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# Gliding arc plasma oxidative steam reforming of a simulated syngas containing naphthalene and toluene

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## ABSTRACT

Conversion of a simulated syngas containing vaporized toluene and naphthalene was studied in a non-equilibrium gliding arc plasma reformer. The reformer was designed for efficient reforming of high temperature syngas (greater than 650 °C) containing heavy hydrocarbons, air, and water vapor. The reactor utilized forward vortex flow, where a preheated simulated syngas containing vaporized naphthalene and toluene tar surrogate was injected tangentially in the flow to ensure effective mixing and reforming of all components. At low tar concentration (30 g/m<sup>3</sup>), over 90% naphthalene and toluene conversion was achieved at the benchmark specific energy input of 0.1 kWh/m<sup>3</sup> and energy efficiencies of 62.5 g/kWh for naphthalene and 215 g/kWh for toluene. At higher tar concentration (75 g/m<sup>3</sup>), over 70% naphthalene and toluene conversion was achieved at the benchmark specific energy input of 0.1 kWh/m<sup>3</sup> and energy efficiencies of 93.6 g/kWh for naphthalene and 369 g/kWh for toluene. Explanations for the results include effective gas mixing and plasma chemistry, such as the very fast reaction kinetics from ions, radicals and active species, specifically hydroxyl.

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## Introduction

The product gas formed from MSW gasification contains the major components CO, H<sub>2</sub>, CO<sub>2</sub>, CH<sub>4</sub>, H<sub>2</sub>O, and N<sub>2</sub>. In addition, organic and inorganic (H<sub>2</sub>S, HCl, NH<sub>3</sub>, alkali metals) impurities and particulates are present. A wide range of organic molecules are present including low molecular weight hydrocarbons (acetylene, ethylene, etc.) to high molecular weight

polynuclear aromatic hydrocarbons (naphthalene, pyrene, etc.). The lower molecular weight hydrocarbons can be used as fuel in gas turbine or engine application. However, the high molecular weight hydrocarbons are generally referred to as “tars,” and can be defined as the downstream condensable hydrocarbon component in the product gas. Tars are problematic to downstream equipment and require more sophisticated methods of conversion/removal than lower molecular weight hydrocarbons. Consequently, tar removal, conversion,

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and/or destruction can be considered as one of the greatest technical challenges that must be overcome for the successful development of commercial advanced gasification technologies.

MSW gasification can be divided into three main stages: moisture evaporation; pyrolysis; and pyrolysis product reforming and gasification. During the first stage, moisture evaporates at around 100 °C. This moisture vaporization lowers the temperature of combustion (absorption of latent and sensible heat) and as a result, slows the combustion process. During the second stage, MSW pyrolysis, the thermal breakdown of long chains of cellulose, hemicellulose and lignin occur, with the main pyrolysis products being H<sub>2</sub>, CO, CO<sub>2</sub>, gaseous hydrocarbons, tar, volatile hydrocarbons and char. The process conditions determine the relative composition of these main products. For example, low temperature decreases the syngas yield (H<sub>2</sub> and CO) and increases tar content. The third stage is reforming and gasification. Reforming is the breakdown of heavy and volatile hydrocarbons into lighter hydrocarbons and CO, H<sub>2</sub>, and CO<sub>2</sub>. In addition, char, which consists of carbon and a small amount of hydrogen and oxygen and ash, mainly yields CO, H<sub>2</sub> and CO<sub>2</sub>.

The predominant location for tar formation is the pyrolysis (thermal) stage of MSW gasification, which occurs in the range of 800–1000 °C. This stage occurs at the optimum temperature for PAH production, where the primary tar components are Tertiary-alkyl and Tertiary-PNA tars which include benzene, toluene, naphthalene, pyrene and indene [1]. Furthermore, at 900 °C, naphthalene has been identified as the major single component of tar content in the produced gas [1]. These tars are problematic because they result in clogging and fouling of downstream equipment. As a result, the reduction of these tars has become a focal point of the gasification industry.

