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Application of non-thermal plasma technology on fugitive methane destruction: Configuration and optimization of double dielectric barrier discharge reactor

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ABSTRACT

Fugitive methane (CH_4) from waste treatment facilities (landfill mining), power industries (oil and gas process plants) and coal mining etc. into atmosphere is an increasing environmental concern. In this study, CH_4 conversion efficiency in double dielectric barrier discharge (DDBD) has been investigated at various operating parameters including input power, feed gas-mixture flow rate, CH_4 initial concentrations, and discharge gap distance between two dielectrics. Increase in input power, decrease in the gas-mixture flow rate and discharge gap distance; results in increases of CH_4 conversion efficiency. In plasma alone, maximum CH_4 conversion efficiency of 76% was obtained using 3 mm plasma discharge gap distance at flow rates of 2 L/min, input power of 65.8 W and is limited by experimental conditions. In addition, CH_4 conversion efficiency in plasma alone and plasma-catalytic is compared by introducing various catalysts includes Pt–Sn/Al_2O_3, BaTiO3 and HZSM-5 in single plasma discharge zone. Results revealed that plasma combined with Pt–Sn/Al_2O_3 showed higher CH_4 conversion efficiency (84.93%) as compare to plasma alone (56.42%) using 6 mm plasma discharge gap distance at flow rates of 2 L/min, input power of 65.8 W. Moreover, maximum energy efficiency of CH_4 conversion (limited by experimental conditions) was 27.24×10^{-12} mol/kJ at 32.6 W observed in plasma-catalyst. Analysis of the exhaust gas showed that DDBD is a promising alternative reactor not only to achieve high CH_4 conversion efficiency, but also to overcome the drawbacks of formation of undesirable byproducts. Moreover, deposition of carbon residues on the surface of internal electrode is not observed, which is often occurred in single DBD reactors.

1. Introduction

Methane (CH_4) emission from power industries (oil and gas process plants), coal mining and waste treatment facilities etc. into atmosphere is an increasing environmental concern (Noor et al., 2013). Its global warming potential is 28 times greater than that of carbon dioxide and is greatly contributing to climatic variability (Montzka et al., 2011; Saikawa et al., 2014; Tian et al., 2012, 2015). Landfills have been found to be a significant source of CH_4 generation and emissions; Some landfills (especially large modern landfills) have landfill gas (LFG) collection systems to use CH_4 for electricity generation (Mønster et al., 2015). Although some large and modern landfills have installed gas collection systems, the notably low landfill gas (LFG) collection efficiency of 30% still lead to a large amount of fugitive CH_4 emitted into the atmosphere (Xin et al., 2016). CH_4 emission from landfill is also related to the stage of landfill. In developed countries, the characteristics of landfills have low organics or in the after-care stage usually has low emission. However, in developing countries, landfill waste is characterized as high organic and at early stage of active CH_4 generation, the CH_4 emission is high. Lando et al. conducted a field measurement of CH_4 emission from landfill in Indonesia. They found that ranges of

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ambient CH_4 concentration 12–4259 ppm (Lando et al., 2017).

In recent times, it has great challenge to decompose CH_4 by conventional thermal catalytic process due to non-occurrence of non-oxidative coupling of CH_4 at room temperature (Chen et al., 2010). High activated energy and high temperature >700 °C is required to break the chemical bonds of CH_4 molecule. So-far, technologies includes thermal decomposition, catalytic decomposition and oxidation are developed to decompose CH_4 (Abbas and Daud, 2010). However, all these techniques may form undesired byproducts during decomposition process (Mahammadunnisa et al., 2013). In the recent decades, Non-thermal Plasma (NTP) has been intensively studied for the treatment of air pollution. NTP has to be proven the promising technique for the treatment of water pollution (Krishna et al., 2016; Magureanu et al., 2011), soil pollution (Aggelopoulos et al., 2016; Lu et al., 2015; Zhang et al., 2017) and air pollution (Ma et al., 2017; Zhang et al., 2016). With the intervention of novel technique, non-thermal plasma (NTP) conversion of CH_4 under low temperature and pressure has got great attention. It has enormous flexibility to adjust the process parameters so that the energy and reactant consumptions can be minimized (Nozaki and Okazaki, 2013; Tsolas et al., 2015).

In NTP, the electrons have high energy with typical electron temperature of upto 10 eV to decompose the inert molecules into ions, free radicals excited atoms and molecules for further reactions (Tsolas et al., 2015). The investigations of CH_4 decomposition in NTP have been made globally by different cold plasma types includes corona discharges, spark discharges, gliding arc or dielectric barrier discharges (DBD). A number of researchers have attempted to insert supporting gasses in CH_4 conversion process, such as argon, air, oxygen (Hoffmann et al., 2013; Holladay et al., 2009; Indarto et al., 2006; Liang et al., 2011; Matsumoto et al., 2012) and noble gas (Jo et al., 2013; Kossyi et al., 2013). Few others used different plasma discharges that could perhaps generate different product distributions (Indarto et al., 2006; Tu and Whitehead, 2012; Xin et al., 2011). Recently, dielectric barrier discharges (DBD) has gain more importance as generators of NTP in a cost-effective way. DBD plasma reactors can operate at higher power values and over an extensive span of discharge gaps. The dielectric properties of feed gases and dielectric materials can transform energy to the system by applying the time derivative of input voltage (Abdel-Salam et al., 2003; Kundu et al., 2012; Valdivia-Barrientos et al., 2006). This has led to numeral of energetic applications including control of environmental pollution, chemical treatment of polymer surfaces, ozone generation for industrial fabrication and plasma enhanced chemical vapor deposition for thin film growth etc (Nikola Bednar and Goran, 2013). However, single DBD reactor cannot completely mineralize the contaminants in environment but also produces noxious pollutants. Meanwhile, in the decomposition of CH_4, solid carbon can easily deposit on the reactor's inner surface and on the surface of inner electrodes, which further reduces the conversion efficiency of DBD reactors. Nevertheless, double-tube DBD can be used as an active reactor to protect the inner electrode from carbon deposition and generation of by-products during the decomposition process by applying high energy to reactor (Zhang et al., 2014).

