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Project ID: **54914**

Project Title: **Atmospheric-Pressure Plasma Cleaning of Contaminated Surfaces**

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Project Title:
Atmospheric Pressure Plasma Cleaning of Contamination Surfaces

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NOTE: AS PER 5/24 HQ GUIDANCE, THE PI WILL BE SUBMITTING THE FINAL REPORT IN SEPTEMBER AND IS NOT REQUIRED TO PROVIDE THE ANNUAL REPORT. (THIS IS THE FINAL YEAR OF THE PROJECT).

I. Research Objective:

The objective of this project is to identify the key physics and chemistry underlying the use of atmospheric pressure plasmas for etching removal of actinides and actinide surrogates. This includes understanding of basic discharge mechanism at atmospheric pressure, gas and surface phase chemistry, and optimization and scale-up effort of atmospheric pressure plasma jet (APPJ).

II. Research Progress and Implications:

This report summarizes work after 1 1/2 year of a 3-year project. There have been several major accomplishments in this project, in which the work is jointly conducted at Los Alamos and UCLA. They are:
1) understanding of discharge physics and its application to a scale-up effort, 2) analysis of gas phase chemistry and characterization of effluent, 3) optimization of the APPJ performance, and 4) development of novel concept to expand the capability of the APPJ. Among them, the findings of (1) and (2) are of great interest to the basic science and we are in the process of writing a paper for widely circulated science magazine such as Science. The successful optimization of the APPJ performance was a major milestone in our program plan for FY 1998. In addition, we are currently in the process of preparing invention disclosures for the novel concepts and the subsequent proto-type designs of the new APPJ. Summaries of these key accomplishments are described below.

1) Understanding of physics and its application to a scale up effort
We have shown that the plasma discharge in the APPJ exhibits characters more akin to the low-pressure discharge than the other atmospheric pressure plasma sources, such as plasma torch, corona and silent discharge. The plasma discharge in APPJ is homogeneous in space and time with the gas temperature being much colder than the electron temperature. In addition, the I-V characteristic of the plasma discharge in APPJ resembles the conventional low pressure DC glow discharge, exhibiting spark breakdown, normal and abnormal glow region and transition to arc. These similarities between the APPJ and low-pressure discharge have led new insights into the discharge physics of the APPJ and addressed some of the very critical questions about the scale-up of the APPJ. Combined with the Paschen curve, it is found that the discharge becomes self-sustainable when the RF voltage between the two electrodes reaches in the range of 100 V to 400 V (rms) depending only on the electrode gap spacing and gas composition. This breakdown voltage is lowest with pure helium and increases with increasing fraction of other gases, such as oxygen, nitrogen, etc. In comparison, it is found that the operation of the APPJ becomes unstable as arcing occurs between the electrodes. The arcing occurs when the RF voltage between the two electrode exceeds a critical voltage in the range of 250 to 500 V (rms) depending strongly on the electrode gap spacing, gas composition and weakly on the gas temperature in the APPJ. This critical voltage for arcing depends strongly on the gap spacing and gas composition. Between the breakdown voltage and the arc voltage, a wide-range of stable operating regime for the APPJ has been identified. In addition, it is

found that this stable operating regime do not depend strong on the surface area of the electrode as neither breakdown voltage nor arc voltage are strong function of the electrode area. This is a very important conclusion since it indicates that the power handling capability of the APPJ will increase in a linear manner with increasing surface area of the APPJ for a given gap spacing and gas composition. A large increase in power handling up to 2 kW has been achieved for the APPJ with the electrode dimension of 10 cm x 10 cm x 0.16 cm with O_2 fraction between 0.5 and 2% and the gap spacing between 0.16 cm and 0.24 cm. Currently an effort is being made to increase the power handling capacity of the APPJ above 10 kW level by increasing the electrode dimension to 60 cm x 10 cm x 0.16 cm.