Unfortunately, once tars are formed, they can be quite difficult to decompose. The thermal requirement varies depending upon process parameters and the individual molecule. Simulations of thermal destruction of benzene, toluene and naphthalene have been reviewed in literature [2]. The authors have indicated very high destruction requires significant residence time and high temperature [3]. To reach very significant tar decomposition in a realistic residence time a temperature of 1250 °C is required [4]. Unfortunately, utilizing this temperature for thermal decomposition has the drawbacks of high cost and production of heavier products and agglomerated soot particles [5]. Thus, conventional thermal decomposition is not an ideal solution for tar removal and reforming.

Although the cause of tar formation has been identified as process conditions, the complicated reactions in such a system are still unclear. From a fundamental standpoint, tar decomposition and growth are called cracking and polymerization, respectively, and are the main reactions to consider in the tar reforming process. Cracking eventually results in the formation of syngas, while polymerization results in the formation of heavy hydrocarbons and the agglomeration of soot particles. Ideally, a solution should be implemented which maximizes cracking potential and minimizes polymerization as this maximizes syngas production and minimizes complications associated with tar.

Some research has been compiled on the polymerization process, and theories for tar polymerization have been suggested including H abstraction/acetylene addition, benzyl radicals and direct combination of intact aromatic rings [6]. Still, the general consensus among researchers is that radical processes are likely the driving force behind heavy hydrocarbon and soot formation and decomposition [6]. As was mentioned earlier, thermal methods are costly and often lead to polymerization, which should be avoided. An interesting alternative is utilizing plasma for heavy hydrocarbon reforming. Plasma can be considered a promising solution because it provides thermal processing of the hydrocarbons with radical generation, which has been identified as a key process, to initiate and drive reactions. As a result, plasma is the main focus of this study. Still, methods other than plasma and thermal tar removal exist and should be mentioned including mechanical methods, catalytic methods, and partial oxidation.

Many review papers have been written on the various methods of tar removal and each method will not be discussed in detail in this paper. Instead, a general overview will be presented below on the various methods with particular focus on the plasma approach.

Mechanical methods of tar removal include both dry and wet gas cleaning. Some dry mechanical methods include: cyclone, rotating particle separators, electrostatic precipitators, bag filters, baffle filters, ceramic filters, fabric filters, sand bed filters, and absorbers [7]. Still, these methods have some drawbacks such as low tar removal, high capital costs, and tar deposition, plugging, and fouling. In addition, wet mechanical methods include: spray towers, packed column scrubbers, impingement scrubbers, venture scrubbers, wet electrostatic precipitators, OLGA, and wet cyclones [7]. These methods include similar drawbacks of low tar removal, high capital costs, large size, poor regeneration efficiency, and tar deposition, plugging, and fouling. Undoubtedly, improvements to mechanical methods will be made in the future; however, incorporation of these methods at large industrial scales will continue to require significant capital and operating costs.

A plethora of catalysts exist and their application for tar conversion has been reviewed in literature [7–11]. In general, catalysts for tar conversion should be evaluated based upon basic criteria including: effectiveness for tar removal, capability of reforming methane and light HC's, suitable syngas ratio production, resistance to deactivation, regeneration difficulty, robustness and cost. Meeting this criteria can be accomplished in multiple ways as synthetic catalysts are comprised of an active catalytic phase, a promoter, and a support all of which can be varied and affect tar conversion. In addition, the catalyst is often used to facilitate steam reforming and partial oxidation. In a recent paper by Anis and Zainal, six categories of catalysts were suggested including: Ni-based catalysts, non-nickel metal catalysts, alkali metal catalysts, basic catalysts, acid catalysts, and activated carbon catalysts. From a practical standpoint, the most effort has been expended in Ni-based catalysts with various supports including alumina, dolomite, olivine, and zeolite. Ni-based catalysts have the potential to reach nearly complete tar removal at high temperatures; however, they are susceptible

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