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Microwave plasma torch for processing hydrocarbon gases

Alex G. Zherlitsyn *, Vladimir P. Shiyan, Paul V. Demchenko
Tomsk Polytechnic University, Russia

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Abstract

We designed and developed an ultrahigh-frequency (microwave) plasma torch with a combined (nitrogen, methane) plasma-forming environment, and microwave output of up to 2 kW, continuously. We demonstrate the possibility of using it in order to process natural and associated petroleum (APG) gas into valuable products (hydrogen and carbon nanomaterial CNM) with up to 70% efficiency. Based on the developed microwave plasma torch, we developed an apparatus capable of converting hydrocarbon feedstock at a capacity of 50 g/h yielding CNM and hydrogen of up to 70 vol. %. In its mobile small-tonnage version, this technology can be used on gas-condensate fields.

Keywords: Microwave plasma torch; Natural gas; Microwave discharge; Plasma; Conversion; Carbon nanomaterial; Hydrogen

1. Introduction

The requirements of today, for the rational use of nonrenewable hydrocarbon-containing natural resources, define the need to search for new effective methods and means of their deep processing. In this aspect, the processing of natural and associated petroleum gas comes in first. One of the options for the deep processing of hydrocarbon-containing gases is converting them to hydrogen and carbon nanomaterial (CNM), which are both in high demand. Significant results in the solution of this problem are obtained when using chemically active plasma of electrical discharges in gases. Among the various types of discharges (arc, barrier, high-frequency, microwave, etc.), the microwave discharge has an obvious advantage as far as impact performance on hydrocarbon gases and other fluids [1–4].

Preliminary studies on the effects of microwave discharge plasma on hydrocarbon gases have shown that there is a lack of stability among the known types of plasma torches. The operational instability is associated with carbon formation during the decomposition of the hydrocarbon gas (methane) in the microwave discharge.

Further studies have been devoted to developing a method and an apparatus that can convert hydrocarbon gases into hydrogen and carbon, based on the combined effects exposure to a metal (Fe, Ni) catalyst and microwave discharge plasma will have on the gas, at atmospheric pressure [5]. A microwave plasma torch with a “passive” initiator of microwave discharge, in the form of disordered stacking of tungsten spirals, is used as the plasma source. The developed method and apparatus have allowed us to achieve a high (up to 70%) degree of conversion of methane and up to (70 vol %) yield of H_2. However, these results were obtained at relatively low gas flow rate of about 0.16 ÷ 0.4 m^3/hr. When increasing the gas flow rate up to 1 m^3/hr and above, there is a problem of stability due to the “burning” of the microwave discharge due to the carbonization of the discharge chamber of the plasma torch, which results in a disruption of the microwave discharge.

In order to solve this problem, a new version of the microwave plasma torch was developed, having a discharge initiator, in the form of an electric gas discharge tube with a nitrogen plasma environment [6]. This gap creates an initial plasma concentration, which is sufficient to initiate the main microwave discharge. The use of this initiator allowed us to partially remove the problem of carburizing the inter-electrode gap of the discharge chamber and thus, increase the time of continuous operation of the microwave plasma torch.

In this paper, the plasma source used to process the hydrocarbon gases is the microwave plasma torch with an active radial discharge initiation system, consisting of four gas arresters arranged in the center plane of the discharge chamber. Experiments have shown that this system ensures a stable

* Corresponding author. Tomsk Polytechnic University, 30, Lenin Avenue, Tomsk 634050, Russia. Tel.: +7 3822 606399.
E-mail address: zherl@tpu.ru (A.G. Zherlitsyn).

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initiation and "burning" of the microwave discharge at elevated (greater than 1 m ^{3}/hr) convertible gas outputs, which are of practical interest.

2. Experimental plasma torch

A diagram of the microwave plasma torch is shown in Fig. 1.

The microwave plasma torch developed by the authors of this study is a waveguide-to-coaxial transition (WCT)*1*, consisting of a rectangular waveguide 2 and a coaxial line. The inner *3* and outer *4* conductor of the coaxial line form the discharge chamber. The discharge chamber has a built-in active discharge initiation system, consisting of four gas dischargers *5*, located in the center line of the cylindrical discharge chamber, with an interval of 90° along the circumference. Each of the dischargers *5* consists of a body *6*, a central electrode *7*, high-voltage input *8*, and is fitted with an individual pipeline *9* for supplying plasma gas (nitrogen) into the discharge gap.

The external appearance of the discharge initiation system is shown in Fig. 2.

The waveguide *2* is equipped with a pipeline *10* for supplying the converted gas. The input of the waveguide *2* through the ferrite valve *11* is connected to a microwave generator (magnetron 12) with a capacity up to 5 kW in a continuous mode and operating frequency (2450 ± 50) MHz. The magnetron is protected from reflected waves by using the ferrite valve. Automobile spark plugs are used as high-voltage gas discharge inputs. Waveguide *2* with a cross-section of 90x45 mm is made of stainless steel. At the output end of the waveguide there is a mounted sliding short circuit 13 so that the plasma torch can be adjusted. The inner conductor *3* of the coaxial line with a diameter of 16 mm and the outer conductor *4* with an inner diameter of 40 mm are also made of stainless steel. In order to form a microwave plasma torch discharge in its hollow inner conductor *3*, the coaxial nozzle is filled *14*.

