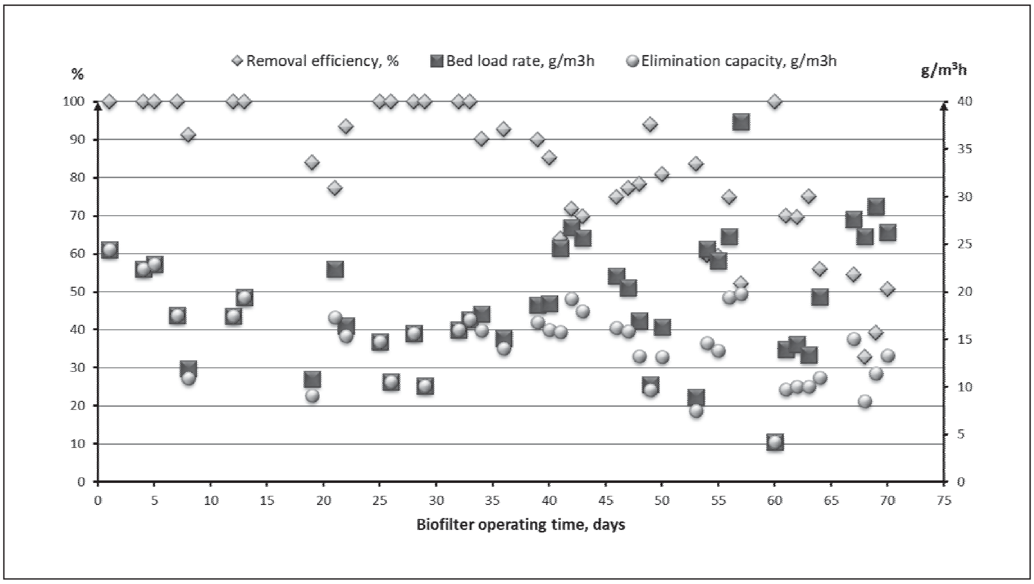

Fig. 2. Dependency between the elimination capacity and removal efficiency and the bed load of toluene for biofilter with Tween 20

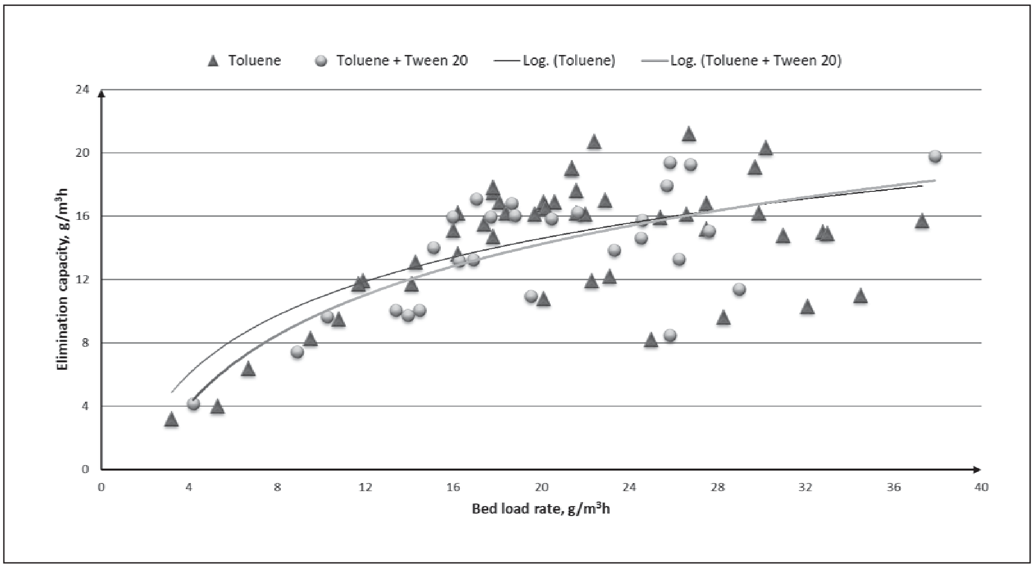

Fig. 3. Dependency between the bed load rate and elimination capacity compared to the control series

microflora (the total number of microorganisms increased to 98 × 10^6 CFU/g d.m.). Further researches should study whether at higher concentrations levels the surfactant improves the solubility of toluene in water without reversely affecting the population density of the bed.

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