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Effective Removal of N-Butyl Methacrylate in Bio-Trickling Filter Packed With Activated Carbon Fibers

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Abstract **-** Removal of n-butyl methacrylate from gas streams was studied in a bio-trickling filter packed with plastic ring coated with activated carbon fibre as a new material. This new material utilized the stronger adsorption capacity of the activated carbon fibre to help the microorganisms in the bio-system capture the contaminants more easily. The performance of the bio-trickling filter was investigated under various operational conditions to better understand the role of activated carbon fibre. Results of the comparison experiments indicated that the use of activated carbon fibre could greatly shorten the start-up time of BTF. The removal efficiency of the bio-trickling filter could maintained at higher than 95% under the empty bed residence time of 44s and inlet concentration of 400 $mg\cdot m^{-3}$, with the corresponding removal loading of 54.3 $g\cdot m^{-3}\cdot h^{-1}$ and 52.9 $g\cdot m^{-3}\cdot h^{-1}$, respectively. When the inlet loading of n-butyl methacrylate was lower than 42.7 $g\cdot m^{-3}\cdot h^{-1}$, a complete removal of n-butyl methacrylate could be achieved in the bio-trickling filter. The good performance of the bio-trickling filter demonstrated that the plastic ring coated with activated carbon fibre is an alternative as the packing material in the full application of the bio-trickling filter.

Keywords**:** N-Butyl Methacrylate, Bio-Trickling Filter, Activated Carbon Fibre, Plastic Ring.

1. Introduction

Methacrylates are widely used as base materials for coating, paper-making, textiles or other binders in various manufacturing procedures (Kano et al., 2011; Graedel, 2011). Researchers have reported that esters were toxic and act as volatile organic compounds (VOCs) which can seriously affect the human health (Sun et al., 2014). Experiments have shown that 1 mL of methacrylate copolymer can be fatal to rats at a concentration of 40 mg mL^{-1} (Eisele et al., 2011). Therefore, the removal of these compounds, including nBM, is undoubtedly necessary.

Among the treating methods, biotechnology is frequently used due to its advantages, such as no secondary pollutants and low operational and investment costs (Krzysztof et al., 2017; Zhou et al., 2016; Lebrero et al., 2014; Padhi and Gokhale, 2016). Bio-trickling filters (BTFs) are one of the effective ways treating VOCs or odour gases (Rachbauer et al., 2016; Goli et al., 2016).

The performance of a BTF can be influenced by various factors, such as the structural type of the BTF, the humidity of the trickling bed, ambient temperature, species of microorganisms and packing materials (Yu et al., 2008; Alvarez-Hornos et al., 2017). Packing materials play an important role in a bio-trickling system; they provide living space for the bacteria, provide mechanical support, store nutrients and maintain a suitable humidity (Lópeza et al., 2016; Ferdowsi et al., 2017). The combined use of the activated carbon fibre and plastic ring may improve both the adsorption capacity and mechanical strength, when compared to activated carbon fibre or plastic ring used singularly (Yu et al., 2008; Wang et al., 2017). The BTF packed with these combinations may occupy a smaller area and run steadily for a longer time in the full application.

In this study, the performance of the BTF packed with the combination of plastic ring and activated carbon fibre in treating the nBM was explored. For comparison, another BTF packed with only plastic ring was used. The two BTFs were operated in parallel to better understand the role of the activated carbon fibre.

