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ANDRIUS TAMOŠIŪNAS

Investigation of Thermo-Hydro-Dynamic Processes in Water Steam Plasma and Its Application for Organic Waste Treatment

Summary of Doctoral Dissertation
Technological Sciences, Energetics and Power Engineering (06T)

2014, Kaunas

KAUNAS UNIVERSITY OF TECHNOLOGY

LITHUANIAN ENERGY INSTITUTE

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KAUNO TECHNOLOGIJOS UNIVERSITETAS

LIETUVOS ENERGETIKOS INSTITUTAS

Andrius Tamošiūnas

TERMOHIDRODINAMINIŲ PROCESŲ TYRIMAS VANDENS GARO PLAZMOJE IR JOS TAIKYMAS ORGANINIŲ MEDŽIAGŲ KONVERSIJAI

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Introduction

Due to limited fossil fuel reserves, crude-oil price variations and international regulations on emissions, the use of new and renewable energy sources has received significant attention. Since the beginning of the last decade of the 20th century, plasma technology has received a significant attention in many scientific and industrial areas such as metallurgy, materials processing, space, chemical and environmental etc. There is no field of human activity where this technology could not be applied. Therefore, one of the most promising technologies, which satisfy the problems mentioned above, could be plasma technology used for waste treatment.

Recently, scientists have noticed that the application of plasma for waste treatment could be improved and successfully implemented in energy, environmental and transport applications as well as in chemistry etc., by using water steam as a plasma-forming gas. Water steam plasma provides an alternative to the conventional neutralization and conversion methods because treated materials are not directly burned but reduced in the anaerobic environment.

The thermal water steam arc plasma is characterized by a high degree of neutralization (99.99%), very high temperatures exceeding up to 3500 K, high energy density, much smaller size, fast startup-shutdown of the equipment, ability to control the process, cheaper installation and is more environmental-friendly compared to the conventional neutralization methods. Therefore, the use of water steam arc plasma could help to solve several problems such as landfill, reduce the ground and air pollution, and recycle any type of organic/inorganic waste allowing to get synthesis gas as a secondary product of the process. Synthesis gas could be further used as a feedstock in energy/heat or chemicals production. This method has a high potential and perspectives to be used as a zero-waste technology.

There are many reference works available in the scientific-technical literature and conference works considering the use of plasma technology for materials processing, fuel conversion and waste treatment etc. On the contrary, there is lack of information published on the conditions of formation of water steam plasma, its diagnostics, and application for the effective waste treatment process, including energy expenditure. Additionally, there is also a gap in a heat and mass transfer research inside the discharge chamber of the plasma torches and plasma-chemical reactors. Therefore, the importance of the investigations in this study is obvious.

The aim of the Doctoral Dissertation

The main goal of the thesis was to design water steam plasma torch for organic waste treatment and investigate thermo-hydro-dynamic processes inside its arc discharge chamber and generated plasma stream.

Tasks of the Doctoral Dissertation

In order to achieve the objective the following tasks should be solved:

1. Design an experimental water steam arc plasma torch used for organic waste treatment. Explore its basic electrical, thermal and operational parameters.
2. Perform the diagnostics of the steam plasma stream at the nozzle exit of the plasma torch.
3. Experimentally and numerically investigate the formation and distribution of concentration of active radicals in the steam plasma depending on its temperature.
4. Experimentally determine the formation of reaction products after the conversion of organic materials in the ambient of water steam plasma. Compare the results with simulated ones.

Scientific novelty

In the thesis, the interaction between the steam, electric arc and electrodes of the plasma torch during the generation process of a low-temperature thermal plasma stream at atmospheric pressure was investigated; the mechanism of heat transfer in the arc discharge channel of the plasma torch was examined; the efficient organic waste treatment method using thermal plasma technology was implemented.

Practical value

The results obtained in the thesis are useful to design water steam plasma torches operating at atmospheric pressure with completely different unique physical properties as water steam is involved to form plasma. Furthermore, the results are of significant value regarding the engineering of plasma-chemical reactors for the organic waste treatment as well as for determining the main parameters of the treatment process, including calculations on process efficiency and energy expenditure.

Statements carried out for defense

1. By changing the parameters of the steam plasma torch, the mean outflow temperature and velocity of the plasma stream increased up to 3300 K and 600 m/s, respectively.

2. The most intensive heat flux exists in the anode, where heat transfer is determined by convection, conduction and radiation. The flow is turbulised in this part of the discharge chamber of the plasma torch.
3. As the temperature of the steam plasma increases up to 3400 K, the concentration of active radicals such as H_2, H, O_2, O and OH, increases gradually.
4. The main reaction products formed after the conversion of organic materials are H_2 and CO gas.
5. As the content of steam in the total steam/treated material ratio increased, the yield of synthesis gas and energy conversion efficiency increased whereas the specific energy requirements decreased.

The structure and the content of the dissertation

The dissertation consists of the following major parts: introduction, literature review, results and discussion, conclusions, references and list of scientific publications. The total size of thesis is 122 pages containing 79 figures, 9 tables and 148 references.

1. LITERATURE REVIEW

This chapter provides an overview of the literature review emphasizing the information on the other author's achievements in the research of electric discharge and plasma formation including their properties, thermo-hydro-dynamical processes occurring in the arc discharge chamber of the plasma torch and the use of water steam plasma for organic waste treatment.

2. EXPERIMENTAL SETUP AND METHODOLOGY

The experimental DC water steam plasma torches designed and used in this study are shown in Fig. 1. The linear plasma torch is made of a 'hot' tungsten-rod cathode, isolating rings for the tangential supply of shielding and plasma-forming gas, and a stair-shaped or confusor-type anode. The electrodes of the plasma torch are made of copper with some parts of steel to prevent the condensation of steam inside the discharge chamber. Argon with flow rate of $0.52 \cdot 10^{-3}$ kg/s is used as a shielding gas to protect a tungsten-rod cathode from erosion, whereas overheated steam to 510 K, with flow rate in the range of $(2.63 - 4.48) \cdot 10^{-3}$ kg/s, as the plasma-forming gas. The electric arc which is ignited in the plasma torch heats up and ionizes the gas tangentially flowing through it.

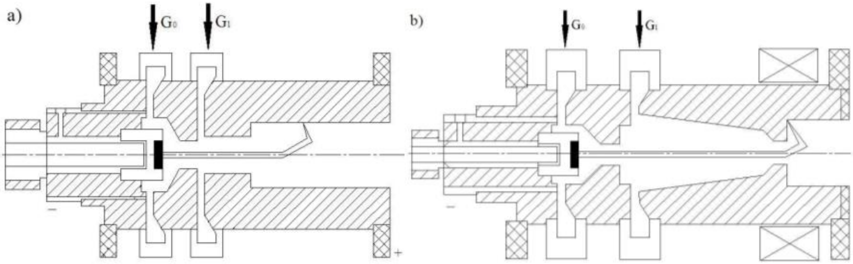

Fig. 1. The schematic of water steam plasma torches, (a) with cylindrical stair-shape anode, (b) with confusor-type anode. G_0, G_1 are shielding and plasma-forming gas, respectively.

For the realization of raised tasks the experiments were performed with several types of plasma-chemical reactors. The plasma-chemical reactor (PCHR) configurations designed for various phase organic waste treatment are shown in Fig. 2. 351 cm^3 volume plasma-chemical reactor with inner diameter of $40 \cdot 10^{-3}$ m and length of 0.28 m was used for gaseous materials conversion. The residence time of treated materials varied from 6 to 8 ms. The conversion of liquid and solid phase materials was performed using a bigger scale PCHR with volume of 0.15 m^3, inner diameter of 0.25 m and length of 1 m. The residence time of treated materials varied from 0.5 to 1 s.

