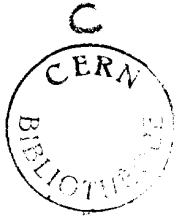


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RD-4 / STATUS REPORT
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STATUS REPORT ON THE RD-4 PROJECT.

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1) INTRODUCTION.

The goal of RD-4, as stated in the proposal DRDC-P6, is the study of using doped liquid argon (LAr) to improve the drift speed and incomplete compensation of pure LAr, while maintaining very good control of the systematic effects.

At the time of the proposal, it was known that the addition of molecular solutes such as hydrocarbons at the level of few tenths of a percent, increases the drift speed by at least a factor 2 above the value of pure LAr at high electric fields. On the other hand, photosensitive dopants at ppm concentrations increase the response to densely ionising particles by converting the UV scintillation light of the argon into detectable charge. It was hoped that a sufficient amount of photosensitive dopant could be added to LAr to improve on both drift speed and compensation simultaneously.

An extensive account of those studies, conducted by RD-4, was presented at the Stockholm Conference on Noble Liquid Detectors, 1991, and has been accepted for publication in NIM. A copy of the CERN preprint containing the main results on this subject is attached. The information contained in this preprint is only partially reproduced in this report.

In this status report we describe the construction of the detector containing the radioactive sources and laser ionization chambers used for the measurements. A vacuum tank and a special cryostat, a specific cryogenic cell dedicated to the infrared absorption spectroscopy of some dopants had to be designed and constructed. These set-ups have been used to measure the response characteristics of LAr doped with ethylene, the most soluble photosensitive dopant known to us, in concentrations of 12.5, 25, 50, 100, 200 and 500 ppm, and 0.1, 0.2 and 0.5%. A good understanding of the behaviour of ethylene-doped LAr has been achieved. This basic behaviour is common to all known dopants.

During the past year, in collaboration with the accordion (RD-3) project, a test of mixing ethylene at a concentration of 0.2% vol was performed in the Helios cryostat to observe any change in the accordion response. This test is also discussed in the present report as it highlights the main difficulties associated with obtaining a precise, correct and uniform

concentration of dopants in large systems filled with LAr, e.g. a LHC calorimeter.

A principal concern at LHC is the destructive effect of radiation on the dopants mixed with LAr. A consequence of the destruction of the dopant molecules is a variation of characteristic parameters such as drift speed and charge. These variations are expected to be highly non-uniform. The question of ensuring in such a context the systematic control of the absolute energy scale to a precision better than a 1% is also discussed.

Radiation tests have not yet been performed by RD-4. A new cryogenic container, dedicated to this purpose, has been built and should be exposed to the CONSERVATOME Co⁶⁰ source at the beginning of 1992.

2) EXPERIMENTAL SET-UP.

Details of the experimental set-up can be found in the attached preprint CERN-PPE/91-171. The multiple-cell detector represents a large step in size (it contains 3.3 liters of LAr instead of cc sizes) and improvement of the cooling system compared to previous cells used for LAr+dopant experiments and permits an extensive measurement of the following parameters:

- the drift speed, α and β charges, and liquid lifetime are measured simultaneously in the same volume.
- the drift speed and lifetime are measured precisely with a UV laser.
- all cathode signals are capacitively decoupled from HV and read out, allowing the measurement of liquid lifetime, essential for correction of the anode signals as well as for the determination of the LAr luminescence attenuation length. The measurement of these parameters proved to be very important for understanding the processes involved.

An independent cryogenic cell was designed for IR absorption measurements. The delicate question of mounting a total of four KRS-5 IR windows, two in the cold detector walls, to withstand vacuum and internal pressure, was resolved using a much simpler technical solution compared to all previous cells published in the literature.

A Bruker IFS-66 FT-IR spectrometer was purchased by the CERN-MT division and loaned to us for the duration of the RD-4 project. The commissioning, both hardware and software, of such a delicate instrument went well. The spectrometer, with its external IR beam passing through the cryogenic cell and MCT detector, is operational in the RD-4 laboratory. It is presently being used to determine the absorption coefficients of hydrocarbons and impurities dissolved in LAr.

At the beginning of the project, data were collected using a 100 MHz digital scope for the drift speed measurements and a multichannel analyser for the charge measurements. This Data Acquisition System (DAS) has been considerably upgraded and automatized, thanks to the RD-12 collaboration which made it possible and successful. The drift time measurements using the laser pulses are now analysed by a 100 MHz Lecroy FADC read out by a Macintosh II fX via a CAMAC-CC module. The charges from sources are digitized by an ADC and read out by the same manner. The RD-12 collaboration wrote a CAMAC driver embedded in the LABVIEW program, and a full software system, including readout, control, display of results, histograms and storage of data on diskettes for later analysis using Kaleidagraph. The system is now fully operational and can be used in any test set-up, or in other R&D collaborations.

Fig.1 shows a view of the cryogenic cell built for radiation studies. The tooling necessary to transfer the irradiated liquid from the cell to the detector is being finalized, so that the experiment can be performed at the beginning of 1992.

In conclusion, concerning the RD-4 experimental set-up, all parts of the proposed apparatus have been built and are operational.

3) RESULTS ON DOPING LAr WITH ETHYLENE.

Ethylene is to our knowledge the only hydrocarbon which is both photosensitive (Q_{eff} around 20%) and soluble in LAr to the level of 1%. All other photosensitive substances are much less soluble (propadiene, propylene,

TMG), whence no substantial increase of the drift velocity can be expected from them. It is our understanding that the following results on ethylene-doped LAr represent a general behaviour common to all unsaturated hydrocarbons. The only difference between one substance and another would be a different quantum efficiency (Q_{eff}) or differences in saturation properties. We do not think that it would change the conclusions of this report.

a) Drift velocity.

At 10 kV/cm, an increase of a factor 2 in drift velocity is observed for ethylene at concentrations $\geq 0.2\%$ vol (see figs. 8 and 9, CERN-PPE/91-171).

b) α and β charges.

Essentially no dependence in the response to β particles from a ^{207}Bi source is observed as a function of ethylene concentration. Such constant response is then to be expected for any minimum ionizing particle (MIP). This behaviour is explained by the fact that the photosensitive substance converts the LAr recombination photons into electrons. The yield of the recombination photons relative to the produced charge is particularly important in the case of a large local ionization density (as created by α 's) but not in the case of the weak ionisation density created by MIP's.

Figs 2a, 2b and 3 summarize our final results on the response to α particles from a ^{241}Am source. Three regions are distinguished :

- C_2H_4 concentration ≤ 25 ppm
the collected charge rises with concentration as more and more UV's are converted into electrons.
- C_2H_4 concentration between 25 and 50 ppm
a plateau region, wherein all UV's are converted in electrons. The exact localization of this plateau is a function of the Q_{eff} of the dopant.
- C_2H_4 concentration > 100 ppm
at large concentrations (the region of increase of the drift velocity), the collected charge decreases well below pure LAr level.

If one supposes that saturation of collected charge in liquids follows a dependance similar to Birk's law for scintillators, the parameter k_{B} can be

computed from the α and β dE/dx values. Fig. 4 represents our results for k_B as a function of electric field and for different C_2H_4 concentrations.

In conclusion, it is not possible to obtain simultaneously: an increased drift velocity and improved response to highly ionizing particles such as α 's. A low dopant concentration (few tens of ppm) is suitable to reach better compensation in a hadron calorimeter with no speed increase whereas a large dopant concentration (few tenths of a percent) can be envisaged for fast electromagnetic calorimeters.

c) Attenuation length of UV photons in LAr.

The previously observed charge increase for highly ionizing particles can be obtained in the small gaps of a LAr calorimeter (for example 1.4 mm as in the accordion), only if the attenuation length of the LAr luminescence photons (λ_{UV}) is much shorter than the gap itself. This attenuation length is a function of the C_2H_4 concentration.

Fig.5 shows a preliminary evaluation of λ_{UV} for various C_2H_4 concentrations. At concentrations > 200 ppm C_2H_4 , λ_{UV} becomes very small; however, in the region where the biggest gain of collected charge due to the photosensitive dopant is expected (around 25 ppm), λ_{UV} is larger than the value of the accordion gap (1.4 mm).

We therefore conclude that the use of small concentrations of photosensitive dopants to improve compensation is not really attractive in LAr calorimeters with small gaps. The principal advantage is the drift speed increase at larger concentrations, such as 0.2% vol C_2H_4 , accepting however a degradation of the collected charge.

4) IMPLICATIONS ON THE STABILITY OF AN LHC CALORIMETER.

The main concern of the monitoring is to ensure the control of the systematics at a level better than 1% on the absolute energy measurement. In the case of a calorimeter filled with pure LAr, the only variable to follow is the change of the lifetime of the liquid due to the presence of O_2 ; usually an

α -source ionization chamber is adequate. In case of doped liquid argon, additional variables have to be monitored: electron drift speed, dE/dx saturation properties and liquid lifetime.

a) Monitoring of the electron drift speed.

The $V_{\text{drift}}(\text{electrons})$ is not saturated in the case of doped LAr. At $E=10$ kV/cm, for a drift gap of 1.4 mm and using (20 ns) shaping, a 1% difference in the charge sampled can result from a variation of ≤ 2 ns in the total drift time of electrons through the gap. Measuring the electron drift velocity to such a precision would be required, which is very difficult.

In collaboration with RD-12, we developed an on-line program able to read the response of a double-gridded ionization chamber to UV laser pulses sent to the cathode. The drift time separating the two grids could be obtained on-line. Figs. 6a and b show single laser pulses and Figs 7a and 7b the histograms of several drift times obtained using a very simple algorithm. One notices that for $E \geq 3.5$ kV/cm the FWHM of the drift time distributions is 80 ns, corresponding to $\sigma=34$ ns. It is then sufficient to accumulate ≥ 300 laser pulses on the cathode to reach a precision of ≤ 2 ns on the mean of the distribution. The monitoring requirement on drift speed can therefore be achieved using a UV laser.

In a large calorimeter, distribution of the necessary UV fibers to the different parts and dedicated feedthroughs through the vacuum tank and detector walls would be required.

For that reason we investigated whether a comparable precision could be obtained using an α -source instead of a laser. Figs. 8a and b show selected single α pulses in a double-gridded ionization chamber similar to that used with the laser. A measurement of the drift time could probably be obtained by a large accumulation of many such events. To reach a precision of below 2 ns on the mean of the distribution could require in addition an on-line rejection of noisy pulses. The method of replacing laser pulses with a source is still under investigations.

b) Monitoring of dE/dx saturation properties.

As mentioned previously, the response of a doped LAr calorimeter to densely and lightly ionising particles is different. It is therefore necessary to

follow the behaviour of α and β sources simultaneously. A precision of 1% on such source signals is certainly within possible goals.

