

Review

Overview of tar reduction in biomass-derived producer gas using non-thermal plasma discharges

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Abstract: Non-thermal plasma is defined as a partially ionised gas where free electrons attain high energy levels and high temperature but where neutral particles and ions carry negligible energy, and the overall temperature of the bulk gas remains low. Non-thermal plasma discharges have been widely applied in various areas such as surface modification, gas discharge lamps, low-temperature plasma chemistry, and pollutant abatement. This work investigates the potential of non-thermal plasma in the treatment of tar. In addition, an overview of recent development in the removal of tar from biomass-derived producer gas is presented with a focus on non-thermal plasma utilisation. The background of tar and non-thermal plasma technology is described and followed by up-to-date progress in applying non-thermal plasma to tar destruction. The effects of the input power and input tar concentration on the destruction efficiency, energy efficiency and gas composition are presented and the performance of several plasma systems is compared and discussed.

Keywords: biomass tar, producer gas, tar reduction, non-thermal plasma discharges

INTRODUCTION

Renewable energy is of growing importance in satisfying environmental concerns over fossil fuel usage. The environmental problems from fossil fuel usage are increasingly serious, manifesting as atmospheric pollution, ozone depletion and climate change. Biomass energy corresponds to the energy derived from plants or plant-derived materials. Hence biomass is a renewable energy source with great future potential since electricity and emissions generated during the thermal conversion of biomass can be considered carbon neutral [1, 2].

Biomass gasification is a proven thermal conversion technology that transforms organic materials into combustible gases. These gases are mainly carbon monoxide (CO), hydrogen (H₂),

methane (CH₄) and higher hydrocarbons in minor quantities. This fuel gas can be utilised in internal combustion engines or gas turbines to produce heat, power and chemicals [3-6].

Tar is generally known to be the main nemesis for biomass gasification. It is a complex mixture of condensable hydrocarbons. The tar content of the producer gas from gasification can range between 1 g/m³ to 100 g/m³, depending on the gasifier type. Problems caused by the presence of tar include blockage, corrosion and clogging of downstream fuel lines, filters, engine nozzles and turbines [7-12]. This results in extra maintenance costs and unplanned plant shut-downs. The impact of high tar content in the producer gas is a major hurdle to the deployment of the technology, particularly where very clean gas streams are required, such as high-efficiency internal combustion engines and fuel cells.

There have been several studies on the post-treatment of tar using physical, thermal cracking, and catalytic cracking methods. Physical cleaning with scrubbers, cyclones, filters and wet-type electrical precipitators are appropriate for large-sized plants, with low environmental contamination [3]. Catalysis is applicable, but this is at the risk of contamination from sulfuric acid, chlorine and nitrogen compounds, as well as coke. Thermal cracking normally requires temperatures greater than 800°C and high energy consumption [6, 13, 14].

Recently, plasma, the fourth state of matter, has been applied to the removal of tar. Plasma is a mixture of ions, electrons and neutral particles. It can be categorised as (i) high-temperature plasma, which is thermodynamically equilibrated plasma, and (ii) low-temperature plasma, which can be subdivided into local thermal equilibrated plasma (thermal plasma) and thermodynamically non-equilibrated plasma (non-thermal plasma) [1]. The non-thermal plasma technology has been shown to have great potential for decomposing tar with relatively low energy consumption. It has been used for hydrogen production through methane reforming and destruction of volatile organic compounds [15, 16]. The technology provides an attractive and promising alternative to the conventional approaches used for removing tar. In non-thermal plasmas, the bulk gas temperature can be as low as room temperature, while the typical energy of the energetic electrons ranges between 1–10 eV. Non-thermal plasmas can easily break most chemical bonds and overcome the disadvantage of high temperature required by thermal and catalytic processes. High reaction rates and the rapid attainment of the steady state in a plasma process enable a quick start and shutdown of the plasma process compared to other thermal treatment technologies. This offers a promising route for clean fuel production [17].

However, there has been relatively little research which focused on the use of the non-thermal plasma gas cleaning for tar removal. Chun et al. [10, 11, 13, 18] studied biomass tar destruction using a gliding arc plasma reactor, and modelled anthracene, benzene and pyrene as tar. They found that the destruction efficiency ranged between 82 - 96%. In addition, Kunnikar and Tippayawong [19] and Tippayawong and Inthasan [20] investigated light tar destruction by conventional gliding arc reactors and used naphthalene to model the tar compound. The destruction efficiency was reported to be more than 68%. Nair [21] and Nair et al. [22] employed a high voltage (80 kV) pulsed corona for the removal of model tar compounds. Nunnally et al. [23] adopted a forward vortex flow gliding arc reactor for the removal of tar. They reported a destruction efficiency of 69–93% for naphthalene and more than 83% for toluene, with a maximum total flow rate of 4.38 m³/h. Zhu et al. [24] studied toluene destruction using a rotating gliding arc reactor. They achieved a destruction efficiency of 95% with a total flow rate of about 0.24 m³/h. Non-thermal plasma seems to have great potential for removing tar from biomass gasification.

In this review paper the application of the non-thermal plasma technique to the tar destruction process for biomass-derived producer gas is analysed. Recent researches on the destruction of biomass tar or volatile organic compounds by this technique are presented. The destruction efficiency of various non-thermal plasma sources and the possibility of using them in biomass gasification are also discussed.

GASIFICATION OF BIOMASS

Gasification is referred to a thermochemical process which converts organic materials or solid fuels into gaseous products, using a gasifying medium such as air, oxygen or steam, either alone or in a mixture. The temperature during the process usually ranges between 800-1,000°C, depending on the operational condition and reactor design. The main combustible composition of the producer gas is carbon monoxide (CO), hydrogen (H₂) and methane (CH₄). Figure 1 shows the conversion of solid fuels into producer gas via gasification. The producer gas has three classifications: a low calorific-value gas (3.8-7.6 MJ/Nm³) which can be used in gas turbines or for direct combustion in heating systems; a medium calorific-value gas (10-16 MJ/Nm³) which can be used for synthesis of fuels and chemicals such as methanol; and a high calorific-value gas (> 21 MJ/Nm³) which is suitable for fuel cells [25-27]. Gasification inside a gasifier involves four sub-processes: drying, pyrolysis, oxidation and reduction, which is summarised in Table 1.

