THE SIMULTANEOUS SYNTHESIS OF HYDROGEN-RICH GAS AND OXIDATION OF FINE METAL PARTICLES IN WATER VAPOUR PLASMA

V. Valinčius, V. Grigaitienė, P. Valatkevičius, A. Tamošiūnas

Lithuanian Energy Institute, Plasma Processing Laboratory, Kaunas, Lithuania

E-mail: vitas@mail.lei.lt

Atmospheric pressure water vapor plasma technology was developed, tested and implemented for the experimental research of fuel conversion processes. The present study offers a methodology for production of hydrogen-rich gas and additional deposition of small metal oxide dispersed particles by the employment of non-equilibrium water vapour arc plasma at atmospheric pressure. The results of the present study enables constructing a specific device which allows a highly efficient production of the synthetic gas containing an increased amount of hydrogen and its use in the production of second generation fuels. The injection of copper dispersed particles causes the oxidation and removal of dissociated oxygen and simultaneous synthesis of fine particles. The relation between the yield of hydrogen in an exhaust gaseous product, the arc current, particle material, and water vapor flow rate in the plasma torch was determined.

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INTRODUCTION

The demand for knowledge on the fuel conversion and production of hydrogen as an alternative energy source has significantly increased in the recent decades. Many researchers emphasized the role of plasma ambient in the possible production of hydrogen-rich gas [1-3, etc.]. It was reported that the inevitable employment of plasma processes enables improving the traditional technology of industrial hydrogen production or creating novel high efficient technology [1, 4]. It is well known that thermal water vapor plasma is an unique ambient and may affect the production of syngas and formation of fine particles, fibers or granules. This process may also occur during plasma spraying and deposition of coatings for wide range of applications. Therefore such technology is very promising and is expected to be applied in the novel approach of hydrogen rich gas production.

The authors of the present paper and the author of [5] assume that the water vapor plasma devices are important tools for technological application and that the essential approach to find a new technical solution for production of hydrogen-rich gas: the development of a novel water vapor plasma technology is probably the most advanced and realistic. So, the present research is devoted to the development and study of a novel water vapor plasma source for fuel conversion and wide range of other applications.

The present study offers a possibility for production of hydrogen-rich gas and additional deposition of small metal oxide dispersed particles by the employment of non-equilibrium water vapor arc plasma at atmospheric pressure.

1. EXPERIMENTAL EQUIPMENT

A linear atmospheric pressure DC electric arc plasma source was developed, manufactured and employed for realization of fuel conversion process (Fig. 1.). The novel water vapor plasma generator (PG) is based on a linear scheme with hot button type tungsten cathode, step-formed stainless steel anode and neutral electrode as described in [6].

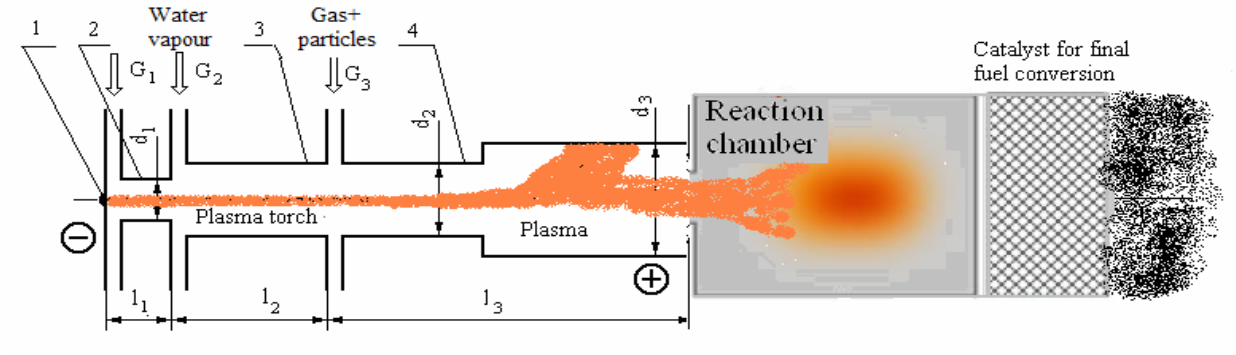

Fig. 1. The schematic presentation of water vapor plasma device for conversion of fuel and production of hydrogen rich gas. 1 – cathode junction; 2 – ignition section; 3 – neutrode; 4 – anode. G_1 is injection of initial ignition gas (argon), G_2 – injection of over-heated water vapor; G_3 – injection of additional gas mixed with Cu dispersed particles

The over-heated water vapor generated in specific equipment was used as plasma forming gas at the arc current of 250 A. The main operational characteristics of the plasma torch were established and are as follows: electric arc current 130…210 A, arc voltage 230…330 V. Power of the plasma source varied in the range of 40…69 kW, water flow rate for the cooling of the torch was $0.1 \cdot 10^{-3}$ kg·s^{-1}, argon (cathode shield) gas feeding $5.2 \cdot 10^{-4}$, water vapor feeding $(1.48…4.48) \cdot 10^{-3}$ kg s^{-1}. The mean temperature of plasma jet was in the range of 2300…3000 K, the mean velocity of plasma jet at the torch exhaust nozzle was 200…600 m·s^{-1}.

