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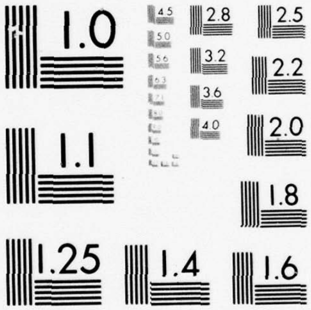
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CROSS SECTIONS FOR THERMAL REACTIONS BETWEEN URANIUM ATOMS AND ATMOSPHERIC SPECIES

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Associative Ionization	Ozone	Lanthanum	SF_4	MoO	Ozone Source
Chemi-Ionization	Sulfur	Gadolinium	CHF_3	MoO_2	Negative ions
Uranium	Thorium	Chlorine	H_2O_2	WO_2	
Oxygen	Neodymium	Bromine	$(CN)_2$	TiO	
Atomic Oxygen	Samarium	SF_6	H_2S	TiO_2	

20. ABSTRACT *(Continue on reverse side if necessary and identify by block number)*

Experimental studies of associative ionization reactions involving uranium and other metal atoms in collisions with O, O_2 and other atmospheric species are described. Absolute reaction cross sections are obtained by comparison with the previously measured cross sections for the U + O and U + O_2 reactions. Negative ions have been observed as products in some of the reactions however these channels account for less than 10% of the total reaction cross section. No reaction products are observed for associative ionization between atomic → next

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oxygen and selected oxides of Mo, W and Ti. Both thorium and uranium have large total cross sections of order 10^{-15} cm^2, for associative ionization with ozone. A simple, efficient ozone source for beam experiments is described.

10 to the -15 power sq. cm.,

I. Introduction

Associative ionization is that type of chemi-ionization[1] wherein two atoms, molecules or radicals, A and B, collide at thermal energies and produce a molecular ion in a process such as

$$A + B \rightarrow AB^+ + e. \qquad (1)$$

In order for the process to proceed it is necessary to provide enough energy to remove the electron from the neutral AB molecule or radical, and this energy comes from (1) chemical binding energy of the AB molecule and (2) internal excitation in either A or B.

Examples of associative ionization reactions which rely on internal excitation have long been known, e.g., the formation of diatomic inert gas ions in discharges and afterglows. The molecular ions formed in this situation can rapidly dissociatively recombine with electrons to produce the initial species in their ground states.

In contrast, if sufficient energy for ionization is provided by the formation of the A-B chemical bond, associative ionization can occur between ground state reactants. Neutralization by dissociative recombination is energetically forbidden for molecular ions formed under these conditions and the lifetime of the ions may be long, even in the presence of a substantial electron density. Since the lifetime against neutralization may be long, the possibility exists that the ion, if formed in an excited state, could radiate even if the transition were in the infrared region where excited state lifetimes are long.

Thus reactants that undergo associative ionization from their ground states offer a source of long-lived electron clouds which can affect radar signal transmission and simultaneously a source of infrared radiation which might be unwanted background in any system involving IR radiation.

We began our investigation of associative ionization reactions, under an earlier research contract[2], by examining the reactions

$$U + O \rightarrow UO^{+} + e \qquad (2)$$

$$U + O_2 \rightarrow UO_2^{+} + e. \qquad (3)$$

An absolute measurement of the cross section for the latter reaction[3] provided a benchmark for measuring other associative ionization cross sections by a simple comparison technique. Further study led to the discovery that ground state associative ionization occurs in a number of metal-oxygen systems[4-8] and a variety of other metal-gas combinations.[2,4,9]

Much of the effort under the present contract has been devoted to the study of various uranium-gas reactions. The observation of dissociative associative ionization[2], in particular the production of UF_2^{+} from U + SF_6, suggested that negative ions might also be formed in some of these reactions. This prompted the development of detection electronics which would permit the observation of negative ions as well as positive ones.

This report summarizes the program as carried out over the past two years, summarizes the present situation with regard to associative ionization processes, and suggests the directions in which future research might profitably move. Included as appendices are a list of associative ionization reactions studied at Extranuclear Laboratories, Inc. and the University of Pittsburgh, abstracts of papers presented at scientific meetings and a preprint of an article on ozone associative ionization with uranium and thorium.

II. Uranium Associative Ionization With Gases

Much of the work performed under the previous contract to examine associative ionization reactions[2] was devoted to reactions between atomic and molecular oxygen and various metals. Reactions were observed between uranium and NO, N_2O and SF_6 producing the positive ions UNO^+, UO^+ and UF_2^+, respectively, although detailed quantitative measurements were not performed. With the observation of dissociative associative ionization in the latter two reactions, the possibility arises that negative ions may also be formed in associative ionization reactions.

We first observed the production of negative ions in the $U + Br_2$ system. Three reaction channels are observed,

$$U + Br_2 \rightarrow UBr_2^+ + e \tag{4}$$

$$\rightarrow UBr^+ + Br + e \tag{5}$$

$$\rightarrow UBr^+ + Br^- \tag{6}$$

By comparison of signal intensities in these reactions with the UO_2^+ signal from reaction (2) under comparable gas beam intensities, we roughly estimate cross sections of 5×10^{-19} cm^2 for (4) and 1×10^{-19} cm^2 for the sum of (5) and (6). The branching ratio betwen (5) and (6) is difficult to estimate, but appears to be of order 10:1. The analogous channels are observed in the reactions of Cl_2 with uranium:

$$U + Cl_2 \rightarrow UCl_2^+ + e \tag{7}$$

$$\rightarrow UCl^+ + Cl + e \tag{8}$$

$$\rightarrow UCl^+ + Cl^- \tag{9}$$

with roughly estimated cross sections of 10^{-17} cm^2 for (7) and 10^{-18} cm^2 for the sum of (8) and (9). The branching ratio appears to be of order unity.

Negative ions are also produced in the U + SF_4 system, where the assumed reactions are

$$U + SF_4 \rightarrow UF_2^+ + SF_2(?) + e \qquad (10)$$

$$\rightarrow UF_2^+(?) + F^- + SF(?) \qquad (11)$$

The cross section for (10) is estimated to be 5×10^{-19} cm^2, while the cross section for (11) is so small as to preclude even a rough estimate. The UF_2^+ product in (11) is shown as uncertain because the F^- signal is sufficiently small that a correspondingly small current of a heavy ion, e.g., UF^+, might easily elude detection. The U + SF_6 system has a comparable cross section for producing UF_2^+, however no negative ions are observed.

Since the uranium-atomic oxygen associative ionization reaction has a relatively large cross section, one might expect the analogous reaction with atomic sulfur, which also has an ns^2 np^4 (^3P) electron structure, to occur. To investigate this possibility an independently heated solids vaporization stage was added to the existing gas dissociation furnace. With this two-stage furance, an atomic sulfur beam can be produced by gently vaporizing molecular sulfur in the relatively cool first chamber and dissociating the vapor in the hotter second chamber.

Uranium and sulfur are found to associatively ionize, producing US^+, an appropriate reaction product to observe during the United States' bicentennial celebration. The reaction cross section is 1.776 x $10^{-17.76}$ cm^2, 50 times smaller than the analogous UO cross section.

A cursory examination of the reaction $U + S_2 \rightarrow US_2^+ + e$ yielded a null result. However, considering beam intensities and the ratio of the O and O_2 cross sections, the US_2^+ product could easily have escaped detection. Insufficient data exist on the dissociation energy and ionization potential of US_2 to determine whether the reaction is even energetically possible.

Several other uranium-gas systems have also been investigated, a number of which, CHF_3, H_2O_2, $(CN)_2$ and H_2S, produced no observable associative ionization signals. The results of these studies are included in Appendix I.

