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J. Va'vra

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REVIEW OF WIRE CHAMBER AGING*

J. VA'VRA

*Stanford Linear Accelerator Center**Stanford University, Stanford, California 94305*

Abstract

This paper makes an overview of the wire chamber aging problems as a function of various chamber design parameters. It emphasizes the chemistry point of view and many examples are drawn from the plasma chemistry field as a guidance for a possible effort in the wire chamber field. The paper emphasizes the necessity of variable tuning, the importance of purity of the wire chamber environment, as well as it provides a practical list of presently known recommendations. In addition, several models of the wire chamber aging are qualitatively discussed. The paper is based on a summary talk given at the Wire Chamber Aging Workshop held at *LBL*, Berkeley on January 16-17, 1986. Presented also at Wire Chamber Conference, Vienna, February 25-28, 1986.

1. Introduction

The wire chamber aging problem is clearly very complex. A few years ago it seemed that it would never yield to rational analysis. The main difficulty was that each experiment or test had very specific conditions and it was very difficult to extrapolate given information to other circumstances. This is still basically true, but nevertheless, it appears that a pattern is emerging and one might even be able to draw some qualitative conclusions. However, a true quantitative understanding of the polymerization process and its control is a long way off. For that we would have to know:

- (a) All cross-sections for collisions of electrons and photons with all species involved, *i.e.*, atoms, ions, molecules, radicals, ionized radicals, *etc.*
- (b) All cross-sections for atom-atom, atom-molecule, atom-radical, radical-radical interactions, *i.e.*, the probabilities for all chemical processes involved.
- (c) All this as a function of electron and photon energy, pressure, temperature, flow rate, chamber design, extremely high electric field ($\sim 200\text{--}300$ kV/cm.atm), *etc.*

Before we can start writing a complex Monte Carlo program simulating the avalanche and its polymerizing effects, we had better know all these parameters. Realizing these difficulties, it might be beneficial to learn as much as possible about the polymerization process from other branches of science. In fact, there does exist a branch of chemistry, called plasma chemistry, which deals empirically with the problem of plasma polymerization and polymer etching. Clearly, we should learn as much as possible about its methods and insights.

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In the traditional polymerization process in chemistry, a necessary prerequisite to form polymers is to start with molecules containing double bonds between two carbon atoms. However, the polymerization in the plasma environment is much more complex.^[1,2] Although the process is complex, it is not hopelessly random or unstable, *i.e.*, one can tune the variables.

In this talk I will concentrate on the following subjects.

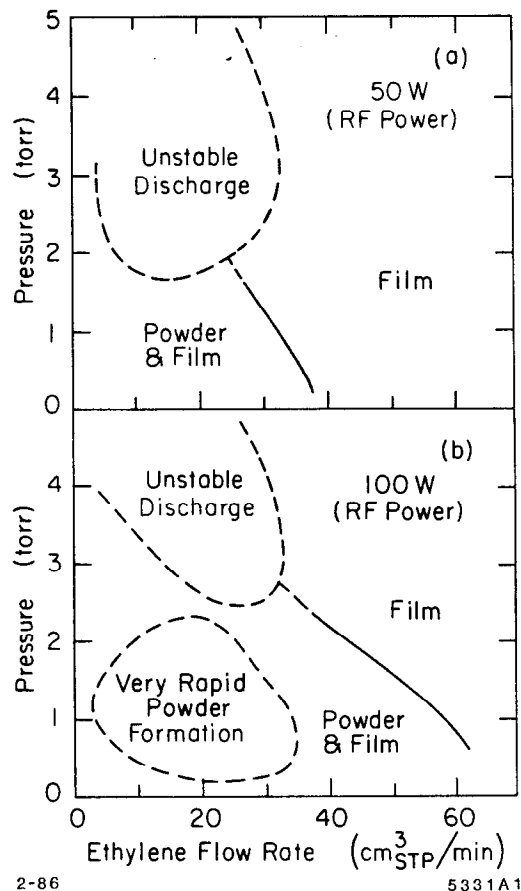
1. The importance of tuning variables.
2. A naive model of electrode coating.
3. Are present plasma chemistry studies in the same range of some basic parameters as we have in our avalanches?
4. Some of the design parameters for building chambers. Is anything known about their effect on aging?
5. Tests of aging in small accelerated tests.
6. A "cook book" of hints and suggestions.

