

SLAC - PUB - 5015

June 1989

(A)

Atomic Oxygen Detection by Silver-Coated Quartz Deposition Monitor *

V. Matijasevic

Department of Physics, Stanford University, Stanford, CA 94305

E. L. Garwin

Stanford Linear Accelerator Center, Stanford, CA 94309

R. H. Hammond

Department of Applied Physics, Stanford University, Stanford, CA 94305

A method for measuring the flux of atomic oxygen utilizing a silver film on a quartz crystal deposition rate monitor is described. Measuring the initial oxidation rate of the silver, which is proportional to the atomic oxygen flux, determines a lower limit on the atomic oxygen flux. This method is more direct than measuring the conductance of the silver film, has an intrinsic flux detection range of 10^{13} - 10^{17} atoms/cm²-sec, and is reversible by exposing the sensor to an atomic hydrogen flux.

(Submitted to Review of Scientific Instruments)

* Work Supported by the Department of Energy
Contract DE - AC03 - 76SF00515

In experiments where an atomic oxygen beam is utilized, it is usually desired to measure the flux of O atoms in the beam. This note describes a simple method to quantify the flux by a variation of a technique described in the literature. Henderson and Schiff¹ first described a procedure to detect atomic oxygen by its reaction with a silver film. Silver reacts rapidly and specifically with oxygen atoms giving a silver oxide. They measured the conductance of the silver film as a function of exposure time to the atomic oxygen beam.

The technique described here uses an oscillating quartz crystal coated with silver. As the silver was exposed to the atomic oxygen beam it oxidized and the mass on the crystal increased, thereby reducing the frequency of the crystal. By monitoring the frequency change, the amount of atomic oxygen diffusing into the silver was determined. Furthermore, by subsequently exposing the silver to an atomic hydrogen beam, the silver was reduced, taking out the oxygen. A similar technique, but oxidizing copper instead of silver, has recently been reported by Clampitt and Hanley.²

A microwave discharge generated the atomic beams. An Ophos Evanson microwave cavity³ was used around a 10 mm o.d. quartz tube through which a flow of gas was maintained. A Tesla coil was used to initiate ionization in the gas. The "cavity" (actually a tunable quarter-wave stub termination for the coaxial feed line) was operated at a frequency of 2.45 GHz. Cooling air was directed at the quartz tube through the body of the cavity. Microwave power input was between 100W and 300W. The cavity was tuned with a tuning stub and a coupling slider in order to match the impedance of the cavity with the discharge to that of the microwave generator and thereby minimize reflected power.

The interior surface of the quartz tube was treated^{4,5} in order to minimize recombination of atomic oxygen at the walls. A boiling concentrated solution of boric acid was poured through the tube after which the tube was heated in a furnace to 600°C for several hours. One end of the quartz tube extended into the vacuum chamber, while the gas was brought in through the other end. Pressure was monitored at both ends of the tube. In the chamber the base pressure was 10^{-6} Torr, while during gas flow it was 10^{-4} Torr. The outer end of the tube was maintained at 0.2 - 1.5 Torr during gas flow. From these pressures and given the pumping speed of the system, we calculate the mass rate of flow as 0.1-0.8 Torr liter/sec. Two inlets into the tube were provided for H_2 and O_2 gases, each with its own leak valve. A shutter was placed at the tube outlet in the chamber in order to precisely control the exposure time of the silver surface to the direct atomic beam.

Standard 5 MHz Sloan crystals for rate monitoring, coated with silver, were used as sensors of atomic oxygen. The silver was at ambient temperature during oxidation, but small amount of heating due to the discharge was discernable from the frequency change. The silver sensor was placed in the chamber facing the outlet of the quartz tube. By flowing O_2 gas without a discharge in the tube we confirmed that molecular oxygen does not affect the silver. However, when the discharge was present in the tube the crystal rate monitor showed a decreasing frequency corresponding to an increase in mass, due to oxidation of silver. The frequency to mass conversion factor is 1.74×10^{-8} g/cm²-Hz. In order to be able to compare results under different conditions, the silver was "cycled", i.e. after each oxidation the silver was reduced with an atomic hydrogen beam, returning the crystal resonance to its original frequency value.

The position of the microwave cavity along the 90 cm long quartz tube, as well as the distance of the silver sensor from the tube outlet were varied. The cavity is easily slid along the tube without breaking vacuum, whereas it was necessary to open the vacuum system in order to reposition the silver coated quartz crystal monitor. These two factors, as well as the gas flow rate and microwave power, determined the atomic O flux at the silver sensor. We note that by moving the cavity (and with it the discharge region) along the tube away from the chamber and into higher O₂ pressure, higher atomic O fluxes were obtained. The flux of atomic species, therefore, did not appear to be limited by recombination along the walls of the tube in this regime.

