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TRD gas regeneration by cryogenic distillation

C. Garabatos[1], F. Hahn[2], G. Tsiledakis[1], and A. Wasem[2]

[1] GSI, Darmstadt, Germany
[2] CERN, Geneva, Switzerland

Abstract

The needs of the ALICE TRD xenon-based gas mixture in terms of regeneration from nitrogen contamination are discussed. Two existing cryogenic plants for the N_2 separation from Xe-CO_2 mixtures have been thoroughly tested and their performance quantified in terms of N_2 removal efficiency and Xe losses. The results of these tests are presented and discussed.

1 Introduction

The ALICE Transition Radiation Detector (TRD) [1] is an array of 540 gas chambers which operate with 28 m^3 of Xe–CO_2 [85–15]. Because of the high cost of xenon (5 €/l), provisions for limiting the losses of gas will be made in the construction of the detectors and in the design of the associated gas system.

Leaks, however small they are, lead to an inevitable loss of xenon. In addition, air diffuses into the gas mixture through the same leaks. Oxygen –and water outgassed by some of the assembly materials of the detectors– are, as usual, filtered out by purifiers such as copper catalysers, which do not trap any xenon or CO_2. Nitrogen and, to a lesser extent, argon, cannot be separated from the mixture with these purifiers. A procedure to separate nitrogen from the mixture must therefore be foreseen and applied with a periodicity determined by the magnitude of the leaks and the maximum tolerable concentration of N_2 in the gas.

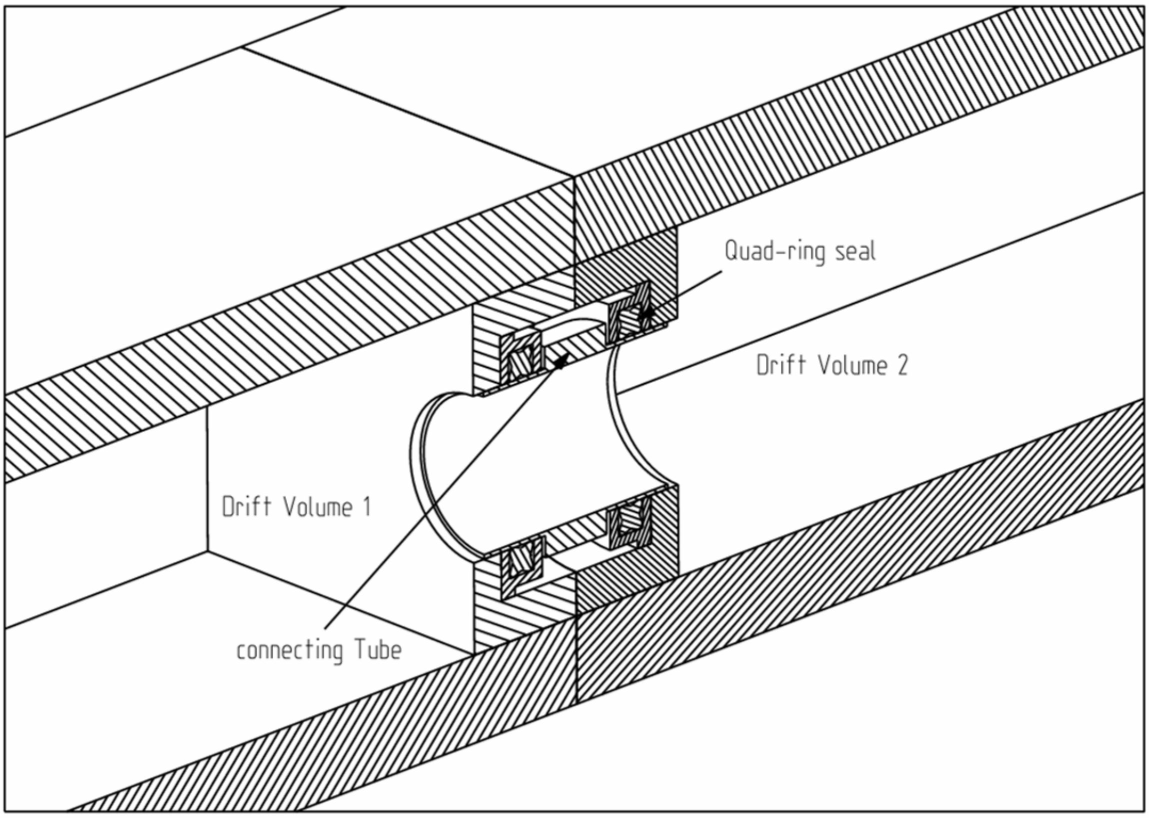

Fig. 1. Drawing of the proposed chamber-to-chamber connection with two 4-fold clover shaped O-rings for sealing.

