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ELECTRICAL BREAKDOWN AND OPTICAL EMISSION PROPERTIES OF HIGH PRESSURE PULSED RF GAS DISCHARGES

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I. INTRODUCTION

A fundamentally new technique for the measurement of the number of subexcitation electrons produced by the passage of ionizing radiation (electromagnetic and high energy charged particles) through a gas has been developed by us during the past few years [1-5]. The new detector is based on the digital characterization of the electrons in the particle track produced by the ionizing radiation where the charged particle track is registered by measuring the number of electrons found in given subvolumes of the gas in the ionization chamber [6-10]. The track is thus characterized by a set of integers in each volume element, from which parameters such as the track length and energy can be measured. We have specifically developed this technique for use in the field of microdosimetry, where the study of the fluctuations in the energy deposition in the charged particle track on the order of the molecular to cellular (i.e., nanometers to microns) dimensions is important in understanding the chemical and biological effects of ionizing radiation [2,4,6]. If the charged particle is a recoil nucleus produced by a neutron interaction, the energy deposited and the linear energy transfer can be inferred from this information, allowing the detector to act as neutron spectrometer [3,11]. In principle, the detector can be used for any application that requires knowledge of the three dimensional locations of single electrons in a gas [1,12].

In this paper the experimental apparatus used to make these measurements and the optical and pulsed RF electric field optimization studies we have performed are described. Images of particle tracks produced by α and β particles, and recoil protons from the interaction of high energy neutrons with hydrogenous materials, are also presented to illustrate the sensitivity and track imaging resolution we have obtained with this detector. The quality of the information gained from the detector (i.e. the uncertainty in the locations of each secondary electron produced by the ionizing particle) is a very sensitive function of the gas mixture constituents, purities and pressures used in the chamber, along with the magnitude, duration, and frequency of the pulsed RF electric field. These considerations are discussed in detail.

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Figure 1. Schematic diagram of the Optical Ionizing Radiation Detector. The Imaging optics in front of the camera were replaced by a spectrometer for the pulsed gas discharge spectroscopy measurements.

II. DETECTOR DESIGN

The detector optically images the secondary electrons produced by the passage of ionizing particles through a gas, and is shown in Figure 1. The technique is an adaption of an experimental method originally developed by Cavalleri [13] to determine the electron density in a low electron concentration gas discharge. More recently the technique has been used to measure thermal electron diffusion coefficients and electron drift velocities in Townsend discharges in several gases [14]. We have previously discussed the use of this technique for the measurement of the locations of the secondary electrons produced by the passage of ionizing particles in a gas [1,4,5] and the method is only briefly outlined here. A pulsed, high-voltage (peak voltages up to 30-40 kV), high frequency RF field ($f = 27.125$ MHz stabilized by a crystal controlled 2000 watt oscillator-amplifier) is temporarily applied to the chamber electrodes, immediately following the passage of the ionizing particle through the chamber (Figure 1). In the present measurements, the chamber was either filled with a 0.5-10 % mixture of N_2 in Ar or a 0.1-0.5% mixture of TMAE (tetrakis(dimethylamine)ethylene) in Ar with total gas pressures in the range 2.5-100 kPa, and a corresponding E/N range of ≈ 10 -100 $V\text{-cm}^2$.

The prompt fluorescence from the decay of the gas molecules excited by the passage of the ionizing particle through the ionization chamber is used to trigger the detector. The light is detected by two fast photomultipliers operating in coincidence mode and the output pulse from the coincidence detector is used to trigger the RF field circuit where a short burst of RF voltage (pulse duration ≈ 1 -3 μs) is applied to the chamber electrodes. Alternatively, the RF field circuit can be operated in a pulsed duty cycle mode (the duty cycle for these measurements was usually 0.1% - 10%) with a pulse duration of typically 0.1-10 ms. The electric field is switched off when the photomultipliers detect a burst of light from the discharge produced by the excited electrons in the particle track. Although both techniques were successfully used to obtain images of the particle tracks in the gas, the particle track results presented in this paper were obtained by the latter technique due to the minimization of the electron diffusion before the application of the high voltage RF field when this technique is used. The maximum RF excitation pulse width in this case was $\approx 0.5 \mu s$.

PIXEL NUMBER

PIXEL NUMBER

Figure 2. Image of an α particle track obtained in a gas mixture of 2% N_2 in Ar at a total gas pressure of 55 kPa.

Figure 3. Alpha particle track imaged under the same conditions as those in Fig 2.

