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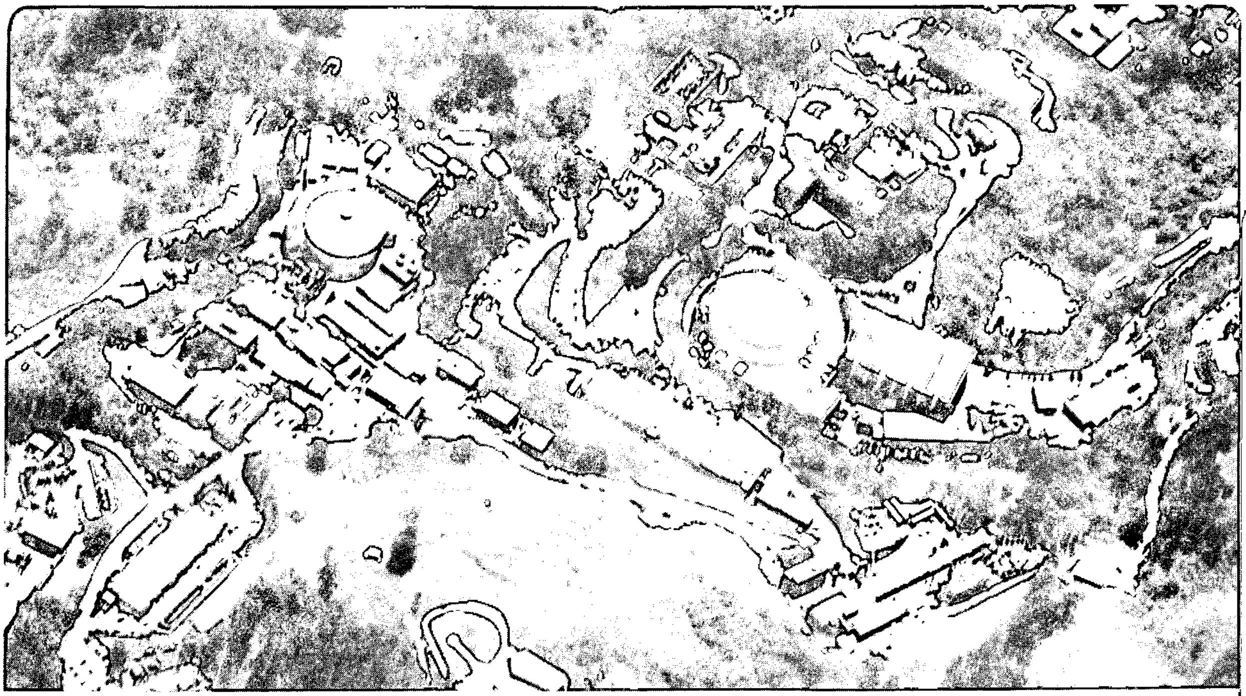

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EFFECTS OF FREONS ON WIRE CHAMBER AGING*

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Abstract

Data are presented to suggest that trace amounts of Freons do not affect the rate of gain loss in wire chambers filled with either dimethyl ether or argon/ethane. Apparent gain loss in dimethyl ether is explained by attachment of primary electrons to a continuously increasing concentration of Freon 11 in the counter gas. An increase in the concentration of Freon 11 in dimethyl ether is caused by a distillation process in the gas supply bottle and is a natural consequence of the unequal volatilities of the two compounds. Degradation of ^{55}Fe pulse-height spectra obtained at the irradiated region of the anode wire is observed, however.

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1. Introduction

There are at least two reasons why Freons might be expected to cause aging in wire chambers. (1) Freons contain halogens, which are known to catalyze plasma polymerization of hydrocarbons in the low-pressure rf-discharge regime [1]. It has been suggested that there may be similarities between the low-pressure rf-discharge plasma regime and that of wire chambers [2]. (2) Some Freons are very electronegative and are therefore expected to attach electrons and be drawn to the anode, where, if they do catalyze polymerization, they could most easily affect the aging rate.

Since the primary electrons lost by attachment to electronegative species are not available to initiate avalanches, however, the observed wire current can change if the concentration of electronegative species in the gas changes. It is possible that an increase in the concentration of a highly electronegative species such as Freon 11 may cause the wire current to drop, a result which may be interpreted as aging [3]. In this report we discuss a mechanism by which trace species that are less-volatile than the main gas may be concentrated in a gas bottle.

2. Experimental

The aging tests in this study were all performed using copper proportional tubes (3/8" ID) with 50-μm-diameter gold-plated tungsten wires (Luma Metall, Kalmar, Sweden). The design of the proportional tube and the techniques used to collect aging data have already been described [3]. The 5.9 keV photons emitted by an ^{55}Fe source were used to simulate particle radiation. Initial current densities were ~1.3 μA/cm, corresponding to wire currents of ~400 nA for an irradiated region 3 mm long. The gas gains were known only approximately, and were in the 20,000-50,000 range. The gas flow rate in all tests was approximately 20 cm^3/min, corresponding to 1.25 chamber volumes/min, or an average linear velocity of 0.47 cm/sec.

We express the aging rate, R, as the normalized rate of gain loss:

$$R = -(1/G_o)(dG/dQ), \tag{1a}$$

where G_o is the initial gas gain, G is the gas gain at a subsequent time, and Q is the accumulated charge per length of wire. If the ionization initiating the avalanches does not change, the current on a wire is linearly proportional to the gas gain, and R can be measured as the normalized rate of current decrease:

$$R = -(1/I_o)(dI/dQ), \tag{1b}$$

where I_o is the initial wire current, I is the wire current at a subsequent time, and R and Q are as defined above. We express R in units of %/C/cm.

3. Aging Tests with Freons in Argon/Ethane

In an attempt to find contaminants that would induce aging in 50% argon + 50% ethane (Matheson grade argon, CP grade ethane, Matheson Gas Products, Newark, CA), a series of aging tests was performed in which gas permeation [4] was used to add halogenated (Freon) contaminants to the argon/ethane. Permeation was effected with a Kin-Tek Precision Gas Standards Generator Model 570-C (Kin-Tek Laboratories, Texas City, TX). These aging tests were run only for relatively short periods in an exploratory mode, looking for easily-observed increases in aging rates. In no case was there evidence that Freon addition resulted in significant aging rates. These results are summarized in Table 1.