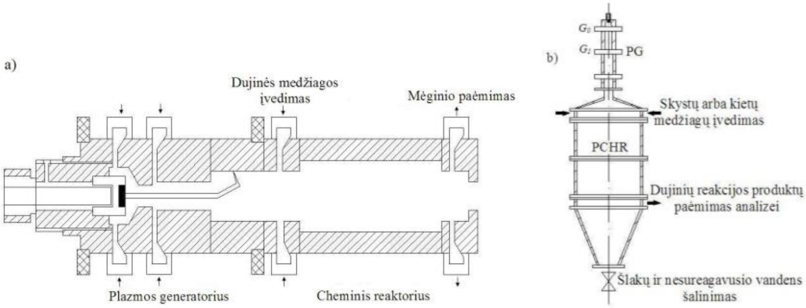

Fig. 2. The schematic of plasma-chemical reactors, (a) for conversion of gaseous organic materials, (b) for conversion of liquid and solid organic materials

The operational parameters and generalized dimensionless regime characteristics of the plasma torch were determined by measuring the current and voltage of the arc, the flow rates of plasma-forming gas, and the flow rate of cooling agent of the plasma torch with simultaneous measurements of its temperature difference. The mean temperature and velocity of plasma stream as well as a heat transfer between the electric arc, heated gas and the walls of the discharge chamber of the plasma torch were calculated from all the measured data using the heat balance equations [1].

The diagnostics of the outflow plasma at the nozzle exit of the plasma torch was carried out using enthalpy probe and optical emission spectroscopy methods [2, 3]. The intrusive enthalpy probe technique was used to measure the local temperature and the velocity distribution lengthwise and crosswise the outflow plasma stream, while the non-intrusive optical emission spectroscopy method was used to determine the chemical composition of steam plasma. Additionally, it helped to identify either the plasma was in a local thermodynamic equilibrium or not.

The plasma-chemical processes occurring in the plasma-chemical reactor were simulated by means of the Chemical Workbench code [4] using a PH-type thermodynamic equilibrium reactor (TER) model. Furthermore, it helped to determine the formation and distribution of concentration of active radicals depending on the plasma temperature.

A few different state organic materials were chosen for the conversion process. Propane, glycerol (99.5% purity) and wood (<8% of moisture) were used as a feedstock, due to the known chemical composition, simple handling and feeding, and non-toxicity.

The thermal plasma reforming system of organic materials was quantified in terms of conversion efficiency, product gas composition and yield, and quality of produced synthesis gas as well as the energy efficiency and specific energy requirements.

3. RESULTS AND DISCUSSION

3.1. Electrical and thermal characteristics of the steam plasma torch

In order to realize the effective plasma conversion process of organic materials, a reliable operation of the water steam plasma torch is required. Reliability is determined by a long and stable plasma torch operation with constant and recurring performance characteristics in a wide arc current and heated steam flow rate ranges. Moreover, the plasma parameters, such as temperature, enthalpy and velocity, directly depend on the stable operation of the plasma torch. Therefore, the operational parameters of the steam plasma torch and generated plasma are shown in Table 1.

Table 1. The operational parameters of the steam plasma torch and generated plasma

Parameter	**Value**
Plasma torch power, kW	34 – 69
Arc current, A	120 – 250
Arc voltage, V	230 – 400
Steam flow rate, 10^{-3} kg/s	2.63 – 4.48
Argon flow rate, 10^{-3} kg/s	0.52
Cooling agent flow rate, 10^{-3} kg/s	130
Plasma torch thermal efficiency, %	53 – 76
Mean temperature of steam plasma, K	2400 – 3300
Mean velocity of steam plasma, m/s	300 – 600
Reynolds number	4000 – 6000

One of the most important electrical characteristics of the plasma torch are volt-ampere characteristics (VACh), which help to choose the parameters of power source and determine the electrical efficiency of the plasma torch. Therefore, the VACh of the steam plasma torch were determined in a wide range of arc current, arc voltage and steam flow rate. The VACh are shown in Fig. 3.

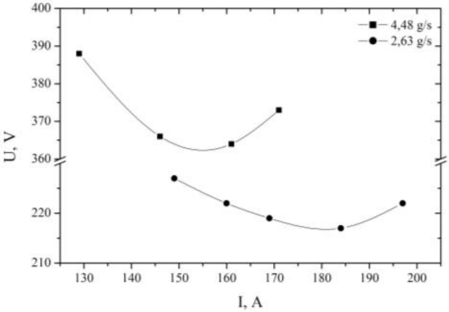

Fig. 3. Volt-ampere characteristics of the steam plasma torch

The VACh in the arc discharge chamber of the plasma torch is U-shaped. The dropping part of the curve determines a lower electrical efficiency of the plasma torch system. Thus, the inclusion of the ballast resistance in the electrical circuit is required. The rising part of the curve ensures more stable arcing in the arc discharge chamber of the plasma torch simultaneously suppressing the large-scale shunting of the arc. Such VACh curve is typical for the steam plasma torches [1, 5].

The existing analytical methods are not capable to describe the arcing processes occurring in the discharge chamber of the plasma torch. Therefore, for the generalization of the VACh and thermal efficiency of the steam plasma torch, the similarity theory was successfully applied. Knowing the plasma torch geometric dimensions (d_2), the arc current (I) and voltage (U), and heated steam flow rate (G), and the mean length of the arc (L), the VACh and integral coefficient of thermal efficiency of the plasma torch were described in equations (1) and (2), respectively.

$$\frac{Ud_2}{I} = 34\left(\frac{I^2}{Gd_2}\right)^{-0.7} \times \left(\frac{G}{d_2}\right)^{0.23} \times \left(pd_2\right)^{0.48}, \qquad (1)$$

$$\tilde{\eta} = \frac{(1-\eta)}{\eta} = 2{,}22 \cdot 10^{-2}\left(\frac{I^2}{Gd_2}\right)^{0.22} \times \left(\frac{G}{d_2}\right)^{-0.38} \times \left(\frac{L}{d_2}\right)^{-0.8}. \qquad (2)$$

The experimental VACh and integral coefficient of thermal efficiency of the steam plasma torch results were compared with theoretical ones when arc burns in the ambient of steam [6]. Figures 4 and 5 show the comparison between the experimental and theoretical results.

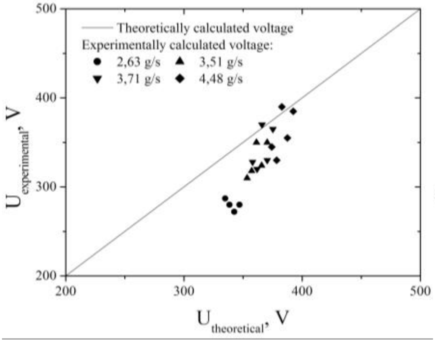

Fig. 4. Comparison between experimental and theoretical VACh results

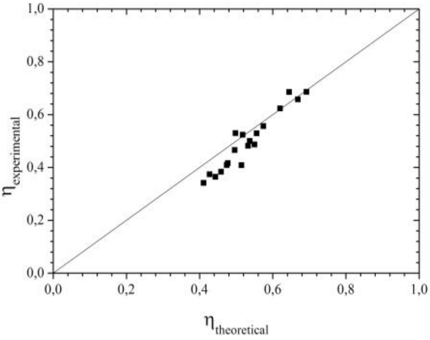

Fig. 5. Comparison between experimental and theoretical integral coefficient of thermal efficiency results

As the flow rate of steam decreased from $4.48 \cdot 10^{-3}$ kg/s to $2.63 \cdot 10^{-3}$ kg/s, the experimentally measured arc voltage increasingly differ from the theoretically calculated. The highest difference was 18%. It could be explained due to the existence of Ar. As the flow rate of steam decreased, the content of argon in the total mass flow rate of plasma-forming gas increased from 10 to 17%. This phenomenon was also reported in [1], when argon content in the total mass flow rate increased to 25%, the voltage decreased in 1/3. The difference between experimentally and theoretically calculated integral coefficients of thermal efficiency was less than 7%.

The knowledge of the main energy and thermal characteristics of the water steam plasma torch enables to select and adjust the required plasma torch operating parameters in organic materials treatment to synthesis gas.

3.2. Heat transfer into the elements of the steam plasma torch

Heat transfer inside the arc discharge chamber of the steam plasma torch between the electric arc, heated gas and the walls of the discharge chamber is very complicated. Due to this, a set of simplifications and presumptions are taken. It is convenient to distinguish three distinct zones, where heat transfer is characterized by separate flow conditions. The initial zone, which length is determined by the contact area of the thermal layer of the arc and the turbulent gas boundary layer developed on the discharge channel wall, the arc burns in the laminar flow. In the transition zone, the arc starts to interact with the turbulent gas flow, and this section is characterized by gradual transition to the developed turbulent flow. Finally, a steady turbulent flow is found. According to this assumption, the heat flux and the heat transfer coefficient were defined experimentally. The results are presented in Figures 6 and 7.