The pulsed RF field excites the subexcitation electrons left along the particle's path leading to ionization of the surrounding gas and to the emission of light in the immediate vicinity of the particle track. Under low electron amplification, the amount of light emitted from a given sub volume of the gas will be directly proportional to the initial number of electrons in that sub volume due to the particle track. The detection of the light emission by one or both of the photomultipliers is used to turn off the RF field in the pulsed duty cycle mode before space charge problems or gas breakdown occurs in the chamber. The emitted light is imaged by a fast microchannel plate intensified digital camera (shutter speed $\approx 0.01\text{-}5\ \mu\text{s}$) and recorded on a 512×512 CCD pixel array within the camera (Photometrics Series 200 camera). The image is digitized and transferred to a computer (PC 486 clone) for later analysis.

III. TRACK IMAGING RESULTS

Images of two α particle events are shown in Figures 2 and 3, and were produced by a ^{232}Th source located at the top electrode of the chamber. These α particle tracks were produced in a gas mixture of 2% N_2 in Ar at a total gas pressure of 55 kPa. The images show two of the general types of α particle tracks that we have detected with this chamber: i.e. conventional tracks with a high degree of ionization near the stopping point of the track (Figure 2), and more unexpectedly, double ended tracks with a high degree of ionization at both the beginning and end of the track (Figure 3). The reason for the track structure observed in Figure 3 is presently not understood, but may be related to charge stripping of the α particle during part of its passage through the gas. The half width of the track image shown in Figure 3 $\approx 0.7\text{-}1.0$ mm, giving a corresponding unit density resolution of $\approx 0.6\text{-}0.9\ \mu\text{m}$.

Images have also been obtained of β particle tracks from the ^{232}Th source, and two of these are shown in Figure 4. These images indicate that, in contrast to the α particle tracks, the β particle undergoes many large angle atomic scattering collisions during its energy decay. The energy deposition for β particles is considerably more irregular than that for the α particles showing regions of dense electron production (and hence high energy deposition) and other regions with very little ionization of the gas (and hence low energy deposition). A Monte-Carlo simulation of an electron track produced by a 20 keV β particle in liquid water is shown in Figure 5. The three tracks shown in Figure 5 are the result of electron diffusion at (a) 10 ps,

Figure 4. Images of two β particle tracks obtained from a ^{232}Th source located within the ionization chamber.

Figure 5. Monte Carlo simulation of a 20 keV β particle in liquid water including electron diffusion at (a) 10 ps, (b) 28 ns, and (c) 280 ns after the passage of the β particle through the water.

(b) 28 ns and (c) 280 ns after the passage of the β particle through the water [6]. The β particle measurements shown in Figure 4 were made several hundred nanoseconds after the passage of the β particle through the gas and correspond approximately to the track in Figure 5c. The similarity between the observed β particle track shown in Figure 4b and the calculated track shown in Figure 5c is, although coincidental, remarkable.

IV. GAS OPTIMIZATION STUDIES

Ideally, we require the sensitivity of the optical detector to be such that the exact location of each secondary electron produced during the passage of the ionizing particle through the gas in the chamber can be found. In reality, the performance of the detector is very dependent on the gas and electric field parameters with which the detector is operated. Strong optical emission has previously been observed from the $\text{C}^3\Pi_u - \text{B}^3\Pi_u$ transition in pulsed 10 MHz RF discharges in N_2 [15], and our initial studies focussed on this gas in dilute mixtures with Ar due the high emission coefficients previously observed from this system [16]. The emission spectrum from a 1% N_2/Ar pulsed gas discharge is shown in Figure 6 in comparison with the spectral response of the intensified CCD camera with a quartz and a standard 50 mm objective lens. The spectrum given in Figure 6 indicates that emission in this gas mixture occurs predominantly at 337.1 nm [$v = 0-0$], 357.7 nm [$v = 0-1$], 380.5 nm [$v = 0-2$] and other higher vibrational levels from the second positive system. Somewhat surprisingly, no emission from the first negative system ($\text{B}^3\Sigma_u^- - \text{X}^2\Sigma_u^+$) was observed in the present studies, while Sato and Haydon [15] have observed strong emission at 391.4 nm [$v = 0-0$]. Plane parallel electrodes were used to create a uniform electric field in the present studies, while the measurements performed by Sato and Haydon were made with a point-plane electrode arrangement. The higher electric fields near the tip of a rod electrode are expected to lead to excitation of higher lying states than for uniform electric field

Figure 6. The quantum efficiency of the intensified CCD camera with a standard optical 50 mm objective lens and a 50 mm quartz objective lens. Also shown are the major lines from the nitrogen emission spectra in a pulsed N_2/Ar gas discharge.

excitation, possibly leading to the observed spectral lines from N_2^+ in the earlier study [15].

