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RESULTS OF WIRE CHAMBER AGEING TESTS WITH CH4- AND DME-BASED GAS MIXTURES

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ABSTRACT

Results are presented of ageing tests performed on single-wire proportional counters under controlled conditions. The rate of the detector ageing with methane mixtures has been found to be independent of the anode and cathode materials used, and also of the purity of the gas. The rate of ageing for DME mixtures, on the other hand, appears to depend on the amount of gas used: it is small when the DME bottle is full, but increases as the cylinder empties. Addition of some water vapour to the Ar–DME mixture provided good lifetime, independently of the amount in the bottle. An explanation of this observation, based on assumptions on the fractional distillation of impurities, is provided.

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1. Introduction

The radiation hardness is an important condition for gaseous detectors expected to operate in high-luminosity collider experiments such as at the Large Hadron Collider (LHC). Detectors designed for this machine are expected to receive, during each year of operation, maximum doses of between 0.02 and 0.5 C/cm of sensing electrode, depending on the gas gain, geometry, and distance from the beam.

Accelerated ageing tests on single-wire proportional counters have been carried out in the laboratory of the CERN R&D Project RD-10 [1], by controlling a number of important parameters, such as the gas mixture and composition, the radiation dose imparted to the detector, the materials in contact with the gas, the gain of the counter under irradiation, and the ambient conditions. The goal of such studies is to find out which sets of conditions can ensure a lifetime of a few years for gaseous detectors operating at the LHC.

2. Experimental set up

A gas system (shown schematically in Fig. 1) that fulfils the requirements of being clean, gas tight, and free of organic materials has been constructed in order to provide suitable gas mixtures. It is made out of stainless-steel tubing, and contains mass flowmeters and oxisorb filters. A vessel that can be kept at a precise temperature is used for the addition of vapours to the gas mixture.

The gas flows into two identical single-wire proportional counters specifically designed, constructed, and cleaned for the ageing tests. The body of the counters is made of stainless-steel, and the material of the wire and the window can be chosen as desired. An aluminium (*Helicoflex*) joint provides gas tightness. The wire is supported in the centre of the counter, that has a square cross section, by glass insulators and crimped in copper inserts. The detector under test (see Fig. 1) can be heavily irradiated with 6.4 keV photons coming from a Fe target X-ray generator. A second counter (the monitor) is moderately irradiated with an attenuated photon flux and is used to allow corrections to the response of the test chamber due to changes in temperature, pressure, or particle flux. During each ageing test, the chambers are continuously irradiated and the current, pulse heights, and counting rates are recorded periodically.

The gas is analyzed, as it flows out of the test chamber, with a gas chromatograph (GC)[1] and either of the two associated detectors: a mass spectrometer

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[1] Hewlett Packard 5890.

(or mass selective device, MSD) and an electron capture device (ECD). The former has been optimized to detect and identify relatively light hydrocarbon molecules, such as those used in typical gas mixtures or which might be present in the gas as impurities. The ECD, instead, is extremely sensitive to electronegative substances such as freons, although the identification of each peak in the resulting chromatograms requires a specific calibration. A second, independent gas line (called the analysis line) is used for the calibration of the GC with gas samples.

3. Ageing studies with Ar–CH$_4$

A number of ageing tests were performed with Ar–CH$_4$-based mixtures. Methane is a rather popular choice for proportional counters, but earlier ageing studies [2–5] have indicated that it is a bad candidate for providing extended lifetimes.

Figure 2 shows the relative gain, i.e. the current drawn by the high voltage power supply when the detector is irradiated with a constant X-ray flux relative to the initial current, as a function of the collected charge per unit length of the anode wire (in this case, stainless steel 35 μm in diameter) of the test chamber. The gas mixture was 10% methane in argon (P10), both of CERN standard qualities (99.95% and 99.996% purity, respectively). The photon flux was 1.4×10^6 Hz/cm and the gas gain 2×10^4. Included in the figure are some pulse-height spectra, obtained by momentarily attenuating the X-ray flux.

The gain in this run drops by 10% after 10 mC/cm, when the pulse-height spectrum has already deteriorated. It should be noted that, owing to the shape of the collimator used to irradiate the chamber (a 1×7 mm^2 slit parallel to the wire), the charge collected at the wire surface is not uniformly distributed around it but concentrates in the forward and backward regions with respect to the beam direction. As a consequence, the deposits on the wire resulting from polymerization in the avalanche plasma will grow faster in these two regions of the wire surface, giving rise to the clear appearance of an extra, lower energy peak in the pulse-height spectrum of the detector, already at a relatively early stage in the ageing evolution.