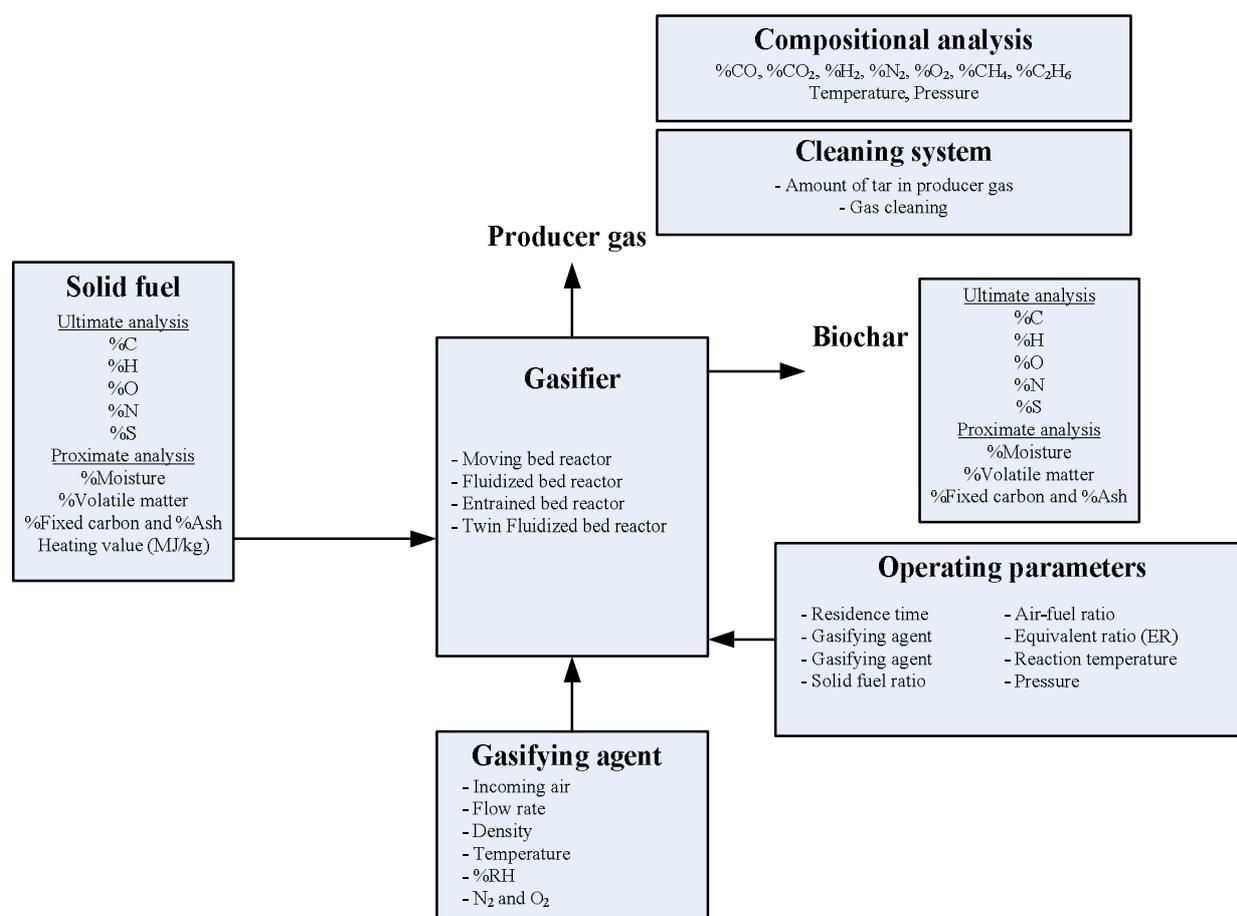


Figure 1. Schematic of solid fuel conversion by gasification to producer gas [26, 28]

The homogeneous reactions (reactions 9-12) are almost instantaneous at high temperature conditions, in contrast to the heterogeneous reactions (reactions 1-8). Three of these reactions are independent; the water gas reaction (6), the Boudouard reaction (7), and hydrogasification (8). In the gas phase, these reactions can be reduced to only two; the water gas shift reaction (11) which is the combination of the reactions (6) and (7), and the methanation reaction (12) which is the combination of the reactions (6) and (8) [29, 30].

Table 1. Overview of biomass gasification reactions [29-32]

Reaction	Reaction equation-Reaction enthalpy (ΔH^a)	
$C_nH_mO_k$ partial oxidation	$C_nH_m + n/2 O_2 \rightleftharpoons m/2 H_2 + n CO$: (exothermic)	(1)
Steam reforming	$C_nH_m + n H_2O \rightleftharpoons (n + m/2) H_2 + n CO$: (endothermic)	(2)
Dry reforming	$C_nH_m + n CO_2 \rightleftharpoons m/2 H_2 + 2n CO$: (endothermic)	(3)
Carbon oxidation	$C + O_2 \rightarrow CO_2$ ($\Delta H^0_{298} = -393.65 \text{ kJ mol}^{-1}$)	(4)
Carbon partial oxidation	$C + 0.5O_2 \rightleftharpoons CO$ ($\Delta H^0_{298} = -110.56 \text{ kJ mol}^{-1}$)	(5)
Water gas reaction	$C + H_2O \rightleftharpoons CO + H_2$ ($\Delta H^0_{298} = +131.2 \text{ kJ mol}^{-1}$)	(6)
Boudouard reaction	$C + CO_2 \rightleftharpoons 2CO$ ($\Delta H^0_{298} = +172.52 \text{ kJ mol}^{-1}$)	(7)
Hydrogasification	$C + 2H_2 \rightleftharpoons CH_4$ ($\Delta H^0_{298} = -74.87 \text{ kJ mol}^{-1}$)	(8)
Carbon monoxide oxidation	$CO + 0.5O_2 \rightarrow CO_2$ ($\Delta H^0_{298} = -283.01 \text{ kJ mol}^{-1}$)	(9)
Hydrogen oxidation	$H_2 + 0.5O_2 \rightleftharpoons H_2O$ ($\Delta H^0_{298} = -241.09 \text{ kJ mol}^{-1}$)	(10)
Water gas shift reaction	$CO + H_2O \rightleftharpoons CO_2 + H_2$ ($\Delta H^0_{298} = -41.18 \text{ kJ mol}^{-1}$)	(11)
Methanation	$CO + 3H_2 \rightleftharpoons CH_4 + H_2O$ ($\Delta H^0_{298} = -206.23 \text{ kJ mol}^{-1}$)	(12)

^a T=298 K, P=1.013 × 10⁻⁵ Pa

TAR

Tar is a complex mixture of condensable hydrocarbons including aromatic compounds with up to five rings (which can be oxygenated) as well as polycyclic aromatic hydrocarbons (PAHs). The International Energy Agency, the Directorate General for Energy of the European Commission and the US Department of Energy met in Brussels in the spring of 1998 and defined tar as all hydrocarbons with a molecular weight higher than benzene. A classification of tar was proposed, based on the solubility and condensability of the different tar compounds. Both of these classification systems are equally important and complement each other, as shown in Table 2 [31, 33-35].