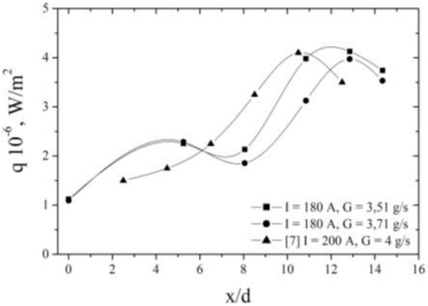

Fig. 6. Distribution of heat flux into the walls of the discharge chamber of the steam plasma torch

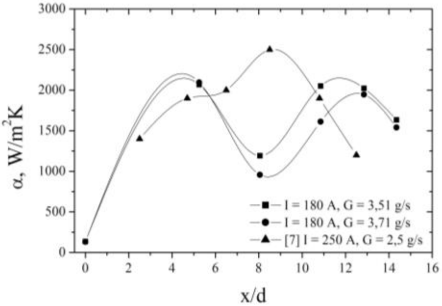

Fig. 7. Distribution of heat transfer coefficient inside the discharge chamber of the steam plasma torch

The experimental results were also compared with reference [7], which shows a good agreement between the magnitudes of calculated values. The curves differ slightly because the distance *x/d*, which depends on the geometrical parameters of the plasma torch, was not the same in both cases. At the distance *x/d* changes from 0 to 8, the initial zone could be observed. Further, the change in distance *x/d* from 8 to 11, matches the characteristics of the transition zone, where the arc starts to interact with the turbulent gas flow, and the heat flux as well as the heat transfer coefficient increases significantly. The highest heat flux and heat transfer coefficient were obtained at the distance of $11 < x/d < 15$, corresponding to the developed turbulent flow zone. A local increase of q and α at the distance of $x/d = 5$ could be explained due to the steam injection. The physical properties of argon such as thermal and electrical conductivity, enthalpy, etc., determined a lower α at the distance of $x/d < 5$.

Total heat losses into the walls of the discharge chamber of the steam plasma torch were generalized according to the equation (3) and are shown in Fig. 8.

$$St = \frac{d}{4l} \cdot \left(\frac{1-\eta}{\eta} \right) = 1.36 \cdot \mathrm{Re}_d^{\,-0.8}. \qquad (3)$$

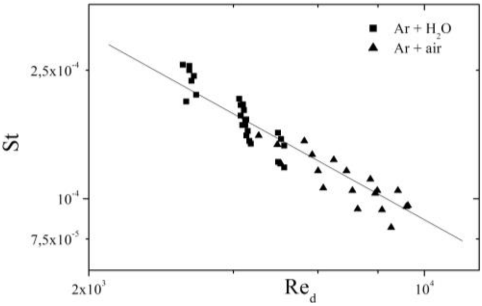

Fig. 8. Total heat losses into the walls of the discharge chamber of the plasma torch

The results were compared with the same plasma torch using air as plasma-forming gas. According to the obtained equation (3), the flow in the entire discharge chamber of the plasma torch was transitional.

Heat transfer into the distinct parts of the plasma torch, such as the cathode, neutral section and anode, were defined by different heat transfer phenomenon. Heat flow into the cathode was determined by radiant and conductive transfer (eqs. 4 and 5). Heat flow into the cathode of the plasma torch comprised 4 – 5% of the total heat flow.

$$Q_{cond} = 4.5I + 23. \qquad (4)$$

$$Q_r = 4{,}9 I^2 . \qquad (5)$$

Heat transfer to the neutral section of the steam plasma torch was defined by convection and radiation. It was hard to distinguish what part of the convective and radiant heat flux was carried to the neutral section separately. Thus, the total heat flow was generalized using the similarity theory and was deduced in equation (6). Heat flow into the neutral section of the steam plasma torch comprised 10 – 15% of the total heat flow.

$$\frac{Q_{n.s.}}{P} = 9 \cdot 10^{-4} \left(\frac{I^2}{G d_1} \right)^{0.17} . \qquad (6)$$

Heat flow into the stair-shaped anode was determined by radiant, conductive and convective transfer. Here, the heat transfer between the arc, heated gas and the walls of the discharge chamber were the most intensive. Total heat flow into the separate d_2 and d_3 parts of the anode was generalized and expressed in equations (7) and (8), respectively.

$$\frac{Q_{d_2}}{P} = 7 \cdot 10^{-9} \left(\frac{I^2}{G d_2} \right)^{0.7} \cdot \left(\frac{G}{d_2} \right)^{-0.96} , \qquad (7)$$

$$\frac{Q_{d_3}}{P} = 9.6 \cdot 10^{-5} \left(\frac{I^2}{G d_3} \right)^{0.38} . \qquad (8)$$

Heat flow into the d_2 part of the anode comprised 17 – 34% of the total heat flow, whereas, heat flow into the d_3 part of the anode was in the range of 50 – 60% of the total heat flow.

In order to describe only the convective heat transfer between the arc, heated gas and the walls of the arc discharge chamber, the simplified assumptions have been made: the arc spot attaches in the narrow d_2 part of the anode, the investigation distance is behind the burning arc, $x/d > 11$, radiation of triatomic gas is uniform in the entire discharge chamber, and the flow temperature is the same the outflow plasma temperature. Therefore, if the radiant heat flux is known and the conductive heat flux is neglected, the convective heat flux into the arc discharge chamber walls could be expressed as follows:

$$q_c = q_{sum} - q_r . \qquad (9)$$

Then, in a wide range of variation of the operational parameters of the steam plasma torch, the heat transfer coefficient was calculated, and, knowing the physical properties of the steam and the geometrical parameters of the plasma torch, the convective heat flow was determined in the equation below and is shown in Fig. 9.

$$Nu_d = 0.0255 \mathrm{Re}_d^{0.8} \mathrm{Pr}^{0.43}. \tag{10}$$

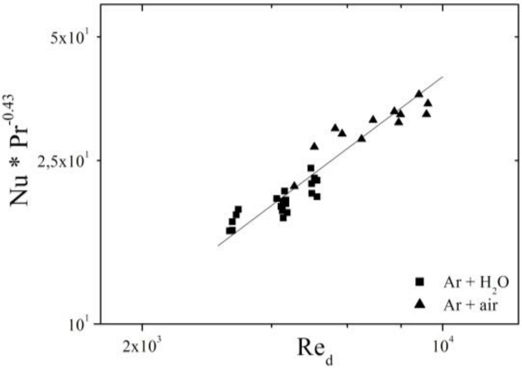

Fig. 9. The convective turbulent heat flow into the anode wall as the distance *x/d* > 11

It could be stated that at the distance of 11 < *x/d* < 15 the flow was turbulent, as the index near *Re* number indicated. Our experimentally determined equation (10), which describes the convective heat transfer in the cylindrical channel with a high-temperature turbulent steam flow, was in a good agreement with the one proposed in [1].

3.3. Diagnostics of the steam plasma at the outflow nozzle of the plasma torch

It is important to know not only the basic parameters of the plasma torch, but also to perform the diagnostics of generated plasma: temperature, velocity and chemical composition, which are crucial factors in the reforming process. Thus, the generated steam plasma was diagnosed using several methods.

3.3.1. The distribution of temperature and velocity profiles of outflow steam plasma by enthalpy probe method

The distribution of the temperature and velocity profiles lengthwise and cross-wise the outflow steam plasma stream was measured by means of enthalpy probe method [2]. The temperature and velocity profiles were measured at the plasma torch power of 57.4 kW (current 200 A, voltage 287 V, steam flow rate $3.71 \cdot 10^{-3}$ kg/s). Additionally, the obtained profiles were compared to the same plasma torch heating air as plasma-forming gas. The results are shown in Figures 10 and 11.

