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SPUTTERING OF CONDENSED GASES BY PROTON BOMBARDMENT

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SPUTTERING OF CONDENSED GASES BY PROTON BOMBARDMENT

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ABSTRACT

Experiments have been carried out on the sputtering of helium argon nitrogen and carbon monoxide condensed layers by 5 keV protons. In the case of helium, yields up to 10^4 atoms/ion were observed but the range of layer thickness which could be investigated was limited, as the minimum target temperature was 2.2° K. The yields of argon nitrogen and carbon dioxide were very much lower as is to be expected on account of their much larger sublimation energy. The yield increased with coverage to quite a sharp maximum of 50-100 atoms/ion at a coverage of about 3×10^{15} molecules/cm^2 and subsequently decreased with increasing coverage to about 5 atoms/ion at 2×10^{16} molecules/cm^2. On the basis of the thermal spike model the maximum substrate temperature is too low for evaporation of the heavier gases and a more direct mechanism of energy transport must be considered.

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1. INTRODUCTION

The desorption of gases from cryogenic surfaces by ion bombardment is of interest in certain types of plasma physics experiment, using either cryopumps or superconducting magnets, and in superconducting accelerators. In previous papers the results of thermal desorption[1] and of ion and electron bombardment of solid hydrogen and deuterium[2] were described. Yields in the range 10^4 - 10^5 atoms/ion were obtained for layers of condensed gas less than 1000 Å thick under proton bombardment and these values were explained in terms of a thermal spike in the copper substrate below the condensed gas layer. It was considered of interest to see how the yields of other gases compared with hydrogen particularly gases with higher heats of sublimation. In this paper we present results of the desorption of heavier gases He, N_2, CO and Ar by 5 keV protons and also the effect on gas yields of making "sandwiches" of condensed gas consisting of H_2, N_2 and Ar in various thicknesses.

2. EXPERIMENT

The details of the experimental technique have been described previously[2]. The gas is condensed on a liquid helium cooled surface whose temperature is measured with a germanium resistance thermometer, fig 1. The target chamber is pumped continuously and the gas desorption rate is measured dynamically with a quadrupole mass spectrometer. The vacuum time constant is approximately 0.05 secs. Gas is introduced either through a palladium-silver leak (for hydrogen isotopes) or through a sintered Si C leak of low conductance. Gas is condensed on the surface for a known time at ambient pressure, from which the thickness of the layer can be deduced assuming a sticking coefficient. The ion beam is then turned on and the yield of gas from the target is measured as a function of time. The experiment is normally started by cleaning the surface with a high current density ion beam ($\sim$ 200 μA cm^{-2}).

3. RESULTS AND DISCUSSION

(a) Argon, nitrogen and carbon monoxide.

A variety of gases were studied in order to compare them with the previous results for hydrogen and deuterium. Measurements of the yield as a function of time for a number of initial layer thicknesses are shown in fig 2. Both nitrogen and argon show similar behaviour to that found previously for hydrogen and deuterium with the difference that the yields are about a factor of 10^3 lower. The layer thickness is quoted in the directly measured units of torr-sec exposure. The sticking coefficient of argon has been measured experimentally and found to be greater than 0.95[3] at 4.2 K over the range of coverages of interest. The layer thickness in molecules cm^{-2} can thus be readily calculated. It has been assumed that N_2 and CO will have similarly high sticking coefficients. For an initial layer thickness greater than 10^{-5} torr sec it is seen that the yield increases with time as the gas layer is eroded. At a certain thickness there is a maximum yield and thereafter the yield decreases as the layer is eroded further. This is illustrated more clearly in fig 3 where the initial yield is plotted as a function of the thickness of the layer from a series of runs as shown in fig 2. The yield increases approximately linearly with layer thickness reaching quite a sharp maximum and then decreasing. The maximum yield is 100 $\pm$ 20% atoms/ion for nitrogen and argon and 50 $\pm$ 20% atoms/ion for carbon monoxide. The absolute yields are measured relative to the hydrogen partial pressure rise when the incident ion beam is allowed to impinge on a saturated palladium target which under suitable conditions has been found to have a yield of 1 atom/ion[4]. A correction is applied for the variation in sensitivity of the mass spectrometer to the various gases.