Tar formed during the condensation of producer gas can be classified as primary, secondary and tertiary compounds. Formation of primary tar during pyrolysis occurs at temperatures below 500°C. During oxidation, when the temperature is above 500°C, the primary tar transforms and begins to rearrange as secondary tar. When the temperature is higher than 800°C, tertiary tar forms. Tertiary alkyl products include methyl derivatives of aromatics such as methyl acenaphthylene, methylnaphthalene, toluene and indene. In addition tertiary products of polynuclear aromatic hydrocarbons (PNA) show the aromatics without substituents (tertiary-PNA) such as benzene, naphthalene, acenaphthylene, anthracene/phenanthrene and pyrene, as shown in Figure 2 [36, 37].

The composition of tar depends on the reaction temperature, the type of reactor and the feed stock. The main components of biomass tar are naphthalene, ethylbenzene and acenaphthylene, as shown in Figure 3 [38].

Table 2. Tar compound classification [31, 33, 35, 39, 40]

Tar class	Class 1	Class 2	Class 3	Class 4	Class 5
Class name	<i>GC-undetectable</i>	<i>Heterocyclic</i>	<i>Light aromatic (1 ring)</i>	<i>Light PAH compounds (2–3 rings)</i>	<i>Heavy PAH compounds (4–7 rings)</i>
Property	<i>Very heavy tar, cannot be detected by GC</i>	<i>Tar containing hetero atoms; highly water-soluble compounds</i>	<i>Usually light hydrocarbons with single ring; it is not a problem regarding condensability and solubility</i>	<i>2- and 3- ring compounds; condense at low temperature even at a very low concentration</i>	<i>Larger than 3 rings; these components condense at high temperatures</i>
Representative compounds	<i>Determined by subtracting the GC-detectable tar fraction from the total gravimetric tar</i>	<i>Pyridine, phenol, cresols, quinoline, isoquinoline, dibenzophenol</i>	<i>Toluene, ethylbenzene, xylenes, styrene</i>	<i>Indene, naphthalene, methylnaphthalene, biphenyl, acenaphthylene, fluorene,</i>	<i>Fluoranthlene, pyrene, chrysene, perylene, coronene</i>

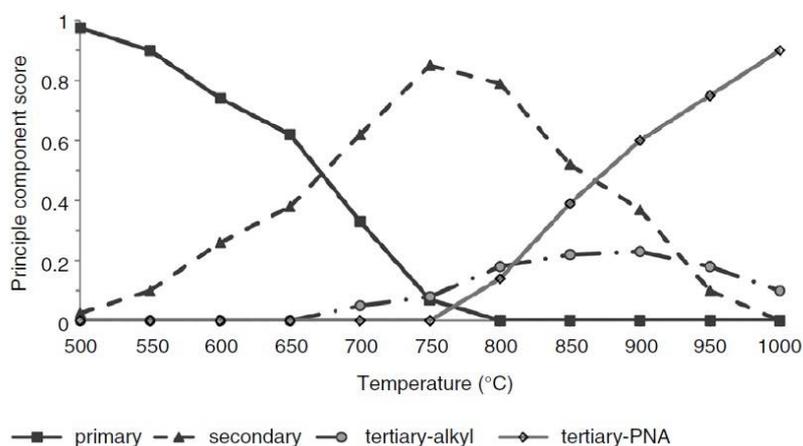


Figure 2. Distribution of different types of tar as a function of temperature [36, 37]

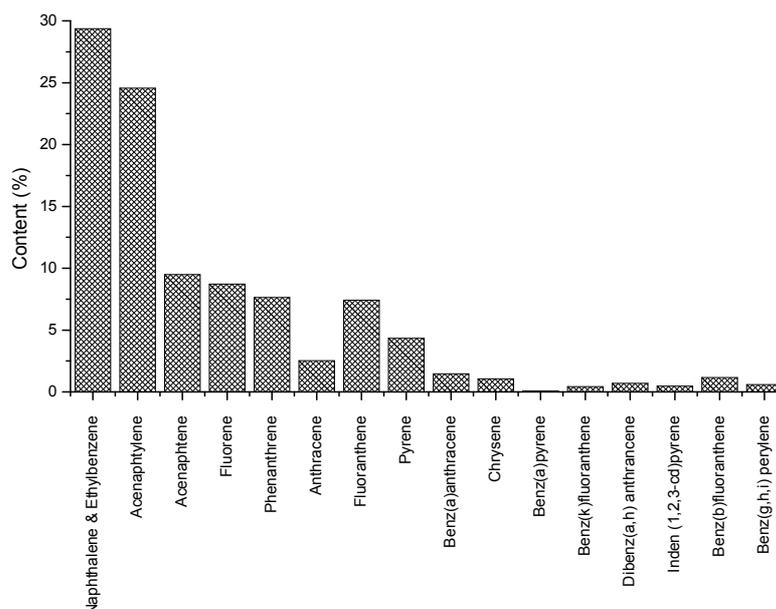


Figure 3. Typical composition of biomass tar from the gasification process [38]

METHODS OF TAR REDUCTION

Most applications of producer gas require low tar content in the order of 50-100 g/Nm³, and several options are available for tar reduction [39]. Two methods are available for tar removal, depending on which stage tar is removed. The primary method refers to treatment of tar inside a gasifier at high temperature. The main solutions proposed in the literature include optimised design of the gasification reactor and its operating parameters (temperature, pressure, oxidising agent/feed ratio, residence time), with addition of catalysts or by plasma treatment. The high gasification temperature (927-1,027°C) minimises the tar quantities and allows the destruction of the aromatics without a catalyst. A reduction of more than 40% has been reported when the temperature was increased from ~ 727°C to ~ 927°C [30, 41].

The secondary treatment approach is a method which uses separate reactors to reduce the tar content in the producer gas. Secondary tar cleaning techniques can be further divided into wet and hot gas cleaning methods. The basic mechanisms used for eliminating tar include thermal cracking, catalytic cracking, mechanical methods (scrubbing, filtration, cyclonic separation, electrostatic precipitation), and plasma cracking method [42]. The plasma method has been widely investigated for pollution control. Several studies show that the plasma method can easily remove CH₄, SO₂ and NO_x from gaseous pollutants. It can also be used as an alternative to the catalytic and thermal treatments [39]. Non-thermal plasma discharges include pulsed corona, dielectric barrier discharges, DC corona discharges, radio frequency plasma, microwave plasma, gliding arc plasma, forward vortex flow and reverse vortex flow gliding arc plasma.

