Atmospheric pressure microwave plasma source for hydrogen production

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Introduction

- Hydrogen is more and more attractive as an efficient and environmental friendly source of energy
- It is considered as a promising fuel of the future
- European roadmap for hydrogen and fuel cells published by European Commission in “Hydrogen Energy and Fuel Cells: A vision of our future” (EUR 20719, EN 2003)
    - from fossil fuel-based economy to hydrogen oriented economy
    - developing and implementing systems for hydrogen production from renewable electricity and biomass
    - research and development of other carbon-free hydrogen sources, such as solar thermal and advanced nuclear should be continued
- United States Department of Energy „Toward a more secure and cleaner energy future for America. National hydrogen energy roadmap” (2002)
    - lower the hydrogen production cost and to improve efficiency
    - improving existing commercial processes (e.g. steam methane reformation) and development new advanced production techniques (e.g. biological methods, nuclear and solar-powered)

Introduction

- Our motivation arises from the growing interest in the hydrogen production technologies
- Investigation concerns microwave (2.45 GHz) atmospheric pressure plasma source (MPS) for hydrogen production via methane conversion
- Microwave plasma applications: hydrocarbons destruction [Jasinski et al., 2004], noble gas purification [Rostaing et al., 2000], surface treatment during cars production, aviation industry, textile and biomedical engineering [Chu et al., 2002, Denes et al., 2004, Tendero et al., 2006, Morent et al., 2008], hydrogen production from decomposition of methanol [Henriques et al., 2010], hydrogen production from water [Pineda et al., 2007]
- Hydrogen production reactions from methane:

$CH_4 \rightarrow C+2H_2$	**(pyrolysis)**
$CH_4+CO_2 \rightarrow CO+2H_2$	**(dry reforming)**
$CH_4+H_2O \rightarrow CO+3H_2$	(steam reforming)
$CH_4+0.5O_2 \rightarrow CO+2H_2$	(partial oxidation)
$2CH_4+O_2+CO_2 \rightarrow 3CO+3H_2+H_2O$	(auto-thermal reforming)
$4CH_4+O_2+ 2H_2O \rightarrow 4CO+10H_2$	(auto-thermal reforming)

Experimental setup

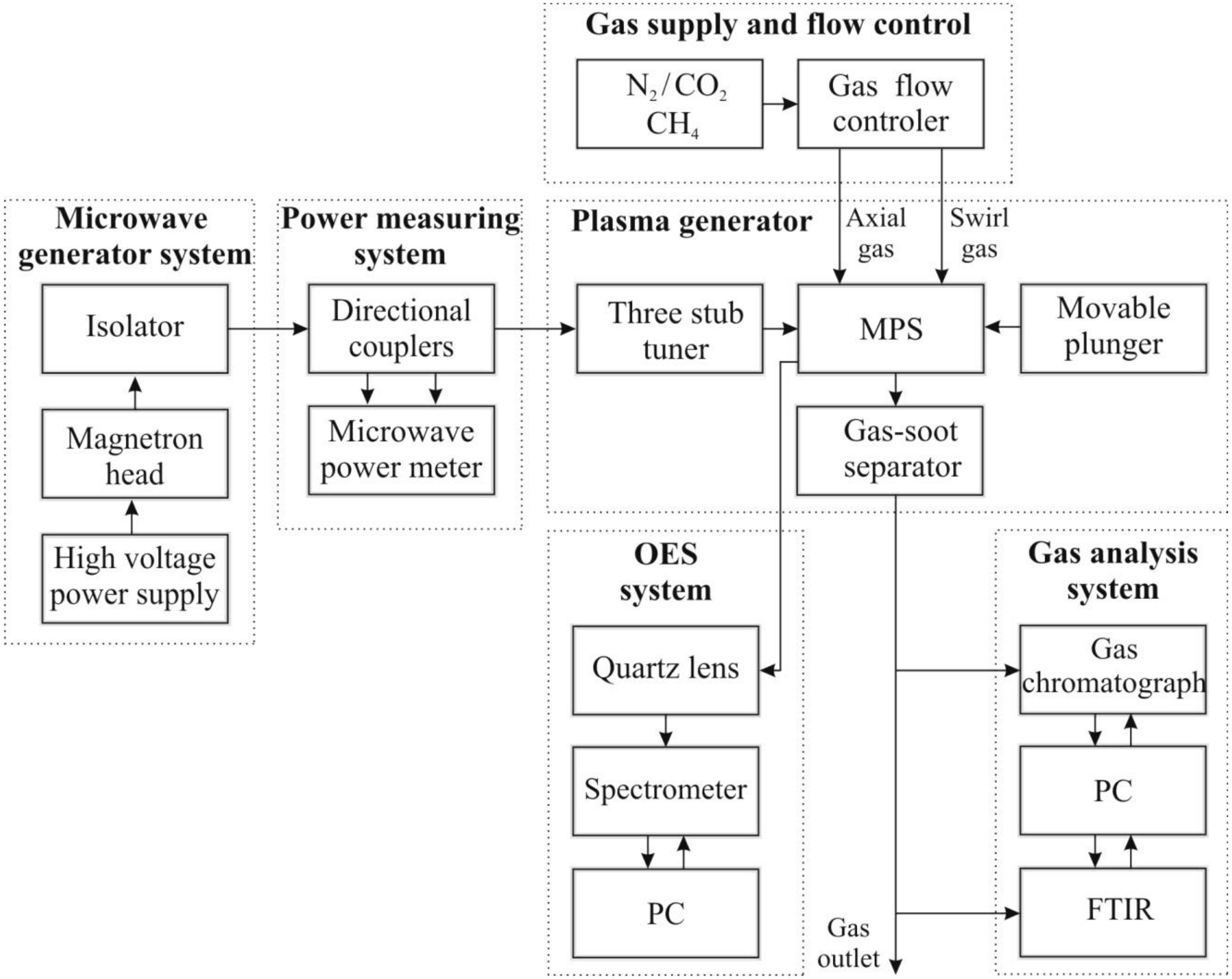


Diagram of the experimental setup

Parameters:

