A benign, low Z electron capture agent for negative ion TPCs

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Abstract

We have identified nitromethane (CH\textsubscript{3}NO\textsubscript{2}) as an effective electron capture agent for negative ion TPCs (NITPCs). We present drift velocity and longitudinal diffusion measurements for negative ion gas mixtures using nitromethane as the capture agent. Not only is nitromethane substantially more benign than the only other identified capture agent, CS\textsubscript{2}, but its low atomic number will enable the use of the NITPC as a photoelectric X-ray polarimeter in the 1-10 keV band.

Key words: Dark Matter, negative ion, X-ray polarimeter, diffusion, ion mobility

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1 Introduction

The negative ion time projection chamber (NITPC) achieves high spatial resolution by transporting charge in the form of negative ions, rather than electrons, thereby reducing diffusion to the thermal limit in both the transverse and longitudinal drift directions\cite{1}. This provides the highest 3-D space-point resolution attainable for long drift distances, particularly where operation at low pressures is required to make very low energy tracks long enough to measure (for example, dark matter WIMP recoil atoms). In relatively low rate

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environments, the NITPC also has the advantage that the ion drift velocities
are about two orders-of-magnitude less than those of electrons. This allows
the use of low-bandwidth electronics that results in lower noise and power
consumption.

The key to reducing diffusion to the thermodynamic lower limit (Equation
1 below) is to prevent net energy transfer from the drift field to the drift-
ing primary ionization[2]. Such “heating” of the drifting charge occurs when
electrons are drifted at reduced fields $E/p > \sim 10-50 \text{ V/m} \cdot \text{Torr}$. Heating is
reflected in an increase of the parameter $\epsilon$ in Equation 1 to values higher than
$\frac{3}{2}kT$, resulting in increased diffusion. For electron drift, gas additives with
large integrated cross sections for inelastic scattering of electrons, such as hy-
drocarbons and CO$_2$, provide channels for dissipation and are known [2,3] to
prevent heating of drifting electrons up to moderate values of the reduced field
($\sim 50 \text{ V/m} \cdot \text{Torr}$).

In contrast, ions drifting in even a high field are very efficiently thermally
coupled to the room-temperature gas, since the ion mass is comparable to
that of the gas molecules. Then, even elastic scattering produces substantial
energy transfer from the drifting ion to the bulk gas. Ion diffusion has been
measured in a number of CS$_2$ mixtures suitable for TPC operation[1,4,5] and
was found to follow the “thermal, low field” limiting behavior[2]:

$$\sigma_D = \sqrt{\frac{4eL}{3eE}} \quad (1)$$

Here $\sigma_D$ is the rms diffusion spread for carriers with charge $e$ after drifting
through a distance $L$ in electric field $E$ with average (thermal) energy $\epsilon$. The
parameter $\epsilon$ (the average thermal energy of a drifting charge carrier) remained
approximately equal to the room-temperature value, up to reduced drift fields
as high as $\sim 2.5 \times 10^3 \text{ V/m} \cdot \text{Torr}$.

Many chemical vapors at STP will capture thermal electrons to form negative
ions[6]. However, to maintain good track resolution the agent must have
a capture mean free path of a hundred microns or less at a relatively low
partial pressure. Since gas gain is desired at the TPC endcaps, the agent
must also relinquish its electrons in the high, strongly inhomogeneous field of
the gain structures, allowing Townsend avalanches to form. In all previous
work, CS$_2$ has been used as the only capture agent fulfilling these requirements
reasonably well.

While the NITPC has been successfully implemented with CS$_2$ as the capture
agent in dark matter observatories[7–9], the utility of the NITPC would be ex-
panded by the identification of new capture agents. CS$_2$ has a combination of
low flash point ($-30^\circ \text{ C}$), high vapor pressure (400 Torr) and low explosive mix-
ture limit in air (1.3%) which require great care in handling the material[10]. Nitromethane[11] has a flash point far above room temperature and a vapor pressure less than a tenth that of CS$_2$. The comparative health effects of the two materials are indicated by the respective ACGIH Threshold Limit Values of 1 ppm for CS$_2$ vs. 20 ppm for nitromethane. CS$_2$ also may form sulfide deposits under conditions of high rate or discharge. More importantly, new applications for the NITPC would be enabled by capture agents with different functional properties. For example, a single-ion counting NITPC with high energy resolution for double beta decay searches has been proposed[12] that would benefit from a capture agent with higher ion mobility than CS$_2$. Another example is photoelectric X-ray polarimetry below 10 keV with a NITPC, which requires capture agents with lower atomic number (see below).

