PPPL--2305

DE86 007531

# LONG-TERM CHANGES IN THE SENSITIVITY OF QUADRUPOLE MASS SPECTROMETERS

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## ABSTRACT

We routinely use quadrupole mass spectrometers (QMS) to monitor vacuum conditions, gas purity, and plasma-wall interactions in the Tokamak Fusion Test Reactor (TFTR) at Princeton. Two QMS systems have been operating on TFTR continuously for a two-year period. Both QMS systems are absolutely calibrated at weekly intervals using a six-part standard gas mixture. The calibration procedure is based on the use of transfer standards (ion gauge and capacitance manometer) that are calibrated against a primary standard (spinning rotor gauge) on an external vacuum system. We have identified variations in the efficiency of the QMS ionizer and drifts in the sensitivity of the electron multiplier ion detector to be the major reasons for the observed changes in overall QMS sensitivity. Weekly variations in sensitivity greater than 100% have been observed following system bakeout at 150°C and with the use of rhenium filaments which were initially in the QMS ionizer. Operation of the QMS systems with tungsten filaments and at constant temperature has yielded more stable operation with weekly sensitivity changes generally being less than 10%.

Presented at the 32nd National Symposium of the American Vacuum Society

(Houston, TX, November 1985)

## I. INTRODUCTION

Quadrupole mass spectrometers (QMS) are important instruments for characterizing the vacuum integrity of fusion devices and for studying plasma-material interactions.[1,2] Applications include monitoring the impurity gas production during high power plasmas, quantitative measurements of H/D exchange processes, and monitoring the effectiveness of the cleaning and impurity control procedures[3,4] used in conditioning of vacuum vessels such as: glow discharge cleaning (GDC), pulse discharge cleaning (PDC), bake-out procedures, and gettering. The successful application of a QMS for these tasks requires an instrument that is well characterized and calibrated.

In this paper we describe some of our experiences at the Princeton Plasma Physics Laboratory (PPPL) with QMS systems. We first discuss the design and operation of the QMS systems on the Tokamak Fusion Test Reactor (TFTR), which are dedicated to continuous monitoring of the vacuum environment of this large (86 $m^3$) magnetic fusion device. In Sec. II we review the calibration procedures which were developed for the TFTR QMS systems and are applied at weekly intervals. Finally, in Sec. IV we discuss some of the long- and short-term variations in QMS sensitivity that we have noted from two years of operation.

## II. QMS DESIGN

There are two identical quadrupole mass spectrometer systems (Balzers M/N 511) on TFTR (Fig. 1), located midway on the two vacuum vessel pumping ducts. The QMS includes a dual filament, cross-beam ionizer, a 20-cm long by 0.8-cm diameter quadrupole mass filter, and a 14-stage Cu-Be electron

multiplier ion detector. Each QMS system includes two turbomolecular-pumped, stainless steel, ultrahigh vacuum chambers which can be isolated from each other, as well as from the torus. Each system also contains two ion gauges, a capacitance manometer, a two-zone heating mantle, two in-line orifice valves for pressure reduction, and a gas injection manifold. The gas manifold, which is used for system calibration, has three gas injection valves, and a variable leak valve with pump-out facilities. The ion gauges, valves, and QMS are connected to the main TFTR control and data acquisition computer through a local microcomputer and an optical, serial data highway.

The normal system configuration for monitoring the torus pressure under high vacuum conditions essentially configures the QMS as an appendage pressure gauge: the QMS turbomolecular pumps (TMP) are valved off and all valves between the QMS and the torus are open. The same setup is used during the vacuum vessel bakeout, pulse discharge cleaning and high power pulses in order to measure the quantity of impurity gasses produced and to monitor vacuum integrity.

During glow discharge cleaning (GDC) the TFTR vacuum pressure is held at about 6 mTorr.[3,4] Therefore, a pressure reduction must be maintained between the QMS and the torus. This is accomplished by the two orifice valves, which were fabricated from pneumatic gate valves with small holes (4.8- and 9.4-mm diameter) drilled in the gate plates. Thus, by introducing either one or both of these low conductance orifices between the QMS and the vacuum vessel, reproducible pressure reduction factors of between 10 to 1000 can be obtained, and impurity gas production during GDC may be monitored. In a third configuration, the QMS systems can be isolated from the tokamak and operated as stand-alone vacuum systems for calibrations, cleaning, and maintenance.

