EMSP Project Annual Report

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

DOE-ID Grant Number: **FG07-00ER45857**

EMSP project number: **73835**

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Research Objective

The objective of this work is to demonstrate a practical, atmospheric pressure plasma tool for the surface decontamination of nuclear waste. Decontamination of radioactive materials that have accumulated on the surfaces of equipment and structures is a challenging and costly undertaking for the U.S. Department of Energy. Our technology shows great promise for mitigating the cost of this clean up effort.

Research Progress and Implications

This report summarizes the work accomplished during the first six months of a three-year project. A low temperature, atmospheric pressure plasma has been developed with initial support from the Department of Energy, Environmental Management Sciences Program (*see references in Information Access*). Now the goal is to demonstrate this technology on actual contaminated structures within the DOE complex. The group at UCLA, headed by Dr. Hicks, is investigating the removal of fission products from surfaces. If successful, a decontamination and decommissioning (D & D) application will be attempted at INEEL. The fission products examined initially are zirconium and cesium deposited on steel coupons (SIMCON II samples). The group at LANL, headed by Dr. Herrmann, is studying plutonium D & D in facilities located at that laboratory.

At UCLA, it has been found that zirconium can be rapidly stripped from surfaces using a chlorine plasma. Rates up to 6.7 μm/min have been observed. Cesium was removed from the surface, but at a slower rate. Figure 1 shows the dependence of the etching rate and afterglow temperature on the RF power. The plasma was operated at 100 ml/min Cl_2, 48 L/min He, 3 mm sample-to-nozzle distance, and an active etching area of 0.8 cm^2. Monel electrodes were used to prevent corrosion of the device. The plasma jet was run at RF powers ranging from 400 to 900 W. Above 900 W, the temperature exceeded 300 °C, causing copper to be etched out of the monel electrodes. It

was observed that the etching rate increased from 0.6 μm/min at 400 W to 4.5 μm/min at 900 W.

The dependence of the zirconium etching rate on the chlorine flow rate is shown in Figure 2. The conditions were 48 L/min He, 270 °C, 3 mm sample-to-nozzle distance and 800 W RF power. The etching rate increased sharply with chlorine flow rate, to a maximum of 6.7 μm/min at 250 ml/min Cl_2. Above this value, arcing was observed.

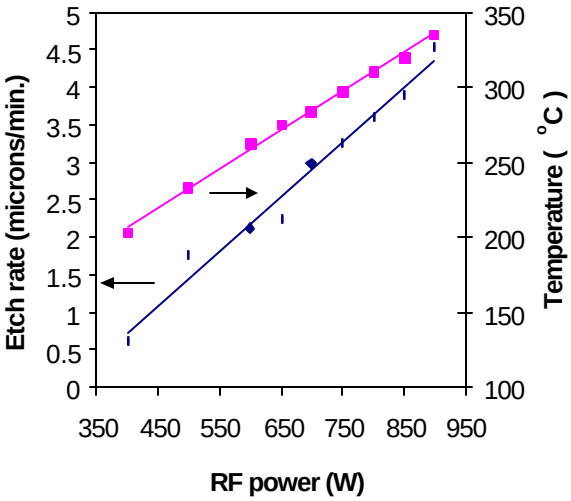

Figure 1. Etch rate of zirconium and afterglow temperature as a function of RF power.

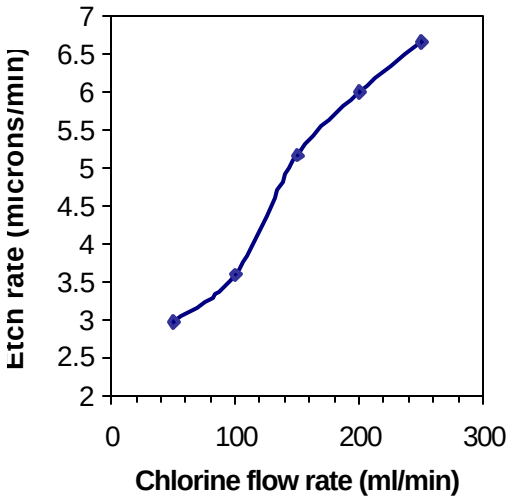

Figure 2. Etch rate of zirconium as a function of chlorine flow rate.

With the Cl_2 plasma source, etching of simulated contamination samples (SIMCON II) was investigated. These samples were steel coupons containing a coating of cesium and zirconium oxide. Metal nitrates were deposited on the steel and converted to metal oxides by heating in an oven to 700 °C for 24 hrs (*Demmer, Development of Simulated Contaminants (SIMCON), WINCO external report #118 (1994)*). Optical microscopy images revealed that the steel surfaces were covered by small oxide crystals ranging from 10 to 30 μm in diameter.

In figure 3, x-ray photoemission spectra (XPS) are presented of SIMCON II samples before and after etching for 10 and 25 min. The reaction conditions were 200 ml/min Cl_2, 48 L/min He, 270 °C, 800 W RF power, and 3 mm sample-to-nozzle distance. The intensities of the Zr peaks decreased sharply following treatment, whereas the intensity of the iron peaks increased. After plasma etching for 25 min, the Zr contamination was eliminated. A closer examination of the cesium 3d peaks, as shown in Figure 4, indicates that most of the cesium was removed as well. In summary, the atmospheric pressure chlorine plasma appears to be an effective tool for dry stripping fission products from nuclear waste.

