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A PHOTOIONIZATION DETECTOR FOR THE DETECTION OF XENON LIGHT

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ABSTRACT

A photoionization detector is described with a high quantum efficiency at wavelengths shorter than about 2000 Å. This instrument, coupled to a xenon gas scintillation proportional counter has an energy resolution of 9.5% FWHM at 6 keV. The vapor pressure and absorption coefficient of TMAE are measured and the prospects of an imaging gas scintillation proportional counter are discussed.

1. INTRODUCTION

There has been a great deal of effort recently to develop an improved method to detect ultraviolet light.¹⁻³ The primary interest is to replace the photomultiplier tube (PMT) used with a xenon-filled gas scintillation proportional counter (GSPC), although there is also much interest in developing an instrument to detect Cerenkov radiation.⁴ Because the emission spectrum of xenon is in the ultraviolet (ranging from about 1500-1950 Å, peaking at about 1670 Å),⁵ PMTs with Spectrosil windows have been used. Although other techniques have been suggested for replacing the PMT to detect xenon light, the most promising is the photoionization detector (PID). The PID is a proportional counter with an optical window and filled with an appropriate gas that converts the incident ultraviolet photons to electrons through the photoelectric effect. The coupling of a PID to a GSPC was first suggested by Pollicarpo,¹ who showed that if a sufficient fraction of the ultraviolet photons produced by the GSPC can be detected in the PID, a substantial improvement in energy resolution should be possible.

The advantages of the PID over the PMT to detect the light from a xenon GSPC are numerous. PIDs can be constructed with almost any dimension and with a variety of windows. They are more rugged and compact than PMTs and,

in principle, can be constructed at a lower cost than PMTs with equivalent window areas. Since the PID is "solar blind," i.e., insensitive to visible radiation, the problem of light leaks is eliminated. The PID would also be a very useful replacement for the PMT for applications in high or varying magnetic fields where the GSPC has been shown to be useful.⁶ Since the light-detecting substance of a PID is a gas, the quantum efficiency across the window should be uniform, unlike that of the PMT. Finally, using the technology of imaging proportional counters (IPC), a PID can be constructed to give event location to high accuracy, which would be useful for Cerenkov radiation imaging and production of an imaging GSPC.

Charpak, Policarpo, and Sauli² have demonstrated a GSPC coupled to a PID (called a Photoionization Proportional Scintillation Chamber, or PIPS, by the authors). A krypton GSPC was coupled to a PID filled with 83% argon, 3% TEA (triethylamine), and 14% methane. An energy resolution of 10.8% FWHM was measured at 5.9 keV. Unfortunately, the photoionization potential of TEA is too high for the detection of xenon light, and the high natural background of krypton limits its usefulness. An additional problem with the use of krypton and TEA is that windows such as LiF_2 or MgF_2 must be used, which are expensive and difficult to work with.

We have recently demonstrated a xenon-filled GSPC coupled to a PID.⁷ To avoid the cumbersome acronyms associated with GSPCs, we suggested the more descriptive name "Scinprop" (scintillator + proportional counter) for a scintillator coupled to a PID.

2. TMAE

2.1 The Tetraaminoethylenes

To make a useful PID, a gas with a low ionization potential, high quantum efficiency for ultraviolet light, and adequate vapor pressure is required. Of all the organic compounds reported to date, the tetraaminoethylenes have the lowest ionization potentials (see Refs. 8-12). Table I gives the photoionization potential in the gas phase (I_g) and the photocurrent threshold (i_{th}) when they are dissolved in tetramethylsilane (TMS) of tetrakis(dimethylamino)ethylene (TMAE), 1,1',3,3'-tetramethyl-2,2'biimidazolidinyliene (TMBI), 1,1,4,4-tetrakis(dimethylamino)butadiene (TMAB), and N,N,N',N'-tetramethyl-p-phenylenediamine (TMPD). TEA is included as a reference.

