

Greenhouse Gas Treatment and H₂ Production, by Warm Plasma Reforming

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ABSTRACT

Carbon dioxide (CO₂) and methane (CH₄) have been identified as the most significant greenhouse gas (GHG) arising from anthropogenic activities affecting the climatic global change. It is of great importance to reduce GHG emissions in order to counteract global warming.

This paper considers dry GHG reforming, involving synthesis gas generation followed by the production of some other solid by-products. CO₂ and CH₄ are both relatively stable compounds with low potential energies. The dry reforming reaction is highly endothermic and external energy must be provided in order to drive it in the forward direction. More recently, applications of plasma gas reforming are highlighted as promising technique for energy saving and environment safe purposes with increasing demand of hydrogen and synthesis gas. In the case of plasma reforming, high energy electron must provide not only radical species, but also enthalpy required for endothermic reaction. The conversion of hydrocarbon in by-products with high added value, is mainly contributed by dissociation and ionization, when a plasma discharge processes is used; therefore, the more energy consumed by these two kinds of reactions, the more energy-effective for hydrocarbon reforming. Although the low pressure plasma, such as radiofrequency or microwave plasma could achieve high hydrocarbon conversion and good H₂ selectivity, the low H₂ production rate and extra energy requirement for vacuum device restrict its practical use, therefore, the plasma reactor here proposed must fulfill two principal characteristics: Environment-friendly and Auto-sustainable. Warm plasma is a transitional discharge which has low specific energy requirement (1-3eV) but still maintaining enough high temperature (1000-3000K) to produce excited species supporting subsequent chemical reactions. Such plasma discharges have significant advantages: Do not require extra cooling systems, since they work with reduced electric current flows and high voltages, this avoid electrodes erosion and stainless steel walls in the reactor designs can be used.

Keywords: Plasma Reforming, Greenhouse gas, Synthetic gas



1. Introduction

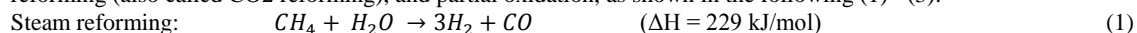
Environmental and energetic problems have enforced researchers into finding and developing alternative and renewable methods for substitute energy production. Most of the chemical energy derived from hydrocarbons today is by a well-known process called combustion. Fuel combustion is an exothermic reaction whereby a fuel is oxidized leading to energy conversion of the chemical species usually present in the GHG, and delivering exhaust gases normally composed by NO_x, CO and CO₂ considered as GHG. Over the latest decades, different laboratories around the world, have investigated the reforming capability in fuel conversion by using all types of plasma, from thermal to non-thermal ones [1-8]. Plasma researchers have utilized various reforming processes, such as Partial Oxidation Reforming (POR), Dry Reforming (DR), Steam Reforming (SR), and Steam-Oxidative Reforming (SOR) to test plasma reactors capabilities in order to replace traditional combustion process or metal catalysis-based systems. The catalytic process has the disadvantages of restricted operating conditions and problems associated with catalytic converters such as cost, sulfur poisoning and extended start up time [9-11].

In warm plasma discharge, the ionization process induced by the electric field, dominates the thermal effect and gives relatively high-energy electrons as well as excited ions, atoms or molecules leading in selective chemical transitions in a very effective manner. The average residence time of the reagents in the reaction zone is about 10⁻³ s, and then very high specific throughputs in the reaction zone are achieved. Because of its high power density, this plasma reactors can be designed for small and large -scale applications, these types of reactors can also work in portable or on-board processes [12].