If the concentration of dopant changes, the difficulty would be to cope with the variation of the compensation properties of the calorimeter. The exact correction of the measured energy would have to rely on extensive test beam measurements and on M.C. simulation.

c) Monitoring of the liquid lifetime.

A correct measurement of the liquid lifetime cannot be extracted from a source measurement due to the contribution of the UV luminescence of LAr. Effectively, the UV photons from LAr extract photoelectrons from the dopant inside the drift gap. The drift distance from the point at which these electrons are extracted to the anode is then unknown and consequently no correct liquid lifetime can be evaluated. The only way to apply a lifetime correction to the data (if needed) is to get it from a laser induced charge measurement.

In addition, the charge collected from an α source is subject to the undistinguishable variations of the dopant concentration and liquid lifetime.

d) Remarks on radiation effects.

An additional, fundamental difficulty for LHC operation would be the secondary effects of radiation on the dopant. In an environment with no radiation, the concentration of dopant is expected to stay constant, as are all monitoring parameters. On the contrary, in a high radiation environment, one expects that all parameters would vary due to the destruction of the dopant. The variation of such parameters would certainly be very inhomogeneous, as a function of rapidity and radius from the interaction point, as the dose varies from the front to the back of a calorimeter as well as from the central barrel to the endcaps.

5) TEST WITH THE ACCORDION, RD-3 PROJECT.

During the summer of 1991 a test was performed in collaboration with the RD-3 project (the accordion), in which 0.2% vol of ethylene was introduced into the cryostat after it was already filled with LAr. Mixing was

achieved by simultaneously heating the bottom of the tank and condensing at the top, generating bubbling during approximately 15 hours. In this test, results were collected simultaneously from an α source ionization chamber, the Helios hadron calorimeter and the accordion.

At $E=10$ kV/cm, the ^{241}Am α signal was 3.04 fC which corresponds well to the value of around 3 fC obtained in the RD-4 test detector for a concentration of 0.2% vol of ethylene. We conclude that the correct amount of ethylene was introduced into the Helios cryostat.

At $E=4$ kV/cm, the electron total drift time measured in the hadron calorimeter was around 510 ns. That agrees well with the value of the drift velocity measured at this particular field with our test set-up in the case of a 0.2% C_2H_4 concentration.

However, two indications lead us to think that the correct mixing of C_2H_4 with LAr was not achieved inside the accordion. Fig.9 shows the drift velocity obtained with the accordion, compared with the results of the test set-up that we estimated from many accumulated pulses on a digital scope. On the other hand, the charge measured at the sampling time of 20 ns shows an increase by a factor of 1.3 compare to pure LAr. In case of a concentration of 0.2% vol C_2H_4 , a factor of 1.8 over pure LAr is expected, which was not observed. Both observations lead us to conclude that correct mixing was not achieved inside the accordion and that the actual average concentration inside was more likely around 500 ppm instead of 0.2%. This consideration indicates the difficulty of achieving uniform mixing of dopant and LAr in a large LHC calorimeter, especially one with a closed geometry, such as the accordion.

Introducing the dopant after the filling of the calorimeter with LAr looks unattractive due to the very slow diffusion of the solute inside and to the uncertainty in reaching an homogeneous mixture. An other possibility would be the filling of the calorimeter with the already prepared mixture, although it would not be easy to realize in practice. The main difficulties are both the large difference between the vapor pressures of ethylene and argon at LAr temperature, and the large difference in their melting points.

6) IR SPECTROSCOPY.

The goal of conducting studies on IR absorption spectroscopy of doped LAr is two-fold: to detect impurities such as CO₂, freons or others, and to measure the correct concentration of the dopant inside the LAr. Both require the knowledge of the corresponding IR absorption coefficients which link the observed IR absorbances to the compound concentrations. We consequently developed a program to measure these coefficients for ethylene and allene mixed with LAr.

For these measurements, we used the Bruker IFS-66 FT-IR spectrometer, property of the MT-division, to measure these coefficients in the gaseous state. This is the subject of the CERN-PPE/91-204 preprint, attached to this report.

The studies on cryogenic liquids are continuing. Fig.10 shows an absorption spectrum of pure LAr in our home-made cell, revealing the definite presence of CO₂. Figs. 11a and b show absorption spectra for ethylene and allene. The very sharp absorption lines, for ethylene and allene, characteristic of cryogenic mixtures, peak above a very smooth background. The method works very well and can be applied to the determination of the absorption coefficients of any dopant or foreseen impurity. In turn, the knowledge of these constants will allow the computation of their exact concentrations in solutions.

7) RADIATION DAMAGE.

The general question of radiation damage is discussed in CERN-PPE/91-171. The main problems associated with the damaging effects of the radiation on dopants are :

- the difficulty of ensuring the monitoring of the systematics at a level of 1% precision, as mentioned before.
- the inhomogeneity of the damage and the fact that monitoring stations cannot be placed everywhere.
- polymerization leaving residues in the calorimeter.

The destruction of the dopant molecules can be estimated from Holroyd [IEEE Trans.on Nucl.Science 37,(1990),513] to be 6×10^{-2} ppm of C_2H_4 destroyed/Gy. A dose of 1 Mrad (10 kGys) would then cause the destruction of 600 ppm of C_2H_4 . This dose value has been computed at cascade maximum in an Lead-LAr EM calorimeter for $\eta=2.5-3.0$. Depending on the radiation level, it is probable that in the long run, large molecules, such as vinyl, will be formed from the radiolysis products of ethylene, which cannot be removed from the calorimeter. The main radiolysis products of ethylene (such as acetylene, n-butane, ethane) are not photosensitive and do not contribute to any conversion of UV light into electrons. The dependence of electron drift velocity on the concentrations of most of the radiolysis products is unknown, so any realistic prediction of the change of the drift velocity under the action of the radiation is hazardous.

It is part of the RD-4 program to perform a radiation test in order to evaluate the consequences of radiation damage on the characteristic parameters of a calorimeter, such as speed and charge collection. High dose irradiation of a cryogenic system is by no means obvious, so we propose a modest program. Only in the event that the results are encouraging, or that the prospects of using doped LAr at LHC are highly attractive because of other considerations, would we consider performing further and more complete tests.

We propose to irradiate the specific cell shown in Fig.1, filled with doped LAr at CERN, with 1 MRad of gamma-rays from a ^{60}Co source (CONSERVATOME). This will involve no *in situ* monitoring, except for the temperature, and will minimize the apparatus which needs to be transported to CONSERVATOME, where no LN_2 is available. The cell, still cold, will be brought back to CERN after irradiation, requiring approximately one full day at maximal dose, and the liquid will be transferred to the RD-4 test detector for measurement. This minimal scheme involves the transportation of the LN_2 and the cold cell, piping and rental of the source.

We intend to perform this test during the first part of 92. Any more elaborate test with on-line monitoring of the relevant parameters (with an electric field applied during radiation), would require an infrastructure substantially beyond that offered at industrial facilities like the CONSERVATOME and should be considered as a new project in itself.

8) CONCLUSION.

The RD-4 project has achieved the following goals :

- construction of a test detector with laser, α and β source ionization chambers; a vacuum tank, cryostat and purification system for Ar and dopants was also built and commissioned.
- development of a DAS system in collaboration with RD-12, and development of a full readout and analysis chain on the Macintosh based on CAMAC and the Labview program.
- construction of a cryogenic cell for the IR absorption spectroscopy on cold liquids.
- successful commissioning of the FT-IR spectrometer.
- completion of the studies on LAr doped with ethylene. The results are thought to be general and valid for any known dopant.
- large scale test in collaboration with RD-3 in doping LAr with 0.2% vol ethylene in the Helios cryostat containing the accordion prototype.
- measurement of IR absorption spectra in LAr of impurities and dopants such as ethylene and allene, and determination of their IR absorption coefficients.

A simple test on radiation damage remains to be done on LAr doped with ethylene. This test should take the first part of 1992. Any more elaborate radiation tests should be envisaged only in the event that the use of dopants for LHC turns out to be very essential because of other considerations.

Several new results have been obtained from the measurements done on LAr doped with ethylene in various concentrations:

- the drift velocity of electrons is multiplied by at least a factor of two for ethylene concentrations $\geq 0.2\%$ vol.
- the amount of charge collected from electrons from ^{207}Bi is mostly unaffected by the dopant. Charge response to MIP's using fast shaping thus depends essentially on the drift velocity of the free electrons in the gap.
- the charge response to α particles from a ^{241}Am source shows a large increase in charge collection at very low concentrations due to the

conversion of the UV luminescence of LAr into electrons by the ethylene. A maximum increase is reached around 25 ppm. However, at large concentrations the signal falls below the response of pure LAr, resulting in an increase in the saturation properties of the mixture.

- in consequence, it is not possible, even with a photosensitive substance such as ethylene, to get at the same time a significant increase of the drift velocity and a better response to highly ionising particles such as α 's, resulting in a better compensation.
- the attenuation length of the UV luminescence from LAr is large. In consequence one will not profit fully from the charge increase given by the photosensitive substance in a calorimeter with small gaps, such as the accordion.

The two last points reduce the attractiveness of using dopants. The principal remaining advantage is the increase in the drift speed at ethylene concentrations $\geq 0.2\%$ for example.

Our studies indicate that the biggest difficulty in monitoring systematics is the precise measurement of the drift velocity. At this moment, the necessary precision can only be guaranteed using a UV laser and optical-fiber system.

A large scale test on the use of dopants in a LAr calorimeter was done in collaboration with the RD-3 (accordion) group. The results indicate that it appears difficult to re-establish a known concentration of dopant after radiation-induced loss by adding gas-phase dopant to the liquid already inside the calorimeter.

In our view the principal difficulty at LHC arises from the effects of radiation destroying the dopant and changing the parameters essential to a proper calibration and the reliability of physics data. RD-4 intends to perform a modest test on radiation damage. If the results are not encouraging, we would consider the use of dopants for LHC calorimetry of only marginal benefit and would not advocate to consider doped LAr calorimeters.

RD-4 PUBLICATIONS AND INTERNAL REPORTS.

P.Cennini et al., CERN DRDC-P6 proposal, 1990.

C.W.Fabjan, V.Vuillemin, Proceedings of the Miniworkshop on Low-Counting and Space-Based Use of liquid Ar and LXe Detectors, Waseda University, Japan (April 5-6, 1990).

F.Vendrell and V.Vuillemin, Technical Note CERN RD-4 TN 90/01.

D.Lacarrere, Technical Note CERN MT-SM/DL/90-095/cl.

V.Vuillemin, Technical Note CERN RD-4 TN 91/01.

P.Cennini et al., Conference on Liquid Noble Gas Detectors and their Applications, Stockholm, Sweden, August 21-23, 1991, presented by V.Vuillemin/CERN-PPE.

V.Vuillemin et al., CERN-PPE/91-171 (to be published in NIM).

M.Moulson et al., CERN-PPE/91-204.