III. Associative Ionization of Metal Oxides With Atomic Oxygen

Qualitative observations have indicated that certain monoxides such as WO may undergo associative ionization with atomic oxygen. Efforts to confirm this were hampered by physical constraints in the experimental apparatus. There is not sufficient space without extensive modification of the apparatus to replace the simple metal beam furnace with a two-stage oven which would be desirable for producing monoxide and dioxide beams from more highly oxidized metal powders. Consequently we had little control of the oxidation state of the beam. However in experiments with atomic oxygen and MoO, MoO_2, WO_2, TiO and TiO_2, no associative ionization products were observed. Based on experimental conditions, this places an upper bound of $\sim 10^{-18}$ cm^2 on the cross sections for associative ionization in these systems, if the reaction occur.

IV. Ozone Associative Ionization Reactions With Uranium and Thorium

In associative ionization reactions involving molecular species, multiple reaction channels are conceivable although for most of the systems which we have investigated, insufficient thermodynamic data are available to determine which channels, if any, are energetically allowed. Ozone is one of the few exceptions; thermochemical experiments[10] indicate that five of the seven possible channels are energetically allowed for ozone associative ionization with both uranium and thorium.

We have investigated both the U-O_3 and Th-O_3 systems. Only two of the possible channels are observed in the uranium reaction, but five distinct channels can be identified with thorium. The observed reactions are summarized in Table I. The cross sections, ranging from 10^{-19} cm^2 to 10^{-16} cm^2, are tabulated in Appendix I; details of the measurements are described in Appendix VIII.

TABLE I

REACTION CHANNELS OBSERVED IN OZONE ASSOCIATIVE IONIZATION WITH URANIUM AND THORIUM

$$U + O_3 \rightarrow UO_2^+ + O + e$$
$$\rightarrow UO^+ + O_2 + e$$
$$Th + O_3 \rightarrow ThO_3^+ + e$$
$$\rightarrow ThO_2^+ + O + e$$
$$\rightarrow ThO_2^+ + O^-$$
$$\rightarrow ThO^+ + O_2 + e$$
$$\rightarrow ThO^+ + O_2^-$$

V. Metal-Oxygen Associative Ionization Reactions

There are three possible associative ionization reaction channels in a metal-molecular oxygen system:

$$M + O_2 \rightarrow MO_2^+ + e \qquad (12)$$

$$\rightarrow MO^+ + O + e \qquad (13)$$

$$\rightarrow MO^+ + O^- \qquad (14)$$

Dr. H. H. Lo at the University of Pittsburgh has observed the production of both MO_2^+ and MO^+ in reactions involving the metals lanthanum and cerium.[11] The production of MO^+ implies that the negative ion channels, (14), is at least energetically allowed, although the MO^+ may be due entirely to reaction (13).

We have investigated the La-O_2 system and observe all three reaction channels. The branching ratio between (13) and (14) is estimated to be of order 10:1, yielding a cross section of $\sim 10^{-18}$ cm^2 for (14) with lanthanum. The cross sections for reactions (12) and (13) with lanthanum are both of order 10^{-17} cm^2. This is consistent with our general observation that even when negative ions are produced in a reaction, the free electron channels dominate.

We have subsequently learned[12] that Dr. R. B. Cohen and his co-workers at the Illinois Institute of Technology have also observed O^- in the La + O_2 reaction and in the Ce + O_2 reaction as well.

Lo and Cohen have also each measured cross sections for a number of metal-atomic oxygen associative ionization reactions. As a consistency check on the different experimental systems and techniques, we have examined the neodynium-oxygen and samarium-oxygen reactions which were studied by both groups. Using the previously measured Gd + O reaction cross section for calibration, we performed direct measurements of the Nd/Gd and Sm/Gd cross section ratios. Setting the ratios of the metal electron impact ionization cross sections equal to unity yields atomic oxygen associative ionization cross sections of 2.7×10^{-15} cm^2 for Nd and 1.7×10^{-16} cm^2 for Sm. A comparison of our results with those of Cohen et al. is shown in Table II. While there is some scatter, the two experiments are in reasonable agreement. Lo has also observed these reactions, however his experiments were not intended to produce absolute cross section measurements.

A simultaneous comparison technique is employed in Cohen's experiments. Two different metals are placed in the beam source and the reaction signals are all observed at a constant source temperature. This approach restricts comparisons to metal pairs having comparable vapor pressures in some temperature range. This requires that the Nd-Gd comparison employ a three element chain, Nd/Tb, Tb/Pr, Pr/Gd while two additional elements, Sm/Ho, Ho/Nd, are required for samarium.

TABLE II

COMPARISON OF MEASURED CROSS SECTIONS OF THE REACTION $M + O \rightarrow MO^+ + e$ FOR THE METALS NEODYMIUM AND SAMARIUM

	Nd	Sm
Cohen[12]	1.3×10^{-15} cm^2	4.3×10^{-17} cm^2
This Work	2.7×10^{-15} cm^2	1.7×10^{-16} cm^2

We have used a sequential comparison method for these experiments. The furnace containing the two metals to be compared is heated slowly and reaction signals are observed as the furnace temperature reaches the appropriate temperature to produce a beam of each metal. Due to the open design of the furnace (described in Apendix III), the lower melting point metal supply is generally exhausted before a beam of the second metal is detected. Thus the signals for the two reactions are measured at different, unknown, furnace temperatures. The geometry of the source also allows a variation in the overlap between the oxygen beam and the two metal beams.

Both techniques have their weak points. We consider our results to be in essential agreement with those of Cohen, and believe that the differences could be reduced by installing a furnace which allows greater control of the metal beam source. For other systems studied by both Lo and Cohen, the agreement is generally satisfactory, less than a factor of ten in most cases.

VI. Technical Developments

In addition to the specific reactions which have been studied, we have developed a number of generally useful experimental techniques. Many of these have been in the area of neutral beam sources. The two-stage furnace which was used to produce an atomic sulfur beam has already been described in Section II. Although the idea of using a two-stage furnace is not new, our experiences have demonstrated that a simple tube-type gas dissociation furnace can be readily adapted to a two-stage design for use with solid samples.

A great deal of effort has been devoted to improving the tungsten foil tube furnace which was used in the early phases of this reasearch project as a uranium beam source. The present design consists of three tungsten rods mounted to form a vee-shaped trough. A chip of uranium is held in the trough by glue which burns away as the furnace is heated, disappearing completely before the uranium begins to vaporize. This furnace has a usable life of five to ten hours with uranium, nearly an order of magnitude longer than the earlier tube furnace.

There are some disadvantages associated with this design. Due to the large physical area over which the beam may be formed, neither the beam geometry nor the beam temperature is well-defined. The source is not well suited for two element beams because the beams of the two metals are produced from different areas of the furnace and are not co-linear.

The ozone beam source design has proved most successful. This all-glass rf discharge source is described in detail in Appendix VIII. The purity of the ozone beam which is produced has not yet been accurately determined due to a lack of data concerning the electron impact ionization cross section and fragmentation pattern of ozone. The source is presently being used, in connection with another project, to measure these parameters.

We have also demonstrated that dc negative ion currents can be routinely measured using an electron multiplier and an Extranuclear Laboratories Negative/Positive Preamplifier. The isolation provided by the preamp allows changing from positive to negative ion detection by simply changing the bias voltages on the ends of the electron multiplier and reversing the polarity of the focusing lens voltages.

VII. Conclusions

The principal conclusion of the present research program is that associative ionization between ground state reactants is not an uncommon process and that it can be a significant source of long-lived electron clouds in the upper atmosphere through the introduction of any of a number of heavy metals. While negative ions may also be produced in some of the reactions, the negative ion channels account for no more than 5% of the total reaction cross section in any of the systems examined thus far.

