TOLUENE AND STYRENE REMOVAL FROM AIR IN BIOFILTERS

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1. INTRODUCTION

BTEX (Benzene, toluene, ethylbenzene and the isomers of xylene) and styrene are among the 50 largest-volume industrial chemicals produced in the world. Each of them is produced at a rate of millions of tons per year [1,2]. They are widely used as fuels and solvents and provide starting materials for the production of resins, polymers, plastics, explosives, agrochemicals, and pharmaceuticals. Because of their natural ubiquitous presence in the environment and their widespread release through industrial and agricultural activities, the biodegradation of these compounds has been widely studied. These compounds are quoted by the US Environmental Protection Agency (EPA) as priority environmental toxic pollutants, due to their substantial toxicity and their carcinogenic potential, even at low concentrations.
The control of volatile organic air toxic emissions from industrial facilities has become critical and expensive to the chemical industries, particularly the small sized ones, in order to meet the more and more severe quality standards. The biotechnological approach to air pollution is now a promising field of research which can supply reliable, simple and cheap technologies for the prevention of air contamination.
The reliability of biological processes and, in particular, of biofiltration for the treatment of waste gas streams containing volatile organic compounds was demonstrated in a very large number of experimental studies [1, 3-6].
The aim of this study is to investigate the ability of the selected strains to remove toluene and styrene vapours in biofilters to get sufficient data for a future macro-kinetic study as well as useful information for industrial application.

2. MATERIALS AND METHODS

2.1 Microorganisms and apparatuses for continuous experiments

Experiments were conducted in two identical continuously operating bench-scale biofilters, one inoculated with the toluene-degrading strain of *Acinetobacter sp*. NCIMB 9689, and the other with the styrene-degrading strain of *Rhodococcus rhodochrous AL* NCIMB 13259. The apparatuses used consisted of 0.65 m long cylindrical glass columns with an inner diameter of 0.05 m and a filter bed height of 0.50 m. The packing material was a mixture of sterilized peat and glass beads (diameter 5 mm) in a 4:1 volume ratio, which was supported by a fine perforated plate of ceramic, placed at the bottom of the column. The moisture content of the packing material was kept between 50 and 60%, either by bubbling the influent synthetic waste air in a humidification unit, or by periodically distributing via nozzle at the top of the packing material a mineral salts solution, flowing counter currently with the gas upward through the column. The biofilter columns were operated at room temperature (20-21°C) and the pH of the packing material was kept around the neutrality. The biofilters were inoculated with a cell suspension of the selected strain. The suspension was mixed with the dry packing material (previously sterilized) in the proportion of 25% by volume. Cell density in the inoculum was 6.0 g l^{-1}.

2.2 Analytical procedures

Toluene and styrene gaseous samples were analyzed with a Carlo Erba Model HRGC 5160 gas chromatograph equipped with a capillary column (25 m x 0.32 mm, Mega Laboratory) and a flame ionization detector connected with a computing integrator.
Cell mass concentration in the liquid phase was determined by filtering 10 ml of culture broth through tared 0.45 μm pore size 47 ± 3 mm diameter cellulose nitrate membrane filters (Sartorius AG. 37070 Goettingen, Germany). After washing with 10 ml 0.9% NaCl solution, the filters were dried at 105°C to constant weight and cooled in a desiccator prior to re-weighing.
Moisture content of the filter bed was determined by the dried weight method.

3. RESULTS

3.1 Continuous biofiltration tests

In order to test the toluene elimination capacity of the lab-scale biofilter, different sets of continuous experiments were made over a period of 10 months by changing both the superficial gas velocity and the toluene inlet concentration. In particular, four series of experiments were performed at 17.8, 35.7, 127.5, and 255 m h^{-1}, and testing, for each given superficial gas velocity, five different inlet toluene concentrations, namely 0.1-0.2, 0.4-0.5, 1.0, 2.0, and 4.0 g m^{-3}. All experiments were performed under non-sterile conditions. During the start-up phase, a control column was used in order to determine the starting abiotic removal due to the physical adsorption by the packing material. Toluene breakthrough occurred after 3h of operation.
Fig. 1 shows the experimental results of continuous tests carried out during the whole

