

Review

Degradation and Removal of Methyl *tert*-Butyl Ether

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Abstract: Methyl *tert*-butyl ether (MTBE) has been used in many countries as an octane-enhancing replacement for lead, primarily in mid- and high-grade gasoline. The prevalent use of MTBE has led to its introduction into environment. MTBE is poorly adsorbed, biologically and chemically stable, making it very persistent in the environment. MTBE has been classified as a possible human carcinogen. Many studies have been carried out to find a better method for the degradation and removal of MTBE in the environment. Activated carbon, clay mineral, resin and zeolite have been used as adsorbents for removal of MTBE. Persulfate, potassium permanganate, Fenton oxidation and photocatalytic oxidation have been studied for chemical degradation of MTBE. In addition, different bacteria have been isolated and identified for biodegradation of MTBE. This review summarizes current knowledge on the adsorption, chemical degradation and biodegradation of MTBE.

Keywords: methyl *tert*-butyl ether; MTBE; adsorption; oxidation; degradation; biodegradation; Fenton; photocatalysis.

1. Introduction

Methyl *tert*-butyl ether (MTBE) has been used as a gasoline additive to improve air quality (Fiorenza and Rifai, 2003). The low cost, easy production, favorable transfer, and blending feature made it a most common oxygenate added to gasoline formulation (Atienza et al., 2005). It could increase the gasoline octane number, and the oxygen rate was also improved. At the same time, the ratio of carbon monoxide and volatile organic compounds in tail-pipe emission for the gasoline containing MTBE was lower than the use of other gasoline (Atienza et al., 2005). With the extension of lead free gasoline, the use of MTBE increases rapidly. The prevalent use of MTBE has led to its

introduction into environment, resulting from spill and leakage of underground storage tanks or petroleum pipelines. MTBE is biologically and chemically stable, poorly adsorbed by soil and has a high solubility in water, making it very persistent in the environment (Ji et al., 2009; Soltani and Moheb, 2010). MTBE may cause dyspnea, asthma, headache, dizziness, insomnia and rash, and the United States Environmental Protection Agency has classified it as a possible human carcinogen (Kamalan et al., 2009; Liang et al., 2010).

In order to remove MTBE from the environment, many techniques have been tested. It is very difficult to remove MTBE from water by aeration due to its low Henry's constant (Atienza et al., 2005; Fiorenza and Rifai, 2003; Lawrence and Larry, 2004), but some methods, such as adsorption, chemical degradation and biodegradation, could be useful for removal of MTBE from the environment. The degradation and removal methods of MTBE have become a research focus in recent years. In this paper, we summarize current knowledge on the adsorption, chemical degradation and biodegradation of MTBE.

2. Adsorption of MTBE

Adsorption is one of the most utilized and effective processes for the removal of some pollutants from water and wastewater. Activated carbon, clay mineral, resin and zeolite are usually used as adsorbents for adsorption of MTBE (Table 1).

As a common adsorbent, activated carbon has been widely studied to wipe off MTBE from water and wastewater (Gironi et al., 2003; Hung et al., 2005; Inal et al., 2009; Li et al., 2001). Activated carbon adsorption of fuel oxygenates MTBE and ETBE from water in a batch system at 27 °C was investigated, and headspace gas chromatography-mass spectrometry was used to determine the oxygenate concentrations (Inal et al., 2009). The adsorption capacity was calculated as 5.50 for MTBE at an equilibrium solution concentration of 1 mg L⁻¹ using Dubinin-Radushkevich isotherm, and Gibbs free-energy change of adsorption was -22.59 kJ mol⁻¹ under the experimental condition studied.

The physicochemical characteristics of three clay minerals (vermiculite, bentonite, and CTAB-bentonite) were studied through batch adsorption tests conducted under various experimental conditions such as time, pH, zeta potential, concentration of solutes and organic matters (Sim et al., 2009). The adsorption of MTBE was increasing in order of vermiculite, bentonite, and CTAB-bentonite. When the humic acid was added, the adsorption of MTBE on these clay minerals significantly increased.