Table 1
Summary of aging results in which halogenated contaminants were deliberately added to 50% argon + 50% ethane. These tests and their respective control tests were run simultaneously and under otherwise identical conditions, except that no contaminants were added to the controls. No significant differences in the aging rates were observed.

additive	additive name	additive concentration, ppm	accumulated charge, mC/cm	R, %/C/cm	control test R, %/C/cm
$CHClF_2$	Freon 22	8	150	5	3
CCl_2F_2	Freon 12	680	180	-1	5
CH_3Cl	methyl chloride	680	380	3	4
CH_3Cl	methyl chloride	550	360	3	no test

A test in which CCl_3F (Freon 11) was added to argon/ethane is shown in Fig. 1. In this test, the concentration of Freon 11 was set at several different levels; at each level the current was stable. The test was initially run without Freon 11 added to the gas (segment A of Fig. 1); 100 mC/cm of charge was collected during this period and the measured aging rate was R~7 %/C/cm. When Freon 11 was added to the gas at varying levels (segments B-F), the current dropped from its initial value due to attachment of primary electrons, but then stabilized. Meaningful values of R could not be calculated for these segments because only small amounts of charge were collected at the reduced currents. The transient spikes between levels are due to the thermal inertia of the permeation device. The cause of the small bump between segments D and E is not understood at present. Segment F was apparently approaching a stable level, but the Freon source was removed prior to the establishment of that level. After the Freon source was removed from the system, the current returned to its initial level (segment G), indicating that little or no deposit had been formed on the wire. A total of about 0.12 C/cm of charge was collected during the exposure to Freon 11. These results were reproduced in subsequent tests.

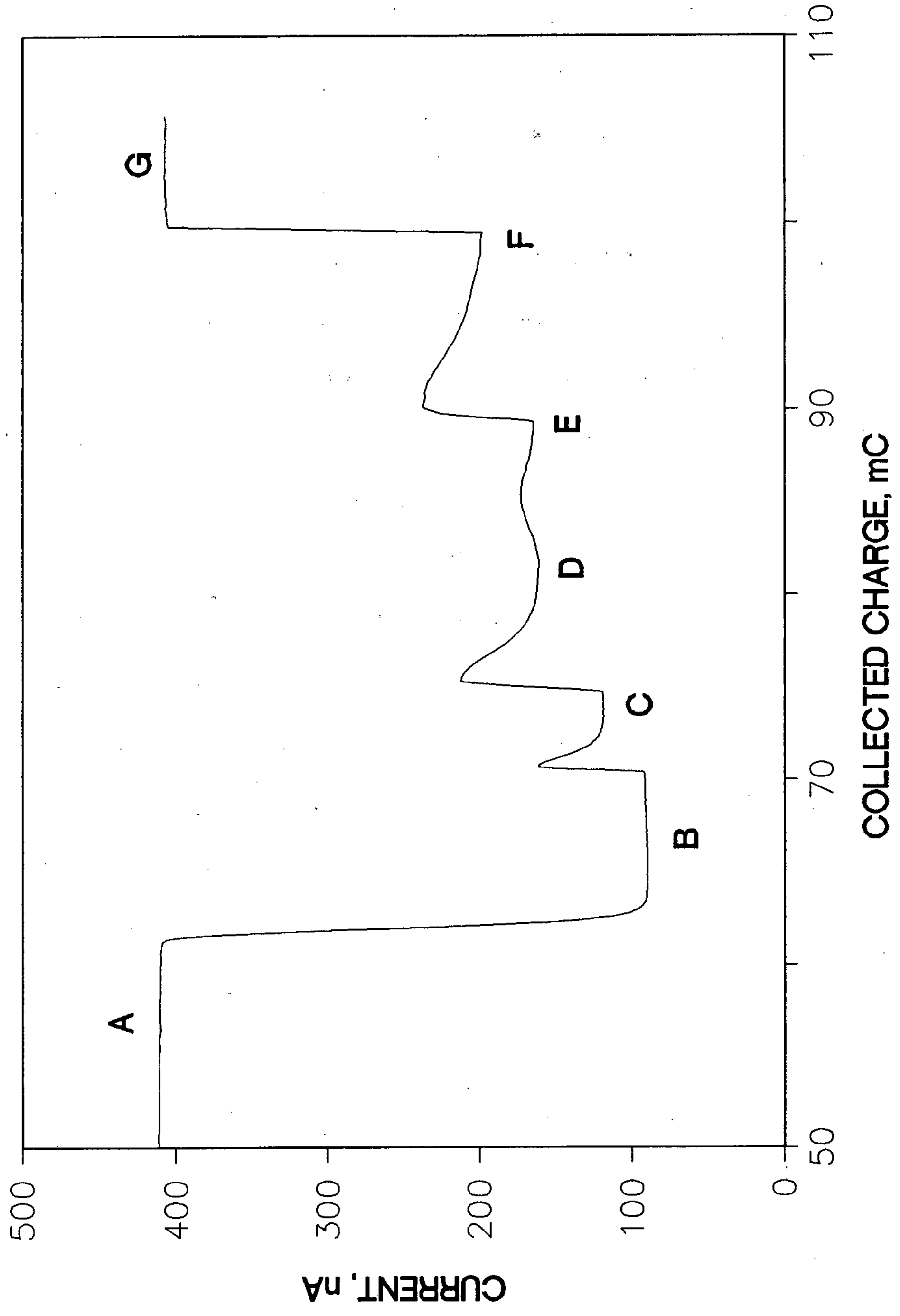

Fig. 1. Aging test in which Freon 11 at varying levels was added to 50% argon + 50% ethane. The irradiated region of the wire was 0.3 cm. Freon concentrations (ppm) were: A, 0; B, 100; C, 70; D, 45; E, 45; F, 34; G, 0.

4. Dimethyl Ether

Dimethyl ether (DME) is an attractive gas with good quenching, a high linear ionization density, a low drift velocity and a low electron diffusion constant [5], properties which make it a good choice for use in high-resolution drift chambers (e.g., vertex detectors). There are, however, several reports [6,7] suggesting that Freon 11 contamination in DME causes rapid aging. Since Freon 11 is a commonly observed [6,7] contaminant in DME, the viability of DME as a useful wire chamber gas has been questioned.

A possible alternative explanation of the above-mentioned observations is that the "aging" is due to a loss of primary electrons by attachment to the very electronegative Freon 11 [3]. If Freon 11 is present in the DME supply, it will be concentrated as the DME, which is more volatile (i.e., has a lower boiling point), evaporates preferentially. This process is known as Rayleigh, or batch, distillation [8]. Results that support this explanation are discussed below.

5. Rayleigh Distillation

Rayleigh distillation describes the equilibrium vaporization process of an initial mass of a binary liquid mixture in a vessel to which heat is added continuously. It is assumed that the liquid is well mixed (i.e., is of uniform composition throughout) and that vapor in equilibrium with the liquid is generated and removed continuously from the vessel. Clearly, the continuous generation and removal of vapor is a valid description of the vaporization of DME in the gas supply bottle. Further, we believe it reasonable to assume that the liquid is well mixed since liquid diffusivities are high.