It is clear that a sharp maximum yield occurs at a coverage of a few monolayers. This is quite different from the case of solid hydrogen. The peak yield is thus a thin film phenomenon, and at greater thickness the yield rapidly approaches the yield of a bulk material. The range of 5 keV protons in N_2 or CO is 1050 Å as calculated from the data of Schiøtt[5]. This range corresponds to $\sim$ 4 x 10^{17} molecules cm^{-2} or a gas exposure of 8 x 10^{-4} torr - ℓ. Judging from the nitrogen case, fig 3b, which is the only one with sufficiently thick layers, there is no marked change in yield when the range of the incident ions is less than the thickness of the condensed gas layer. This again is in contrast to the results for hydrogen.

There are thus two main results to explain - the reason for the maximum yield of $\sim$ 100 atoms/ion and the yield from the thick layers $\sim$ 5 atoms/ion. The first obvious check is to see whether the thermal spike mechanism, which was successful in explaining the high hydrogen yields, would apply in this case. However it is soon clear that there is no possibility of this mechanism explaining the yields for the heavier gases. For thin condensed gas layers the incident protons will slow down and give up their energy in the copper substrate, thus the calculation is identical to that described previously for hydrogen[2]. The spike radius is $\simeq$ 250 Å, the temperature 30 K and the lifetime $\sim 10^{-10}$ sec. From vapour pressure data this leads to yields of $\sim 10^{-4}$ atoms/ion for nitrogen and less for Ar and CO. The yield from a conventional sputtering mechanism assuming a thick layer of condensed gas has also been calculated. Estimates from both the theory of Sigmund[6] (which is recognized not to apply too well for light ions and tends to give an over estimate) and also from the simpler theory of Pease[7] leads to yields from nitrogen in the range 0.1 to 1.0 atoms/ion. It is thus difficult to explain the observed yields by this process.

A further possibility is release of gas by dissociation or ionization in a way similar to the process of electron desorption of adsorbed gas[8]. There is then the possibility of cluster formation around ions as has been observed in the electron bombardment of solid hydrogen[9] and in the ion bombardment of water[10]. However there is little information available to confirm whether such a mecha: sm can explain the observed yield. It is rather unlikely that such a mechanism can explain the sharp maximum in the yield curve. As discussed earlier this appears to be a thin film phenomenon and one possibility is that phonons produced in the copper by the incident ion cannot propagate across the boundary from the copper to the condensed gas when the gas layer is thick, because the phonon spectra in the two materials are radically different. However when the condensed gas film is thinner than the phonon mean free path then the condensed gas layer will not have a well defined phonon spectrum and thus energy transfer across the boundary may be allowed.

(b) Helium

Because of the relatively high temperature of the substrate and hence the high vapour pressure of helium the thickness of the helium with increasing exposure will probably increase by physisorption only to a few monolayers before the sticking coefficient will become effectively zero. The surface concentration will then be constant with further increase in time at constant pressure, the rate of arrival of atoms at the surface being in equilibrium with the rate of desorption. The results with 10^{-8} torr helium present are shown in fig 4a. It is apparent that this curve is much closer to the behaviour of the hydrogen results described earlier[2] than to those of the heavier gases. The yield increases linearly to a maximum which is of the same order of magnitude as that for hydrogen. Since the substrate temperature must again be $\sim$ 3OK vapour pressure of the helium liquid will be high (since it is atmospheric pressure at 4.2 K), and so the yield can be readily explained by the thermal spike mechanism. The fact that the helium yield is lower than hydrogen (which has a heat of sublimation about 10 times greater) may be due to the lower equilibrium coverage, or to the physisorption of He on copper at these coverages having a higher heat of adsorption than the heat of evaporation of bulk hydrogen.

A further experiment with helium is shown in fig 4b where the yield is measured as a function of substrate temperature for a constant exposure of 10^{-6} torr secs. No quantitative measure is available of the layer thickness but the results illustrate the decrease in yield as the surface coverage decreases with increasing temperature.