2. Materials and Methods
2.1. Bio-Trickling Filter System

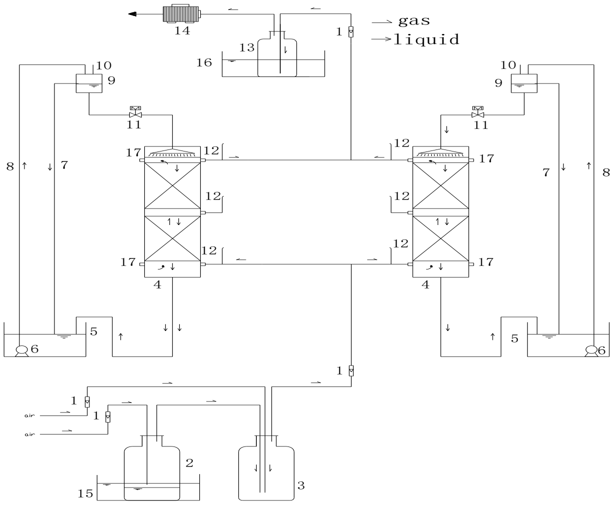

Fig. 1: Schematic of the experimental setup for the elimination of nBM (1. Flow metre; 2. nBM container; 3. Expansion tank; 4. Bio-trickling filter; 5. Nutrient solution; 6. Peristaltic pump; 7. Upflow tube; 8. Overflow pipe; 9. Surge tank; 10. Solenoid valve; 11. Gas sampling port; 12. Surge tank; 13. Air compressor; 14. Thermostatic waterbath).

The experimental setup contained two parallel BTFs which were made up of Plexiglas columns (0.085 m inner diameter × 1.3 m height). The trickling bed was divided into two segments with a working bed volume of 2.84 L (Fig. 1). Top and bottom free spaces (15 cm) were buffer zones. A nutrient tank was connected to the bottom of each trickling bed by a return line while a water pump was used to lift the nutrient solution to the top of the BTF to form a counter current mode with the gas flow. A nutrient solution was periodically sprayed over the medium bed at 150 ml h^{-1} from the top of each BTF. Simulative waste gas was generated by purging the target contaminant with an air compressor.

2.2. Packing Materials

Plastic ring was used as the packing material of BTFa while both the plastic ring and plastic ring wrapped in activated carbon fibre were used as the packing material of BTFb. Packing materials in BTFb were shaped to be cuboid with the length, width and height of 50 mm, 30 mm and 30 mm, respectively. The crevices between the cuboid materials were filled with plastic ring. Both the two packing materials were affordable in practical applications. The physical properties of the packing materials were tested and the results are shown in Table 1.

Table 1: Physical properties of packing materials.

Packing materials	Exterior	Diameter/thickness (mm)	Specific surface area ($m^2 \cdot g^{-1}$)	Average aperture (nm)	nBM adsorption capacity (mg ml^{-1})
Plastic ring	Spherical Microporous	Φ8	1.33	50	184.3
Activated carbon fibre	Thin sheet microporous	Th3	577.69	2.5	976.4

2.3. Microbes and Nutrient Solution

Nutrients were supplied to the microbes by spraying the nutrient solution onto the packing materials. The contents of the nutrient solution per litre of tap water were as follows, which was named Minimal Salt Medium (MSM): 0.11 g $K_2HPO_4{\cdot}H_2O$，0.16 g $Na_2HPO_4{\cdot}12H_2O$，0.04 g KH_2PO_4，0.03 g NH_4Cl，0.07 g $MgSO_4$，0.04 g $CaCl_2$，0.04×10^{-3} g $MnSO_4{\cdot}H_2O$，0.25×10^{-3} g $FeCl_3$，0.04×10^{-3} g $ZnSO_4{\cdot}H_2O$ and 0.03×10^{-3} g $Na_2MoO_4{\cdot}4H_2O$. All the chemicals were of analytical grade.

2.4. Analytical methods

N-butyl methacrylate concentrations in the gas flow at the inlet and outlet sampling ports were determined using a gas chromatography (GC) (HP 6890N, Agilent, USA) equipped with a flame ionization detector (FID). A HP-5 capillary column (30 m×2.5 µm, Agilent, USA) was used for the analysis. The injector, detector and oven temperatures were set at 100°C, 300°C and 60°C, respectively. All the data obtained were the average of three tests.

The pressure drop was measured using a U-tube water manometer connected to the top and the bottom of the BTFs, with an operational range of 0–10 kPa (0–100 cm H_2O).

The biomass was measured using the method of dry weight using maffle (SX-G16105) (APHA, 1998). The samples were randomly selected from different sections of the trickling bed and the biomass values were calculated from the average of three measurements.

