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Compounds interaction on biodegradation of toluene and methyl ethyl ketone mixtures in a composite biofilter

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Compounds interaction on biodegradation of toluene and methyl ethyl ketone (MEK) mixtures in a composite bead biofilter was investigated. The biodegradation rate of two compounds in the exponential growth phase and stationary phase for the single compound and two compounds mixing systems was determined. For the single compound system, the biodegradation rate of two compounds in the microbial growth phase and stationary phase was inhibited with increasing inlet compound concentration. The inhibitive effect resulting from increasing the inlet compound concentration was predominated at higher inlet compound concentrations. For the two compounds mixing system, the microbial metabolic activity of biodegraded toluene in the microbial growth process and biochemical reaction process was inhibited as MEK was introduced. The inhibitive effect resulting from compounds interaction was more pronounced at higher inlet concentrations of both compounds. The maximum elimination capacity of toluene was reduced as MEK was introduced and that of MEK was not affected as toluene was introduced. Increasing concentration of MEK in the influent mixture strongly suppressed the biodegradation efficiency of toluene, while increasing concentration of toluene in the influent mixture had no effect on the biodegradation efficiency of MEK.

Key words: Toluene, methyl ethyl ketone, biodegradation, compounds interaction, composite bead biofilter.

INTRODUCTION

The removal of volatile organic compounds (VOCs) from a polluted air stream using a biological process is highly efficient and has low installation and operation/ maintenance costs. A spherical polyvinyl alcohol (PVA)/ peat/KNO₃/GAC) composite bead was prepared and was proven suitable as a filter material in the biofiltration process in our previous works (Chan and Lin, 2006; Chan and Peng, 2008). Acetone, methyl ethyl ketone (MEK), methyl isobutyl ketone (MIBK), methyl isopropyl ketone (MIPK) and toluene are widely used industrial chemicals. These compounds were designed as high-priority toxic chemicals. Large volumes of these compounds are released into the atmosphere during manufacturing processes every year, possibly endangering air quality and public health.

The industrial effluent air stream almost contains VOCs mixtures. The interaction resulting from various physical and chemical properties of VOCs would affect the removal efficiency of VOCs by biolfiltration. No noticeable adverse effect on removal of n-butyl acetate was seen, while it was impacted for a few hours in the case of MEK and methyl propyl ketone (MPK). Toluene removal was adversely impacted for a few days if the loading condition of biofilter was made on the basis of daytime operation with weekend shutdown (Moe and Qi. 2004). N-Butvl acetate and MEK were more rapidly biodegraded than toluene and o-xylene in a coal based biotrickling filter for the removal of paint solvent mixtures (Mathur and Majumder, 2008). The biofilter easily acclimated to the oxygenated compounds (MEK and MIBK), while reacclimation was delayed for the aromatic compounds

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(toluene and styrene) (Cai et al., 2006). The elimination capacity of styrene, MEK and MIBK was not affected significantly by the presence of the other VOCs, but it was not so in the case of toluene; it was significantly affected (Cai et al., 2007). The presence of ethyl acetate in the system significantly reduced the removal capacity of toluene in the biofiltration of ethyl acetate and toluene mixtures. However, the removal efficiency of ethyl acetate was not affected by the presence of toluene in air streams. The biofilter acclimated with ethyl acetate had higher elimination capacity for ethyl acetate than the biofilter acclimated with toluene (Liu et al., 2002). In the biofiltration of ethyl acetate, isopropanol, and toluene mixtures, their maximum elimination capacity (EC) was 350, 150 and 20 g h⁻¹ m⁻³, respectively. The lower removal rate of toluene resulted from the inhibition by the presence of ethyl acetate and isopropanol (Liu et al., 2005). Ketone compounds were more rapidly degraded than aromatic compounds for the biofilter treating a gas mixture containing acetone, methyl ethyl ketone, toluene, ethylbenzene and p-xylene (Qi and Moe, 2006). The biodegradation of hydrophobic toluene was interfered by hydrophilic ethanol dissolved in the biolayer. The degree of interference was proportional to the inlet load of toluene and ethanol, and it was inversely proportional to the solubility of ethanol. The existence of toluene could hardly inversely affect the removal of ethanol in the biofilter (Lim, 2005). The increasing concentration of aromatic compounds (toluene and xylene) in the biotrickling filter influent mixtures had no effect on the degradation efficiency of ketones and ester compounds (methyl ethyl ketone, methyl isobutyl ketone and n-butyl acetate), but it caused a significant drop in the removal efficiency of aromatic compounds (Paca et al., 2007).