Knowing the absolute cross sections for the U + O and U + O_2 reactions has made it straightforward to obtain cross sections for other associative ionization reactions. These cross section measurements all contain at least one factor which is a ratio of usually poorly known electron impact ionization cross sections, pointing out a need for improved electron impact cross section data. Presently stated results explicitly show this factor so that improved values for these associative ionization cross sections can be generated as soon as improved EI cross sections become available.

It is not yet possible to predict the combinations of reactants which will associatively ionize. Additional data on the ionization potentials and dissociation energies of heavy metal radicals appear to be a necessary, although probably not sufficient, requirement for developing a predictive capability. Not all of the reaction channels known to be energetically allowed have been observed, however for every combination or reactants known to have one or more allowed channels, at lease one channel has been observed.

There remains much to be learned about associative ionization. The possibilities of internal excitation of the product positive ions and the energy distributions of the electrons have not yet been fully examined. There is no doubt as to the basic scientific interest of this entire field and it is quite possible that the processes may also have significant bearing on military communications problems, absolute neutral atom detection, and perhaps isotope separation and other processes in which chemi-ionization between ground state reactants represents an energetically economical means of ionization.

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APPENDIX I

ASSOCIATIVE IONIZATION REACTIONS STUDIED AT EXTRANUCLEAR LABORATORIES, INC. AND THE UNIVERSITY OF PITTSBURGH UP TO DECEMBER 31, 1976

	Reaction	Observers	Method	Cross Section (cm^2) See Note 1
1.	$U + O \rightarrow UO^+ + e$	1,2,3	A	1.6×10^{-15}
2.	$Th + O \rightarrow ThO^+ + e$	1,3	A	1.0×10^{-15}
3.	$Zr + O \rightarrow ZrO^+ + e$	1,3	A	1.3×10^{-15}
4.	$Gd + O \rightarrow GdO^+ + e$	1,3	A	8.4×10^{-16}
5.	$Ti + O \rightarrow TiO^+ + e$	1,3	A	4.5×10^{-15}
6.	$La + O \rightarrow LaO^+ + e$	3	A	5.1×10^{-15}
7.	$Sm + O \rightarrow SmO^+ + e$	3,6	A	1.7×10^{-16}
8.	$Nd + O \rightarrow Nd\ O^+ + e$	3,6	A	2.7×10^{-15}
9.	$Y + O \rightarrow YO^+ + e$	3	A	3×10^{-16}
10.	$Ce + O \rightarrow CeO^+ + e$	3	A	1×10^{-16}
11.	$Pr + O \rightarrow PrO^+ + e$	3	A	2×10^{-15}
12.	$Dy + O \rightarrow DyO^+ + e$	3	A	1×10^{-17}
13.	$Er + O \rightarrow ErO^+ + e$	3	A	1×10^{-18}
14.	$U + O_2 \rightarrow UO_2^+ + e$	1,2,3	A	1.7×10^{-17}
15.	$Th + O_2 \rightarrow ThO_2^+ + e$	1,3	A	1.5×10^{-17}
16.	$La + O_2 \rightarrow LaO_2^+ + e$	3	A,B	1.7×10^{-17}
17.	$\rightarrow LaO^+ + O + e$	3	A,B	1.2×10^{-17}
18.	$\rightarrow LaO^+ + O^-$	6	A	10^{-18}
19.	$Ce + O_2 \rightarrow CeO_2^+ + e$	3	A	3×10^{-19}
20.	$\rightarrow CeO^+ + ?$	3	A	1×10^{-20}

Reaction	Observers	Method	Cross Section (cm^2) See Note 1
21. $Pr + O_2 \rightarrow PrO_2^+ + e$	3	A	4×10^{-19}
22. $U + O_3 \rightarrow UO_2^+ + O + e$	5,6	A	2×10^{-16}
23. $\rightarrow UO^+ + O_2 + e$	5,6	A	2×10^{-16}
24. $Th + O_3 \rightarrow ThO_3^+ + e$	5,6	A	2×10^{-19}
25. $\rightarrow ThO_2^+ + O + e$	5,6	A	1×10^{-17}
26. $\rightarrow ThO_2^+ + O^-$	5,6	A	2×10^{-18}
27. $\rightarrow ThO^+ + O_2 + e$	5,6	A	4×10^{-16}
28. $\rightarrow ThO^+ + O_2^-$	5,6	A	1×10^{-18}
29. $U + Cl_2 \rightarrow UCl_2^+ + e$	5,3	A,B	1×10^{-17}
30. $\rightarrow UCl^+ + Cl + e$	5,3	A,B	5×10^{-19}
31. $\rightarrow UCl^+ + Cl^- + e$	5	A	5×10^{-19}
32. $La + Cl_2 \rightarrow LaCl_2^+ + e$	3	B	1.1×10^{-16}
33. $\rightarrow LaCl^+ + ?$		B	1.2×10^{-16}
34. $Y + Cl_2 \rightarrow YCl_2^+ + e$	3	B	9×10^{-18}
35. $\rightarrow YCl^+ + ?$	3	B	4×10^{-19}
36. $U + Br_2 \rightarrow UBr_2^+ + e$	5	A	5×10^{-19}
37. $\rightarrow UBr^+ + Br + e$	5	A	1×10^{-19}
38. $\rightarrow UBr^+ + Br^-$	5	A	1×10^{-20}
39. $U + NO_2 \rightarrow UO_2^+ + N + e$	3	B	See Note 2
40. $U + N_2O \rightarrow UO^+ + N_2 + e$	4,5,3,6	A,B	3×10^{-18} See Note 2
41. $La + N_2O \rightarrow LaO^+ + N_2 + e$	3	B	1.5×10^{-15}

	Reaction	Observers	Method	Cross Section (cm^2) See Note 1
42.	$U + NO \rightarrow UNO^+ + e$	4,3	A,B	4.4×10^{-19}
43.	$U + SF_6 \rightarrow UF_2^+ + SF_4 + e$	4,5	A,B	Small
44.	$U + SF_4 \rightarrow UF_2^+ + SF_2 + e$	5	A	5×10^{-19}
45.	$\rightarrow UF_2^+ + SF + F^-$	5	A	10^{-20}
46.	$U + S \rightarrow US^+ + e$	5,6	A	3×10^{-18}
47.	$Ba + OH \rightarrow BAOH^+ + e$	3	A	Confirming Cohen *et al.*

Observers:

1. W. L. Fite
2. P. Irving
3. H. H. Lo
4. B. N. Kim
5. M. W. Siegel
6. T. A. Patterson

Methods:

A. Crossed Beams
B. Beam through gas or magnetic bottle

Note 1: All of the cross sections must be multiplied by the ratio of the electron impact ionization cross section of the metal to that of uranium. The cross sections for reactions 22-47 must also be multiplied by the ratio of the ionization cross section of the gas to that of O_2.

Note 2: $U + N_2O$ in a crossed beam experiment gave a value of 3×10^{-18} cm^2. However, the value which came from a magnetic bottle experiment was over 10^{-17} cm^2. We are now trying to understand this discrepancy in terms of systematic errors, probably in the bottle experiment, since a repeat of the $U + N_2O$ crossed beam experiment by a different observer under different conditions also gave 3×10^{-18} cm^2. Only the bottle experiment has been used to date for the $U + NO_2$ reaction so we are reluctant to cite a value at this time.

