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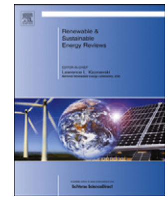

A review on the evolution of ethyl *tert*-butyl ether (ETBE) and its future prospects

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ABSTRACT

The environmental pollution crisis and the new environmental legislations have facilitated the need for cleaner-burning gasoline. Oxygenate additives, which can increase the octane rating and induce complete fuel combustion, play increasingly important roles in the development of a greener and more sustainable environment. In this short review, one of the better-known oxygenate additives, ethyl *tert*-butyl ether (ETBE), is discussed; this compound gained interest after the limitations and negative impacts caused by the addition of methyl *tert*-butyl ether (MTBE) to gasoline were discovered. The discussion focuses on the trends in ETBE production and on the evolution of conventional separation techniques toward the development of hybrid processes. Moreover, a new concept that involves the use of nanomaterials (carbon nanotubes (CNTs) and graphenes) in the production and separation of ETBE is proposed and discussed.

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Abbreviations: A-15, amberlyst-15; A-35, amberlyst-35; ACGHI, American Conference of Governmental Industrial Hygienists; ARCO, Atlantic Richfield Company; [bmin][OTf], 1-butyl-3-methylimidazolium trifluoromethanesulfonate; BRvp, blending Reid vapor pressure; CA, cellulose acetate; CAB, cellulose acetate butyrate; CAP, cellulose acetate propionate; CNTs, carbon nanotubes; CO, carbon monoxide; CSTR, continuous stirred tank reactor; CT-124, CT-145H, CT-151, CT-175, CT-275, ion-exchange resin; DWCNTs, double-walled carbon nanotubes; [emim][EtSO$_4$], 1-ethyl-3-methylimidazolium ethylsulfate; EPA, Environmental Protection Agency; ER, Eley–Rideal; ETBE, ethyl *tert*-butyl ether; EtOH, ethanol; FCSA, flurocarbonsulfonic acid; $H_6P_2W_{18}O_{62}$, Wells-Dawson acid heteropolyacid; $H_6P_2W_{18}O_{62} \cdot 27H_2O$, diphosphooctadecatungstic acid; HC, total hydrocarbon; H-ZSM-5, H-zeolite socony mobil-5; IARC, International Agency of Research on Cancer; IB, isobutene; KU-2–8, gel type ion-exchange resin; LHHW, Langmuir–Hinshelwood–Hougen–Watson; MR, molar ratio; MTBE, methyl *tert*-butyl ether; MWCNTs, multi-walled carbon nanotubes; NO_x, nitrogen oxides; NSTC, National Science and Technology Council; OH^-, Lewis base group; PFR, plug flow reactor; PLA, polylactic acid; PM, particulate matter; PPA, phenylphosphonic acid; PVP, polyvinylpyrrolidone; QH, Quasi-homogeneous; S-115, union carbide zeolite catalyst; $SiO_2 \cdot xH_2O$, silicic acid; $-SO_3H$, sulfonate group; STA, silicotungstic acid; SWCNTs, single-walled carbon nanotubes; *T*, temperature; TBA, *tert*-butyl alcohol; TiO_2, titanium oxide; TPA-K, Keggin-type tungstophosphoric acid; U.S., United States; VA, vinyl acetate; VOC, volatile organic compounds; VP, vinyl pyrrolidone; WHSV, weight hourly space velocity; ZSM-5, zeolite socony mobil-5

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1. Introduction

1.1. History and development of tertiary alkyl ether industry

In the 1970s, the issue of environmental air pollution and the health problems that are caused by automotive vehicle emissions reached a limit that induced great societal concern and the need for strong legislative efforts to reduce automobile emissions [1]. As a result, the exhaust compounds, such as carbon monoxide (CO), nitrogen oxides (NO_x), total hydrocarbon (HC) and particulate matter (PM), were legally regulated. In general, the exhaust compounds can be reduced to an acceptable level in one of three ways: (i) the invention of new engine technologies, (ii) the treatment of the fuel exhaust and (iii) the development and reformulation of existing fuels through the use of additives.