Toluene and MEK are commonly mixed together in the effluent stream of the chemical plants, paint and ink, semi-conductor and optic-electronic manufacturing. Toluene and MEK are designated as hydrophobic and hydrophilic compounds, respectively. We had indicated that the exponential growth (EG) and stationary phases (SP) were important for controlling the removal efficiency of a biofilter. Hence, we studied the biodegradation kinetic behaviors of the EG/SP for a single compound in a composite bead biofilter (Chan and Lin, 2006; Chan and Peng, 2008). It is well recognized that the metabolic activity on the biodegradation of two or more pollutants may involve the mechanism of induction, inhibition or cometabolism, depending on the substrates and microbial species present. However, details of the biodegradation kinetic behaviors of hydrophobic and hydrophilic compounds mixture in a biofilter are scant. This article investigates the biochemical kinetic behaviors of toluene and MEK mixtures in a composite bead biofilter which is composed of spherical PVA/peat/KNO₃/GAC composite bead. The effect of inlet compound concentration and compounds interaction on the microbial growth rate and studied. The biochemical reaction rate was inlet concentration of toluene and MEK varied from 500 to

2000 ppm, respectively.

MATERIALS AND METHODS

Peat (industrial grade from KekkilaOyj, Tuusula, Finland) was dried at 105°C before use. It has a dry density of 90 kg m⁻³, a pH of 5.5, a pore volume of 96%, and an organic substance content of 91%. Boric acid, sodium monobasic phosphate, sodium dibasic phosphate, potassium nitrate, methyl ethyl ketone and toluene (extra pure grade from Union Chemical, Hsinchu, Taiwan) were used as received. Poly vinyl alcohol (PVA) powder (industrial grade from Chung Chun Petrochemical, Hsinchu, Taiwan) and granular activated carbon (GAC) (industrial grade from Taipei Chemical, Hsinchu, Taiwan) were also used as received.

The procedures for preparing PVA/peat/GAC/KNO3 composite beads and the apparatus and operation of the biofilter system were described in Chan and Lin (2006) and Chan and Peng (2008). Toluene and methyl ethyl ketone were used as VOCs. They were individually poured into each Erlenmeyer flask. Before packing, the filter material was immersed in 0.384 M KNO₃ aqueous solution to adsorb KNO₃ and to reach equilibrium (approximately 12 h). The bead moisture content was humidified to more than 1.5 g water g⁻¹ dry composite bead and the seeding was performed with activated sludge obtained from the sludge thickener of an industrial wastewater plant. The inlet gas stream was designed to a single compound system and a two compounds mixing system. The single compound system was only one compound at a desired inlet concentration in the gas stream. The two compounds mixing system was one compound at a fixed concentration mixed with another compound at various concentrations in the gas stream. Each desired inlet VOCs concentration was obtained by adjusting the amount of evaporated VOCs using mass flow controllers. It was maintained at this concentration during the period of biofilter operation. The gas flow rate was maintained at 0.024 m³ h⁻¹ for all experiments and consequently, the empty bed residence time (EBRT) of biofilter column was 120.6 s. As the stationary phase had maintained more than 3 days, the operation of biofilter was stopped according to the variations of the removal efficiency of VOCs. Then, new filter material was repacked and the operation procedures afore described were carried out to start another experiment with the desired inlet concentration. The inlet and exit air streams of each section were sampled using an air auto-sampling system equipped with a 10-port selector and a 1 ml loop 6-port injector (Valco Instrument, Huston, USA), and injected into a gas chromatography. The concentration of VOCs in the air stream was measured online and analyzed using gas chromatography (GC) (Model GC-8A from Shimadzu, Tokyo, Japan) equipped with a flame ionization detector (FID) and J & W DB-5 fused silica capillary column (30 m × 530 µm × 5 µm). The temperature of the injector, oven and detector were 200, 100 and 220°C, respectively. Nitrogen was used as a carrier gas. The calibration curves with the maximum concentration of 2500 ppm were established for each toluene and MEK measurement. The removal efficiency of the VOCs was calculated by the difference of the VOCs concentration between the inlet and exit gas streams. The relative standard deviation and relative error of the experimental measurements were less than 2 and 5%, respectively.

