

Characterization of Ar/N₂ gaseous mixtures for MPGD-based detectors

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Abstract

We report a spectroscopic study of primary scintillation of Ar/N₂ mixtures obtained under X-ray irradiation, for different concentrations of N₂ (0%-100%) and pressures (1-5 bar). Special care was taken with respect to Nitric Oxide (NO) contamination in the chamber, which was found to produce strong scintillation at the ppm level, in accordance with previous works. Scintillation of N₂ was determined to be independent of the existence of NO. We show the good performance of N₂, in comparison to CF₄, as VUV-quencher and wavelength shifter, where the highest scintillation yield was measured in the range of 0.5% - 10% of N₂ concentration.

I Introduction

Micro-Pattern Gas Detectors (MPGDs) amplify weak signals produced by the ionization of gas atoms caused by charged particles passing through the detector. Electrons released during primary ionization are accelerated by strong electric fields, triggering avalanche multiplication that boosts the signal to detectable levels. Traditionally, these signals are read out electronically, which allows efficient coverage of large detector areas at high rates. However, electronic readout systems face limitations in spatial granularity and event reconstruction complexity.

An emerging alternative is the optical readout of gaseous scintillation. This technique captures light emitted during the de-excitation of ionized atoms or molecules, enabling high-spatial-resolution imaging and offering enhanced spatial and temporal discrimination capabilities complementary to electronic readout.

Noble gases are commonly used in gaseous detectors due to their simple interaction mechanisms at low energies, predominantly elastic scattering, making primary as well as secondary ionization simpler to obtain, see Figure 1. Ionization in noble gases produces scintillation light in the vacuum ultraviolet (VUV) range, which presents two main challenges: its detection requires specialized equipment, and it can induce photoelectric emission from detector surfaces —known as *photon feedback*— which may destabilize the detector. To address this, VUV-quenchers are added; these eliminate the VUV photons and suppress *photon feedback*. Some quenchers, like carbon tetrafluoride (CF₄), also scintillate at longer wavelengths through *wavelength-shifting*, allowing optical readout in the visible range [1; 2].

Despite their effectiveness, many commonly used VUV-quenchers have high global warming potential (GWP) [3; 4], motivating the search for environmentally friendly alternatives [5]. Nitrogen (N₂), an abundant and benign gas, is a promising candidate due to the existence of quasi-resonant transitions between argon atomic and excimer

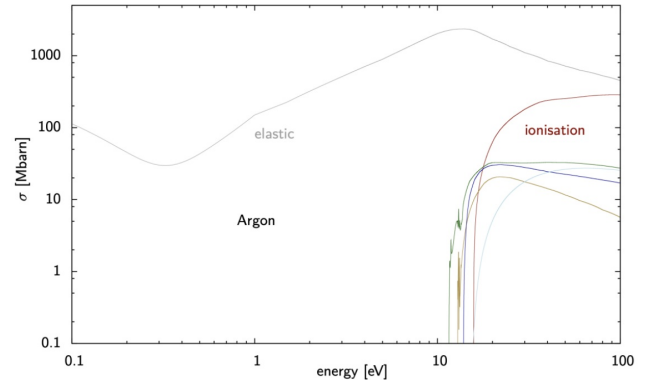


Figure 1: Interaction channels of argon (Ar) at low energies.

states and the C³Π_u level of N₂. The subsequent decay to the metastable B³Π_g state results in scintillation emission around 300–400 nm, effectively wavelength-shifting the VUV light from argon (Figure 2) [6; 7]. Prior studies on Ar/N₂ mixtures have mainly explored electrical readout, see G. Croci *et. al.* [8].