Figure 1 shows two typical data sets showing the change in mass of the silver as a function of beam exposure time. Initially the mass change is linear in time. As an oxide layer is built up the mass increases as the square root of the time. This latter behavior is indicative of a diffusion-limited process. The initial behavior, however, is limited only by the product of the flux impinging on the silver sensor and the sticking coefficient. From the slope of the initial linear mass increase we derive a lower bound for the flux.

When the silver sensor was 15 mm away from the end of the tube, Fig. 1(a), a large part of the atomic oxygen coming out of the tube was incident on the silver sensor. In this position the oxygen absorption into the silver seems to be almost immediately limited by the in-diffusion of oxygen. The mass change very quickly develops a square root dependence on the time. The initial slope of the data in Fig.1(a) is $\sim 3.5 \times 10^{16}$ atoms/cm²-sec. The total O flux through the tube was $\sim 5 \times 10^{18}$ molec./sec. If we assume a uniform 2x distribution in angle at the end of the tube, then the O₂ flux on the silver sensor would be $\sim 3 \times 10^{17}$ molec./cm²-sec. This

gives an upper bound of about 10% for the efficiency in dissociation of O_2 in the discharge.

When the silver sensor is moved out to 80 mm away from the end of the tube, thereby reducing the incident flux by a factor of 25 simply by the geometry, as shown in Fig.1(b), initially there is a linear region which turns into a square root after some time. Here again we used the slope of the mass change in the initial linear region to get the lower bounds on the incident atomic oxygen flux.

Fig. 2 shows the values of the initial rates as a function of flow rate or, equivalently, the pressure at the end of the tube. We observed a saturation of the flux at a pressure of -2 Torr in the discharge region. Ion current, both positive and negative, was measured using a Faraday cup at the position of the silver sensor. Negligible ion current was present ($<10^{-9}A$ or 10^{10} ions/cm²-sec) and by applying a magnetic field across the tube whatever current had existed was removed. This had no effect on the measured oxidation rate of the silver.

Measuring the oxidation rate of silver with a deposition rate monitor provides a method for determining a lower limit on the flux of atomic oxygen species. Diffusion limited processes place a limit on the detection rate to below 10^8 atoms/cm²-sec.

We are grateful to J. Gregory (Univ. of Alabama), T. Raymond (Sandia Laboratories), and K. Gillen (SRI) for initial suggestions regarding this procedure. Since this work, the procedure has been used extensively by others⁶ for determining atomic oxygen concentrations and we acknowledge useful collaboration and conversations with N. Missert, J.E. Mooij, P. Rosenthal, C.-B. Eom, K. Yamamoto,

and K. Shinohara at Stanford. We would like to thank G.J. Collet of SLAC for technical assistance. Work at SLAC was supported by the Department of Energy, Contract # DE-AC03-76SF00515.

References

1. W.R. Henderson and H.I. Schiff, *Planet. Space Sci.* **18**, 1527 (1970).
2. R. Clempitt and P.E. Hanley, *Supercond. Sci. Technol.* **1**, 5 (1988).
3. F.C. Fehsenfeld, K.M. Evenson, and H.P. Broida, *Rev. Sci. Instrum.* **36**, 294 (1965).
4. J.A.R. Samson and P.N. Pareek, *Phys. Rev. A* **31**, 1470 (1985).
5. D.M. De Leeuw, R. Mooyman, and C.A. De Lange, *Chem. Phys. Lett.* **54**, 231 (1978).
6. N. Missert, R. Hammond, J.E. Mooij, V. Matijasevic, P. Rosenthal, T.H. Geballe, A. Kapitulnik, M.R. Beasley, S.S. Laderman, C. Lu, E.L. Garwin, and R. Barton, *IEEE Trans. Magn.* **25**, 2418 (1989).

Figures

Fig. 1. Change in mass of the silver as a function of atomic oxygen beam exposure time: a) silver sensor 15 mm from tube; b) silver sensor 80 mm from tube.

Fig. 2. Initial slopes of the mass change curves as a function of pressure at the discharge. The numbers by the data points indicate the incident and reflected microwave power in W.