3. Results and Discussion

3.1. Start-Up of Bio-Trickling Filters

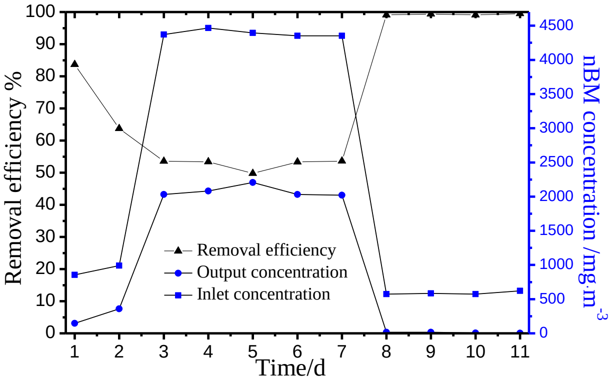

Fig. 2: Time course of the inlet and outlet concentration and the Re during start-up period.

A “three step immobilization method” was adopted to start-up the trickling filters in this study (Liu et al., 2013). At the initial start-up stage, the inlet gas flow was set at 0.1 $m^3\ h^{-1}$ and the corresponding EBRT was 106 s, with a concentration of 1000 mg m^{-3}. The nBM Re variation trends of the BTFs are illustrated in Fig.2. The high nBM Re and downward trend of nBM Re in the first three days revealed the slight adsorption capacity of the bio-trickling systems. A similar situation also occurred in a study about H_2S abatement in BTF packed with plastic ring (Liu et al., 2013). The more than 10% higher Re of BTFb at the first day showed the better adsorption capacity of activated carbon fibre. To accelerate the packing material adsorption rate and growth rate of the microorganisms in the BTFs, a high concentration of nBM was introduced into the bio-trickling system (Wu et al., 2008; Xue et al., 2013). The increase of the Re on the following days added further evidence that nBM sorption was dominant during the first three days, but was rapidly surpassed by biological degradation, which became the dominant mechanism for nBM abatement from Day 3 onwards. Concurrent with the packing materials reaching adsorption capacity saturation, the inlet concentration of nBM was adjusted to 700 mg m^{-3} on Day 8 to observe the performance of the BTFs. The EC of BTFb was considerably greater than that of BTFa, with removal loadings as high as 21.9 g nBM $m^{-3}h^{-1}$ being maintained (100% Re), compared with 15.0 g nBM $m^{-3}h^{-1}$ in BTFa. Higher Re of BTFb suggested that the larger surface area of the activated carbon fibre allowed more activated microorganisms to grow and correspondingly shortened the acclimation period.

3.2. Performance of BTFs under Different EBRT

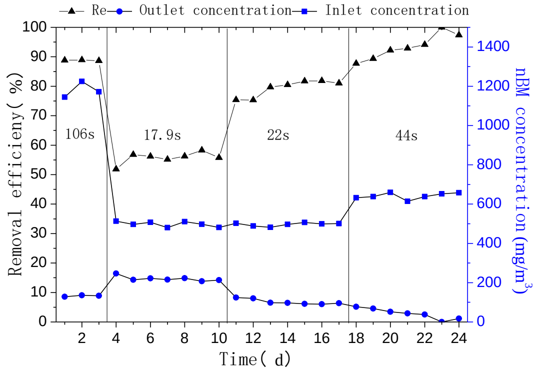

Fig. 3: Effect of EBRT on the removal efficiencies of the bio-trickling filter.

From Fig.3, it can be seen that the nBM removal efficiency of the biotrickling filter can reach 90% at the first day during the start-up period, indicating that the purification and acclimation of pure strains and activated sludge have a good degradation effect on nBM. And the microorganisms can grow quickly in the biotrickling filter. After 3 days of continuous operation, the purification efficiency of the biotrickling filter remained stable, indicating that the microorganisms can adapt to the environment of the biotrickling filter system and grow stably. The inlet concentration was reduced to 500 mg/m^3, and the EBRT was adjusted to 17.9 s. The change in the purification efficiency of the biotrickling filter over time was observed. As can be seen from Fig.3, after shortening the EBRT, the purification efficiency of the biotrickling filter is greatly affected, dropping to about 56%. It shows that under the lower EBRT, the biotrickling filter can not obtain better performance. With the EBRT adjusted to 22s, and the efficiency of the biotrickling filter rose to about 80%. For the comparison with the previous stage of experiments, the packing of activated carbon fiber wrapped plastic ring was continued to increase the EBRT to 44 s, and the change of the removal efficiency was observed. It can be seen that when the EBRT was 44 s, the nBM purification efficiency of the biotrickling filter was above 90%, and the maximum removal efficiency can reach 100%. The packing material brought the good performance of the bio-trickling filter.

