# Dissociation degree of $N_2$ in dc discharges from Schlieren measurements.

G. Cicala [1], A. C. Rainò[2] and A. Boggia[2]

[1] IMIP-CNR (Istituto di Metodologie Inorganiche e dei Plasmi, Consiglio Nazionale delle Ricerche), Via Amendola 122/D, 70126 Bari, Italy

[2]Dipartimento di Fisica/INFN Sezione di Bari, University of Bari, Via E. Orabona 4, 70126 Bari, Italy

Acoustic shock waves generated *in-situ* by a spark gap are propagated in weakly ionized plasmas of atomic Ar or molecular $N_2$ gas, at low pressure of 3 Torr. The $N_2$ dissociation percentage in dc discharge is determined by Schlieren method combined with power measurements and calculation of gas temperatures and enthalpies and it is found to be of 0.25 %. This value is in good agreement with the experimental results of mass spectrometry in discharge and of electron paramagnetic resonance or nitrogen atom titration in post-discharge as reported by some authors in similar dc discharge conditions.

## 1. Introduction

The dissociation of $N_2$ in dc discharges has been extensively investigated from both experimental [1-3] and theoretical [4,5] viewpoints. The nitrogen atom (N) concentration has been measured in discharge and/or in post-discharge regions. Experimentally, many efforts have been focused on the quantitative measurements of N atoms in the discharge by mass spectrometry [2] and in the after-glow regions by titration with NO gas [1, 2], or by the Electron Paramagnetic Resonance (EPR) [3]. The authors of ref. 2 have found, in active positive column at 3 Torr, an increase of dissociation percentage (d %) from 0.12 to 0.41 %, whereas in after-glow from 0.11 to 0.31 % by increasing the current from 10 to 80 mA.

This paper deals with shock waves (SW) generated *in-situ* by a spark gap and propagated either in atomic argon or molecular nitrogen at dc discharge *off* and *on*. The aim of this paper is the determination of the $N_2$ dissociation percentage on the basis of: 1) the propagation SW velocity in Ar and $N_2$ measured by laser deflection technique and 2) the power of Ar and $N_2$ discharges determined by electrical measurements. A joint analysis of experimental (such as local SW velocity and dc power) and calculated results (such as gas temperature [6] and enthalpy [7]) permits the determination of the $N_2$ dissociation, by utilizing the Ar plasma as a standard for this new proposed method.

## 2. Experiment

The experimental apparatus has been reported elsewhere [8]. It comprises a pyrex tube (80 cm long and 4.3 cm inner diameter), in which a non equilibrium dc discharge has been ignited by a dc supplier (Alintel SHV 1000, 200mA/5kV), between a pair of annular electrodes 38.5 and 58.5 cm, respectively, far from the spark gap. The discharge current has been changed in the range between 10 and 80 mA. The power has been evaluated by the discharge voltage measured by a voltage divider and the discharge current. Acoustic shock waves have been produced by a spark gap (located at one end of the pyrex tube) driven by a high voltage capacitor (0.5μF, 12.5kV), a triggered home-made spark switch and a dc supplier (50 kV). The Ar and $N_2$ amount has been fixed at 200 sccm by independent MKS gas flowmeters, by flowing in opposite direction to that of shock wave propagation, and the pressure has been maintained constant at 3 Torr.

The laser deflection measurements have been carried out by an optical deflectometer based on the Schlieren principle i.e. on the density gradient [9] induced by the passage of a shock wave transverse to a beam laser path. In the present study the Schlieren optical system consists of two He-Ne lasers (Melles Griot Mod.25-LHR-991) L1 and L2, two amplified Silicon detectors (Thorlabs PDA155-EC) PD1 and PD2, an oscilloscope (Tektronix Mod.TDS2014) and a computer. The two laser beams explore two distinct zones (41 and 51 cm far from the spark gap) of the dc discharge.

## 3. Results and Discussion

The discharge power values, estimated by the discharge voltage and current measurements, are reported as a function of discharge current in Fig. 1. The discharge power increases by increasing the current and in the entire range of current the power of Ar discharge is always found to be lower than that of $N_2$, as shown in Fig. 1. Thus, the Ar discharges are less energetic than $N_2$ ones.

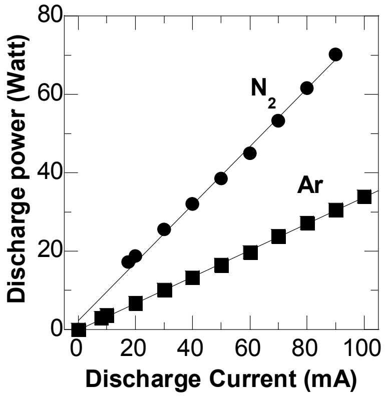


Fig. 1 Power of $N_2$ and Ar plasmas _vs_ discharge current.