2. Importance of Tuning of Variables

Plasma chemistry is a branch of science which deals with very complex reactions on a qualitative level of understanding and basically tunes a multi-variable process empirically. Typical variables are the choice of polymerizing gas, pressure, gas flow, *rf*-power density, impurity content, *etc.*, (*i.e.*, the variables which we typically also have to control). A typical example of such empirical tuning is shown in Fig. 1, the plasma polymerization of ethylene under various conditions. One can create film, powder or oil depending on operating conditions. Both the powder and the film are insoluble in common organic solvents, indicating a high degree of cross-linking of molecular chains. The oily products, on the other hand, are soluble in both acetone and xylene. (The possible structure of the molecular chains generated in a plasma condition can be seen on Fig. 2.)

These observations are not dissimilar to what many people have observed in our field, with one basic exception. In our field (wire chambers) we tend to choose the operating conditions rather arbitrarily from the aging point of view. In fact, one should always tune the additives level, the gas flow, the gain on the wires, tolerable level of impurities, *etc.* To illustrate this we cite as an example adding 1% of hydrogen to a gas mixture to improve aging. First of all, 1% is completely arbitrary. What we have to do is to tune the hydrogen content for the gas mixture, gain, gas flow, *etc.* If we change any of these variables we have to retune the hydrogen content. In comparison, the detailed understanding of the anatomy of the polymerized chains (Fig. 2) is perhaps not that important and should be left to specialists.

Fig. 1. Examples from plasma chemistry. Polymerization of ethylene and importance of tuning power, pressure and gas flow on a final appearance of polymerized species.^[51]



3. Naive Model of Electrode Coating

The avalanche can clearly produce a large variety of molecular species. However, as one can see in Table 1, to break a typical covalent molecular bond takes some 2–5 times less energy than to ionize the same molecule. When an electron or photon breaks a covalent bond, a radical molecule is formed. Such a molecule has no net electrical charge, but it typically has a large dipole moment, because it is frequently distorted. Because of a very high electric field near the anode, these radical molecules will be attracted towards the anode surface. They are chemically very active and will either recombine back to original molecules or form new molecular species.^[1,2] The concentration of these radicals in a typical avalanche is not known. To get a true distribution, the analysis of dissociated molecules would have to be done immediately during and inside the avalanche (of course, impossible). If we do it outside the chamber, many radicals are probably recombined by then. However, under plasma chemistry conditions, the typical concentrations of free radicals in the plasma is usually 5-6 orders of magnitude higher than that of ions.^[3]

We will make a simplifying working assumption, that the polymerization rate is proportional to the density of radical molecules. The density of radicals is in turn proportional to the density of electrons (and photons) in the avalanche and their energy distribution.

Unfortunately, we do not know how to calculate the density of electrons and their