Similar tests were performed on new counters where the gas purity, wire and/or cathode material, detector gain, and X-ray flux, were varied. Figure 3 shows, as an example, the ageing curve of a counter with a gold-plated wire, 40 μm thick, and graphite [6] cathodes mounted on the chamber walls and window. Measurements of current are made here with ≤ 1 nA accuracy, and the data are corrected by making use of the recorded gain in the monitor chamber.

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The rate of ageing has been expressed in previous works as $R = -(dG/dQ)/G_0$ [5]; we found it more convenient, however, to define a new quantity R', whose value appears not to depend on the particular dose accumulated in the corresponding ageing run, through the following phenomenological expression:

$$\frac{G}{G_0} = a \cdot e^{-R'\sqrt{Q}},$$

where G/G_0 is the gas gain, relative to the initial one, for a given accumulated charge Q (expressed in C/cm), and a is a constant of value ≈ 1. As seen in Fig. 3, R decreases as the charge increases, whereas R' tends to a constant value, and is therefore a more convenient parameter to characterize ageing. In Fig. 4 the expected gain drop is shown as a function of the total accumulated charge computed for some values of R'.

The R' values computed for different runs using argon–methane mixtures, but with varying operational conditions, have been plotted in Fig. 5 as a function of the initial current (basically the product of the flux and the gain of the counter).

Two conclusions can be inferred from the figure: the first one is that the independence of the rate of ageing of the materials of the electrodes used and, more importantly, of the purity of the methane, suggests that it is the methane itself that polymerizes in the avalanche plasma and produces the deposited layer on the wire surface.

This is confirmed by the results of surface analysis realized on aged wires[2], shown in Fig. 6. The deposits are mainly C_nH_m and aromatic hydrocarbons that cover the wire surface metal (in this case, stainless-steel). We have also observed that the addition of water somewhat decreases the rate of ageing, in accordance to the idea [3,5,7] that oxygen-containing molecules, among others, prevent to some extent the polymer chains from growing. Since the source of active monomers is presumably much more abundant than is the water, the improvement due to the addition of water is limited: a value of 0.2 of R', for example (see Fig. 5), corresponds to a 20% drop in gain after 1 C/cm of charge, not to speak of the complete degradation of the observed pulse-height spectra. It should also be mentioned that, when no vapour is added to the gas mixture, the amount of water and air in this particular gas system did not exceed 50 ppm in total.

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The second conclusion that can be derived from Fig. 5 is the tendency, already noticed elsewhere [2], to have higher values of R' for lower charge collection rates. Indeed, the points on the left most side of the figure correspond to rates comparable to those expected for minimum ionizing particles in the LHC (a few nanoamperes for a typical wire chamber geometry and gain). The results of the most accelerated tests (where 1 C/cm can be accumulated in a few weeks) are then optimistic but not by a large factor. In plasma chemistry, where some parameters such as gas pressure, gas flow, and energy available in the plasma are quite different from the case of wire chambers, it is known that the rate of the radiation-induced polymerization increases with the square root of the dose [8]. Therefore, the observed faster polymerization at lower charge rates could be a consequence of the particular phenomena that take place in the avalanche plasma; for example, the larger volume where the charge is distributed at higher rates owing to the distortion of the electric field in the counter.

4. Ageing studies with argon–dimethylether

A more promising gas mixture, Ar–DME [90–10], was flushed through the gas system to make ageing tests. The DME[3] is delivered in 150 litre cylinders, and its vapour pressure is 5.4 bars at room temperature. Starting the operation with a new cylinder of DME, the first result obtained is shown in Fig. 7, where a counter with a gain of 2×10^4 and all stainless-steel electrodes was irradiated during 6 weeks with a flux of 1.6×10^6 photons/s·cm, up to a total collected charge of 1.1 C/cm. Although the rate of ageing, R', is only 0.067 $(C/cm)^{-1/2}$ (or a 5% gain drop after 1 C/cm) the pulse-height of the chamber shows signs of deterioration at about 0.7 C/cm. A similar result was obtained with a second, full DME bottle and a gold-plated tungsten wire.