experimental investigation. Each test, under a given set of operative conditions, lasted a period of about 10-12 days. In order to prevent a shock to the microorganisms due to excess substrate, the first set of experiments was performed at the lowest superficial gas velocity (17.8 m h^{-1}) as well as at the lowest range of inlet toluene concentration (0.1-0.2 g m^{-3}). During this period, a removal efficiency close to 100% was initially observed, which was followed by a gradual decrease and a final restoration of the high starting values. This behaviour is the result of the preliminary adsorption of the pollutant by the filter bed and of the subsequent biological action of the microbial cells.
Nearly constant values of degradation efficiency were reached in about 3 days of continuous operation at the lowest concentration range and the highest residence time. This short start-up period was likely due to the inoculation of the biofilter with a specific and adapted microorganism.
A final average degree of conversion close to 100% has been assured, which indicated biofiltration is a really efficient technique in the control of waste gases containing toluene at low concentrations.
After the efficiency of the system had been tested at low pollutant levels, the inlet toluene concentration was progressively increased to 0.4-0.5, 1.0, 2.0, and 4.0 g m^{-3}, while the residence time was decreased from 202 to 14 s, with the aim of evaluating the actual possibility of employing this system in the presence of variable and high toluene loads in waste gases.
The average degree of conversion calculated in these subsequent phases were surprisingly high and the system proved very stable during the whole continuous experimentation. This indicates the high capacity of the cells to adapt themselves to large variations of pollutant concentrations as well as an unexpected ability to survive at relatively high feed concentrations.
The lab-scale biofilter used for styrene vapours removal was run for a period of six months with styrene volumetric loading rates varying from 6.14 to 588 g $m^{-3}_{\text{packing material}}$ h^{-1}, corresponding to superficial gas velocities of 61.2 to 245 m h^{-1} and influent styrene concentrations of 0.05 to 1.2 g m^{-3}.
The experimental procedure followed for the continuous tests of styrene removal was similar to that followed for toluene. Thus, also in this case, in order to prevent a shock to the microflora, the first series of runs was conducted at the lowest superficial gas velocity (61.2 m h^{-1}) as well as at the lowest inlet styrene concentration (0.05 g m^{-3}). After twenty days of operation, the superficial gas velocity was progressively increased to 122 and 245 m h^{-1}, while the inlet concentration (0.05 g m^{-3}) was kept constant. Every series of tests lasted 10 days, during which a pseudo-steady state conditions were normally achieved within 10 h.
The same experimental procedure was followed for the other influent styrene concentrations investigated (0.2, 0.4, 0.8 and 1.2 g m^{-3}), in order to study the influence of both parameters on the elimination capacity. The performance of the bench-scale biofilter over a period of six months is reported in Fig. 2.
In this study, the deodorization of both toluene and styrene waste-gases was also considered. The deodorization is defined as the achievement of an outlet pollutant concentration below the olfactory threshold values, that are 8.8 10^{-3} g m^{-3} and 0.2 10^{-3} g m^{-3} for toluene and styrene respectively.

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4. DISCUSSION

4.1 Continuous tests of toluene and styrene biofiltration

The satisfactory removal capacity of the biofilters is demonstrated in Figs. 3 and 4, where the elimination capacity of toluene and styrene, respectively, are plotted versus the organic load at different inlet concentrations of these pollutants. From these results, it is evident that this parameter increases regularly with the organic load, with scarce relevance whether such a dependence is the result of an increase in C_{go} (inlet pollutant concentration) or of a decrease in residence time. In particular, a gradual and linear increase of the elimination capacity occurs up to a value of the organic load corresponding to about 100 g m^{-3} h^{-1} for toluene and 50 g m^{-3} h^{-1} for styrene. Beyond these values, the elimination capacity increases more slowly and reaches maximum values of about 220 and 60 g m^{-3} $_{\text{packing material}}$ h^{-1} for toluene and styrene, respectively. These thresholds were obtained for toluene at an inlet concentration of 4.0 g m^{-3} and at a superficial gas velocity of 127.5 m h^{-1}, and for styrene at 0.8 g m^{-3} and 245.0 m h^{-1}.

