Chemical Engineering Journal 155 (2009) 874–880

Contents lists available at ScienceDirect

Chemical Engineering Journal

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

Plasma-catalytic reforming of methane in AC microsized gliding arc discharge: Effects of input power, reactor thickness, and catalyst existence

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ARTICLE INFO

Article history:
Received 30 June 2009
Received in revised form
24 September 2009
Accepted 2 October 2009

Keywords:
Non-thermal plasma
Gliding arc discharge
Microreactor
Methane conversion

ABSTRACT

A new design of a microsized gliding arc discharge reactor was preliminarily investigated for the reforming of methane to various useful products. In methane reforming by the gliding arc (GA) microreactor under ambient conditions, hydrogen and acetylene were dominantly produced, with high selectivities of ~75% and ~70–90%, respectively, which were much higher than those in conventional GA reactors because of the high power density and the extremely short residence time of the GA microreactor, as compared to conventional GA reactors. Small amounts of other products (including ethylene, ethane, butadiene, and coke) were also formed. The results showed that the methane conversion strongly depended on input power and reactor thickness. A Ni-loaded porous alumina-silica catalyst, prepared by wet impregnation, was used to investigate the catalytic effect on the microreactor performance for methane reforming. A considerable enhancement of methane conversion, but not hydrogen and other product selectivities, was achieved in the combined plasma-catalytic system. Furthermore, heating the surface of the Ni-loaded catalyst brought about a significant change in product selectivity, but not methane conversion.

1. Introduction

Natural gas is the cleanest and environmentally safest primary fossil fuel, in terms of its lowest emission of CO_2. Interestingly, numerous investigations have focused on upgrading the conversion process of methane (CH_4), which is a main constituent in natural gas in all reserves around the world, to produce higher value-added chemicals by reforming reactions. Non-thermal plasma has reportedly been used for several chemical conversion processes, also involving either non-oxidative methane reforming [1–7], oxidative methane reforming [8–15], or hybrid plasma-catalytic methane reforming [16–29]. Due to its non-equilibrium property, non-thermal plasma, with high-energy electrons, creates principally a large number of chemically active species through electronic and ionic collision processes, and then immediately induces subsequent chemical reactions under ambient temperature and atmospheric pressure. Hence, highly stable methane molecules can potentially be converted by means of non-thermal plasma. For further development of the non-thermal plasma processing, some researchers have integrated it with the novel idea of microreactor technology [30–33]. In a microreactor, the extremely small volume and large specific surface area provide several significant process advantages, such as better heat distribution and heat transfer, precise control of a shorter residence time, a higher chance for reactant molecules to collide with electrons, and small processing amounts of reactants [30–35].

This present work focuses on studying the performance of a gliding arc discharge microreactor, based on a concept of integrating a non-thermal plasma with a microreactor technology, by using the methane reforming as a model reaction for the preliminary test. The effects of input power, reactor thickness, and nickel metal loaded on a porous alumina-silica catalyst plate on the performance of non-oxidative methane reforming were investigated. Nickel was used as the studied reforming catalyst due to its good reforming capability, as reported in several articles [18,24,36]. Moreover, the effect of heating the catalyst surface on the plasma reaction performance was also investigated.

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doi:10.1016/j.cej.2009.10.009