PLASMA-ASSISTED REDUCTION OF TAR

The mechanisms of tar removal in a plasma discharge reactor may be explained as plasma cracking and carbon black formation reaction by equations (13) and (14). With steam introduced into the plasma reactor, radical formation, conversion, depletion and reaction mechanisms for carbon black can be expressed with the following equations (15) to (21).

Cracking reaction



Formation reaction of carbon black



Radical formation reaction



Radical conversion reaction



Radical depletion reaction



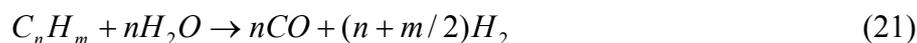
Carbon black conversion reaction



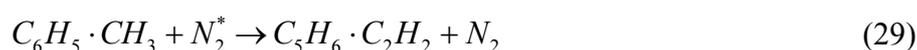
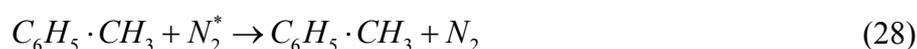
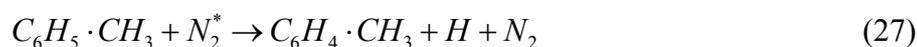
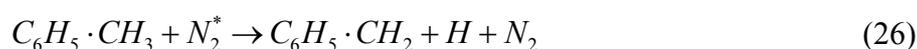
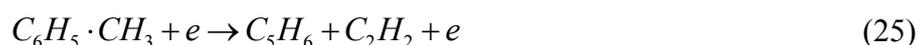
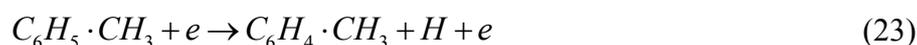
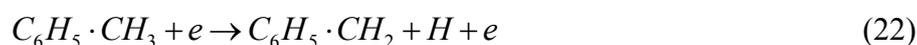
Steam conversion reaction



Hydrocarbon steam reforming reaction



Moreover, Zhu et al.[24] and Liu et al.[8] showed that the destruction of low-concentration toluene in nitrogen plasma can be initiated through two major reaction pathways: H-abstraction of the methyl group and breaking of the benzene ring through reactions induced by energetic electrons and excited N₂ states such as N₂ (A³) and N₂ (a') shown in equations (22)-(29). N₂^{*} in equations (26) and (29) refers to an excited nitrogen molecule. Chun et al. [13] used pyrene as a model tar and reported that the gases after tar destruction included CO, CO₂, H₂, CH₄, C₂H₄ and C₂H₆. The destruction of naphthalene, anthracene and benzene was found to result in CO, CO₂, H₂, CH₄, C₂H₂, C₂H₄, C₂H₆ and C₃H₈ [9, 13, 14].



PLASMA DISCHARGES

For any physical substance, as temperature increases, its molecules become more energetic and transform its phase in the following sequence: solid, liquid, gas and plasma [16]. Plasma is thermally and electrically conductive due to the charged particles and can be partially ionised or fully ionised [43]. The key process in plasma is ionisation, i.e. the conversion of neutral atoms or molecules into electrons and positive ions. Ionisation is the first elementary plasma-chemical process to be considered. Being highly energetic, plasma can enhance chemical reactions. The energy needed to form the plasma can be electrical, thermal or optical. In industry plasma is currently used in many applications such as coating, thin films, gasification, flow control, and sterilisation [43, 44]. The temperatures of the species such as the electrons, ions and gases determines the type of plasma as high- or low-temperature [44] whose relationship is shown in Figure 4.

Plasma can be classified as thermal and non-thermal, the primary difference being how energy is deposited onto the plasma gas stream. Their characteristics are shown in Table 3 [45]. In thermal plasma the electrons and the heavy particles are at the same temperature, i.e. in thermal equilibrium. Likewise, if all species are at high temperatures such as between 4,000 K (for easily ionised elements) and 20,000 K (for difficult ionisation), the plasma is called high-temperature plasma. For non-thermal plasma, the temperatures of the plasma components are not the same, the electrons often having a greater temperature than that of the other gas components. The gas molecules in non-thermal plasma are often at or near room temperature and the plasma is referred to as non-equilibrium plasma. Likewise, if the electron temperature is much higher than that of the ion and neutral gas species, the plasma is called low-temperature plasma [43, 44].

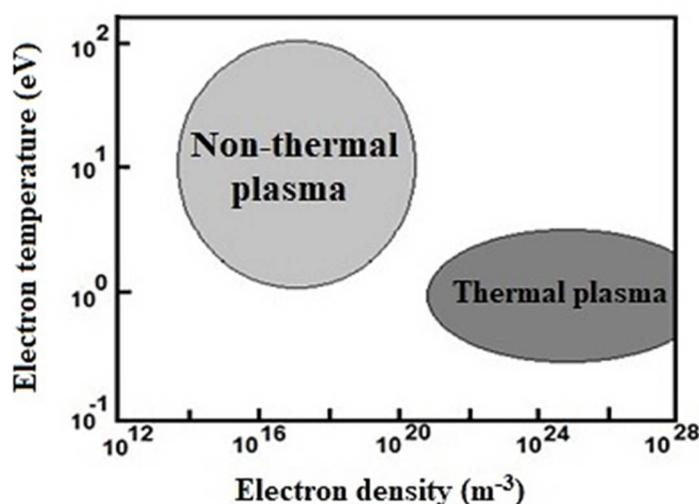


Figure 4. Classification of plasma in terms of electron density and electron temperature

Table 3. Typical characteristics of thermal and non-thermal plasma [45]

Equilibrium plasma / Thermal plasma	Non-equilibrium plasma / Non-thermal plasma
$T_e = T_h$	$T_e > T_h$
High electron density: 10^{21} - 10^{26} m ⁻³	Lower electron density: $<10^{19}$ m ⁻³
Inelastic collision between electrons and heavy particles creates plasma reactive species, whereas elastic collision heats heavy particles (energy of electrons is thus consumed).	Inelastic collision between electrons and heavy particles includes plasma chemistry. Heavy particles are slightly heated by a few elastic collisions (which is why energy of electrons remains very high).
$T_e = T_h \sim 10,000$ K	$T_e \sim 10,000$ - $100,000$ K $T_h \sim 300$ - $1,000$ K

Note: T_e represents electron temperature; T_h represents heavy particle temperature.