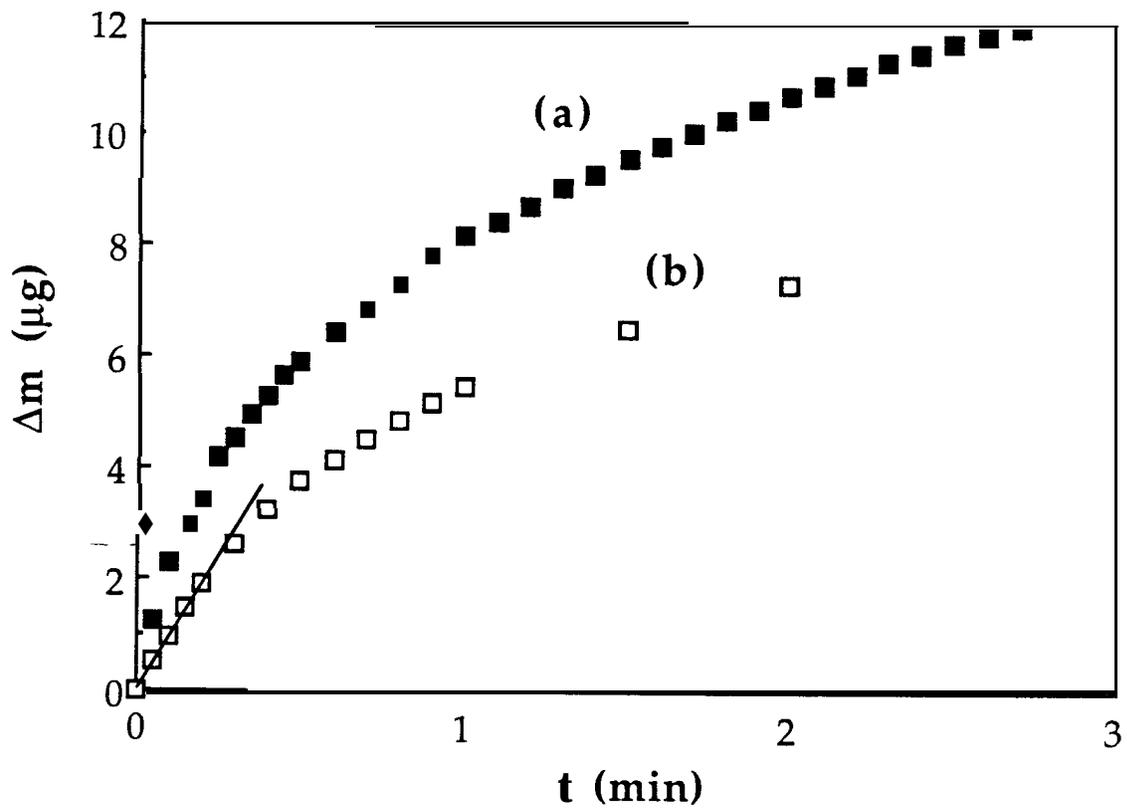


Fig. 1

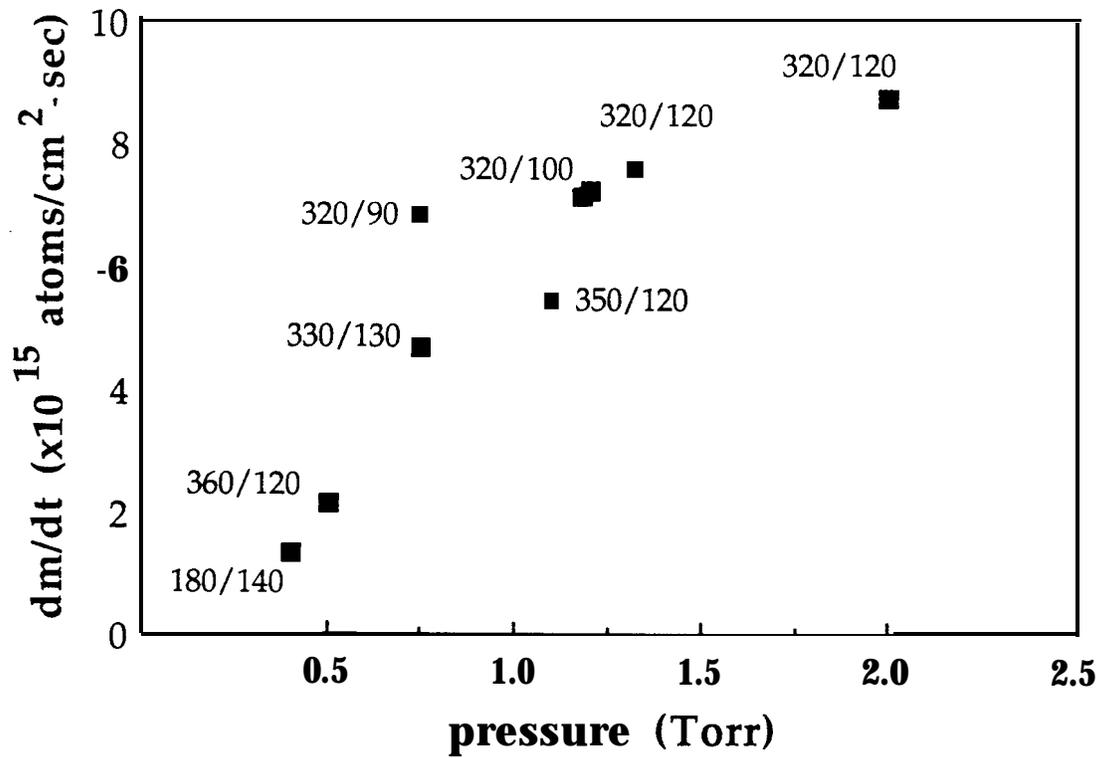


Fig. 2