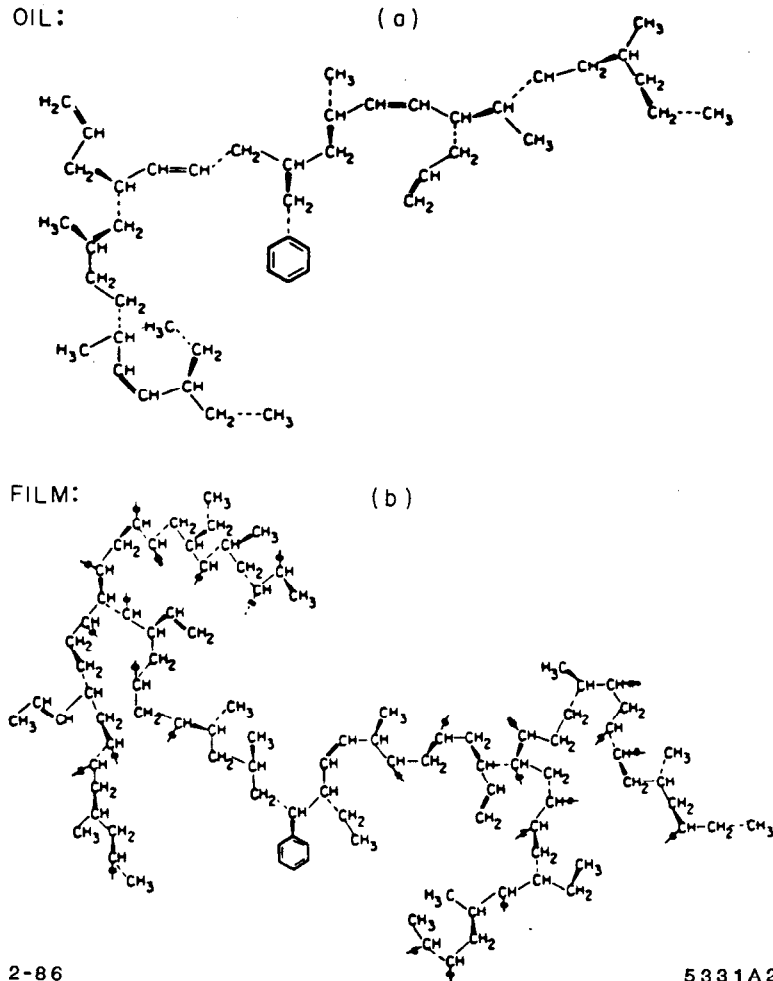


Fig. 2. A predicted structure of polyethylene produced in the plasma environment: a) oily product, b) film coating.^[52]

energy distribution. Recently, there has been an attempt to simulate an avalanche in Argon only^[4] The authors considered electron-Argon elastic, excitation and ionization cross-sections with a proper electron motion between the collisions and a proper treatment of energy balance and direction change after each collision. However, they neglected the photon emission, the photo-ionization processes and the effect of space charge within the avalanche. Nevertheless, as one can see in Fig. 3, they solved the problem in three dimensions and they predicted the electron density and energy distribution. We will use these predictions in the next section.

Figure 4 shows schematically a formation of polymers on the anode surface as caused by a large concentration of free radicals. Initially the polymer is held to the surface very weakly unless some additional chemical reaction takes place between the polymer atoms and atoms of the material of the wire, or unless the polymer completely surrounds the anode surface. That would mean that the polymers or free radicals can be blown away from the anode surface by a large gas flow. If a large polymerized macroscopic piece

TABLE 1 [74]

(1) Bond Energies of Typical Covalent Bonds:

$C - H$	4.3 eV	$C = C$	6.4 eV
$C - C$	3.6	$C = N$	9.3
$C - O$	3.7	$C = O$	7.8
$C - N$	3.2	$N \equiv N$	9.7
$N - H$	4.0	$C \equiv C$	8.6
$O - H$	4.8		
$O - O$	1.5		
$F - H$	5.8		
$F - C$	5.4		
$C - Cl$	3.5		

$\left[CO_2 \right] \rightleftharpoons \left[O = C = O \right]$
 is very stable

(2) Dissociation Energy and Ionization Energy for Some Gases:

	<u>Dissociation Energy</u>	<u>Ionization Energy</u>
Ar	-	15.8 eV
Xe	-	12.1
H_2	4.5 eV	15.6
N_2	9.7	15.5
O_2	5.1	12.5
Ethanol	≥ 3.2	10.5
Iso-Propanol	≥ 3.2	10.2
DME	≥ 3.2	9.98
C_2H_6	≥ 3.6	11.5
H_2O Vapor	4.8	12.6
Methylal	≥ 3.2	10.0
CO_2	7.8	13.8
Iso-Buthane	≥ 3.2	10.6
CH_4	4.3	12.6

receives a positive charge in a subsequent avalanche, it will slowly drift toward the cathode (it is heavy). Again a large gas flow can successfully, according to this naive picture, prevent coating of the cathode. As F. Sauli^[5] pointed out, the deposit layer on the anode probably tends to carbonize under a heavy electron bombardment (he has always found only conductive black anode deposits).

