SEEDING AGENTS FOR SIMULATION OF IONIZING TRACKS BY TWO-PHOTON IONIZATION ABSORPTION

A. Bamberger, R. Isele, J. Schlüpmann and M. Stegle
University of Freiburg, Germany

ABSTRACT

Strong ionization yield has been observed for counting gases doped with aromatic substances using a quadrupled Nd-YAG laser at 266 nm. The yield of benzene, toluene, M.xylene, 1,3,5-mesitylene, isopropylbenzene, and n-butylbenzene varies within one and a half orders of magnitude, benzene being the lowest of the investigated substances. The signals for admixtures of 1 ppm exceed the background level by several orders of magnitude. Minimum ionizing particles can be simulated with less than 100 ppb admixtures of cumene at laser intensities as low as 0.1 μJ/mm². In any normally operated counter, signals are observed at moderate intensities. We suggest as an explanation, that some benzene derivative being present could be responsible for the signal at 266 nm.

(Submitted to Nuclear Instruments and Methods in Physics Research)
1. INTRODUCTION

The track simulation by laser ionization for calibration purposes has been investigated by several authors [1-8]. The two photon absorption leading to ionization has been proven to work successfully at convenient photon energies and reliable laser systems. A requirement for the calibration of large track detector systems is the quantitative control of the yield, which depends among other factors on the amount of ionizable substances in counting gases. The additives used up to now have the disadvantage of being either sticky because of their low vapour pressure (phenol or TMPD) [7, 20] or not very convenient to handle (TMA)[8]. Signals due to laser ionization have been observed in undoped counting gases. The search for the identification of the contaminants responsible for the background signal is continuing at present [7-9]. This work gives a possible clue for their identification. It also gives the yield of the ionization signal and describes the properties of aromatic substances that could be used as seeding agents in counting gases.

2. APPARATUS

The layout of the experiment is shown in figure 1. A quadrupled Neodym-YAG pulsed laser (Quanta Ray DCR-2) is used for the experiment. According to the specifications the pulse length is about 4-5ns. After the frequency selector the beam is defined by an aperture of 1.95 mm diameter and enters the chamber through a quartz window. The laser beam transverses the chamber parallel to the signal wire and can be displaced within the sensitive volume of the chamber by a movable optical bench. The laser intensity is measured pulse by pulse by a pyroelectric joulemeter (GEN TEC Ed 100) mounted behind the chamber. Additionally, a system consisting of a beam splitter, a frequency analyzer, a filter and a photo diode serves to monitor the beam intensity. The correspondence between photo diode and joulemeter is verified for the intensity interval investigated. Since the relation between photo diode and joulemeter is the same both for evacuated and pressurized counter the attenuation of the beam within the sensitive volume of the chamber is negligible.
The chamber has a sensitive volume of 1 meter length and 10 cm diameter. The thickness of the central signal wire is 100 μm. The chamber can be operated both in the ionization and in the proportional mode at gas pressures ranging from 4 mbar to 4 bar. The measurements are done in the ionization mode. The calibrated charge signal is checked in the proportional mode with X-rays of a ¹⁸⁵Cd source and with cosmic rays traversing the apparatus. The correct dependence of the charge signal on the radial position of the laser beam and on the high voltage at both polarities is verified. In this way also the effect of electron attachment is found to be negligible.

The gas system is shown in figure 2. The essential part is a device for filling the seeding agent into the system. The substances are liquid at normal conditions. They are introduced by an injection needle into the evacuated vessel through a Si-rubber septum. For a concentration of 1 ppm at 2 bar, liquid of about one microliter is needed (the total volume being about 52 l). The air leaking into the system during the operation of seeding does not influence the background signal.

A purification system similar to that used for the prototype of the OPAL detector [11] is connected with the counter. The main components are a Cu-catalyst (BASF, R3-11), a molecular sieve (Union Carbide, Type 3A) and a spiral for freezing out components of the counting gas. The circulation is provided by a double membrane pump (Neuberger, NK 143.AN12E). The \( O_2 \)- and \( H_2O \)-contents in the system are measured by an oxymeter (AMS, OA1440) and a hygrometer (Panametrics, System IV) respectively. The purification system is used to achieve low background conditions. This can also be achieved by pumping and exchanging gases. A mixture of 90% argon and 10% methane is used as counting gas.