RESOURCES REQUESTED FOR 1992. **kSFr.**
[RD-4 PROJECT]

Industrial support. 20
 [This sum corresponds to the rental of a full-dedicated week
 of the CONSERVATOME Co⁶⁰ source.]

Travel related to the project. 5

Purchases for the radiation damage cell. 5

— Large pathlength gas cell for precise IR absorption measurements. 25
 [For IR analysis of decay products after irradiation.]

Total [project budget] 55

RESOURCES REQUESTED FOR 1992.
[RD-4 EXPLOITATION]

— Running expenditures, travel, electronic pool. 21

Multichannel analyser with CAMAC interface. 10

Gases, liquid Argon, tools. 9

Total [exploitation budget] 40

TOTAL **95**

LIST OF FIGURES.

- Fig.1 Cryogenic container for radiation damage studies.
- Fig.2 Charge collected on the cathode (a) and on the anode (b) from an ^{241}Am α source in LAr doped with ethylene as a function of electric field for several concentrations. In both figures superimposed curves are simple power-law fit lines.
- Fig.3 Charge from an ^{241}Am α source in LAr doped with ethylene as a function of dissolved ethylene concentration.
- Fig.4 Values of Birk's parameter obtained from α and β collected charges in LAr doped with ethylene as a function of electric field for several concentrations.
- Fig.5 Values of the attenuation length of the LAr UV luminescence as a function of ethylene concentration.
- Fig.6 a,b Direct response of a double-gridded ionization chamber to a single UV laser pulse extracting free electrons into the gap from the cathode plane.
- Fig.7 a,b Histograms of drift time obtained from 500 UV laser pulses hitting the cathode. The algorithm used to compute the drift time is very preliminary and simple.
[1 channel = 10 ns]
- Fig.8 a,b Direct response of a double-gridded ionization chamber to a selected single ^{241}Am α source pulse.
- Fig.9 Drift velocity of electrons in LAr doped with ethylene as a function of the electric field for several concentrations. Superimposed is the estimation of the drift velocity measured by the accordion during the HV scan of the summer 91 test.
- Fig.10 Pure LAr IR absorption spectrum. The prominent peak at

2360 cm^{-1} is characteristic of CO_2 , while the features about 3000 cm^{-1} and 600 cm^{-1} may come from particulate water in the sample.

Fig.11 IR absorption spectrum of ethylene (a) and allene (b) dissolved in LAr (7.5 ppm vol concentration).