By using a properly designed plasma stream reactor connected to the PG, heated by energy of electric discharge and keeping the fluid at 3000 K, it is possible to produce dissociated water vapor plasma, avoid the reverse fusion and extract hydrogen or hydrogen-rich gas. Propane-butane gas and dispersed particles of copper were used as additional oxygen and ozone acceptors affected the specific conditions to reduce amount of oxygen molecules in the fluid gas. Substantial amounts of fluid gas could be converted into hydrogen and carbon monoxide with production of additional amounts of CuO or Cu_2O which could be used for further wide range applications. Voltage-current characteristics (VCC) of the PG were determined from the heat conservation calculations while measuring current strength in the circuit, voltage drop, and gas amounts. The generalized electric and thermal characteristics of the PG permit the determination of the operating regime needed for plasma technologies and selection of optimal operating modes.

The emission spectra of exhaust Ar/water vapor plasma jet at the exit nozzle of the PG was measured for the determination of elemental composition. This has been done by means of AOS4-1 spectrometer. The schematic presentation of the measurement set-up is shown in Fig. 2.

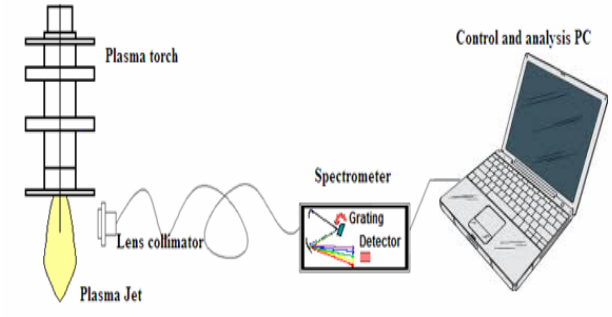

Fig. 2. A scheme of experimental set-up for optical emission spectroscopy

2. RESULTS AND DISCUSSION

Two regimes of PT were used for experimental investigation. They differed only in respect of arc current and water vapor flow rate. Overheated water vapor was injected in the anode side and was dissociated and ionisated in the arc channel. A large amount of ozone, which occurred as a very strong oxidizer, formed over the cooled anode surface. So, during the operation of the system, a strong oxidation of pure copper powder was observed and the outflowing plasma jet assumed a green color. To avoid the oxidation of the former copper anode, the stainless steel was applied instead of copper. Thus, the influence of current, flow rate and discharge channel diameter on the VCC for such vortex PG with both copper and stainless steel anode was necessary to know.

After the analysis of the primary experimental results, VCC has been generalized on the basis of the principles of dynamic similarity theory [7].

The measurements suggests that VCC strongly depend on several factors as showed in equation (1) for stainless steel anode.

$$\frac{Ud_2}{I} = 2{,}9 \cdot 10^4 \left(\frac{I^2}{Gd_2}\right)^{-0.7}.$$

The relationships between the operating characteristics employing dimensionless equations describe physical phenomena in the PG.

The investigation of chemical processes during the water vapor decomposition and conversion was performed using analytical equipment with optical spectrometer and gas chromatograph in a specific experimental setup (see Fig. 2). It was found that copper particles effectively removes the oxygen and improves the output of hydrogen in the gasification products. The hydrogen conversion degree reaches up to 75%.

As a conclusion it can be stated that the employment of results of the present study enables constructing a specific device which allows a highly efficient production of synthetic gas containing an increased amount of hydrogen and its use in the production of second generation fuels. The injection of carbon or copper dispersed particles is considered for a better removal of dissociated oxygen.

The optical emission spectra were measured for the argon/water vapor plasma jet in the range of wavelength from 300 to 800 nm (Figs. 3 and 4) at the 7 mm distance from the exhaust nozzle of the PG. The experiment conditions in the emission spectra measuring (see Fig. 3) were the following: the power of plasma torch P=55.2 kW, the electrode of stainless steel was used, argon flow rate $5.2 \cdot 10^{-4}$ kg·s^{-1} and water vapor flow rate $3.51 \cdot 10^{-3}$ kg·s^{-1}. In another case (see Fig. 4), the power of plasma torch P=69.1 kW, argon flow rate $5.2 \cdot 10^{-4}$ and water vapor flow rate $4.48 \cdot 10^{-3}$ kg·s^{-1}.