Different resins, such as ion exchange resin, carbonaceous resin and solvent impregnated resin, have been studied for adsorption of MTBE (Bi et al., 2005; Burghoff et al., 2010; Hung et al., 2006; Ji et al., 2008). Ion exchange resin could adsorb and hydrolyze MTBE (Ji et al., 2008). Both Na-D008 and H-D008 ion exchange resins exhibited strong MTBE adsorption, and the adsorption followed the linear adsorption isotherm. MTBE hydrolysis over H-D008 was easy because the activation energy of the reaction was relatively low (124.8 kJ mol⁻¹). The hydrolysis reaction rate constants were independent on MTBE initial concentrations, but they increased with the increase of reaction temperature. In another study, two kinds of carbonaceous resin, Amborsorb 563 and 572, were tested, and both of them had the high adsorption capacities for MTBE (Hung et al., 2006).

Table 1. The techniques for the adsorption, chemical degradation and biodegradation of MTBE

Technique	Reference
adsorption	
activated carbon	Gironi et al., 2003; Hung et al., 2005; Inal et al., 2009
clay mineral	Sim et al., 2009
ion exchange resin	Ji et al., 2008
carbonaceous resin	Hung et al., 2006
solvent impregnated resin	Burghoff et al., 2010
zeolite	Hung et al., 2006; Lu et al., 2009; Yazaydin et al., 2006
chemical degradation	
persulfate	Chen et al., 2009; Huling et al., 2011; Liang et al., 2010
potassium permanganate	Damm et al., 2002
Fenton oxidation	Burbano et al., 2005 ; Han et al., 2009; Xu et al., 2004
photocatalytic oxidation	Hung 2006; Katsanou et al., 2008; Xu et al., 2006
sonolytic degradation	Neppolian et al., 2002
biodegradation	
aerobic biodegradation	Bianchi et al., 2009; Munoz-Castellanos et al., 2006
anaerobic biodegradation	Raynal et al., 2010; Youngster et al., 2010

Many zeolite composites have been studied for adsorption of MTBE from water and wastewater (Anderson, 2000; Hung et al., 2005 & 2006; Li et al., 2003; Lu et al., 2008 & 2009; Yazaydin et al., 2006). Most zeolites showed effective results for the sorption of MTBE, and some hydrophobic zeolites have shown a better performance than granular activated carbon, particularly in the $\mu\text{g L}^{-1}$ level (Yazaydin et al., 2006). Another study showed that the Silicalite-1/fly ash cenosphere zeolite composite could adsorb approximately 93-95% MTBE with an initial concentration of $1000 \mu\text{g L}^{-1}$ (Lu et al., 2009). In the adsorption process, many factors, such as pore volume, pore structure, aperture size, ratio of composition, adsorption time and temperature, and initial MTBE concentration, could play an important role (Anderson, 2000; Hung et al., 2005; Li et al., 2003; Lu et al., 2009; Yazaydin et al., 2006). The zeolite composites silicalite-1/fly ash cenosphere had a higher maximum adsorption capacity than silicalite-1/diatomite, due to the higher amount of silicalite-1 coated on its outer surface (21%) than that on silicalite-1/diatomite (12%) (Lu et al., 2008). The amount of MTBE adsorbed on allsilica beta increased linearly with MTBE concentration from 9.4 to $590 \mu\text{g L}^{-1}$ (Li et al., 2003).

3. Chemical Degradation of MTBE

Persulfate, potassium permanganate, Fenton oxidation, photocatalytic oxidation and sonolytic degradation have been widely studied for the degradation of MTBE (Table 1) (Chen et al., 2009; Damm et al., 2002; Gonzalez-Olmos et al., 2009; Hetflejs et al., 2007; Huling et al., 2011; Lee et al., 2010; Liang et al., 2010).

