*DOE Plasma Science Center*

**Visiting Graduate Student/Post-Doctoral Researcher Fellowship Report**

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| **Title of Project:** | Detection of O, N, and NO produced by an atmospheric pressure plasma jet by laser induced fluorescence | |
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| **Institution Visited:** | The Ohio State University | |
| **Hosts:** | Profs. Walter Lempert and Igor Adamovich | |
| **Host email:** | Lempert.1@osu.edu | |
| **Dates of Visit:** | **Start: 31 July 2013** | **End: 8 September 2013** |

## I. Description and Importance of Research Issues Investigated During Visit

The atmospheric pressure plasma jet used in Prof. Oehrlein's group (University of Maryland, UMD) has been shown to modify and deactivate biomolecules. X-ray photoelectron spectroscopy and an enzyme-linked immunosorbent assay have measured changes in surface chemistry and biological activity, respectively. These surface sensitive techniques demonstrated that the greatest surface modifications occurred when the plasma was composed of Ar with low $O_2$ admixtures (< 2%).[1] The facilities at UMD effectively characterize the surface, but opportunities to characterize the plasma jet are limited. The optical diagnostics available in Prof. Lempert's group (the Ohio State University, OSU) naturally complement the surface studies conducted at UMD as they are able to detect a variety of reactive species produced in the plasma. This visit has fostered future collaborations between UMD and OSU.

At the start of the collaboration, we sought to use laser-induced fluorescence (LIF) to detect ground state NO and two-photon absorption laser-induced fluorescence (TALIF) to detect O and N atoms. During the visit, planar LIF was used to image NO production both in open air and in a controlled environment. TALIF measurements were not performed due to hardware issues. Instead, spatially-resolved optical emission measurements were performed using narrow bandpass filters and an iCCD camera. Aside from the measurements, graduate student Elliot Bartis had the opportunity to work hands-on with advanced plasma characterization techniques that are not available at UMD. The collaboration also provided a framework for the OSU team to pursue research in the field of plasma medicine.

## II. Discussion of Research Outcomes and Findings Resulting from Visit

Spatially-resolved optical emission at 340 ± 10 nm and 750 ± 10 nm was performed by imaging the plasma jet with an iCCD camera in combination with narrow bandpass filters. $N_2$* ($C \rightarrow B$, second positive system) emits strongly at 337 nm while Ar* ($3s^2 3p^5 (^2P°_{1/2}) 4p \rightarrow 3s^2 3p^5 (^2P°_{1/2}) 4s$) emits strongly at 750 nm. The latter upper state is chiefly excited by electron impact excitation of ground state Ar and indicates where higher electron densities are found. Measurements were performed in open air for Ar plasmas at applied voltages ranging from 6-9

kV, flow rates from 2-5 liter/min, and molecular gas admixtures up to 2% for $N_2$, $O_2$, and dry air. For pure Ar discharges, the plume length increases for higher applied voltages and gas flows. At an applied voltage of 6 kV, the plasma jet has a faint glow at the nozzle, which extends into a long, narrow plume as the voltage is increased. Figure 1a shows the cross sectional emission intensities for corresponding positions along the jet axis. The image is shown for comparison. In Figure 1b, the $N_2$* emission was isolated from a pure Ar discharge. The jet's interaction with ambient $N_2$ is clear as a localized cluster of $N_2$* emission occurs at approximately 1 cm downstream of the nozzle. In contrast, Ar* emission downstream from the jet nozzle is concentrated into a narrow channel. As the feed gas flow increased, the localized $N_2$* emission occurs farther from the nozzle and weakens in intensity. At the highest flow rate measured, the $N_2$* emission cluster was gone. We speculate that increased flows enhance the recombination and relaxation of reactive species that contribute to the formation of $N_2$(C).