(c) Gas mixtures

Finally some measurements were made of the yield of one gas as the thickness of another gas on top or underneath it was varied. In fig 5 the results obtained for the yield from a fixed thickness of argon is shown as the thickness of a hydrogen layer on top of the argon is increased. The argon yield remained constant until the hydrogen layer on top of it increased to the point where the hydrogen layer thickness exceeded the range of the incident proton ($\sim 2 \times 10^{-4}$ t-s). It is interesting to note that the yield did not decrease significantly until this point, indicating that the argon desorption could take place through the hydrogen layer. The corresponding yield of hydrogen in a similar sandwich is shown in fig 6. In this case in successive runs a constant thickness of hydrogen is condensed on top of various thicknesses of argon. It is found that the hydrogen yield is initially at the same level as obtained when it is condensed on a simple copper substrate. As the argon thickness is increased above 10^{16} molecules/cm^2 the hydrogen yield decreases. At this layer thickness some of the incident proton energy is being lost in the argon. However no calculations have yet been made of thermal spikes in argon, so it is not yet clear whether the temperature spike is simply lower in argon than copper or whether the argon layer is acting as a thermal barrier to the energy which is deposited in the copper substrate. A further possible explanation is that the hydrogen actually diffuses into the argon rather than staying on top after condensation. Some evidence for this has been obtained when the condensed gas sandwich is produced with nitrogen on top and hydrogen underneath. It is found that hydrogen will diffuse through the nitrogen layer at a detectable level in our apparatus at 2.8 K. The rate of diffusion increases with temperature and a plot of the flow rate vs 1/T (for constant nitrogen thickness of 10^{17} molecules/cm^2) indicated an activation energy for diffusion of 170 cals/mole.

CONCLUSIONS

The desorption of heavy gases such as Ar, N_2 and CO by 5 keV protons is a much less probable process than the desorption of the light gases such as hydrogen and helium. This is in general agreement with the very large difference in their heats of sublimation. From the present results helium appears to behave in a similar way to hydrogen with yields of $\sim 10^4$ atoms/ion and this can be explained qualitatively on the basis of the thermal spike mechanism previously developed for hydrogen. However a detailed explanation is not yet possible because in the present experiments the helium is only adsorbed rather than condensed, and its thickness has not been well defined.

It appears that the yield of the heavier gases cannot be explained either by the thermal spike mechanism or by a conventional momentum transfer sputtering theory. It has been put forward as a hypothesis that an ionization or atomic excitation mechanism is the explanation, but further experimental work, particularly on the mass distribution of the desorbed species, would be necessary to establish this hypothesis.

Finally the measurements on gas mixtures have produced some interesting results which may be useful in assessing practical applications of cryosurfaces in the presence of ions. Although no clear explanation has yet been found for these results it is possible that this approach could be a useful method of further investigation of mechanisms since it is a simple way of looking at a variety of substrates.