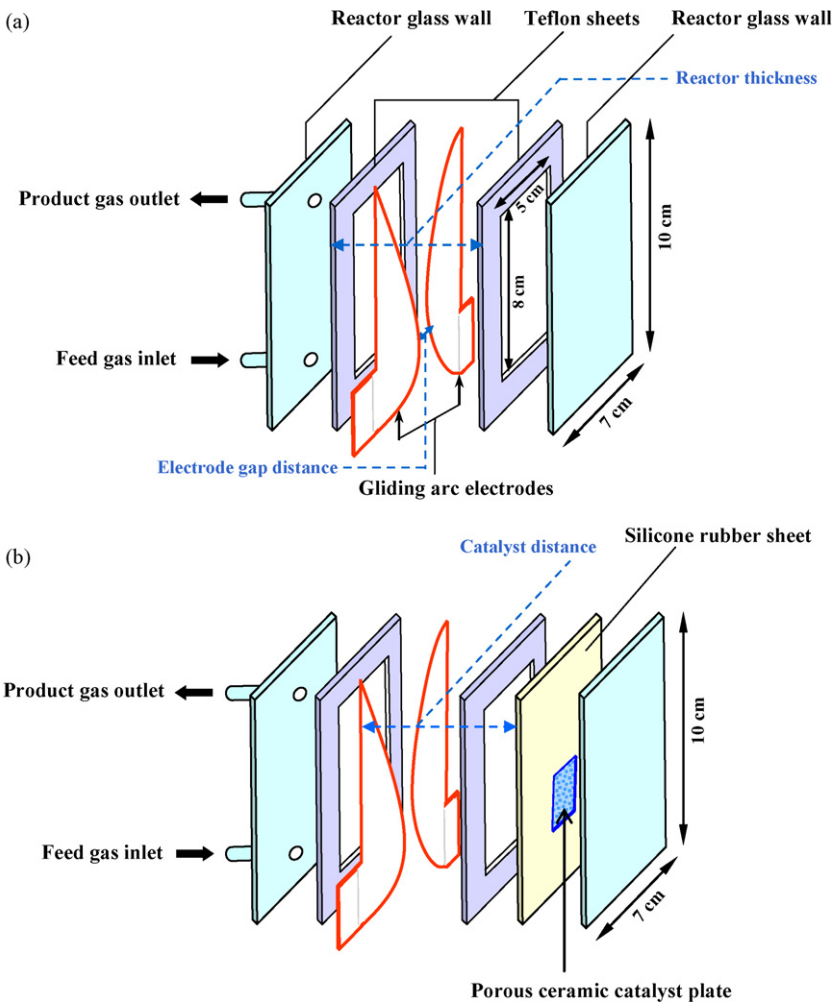

Fig. 1. Configuration of a gliding arc discharge microreactor (a) without a catalyst and (b) with a catalyst.

2. Experimental

2.1. Gliding arc microreactor system

The configuration of the gliding arc (GA) microreactor used in this study is illustrated in Fig. 1. The GA microreactor was made of two glass plates with a width of 7 cm, a length of 10 cm, and a thickness of 0.5 cm, and had two diverging knife-shaped electrodes fabricated from two stainless steel sheets with a thickness of 0.25 mm. These electrodes were vertically positioned inside the reactor and connected to the AC power supply. Two pre-drilled holes on the glass cover (reactor wall) served as the gas inlet and outlet. Two Teflon sheets with different thicknesses were placed between the reactor glass walls and the electrodes on both sides (see Fig. 1(a)) to create various reactor thicknesses (reactor thickness = electrode thickness + 2 × Teflon sheet thickness) and to seal the microreactor. To examine the effect of catalyst in this studied plasma microreactor system, a 2 cm × 3 cm catalyst plate was inserted into a holder made of a silicone rubber sheet, and then was packed between the reactor glass wall and the Teflon sheet on the back side of the microreactor (see Fig. 1(b)). In Fig. 1(b), the catalyst distance is defined as the distance between a silicone rubber sheet and a pair of electrodes—in other words, a right-hand-side Teflon sheet thickness. Additionally, to study the effect of catalyst surface temperature, the catalyst plate was heated to 210 °C by being externally irradiated with an infrared (IR) beam equipped with a halogen lamp (LCB50) and an IR beam heater (LCB-PS12, Inflidge Industrial Ltd.). The temperature on the surface of the catalyst plate was measured by using a thermal label indicator.

2.2. Catalyst preparation procedure

A porous ceramic plate with a chemical composition of 23% SiO_2, 75% Al_2O_3, and 2% others (Nikkato Corporation) was used as a catalyst support. Its Brunauer–Emmett–Teller (BET) specific surface area and specific pore volume were 2.6 m^2/g and 5.9 mm^3/g, respectively. $Ni(NO_3)_2{\cdot}6H_2O$ (Wako Pure Chemical Industries, Ltd.) was used as a nickel precursor. To prepare the nickel loaded on the catalyst support by using the wet impregnation method, the porous silica-alumina plate was immersed in an aqueous solution of 2.0 M $Ni(NO_3)_2{\cdot}6H_2O$ for 10 h. After that, the catalyst was dried at 110 °C for 2 h, and then calcined in air at 500 °C for 5 h. The calcined catalyst was further reduced in a H_2 atmosphere at 500 °C for 3 h prior to use. Both unloaded and Ni-loaded catalyst plates were used comparatively for testing the plasma reaction performance. An energy dispersive X-ray analyzer (EDX, Link ISIS, Series 300) was used to analyze the Ni content in the prepared catalyst, and it was found to be approximately 5 wt.%.

2.3. Reaction testing experiments

In this work, a mixed feed gas having 5% CH_4 with Ar balance was introduced upward to the GA microreactor system. The flow rates of CH_4 and Ar were regulated by rotameters. After the com-