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Original research article

Biodegradation Kinetics of Odorants from Block Rubber Drying Process Using Laboratory Scale Biofilter

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ABSTRACT

The aim of this study is to define the biodegradation kinetics of odorants from the block rubber drying process and the toleration of the biofilter system according to the inlet concentrations of the odorants. In this study, a lab-scale biofilter was applied to study the degradation of the synthetic odorant gas generated from the block rubber drying process. *Bacillus megaterium* bacteria was fixed on polyethylene pall ring media which was used as a fixed bed in the 105.98 liter lab-scale biofilter. Vapor of butyric acid, propionic acid and methyl mercaptan mixed with clean air at a ratio matching the exhaust gas from the block rubber drying process was used as a synthetic odorant gas stream. Moisture and nutrients were periodically added into the system.

The synthetic odorant gas with concentrations range of 130.60-499.79 ppm was introduced into the biofilter to analyze the degradation efficiencies and the kinetics of the system. The result shows that the lab-scale biofilter can remove odorants with 79.94% to 80.89% efficiencies. The concentration of odorants up to 305.44 ppm did not affect the removal efficiency at the same EBRT. The result indicated that the kinetic of the biodegradation of the synthetic odorants from block rubber drying process, having the coefficient of 0.0144, complied with the first order reaction.

Keywords: Kinetics; Biodegradation; Block rubber drying process; Biofilter

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1. Introduction

Block rubber is a product from the rubber drying process. In block rubber manufacturing, cup lump rubber is brought in as a raw material. The cup lump rubber, after cleaning, is shredded into small pieces and pressed into sheet rubber. The crepe rubber is then dried to eliminate moisture. In the last step, the rubber is cut and packaged. The final product is called block rubber [1-2]. Examples of environmental pollution from the block rubber manufacturing are wastewater from the washing process, and odors from the drying process. Although wastewater can be effectively treated, odor is still a problem to be resolved [3-6].

Exhaust gas from the block rubber drying process were analyzed for chemical compositions and odorants. Polycyclic aromatic hydrocarbons (PAHs), volatile fatty acids (VFA) and sulfur compounds were found [7]. Propionic acid, butyric acid and methyl mercaptan were the main concern compounds due to the high level of concentration and strong odor [8-9]. Odorants from the block rubber drying process are released to the environment impacting people in communities. The symptoms from odorants exposure are irritations to the respiratory system, sneezing, suffocating dizziness, nausea, headache, sleeplessness, etc. [10].

In Thailand, the highest complaints about pollution problems during 2003-2018 were odor. In 2017, odor contributed up to 45 % of the complaints. In Udon Thani province, where block rubber manufacturing is prevalent, 7 out of 13 complaints were about odor problems [11].

Biofiltration, an air pollution control technology, uses microorganisms in biofilm to cover packing media in order to degrade air pollutants. This technology costs less than other alternatives and can remove even very low concentration of odorants [12].

2. Methodology

2.1 Experiment

The operation of lab-scale biofilter starts from a zero air generator that generates clean air to supply 4 lines of the system. Zero air in line No. 1, 2 and 3 was passed through needle valves and flow meters to the impingers containing butyric acid, propionic acid and methyl mercaptan, respectively. This air stream will then carry vapor of pollutants to the mixing chamber. Line No.4, zero air, was passed to the mixing chamber to adjust the concentrations.

Pall ring media has a rather specific surface area and a high porosity that are suitable for microorganism growth and development of biofilm. *Bacillus megaterium* is recognized in a master's degree thesis of a student in the Faculty of Science and Technology, Thammasat University. *Bacillus megaterium* was isolated and selected from the wastewater sludge of the block rubber drying process in Udon Thani province, Thailand [13].