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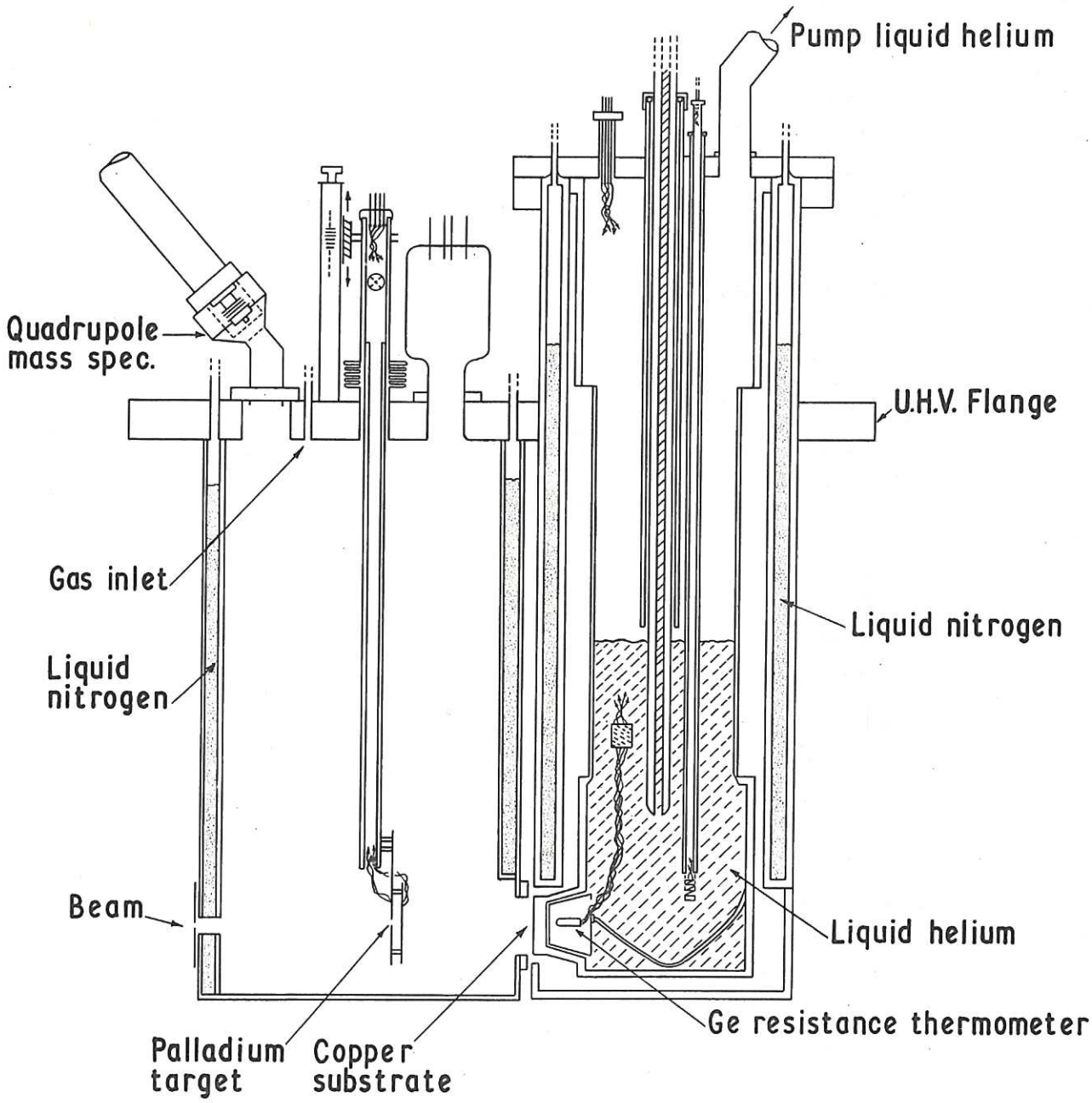


Figure 1. Schematic diagram of target chamber showing He cryostat.