The present measurements show that the quartz lens is required if the full optical emission from the discharge is to be used to image the particle tracks in this gas mixture. In practice, the standard crown glass/flint glass lens must be used even though light below about 350 nm is not transmitted. Spherical aberration problems (leading to a loss in image resolution) were found to be too severe with the quartz objective. As a result, only approximately 10% of the total light generated by the N_2/Ar gas discharges is used to obtain the images in Figure 2-4.

Typical RF electric field voltage waveforms for a gas mixture containing 1.0 torr of N_2 in Ar are shown in Figure 7a at a total gas pressure of 200 torr, and at a total gas pressure of 300 torr in Figure 7b. Also shown is the light output from the 337.1 nm line in N_2 in these two gas mixtures. These waveforms illustrate the major problem with applying the RF voltage after the passage of the ionizing particle through the detector. With the tank excitation circuit we use to produce the pulsed RF electric field [5], it takes 2-4 μs for the voltage level across the discharge chamber electrodes to rise to a point where significant ionization and excitation in the gas can occur and thus before significant light emerges from the discharge. Significant thermal diffusion of the secondary electrons on the particle track can occur during this time leading to a loss of resolution of the initial track and to reduced information on the energy deposition in the particle track (see Figure 5).

The fluorescence decay times observed in the light output measurements in Figure 7 were 6.4×10^{-7} sec. at 200 torr and 4.4×10^{-7} sec. at 300 torr. These decay times are much longer than the expected 9 ns decay time for the $C^3\Pi_u$ state of N_2 at these gas pressures [15, 17]. A significant fraction of the emission from the N_2/Ar gas mixtures is expected to come from the collisional quenching of high lying metastable states in Ar by the N_2 leading to additional emission from the second positive group [18]. The apparent decay times decrease with increasing gas pressure primarily due to the loss of Ar metastables from the formation of Ar excimers in three body collisions with Ar and N_2 [17].

Figure 7. Typical RF voltage waveforms and 337.1 nm light output from pulsed RF 27 MHz discharges in a 1.0 torr N_2 gas mixture with Ar at a total pressure of (a) 200 torr and (b) 300 torr.

Figure 8. Prominent N_2 emission lines from the RF discharges in N_2 /Ar gas mixtures as a function of the percentage of N_2 .

Figure 9. Prominent emission lines from N_2 in a 0.5% N_2 /Ar gas mixture as a function of the total gas pressure.

Optical emission measurements have been performed in N_2 /Ar gas mixtures as a function of the partial pressure of N_2 and as a function of the total gas pressure to find the optimum gas mixture which maximizes the light output from the discharge, thus giving the highest detector sensitivity. These results are shown on Figures 7 and 8 for several of the strongest emission lines in N_2 . At very low partial N_2 pressures, the second positive band emission is a simple function of the N_2 concentration. At higher N_2 concentrations, energy loss due to vibrational excitation of N_2 alters the high energy tail electron energy distribution function in the discharge, reducing the $C^3\Pi_u$ excitation coefficient. At the low total gas pressures, the nitrogen emission increases with the N_2 (direct excitation) and Ar (collisional quenching) concentrations. At the higher gas pressures, Ar excimer formation and N_2 collisional quenching compete with N_2 emission to reduce the rate of increase in the $C^3\Pi_u$ emission. These measurements indicate that to optimize the light output from the detector, gas mixtures composed of $\approx 0.5\%$ N_2 at total gas pressures above 200 torr are required.

V. CONCLUSIONS

We have developed a new type of ionizing radiation detector which can be used to visualize the particle tracks produced by an ionizing particle as it traverses an ionization chamber. We have shown that the detector is capable of imaging α and β particles with a resolution of < 1 mm. The detector has also been shown to have the sensitivity to resolve single electrons. Measurements of the detector's performance have been made in N_2/Ar and TMAE/Ar gas mixtures (only the results for the N_2/Ar gas mixtures have been presented in this paper). Future improvements will give a detector resolution down to ≈ 0.1 mm by reduction in the electronic delays, and the proper choice of gas mixtures, and RF electric field strength, frequency, and duration.

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