NON-THERMAL PLASMA TECHNIQUES

Non-thermal plasmas are an appealing and relatively new alternative for the synthesis of fuels and chemicals, offering a unique way to enable thermodynamically unfavourable reactions (e.g. biogas reforming) to occur at atmospheric pressure and low temperatures. Non-thermal plasmas show a unique non-equilibrium character and can produce highly energetic electrons and a variety of chemically reactive species, notably radicals, excited atoms, molecules and ions, which are capable of initiating many physical and chemical reactions. Non-thermal plasmas including dielectric barrier discharge [46-48], corona discharge [6, 49, 50], microwave discharge [51-53], gliding arc [8, 10, 13, 18] and vortex flow gliding arc plasma [23, 54, 55] have been investigated for reforming biogas into syngas. There are many researchers that have studied and developed tar destruction using these plasma techniques.

Dielectric Barrier Discharge

Dielectric barrier discharge (DBD), known as silent discharge, is used widely in ozone production on a large industrial scale. DBD is one of the most commonly used discharge types to produce plasmas and has been employed to treat various gaseous pollutants such as volatile organic compounds (VOCs), sulfides (CS₂, H₂S and SO₂), nitrogen oxides and some emerging and special contaminants (e.g. SO₂F₂, perfluorocarbons and CH₂Cl₂) [56]. Two different arrangements of DBD have been used (Figure 5) [48]. The advantage of this non-thermal plasma lies with the ease of its operation at atmospheric pressure. Nonetheless, DBD cannot treat air pollutants completely and may lead to the generation of potentially harmful by-products. At prolonged reaction time and high pollutant concentrations, carbon deposition can easily occur on the surface of the reactor walls and electrodes, negatively affecting the performance of air purification [57].

In a DBD reactor an AC high voltage, typically of 10 - 20 kV and 50 - 2000 Hz, is applied to electrodes, one or both of which are covered with a thin dielectric layer such as glass. The gap between the electrodes is in the order of a few hundred μ m to several mm. The current density of the micro discharge is approximately 1 kA/cm². The diameter is about 0.1 mm and the pulse duration is about 3 ns. Because energetic electrons are generated in this micro discharge, various radicals and ions are produced by electron collisions with gas molecules. These radicals diffuse into

the barrier discharge space and react with background gases. As a result, ozone generation and NO_x or VOCs removal are realized [48].

Abdelziz et al. [46] studied the effect of voltage waveforms on the performance of surface DBD reactor for naphthalene destruction. Specifically, the waveform that achieved a destruction efficiency between 30-60% consumed power between 0.15-0.4 W. Mohanty et al. [47] investigated the destruction of nitrobenzene and chlorobenzene using a DBD reactor. The electron temperature was found to be about 20,000 K or ~1.8 eV equivalent for an applied voltage of 7 kV, a mass flow rate of 1 L/min and a current of 0.3 mA. The electron temperature and electron density increased as the applied voltage increased, showing enhancement of ionisation as well as increased nitrobenzene destruction.

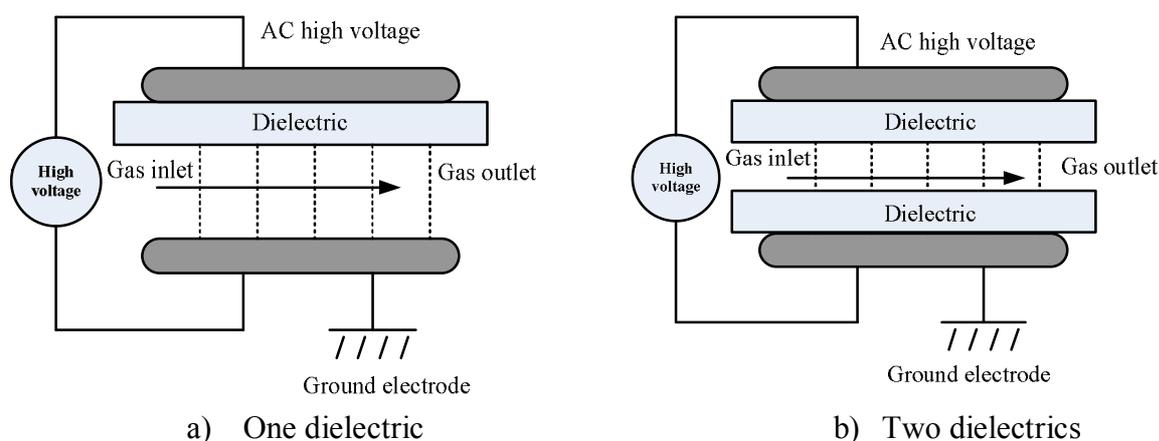


Figure 5. Schematic of dielectric barrier discharge [48]

Corona Discharge

A non-thermal corona discharge is a weak luminous discharge which occurs at atmospheric pressure near sharp tips, edges or thin wires when the electric field is sufficiently large. The corona discharge is always non-uniform; strong electric fields, ionisation and luminosity are located in the vicinity of one electrode [16]. The corona discharge can be positive or negative, depending on the polarity of the field and the electrode geometrical configuration. Positive corona in the needle-plate electrode configuration is shown in Figures 6 and 7.

The application of continuous coronas is restricted by low current and power, resulting in a low rate of treatment. DC corona discharges are used in electrostatic precipitators. For environmental applications (e.g. VOC removal, water purification), corona discharges operated by pulsed high voltages are usually adopted since higher densities of reactive species can be achieved at higher voltages. Pulsed corona discharges are characterised by plasma regions which fill a much larger fraction of the discharge gap than those generated by DC or low corona discharges [49]. Nair et al. [22] investigated the destruction of tar using pulsed corona discharges, showing a conversion of 39% with an energy density of 148 J/L. Dust removal efficiency between 72-95% was reported for the wood gasifier test and conversion of heavy tar into lighter tar was shown at an energy density of about 200 J/L [22]. Pulsed corona plasma system was also studied for gas cleaning at a temperature of about 500°C. The power discharge, mass flow rate, type of corona reactor and type of catalysts were reported to have a slight effect on tar destruction [6, 22, 26, 50].