Clean air, propionic acid, butyric acid and methyl mercaptan in gaseous form are mixed as synthetic odorant in the mixing chamber. Afterwards, the synthetic odorant from the mixing chamber was passed through the impinger containing water and flowed to the inlet of the lab-scale biofilter. The synthetic gas passes through the packing media in the biofilter that was fixed with microorganism. The treated gas was measured by the MiniRAE 3000 analyzer for odorants concentrations through the sampling outlet at different heights. The nutrients solution in the bottle was periodically fed into the biofilter by a pump. The results of the experiment were determined for the empty bed residence times (EBRT) and the removal efficiencies (RE). EBRT and RE were analyzed for the kinetics of the biodegradation.

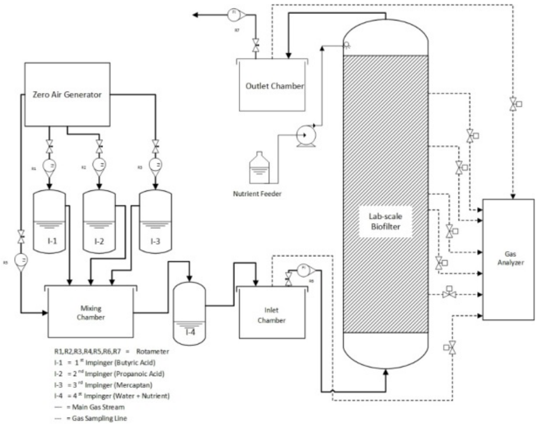

Fig. 1. Laboratory scale biofiltration system.

2.2 Kinetics of biodegradation

In a biofilter, synthetic odorant gas is transferred to the biofilm by a diffusion process. The microorganism then digests the synthetic odorant gas. The biodegradation rate depends on the removal efficiency (RE), the empty bed residence time (EBRT) and the inlet pollutant concentration (C_{in}). There are 3 types of kinetics as follows:

2.2.1 First-order reaction

Biodegradation rate is in a steady state condition while the synthetic odorant gas concentration is sensitive to the biodegradation rate. The removal efficiency of biofiltration depends on the empty bed residence time (EBRT). The first-order kinetic equation is developed as follows:

$$C_{out} = C_{in} exp(-K_1 EBRT), \quad (1)$$

$$\frac{C_{out}}{C_{in}} = exp(\frac{-EBRT \cdot A_s \cdot D_e}{m\delta} \varnothing \tanh \varnothing), \quad (2)$$

$$\varnothing = \delta \sqrt{\frac{K_1}{D_e}}, \quad (3)$$

$$RE = \frac{(C_{in} - C_{out}) \times 100}{C_{in}}, \quad (4)$$

$$RE = \left(1 - \frac{C_{out}}{C_{in}}\right) \times 100, \quad (5)$$

$$RE = \left(1 - exp\left(\frac{-EBRT \cdot A_s \cdot D_e}{m\delta} \varnothing \tanh \varnothing\right)\right) \times 100, \quad (6)$$

$$\ln(RE) = \ln 100 - (\frac{-EBRT \cdot A_s \cdot D_e}{m\delta} \varnothing \tanh \varnothing), \quad (7)$$

where C_{in} = The inlet pollutant concentration (g pollutant m^{-3}),
C_{out} = The outlet pollutant concentration (g pollutant m^{-3}),
K_1 = First-order rate coefficient (s^{-1}),
EBRT = The empty bed residence time (min),
RE = Removal efficiency (%),
A_s = Biofilm surface area,
D_e = Effective diffusion in the biofilm,
δ = Biofilm thickness.

First order reaction is considered by plotting-ln(*RE*) with (*EBRT*). If a straight line is formed, the first-order reaction is expected.

2.2.2 Zero-order reaction with reaction-limited

Biodegradation rate is in a steady state condition while the synthetic odorant gas concentration does not affect the biodegradation rate. The process of biodegradation rate is reaction- limited. Removal efficiency of the biofiltration depends on empty bed residence time (EBRT). The kinetic is zero-order reaction with reaction limited. The equation is developed as follows:

$$\frac{C_{out}}{C_{in}} = 1 - \frac{K_0 \cdot A_s \cdot \delta \cdot EBRT}{C_{in}}, \quad (8)$$

$$RE = \left(\frac{K_0 \cdot A_s \cdot \delta \cdot EBRT}{C_{in}}\right) \times 100, \quad (9)$$

where K_o = Zero-order rate coefficient.