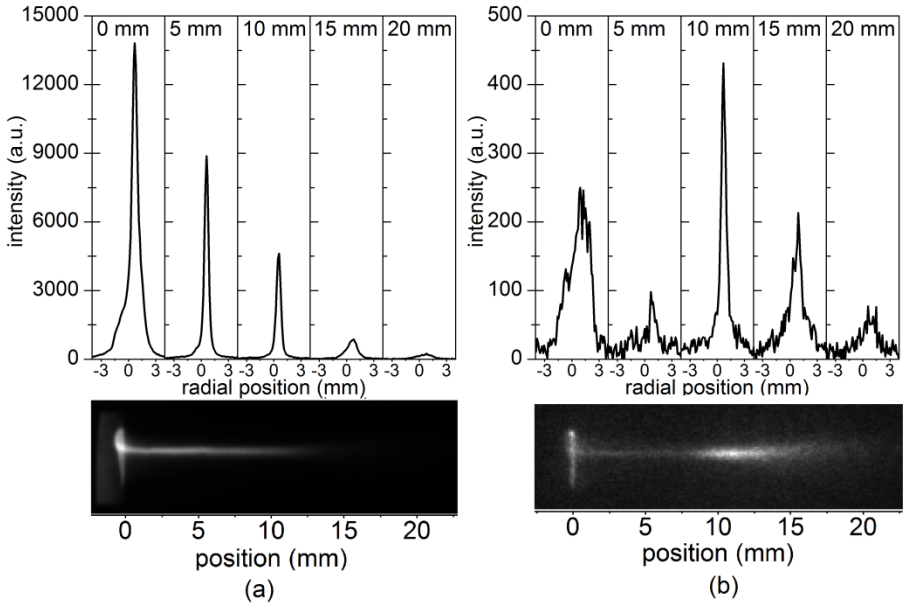


*Figure 1: Emission intensity and images of atmospheric pressure plasma jet for a) full emission and b) isolating $N_2$* emission.*

Figure 2 shows the effect of molecular gas admixtures to Ar plasma. $N_2$ addition to the plasma (Fig. 2a) causes the discharge region to expand radially. The intensity at the nozzle decreases when 0.25% $N_2$ is added, but increasing the admixture up to 2% does not significantly decrease the intensity further. Additionally, we found that $N_2$/Ar mixtures enhance the plume length. At distances greater than 5 mm from the nozzle, 1% $N_2$ in Ar plasmas emit more than 0.25% $N_2$ in Ar plasmas. Air and $O_2$ admixtures also expand the discharge region radially, but differ from $N_2$ admixtures in that increased admixtures decrease the overall emission (Fig. 2b, 2c). The plume length also decreases with increasing admixtures more rapidly than the Ar and $N_2$/Ar plasma; hardly any emission is detected 15 mm from the nozzle. $O_2$-containing discharges also do not form a narrow plume downstream from the nozzle and the discharge appears as a glow. These changes demonstrate the strong effect of $O_2$ on the plasma properties.

