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## Progress and perspectives in converting biogas to transportation fuels

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## ABSTRACT

The discovery of abundant natural gas resources has greatly increased the study of using methane as a feedstock to produce transportation fuels. Biogas (primarily containing methane and CO<sub>2</sub>), which is generated from waste biomass via anaerobic digestion or landfills, is regarded as a renewable source of methane, and has the potential to achieve sustainable production of transportation fuels. Since biogas also contains a significant amount of impurities (e.g., H<sub>2</sub>S, NH<sub>3</sub>, and siloxane), a cleaning procedure is generally required prior to conversion to transportation fuels. Physical approaches, mainly compression and liquefaction, have been commercially applied to upgrade biogas to bio-compressed natural gas (CNG) and liquefied biogas (LBG). For chemical approaches, catalytic reforming is the dominant method for converting methane to syngas, followed by Fischer–Tropsch synthesis (FTS) or fermentation of syngas to a variety of alcohols (e.g., methanol, ethanol, and butanol) and liquid hydrocarbon fuels (e.g., gasoline, diesel, and jet fuels). High purity hydrogen, a clean fuel, can also be produced via reforming. Methanol can be produced by direct oxidation of methane, while interest in the biological conversion of methane to methanol has grown recently due to its mild operating conditions, high conversion efficiency, and potential for using raw biogas. The derived methanol can be further converted to gasoline via a methanol to gasoline (MTG) process. This paper provides a comprehensive review of major research progress on technologies for converting biogas/methane into transportation fuels, and discusses the principles, kinetics, operating conditions, and performance of each technology. Efficient direct conversion of biogas into ethanol and higher alcohol fuels (e.g. butanol), which is envisaged to be the focus of research pursuits in the near future, is also discussed, with emphasis on the development of methane-utilizing microbes through genetic engineering.

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**Abbreviations:** AD, anaerobic digestion; AOB, ammonia oxidizing bacteria; ATR, autothermal reforming; CBG, compressed biogas; CFC, chlorofluorocarbons; CNG, compressed natural gas; CHP, combined heat and power; COE, crude oil equivalents; ctl, coal-to-liquid; DEA, diethanolamine; DGE, diesel gallon equivalent; FTS, Fischer Tropsch synthesis; GGE, gasoline gallon equivalent; GHG, greenhouse gas; GTL, gas-to-liquid; HAS, higher alcohols synthesis; IEA, International Energy Agency; LBG, liquefied bio-gas; LPG, liquid petroleum gas; LNG, liquid natural gas; MDEA, methyldiethanolamine; MDH, methanol dehydrogenase; MEA, monoethanolamine; MMO, methane monooxygenase; MTG, methane-to-gasoline; NADH, nicotinamide adenine dinucleotide hydrogen; OFMSW, organic fraction of municipal solid waste; POM, partial oxidation of methane; POR, partial oxidative reforming; PSA, pressure swing adsorption; PWS, pressurized water scrubbing; TSA, temperature swing adsorption; WGS, water gas shift

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## 1. Introduction

Concerns about depletion of fossil fuels, energy security, and emission of greenhouse gas (GHG) have prompted renewable energy studies. Biogas, generated from anaerobic digesters or landfills via biological degradation of organic compounds, is considered a renewable energy carrier. Production of biogas via anaerobic digestion (AD) involves a series of biochemical processes, primarily comprised of four steps: hydrolysis, acidogenesis, acetogenesis, and methanogenesis. Organic materials, such as food processing wastes, lignocellulosic biomass, and animal manure, which contain carbohydrates, fats and proteins, are commonly used as feedstocks. These feedstocks are usually available at a low cost or may even generate a tipping fee, thus making biogas production economically favorable. According to an AgSTAR report, a 1 MW AD facility can produce approximately 3 million kWh of electricity per year, which is enough to supply power to more than 200 homes [1]. Another study evaluated energy crops for biogas production in the EU-25 (the 25 Member States of the European Union). It showed that 320 million tonnes of crude oil equivalents (COE) could be produced with crop rotations that integrate the production of food, feed, raw materials (e.g. oils, fats, organic acids), which would provide up to 96% of the total energy demand of cars and trucks in the EU-25 [2]. In China, biogas production from small-scale biogas digesters has increased from approximately  $1.8 \times 10^9 \text{ m}^3$  in 1996 to  $1.0 \times 10^{10} \text{ m}^3$  in 2007 (equivalent to  $1.1 \times 10^{11} \text{ kWh}$  electricity), while biogas production from medium- and large-scale biogas projects has increased from approximately  $1.2 \times 10^{11} \text{ m}^3$  in 1996 to  $6.0 \times 10^{12} \text{ m}^3$  in 2007 (equivalent to  $6.3 \times 10^{13} \text{ kWh}$  electricity) [3]. Given the tremendous amount of organic wastes from agriculture and food processing, there is growing interest worldwide in employing AD as a waste treatment method as well as an energy production technology.

