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REACTIONS OF UO_2 WITH ATMOSPHERIC GASES.(U)
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REACTIONS OF UO^+ WITH ATMOSPHERIC GASES

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20. ABSTRACT (Continue on reverse side if necessary and identify by block number) The rate coefficient for the ion-molecule reaction of uranium monoxide ions with molecular oxygen at thermal energies has been measured using a magnetic bottle apparatus. The UO_2^+ ions were produced by the associative ionization reaction between U and N_2O . The primary (UO_2^+) and secondary (UO_2^+) ions were selected by a quadrupole mass spectrometer and their variations vs. O_2 pressure were plotted on a X-Y recorder. The rate coefficient was determined to be $2.03 \pm 0.44 \times 10^{-9} \text{ cm}^3/\text{sec}$. CUBIC CM TEN TO THE MINUS 9TH POWER PLUS OR MINUS Unclassified 284 135		

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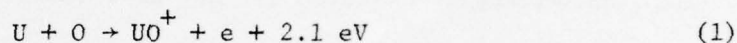
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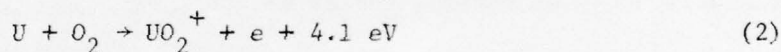
REACTIONS OF UO^+ WITH ATMOSPHERIC GASES

I. Introduction

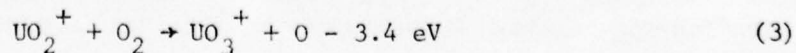
When uranium atoms are released in the upper atmosphere where atomic and molecular oxygen are present, the associative ionization reactions



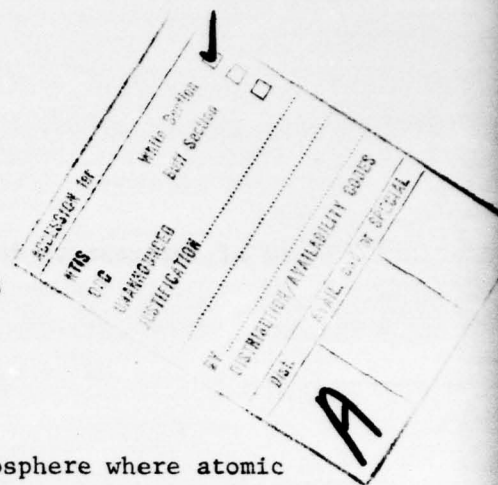
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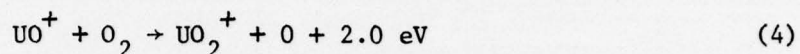
occur. The plasma produced by these reactions cannot be neutralized by the normal dissociative recombination reactions because of the endoergicity of the reactions inverse to Reactions (1) and (2), and hence a long-lived plasma which can cause interference to radar communications can be produced. Further, because of the exoergicity of Reactions (1) and (2), it is expected that both the UO^+ and UO_2^+ ions will be formed in excited states and subsequently radiate causing interference with optical and infra-red communication systems. There is firm experimental evidence from this laboratory that of the order of 1% of the UO_2^+ is formed with internal energies higher than 3.4 eV from the observation that the reaction



proceeds when the UO_2^+ is formed by Reaction (2).



It is of interest to determine the rate at which the reaction



proceeds. Where there are both O and O₂ present in the atmosphere this reaction can reduce the UO⁺ spectrum with time and enhance the UO₂⁺ spectrum. The exoergicity of Reaction (4) may also cause an alteration of the UO₂⁺ spectrum, through population of excited states in addition to those populated by Reaction (2).

This report describes the experimental measurement of the rate coefficient for Reaction (4) at thermal energies.

II. Experimental Approach

The approach employs a "magnetic bottle" apparatus¹ which is shown in Fig. 1. Within the vacuum system are coils which produce a magnetic field bounded by two magnetic mirrors. A beam of uranium atoms crosses the magnetic field at the center of the bottle and ions are formed along the atomic beam's length by associative ionization reactions with gases admitted at very low pressure into the vacuum chamber. Those ions that are formed within the magnetic bottle are trapped in the bottle, moving back and forth between the mirrors on helical trajectories along the magnetic field lines. Eventually elastic collisions of the ions with the added gas change the pitch angle of the ions to a sufficiently low value that the ions can escape through the mirrors bounding the bottle. A quadrupole mass filter is placed outside one of the mirrors and detects the escaping ions.