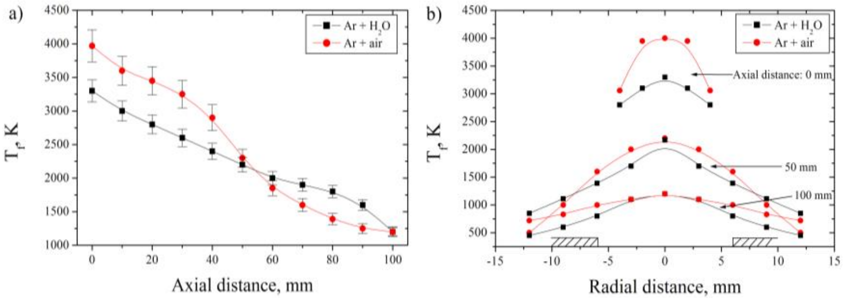

Fig. 10. Axial (a) and radial (b) distribution of the steam plasma temperature

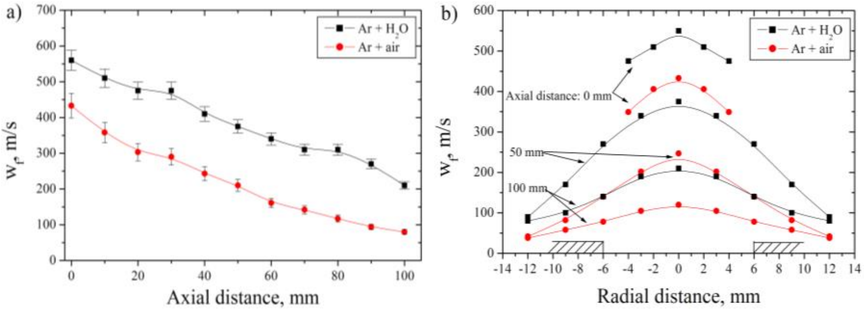

Fig. 11. Axial (a) and radial (b) distribution of the steam plasma velocity

The highest measured local temperature of the steam and air plasma at 0 mm axial distance was 3300±100 K and 4000±150 K, respectively, whereas, the highest local velocity of the steam and air plasma was 560±30 m/s and 430±30 m/s, respectively. The difference between the measured temperatures and velocities could be explained due to the different physical properties of the steam and air, especially enthalpy. Therefore, the enthalpy of steam plasma was three times higher the enthalpy of air plasma at such temperatures. Thus, the treated organic material would receive a three times higher energy density using steam as plasma-forming gas instead of air and would be converted to synthesis gas much faster. Additionally, the Reynolds number was calculated for both cases, which was $Re_d = 4900$ and $Re_d = 5000$ for the steam and air plasma, respectively. This indicates that the out-flowing high-temperature steam and air plasma was turbulent providing a better mixing with the treated organic materials.

3.3.2. Qualitative-quantitative analysis of steam plasma by optical emission spectroscopy

The qualitative-quantitative analysis of the steam plasma was performed by optical emission spectroscopy (OES) method. The qualitative analysis revealed the composition of the steam plasma, whereas, the quantitative analysis led to evaluate the temperature of plasma radicals and to determine either the steam plasma was in a local thermodynamic equilibrium or not. The emission spectra was measured at the plasma torch power of 47.6 kW (current 170 A, voltage 280 V, steam flow rate $3.71 \cdot 10^{-3}$ kg/s) and 62 kW (current 250 A, voltage 248 V, steam flow rate $3.71 \cdot 10^{-3}$ kg/s), respectively. The measured emission spectra are shown in Figures 12 and 13.

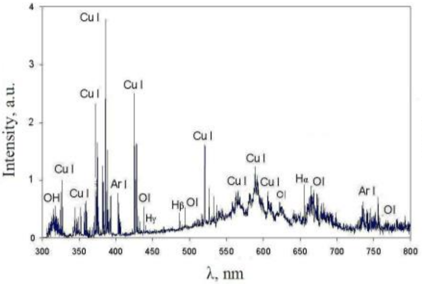

Fig. 12. The emission spectra in Ar-steam plasma at the plasma torch power of 47.6 kW

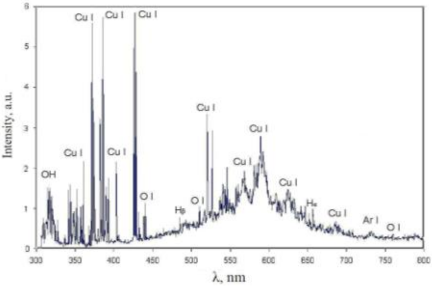

Fig. 13. The emission spectra in Ar-steam plasma at the plasma torch power of 62 kW

The emission spectra have showed that dominant chemical elements in Ar-steam plasma were excited Ar (I), O (I), Cu (I), OH. Additionally, Balmer emission line profile corresponding to H_α, H_β and H_γ were detected. The formation of O, H and OH radicals is very important in the reforming process of organic materials as their ability to initiate the oxidation-reduction chemical reactions and accelerate reaction kinetics much faster compared to conventional reforming methods.

Table 2. Experimental and calculated data

***I*, A**	***U*, V**	***P*, kW**	**G_{steam}, g/s**	**$T_{balance}$, K**	**T_{rot}, K**	**T_{exe}, K**
170	280	47.6	3.71	2500±6%	~1700	~4000
250	248	62	3.71	3100±6%	~4600	~4800

According to the spectra, there was the ability to roughly estimate the mean plasma temperature and to determine whether the steam plasma was in a local thermodynamic equilibrium or not. The OH (A-X) band spectra were used to calculate the gas temperature. It enabled to determine rotational T_{rot} temperature, which is considered to be equal to gas temperature ($T_{rot} \approx T_g$), by fitting the experimental spectra with the simulated one. LIFBASE [8] software for spectral simulation was used. The mean gas temperature was calculated from the heat balance equation. The excitation temperature (T_{exe}) was roughly calculated using a two-line method (considering Cu I line transitions between

510.554 nm and 515.324 nm, as well as among 515.324 nm and 521.820 nm) [9]. The results are shown in Table 2.

It could be seen that at current intensities of 170 A and 250 A, the difference between the temperatures ($T_{balance}$ and T_{rot}) was significant. The difference may appear due to the water steam used as the plasma-forming gas. Non-dissociated steam causes measurement uncertainty in spectra measurements. In the very humid environment, some portion of energy was spent for water evaporation and dissociation. In the second case, the T_{rot} was much higher than $T_{balance}$. It could be explained that the rotational temperature measured from OH spectra may be overestimated due to the fact that these radicals are the result of chemical reactions and residual chemical energy can manifest itself by elevating temperatures [10].

In order to determine whether the steam plasma was in a local thermodynamic equilibrium or not, the rotational and excitation temperatures should be equal to ($T_{rot} \approx T_{exe}$). At current intensity of 170 A, a difference between the rotational and excitation temperatures was significant. It indicated that our plasma was not in a local thermal equilibrium (non-LTE). On the contrary, at current intensity of 250 A, the obtained rotational and excitation temperatures were close to each other, indicating that the local thermodynamic equilibrium conditions in our plasma were fulfilled.

3.4. The formation and distribution of concentration of active radicals in the steam plasma

In spite of the fact that water steam has been directly involved in the plasma-chemical processes as the plasma-forming gas, a heat carrier and reagent, it is important to determine the dominant active radicals not only qualitatively but also quantitatively by evaluating its concentrations. Therefore, the formation and distribution of concentration of active radicals in the steam plasma was simulated by Chemical Workbench code [4] using a PH-type thermodynamic equilibrium reactor (TER) model. The simulation of the conversion process of organic materials and comparison with experiments will be discussed later.

The simulation was performed at the steam flow rate of $3.71 \cdot 10^{-3}$ kg/s, initiative temperature of 373 K, and pressure of 10^5 Pa in the TER. The PH-type TER model indicated that the simulation process is followed by constant pressure and enthalpy. The calculated concentrations of active radicals depending on the plasma temperature are shown in Fig 14.

As the temperature of the plasma increased from 400 K to 4100 K, the concentration of steam in the reactor decreased gradually and reached ~1% at 4100 K. Meanwhile, the concentrations of H_2, O_2 and OH increased gradually reaching their maximal values at 3400 K, and later decreased. The concentrations of O and H increased continually up to 4100 K. Additionally, the formation and distribution of concentrations of active radicals in the steam plasma were simulated depending on enthalpy. The obtained results are shown in Fig 15.