Before the 1970s, gasoline was reformulated through the addition of tetraethyl lead as an antiknock agent to increase the octane rating. However, the utilization of this antiknock agent was terminated when the Clean Air Act of 1970 was enacted. This legislation required drastic reductions in automobile emissions and led to the invention of a catalytic converter by the automobile manufacturers to meet the needs of the legislation. As a result, the vehicles with the catalytic converters required unleaded gasoline to prevent the lead in the tetraethyl lead from poisoning the catalyst system [2]. Methyl *tert*-butyl ether (MTBE) was thus introduced in low concentrations (1–3 vol%) into gasoline as a replacement for tetraethyl lead as an octane booster. In 1979, MTBE was widely used as an octane booster in gasoline component [3].

In 1990, Clean Air Act Amendments were enacted to emphasize the utilization of the oxygenated reformulated gasoline to achieve a better fuel combustion and improve air quality [4]. These amendments set minimum oxygen contents of 2.0 wt% [5] and 2.7 wt% [6] for reformulated gasoline in ozone-nonattainment areas and CO-nonattainment areas in the United States (U.S.), respectively. Moreover, the volatile organic compounds (VOC) emissions can be reduced by setting the blending Reid vapor pressure (bRvp) to a value not higher than 9.0 psi. Thus, these amendments catalyzed the development of MTBE industries, which changed the role of MTBE from an octane booster to a gasoline oxygenate [2].

In 1996, a U.S. Geological Survey study reported that MTBE was frequently found in the urban groundwater supplies sampled [2]. Leakages from the underground storage tanks and pipelines were the primary sources of MTBE contamination. MTBE is quite water soluble and difficult to biodegrade and does not readily adsorb to soil particles; thus, MTBE travels faster and farther than other gasoline constituents if a leakage happens. Moreover, the International Agency of Research on Cancer (IARC) and the U.S. Environmental Protection Agency (EPA) classified MTBE as a health risk threat in 2000 [7]. Although the utilization of MTBE as an oxygenate additive could reduce the amount of pollutants emitted into the atmosphere due to a more complete combustion, its negative effects on water quality have become an issue of debate. A global market report showed that the demand for MTBE increased gradually from 20.6 million tons in 1994 [8] to 21.0 million tons and 22.0 million tons in 1999 and 2002, respectively [2,9]. However, because of the negative impacts that MTBE has on the environment, ETBE has become a popular alternative oxygenate additive for gasoline. ETBE has superior properties compared with MTBE. ETBE was first used in France in 1992. In 2002, France and Spain contributed a total of 568,000 t to the ETBE production capacity in the European Union [10]. The ETBE production capacity increased from 2 million tons to 4 million tons from 2005 to 2007 [9,11]. ETBE is mainly used as a fuel additive in several European countries, including France, Netherlands, Germany, Spain and Belgium [11–13]. Japan tends to produce ETBE using bio-ethanol as one of the reactants [14]. Japanese oil industries started to blend 7% bio-ETBE into automobile fuel in 2010 in accordance with the Kyoto Protocol Achievement Plan [15].

1.2. Fuel additive and oxygenate additive

A fuel additive is a chemical substance that assists the cleaning of engine parts, such as the carburetor, the intake valve and the fuel injector. Fuel additives also play an important role in the reduction of greenhouse gas emissions through complete combustion, the prevention of temper fuel gelling and nozzle choking and the protection of engine parts from corrosion [16]. Fuel additives can be classified into hybrid compound blends, oxygenates (alcohol and ether), antioxidants, antiknock agents, lead scavengers and fuel dyes.

Oxygenate additives contain oxygen as part of their chemical structure and are usually employed as gasoline additives. These additives increment the octane rating and combustion quality and reduce particulate emission and carbon monoxide production [16]. Alcohol and ether are common oxygenate compounds. Alcohols, such as methanol and ethanol, are widely used in automobile fuels. Whereas tertiary alkyl ethers, such as MTBE and ETBE, are commonly used as octane improvers for liquid fuels, their low vapor pressure characteristic reduces the vapor pressure of gasoline [17].

1.3. Advantages and disadvantages of MTBE and ETBE

After the disadvantages of MTBE were revealed, ETBE was found to be an alternative and environmentally friendly octane oxygenate. When the Clean Air Act of 1970 was enacted, MTBE was the most reliable oxygenate additive due to its characteristic behavior. MTBE is non-toxic, has a high octane value and is compatible with the hydrocarbons found in gasoline [2]. However, its impact on health was first revealed after high levels of MTBE were discovered in water wells and groundwater, mainly in North America [1]. MTBE is highly soluble in water (42 mg/L) [9,18] and difficult to biodegrade [9].

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