## III. CALIBRATION PROCEDURE

In order to trend the changes in sensitivity in the analyzers and to obtain quantitative results during TFTR operations, the two QMS systems are calibrated at weekly intervals. The first step of a calibration consists of isolating the QMS system from the tokamak and pumping and purging the gas manifold with a calibration gas mixture. The calibration gas is an equi-molar mixture of six gasses ($H_2$, $^4He$, $CH_4$, Ne, $N_2$, Ar). With the gas injection line evacuated, one of the gas injection valves is opened and a flow of calibration gas is established in the QMS system by varying the flow rate through the leak valve. When the flow rate is set, the system turbomolecular pumps are isolated and the pressure is allowed to increase to an appropriate level. The gas injection valve is then closed, and a zero pumping speed measurement of the capacitance manometer and ion gauge is made. The calibration gas is then pumped out to high vacuum and the gas flow re-established. This procedure is repeated a few times for pressures from the high $10^{-5}$ to low $10^{-4}$ Torr range in order to obtain an ion gauge factor for the calibration gas by comparing the ion gauge reading to the capacitance manometer. Once the gauge factor is acquired, the calibration gas is injected at lower pressures ($10^{-6}$ to $10^{-5}$ Torr) while monitoring the current levels generated in the QMS for the six different gasses, and simultaneously recording the ion gauge pressure. The gas flow rate and QMS sweep speed are set such that a complete calibration spectrum is obtained in less than 10 seconds from the time the QMS turbomolecular pumps are valved off, in order to minimize the error in measurement from wall pumping and degassing effects. The sensitivity of the QMS (in A/Torr) is derived for each of the six gasses from the linear portion of a plot of partial pressure vs. QMS output current. Comparisons were made between calibration spectra obtained for a full bottle of the calibration gas

and a nearly empty bottle with no perceptible difference observed.

A calibration facility[5] has been set up at PPPL for the calibration of capacitance manometers and ion gauges against a spinning rotor gauge. Periodically the capacitance manometers are recalibrated in order to maintain the accuracy of the QMS calibrations within the estimated range of ± 10% at $10^{-6}$–$10^{-5}$ Torr.

## IV. LONG- AND SHORT-TERM SENSITIVITY TRENDS

The variations in sensitivity of the two TFTR quadrupole mass spectrometers have been trended since they were placed into service on TFTR. The sensitivity for argon and nitrogen extending over a two-year period for one system and a one-year period for the second system is shown in Figs. 2a and 2b, respectively.

For clarity the sensitivity variations for only two of the calibration gas components ($N_2$, Ar) are shown in Fig. 2. The sensitivity trends of the other four gasses track the plotted trends, and no mass dependence of the sensitivity changes is observed. Initially rhenium filaments were used in the ionizers of the spectrometers. As can be seen from the data in Fig. 2, there were large excursions in the QMS sensitivity during this period of time. Changes in sensitivity of more than 100% have been observed from one calibration to the next. The largest changes occurred after bake-out and glow discharge cleaning sessions, when the QMS was held at relatively high $H_2$ pressures ($> 10^{-5}$ Torr) with large partial pressures ($< 10^{-7}$ Torr) of active gasses such as CO and $H_2O$ for extended periods. Throughout this period the analyzers exhibited both positive and negative changes in sensitivity. We attribute this variability to changes in the ion optics within the QMS

ionizer, caused by excessive evaporation and chemical reactivity of the Re filaments in the high pressure, active gas environment to which the QMS was periodically exposed. We replaced the rhenium filaments with tungsten and obtained a significant improvement in the stability of the QMS: subsequently observed changes in sensitivity were usually less than 10% between calibrations at constant temperature (Fig. 2). The trend of slowly decreasing sensitivity with time (after the filament change to tungsten) is consistent with our previous experience with QMS devices with Cu-Be electron multiplier detectors.[1] Such detectors show an initial large drop (x 0.1) in gain over the first few weeks of operation, but then show only a slowly decreasing gain over many months of subsequent operation in our vacuum environments.

After observing the above behavior with the on-line QMS system, laboratory experiments were performed to examine QMS sensitivity variations in more detail. A Balzers (M/N 511) QMS was installed on a separate turbomolecular-pumped, 304 stainless steel, ultrahigh vacuum system similar to the TFTR configuration, and calibrations were made using the procedure already described. During these experiments, all ionizer voltages were held constant, the emission current was maintained at 1 mA and the secondary electron multiplier (SEM) was kept at 1.4 kV unless otherwise specified.

The long-term trend in sensitivity of this off-line QMS for $H_2$ is shown in Fig. 3, for both SEM and Faraday cup signal outputs. Each data point represents the average of a day-long scan in the pressure range $2 \times 10^{-6}$ to $4 \times 10^{-5}$ Torr. Initially, a rhenium filament was used, which was later replaced by tungsten. As shown in Fig. 3 no significant long-term change (over 65 days) was observed in the sensitivity of this QMS when measurements were made with the Faraday cup, whereas the expected slowly decreasing sensitivity with the SEM output is exhibited. We also used this off-line QMS

to confirm the difference in stability observed on the on-line systems when the filament material was changed from rhenium to tungsten.

