Journal of CO_2 Utilization 19 (2017) 221–229

Contents lists available at ScienceDirect

Journal of CO_2 Utilization

journal homepage: www.elsevier.com/locate/jcou

Reforming of methane to syngas in a microwave plasma torch at atmospheric pressure

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ARTICLE INFO

Article history:
Received 30 November 2016
Received in revised form 27 January 2017
Accepted 19 March 2017
Available online 13 April 2017

Keywords:
Methane reforming
Microwave plasma
Synthesis gas
Syngas

ABSTRACT [1]

Carbon dioxide was converted to synthesis gas (syngas) in a microwave plasma torch by methane reforming at atmospheric pressure. The microwave plasma torch converts $CO_2 + CH_4$ into synthesis gas. This study examined conversion rates as a function of gas temperature for dry methane reforming. The temperature of the torch flame is measured 6760 K by making use of optical spectroscopy. The CO_2/CH_4 reforming can be completely converted into synthesis gas (conversions: 68.4%, CO_2; 96.8%, CH_4) through their reforming reactions at a microwave power of 6 kW. When the reforming gas (CO_2:CH_4) mole ratio was 1:1, the resulting synthesis gas (H_2:CO) mole ratio was 0.9:1.1. The H_2 and CO mass yield rates increased to 0.24 kg/h and 1.86 kg/h, respectively. Also, the energy yield are 240 g/h and 41.4 g/kWh. The CO_2 microwave plasma torch not only exhibited noteworthy results for CO_2 reduction and syngas production, but the H_2:CO mole ratio of the gas produced is easily controlled by adjusting the CO_2:CH_4 ratio during the feeding process.

1. Introduction

Atmospheric concentrations of carbon dioxide (CO_2) have increased dramatically since the industrial revolution, an issue acknowledged to be a significant contributing factor to the greenhouse effect. Given that reducing greenhouse gas concentrations is very important to the environment, it is important to find solutions that achieve zero-emissions of CO_2, an important greenhouse gas. Recently, CO_2 capture and storage (CCS) [1,2], CO_2 capture and utilization (CCU) [3,4], and the CO_2 reforming of methane (CH_4) [5], have all been used as technologies to reduce CO_2 levels. However, the CCS technology is not a fundamental solution for reducing CO_2 emitted by industrial processes, despite the fact that CCS is the primary technology used for CO_2 capture by industry. Methods that utilize CO_2 in energy sources and chemical processes are better suited to resolving these environmental problems. An alternative and more effective method involves the material conversion of CO_2 into another useful substance. CO_2 and CH_4 reforming processes in CCU technologies are becoming more attractive because they not only reduce atmospheric emissions of CO_2 while consuming CH_4, they also obtain a mixture of hydrogen (H_2) and carbon monoxide (CO). In industry, CH_4 or natural gas reforming processes are widely used to obtain H_2 or synthesis gas (syngas), which are versatile intermediates for many chemical processes, such as methanol synthesis, ammonia synthesis, carboxylation, hydrogenation [6] and the production of liquid hydrocarbons through the Fisher-Tropsch (F-T) process [7]. Syngas and H_2 are also regarded as major sources of clean energy for power generation. Since CH_4 is the major component of natural gas, it is therefore also a possible alternative energy source, especially in view of predicted oil shortages. In other words, the CO_2 and CH_4 reforming process can convert these two greenhouse gases into syngas and, in doing so, can contribute to slowing atmospheric global warming. In recent years, many processes have been used for CO_2 and CH_4 reforming. These conversion methods have used catalysts [8,9], corona plasma [10], dielectric barrier discharge [11], gliding arc gas discharge [12], microwave plasma, [13,14] glow discharge plasma [15] and thermal plasma [16]. There are intractable problems with the catalyst in various reforming processes, including carbon deposition and deactivation of the catalyst, but these catalytic methods are inexpensive and efficient. On the other hand, CO_2 plasma can easily be converted into syngas without expending the additional energy required by a thermo-catalytic shift reaction, but these plasma methods are highly

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[1] CO_2 capture and storage (CCS), CO_2 capture and utilization (CCU), CO_2 dry reforming (CDR).

http://dx.doi.org/10.1016/j.jcou.2017.03.016
2212-9820/

energy consuming. To utilize CO_2 as a feedstock and solve the intractable problems associated with catalytic or plasma processes, we considered CO_2 dry reforming (CDR) using a microwave plasma torch at atmospheric pressure. The CDR reaction strong depend on the applied temperature and chemical species. In this article, we investigated the dissociation properties of CO_2 molecules at high-temperature, as well the syngas produced, in CDR reactions using a CO_2 microwave plasma torch. This torch can be operated at atmospheric pressure at a frequency of 2.45 GHz with a microwave power of 6 kW and a total gas flow rate of 30 L/min. Spectroscopic diagnostics indicate that high temperatures, together with the active species from CO_2 produced in the microwave plasma, induce the CDR reaction. The production of synthesis gas through dry reforming confirms that a CO_2 microwave plasma torch can be used to effectively achieve methane conversion at atmospheric pressure.

2. Experimental set-up for the CO_2 microwave plasma torch

The proposed CO_2 microwave plasma torch for syngas production via CH_4 conversion was operated at a frequency of 2.45 GHz at atmospheric pressure. The experimental parameters included variable power (3 ~ 6 kW) and a total flow rate of 30 L/min (CO_2 15 L/min and CH_4 15 L/min, CO_2:CH_4 = 1:1). Experimental results included spectroscopic diagnostics as well as the reforming efficiency of the CO_2 microwave plasma. The design and operation of microwave plasma torch have been reported in detail in previous work [17–19].