It was found, in further ageing tests with the same gas mixture, that the rate of ageing increases significantly as the DME in the cylinder is used, as can be seen in Fig. 8. This behaviour could be reproduced, starting the measurements again with a new bottle of DME.

As was already pointed out [9], there exists the possibility that certain impurities in the liquefied DME enter, by selective or Rayleigh distillation [10], into the gas phase of the cylinder (and flow out of it) with a rate that depends on the percentage of DME left in the bottle.

For those impurities whose boiling points are higher than that of DME, an expression to calculate their relative concentration in the gas flow as a function of the

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fraction of gas used is given in Ref. [9]. As an example, we show in Fig. 9 the results of this calculation applied to freon 11 (trichlorofluoromethane), two kinds of C_4 hydrocarbons, and water. After 25% of the DME has been used, the concentration of water in the gas drops by a factor of ≈ 2, whereas the abundance of the other impurities increases by 10 to 30%. This is compatible with an increasing rate of ageing as the DME is being used.

The analysis of the gas mixture made with the GC shows small traces of freon 12 (dichlorodifluoromethane) and freon 11, detected with the ECD (Fig. 10). With the use of calibration gases we estimate that the content of freon 12 in the mixture was of the order of 0.1 ppb, and the content of freon 11 less than 1 ppt. These values were measured when the cylinder contained 70% of the total amount of DME. The area of the freon 12 peak normalized to the area of the DME peak is plotted as a function of the percentage of the DME used in the bottle in Fig. 11, showing a decrease in the concentration of this pollutant as the DME is used. The abundance of freon 11 in the DME is, on the other hand, quite low. It has been previously demonstrated [9] that this type of freon, when added to ethane, does not affect ageing.

A few hydrocarbon molecules were detected and identified using the mass spectrometer, as shown in Fig. 12. Some of them correspond to chains of 3–4 carbons with at least one double bond, which facilitates polymerization. The equipment has not been calibrated for these species in terms of concentration. These pollutants, and/or the contents of water in the bottle, could cause the increasing rate of ageing observed in DME, although this point has not been proved at the present time.

A successful way to prevent our proportional counters from ageing in Ar–DME is the addition of a fraction of water vapour to the gas mixture. Figure 13 shows the ageing curve obtained with Ar–DME–H_2O [90–10–0.8] in a counter with a gold-plated wire up to an accumulated charge of 0.4 C/cm. The extrapolated gain drop after 1 C/cm, according to the fit of R', is 1%. This ageing test was carried out when more that one quarter of the DME in the bottle had been used (see Fig. 8).

7. Conclusions

We have performed ageing tests on proportional counters using CH_4 and DME gas mixtures with argon, under different conditions. Severe ageing has been found to occur in methane, and evidence was obtained that the hydrocarbon itself is responsible for this effect. Better results have been obtained with DME, although an increasing rate of ageing has been seen as the gas bottle is being used. This phenomenon, not yet well

understood, can be cured by the addition of a small quantity of water vapour in the gas mixture; this solution, however, might not be easily applicable in all types of detectors.

9. Acknowledgements

The authors would like to thank J. Kulasa, G. Million, M. Perä, Y. Pestov, M. Price, L. Ropelewski and L. Shekhtman for their interest and the ways they contributed to the work presented here.

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Figure captions

Fig. 1. Schematic of the RD-10 gas system, in its present configuration, showing the gas-analysis equipment and the chambers used in the ageing tests.

Fig. 2. Ageing curve for an argon–methane mixture measured in a proportional counter with a stainless steel-wire and body. The initial current in this run was 445 nA.

Fig. 3. Relative gain (G_r) and two different characterizing parameters, R and R', (see text), as a function of the accumulated charge per centimetre of wire for a chamber with graphite cathodes and gold-plated wire, irradiated with a photon rate of 6.4×10^4 Hz/cm (initial current = 170 nA). The gas filling is Ar–CH_4 [90–10].

Fig. 4. Computed gain drop as a function of the total accumulated charge for values of R' ranging from 0.01 to 10 $(C/cm)^{-1/2}$.

Fig. 5. Values of the rate of ageing, R', as a function of the initial current drawn in the counter for various tests carried out under different conditions, including a higher (99.995%) purity methane (triangle data point).

Fig. 6. TOF-SIMS surface analysis performed in two regions of a stainless-steel wire aged in P10.