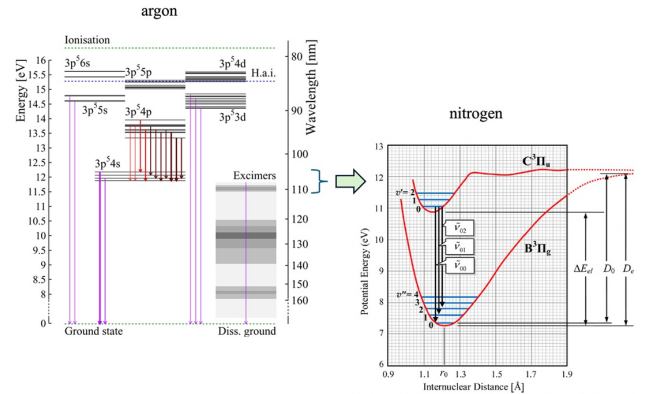


Figure 2: Energy-level diagram illustrating quasi-resonant transitions between argon and nitrogen, enabling VUV quenching and wavelength-shifting. [6; 7]

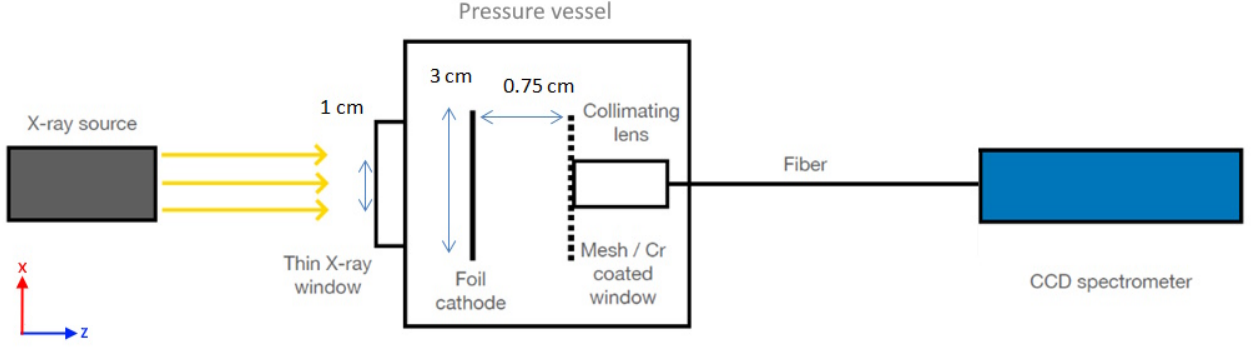


Figure 3: Schematic of the experimental setup: X-ray source, thin aluminium (Al) window, foil cathode, gas gap of 0.75 cm, and anode with collimator.

In this work, we investigate the primary scintillation of Ar/N₂ mixtures over nitrogen concentrations from 0% to 100% and pressures from 1 to 5 bar. Our goal is to evaluate the scintillation efficiency of N₂ as a VUV-quencher and wavelength shifter compared to standard CF₄ mixtures, characterizing these environmentally friendly alternatives for gaseous detector applications.

II Methods and Materials

Figure 3 shows a schematic drawing of the experimental setup. Measurements were performed on a CF63 aluminium-cube serving as a vessel, irradiated with X-rays from a copper tube at 40 kV. The chamber had an entrance window of 1 cm-diameter made of a thin aluminium foil of 50 μm thickness which was facing the tube. Inside the chamber, an electrically biased cylindrical volume was placed, with 3 cm in diameter and 0.75 cm in height. Its upstream electrode served as a cathode and was made from the same foil as the window. A semitransparent Cr served as the anode (OceanOptics 74-UV) leading to a multi-mode optical fiber (UV-VIS, 600 μm core) and finally coupled to an OceanOptics FX UV-VIS CCD spectrometer sensitive in the 210-800 nm range. The photon spectrometer was calibrated using a lamp with reference light sources for the UV and the visible regions, coupled to the anode mesh. Both calibrations were merged at around the 300 nm mark.