With a rhenium filament in the ionizer, large short-term changes in the sensitivities were observed, particularly for $H_2$ for which 50% changes were measured over eight-hour periods (Fig. 4). By replacing the rhenium filament with a tungsten, the sensitivity variations were reduced to $\leq \pm 10\%$ (Fig. 5, open data points). Figure 5 also shows that the pressure limit for constant sensitivity (within ± 10%) is in the mid $10^{-5}$ Torr range for $H_2^+$ signals with ionizer emission currents of 1 mA. The range of constant sensitivity may be extended by decreasing the emission current (Fig. 6). This is a well-known technique[7] for extending the range of linearity in electron-bombardment ion sources that minimizes ion-molecule reactions within the ionization and extraction volumes.

Other factors which are known to affect the measured QMS sensitivity for a particular gas are gas-surface reactions within the ionizer[6] or with the vacuum vessel wall. If the wall is heavily loaded with $H_2$, desorption will occur as shown in Fig. 7 (case 2). However, if the walls are not heavily loaded, the injected $H_2$ may be dissociated by a hot filament (e.g., an ion gauge) and pumped by the walls as shown in case 1. In both cases, $CH_4$ reacted with hot filaments and was pumped by the walls. The result is that the measured sensitivities are in error, since the calibration gas mixture no longer contains equal amounts of each component gas. These effects were minimized during QMS calibration by switching off the ion gauge after measuring the total pressure of the system (Fig. 5).

## V. SUMMARY

The calibration data from two years of operation of the TFTR quadrupole mass spectrometer systems have been trended. These calibrations were done on a weekly basis during tokamak operation. Large sensitivity variations, some greater than 100%, in the initial QMS performance have been observed, particularly after exposure of the QMS systems to high pressures of $H_2$ and bakeout cycles during discharge cleaning operations These large sensitivity variations were attributed to process gas reactions with the rhenium filament in the ionizer affecting the ionizer efficiency. Week-to-week sensitivity variations were reduced to less than 10% at constant temperatures by replacing the rhenium filaments in the QMS ionizers with tungsten. The QMS sensitivities were subsequently observed to decrease slowly with time. These sensitivity changes are attributed to gain changes in the Cu-Be secondary electron multiplier detector.

## ACKNOWLEDGEMENTS

The authors wish to thank J. Dong, T. Provost, M. McGowan, and D. Doersam for their technical support. This work was supported by US DOE contract No. DE-AC02-76-CH03073.

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## FIGURE CAPTIONS

Fig. 1 Schematic drawing of one of the two quadrupole mass spectrometer (QMS) systems used for residual gas analysis on the Tokamak Fusion Test Reactor (TFTR). (IV-interface valve, HVV-high vacuum valve, OV-orifice valve, GIV-gas injection valve, LV-leak valve, IG-ion gauge, CM-capacitance manometer, TMP-turbomolecular pump, MP-mechanical pump).

Fig. 2a,b Quadrupole mass spectrometer sensitivities for $N_2$ and Ar for each of the TFTR QMS systems. At points A, B the system was baked at 150°C and at point C the rhenium filaments were replaced with tungsten.