The main species observed in Ar/water vapor plasma were: Ar (I), Ar (II) OH, H, O (I), Cu (I), Cu (II). The emission spectra showed the peaks of hydrogen atoms: H_α (656.2 nm), H_β (486.1 nm) and H_γ (434.1 nm), which belong to Balmer series of H_2.

The high instability ratios of the plasma flow were observed. Therefore, the performed emission spectra measurements confirms that water vapor was decomposed into H, O and OH radicals by high voltage DC electric arc.

Visually, the H_α line tends not to be seen well due to the lack of sensitivity of the human eye in the deep red part of the spectrum, but the H_β line tends to come fairly intense. The emission intensities of the obtained peaks are quite low, because of low energies of exited hydrogen atoms at the atmospheric pressure conditions. Consequently, the temperature of plasma jet at the exhaust nozzle of the plasma torch is high due to the

intense collisions between molecules, but the energies of the exited species are low, because the mean free path is short. The experiments on the observation of plasma jet behaviour showed that the synthesis of hydrogen is occurring mainly in the central region of plasma column inside the anode chamber. This hypothesis was tested by comparing the emission spectra of both PG regimes using the stainless steel anode.

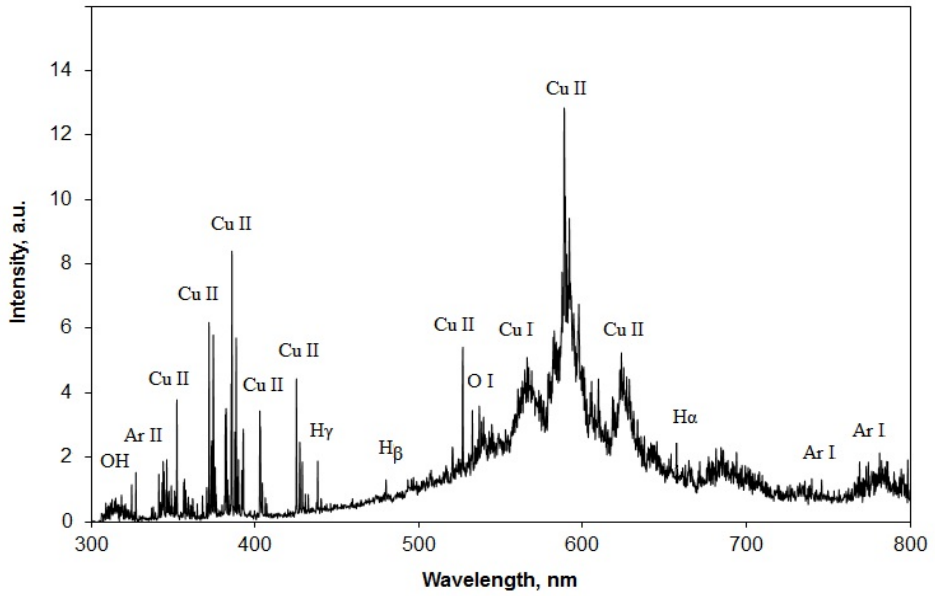

Fig. 3. Optical emission spectra of Ar (as shield gas)/water vapor plasma at P = 69.1 kW, water vapor flow rate $4.48 \cdot 10^{-3}$ kg·s^{-1}

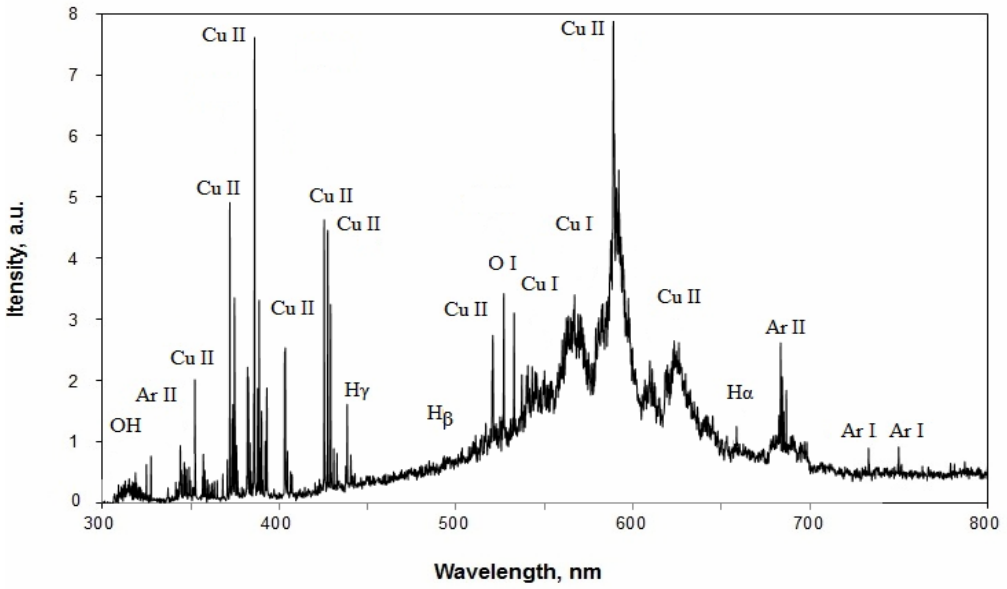

Fig. 4. Optical emission spectra of Ar (as shield gas)/water vapor plasma at P = 55.2 kW, water vapor flow rate $3.51 \cdot 10^{-3}$ kg·s^{-1}