Warm plasma is a transitional discharge, which lies between conventional thermal plasma and nonequilibrium plasma conditions, working under moderate power density; but still maintaining enough high gas temperature (1000–3000 K) to produce excited species supporting subsequent chemical reactions. Such plasma discharges have significant advantages: they do not require extra cooling systems, since they work with reduced electric currents and high voltages, thus avoiding electrode erosion, and the reactor has simple construction [13]. Warm plasmas are also characterized by high chemical selectivity, and has found applications in fuel conversion for syngas production, hydrogen sulfide dissociation, carbon dioxide dissociation among others. Examples of warm plasma discharges include gliding arc plasma discharge or Jet Atmospheric Pressure Plasmas.

The focus of this work consists in research the warm plasma discharge, applied for GHG reforming optimizing the electrical energy cost to produce higher syngas GHG conversion and obtaining a high yield and selectivity rates of H₂. However, hydrogen as a fuel, has significant drawbacks, especially those related with its storage. Despite hydrogen's high mass heating value (120 kJ/g), it has a very low volumetric heating value (11 kJ/l), compared to 16000 kJ/l for methanol for instance.

There are many methods to generate syngas having the desired H₂/CO ratio [14]. These methods include steam reforming, dry reforming (also called CO₂ reforming), and partial oxidation, as shown in the following (1)– (3):



The merits of dry reforming (2), compared with these other processes, include the use of carbon dioxide, which is an important contributor to global greenhouse gas inventories, as well as a theoretical product yield ratio of H₂/CO = 1. Reforming reactions (1 - 3) are not only a process of substance conversion, but a process of energy conversion, it is important that the novel technology for CH₄-CO₂ reforming should be with high conversions, large treatment capacity and easy to enlarge. To discuss the convenience of specific energy and energy conversion efficiency these concepts will be treated in section 2.1.

The real energy needed for such process cannot solely be deduced from thermodynamic calculations, since a significant amount of extra energy will be needed to overcome the energy barrier of the reaction. This amount of extra energy is not reflected in the thermodynamic enthalpy values. Moreover, these enthalpy values are defined only for a specific reaction, whereas in a plasma conversion reactor not all energy can be directed selectively into a specific reaction in a specific place; then a lot of energy is lost due to heat loss, competing chemical reactions; therefore, there will always be a large difference between the thermodynamic ideally required energy and the required energy for a real process. Besides, the rotationally and vibrationally excited species are generally considered useless for hydrocarbon reforming because of the short lifetimes (several ns) and low threshold energy (<2 eV). It is noted that the bonding energy for most hydrocarbons are generally between 3 and 6 eV. As a result, the decomposition of hydrocarbons mainly attributes to electron-impact dissociation and



ionization. As expected, a higher electric field leads to higher electron energy, which is more favorable for the electron-impact reactions responsible for hydrocarbon reforming. In other words, the energy consumed by the electron-impact reactions would be reduced while the electric field is increased. That is why; we have researched and developed warm plasma reactors for this application in GHG reforming.

1.2.- Dry reforming or Dry CO₂ reforming (DR)

Gasification is the thermochemical conversion of organic matter (i.e. hydrocarbons) into a gaseous product, which could be used directly for combustion or synthesized into fuel or chemicals. The main exhaust gas is the syngas composed by H₂ and CO in majority, including lower concentrations of CO₂, H₂O, CH₄, higher hydrocarbons and N₂.

Dry CO₂ reforming can be described as a reforming technique whereby the reaction between carbon dioxide and a hydrocarbon yields a combination of hydrogen and carbon monoxide as in eq. (2). As an alternative, several authors [6,12] have proposed to use the CO₂ produced by various industrial facilities: cement plants, blast furnaces and to conduct studies on the Dry CO₂ reforming of hydrocarbons into synthesis gas. The dry reforming produces synthesis gas with H₂/CO ratio equal to 1.

The ideal scenarios where dry reforming might be considered is where the supply of methane is contaminated with carbon dioxide and there are no ready facilities for separation, or where a product syngas with a H₂/CO ratio of unity is desired when a second process i. e. Fischer-Tropsch is required to transform syngas into synthetic oils [15].