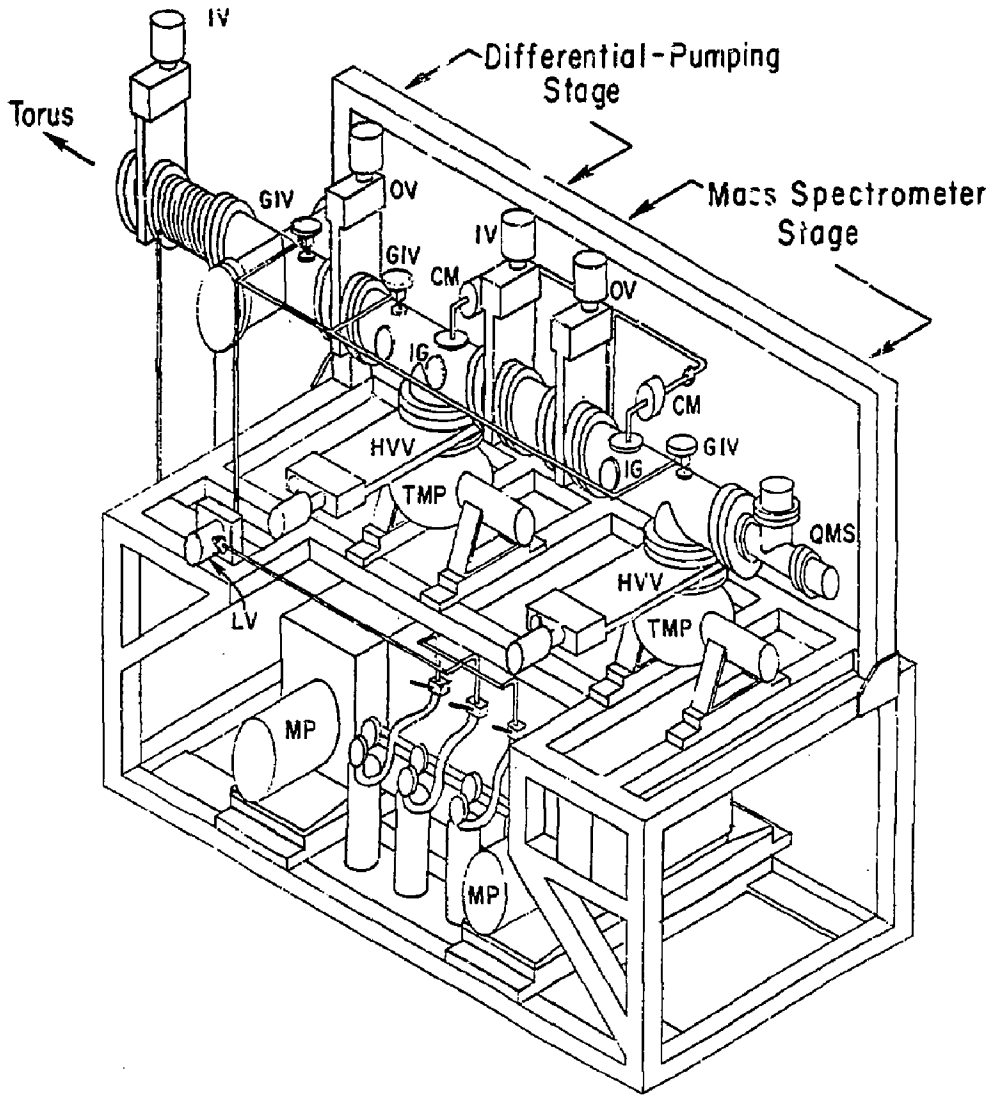
Fig. 3 Comparison of QMS sensitivity trends for Faraday cup and secondary electron multiplier (SEM) output signals.

Fig. 4 Short-term variation of sensitivity for $H_2$ for the QMS with rhenium filaments in the ionizer.

Fig. 5 Comparison of QMS calibration stability for rhenium vs. tungsten filaments in the ionizer. The interfering effects of a nearby ion gauge on the measured QMS sensitivity are also shown.

Fig. 6 Pressure dependence of the QMS sensitivity for $H_2$ as a function of the electron emission current in the ionizer.

