Pulsed laminar arc jet with synchronized suspension injection-spectroscopic studies

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Abstract. The uncontrolled arc plasma instabilities are one of the difficulties encountered in in suspension plasma spraying. The improvement of this process is usually attempted by means of the reduction of arc fluctuations. The following paper presents a new approach to overcome these arc instabilities. The principle is to produce a pulsed laminar plasma jet combined with phased injection of liquid droplets. This is achieved by the particular design of the plasma torch which allows coupling two modes of the plasma oscillations, restrike and Helmholtz. The plasma produced in a new mode is laminar and pulsed, characterized by a modulated enthalpy, what permits to make the synchronization with the suspension injection. The droplets are injected using a piezoelectric device, based on drop-on-demand method, triggered by the voltage signal. The results are evaluated by time-resolved imaging technique and Time-Resolved Optical Emission Spectroscopy.

1. Introduction
Suspension plasma spraying is a well-established process which allows producing finely structured and dense coatings. However, in spite of the fact that the plasma torch is supplied by a direct current (DC) power source, the plasma jet presents unsteady characteristics. The research studies have verified that this time-fluctuating momentum of the plasma have a strong influence on the droplet fragmentation and vaporization process [1, 2]. It results in a poor reproducibility and reliability of the method, what causes the limited applications of suspension plasma spraying in advanced processing. Therefore, for many years, the special efforts have been devoted to understand the arc behavior in dc plasma torch. One of the sources of the plasma instabilities is the ‘stick and slip’ motion of the arc inside the nozzle [3-4]. This kind of oscillations, known as ‘restrick’ mode, gives an irregular voltage signal approximately saw-tooth in shape. The other major source of the plasma fluctuations has been more recently identified in conventional plasma torches. Delair et al. have suggested that Helmholtz oscillation in the arc chamber can be the reason for high frequency fluctuations of the arc voltage [5]. Rat and Coudert have shown that the plasma instabilities are related to compressibility effects of plasma forming gas in the cathode cavity [6, 7]. These instabilities appear in power spectrum of the arc voltage as a strong, sharp peak (2–5 kHz), and have been referred to Helmholtz mode. The following work highlights the possibility to control the instabilities of plasma. The arc jet working in a pulsed laminar regime, obtained by coupling Helmholtz and restrick modes in a mechanism of phase-locked loop, is presented. This pulsed plasma, provided the use of synchronous injection, is expected

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to improve the control of dynamic and thermal interaction between the plasma and injected material. The synchronization of suspension injection with the plasma is possible by the use of an innovative injection system based on drop-on-demand (DOD) technique. The results obtained by this new suspension plasma spraying system are evaluated by time-resolved imaging technique and Time-Resolved Optical Emission Spectroscopy.

2. Experimental setup
As has been highlighted above, in the plasma jet generated by dc torch the oscillations modes, restrike and Helmholtz, have been identified. The following paper shows the possibility of coupling these two modes to obtain pulsed laminar arc plasma jet [8, 9]. It is possible to obtain by a special torch characterized by a larger cathode cavity \((V_g=17.8 \text{ cm}^3)\). The study of Helmholtz mode shows that the resonance frequency, \(f_H\), linked to this oscillation, depends on: \(f_H = 1/2 \pi \left( \gamma_g p_g/\rho_p \right) \left( S/L_p \cdot V_g \right) \), where: \(\gamma_g\), \(p_g\) are, respectively, the isentropic coefficient of the cold gas, the mean pressure in cathode cavity, \(\rho_p\) is the plasma density, \(S\), \(L_p\) and \(V_g\) are, successively, the cross section area, the length of the nozzle channel and the volume of the cathode cavity. According to the above equation, the enlargement of the cathode cavity reinforces the Q factor, which is linked to the band pass of resonator, \(\Delta f\), and to the damping factor, \(\xi\), as follows: \(Q = f_H/\Delta f = 1/2\xi\), what decreases the Helmholtz mode frequency. Nitrogen is used as plasma gas with mean mass flow rates, \(\dot{m}\), between 0.042 and 0.104 g.s\(^{-1}\) and different nozzles are tested with channel diameters, \(d\), of 2.5, 3, 3.5 and 4 mm. The regulation of the above mentioned parameters and the arc current results in a new oscillation mode, called mosquito mode because of the sound produced by a torch that recalled the flight of a mosquito, characterized by a very regular voltage signal, presented in figure 1.