(2) Analysis of gas phase chemistry and characterization of effluent

Significant progress has been made in analysis of gas phase chemistry and characterization of effluent by measuring absolute concentration of two key chemical species, Ozone by UV absorption spectroscopy and metastable O_2(b) by absolutely calibrated optical emission spectroscopy. By measuring the concentrations of these species both in the discharge and in the effluent, we now have a better understanding of the gas phase chemistry. In the discharge region, a large amount of chemically reactive species are produced by electron impact collision, e.g. atomic oxygen of up to $1x10^{16}$ cm^{-3}, metastable O_2(b) of up to $5x10^{12}$ cm^{-3} and Ozone of up to $5x10^{15}$ cm^{-3}. As these reactive species leaves the discharge region, recombination and de-excitation changes the composition of reactive species in the effluent as a function of distance from the jet exit. In a short range of less than 1 cm, most of metastable O_2(b) is collisionally quenched by ozone. In a medium range of between 0.5 cm and 2 cm, there is sufficient atomic oxygen (about $1\text{-}5x10^{15}$ cm^{-3}) which can initiate many chemical reactions at a considerable rate, while the atomic oxygen recombines with O_2 molecules to form ozone. In a long rage of between 2 cm and up to 10 cm, there is a large concentration of chemically reactive ozone (up to $1.2x10^{15}$ cm^{-3}). In addition, it is expected that a significant concentration of metastable O_2(a) of about $1x10^{16}$ cm^{-3} will persist up to 10 cm in the jet effluent from the plug-flow model by a graduate student in UCLA (James Y. Jeong). Thus, it is now possible to selectively utilize or maximize the chemical reactions based on the dominant chemical species in the effluent.

3) Optimization of the APPJ performance

Substantial performance improvement has been achieved for the flat jet. By optimizing the flow rate and volume fraction of O_2 content, polyimide (kapton) etch rates of 3 to 5 mg per minute have been obtained for an input power range of 275 watts to 700 watts. These etch rates are comparable to or faster than the previously optimized etch rate of a round jet. Thus, these results with a greater power handling capability of the flat jet strongly support the flat jet as a platform for scale-up of the APPJ.

4) *Development of novel concept to expand the capability of the APPJ*

As a by-product of our ongoing research effort, we have successfully explored novel concepts to maximize the potential of the APPJ. These concepts are: (1) stable jet operating at a very low helium flow rate, (2) in-situ material treatment in the plasma discharge region, and (3) treatment and removal of thin films on the electrode surface. Two proto-type designs based on these novel concepts were developed and tested successfully.

III. Planned Activities

- Computer modeling of APPJ: To investigate the basic plasma physics issues of APPJ, Dr. Jaeyoung Park will work on the fluid-based computer model for about 3 months (June - Aug, 1998), part of which (May 21st ñ July 10th) he will be working at NYU.
- Test on removal of the depleted Uranium from the metal surface: Preliminary tests on removal of the depleted Uranium from metal surface using APPJ will be conducted in the latter part of 1998 calendar year. This test will last about 2-4 weeks and provide the guideline for the research during FY 1999. Currently, regulatory issues of dealing radioactive material are being addressed.
- Scale-up of APPJ: To increase the speed and performance of the APPJ, up to 2 feet wide jet is currently being designed to accommodate up to 10 kW of CW power. We expect the construction of the large-scale jet by the end of FY 1998, and the test and optimization during the first half of FY 1999.

IV. Information Access

- Two technical papers are accepted for publication, while an invited review paper was submitted. In addition, this project was featured in an article in the October 1997 issue of Discover magazine.

1. J. Y. Jeong, S. E. Babayan, V. J. Tu, J. Park, R. F. Hicks and G.S. Selwyn, Plasma Source Science and Technology, accepted for publication in 1998.
2. S. E. Babayan, J. Y. Jeong, V. J. Tu, J. Park, G.S. Selwyn and R. F. Hicks, Plasma Source Science and Technology, accepted for publication in 1998.
3. A. Schutze, J. Park, J. Y. Jeong, S. E. Babayan, G.S. Selwyn and R. F. Hicks, submitted for special issue for IEEE transaction on plasma science.

- Together with our sister project at UCLA, we are maintaining a web page on APPJ at http://prosurf.seas.ucla.edu/memc/plasma.htm.