RESULTS AND DISCUSSION

The variations of VOCs removal efficiency with operation time for two compounds mixing system are shown in Figure 1 (only the MEK inlet concentration of 1000 ppm and the toluene inlet concentration of 1000 ppm are shown). The variations of VOC removal efficiency with



Figure 1. The variations of VOCs removal efficiency with operation time (t) for two compounds mixing system at their inlet concentration of 1000 ppm: (\blacktriangle) toluene, (Δ) MEK.

operation time appeared in three phases: Lag phase (phase I), exponential growth phase (phase II) and stationary phase (phase III) (Chan and Lin, 2006; Chan and Peng, 2008). Only the biochemical kinetic behaviors in the exponential growth phase and stationary phase was studied in this work.

Microbial growth process

In the exponential growth phase (phase II), the microbial growth rate increased exponentially and was represented by the following equation (Chan and Peng, 2008; Valsaraj, 1995):

$$\ln(C/C_0) = -k_g t \tag{1}$$

Where, C and C₀ are the concentration of VOCs in the exit and inlet air stream, respectively. A plot of $In(C/C_0)$ versus t should correspond to a straight line and k_g can be determined. The microbial growth rate (k_g) of two compounds in the single compound system and two compounds mixing system at various inlet concentrations was calculated from the data in phase II and Equation 1.

The variations of the ratio values of microbial growth rate of MEK to toluene ($k_{g, MEK}/k_{g, Toluene}$) with inlet MEK concentration (500 to 2000 ppm) at a fixed inlet toluene concentration of 1000 ppm is shown in Figure 2. The ratio

values of $k_{g, \text{ MEK}}/k_{g, \text{ Toluene}}$ of the single compound system decreased from 5.025 to 2.425 as the inlet MEK concentration increased from 500 to 2000 ppm. An increase in the inlet concentration generally, would enhance the transfer rate of the VOCs from the gas phase to the biofilm. This phenomenon leads more microorganisms to participate in the biodegradation. However, high concentrations of some recalcitrant VOCs may produce inhibitive effects on the metabolic activity of the microbial population (Leson and Winer, 1991). Therefore, the result indicated that the microbial metabolic activity of biodegraded MEK was inhibited and the inhibitive effect resulting from increasing the inlet MEK concentration predominated at higher inlet MEK concentration for the single MEK system. The result closely corresponded to the result reported in Chan and Peng (2008).

The ratio values of $k_{g, MEK}/k_{g, Toluene}$ of the two compounds mixing system increased from 8.041 to 40.528 as the inlet MEK concentration increased from 500 to 2000 ppm. This phenomenon was opposite to that in the single compound system. The result indicates that the $k_{g, Toluene}$ values decreased with increased inlet MEK concentration and the degree of $k_{g, Toluene}$ values decreased resulting from increasing the inlet MEK concentration was greater than that of $k_{g, MEK}$ values decreased resulting from increasing the inlet MEK concentration. The metabolism



Figure 2. The variations of $k_{g, MEK}/k_{g, Toluene}$ with inlet MEK concentration (500 to 2000 ppm) at a fixed inlet toluene concentration of 1000 ppm: (\Box) single compound system, (**•**) two compounds mixing system.



Figure 3. The variations of $k_{g, Toluene} / k_{g, MEK}$ with inlet toluene concentration (500 to 2000 ppm) at a fixed inlet MEK concentration of 1000 ppm: (\Box) single compound system, (**a**) two compounds mixing system.

of aromatic compounds (toluene and xylene) is much more complex when compared to that of oxygenated compound (MEK and MIBK) which are a more easily biodegradable source of carbon and energy for the cells (Paca et al., 2007). Ketone compounds (acetone and methyl ethyl ketone) were more rapidly degraded than aromatic compounds (toluene, ethylbenzene and pxylene). This may be because ketone compound has higher aqueous partitioning coefficients leading to faster mass transfer (Qi and Moe, 2006). Therefore, the microbial metabolic activity of biodegraded toluene would be inhibited as MEK was introduced and the inhibitive effect resulting from compounds interaction was more pronounced at higher inlet MEK concentration for the two compounds mixing system.