Another group that may prove useful for the detection of ultraviolet light is the organo-metallic compounds known as the ocenes which have low ionization potentials and possibly acceptable vapor pressures.¹³ Table I also includes the ionization potentials of cobaltocene, chromocene, nickelocene, and ferrocene. Of this group, we have only made an unsuccessful test of ferrocene.

The most promising of the tetraaminoethylenes is TMAE (pronounced "Tammy"). It has the lowest ionization potential and appears to be the most available. Nakato, Ozaki, and Tsubomura¹¹ have measured the relative photo-ionization cross section of TMAE shown in Fig. 1. As a reference, the emission spectrum of xenon gas and the approximate xenon spectra after passing through a 4 mm thick Spectrosil window and a 5 mm thick CaF₂ window are included. The cross section of TMAE is at a maximum over the Spectrosil-xenon spectrum. There are no TMAE cross section data below 1675 Å, but it is likely that the cross section of TMAE remains high over the entire xenon spectrum. It appears that the spectral response of TMAE may be well matched to the CaF₂-xenon spectrum.

2.2 Handling and Purification

TMAE has proved to be quite easy to work with when a few precautions are taken. It reacts with the oxygen in the air so that it should be handled in an inert environment, although handling it in air seems to do little harm if it is later purified. TMAE, or its decay products, also react with such materials as vacuum grease and O-ring materials. (The degradation of Viton has been found to be much less than that of Neoprene.) It does not seem to react with Teflon. Thus, in the design of an instrument, organic materials should be avoided. When these precautions are taken, the TMAE is stable.

There are several techniques for purifying TMAE:

Method 1.¹⁴ TMAE is shaken under nitrogen with a small amount of water and separated with a separating funnel. This is repeated until the water is clear. This procedure removes the oxidation products of tetramethyl oxide, tetramethyl urea, and dimethyl amine. The TMAE is then dried with a 4 Å or 5 Å molecular sieve.

Method 2.⁹ Alumina may be added directly to the TMAE to remove the products of oxidation.

Method 3.¹¹ Distillation.

Method 4. The TMAE is placed in a small bottle connected to an all-metal bellows valve. The TMAE is exposed to a vacuum until a noticeable amount (~10%) has been removed. The TMAE is then ready for use. We have developed this method.

2.3 Vapor Pressure

Early measurements of the vapor pressure of TMAE were quite discouraging, giving pressures of only 0.01 torr and 0.9 torr at 30°C and 90°C respectively.¹⁵ These pressures were not consistent with our experience so we made our own measurements.

In our vapor pressure measurements, a calibrated MKS Baratron with a 3 torr head and a calibrated thermometer with 0.1°C divisions were used. The system was kept at an elevated temperature (≥50°C), and the TMAE sample was kept in a water bath to control the temperature of the sample. Three sets of data were taken, using two samples of TMAE. Fig. 2 shows the vapor pressure (P) plotted versus the reciprocal of the absolute temperature (T) and the temperature in degrees Centigrade.

The vapor pressure of TMAE is expected to obey the Clausius-Clapeyron equation:

$$P = P_0 \exp \left[\frac{\Delta H}{R} \left(\frac{1}{T_0} - \frac{1}{T} \right) \right] \quad (1)$$

Here, P_0 and T_0 are the reference pressure in torr and temperature in Kelvin, respectively. ΔH is the molar heat of vaporization and R is the molar gas constant.

The best fit to the data is

$$P = 0.577 \exp \left[6454 \left(\frac{1}{300} - \frac{1}{T} \right) \right] \quad (2)$$

where $T_0 = 300$ K was chosen for convenience. This yields a vapor pressure at room temperature (20°C) of 0.35 torr.

2.4 Absorption Cross Section

The experimental arrangement used to measure the absorption cross section of TMAE is shown in Fig. 3. It consisted of a PID with 2 CaF_2 windows separated by 5 mm, coupled to a xenon-filled GSPC with a Spectrosil

window. The PID was filled with 600 torr of P-10 (90% argon, 10% methane) and TMAE. The space between the CaF_2 windows was connected to a vacuum system and a bottle of TMAE. The TMAE was prepared according to Method 4, described above, and kept in a water bath.