The main potential source of leaks in the whole TRD assembly is the chamber-to-chamber gas connection, since they amount to 2×540 sealings. The gas system itself and the sealed gas enclosures of the chambers themselves are supposed to be ideally tight. The chamber-to-chamber connection is conceived as a short piece of peak tube inserted into the side walls of each chamber and sealed with a special O-ring as shown in Fig. 1. A leak test of this union has been carried out by flushing 10 l/h of argon through two stesalit sealed volumes connected with this gas feed-through concept. Fig. 2 shows the oxygen increase through the system as a function of the tilt angle of one of the chambers, both in the ALICE azimuth and beam direction orientations. The maximum recorded oxygen contamination of 10 ppm corresponds to a leak rate $L = 10^{-4}$ l/h. The extrapolated leak rate in ALICE is simply this number times 540 chambers, i.e. $L_{ALICE} = 0.054$ l/h. This number translates into 260 l/year of xenon lost through these leaks, i.e. less than 1500 €/year.

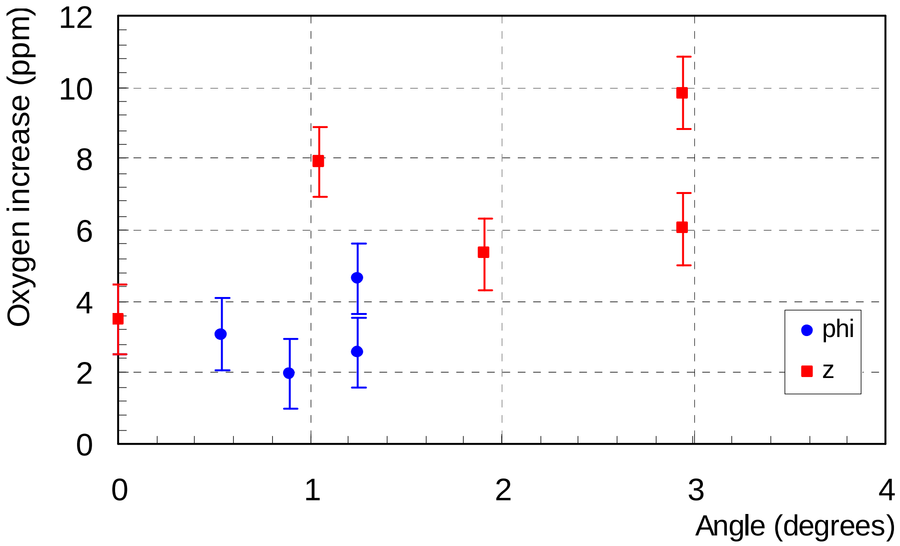

Fig. 2. The measured oxygen leaking through the chamber joint as a function of the tilt angle between chambers, for a gas flow of 10 l/h.

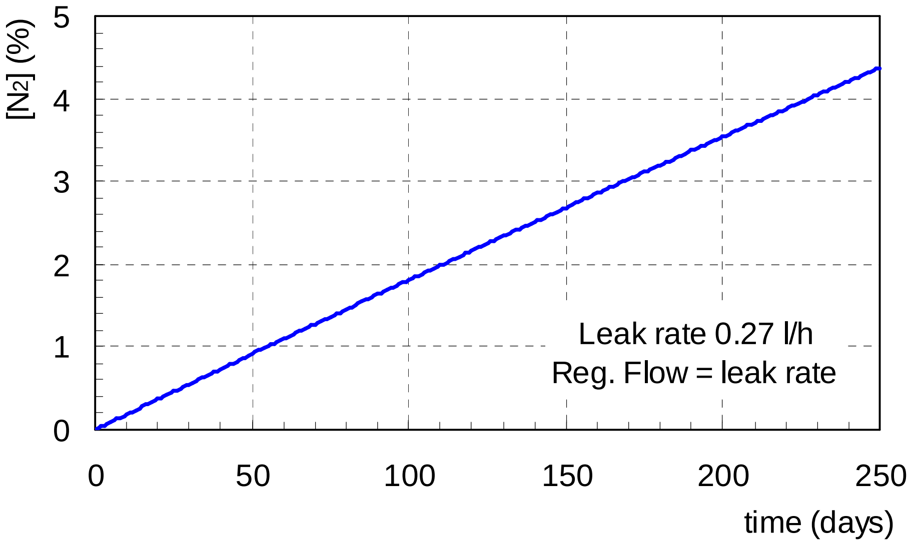

Fig. 3. Estimated evolution of the nitrogen concentration in the TRD gas mixture for a leak rate 5 times higher than expected from the contribution of the chamber-to-chamber connection.

