A Long-Lived Reaction Product from Helium Discharges[1]

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Fowler and Jones (1963) have described a method by which a simple ternary mixture of electrons, virgin molecules, and ions from those molecules can be obtained in glow discharges. It is expected that the method will provide truly quantitative data on the glow discharge, especially with reference to such unstable effects as the constriction under simple density increase. Briefly the method consists in operating a pulsed discharge for an interval long enough to allow primary ionization and diffusion to stabilize, but short enough to eliminate chemical reactions between the products of the discharge.

The problem under study in this paper was the determination of the time required for appearance of the first contaminating species, and the visibility of this species. It had previously been observed (Fowler and Jones, 1963) that the effect of contaminants was to alter the state of constriction of partially constricted columns. Therefore the partially constricted column was chosen as a test of the appearance of contaminant species.

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The experimental apparatus consisted primarily of a 9 μf capacitor bank, a current limiting resistor, and a discharge tube. The tube was constructed of 5 cm.-diameter Pyrex tubing with a central section of 5 cm. x 5 cm. square tubing. The diameter of the plane, circular, nickel electrodes was 5 cm. Their separation was 100 cm. Associated circuitry included the capacitor charging circuit, a hydrogen thyratron (5C22) to switch the capacitor bank into the discharge tube, an ignitron (GL-7171) to crowbar the capacitor bank and stop the discharge, and the necessary trigger timing circuits to coordinate these elements. Current and voltage waveforms were monitored on an oscilloscope. A cross section of the positive column was photographed through the side on 35 mm Tri-X film and scanned with a Leeds and Northrup recording densitometer (Fowler and Jones, 1963).

The capacitor bank was first charged to the desired voltage. A triggering circuit initiated the discharge by firing the thyratron. It also activated a timing circuit which, a short time (1 millisecond) later, triggered the ignitron which cut off the discharge sharply. The camera shutter was opened before initial triggering and closed after the discharge was over. The film was calibrated with a Jarrell-Ash, 7-step filter and the normalized density plotted as a function of tube diameter. In this study, the tube was fired every 30 seconds and every twentieth discharge was photographed. After 200 discharges, the repeated discharges were stopped. Fifteen minutes later the tube was "inspected" by discharging it once. The quiescent time was doubled and again the tube was discharged for inspection purposes. This continued for about 32 hours, with the quiescent time being doubled after each "inspection" discharge.

The results are shown in Figs. 1 and 2. Fig. 1 shows the constriction of the positive column as a function of the number of discharges. There is a marked increase in the constriction of the positive column as the total discharge time (number of discharges times the duration of each) increases. The increase in constriction ceases when the repeated discharges cease. Fig. 2 shows that the constricted column returns very slowly to normal, indicating a long life for the contaminating species. Two days were required for 65% recovery.

There are several possible explanations for this effect. First, the discharge might have become contaminated by impurities from the glass and electrodes of the discharge tube. This is very unlikely, for previously the electrodes were thoroughly heated in a vacuum to dull red heat by an induction heater, and the glass section was degassed simultaneously with gas burners and a heat gun, until the pressure of the gases being driven out had decreased while the system was hot to about 10^{-4} Torr as measured by a McLeod gauge. After this, the system was pumped to 10^{-6} Torr before the helium was admitted to 2.9 Torr through a liquid nitrogen cold-trap which prevented any stopcock or mercury vapors from entering the discharge tube. Furthermore, careful spectrographic analysis of discharges from similar tubes showed no trace of such impurities.

Secondly, the wall of the tube might have become electrically charged and thus caused an increase in the constriction. This possibility was disposed of by doing the same sequence of 200 discharges, pumping the tube to 10^{-6} Torr and admitting clean gas within about 4 minutes. The next discharge was identical to the initial one of the first series.

Finally, the increase in constriction might be due to the presence of a helium molecular-complex. This could be a helium-nickel compound from the electrodes, or some compound with the constituents of Pyrex. A more plausible suggestion is that it is a helium molecule produced by three-