In operation, the GSPC was irradiated with 5.9 keV x rays to give a pulsed ultraviolet light source between about 1950 Å and 1700 Å. The absorption cell was evacuated and a pulse height spectrum was taken with the PID, yielding a pulse height I_0 . The absorption cell was then exposed to the TMAE, which is held at a fixed temperature, and another pulse height spectrum taken, yielding I . The ratio of the two pulse heights is related to the linear absorption coefficient (μ) by

$$\frac{I}{I_0} = \exp(-\mu x), \quad (3)$$

where x is the cell depth. The results of the measurements of the absorption coefficient (μ) for TMAE, plotted versus the reciprocal of the absolute temperature and the temperature in degrees Centigrade, are shown in Fig. 4. These data have not been corrected for the small inaccuracy due to the different temperatures and thus different densities in the system. The best fit is

$$\mu = 0.731 \exp[6510(\frac{1}{300} - \frac{1}{T})] . \quad (4)$$

The absorption coefficient should be proportional to the pressure and, in these measurements, the exponent ($\frac{\Delta H}{R}$) for the fits of μ and P differ by less than 1%. These two measurements may be considered to confirm each other since they are independent measurements. The 1/e absorption depth in TMAE is about 23 mm and 11 mm at 20°C and 30°C, respectively.

The absorption cross section per atom (σ) for TMAE in the spectral region measured is equal to

$$\sigma = \frac{\mu}{n}, \quad (5)$$

where n is the number of atoms per unit volume. Evaluating expressions (2) and (4) at 20°C and substituting into (5), we get

$$\sigma = 38 \text{ Mb} \quad ,$$

(6)

which is a reasonable result.

3. THE SCINPROP

3.1 Description

A schematic of the scinprop is shown in Fig. 5. The GSPC is a sealed, curved grid counter similar to instruments reported earlier.¹⁶ It has a beryllium x-ray window and a 4.8 mm thick Spectrosil window to pass the ultraviolet photons. The filling was 760 torr of research grade xenon with gas purity maintained by a getter (SAES model ST 171/HI/16-10/300).

The PID is a multiwire proportional counter consisting of twelve square cells, 13 mm on a side and 15 cm long. The anodes are 20 μm , gold-coated, stainless steel wires connected at one end to form a single sensor. The cathode wires are 76 μm , gold-coated stainless steel wires. The window is a 96.5 mm diameter Spectrosil disk, 4.8 mm thick. A grounded wire mesh is placed under this window to complete the cathode plane. An x-ray window, made of 13 μm aluminized Kapton, is provided to test the performance of the PID directly with an x-ray source. The counter is assembled with Viton O-rings. An appendage is provided, as shown, to hold the TMAE. It consists of a capped stainless steel tube and valve. The two parts of the scinprop are coupled without use of optical grease. Since TMAE is solar blind, no effort was made to make the instrument light tight. The PID was filled with TMAE and 380 torr of P-10.

3.2 Performance

A useful parameter for describing the performance of the scinprop is

$$\eta = \frac{P_1}{P_2} \quad . \quad (7)$$

Here, P_1 is the signal pulse height for x-ray events in the GSPC, and P_2 is the pulse height when the same energy x rays are detected by the PID directly (i.e., used as a standard proportional counter).