The magnetic bottle used was produced by a set of ten water-cooled coils, each being 5.1 cm i.d., 15.2 cm o.d., and 2 cm thick. A gold-plated

brass cylinder of 5 cm diameter was inserted in the column of the coils and maintained at -3V. The bottle provided a field strength of 1500 G at the center and 2500 G at the mirrors.

The uranium atom beam was produced in a source consisting of a tungsten tube of 5 cm length and 6 mm diameter, made by rolling a 5.7 cm x 10.2 cm tungsten foil 0.025 mm thick. A hole of 1.6 mm diameter was placed in the side wall of the tube at the center along its length although we have found in previous experiments that the hole is actually unnecessary due to the ability of U atoms to diffuse through the hot tungsten foils. The hole was made for the convenience of recharging U supply into the W tube. Two pieces of 1.6-mm diameter U rod, each 5 mm long, were confined in the central portion of the tube by two waddings of Ta fine wires. The tube was fitted into molybdenum caps at each end which in turn were bolted onto a water-cooled copper jacket which both carried the electrical current and dissipated furnace heat. A current of about 200 A at 2.5 V heated the central portion of the tube to about 2100°K.

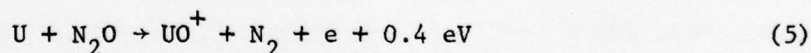
The U atom beam was formed by passing through two collimators attached to the copper cooling jacket before entering the reaction region through a 4 mm x 8 mm entrance aperture in the side of the gold-plated brass cylinder. Proper biasings (the furnace tube at +6V, the first collimator at the ground potential, and the second at +70V) prevented both ions and electrons produced in the U atom source from reaching the reaction region.

A pair of parallel plates situated on the other side of the magnetic coil column and across from the U atom source was used to monitor UO_2^+ formed by Reaction (2) when the U beam intersected a known amount of O_2 admitted into the vacuum. The UO_2^+ ion current measured by the condenser method at a fixed

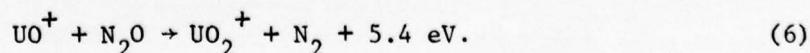
O_2 pressure indicates the U atom beam current and is important in the measurements because the U atom beam current tends to gradually decrease with time.

The primary and secondary ions were extracted through a 9.9-mm exit aperture at one end of the magnetic bottle, some 16 cm from the reaction region, but an extractor normally biased at -2V. The ions were then m/e selected by a quadrupole mass spectrometer and detected by an EMI 21-state venetian blind electron multiplier. The ion current output from the multiplier was measured by an Extranuclear electrometer and the output of the latter was fed into the y-axis of a X-Y recorder. The pressures of the reactant gases were monitored on the x-axis of the X-Y recorder. The pressure readings of N_2O and O_2 added to the vacuum were furnished by a Hughes type 6578 ionization gauge which was calibrated for N_2O and O_2 against a Consolidated GM-110 McLeod gauge.

In the present experiments we added two gases, N_2O and O_2 , in the vacuum. The primary ions were UO^+ and UO_2^+ formed by the reaction



and Reaction (2). The respective rate coefficients, k_5 and k_2 , are known^{2,3} to be $1.51 \times 10^{-13} \text{ cm}^3/\text{sec}$ and $1.04 \times 10^{-12} \text{ cm}^3/\text{sec}$. UO_2^+ can be produced additionally by the reaction



The rate coefficient, k_6 , has also been determined to be $2.73 \times 10^{-10} \text{ cm}^3/\text{sec}$ by the authors in a previous magnetic bottle experiment. If the rate coeffi-

cient for the reaction of interest, Reaction (4), is designated as k_4 , we can write down the rate of change of the concentrations n_1 (for UO^+) and n_2 (for UO_2^+) as

$$\frac{dn_1}{dt} = k_5 n_u n_a - (k_6 n_a + k_4 n_b) n_1 - L n_1 \quad (7)$$

and

$$\frac{dn_2}{dt} = k_2 n_u n_b + (k_6 n_a + k_4 n_b) n_1 - L n_2 \quad (8)$$

where n_u is the concentration in the U atom beam, and n_a and n_b are the number densities of N_2O and O_2 , respectively, admitted into the vacuum. L is the loss rate due to escape of the ions from the mirrors, which is proportional to $n_a + n_b$.

