Plasma temperature diagnostic methods of large size RF APPJ for biomedical application

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Abstract: Main focus of the work is gas temperature characterization of atmospheric pressure plasma jet for safe skin treatment in content of wound healing. Three techniques were used in order to validate correctness of plasma temperature measurements at atmospheric conditions: OES, Rayleigh and Raman scattering spectroscopy. It was shown that gas temperature obtained via OES based on OH rotational temperature (310-350 K depending on power) is in good agreement with both Rayleigh and Raman scattering techniques.

Keywords: APPJ, gas temperature diagnostics, OES

1.Introduction

Atmospheric pressure, radio-frequency driven plasmas are showed to be cold, safe and reproducible, generating active oxygen and nitrogen species and are suitable for surface treatment of heat-sensitive substrates. RF (13.56 MHz) APPJ in co-axial geometry with possibility of aerosol introduction has been designed for biomedical purposes such as plasma assisted wound healing and topical drug introduction. The focus of this work is on testing APPJ applicability for skin treatments in the terms of human-safe gas temperature as a key parameter. For precise temperature measurements three different techniques were applied based on different physical interpretation of gas temperature in plasma, nevertheless application of specific temperature diagnostic method has to be evaluated for atmospheric conditions. Optical emission spectroscopy is fast, easy and cheap, non-invasive technique especially suitable for temperature measurements once aerosol is introduced in plasma jet. OES measurements were based on the fact that rotational temperature of molecules is good represent of translational temperature due to high frequency collisions leading to gas thermalization [1]. Rotational temperatures were determined from partially resolved rotational-vibrational bands OH $A^2\Sigma+ \rightarrow X^2\Pi_i$ (0,0) and N_2 $C^3\Pi_u \rightarrow B^3\Pi_g$ (0,2) both showing Boltzman behavior. In order to validate OES results laser scattering techniques were applied: Rayleigh and Raman scattering spectroscopy, both spatially and temporally resolved.

Intensity of Rayleigh scattered signal is directly proportional to density of heavy species in plasma, and so gas temperature in plasma can be obtained (*Equation 1.*) :

$$I \sim \sum_i \sigma^i n_g^i = \sum_i \sigma^i \frac{p^i}{k_B T_g} \quad (1)$$

Where σ^i is scattering cross-section, n_g^i-density, p^i-partial pressure of species i and T_g gas temperature. However, intensity of scattered light depends on particle pressure. In plasmas operating in ambient air property of plasma flow control has been noted [2], known as plasma actuators thus temperature diagnostic method independent on gas dynamics is desirable in order to prove applicability of Rayleigh spectroscopy in APPJ.

Rotational Raman scattering spectroscopy has been chosen to validate both OES and Rayleigh technique, since it is independent on ambient air entrainment in the jet effluent. Intensity of the emitted line $I_{J\text{-}J'}$ corresponding to rotational transition *J-J'* is directly proportional to density of molecules n_J in initial state J (see *Equation 2.*) what further can be related to rotational temperature if Boltzman distribution of rotational states population is assumed (*Equation 3.*).

$$I_{J-J'} = C n_J \frac{d\sigma_{J-J'}}{d\Omega} \quad (2)$$

$$n_j = \frac{n}{Q}(2J+1)\, g_J\, e^{-\frac{B_v J(J+1)}{kT}} \quad (3)$$

Where C is experimental constant, $\frac{d\sigma_{J-J'}}{d\Omega}$ differential cross section of transition *J-J'*, n density of Raman active scatterers, Q partition function, g_j nuclear spin degeneracy and B_v rotational constant of molecules.

In order to obtain gas temperature, theoretical spectrum of Raman active species in the effluent, N_2 and O_2, was fitted to experimental one with rotational temperature as a fitting parameter for transitions with $\Delta J=\pm 2$.

2. APPJ characterization

Plasma jet operates in stable diffuse mode in range of transferred power 10-30 W in 2-4 SLM of argon thus all temperature measurements were carried out for these conditions. Effluent length of the jet can be controlled in range of 2 mm-5 mm by variation of the transferred RF power as shown in *Figure 1*.