3. ELECTRONICS AND DATA ACQUISITION SYSTEM

The signal of the counter is amplified by a charge sensitive preamplifier (Tennelec Model 142A) followed by a shaping amplifier (Ortec Model 450). The signal of the photo diode is also shaped in a main amplifier and delayed according to the delay of the counter signal due to the drift time of the charge carriers. The joulemeter signal is amplified
by a factor 100 in a voltage amplifier. The signals of the chamber and
the photo diode are digitized by peak sensing ADCs (LeCroy 2259) and the
signal of the joulemeter by a charge sensing ADC (LeCroy 2249A). The
CAMAC-system is interfaced with a PDP 11/40 by a GEC-ELLIOT system.

4. OPERATION

All seeding agents investigated in this work are liquid at normal
temperature and pressure. We found it most convenient to seed the counter
with a given amount of the fluid. The gas system is evacuated together
with an appendix consisting of a valve followed by a short tube which is
sealed with a Si-rubber septum at the other end. During normal operation
of the counter the valve remains closed. To inject the seeding agents a
needle is inserted into the septum and the liquid is sucked into the
vessel. Typically, after several minutes the equilibrium of the pressure
within the system and the stability of the ionization signal are
established. Then the system is filled up to 2 bar with a mixture of 90/10
argon-methane. Successive dilution of the doped gas with fresh gas varies
the partial pressure of the seeding agent appropriately.

The data of a given mixture is recorded for one or several different
settings of the flash light intensity of the laser. For each setting 1000
laser pulses are taken. Typically the intensity varies between 0.1 and
10\,\mu\text{J} per pulse. The inevitable variation from pulse to pulse is almost
one order of magnitude. These measurements are used to establish the
functional dependence of the ionization signal on the intensity of the
laser light. The photo diode is used as the reference for the light
intensity because its signal to noise ratio is better than the one of
the joulemeter. The absolute energy calibration of the photo diode is
established by the joulemeter.

5. BACKGROUND CONDITIONS DURING THE MEASUREMENTS WITH SEEDING AGENTS

Between measurements with different seeding agents the counter is
evacuated by a rotary pump and a turbo molecular pump. It is observed that
the background signal decreases every time the counting gas is exchanged.
Nevertheless, there is always enough laser power available to create a measurable ionization signal even without gas amplification.

6. RESULTS FOR AROMATIC SUBSTANCES

The investigated substances are listed in table 1 along with the main properties like the ionization potential, the melting point, the boiling point and the prominent UV-absorption lines near 266 nm. They were selected from 71 hydrocarbons containing only hydrogen, carbon, nitrogen and oxygen. For all the considered hydrocarbons the ionization potentials are known, except for ethylvinylether, which is a homologue of methylvinylether having an ionization potential of 8.93 eV and unpleasant polymerizing properties. The latter is therefore discarded as possible seeding agent. Out of the above class of hydrocarbons those being liquid under normal conditions and having an ionization potential below 9.3 eV are selected.

The results of the ionization signal for aromatic molecules at various laser intensities are collected in figure 3. The concentration is 1 ppm at a pressure of the counting gas of 2 bar. The first six agents of table 1 are shown. Except for benzene the yields at a given laser intensity vary within one order of magnitude, isopropylbenezene (Cumene) being the most efficient agent. Toluene, n-butylbenzene, xylene and 1,3,5-mesitylene are less efficient, followed by benzene.

The uncertainty of the ionization signal is governed by the uncertainty of the dosage of the liquid. The reproducibility is estimated not to be better than 20% in general. A special problem was encountered for xylene where the reproducibility was somewhat worse than for the other aromatic substances. The general uncertainty of the abscissa is governed by the determination of the energy density. It is simply derived from the quotient of the pulse energy and the area of the aperture. By inspection, however, it is seen that the intensity of the beam is not homogeneous. Without recording the profile this systematic uncertainty is difficult to estimate. Since no focusing system was employed, our estimation of the effective area of 3mm² might still be useful for the comparison of different experimental arrangements.
In figure 3 the continuous line represents a fit according to the formula

\[ N = A I^n, \]  

(1)

where \( N \) is the number of charge carriers, \( I \) the energy density of the laser beam. The proportionality constant \( A \) and the exponent \( n \) are fit parameters. This exponent varies between \( n = 1.85 \) and \( n = 2.03 \).