To describe a Rayleigh distillation, it is necessary to relate the amount of liquid remaining to its composition. This is done by means of a mass balance relating changes in the amount and composition of the liquid to the amount and composition of the vapor removed. In its most general form, the Rayleigh equation (for component i) is

$$\ln(L/L_o) = \int_{x_{io}}^{x_i} dx_i/(y_i - x_i), \qquad (2)$$

where L_o and x_{io} are the initial number of moles of liquid and liquid mole fraction, and L, x_i, and y_i are the number of moles of liquid, liquid mole fraction, and vapor mole fraction at any subsequent time. Integration of equation (2) requires a relation between y_i and x_i; the equilibrium relation, as defined by the relative volatility, is used.

The relative volatility, or separation factor, α, between two phases in equilibrium is defined as

$$\alpha = (x_A/x_B)_{\text{phase }1}/(x_A/x_B)_{\text{phase }2}. \qquad (3a)$$

It is conventional to choose the phases so that $\alpha>1$. For the case at hand, this means that phase 1 is the gas, phase 2 is the liquid, the subscript A refers to the light component

(DME), and the subscript B refers to the heavy component (Freon 11). Following the common usage that x_i refers to a liquid-phase mole fraction and y_i to a gas-phase mole fraction,

$$\alpha = (y_A/y_B)/(x_A/x_B). \tag{3b}$$

Because we are interested in very dilute solutions (Freon mole fractions less than 10^{-4}), we can assume ideal behavior. This means that the solution obeys Raoult's law, which states that the vapor pressure exerted by a component in the solution is equal to the vapor pressure of the pure component weighted by the mole fraction of that component in the solution, and that the vapor phase obeys Dalton's law, which states that the partial pressure of a component in a gas mixture is equal to the total pressure of the gas weighted by the mole fraction of that component in the gas. Combining Raoult's and Dalton's laws,

$$y_iP = p_i = x_iP_i^\circ, \tag{4}$$

where P is the total pressure of the vapor phase, p_i is the partial pressure of component i, and P_i° is the vapor pressure of pure component i.

A consequence of Raoult's and Dalton's laws is that α is simply the ratio of the vapor pressures of the two pure components at a given temperature:

$$\alpha = P_A^\circ/P_B^\circ. \tag{5}$$

It can be seen that α is independent of pressure and composition. For this situation, it is further argued that α is only a weak function of temperature [9]. As the temperature of the gas supply bottle is expected to undergo little or no fluctuation, we are justified in treating α as a constant.

Making use of the fact that $x_A + x_B = y_A + y_B = 1$ for a binary mixture, equation (3b) can be rewritten as

$$y_A = \alpha x_A/[1 + (\alpha-1)x_A]. \qquad (6)$$

Substituting equation (6) into equation (2), using the fact that α is constant, and integrating with respect to component A gives

$$\ln(L/L_o) = (1/\alpha-1)\ln[((1-x_B)/x_B)/((1-x_B)/x_B)_o] - \ln(x_B/x_{Bo}). \qquad (7)$$

With an appropriate value for α (see the following section), this equation can be used to predict the Freon 11 concentration as a function of the amount of liquid DME remaining in the bottle.

6. Simulation of Rayleigh Distillation

For general applicability, we refer to the simulation results in terms of the <u>enrichment multiple</u> (x_B/x_{Bo}), which is the heavy-component mole fraction normalized to its initial value, and the fraction of the initial amount of liquid that has been used ($1-L/L_o$). Equation (6) relates the vapor- and liquid-phase concentrations of the light component, but can be rewritten to relate the concentrations of the heavy component:

$$y_B = x_B/[\alpha + (1-\alpha)x_B]. \tag{8}$$

Noting that we are in a regime in which $x_B \ll 1$, and hence $(1-\alpha)x_B \ll \alpha$, equation (8) can be approximated as

$$y_B = x_B/\alpha. \tag{9}$$

The enrichment multiple therefore applies to both the vapor and liquid phases.

The pure-component vapor pressures needed to determine α can be calculated from tabulated data [10]. The predicted relative volatility of DME/Freon 11 near room temperature is shown in Table 2.

Table 2
Predicted relative volatility (α) of DME/Freon 11 near room temperature [10].

T, °C	α
18	5.78
19	5.74
20	5.70
21	5.66
22	5.62
23	5.59

In the regime of relative volatility ($5.5<\alpha<5.8$) and initial Freon concentration ($x_{Bo}<10^{-4}$) in which we are interested, the solution of equation (7) is not sensitive to variations in either of these parameters: the Freon 11 concentration doubles each time 57% of the liquid remaining in the bottle is used. The prediction for $\alpha=5.63$ and $x_{Bo}=10^{-7}$ is shown in Table 3.

Table 3
Numerical solution to equation 7 for $\alpha=5.63$ and $x_{Bo}=10^{-7}$.

enrichment multiple	fraction of liquid used	enrichment multiple	fraction of liquid used
1.0	0.000	2.8	0.714
1.1	0.109	3.0	0.737
1.2	0.199	3.5	0.782
1.3	0.273	4.0	0.815
1.4	0.336	4.5	0.839
1.5	0.389	5.0	0.859
1.6	0.435	6.0	0.887
1.7	0.475	7.0	0.906
1.8	0.511	8.0	0.920
1.9	0.542	10.0	0.939
2.0	0.570	12.0	0.951
2.2	0.617	14.0	0.960
2.4	0.655	16.0	0.966
2.6	0.687	32.0	0.985

7. Aging Tests with Freon 11 in Dimethyl Ether

We previously reported [3] that very low aging rates can be achieved with DME using gas plumbing that is rigorously free of plastics. The tests described below were run using such a plumbing system.

To investigate the effect of a constant level of Freon 11 in DME, a Kin-Tek SRT-type source was used with the gas permeation system mentioned above to add ~5 ppm of

Freon 11 to otherwise pure DME (Dymel A, DuPont, Wilmington, DE). (We measured the Freon 11 concentration in this DME to be ~5 ppb.) The SRT source is a ~10-cm length of Teflon tube that contains liquid Freon 11. Although Teflon has been reported to cause signal loss [7], we believe that it did not affect this test. Indeed, low aging rates have been achieved using DME in systems with certain plastics, including Teflon [11]. To ensure that the Freon 11 concentration in the DME supply would not change appreciably due to a Rayleigh distillation, less than 30% of the liquid in the DME supply bottle was used over the course of the test. The test was started without any added Freon 11; as shown in Fig. 2, the current dropped to ~25% of its initial value upon addition of the Freon. The permeation source was bypassed several times during the test, and the current returned to its initial level each time. A total of 0.45 C/cm of charge was collected during the exposure to Freon 11, and the measured aging rate during this period was consistent with a value of zero. ^{55}Fe pulse-height spectra taken at the end of the test were somewhat degraded, however (Fig. 3), while those taken at the end of a comparable test without added Freon 11 were not (Fig. 4). The spectra shown in Fig. 3 were taken at successive positions along the irradiated region of the wire and display the classic degradation pattern [12]. These data indicate that the presence of Freon 11 in DME does not result in measurable