The local SW velocities, estimated by dividing the distance (10 cm) between the two laser beams by the difference of the SW arrival times measured by the corresponding Schlieren signals [10], are reported in Fig. 2 versus discharge current. The velocity of shock wave in $N_2$ and Ar plasmas increases by increasing the discharge current, as already observed by many authors [11-13].

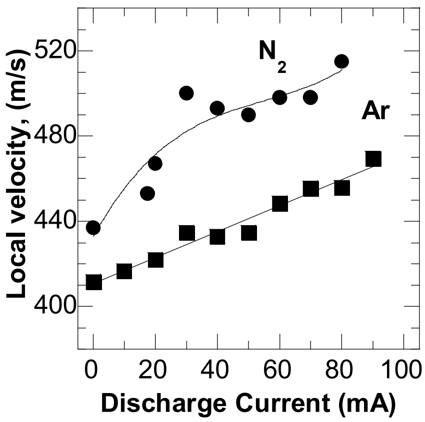


Fig.2 Local velocity of shock wave propagating in $N_2$ and Ar plasmas _vs_ discharge current.

The local SW velocities in ionized $N_2$ gas are higher than those in ionized Ar because the $N_2$ plasmas are more energetic than Ar ones.

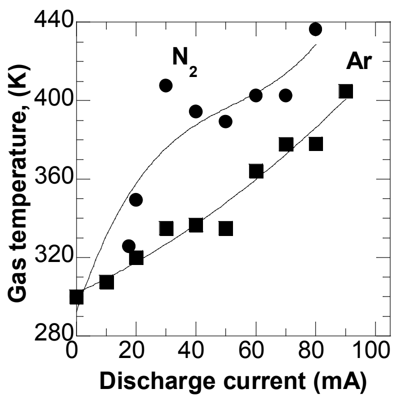


Fig. 3 Gas temperature in $N_2$ and Ar plasmas _vs_ discharge current.

The gas temperatures, calculated from the following equation [6]:

$$\frac{2}{\gamma_1+1}\cdot M_1\cdot(1-\frac{1}{M_1^2})+\frac{2}{\gamma_1-1}\cdot\left\{\frac{\left[2\gamma_1 M_1^2-(\gamma_1-1)\right]\cdot\left[(\gamma_1-1)M_1^2+2\right]}{(\gamma_1+1)^2\cdot M_1^2}\right\}^{1/2}\cdot$$

$$\cdot\left\{1-\left[\frac{\frac{2\gamma_2}{\gamma_2+1}M_2^2-\frac{\gamma_2-1}{\gamma_2+1}}{\frac{2\gamma_1}{\gamma_1+1}M_1^2-\frac{\gamma_1-1}{\gamma_1+1}}\right]^{\frac{\gamma_1-1}{2\gamma_1}}\right\}=\frac{2}{\gamma_2+1}\sqrt{\frac{T_2}{T_1}}M_2\left(1-\frac{1}{M_2^2}\right) \quad (1)$$

and from the condition that the local SW velocity ($v_{SW}$) of $N_2$ and Ar is given by:

$$v_{SW}=M_2\cdot v_{sound}\cdot\left(\frac{T_2}{T_1}\right)^{1/2} \quad (2)$$

where:

$M_1$ and $M_2$ are the Mach numbers at temperatures $T_1$ and $T_2$,
$T_1$ is 300 K the initial (ambient) gas temperature,
$T_2$ is the gas temperature dependent on the discharge current,
$\gamma_1$ and $\gamma_2$ equal to $\frac{N+2}{N}$, with N the number of freedom degrees,
$v_{sound}$ is the sound velocity,

are reported in Fig. 3 as a function of discharge current. It shows that the gas temperatures increase by increasing the discharge current and in $N_2$ the gas temperature reaches values higher than that in Ar.

The thermodynamic equations of ref. [7] are useful to calculate the enthalpy of atomic Ar ($H_{Ar}$) and molecular nitrogen ($H_{N2}$).

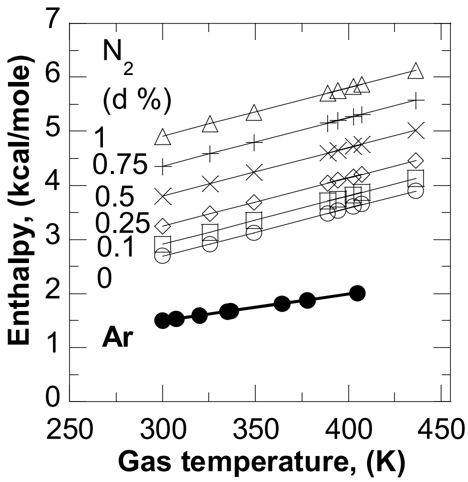


Fig. 4 Enthalpy of Ar and $N_2$ at variable dissociation percentage, (d %) _vs_ gas temperature.