The background is measured with a fresh gas mixture of 90/10 argon-methane. The purity of argon is 99.999% and of methane 99.995%. This gas is introduced after a series of measurements with seeding agents and after the evacuation of the vessel for several hours with the turbo molecular pump. In figure 4 the ionization signal of the background is shown. The exponent of the formula (1) is compatible with \( n = 2 \) over 2 1/2 orders of magnitude in the charge signal. No change of the exponent within the investigated intensity interval is observed.

Figures 5a and 5b show examples of approximate proportionality of the charge signal with the concentration of the seeding agent. One realizes that background conditions as described above are reached at a concentration of the seeding agent of only 10 ppb approximately.

The stickiness of the investigated substances is small, as can be inferred from the dependence of the ionisation signal on the concentration of these substances. This dependence is shown in figure 6 for a laser intensity of \( 1 \mu J/mm^2 \). The ionization signal of the agents \# 1 to 6 of table 1 seem to be proportional to the concentration. The high vapour pressure and small polarizability of the aromatic substances used in this work explains this proportionality, in contrast to aromatic substances like phenol having a polar -OH group.

A proportionality between the total gas pressure at a given concentration of the seeding agent is observed down to 0.5 bar. In figure 7 this is shown for 1 ppm toluene in the counting gas at pressures between 0.5 to 4 bar. As indicated below the pressure dependence might be different at still lower pressure.
It is observed that the $\text{O}_2$-catalyst as well as the molecular sieve remove the seeding agents effectively. It is conceivable that the $\text{O}_2$-catalyst operates also as an adsorbent since 1 g of the material has a surface of 200 m$^2$.

7. RESULTS FOR NON AROMATIC HYDROCARBONS

The substances listed in table 1 under item 7 through 9 produce ionization yields inferior to the background of the present experimental arrangement, which is not believed to be minimized. The upper limit of the ionization charge signal is 150 e$^-$/cm/mm$^2$ at a laser intensity of 1 $\mu$J/mm$^2$ for furan, tetramethylethylene and ethylvinylether at concentration of 50 ppm at 2 bar.

8. INTERPRETATION OF THE RESULTS

The marked difference of the ionization yield between the aromatic and the other substances can be interpreted in the following way. The two photon absorption ionization proceeds through an intermediate state, which is either nearly real or virtual. The inspection of the last column of table 1 reveals that the UV-absorption near 266 nm is only possible for the aromatic substances. The levels populated in this case result from the electronic excitation of the benzene ring. On the other hand, strong UV-absorption for furan and tetramethylethylene is observed at shorter wavelengths (at about 200 nm). The catalogue of UV-absorption spectra states an optical thickness at 266 for furan, which is two orders of magnitude lower than for xylene. A similar statement can be made in the case of tetramethylethylene by extrapolating existing data to 266 nm.

The exponent in formula (1) is close to $n = 2$. This suggests that the basic process is two photon resonance absorption into the continuum, see also ref. [18]. Even if the energy absorbed with the two photons exceeds the ionization threshold by only 0.1 eV, as for benzene, the ionization yield is still high compared with non resonant processes like for furan or tetramethylethylene.
Preliminary measurements indicate that there might be some additional enhancement of the signal due to collision broadening \[19\]. We fill 0.4 µl of xylene and some methane as quencher into the counter, as a result the total pressure is 4 mb. At this pressure the ionization signal is lower by a factor 10 than at 2 bar.

9. BACKGROUND

The ionization signal in the counter without additives is typically between 150 and 500 e⁻/cm/mm² at 1 µJ/mm². The observed slope is close to \( n = 2 \). The signal depends much on the history of the treatment of the vessel. The more often the counting gas is exchanged the lower the background signal becomes. It is higher again when the vessel has been opened or has not been used for some time.

The following observations of the variation of the background signal with pressure and temperature give some indications on the properties of the contaminants being responsible for the background signal.

i) Figure 8 shows the independence of the ionization signal on the total gas pressure at two different laser energies.

ii) The purification system reduces the signal by a factor 2 to 6 depending on the initial amplitude of the signal. The signal drops very rapidly in the beginning and levels out after 10 to 30 minutes. It rises again when the purification system is bypassed.

iii) As shown in figure 9 the heating of the vessel surface to about 60°C increases the signal by a factor 3 and the reverse effect happens if the vessel is cooled down again to room temperature. During this operation other components of the experiment are kept at room temperature.