Persulfate is one of the strongest oxidants. However, reactions of persulfate with organic pollutants are usually slow at ambient temperature, and activation of persulfate is necessary to

accelerate the process. Persulfate can be activated to generate sulfate free radicals by thermal, chemical and photochemical techniques, such as heat, UV light, transitional metal, activated carbon, microwave and integrated activation techniques (Chen et al., 2009; Huling et al., 2011; Liang et al., 2010; Xu et al., 2012). Thermal activation of persulfate was effective and resulted in greater MTBE removal than either alkaline activation or H₂O₂-persulfate binary mixture (Huling et al., 2011). In another study, pyrite (FeS₂) was used to activate persulfate (Liang et al., 2010). The result indicated that MTBE might be completely degraded within 4 hr. if that does of FeS₂ and persulfate were sufficient. In addition, effects of various factors on the efficiency of MTBE degradation by ferrous ion-activated persulfate were studied, including concentrations of persulfate and ferrous, and persulfate coupled with hydrogen peroxide (Chen et al., 2009). Excess addition of ferrous ion might result in the decrease of MTBE degradation rate, most likely because of competition for sulfate free radicals between ferrous ion and MTBE. The intermediate products of MTBE degradation, including *tert*-butyl formate, *tert*-butyl alcohol, methyl acetate, and acetone, could be degraded subsequently (Chen et al., 2009; Liang et al., 2010). In the literature, potassium permanganate was also tried for degradation of MTBE, and the oxidation rate was slower than other oxidants (Damm et al., 2002). The slower rate of MTBE oxidation by permanganate limited the applicability of this process for rapid MTBE cleanup.

Fenton's reagent is a mixture of H₂O₂ and Fe²⁺, which can generate great amount of hydroxyl free radicals with powerful oxidizing ability. Degradation of MTBE in aqueous solution by Fenton's reagent was investigated (Xu et al., 2004). The initial 1 mM MTBE solution was reduced by 99% within 120 min under the optimum condition, and several compounds including *tert*-butyl formate, *tert*-butyl alcohol, methyl acetate and acetone were identified to be the primary intermediates and by-products of the degradation process. In another study, effect of pH on MTBE degradation in the batch reactors under initially anaerobic conditions was investigated (Burbano et al., 2005). The experiments performed at acidic pH exhibited the best degradation results, while at neutral pH the degradation rates dropped significantly. In addition, two types of Fe-containing zeolites (Fe-ZSM5 and Fe-Beta) as catalysts for chemical oxidation of MTBE in water with H₂O₂ were studied (Gonzalez-Olmos et al., 2009). The Fe-Beta zeolite showed a lower catalytic activity for hydrogen peroxide, but in terms of utilization of hydrogen peroxide for MTBE degradation Fe-Beta is better. Maybe Fe-Beta zeolite owns a stronger adsorb ability of MTBE. Moreover, the initial concentrations of MTBE and hydrogen peroxide, or the presence of some inorganic anions also could affect the MTBE degradation differently (Han et al., 2009). The accretion of MTBE played a negative role in the degradation process, while enlargement of the hydrogen peroxide amount had a positive affection to the degradation. Some anions, such as PO₄³⁻, HPO₄²⁻, H₂PO₄⁻, Cl⁻ and SO₄²⁻, could suppress the decomposition of MTBE. In the literature, effects of temperature and acidic pre-treatment on Fenton-driven oxidation of MTBE-spent granular activated carbon (GAC) were also reported (Kan and Huling, 2009). The Fenton-driven reaction contributed to greater oxidation and removal of MTBE in GAC, and hydrogen peroxide reaction and MTBE removal in GAC increased with the increasing temperature.

