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A gliding discharge reactor supplied by a ferro-resonance system for liquid toluene decomposition

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ABSTRACT

The paper presents results of toluene decomposition performed in a gliding discharge reactor with a conical chamber and six high-voltage electrodes. The electrodes of this plasma reactor are supplied by a 6-phase ferro-resonance system composed of resonant capacitors and stray inductances of the high voltage transformers supplied from a digital inverter. The overall toluene conversion mainly depended on the initial C_7H_8 concentration and gas flow rate. It was possible to achieve a complete toluene conversion. Moreover, soot or higher hydrocarbons were not formed in this large-scale gliding discharge reactor.

1. Introduction

A gliding discharge (GD) is characterized by a combination of equilibrium plasma at the beginning of, and non-equilibrium plasma in the further part of the discharge process. Therefore, this type of atmospheric pressure discharges can produce relatively high levels of electron energy, density, current and power with comparatively low temperature of the gas. These non-equilibrium properties of the gliding discharge are the reason why GD has been the focus of scientific interest and is facilitated in practical applications in waste treatment, as well as environmental control (Fridman et al., 1999). Non-equilibrium plasma is used for decomposition of substances particularly hazardous for the environment (Ulejczyk et al., 2013; Schmidt-Szałowski et al., 2011) because the reagents are easily activated by the high energy electrons, so even reactions with high activation energy may proceed (Krawczyk et al., 2013a).

The practical application of the gliding discharge was first demonstrated by Chapelle and Czernichowski (Krawczyk et al., 2013a) and is currently being tested in many research centres (Chapelle and Czernichowski, 1992; Brisset et al., 2008; Kalra et al., 2005; Fridman et al., 2005; Sreethawong et al., 2007). The advantages of this type of discharge are: simple construction of the reactor and ability to supply high energy to the plasma zone. A typical gliding discharge system consists of two electrodes positioned opposite to each other, which form an acute angle. In the vicinity of the gas inlet, the gap between the electrodes is very narrow. Therefore, it is possible to obtain subsequent electric breakdowns. Multiple discharge channels move towards the ends of the electrodes, grow while travelling upwards and break when a critical value of supplied

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energy is attained. This is determined by the power supply capability.

A discharge occurs when a rapid stream of gas is passed along the edges of two or more diverging electrodes. When a channel extends due to the movement of the gas stream, the gas temperature decreases and the plasma changes from equilibrium plasma to non-equilibrium plasma. Up to 80% of the electrical energy may be directly introduced into endothermic chemical reactions (Kalra et al., 2005). In that phase, the mean temperature of the gas usually does not exceed 800 K, often being much lower. In such a system, a series of periodic phenomena takes place with a high frequency, including the discharge ignition, displacement of the discharge channel, and its disappearance. Because of the high energy of electrons in the discharge channels (1–10 eV) gas molecules may be activated and active species can be generated. It should be noted that the density of energy in the GD channels is high and the duration of a single discharge much longer than those in e.g. dielectric barrier discharges. The gliding discharge can be effectively used for chemical processes in which non-equilibrium plasma initiates the chemical reactions at atmospheric pressure (Fridman et al., 1999, 2005; Czernichowski, 2001, 1994; Czernichowski et al., 1996; Mustaf-Yardimci et al., 2000; Lesueur et al., 1988).

Currently four ways for generating gliding discharge may be distinguished:

- discharge occurs between two, three or six knife-shaped vertical electrodes (Chapelle and Czernichowski, 1992)
- discharge occurs between the electrodes and a rotating disc (Czernichowski et al., 2006)
- discharge occurs in the reactor resembling a cyclone (Czernichowski, 2009)
- discharge takes a tornado-shaped form (Kalra et al., 2005; Yu et al., 2011).

In recent years different kinds of gliding discharge reactors, with either 2 or 3 electrodes, were used for decomposition of VOCs (Vandenbroucke et al., 2011; Indarto et al., 2007), nitrous oxide (Krawczyk and Młotek, 2001) and conversion of methane to higher hydrocarbons (Sreethawong et al., 2007; Młotek et al., 2009). These studies were conducted on a laboratory scale and concern the conversion of gaseous reagents in gliding discharge plasma or conversion of substances dissolved in water by active species produced in the plasma zone located above the surface of the solution (Chapelle and Czernichowski, 1992; Sreethawong et al., 2007; Vandenbroucke et al., 2011; Indarto et al., 2007; Krawczyk and Młotek, 2001; Młotek et al., 2009; Bo et al., 2007; Du et al., 2007).