Presumably some of the laser active contaminants are deposited on the walls of the vessel and can evaporate into the sensitive volume. Independently of the total gas pressure, the partial pressure increases with temperature and decreases during the cleaning operation until equilibrium is reached.
From our measurements it can be inferred that all aromatic substances have similar yields for laser induced ionization 266 nm. Some are sticky like phenol [20] and others are volatile as shown in our investigation. It is known that traces of aromatic substances occur in tracking chambers.

10. CONCLUSION

This work describes the use of aromatic seeding agents to simulate particle tracks with laser induced ionization. It demonstrates:

i) A high yield of ionization of aromatic substances which are commonly used as solvents and for cleaning purposes. Therefore they are not expected to influence even the long term operation of a tracking chamber.

ii) How to control the quantity of the seeding agent filled into the counter.

iii) How to remove the agent if desired.

Moreover the high yield of aromatic substances and the stickiness of some of them suggest that the background signal at 266 nm is, at least partially, due to aromatic substances.

11. ACKNOWLEDGMENT

The authors would like to acknowledge the help of W. Eberbach, Department of Chemistry, Freiburg, and to thank H. Börner, CERN, and K.M. Smith, Glasgow, for enlightening discussions.
REFERENCES


Table I

<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>benzene</td>
<td>C₆H₆</td>
<td>9.24</td>
<td>5.3/80.1</td>
<td>259</td>
<td>[12]</td>
</tr>
<tr>
<td>2</td>
<td>toluene</td>
<td>C₇H₈</td>
<td>8.82</td>
<td>-95/110</td>
<td>267</td>
<td>[13]</td>
</tr>
<tr>
<td>3</td>
<td>m-xylene</td>
<td>C₈H₁₀</td>
<td>8.56</td>
<td>-48/139</td>
<td>267</td>
<td>[14]</td>
</tr>
<tr>
<td>4</td>
<td>1,3,5-mesitylene</td>
<td>C₉H₁₂</td>
<td>8.40</td>
<td>-45/165</td>
<td>(266)</td>
<td>[15]</td>
</tr>
<tr>
<td>5</td>
<td>isopropylbenzene(Cumene)</td>
<td>C₉H₁₂</td>
<td>8.69</td>
<td>-96/152</td>
<td>266</td>
<td>[16]</td>
</tr>
<tr>
<td>6</td>
<td>n-butylbenzene</td>
<td>C₁₀H₁₄</td>
<td>8.69</td>
<td>-88/183</td>
<td>(265,268)</td>
<td>[17]</td>
</tr>
<tr>
<td>7</td>
<td>furan</td>
<td>C₄H₄O</td>
<td>8.89</td>
<td>-86/31</td>
<td>(208)</td>
<td>[15]</td>
</tr>
<tr>
<td>8</td>
<td>tetramethylethylene</td>
<td>C₆H₁₂</td>
<td>8.30</td>
<td>-74/73</td>
<td>(198)</td>
<td>[15]</td>
</tr>
<tr>
<td>9</td>
<td>ethylvinylether</td>
<td>C₄H₈O</td>
<td>?</td>
<td>-116/35</td>
<td>?</td>
<td>--</td>
</tr>
</tbody>
</table>

TABLE CAPTION: Properties of seeding agents.
FIGURE CAPTIONS

Fig. 1 Layout of the experiment.

Fig. 2 Layout of the gas system.

Fig. 3 Charge signal versus laser intensity for different aromatic substances.

Fig. 4 Charge signal versus laser intensity for the background condition after the measurements with the aromatic seeding agents.

Fig. 5a Charge signal versus laser intensity for Cumene (isopropylbenzene) at different concentrations at 2 bar.

5b Charge signal versus laser intensity for toluene at different concentrations at 2 bar.

Fig. 6 Charge signal versus seeding agent concentration at 1 μJ/mm².

Fig. 7 Charge signal versus gas pressure of the counting gas added with toluene of 1 ppm concentration.

Fig. 8 Charge signal of the background at different gas pressures.

Fig. 9 Dependence of the charge signal on the temperature of the vessel.
OPTICS and ELECTRONICS

FIG. 1
FIG. 3
FIG. 4
FIG. 5b
FIG. 6
PRESSURE DEPENDENCE OF IONIZATION IN
Ar/CH₄ + 1ppm TOLUENE

IONIZATION (e⁻/cm² mm²)

GAS PRESSURE (bar)

laser intensity: 0.6μJ/mm²

FIG. 7