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Destruction of toluene by rotating gliding arc discharge

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HIGHLIGHTS

- A rotating gliding arc (RGA) discharge reactor is developed for tar destruction.
- The highest destruction efficiency could exceed 95% using toluene as tar surrogate.
- The two major gaseous products are hydrogen and acetylene.
- The liquid and solid byproducts are collected and determined.

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ABSTRACT

Non-thermal plasma is considered as an alternative treatment of tar present in the effluent from gasification processes. In this study, a novel rotating gliding arc (RGA) discharge reactor was developed for tar destruction. Toluene in nitrogen flow was used as a tar surrogate. The physical features of RGA discharge and its application to toluene destruction are investigated at different input concentrations and total gas flow rates. As a result, the highest destruction efficiency could exceed 95%, with a toluene concentration of 10 g/N m^3 and a total flow rate of 0.24 N m^3/h. The two major gaseous products are H_2 and C_2H_2, with maximum selectivity of 39.35% and 27.0%, respectively. A higher input concentration slightly reduces this destruction efficiency but the energy efficiency further expanded, with a highest value of 16.61 g of toluene eliminated/kW h. In addition, the liquid and solid byproducts are collected downstream of the RGA reactor and determined qualitatively and semi-quantitatively. The amount and structure of these by-products is instructive for reaching a better comprehension of the chemical consequences of plasma treatment to the model compound and to the carrier gas nitrogen.

1. Introduction

The use of biomass and municipal solid waste (MSW) as alternative fuel has received considerable attention, due to the steady depletion of fossil fuels. Gasification is regarded as one of the most promising techniques to convert biomass or MSW to fuel gas or even to syngas, which – after thorough cleaning – can not only be used as fuel in gas turbines and gas engines, but also offers some basic building blocks for producing valuable chemicals, liquid fuels and hydrogen [1–3]. During gasification, besides bringing useful gaseous products, also many undesirable byproducts, such as particulates, alkali metal salts, acid gases and tar are formed [4,5]. Among these byproducts, tar is the most troublesome contaminant, as upon cooling and condensing it can cause fouling, clogging and corrosion problems in downstream equipment [6,7].

Generally, the removal or destruction of tar is implemented after a gasifier and its successive steps can be categorized into (i) physical particle and aerosol removal (cyclone, filter, electrostatic precipitator) and wet scrubbing methods and (ii) thermochemical methods, i.e. thermal or catalytic cracking [2]. Physical methods are unattractive, as they transfer the tars separated into a liquid, paste-like, or solid phase, which has the dual drawback of causing secondary pollution and losing the chemical energy contained in tars [6]. The only proven method has been the scrubbing with oil and the energetic use of the spent oil, as in a project of the Energy research Centre of the Netherlands (ECN) [8]. For fast thermal tar cracking, a high reaction temperature, usually well over 1000 °C is needed, involving large energy expenditure [9]. Catalytic cracking seems suitable to convert tar into valuable cracking products and also operates at lower temperatures than thermal tar cracking

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[10]. However, various catalysts have high affinity for sulfur, chlorine, and may become poisoned, or else fouled by coke. Coking of catalysts is difficult to avoid unless the gasifier feed is well tested and dependable in its characteristics and composition. Various sulfur, chlorine and nitrogen compounds are major contaminants of the producer gas resulting from the gasification process [11]. Thus, complete tar removing is still a big challenge, due to the complex composition of tar, the various cleaning duties to be combined, and a general lack of proven, highly efficient technology.

Considering the properties and possible cracking mechanisms of tar, a non-thermal plasma is regarded as an alternative solution with the major merits of fast ignition, compact design and high efficiency. Basic study on low temperature pulsed plasma [12,13] and microwave plasma [14,15] for tar destruction has been conducted. Compared to conventional thermal and catalytic cracking, plasma discharge shows higher removal efficiency (>95%). However, a high specific energy input, a low electron density and the heterogeneous distribution of radicals are the key factors that – until now – have limited the industrial scale application of the plasma method in its numerous variants.

An atmospheric pressure gliding arc discharge (GAD) is considered as an attractive and promising route to address the aforementioned problems. Such a gliding arc has been widely studied for environmental clean-up (e.g. removal of volatile organic compounds) [16,17] and for fuel production applications (e.g. reforming of hydrocarbons) [18–21]. The main part of the gliding arc power consumed (up to 75–80%) is dissipated in the non-equilibrium zone and the dissipated power reaches 40 kW per electrode pair [22]. This equipment offers high energy efficiency and selectivity for activating chemical reactions, and appears to have a great potential for cracking tars from gasification [23].

