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Conversion of petroleum to methane by the indigenous methanogenic consortia for oil recovery in heavy oil reservoir

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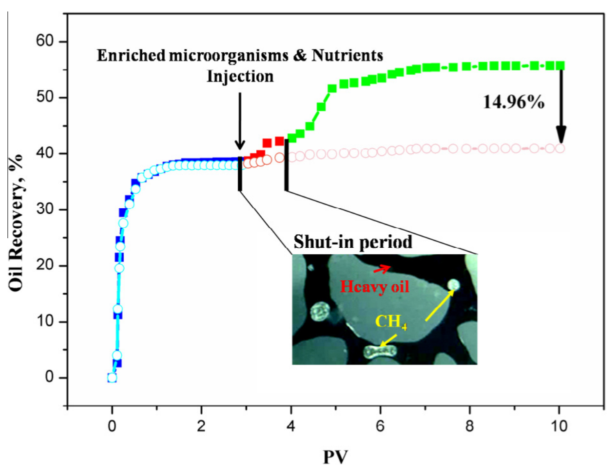
HIGHLIGHTS

- Heavy oil was degraded by the enriched methanogenic consortia.
- Methane production during the biodegradation of heavy oil.
- Viscosity of the degraded heavy oil was reduced with the dissolution of methane.
- In-situ bioconversion of heavy oil to methane has great potential on EOR.

GRAPHICAL ABSTRACT



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ABSTRACT

Microbial enhanced oil recovery has been well acknowledged and becoming an advanced technology for oil recovery. Compared with current techniques, a newly technical strategy of the in-situ heavy oil gasification to methane for oil viscosity reduction was proposed and successfully proved via enriching the methanogenic consortia from the brine of oil reservoir with heavy oil as carbon source. During 200 days anaerobic culturing, 2.34 g of heavy oil was degraded coupling with 1514 μmol of methane production. Phylogenetic diversity analysis showed that the enriched consortia composed with sequences affiliated with the *Firmicutes*, *Proteobacteria*, *Deferribacteres* and *Bacteroidetes*. The recovered archaeal phylotypes were close to the *Methanobacteriales* and *Methanosarcinales*, which could convert the produced small molecules (formic acid and acetic acids) to methane. The viscosity of the degraded heavy oil was reduced by 72.45% at 20 MPa after the dissolution of the produced methane. Core flooding tests finally showed that the oil degradation and methane production by the enriched methanogenic consortia made 14.7% of the tertiary enhanced oil recovery. These results demonstrated a promising and practical strategy of microbial technology on oil recovery by activating the methanogens in heavy oil reservoir.

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1. Introduction

Worldwide energy demand is continuously increasing with the current pace of development, and oil continues to play crucial role in total energy consumption. Exploration of petroleum resources traditionally follows a pattern where the better quality and more accessible resources, like light oils, are extracted firstly before programming to general lower quality, less accessible resources that require more efforts and have higher energetic, economic, and environmental costs [1]. Reserves of heavy oil, ultra-heavy oil and natural asphalt all over the world are about 100×10^9 t [2]. With the depletion of conventional oil reserves, the production of these unconventional fossil resources achieved a boost especially with the significant increases in production from tar sands and heavy oil reservoir around the world. Unparalleled attentions are shifting toward the development of heavy oil. However, to improve oil recovery on this type of unconventional resource is commonly facing great technical challenging due to high viscosity and density, poor fluidity, and easy absorption. Conventionally, thermal recovery techniques including steam stimulation and steam drive methods [3–5], and cold production techniques including alkaline drive, polymer drive and miscible flooding [6–8], have been well developed and widely applied for the heavy oil recovery. In these traditional techniques, reducing viscosity has become the key strategy in heavy oil exploitation, transportation and refining. At present, the main ways to reduce the viscosity of heavy oil are thermal recovery (by means of heating cables, electric heating oil-pumping rod, and heat-conducting oil), dilution method by using light oil, and phase-behavior by emulsification. However, these techniques generally have extreme high cost, and negative effects on the surrounding environment [4,5]. Therefore, extensive interests in microbial enhanced oil recovery (MEOR) for heavy oil recovery have been increasing as cost-effective and environmentally friendly candidate [9].