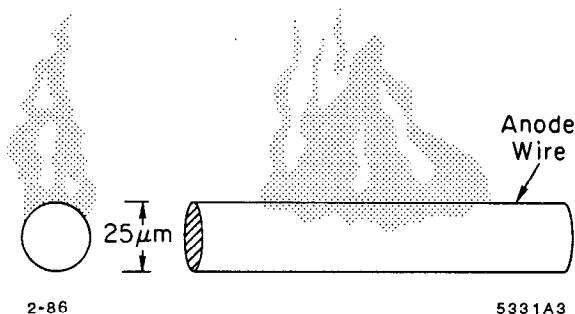


Fig. 3. Monte Carlo prediction of avalanche formation in pure argon.^[4]

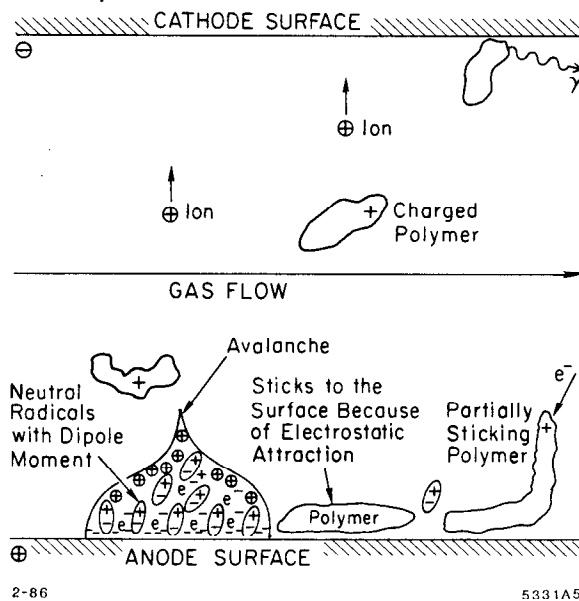


Fig. 4. A naive model of formation of polymers out of free radicals on the anode surface and an effect of gas flow on heavy positively charged species slowly drifting towards the cathode.

S. Majewski^[6] pointed out that a typical “wire pattern” image of deposits frequently found on the cathode foil^[5,6,40] is an important message any model should be able to explain. Because we argue that mainly very large molecules peel off and drift toward the cathode, we expect that the most likely flow direction will be perpendicular towards the cathode and therefore the cathode deposits will show the “wire pattern” structure.

There are, of course, a variety of other processes which could contribute to aging—see Table 2 for a summary. Notice, for instance, that above ~ 10 kV/cm one can multiply the number of electrons near the cathode surface (gain). CO_2 mixtures are known for carbon build-up, which occurs specifically at the cathode.^[33] We should mention the Malter effect^[31] which is an emission of electrons and occurs after the cathode has been coated by a thicker insulating film (oxides, polymer deposits, fingerprints^[5]). Finally, it appears that extremely thin cathode deposits or oxides can be very photosensitive,^[5,31,21,67,46] presumably due to lowering of electron work function, allowing even very low energetic photons to liberate electrons from the cathode. This may occur because the extremely thin layers of deposits or oxides may have electric dipole moment which helps to “pull” the electrons

from the metal.^[64] Clearly, if this happens, the chamber can deteriorate very quickly because of electron emission at the cathode and a subsequent positive feedback between this effect and the anode amplification. But we still believe that the primary process is polymer building at the anode according to the previously mentioned naive model.

We should also mention that the polymerization can be “primed” by a previous exposure of the wires to an atmosphere containing tiny droplets of oil and silicon dust. H. Hilke mentioned results of Christy^[65], who demonstrated that even 10^{-6} ppm (not a mistake) of silicon oil vapor can cause significant aging. These oil molecules get attached to the wires presumably due to their dipole moments. The details of manufacturing wires should also be closely examined,^[59] as well as gas processing procedures.

