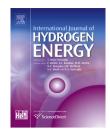
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Effect of microwave plasma torch on the pyrolysis fuel oil in the presence of methane and ethane to increase hydrogen production

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ABSTRACT

Pyrolysis fuel oil (PFO) processing by microwave plasma torch was developed for the production of hydrogen. The PFO cracking process was performed at atmospheric pressure in the absence of catalyst and effect of plasma gas on the production rate of hydrogen and light hydrocarbons (C_2-C_4) was evaluated. In the first step, effect of the applied power and the working gas flow rate was investigated. In the second step, the applied power and working gas rate were set to 650 W and 4000 sccm, respectively, which were provided by combining methane or ethane as 0%, 2.5%, 7.5%, and 20% with argon. By increasing the percentage of the existing methane in argon, production rate of the sum of the light hydrocarbons was increased and that of hydrogen was reduced, but it was more than the case when argon was applied alone. By increasing ethane percentage, hydrogen production and light hydrocarbon rate were increased. The best conditions of the plasma gas for producing hydrogen at the power of 650 W were obtained as 5CC PFO feed, 2500 sccm (80%) argon, and 500 sccm (20%). The hydrogen production rate in optimized conditions was 2343.16SCCM with selectivity of 84.41%. Sum of the obtained hydrocarbons in this test was 434.25 sccm. Another parameter in the present study was the feed volume processed by plasma. In this case, 5 cc, 3 cc, and 1 cc of the feed were tested when the plasma gas was 3000 sccm argon with the power of 650 W. The results showed that, by increasing the feed, the products were increased. In the processing of 5 cc feed with plasma, 896.41 sccm hydrogen and 61 sccm light hydrocarbon were produced.

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Introduction

Need of humans for energy and environmental pollution of expensive fossil fuels have motivated researchers to find new energy alternatives. Hydrogen is an alternative for nonrenewable energy resources, which is compatible with fuel cells. Based on its importance, researchers have attempted to investigate many methods for producing hydrogen. Most of the works have been carried out on natural hydrocarbons. Since the supplies of the natural hydrocarbons are limited, conversion (reforming, cracking and upgrading) of

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inexpensive fuel oil into valuable petroleum products such as hydrogen and lighter hydrocarbons with a greater economic value is very important for a variety of applications.

In recent studies, many works have been done to generate hydrogen by different types of plasma reactors. Results have shown that plasma has a promising effect on the heavy hydrocarbons [1–4] or methane [5–8] to produce hydrogen. Kado et al. [9] dissociated methane by means of dielectric barrier discharge (DBD), corona and spark discharge at atmospheric pressure and reported that methane was easily ionized through ions impact on carbon and hydrogen atoms at temperatures between 420 and 460 K. Using high-frequency pulsed plasma (HFPP) to convert methane into ethylene and hydrogen, Yao et al. [10] revealed better conversion efficiency and energy consumption of HFPP (high-frequency pulsed plasma) compared to traditional methods of arcing and partial oxidation/combustion (POC). Zhang et al. [11] used a DBD plasma system to convert methane and carbon dioxide and found that the ratio of produced hydrogen to carbon monoxide was increased with increasing the concentration of methane intake. Fincke et al. [12] used the thermal effect from directing methane into a reaction chamber heated at 1600 °C to form high purity hydrogen and carbon black.

There are few works which have used plasma for the conversion of liquid hydrocarbons into hydrogen. Rahimpour et al. [13,14] investigated the heavy naphtha conversion into hydrogen and light hydrocarbons by the dielectric barrier discharge plasma reactor. In more recent works, microwave plasma reactors have been investigated for hydrogen production via hydrocarbons conversion. Jasinski et al. [15] studied the hydrogen production at atmospheric pressure by the microwave plasma source. They investigated two types of an atmospheric pressure microwave plasma source (MPS) for hydrogen production via methane conversion. Their experiments showed that MPSs had a high potential for hydrogen production via hydrocarbon conversion. Ogungbesan et al. [16] investigated the experimental validation of local thermal equilibrium in an MW plasma torch for hydrogen production during methane dissociation. Ethanol conversion into hydrogen by microwave plasma reactors is also a very interesting topic in recent studies. Bundaleska et al. [17–19] studied the microwave plasma steam reforming of ethanol under vortex gas flow and atmospheric pressure conditions. The main gas products of the steam reforming were H and CO as detected by mass spectrometry and Fourier transform infrared spectroscopy.