The variations of the ratio values of microbial growth rate of toluene to MEK ($k_{g, Toluene}/k_{g, MEK}$) with inlet toluene concentration (500 to 2000 ppm) at a fixed inlet MEK concentration of 1000 ppm is shown in Figure 3. The ratio values of $k_{g, Toluene}/k_{g, MEK}$ of the single compound system decreased from 0.243 to 0.217 as the inlet toluene concentration increased from 500 to 2000 ppm. The

result indicated that the microbial metabolic activity of biodegraded toluene was also inhibited and the inhibitive effect resulting from increasing the inlet toluene concentration predominated at higher inlet toluene concentration for the single toluene system. The result closely corresponded to the result reported in Chan and You (2010).

The ratio values of $k_{g, Toluene}/k_{g, MEK}$ of the two compounds mixing system decreased from 0.107 to 0.021 as the inlet toluene concentration increased from 500 to 2000 ppm. This phenomenon is the same as that in the single compound system. The result indicates that the k_{g, Toluene} value also decreased with increasing the inlet toluene concentration for the two compounds mixing system. The ratio values of $k_{g, Toluene}/k_{g, MEK}$ of the two compounds mixing system was smaller than that of the single compound system and the difference of the ratio values of $k_{\text{g, Toluene}}/k_{\text{g, MEK}}$ between the two systems was increased from 0.136 to 0.196 as the inlet toluene concentration increased from 500 to 2000 ppm. The result indicates that the microbial metabolic activity of biodegraded toluene would be inhibited as MEK was introduced and the inhibitive effect resulting from compounds interaction was more pronounced at higher inlet toluene concentration for the two compounds mixing system. Therefore, increasing the content of toluene (aromatic compounds) in the mixing system did not affect the biodegradation rate of MEK. The slope of the linear profiles in this concentration range for the single compound system and the two compounds mixing system were 1.920×10^{-5} and 6.372×10^{-5} ppm⁻¹, respectively. The result indicates that the inhibitive effect for toluene resulting from increasing the inlet toluene concentration in the two compounds mixing system was more pronounced than that in the single compound system.

Biochemical reaction process

In the stationary phase, the population of viable cells was at a relatively constant value. A biofiltration model that described elimination of individual carbon energy substrates steady state was proposed by Ottengraf. The three basic situations of the Ottengraf model was firstorder kinetics, zero-order kinetics with reaction limitation, and zero-order kinetics with diffusion limitation (Ottengraf and Oever 1983; Ottengraf, 1986). The corresponding equations expressed the rates of biochemical reaction for each situation as follows:

1. First-order kinetic:

 $\ln(C/C_0) = -k_1 \theta \tag{2}$

2. Zero-order kinetic with reaction limitation:

$$C_0 - C = k_0 \theta \tag{3}$$

3. Zero-order kinetic with diffusion limitation:

$$I - (C/C_0)^{1/2} = k_d \theta \tag{4}$$

Where, k_1 , k_0 and k_d are the rate coefficient of first-order kinetic, zero-order kinetic with reaction limitation and zero-order kinetic with diffusion limitation, respectively (Yang and Allen, 1994).

The substrate utilization rate by microbial activity was generally expressed by the Michaeilis-Menten relationship. Under the state of microbial growth, population does not change with time, three possible situations may be encountered in a biochemical reaction system (Yang and Allen, 1994): situation 1, if the substrate concentration was very low ($K_s >> C_0$), the reaction rate expression could be simplified to first-order kinetic; situation 2, if the substrate concentration was very high ($K_s << C_0$), the reaction rate expression could be simplified to zero-order kinetic; situation 3, if the substrate concentration C_0 was comparable with K_s , the reaction rate expression could not be simplified and it followed fractional-order kinetic, and the Ottengraf diffusion limiting model was found to be the most approximate expression.