DR is an endothermic reaction that requires a significant amount of energy in order for the reforming reaction to progress. This energy must be delivered by the warm plasma reactor.

1.3.- On Board Reforming or Partial Oxide Reforming:

A disadvantage of reforming is the thermodynamic cost of extracting hydrogen from hydrocarbon fuels; therefore the partial oxidation is an alternative to produce hydrogen at high yield rates with low spent of energy, for instance, in research scale a rate up to 30 standard LPM, is sufficient for a fuel cell to generate 3kW of electrical power. The general idea behind partial oxidation consists in using air to balance the energy required by oxidizing some of the fuel (CH₄). The N₂ participates as an inert gas in the 800-2000K temperature range; that's why the warm plasma reactor was selected because the operating temperature is not bigger than 3000K (as in thermal plasmas torch) neither lower than 500K (as in cold plasmas reactor).

If only one half of air mole is present and considering N₂ as inert gas, then chemical equilibrium predicts that the exhaust gas mixture is mostly composed by CO and H₂ with some traces of CO₂ and H₂O vapor. For a general hydrocarbon fuel, that's means that



Partial oxidation reactions proceed in two distinct phases; a rapid, highly exothermic combustion phase where all free oxygen is consumed to form heat and the products of CO₂ and H₂O; and a much slower endothermic reforming phase where residual unconverted fuel reacts with the CO₂ and H₂O plus the reaction heat to form syngas products H₂ and CO

In addition, hydrogen selectivity is an important parameter in partial oxidation processes, since the selectivity is directly related with process efficiency. In methane reforming using the partial oxidation process, hydrogen selectivity is defined as below

$$H_2 \text{ selectivity}(\%) = 0.5 * \frac{\text{mole produced } H_2}{\text{mole converted } CH_4} * 100\% \quad (5)$$

100% of hydrogen selectivity means that all hydrogen atoms of the methane molecules are converted into hydrogen molecules.



2. Experimental

The flux rate of GHG ($\text{CH}_4 + \text{CO}_2$) and the plasma gas (N_2) are previously measured and mixed, and then introduced to the reactor (See Fig. 1), which is constructed in double wall stainless steel, the gas mixture enters by the bottom side and ascends between the space formed by a double wall; having a dual purpose: to maintain the reactor wall at low temperature and to preheat the gas mixture before to be treated. The plasma discharge is generated between an external electrode, in this case the reactor wall and a central conical electrode; then it moves down to the end of conical electrode and does not detach from it, formerly, the arc column length increases slowly, just rotating by the flow, during a time represented by $(1/\omega_c)$. The arc discharge continues the spiral motion descending along the chamber reactor increasing the column length (during time $1/\omega_d$) and a new cycle is recommenced each 25 ms. The plasma jet is expelled out the reactor chamber and can flow in a free space or in a container. In our case, the post-chamber elongates the plasma jet and has a shield effect, keeping away the atmospheric air surrounding the jet, therefore, the plasma jet is spread out with a very long volume, as it was reported in [13]. Contrary, when the plasma jet blows directly in air, it has a smaller volume.

The residence time t_r of the gas inside the reactor could be calculated by the following relation:

$$t_r = [\pi(R_1 - r)^2(\ell_1) + (\pi(R_2)^2(\ell_2))] \text{ (ml)/flow rate (ml/s)} \quad (6)$$

where, R_1 the radius of the discharge chamber, R_2 the radius of the post-discharge chamber, r the radius of the internal electrode, and ℓ_1 and ℓ_2 the discharge and post discharge length respectively. As the arc length expands, the heat dissipation increases. This results in a lowered temperature, and the plasma tends to be in nonthermal plasma discharge.