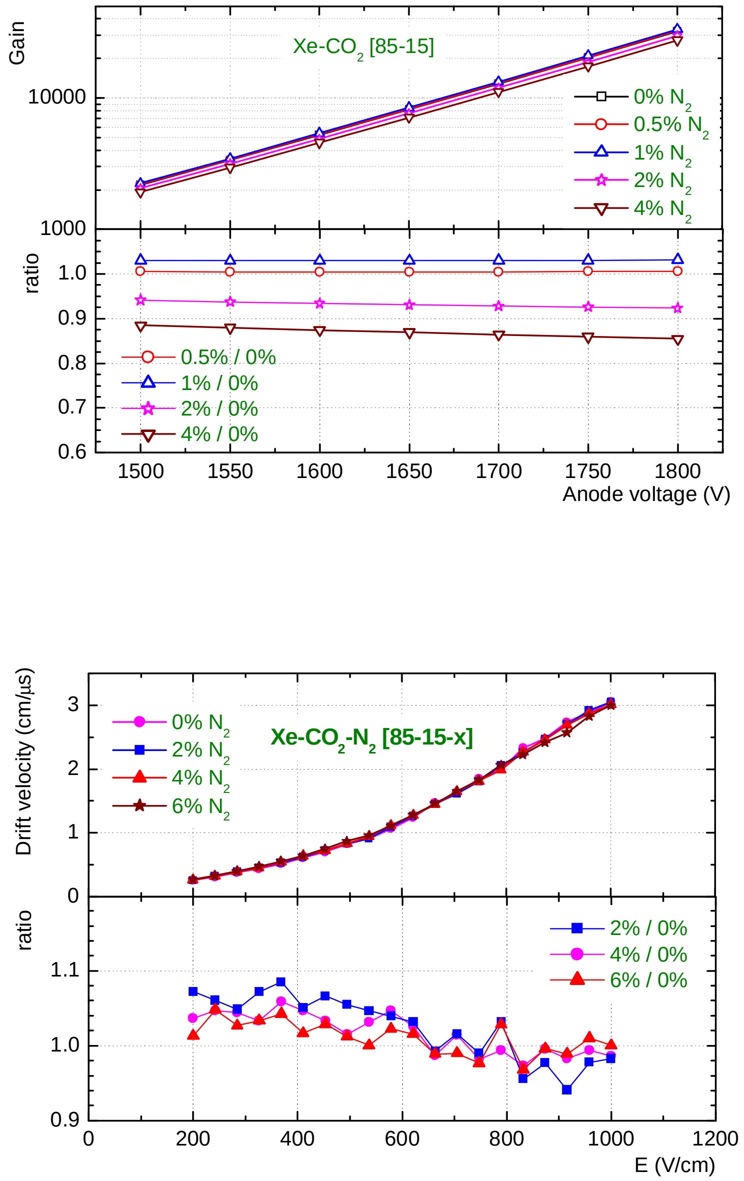

Fig. 4. Dependence of the gain (top) and drift velocity (bottom) as a function of the relevant potential for different concentrations of nitrogen in the Xe-CO$_2$ [85-15] mixture, in a TRD module.

The fraction of nitrogen contained in the mixture at any time t is given by

$$X_{N2}(t) = \frac{L_{N2} \cdot t}{R \cdot t + V} = \frac{0.8 \cdot L \cdot t}{R \cdot t + V},$$

where R is the regeneration flow –in this case, the fresh gas injection–, which is designed to be equal to the absolute leak rate L, and V is the volume of the system. We have assumed that the initial nitrogen concentration is zero. At infinite time, the fraction of nitrogen tends to its partial pressure in air, 80%. The evolution of this concentration along one ALICE year (8 months) is shown in Fig. 3 for a leak rate 5 times higher than the value quoted above. The nitrogen builds up almost linearly reaching, under the present assumptions, 4.5% by the end of the running period. Whether this value, and its variation along time, are tolerable or not is determined by the effect of the nitrogen contamination on the detector performance, in particular on the gas gain and the gas drift velocity. Simulations carried out with the GARFIELD [2], MAGBOLZT and IMONTE [3] packages suggest, as shown in Fig. 4, that a few percent of N_2 in the mixture can be tolerated. The deviations from nominal performance could be compensated for by readjusting the voltages in the drift and sense electrodes.