Photoelectric polarimeters derive their information by determining the photoelectron emission direction, which is strongly correlated with the electric field vector of the incident photon. The most sensitive photoelectric polarimeters image the tracks of the photoelectrons with pixels small compared to the photoelectron track length and reconstruct the emission direction on an event-by-event basis. Such polarimeters have been realized with optical avalanche chambers[13,14], micropattern gas detectors with direct pixel readout[15–17], and a micropattern TPC[18].

X-ray polarimetry below 10 keV with a NITPC would benefit from low atomic number capture agents for several reasons. First, the correlation between the photoelectron emission direction and photon electric field is strongest for emission from s-orbitals[19], so it is preferable to use gases whose K-shell energies are well below the X-ray energy. Second, the photoelectron is emitted with a kinetic energy that is the difference between the binding energy and the X-ray energy so that lower Z gases result in photoelectrons with larger kinetic energies and longer track lengths. Finally, lower Z gases have less multiple Coulomb scattering, which results in straighter tracks and a better correlation between the photoelectron emission direction and photon electric field.

Therefore, motivated by the requirements of low energy X-ray polarimetry and guided by the chemical literature on electron capture, we have explored a number of compounds as alternative NITPC capture agents. We have identified nitromethane (CH$_3$NO$_2$) as a suitable capture agent for X-ray polarimetry which is also relatively benign. We present measurements of drift velocity, gas gain and longitudinal diffusion in gases with nitromethane as the capture agent.
Capture agent screening apparatus

Capture agents were screened for negative ion drift velocity, gas gain and longitudinal diffusion at low to moderate fields using a single-wire proportional counter attached to a homogeneous-field drift region. The apparatus was enclosed in a stainless steel bell jar with a simple gas manifold. Drift fields up to $4.0 \times 10^4 \text{ V/m}$ were used with this apparatus at reduced pressure. Photoelectrons were generated at an Sn photocathode attached to the drift-cathode. The photocathode was arranged so that it could be cleaned between runs using a glow discharge in pure argon. This was essential for maintaining the photoelectron yield.

UV light flashes from an EG&G Flash-Pak[20] were admitted into the bell jar through a quartz window, passed through a hole in the proportional counter wall, and struck the photocathode producing photoelectrons. The standard internal capacitors of the Flash-Pak were augmented with additional HV capacitors to give a stored energy of about 0.2 Joule per pulse. The Flash-Pak was triggered by an external pulser, from which a time-zero signal was also derived. The proportional wire signal was read out through an ORTEC 142PC preamp and an ORTEC 579 shaping amplifier. Drift times to the proportional wire and wire-signal time widths were measured with a digital oscilloscope.

Drift velocities and gas gain at moderate to high fields ($1.0 \times 10^5 - 4.0 \times 10^5 \text{ V/m}$) were measured in various gas mixtures with a micropattern gas detector (MPGD) having the same electrode structure as the gas electron multiplier[21], but assembled from two etched stainless steel meshes[18] separated by a 100 µm thick teflon spacer. A drift electrode, also of stainless steel, was placed 10 mm above the MPGD cathode. The MPGD was operated at a gain of about 3000. The MPGD cathode was instrumented with a charge-sensitive preamplifier followed by a bipolar shaping amplifier with a six microsecond shaping time constant.

The MPGD cathode and drift electrode were simultaneously illuminated with a xenon flashlamp (Perkin-Elmer PAX-10), which produced photoelectrons from both surfaces. Photoelectrons from the cathode produced a prompt pulse, while the photoelectrons from the drift electrode produced negative ions and gave a pulse delayed by the drift time across the 10mm drift distance. The drift time was measured by averaging 100 pulses on a digital oscilloscope and taking the difference between the zero-crossing times of the prompt and the delayed pulses.
3 Results and Discussion

Nitromethane is known to have a large capture cross section for thermal electrons[6]. However, at moderate to high drift fields the pure near-saturated vapor at 20 Torr produces both electron-drift and negative-ion-drift signals. This is understandable since a drift field of $5.0 \times 10^4$ V/m at 20 Torr is already a reduced field of 2500 V/m-Torr, easily high enough to raise the energy of drifting electrons out of the thermal range before they could be captured. CO$_2$ was therefore added to the nitromethane vapor. CO$_2$ is known to be very effective in thermalizing drifting electrons, due to its large inelastic scattering cross sections[3]. Mixtures of 20 Torr nitromethane with 50 Torr or more of CO$_2$ were found to have satisfactory characteristics as negative ion drift mixtures.