![Figure 1. Arc voltage signal for: \(d = 4 \text{ mm}, I = 14.9 \text{ A}, N_2: 0.042 \text{ g.s}^{-1}, \overline{V} = 74 \text{ V}\).](image)

The measurements of the heat losses, \(Q_{\text{loss}}\), to the electrodes and the electric power supplied to the torch, \(P_{\text{elec}}\), permitted to determine the specific enthalpy of the plasma by the equation: \(h = (P_{\text{elec}} - Q_{\text{loss}}) / \dot{m}\). The thermal losses are proportional to the arc current \(I\), so that the ratio \((Q_{\text{loss}}/I)\), that is homogeneous to a voltage, \(V_{\text{elec}}\), represented about 35 V. That means that the recorded voltage, \(V(t)\) must be reduced by 35 V before the conversion into time dependent specific enthalpy, following the relationship, \(h(t) = (V(t) - V_{\text{th}}) / \dot{m}\). Then it can be predicted that the specific enthalpy in the mosquito mode is modulated in the proportion, \(h_{\text{max}}/h_{\text{min}}\), up to 16, with a mean value of 13.3 MJ.kg\(^{-1}\). That last value is comparable to what is currently obtained for conventional plasma spraying torches. Figure 2 presents the suspension plasma spraying system used in the experiment. It consists of a new home-made torch, described above, synchronized with an external injector based on the drop-on-demand (DOD) method (Ceradrop, Limoges, France).
The aqueous suspension, stored in a buffer tank and composed of TiO$_2$ crystallites (42 wt.%), is injected into the plasma jet using a piezoelectric head actuated by voltage pulses. This piezoelectric injector allows obtaining a single calibrated droplet with a diameter of 50 µm and a velocity between 2 and 10 m.s$^{-1}$. The principle of synchronous injection with the pulsed plasma jet consists in the insertion of the suspension droplet at the chosen moment of the periodic plasma jet oscillations, providing that the injection frequency is equal to that of the Helmholtz resonant mode. The emission of each droplet is triggered from sampling of the torch voltage after an adjustable time delay, $\tau$. The time-resolved imaging system allows to observe the interaction between the plasma and the suspension droplet. It is composed of a fast shutter camera (PCO, Kelheim, Germany), characterized by high resolution (1392 x 1040 pixels), coupled with a laser (HiWatch, Oseir, Tampere, Finland). The pulsed laminar arc jet is diagnosed by the Time-Resolved Optical Emission Spectroscopy (TROES) which consists of an IsoPlane spectrograph (Princeton Instruments, Trenton, New Jersey) equipped with 1200 g/mm gratings. The spectra acquisition is realized by using a PI-MAX4 ICCD camera (Princeton Instruments) connected to the PC and controlled by LiathField software. Both the time-resolved imaging system and TROES are synchronized with the injection by the synchronization box.

Two different moments, $\tau_1$ and $\tau_2$ presented in figure 3, of the periodic arc voltage have been chosen to trigger the suspension injection, the camera, laser and spectroscopy recording.

3. Results
The synchronization of the time-resolved imaging system with the arc voltage signal allows to observe the plasma jet in two triggering moments, shown in figure 3. Figure 4 presents the photo of a pulsed plasma jet taken in: a) $\tau_1$ moment at the arc voltage level of 60 V, characterized by a low specific enthalpy, b) $\tau_2$ moment characterized by the highest level of the enthalpy, corresponded to the
maximum voltage level of 100 V. These following results show the effect of the coupling restrike and Helmholtz modes- periodic modulated plasma jet.

Figure 4. Pulsed arc jet with: a) low level of local specific enthalpy, corresponded to the arc voltage level of 60 V, b) high level, corresponded to 100 V.

To better analysis the periodicity of this new pulsed plasma jet and the modulation of the specific enthalpy the spectroscopic measurements have been carried out for the two moments of the plasma jet presented in figure 4. The temperature of the plasma flow is commonly determined by the rotational lines of the first negative system \( B^2\Sigma_u^+ \rightarrow X^2\Sigma_g^- \) [10, 11]. Therefore, the emission spectrum of the first negative system of the \( \text{N}_2^+ \) molecule have been measured at a distance of 1 mm from the nozzle. Figure 5 presents the spectrum of the nitrogen plasma obtained in the triggering moment \( \tau_2 \). The spectroscopy system has been calibrated, both in the intensity and wavelength axis.

Figure 5. Comparison of the measured and simulated by Specair software (N\(_2^+\)) first negative spectra.

The rotational temperature, \( T_{rot} \), has been calculated by using Specair software, which allows comparing the experimental and simulated spectra. Bruggeman et al. have shown that the rotational temperature is strongly determined by the slit function [12]. Therefore, to ameliorate the accuracy, this instrument function has been carefully chosen. It has been determined by recording the line profile of the emission from a low-pressure Hg lamp at the wavelength of 404.7 nm.
The measured spectrum has been fitted to the theoretical one, what is presented in figure 5. Well-fitted rotational structures permit to determine the rotational temperature with good precision.

Figure 6. \((\mathrm{N}_2^+)\) first negative spectra for two different moments, \(\tau_1\) and \(\tau_2\) of the periodic arc voltage.