From this discussion about the leaks in the TRD system, we can conclude that the driving criterion to limit them as much as possible is indeed the cost of the lost xenon which must be replaced. The need for continuous separation of the diffused nitrogen from the mixture will be determined by the actual leak rate of the final system .

On the other hand, the filling –and emptying before maintenance periods– of the detectors with the operating gas mixture requires the separation of xenon from either nitrogen or carbon dioxide, thus ensuring that no xenon is vented out.
The baseline approach for this process is to first flush the detector with nitrogen, and then start the recirculation of the Xe-CO_2 mixture and at the same time to dump the outlet gas into a cryogenic recovery plant. In this plant, the gas is cooled down to temperatures approaching that of liquid nitrogen (LN_2), such as to freeze both the CO_2 and the xenon (see Table 1). The non-condensable gases, essentially nitrogen, stay in the gas phase and are pumped out at this point. The remaining solid species –the operating gas mixture free, to a certain extent, of impurities– are then warmed up and either put into circulation again or compressed into an appropriate tank for further use. This separation procedure has been previously utilized for other xenon-based detectors in experiments such as NOMAD [4] and ALEPH [5].

Table 1. Freezing and boiling points of the gases relevant for the cryogenic recovery.

Gas	Freezing point (^{o}C)	Boiling point (^{o}C)
N_2	-209.86	-195.8
Xe	-111.9	-108.1
CO_2	-78.4 (sublim.)	

In this note we report on experience and results obtained in the use of the very same recovery plants of these two experiments. The NOMAD system, more modest, was used under realistic conditions during a test beam with prototype detectors at the GSI SIS in June, 2002. The second plant, which belonged to the ALEPH electromagnetic calorimeter system, was tested at CERN in collaboration with the EP-TA1-GS group.

2 Experience with the NOMAD recovery plant

The NOMAD cryogenic system consists of a standard 50 l gas cylinder surrounded by insulating material and an aluminium cover. A copper pipe, in thermal contact with the bottle, runs around it in order to cool down the contents by flushing LN_2 through the pipe. Provisions for heating the bottom of the cylinder are also implemented.

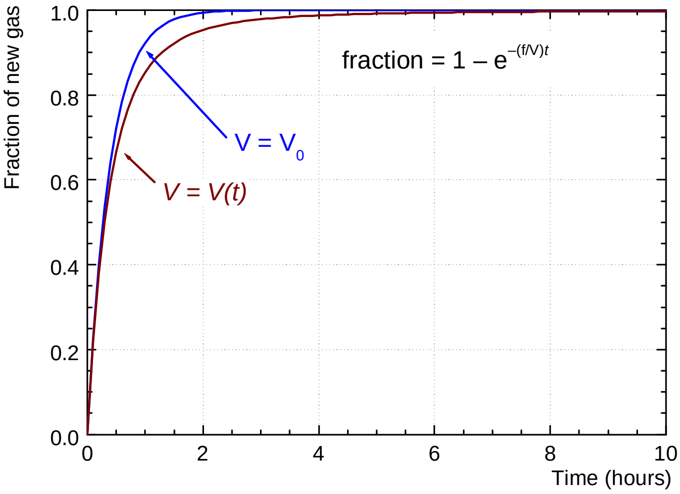

Fig. 5. Evolution with time of the filling of the detector with the proper gas mixture, for a 45 l system volume and an injection gas flow of 115 l/h. If the system was open (constant volume), the mixture would be established in about 2 hours. The gradual increase of the pressure in the volume behind the compressor (increasing volume with time), needed to dump the gas into the recovery bottle, leads to a slower filling, and results in 1 % N_2 in the mixture after 5 hours.

Prior to the operation of the detectors in the beam, the whole system is flushed with N_2 until the air (oxygen) content is negligible. At this point the operation mixture Xe-CO_2 is injected and the recirculation system is started. The outlet gas from the detectors is thereafter dumped into the recovery bottle, which was previously evacuated, with the use of the compressor. After 5-6 volume exchanges the operating mixture is established in the detectors, which are then ready to run. The time needed to achieve this operating point depends on the injection gas flow, but also on the fact that, the recovery cylinder being at room temperature, the pressure at the outlet of the compressor (*high pressure* volume) gradually increases. Fig. 5 shows how the filling of the system with the proper mixture evolves with time, both at atmospheric pressure and at a gradually increasing post-compressor pressure, for a given injection flow. At the end of this operation, about 20% of the 250 l of gas contained in the recovery cylinder is nitrogen. A similar operation could have been performed at the end of the beam period, although it was skipped because the volume of the detectors was fairly small (10 l).