4. Is Plasma Chemistry in the Same Range as Some Basic Parameters We Have in Our Avalanches?

As we said there is considerable experience in plasma chemistry with the polymerization problem. The question is, are we operating in the same regime of variables? Table 3 shows this comparison, assuming anode diameters of $20 - 50 \mu\text{m}$, pressure of 1 atm, mean free path between the electron collisions, $\lambda \sim 1 \mu\text{m}$, mean free time between the collisions^[7] $\tau \sim 1$ psec, an average electron energy of about 5 eV and an effective volume according to naive pictures as shown in Fig. 5. We conclude that many parameters in the two fields are rather different. Nevertheless, in the absence of better information, we will continue to quote many examples from plasma chemistry and we will see that many are in agreement with what is observed in wire chambers.

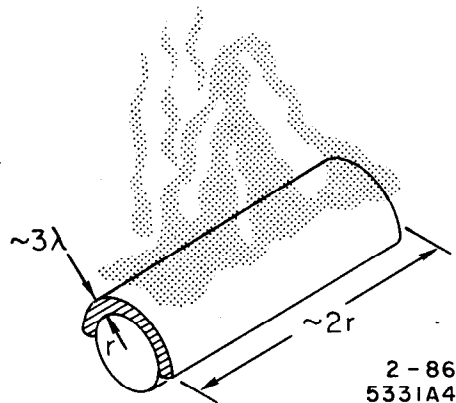


Fig. 5. A naive assumption about the avalanche effective volume needed to predict approximately effective volume of avalanche in Table 3.

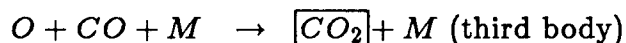
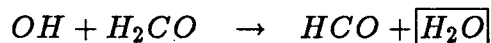
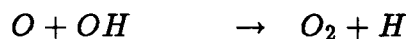
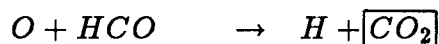
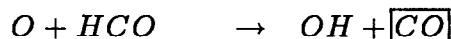
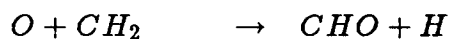
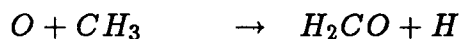
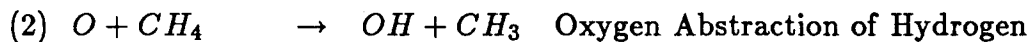
5. The Design Parameters for Building Chambers and Their Effect on Chamber Aging

In this section we will make a survey of what is known about the correlation between aging and chamber design parameters such as: gas [type (resistant or susceptible to

TABLE 3
Comparison of Basic Operating Conditions

Parameter	Plasma Chemistry [1]	Wire Chambers
Average Electron Energy	1 – 10 eV	5 – 10 eV (Ar)
Effective Volume	100 – 1000 cm ³	10 ⁻¹⁰ – 10 ⁻⁸ cm ³
Typical Electron Density	10 ⁹ – 10 ¹² e/cm ³	10 ¹⁴ – 10 ¹⁷ e/cm ³ /avalanche
Typical Power Density	0.01 – 10 watts/cm ³	10 ⁸ – 10 ¹² watts/cm ³ /avalanche
Gas Pressure	0.01 – 10 Torr	≥ 760 Torr
<i>E/p</i>	10 – 50 V/cm·Torr	100 – 400 V/cm·Torr (on the surface of the anode)
Type of Electric Field	RF	DC
Typical Gas Flow	~ 1 Gas Volume/1-10 minutes	~ 1 Gas Volume/1-8 hours

TABLE 4
*CH*₄ Gas in Oxygen Plasma [70]



polymerization), impurities (undesirable), additives (desirable), flow rate, pressure, tubing material and temperature]; anode and cathode wire material; cathode type (wire versus continuous); electric field on anode and cathode surface; anode-cathode distance; chamber body material; glues and adhesives; and gain on the anode wires.