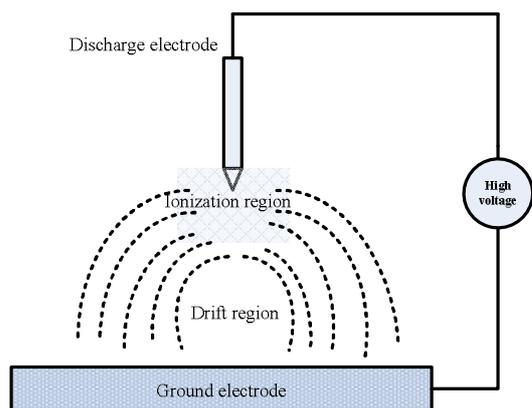


Figure 6. Schematic of a corona discharge [45]

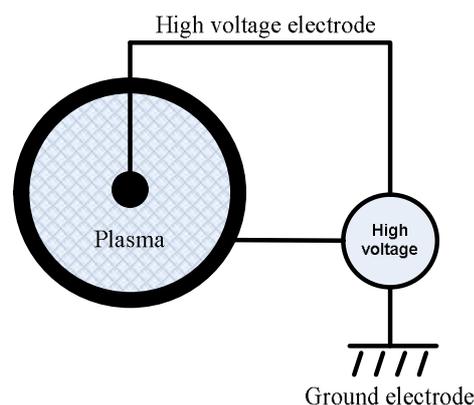


Figure 7. DC-corona discharge reactor [47]

Microwave Discharge

Microwave has great potential for gas reforming and elimination of volatile matter. Microwave plasma works on GHz frequency ranges. A microwave plasma reactor consists of a generator, wave guides and a resonance cavity inserted into a quartz tube [58]. The schematic of a microwave plasma source is shown in Figure 8. Microwave plasma is unique for volumetric heating which enables fast heating, thus accelerating and enhancing chemical reactions through efficient energy transfer [59]. Microwave irradiation affords a means of increasing the yield of a chemical reaction and of achieving significant energy savings, shorter processing times, and improving yields.

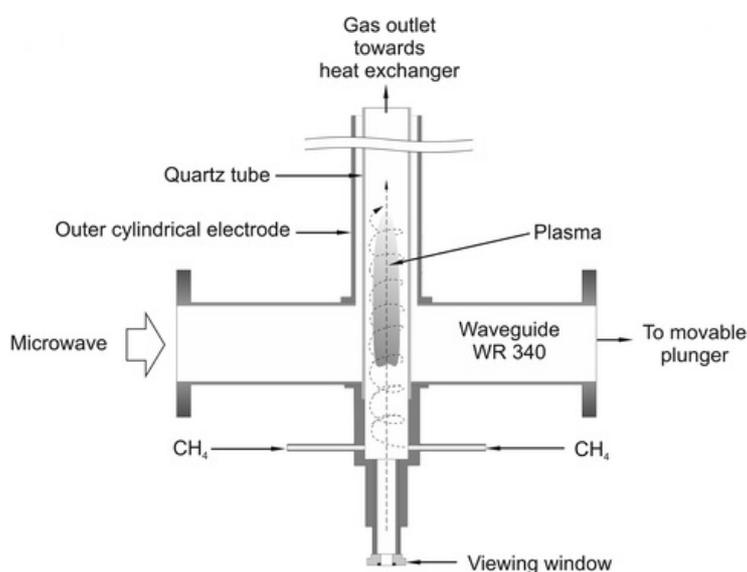


Figure 8. Schematic view of a microwave plasma set-up [63]

The application of heating by microwave irradiation to the thermo-catalytic treatment of toluene has been comprehensively investigated. Microwave irradiation technology has demonstrated spectacular success in tar cracking with good efficiency and low energy consumption [51, 60, 61]. There were studies applying microwave plasma to the destruction of VOCs and the reforming of methane into hydrogen [51, 53, 58]. Jamroz et al. [62] studied a microwave plasma reactor for the conversion of various tar model compounds (e.g. benzene, toluene and 1-

methylnaphthalene). They showed that a process efficiency of 98% could be achieved with an initial tar concentration of 10 g/Nm³ and a nitrogen gas flow rate of 30 L/min. Sun et al. [52] investigated the application of microwave plasma to tar destruction using toluene. Uniform tungsten electrodes were deployed and optimised to provide a fast ignition and a relatively stable and sustainable discharge process. Microwave tungsten discharge could effectively destruct toluene into useful gases of H₂, C₂H₂ and CH₄ and solid carbon, with a high conversion efficiency of more than 90%.

Gliding Arc Plasma Discharge

Gliding arc (GA) plasma is an auto-oscillating discharge between at least two diverging electrodes submerged in a gas flow. Self-initiated in the upstream narrowest gap, the discharge forms the plasma flame connecting the electrodes. This flame is drawn by the gas flow towards the diverging section downstream, and the plasma flame grows with increasing electrode distance until it is extinguished. The plasma flame reignites itself at the minimum distance between the electrodes to start a new cycle. The discharge starts at an electrode gap of about 1-2 mm [16]. Figure 9 shows a flat GA discharge.

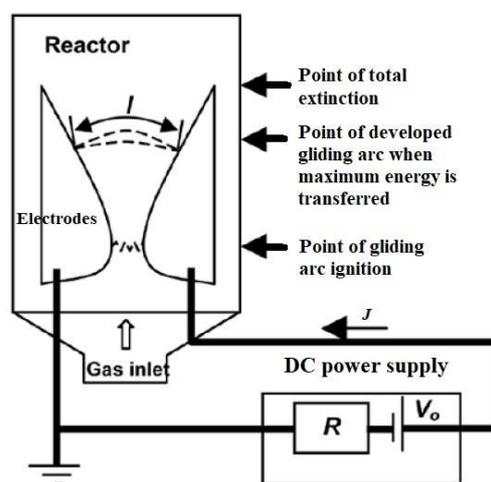


Figure 9. Characteristic of a flat GA discharge [16]

The GA plasma is compact, quick starting and has a fast response. The GA discharge has been used in developing a GA plasma reformer for tar reduction. In a previous study, Tippayawong and Inthasan [20] applied a GA plasma reactor to tar destruction using naphthalene as a tar model compound. The GA plasma reactor was made of stainless steel with two knife-shaped electrodes with a gap distance of 5 mm between them. The maximum supply voltage and current were 15 kV and 30 mA respectively. A naphthalene destruction efficiency of 70-95% was reported at an applied power density of 0.95-2.9 kWh/m³. A similar study with a flat-shape GA and a DC-pulse 20 kV voltage showed that a high destruction efficiency of 60-80% could be achieved with a single pass, and an efficiency of more than 90% could be achieved with two-stage reactors [19, 20]. Another study with three-blade electrodes of the GA plasma reactor showed a tar destruction efficiency of 96% with an energy utilisation efficiency of 1.14 g/kWh [10, 18]. Moreover, Yang and Chun [14] investigated effects of electrode length on the destruction of naphthalene. The optimum length of the electrode was found to be 95 mm for a three-blade GA plasma reactor. Benzene was used as model compound and the destruction efficiency and energy utilisation efficiency were 82.6% and

20.9 g/kWh respectively [11, 13].