In order to verify the biochemical reaction kinetic model, assume there was a plug air flow in the biofilter column and the following equation was derived from the Michaelis-Menten equation (Valsaraj 1995):

$$(C_0-C)/\ln(C_0/C) = V_m (\theta/\ln(C_0/C)) - K_s$$
 (5)

Where, K_s is the half-saturation constant and V_m is the maximum reaction rate. A plot of $(C_0-C)/\ln(C_0/C)$ versus $\theta/\ln(C_0/C)$ should correspond to a straight line, and K_s and V_m can be determined. The plot of $(C_0-C)/\ln(C_0/C)$ versus $\theta/\ln(C_0/C)$ for a single compound system is shown in Figure 4. The calculated Ks values for toluene and MEK were 262.08 and 131.36 ppm, respectively. The V_m values for toluene and MEK were 17.14 and 23.10 ppm s , respectively. The ratio values of C₀/K_s for toluene and MEK were found to be 1.91 to 7.63 and 3.81 to 15.23, respectively. The results indicate that the relationship of C₀ and K_s does not correspond to situations 1 or 2, and it corresponds to situation 3 for two compounds. Therefore, the concentration C₀ was comparable with K_s, and zeroorder kinetic with diffusion limitation was regarded as the most adequate biochemical reaction kinetic model in this study. The k_d value of two compounds in the single compound system and the two compounds mixing system at various inlet concentrations was calculated from the data in phase III and Equation 4.

The variations of the ratio values of biochemical reaction rate of MEK to toluene ($k_{d, MEK}/k_{d, Toluene}$) with inlet MEK concentration (500 to 2000 ppm) at a fixed inlet toluene concentration of 1000 ppm is shown in Figure 5. The ratio values of $k_{d, MEK}/k_{d, Toluene}$ of the single compound system decreased from 2.530 to 1.249 as the inlet MEK concentration increased from 500 to 2000 ppm. The



Figure 4. The plot of $(C_0-C)/\ln(C_0/C)$ versus $\theta/\ln(C_0/C)$ for single compound system: (•) toluene, (**■**) MEK.



Figure 5. The variations of k_{d, MEK}/k_{d, Toluene} with inlet MEK concentration (500 to 2000 ppm) at a fixed inlet toluene concentration of 1000 ppm: (\Box) single compound system, (**•**) two compounds mixing system.



Inlet toulene concentration (C₀, ppm)

Figure 6. The variations of $k_{d, Toluene}/k_{d, MEK}$ with inlet toluene concentration (500 to 2000 ppm) at a fixed inlet MEK concentration of 1000 ppm: (\Box) single compound system, (**•**) two compounds mixing system.

result indicates that the biochemical reaction rate of biodegraded MEK was also inhibited and the inhibitive effect resulting from increasing the inlet MEK concentration predominated at higher inlet MEK concentration for the single MEK system. The result closely corresponded to the result reported in Chan and Peng (2008).

The ratio values of $k_{d, MEK}/k_{d Toluene}$ of the two compounds mixing system increased from 2.710 to 7.485 as the inlet MEK concentration increased from 500 to 2000 ppm. This phenomenon is opposite to that in the single compound system. The result indicated that the k_d Toluene values decreased with increasing the inlet MEK concentration and the degree of kd, Toluene values decreased resulting from increasing the inlet MEK concentration was greater than that of kd, MEK values decreased resulting from increasing the inlet MEK concentration. Therefore, the biochemical reaction rate of biodegraded toluene would be inhibited as MEK was introduced and the inhibitive effect resulting from compounds interaction was more pronounced at higher inlet MEK concentration for the two compounds mixing system.

Therefore, the biochemical reaction rate of biodegraded toluene would be inhibited as MEK was introduced and the inhibitive effect resulting from compounds interaction was more pronounced at higher inlet MEK concentration for the two compounds mixing system.

The variations of the ratio values of biochemical reaction rate of toluene to MEK ($k_{d, Toluene}/k_{d, MEK}$) with inlet toluene concentration (500 to 2000 ppm) at a fixed inlet MEK concentration of 1000 ppm is shown in Figure 6. The ratio values of $k_{d, Toluene} / k_{d, MEK}$ of the single compound system decreased from 0.551 to 0.470 as the inlet toluene concentration increased from of 500 to 2000 ppm. The result indicates that the biochemical reaction rate of biodegraded toluene was also inhibited and the inhibitive effect resulting from increasing the inlet toluene concentration for the single toluene system. The result closely corresponded to the result reported in Chan and You (2010).