Fig. 1 depicts a schematic diagram for syngas production using the CO_2 microwave plasma torch. The principal components of the experimental configuration comprise a microwave generator, a microwave plasma torch, gas supply, and an optical emission spectroscopy and synthesis gas analyzer. Efficient microwave power transfer from the magnetron to the reactive gas can be achieved via the microwave generator, which basically consists of a magnetron head, an isolator, a directional coupler, and a three-stub tuner. Microwave power from the magnetron head is transmitted via a WR340 standard rectangular waveguide to the microwave plasma torch. The isolator protects against damage to the magnetron by reflected microwaves. The power induced by microwave radiation in the quartz tube can be adjusting by the three-stub tuner. In addition, reflected power can be minimized by adjusting the three-stub tuner to less than 1% of the forward power. A directional coupler is used to measure the injected and reflected microwave power in the microwave plasma torch.

As shown in Fig. 2, the CO_2 microwave plasma torch is used to produce syngas via CH_4 conversion at atmospheric pressure; this plasma torch uses CO_2 (99.99%) to maintain working, stable plasma. The torch mainly consists of a tapered waveguide to provide a maximized electric field, and a quartz supporter for injecting the CO_2 ignition gas. The quartz tube (outer diameter: 30 mm, thickness: 2 mm, and length: 90 mm) pierces perpendicularly through the wide wall of the tapered waveguide. The center axis of the quartz tube is located at one-quarter wavelength from the short end of the tapered waveguide wall. The induced electric field is maximized at the center of the quartz tube axis before the plasma is turned on. Microwaves from the magnetron propagate through the waveguide, concentrating their power in the tube and generating plasma. CO_2, as the ignition gas, was injected via the quartz supporter into the plasma torch through four small tangential holes, resulting in a flow that swirls inside the quartz tube. In addition, CH_4 gas (99.99%), as the reforming gas, was also introduced into the plasma via the quartz supporter. The main purpose of gas swirling is the stabilization of the CO_2 microwave

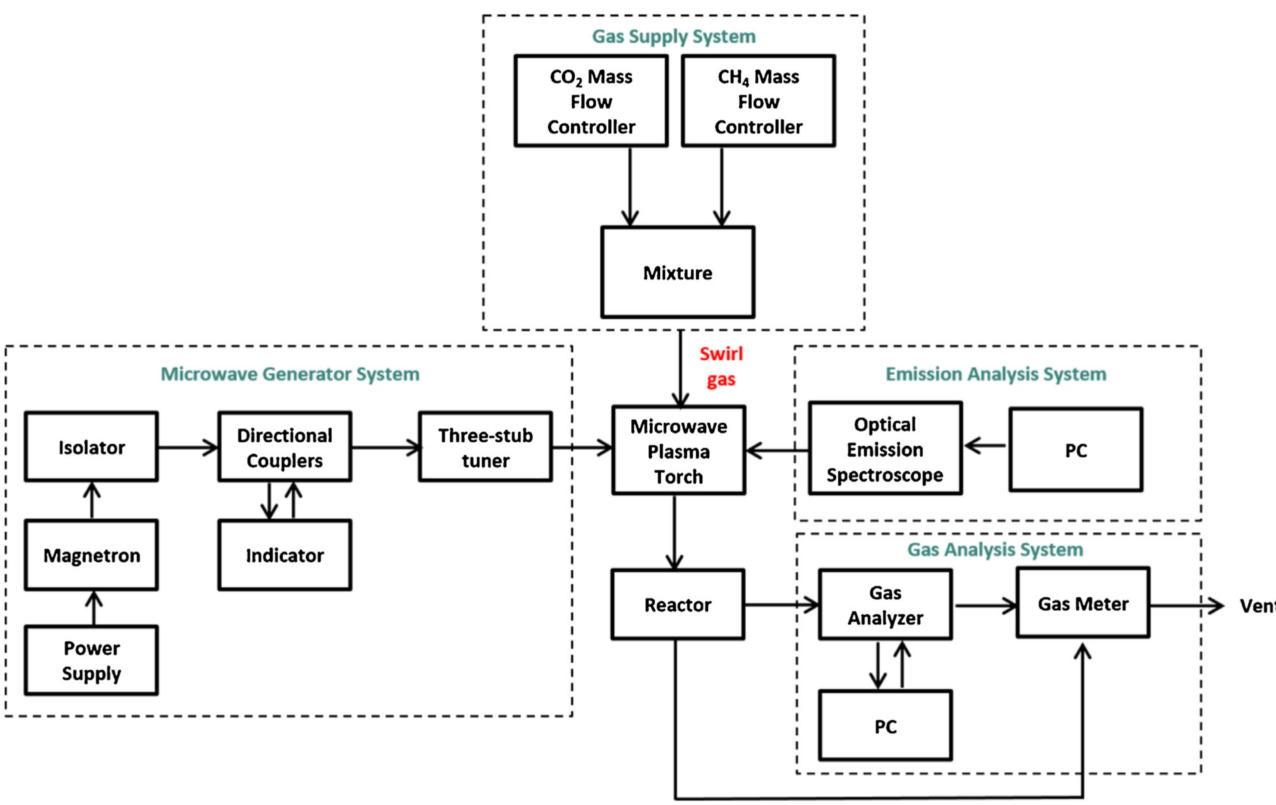

Fig. 1. Schematic diagram of the experimental set up for syngas production. The microwave plasma torch consists of a 2.45 GHz magnetron, a power supply and WR-340 waveguide components that consist of a circulator, a coupler, a 3-stub tuner and a tapered waveguide. Also included is a schematic of the experimental setup for optical emission spectroscopy and the gas analysis system.