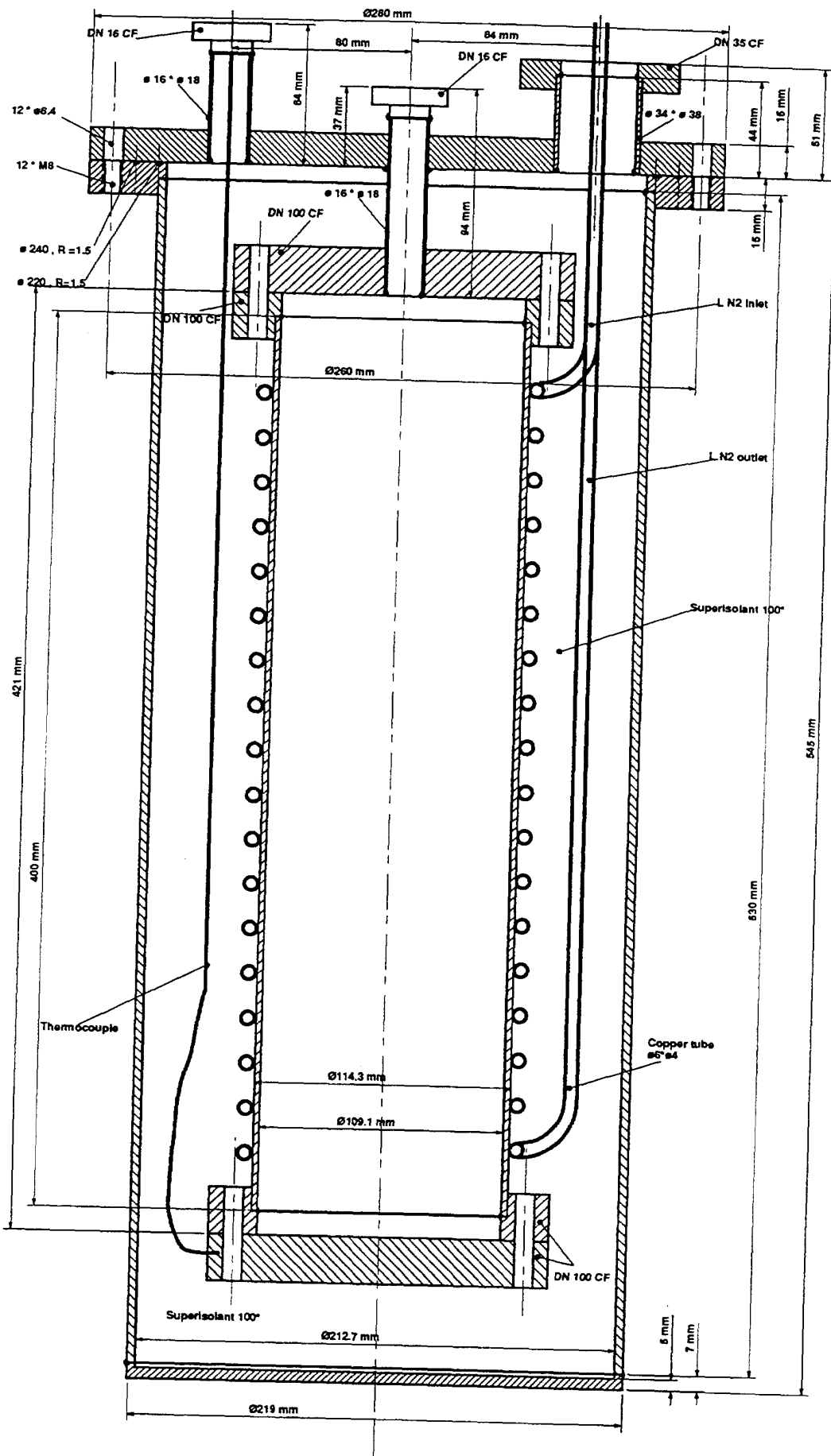


FIGURE 1

DESCRIPTION		POS.	MATIERE	OBSERVATIONS		
ENSEMBLE						
RADIATION TEST SETUP				1:1	DATE	0.02

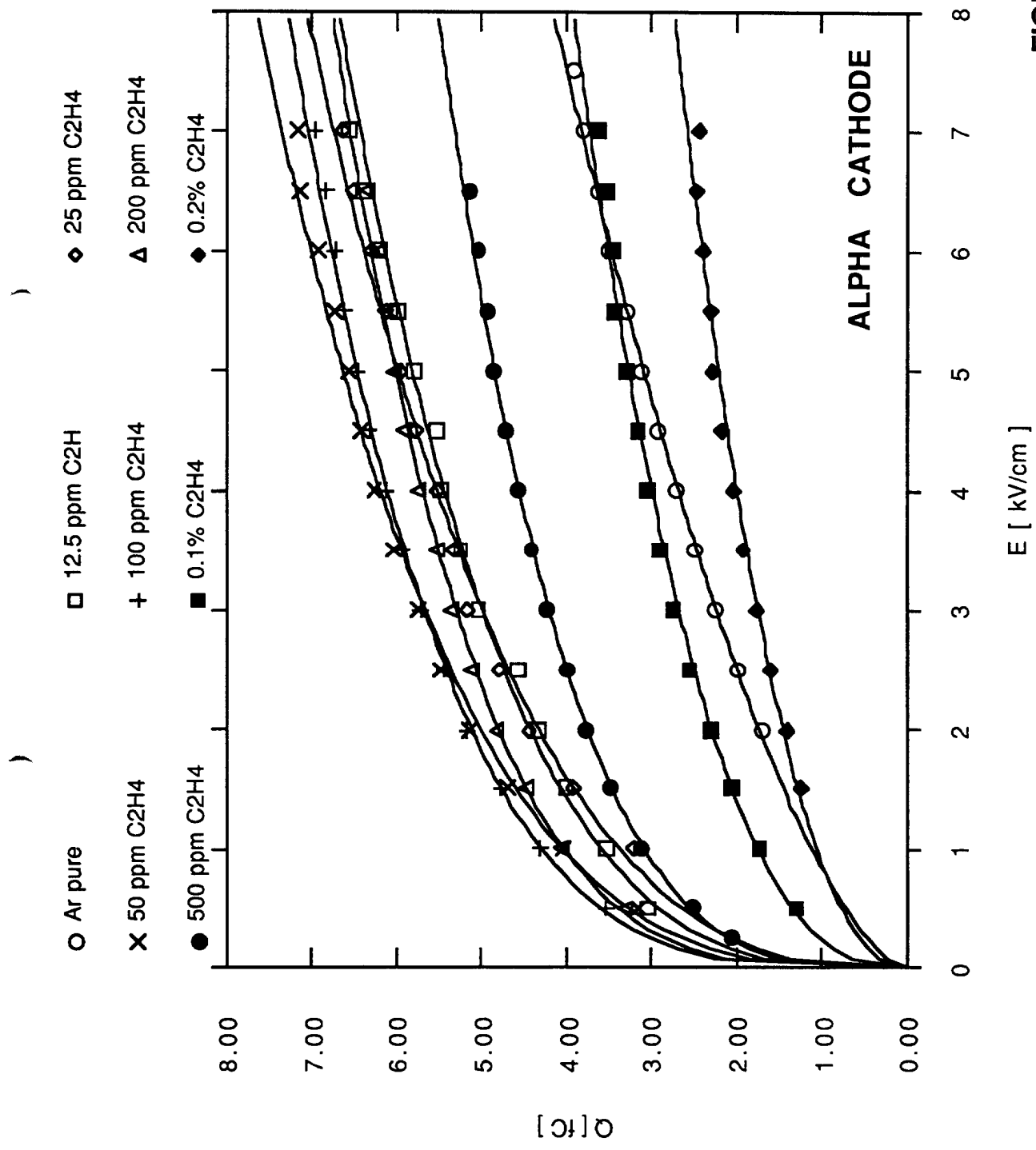


FIGURE 2a

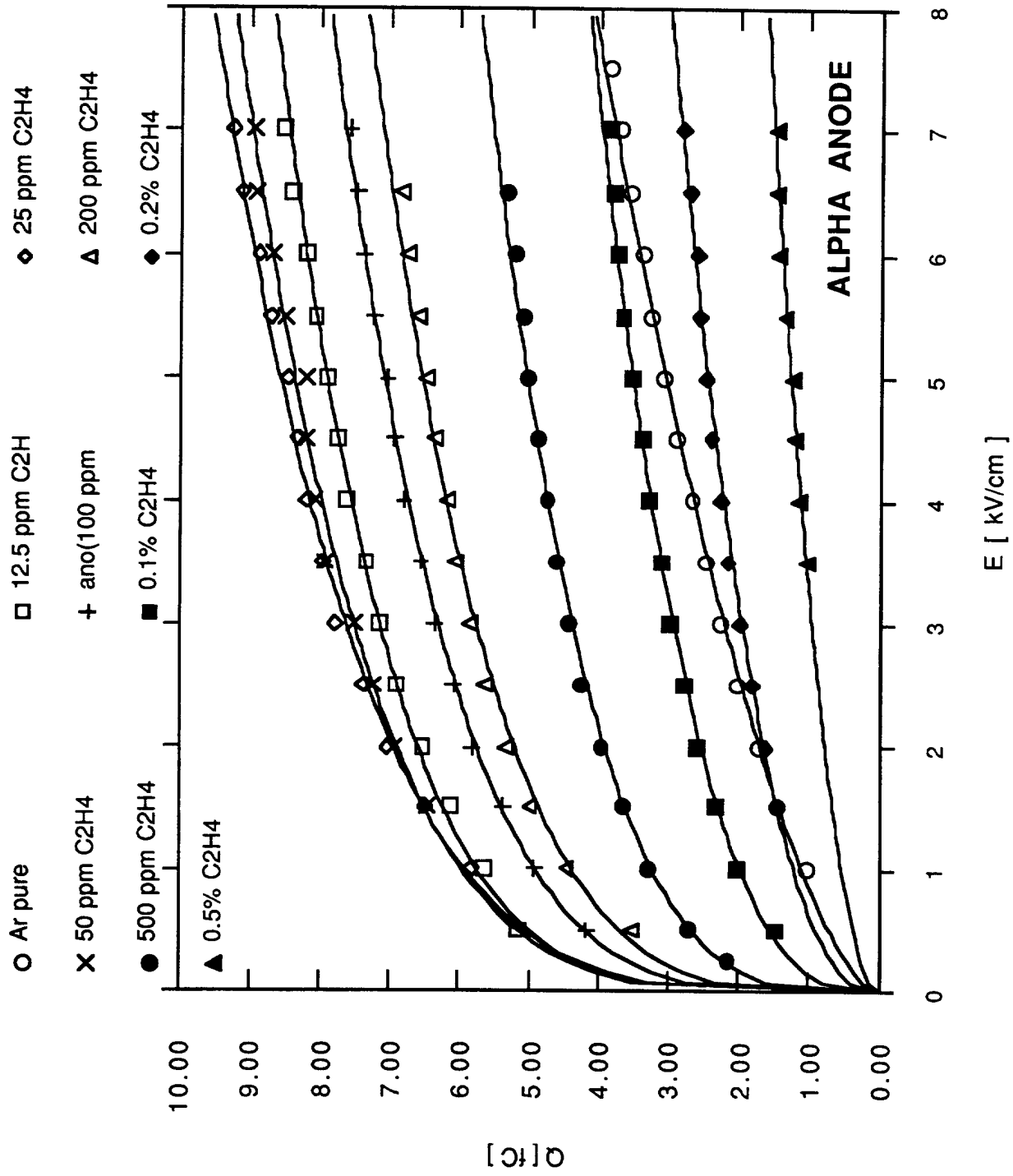


FIGURE 2b

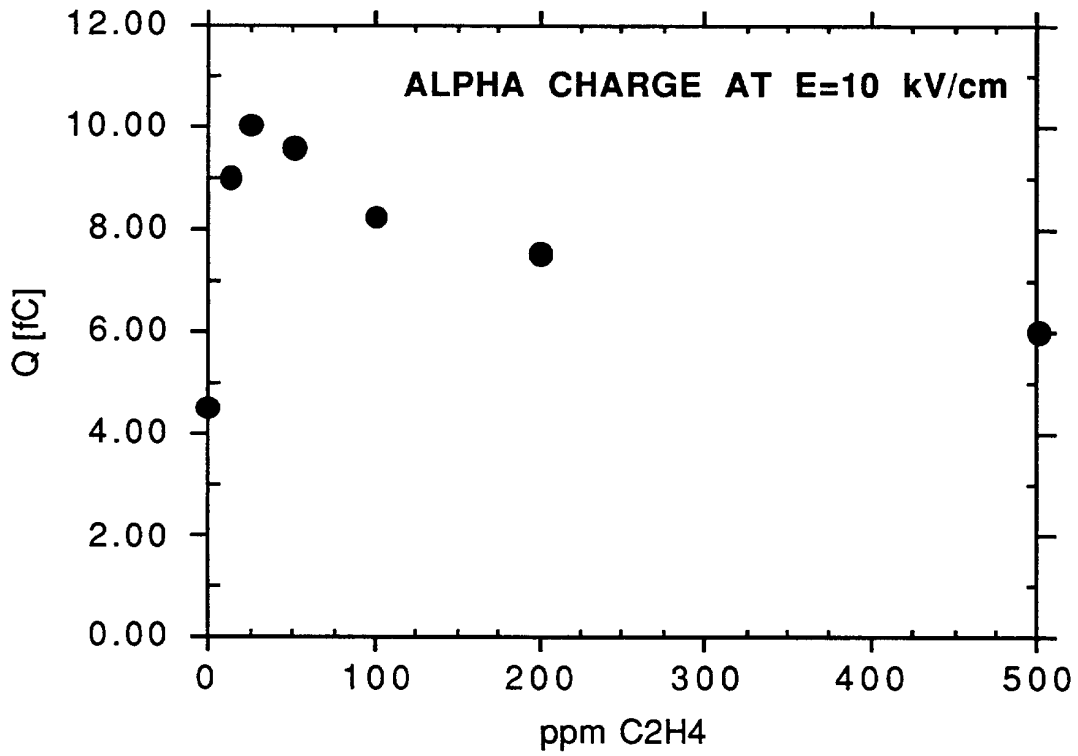
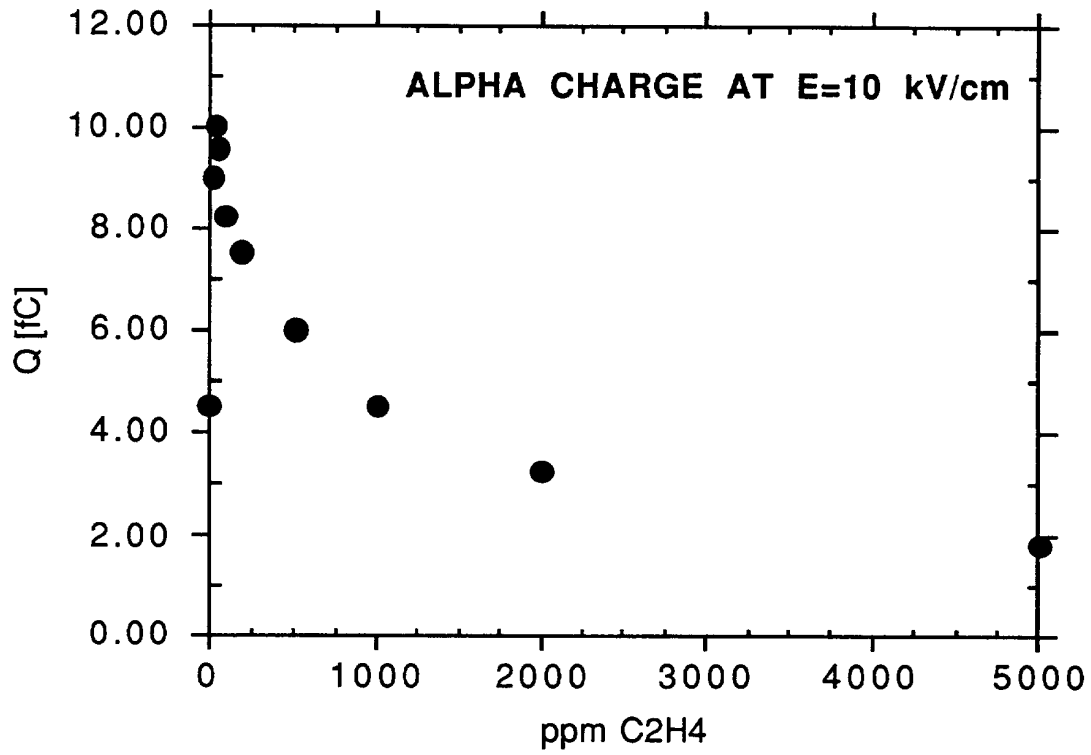
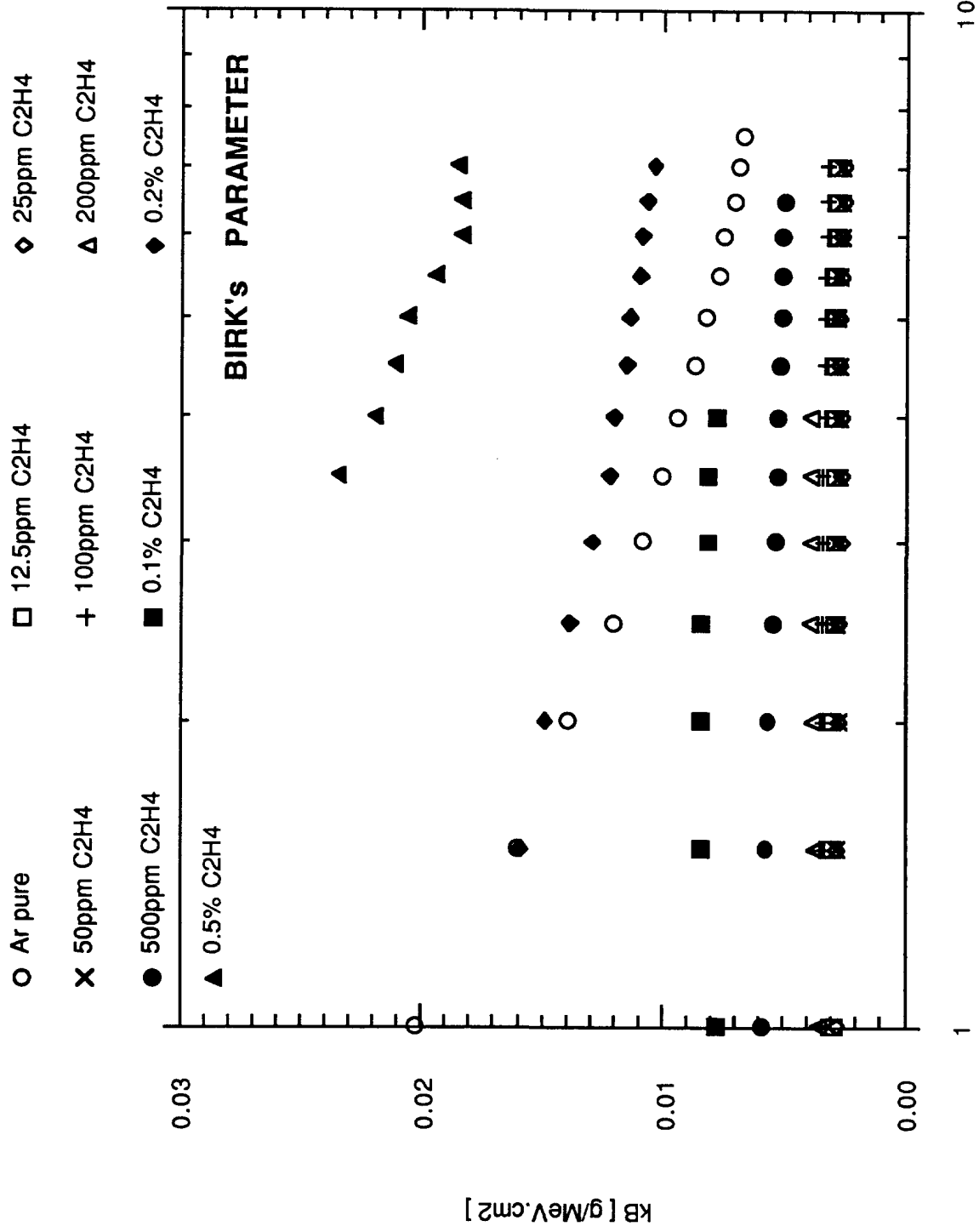