Photocatalytic oxidation is another advanced oxidation process, and has been widely studied for degradation of MTBE (Eslami et al., 2008; Hung 2006; Katsanou et al., 2005 & 2008; Xu et al., 2006). In the UV/TiO₂ system, alkaline solution supplied a better place for the reaction of MTBE degradation, while the H₂O₂-UV/TiO₂ system put up faster degradation rate in acidic solution (Hung, 2006). In addition, the addition of H₂O₂ to UV/TiO₂ system could improve the degradation rate of MTBE

because H_2O_2 could be a more efficient electron acceptor than oxygen for retarding recombination of electron-hole pairs. Furthermore, the reaction was more enhanced when the pH was below 5. However, after adding too much H_2O_2 to the system, the rate would independent of H_2O_2 dosage. In another study, two important environmental pollutants, MTBE and hexavalent chromium, were simultaneously decontaminated by UV/TiO₂ process (Xu et al., 2006). Under UV irradiation, Cr(VI) reduction was observed in Cr(VI)/TiO₂ system, and MTBE oxidation was observed in MTBE/TiO₂ system. Moreover, the system containing Cr(VI) and MTBE by UV/TiO₂ process demonstrated the synergistic effect between oxidation of MTBE and reduction of Cr(VI).

4. Biodegradation of MTBE

Many studies have been carried out to isolate and identify microorganisms capable of degrading MTBE, and to investigate effects of environment factors on biodegradation of MTBE (Table 1). Different kinds of microorganisms for biodegradations of MTBE have already been reported in recent years. The common aerobic classes include *Bacillus*, *Rhodococcus* and *Micrococcus*, and the usual anaerobic types contain *Proteobacteria*, *Pseudomonas* and *Thermotogae*.

4.1. Isolation of Microorganisms for Biodegradation of MTBE

Many studies have been carried out to find microorganisms that can degrade MTBE (Mang et al., 2007; Nikpey et al., 2006; Raynal et al., 2010; Youngster et al., 2010). The usual method to obtain MTBE-degrading strains was enrichment culture and 16S rRNA genes cloned and sequenced. Munoz-Castellanos et al. (2006) isolated 59 different bacterial strains by enrichment culture with three consecutive selective transfers from a gasoline-contaminated soil at a service station, and identified them by biochemical and morphological characterization. They were classified as *Bacillus*, *Rhodococcus*, *Micrococcus*, *Aureobacterium* and *Proteus*. Six strains could significantly reduce MTBE. In another study, Youngster et al. (2010) analyzed the bacterial communities in anaerobic-enriched cultures originating from three different contaminated sediments that have retained MTBE-degrading activity for over a decade. Terminal restriction fragment length polymorphism (T-RFLP) analysis of bacterial 16S rRNA genes showed that multiple species are capable of MTBE biodegradation. In addition, Arabi et al. (2007) got two pure bacteria strains that could degrade MTBE rapidly under aerobic conditions. The strains were identified as *Bacillus cereus* and *Klebsiella terrigena*. Both strains could be able to grow in the presence of high concentration, even in the maximum concentration of MTBE (e.g. 48 g L⁻¹) in water. The low concentration (e.g. 10 g L⁻¹) of MTBE was able to degrade completely in less than a day. At optimum conditions the specific degradation rate of MTBE were 5.89 g (MTBE) g (cells)⁻¹ h⁻¹ for *B. cereus* and 5.78 g (MTBE) g (cells)⁻¹ h⁻¹ for *K. terrigena*. The cultivations were carried out successfully at 25, 30 and 37 °C while they showed the best performance at 37 °C. Both the strains are aerobic bacteria, and were not able to grow and degrade MTBE anaerobically. Moreover, Lin et al. (2007) got three pure strains from a petrochemical wastewater treatment plant through a series of screening and isolation. These microorganisms were identified as *Ralstonia sp.* (YABE411), *Pseudomonas sp.* (YATO411), and *Pseudomonas sp.* (YAET411). And the three strains could be grown in the mineral salts medium at pH 6.8 ± 1.0.