Microbial enhanced oil recovery (MEOR) technology has been well recognized and successfully applied in the development of the conventional oil, which is a way to utilize microorganisms or their metabolites to improve the recovery of crude oil from reservoirs [10,11]. The main mechanisms of MEOR involve [12]: (1) biosurfactants production could reduce the surface/interfacial tension and alter the wettability of solid surface; (2) degrade the heavy component of crude oil to improve the oil quality; and (3) gas production could dissolve in crude oil and then reduce its viscosity, or increase reservoir pressure. Most of microorganisms that have performed successfully in numerous researches or field pilots for oil recovery, no matter in conventional or unconventional reservoir, are aerobic or anoxic bacteria. Few efforts have been relatively involved into the application of anaerobic microorganism on improving oil recovery, significantly due to that aerobic bacteria have showed greater performance on biosurfactant production and crude oil degradation than anoxics or anaerobics [11,13]. However, these bacteria always cannot fulfill the expecting functions after injected or stimulated in subterranean of oil reservoirs. This is also the key reason that the mechanism of MEOR has not been qualitatively and quantitatively characterized, resulting in MEOR technology has not been widely spread throughout the whole petroleum industry. Recently increasing studies demonstrated that anaerobic microbes dominate subsurface environments, despite slow reaction kinetics and uncertainty as to the actual metabolic activities (such as degradation and emulsification of heavy oil, methane generation) occurring in oil reservoirs [14–18]. However, most of laboratory researches or oilfield trials are limited to the utilization of anaerobic biosurfactant-producing bacteria and its metabolites to form the oil-in-water emulsion, which has limited effects on viscosity reduction at subterranean [19].

Petroleum reservoirs are characterized as extreme environments by the wide range of temperature and salinity, high pressure, and anoxic/anaerobic conditions, and coupled with multiphase fluids of oil, gas and water. Interests in microbiosphere at deep subterranean petroleum-rich strata for MEOR have been driven by the potential presence and multi functions of living microorganisms within. Recent decades, numerous types of microorganisms from various oil reservoirs and their functions has been recognized, including nitrate reducers, sulfate reducers, fermentative bacteria, iron reducers, acetogens and methanogens under anaerobic condition [18,20–23]. Among them, the conversion of hydrocarbons to methane in oil reservoirs is a typical ultimate anaerobic biodegradation process [23,24], and the generated methane is able to dissolve in oil under subterranean pressure to thereby reduce the oil viscosity dramatically [23,25]. Therefore, the converting the residual oil to methane by anaerobic microorganisms under methanogenic condition could be suggested as a newly way to improve the exploitation and development of heavy oil through heavy oil degradation and in-situ methane dissolution. Under this strategy, heavy oil reservoirs could be regarded as "bioreactors" in which diverse physiological types of microorganisms acting in syntrophic association can be stimulated with specific nutrients and significantly degrade heavy oil to methane [26,27]. Although methane production from crude oil by microbial consortia in oil reservoir has been well documented [28,29], none of researches focus their efforts on the in-situ methane dissolution into the degraded heavy oil and its influence on the viscosity reduction as well as on the final heavy oil recovery. Therefore, this study will investigate the anaerobic microorganisms and their activities on heavy oil degradation under methanogenic condition; quantify the characterization of the in-situ methane production and dissolution in the degraded oil; calculate the comprehensive effects on viscosity reduction; and finally evaluate the potential of heavy oil recovery. Compared with the previous attempts to apply MEOR and other approaches on improving heavy oil recovery, this study will not only extend the theoretical understanding of the mechanism of MEOR, and but also create a practical, high-efficiency and low-cost technique to improve heavy oil recovery. In terms of the current oil price, it is obviously more and more sensible and economical to apply this green strategy to the heavy oil recovery.

2. Materials and methods

2.1. Sample collection

Oil and brine samples were collected from Xing block of Daqing oilfield, and were immediately sealed in sterile 150 ml bottles and transported to the laboratory at 4 °C for further research. The reservoir located at the Chinese northwest (latitude 46.79, longitude 125.04) with depths of 850 m to 900 m subterranean, and characterized with temperature of 50 °C, permeability of 375×10^{-3} μm^2, dehydrated dead oil viscosity of 1823.86 mPa s at 50 °C, and total salinity of 3852.26 mg/L.

2.2. Anaerobic enrichments

Pre-enrichment culture of the methanogenic consortia was carried out by the incubation of 1 g oil and 150 ml enrichment-medium with 50 ml of brines at 50 °C until the detection of methane. The pre-enrichment medium contains (g/L): NH_4Cl 2.0, K_2HPO_4 1.5, KH_2PO_4 1.5, $MgCl_2$ 0.2, CH_3COONa 2.0, HCOONa 2.0, citrate 1.0, yeast extract 0.05, cysteine 0.05, resazurin 0.002, pH 6.0–7.0, supplemented with 1 ml of the trace element solution and 2 ml vitamins solutions previously developed [30]. After