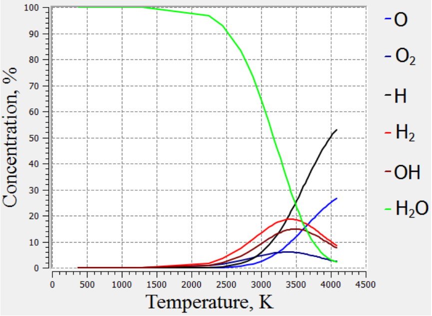

Fig. 14. The calculated concentrations of the chemical elements depending on the plasma temperature at thermodynamic equilibrium

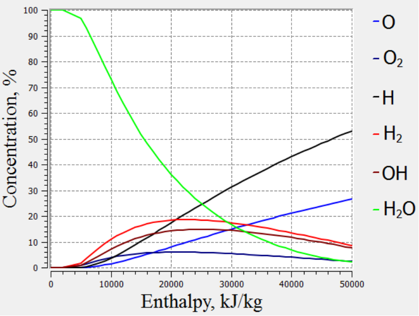

Fig. 15. The calculated concentrations of the chemical elements depending on the plasma enthalpy at thermodynamic equilibrium

As can be seen from the above figures, the highest concentrations of active radicals O_2, H_2 and OH were calculated at 3400 K or 24 MJ/kg. This means that the mean temperature of the plasma should be 3400 K in order to reach the best production of active radicals. Furthermore, in order to check whether the calculated concentrations match the experiment, H_2 concentrations were measured experimentally at 2700 K and 3000 K by gas chromatography method. It was determined that H_2 concentrations were 9%±1% and 12.5%±1 %, respectively, while those according to the model were 8% and 13%, respectively. As conclusion it could be stated that the model used in this study is suitable for the simulation of conversion processes of organic materials at thermodynamic equilibrium.

3.5. The conversion of organic materials in water steam plasma

3.5.1. Propane conversion

Propane conversion in the ambient of water steam plasma was carried out at two distinct experimental conditions:

- At constant C_3H_8 flow rate of $1.48\cdot10^{-3}$ kg/s, and various flow rates of steam of $(2.63 - 4.48)\cdot10^{-3}$ kg/s as well as the plasma torch power in the range of 56 – 63 kW. The H_2O/C_3H_8 ratio changed from 1.8 to 3, $T_f = 2800$ K.
- At constant H_2O flow rate of $3.51\cdot10^{-3}$ kg/s and plasma torch power of 56 kW, and various rates of C_3H_8 of $(0.66 - 1.48)\cdot10^{-3}$ kg/s. The C_3H_8/H_2O ratio changed from 0.18 to 0.42, $T_f = 2800$ K.

3.5.1.1. Effect on the steam content

The effect of the various steam flow rates on the reforming characteristics of propane was investigated. The produced gas concentrations, conversion degree of propane and H_2/CO ratio, H_2 and CO yield and selectivity, energy conversion efficiency with specific energy requirement depending on the H_2O/C_3H_8 ratio are shown in Figures 16, 17, 18, 19.

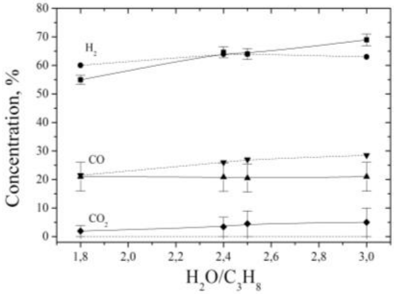

Fig. 16. Effect of the H_2O/C_3H_8 ratio on the produced gas composition. Full line represents the experimental results, dashed line – calculated

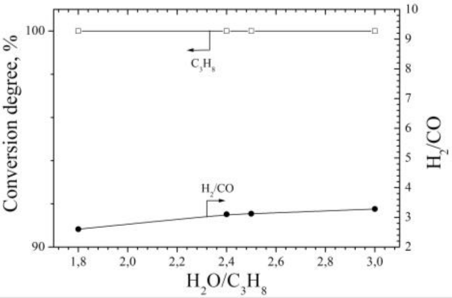

Fig. 17. Effect of the H_2O/C_3H_8 ratio on the conversion degree of propane and H_2/CO ratio.

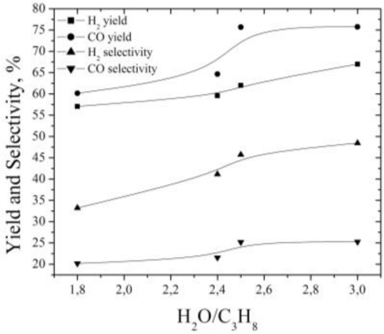

Fig. 18. Effect of the H_2O/C_3H_8 ratio on the H_2 and CO yield and selectivity

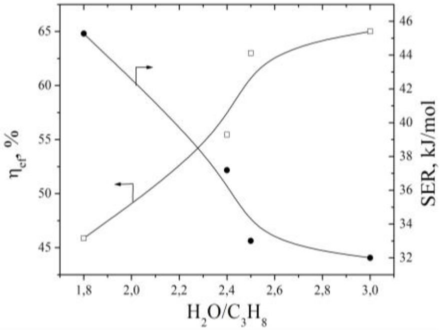

Fig. 19. Effect of the H_2O/C_3H_8 ratio on the energy conversion efficiency and specific energy requirement

As the H_2O/C_3H_8 ratio increased from 1.8 to 3, the H_2 and CO_2 concentrations increased, whereas CO concentration remained stable. The experimentally obtained H_2 and CO concentrations from the calculated at a thermodynamic equilibrium differ 3% and 24%, respectively. The increase of H_2

and CO_2 concentrations could be explained by the dominance of the exothermic water-gas-shift reaction.

The residence time varying from 6 to 8ms was sufficient to fully reform the propane to other gas as no trace of it was found after the reaction as shown in Fig. 17. The H_2/CO ratio, showing the quality of the synthesis gas produced, increased, as the H_2O/C_3H_8 increased.

The H_2 and CO yield as well as the selectivity increased as the H_2O/C_3H_8 changed from 1.8 to 3. It could be affected by the increased enthalpy (temperature) inside the plasma-chemical reactor as the content of steam in the steam/glycerol ratio increased. In order to quantify the reforming process, the energy conversion efficiency and the specific energy requirements were calculated, which are one of the major parameters for the evaluation of the process efficiency. As the H_2O/C_3H_8 ratio increased with simultaneous increase of the plasma torch power, the energy conversion efficiency increased and the specific energy requirement decreased. The best efficiency of the conversion process was obtained at the η_{ef} of 65% and the *SER* of 32 kJ/mol (equal to 0.31 kWh/kg) when the flow rate of steam and propane were $4.48 \cdot 10^{-3}$ kg/s and $1.48 \cdot 10^{-3}$ kg/s, respectively, and the plasma torch power of 63 kW.

3.5.1.2. Effect on the propane content

The effect of the various propane flow rates on the reforming characteristics was investigated. The produced gas concentrations, the conversion degree of propane and H_2/CO ratio, the H_2 and CO yield and selectivity, the energy conversion efficiency with specific energy requirement depending on the C_3H_8/ H_2O ratio are shown in Figures 20 – 23.

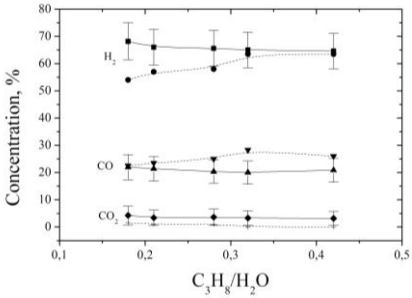

Fig. 20. Effect of the C_3H_8/ H_2O ratio on the produced gas composition. Full line represents the experimental results, dashed line – calculated

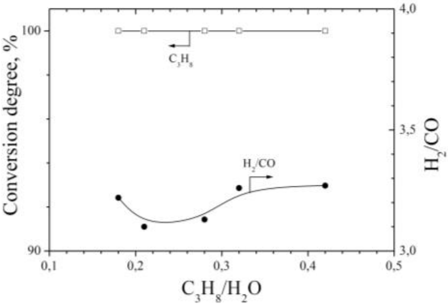

Fig. 21. Effect of the C_3H_8/ H_2O ratio on the conversion degree of propane and H_2/CO ratio.