APPENDIX II

PRESENTED AT THE IX INTERNATIONAL CONFERENCE ON THE PHYSICS OF ELECTRONIC AND ATOMIC COLLISIONS, SEATTLE, JULY 1975

STUDIES OF POSITIVE AND NEGATIVE ION ASSOCIATIVE IONIZATION IN CROSSED BEAMS OF URANIUM AND N_2O, SF_6, Br_2, H_2O_2, CHF_3 AND O_2*

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We have recently surveyed six gas-uranium systems seeking associative ionization processes leading to positive and negative ions. In two cases we have been able to determine estimates for associative ionization cross sections relative to the 1.68 ± 0.27 x 10^{-17} cm^2 cross section for UO_2^+ from U and O_2, previously investigated by Fite _et al._[1] The apparatus and technique used herein are essentially identical to those described in Ref. 1. The only significant change is the addition of a two port gas handling system to allow rapid interchange of gases for normalization purposes.

Quantitative measurements have been made on the UO^+ from U and N_2O system, where the reaction is

$$U + N_2O \rightarrow UO^+ + e$$

The cross section for this process is estimated at 14% of the cross section for UO_2+ from U and O_2. This measurement does not yet include several corrections (e.g., O_2 and N_2O beam intensities by electron bombardment, mass discrimination effects, etc.) which will have to be carefully determined before a precise result can be stated. As would be expected, no negative ions were detected.

In the U + SF_6 system the UF_2^+ ion is observed, but no negative ion is found. The process involved is thus presumed to be

$$U + SF_6 \rightarrow UF_2^+ + SF_4 + e$$

Its cross section appears to be small. The UF_2^+ is detected at a convenient signal level when the reaction chamber partial pressure due to the SF_6 beam is of the order of 5×10^{-6} torr. When an O_2 beam producing a pressure increase of 1×10^{-6} torr is mixed with the SF_6 beam, the UO_2^+ signal dominates the UF_2^+ signal by a factor of approximately 7, apparently indicating that the cross section for producing UF_2^+ from U and SF_6 appears to be about 3% of the cross section for producing UO_2^+ from U and O_2.

In the U + Br_2 system the UBr_2^+ ion was observed but the UBr^+ was not. Again, negative ions were sought and none observed.

Positive ions (only) were sought in the U + H_2O_2 system, but none were observed. Obtaining this null result required a variation on the usual procedure, necessitated by the inseparable admixture of O_2 with the H_2O_2 beam; thus an associative ionization signal is always observed, and it was at first believed to be due to UO_2^+ (or perhaps UO_2H^+) from the U + H_2O_2 reaction. To distinguish signal from H_2O_2 from signal O_2 we used a mass stepping device to switch approximately once per second between UO_2^+ and O_2^+; the electron beam was switched on only during the O_2^+ dwell periods. The signals were demultiplexed to sample-and-hold amplifiers, and an x-y plot was made of the UO_2^+ vs. O_2^+ signals as the source pressure of O_2 and (H_2O_2 + O_2) were separately varied. These plots had identical slopes, indicating that all of the UO_2^+ signal was due to the O_2 reaction.

Positive ions (only) were sought in the U + CHF_3 (Freon 23) system, but a null result was obtained.

In addition to the above newly investigated systems, negative ions were sought in the U + O_2 system; again, as expected a null result was obtained.

The null results in our search for negative ions from N_2O, O_2, Br_2, and SF_6 + U are disappointing, especially in the latter two cases. We will have to continue to doubt the proper functioning of the apparatus until such time as some associative ionization negative ion is observed. However the experimental procedure is entirely symmetric for negative and positive ions, and we believe that if any of these systems do lead to negative ions, the cross sections are more than an order of magnitude below the smallest positive ion cross section observed to date.

We anticipate that by conference time we will be able to discuss one or two of these new results in quantitative detail; our search for negative ions will also continue, and we hope to be able to report at least one non-null result.

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[1] W. L. Fite, H. H. Lo, and P. Irving, J. Chem. Phys. 60, 1236, (1974)

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APPENDIX III

PRESENTED AT THE AMERICAN SOCIETY FOR MASS SPECTROMETRY CONFERENCE, HOUSTON, MAY 1975

ASSOCIATIVE IONIZATION AND SECONDARY REACTIONS IN $U + O_2$ AND $U + N_2O$ SYSTEMS

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Crossed beam experiments have yielded the cross section for $U + N_2O \rightarrow UO^+ + N_2 + e$, from the previously measured cross section for $U + O_2 \rightarrow UO_2^+ + e$. In a second experiment a beam of U atoms crossed a magnetic bottle field where gas was added. Ions formed and trapped in the bottle field were examined. With N_2O, UO_2^+ was also present, arising from the ion-molecule reaction $UO^+ + N_2O \rightarrow UO_2^+ + N_2$. With O_2, UO_3^+ was also present but arising, we believe from energy considerations, from the associative ionization process $UO^* + O_2 \rightarrow UO_3^+ + e$, with the UO coming from $U + O_2 \rightarrow UO^* + O$. The appearance of UO_3^+ suggests possible errors in the known energetics of the uranium-oxygen system. The experiments are described and the results discussed. Possible uses of magnetically confined plasmas of such ions are considered.

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APPENDIX IV

PRESENTED AT THE V INTERNATIONAL CONFERENCE ON
ATOMIC PHYSICS, BERKELEY, JULY 1976

STUDY OF THE ASSOCIATIVE IONIZATION REACTION BETWEEN URANIUM AND SULFUR*

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We are continuing our survey of associative ionization reactions involving uranium atoms by examining the reaction

$$U + S \rightarrow US^+ + e. \qquad (1)$$

Since the analogous reaction with atomic oxygen, which like atomic sulfur has an $ns^2\ np^4\ \{^3P_2\}$ structure, proceeds with a relatively large cross section,[1] $1.62 \pm 0.41 \times 10^{-15}$ cm^2, it is reasonable to expect that this reaction would occur.

The experiment is performed in a crossed beams configuration using an apparatus and technique which are essentially identical to those described in Ref. 1. A two-stage oven has been added to the system to produce atomic beams from molecular solids. The solid charge is vaporized in the first stage and then dissociated in the hotter second stage. Generally, a single stage oven hot enough to dissociate a molecule will vaporize the charge too rapidly to produce a usable beam.

The cross section for reaction (1) is measured to be of order 3×10^{-18} cm^2, based on a comparison with the $1.68 \pm 0.27 \times 10^{-17}$ cm^2 cross section for UO_2^+ from U and O_2, previously investigated by Fite et al.[1] This value does not include corrections for such factors as the relative efficiencies of the O_2 and S beam intensity measurements by electron bombardment, detailed examination of the S_2 dissociation fraction and mass discrimination effects. These factors will have to be determined to yield a more accurate result.

A cursory examination of the reaction

$$U + S_2 \rightarrow US_2^+ + e \qquad (2)$$

yielded a null result. However, considering beam intensities and the ratio of the O and O_2 cross sections, the US_2^+ product could easily have escaped detection. Insufficient data exists on the dissociation energy and ionization potential of US_2 to determine whether the reaction is even energetically possible.