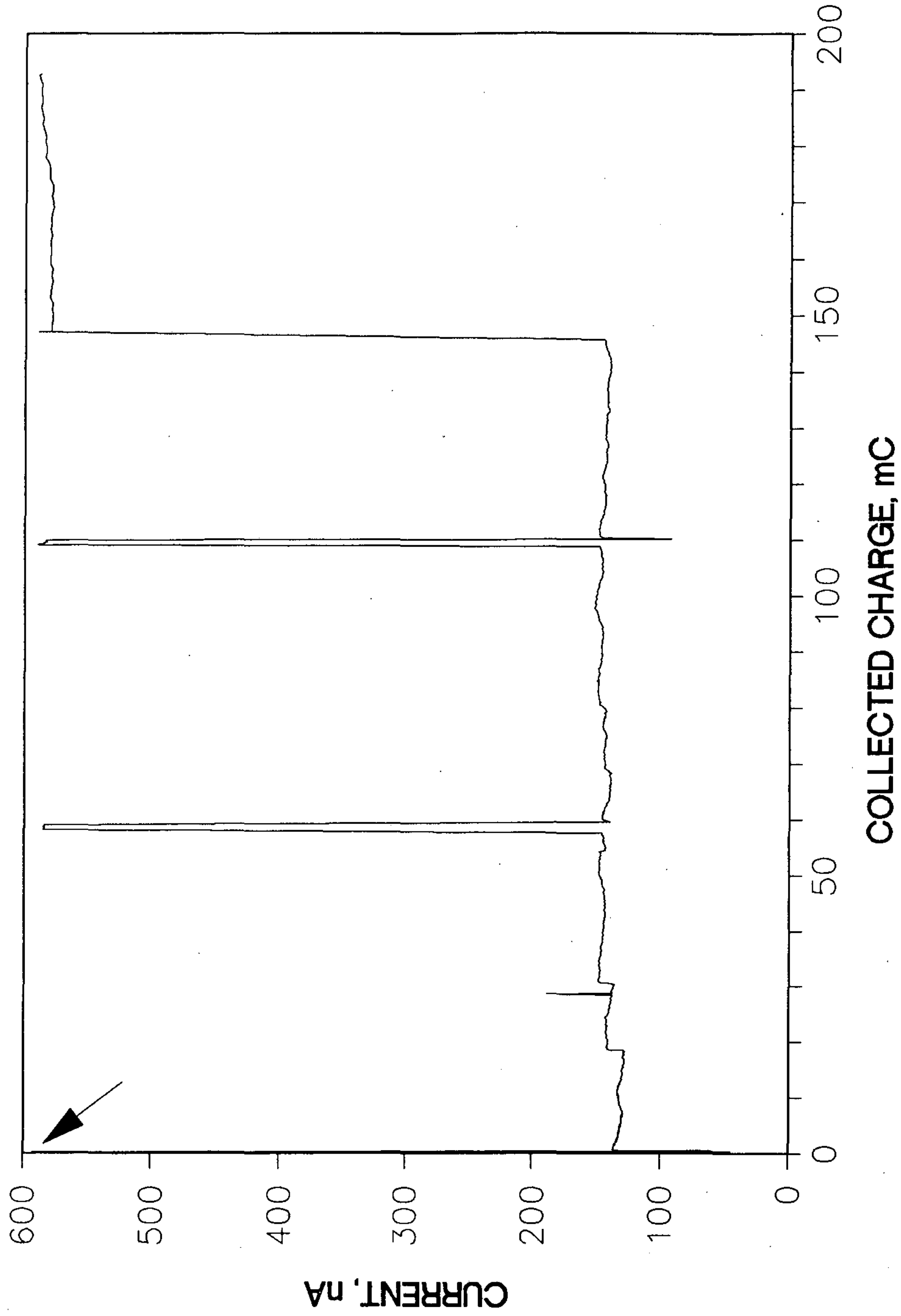

Fig. 2. Aging test in which ~5 ppm Freon 11 was added to DME. The current dropped ~75% from its initial value (indicated by the arrow) upon addition of the Freon, but no aging was observed. The irradiated region of the wire was 0.3 cm.