3.3. Pressure Drop and Biomass

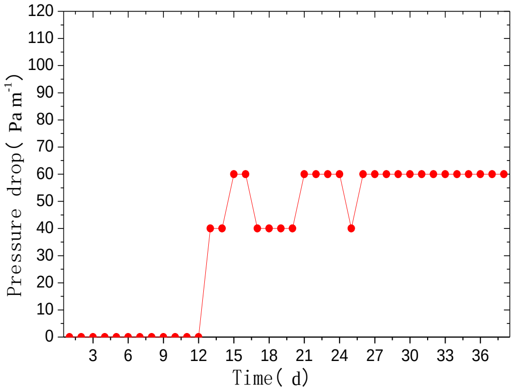

Fig. 4: Connection between biomass and removal efficiencies during different stages.

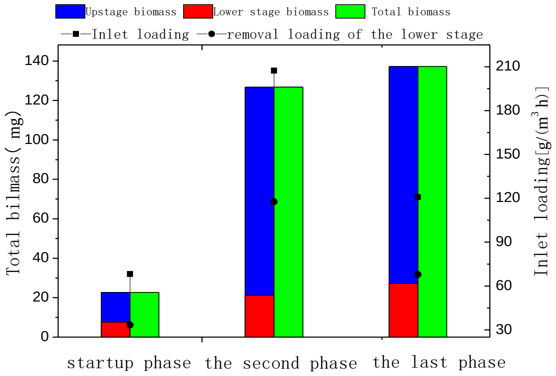

Fig. 5: nBM removal efficiencies of lower and upper stages.

Pressure drop is one of the key parameters that marks the performance stability of a BTF for long-term operation (Chen et al., 2016). Variations of pressure drops could indicate the biomass information, spaces among the packing materials and gas flow distribution of a BTF (Xue et al., 2013). As can be seen in Fig.4, it was obvious that pressure drop across the trickling bed was not detected during the first 12 days. However, the pressure drop exhibited the increasing trend from Day 13 onwards. This may be attributed to the greater biomass in the BTF, resulting in partial plugging of the trickling bed. An explanation for the excessive biomass growth in the BTF is that the better water-holding ability of activated carbon fibre supplied plenty of nutrients for the bio-film which resulted in the excessive growth of microorganisms and a rapid increase in the pressure drop. At the same time, the larger surface of the activated carbon fibre allowed more microorganisms to grow in the trickling bed.

From Fig.5, it can be seen that there is greater amounts of biomass in the lower layer compared to the upper layer in both the BTFs during different stages. A possible explanation for this is that there was a higher concentration of nBM in the lower layer, and a lower concentration in the upper layer, when the gas flowed through the trickling bed, as a result of the microorganisms in the lower layer degrading part of the substrate, which stimulated the growth of the degraders in the lower layer of the BTF (Chen et al., 2016). The degraders in the lower layer of the BTF took advantage of the favourable conditions which benefitted their growth. The lower layer of the BTF contributed the larger percentage of the total removal efficiencies. This phenomenon suggested that the lower section of the BTFs was more active.

4. Conclusions

This study investigated the performance of the BTF packed with plastic ring coated with activated carbon fibre as a novel material in treating nBM. The addition of the activated carbon fibre can shorten the start-up time of the BTF significantly. The Re of the BTF can be increased significantly at the EBRT lower than 44 s with a greater biomass. The larger biomass caused a greater pressure drop but still a higher Re of the BTF. Plastic ring coated with activated carbon fibre is a promising alternative as the packing material in the practical application of the BTFs.

Acknowledgements

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