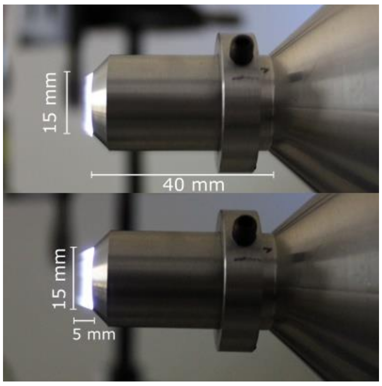

Figure 1. Visual view of effluent in ambient air for 10 W and 30 W of transferred power, respectively

The discharge is found to be capacitive coupled operating in glow regime, sustained by Ohmic electron heating, indicating that main ionization processes are due to fast oscillating electrons following RF field.

3. Temperature measurement.

Emission spectroscopy has an advantage of use for diagnostic purposes once plasma jet is in contact with substrate or liquid, but must be validated for applicability at atmospheric conditions. Space averaged spectra of bands OH $A^2\Sigma+ \rightarrow X^2\Pi_i$ (0,0) and N_2 $C^3\Pi_u \rightarrow B^3\Pi_g$ (0,2) were recorded with 0.05 nm resolution spectrometer in order to resolve rotational structure. OH (A-X) rotational temperature was obtained as a fitting parameter from software LIFBASE, while N_2 (C-B) rotational temperature was estimated from Boltzmann plot slope. For diagnostic purposes 0.05-0.2% of N_2/air was added to gas mixture. LIFBASE simulations show OH rotational temperature in range of 310-350±25 K, independent on input power. The temperature decreases with higher argon flow that is explained by gas flow cooling. Nevertheless Boltzmann plot method applied to nitrogen second positive system gives always 100 K higher value of N_2 rotational temperature in the range of 450-480±25 K. Due to high difference in gas temperature measurements, laser techniques were applied in order to check what method is applicable for conditions of APPJ. For both elastic Rayleigh and inelastic Raman scattering detection pulsed Nd: YAG laser light of 532 nm was utilized with a frequency of 10 Hz, duration of pulse 8 ns and energy per pulse 20-30 mJ. With Rayleigh scattering spectroscopy we were able to space resolve and obtain radial temperature map of the jet 1.5 mm beneath the nozzle. Elastic scattered light 532 nm was recorded with fast imaging camera, with an example of measured signal shown in *Figure 2*. Gas temperature in discharge was based on relative calculation of referent signal intensity and temperature as shown in *Equation 4*.

$$T_g = \frac{I_0}{I_P} T_0 \qquad (4)$$

Where I_0 , I_P and T_0 are respectively intensity of referent signal, intensity of scattered light when plasma is on and referent gas temperature.

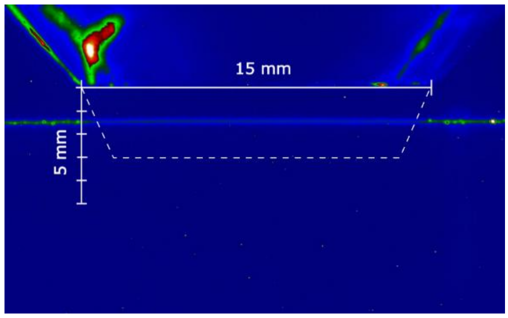

Figure 2.Example of image taken with fast imaging camera 1.5 mm beneath the nozzle

Results of Rayleigh scattering experiments are in good agreement with OH rotational temperature results, radial averaged temperature obtained in pure argon is 300-350±15 K and in mixture Ar +0.05% N_2 is 300-380±15 K depending on power. Gas temperature is almost linearly increasing with RF power. Space resolved measurements of plasma jet temperature gives almost uniform distribution of plasma temperature as presented in *Figure 3*.