Microwave frequency: 2.45 GHz

Microwave power: up to 6 kW

Working gas: N_2, Air, CO_2, CH_4,

Gas flow rate: up to 200 l/min

Pressure: atmospheric

OES–Optical Emission Spectroscopy

Experimental setup

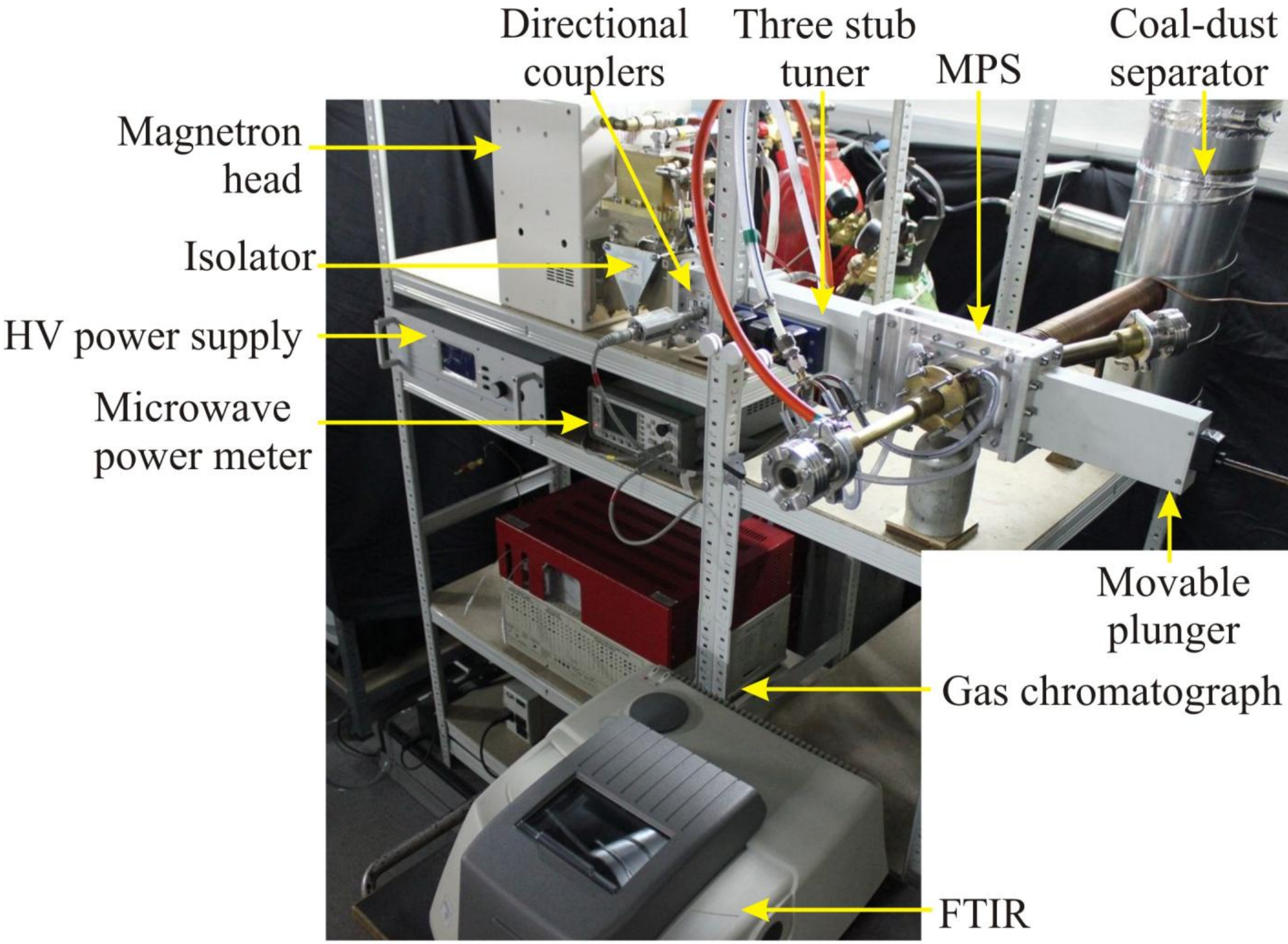
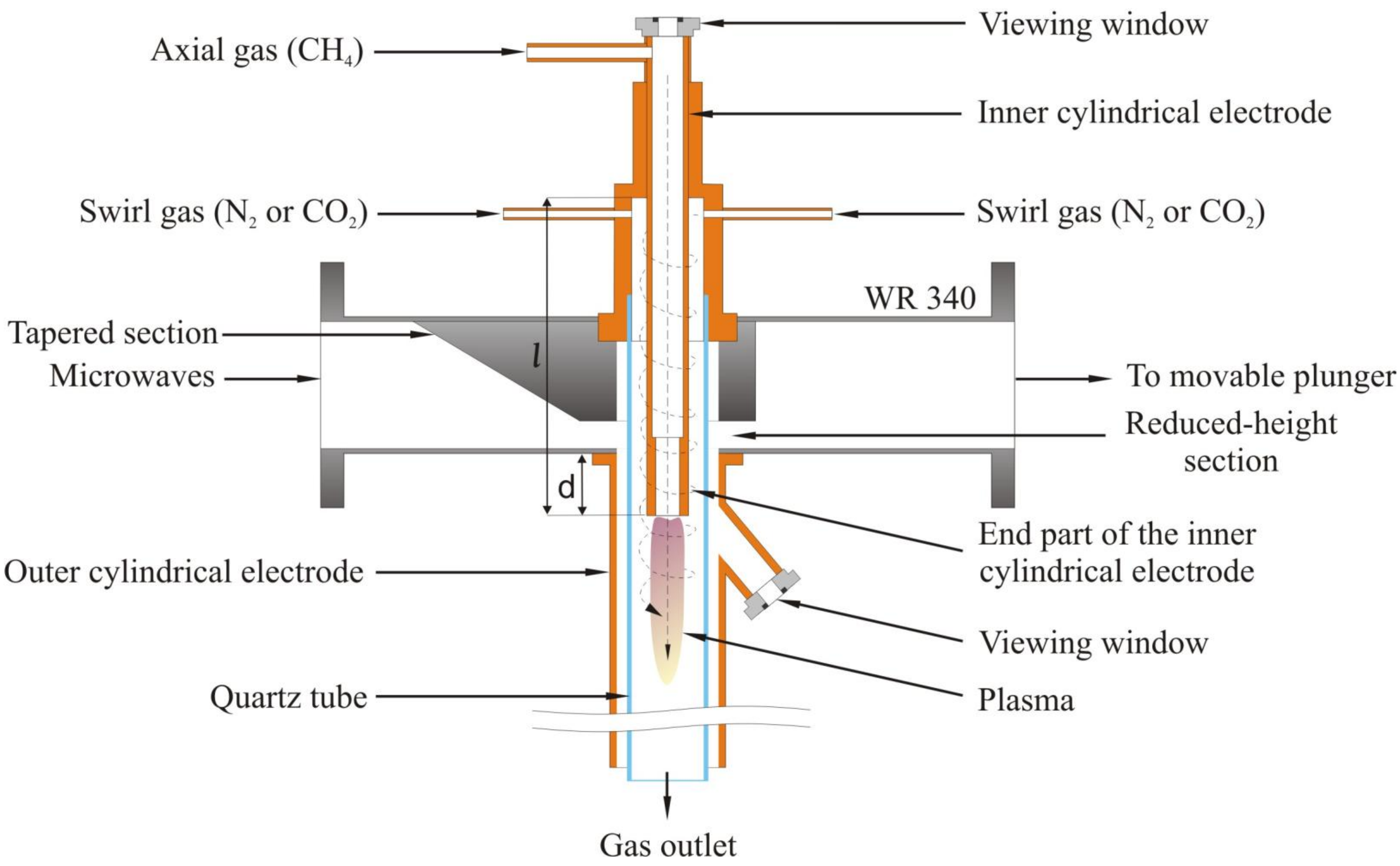