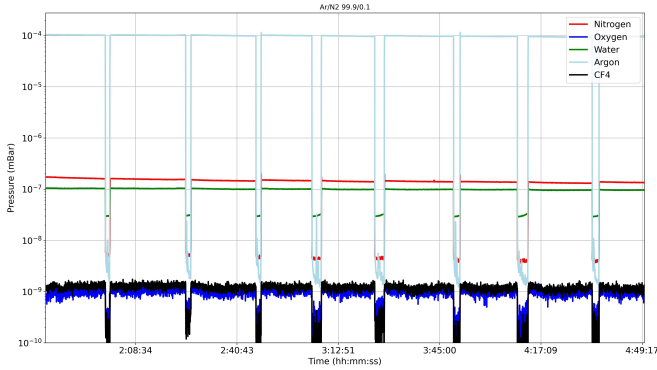


Figure 4: Relative pressures analysed by the RGA during spectroscopy measurement. Ar, N₂, H₂O, CF₄ and O₂.

Upon excitation and ionisation of the gas, electrons and ions were collected by means of a uniform electric field,

the current being read at the anode with a Keithley picoammeter (model 6487). Previous work using the same arrangement, shows that inside the collimated region, ionization can be regarded as uniform throughout these measurements. [9]

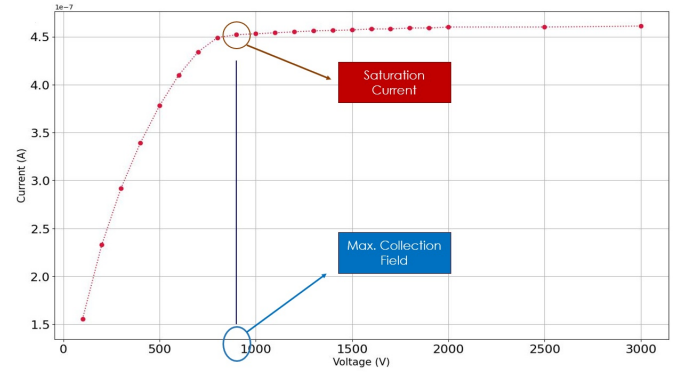


Figure 5: Curve of collected current as a function of the field applied. The saturation current, as the maximum collection of electrons in the ionised gas, is determined as the first point of the plateau. The correlated voltage applied is called maximum collection voltage (MC).

Prior to the measurements, the chamber was pumped down to below 5×10^{-4} mbar. The Ar/N₂ mixtures were studied at N₂ volume fractions of 0%, 0.1%, 0.5%, 1%, 5%, 10%, 20%, 50%, 100%, and pressures from 1 to 5 bar. The chamber was first filled with N₂ until the desired partial pressure, using two pressure/vacuum gauges, namely a Pfeiffer Vacuum PCR 280 Pirani/Capacitance and an MKS pressure transducer, for the readings. Afterwards, argon was admixed. Gas circulation was deemed unnecessary for proper mixing, as the concentration could be verified by sampling the gas into a residual gas analyzer (RGA) and waiting for the ratio of the pressures of the species to stabilize. This agrees with the notion that the forced flow of argon gas, being dominant by at least a factor 10 in volume, drives the mixing in such a small chamber. For the lowest N₂ concentrations, that would be limited by the accuracy of the sensor, the filling was done at high pressure and diluted until the target concentration was achieved.

Purity was monitored continuously with a residual gas analyser (RGA) coupled to the main system through a leak valve (see Figure 4). The RGA region was kept at a constant pressure of 3×10^{-5} mbar throughout the mea-

surements by adjusting the leak-valve opening. The main impurities in the system were water (H_2O), oxygen (O_2), Nitric Oxide (NO) and Carbon tetrafluoride (CF_4). Their concentrations were estimated to be below 100 ppm, with NO being a particular case of study (see Section III), since its scintillation at the ppm level is very strong.[10; 11]

The final scintillation spectrum was divided by the current at full collection field. Figure 5 shows the determination method of the saturation current (SC) and the correlated field applied (maximum collection voltage). As the *plateau* of the current for greater fields than the maximum collection voltage is not strictly constant, the uncertainties in the determination of SC were taken as the difference between the first point in the *plateau* and the current measured at 3kV applied.