The mean energy per ion pair for argon-TMAE mixtures has been measured to be about 21 eV.¹⁷ This is a significant improvement over the 26.0 eV per ion pair reported for argon-methane mixtures. Thus, the number of

ultraviolet photons detected per x-ray event is

$$N = 281 \eta \text{ (at 5.9 keV).} \quad (8)$$

For voltage settings of 300 V and 7500 V on the GSPC grids G1 and G2, respectively, and 900 V on the PID anode, η typically equaled 10 for different fillings, although earlier fillings gave values of η as low as 0.3 when the TMAE had not been adequately purified. Thus, about 2800 photons were detected for 5.9 keV x-ray events. Using a ^{55}Fe source, the best resolution measured was 9.5% FWHM, shown in Fig. 6. For comparison with the resolution predicted in the work by Policarpo,¹ the ratio of the number of ultraviolet photons detected by the PID to the number of primary electrons produced in the GSPC per x-ray event, defined in the above work, is

$$L \approx \eta. \quad (9)$$

The predicted resolution for a value of $L = 10$ is about 9% FWHM at 6 keV. This is in fairly good agreement with our measured resolution of 9.5% FWHM at that energy. The discrepancy is probably due to all the anode wires being connected together with different anodes very likely having slightly different gains, degrading the energy resolution. The work of Policarpo shows that if L can be increased by about a factor of 2, the expected resolution at 6 keV would be 8% FWHM. This should be easily obtainable by replacing the Spectrosil window with CaF_2 , which passes approximately 2.5 times as much of the xenon light.

To determine the quantum efficiency of TMAE, the number of photons detected by the PID was compared to the number of photons detected by a PMT. From earlier work,¹⁸ using the same GSPC counter design and gas pressure, and a PMT, it was found that about 880 photons were detected per 5.9 keV x-ray event, with a potential between the grids of the GSPC of 4050 V. The alkali photocathode used had a quantum efficiency of about 24% between 1700-1950 Å, with the reduction in PMT quantum efficiency due to the Spectrosil window.¹⁹ For the PID, which has a window similar to that of the PMT, the average of two measurements made at the same GSPC voltage settings is $\eta = 4$. Thus, about 1120 photons are detected per 5.9 keV x-ray event. This would imply that about 30% of the ultraviolet photons entering the PID are detected. Using the value of η given by Eq. (4), and assuming a

temperature of 22°C, about 50% of the light is absorbed in the 13 mm depth of the counter. This indicates that for a PID with an infinite absorption depth, the absolute quantum efficiency of TMAE to the Spectrosil-xenon spectrum is about 60%, much higher than the PMT efficiency. This number may also increase when measurements are made with a cleaner PID.

4. AN IMAGING SCINPROP

With the coupling of an imaging proportional counter (IPC) to a GSPC, one would hope to have an instrument that combines the good energy resolution of a GSPC with the position sensitivity of the IPC. The energy resolution of a GSPC is a factor of two better than that of the IPCs used in x-ray astronomy. Such an instrument would be very useful at the focal plane of an x-ray telescope where the high resolution is needed to resolve emission lines. A high pressure imaging scinprop would also find use in medical applications such as fluorescent thyroid imaging.

It is possible to estimate the ability of an imaging scinprop to determine the location of absorption of a low energy x ray (<5 keV). For an instrument using an IPC that employs the center of gravity technique,²⁰ the uncertainty in the location determination is given by

$$\sigma_p^2 = \sigma_R^2 + \sigma_1^2 + \sigma_2^2, \quad (10)$$

where

σ_R is the range of the photoelectron in the GSPC,

σ_1 is the fluctuation in the spatial distribution of the ultraviolet photons,

and

σ_2 is related to the size of the fundamental detection unit in the IPC.

It has been shown²¹ that

$$\sigma_1 \approx \frac{h}{2\sqrt{N}}, \quad (11)$$

where h is the average distance of the light production region from the light detection region and N is the number of ultraviolet photons detected per x-ray event. The value of σ_2 is approximately

$$\sigma_2 \sim \sqrt{\frac{d}{N}} \quad (12)$$

where d is the distance between anode wires. For a GSPC filled with 1 atm of xenon, σ_R can be neglected for low energy x rays.