Fig. 7. Ageing curve obtained with Ar–DME [90–10] on a counter with stainless-steel electrodes, when the DME bottle is full. The DME used for this run was 2% of the contents of the bottle.

Fig. 8. Rates of ageing measured with Ar–DME as a full DME bottle empties. The square data point corresponds to the addition of water to the gas mixture (see later in the text).

Fig. 9. Calculation of the relative enrichment in the gas phase of some species with boiling points higher than that of DME, as the gas flows out of the cylinder.

Fig. 10. Detail of an ECD chromatogram showing some traces of freons in DME.

Fig. 11. Evolution of the relative concentration of freon 12 in DME as the bottle is being used.

Fig. 12. Detail of the gas chromatogram of the DME showing the presence of impurities, some of them identified by the MSD as unsaturated hydrocarbons. Methyl chloride is also present in the gas.

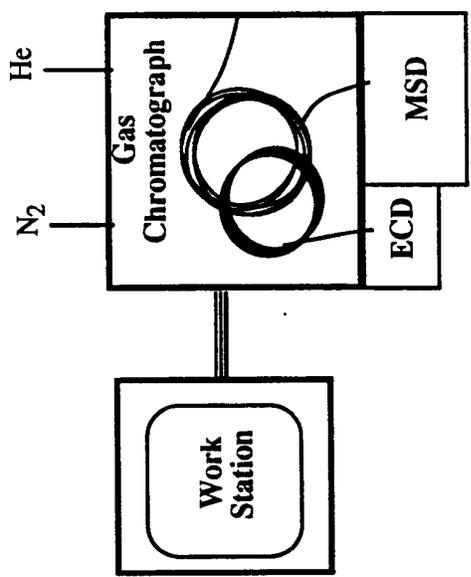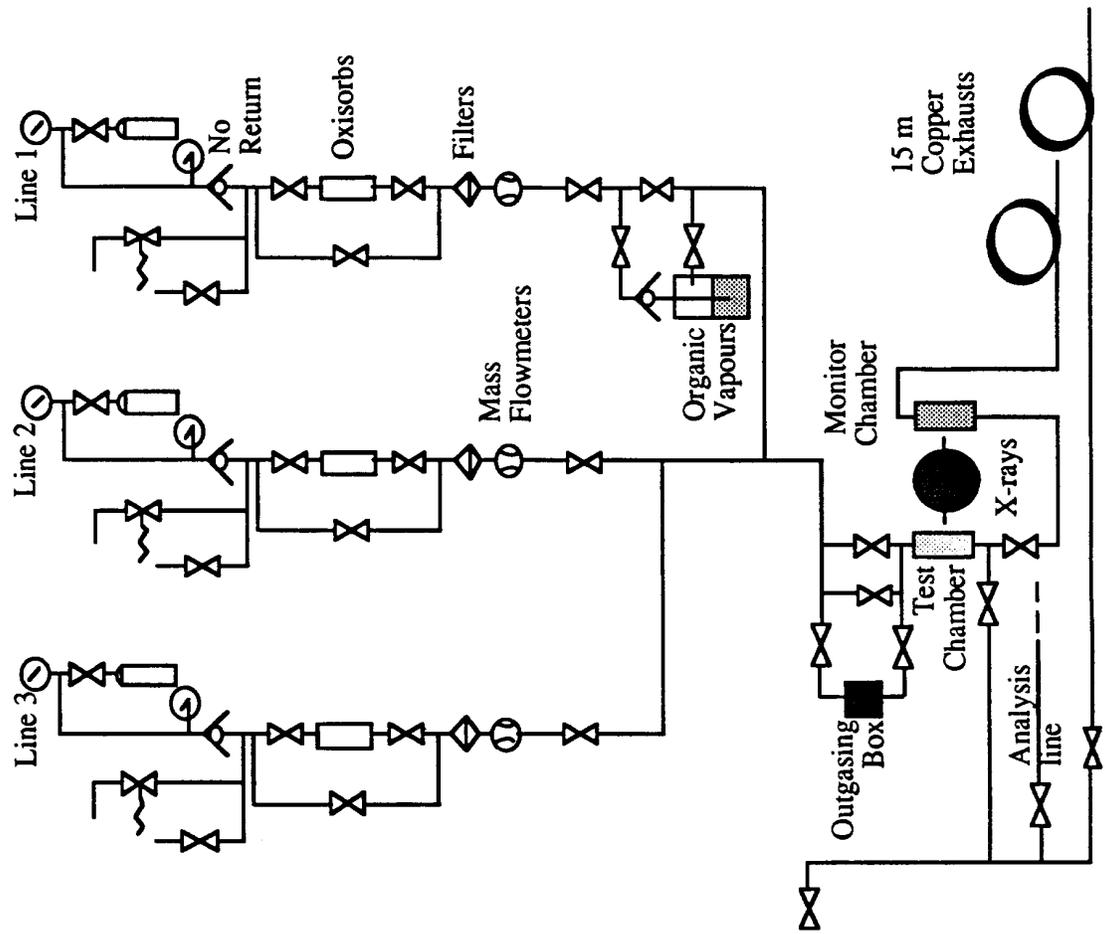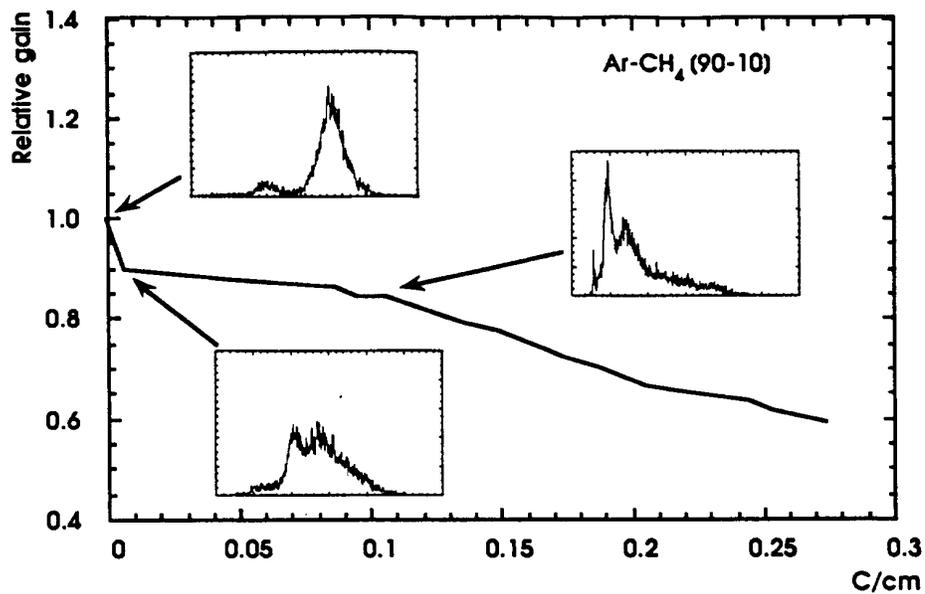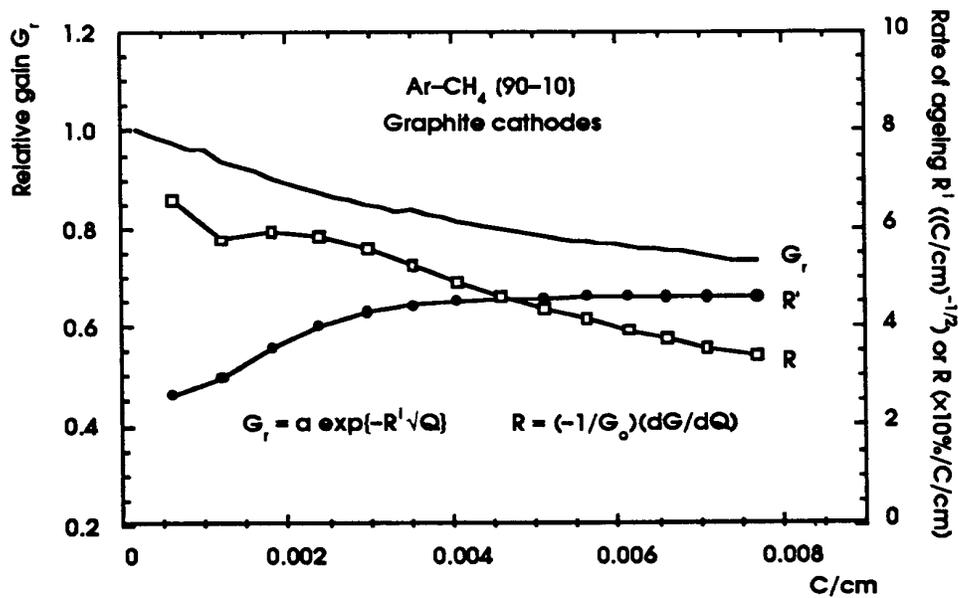Fig. 13. Ageing curve of Ar–DME–H_2O [90–10–0.8].