Figure 6 presents the molecular emission spectra of the first negative system of the \(\mathrm{N}_2^+\) molecule of the nitrogen plasma obtained at the moments, \(\tau_1\) and \(\tau_2\), of the periodic arc voltage. Analyzing the \(\mathrm{N}_2^+\) molecule emission at 391 nm, it can be stated that the spectrum measured at the moment \(\tau_2\), corresponded to the maximum voltage level of 100 V, is characterized by the higher value of the intensity than the spectrum detected in the moment \(\tau_1\). The rotational temperature determined for the plasma measurement at the moment \(\tau_2\) is much higher and equals to 7500 ± 150 K. The spectroscopic estimation of the rotational temperature of the nitrogen plasma permits to obtain the values of the specific enthalpy of the plasma jet measured at different moments, \(\tau_1\) and \(\tau_2\).

Figure 7. The nitrogen specific enthalpy [13].

The comparison of the obtained results with the nitrogen enthalpy graph, presented in figure 7, shows that the plasma jet analyzed at the moment \(\tau_2\) is characterized by the enthalpy value of around 35 ± 3 MJ/kg and at the moment \(\tau_1\) by the enthalpy of 15 ± 2 MJ/kg. Figure 8 presents the material injection to the pulsed plasma jet.
Figure 8. Influence of the local instantaneous specific enthalpy on droplet thermal treatment: a) high level of local specific enthalpy, b) low level.

In figure 8 a) the droplet enters the plasma 4 mm downstream of the nozzle exit, at the moment $\tau_2$ characterized by a high level of specific enthalpy, around 35 MJ/kg. The almost immediate vaporization process of the droplet has been observed. Another situation is shown in figure 8 b). The droplet is injected into the plasma with a medium level of specific enthalpy. At that time the vaporization-seeding process does not concern this droplet but that one which was injected one period earlier. These two pictures are observed through a narrow band-pass filter centered on the laser wavelength. The emission wavelength of the laser is in the range of 801 ± 2 nm. Therefore, the interferential filter of 801 nm has been applied. This configuration permits to eliminate on the image the light coming from the pure nitrogen plasma, compared to figure 4. Only the particles that reflected the laser light are seen, together with the light ball that resulted of the interaction of the nitrogen plasma with the material contained in the droplet that produced a strong increase of the brightness and an enrichment of the spectrum.

4. Conclusions
A new approach to overcome the plasma instabilities has been presented, obtained by a phase locking Helmholtz oscillation and rearcing events in the nozzle. The torch works in a new resonant mode, called mosquito mode, characterized by very regular, saw-tooth shaped arc voltage signal. The plasma produced by this new mode is periodic, pulsed with the enthalpy highly modulated ($h_{\text{max}}/h_{\text{min}}$=13). This periodicity has been investigated by using Time-Resolved Optical Emission Spectroscopy synchronized with the arc voltage signal. The spectroscopic diagnostics allow to determine the rotational temperature of the pulsed nitrogen plasma and demonstrate the significant modulation of the plasma specific enthalpy. The modulation of the arc voltage has been used to synchronize the plasma with the injection system, based on drop-on-demand method, which enables to obtain the suspension injection in a chosen moment. The interaction between the periodic plasma flow and the droplets has been examined by the time resolved imaging triggered by arc voltage signal and synchronized with the suspension injection. The thermal treatment of injected material is shown to be very sensitive on the moment at which the droplet penetrates the plasma. This type of synchronous injection with a periodic pulsed arc jet is supposed to control the dynamic and thermal interaction between plasma and material.

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6. References
[1] Vardelle A, Chazelas C, Marchand C and Mariaux G 2008 Pure Appl. Chem. 80 1981-1991
[2] Etchart-Salas R, Rat V, Coudert J-F, Fauchais P, Caron N, Wittman K and Alexandre S 2007 J. Therm. Spray Techn. 16 857-865
[3] Duan Z and Heberlein H 2002 J. Therm. Spray Techn. 11 45-51
[4] Rat V and Coudert J-F 2008 J. Phys. D: Appl. Phys. 4 205208
[5] Delair L, Tu X, Bultel A and Cheron B G 2005 High Temp. Mater. Processes 9 583-597

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[6] Rat V and Coudert J-F 2010 J. Phys. D: Appl. Phys. 108 043304
[7] Rat V and Coudert J-F 2010 Appl. Phys. Lett. 96 101503
[8] Krowka J, Rat V and Coudert J F 2013 J. Phys. D: Appl. Phys. 46 224018
[9] Krowka J, Rat V, Goutier S and Coudert J F 2014 J. Therm. Spray Techn. 23 786-794
[10] Laux C O, Gessman R J, Kruger C H, Roux F, Michaud F and Davis S.P. 2001 J. Quant. Spectrosc. Radiat. Transfer 68 473-482
[11] Parigger C, Plemmons D H, Hornkohl J O and Lewis J W L 1995 Appl. Opt. 34 3331-5
[12] Bruggeman P J, Sadeghi N, Schram D C and Linss V 2014 Plasma Sources Sci. Technol. 23 023001
[13] Boulos M I, Fauchais P and Pfender E 1994 Thermal plasmas: fundamentals and applications, Volume 1 (New York: Plenum Press)