FIGURE 3



E [kV/cm]

FIGURE 4

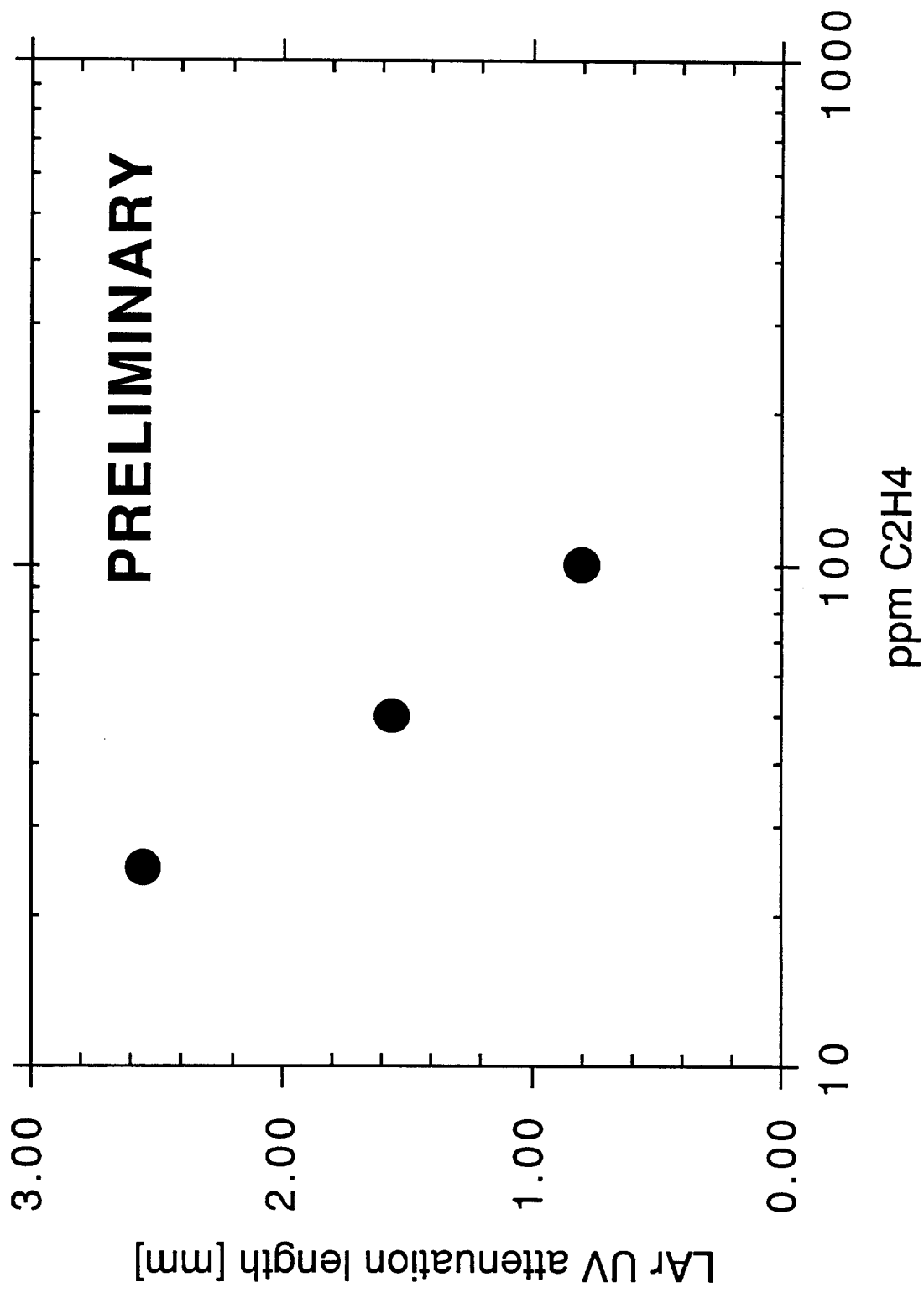


FIGURE 5

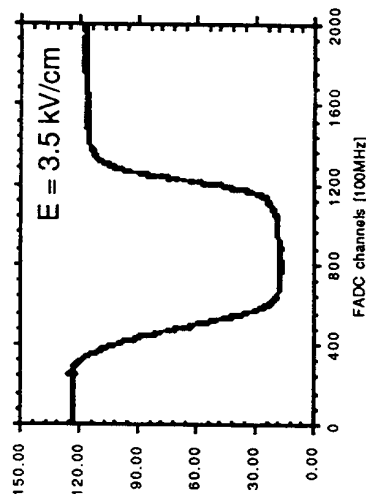
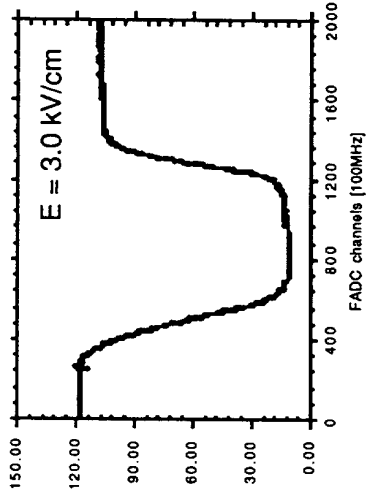
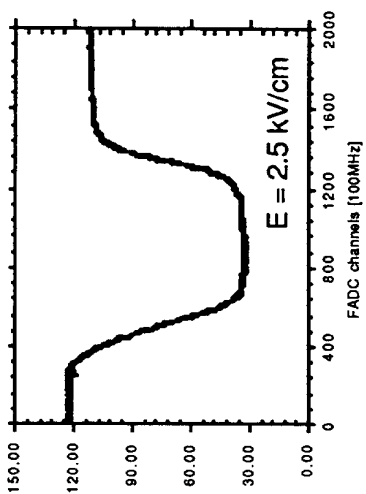
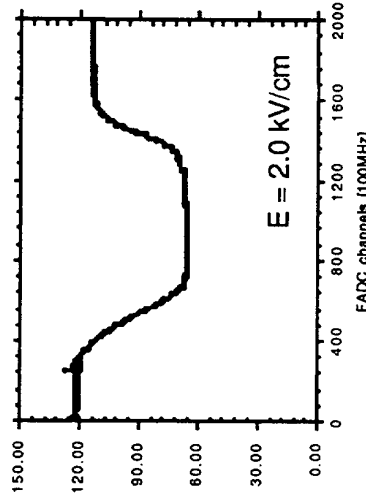
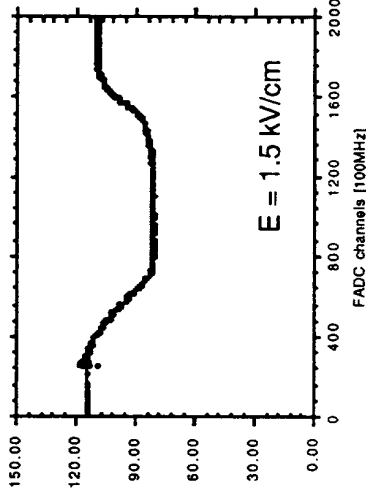
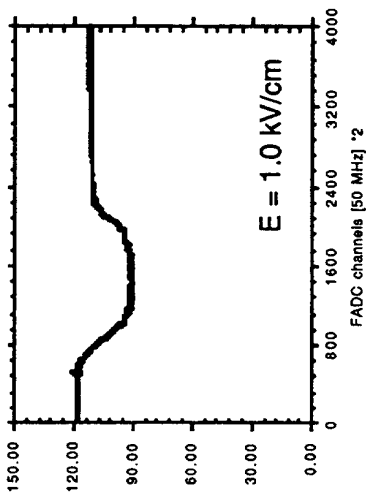