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[1] W. L. Fite, H. H. Lo and P. Irving, J. Chem. Phys. 60 1236 (1974)

*This research was supported by the Defense Nuclear Agency and monitored by the Air Force Geophysics Laboratory under Contract Number F 19628-74-C-0215

APPENDIX V

PRESENTED AT THE V INTERNATIONAL CONFERENCE ON ATOMIC PHYSICS, BERKELEY, JULY 1976

STUDIES OF POSITIVE AND NEGATIVE ION ASSOCIATIVE IONIZATION IN CROSSED BEAMS OF URANIUM AND Br_2, Cl_2, NO_2, SF_4, $(CN)_2$ AND H_2S*

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In a previous report[1] on our crossed beam studies of associative ionization between uranium atoms and various gases we reported observing the reaction

$$U + Br_2 \rightarrow UBr_2^+ + e \qquad (1a)$$

with a null result for the channels

$$U + Br_2 \rightarrow UBr^+ + Br + e \qquad (1b)$$

and

$$U + Br_2 \rightarrow UBr^+ + Br^- \qquad (1c)$$

Further investigation under more favorably experimental conditions now indicates that reactions (1b) and (1c) do indeed occur. This appears to be the first reported associative ionization reaction leading to a negative ion product. By comparison of observed signal intensities in these cases with the UO_2^+ signal from the reaction $U + O_2 \rightarrow UO_2^+ + e$ under comparable gas beam intensity conditions, we roughly estimate cross sections of 5×10^{-19} cm^2 for (1a), and 1×10^{-19} cm^2 for the sum of (1b) and (1c). The branching ratio between (1b) and (1c) is difficult to estimate, but appears to be the order of 10:1.

We have similarly investigated the reactions of Cl_2 with uranium and observed

$$U + Cl_2 \rightarrow UCl_2 + e \qquad (2a)$$

$$U + Cl_2 \rightarrow UCl^+ + Cl + e \qquad (2b)$$

and

$$U + Cl_2 \rightarrow UCl^+ + Cl^- \qquad (2c)$$

with roughly estimated cross sections of 10^{-17} cm^2 for (2a), and 10^{-18} cm^2 for the sum of (2b) and (2c). The branching ratio between (2b) and (2c) is, as in the analogous bromine case, difficult to estimate, but appears to be of order unity.

The reaction

$$U + NO_2 \rightarrow UO_2^+ + N + e \qquad (3)$$

previously reported by Fite, Lo and Vasu[2] in a magnetic bottle configuration, has now been observed in the crossed beam configuration. Its cross section is estimated at 3×10^{-18} cm^2. No negative ion channel is possible.

The reactions

$$U + SF_4 \rightarrow UF_2^+ + SF_2\ (?) + e \qquad (4a)$$

and

$$U + SF_4 \rightarrow UF_2^+\ (?) + F^- + SF\ (?) \qquad (4b)$$

have been observed, in interesting contrast to the previously reported[1] reaction $U + SF_6 \rightarrow UF_2^+ + SF_4 + e$ where no negative ion channel is observed. The cross section for (4a) is estimated at 5×10^{-19} cm^2, while the cross section for (4b) is so small as to preclude even a rough estimate at this time. The positive ion product in (4b) is shown as uncertain because the F^- signal is sufficiently small that a correspondingly small current of a heavy positive ion, e.g., UF^+, might easily elude detection.

The systems U + $(CN)_2$ and U + H_2S were examined and not found to yield ionized products.

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2. W. L. Fite, H. H. Lo and P. Vasu, Ibid., p. 1086.

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APPENDIX VI

PRESENTED AT THE APS DIVISION OF ELECTRON AND ATOMIC PHYSICS MEETING, LINCOLN, DECEMBER 1976

OZONE ASSOCIATIVE IONIZATION WITH THORIUM AND URANIUM*

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Associative ionization reactions between ozone and both uranium and thorium have been studied in a crossed beams experiment. The positive ion products MO_3^+, MO_2^+, MO_2^+ and MO^+, the neutral products O and O_2, and the negatively charged products e, O^- and O_2^- group into seven associative ionization channels. In the Th-O_3 system all five ion products are observed, indicating that at least three channels are open. Relative intensities of positive and negative ion signals indicate that at least five channels are actually open. The channels leading to ($ThO^+ + O + O + e$) and ($ThO^+ + O + O^-$) can probably be excluded on energetic grounds, so cross sections for the five distinct channels can be obtained from the data. In the U-O_3 system, UO^+ and UO_2^+ but not UO_3^+ are observed, in agreement with known energetics. Neither O_2^- nor O^- are observed, although energetically allowed. Measured cross sections span the range 10^{-19} to 10^{-16} cm^2.

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APPENDIX VII

PRESENTED AT THE APS DIVISION OF ELECTRON AND ATOMIC PHYSICS MEETING, LINCOLN, DECEMBER 1976

LANTHANUM-OXYGEN ASSOCIATIVE IONIZATION*

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Multiple reaction channels have been observed in heavy metal-molecular halide associative ionization reactions.[1] We report the first observation of multiple channels in a metal-molecular oxygen system. Associative ionization between lanthanum and oxygen has been studied in a crossed beams experiment. In addition to the reaction

$$La + O_2 \rightarrow LaO_2^+ + e$$

which has been observed in many similar metal-oxygen systems,[2] LaO^+ and O^- are also produced. The cross sections for the first two reactions are of order 10^{-17} cm^2, while that of the third is significantly smaller.

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*Supported by the Defense Nuclear Agency and monitored by the Air Force Geophysics Laboratory under Contract Number F 19628-74-C-0215 and NSF

[1] M. W. Siegel and W. L. Fite, Abstracts of the V ICAP (Berkeley, CA, 1976), R. Marrus, M. H. Prior and H. A. Shugart (Eds.), p. 135.

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APPENDIX VIII

ASSOCIATIVE IONIZATION OF URANIUM AND THORIUM IN COLLISIONS WITH OZONE*

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ABSTRACT

Associative ionization cross sections for U and Th with O_3 have been measured using a crossed beams technique. Thermochemical data indicate five energetically allowed reaction channels for each metal. Only two of these channels are observed in the U + O_3 system. All five channels, including two leading to the production of negative ions, are observed in the Th-O_3 system. The total reaction cross section for both metals is about 4×10^{-16} cm^2, with individual channel cross sections spanning the range 2×10^{-19} cm^2 to 4×10^{-16} cm^2.

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ASSOCIATIVE IONIZATION OF URANIUM AND THORIUM IN COLLISIONS WITH OZONE

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I. Introduction

Associative ionization reactions between lanthanide and actinide heavy metals and a variety of atomic and simple molecular species have been investigated recently.[1-8] Metal-atom associative reactions have the simple form

$$M + A \rightarrow MA^+ + e \qquad (1)$$

Metal-molecule systems are more complex because multiple dissociative-associative reactions channels, including some leading to negative ions, are possible in addition to the simple associative reactions. For many of the heavy metal systems, insufficient molecular binding energy data are available to predict which, if any, of the associative ionization channels are energetically allowed. Turning this deficiency around, the existence or absence of open associative ionization channels may be used to place bounds on certain ionization potentials and dissociation energies.

In this paper we report several associative ionization reactions between ozone and uranium and thorium. Seven associative ionization reaction channels are in principle possible:

$$M + O_3 \rightarrow MO_3^+ + e \qquad (2)$$

$$\rightarrow MO_2^+ + O + e \qquad (3)$$

$$\rightarrow MO_2^+ + O^- \qquad (4)$$

$$\rightarrow MO^+ + O_2 + e \qquad (5)$$

$$\rightarrow MO^+ + O_2^- \qquad (6)$$

$$\rightarrow MO^+ + O + O + e \qquad (7)$$

$$\rightarrow MO^+ + O + O^- \qquad (8)$$

Reactions (2)-(6) are known to be exothermic for both uranium[9] and thorium[10] while (7) and (8) are known to be endothermic by more than 1.5 eV for both metals. Table I summarizes the exothermicity of the reactions. Exothermicity provides only a measure of whether the reaction is permitted, but does not guarantee that the reaction proceeds with sufficient cross section to be observable. Experience has indicated that most but not all allowed reactions are in fact observed. This would be expected on the simple grounds that an enormous number of low lying molecular states exist in these systems, almost assuring one or more appropriate curve crossings in each case.