A variety of feedstocks have been used for AD. At mesophilic conditions (35–40 °C), one tonne dry feedstock can produce 13–635  $\text{m}^3$  of  $\text{CH}_4$  gas, depending mainly on the composition of the feedstock [4–8]. Municipal wastes usually have higher  $\text{CH}_4$  yields than lignocellulosic biomass such as yard trimmings. For instance, one tonne of dry organic fraction of municipal solid waste (OFMSW) showed a high  $\text{CH}_4$  yield of 635  $\text{m}^3$  [5], which is

equivalent to  $2.39 \times 10^{10} \text{ J}$  energy or 751 l gasoline. Assuming fuel consumption is 10.6 km/L (25 mile/gallon) for a passenger car, energy produced from one dry tonne of OFMSW would allow a passenger car to travel 7963 km. To date, efforts have been made to improve the  $\text{CH}_4$  yield during anaerobic digestion. Feedstock selection, process design and operation, digestion enhancement, and co-digestion with multiple substrates have been extensively studied, and several reviews are available [9–13].

Currently, biogas is primarily used for: (1) burning biogas in a combined heat and power (CHP) unit for heat and electricity generation; (2) upgrading biogas for natural gas pipeline injection; and (3) converting purified biogas to compressed biogas (CBG) or liquid biogas (LBG) for a variety of fuel applications (Fig. 1). CBG is considered to be the same as compressed natural gas (CNG), and is often referred to as Bio-CNG. Heat and electricity production systems are usually adopted for on-site usage of biogas. Purified biogas is almost the same as natural gas in terms of heating value; therefore, injection of clean biogas into a natural gas pipeline becomes an option. Biogas was used as a transportation fuel in Germany in the 1930s and the interest was renewed in the 1990s. Nowadays, biogas is provided as a transportation fuel in gas filling stations in Europe, especially Germany and Sweden, either as 100% methane (CBG100) or blended with natural gas (e.g. CBG10 and CBG50) [14]. Using biogas as a transportation fuel has the advantage of generating low GHG emissions compared to conventional fossil fuels. Vehicles using CNG/Bio-CNG generate approximately 8–22 g  $\text{CO}_{2\text{eq}}$  per MJ, which is over 80% lower than those using petroleum based fuels [15]. Lower particulate matter (e.g. metals and soot) emissions are another advantage. Use of LBG is also growing. In 2005, a commercial LBG filling station, which used landfill gas, was opened in Los Angeles, California. Commercial LBG filling stations are also available in China and a few European countries.

Besides Bio-CNG and LBG, biogas may be converted to other transportation fuels (Fig. 1). Clean biogas can go through a catalytic reforming process, from which syngas or high purity  $\text{H}_2$  can be obtained.  $\text{H}_2$  is a clean transportation fuel, and syngas can be used as a substrate for alcohol production. Previously, studies mainly focused on the reforming of  $\text{CH}_4$ , while recent studies reported that both  $\text{CH}_4$  and  $\text{CO}_2$  can be converted to syngas via dry reforming or steam reforming, thus enabling direct use of biogas

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