In this study, a novel rotating gliding arc (RGA) discharge reactor was specifically developed for the destruction of tars. Compared with the typical knife-shaped GAD [22], the RGA discharge provides a stable and wide three-dimensional plasma region. Tests are conducted, using toluene in nitrogen flow as a model tar. Preliminary experiments are carried out to investigate the influence of the toluene concentration levels and the gas flow rate on the toluene destruction efficiency and the RGA energy efficiency. In addition, the gaseous, liquid and solid products are collected downstream the RGA reactor and determined qualitatively and semi-quantitatively. The identity of the dense product fraction yields a key towards the nature and relative importance of the reactions responsible for removing the tar surrogate, toluene. Toluene was selected as a suitable tar surrogate, since it shows a simple structure and high thermal stability, even though – compared to typical biomass tar components, e.g. levoglucosan – its boiling point is much lower, its thermal stability much higher and its chemical structure relatively simple. Such simple structure assists considerably in identifying and quantifying the reaction products.

2. Experimental

2.1. Experimental setup

The experimental setup for tar destruction is shown schematically in Fig. 1. It comprises a RGA reactor, a power source, gas delivery system and analysis system. Fig. 2 shows the configuration of the RGA plasma reactor. The RGA reactor proper has outer dimensions of 36 mm diameter and 180 mm height. It is composed of a conical inner electrode (anode) and a grounded cylindrical outer electrode (cathode), both made of stainless steel (18/8 Cr–Ni). A ring magnet is situated coaxially with the electrodes to provide a magnetic field (approximately 2000 G) oriented along the reactor axis. The narrowest gap between the two electrodes, meant for arc ignition, is 2 mm. The feed gas is injected into the reactor through three tangential inlets, to form a swirling flow. The arc is initially ignited at the narrowest gap and then moves down to the lower point of the conical electrode. Finally, it is anchored near the tip of the inner electrode and there rotates periodically. The arc discharge zone is assumed to be cylindrical in shape, with a height of 20 mm. It is marked in Fig. 2, to estimate the residence time.

The flow rate of carrier gas (N_2) is controlled by a mass flow controller (MFC, Sevenstar D07-series). Toluene is injected into the feeding line by a high resolution syringe pump (Harvard, 11 plus) and mixed with the carrier gas in a stainless steel pipe, preheated to 200 °C. The mixture of nitrogen and toluene vapor is then sprayed into the RGA reactor. A high voltage DC power source (Teslaman TLP2040) is connected to the RGA reactor to generate plasma, and a 40 kΩ resistance is used as current limiter. The electrical parameters are measured by means of an oscilloscope (Tektronix DPO4034B) and the dynamic behavior of the arc is recorded by a high-speed camera (HG-100K).

The gas flow leaving the tar destruction unit passes through three successive absorption bottles with the first two containing hexane and the third empty, to collect entrained droplets and all are placed in an iced water bath. The gaseous products are determined qualitatively and quantitatively by gas chromatography

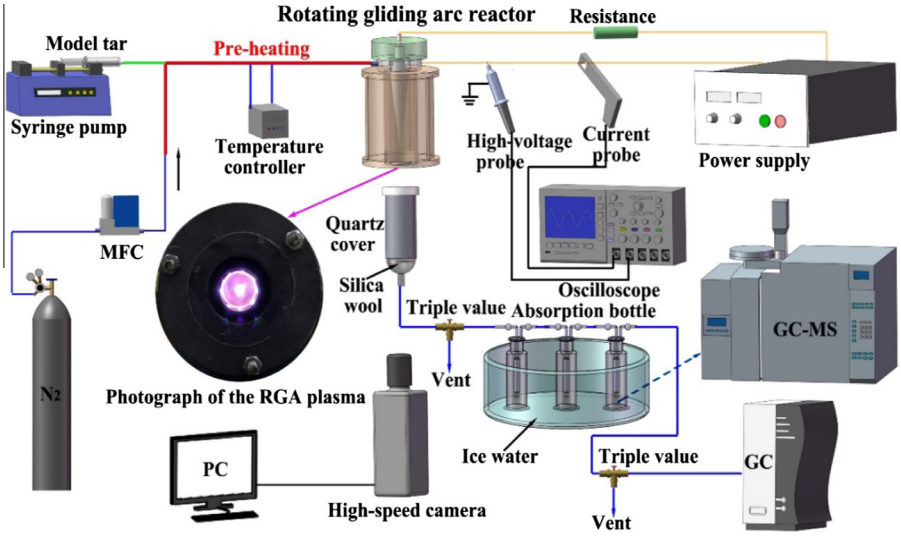

Fig. 1. Schematic of the RGA plasma experimental setup for tar destruction.