However, the most important variables probably are (apart from the gases):

- (a) effective volume of the avalanche (cm^3),
- (b) average electron and photon energy in the avalanche (eV), and
- (c) average electron and photon density ($\#/\text{cm}^3$).

All three variables then define the average power density (Watts/cm^3). These are the important variables because they control the density of the radicals, which according to our naive model, controls the rate of polymerization. Unfortunately, unlike in the plasma chemistry investigations, we really do not know how to calculate these basic variables and this probably contributes to our general confusion.

Before we start discussing good additives and bad impurities, we will review the electronegativity concept. In a bond between two atoms, the electron will be attracted more

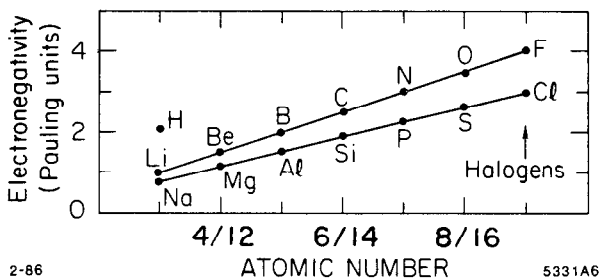


Fig. 6. Relative electronegativity of some of the more common elements.

strongly to the atom with larger electronegativity (or electron pulling power). As one can see in Fig. 6 the most electronegative atoms are halogens, however, oxygen is immediately behind fluorine in its reactivity. Notice also that carbon is similar in electronegativity to silicon. Figure 7 shows the fluorine atom has one missing electron in the valence shell and oxygen has two empty slots. As a result, the fluorine atom can make only a single bond, which as we can see in Table 1 is strong, but not as strong

as double bonds (two pairs of electrons in the covalent bond) which can be made, for instance, in oxygen reactions. This allows oxygen to make stronger species in the plasma reactions which will tend to terminate polymerization because the density of radicals is reduced. Silicon and carbon have similar valence shells (see Fig. 8) the only difference being that the silicon atom is heavier and its molecules are less volatile.

5.1 GOOD ADDITIVES

We will now discuss examples of additives which are believed to prolong chamber life.

5.1.1 Good additives - Oxygen

It is a well known fact in plasma chemistry that atomic oxygen O reacts with hydrocarbon radicals and it is generally found that the end products are CO, CO₂, H₂O and H₂. As we said earlier they are more stable molecules and they are volatile and can be blown away by a sufficient gas flow. In addition, the rate of removal of organic polymeric

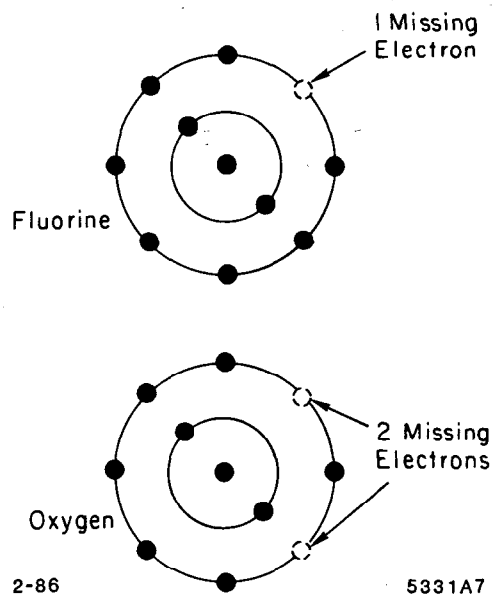


Fig. 7. A valence shell structure of fluorine and oxygen.