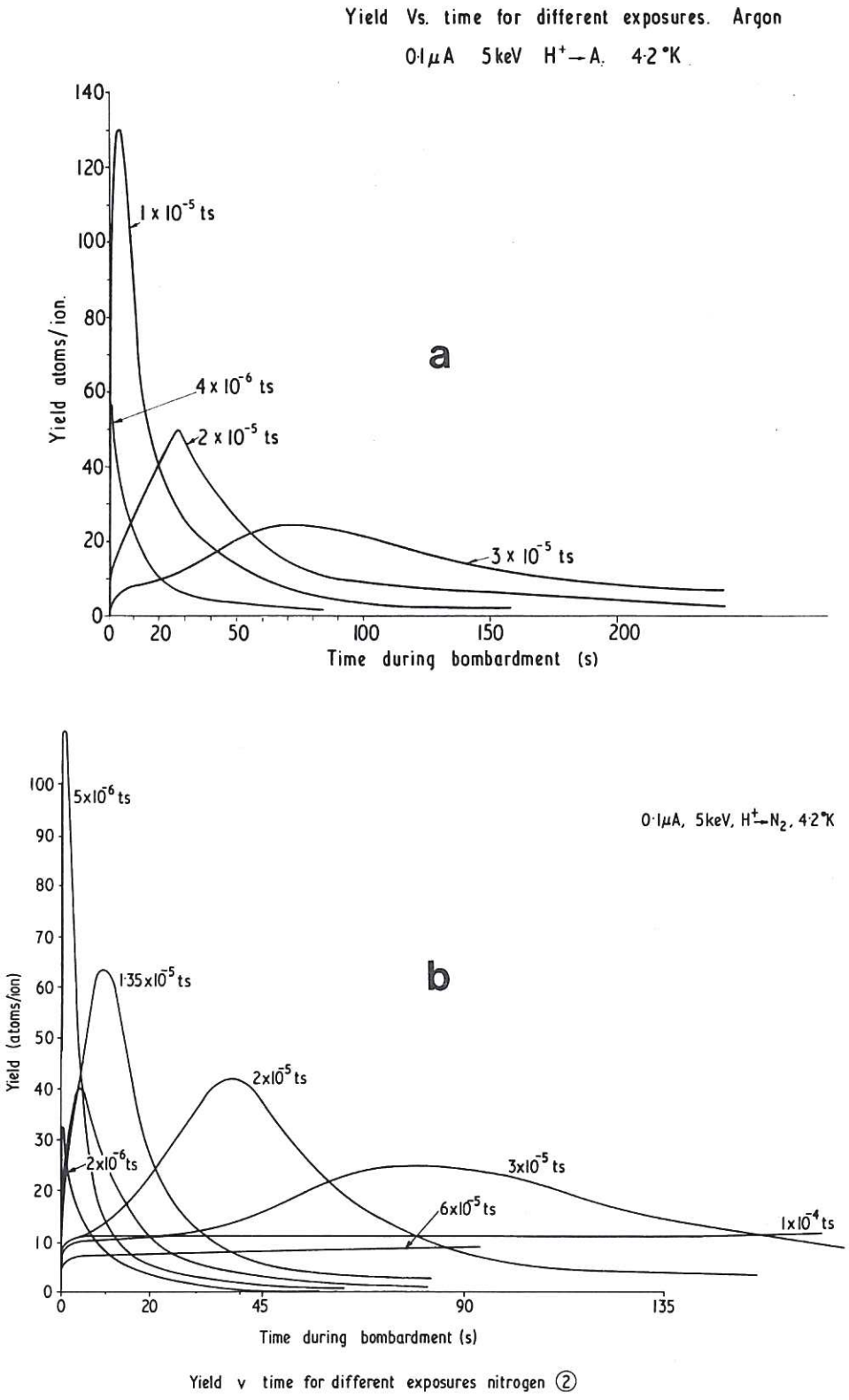


Figure 2. Yield of gas desorbed from the target by 5 keV protons as a function of time for different initial thickness of condensed gas layer at 4.2 K