FIGURE 6a

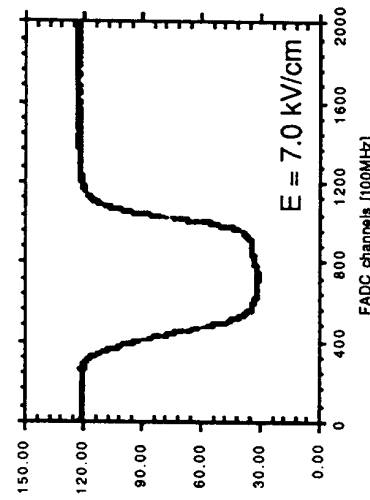
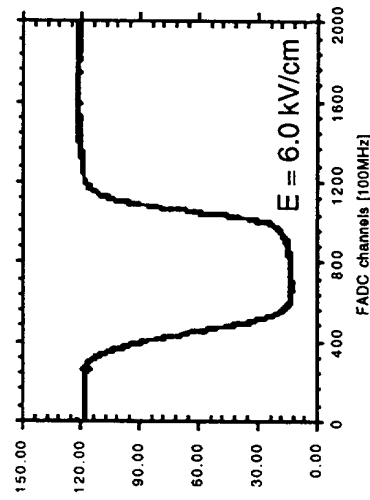
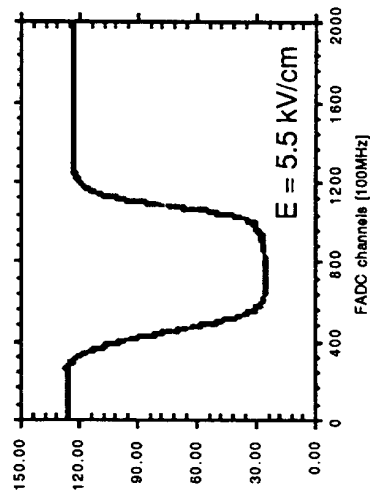
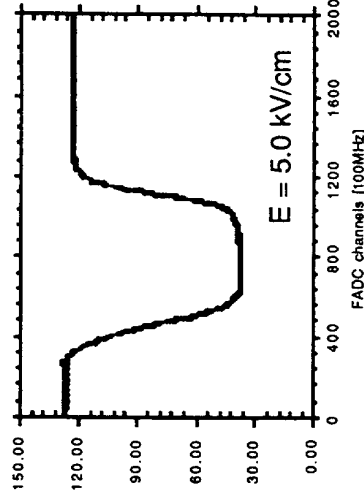
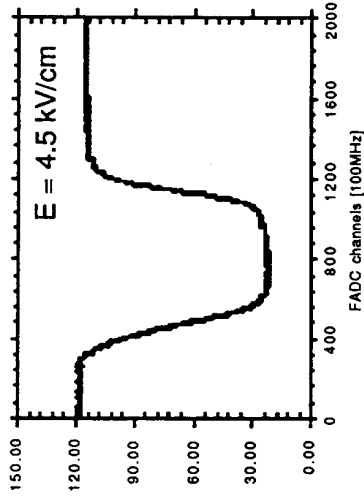
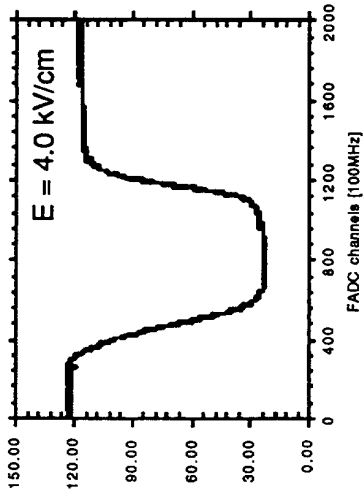


FIGURE 6b

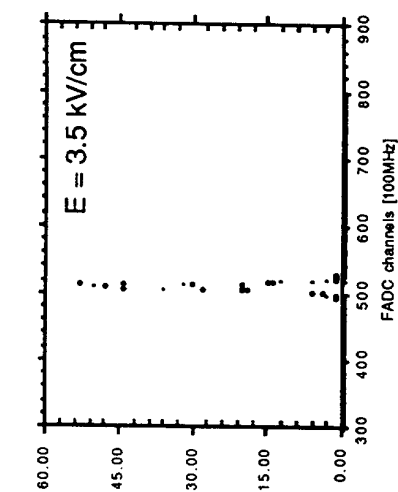
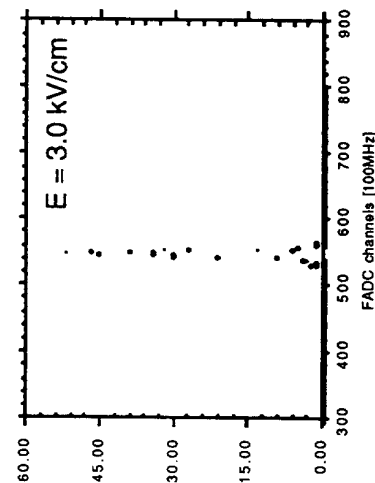
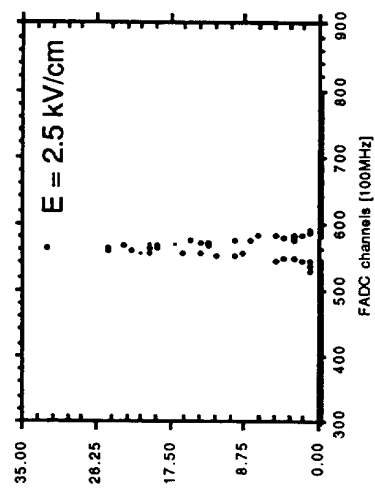
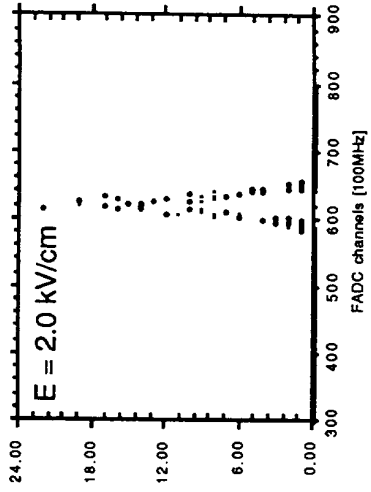
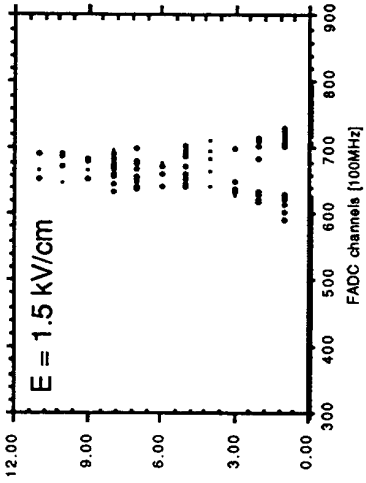
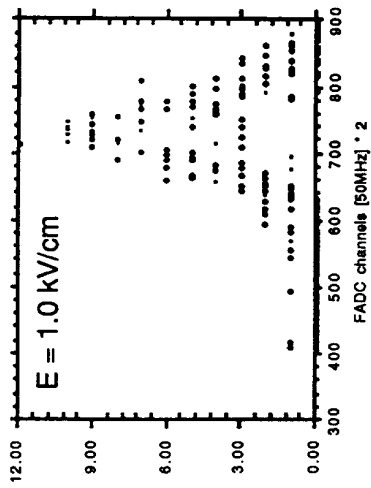


FIGURE 7a

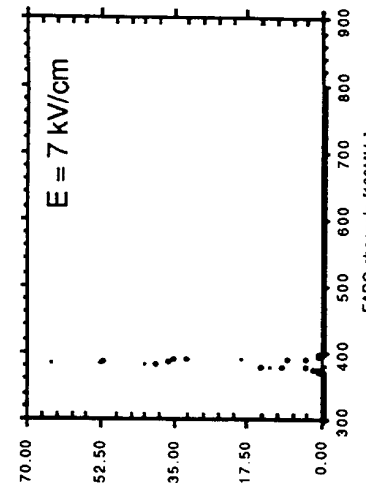
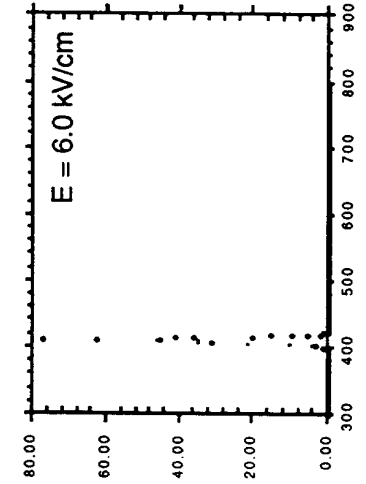
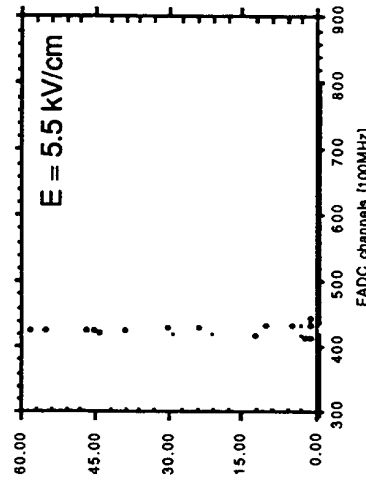
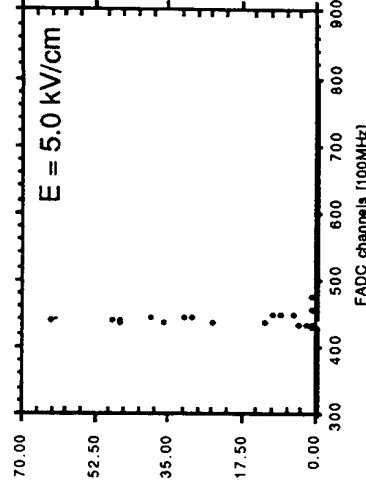
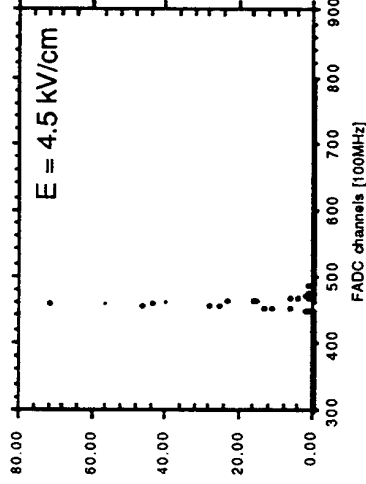
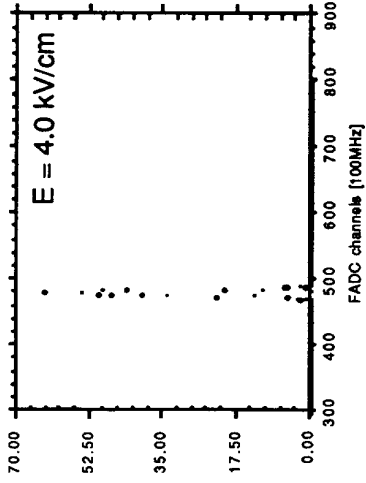