Rotating Gliding Arc Plasma Discharge

A rotating gliding arc (RGA) discharge reactor (Figure 10) is a relatively novel plasma generator and has been specifically developed for the destruction of tar. Normally, a GA reactor is characterised by a plane geometry, but in many applications three-dimensional geometry is preferred. The cylindrical RGA reactor can increase the interface between the treated fluid and the active species [24]. In addition, the RGA is driven by both the magnetic fields and the tangential flow for hydrogen production and methane conversion. Compared with a conventional GA reactor, the RGA reactor enables the arc to rotate quickly without extinction, supporting a wide range of gas flow rates between 0.05-40 L/min. The advantage of the RGA reactor is an increased retention time of the reactants in the chemical reactions [64].

Recently, a toluene destruction efficiency of 95% was achieved with an RGA reactor, with a total flow rate of 0.24 m³/h and a toluene concentration of 10 g/Nm³. The power source was from a DC high voltage supply with 40 k Ω resistance. The products from the destruction of toluene were H₂, C₂H₂ and solid carbon [24]. Ren et al. [65] studied the destruction of hexachlorobenzene, a persistent organic pollutant present in flue gas from municipal solid waste using GA plasma treatment coupled with a V₂O₅-WO₃-TiO₂ catalyst downstream. The removal efficiency of 76% was reported at the gaseous concentration of about 71.6 ng/Nm³.

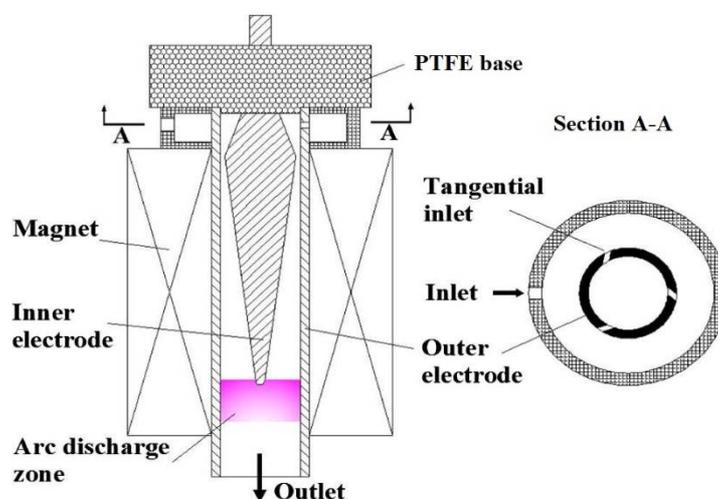
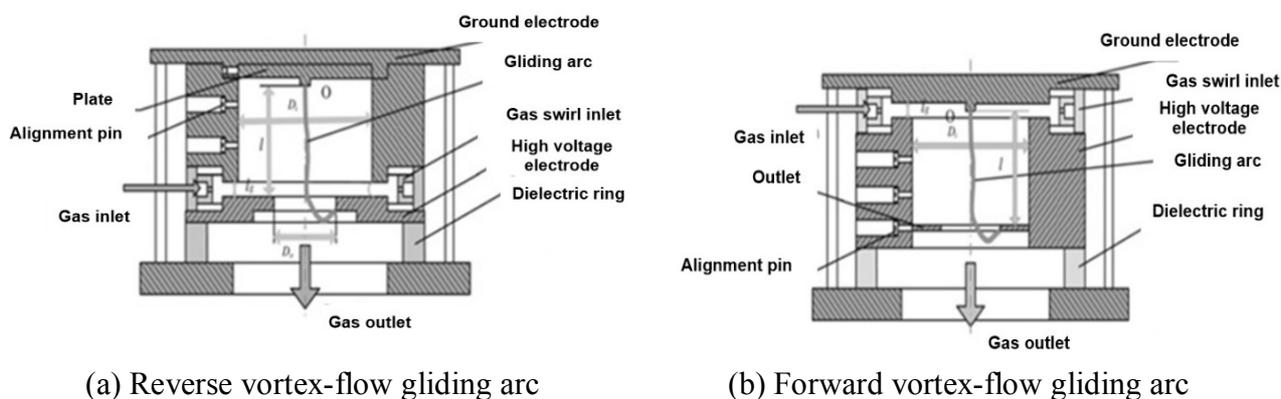


Figure 10. RGA plasma reactor showing side and top views [24]

Forward and Reverse Vortex Flow Gliding Arc Discharge

A GA plasma system for hydrocarbon conversion into synthesis gas was first developed with a conventional GA geometry. However, its limitations included an unsteady discharge due to the high gas velocity and the inefficient mix between the plasma and feed gas. There were two methods of overcoming these limitations: optimising the shape of the electrodes and improving the flow field in the reactor. A new reverse vortex flow of the GA plasma was developed for chemical processes, combining the GA discharge with the vortex flow [55]. The GA plasma in cylindrical geometry with a vortex provides better mixing and greater efficiency [54, 55]. More and more chemical processes have been tested with GA discharges, particularly with the vortex flow. Depending on the relative locations of the feed gas inlet and product outlet, the vortex-based GA

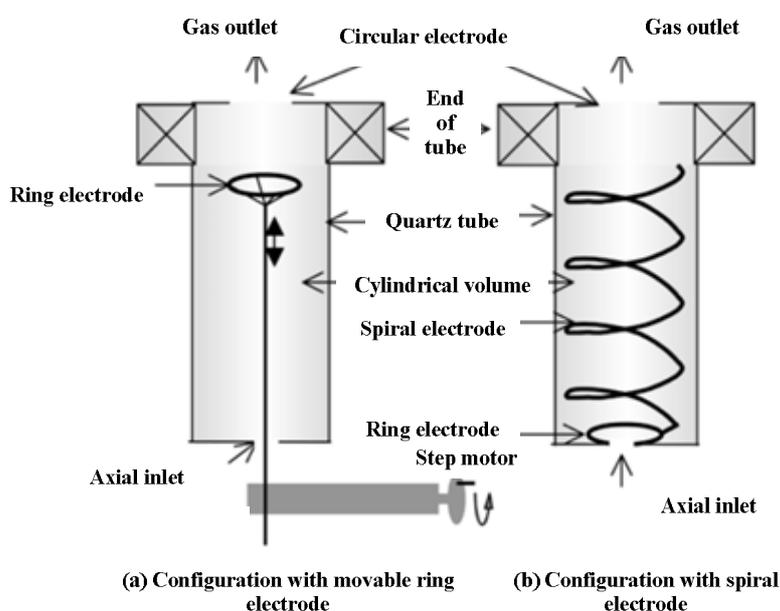
reactor can be categorised as reverse vortex flow (RVF) reactor or forward vortex flow (FVF) reactor, as shown in Figures 11 and 12.