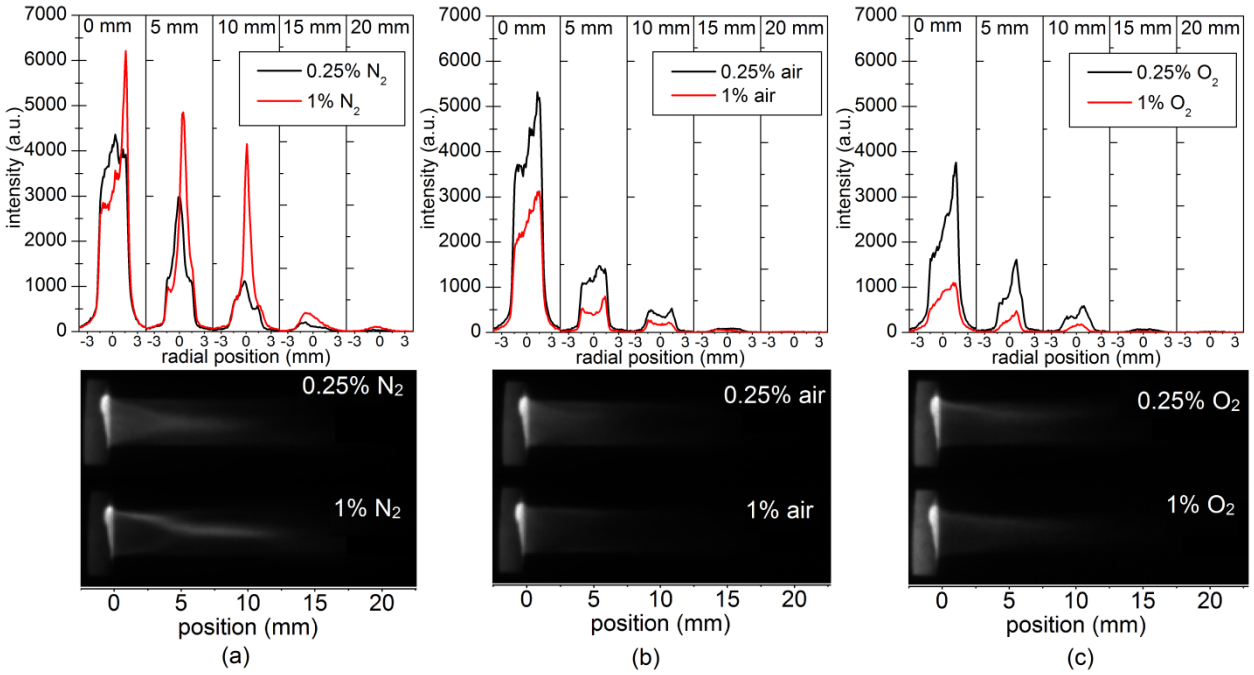


*Figure 2: Emission intensity and images of atmospheric pressure plasma jet for a) $N_2$, b) air, and c) $O_2$ addition.*

LIF of ground state NO was performed in a controlled Ar environment with the plasma confined within the alumina tube. The resulting images of NO emission are shown in Figure 3. As a positive control, 100 ppm NO in $N_2$ was flowed through the alumina tube with the plasma off. A clear signal was measured. Pure Ar plasma in the Ar environment showed a weak signal. As NO should not be produced in these conditions, 1% air was added to the plasma to check if the signal was due to trace amounts of air in the system. With the air admixture, an equally weak signal was observed. In contrast to Ar-based discharges, pure He plasma does not generate a weak signal while a strong signal is observed for 1% air in He plasma. Van Gessel et al. have measured NO in an rf-driven air/Ar atmospheric pressure plasma jet by LIF and report densities as high at 7.5E20 NO $m^{-3}$.[2] The geometry of their jet differs from ours, which might explain the differences in NO density. NO production also varies with temperature. The difference in NO density encourages future research as it suggests that NO production can be regulated. The ability to regulate NO density is crucial to the development of atmospheric pressure plasma sources, as described in a recent review by Graves.[3] The weak emission in pure Ar plasma only occurs when the plasma and laser are on, motivating a new research direction for studying LIF of Ar species.

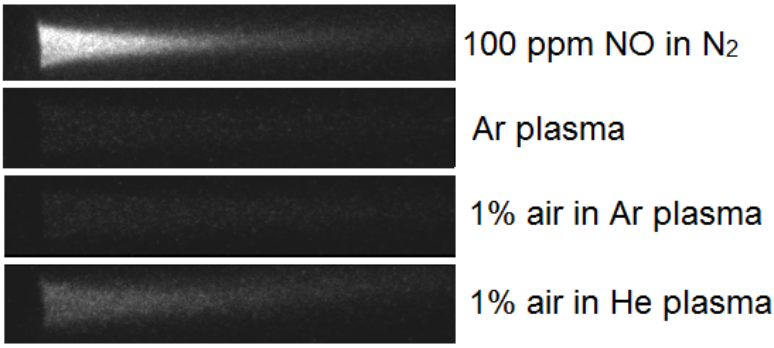


*Figure 3: Planar LIF imaging of NO. Pure Ar plasma produces a weak signal that could be due to LIF of Ar species. Air/Ar plasma does not generate significant NO compared to air/He plasma.*

**III. Follow-up to Visit**

Future work involves measuring reactive oxygen species such as atomic O using TALIF and singlet delta oxygen using infrared optical emission spectroscopy. We also seek to understand how we can modify the APPJ so that appreciable amounts of NO are produced in Ar discharges. The interaction of the jet plume with the environment will also be studied by using controlled environments of $N_2$/Ar, $O_2$/Ar, and air/Ar mixtures to study the production and destruction of reactive species in the environment.

**References:**

1. E. A. J. Bartis, D. B. Graves, J. Seog and G. S. Oehrlein, J. Phys. D, Appl. Phys. **46** (31) (2013).
2. A. F. H. van Gessel, K. M. J. Alards and P. J. Bruggeman, J. Phys. D, Appl. Phys. **46** (26) (2013).
3. D. B. Graves, J. Phys. D, Appl. Phys. **45** (26) (2012).