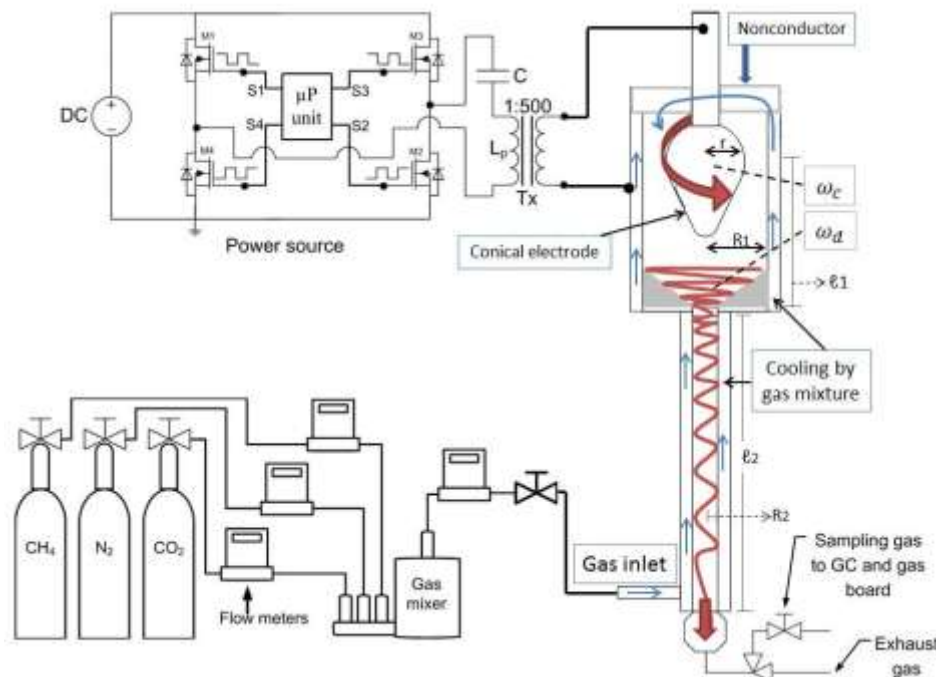


Fig 1.- Warm Plasma Reactor with power source and GHG system.

The power source consists on a high-frequency series resonant inverter [16] constituted by one setup ferrite core transformer, capable to provide 15 kV at high frequencies in the secondary winding. Four sequential control pulses ($S_1 - S_4$) generated by a simple logic circuit active the high-power MOS transistor ($M_1 - M_4$) in order to command the transformer primary side. The power supply handles a wide frequency range (5–200 kHz). The duty cycle in each phase is adjusted to 50% providing a soft start in the MOS and with the intention of avoiding unnecessary power consumption in the plasma discharge. The high frequency transformer was assembled to attain the maximum energy transfer towards the plasma discharge; besides, it also



functions as a stabilizer, because a natural negative feedback allows the plasma discharge ignition, and when the impedance charge descends, then the voltage is automatically adjusted to sustain a stable plasma discharge, and consequently an electrical field of about 3 kV/mm is applied in air between electrodes, as a result, a low-current discharge streaks between the closest points of the electrodes, preionizes the gas gap, provoking the formation of a stronger current to sustain the warm plasma discharge at atmospheric pressure, using nitrogen or the GHG as plasma gas; the gliding discharge on the central electrode is reflected in the electrical signals [13]. The gliding period (T_g) can be determined from the electrical signals, in this case, a T_g of 25 ms is obtained, so the gliding frequency corresponds to 40 Hz. The arc column length is a function of power applied to the discharge, reactor geometry, and nature of the gas to be treated.