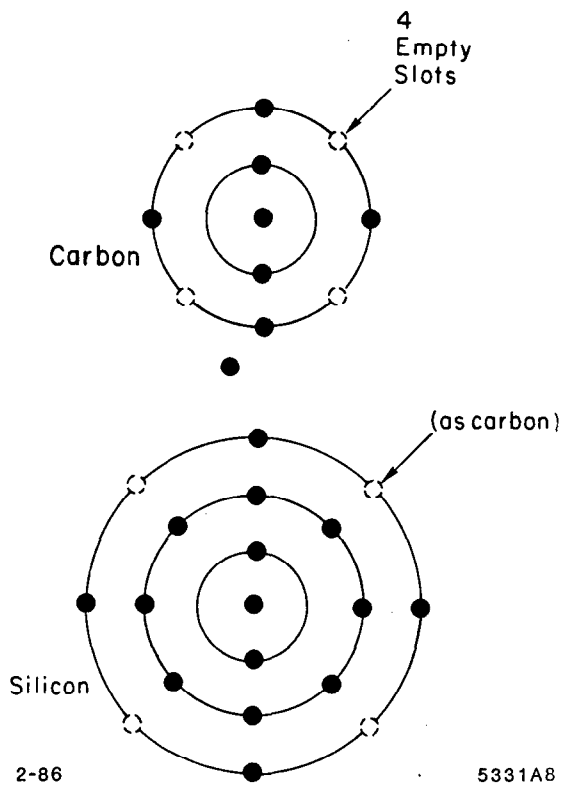


Fig. 8. A valence shell structure of carbon and silicon.

material can often be increased in an oxygen plasma by the addition^[9] of H_2O , H_2 , N_2 , NO , N_2O . These are empirical facts supported by qualitative models. Table 4 shows an example of such chain of reactions. The input is CH_4 gas and its radicals CH_3 , CH_2 , and the atomic oxygen produced in the plasma either from O_2 or from a molecule containing oxygen. Another example from plasma chemistry is the cleaning of contaminating films from mirrors in an oxygen plasma.^[8]

Yasuda^[13] in fact argues that in plasma chemistry most organic compounds with oxygen containing groups such as $-COOH$, $-CO-$, $-OCO-$, $-OH$, $-O-$, $-C=O$ are generally reluctant to form polymers in the plasma environment. This is not to say that it cannot happen. For instance $CO + H_2 + N_2$ in a discharge can form structures resembling proteins.^[14] Also, as we will see later, oxygen polymerizes with silicon to form various types of silicates (for instance a quartz).

In our field, I would like to mention two examples. The first one comes from the MARK II drift chamber,^[10] which ran for a year with 0.6% oxygen added to 50% Ar + 50% C_2H_6 gas. The oxygen is believed to stabilize the operation. The second example comes from Turala's talk,^[11] where he mentioned that excellent lifetimes were achieved with Ar + 8% N_2 mixture (~ 10 C/cm).

It is tempting to suggest that we can clean the wires if we simply run humid air in the chamber at large enough voltage so that there is a chance of producing atomic oxygen.

However, this may be practical only for chambers designed for high pressure operation, because typical feedthroughs would fail long before there was a gain on the wires. Another important point is that the gas flow has to be adequate, as one can see on Fig. 9, where nylon is removed in the oxygen plasma. Notice also that power is an important variable.