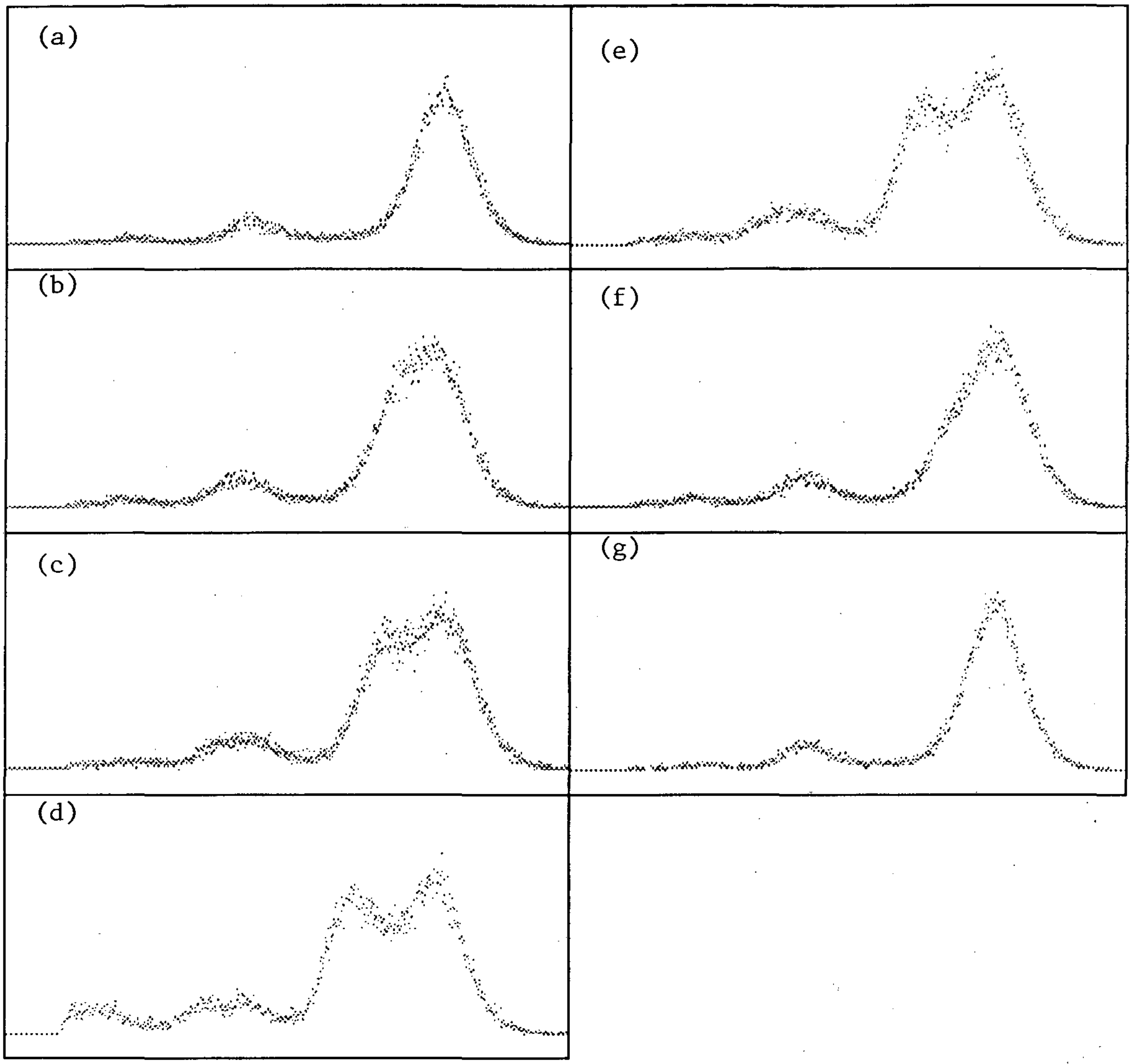

Fig. 3. ^{55}Fe pulse-height spectra taken after DME aging test with ~5 ppm Freon 11. The ^{55}Fe line source was perpendicular to the wire. The classic pattern of degradation of the spectrum is seen [12]. The positions of the ^{55}Fe line source with respect to the approximate center of the irradiated region were: a, -3; b, -2; c, -1; d, 0; e, +2; f, +3; g, +4, where distances are in millimeters, and negative positions are upstream and positive positions are downstream with respect to the direction of gas flow.

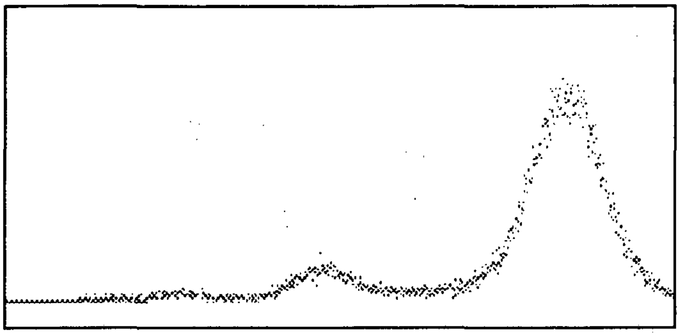

Fig. 4. ^{55}Fe pulse-height spectrum taken at the center of the irradiated region of the wire after DME aging test with no added Freon 11. The ^{55}Fe line source was perpendicular to the wire. No significant degradation of the spectrum is observed.

gain loss, but does cause some degradation of energy resolution.

To investigate the possibility of Freon 11 being concentrated by a Rayleigh distillation, two aging tests were performed in which all of the liquid DME (Semiconductor grade, Matheson Gas Products, Newark, CA) in the bottle was used. It was necessary to use all of the liquid because large changes in the enrichment multiple are predicted to occur as 80-100% of the liquid is used (Table 3). The initial gas-phase Freon 11 concentration was approximately 0.2 ppm in the first test and approximately 0.04 ppm in the second test.

In each of these tests, one of which is shown in Fig. 5, the current was initially stable, but dropped rapidly toward the end of the test when most of the DME had been used. The point at which no liquid DME remained is indicated in the figure. Although no liquid DME remained,

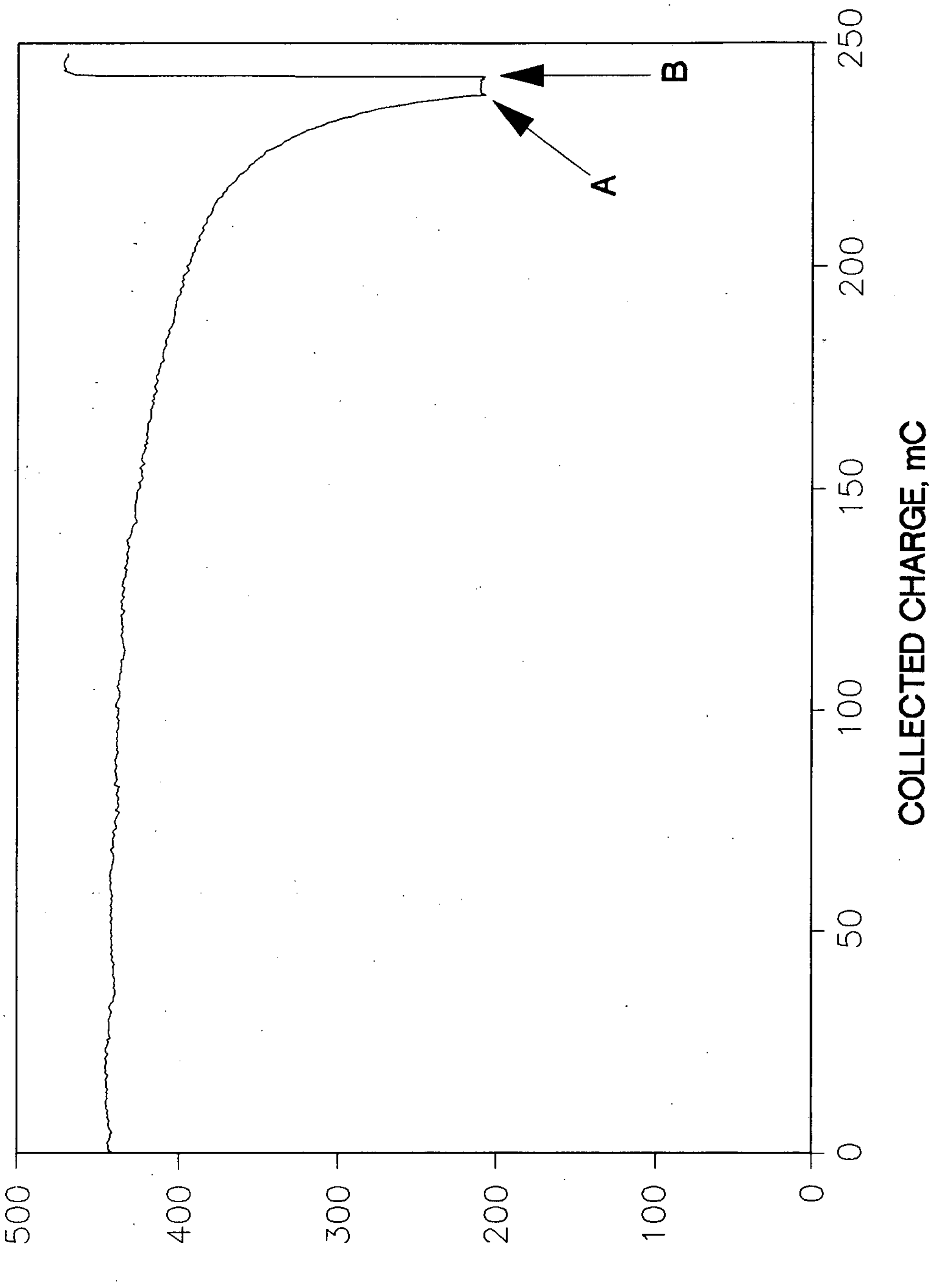

Fig. 5. Aging test in which all of the liquid DME was used. No liquid DME remained at point A. The liquid-depleted bottle was replaced with a full bottle at point B. The enrichment multiple of the Freon 11 in this test is shown in Fig. 7b.