2.1.- Energy Consumption Analysis

By using the same procedure indicated in [17], based in the instantaneous values intensity of voltage and current waveforms; the root medium square power P_{RMS} can be deduced, and finally the energy consumption during operational experiences can be obtained. The operational condition in the power supply was 450W of power and a frequency of 140 kHz, and flow rates were 2LPM for CH_4 and 1LPM for CO_2 . According these operating conditions, the specific energy (SE) required generating CO and H_2 (syngas) can be calculated with the following expression [14, 17]:

$$SE = \frac{P}{[CO+H_2]_{produced}} \quad (7)$$

where, P refers to the input power of plasma in kW (kJ/s); $[CO + H_2]_{produced}$ refers to moles of (CO + H_2) produced per second (mol/s). Equation (7) is referred as the energy consumption needed to produce a mole of CO and H_2 (a syngas mole). This calculation includes the energy input as electrical power from the plasma discharge. The SE is optimal when the energy consumption of the reforming process is lower. According (7) a value of 310.79 kJ/mol of syngas is obtained; not so far from the thermodynamically value (2).

In addition, to determine the energy conversion efficiency (ECE) applied to the gas mixture entering to the plasma reactor; the expression (8) is used:

$$ECE = \frac{n_{H_2} * LHV_{H_2} + n_{CO} * LHV_{CO}}{P + n_{CH_4} * LHV_{CH_4}} \quad (8)$$

From [18] and [19], next values can be obtained: 1) $LHV_{H_2} = 242.056 \text{ kJ.mol}^{-1}$; 2) $LHV_{CO} = 283.179 \text{ kJ.mol}^{-1}$; and 3) $LHV_{CH_4} = 802.933 \text{ kJ.mol}^{-1}$ @ 298.16 K. By making the pertinent calculations, the ECE obtained is 22.05%. It's worthy to note, that the maximal ECE for these operations conditions will be 34%, therefore the warm plasma reactor is a profitable alternative method to confirm the destruction efficiency.

In [14], a comparison for ECE and SE in syngas production is reported for different plasma configurations. Taking this into consideration, the SE, and ECE results obtained in this paper for syngas generation, places the Warm Plasma discharge as one of the most efficient methods for the syngas production.

The conversion percentage for CH_4 and CO_2 can be calculated by

$$\text{Conversion percentage} = \frac{X_0 - X}{X_0} \times 100\% \quad (9)$$

where X_0 is their initial concentration and X is the final concentration.

A chemical mechanism of hydrogen and carbon monoxide generation was studied in [20]. In general, the H_2 production can be explained by the interaction of CH_4 with O^* and H^* radicals. CO formation could be mostly explained by the collision of CO_2 with electrons and a three-body reaction with N_2 ; consequently, the selectivity coefficient can be obtained with the subsequent equations [20]:

$$S_{CO} = \left\{ \frac{[CO_{produced}]}{[CO_2_{consumed}]} \right\} \times 100\% \quad (10)$$

$$S_{H_2} = \left\{ \frac{[H_2_{produced}]}{[CH_4_{consumed}]} \right\} \times 100\% \quad (11)$$



Based on the operating conditions already described, and using (9) - (11), the CO and H₂ selectivity respectively was 63% and 99.44%. The conversion capacity of CO₂ and CH₄ correspondingly was 29.41% and 98.44%.

3. Results and discussion

For the case of Partial Oxidation the CH₄ and Air mixture has a flow rate of 2:1 LPM respectively. This mixture is introduced into the bottom reactor and immediately at the reactor exit, a gas analyzer is connected (Gas Analyzer Board®) to measure real-time concentration of up to 6 different gases: CO₂, CH₄, CO, O₂, C₂H₆ and H₂.

The plasma is applied (Zone 1, in Fig 2); the power source supplies a sinusoidal voltage at a frequency of 140 kHz and a power of 250W. Fig. 2, shows the results obtained from the reforming gas analyzer expressed in % volume versus time, two zones are clearly revealed: Zone 1 presents conditions under plasma action, and zone 2 represents the period of time when the plasma discharge is turned off, a change of gas concentration is rapidly exposed; the gas mixture (GHG) increase to the initial conditions, whereas the syngas concentration descends to the lower level.