Figure 6 shows the scintillation spectra of Ar/ N_2 99.9/0.1 at different pressures, as an example of the spectrum measured with the setup. As it can be seen, scintillation of Nitric Oxide (NO) is measured inside the chamber, even though it was not placed there on purpose. Contamination of this source, estimated at the ppm level, was measured in previous works [10; 11]. The range of emission from this source (from 200nm to 300nm) is, in principle, undesirable as it can produce *photon feedback* and damage the stability of the detector, depending on the metal from which it is made. In the range from 300 - 450 nm, the scintillation of N_2 takes place and finally the infrared scintillation of Ar, in the range from 680nm to 800nm. The pink spectra corresponds to Ar/ CF_4 95/5 mixture, measured in this setup. This is used as the reference spectra throughout this study, as it is one of the most used gaseous wavelength-shifters nowadays. This mixture produces scintillation in the range of 500 to 750nm and its spectra is pressure-invariant, as previous works claim. [1; 2]

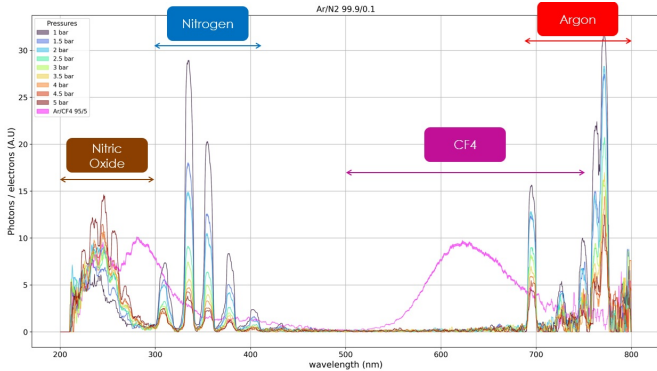


Figure 6: Different scintillation spectra. We see the scintillation spectra of Ar 99.9% N_2 0.1% (Ar/ N_2 99.9/0.1), for different pressures. In pink, the scintillation spectra of Ar 95% CF_4 5%, used as the reference. From left to right: the scintillation of NO contamination, scintillation of N_2 , visible scintillation of CF_4 and infrared scintillation of Ar.

III Results and Discussion

Systematic measurements of different Ar/ N_2 mixtures were carried out for pressures from 1 to 5 bar. Results are shown in figure 8. An increment can be seen in the scintillation of N_2 (300nm - 450nm) from 0.5% to 10% of N_2 in

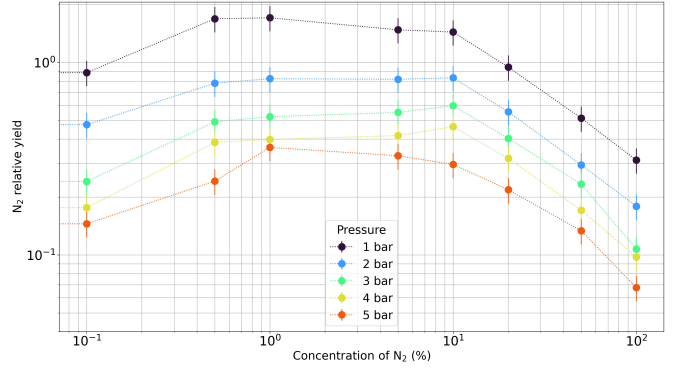


Figure 7: Scintillation yield ratio as function of the N_2 concentration. The scintillation yield ratio between Ar/ N_2 mixtures from 300 to 450 nm and Ar/ CF_4 95/5 from 500 to 750 nm is shown. A *plateau* is seen in the range of 0.5% - 10% of N_2 concentration. The scintillation yield is pressures dependent, being 1 bar where the best scintillation performance is seen.

