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Research review paper

Biological conversion of methane to liquid fuels: Status and opportunities

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ABSTRACT

Methane is the main component of natural gas and biogas. As an abundant energy source, methane is crucial not only to meet current energy needs but also to achieve a sustainable energy future. Conversion of methane to liquid fuels provides energy-dense products and therefore reduces costs for storage, transportation, and distribution. Compared to thermochemical processes, biological conversion has advantages such as high conversion efficiency and using environmentally friendly processes. This paper is a comprehensive review of studies on three promising groups of microorganisms (methanotrophs, ammonia-oxidizing bacteria, and acetogens) that hold potential in converting methane to liquid fuels; their habitats, biochemical conversion mechanisms, performance in liquid fuels production, and genetic modification to enhance the conversion are also discussed. To date, methane-to-methanol conversion efficiencies (moles of methanol produced per mole methane consumed) of up to 80% have been reported. A number of issues that impede scale-up of this technology, such as mass transfer limitations of methane, inhibitory effects of H_2S in biogas, usage of expensive chemicals as electron donors, and lack of native strains capable of converting methane to liquid fuels other than methanol, are discussed. Future perspectives and strategies in addressing these challenges are also discussed.

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Introduction

Methane (CH_4) is an abundant energy source which has attracted attention due to rapidly growing energy demand. Proven global natural gas (containing 99–100% CH_4) reserves have increased from 113 trillion m^3 in 1990 to 194 trillion m^3 in 2012, and more is likely to be discovered in the future. The energy potential of these reserves is roughly 2.0×10^{15} kWh, more than ten times the predicted world energy consumption in 2020 (U.S.EIA, 2013). Biogas (containing 60–70% CH_4) produced from anaerobic digestion (AD) of organic matter is another source of CH_4 (Yu and Schanbacher, 2010). Unlike natural gas, which was formed millions of years ago (Mann et al., 2003), biogas can be produced from abundant and renewable organic feedstocks in a short period of time and is considered a carbon-neutral energy source. Therefore, CH_4 is not only a crucial part of the current energy portfolio, but it will also play an important role in achieving long-term energy sustainability.

Methane is in gaseous form at ambient temperatures with a boiling point of −164 °C, making it costly to store, transport, and distribute. Although CH_4 can be compressed into compressed natural gas (CNG) or liquefied natural gas (LNG), these processes require high energy inputs and large capital expenditures. Both CNG and LNG are hazardous due to their high pressure (21–25 MPa) and low temperature (−160 °C). In order to address these issues, it is desirable to convert CH_4 into easily handled liquid fuels such as methanol, ethanol, and butanol. At present, the most commonly used CH_4-to-liquid fuel technology is the two-step thermochemical conversion process with syngas as the intermediate (Chuang, 2012; Park and Lee, 2013). Alternatively, one-step thermochemical conversion of CH_4 to liquid fuels has also been studied (Ho et al., 1996; Park and Lee, 2013). Owing to its high C-H bond strength, high ionization potential, and low acidity, the CH_4 molecule is difficult to activate. As a result, thermochemical conversion usually encounters a number of barriers, such as high pressure and/or temperature, expensive chemical catalysts, and low efficiencies (Park and Lee, 2013). Also, since biogas contains contaminants such as H_2S and NH_3 that can deactivate chemical catalysts, costly purification processes are required prior to thermochemical conversion (Kohn, 2012; Navarro et al., 2013).