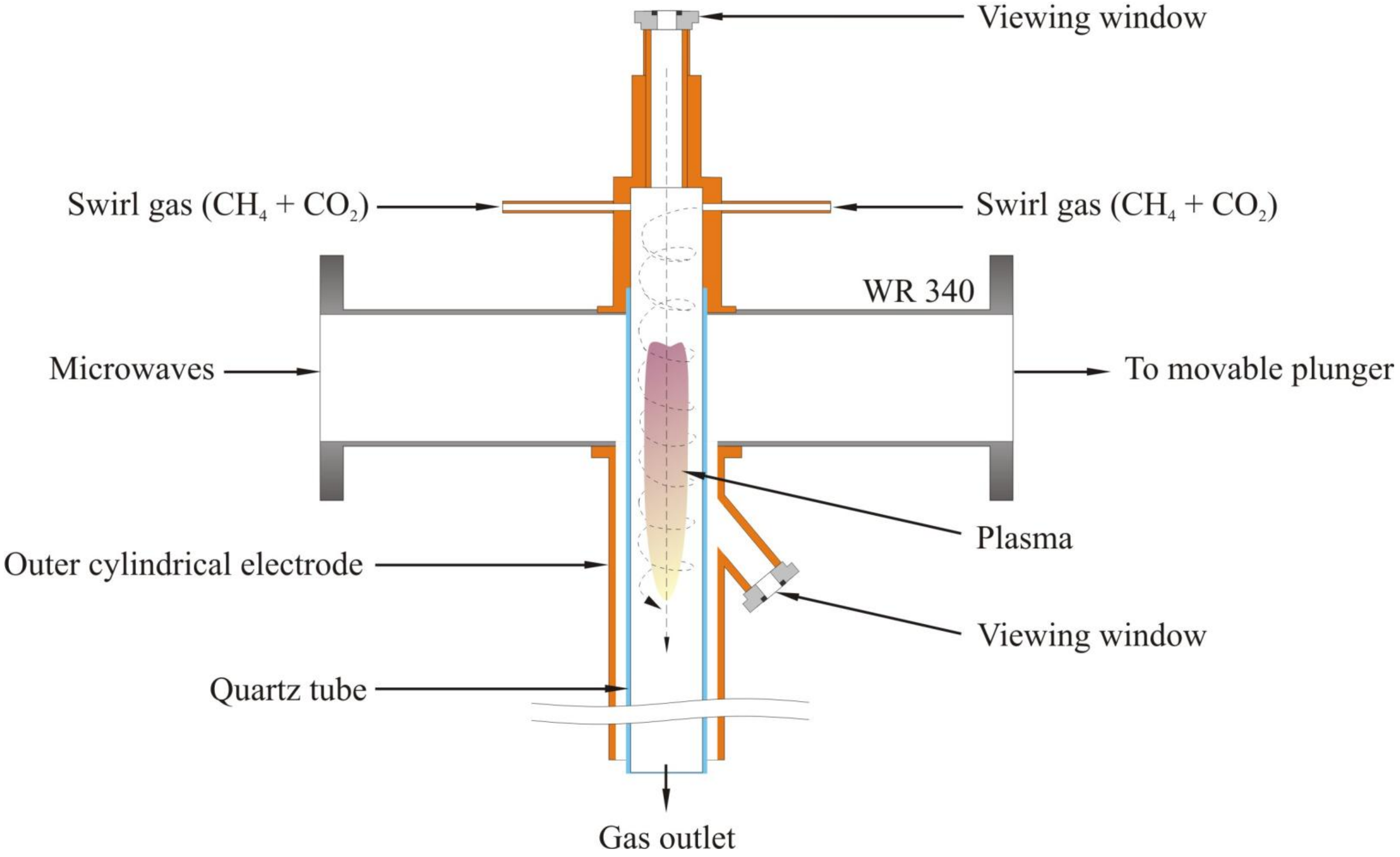
Photo of the experimental setup

Waveguide-supplied coaxial-line-based MPS



Schematic view of the waveguide-supplied coaxial-line-based MPS

Waveguide-supplied metal-cylinder-based MPS



Schematic view of the waveguide-supplied metal-cylinder-based MPS

Waveguide-supplied MPS

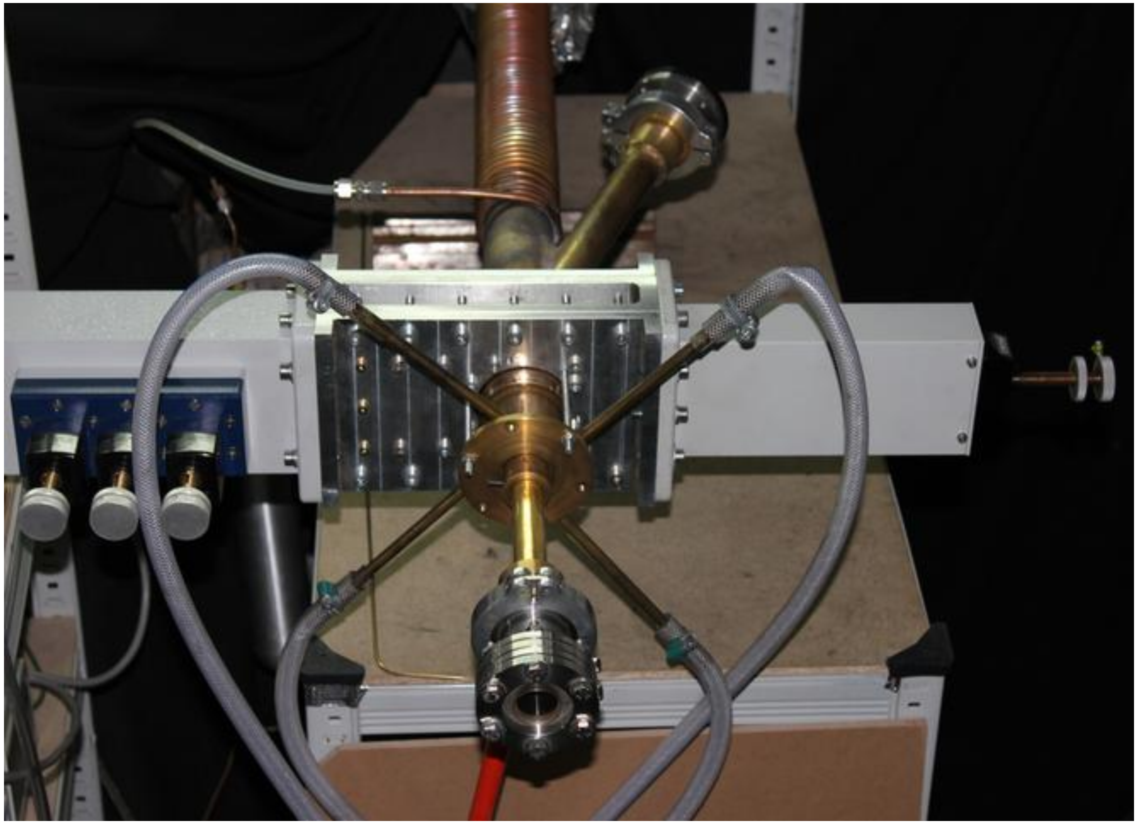
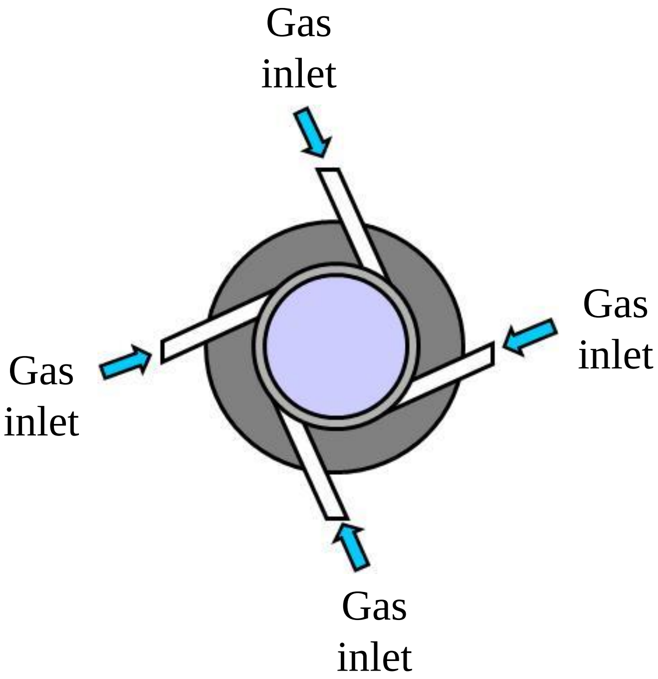