the mixture, and then a decrease until 100%; corresponding to the scintillation of pure N_2 inside the chamber. The filled area in Figure 8 represents the scintillation yield of Ar/ N_2 mixtures and the reference. This yield is the integral from 300 to 450 nm for Ar/ N_2 mixtures, and from 500 to 750 nm for the reference, that corresponds to the visible emission of Ar/ CF_4 . The relative scintillation yield of Ar/ N_2 mixtures to Ar/ CF_4 95/5 mixtures, defined as the ratio between its yields, as a function of the N_2 concentration is shown in Figure 7. There exists a *plateau* between 0.5% and 10% of N_2 concentration, this range being the best operating region of Ar/ N_2 mixtures. Furthermore, the scintillation yield ratio is pressure-dependent, offering 1 bar the best scintillation performance for Ar/ N_2 mixtures for all concentrations. At 1 bar and with concentrations of N_2 in the range 0.5% to 10%, the scintillation yield of Ar/ N_2 is comparable to the Ar/ CF_4 95/5, as the ratio is close to 1. It should be noted that the scintillation yield of Ar/ N_2 at very small quantities of N_2 such as 0.1% is greater than the scintillation yield of pure N_2 for all pressures.

A study on the effect of the N_2 scintillation yield due to the NO contamination was carried out. Figure 9 left-hand plots show the subtraction of the contaminated spectra and the clean one for pressures from 1 to 5 bar and concentrations of 5%, 20%, and 50%. On the right-hand plots, the ratios of the contaminated and clean spectra scintillation yields ($Y_{\text{Cont.}}$ and Y_{Clean} respectively), taken as the integral between 300 to 450 nm, are shown. In the studied range, both yields seem comparable within an overall 20% systematic shift, which is compatible with the systematic variations that are typical in our setup and have been reported earlier [2; 9]. This makes the scintillation of NO an independent process of the scintillation of N_2 [12]. The concentration of NO is not well known. Based on previous works [10; 11] and the relative scintillation of NO with respect to N_2 in this experiment, the limits of NO concentration are set between 3 and 30 ppm for the contaminated spectra (see Appendix A).

Measurements on the field-dependence of Ar/ N_2 mixtures were done by measuring the spectra without drift field applied and with maximum collection field applied. Figure 10 left-hand plots, show the subtraction of this