Drift velocity and diffusion results for the nitromethane:CO$_2$ 20:50 mixture are shown in Figures 1 and 2. The drift velocity rises linearly with drift field, with mobility $(4.27 \pm .03) \times 10^{-4}$ m$^2$/V·sec and a small positive intercept $(1.44 \pm .08$ m/s). The diffusion is studied by plotting the square of the pulse width in time as a function of $1/(v_d^2 E_d)$. The pulse width is expected to be the quadrature sum of the amplifier shaping time (10 µsec) and the broadening due to diffusion. If the diffusion is governed by Equation 1, this plot will be linear in the region where diffusion dominates (essentially the entire plot), with slope. Fitting Figure 2 to a straight line gives $T = 313 \pm 25$ K.

Drift velocities for higher pressure gas mixtures of nitromethane-CO$_2$-neon, nitromethane-CO$_2$-argon and nitromethane-CH$_4$ were measured up to drift fields of $4.0 \times 10^5$ V/m. Results are shown in Figure 3. The linear dependence of drift velocity on drift field persists up to these very high fields. Results of linear fits for the mixtures studied are shown in Table 1. Shown in Figure 4 are gas gain curves for higher pressure mixtures of CO$_2$-neon, and neon-CO$_2$-nitromethane. Gains as high as several thousands were obtained in all mixtures demonstrating the ability of nitromethane as a feasible, productive detector gas.

4 Conclusion

A negative ion drift mixture of nitromethane with CO$_2$ is found to have linear drift velocity vs. field and to exhibit thermal-limit longitudinal diffusion up to drift fields as high as $2.8 \times 10^4$ V/m. Higher pressure mixtures with argon, neon, CH$_4$ and CO$_2$ also show linear drift velocity up to fields as high as $4.0 \times 10^5$ V/m. All mixtures exhibit stable operation as NITPCs.
The introduction of nitromethane as an electron capture agent will enable the use of the NITPC as an X-ray polarimeter in the 1-10 keV band. The low drift velocity of the NITPC greatly eases the difficulty of calibrating drift velocity and substantially reduces power consumption, which is particularly important for satellite-borne astronomical instruments. The low diffusion of the NITPC will also enable large-volume photoelectric polarimeters that could be used, for example, as wide field-of-view instruments for unpredictable astronomical transients, such as gamma-ray bursts, or with rotation modulation collimators for high-resolution imaging polarimetry of solar flares[22].

Acknowledgements

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Fig. 1. Drift velocity $v_d$ vs. drift field $E_d$ for Nitromethane:CO$_2$ 20:50
Fig. 2. Longitudinal diffusion data for Nitromethane:CO$_2$ 20:50, plotted as the square of the pulse time width vs. $1/v_d^2 E_d$ for 80 mm drift.
<table>
<thead>
<tr>
<th>Mixture</th>
<th>CO₂ Torr</th>
<th>Neon Torr</th>
<th>Argon Torr</th>
<th>Methane Torr</th>
<th>Mobility ([m^2/V\cdot s])</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>510</td>
<td>170</td>
<td>–</td>
<td>–</td>
<td>8.43 ± .01 × 10⁻⁵</td>
</tr>
<tr>
<td>B</td>
<td>170</td>
<td>-</td>
<td>510</td>
<td>–</td>
<td>9.69 ± .01 × 10⁻⁵</td>
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<tr>
<td>C</td>
<td>340</td>
<td>340</td>
<td>–</td>
<td>–</td>
<td>10.5 ± .01 × 10⁻⁵</td>
</tr>
<tr>
<td>D</td>
<td>170</td>
<td>510</td>
<td>–</td>
<td>–</td>
<td>13.1 ± .02 × 10⁻⁵</td>
</tr>
<tr>
<td>E</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>380</td>
<td>21.0 ± .07 × 10⁻⁵</td>
</tr>
</tbody>
</table>

Table 1
Fitted slopes for ion drift in mixtures of 20 Torr Nitromethane with CO₂, Neon, Argon and Methane.

Fig. 3. Drift velocity at high fields in mixtures of Nitromethane and CO₂ and/or Neon, Methane mixtures A-E described in Table 1. Circles: A, Plusses: B, Squares: C, Crosses: D, Triangles: E
<table>
<thead>
<tr>
<th>Mixture</th>
<th>CO₂ Torr</th>
<th>Neon Torr</th>
<th>Nitromethane</th>
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<tbody>
<tr>
<td>A</td>
<td>210</td>
<td>490</td>
<td>-</td>
</tr>
<tr>
<td>B</td>
<td>170</td>
<td>510</td>
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</tr>
<tr>
<td>C</td>
<td>340</td>
<td>340</td>
<td>20</td>
</tr>
<tr>
<td>D</td>
<td>510</td>
<td>170</td>
<td>20</td>
</tr>
</tbody>
</table>

Table 2

Fig. 4. Gas Gain in mixtures containing Neon and CO and/or Nitromethane A-D described in Table 2. Plusses: A, Triangles: B, Circles: C, Squares: D
References