FIGURE 7b

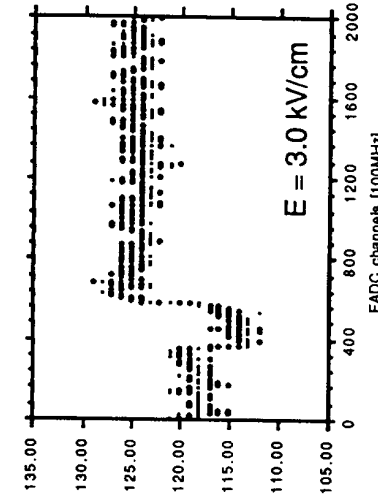
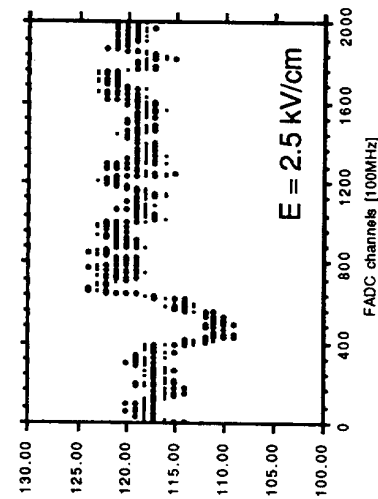
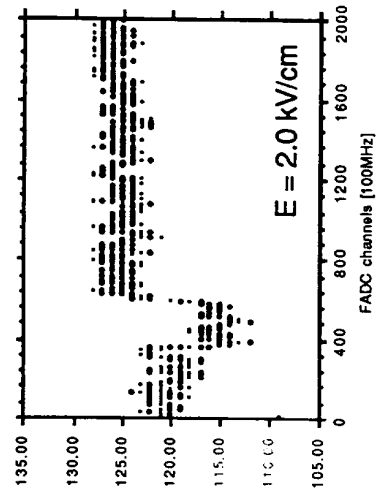
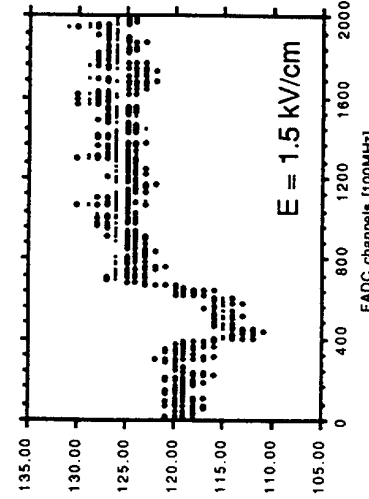
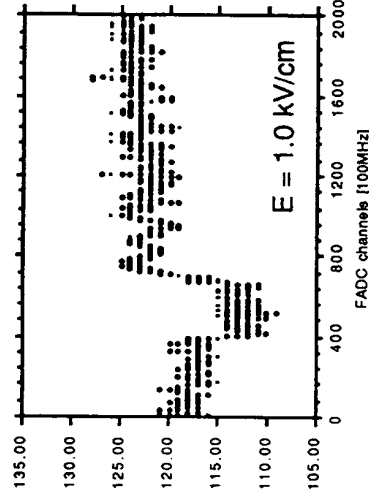
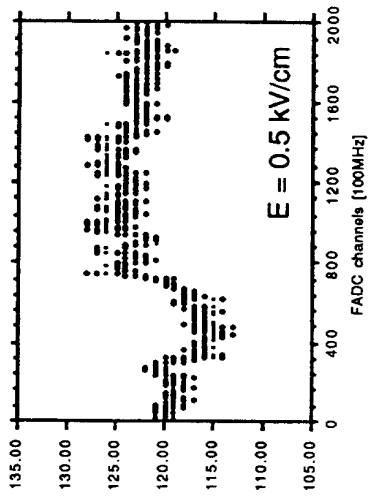


FIGURE 8a

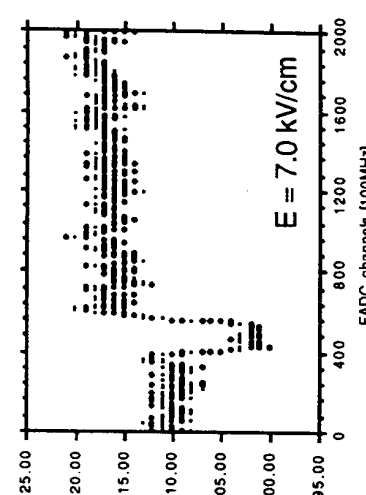
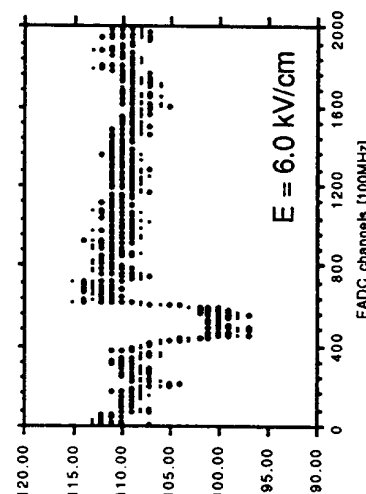
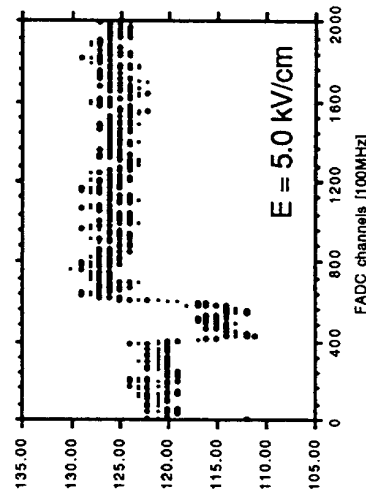
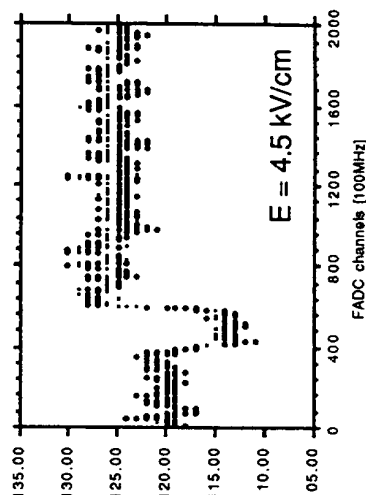
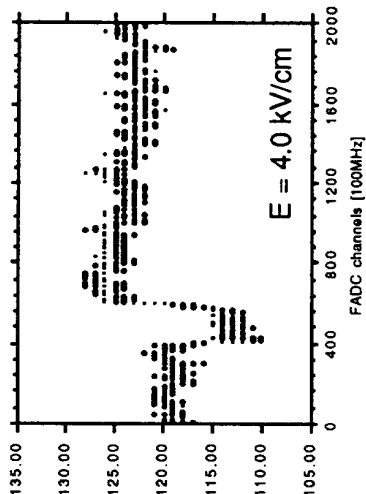
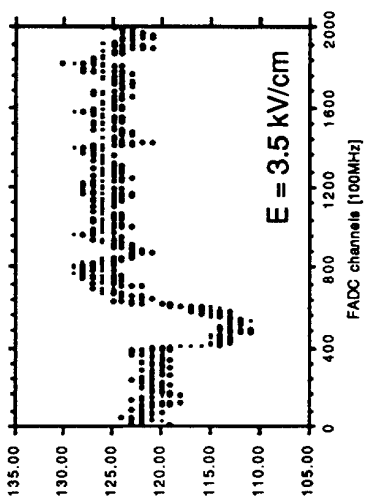