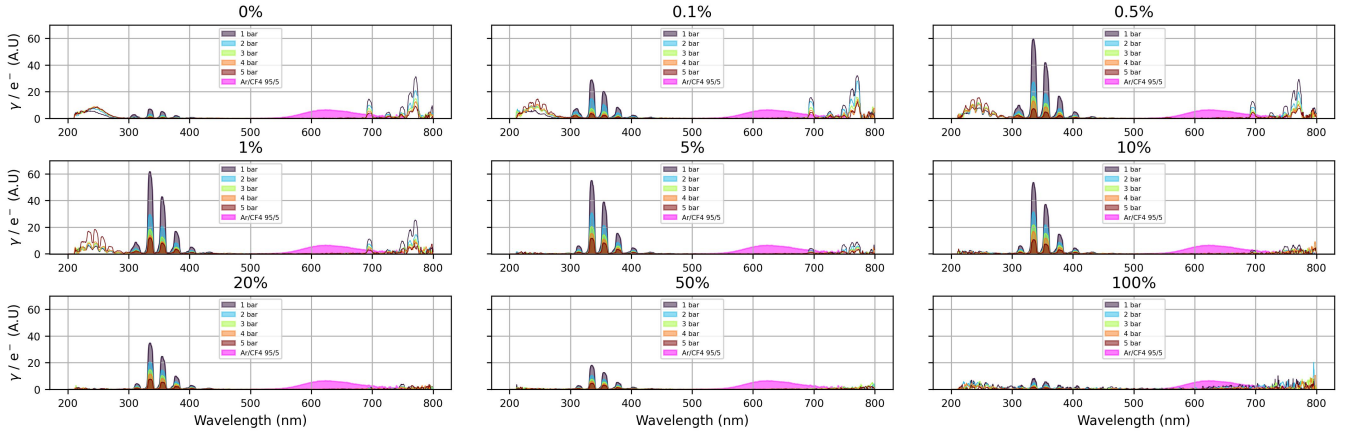


Figure 8: Systematic measurements of Ar/N₂ mixtures scintillation spectra. Each plot corresponds to a certain concentration of N₂, and presents the pressure dependence of this spectra, in the range of 1-5 bar. In pink, the reference Ar 95% CF₄ 5%. An increment of the scintillation of N₂ in the region of 0.5% - 10% can be seen. The filled area indicates the scintillation yield for Ar/N₂ mixtures and for the reference. For N₂ the accounted range is 300-450 nm and 500-750 nm for the reference. The scintillation in 0% plot corresponds to N₂ contamination.

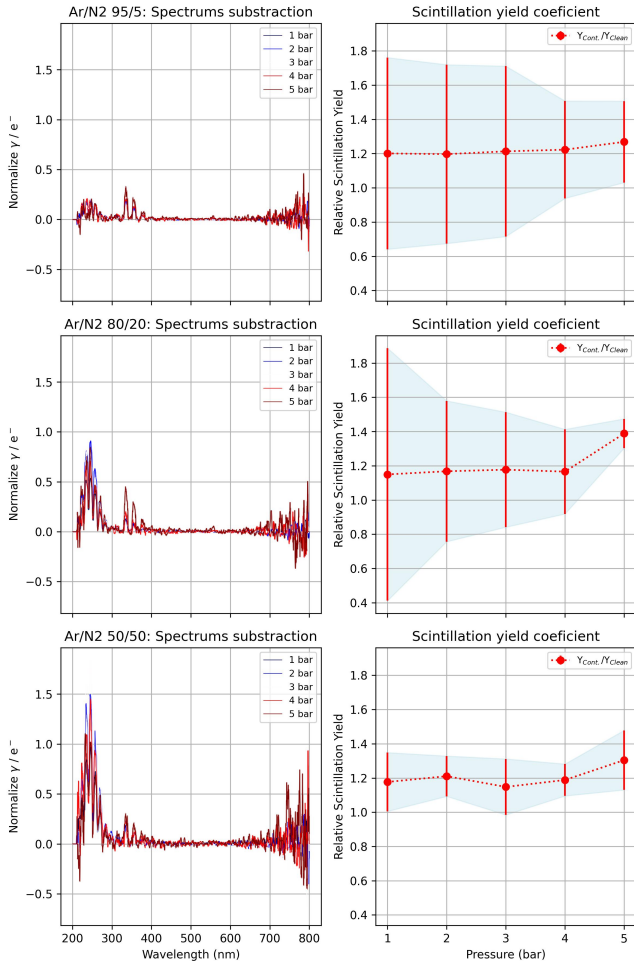


Figure 9: Relative difference scintillation in contaminated and clean spectra. Left column: relative difference between NO contaminated spectra and clean spectra, to the clean scintillation spectra for different concentrations. Right column: ratio of scintillation yield of NO contaminated spectra ($Y_{Cont.}$) and the clean one (Y_{Clean}).

two spectra for each pressure, and for the same concentrations mentioned above. On the right-hand plots, the scintillation field ratio between maximum collection voltage applied and with 0V field, as the integral between 300 to 450nm, is shown. For the three concentrations stud-

ied, the ratio is similar to 1 for all pressures; this result is compatible with the non-existence of space charge and the consequent absence of scintillation from recombination processes.