Photo of the MPS

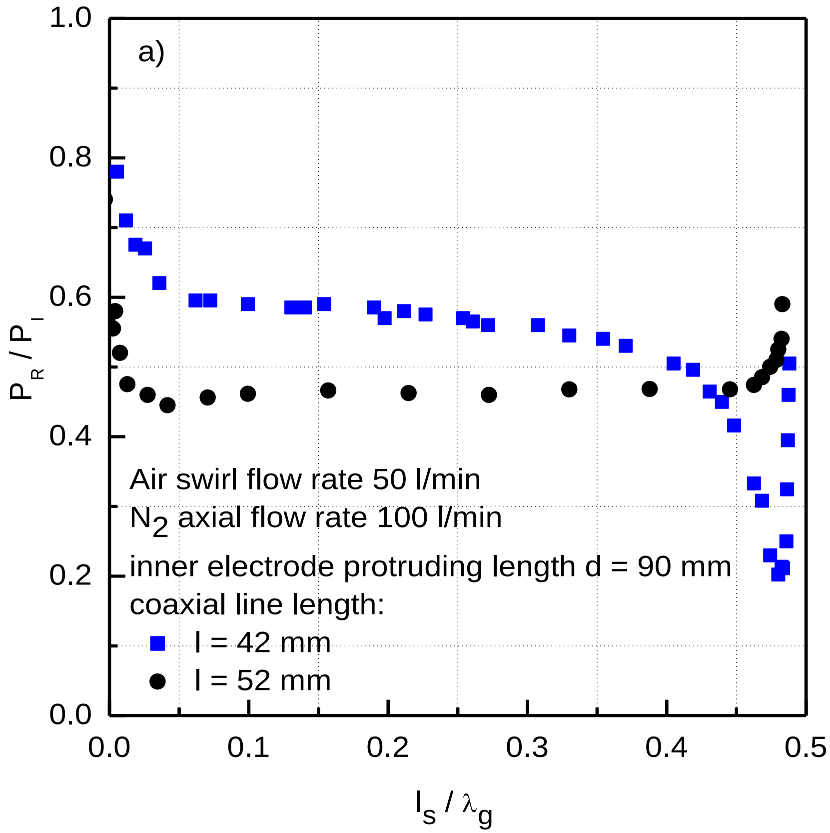


Scheme of the swirl gas inlets

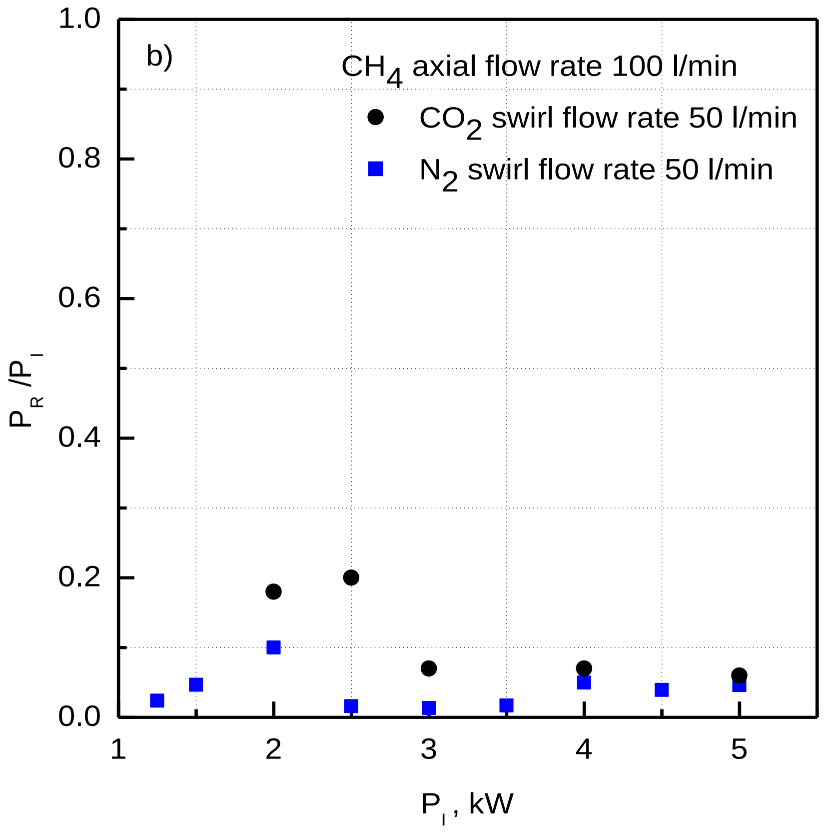
Results

- Tuning characteristics of the MPSs – to determine the efficiency of microwave power transfer to the plasma and stability of MPSs operation
- Visualization of the plasma flame – photos of the methane plasma flame for different working conditions
- Spectroscopic diagnostics – in the meaning of Optical Emission Spectroscopy, to determine the gas temperature
- Hydrogen production via methane conversion – hydrogen production rate and energy efficiency