In the steady state, Equations (7) and (8) solve to give the ratio of UO_2^+ and UO^+ as

$$\frac{n_2}{n_1} = \frac{1}{L} \left[k_6 n_a + \left(k_4 + \frac{k_2 k_6}{k_5} + \frac{k_2}{k_5 n_a} \cdot L \right) n_b + \frac{k_2 k_4}{k_5 n_a} n_b^2 \right]. \quad (9)$$

Since k_6 is known and n_a is measured in the experiment, the loss coefficient, L , is determined for $n_b = 0$ from the ratio of the secondary to primary ions when only the N_2O is present. The loss coefficient with added O_2 is then given by $L = L_0 (1 + (n_b/n_a))$ and this can be used to obtain k_4 from the linear portion of a curve of n_2/n_1 , for fixed n_a , as a function of n_b , i.e., the slope as the added oxygen gas pressure approaches zero.

Because of cumulative uncertainties in the linear coefficient expression, however, it is more satisfactory to use the coefficient of the quadratic term in Equation (9) to obtain k_4 .

III. Method

In the present experiment, the UO^+ ions that were produced by Reaction (5) at a fixed N_2O pressure reacted with O_2 , which was added to the vacuum chamber in addition to the fixed N_2O pressure, forming UO_2^+ ions that were also produced simultaneously by Reactions (2) and (6). Figure 2 shows the typical curves of UO_2^+ and UO^+ ion currents vs. O_2 pressure obtained on a X-Y recorder for a fixed N_2O pressure. The pressure of N_2O was typically 3×10^{-5} Torr while the pressure range of O_2 was from 0 to 1.5×10^{-4} Torr. The measurements were repeated for a number of N_2O pressures.

Using data from these two curves, a plot of n_2/n_1 (ratio of UO_2^+ to UO^+ currents) as a function of the number density of O_2 , n_b , was drawn for each fixed number density of N_2O , n_a . The curve of n_2/n_1 vs. n_b is a parabola, as expected by Equation (9). The linear term of Equation (9) can be obtained from the tangent to this curve at $n_b = 0$. Next, the square root of the difference for n_2/n_1 between the parabola and the tangent was plotted as a function of n_b , forming a straight line. The slope, S , of this straight line was used to determine the rate coefficient of Reaction (4) using the relation

$$k_4 = \frac{k_5}{k_2} n_a L S^2. \quad (10)$$

IV. Results and Discussion

The rate coefficient for the ion-molecule reaction of $\text{UO}^+ + \text{O}_2 \rightarrow \text{UO}_2^+ + \text{O}$ at thermal energies was found to be $2.03 \pm 0.44 \times 10^{-9} \text{ cm}^3/\text{sec}$.

Under different experimental conditions, such as changes in the uranium atom source temperature (ranging from 2050 to 2200°K) and variations in the biasing of the reaction region (at -3V and +0.6 V), no obvious difference was observed for the rate coefficient within the experimental errors.

At thermal energies, it appears that the ion-molecule reaction of $\text{UO}^+ + \text{O}_2 \rightarrow \text{UO}_2^+ + \text{O}$ is almost one order of magnitude faster than that of $\text{UO}^+ + \text{N}_2\text{O} \rightarrow \text{UO}_2^+ + \text{N}_2$, the rate coefficient of the latter being $2.73 \times 10^{-10} \text{ cm}^3/\text{sec}$.

Since the loss coefficient, L , is related to the number density of oxygen, n_b , by $L_o(1 + \frac{n_b}{n_a})$, Equation (10) indicates that the rate coefficient, k_4 , for $\text{UO}^+ + \text{O}_2 \rightarrow \text{UO}_2^+ + \text{O}$ is not independent of n_b . Hence the rate coefficient of $2.03 \times 10^{-9} \text{ cm}^3/\text{sec}$ may be considered as the lower limit of the reaction investigated.