In another work by Zhivotov et al. [20] the process of partial oxidation of kerosene ($C_{11}H_{22}$) with air was examined by the atmospheric pressure microwave plasma torch. The results showed that microwave discharge had a better effect on the process of kerosene conversion than the equivalent thermal energy input via the combustion of a portion of the fuel. Sekiguchi and Muri [21] used a microwave plasma reformer to reform the mixture of steam and hexane for hydrogen forming. They investigated the performance of a fuel cell power system of a steam plasma reformer in term of energy efficiency.

In the mentioned works, different types of plasma have been used to convert gases and pure liquid hydrocarbons. On the other hand, PFO is a very complex composition of olefins, diolefins, triolefins, aromatics, polynuclear aromatics, etc., which is called waste in petrochemical industry. Therefore, PFO was chosen as feed in the present work. In the previous work, the MW torch was designed and effect of the applied power and working gas flow rate on the cracking of pyrolysis fuel oil was investigated [22]. The results showed that microwave plasma has a good ability to convert PFO to valuable products such as: hydrogen, methane, ethane and butane. Also, increasing the applied power and working gas flow rate, increase the production rate of products. We showed when power increase from 350 W to 850 W (argon flow rate was set to 3000 sccm), the production rate of hydrogen and hydrocarbons increase from 116.5 and 16.55 sccm to 405.6 and 37.82, respectively. Also, by increasing the flow rate from 1000 sccm to 4000 sccm the production rate of hydrogen and hydrocarbons increase from 71.67 and 4.09 sccm to 446.31 and 46.78 sccm, respectively. The approach of the present work was to present a microwave plasma torch (MW torch) to increase the conversion of PFO into hydrogen and valuable petrochemical products and investigate the usage of ethane and methane as an auxiliary gas with argon for the first time. The volume of the feed was also studied. This detailed study proposed new insights into the cracking process of these parts of heavy fuel oils. The present study aimed to explain the hydrogen production process and also elaborate on the cracking of waste generated (PFO) in petrochemical industry.

Experimental

Experimental analysis of pyrolysis fuel oil

PFO feed was analyzed by gas chromatography (GC-DHA; Varian CP-3800, capillary column SilPona CB) equipped with a flame ionization detector (FID) and GC/mass spectroscopy (GC-MS; Varian CP-3800, Column CP-Sil 8). Detailed hydrocarbon analysis (DHA) is a separation technique used by a variety of laboratories involved in the petrochemical industry for analysis and identification of individual components as well as for bulk hydrocarbon characterization of a particular sample. Therefore, the results are shown in Table 1.

Microwave torch reactor setup

As shown in Fig. 1, the setup consisted of a 2.45 GHz power supply, a waveguide, a torch, a mass flow controller, and a quartz chamber. In all experiments in Sections 3.1 and 3.2, 1 cc of the feed was injected into the reactor and in Section 3.3 the

Table 1 $-$ The weight percent of the main components of fuel oil.		
Components	Wt %	Carbon number
Aromatics	40.586	C ₆ -C ₁₈₊
Iso-Paraffins	10.896	C ₆ -C ₁₁
Naphthenes	1.496	C7-C13
Olefins	9.977	C ₆ , C ₈
Paraffins	1.773	C ₁₀ -C ₁₆
Unknown residue	35.272	$C_{14} - C_{>40}$
Total	100	

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