4.2. Aerobic Biodegradation of MTBE

The MTBE could be aerobically degraded by pure bacteria (Munoz-Castellanos et al., 2006) or by a bacterial consortium (Bianchi et al., 2009). The microorganisms that are capable of reduce the concentration of MTBE could use MTBE as a source, some kinds as the sole source, of carbon and energy (Ferreira et al., 2006; Munoz-Castellanos et al., 2006). Many studies focused on the relationship between MTBE-degrading activity and some environment factors in aerobic biodegradation, such as concentration of MTBE, temperature, dissolved oxygen concentration, soil type and initial microbial conditions (Bianchi et al., 2009; Rosell et al., 2010). Shah et al. (2009) compared the degradation of MTBE in uncontaminated and contaminated fractured chalk aquifer under aerobic condition. In the uncontaminated aquifer, with MTBE concentration between 0.1 and 1.0 mg L⁻¹ and dissolved O₂ concentration between 2 and 10 mg L⁻¹, could attenuate MTBE, and the biodegradation rate increased threefold because of the subsequent additions of MTBE. The results demonstrated the aquifer had an intrinsic potential to biodegrade MTBE aerobically. In the contaminated aquifer, the lag before MTBE degradation was only 15 or 33 days, and the biodegradation rates were higher (max. 27.5 µg L⁻¹ day⁻¹) than uncontaminated condition, the possible reason was the presence of acclimated microbial population in the contaminated environment. The initial concentration of MTBE had positive effect to the biodegradation, while the availability of dissolved O₂ did not affect MTBE biodegradation significantly. Rosell et al. (2010) also demonstrated that the biodegradation rate was not significantly or only slightly affected by dissolved O₂ concentration. In addition, Bianch et al. (2009) reported the MTBE degradation in an upflow fixed bed reactor by a bacterial consortium, and MTBE could be reduced by 99.8% when this system ran at 18 °C, MTBE concentration of 27.8 mg L⁻¹ and hydraulic retention time of 5 h.

Mycobacterium austroafricanum IFP 2012 is able to slowly grow on MTBE, and it could be used for aerobic MTBE degradation with isopropanol as a secondary carbon source (Maciel et al., 2008). The biofilter was operated for 85 days at an influent flow rate of 20 mL h⁻¹ by varying the MTBE concentration from 10 to 20 mg L⁻¹. The hydraulic retention time was fixed at 5 days. The removal of MTBE depended on the inlet MTBE concentration, and the MTBE removal efficiency higher than 99% was obtained for MTBE concentrations up to 15 mg L⁻¹.

Many studies reported that some microorganisms could degrade MTBE through co-metabolism (Chang et al., 2001; Deeb et al., 2000; Haase et al., 2006; Piveteau et al., 2000; Stephanie et al., 2003). Other hydrocarbons might be needed as the source of carbon and energy in the primary growth period. Piveteau et al. (2000) used two strains to attenuate MTBE in the laboratory experiments. *Gordonia terrae* IFP 2007 grown on ethanol or other substrates could degrade MTBE to *tert*-butyl alcohol (TBA) and formate, while *Burkholderia cepacia* IFP 2003 could translate the intermediate product to CO₂. When the MTBE/ethanol ratio was 0.5 (w/w) in laboratory scale fed-batch fermentation, MTBE could be totally degraded, without any accumulation of TEA. In another study, the butane-utilizing microorganisms could co-metabolically degrade MTBE and other gasoline oxygenates (Chang et al., 2001) In addition, three methane-, three propane- and four butane-utilizing cultures, which could degrade MTBE cometabolically, were enriched (Haase et al., 2006). In experiments with resting cells at 20 °C, enrichment cultures grown on methane could degrade MTBE at a maximum rate of 1.0 mg MTBE (g biomass h)⁻¹. The other two enrichment cultures were able to co-metabolically degrade

MTBE at temperatures of 20 °C and the intermediate was *tert*-butyl alcohol. The maximum rate was 7.2 mg MTBE (g biomass h)⁻¹ for propane-utilizing and 5.2 mg MTBE (g biomass h)⁻¹ for butane-utilizing culture, respectively. MTBE and TBA could also be degraded consecutively at a lower rate if propane-utilizing cultures grow on 1-propanol.