II. Apparatus

The experiments are performed in the crossed beams apparatus shown schematically in Figure 1. The metal vapor beam and the modulated ozone beam intersect at right angles in the analyzer chamber of a two-chamber, differentially pumped vacuum system. Ions produced by associative ionization are electrostatically extracted from the interaction volume and focused into the entrance of a quadrupole mass filter. Ions of a selected m/e are detected with a continuous dynode (Channeltron®) multiplier. An electron beam can also be turned on to pass through the interaction volume at right angles to the gas and metal beams. The beam intensities can thus be measured by electron bombardment ionization mass spectrometry.

The metal vapor beam is formed by placing a chip of the metal in a horizontal trough formed by three tungsten rods. The rods are resistively heated to a normal operating temperature of 2000-2200 K. There are three collimating aperatures between the beam source and the interaction volume. This very simple technique has been found to be a more satisfactory beam source than the tungsten foil tube furnaces which were used in earlier experiments.[1] The source has an operating life of 8-12 hours with uranium and can easily be replenished many times when used with a less reactive metal.

Ozone is produced in an rf discharge in flowing oxygen at a pressure of 1-10 Torr. The all-glass and teflon ozone source is illustrated in Figure 2. In order to keep O and O_2 discharge products out of the beam and to eliminate electrical noise interference, the ozone is produced and released cyclically. The ozone produced in the discharge is first collected as a solid by cooling the discharge bulb in liquid nitrogen. After collecting approximately .5 cm^3 of solid ozone, the discharge and oxygen flow are turned off. The ozone beam is produced by allowing the solid ozone to evaporate. The ozone effuses from the nozzle of the source into the source chamber of the vacuum system. The nozzle is approximately 1 cm from the beam defining aperture which connects the source chamber with the analyzer chamber.

One of the difficulties in working with a beam of an unstable species such as ozone is to assure that the beam survives the transit from the source to the interaction region. This is particularly important in this case since the ozone dissociation product impurities, O_2 and possibly O, will also react with the metal beam, producing some of the same ions which are expected in the metal-ozone reaction. The only method available for monitoring the ozone beam is to observe the ions produced by electron impact ionization. This introduces a problem in that the ions produced from the impurities by electron impact are also ozone dissociative ionization products. Thus a large O_2^+ signal is not necessarily an indication of an O_2 impurity in the beam. Because the electron impact ionization cross section and fragmentation pattern of ozone is not known, it is not possible to determine the relative concentrations of O, O_2 and O_3 in the beam by observing the ratios of the O^+, O_2^+ and O_3^+ ion signals. Mass discrimination effects, to be discussed later, would complicate this approach even if the necessary cross section data were available.

Modulating the beam with a rotating toothed wheel permits some information about the purity of the beam to be extracted. By using a frequency selective detection system tuned to the modulation frequency, the ion signals produced from beam components can be separated from ion signals produced from the background gas. If the phases of the ion signals, relative to some reference signal generated by the modulator, are also monitored, information about the parent neutral(s) may also be obtained, a technique known as phase spectrometry.[11]

Phase spectrometry is a form of time-of-flight beam analysis. Since neutrals of different masses coming from a source at a given temperature have different velocities, they will have different times of flight in traversing the distance between the modulator and the ionization region. Hence the signal phase of an ion formed by a simple ionization of a neutral will be different from the phase of the same ion formed by dissociative ionization of a more complex neutral. The phase of a particular ion signal depends on the total time of flight, i.e., the neutrals' flight time from the modulator to the ionization region plus the ions' time from the ionization region to the detector. Although the phase is not quite a linear function of the time of flight characteristic of a molecule with the mean velocity, it can be calculated[12] using the path lengths, the source temperature and the ion energy.

The ion signals are treated as two dimensional vectors characterized by an amplitude and a phase. The expected phases of O_3^+ from O_3, O_2^+ from O_3 and O_2, and O^+ from O_3, O_2 and O are calculated and compared with the observed signal phases. In principle, the observed signal vector of an ion can be resolved into components along the possible source phase axes. This information, together with the relative cross sections for the various ionization reactions, yields a complete description of the neutrals present in the beam.

In practice, the requirement that the ozone beam path length be kept short to maximize the beam intensity in the interaction region leads to the situation that the uncertainty of a phase measurement, typically ± 1-2°, is 10-20% of the phase difference to be detected. The observed signal phases indicate that there are probably small amounts of O_2 in the ozone beam, but without increased phase separation and knowledge of the relative ionization cross sections, it is difficult to make estimates of the impurity concentrations.

III. Procedure

Absolute cross sections are obtained by comparing the product ion current with the product ion current for a similar reaction whose absolute cross section has been carefully measured in another apparatus. The uranium[3] and thorium[4] associative ionization reactions with oxygen,

$$M + O_2 \rightarrow MO_2^+ + e \qquad (9)$$

whose cross sections are $(1.68 \pm 0.27) \times 10^{-17}$ cm^2 and 1.5×10^{-17} cm^2 respectively, are used as the calibration reactions. The effective associative ionization cross section is proportional to the resultant ion current divided by the product of the ion currents produced by electron impact ionization of the neutral reactant beams.

In practice an oxygen beam is first generated by flowing O_2 through the ozone source. The electron beam is turned on and the modulated O_2^+ and O^+ (from dissociative ionization) signals and the unmodulated M^+ signal are recorded. The electron beam is then turned off and the modulated MO_2^+ associative ionization signal is recorded. The phases of the modulated signals are also noted for later consistency checks.

Ozone is then generated and collected. Using the collected solid ozone at liquid nitrogen temperature as the beam source, the unmodulated M^+ signal and the modulated ozone signals, O_3^+, O_2^+ and O^+, produced with the electron beam on, are recorded. The electron beam is then turned off and the observable metal-ozone associative ionization positive ion product signals are recorded. Phases of the modulated signals are again noted. This technique assures reasonably consistent beam overlaps and allows reliable measurement of relative cross sections for the more intense channels.

By removing the liquid nitrogen bath from the ozone collection bulb, the ozone beam intensity can be increased, allowing observation of ion products from other reaction channels having smaller cross sections. As the solid ozone warms, the gas load presented by the source causes the vacuum system pressure to rapidly approach the upper safe operating limit making it necessary to reimmerse the source in liquid nitrogen. This transient nature of the more intense beam requires a modified data acquisition technique.

The quadrupole mass filter is programmed to alternately transmit two different masses, switching masses at one second intervals. The transmitted ion signal is demultiplexed and displayed as two traces on a chart recorder. A previously observed ion product and, at higher signal gain, the mass corresponding to another possible ion product are monitored as the ozone beam intensity is increased. If a new product is observed, the ratio of the reaction channel cross sections is equal to the ratio of the observed ion signals.

This technique is also used to observe the negative ions, O^- and O_2^- which are produced in the thorium-ozone associative ionization reaction. A simultaneous comparison of positive and negative ion yields is not possible with the present system; however the metal atom beam is sufficiently stable and the ozone pressure buildup sufficiently reproducible that reasonably accurate results can be obtained.