Fig. 1

Fig. 2

Fig. 3

Fig. 4

Fig. 5

Non-irradiated region

Irradiated region

Fig. 6

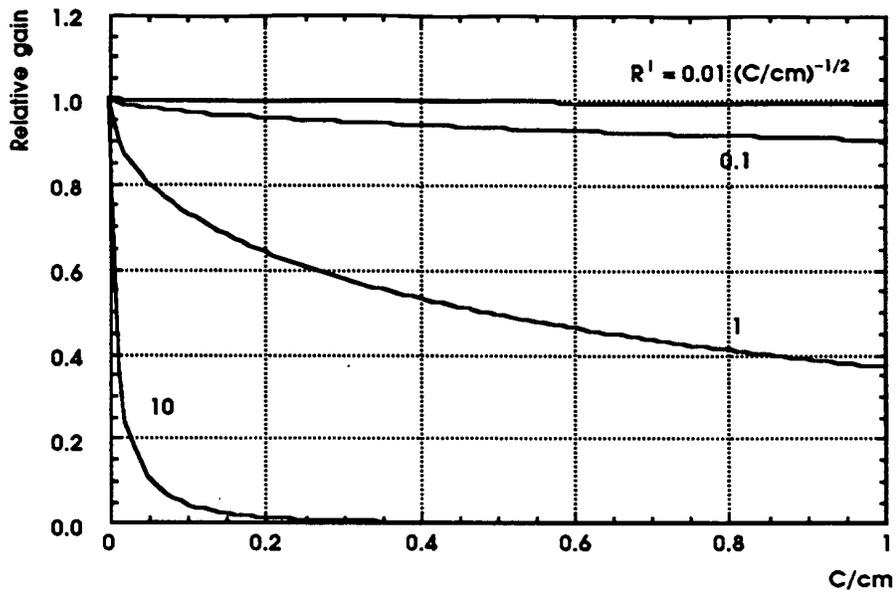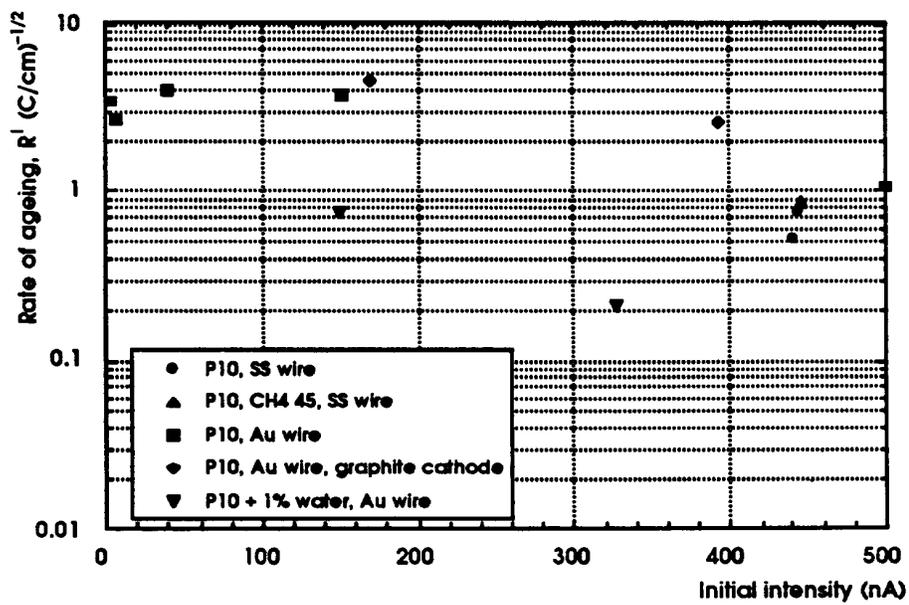