Once *off-line*, the recovery procedure starts: a 50 l Dewar full of LN_2 is connected to the cooling inlet of the recovery bottle. Although possible to implement, there were in this occasion no measurement of the temperature of the bottle, and only an approximate reading of the pressure. However, samples of the gas contained in the bottle were analysed periodically in order to asses whether the xenon was totally frozen or not. The Dewar had to be refilled twice before basically all gas was nitrogen only. Fig. 6 shows several gas chromatograms of samples of the gas taken at different stages. The latest analysis revealed a concentration on xenon in the gas of 40 ppm. This amount, extrapolated to one volume of the full TRD system, represents only 1.1 l. Our gas chromatograph at GSI, in the configuration used here, is insensitive to CO_2.

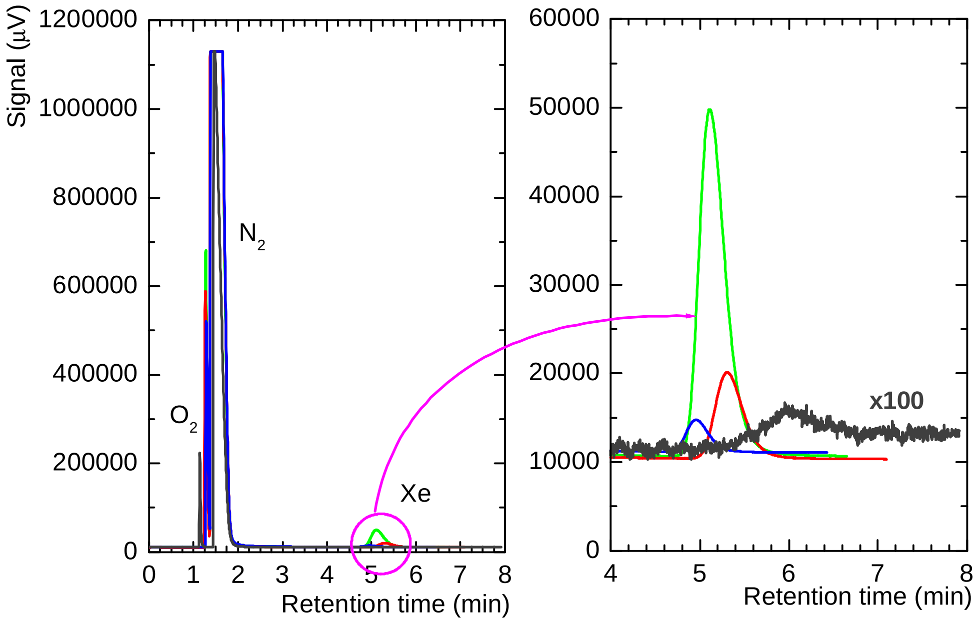

Fig. 6. Overlaid gas chromatograms of the gas contained in the NOMAD recovery bottle while being cooled down. The oxygen comes from the air contamination in the glass recipient that contains the sample. The peak of xenon, expanded in the right panel, decreases as it freezes. The manual injection and start-up of the gas analysis lead to time offsets.

One can then proceed to pump out the nitrogen while the LN_2 is still flushing. Naively, one expects that the longer one pumps the cleaner the mixture will be. After 45 minutes of pumping, the LN_2 flow was stopped and the recovery bottle was heated up. Two days later the remaining gas, now at room temperature, was analysed again. The resulting chromatogram, shown in Fig. 7, revealed that there was still 2% N_2 left in the Xe-CO_2 mixture. The entire cryogenic procedure was repeated once more. The remaining nitrogen, after proper subtraction of the air, was 1%.