IV. Analysis

The observed associative ionization molecular positive ion signal is given by

$$S_3(MO_i^+) = K_3 \, Q_3(MO_i^+) \, I_3 \, n_3 \tag{10}$$

where $Q_3(MO_i^+)$ is the effective cross section for the production of MO_i^+ (i = 1,2 or 3) in the M-O_3 reaction. I_3 and n_3 are, respectively the neutral ozone current and the metal number density in the neutral beams. The apparatus constant, K_3, includes the overall detection efficiency for the product ions and overlap intergral of the neutral beams. Similarly, the calibration reaction ion signal can be expressed as

$$S_2(MO_2^+) = K_2 \, Q_2(MO_2^+) \, I_2 \, n_2. \tag{11}$$

The apparatus constants K_3 and K_2 are assumed to be equal. The ion detection efficiency includes the efficiency with which the ions are drawn into the mass filter, the transmission efficiency for the ions and the efficiency of detection at the electron multiplier. Mass discrimination effects in the ion detection can be neglected because the differences in product ion masses are small. Since the source geometry is the same for the oxygen and ozone beams and the metal atom beam is relatively stable, the overlap integrals are approximately equal.

Combining Equations (10) and (11), the ratio of the effective cross section is

$$\frac{Q_3(MO_i^+)}{Q_2(MO_2^+)} = \frac{S_3(MO_i^+)}{S_2(MO_2^+)} \frac{n_2}{n_3} \frac{I_2}{I_3}. \qquad (12)$$

The first term is simply the ratio of the observed ion signals. The second term is equal to the ratio of the electron impact ionization metal ion signals recroded during the data runs. This ratio corrects for the small drift in metal beam intensity during the course of the measurements. The third term is the ratio of the O_2 and O_3 neutral beam currents which can be expressed in terms of the number densities and mean molecular speeds as

$$\frac{I_2}{I_3} = \frac{n_2}{n_3} \frac{\bar{v}_2}{\bar{v}_3} . \qquad (13)$$

By considering the source temperature and pressure and the length and diameter of room temperature tubing connections the source to the vacuum chamber, it can be shown that the ozone is thermalized from liquid nitrogen temperature to room temperature by the time it enters the beam. Thus the mean molecular speeds are inversely proportional to the square roots of the masses, yielding

$$\frac{I_2}{I_3} = \frac{n_2}{n_3}\left(\frac{M_3}{M_2}\right)^{1/2} = \sqrt{\frac{3}{2}}\ \frac{n_2}{n_3}\ . \tag{14}$$

When the ion optics are optimized for maximum sensitivity to the higher mass reaction product ions, significant mass dependent variations in ion transmission may occur at low masses. To avoid transmission factors which are difficult to evaluate, both n_2 and n_3 are expressed in terms of O_2^+ ion signals. Assuming that the ozone beam contains no O_2, the ratio of the neutral beam currents becomes

$$\frac{I_2}{I_3} = 1.2\ \frac{S_2(O_2^+)}{Q_2^e(O_2^+)}\ \frac{Q_3^e(O_2^+)}{S_3(O_2^+)} \tag{15}$$

where $Q_2^e(O_2^+)$ and $Q_3^e(O_2^+)$ are the cross sections for the production of O_2^+ by electron impact on O_2 and O_3, respectively.

Combining Equations (12), (14) and (15), the expression for the ratio of the effective associative ionization cross sections reduces to

$$\frac{Q_3(MO_i^+)}{Q_2(MO_2^+)} = 1.2\ \frac{S_3(MO_i^+)}{S_2(MO_2^+)}\ \frac{S_2(M^+)}{S_3(M^+)}\ \frac{S_2(O_2^+)}{S_3(O_2^+)}\ \frac{Q_3^e(O_2^+)}{Q_2^e(O_2^+)} \tag{16}$$

All but the last term can be evaluated with the experimental data. Dissociative ionization cross sections for ozone are not presently available. For future reference when this cross section becomes available, the electron energy was 50 eV in these experiments. The electron energy for the ratio of the metal ionization cross sections which appears in the thorium-ozone reaction[4] results was 100 eV.

V. Results

Two uranium-ozone and five thorium-ozone associative ionization reaction channels are observed. The reaction and the measured cross sections are presented in Table II. The quoted cross sections are based on the observations of the first reaction product. Because electron and neutral products are not monitored, the reactions are balanced with the most probable additional products.

The run-to-run scatter in the data indicates an experimental uncertainty of ± 30-40%. This is small compared with the estimated errors associated with the assumptions made in the interpretation of the data. Assuming equal apparatus constants for the O_2 and O_3 reactions introduces no more than a 5% uncertainty. The most significant assumption is that the ozone beam contains no O_2 or O.

While phase spectroscopy provides some support for this assumption, apparatus constraints limit the sensitivity of this technique. The sign and the magnitude of the effects of 5-10% O and 10-20% O_2 impurity concentrations depend on the reaction product, the reaction cross section and the cross sections for the M-O and M-O_2 reactions. These effects can produce changes of a factor of 2-4 in the positive ion cross sections in Table II.

The thorium negative ion cross sections are, at best, order of magnitude estimates. Since the negative ion products are observable only in the transient ozone beam mode, simultaneous comparisons with positive ion signals are not possible. The cross sections are based on comparing the negative ion signals with positive ion signals obtained at another time. In addition to uncertainties associated with transmission factors for positive and negative ions which differ in mass by an order of magnitude, the run-to-run reproducibility of the transient ozone beam is difficult to monitor. A conservative interpretation of the data would be that they do no more than confirm that the reactions occur. Because of this large uncertainty, no attempt has been made to subtract the negative ion contributions to the third and fourth reactions.

VI. Conclusions

There are no apparent correlations between the individual reaction channel cross sections and product ion complexity or reaction exothermicity. The total ozone associative ionization cross sections for both thorium and uranium are of order 10^{-16} cm^2. These can be compared with atomic oxygen cross sections which are $\sim 10^{-15}$ cm^2 and molecular oxygen cross sections, $\sim 10^{-17}$ cm^2. As with previous systems in which negative ion products are observed, the thorium-ozone negative ion channels appear to account for less than 1% of the total cross section. The major negatively charged associative ionization product is a free electron.

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TABLE I

REACTION EXOTHERMICITY (eV)

		U	Th
$M + O_3$	$\rightarrow MO_3^+$	+ 4.9	$\geq$ 1.8
	$\rightarrow MO_2^+ + O + e$	+ 3.2	+ 1.8
	$\rightarrow MO_2^+ + O^-$	+ 4.7	+ 3.3
	$\rightarrow MO^+ + O_2 + e$	+ 1.1	$\leq$ 1.8
	$\rightarrow MO^+ + O_2^-$	+ 2.6	+ 2.3
	$\rightarrow MO^+ + O + O + e$	- 4.0	- 3.3
	$\rightarrow MO^+ + O + O^-$	- 2.5	- 1.8

TABLE II

REACTION CROSS SECTIONS

Reaction	Cross Section (cm^2) See Note 1
$U + O_3 \rightarrow UO_2^+ + O + e$	2×10^{-16}
$\rightarrow UO^+ + O_2 + e$	2×10^{-16}
$Th + O_3 \rightarrow ThO_3^+ + e$	2×10^{-19}
$\rightarrow ThO_2^+ + O + e$	1×10^{-17}
$\rightarrow ThO^+ + O_2 + e$	4×10^{-16}
$\rightarrow O_2^- + ThO^+$	1×10^{-18}
$\rightarrow O^- + ThO_2^+$	2×10^{-18}