(a) Argon (b) nitrogen

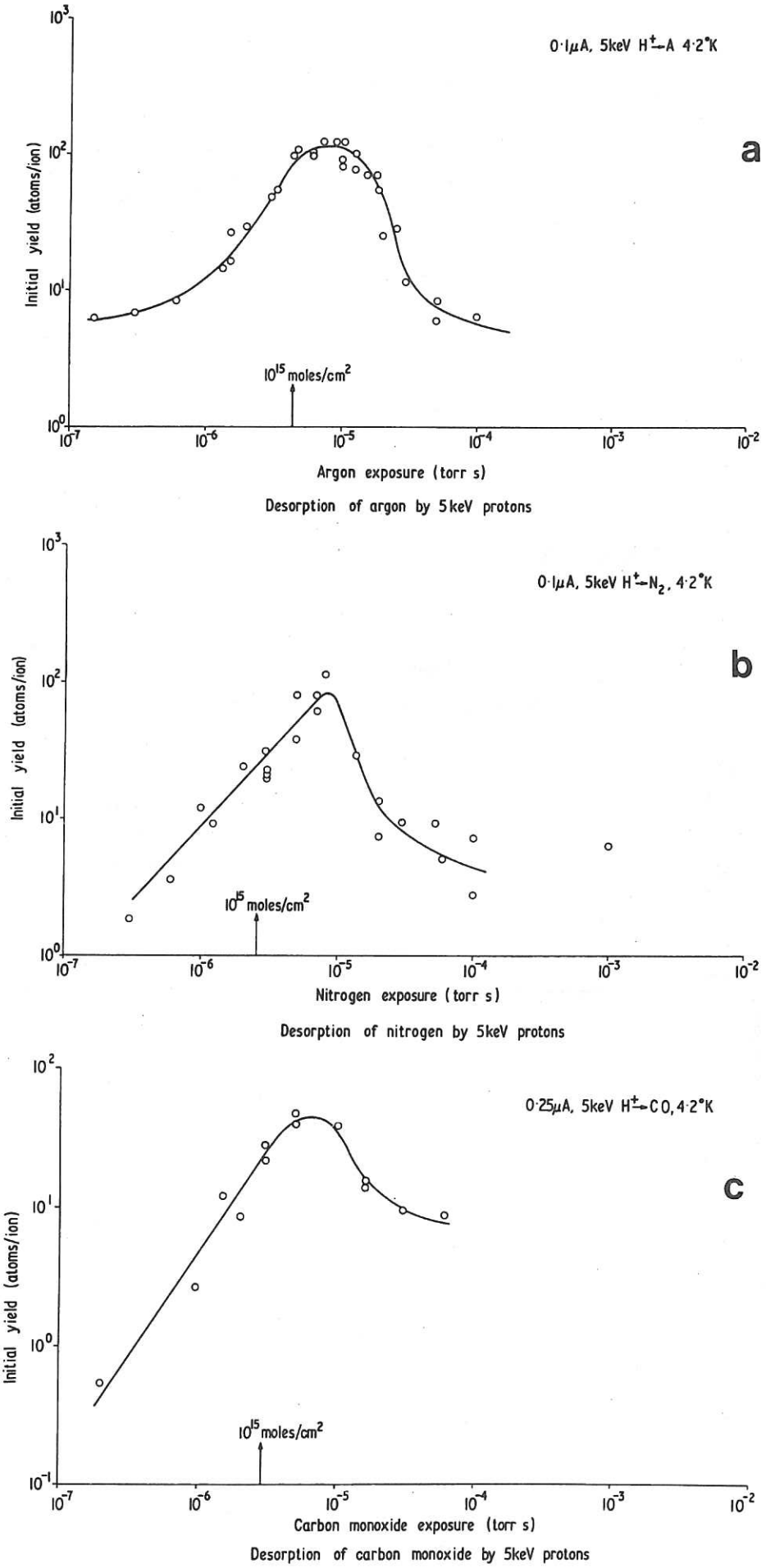