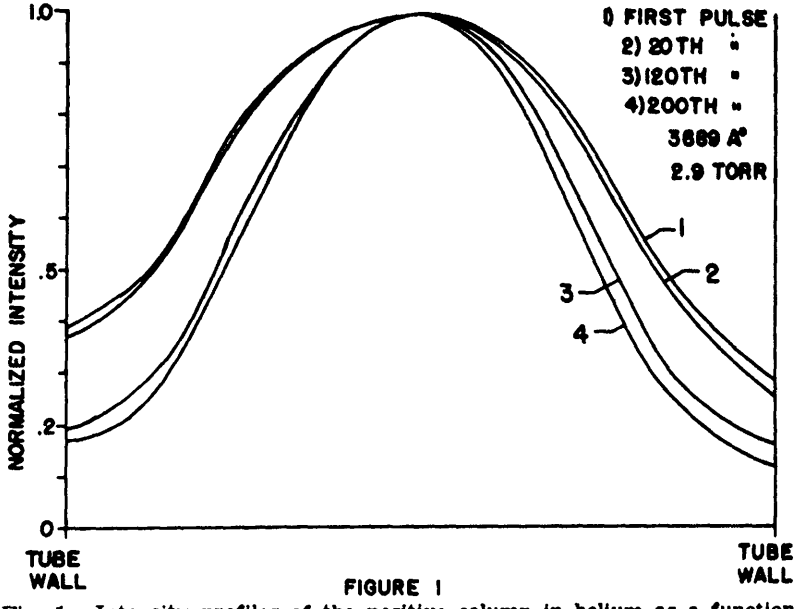

FIGURE 1

Fig. 1. Intensity profiles of the positive column in helium as a function of number of successive discharges.

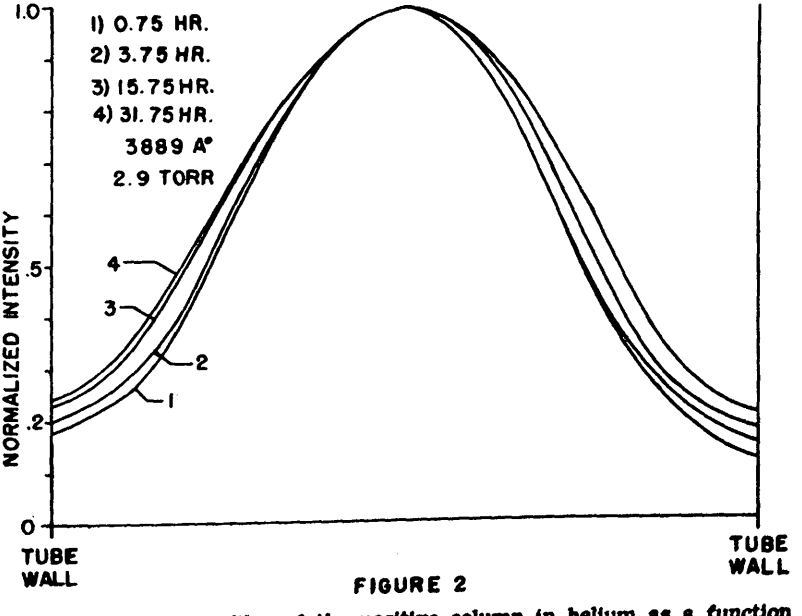

FIGURE 2

Fig 2. Intensity profiles of the positive column in helium as a function of decay interval.

body collisions between two helium atoms and a helium metastable atom:

$$He + He + He^* \rightarrow He_2^* + He.$$

This reaction continues after the current is cut off until the metastable atoms have diffused to the wall. Knowing the distribution of He* at cutoff and assuming zero population of He* at the walls, one may solve the diffusion equation in cylindrical symmetry (as an approximation to the experimental tube which used a combination of cylindrical and square tubing) to get the distribution as a function of time after the current cutoff. The time constant of the exponential decay thus found was about 4 milliseconds, making a total reaction time of about 5 milliseconds per pulse.

We may calculate the production rate of He_2^* in the following manner.

$$\left(\frac{dn}{dt}\right)_{He_2^*} = N^2 N^* \sigma \bar{c} \Delta V$$

WHERE:

N = NUMBER DENSITY OF NEUTRAL HELIUM.

N^* = NUMBER DENSITY OF He^*.

σ = MOLECULAR CROSS SECTION OF NEUTRAL HELIUM.

$\bar{c}$ = MEAN THERMAL VELOCITY OF He ATOMS AT 20° C.

$\Delta V = 4/3\pi (3\sqrt{\sigma/\pi})^3$, THE "ACTION VOLUME."

All of these are well known, except N*, which may be calculated from the density of electrons and knowledge of the production rate of He* by electrons. The electron density may be found from the current through the tube (0.3 ampere). He* is produced by downward transitions from higher energy levels. The production coefficient by downward transition after electron excitation is approximately 0.1 metastable atom per electron per cm. drift in the field (St. John, 1963). If we assume that one He_2^* is produced per collision we get 4.6×10^{13} molecules per cubic centimeter per pulse. After 200 pulses of current, the concentration can rise to about 0.006 of the total gas concentration. This would be sufficient to cause additional constriction in the discharge.

The decay time of these molecules is consistent with their annihilating each other in pairs with the formation of an ion, with a cross-section 10^{-4} times as large as the elastic cross-section. The decay time is too long for destruction to occur by wall collisions.

Literature Cited

Fowler, R. G., and L. W. Jones. 1963. Investigations on constriction of the positive column using glow discharges of limited duration. J. Appl. Phys. (in press).

St. John, R. M. (University of Oklahoma). 1963. Private communication.