The ratio values of $k_{d, Toluene}/k_{d, MEK}$ of the two compounds mixing system decreased from 0.461 to 0.156 as the inlet toluene concentration increased from 500 to 2000 ppm. This phenomenon is the same as that in the single compound system. The result indicated that the $k_{d, Toluene}$ values also decreased with increasing the inlet toluene concentration for the two compounds mixing system. The ratio values of $k_{d, Toluene}/k_{d, MEK}$ of the two compounds mixing system was smaller than that of the single compound system and the difference of the ratio



Figure 7. The variations of elimination capacity (EC) with load for (A) toluene and (B) MEK : (\Box) single compound system, (•) two compounds mixing system at a fixed inlet MEK concentration of 1000 ppm, (\blacklozenge) two compounds mixing system at a fixed inlet toluene concentration of 1000 ppm, (---) 100% removal.

values of $k_{d, Toluene}/k_{d, MEK}$ between two systems was increased from 0.090 to 0.314 as the inlet toluene concentration increased from 500 to 2000 ppm. The result indicated that the biochemical reaction rate of biodegraded toluene would be inhibited as MEK was introduced and the inhibitive effect resulting from compounds interaction was more pronounced at higher inlet toluene concentration for the two compounds mixing system. The slope of the linear profiles in this concentration range for the single compound system and the two compounds mixing system were 5.568×10^{-5} and 2.268×10^{-4} ppm⁻¹, respectively. The result indicated that the inhibited effect resulting from increasing the inlet toluene concentration in the two compounds mixing

system was more pronounced than that in the single compound system.

Elimination capacity

Elimination capacity and load were calculated according to the following equations (Deviney et al., 1999): $EC = Q (C_0-C)/V$ (6)

$$Load = Q C_0 / V$$
 (7)

Where, Q is the flow rate of inlet air steam and V is the bed volume of filter material as packed. The relationship of elimination capacity (EC) of biofilter versus load for the

single compound system and two compounds mixing system is shown in Figure 7. The EC of toluene reached the maximum value and then it decreased with increasing the load of toluene while the EC of MEK reached the maximum value and then it maintained this value as the load of MEK increased for the two compounds mixing system. The result indicated that the removal capacity of toluene would be inhibited in the presence of MEK and the removal capacity of MEK was not affected in the presence of toluene. Therefore, the presence of MEK had a negative effect on the removal of toluene and the existence of toluene had no effect on the removal of MEK.

The maximum elimination capacity of toluene and MEK were 180.9 and 144.0 g C h⁻¹m⁻³ bed volume, respecttively for the single compound system. The maximum elimination capacity of MEK was 145.3 g C h⁻¹m⁻³ bed volume for the two compounds mixing system with a fixed inlet toluene concentration of 1000 ppm and inlet MEK concentration was in the range of 500 to 2000 ppm. The result indicated that the maximum EC of MEK in this mixing system was almost the same as that in the single MEK system in the same MEK inlet concentration range. The maximum elimination capacity of toluene was 144.7 g C h⁻¹m⁻³ bed volume for the two compounds mixing system with a fixed inlet MEK concentration of 1000 ppm and inlet toluene concentration was in the range of 500 to 2000 ppm. The result indicated that the maximum EC of toluene for this mixing system was less than that for the single toluene system in the same toluene inlet concentration range.

Conclusions

This work was to test the biodegradation of toluene and methyl ethyl ketone (MEK) individually, and as mixtures at various concentrations, and to investigate the interaction between the two compounds during their biodegradation in a composite bead biofilter. For the single compound system, the biodegradation rate of the two compounds in the microbial growth phase and stationary phase was inhibited with increasing inlet compound concentration. The inhibitive effect resulting from increasing the inlet compound concentration was predominated at higher inlet compound concentrations. The microbial metabolic activity of biodegraded toluene in the microbial growth process and biochemical reaction process was inhibited as MEK was introduced and the inhibitive effect resulting from compounds interaction was more pronounced at higher inlet concentrations of both compounds for the two compounds mixing system. The removal capacity of toluene was inhibited in the presence of MEK and that of MEK was not affected in the presence of toluene. Increasing concentration of MEK in the influent mixture strongl suppressed the biodegradation efficiency of toluene, while increasing concentration of toluene in the

influent mixture hardly affected the biodegradation efficiency of MEK.

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