the bottle still contained gaseous DME at its room-temperature vapor pressure, ~62 psig. Subsequent to this point, the distillation process ceased, and the gas composition and wire current were nearly stable. The gas flow rate, which is a function of the pressure in the bottle, dropped during this period, confirming that no liquid DME remained. When another bottle of DME was connected to the system, the current rose somewhat above its initial level. This was because the Freon 11 concentration in the new bottle was lower than what it had been in the previous bottle at the beginning of the test.

Both of these aging tests were performed using the same proportional tube and the same wire, and a total of 1.4 C/cm of charge was collected. ^{55}Fe pulse-height spectra taken at the end of the second test were severely degraded, however (Fig. 6), indicating the presence of some deposit on the wire. (See discussion in section 8.2.)

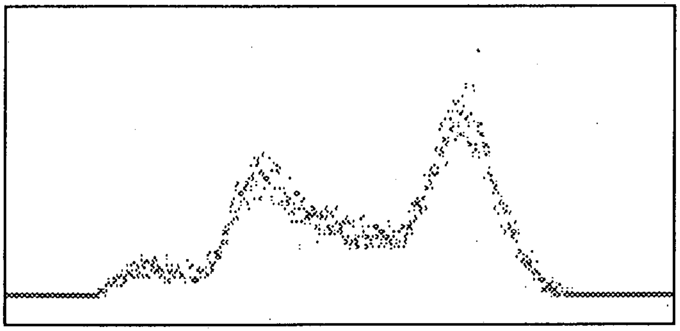

Fig. 6. ^{55}Fe pulse-height spectrum taken after two DME aging tests in which all of the liquid DME was used. The ^{55}Fe line source was perpendicular to the wire. Rather severe degradation of the spectrum is observed.

To evaluate the effect of this deposit on the gain, a bottle of DME that had been used previously and was known not to cause aging was connected to the proportional tube. The ^{55}Fe source that was used for the aging tests was moved along the wire and the variation in current drawn was observed to be only 1.5% between the coated and an uncoated region, corresponding to an overall aging rate of R~1 %/C/cm. During this test, however, an aging rate of R~400 %/C/cm was observed. A new proportional tube with a new wire was installed: the rapid aging continued. When all gas plumbing between the DME bottle and the proportional tube was replaced with new components, however, the aging rate returned to a value consistent with zero. These data suggest that some compound remained in the plumbing, but subsequently outgassed and induced rapid aging in DME.

The validity of using the Rayleigh model to describe this system was tested by comparing the predicted and the experimental values of the enrichment multiple as a function of the fraction of the liquid DME used. The enrichment multiple was determined by measuring the concentration of the Freon 11 in the gas throughout the tests. A Varian 3400 gas chromatograph equipped with a Varian model 02-001972-00 electron capture detector (Varian Instrument Group, Walnut Creek, CA) was used for this purpose. The fraction of the liquid used was taken to be equivalent to the fractional elapsed time of the test because the gas flow rate, which was monitored, remained constant throughout the test.

(Fluctuations in the flow rate were determined to be insignificantly small.)

The GC analysis was performed with a DB-5 capillary column of 30 m length, 0.25 mm inside diameter, and 0.25 μm phase thickness (J&W Scientific, Folsom, CA). To effect the desired separation, the GC temperature program consisted of a ramp to 50°C at 10°C/min after an initial three-minute hold at -30°C. A helium carrier gas pressure of 15 psig was used. The injector and detector were kept isothermal at 100°C and 200°C, respectively. Under these conditions, Freon 11 eluted in 4.7±0.02 min. The analyzed DME samples (10 μl each) were withdrawn from the downstream side of the proportional tube.

Some deviation from the Rayleigh prediction can occur as consequence of the nonzero mass of vapor in the DME bottle, which is not accounted for in the model. Only a small fraction of this vapor (that near the vapor/liquid interface) is in equilibrium with the liquid in the bottle; the remainder is less enriched in Freon 11 because it was produced earlier, from liquid that was less enriched in Freon 11. Therefore, the concentration of Freon 11 in the vapor leaving the bottle, which is what we measured, is always lower than the prediction, and the Rayleigh prediction consequently represents an upper limit on the Freon concentration.

A lower limit on the Freon 11 concentration can be estimated from the time required for vapor-phase concentration changes to propagate from the liquid surface to the point at which samples are withdrawn for analysis. This time is given by the ratio of the corresponding vapor volume to the volumetric gas flow rate. The propagation delay is most conveniently expressed as an offset in the fraction of the liquid used. This limiting offset is numerically equal to the ratio of the propagation time to the total elasped time of the test, and a line representing the lower limit on the Freon concentration can therefore be plotted by offsetting the Rayleigh prediction by this ratio (Fig. 7). This represents a lower limit because any diffusion in or mixing of the vapor will reduce the effective propagation time. In addition, when all of the liquid is used, mixing of the vapor in the bottle dilutes more-highly-enriched vapors, thereby causing the Freon 11 concentration to remain finite, a behavior not predicted by the model.

As shown in Fig. 7, there is good agreement between prediction and experiment. Within the experimental uncertainty, the data lie between the limiting cases, indicating that the Rayleigh distillation model accurately describes the vaporization of DME in a gas bottle.

While analyzing the DME for its Freon 11 content during these tests, it was observed in the chromatograms that several species were present in addition to Freon 11, and that the concentrations of all species increased as the liquid DME was used. Further, as the liquid DME was used, progressively larger numbers of distinct species were observed in the chromatograms. These species have not been identified.