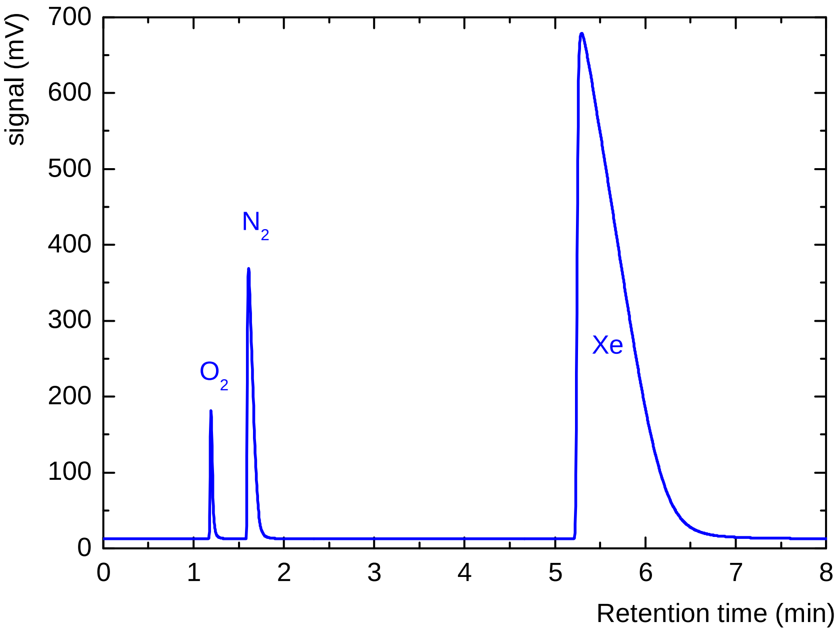

Fig. 7. Gas chromatogram of the regenerated, warm gas after nitrogen removal. The air comes from the sampling technique. The nitrogen left, after subtracting the air contribution through the oxygen peak, is 2%. A second regeneration cycle resulted in 1% N_2.

3 Experience with the ALEPH recovery plant

The ALEPH separation plant [6] is schematically shown in Fig. 8. It has been built by l'Air Liquide for the electromagnetic calorimeter of the ALEPH experiment, which ran with a Xe-CO_2 [80-20] mixture. It consists of a 100 l distillation column divided into three compartments, E01, E02 and C01, a sample volume R01, a LN_2 cooling circuit with temperature regulation, provisions for heating up the bottom of the column, and several pneumatic and manual valves for the operation of the plant.

The principle of operation has two steps, although only the first one has been carried out in the history of the plant. This procedure consists of first cooling the evacuated column volumes down to -170 ^{o}C with LN_2 prior to the injection of gas. Keeping the temperature in the volumes E01 and E02 regulated to the value indicated above, the injection of the gas to be regenerated takes place. By doing so, both the xenon and the CO_2 freeze as they enter the column through E01, whereas the nitrogen stays in the gas phase. Therefore, by ensuring enough flow of LN_2, one can in principle inject a large amount of gas in its volume, which is rated for 15 bars maximum pressure. Note that even large concentrations of nitrogen in the gas will result in a relatively low pressure in the column, since pressure scales with temperature. When all the gas to be regenerated has been injected, one closes the input valve and pumps out, through the top outlet, the non-condensable species, namely the nitrogen.