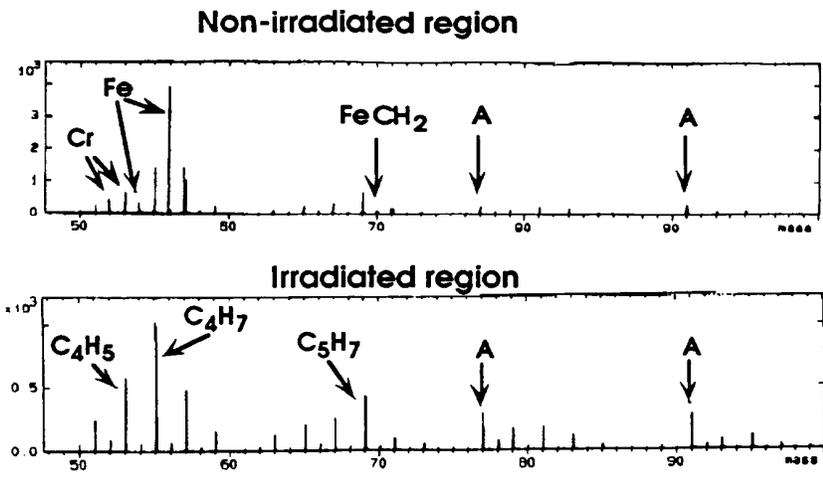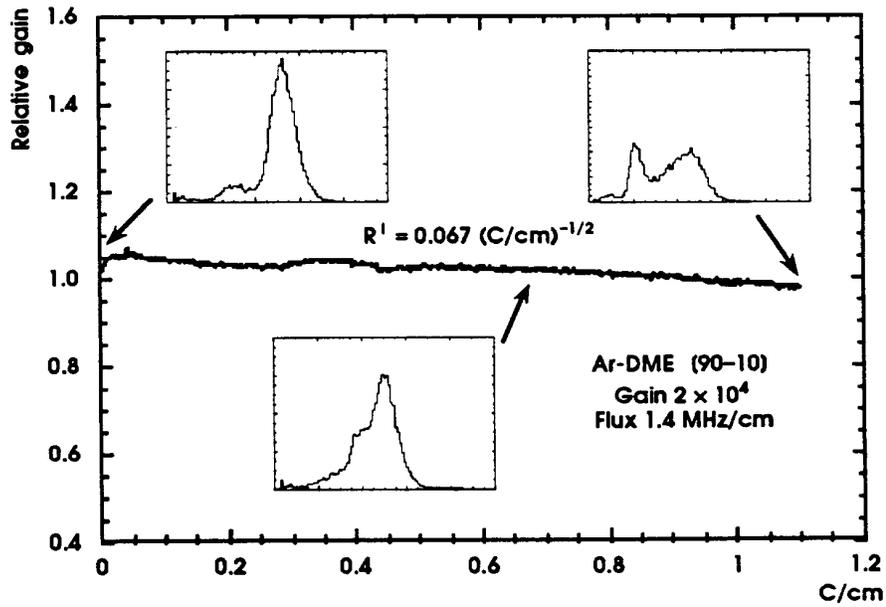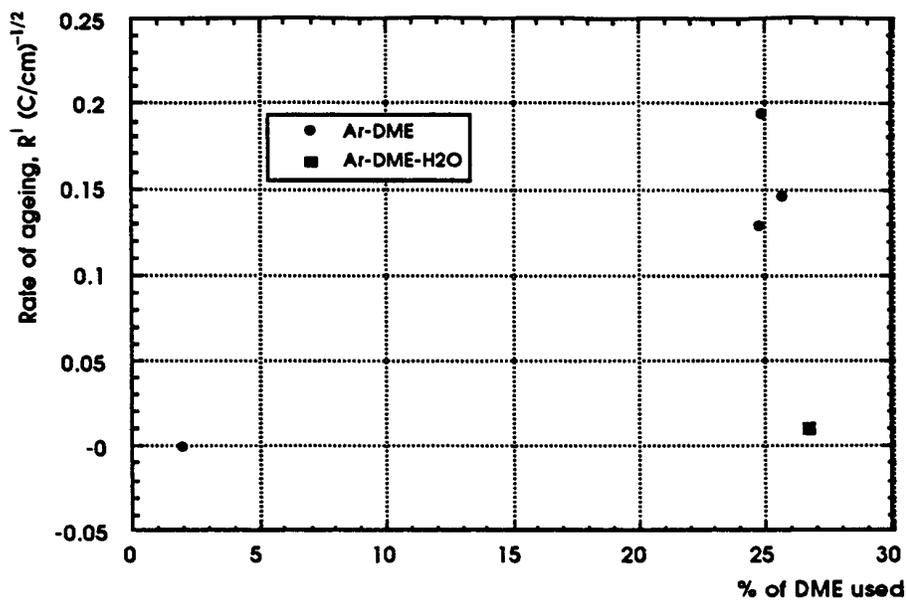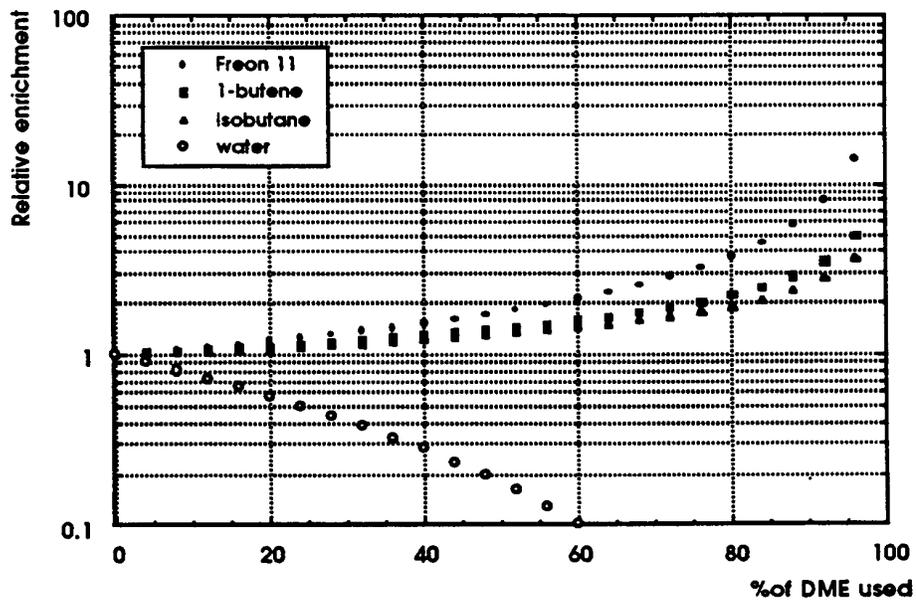$R^{\text{I}} = 0.067 \ (C/cm)^{-1/2}$