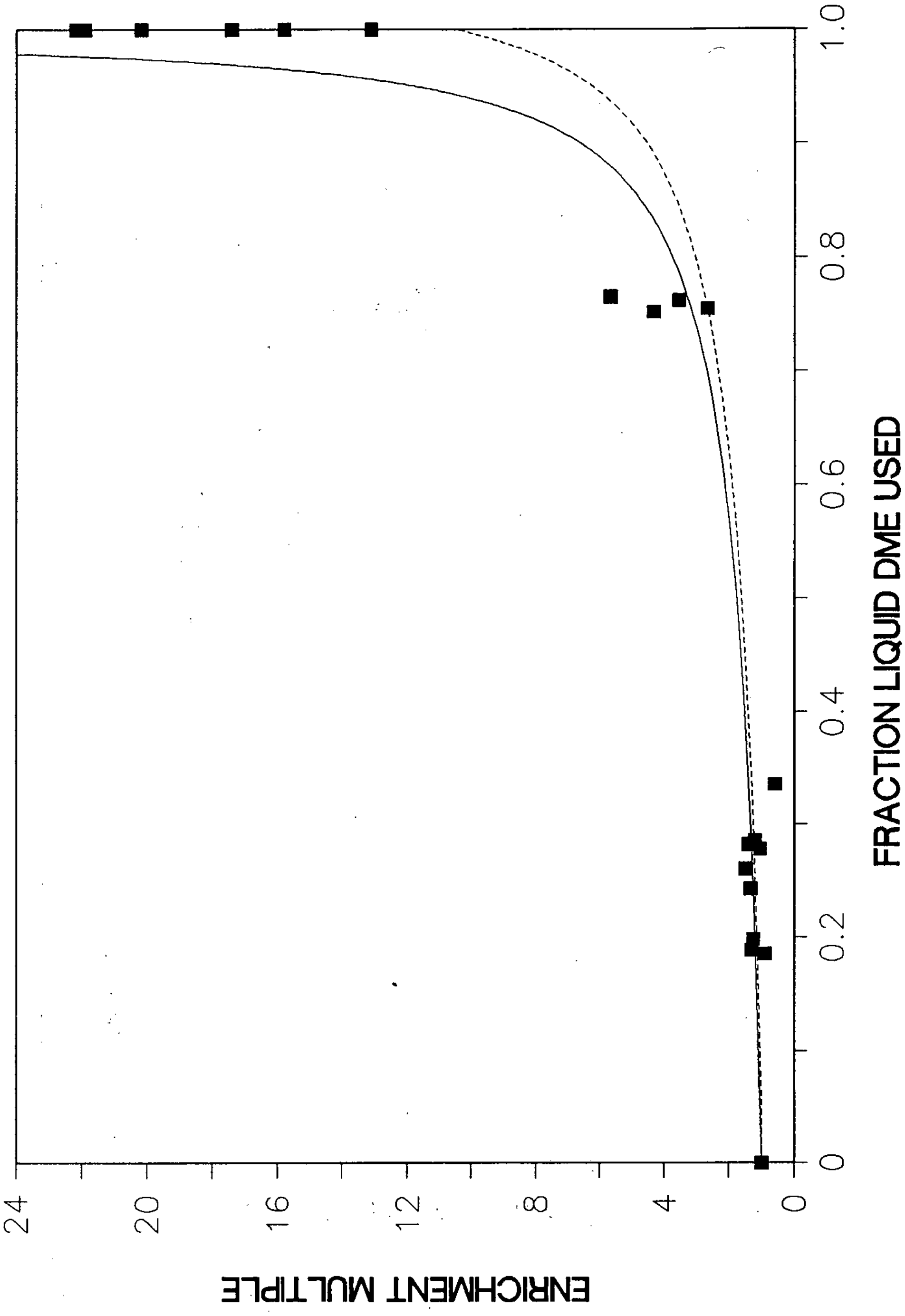

Fig. 7a. Predicted and experimental (■) enrichment multiples of Freon 11 in DME. The solid line represents the Rayleigh prediction; the dashed line represents an offset of 0.058 (= 5 hrs propagation time/86.6 hrs total time). The initial amount of DME in this test was 255 g; 60 g of gas remained after all of the liquid had vaporized. The initial gas-phase Freon 11 concentration was ~0.2 ppm.