In this study, double dielectric barrier discharge (DDBD) reactor with one input power source has been developed for the first time to examine fugitive CH_4 destruction and also to overcome the drawbacks of single DBD in terms of generation of higher hydrocarbons. Parameters including effect of input power, feed gas-mixture flow rate, CH_4 initial concentrations as well as plasma discharge gap distance on CH_4 conversion efficiency in inert atmosphere have been studied in a DDBD reactor. Moreover, combination of NTP with bi-metallic catalyst has also been investigated to scrutinize the effect on CH_4 conversion efficiency.

2. Materials and methods

2.1. Plasma rector

The DDBD reactor used in this study has a tubular structure as shown in Fig. 1. Two cylindrical quartz tubes were used as dielectrics; each one is positioned in contact with one electrode. The inner tube (outer diameter 8 mm, thickness 1 mm, length 360 mm) is adjacent to the ground electrode; the outer surface of outer tube (outer diameter 25 mm, thickness 2.5 mm, length 360 mm) is contacted with the discharge electrode. The discharge electrode made of a steel mesh (length 205 mm) is connected to power supply system. The copper metal rod was used as ground electrode inside internal dielectric quartz tube. DDBD reactor configuration serves to chemically isolate inner and outer electrodes from plasma discharge zone and ensures that all gas-mixture pass through the single plasma reaction zone and are homogeneously treated. Feed gas-mixture enters the space bounded by two dielectrics via inlet, pass through the plasma reaction zone and then exits via outlet.

2.2. Input gas and controlled parameters

Alternating current plasma generator (Nanjing Suman Electronics Corp. CTP-2000K) was used to generate plasma discharge. The fixed frequency of 9 kHz was maintained and recorded by a digital oscilloscope (Tektronix DPO 4034). Eleven different levels of input power including 8.1, 11, 16, 23.1, 32.6, 39.3, 44.3, 49.5, 53.7, 57.6 and 65.8 W; eight different gas-mixture (CH_4+ N_2) flow rates involving 0.3, 0.6, 1, 1.3, 1.6, 2, 2.3 and 2.6 L/min were chosen to analyze the effect of input power and feed gas-mixture flow rate on CH_4 conversion efficiency. Numerous studies have been carried out with the diverse flow rates and input powers for CH_4 conversion. Khalifeh et al. conducted a study with flow rate ranging from 0.025 L/mint to 0.2 L/mint (Khalifeh et al., 2016). Xu and Tu, carried out an investigation on the CH_4 decomposition in DBD reactor with a total flow rate varied from 0.05 L/mint to 0.3 L/mint (Chao Xu, 2013). Similarly, Liu et al. conducted a study on the CH_4 decomposition with varied discharge power from 15 to 75 W and different flow rates ranges between 0.05 L/mint to 0.3 L/mint (Liu et al., 2014). Therefore, flow rate from 0.3 L/min to 2.6 L/min and input power from 8.1 W to 65.8 W in current study was set based on the literature review. In addition, for the safety measures of used DDBD reactor, further increase of input power is not applied. Furthermore, the initial concentration of CH_4 diluted in pure nitrogen (N_2) was set at 1000 ppm (% ratio CH_4:N_2 = 0.1:99.9) and plasma discharge gap distance of DDBD reactor fixed at 6 mm. Furthermore, CH_4 conversion efficiency was compared with other two different CH_4 initial concentrations including 5000 ppm (% ratio

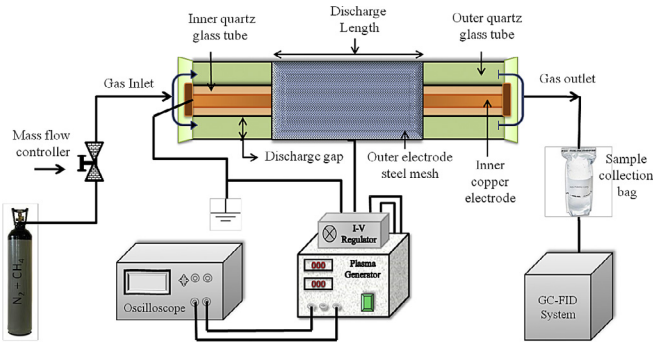

Fig. 1. Schematic illustration of DDBD reactor setup for methane destruction.