Thus, Eq. (10) becomes

$$\sigma_p^2 = \left(\frac{h}{2\sqrt{N}}\right)^2 + \left(\frac{d}{\sqrt{N}}\right)^2 \quad (13)$$

Referring to Fig. 7, Eq. (13) can be written

$$\sigma_p^2 = \frac{1}{4N} [(S/2 + t + \bar{x})^2 + 4d^2] \quad (14)$$

where S is the distance between the grids of the GSPC, t is the window thickness, and $\bar{x}$ is the mean absorption depth of the ultraviolet light in the IPC. Defining:

$$x = t + \bar{x} \quad (15)$$

and letting n equal the number of photons detected in the IPC per keV of x-ray energy per unit distance between the grids, and E_x equals the photon energy, we have

$$\sigma_p^2 = \frac{1}{4nE_x S} \left[\frac{S^2}{4} + Sx + x^2 + 4d^2 \right] \quad (16)$$

The minimum of Eq. (16) is

$$S' = 2\sqrt{x^2 + 4d^2} \quad (17)$$

$$2x \quad (4d^2 - x^2) \quad .$$

Thus, there is an optimum S for a given IPC geometry and temperature, although it can be shown that σ_p is quite insensitive to S as long as S is greater than this optimum value. Unfortunately, S cannot be made equal to $2x$ for reasonable temperatures because of the high voltages required. For a practical counter, S will probably be limited to about 1.5 cm.

To calculate $\bar{x}$, one must evaluate

$$\bar{x} = \frac{\int_0^D x e^{-\mu x} dx}{\int_0^D e^{-\mu x} dx} \quad (13)$$

where D is the depth of the IPC. The solution to Eq. (13) is

$$\bar{x} = \frac{[e^{-\mu D} (D\mu + 1) - 1]}{\mu [e^{-\mu D} - 1]} \quad (19)$$

For our calculation, we take D to be 3 cm.

From work done elsewhere²² we can take

$$n = 800 \text{ photons cm}^{-1} \text{ keV}^{-1}, \quad (20)$$

for a Spectrosil window. Assuming a factor of 2.5 greater transmission for a CaF_2 window, we have

$$n = 2000 \text{ photons cm}^{-1} \text{ keV}^{-1} \quad (21)$$

Equation (13) may be evaluated, assuming the values of n given by Eqs. (20) and (21), $t = 4$ mm for Spectrosil and $t = 6$ mm for CaF_2 , and $S = 1.5$ cm. Curves (c) and (d) of Fig. 8 show the calculated position resolution for a scinprop, between the temperatures of 20°C and 30°C, with Spectrosil and CaF_2 windows, respectively. The higher the temperature the better the position resolution. The performance of an imaging GSPC using PMTs developed by the European Space Agency, curve (a), and a proposed design, curve (b),²³ are included along with the position resolution of an IPC,²⁴ curve (e). As a better reference, the position resolution of these five instruments at

1 keV given in Table II. Although the position resolution of an IPC is a factor of 2 better than that calculated for an imaging scinprop, the greater stopping power of xenon and the factor of two better energy resolution makes the scinprop a very useful instrument.

5. HIGH ENERGY X-RAY DETECTION

There is a great need in fields such as medicine, industrial radiography, nuclear safeguards, and gamma-ray astronomy for an electronic alternative to film for the detection and imaging of high energy x rays. Although there has already been some good work done along these lines with gas counters,^{25,26} the stopping power of gas is too low to be useful above 100 keV where many of the important applications lie. The primary instrument working above 100 keV is the Anger camera which consists of a thin NaI(Tl) crystal coupled to an array of PMTs. The state-of-the-art position resolution for such instruments is about 2.5 mm.

A great improvement in high energy x-ray imaging would be possible if a crystal could be found that produced scintillations detectable by the TMAE. With even a moderate number of photons detected per event, position resolution of less than a millimeter should be possible.