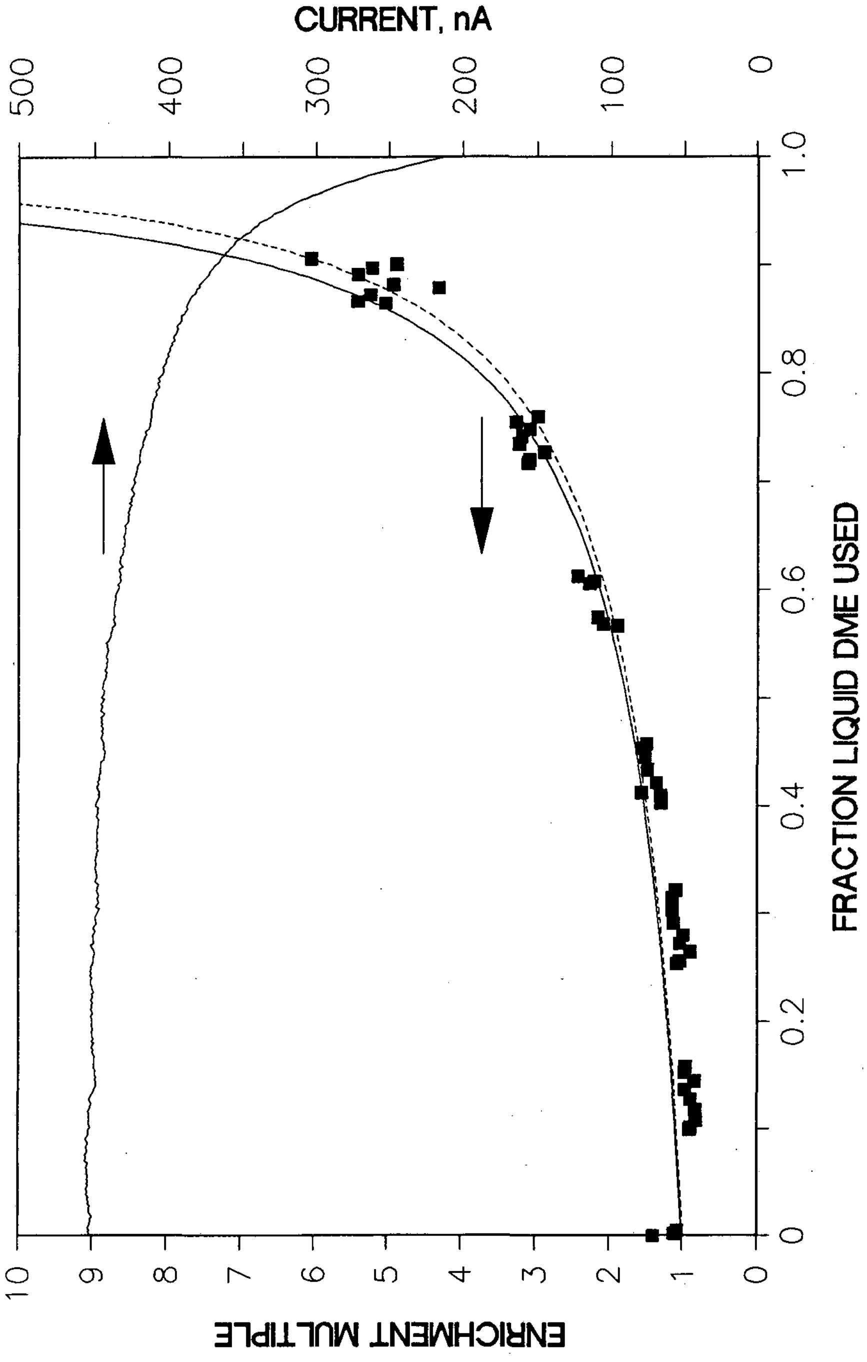

Fig. 7b. Predicted and experimental (▪) enrichment multiples of Freon 11 in DME. The solid line represents the Rayleigh prediction; the dashed line represents an offset of 0.018 (= 2.9 hrs propagation time/159.6 hrs total time). The initial amount of DME in this test was 670 g; 60 g of gas remained after all of the liquid had vaporized. The initial gas-phase Freon 11 concentration was ~0.04 ppm. The wire current as a function of the fraction of liquid used is also shown.

8. Discussion

8.1. Aging Results

We noted that increases in the concentration of Freon 11 can give the appearance of aging due to electron attachment. Because the concentration of Freon 11 in DME can increase as a result of Rayleigh distillation, it is important to know the initial Freon 11 concentration: the effect of electron attachment may be evident very early (if the initial Freon 11 concentration is large) or it may not be evident until nearly all of the liquid has been used (if the initial Freon 11 concentration is small). By using an appropriately small fraction of the DME in the supply bottle, however, the variation in the Freon 11 concentration, and hence the variation in wire currents, can be kept within any desired limit.

We also noted that rapid aging was observed in DME after using all of the liquid in two bottles. It has not yet been possible to identify the compound (or compounds) responsible for this aging, but we believe it to be less volatile than Freon 11, as (1) it did not appear until most, if not all, of the liquid DME had been used, and (2) it appeared to remain in the plumbing, unlike Freon 11, which is quite volatile at room temperature. It is worth noting that both DME bottles used in these tests had been used in previous aging tests in which little aging was observed [3]. Assuming that the compound inducing the aging originated in the DME bottle, which we believe to be the case, it must have been present in the earlier tests, but at too low a concentration to have had an effect. This would be further reason to use only a fraction of the DME in a given supply bottle.

8.2. Degradation of Energy Resolution

We noted that some degradation of the ^{55}Fe pulse-height spectrum was observed in the DME aging tests in which Freon 11 was added. Although this degradation indicates the presence of deposits on the wire, we believe it to be of relatively minor significance because the pulse-height spectrum is a far more sensitive indicator of wire deposits than is the change in gain. Indeed, the severe degradation of the pulse-height spectrum shown in Fig. 6, corresponded to only a 1.5% change in gain after 1.4 C/cm of charge was collected.

We expect that the degree to which the pulse-height spectrum is degraded may depend on both the concentration of the species inducing the aging and the amount of charge collected. If we take the product of these quantities as a measure of the amount of degradation expected, we find that much more degradation is expected for the wire exposed to the 5 ppm of Freon 11 (Fig. 3) than for the wire from the tests in which all of the liquid in two DME bottles of DME was used (Fig. 6):

Tests with direct addition of Freon 11 using the permeation device:

(0.45 C/cm)*(5 ppm Freon 11 concentration) = 2.25.

Tests in which all liquid DME was used:

(1.4 C/cm)*(0.3 ppm average Freon 11 concentration) = 0.42.

That the observed degradation of the spectra is actually much less in the case of the direct Freon 11 addition, rather than much more, as the above estimate predicts, can be taken as an indication that some species other than Freon 11 induces aging in DME. As previously noted, many species, any of which could potentially induce aging, were detected in the DME.

We note, however, that energy resolution is of major importance only when using energy-loss (dE/dx) measurements for particle identification, and it is expected that this technique will not be very useful at the SSC because the particles of interest will generally have very high energies, outside the range of this technique. For particle tracking only, degradation of energy resolution, as distinct from loss of gain, is not a significant issue or limitation.

8.3. Rayleigh Distillation

As a result of Rayleigh distillation, concentration changes of trace species will occur whenever there is a liquid phase in the gas bottle. Indeed, it was noted in aging tests with propane, which has a liquid phase in the gas bottle, that the gas purity varied not only from bottle to bottle, but also within a bottle as it was used [13]. The concentrations of species less volatile than the main gas will increase while those of species more volatile than the main gas will decrease. This being so, the potential effects of changes in gas purity on wire chamber operation must be considered.

The degree to which the heavier component in a binary mixture will be concentrated as a result of Rayleigh distillation depends primarily on the relative volatility of the two components: the closer this value is to unity, the less noticeable is the concentration effect. For example, the apparent loss of gain cause by Freon 11 will be more severe in propane ($\alpha_{\text{propane-Freon 11}} \approx 9.4$ [10]) and less severe in isobutane ($\alpha_{\text{isobutane-Freon 11}} \approx 3.8$ [10]) than in DME ($\alpha_{\text{DME-Freon 11}} \approx 5.6$).

As we have shown, increases in the concentration of highly electronegative species can give the appearance of aging. This work investigated only the effects of Freon 11, but the possibility exists that other electronegative species are of the correct volatility to give the same effect. As suggested above, it is also possible that species that induce wire degradation (as contrasted with effects of electronegativity) may be concentrated so that their effect is noticeable. An alternative, less-plausible, explanation is that certain species that inhibit aging are depleted because they are more volatile than the main gas. In any case, concentration changes caused by Rayleigh distillation complicate the interpretation of aging results. The concentration changes and their consequences can be minimized, however, by using a suitably small fraction of the liquid initially in the gas bottle.

9. Conclusions

We have found no evidence that Freons in general, and Freon 11 in particular, affect the rate of gain decrease in either argon/ethane or dimethyl ether. Aging tests with both gases in which controlled amounts of Freons were added showed no gain loss in excess of that expected without the Freons. Degradation of energy resolution, as characterized by the ^{55}Fe pulse-height spectrum, seems to be accelerated by the presence of Freon 11, however.

The large electronegativity of Freon 11 causes signal loss by attachment of primary electrons and so can give the appearance of gain loss if its concentration increases. We showed by experiment and simulation that apparent aging observed while using dimethyl ether can be explained by the natural increase in the concentration of Freon 11 that occurs in a Rayleigh distillation process. Large changes in the concentration of Freon 11 in DME and the consequent changes in wire currents can be avoided by not using all of the liquid DME in a given supply bottle.

Concentration changes of trace species are expected to occur whenever there is a liquid phase in a gas bottle. It is therefore prudent not to use all of the liquid in such cases regardless of the type of gas.

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