Results: Tuning characteristics of the MPS

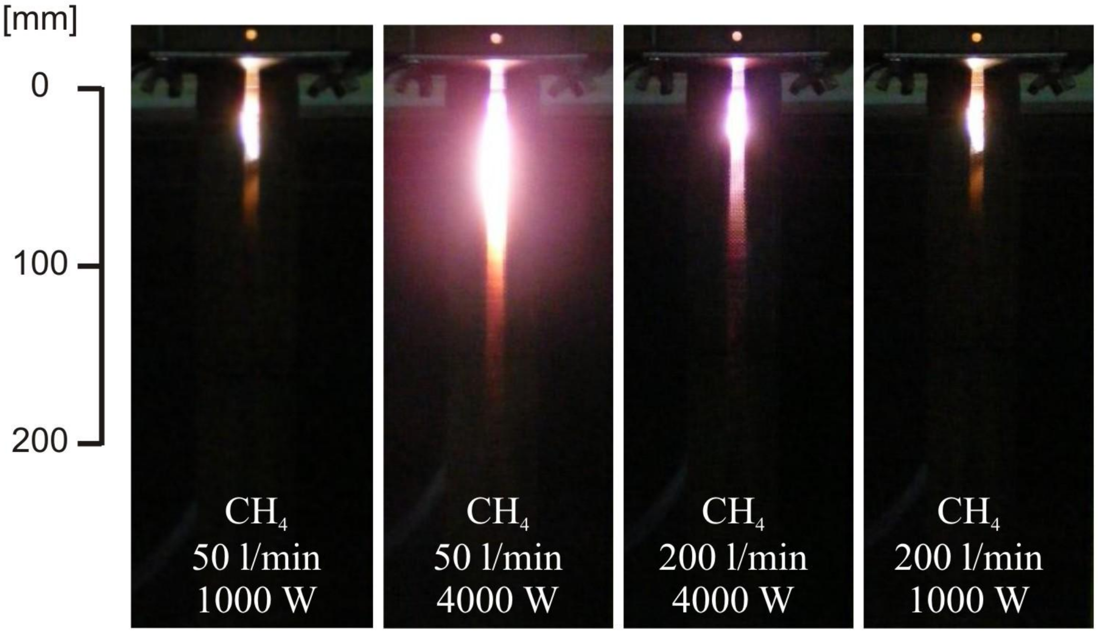


Tuning characteristics of the waveguide-supplied coaxial-line-based MPS measured for inner cylindrical electrode protruding length d=90 mm and incident microwave power P_I=2000 W



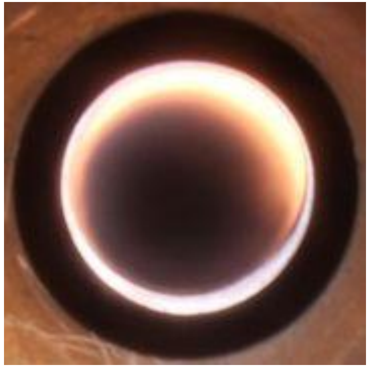
Dependence of the reflection coefficient on the incident microwave power of the MPS with nitrogen and carbon dioxide swirl flow after tuning by using three stub tuner

Results: Visualization of the plasma flame

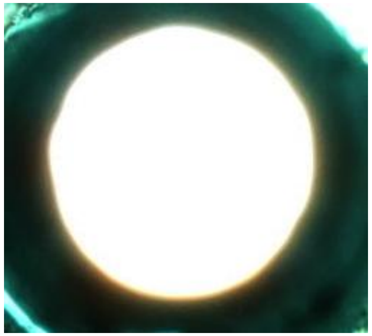


Front view of the methane plasma generated in the waveguide-supplied coaxial-line-based MPS at different flow rates and absorbed microwave powers

Bottom view of the plasma :

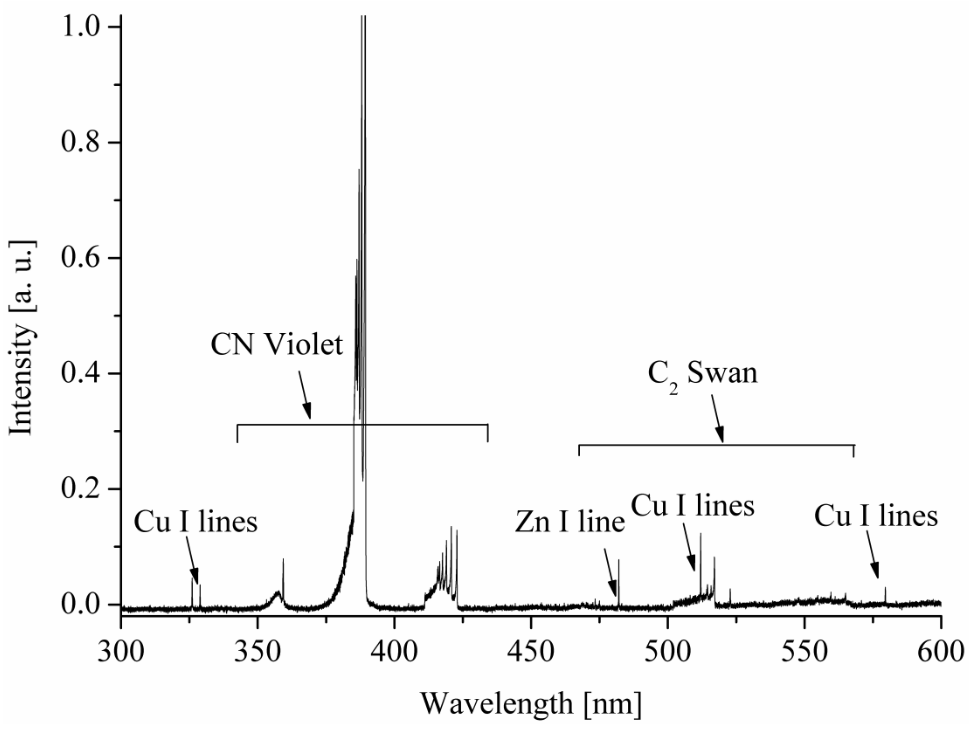


in the waveguide-supplied coaxial-line MPS
CH_4 -100 l/min (axial), N_2 – 50 l/min (swirl)
P_A = 3500 W



in the waveguide-supplied metal-cylinder MPS
CH_4 - 50 l/min (swirl), CO_2 - 50 l/min (swirl)
P_A = 2800 W

Results: Optical Emission Spectroscopy (OES)



Parameters:

spectrometer type: DK-480 (CVI)