Zero-order reaction with reaction limited is determined by plotting RE with EBRT/C_{in}. If a straight line is formed, zero order diffusion-limited is expected [12].

2.2.3 Zero-order reaction with diffusion-limited

The biodegradation rate depends on diffusion rate of the odorants to the biofilm on packing media. The equation is developed as follows:

$$\frac{C_{out}}{C_{in}} = \left(1 - EBRT \cdot A_s \sqrt{\frac{K_0 \cdot D_e}{2mC_{in}}}\right)^2, \tag{10}$$

$$\ln(RE) = \ln 100 + 2\ln 1 - \ln\left(EBRT \cdot A_s \sqrt{\frac{K_0 \cdot D_e}{2mC_{in}}}\right), \tag{11}$$

where $m =$ Air/biofilm distribution coefficient,

Zero-order reaction with diffusion limited is determined by plotting $\ln(RE)$ with $\ln\left(EBRT/\sqrt{C_{in}}\right)$. If a straight line is formed, then zero-order reaction with diffusion-limited is expected [14].

3. Results and Discussion

3.1 Experimental data

The result data from the experiment was used to determine the kinetics of the biodegradation of odorants from the block rubber drying process. Removal efficiencies of the system were found in the range of 43.87-79.94% with EBRTs of 3.53 - 21.20 s. The concentration of odorants in the range of 130.60-305.44 ppm did not affect the removal efficiency of the system. The results of the experiment are shown in Table 1.

Table 1. The result of concentration, EBRT and efficiency of the biodegradation for synthetic odorant from block rubber drying process.

Day	C_{in} (ppm)	EBRT (s)	Efficiency (%)
15	130.60	3.53	43.87
		7.07	50.23
		10.60	52.30
		14.13	58.50
		17.66	68.15
		21.20	79.94
16	211.26	3.53	43.25
		7.07	48.46
		10.60	52.77
		14.13	58.47
		17.66	69.15
		21.20	80.80
17	305.44	3.53	45.67
		7.07	48.37
		10.60	51.91
		14.13	60.72
		17.66	69.63
		21.20	80.89
18	405.95	3.53	20.82
		7.07	23.86
		10.60	25.91
		14.13	29.86
		17.66	42.84
		21.20	46.27
19	499.79	3.53	9.52
		7.07	10.92
		10.60	11.55
		14.13	12.97
		17.66	15.56
		21.20	20.82
20	499.79	21.20	1.04

However, when the concentration of the synthetic odorant increased to 405.95-499.79 ppm, the removal efficiency decreased to 20.82-46.27 %. The result showed that concentration of the synthetic odorant from 405.9 ppm and above may be toxic to the lab-scale biofilter, so, the concentration of synthetic odorant from block rubber drying process should not higher than 305.44 ppm.

3.2 Kinetics of the biodegradation of the synthetic odorant from block rubber drying process

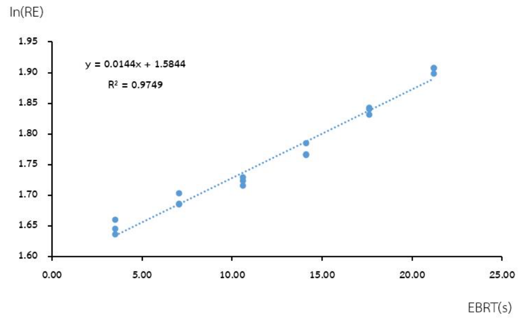

Fig. 2. Plotting between ln(RE) and $EBRT$ of the biodegradation for synthetic odorants from block rubber drying process.