4.3. Anaerobic Biodegradation of MTBE

Anaerobic degradation of MTBE has been widely studied, and some anaerobic microorganisms capable of degrading MTBE have been reported, such as *Proteobacteria*, *Bacteroidetes*, *Firmicutes*, *Chloroflexi*, *Thermotogae*, *Pseudomonas sp.*, *Sphingomonas*, *Achromobacter*, and *Rhodococcus* (Raynal et al., 2010; Waul et al., 2009; Youngster et al., 2010). Raynal et al. (2010) studied the anaerobic cultures with Fe(III) or SO₄²⁻ as the electron acceptors for degradation of MTBE. The MTBE biodegradation rates ranged from 3.80 to 9.20, 1.13 to 1.71, 1.46 to 1.70 mg L⁻¹ d⁻¹ when Fe(III), SO₄²⁻, and Fe(III) & SO₄²⁻ were the electron acceptors, respectively.

4.4. Biodegradation of MTBE in Mixture

At most contaminated environments, MTBE is not the sole contaminant, and other compounds (such as BTEX and TBA) could exist. The biodegradation of other pollutants may have positive or negative effects on MTBE biodegradation (Stephanie et al., 2003). Raynal et al. (2008) monitored MTBE biodegradation in the presence and absence of BTEX in duplicate batch reactors inoculated with distinct enrichment cultures. The result showed that the presence of BTEX was able to inhibit the degradation of MTBE severely or even completely, which depended on the kind of the reactor. The rate of semi-batch reactor was almost three times as high as that of the batch reactor. In another study, Lee et al. (2011) used batch microcosm to monitor bacterial growth associated with toluene and/or MTBE degradation using *Pseudomonas putida*. The concentration of either toluene or MTBE as low as 25 mg L⁻¹ could enhance degradation of another compound, but the concentration higher than 25 mg L⁻¹ tended to result in negative or no effect. The negative effect was more pronounced for toluene since MTBE degradation was inhibited at a concentration of 50 mg L⁻¹ or higher toluene. The results indicated that the concentrations of co-contaminants become a limiting factor for the bioremediation of groundwater. Therefore, for higher degradation efficiency, care should be taken in the selection of concentration ranges of toluene and MTBE. Moreover, Wilson et al. (2001) reported MTBE degradation in a continuously stirred tank reactor. The rate of MTBE removal exceeded 99.99% when the VSS concentration in the reactor was over 600 mg L⁻¹. The biodegradation of MTBE and TBA in the reactor were higher for initial concentrations of 15 mg L⁻¹ than for concentrations of 5 mg L⁻¹. The presence of TBA at lower concentrations did not affect the rate of MTBE degradation, but higher concentrations of TEA did reduce the rate of biodegradation of MTBE. In addition, Mang et al. (2007) got a bacterium culture *Chryseobacterium sp.* that could degrade MTBE best at 25-30 °C, pH 7.0, initial concentration of MTBE from 50 to 100 mg L⁻¹. The additional TBA would lead to the decrease of the initial MTBE degradation rate, and the inhibitory effect of TBA increased with the increase of TBA concentration.

5. Conclusions and Prospects

Methyl *tert*-butyl ether has been used in many countries as an octane-enhancing replacement for lead. The widespread application of MTBE has resulted in its introduction into environment, and MTBE has been classified as a possible human carcinogen. Many techniques, including adsorption, chemical degradation and biodegradation, have been studied for the degradation and removal of MTBE in the environment. Activated carbon, clay mineral, resin and zeolite could be suitable adsorbents for removal of MTBE. Persulfate, Fenton oxidation and photocatalytic process could be used for chemical degradation of MTBE. Maybe, the biodegradation was the best way to eliminate MTBE from environment. In the future, new microorganisms capable of effectively degrading MTBE should be isolated and identified, and effects of environment factors on the biodegradation of MTBE should be investigate further. In addition, the integrated technique combining adsorption, chemical degradation and biodegradation should be studied for highly effective degradation and removal of MTBE.

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