4.2 Deodorization tests

The average toluene and styrene inlet concentrations, at which the deodorization of the gas was obtained, were also determined. The maximum concentrations, at which the outlet pollutant concentration was reduced below the olfactory threshold values, were 1.99 g m^{-3} and 0.2 g m^{-3}, and were obtained at a superficial gas velocity of 17.8 m h^{-1} for toluene and 122.0 m h^{-1} for styrene, respectively. These seem to be very interesting results if one consider that biofiltration is usually considered a particularly effective technique for treating gaseous emissions containing contaminants only in relatively low concentrations (< 1.0 g m^{-3}).

References

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Figure 1 - Experimental results of continuous tests of toluene removal from air stream (g m^{-3}): (●) inlet toluene concentration; (○) outlet toluene concentration.

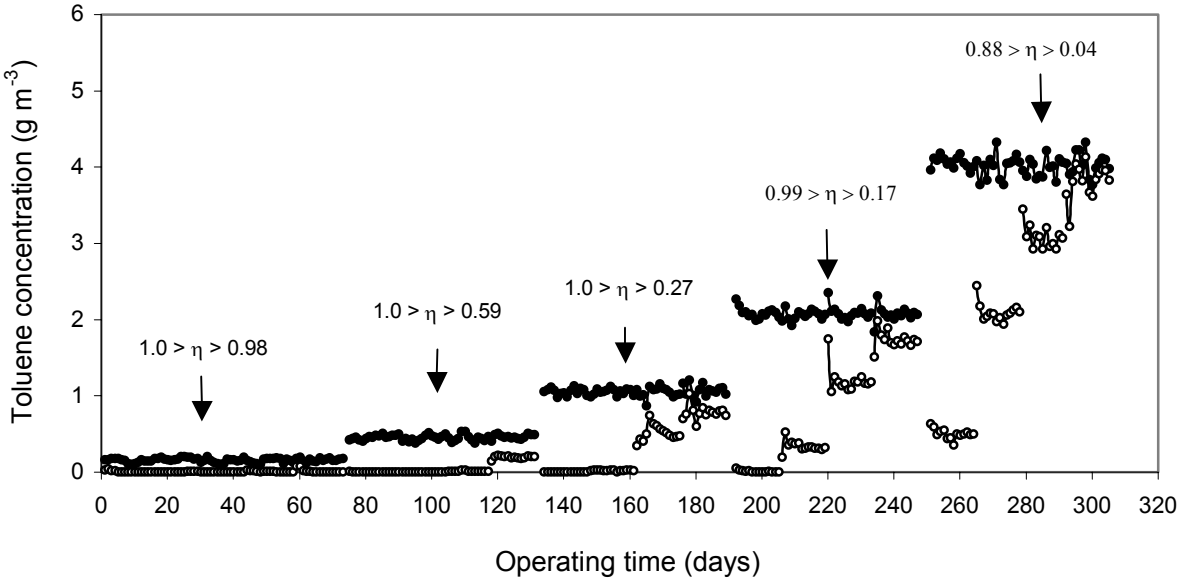

Figure 2 - Experimental results of continuous tests of styrene removal from air stream (g m^{-3}): (●) inlet styrene concentration; (○) outlet styrene concentration.