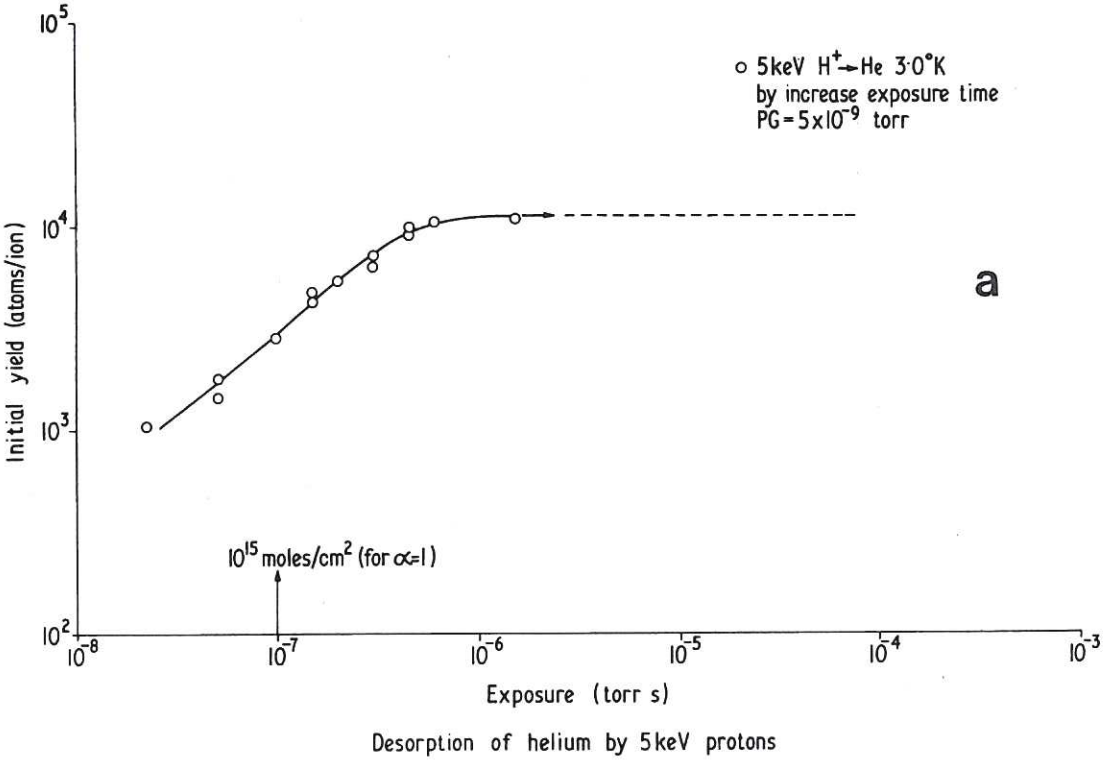
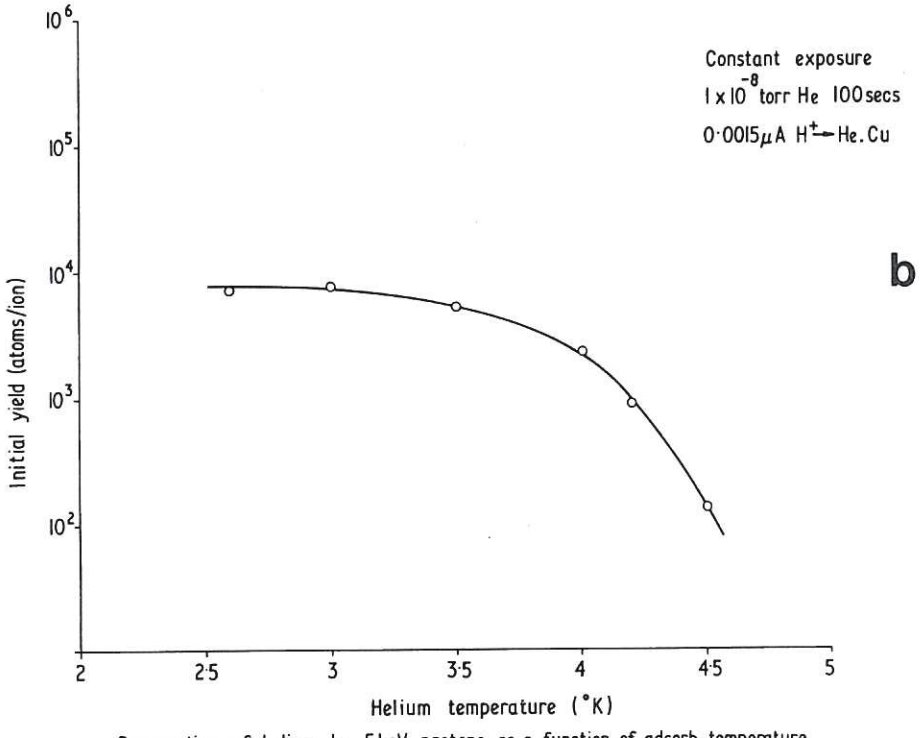