It was also observed that the intensity of copper (Cu) was much higher compared to the rest species detected. It is considered that copper emits Cu radicals in the plasma due to the oxidation of the copper, occurring in the discharge chamber where the electric arc spot burns. The presence of reactive atomic oxygen O(I), which forms during the ionization of water vapor, also amplifies the oxidation process. This could be explained by the fact that oxygen molecule is much heavier compared to hydrogen and tries to settle down to the periphery of the particle, where it interacts with the inner surface forming the copper oxide (CuO). Hard metallic material, emitted from the outflow nozzle of the PG was collected and analyzed by X-Ray scattering technique The main components that were found are the following: copper (Cu), cuprite (Cu_2O) and gray-to-black metallic mineral – tenorite (CuO). Naturally, tenorite occurs in addition to the hydrothermal oxidized environment, usually as a sublimation product on volcanic lavas. Therefore, the performed spectral measurements confirmed that water vapor was decomposed into H, O and OH radicals by DC electric arc. Beside these important groups of spectra lines, the

spectrum contains a large number of other lines with variable intensities, making the plasma emission spectrum rather complicated.

For further, more effective extraction of moecular hydrogen without reversible recombination to water vapor could be performed using an original catalytic converter (see Fig. 1) created on the basis of the catalytically active CuO powder deposited during the described above process.

CONCLUSIONS

The performed experimental investigation demonstrated that water vapor plasma is a potential raw material for the hydrogen-rich gas production. The regularities of the PG operating regimes influence the process of hydrogen synthesis and affect the yield of the product.

The injection of carbon or copper dispersed particles is considered for a better removal of dissociated oxygen and simultaneous synthesis of fine particles for wide range of applications.

The copper powder enables increasing the amount of hydrogen in the exhaust gas. The synthesis of hydrogen is occurring mainly in the central region of plasma column inside the anode chamber.

ACKNOWLEDGEMENTS

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ОДНОВРЕМЕННЫЙ СИНТЕЗ ОБОГАЩЕННОГО ВОДОРОДОМ ГАЗА И ОКИСЛЕНИЕ МЕЛКИХ ЧАСТИЦ МЕТАЛЛА В ПЛАЗМЕ ВОДЯНОГО ПАРА

В. Валинчюс, В. Григайтене, П. Валаткявичюс, А. Тамошюнас

Разработана, испытана и внедрена плазменная технология атмосферного давления водяного пара для экспериментального исследования процессов преобразования топлив. Данное исследование представляет метод для производства обогащенного водородом синтез-газа, а также одновременное осаждение дисперсных частиц окиси металла при использовании атмосферного давления неравновесной электродуговой плазмы водяного пара. Результаты исследования позволяют создать установку для высокоэффективного производства синтетического газа, содержащего повышенное количество водорода, для его использования в производстве второго поколения топлив. Исследования показали, что введение в плазменный поток дисперсных частиц меди вызывает их окисление и удаление диссоциированного кислорода. Установлено, что существует связь между выходом водорода в выхлопных газах, силой тока дуги, материалом частиц и паров, расходом потока через плазмотрон.

ОДНОЧАСНИЙ СИНТЕЗ ЗБАГАЧЕНОГО ВОДНЕМ ГАЗУ ТА ОКИСЛЕННЯ ДРІБНИХ ЧАСТИНОК МЕТАЛУ В ПЛАЗМІ ВОДЯНОЇ ПАРИ

В. Валінчюс, В. Грігайтене, П. Валаткявічюс, А. Тамошюнас

Розроблена, випробувана і впроваджена плазмова технологія атмосферного тиску водяної пари для експериментального дослідження процесів перетворення палива. Дане дослідження представляє метод для виробництва збагаченого воднем синтез-газу, а також одночасне осадження дисперсних частинок окису металу при використанні атмосферного тиску нерівноважної електродугової плазми водяної пари. Результати дослідження дозволяють створити обладнання для високоефективного виробництва синтетичного газу, що містить підвищену кількість водню, для використання його у виробництві другого покоління палив. Дослідження показали, що введення в плазмовий потік дисперсних частинок міді викликає їх окислення та видалення дисоційованого кисню. Встановлено, що існує зв'язок між виходом водню у вихлопних газах, силою струму дуги, матеріалом частинок і парів, витратами потоку через плазмотрон.