spectrometer: grating1200 gr/mm, 3600 gr/mm

CCD camera type: SBIG ST-6, 750 × 242

quartz lens: 50 mm in diameter, focal length – 75 mm

width of the entrance slit of the spectrometer: 50 μm

height of the entrance slit of the spectrometer: 20 mm

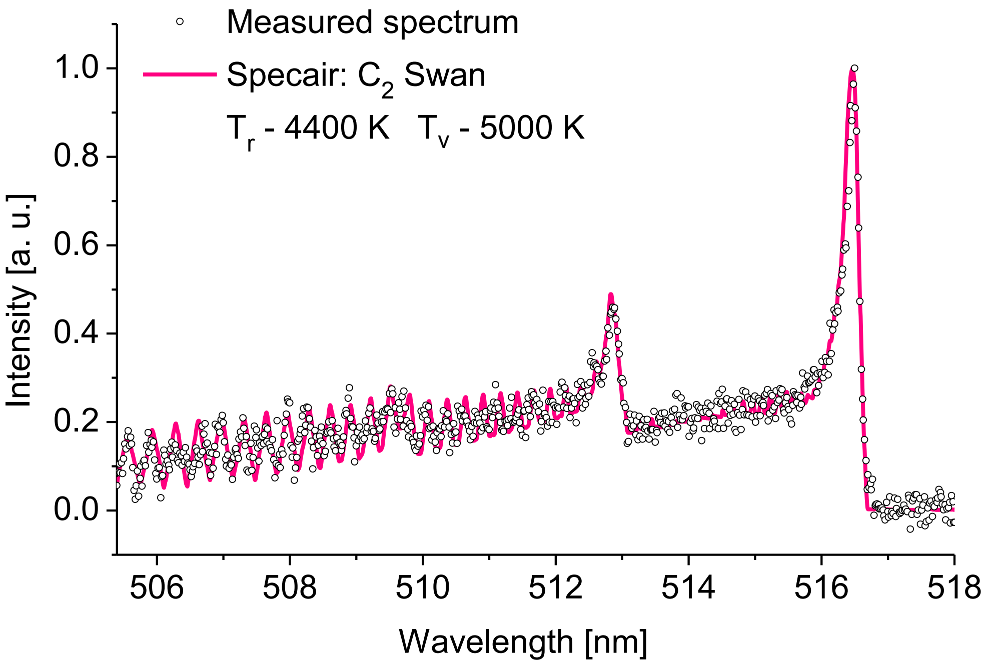
opaque screens pinholes diameter: 1 mm

C_2 Swan system ($A^3\Pi \rightarrow X^3\Pi$, 506–518 nm band)

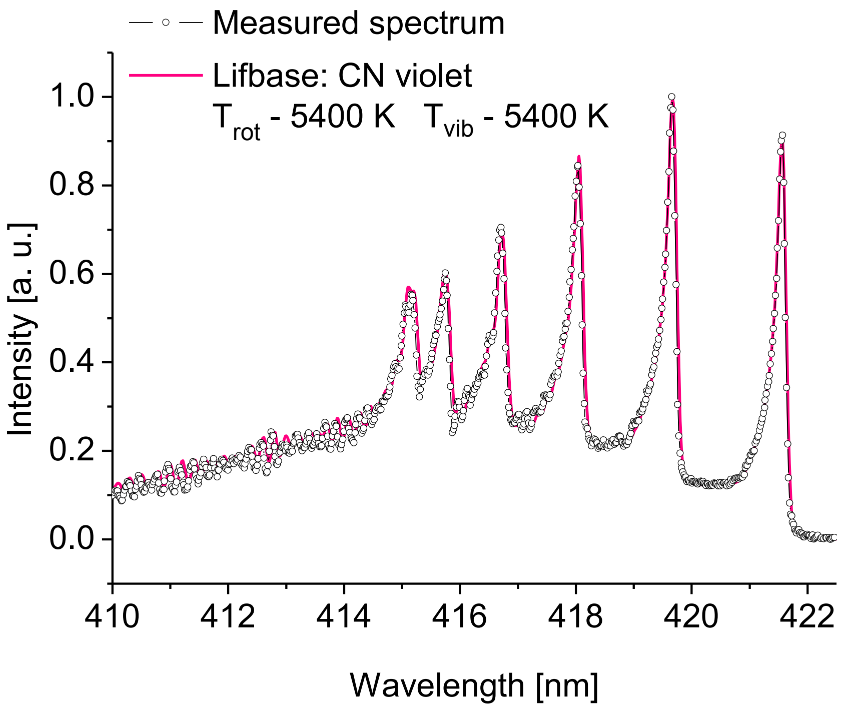
CN violet system ($B^2\Sigma \rightarrow X^2\Sigma$, 410–422 nm band)

Measured emission spectrum of methane plasma (absorbed microwave power P_A =3 kW, methane flow rate - 50 l/min, 15 mm below the inner electrode end)

Results: Optical Emission Spectroscopy (OES)

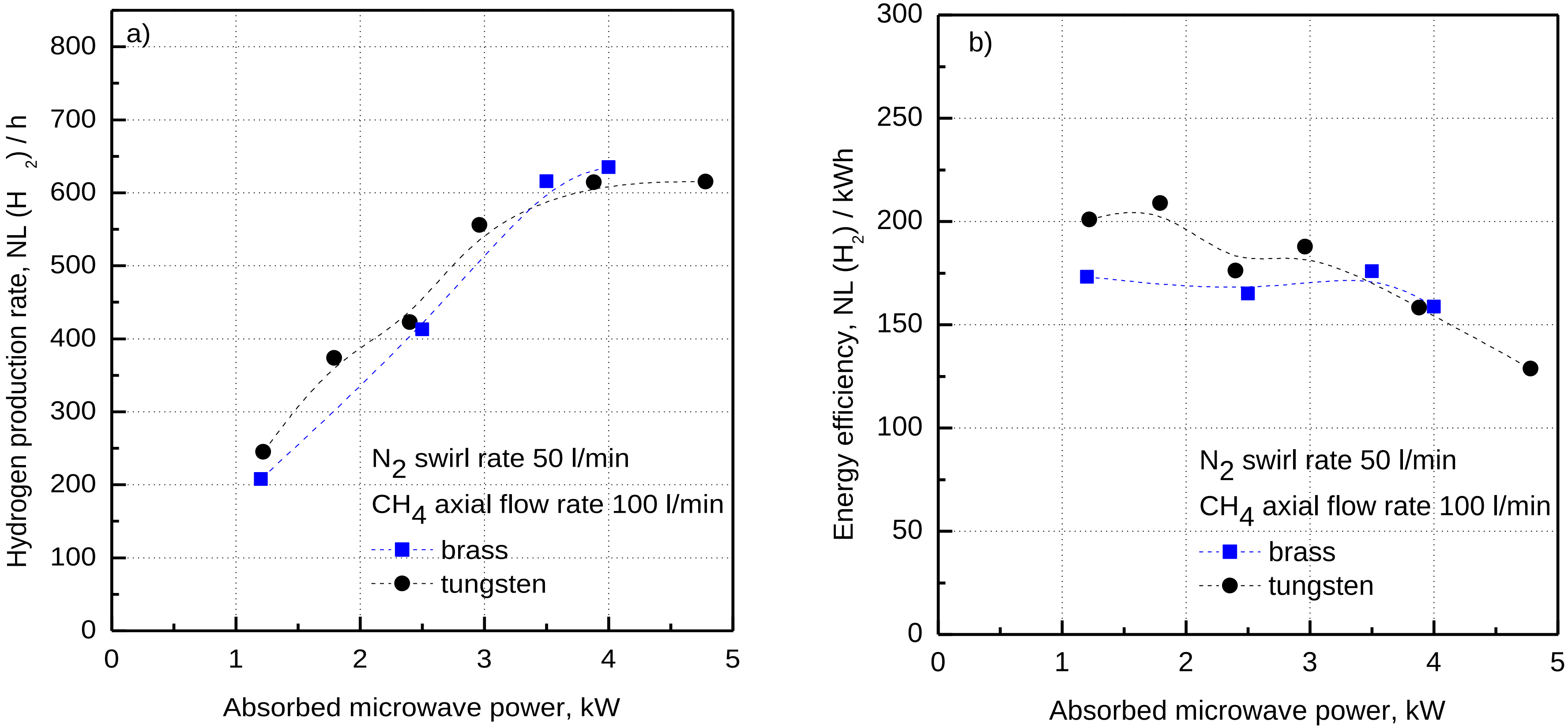


Comparison of the measured and simulated emission spectra of C_2 Swan band in methane plasma (absorbed microwave power P_A = 3 kW, methane flow rate - 88 l/min, 15 mm below the electrode end)