Figure 3. Initial yield of gas as a function of condensed layer thickness. 5 keV protons on condensed gas at 4.2 K

(a) Argon (b) Nitrogen (c) Carbon monoxide



Desorption of helium by 5keV protons



Desorption of helium by 5keV protons, as a function of adsorb temperature

Figure 4. Desorption of adsorbed helium by 5 keV protons.

(a) Yield vs exposure Temperature 3.0 K. Pressure during adsorption 5 x 10^{-9} torr.

(b) Yield vs temperature. Exposure 10^{-6} torr sec. Pressure during adsorption 10^{-8} torr.

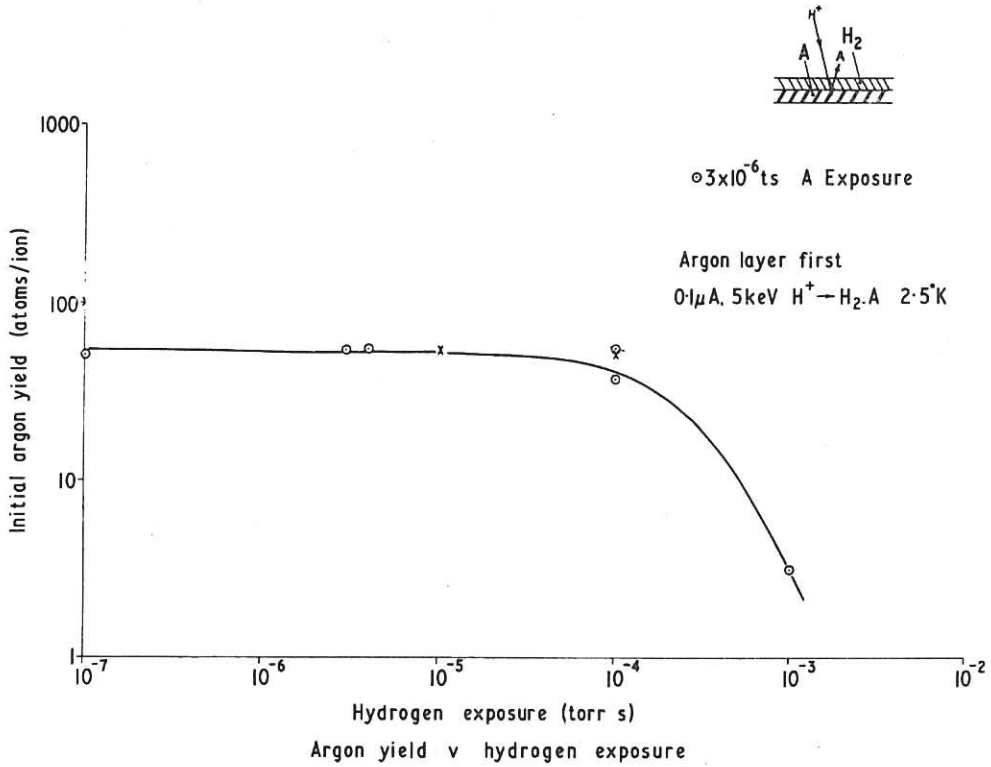


Figure 5. Desorption of argon from a layer of condensed argon covered by a layer of hydrogen of varying thickness by 5 keV protons. Temperature 2.5 K.

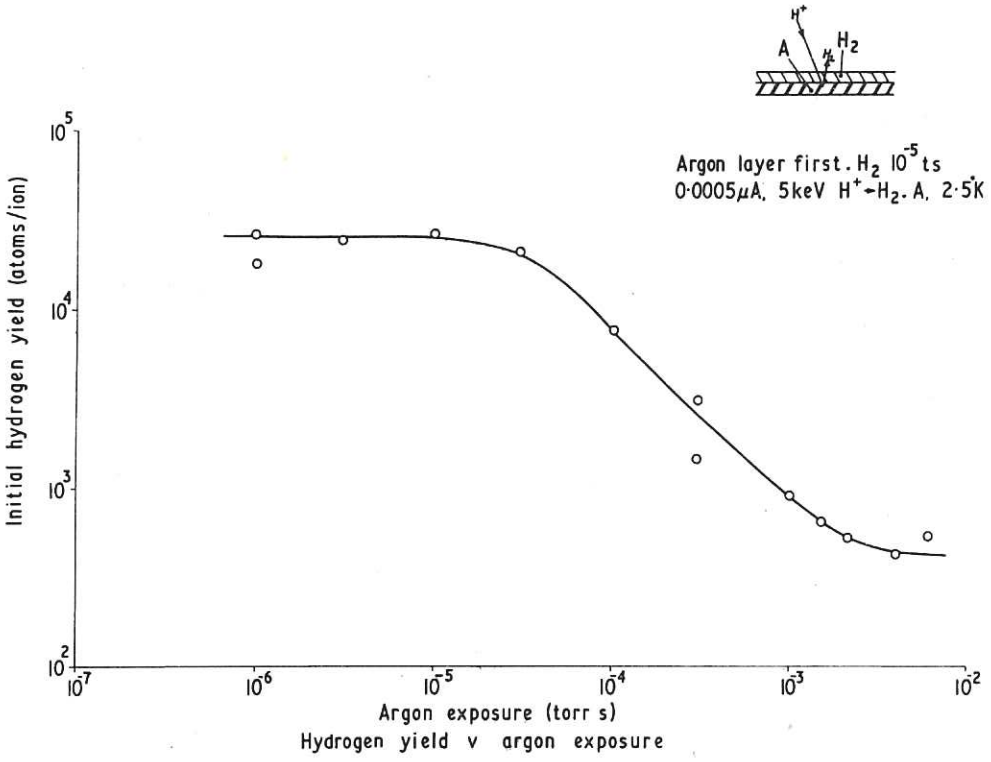


Figure 6. Desorption of hydrogen from a layer of hydrogen condensed on top of a layer of argon of varying thickness by 5 keV protons. Temperature 2.5 K. Hydrogen layer thickness $\sim 2 \times 10^{16}$ molecules/cm^2.