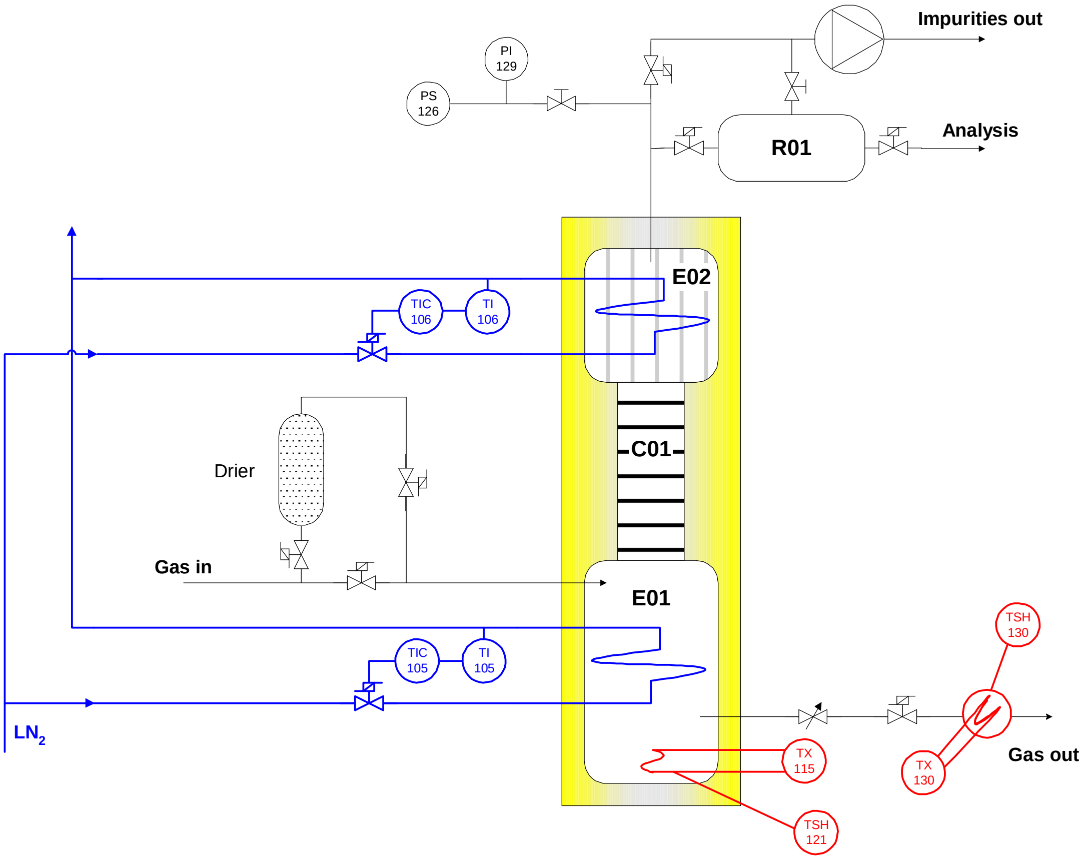

Fig. 8. Simplified diagram of the ALEPH recovery plant.

A second, foreseen regeneration step, which explains the complexity of the column, consists on a total reflux distillation of the gas: by heating the bottom volume (E01) and keeping the top one (E02) cold, the condensable species that evaporate at E01 condense into liquid in the C01 part of the column, which is an assembly of so-called Dixxon rings, whereas the remaining nitrogen –and possibly some gaseous xenon and CO_2 as well– stay at the top compartment. By analysing samples of gas extracted into the buffer R01, one can estimate how clean the regenerated gas is, and decide to either pump out the contents of R01 and continue the reflux distillation or stop the process.

In this note we report on the operation and results of the first step only. Both the pressure and the temperature of the column were known at all times, the latter being regulated by a control system. Samples of the gas were analysed in a gas chromatograph.

A mixture of Xe-CO2 [80-20] was in turn mixed with 59 % of N_2 (fig. 9, top panel) and injected into the pre-cooled (-170 ^{o}C) column at a flow of 500 l/h, while running the LN_2 cooling regulation. After accumulating about 1.8 m^3 total gas (volume referred to ambient T and p), the injection was stopped. The pressure in the column was at this point 2.7 bars. An analysis of the gas in these conditions showed that the majority of it was nitrogen, although traces of Xe (0.06 %) and CO_2 were still visible, as shown in the second chromatogram in Fig. 9. The pump was then run for about 10 min, until the pressure decreased to 50 mbar. The operating manual of the plant advised not to pump any further since due to the finite vapour pressure of the xenon at this temperature, some of this gas is anyway lost. The gas contained, at this stage (third panel in Fig. 9), 6.5 % Xe. The LN_2 cooling was then stopped, and the gas warmed up to ambient temperature. The pressure went up to about 7 bars, which means that there were about 700 l left in the column, in good agreement with the estimation of the injected Xe-CO_2. The subsequent gas analysis (Fig. 9, bottom) revealed that the nitrogen left in the mixture was 1.4 %, i.e. about 10 l. The pressure in the column when this sample was extracted was 20 mbar.