(a) Reverse vortex-flow gliding arc

(b) Forward vortex-flow gliding arc

Figure 11. Configurations of forward and reverse vortex-flow gliding arcs [54, 55]



(a) Configuration with movable ring electrode

(b) Configuration with spiral electrode

Figure 12. Schematic of reverse vortex reactor design for GA stabilisation [66]

The FVF GA plasma has also been applied to the destruction of naphthalene and toluene. Nunnally et al. [23] studied oxidative steam reforming of a simulated producer gas combined with the naphthalene and toluene generation process. The heavy hydrocarbons, air and water vapour were fed into the reactor at a temperature above 650°C. It was reported that the naphthalene and toluene conversion efficiencies were more than 90% at a specific energy input of 0.1 kWh/m³ and an energy consumption of 62.5 g/kWh for naphthalene and 215 g/kWh for toluene. At the maximum concentration (75 g/m³), over 70% conversion of naphthalene and toluene were achieved. This reactor used a DC high voltage power supply with an operational maximum output voltage of 10 kV, a current of 1 A and an input frequency of 50/60 Hz. The flow rate was 4-4.38 m³/h for tar concentration and the specific energy input was varied from 0.06 to 0.27 kWh/m³. However, the efficiency of the RVF reactor was much better than that of the FVF system. The tornado flow

obtained in the RVF ensures high gas velocities necessary for the GA and very effective heat and mass exchange at the central zone of the plasma. Thus, the development of GA in a tornado optimizes the properties of the RVF for enhancing non-equilibrium low-current GA plasma.

Kalra et al. [66, 67] designed an RVF GA plasma using two kinds of reactor based on different cathode shapes: ring electrode reactor and spiral electrode reactor. The former required the ability to move and increase the distance between the ring electrodes to increase the length of the arc. Ren et al. [68] designed and investigated a tornado reactor by recording the arc voltages and plasma images that were strongly dependent on the gas flow configuration in the reactor. A stable and large plasma flame was generated with a tangential gas flow of 12 L/min. and an axial gas flow rate of 2 L/min., the gap between the anode end and cathode ring being 2-3 mm. The gas broke down under a high input voltage of 10 kV. The generated electric arc was driven using a tangentially injected gas between 7-12 L/min. through four tangential inlets at the bottom part and an axially injected gas between 0.8-1.5 L/min. from the inlet at the top part. The outlet was fixed at the bottom of the reactor.

In a tar destruction study using RVF plasma, Yu et al. [69] investigated the effect of tangential and axial gas injection into the plasma reactor. The power supply was a DC high voltage of 10 kV or approximately 800 W, and the current was limited by a resistor of 40 k Ω . Gallagher et al. [70] investigated a plasma-assisted conversion of heavy hydrocarbons into synthesis gas using an RVF GA reactor and a GA plasmatron. The power consumption was kept constant at approximately 250W at a tetradecane feed rate of 0.15 g/s. Energy conversion efficiency as high as 80-90% was reported.

Table 4 shows preliminary results of the developed plasma-assisted tar destruction systems as compared with other techniques. The destruction efficiency is between 65-100%. The main differences are in the types of GA reactors, gas flow rates and carrier gas components used. The conventional GA two-blade electrode in a flat box shows a high destruction efficiency up to 100% when two successive stages are employed. The three-blade GA plasma is shown to have a higher total gas flow rate and concentration of tar because of the cylindrical geometry. Both the FVF and rotating GA are also shown to handle higher gas flow rates than the conventional GA reactor. Vortex flow in a reactor appears to give good mixing and increase the retention time for tar destruction. So far, non-equilibrium plasma reactors still consume relatively high energy input. Consequently, more research is required to reduce the input energy demand and to perform tests with real biomass tar rather than just model compounds.

Table 4. Comparison between different GA configurations for removal of tar

Reference	Type of reactor	Tar model	Power source	Concentration (g/m ³)	Gas flow rate (L/min)	Removal efficiency (%)	Specific energy input (kWh/m ³)
[20]	Conventional GA, flat	C ₁₀ H ₈	AC-HV 0.45kVA	0.13-0.69	5-15	90-100	0.95-2.9
[19]	Conventional GA, flat	C ₁₀ H ₈	Pulsed-HV DC 20kV	0.21-0.81	5-15	64.04-87.12	0.014
[10]	3-blade GA, cylindrical	C ₁₆ H ₁₀	AC-HV 15kVA	0.13	12.05	88.30	0.91
[18]	3-blade GA, cylindrical	C ₁₄ H ₁₀	AC-HV 15kVA	0.21	12.05	96.10	1.14

Table 4. (Continued)

Reference	Type of reactor	Tar model	Power source	Concentration (g/m ³)	Gas flow rate (L/min)	Removal efficiency (%)	Specific energy input (kWh/m ³)
[9]	3-blade GA, cylindrical	C ₁₀ H ₈	AC-HV 15kVA	57.22	18.40	79	1.0
[24]	Rotating GA, cylindrical	C ₇ H ₈	DC-HV 10kVA	4-14	4-16	95	0.17-0.69
[23]	FVF GA, cylindrical	C ₁₀ H ₈	DC-HV 10kVA	7-31	71.67	69-93	0.1

Note: C₁₀H₈= naphthalene, C₁₆H₁₀= pyrene, C₁₄H₁₀= anthracene, C₇H₈= toluene, HV = high voltage

CONCLUSIONS

Non-thermal plasma has been shown to be effective in removing tar from biomass-derived producer gas. Gas temperature in the plasma is crucial for product formation and distribution because it affects the recombination reaction and determines which reactions are activated. GA technology is very promising for tar removal with high destruction efficiency and low energy requirement, while DBD and corona discharges are inferior. GA technology can achieve high conversion of tar, sufficient for downstream use in internal combustion engines. Despite promising prospects, some technical challenges such as the cost and lifetime of the plasma power supply, the change in the physico-chemical property of the remediation process and the scale-up of this technology currently limit its practical use.

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