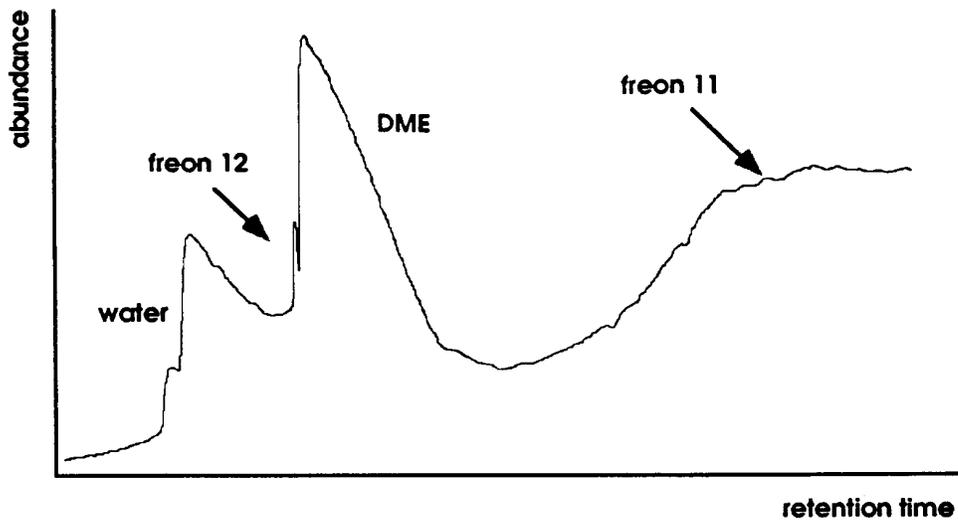Ar-DME (90–10)
Gain 2×10^4
Flux 1.4 MHz/cm

Fig. 7

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Fig. 8

Fig. 9

Fig. 10

Fig. 11

Fig. 12

Fig. 13

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