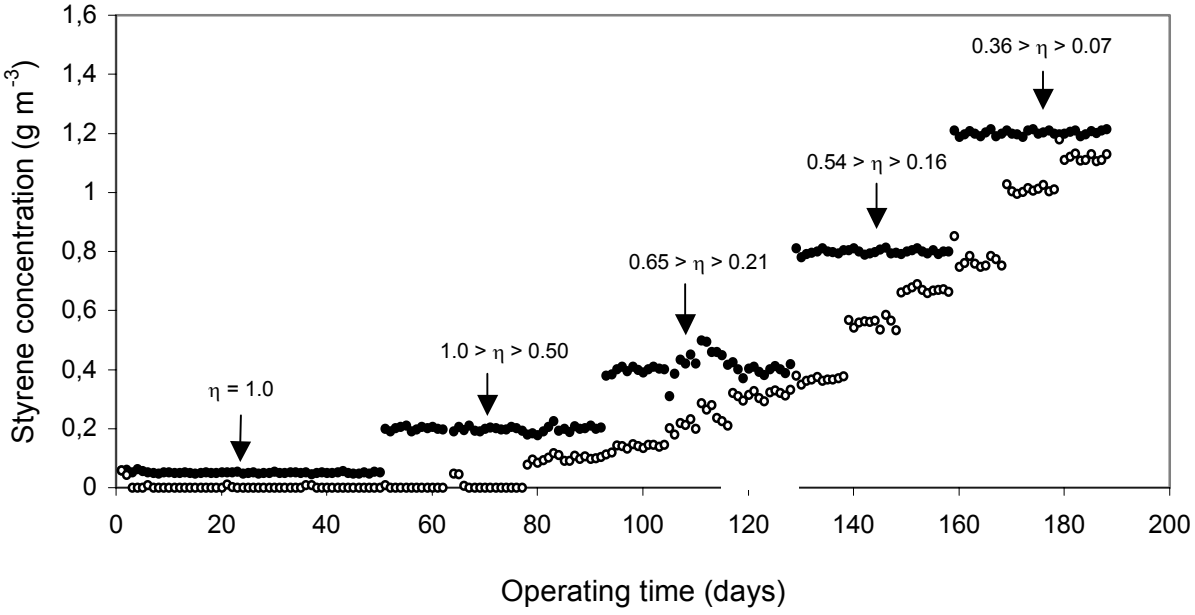

Figure 3 – Toluene elimination capacity of the biofilter versus the organic load. Inlet concentration (g m^{-3}): (▲) 0.1–0.2; (□) 0.4–0.5; () 1.0; (Δ) 2.0; (■) 4.0.

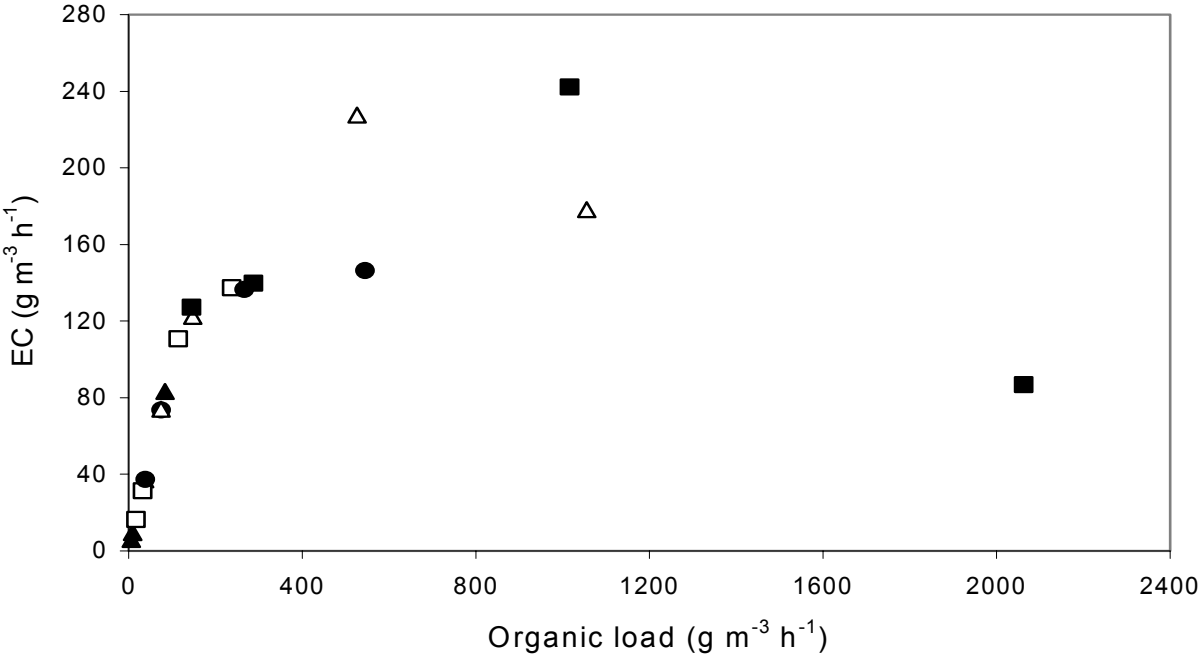

Figure 4 – Styrene elimination capacity of the biofilter versus the organic load. Inlet concentration (g m^{-3}): (▲) 0.05; (□) 0.2; (●) 0.4; (Δ) 0.8; (■) 1.2.