We have examined CsI(Tl), NaI(Tl), NaI, and BaF₂ in the search for such a crystal. Of the crystals tested only BaF₂, with an emission maximum at 3250 Å,²⁷ gave other than a null result. A 25 mm diameter, 1 mm thick crystal was aluminized on all but one face. This unaluminized face was then coupled to a TMAE and P-10 filled PID without an intermediate window. The pulse height spectra of several sources are shown in Fig. 9 where the abscissa is in ultraviolet photons per event, as determined by using 6 keV x rays directly in the PID. Where more than one source is listed for a curve, the second source gave a pulse height spectrum similar to the first. Table III lists the sources used, the primary photon energies, the half-maximum number of ultraviolet photons per event, and the approximate detection efficiency. As the energy of the γ rays increases, the number of ultraviolet photons and the detection efficiency also tend to increase. Thus, as the γ -ray energy increases, more photons are in the high energy tail of the emission spectrum detected by the TMAE.

Although the results with BaF₂ are discouraging, the principle of detecting the light from a scintillator with a PID is demonstrated. The search will continue for the proper scintillator.

6. CONCLUSION

The chemical IMAL has proved to have a high quantum efficiency at the wavelengths of xenon light, making possible a wide range of improvements in GSPCs. Scinprops should be possible with larger areas and better energy resolution than was possible with PMTs. Useful imaging instruments are possible with both good energy resolution and good position sensitivity. Because IMAL extends the usefulness of the PID into the longer wavelengths, there is none that a crystal scintillator will be found that has a sufficient fraction of its spectrum overlapping the photoionization of IMAL to produce high energy x-ray imaging instruments.

7. התאחדות העובדים – ארגון העובדים הראשון שהוקם, שייך
 למפלגה העבודה.

(.) :הערה, א, (ב) :הערה, ב, (ג) :הערה, ג.

**The 10 Best Ion Beam Unit
at Various Imaging X-ray Unit and 1 keV**

3. . וויסא יאג. (11), 4. . וויסא יאג. (11)

Table III

The Radioactive Source, Primary Energy,
the Half-Maximum Number of Ultraviolet Photons,
Detected per Event, and Efficiency of a BaF₂
Crystal Coupled to a PID.

Source	Primary Energy (keV)	Half-Maximum Number of UV Photons	Detection Efficiency (%)
⁵⁷ Co	125	3.3	0.5
¹³³ Ba	356	4.3	2.0
¹³⁷ Cs	662	5.4	2.5
⁵⁴ Mn	835	5.0	2.6
⁶⁰ Co	1173 & 1332	10.7	3.0
BKγ	1836 & 2038	10.5	5.0

Figure Captions

1. A compilation of the relative photoionization cross section of TMAE (Ref. 11), the emission spectrum of xenon, and the approximate xenon spectra after passing through 4 mm of Spectrosil and 5 mm of CaF_2 as a function of photon wavelength.
2. TMAE vapor pressure as a function of the inverse of the absolute temperature and the temperature in degrees Centigrade.
3. The experimental arrangement used to measure the absorption of xenon light by TMAE.
4. The absorption coefficient of TMAE as a function of the inverse of the absolute temperature and the temperature in degrees Centigrade.
5. Schematic of the scinprop.
6. The measured ^{56}Fe x-ray spectrum. The Mn 6.5 keV K_α line is beginning to stand out on the right hand side of the 5.9 keV K_α line.
7. Definition of terms used in calculating the position resolution of an imaging scinprop.
8. The position resolution, as a function of x-ray energy: (a) measured for an existing GSPC coupled to an array of PMTs, (b) calculated for an improved GSPC/PMT design (See Ref. 23), (c) calculated for a scinprop with a Spectrosil window, (d) calculated for a CaF_2 window, and (e) measured for an IPC (see Ref. 24).
9. The pulse height spectra of several sources detected with a BaF_2 crystal coupled to a TMAE filled PID. Where more than one source is listed for a curve, the second source gave a similar response.

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Figure 1

Figure 2

Figure 3

Figure 4

Figure 5

Figure 6

Figure 7

Figure 8

Figure 9