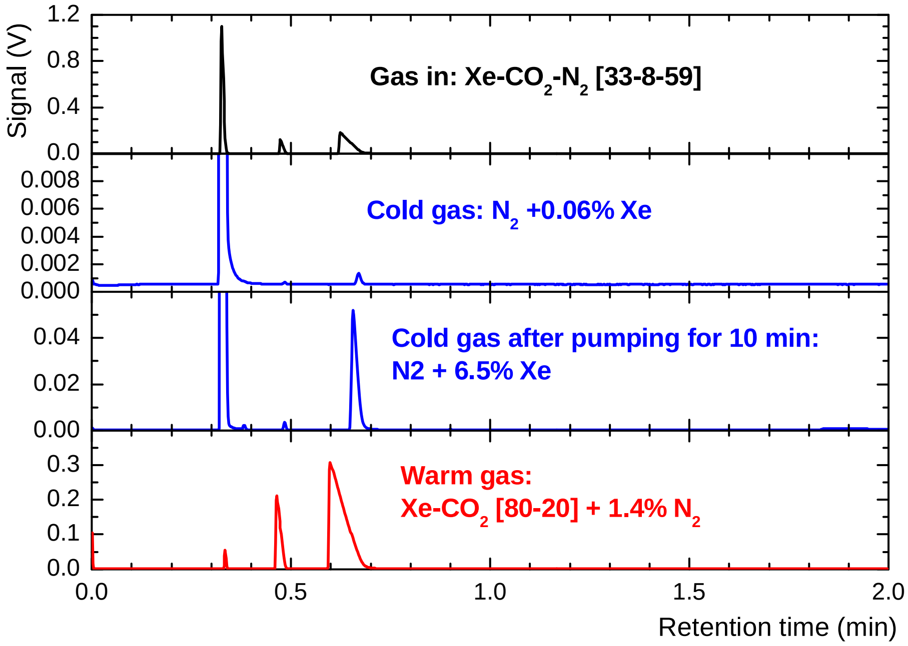

Fig. 9. Gas chromatograms taken at the different phases of the distillation process with the ALEPH recovery plant: the injected gas mixture (top panel), the analysis of the gaseous residuals in the cold column (second panel), the contents of the cold column after removing the nitrogen (third panel), and the resulting warm gas (bottom panel).

Let us try now to estimate the rate of xenon lost during the pumping period. The absolute flow, referred to ambient pressure, of the pump is

$$F(t) = P(t)\theta = (P_{Xe} + P_{N2}(t))\theta$$

where θ is the constant capacity of the pump per unit time (or the flow through it at atmospheric pressure) and P_{Xe}, P_{N2} are the partial pressures of Xe and N_2 in the column, respectively.

The partial pressure of xenon is also constant, and can be derived from the measured concentrations in the chromatograms before and after pumping:

$$P_{Xe} = [Xe] \cdot P = 0.0006 \times 2.7 \approx 0.065 \times 0.02 \approx 1.5 \text{ mbar}.$$

In order to estimate θ, we make use of the knowledge that a total of about 1050 l of nitrogen –disregarding the xenon– has been evacuated from the column in an exponential fashion, since this is how the pressure behaved. An exponential fit to two points, namely the pressure before and after pumping, as a function of time, gives us the pressure dependence of the flow. The total (nitrogen) volume extracted from the column is the integral of this flow:

$$\int F(t)dt = \int P(t)\vartheta dt \approx \vartheta \int P_{N2}(t)dt = \vartheta \int \mathrm{a} \cdot \exp\{-\mathrm{b}t\}dt = 1050\,\mathrm{l}$$

which yields θ = 160 l/min·bar. The rate of xenon loss is then $P_{Xe} \cdot \theta$ = 0.24 l/min. This modest rate, even if it might be affected by a large error, would allow one to purify even further the TRD mixture at a reasonable loss of xenon.

4 Discussion

We have shown that the separation of N_2 from the Xe-CO_2 mixture by cryogenic distillation performs satisfactorily: most of the nitrogen has been successfully removed from the mixture at a moderate loss of xenon. Nitrogen levels down to 1% have been achieved. These results have been accomplished with two different plants, one of them lacking basic diagnose functions. In addition, it has been shown that lower contamination levels are possible at linear extra loss of xenon, by just evacuating the nitrogen for a longer time while most of the rest of the gas is frozen.

However, the following disadvantages of this method should be considered:

- Long regeneration times. The time to fill the distillation column with the gas to be regenerated can reach up to a few days for large amounts, as will be needed in the real scenario. Furthermore, the warming-up time of the regenerated mixture to ambient temperature was two days even with active heating of the column, and would be longer for larger amounts.
- Repetition of the procedures. For the full TRD system (28 m^3) a minimum of 140 m^3 of gas must be regenerated in order to replace 5 volumes of the detector gas with the operating mixture (and back to nitrogen for the maintenance periods). On average, 20% of this volume would be nitrogen. This regeneration will most probably happen once per year. The ALPEH recovery plant could hold up to 25 m^3 of the average mixture at low temperatures only, which would imply 6 regeneration cycles and perhaps up to 6 weeks altogether.
- Safety. In the scenario described above, the flow of LN_2 must be guaranteed while the process takes place. Furthermore, the regenerated mixture should be evacuated as it evaporates in order not to overcome the maximum pressure of the (safety valve of) the column. Failure to do so, for example due to a late delivery of LN_2, would result in the loss of potentially large amounts of xenon.

For all this reasons the regeneration procedure *à la* ATLAS TRT will be looked at. This procedure consists on the separation of nitrogen from a Xe-CO_2 mixture with passive membranes. The separation membranes are bundles of capillary tubes of a porous material (kapton or carbon fibre) through which the nitrogen diffuses, but not the xenon or the carbon dioxide. Some R&D will be necessary in order to determine the efficiency and losses of this technique under our system conditions.

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