Fig. 7 Observed changes in the partial pressures of $H_2$ and $CH_4$ from an




TFTR Residual Gas Analyzer

Fig. 1

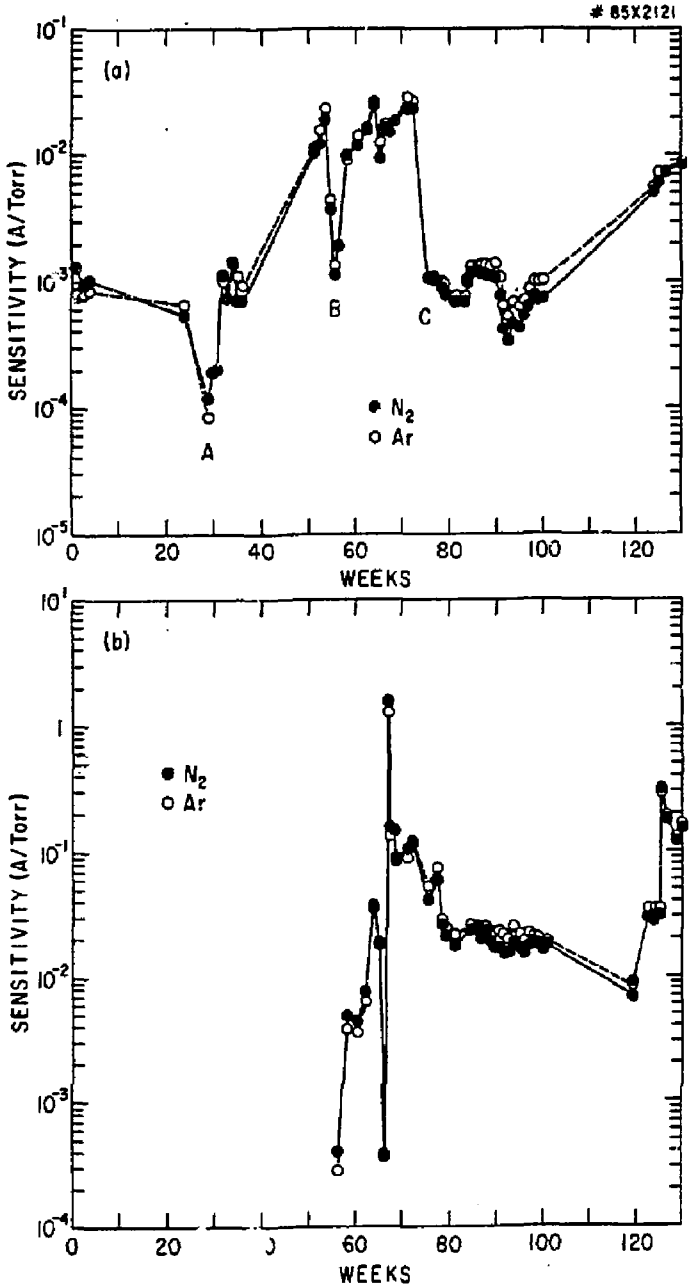
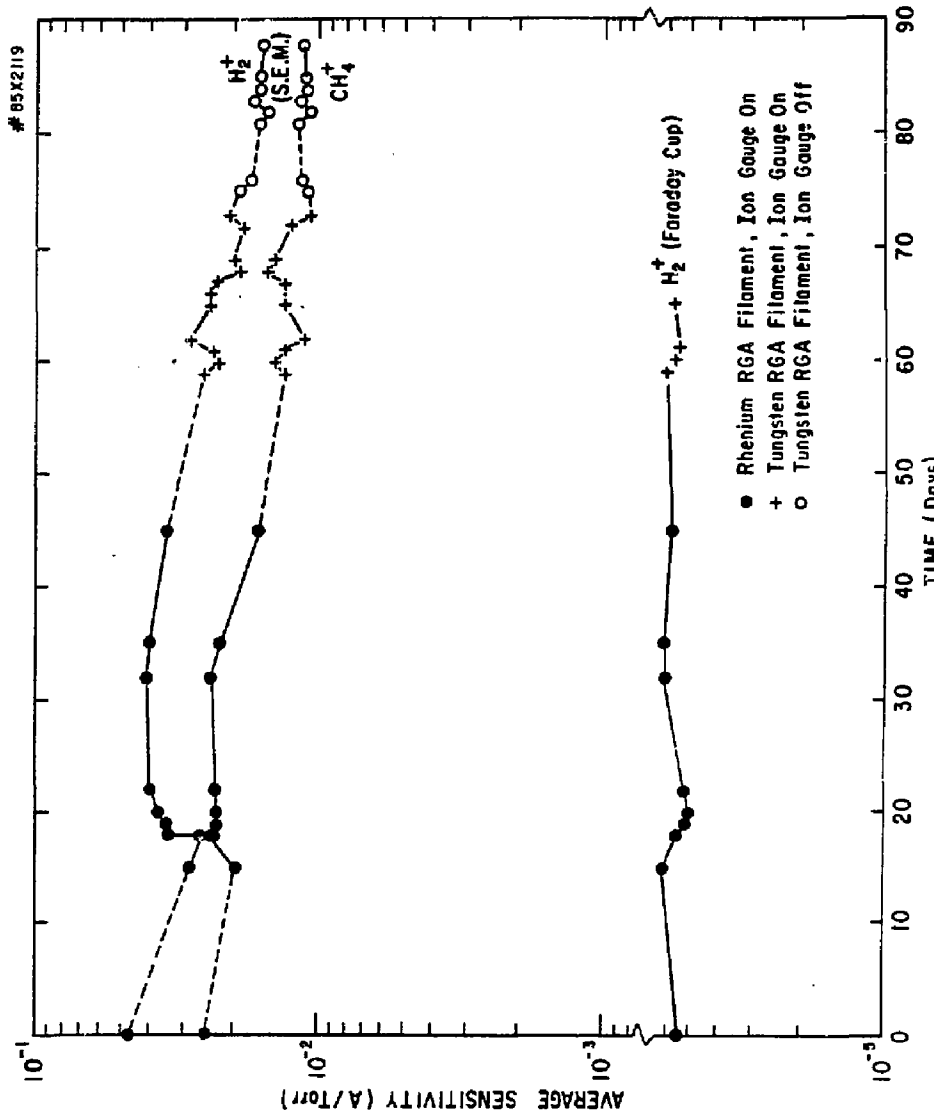