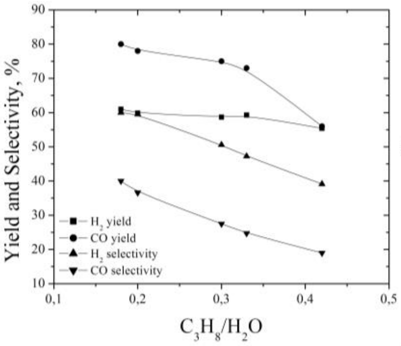

Fig. 22. Effect of the C_3H_8/ H_2O ratio on the H_2 and CO yield and selectivity

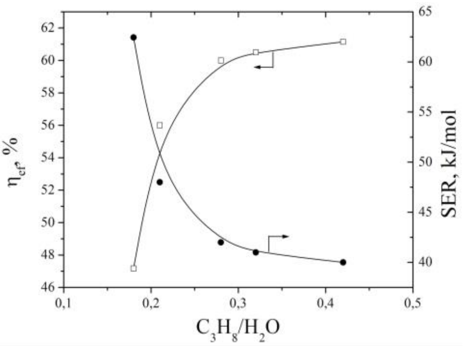

Fig. 23. Effect of the C_3H_8/ H_2O ratio on the energy conversion efficiency and specific energy requirement

As the C_3H_8/ H_2O ratio increased from 0.18 to 0.42, the H_2, CO and CO_2 concentrations were almost stable or slightly decreased. The experimentally obtained H_2 and CO concentrations from the calculated at a thermodynamic equilibrium differ 10% and 21%, respectively.

The residence time was sufficient to fully reform the propane, and the H_2/CO was close to stable 3.1 – 3.3. This stability could be explained by the carbon black formation during the process of the CO methanation reaction.

The H_2 and CO yield as well as the selectivity decreased as the C_3H_8/H_2O ratio increased. The energy conversion efficiency increased with simultaneous decrease of the specific energy requirement. The best process efficiency was obtained at the η_{ef} of 61% and the *SER* of 40 kJ/mol (equal to 0.37 kWh/kg) when the flow rate of propane and steam were $1.48 \cdot 10^{-3}$ kg/s and $3.51 \cdot 10^{-3}$ kg/s, respectively, and the plasma torch power of 56 kW.

3.5.2. Glycerol conversion

Glycerol conversion in the ambient of water steam plasma was performed at the following experimental conditions: glycerol flow rate of $2 \cdot 10^{-3}$ kg/s, steam flow rate of $(2.63 – 4.48) \cdot 10^{-3}$ kg/s, and the plasma torch power of 48 – 56 kW. The $H_2O/C_3H_8O_3$ ratio changed from 1.3 to 2.2, T_f = 2800 K. Additionally, the flow rate of glycerol was increased to $4 \cdot 10^{-3}$ kg/s at the same experimental conditions.

The produced gas concentrations, the conversion degree of glycerol and reaction products yield, the glycerol conversion to gas and char, the H_2 and CO yield with H_2/CO ratio, and the energy conversion efficiency with specific energy requirement depending on the $H_2O/C_3H_8O_3$ ratio and plasma torch power are shown in Figures 24 – 28.

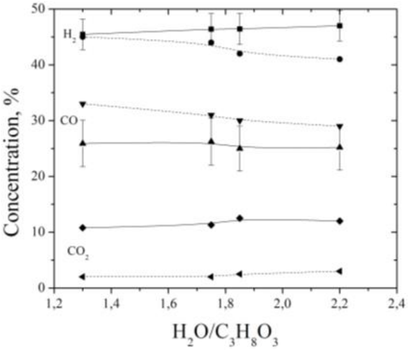

Fig. 24. Effect of the $H_2O/C_3H_8O_3$ ratio on the produced gas composition. Full line represents the experimental results, dashed line – calculated

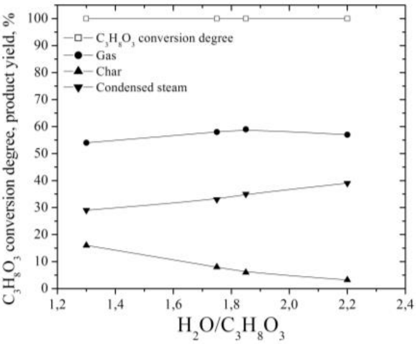

Fig. 25. Effect of the $H_2O/C_3H_8O_3$ ratio on the glycerol conversion degree and product yield

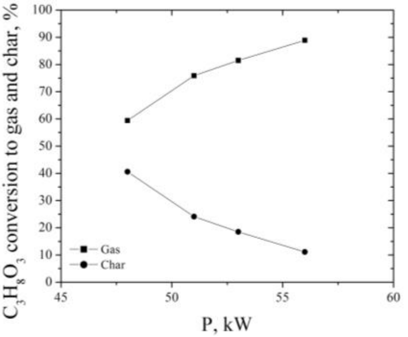

Fig. 26. Effect of the plasma torch power on the glycerol conversion to gas and char

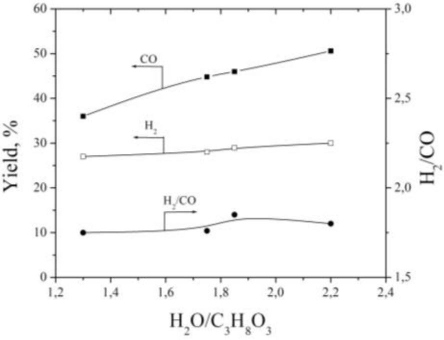

Fig. 27. Effect of the $H_2O/C_3H_8O_3$ ratio on the H_2 and CO gas yield and H_2/CO ratio

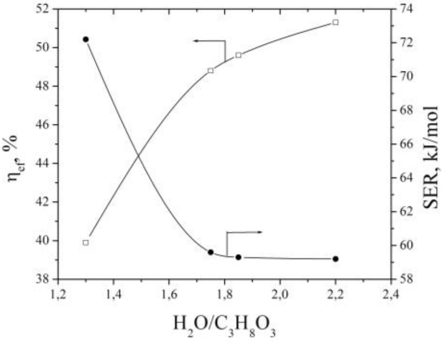

Fig. 28. Effect of the $H_2O/C_3H_8O_3$ ratio on the energy expenditure

As the $H_2O/C_3H_8O_3$ ratio increased from 1.3 to 2.2, the H_2 and CO concentrations were stable, whereas that of CO_2 slightly increased. The experimentally obtained H_2 and CO concentrations from the calculated at a thermodynamic equilibrium differ 6% and 17%, respectively. Glycerol conversion mechanism in the ambient of water steam plasma is complicated and mostly based on the steam reforming, cracking, water-gas-shift, methanation, and hydrogenolysis reactions, etc. Thus, the reaction pathway depends on temperature, active radicals' concentration, residence time, etc.

No traces of the glycerol after the conversion had been detected. It was decomposed into gas and liquid mostly and little amount of char as is indicated in Figs 25, 26. Therefore, as the $H_2O/C_3H_8O_3$ ratio and plasma power increased, the yield of gas in the total reaction products mass balance increased, while the yield of char decreased significantly. The yield of liquid products, mostly water, increased due to excess steam and methanation, and hydogenolysis of glycerol. On the other hand, it determined a lower H_2 yield and a slight increase of it. However, the CO yield was high enough and increased significantly, indicating that the glycerol was mainly converted to gas. The H_2/CO ratio was close to stable, showing the ability of the synthesis gas produced to be directly used for energy/heat or chemicals production. The best process efficiency was obtained at the η_{ef} of 51% and the *SER* of 59 kJ/mol (equal to 0.54 kWh/kg) when the flow rate of glycerol and steam were $2 \cdot 10^{-3}$ kg/s and $4.48 \cdot 10^{-3}$ kg/s, respectively, and the plasma torch power of 56 kW.

As the content of glycerol in the $H_2O/C_3H_8O_3$ ratio increased to $4 \cdot 10^{-3}$ kg/s, the content of produced gas increased up to 6% in the total reaction products mass balance. However, the char content increased to 15% giving a lower carbon conversion efficiency of 63%, but the liquid product concentration decreased more than 20%. Despite the increased char content, the energy conversion efficiency slightly increased up to 52% and the specific energy requirement decreased to 44.7 kJ/mol compared to 51% and 59 kJ/mol, respectively, at $2 \cdot 10^{-3}$ kg/s glycerol flow rate. Therefore, these results suggest that there is a potential for further increase of flow rate of glycerol in order to achieve a better reaction performance as well as the process efficiency. However, char formation could be minimized by improving the reactor design, i.e. installing the glycerol spray nozzles closer the plasma jet or by increasing the reaction temperature. Thus, the glycerol could be pyrolized even much faster by high-temperature steam plasma with higher active radicals' concentration inside.