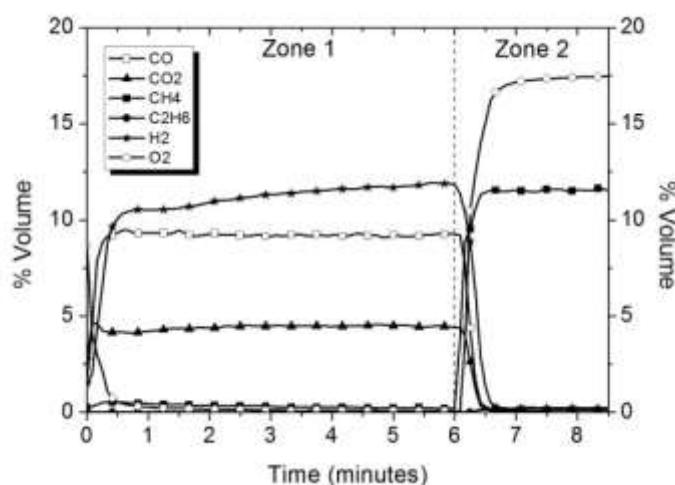


Fig. 2.- GHG and Syngas Concentration Evolution (% Vol).

The experimental results are shown in Table I indicating the initial and resultant concentration in % volume for the GHG and syngas components.

Table I.- GHG and Syngas compounds after and before plasma treatment

Compound	Before treatment (%vol)	After treatment (%vol)
Oxygen	17.5	3.4
Nitrogen	71	71
Methane	11.5	0.2
Carbon monoxide	0	9.2
Carbon dioxide	0	4.4
Hydrogen	0	11.8

Once the plasma discharge is ON, instantly an increase in H₂ concentration around 12% volume occurs, at the same time, the carbon monoxide reach 9.2% and carbon dioxide touch 4.4%; getting, according (9), a conversion of CH₄ of 98.6%. These values were also confirmed by Gas Chromatography (TRACE GC ULTRA®). In Fig. 3a), three significant peaks were previously identified with sample gas standards to confirm the presence of O₂ (first peak at 2.8m), N₂ (at 3.5m) and CH₄ (at 4.3m) of time retention. In the case of output components (Fig 3b) the CG results shows the presence of H₂ at 2.4m, O₂ at 2.8m, N₂ at 3.5m, and CO at 5.0m, the occurrence of CH₄ no longer arises.



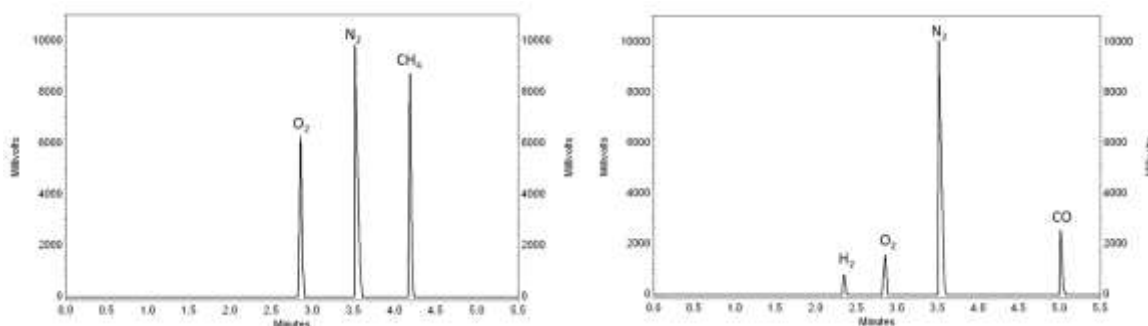


Fig. 3.- Chromatogram of the sample before (a) and after (b) warm plasma treatment.