#85X2121
(a)
SENSITIVITY (A/Torr)
WEEKS
A
B
C
N2
Ar
(b)
SENSITIVITY (A/Torr)
WEEKS
N2
Ar

Fig. 2a, b


#85X2119
AVERAGE SENSITIVITY (A/Torr)
TIME (Days)
$H_2^+$ (S.E.M.)
$CH_4^+$
$H_2^+$ (Faraday Cup)
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+ Tungsten RGA Filament, Ion Gauge On
o Tungsten RGA Filament, Ion Gauge Off

Fig. 3

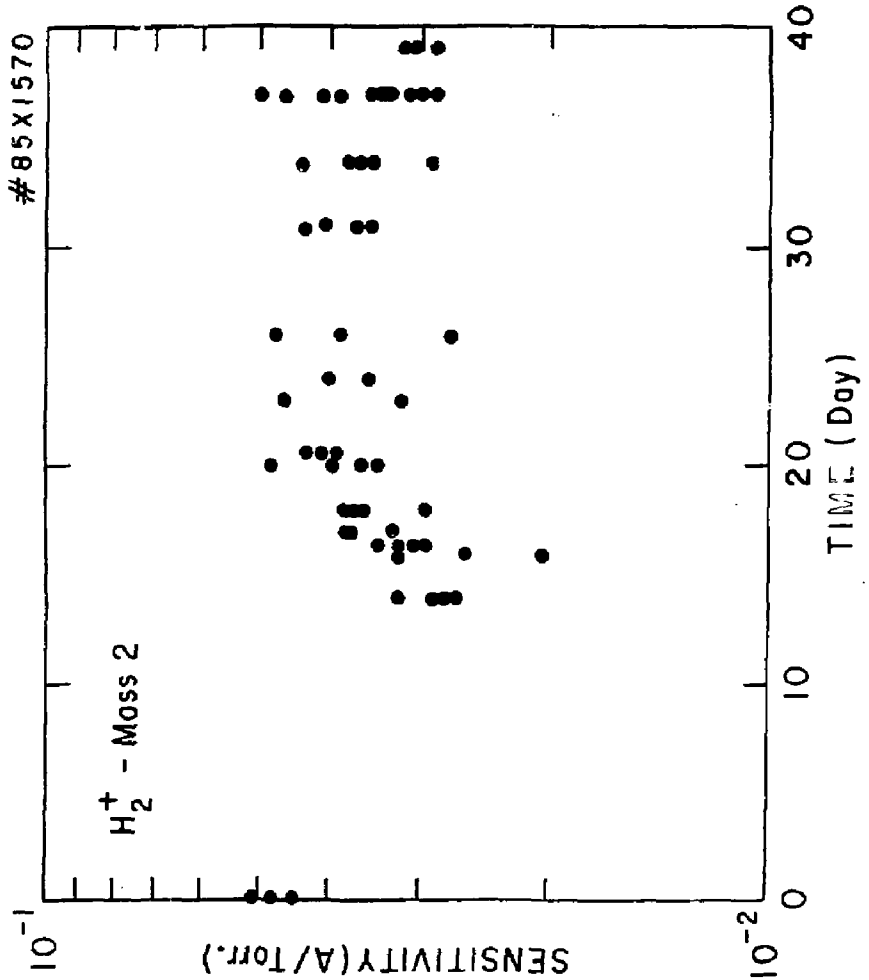
#85X1570
$H_2^+$ - Moss 2
SENSITIVITY (A/Torr.)
$10^{-1}$
$10^{-2}$
0
10
20
30
40
TIME (Day)