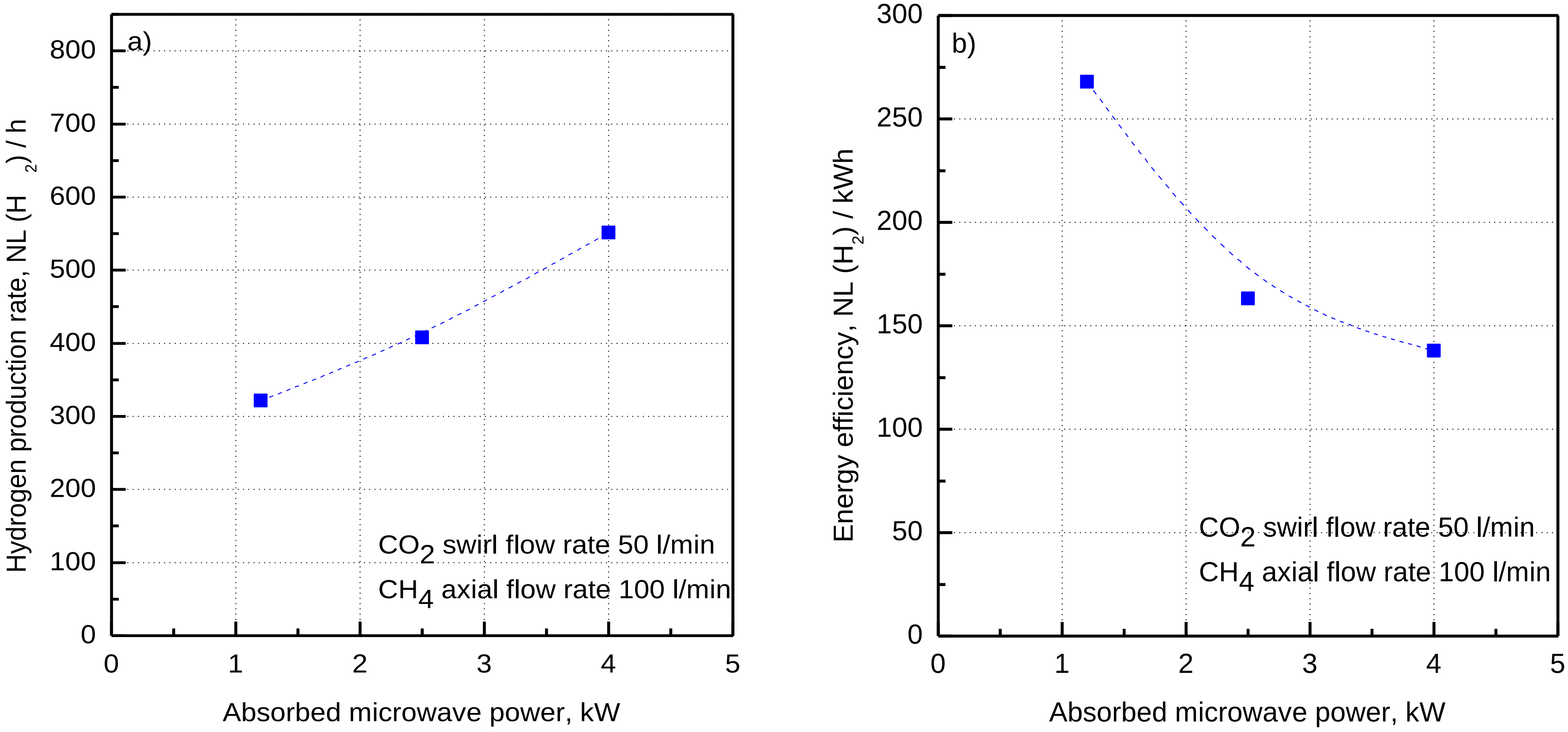
Comparison of the measured and simulated emission spectra of CN violet band in methane plasma (absorbed microwave power P_A = 2 kW, methane flow rate – 88 l/min, 15 mm below the electrode end)

Results: Hydrogen production using coaxial-line-based MPS



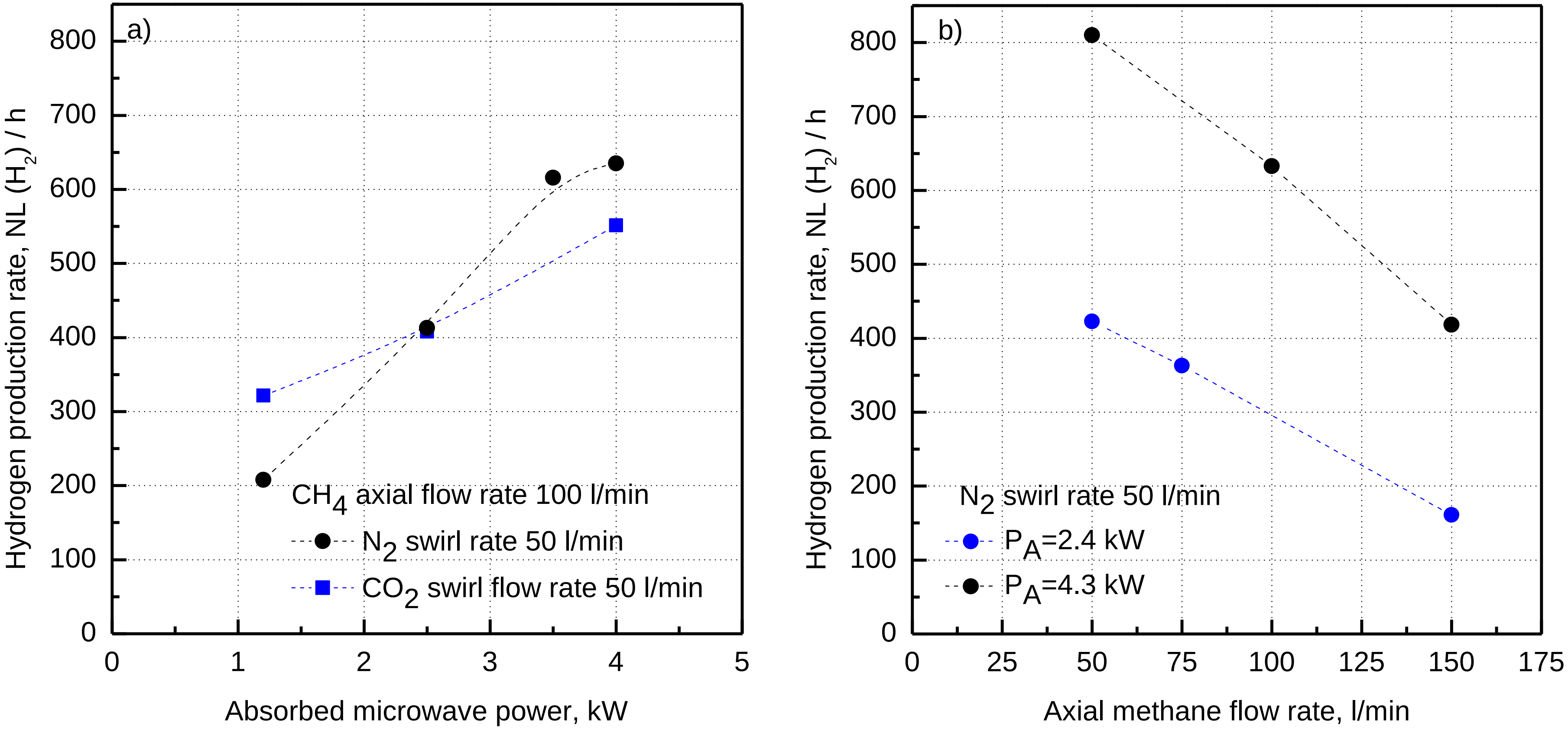
Hydrogen production rate (a) and energy efficiency of hydrogen production (b) as a function of absorbed microwave power. The waveguide-supplied coaxial-line-based MPS with nitrogen swirl flow and inner cylindrical electrode end part made of brass and tungsten.