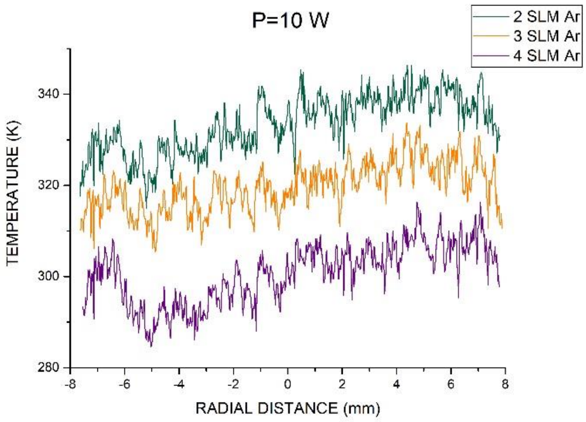

Figure 3. Example of temperature profile of Ar plasma jet generated at the lowest input power of 10 W. Visible slope in temperature profile is due to slight asymmetry of electrodes design.

Considering possible effect of plasma on gas flow dynamics and so on correctness of Rayleigh measurements the rotational Raman spectroscopy was applied to cross-check results of other techniques. Raman spectroscopy was used to measure rotational temperature of Raman active molecules in atmospheric plasma jet, N_2 and O_2. Besides rotational Raman spectrum, Rayleigh scattered light was also appearing so in order to avoid detector and detect 10^4 times lower intensity Raman spectrum, Rayleigh peak should be filtered out [3] that was done with volumetric Bragg grating notch filter. The temperature was set as the fitting parameter for simulated and example of fitting for Stokes component is shown in *Figure 4.*

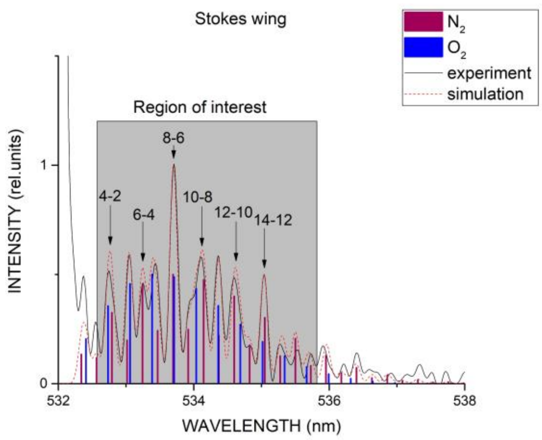

Figure 4. Example of Stokes wing fitting with noted rotational transitions

Raman spectras were recorded for the lowest feeding gas flow where effect of air entrainment and plasma impact on gas flow dynamics should strongest. However, obtained results are in good agreement with both OH rotational spectroscopy and Rayleigh scattering measurements, proving applicability of these methods in terms of atmospheric pressure plasma jets temperature diagnostics.

4.Conclusions

Gas temperature of argon RF APPJ has been measured by the meaning of three different techniques. Optical emission spectroscopy has been used for rotational temperature measurements of species OH and N_2. Bands OH $A^2\Sigma+ \rightarrow X^2\Pi_i$ (0,0) and N_2 $C^3\Pi_u \rightarrow B^3\Pi_g$ (0,2) are utilized for gas temperature measurements, assuming thermalization between rotational and translational temperature in non-equilibrium plasmas. OH rotational temperature was found in the range of 310-350±25 K independent on power and decreasing with increase of the gas flow. Temperature independence on input power can be related to constant energy density and expand of discharge volume with increased power. Rotational temperature based on nitrogen second positive system analysis was 100 K higher in range 450-480±25 K, slightly increasing with transferred power. Laser scattering techniques were used as alternative to OES for improvement of temperature measurements and validation of the emission spectroscopy. Rayleigh scattering spectroscopy gave space resolve temperature map, indicating space averaged temperature close to one obtained from OH rotational spectrum. Gas temperature was in the range 300-350±15 K in case of pure argon as feeding gas and in the range 300-380±15 K in case adding 0.05% of N_2. Finally, gas dynamics independent diagnostics method was carried out to evaluate both OES and Rayleigh method. Rotational Raman spectroscopy has been done for the lowest flow when is assumed that penetration of ambient air into the plasma has impact on temperature measurements. However, no significant difference in results was noticed, proving both OES OH based measurements and Rayleigh scattering spectroscopy as valid tool for cold atmospheric pressure plasma temperature diagnostic.

5.References

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