Fig. 4

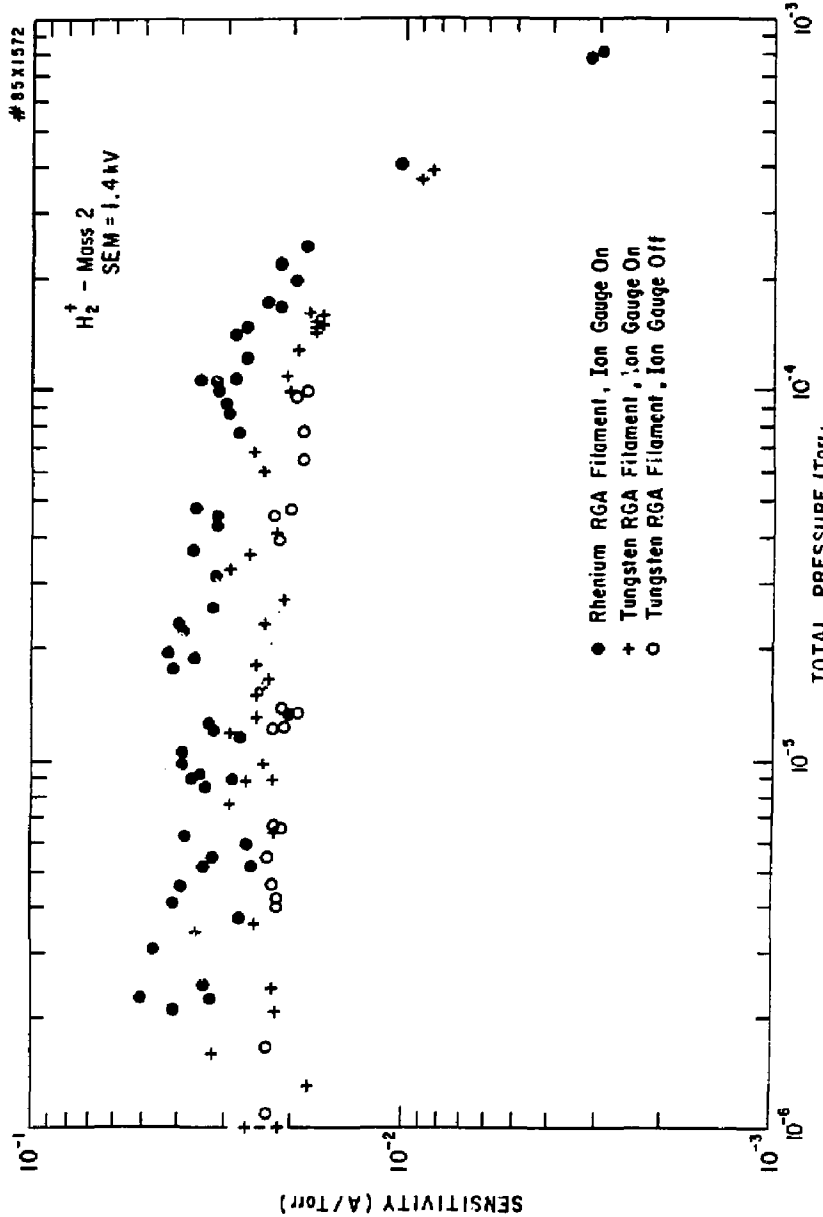
#85X1572
$H_2^+$ - Mass 2
SEM = 1.4 kV
Rhenium RGA Filament, Ion Gauge On
Tungsten RGA Filament, Ion Gauge On
Tungsten RGA Filament, Ion Gauge Off
SENSITIVITY (A/Torr)
TOTAL PRESSURE (Torr)
$10^{-1}$
$10^{-2}$
$10^{-3}$
$10^{-3}$
$10^{-4}$
$10^{-5}$
$10^{-6}$

Fig. 5

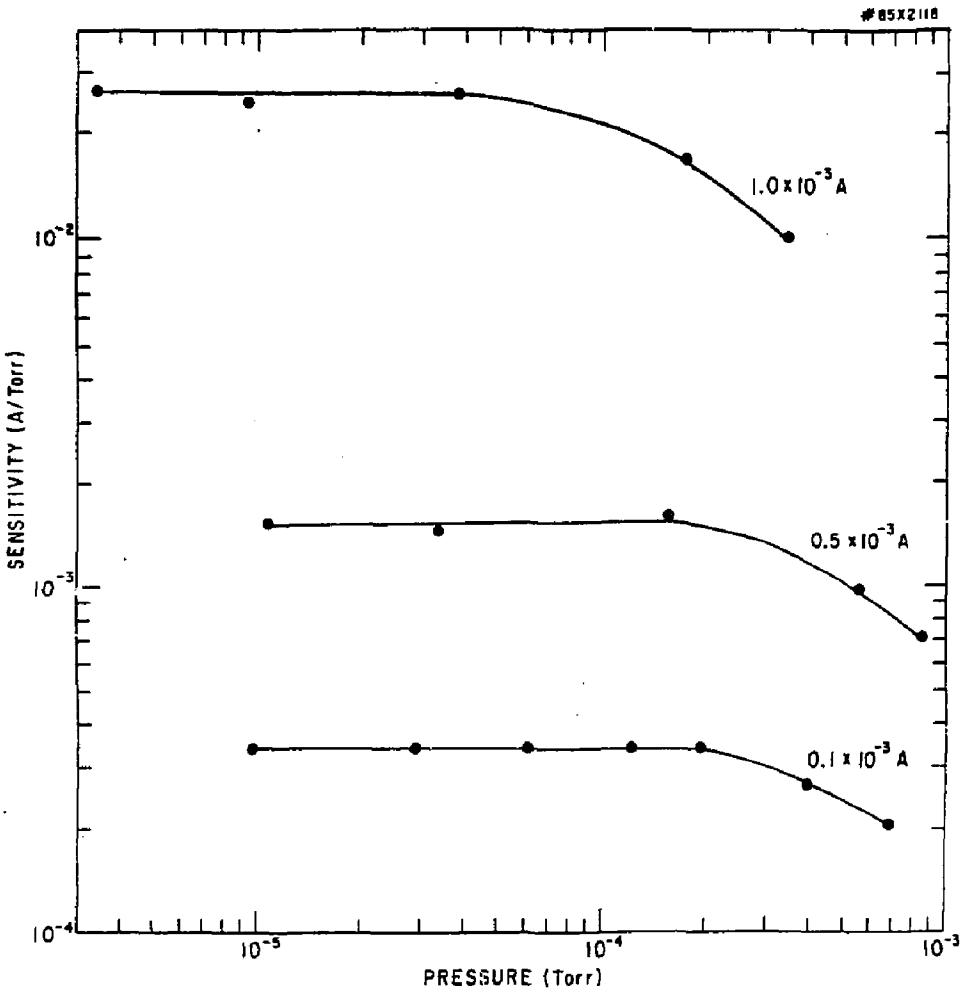
#85X2118
1.0 x 10⁻³ A
0.5 x 10⁻³ A
0.1 x 10⁻³ A
SENSITIVITY (A/Torr)
PRESSURE (Torr)
10⁻²
10⁻³
10⁻⁴
10⁻⁵
10⁻⁴
10⁻³