Figure 4, displays a bar graph derived from the GC analysis and indicates the concentration of the mixture before and after treatment; the black bars indicate the mixture composition before treatment (O₂, N₂ and CH₄). The gray bars correspond to the exhaust gas components which are CO₂, H₂, O₂, N₂ and CO. In the case of N₂ the peak intensity is fairly similar before and after treatment, this confirms that it functions like a plasma gas and does not react with other compounds; hence any NO_x formation is produced during GHG reforming. It is noteworthy that methane concentration during the reformation process, descends to 0%, therefore a high CH₄ conversion and a high production of hydrogen is obtained. Additionally, a second reliable result is related with nitrogen behavior, it's clearly shown that it does not react with other gas, because its intensity level is quite the same before and after treatment.

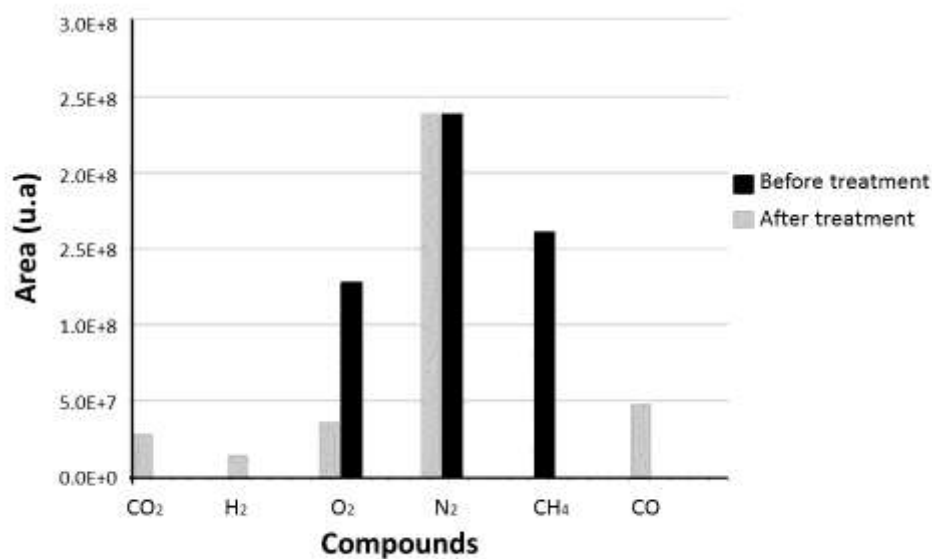


Fig. 4.- Concentration of input and output gas in warm plasma reactor



4. Summary and perspectives

Warm plasma fuel reforming technology has several advantages over traditional catalytic or thermal processes including fast start time, high productivity with a relatively low electrical energy costs operation.

The results show that warm plasma reactor is capable of reforming heavy hydrocarbon fuels with high conversion efficiency and is an important piece of technology for on-board vehicular reforming systems that should be further developed and optimized.

In plasma reforming process, considering electricity coming from methane combustion (combustion value of 890 kJ/mol), only 42% of combustion energy is converted to electricity, and only 67% of electric energy is utilized by the reforming reaction, therefore, to get 1 mol CO, it would burnt 0.5 mol of methane and 0.5 mol CO₂ released in the power plant. However, these CO₂ would be balanced by that removed during the reforming process.

In one word, to obtain 1 mol CO in CH₄ + CO₂ reforming by plasma process, it expends 1 mol of methane without CO₂ exhaust; in steam reforming of CH₄, it expends 1.33 mol of methane and 0.33 mol of CO₂ released.

The growing demand for chemicals derived from syngas has led to the development of several technologies (e.g. Fischer-Tropsch synthesis, NH₃ or oxygenate production). These processes are of strategic importance for any country.

In this paper, a Resonant Electronic Converter has been developed and applied assuring the energy consumption with high transfer efficiency.

The system presented here is able to provide enough energy required to break GHG gas chemical bonds, under atmospheric pressure without using supplementary inert gases.

Electric analysis was performed to determine the current, voltage, power and energy required for the GHG treatment.

Reforming conversion of GHG by warm plasmas offers a promising route to generate higher value products such as syngas at low energetic cost.

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