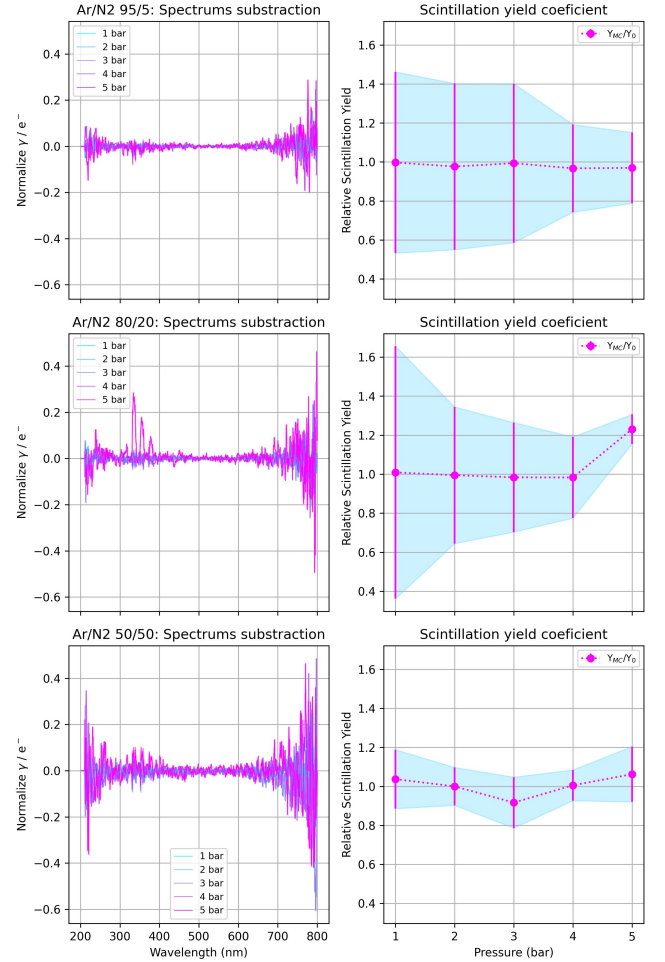


Figure 10: Relative difference scintillation of 0V and maximum collection voltage (MC) applied to Ar/N₂ spectra. Left column: relative difference between MC spectra and 0V spectra, to the 0V scintillation spectra for different concentrations. Right column: ratio of scintillation yield of MC field applied spectra (Y_{MC}) and 0V field applied (Y_0).

IV Conclusions

In this work a spectroscopic study on primary scintillation of Ar/N₂ mixtures, for different concentrations of N₂ and pressures from 1 to 5 bar, was carried out in order to determine the scintillation yield of these mixtures and the quality of N₂ as a VUV-quencher and wavelength-shifter. All spectra were compared with the scintillation spectra of Ar/CF₄ 95/5 used as the reference. This mixture is widely used in gaseous detectors and greatly studied in the literature.

The effect of oxygen (O₂) contamination in the chamber was studied. It was found that the consequent production of Nitric Oxide (NO) allows a transition from the metastable state of N₂ to the first excited state of NO, which then decays to its ground state, producing scintillation in the 200-300 nm range. This contamination produces strong scintillation, making NO a possible scintillation source. Nonetheless, this emission can produce *photon feedback*, damaging the stability of the detector. Further investigation is required in this field. The NO scintillation mechanism and the scintillation yield of Ar/N₂ mixtures were measured to be independent.

Systematic measurements of scintillation spectra of Ar/N₂ mixtures, with concentrations of N₂ from 0% to 100% and pressures from 1 to 5 bar, were carried out. The scintillation yield of N₂, defined as the integral in the range from 300 to 450 nm, presents a *plateau* between 0.5% and 10% of N₂ concentration, where the highest scintillation is observed. It also presents a dependence on pressure, with best performance at 1 bar.

A Concentration of NO

After establishing that the scintillation in the 200–300 nm range originated from the presence of NO inside the chamber, the spectra of pure N₂ at 5 bar (see Fig. 11) were recorded to quantify the NO contamination. During this measurement, the concentration of NO was estimated to be 30 ppm, based on the ratio of the partial pressures of the main NO and Ar peaks with the RGA.