First-order reaction was determined by plotting-ln(RE) with ($EBRT$). The result shows a coefficient of multiple determination (R^2) = 0.9747. The straight line equation was y = 0.0144x+1.5844 as shown in Fig. 2.

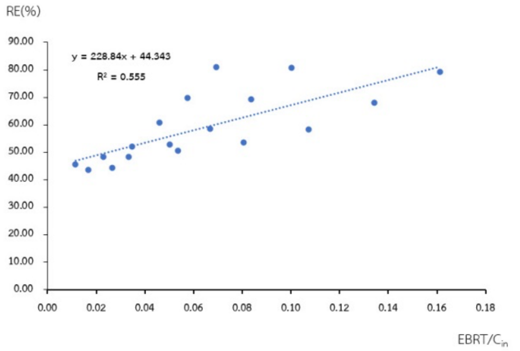

Fig. 3. Plotting between RE and EBRT/C_{in} of the biodegradation for synthetic odorant from block rubber drying process.

Zero-order reaction with reaction-limited was determined by plotting RE with $\left(EBRT/C_{in}\right)$. The result shows a coefficient of multiple determination (R^2) = 0.555. The straight line equation was y = 228.84x + 44.343 as shown in Fig. 3.

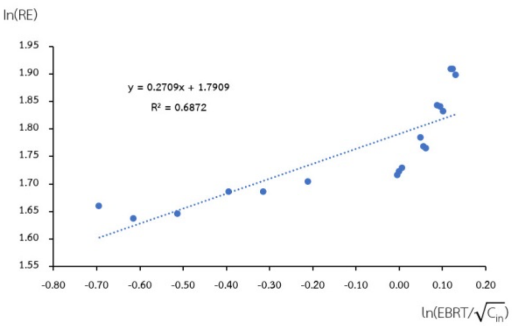

Fig. 4. Plotting between $\ln(RE)$ and $\ln\left(EBRT/\sqrt{C_{in}}\right)$ of the biodegradation for synthetic odorant from block rubber drying process.

Zero-order reaction with diffusion-limited was determined by plotting ln(RE) with $\ln\left(EBRT/\sqrt{C_{in}}\right)$. The result shows a coefficient of multiple determination (R^2) = 0.6872. The straight line equation was y = 0.2709x + 1.7909 as shown in Fig. 4.

When considered by Fig. 2 to Fig. 4, the straight line in Fig. 2 was the best fit. The result showed that the kinetic of the biodegradation of the synthetic odorants from the block rubber drying process complied with the first order reaction. Similar result of first order reaction was found by Shareefdeen et al. (2011). They studied the kinetics and modeling of H_2S removal in a novel biofilter by using a pilot biofilter column set-up at a pumping station located in the University City, Sharjah, UAE [15]. The first order reaction was also found and reported by Wang, Kolar, Kastner and Herner (2008). They studied biofiltration kinetics of a gaseous aldehyde mixture using a synthetic matrix as packing material. hexanal, 2-methylbutanal, and 3-methylbutanal in a bench-scale biofiltration [16].

For this kind of kinetics, the concentration of the odorants is sensitive to the efficiency of the biofilter. Therefore, the inlet concentration should be carefully maintained and controlled in the range that this study performed.

Besides, in this study, the experiment was performed in a laboratory. The condition in the lab-scale biofilter was maintained according to literature reviews. Temperature, relative humidity and nutrients in the lab-scale biofilter were controlled. If the result of this experiment is applied for scaling up to the actual use and operation, the condition of the operation must be controlled in the ranges that have been performed in this study. The temperature was in a range of 25-30°C and the relative humidity was in a range of 80-95%.

4. Conclusion

The kinetic of the biodegradation of the synthetic odorants from block rubber drying process is found to be a first-order reaction. The first-order rate coefficient was 0.0144.

Biofiltration technology can remove the odorants from block rubber drying process with a concentration up to 305.44 ppm. The concentration that affects the efficiency of the lab-scale biofilter for synthetic odorant from block rubber drying process is 405.95 ppm.

Acknowledgements

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