Results: Hydrogen production using coaxial-line-based MPS



Hydrogen production rate (a) and energy efficiency of hydrogen production (b) as a function of absorbed microwave power. The waveguide-supplied coaxial-line-based MPS with carbon dioxide swirl flow and inner cylindrical electrode end part made of brass.

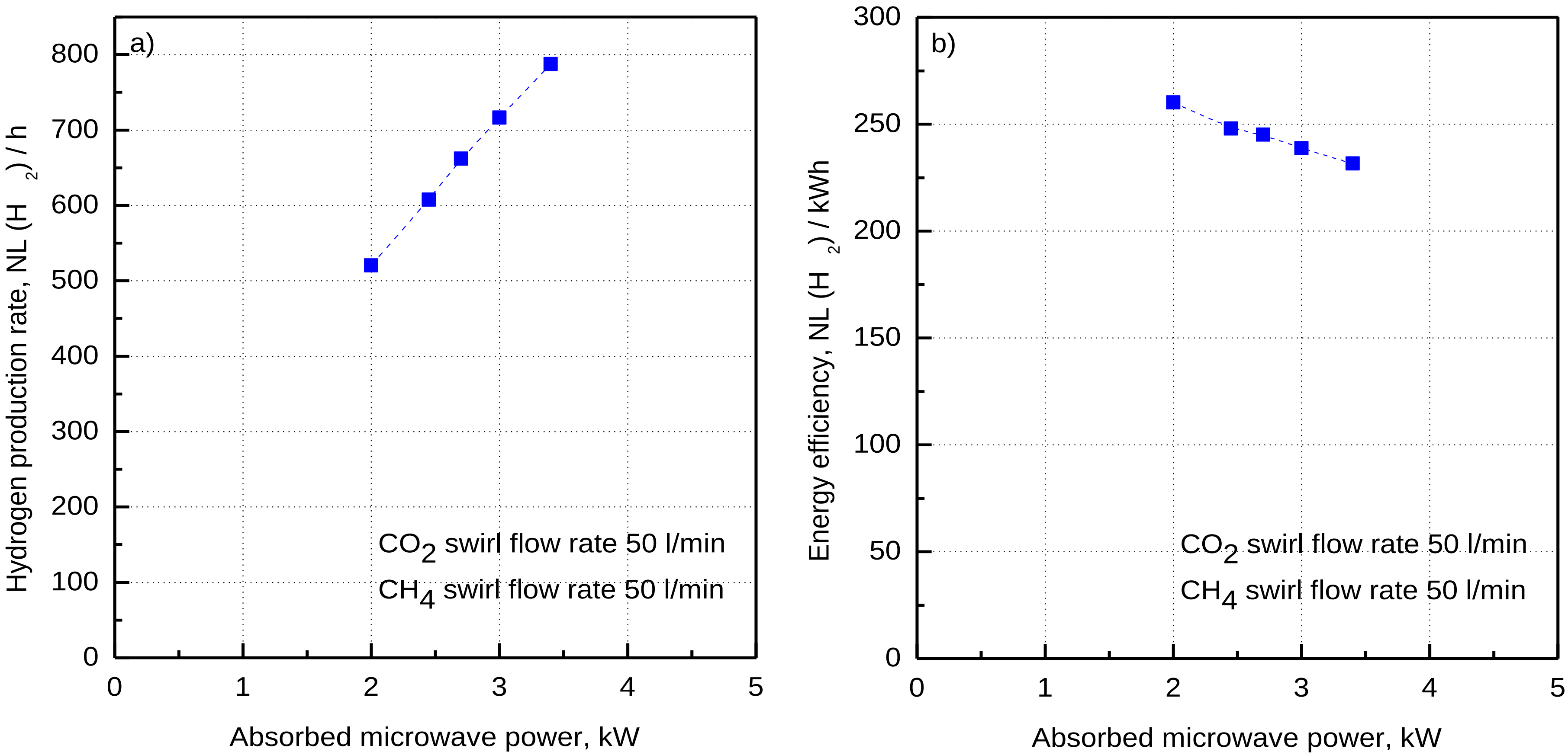
Results: Hydrogen production using coaxial-line-based MPS



Comparison of the hydrogen production rate in the case of the waveguide-supplied coaxial-line-based MPS with nitrogen and carbon dioxide swirl flow. The inner cylindrical electrode end part made of brass.

The hydrogen production rate as a function of axial methane flow rate for two values of absorbed microwave power. The MPS with nitrogen swirl flow and inner cylindrical electrode end part made of tungsten.

Results: Hydrogen production using metal-cylinder-based MPS



Hydrogen production rate (a) and energy efficiency of hydrogen production (b) as a function of absorbed microwave power in the case of the waveguide-supplied metal-cylinder-based MPS

Summary and conclusions

- The investigated MPS (coaxial-line-based and metal-cylinder-based) can be operated with a good power efficiency and stability

- The MPS can be operated in different gases like nitrogen, air, carbon dioxide, methane with microwave power of a few kW with high gas flow rates

- The spectroscopic measurements proved the high gas temperature (4000-6000 K) what makes the MPS an attractive tool for hydrogen production via hydrocarbon conversion

- Results concerning conversion of methane give the hydrogen production rate and energy efficiency up to:
  - 800 NL[H_2]/h and 250 NL[H_2]/kWh

- The obtained experimental results allow to conclude that our MPS had a high potential for hydrogen production via hydrocarbon conversion

- Further test are under preparation

Thank you for your attention

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Acknowledgment

This research was supported by The National Center for Research and Development under the program NR14-0091-10 /2010 and The Szewalski Institute of Fluid-Flow Machinery, Polish Academy of Sciences under the program IMP PAN O3Z1T1