The enthalpy trend of Ar and those of $N_2$ at variable dissociation percentage are reported in Fig. 4 as a function of Ar and $N_2$ gas temperature, respectively. The molecular nitrogen enthalpy is higher than that of atomic argon and it becomes much higher with increasing its dissociation percentage.

Figure 5 shows the profiles of $N_2$ enthalpy at variable dissociation percentage (from 0 to 1 %) and

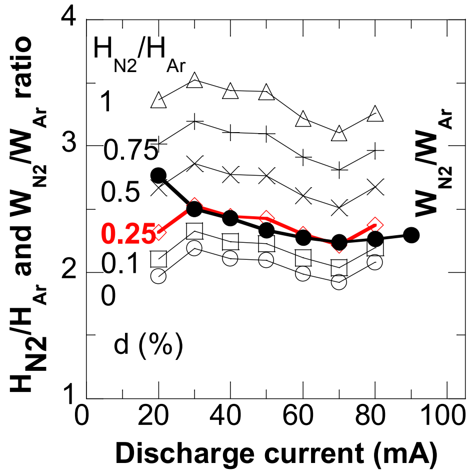


Fig. 5 Enthalpy ($H_{N2}/H_{Ar}$) ratio at variable dissociation percentage, (d %) of $N_2$ and power ($W_{N2}/W_{Ar}$) ratio *vs* discharge current.

the $N_2$ power (both normalized by the enthalpy and power, respectively, of atomic inert Ar utilized as a standard) against discharge current.

The match between the values of power ratio ($W_{N2}/W_{Ar}$) and the values of enthalpy ratio ($H_{N2}/H_{Ar}$) is established only if some energy of the $N_2$ discharge is utilized for its dissociation, found to be of 0.25%. Thus, the above-described method represents a way to evaluate the dissociation degree of $N_2$ by employing Schlieren measurements. Additionally, the value 0.25% is in good agreement with that determined in discharge by mass spectrometry [2] and in post-discharge by electron spin resonance [3] and/or N atom titration [2].

## 4. Conclusions

$N_2$ plasma is found to be more energetic than that of Ar. This is caused by the molecular nature of the nitrogen which strongly affects the plasma parameters such as discharge voltage and discharge power (see Fig. 1). In fact, in $N_2$ the surplus energy is distributed in vibrational freedom degree and utilized for a dissociation degree of 0.25%, as determined by the simple, *in-situ* and non invasive Schlieren method combined with dc power measurements and the calculation of gas temperatures and enthalpies.


## Acknowledgement

The authors would like to thank Prof. M. Capitelli for helpful discussions.


## References


[1] I. M. Campbell, B. A. Thrush, Proc.R. Soc: A 296 (1967) 201.
[2] S. Kumar, P.K. Ghosh, J. Phys. D: Appl. Phys. **26** (1993) 1419.
[3] I. N. Brovikova, E. G. Galiaskarov, High Temp. **39** (2001) 809.
[4] V. Guerra, E. Galiaskarov, J. Loureiro, Chem. Phys. Lett. **371** (2003) 576.
[5] M. Capitelli, M. Dilonardo, Chem. Phys. **24** (1977) 417.
[6] A.F. Aleksandrov, N. G. Vidyakin, V. A. Lakutin, M. G. Skvortsov, I. B. Timofeev, and V. A. Chernikov, Sov. Phys. Tech. Phys **31** (1986) 468.
[7] M. Capitelli, E. Molinari, J. Plasma Phys. **4** (1970) 335.
[8] G. Cicala, A. Russo, M. Capitelli, O. De Pascale, S. Longo, D. Bruno, P. Minelli, A. Boggia, A. C. Rainò, V. Nassisi, XVIII ESCAMPIG, Lecce (Italy), Vol. 30, 207, 12-16 July 2006, published by European Phys. Soc.
[9] L. Syrova, Meas. Sci. Rev. **1** (2001) 107.
[10] G. Cicala, A. Russo, M. Capitelli, O. De Pascale, S. Longo, D. Bruno, P. Minelli, A. Boggia, A. C. Rainò, V. Nassisi, XXV International Symposium on Rarefied Gas Dynamics, Saint-Petersburg (Russia) 21-28 July 2006, p. 46.
[11] P. Bletzinger and B. N. Ganguly, Physics Lett.A. **258** (1999) 342.
[12] A. R. White, V. V. Subramaniam, J. Thermophys. & Heat Transfer **15** (2001) 491.
[13] Y. Z. Ionikh, N. V. Chernysheva, A. V. Meshchanov, A. P. Yalin and R. B. Miles, Phys.Lett. A. **259** (1999) 387.