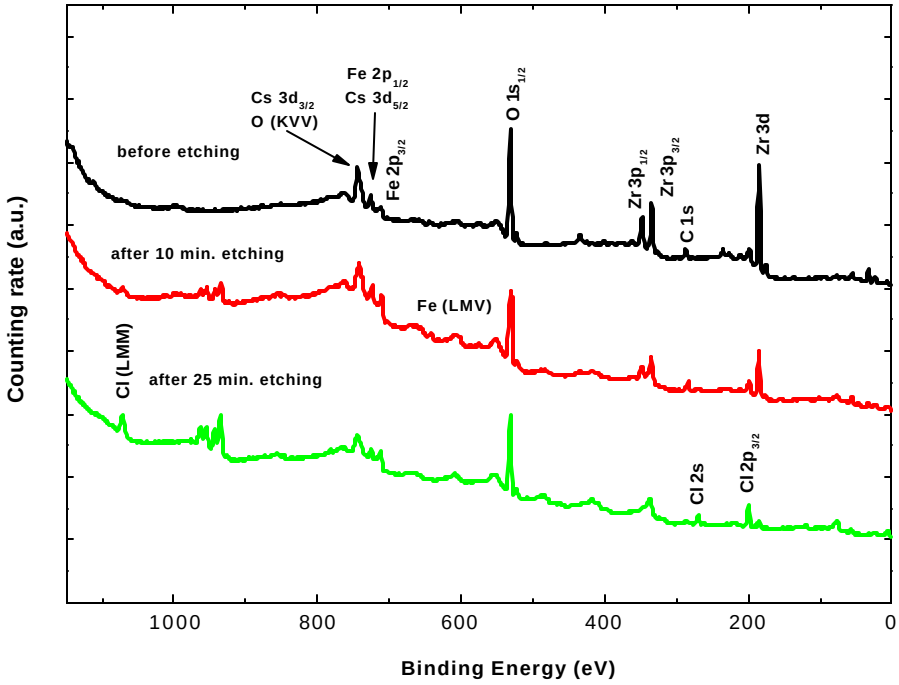

Figure 3. XPS of SIMCON II samples before and after etching.

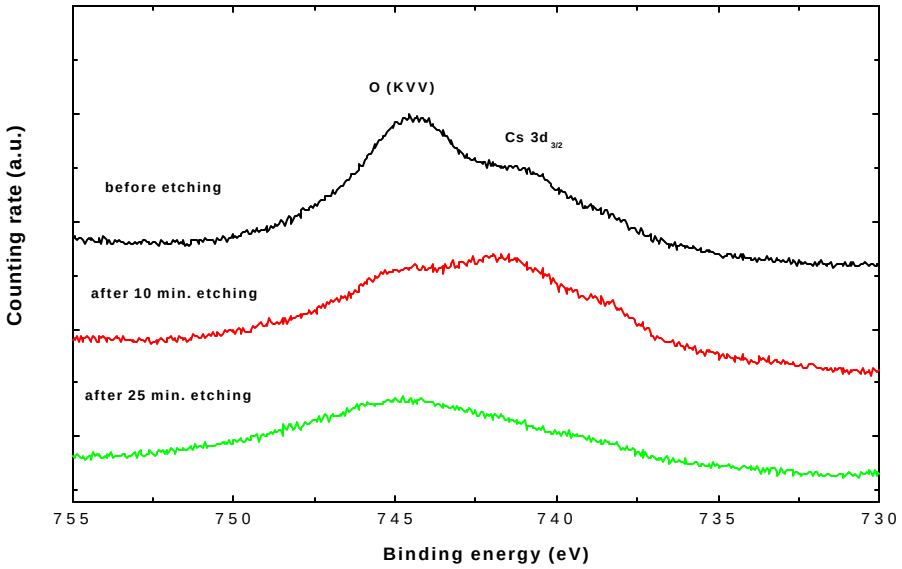

Figure 4. XPS of SIMCON II samples before and after etching.

At LANL, the etching of tantalum, a surrogate for plutonium, was examined with the Atmospheric Pressure Plasma Jet (APPJ). In this case, a mixture of carbon tetrafluoride, oxygen and helium is fed to the gas discharge. Further process optimization has allowed us to achieve etch rates more than double the previous record. This has been achieved primarily through increased RF power stability limits. Work is underway on the design of the APPJ system that will be placed inside a glove box for plutonium etching. We have also succeeded in developing a stable discharge in ambient air, without helium, and have been diagnosing this plasma to determine its suitability for D & D applications.

Planned Activities

We plan to replace the monel electrodes with a more resistant material in order to run the plasma at higher power. Higher radio frequencies will be used to prevent arcing at chlorine flow rates above 250 ml/min. Zirconium foils and SIMCON II samples will be etched with plasmas containing mixtures of chlorine, oxygen, carbon tetrafluoride and argon. We also plan to combine plasma etching with water rinsing to completely remove

the cesium. Once an effective decontamination procedure has been developed, we will explore the possibility of demonstrating the technology at the INEEL site.

At LANL, an APPJ system will be built for insertion into a Pu-contaminated glove box. This device will be deployed and tested in fiscal year 2002. We will also continue to look for ways to improve the decontamination process as well as make it cheaper.

Information Access

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2. J. Park, I. Henins, H.W. Herrmann, G.S. Selwyn, J.Y. Jeong, R.F. Hicks, D. Shim, C.S. Chang, "An Atmospheric Pressure Plasma Source," *App. Phys. Lett.*, **76**, (Jan 2000), 288.

3. J. Park, I. Henins, H.W. Herrmann and G.S. Selwyn, "Neutral Bremsstrahlung Measurement in an Atmospheric-Pressure Radio-Frequency (RF) Discharge," *Physics of Plasmas*, **7**, (Aug 2000), 3141.

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