Project Summary

Project ID: 54914
Project Title: Atmospheric-Pressure Plasma Cleaning of Contaminated Surfaces
Publication Date: February 16, 2000. **Number of graduate students:** 2

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Specific DOE problems that are being addressed: The science undertaken through this project has addressed a critical need within the U.S. Department of Energy. The decontamination and decommissioning of buildings, structures and materials within the nuclear weapons complex is one of the most expensive and challenging components of the environmental restoration effort. The atmospheric-pressure plasma jet shows real promise as a cost-effective and environmentally benign method of removing radioactive contamination. The research funded by the Environmental Management Sciences Program has put this technology on a firm scientific foundation, and has laid the groundwork for transfer into practice. Through the continued support of the Department of Energy, this exciting science will yield new technologies that significantly reduce costs and accelerate the cleanup effort.

Research Objective: The purpose of this project is to develop a low-cost, environmentally benign technology for the decontamination and decommissioning of transuranic waste. In order to accomplish this goal, an understanding of the scientific principles of operating the atmospheric-pressure plasma jet must be achieved. This knowledge can then be applied to the design of a working tool for D & D applications within DOE.

Research Progress and Implications: Throughout the duration of this three-year project, this research has led to a fundamental understanding of high-pressure plasmas and their interaction with metal surfaces. We have characterized the plasma physics of an exciting new discharge that operates at atmospheric pressure. At the same time, we have identified the gas-phase chemical reactions occurring in oxygen-containing plasmas, and determined the surface kinetics of etching metals with plasmas. In addition to significantly advancing plasma and materials science, this research project should lead to new applications of plasmas in the ambient pressure regime.

We have demonstrated that by the addition of helium and by altering the electrode design, a plasma may be generated at atmospheric pressure that is weakly ionized with a neutral temperature of 75 to 125 °C. The electrons in this plasma are not in equilibrium with the rest of the gas: their energy is 2 to 3 electron volts, or more than 20,000 °C. The electrons are present in very low concentration. Nevertheless, they cause the dissociation of a significant fraction of the molecules present. For example, at an applied power of 24.4 Watts/cm^3 and 10 Torr O_2 in the feed, the plasma dissociates about 1% of the oxygen molecules into atoms (a concentration of ~10^{16} cm^{-3}). We found that by adding a few Torr of carbon tetrafluoride with the oxygen, the plasma generates a large flux of fluorine atoms. These fluorine atoms etch tantalum and other metals at several microns per minute. An examination of the surface chemistry has revealed that the etching rate is limited by the reaction of fluoride species with a metal fluoride layer on the tantalum substrate.

Planned Activities: Future work needed to implement this device for D&D applications include (a) identification of the reactive species in CF_4/He and CF_4/O_2/He plasma jets, (b) extension of the APPJ etching process to radioactive materials, (c) identification of volatile products and development of filtration techniques. Identification of the reactive species in the plasma jet is extremely important because we need this knowledge to increase metal etching efficiency, and to better understand how the volatile products are formed. It is also necessary to perform etching experiments with radioactive elements to determine tool effectiveness.

We have chosen to study the plasma etching of tantalum, because this element exhibits chemistry similar to plutonium and is a good surrogate material for the actinides. However, we expect there to be differences between the surface chemistry of etching tantalum and actinide metals, and these differences should be explored. Therefore, in the future, we propose to investigate the atmospheric-pressure plasma etching of uranium oxide and metal films. We also propose to collaborate with scientists at Los Alamos National Laboratory to demonstrate the use of the plasma jet for an actual decontamination problem. An excellent test case would be to remove plutonium from the surfaces of glove boxes. This work would be carried out in the specialized facilities available at the Los Alamos laboratory.

Information Access:

1. Jeong, J.Y., Babayan, S.E., Tu, V.J., Henins, I., Velarde, J., Selwyn, G.S. and Hicks, R.F., "Etching Materials with an Atmospheric-Pressure Plasma Jet," *Plasma Sources Sci. and Tech.* **7**, 282-285, 1998.

2. Schütze, A., Jeong, J.Y., Babayan, S.E., Park, J., Selwyn, G.S. and Hicks, R.F., "The Atmospheric-Pressure Plasma Jet: A Review and Comparison to other Plasma Sources," *IEEE Trans. Plasma Sci.* **26**, 1685-1694, 1998.

3. Jeong, J.Y., Babayan, S.E., Schütze, A., Tu, V.J., Park, J., Henins, I., Selwyn, G.S. and Hicks, R.F., "Etching polyimide with a non-equilibrium atmospheric-pressure plasma jet," *J. Vac. Sci. Technol. A* **17**, 2581-2585, 1999.

4. Park, J., Henins, I., Herrmann, H.W., Selwyn, G.S., Jeong, J.Y., Hicks, R.F., Shim, O., and Chang, C.S., "An Atmospheric Pressure Plasma Source," *Appl. Phys. Lett.*, **76**, 288-290 (2000).

5. J. Park, I. Henins, H.W. Herrmann and G.S. Selwyn, "Neutral Bremsstrahlung Measurement in Radio-Frequency (RF) Atmospheric-Pressure Discharge", submitted to *Phys. Rev. Lett.*, July 1, 1999.

6. Tu, V.J., Jeong, J.Y., Schütze, A., Babayan, S.E., Selwyn, G.S. and Hicks, R.F., "Tantalum Etching with a Non-Thermal Atmospheric-Pressure Plasma," submitted to *J. Vac. Sci. Technol. A*, February 11, 2000.