Compared to thermochemical processes, biological conversion is highly attractive due to efficient conversion reactions under mild operating conditions. Furthermore, microorganisms have an extraordinary biological diversity which enables them to adapt to various environments. In nature, two groups of bacteria have been found that can activate the stable C-H bond of CH_4 under ambient conditions. One is aerobic methanotrophic bacteria, which use methane monooxygenase (MMO) to activate and then utilize CH_4 as their source of carbon and energy (Hanson and Hanson, 1996). The other group is ammonia-oxidizing bacteria (AOB), which can partially oxidize CH_4 to methanol when using ammonia as an energy source (Taher and Chandran, 2013). Besides, some methane oxidizing archaea and bacteria can also carry out anaerobic oxidation of methane (AOM) (Beal et al., 2009; Boetius et al., 2000; Ettwig et al., 2010; Haroon et al., 2013). Although no native strains have been reported to be able to directly convert CH_4 into other liquid fuels, some acetogens, as defined by Drake et al. (2006), have the potential to convert methanol into acetate, acetone, isopropanol, butanol, ethanol, butyrate, and 2,3-butanediol (Abrini et al., 1994; George et al., 1983; Köpke et al., 2011; Tracy et al., 2012). Conversion of CH_4 to liquid fuels other than methanol may be fulfilled by an integrated bioprocess (CH_4 to methanol and then methanol to other liquid fuels) using different microorganisms, or by a genetically engineered organism (using either methanotrophs, AOB, or acetogens as host cells) that contains the two pathways (Fig. 1).

This paper reviews the habitats, biochemical pathways, liquid fuel yields, and genetic engineering of these microorganisms, which are crucial for CH_4-to-liquid fuel conversion. Some of the technical issues in industrial applications and possible strategies to address them are also discussed.

Methanotrophs: the most extensively studied methane-oxidizing microorganisms

Methanotrophs are a group of microorganisms that can use CH_4 as their sole source of carbon and energy (Hanson and Hanson, 1996). This unique natural ability makes methanotrophs a valuable candidate for bioconversion of CH_4 to liquid fuels.

Classification and habitats of methanotrophs

The first methanotrophic bacterium was identified in 1906 by Söhngen (Hanson and Hanson, 1996). Extensive isolation and characterization of more than 100 CH_4-utilizing bacteria was performed by Whittenbury et al. (1970), who established the basis for the classification of methanotrophs. Methanotrophs were initially classified into three types (I, II, and X) based on their characteristics, including, but not limited to, location of introcytoplasmic membranes, carbon assimilation pathways, major phospholipid fatty acids, and growth conditions (Table 1) (Hanson and Hanson, 1996; Semrau et al., 2010).

Methanotrophs have been isolated from a multitude of environments where CH_4 is emitted. These environments vary dramatically in physical and chemical conditions, such as temperature, pH, and salinity (Table 2). All the isolated methanotrophs reported hereto are aerobic methanotrophic bacteria. Based on their 16S rRNA gene sequences, the currently known aerobic methanotrophic bacteria are classified into two classes of *Proteobacteria* (*α-Proteobacteria* and *γ-Proteobacteria*) as well as one phylum, *Verrucomicrobia*. The methanotrophs belonging to *α-Proteobacteria* are classified to four genera: *Methylosinus*, *Methylocystis*, *Methylocapsa*, and *Methylocella*. The methanotrophs within *γ-Proteobacteria* are more diverse, representing 12 genera, including *Crenothrix*, *Clonothrix*, *Methylobacter*, *Methylocaldum*, *Methylococcus*, *Methylomicrobium*, *Methylohalobius*, *Methylomonas*, *Methylosarcina*, *Methylosphaera*, *Methylosoma*, and *Methylothermus*. The verrucomicrobial methanotrophs are only found in the genus *Methylacidiphilum* (Semrau et al., 2010).

Anaerobic methanotrophs (ANME) have also been identified in marine and freshwater sediments where AOM occurs, but no ANME have been isolated either in pure culture or in a consortium (Haynes and Gonzalez, 2014). Consortia of bacteria and ANME archaea can carry out AOM coupled to reduction of iron (Beal et al., 2009), manganese (Beal et al., 2009), and sulphate (Boetius et al., 2000). Besides, Ettwig et al. (2010) reported a ANME bacterium, *Candidatus* '*Methylomirabilis oxyfera*', that can couple AOM to nitrite reduction. This bacterium is affiliated with the candidate phylum NC10 which does not have any cultured representatives so far (Deutzmann and Schink, 2011). More recently, Haroon et al. (2013) has demonstrated a novel ANME lineage,

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