3.5.3. Wood conversion

Wood conversion in the ambient of water steam plasma was carried out at the following experimental conditions: wood flow rate of $1.2 \cdot 10^{-3}$ kg/s, steam flow rate of $(2.63 – 4.48) \cdot 10^{-3}$ kg/s, and the plasma torch power of 49 – 56 kW. The steam/wood ratio was in the range of 2 – 3.4, $T_f = 2800$ K.

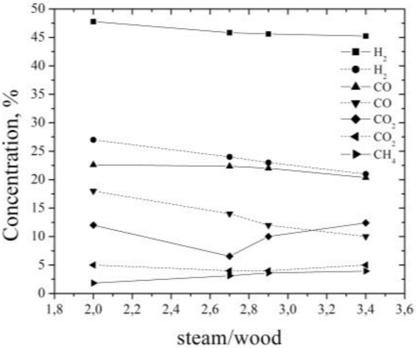

Fig. 29. Effect of the steam/wood ratio on the produced gas composition. Full line represents the experimental results, dashed line – calculated

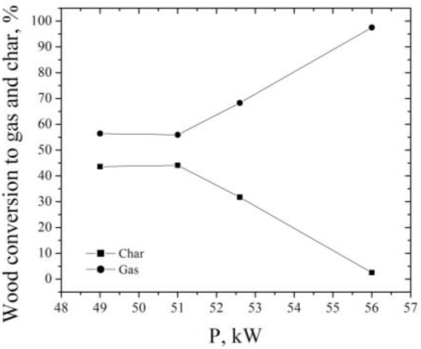

Fig. 30. Effect of the plasma torch power on the wood conversion to gas and char

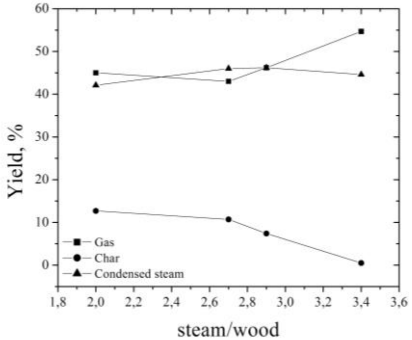

Fig. 31. Effect of the steam/wood ratio on the product yield

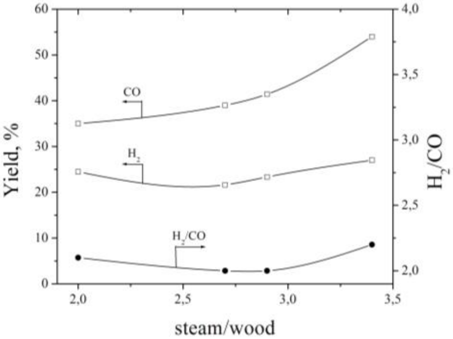

Fig. 32. Effect of the steam/wood ratio on the H_2 and CO gas yield and H_2/CO ratio

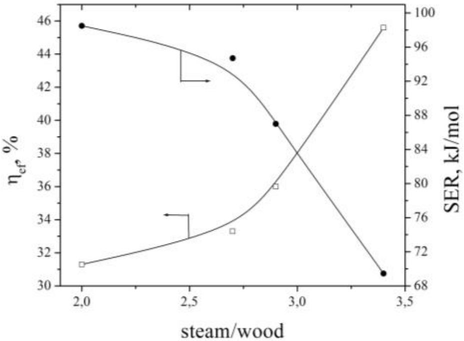

Fig. 33. Effect of the steam/wood ratio on the energy expenditure

The produced gas concentrations, the wood conversion to gas and char, the reaction products yield, the H_2 and CO yield with H_2/CO ratio, and the energy conversion efficiency with specific energy requirement depending on the steam/wood ratio and plasma torch power are shown in Figures 29 – 33.

As the steam/wood ratio increased from 2 to 3.4, the H_2 and CO concentrations slightly decreased, whereas the CH_4 content increased. The CO_2 concentration decreased while the steam/wood ratio increased to 2.7 and began to increase after the steam/wood ratio of 2.7. This could be affected by the dominance of the exothermic water-gas-shift reaction. The experimentally obtained H_2 and CO concentrations from the calculated at a thermodynamic equilibrium differ more than 40 – 50% and 25 – 50%, respectively. It shows that the simulation program used was not suitable for the modeling of the wood conversion process. Wood pyrolysis/gasification in the ambient of water steam plasma is complicated and mostly based on the water-gas, Boudouard hydrogasification, water-gas-shift, methanation reactions etc.

The wood was completely converted to gas, liquid and chars, as it is shown in Figures 30 and 31. At the plasma torch of 56 kW and the steam/wood ratio of 3.4, the residence time was sufficient for the wood conversion to gas and liquid. The yield of char in the total reaction products mass balance decreased almost to 0%, while the gas yield increased. As the wood consists of moisture and H_2, the yield of liquids, mostly condensed water, slightly increased due to excess steam used in the process and wood methanation reaction. The H_2 yield slightly increased, while that of CO increased significantly. Much higher CO yield could be explained due to that the wood is mainly composed of C and O, and therefore, after the reaction in the plasma-chemical reactor, the wood was mainly decomposed into the gas leading to the increase of the CO yield. The H_2/CO ratio was close to stable, indicating that the produced synthesis gas was of high quality. The best process efficiency of the wood conversion was obtained at the η_{ef} of 45.6% and the *SER* of 69 kJ/mol (equal to 0.64 kWh/kg) when the flow rate of wood and steam were $1.2 \cdot 10^{-3}$ kg/s and $4.48 \cdot 10^{-3}$ kg/s, respectively, and the plasma torch power of 56 kW.

3.5.4. Summary of the results

Once again the results of the conversion of various phase organic materials to synthesis gas have been summarized and are shown in Table 3 at the best process efficiency.

Table 3. Summary of the conversion results

Material	**Ratio with steam**	**P, kW**	**Yield, %**		**H_2/CO**	**η_{ef}, (%)**	**SER, (kJ/mol)**
			H_2	**CO**			
Propane	3	63	67	75	3,3	65	32
Glycerol	2,2	56	30	50	1,85	51	59
Wood	3,4	56	27	54	2,2	45,6	69

As it is presented in the above, the best efficiency of the conversion process was get using propane as organic material as compared to the cases of glycerol and wood used. Therefore, gaseous materials do not require additional energy to be used for their decomposition into gas, as liquid and solid do. Thus, the overall efficiency of the conversion process depends not only on the plasma parameters but also on the flow rate of treated materials ant their physical properties such as physical state, caloric value, preparation etc.

In order to get higher process efficiency, wastes with higher caloric value are required. Therefore, considering the environmental regulations on emissions, the water steam arc plasma method could be successfully implemented as a technologically advanced solution avoiding direct organic/inorganic waste incineration with simultaneous extraction of valuable reaction products. Thus, the water steam arc plasma technology offers a possibility to reach the neutralization efficiency up to 99.99%, is characterized by a smaller size of the equipment, fast start-up–shutdown and control of the process, ability to reach very high temperatures which are limited in conventional neutralization methods, waste volume reduction to 3 – 5% of their initial volume, ability to get synthesis gas as a by-product of the process, which could be further used as a feedstock for energy/heat, chemicals, such as methanol, hydrogen, etc., production or liquefied and used in the internal combustion engines or fuel cells.

CONCLUSIONS

In this thesis, the thermo-hydro-dynamic processes in the arc discharge chamber of the newly designed water steam plasma torch and generated plasma stream were investigated. The plasma torch was successfully used for the organic waste treatment extracting synthesis gas as the main product of the process.

