III. PHYSICAL ELECTRONICS AND SURFACE PHYSICS

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A. INTERACTION OF GASES WITH SOLIDS

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Within the last six months we have nearly completed a mass spectrometric study of the transport of tungsten that is due to chemical reactions with CO_2 and D_2O at high temperatures and low pressures. We have covered a pressure range of 10^{-5}-10^{-3} Torr for each gas and surface temperatures from 1400°K to 3200°K. We have measured the probability of reaction and the rate of formation and evaporation of a variety of volatile tungsten oxide species. Typical reduced data are presented in Figs. III-1 and III-2. We have compared these results with a quasi-equilibrium model of gas-surface reactions formulated by Batty and Stickney.[1]

We are now working on refinements to the quasi-equilibrium model and on the experimental techniques in order to test quantitatively the idea of equilibrium,[2] in particular, we are incorporating a double ionizer into our detection system. As can be observed from the form of the tungsten curves in Figs. III-1 and III-2, a substantial amount of fragmentation occurs during the ionization process used to detect the products. This is unavoidable at the electron energies that are necessary to obtain reasonable signals and get good estimates of the ionization probabilities, and it is the cause of considerable uncertainty in general for reaction rates and thermochemical tables of high-temperature species. By introducing a double ionization technique,[2] we can avoid the problems associated with fragmentation and obtain a meaningful quantitative comparison between the quasi-equilibrium model and experiment.

We are also developing an analogous analytical scheme to evaluate the effect of the competitive formation of neutral species in the theory of the production of negative ions at surfaces.

On another apparatus in which we incorporate mass spectrometry, Auger electron spectroscopy, and nozzle molecular beams to study gas-surface reactions we are looking at the reaction of a variable energy oxygen molecular beam with a Si (111) crystal. We have constructed a rotatable stage to support a new Extranuclear Laboratories ELFS high-resolution quadrupole and we are completing the measurement of velocity
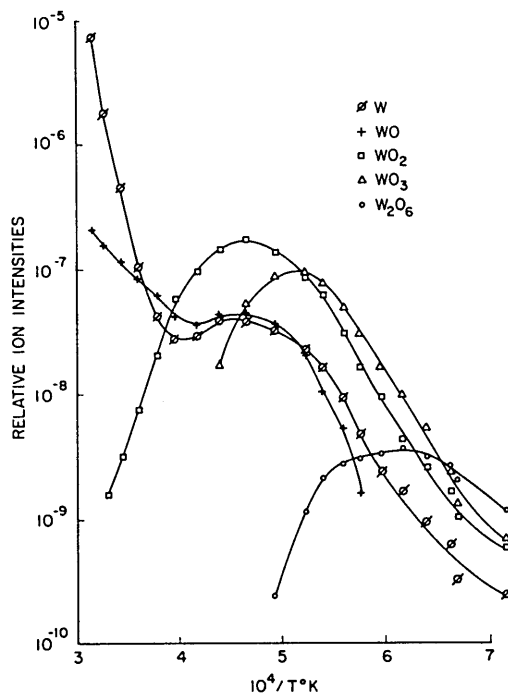
Fig. III-1.

Relative flux signals of reaction products desorbing from a tungsten ribbon subjected to a molecular beam of D_2O. Effective pressure of the beam on the ribbon, 1.9×10^{-4} Torr. Humps in the W and WO curves give clear indication of dissociative ionization occurring in the mass spectrometer ionizer.
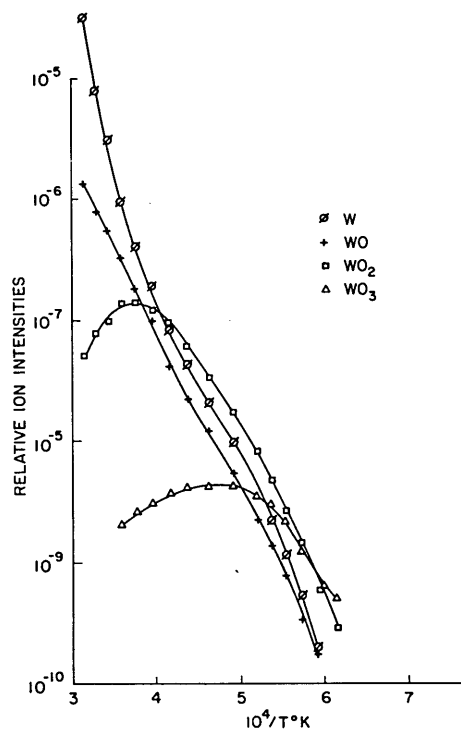
Fig. III-2.

Relative flux signals of reaction products desorbing from a tungsten ribbon subjected to a CO_2 flux with an effective pressure of 1.6×10^{-3} Torr.

distributions of mixed beams of He and O_2. We are using He to obtain collisional acceleration of the O_2 to high velocities by extracting the center line of high-pressure free jet expansions. This will provide O_2 incident energies over a range of .01-2 eV. We are looking at the effect of this variation in kinetic energy, the target surface structure, and surface temperature on the reaction probabilities of O_2 with Si and Ge. At the higher crystal temperatures employed and low steady-state oxygen coverages (less than 1 monolayer), we can monitor the reaction probability by measuring the rate of formation of the volatile monoxide.

References

1. J. C. Batty and R. E. Stickney, J. Chem. Phys. 51, 4485 (1969).
2. C. K. Crawford, Proceedings of Electron Beam Symposium, Boston, Mass., March 1963, J. R. Morley (Ed.).