Fig. 6

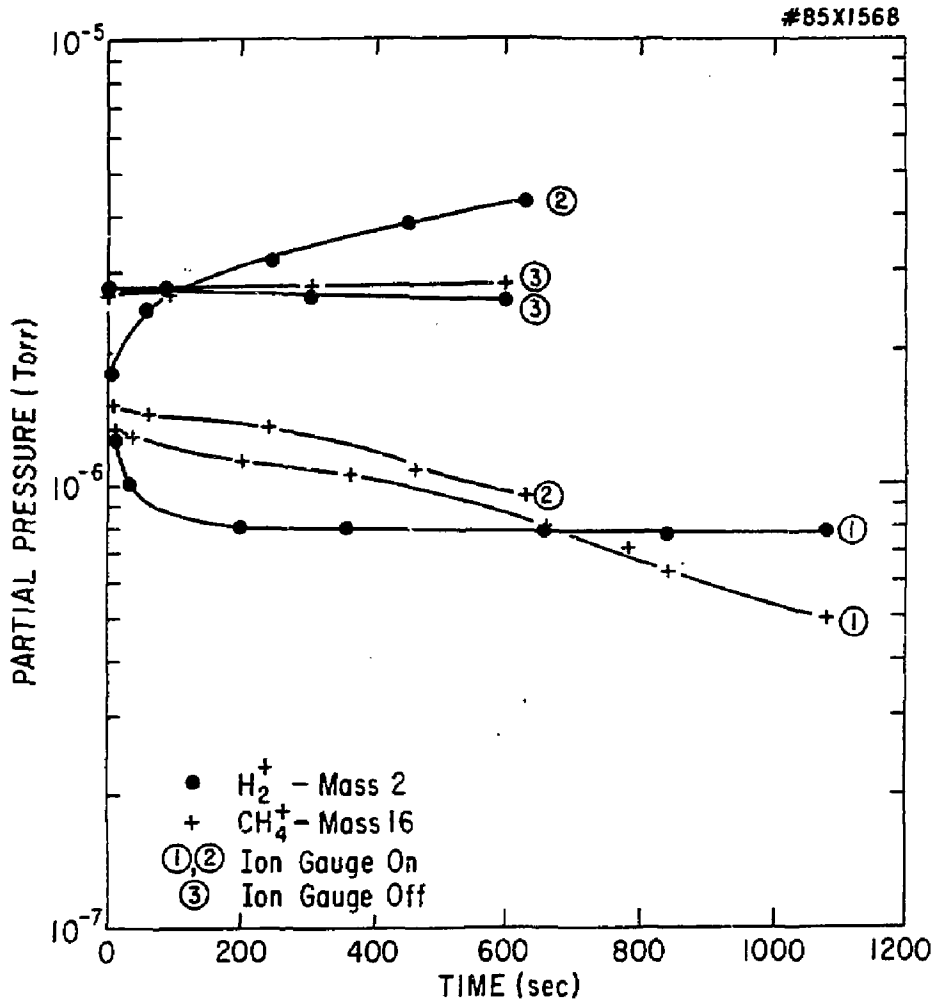
#85X1568
PARTIAL PRESSURE (Torr)
$10^{-5}$
$10^{-6}$
$10^{-7}$
0
200
400
600
800
1000
1200
TIME (sec)
$H_2^+$ – Mass 2
$CH_4^+$ – Mass 16
①,② Ion Gauge On
③ Ion Gauge Off

Fig. 7

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