FIGURE 8b

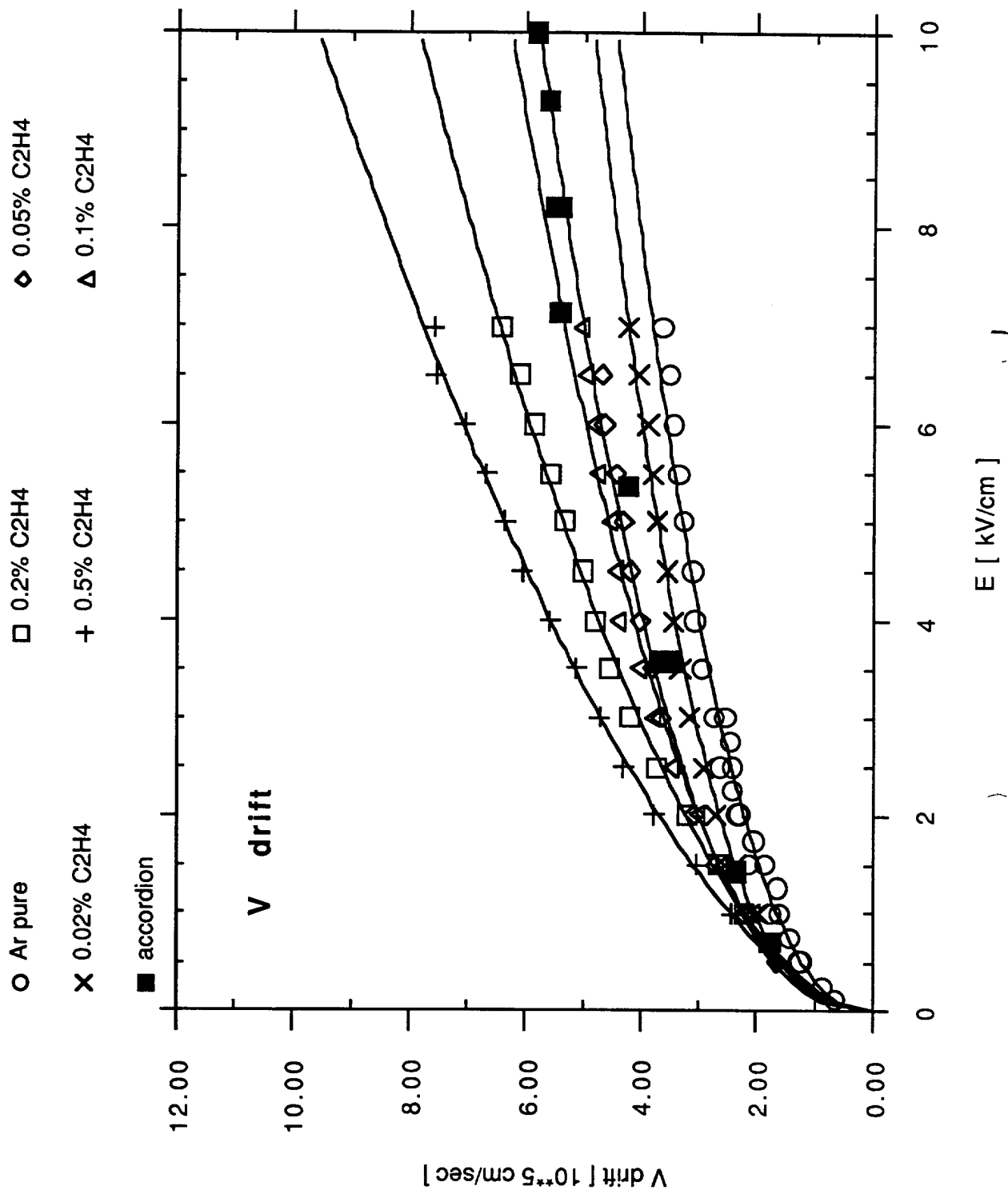


FIGURE 9

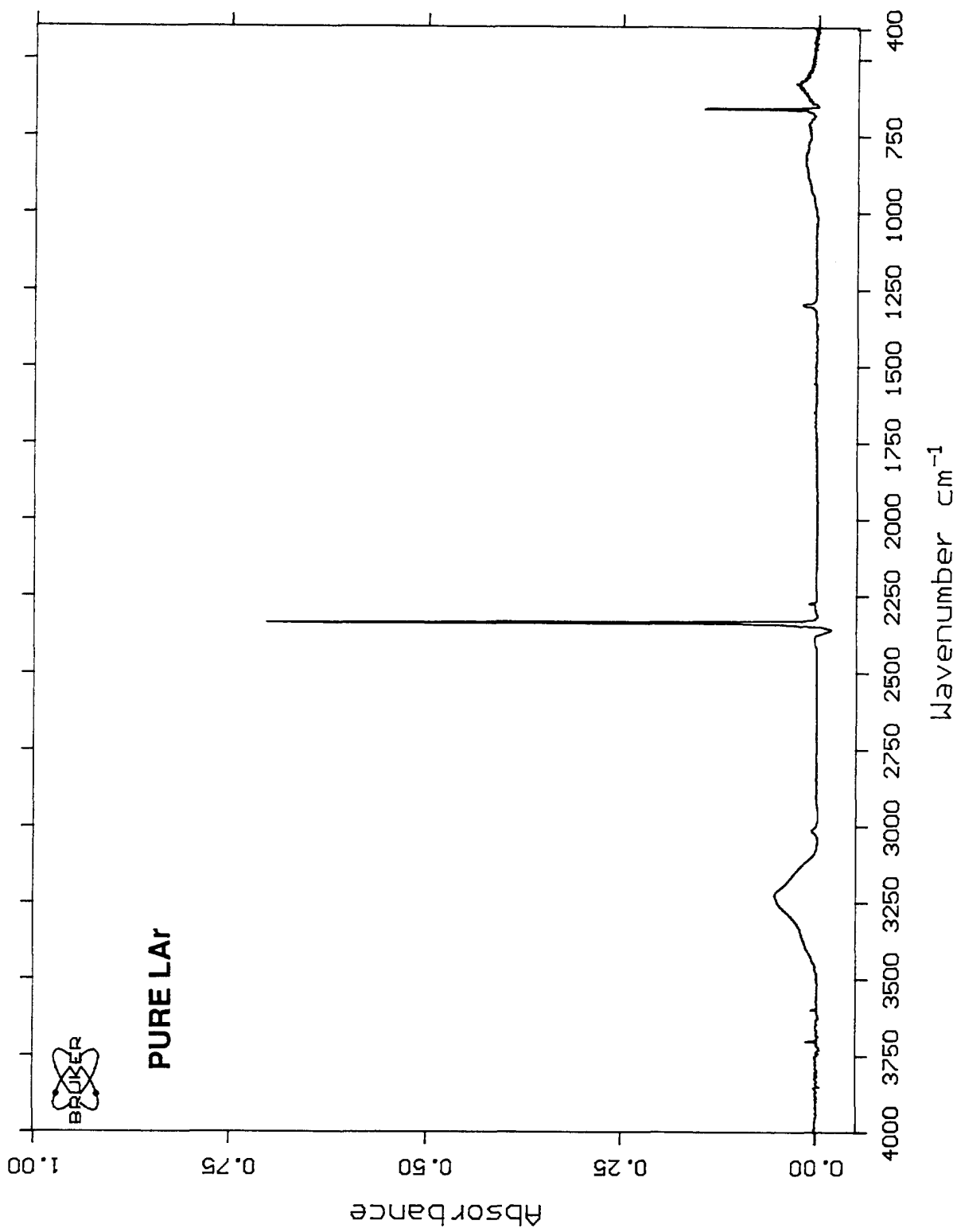


FIGURE 10

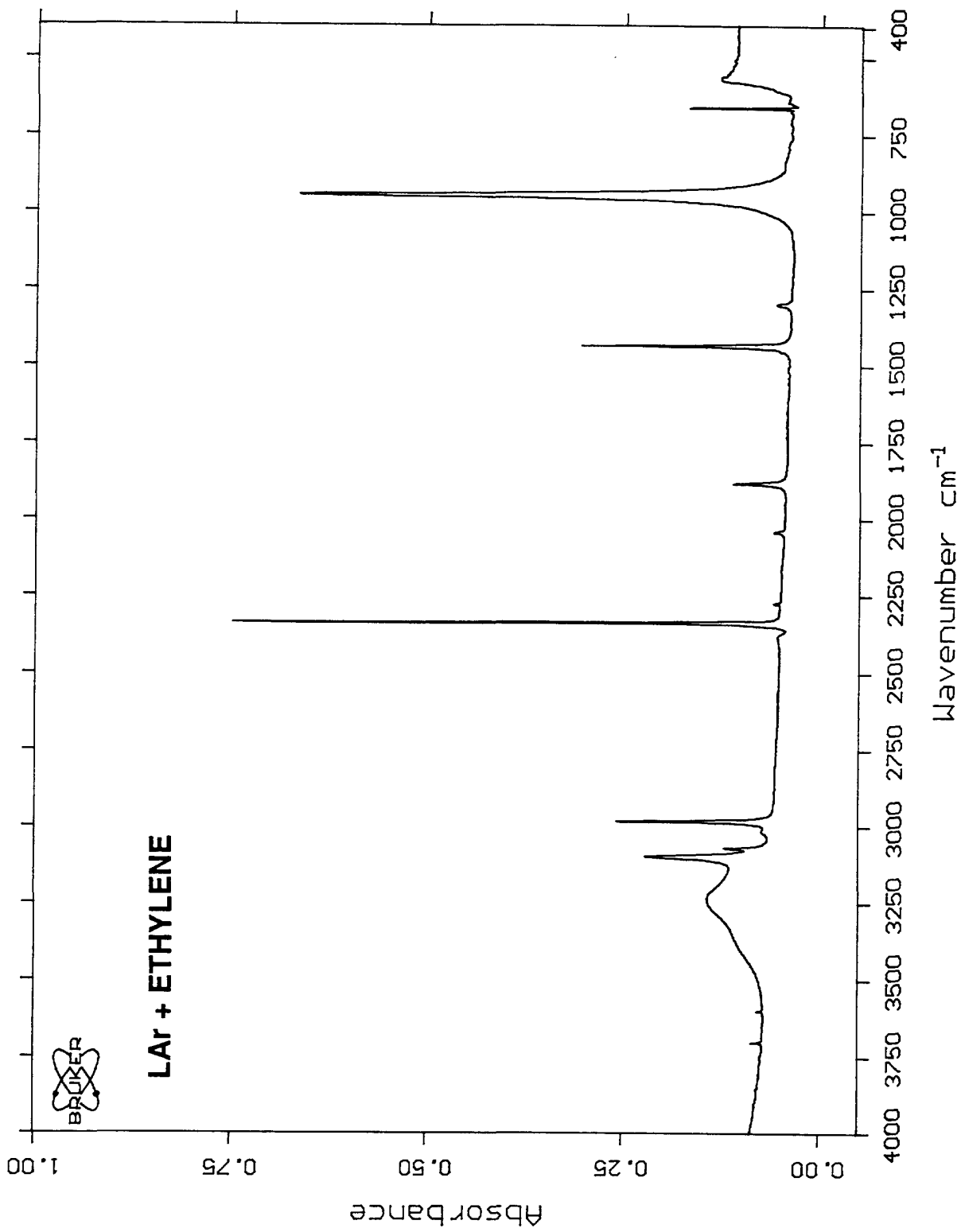


FIGURE 11a

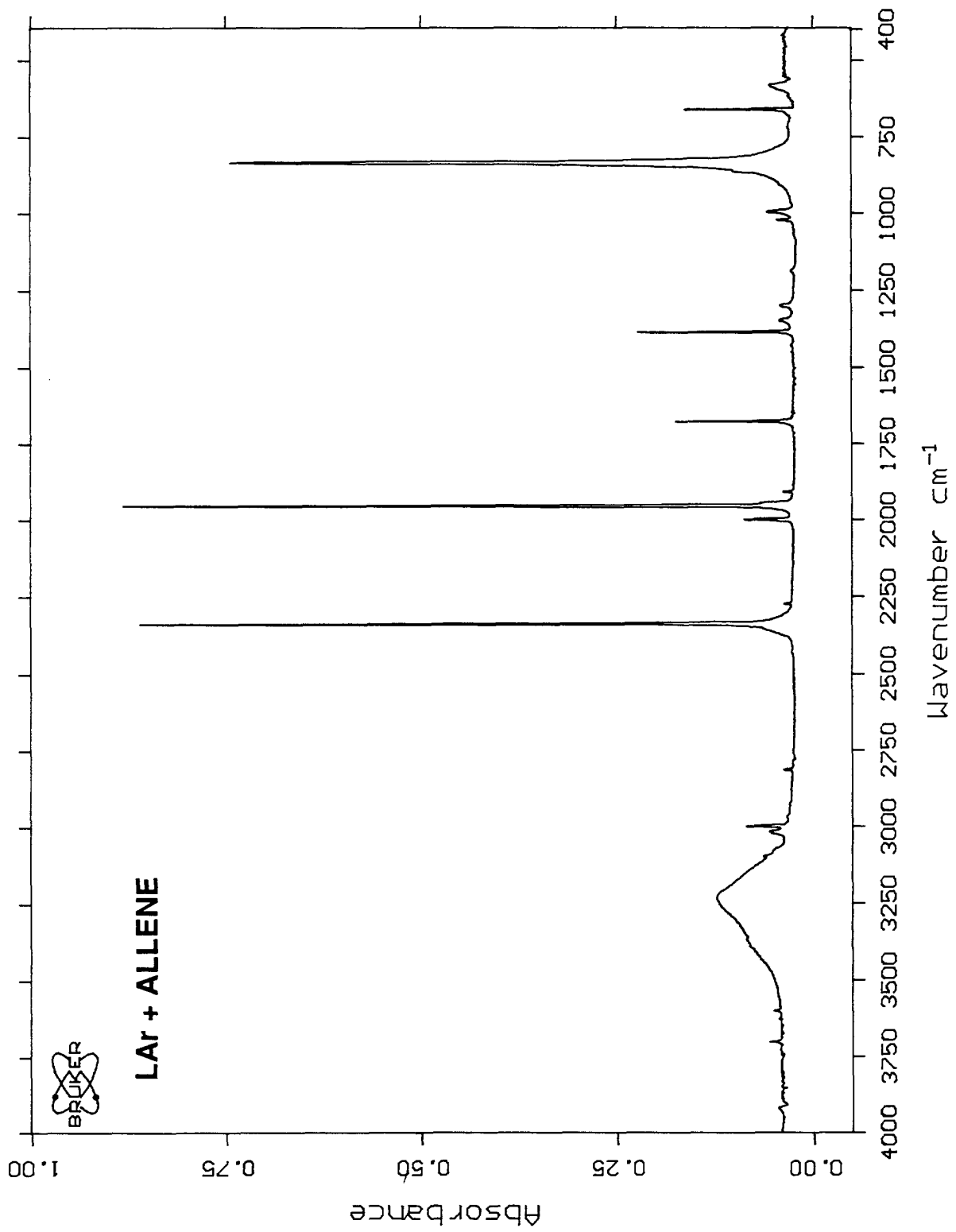


FIGURE 11b