In the commonly-used gliding discharge reactors, the fast gas stream is injected between the electrodes. As a result, the residence time of the reagents in the plasma zone might not be enough to achieve the expected conversion or product yields. In order to expand the discharge chamber, high voltage and turbulent gas streams are usually applied. However, this solution does not solve the problem when scaling-up of the reactors is required. The presented article shows a new, unique reactor and power system, which can be used for decomposition of liquid or toxic gases with high flow rates. In this paper, a large-scale gliding plasma reactor was used to decompose liquid toluene.

The first step of the reaction in an air–toluene mixture consists in radical formation and attachment of an electron, resulting in the formation of C_6H_5, CH_3, C_3H_4, and of hydrogen radicals (reactions (1)–(4)) (Bo et al., 2007; Du et al., 2007; Guo et al., 2007; Kohno et al., 1998):

$$C_6H_5{\cdot}CH_3 + e \rightarrow C_6H_5 + CH_3 + e \qquad (1)$$

$$C_6H_5{\cdot}CH_3 + e \rightarrow C_6H_4{\cdot}CH_3 + H + e \qquad (2)$$

$$C_6H_5{\cdot}CH_3 + e \rightarrow C_5H_6 + C_2H_2 + e \qquad (3)$$

$$C_6H_5{\cdot}CH_3 + e \rightarrow C_3H_4 + C_4H_4 + e \qquad (4)$$

The active species present in the air plasma (O(^{1}D), O(^{3}P), and N) can also initiate the toluene decomposition process. The O atom can also react with the C atom to break $C_6H_5—CH_3$ bond (reactions (5) and (6))

$$C_6H_5CH_3 + O^+; O_2{}^+; N; N_2{}^+ \rightarrow C_6H_5CH_3{}^+ + O; O_3; N; N_2 \qquad (5)$$

$$C_6H_5CH_3{}^+ + e \rightarrow C_6H_5 + CH_3 \qquad (6)$$

The C_6H_5 radicals may react with other radicals to form soot precursors (reactions (7)–(9)).

$$C_6H_5 + 6CH_3 + H \rightarrow C_{12}H_{26} \qquad (7)$$

$$C_6H_5 + H \rightarrow C_6H_6 \qquad (8)$$

$$C_6H_5 + H \rightarrow 3C_2H_2 + e \qquad (9)$$

Benzene and C_2 hydrocarbons are the intermediate step of toluene oxidation to CO and CO_2. Under plasma conditions, the intermediate products can also be decomposed to soot (reactions (10) and (11)) (Kado et al., 2003) and after that oxidized to CO and CO_2.

$$C_6H_6 \rightarrow 6C + 3H_2 \qquad (10)$$

$$C_2H_n \rightarrow 2C + n/2H_2 \qquad (11)$$

2. Experimental

In this study a new construction of the reactor (Fig. 1) (Schmidt-Szałowski et al., 2013) and power supply were used (Krawczyk et al., 2013b). The reactor has two chambers: a conical chamber, where the six high voltage electrodes were placed, and a cyclone chamber. The gas inlet to the cyclone chamber was located tangentially to the inner wall. In this chamber the gas was introduced into a vortex flow by three nozzles. The gas velocity was in range 29–44 m/s in every nozzle. Then the gas passed into the conical chamber through the Venturi orifice. The ratio of diameters of the cyclone chamber and the Venturi orifice was 12 to 1, which resulted in a very high vortex velocity of the gas. The liquid reagent was introduced into the reactor by a nozzle placed in the upper part of the cyclone chamber. The nozzle sprayed the liquid directly into the Venturi orifice. This arrangement of gas and liquid insertion systems led to very effective mixing of reagents. Moreover, thanks to this configuration, the residence time of the reagents in the discharge zone became appreciably longer (Fig. 1).

GD power supply systems are based on high voltage transformers. Many different constructions of GD power sources have been developed. One of the more advanced is using five-limb transformers for a three-phase power