Note 1

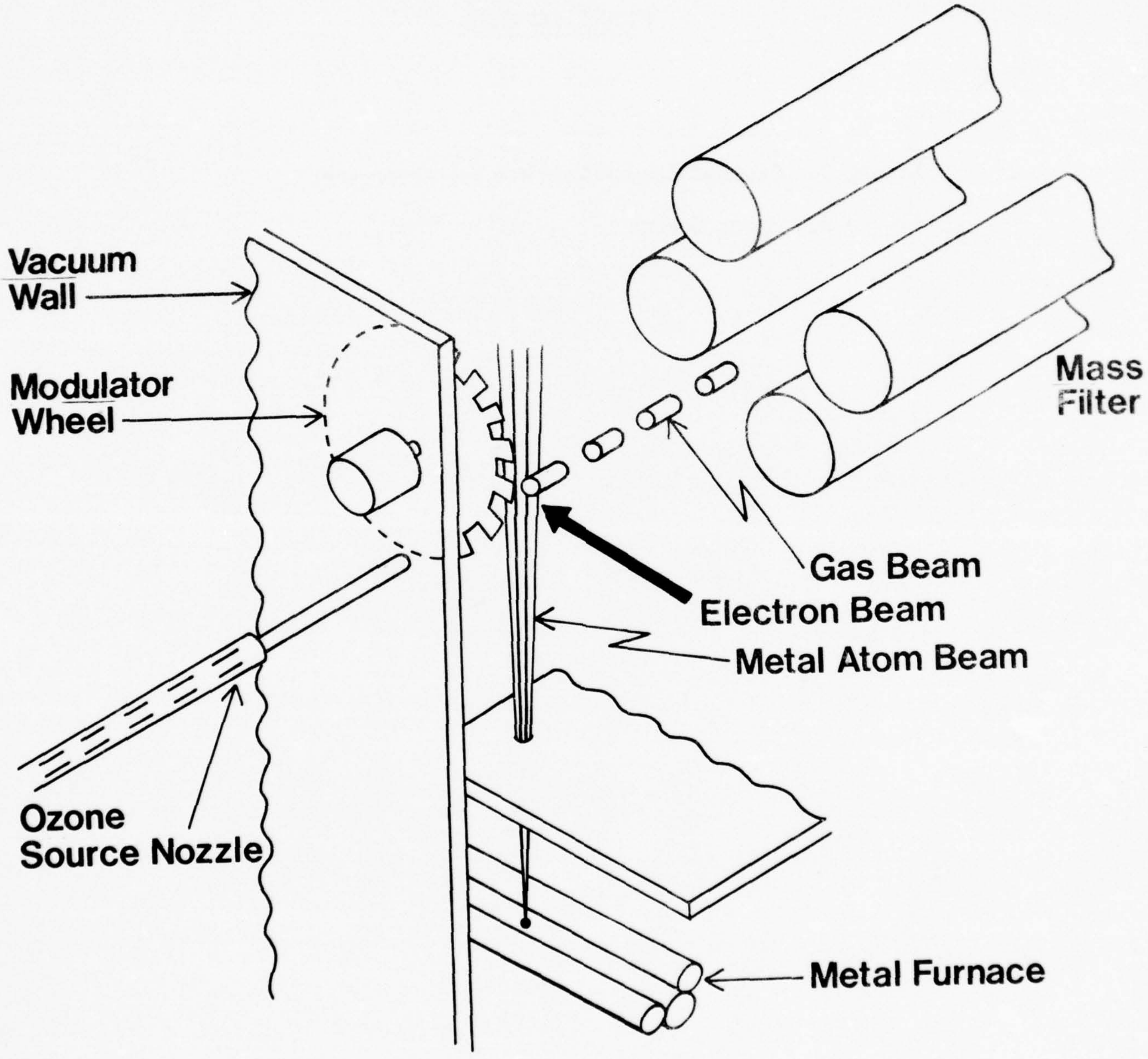
All cross sections must be multiplied by the ratio of the cross sections for the production of O_2^+ by electron impact on O_3 and O_2. The thorium cross sections must also be multiplied by the ratio of the thorium and uranium electron impact ionization cross sections.

FIGURE CAPTIONS

Figure 1. Crossed Beams Experimental Arrangement

Figure 2. Ozone Source

FIGURE 1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Modulator Wheel
Mass Filter
Gas Beam
Electron Beam
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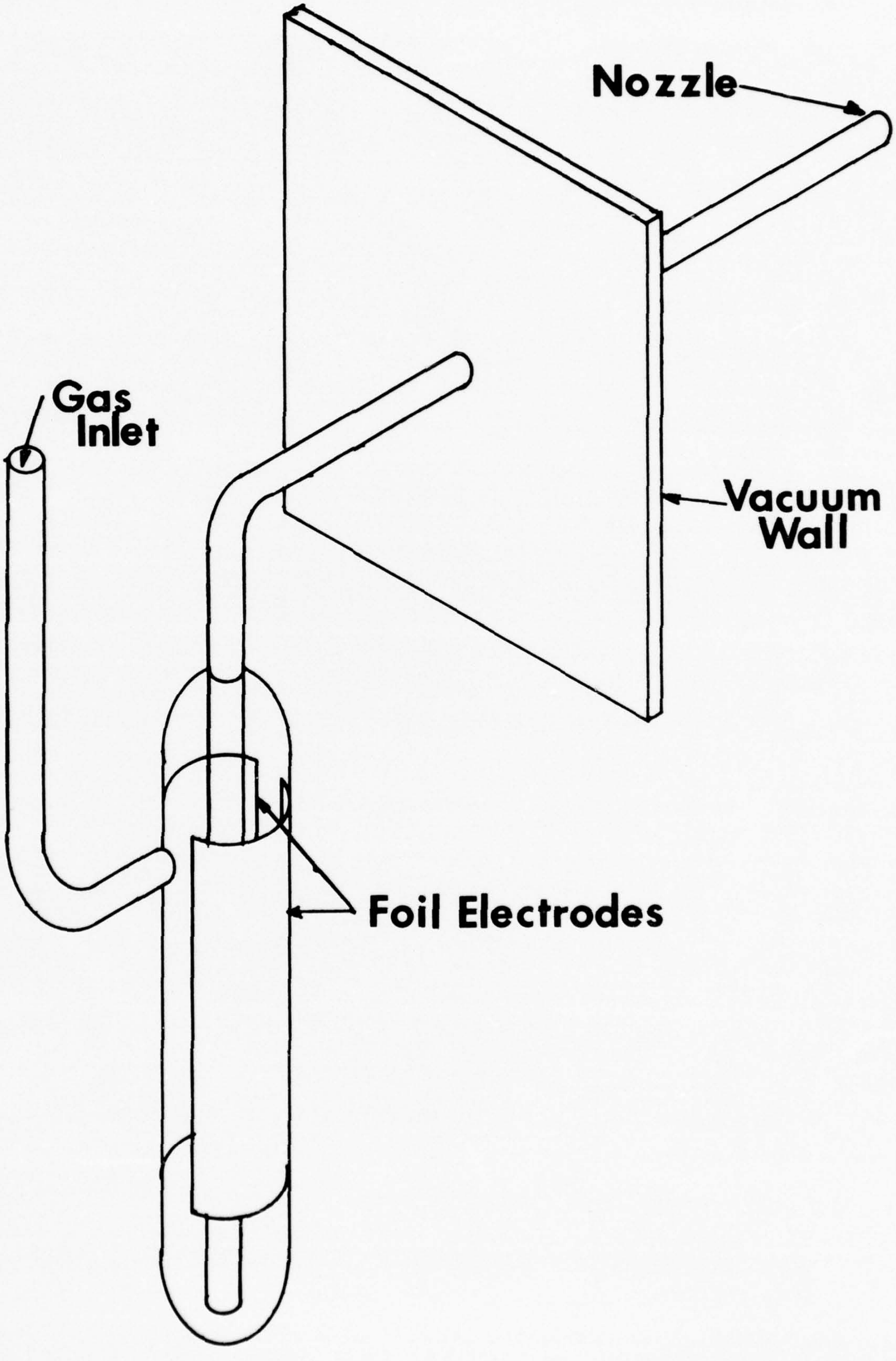
Ozone Source Nozzle
Metal Furnace

FIGURE 2



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