The microwave plasma torch operates as follows. At the first stage in the area of the discharge gap of each of the arresters *5*,

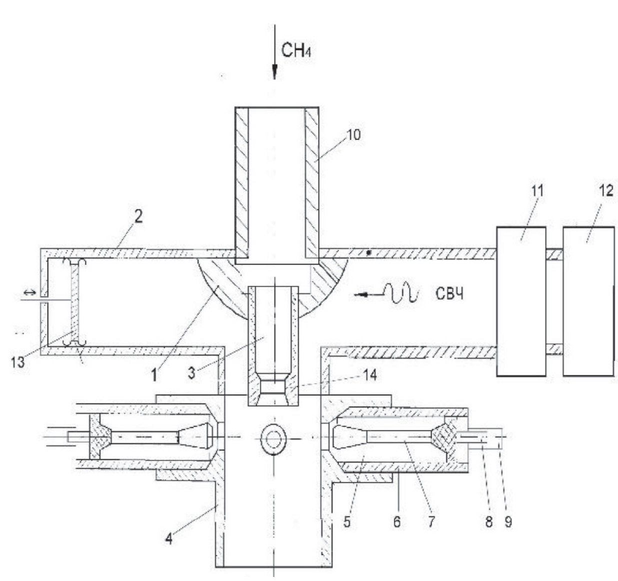

Fig. 1. Microwave plasma torch with a radial discharge initiation system.

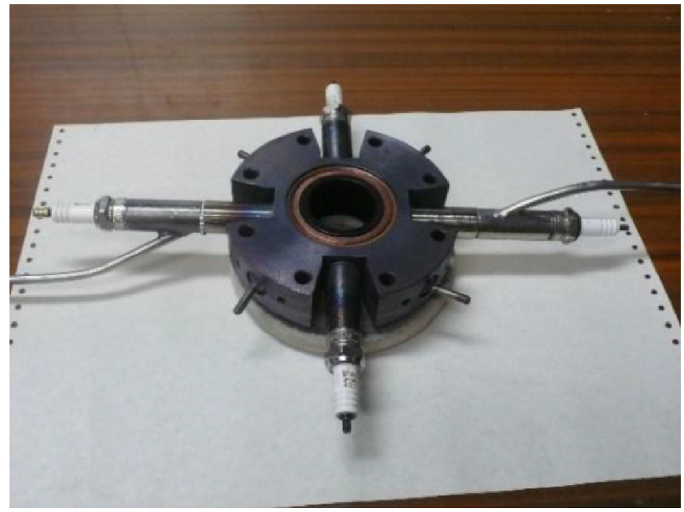

Fig. 2. The external appearance of the microwave discharge initiation system.

each individual pipeline *9* is fed nitrogen, and the central electrode *7*, through a high-voltage input *8* gets pulses of amplitude up to 15 kV at a repetition rate of 100 Hz. Pulses are generated by a high-voltage source (not shown). The electrical discharge is ignited under the influence of these impulses, which creates plasma, the concentration of which is sufficient to initiate the ground (microwave) discharge. Then, the natural gas is fed into the discharge chamber containing about 95 vol. % methane (CH_4), through the hollow inner conductor *3* of the coaxial WCT, as well as microwave power from generator *12*. The coaxial WCT line turns into the circular waveguide by virtue of its own continuation, which is the outer conductor of 4 coaxial with a diameter of 40 mm. The waveguide of this diameter is the limit for the working wave of the generator $\lambda_0 = 12,45$ cm. Due to this, in the end zone of conductor *3*, there is an increase in the electric field strength, and the microwave discharge is ignited at atmospheric pressure. Along with this, the conditions for the initiation and further maintenance of stable combustion of the microwave discharge are ensured by using the radial discharge initiation system. The process is such that the trigger (auxiliary) discharges burn in the zone having the predominant concentration of nitrogen, and the main (microwave) discharge is in the zone where the methane concentration is dominant. With this division of gas flow, we can ensure stable combustion of the microwave discharge in a broad range of convertible gas flow and levels of microwave power input into the discharge. Due to this construction, discharge initiation thereby decreases the likelihood of forming carbon "bridges" that would overlap with the discharge intervals, the reaction volume is increased, and the spatial uniformity of plasma formation is improved, which also increases the degree of methane conversion.

The methane conversion products (hydrogen and carbon) that are formed using microwave discharge plasma are removed from the discharge chamber as gas flow, which passes through a filter system after which the products are collected by their respective accumulators.

An apparatus was created based on the developed microwave plasma torch, and a series of experiments were conducted, to