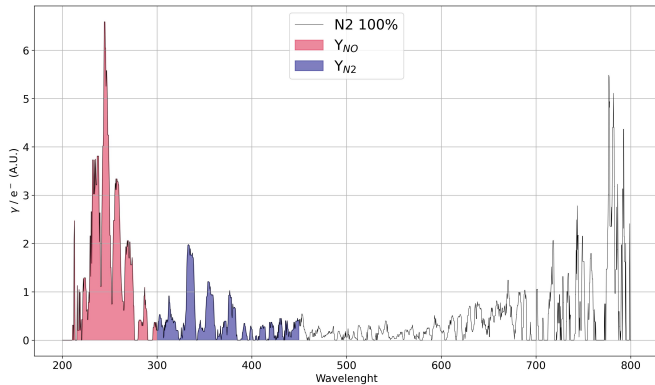


Figure 11: Scintillation spectra of pure N₂ at 5 bar. The concentration of NO measured by the RGA was 30 ppm. The red curve corresponds to the scintillation yield of NO (Y_{NO}), determined between 200 and 300 nm, while the blue curve corresponds to the scintillation yield of N₂ (Y_{N_2}) from 300 to 450 nm.

The work of T. Kerst and J. Toivonen [10] presents a model of NO concentration based on the ratio of the

scintillation yields of N₂ and NO. Figure 12 shows the estimated NO concentration, according to this model, for the contaminated spectra of Ar/N₂ mixtures with 5%, 20%, and 50% N₂.

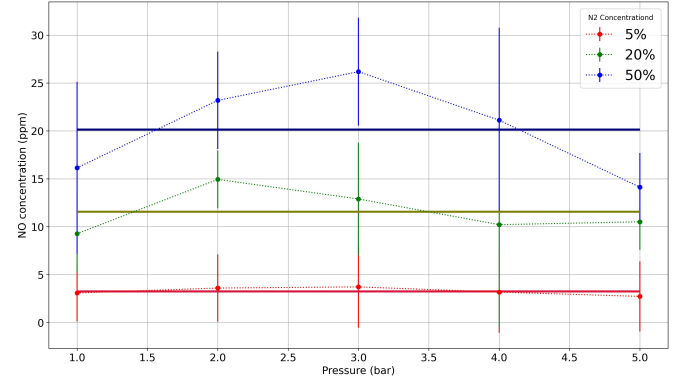


Figure 12: Concentration of NO for Ar/N₂ mixtures at 5%, 20%, and 50% N₂, for pressures from 1 to 5 bar. The concentrations for each mixture are obtained from the relation between the scintillation yield ratio (NO/N₂) in the given mixture and the one obtained for 100% N₂, for which 30ppm were directly measured with the RGA (see Figure 11). The horizontal lines indicate the mean value of NO contamination across all pressures, for each mixture.

The oxygen (O₂) contamination inside the chamber originated from the presence of a plastic pipe in the N₂ line, connecting to another setup, in parallel, however closed at its end. Consequently, it might be expected that the amount of O₂ carried by the N₂ flux is proportional to the amount of N₂ introduced into the chamber. Table ?? compares the expected and measured contamination levels for Ar/N₂ mixtures at 5%, 20%, and 50% N₂, for which contamination was performed in a controlled manner. The results are consistent with this interpretation.

N ₂ Concentration	Expected (ppm)	Estimated (ppm)	Combined (ppm)
100%	30		
50%	15	20	17(8)
20%	6	11	8(7)
5%	1.5	3	2(3)

Table 1: Comparison between NO contamination, expected (under the assumption that the amount of O₂ contamination remains constant in the N₂ line), and estimated (based on the transfer model sketched in [10]). The estimation of 30 ppm was based on the ratio of the partial pressures of the main NO and Ar peaks, measured with the RGA.

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