1. The highest mean outflow temperature and velocity of the steam plasma stream was 3300 K and 600 m/s, respectively, obtained at the experimental conditions in the range of current intensity of 120 – 250 A, voltage of 230 – 400 V, and steam flow rate of $(2.63 - 4.48)\cdot10^{-3}$ kg/s.
2. The highest heat flux was determined into the walls of the (d_3) part of the anode of the plasma torch were it was transferred by convection, radiation and conduction, and comprised 50 – 60% of the total heat flow. The flow in the discharge chamber of the plasma torch at the distance of $11 < x/d < 15$ was observed to be turbulent.
3. The numerical simulation revealed that as the temperature in the reactor increased to 4100 K, the concentration of steam decreased to 1%, while those of H and O increased gradually. The maximal H_2, O_2 and OH concentrations were calculated at 3400 K. The experimentally determined H_2 concentration at 2700 K and 3000 K was 9±1% and 12.5%, respectively, whereas according to the simulation it was 8% and 13%, respectively.
4. It was determined that after the thermal steam plasma conversion of various phase organic materials, the main reaction products were H_2 and CO gas. The concentration of synthesis gas was over 55% in the total mass balance.
5. The experimental studies revealed that, as the content of steam in the total steam/treated material ratio increased from $2.63\cdot10^{-3}$ kg/s to $4.48\cdot10^{-3}$ kg/s (as well as the power of the plasma torch), the yield of synthesis gas and energy conversion efficiency increased, whereas the specific energy requirement decreased.

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Reziumė

Termohidrodinaminių procesų tyrimas vandens garo plazmoje ir jos pritaikymas organinių medžiagų konversijai

Didėjant iškastinio kuro ir iš jo gaunamų medžiagų poreikiui, didėja su jo vartojimu susijusios problemos, t.y. teršalų patekimas į atmosferą sukeliantis „šiltnamio“ reiškinį, sąvartynų ploto didėjimas, ozono sluoksnio storio plonėjimas, fotocheminis smogas ir t.t. Todėl būtina kurti naujas technologijas, užtikrinančias švaresnį, racionalesnį bei efektyvesnį energijos generavimą, tenkinant aplinkosaugos reikalavimus, bei mažinant priklausomybę nuo smarkiai senkančių iškastinio kuro išteklių. Viena pagrindinių technologijų, lemiančių švaresnį, racionalesnį ir efektyvesnį energijos generavimą yra plazminė konversija. Ji jau plačiai naudojama įvairių rūšių atliekų nukenksminimo procesuose, medžiagų perdirbimo ir modifikavimo srityse. Nuo XX a. paskutiniojo dešimtmečio plazminėms technologijoms pasauliniame moksle ir praktikoje skiriamas ypatingas dėmesys. Jos tapo pagrindinėmis priemonėmis energijos taupymo, aplinkos taršos mažinimo bei naujų medžiagų sintezės srityse ir šiuo metu vaidina svarbų vaidmenį žmogaus gyvenime.

Neseniai paaiškėjo, kad egzistuoja galimybė pasiekti dar daug geresnių rezultatų energetikos, aplinkosaugos, chemijos, transporto ir kt. pagrindinėse srityse panaudojant vandens garo plazmą. Tai yra alternatyva tradiciniams perdirbimo ir pavojingų medžiagų nukenksminimo metodams. Naudojant vandens garo plazminę technologiją apdorojamos medžiagos ir atliekos nėra tiesiogiai deginamos. Skaidymas vyksta anaerobinėje aplinkoje, todėl nesusidaro kenksmingų junginių.

Plazminis metodas pasižymi ypatingai aukštu nukenksminimo efektyvumu (99,99 %), aukšta temperatūra (2000–3500 K), dideliu energijos tankiu, nedidelių gabaritų įrengimais, greitu proceso paleidimu ir sustabdymu, pigesne instaliacija ir yra ekologiškai švaresnis nei tradiciniai apdorojimo metodai. Taigi, taikant vandens garo plazminę technologiją galima išspręsti daugelį problemų: sumažinti tinkamų perdirbti atliekų deponavimą sąvartynuose, sumažinti kenksmingų atliekų emisiją, perdirbti įvairių rūšių kurą bei gauti vertingas sintetines dujas, kurios plačiai naudojamos kaip kuras katiluose, vidaus degimo varikliuose ar vertingų cheminių medžiagų, vandenilio, metanolio, acetileno ir kt., gamyboje. Šis būdas yra itin perspektyvus, nes gali būti plėtojama beatliekinė (*zero waste*) arba mažai atliekų išskirianti technologija.

Atlikus pasaulinės mokslinės-techninės literatūros analizę pastebėta, kad specialiuose mokslo žurnaluose gausu informacijos apie plazmines medžiagų, kuro ir atliekų apdorojimo technologijas, nuolat vyksta stambios pasaulinės mokslinės konferencijos. Tačiau literatūroje visiškai nėra duomenų apie vandens garo plazmos srauto tekėjimą, elektrinius ir šiluminius procesus bei sąveiką su apdorojamomis medžiagomis. Literatūroje mažai duomenų apie šilumos ir masės

pernešimo procesus plazmos generatoriuje ir plazmocheminiuose reaktoriuose. Taigi, efektyviam metodo taikymui reikalingi išsamesni moksliniai tyrimai projektuojant ir konstruojant vandens garo plazmos generatorių, įvertinant jo konstrukciją, šilumines ir elektrines charakteristikas, jame vykstančius šiluminius ir dinaminius procesus. Būtina atlikti vandens garo plazmos srovės diagnostiką, parinkti optimalius darbo parametrus nepertraukiamam organinių medžiagų neutralizavimo ir skaidymo procesui. Todėl šiame darbe atliekamų tyrimų svarba yra akivaizdi.

Darbo tikslas – sukurti atmosferinio slėgio vandens garo plazminę technologiją, skirtą organinių medžiagų konversijai, ir atlikti termohidrodinaminių procesų tyrimus vandens garo plazmos generatoriuje ir plazmos sraute.

Darbe nustatytos vandens garo plazmos sudarymo sąlygos naujai suprojektuotame ir pagamintame plazmos generatoriuje; pirmą kartą detaliau ištirti jo lankinio išlydžio kameroje vykstantys termohidrodinaminiai procesai; parodyta, kad vandens garo plazminė technologija gali būti efektyvi priemonė įvairios kilmės organinių medžiagų konversijos procesui atlikti.

Pagrindiniai darbe gauti rezultatai parodė, kad sukurto vandens garo plazmos generatoriaus, skirto organinių medžiagų konversijai, darbo pobūdis priklauso nuo jo geometrinės konstrukcijos ir režimo parametrų. Naudotas nuolatinės srovės linijinis PG su laiptuotu ar konfuzoriaus tipo anodo kanalu, stabiliai dirbo 120–250 A srovių, 230–400 V įtampos ir $(2{,}63–4{,}48)\cdot10^{-3}$ kg/s vandens garo srauto ribose. Jo voltamperinės charakteristikos yra parabolės formos, o šiluminis naudingumo koeficientas kinta 53–76 %. Ištyrus šilumos srautus į atskiras vandens garo PG elektros lanko išlydžio kameros dalis nustatyta, kad didžiausias šilumos srautas susidaro plačiojoje anodo dalyje ir siekia 50–60 % visų šilumos nuostolių. Srautas, tekėdamas elektros lanko išlydžio kamera, yra turbulizuojamas atstume $11 < x/d < 15$. Skaitiniais metodais išanalizavus aktyvių radikalų susidarymą vandens garo plazmoje nustatyta, kad, didėjant garo temperatūrai iki 4100 K, jo koncentracija sumažėja iki 1 %, o jį sudarančių atominio H ir O elementų koncentracija nuolat didėja. Nustatyta, kad maksimalios H_2, O_2 molekulių ir OH radikalo koncentracijos pasiekiamos, esant vidutinei plazmos srauto temperatūrai artimai 3400 K. Eksperimentiniu metodu gauta, kad 2700 K ir 3000 K temperatūros vandens gare, H_2 koncentracija atitinkamai siekė 9 ir 12,5 % ± 1 %, o apskaičiuota pagal modelį 8 ir 13 %. Realizavus organinių medžiagų konversijos procesą vandens garo plazmoje ir ištyrus pagrindinių produktų sintezės dėsningumus nustatyta, kad pagrindiniai reakcijos produktai yra H_2 ir CO dujos. Nustatyta, kad didėjant vandens garo kiekiui, bendrame H_2O/skaidomos medžiagos sraute, sintetinių dujų (H_2 + CO) išeiga ir energijos konversijos efektyvumas didėja, o savitosios energijos sąnaudos mažėja.

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