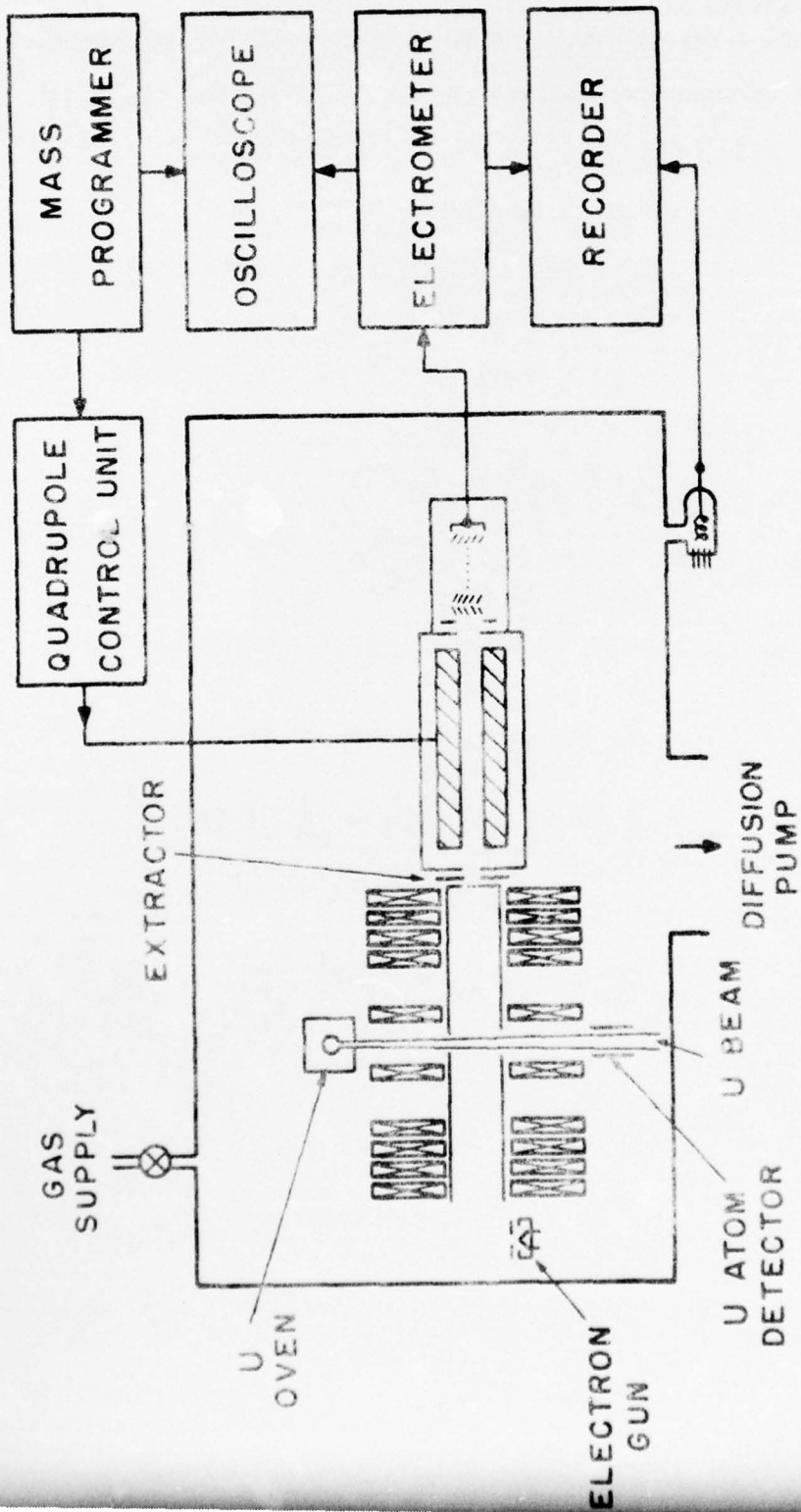
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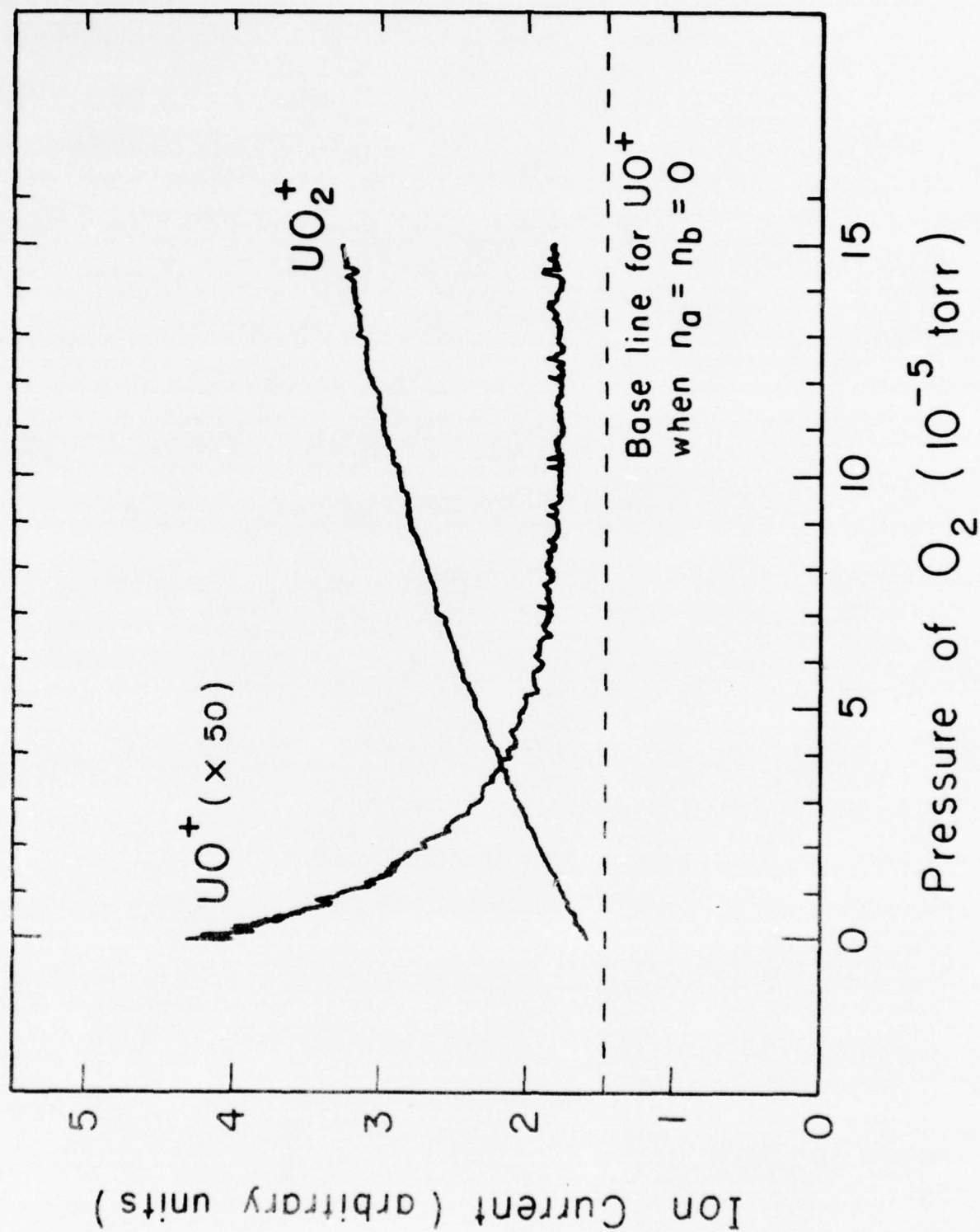
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Figure Captions

Figure 1. "Magnetic Bottle" apparatus.

Figure 2. Typical curves of UO_2^+ and UO^+ ion currents vs. O_2 pressure for a fixed number density of N_2O at $1.07 \times 10^{12} \text{ cm}^{-3}$.





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