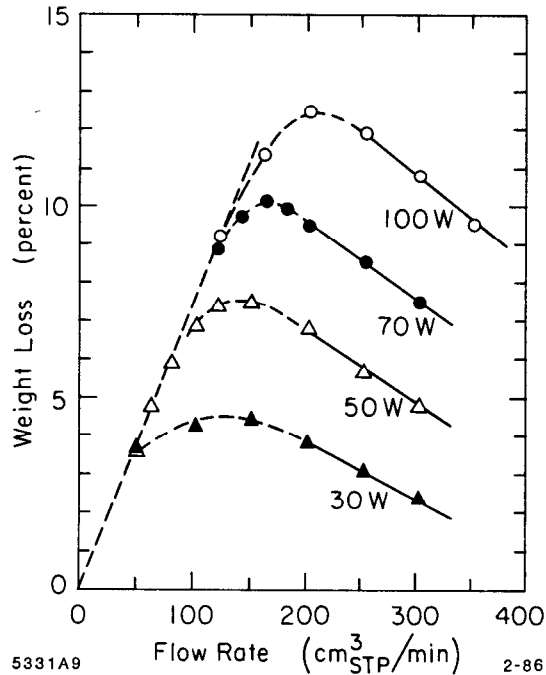
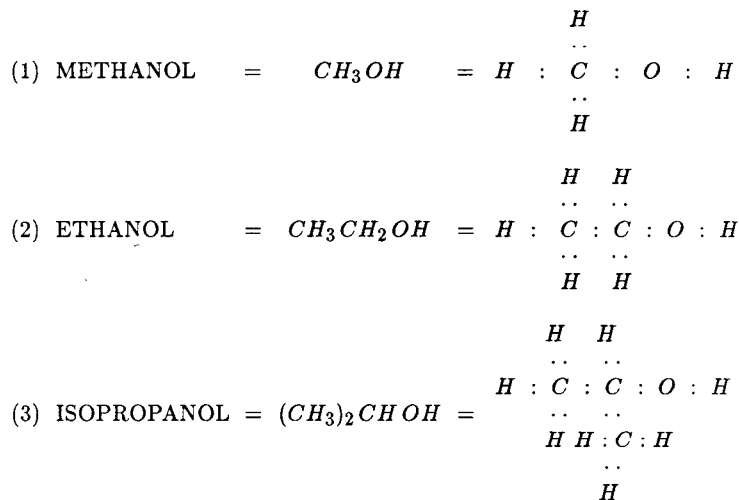


Fig. 9. Etching of nylon in an oxygen plasma under various conditions of flow rate and power in a plasma chemistry environment.^[71]

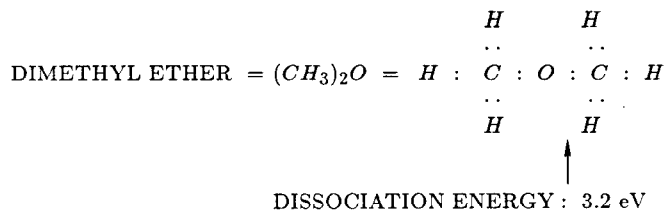
5.1.2 Good additives – H₂O, alcohols, ethers, methylal

There is more than one mechanism by which these additives help aging. Figure 10 and Table 5 show the molecular structure of these additives. As we can see they all have large dipole moment and as a result they tend to have large cross-sections for electron-molecule scattering, *i.e.*, the mean value of electron energy will get smaller in the avalanche, which is an important factor. As M. Atac pointed out a long time ago,^[12] they also have large cross-sections for absorption of UV photons. Because of the large dipole moment, these molecules will tend to concentrate near the anode and cathode surfaces. As we can see in Table 1, the ionization potential of these additives is lower than the ionization potential of typical hydrocarbon molecules (with the exception of water). Therefore, charge exchange is possible which will tend to protect the hydrocarbon molecule during its ionic neutralization at the cathode.^[5] Finally, if these molecules of the additives are broken, the electronegativity of oxygen will tend to repair them immediately, or, as we discussed in the previous section, the final oxygen products will tend to be volatile.

ALCOHOLS:



ETHERS:



METHYLAL:

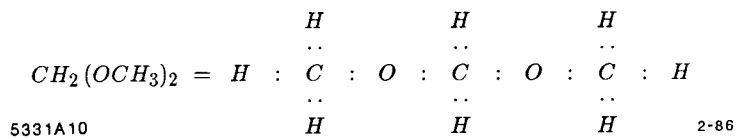


Fig. 10. A schematic molecular structure of some alcohols, dimethyl ether and methylal.

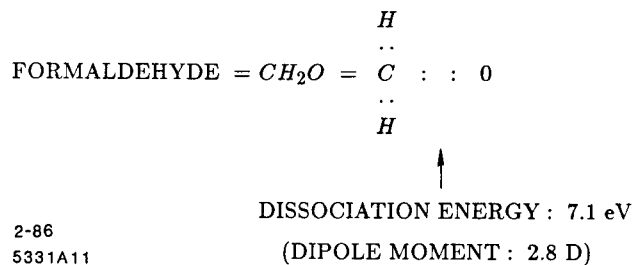


Fig. 11. A schematic molecular structure of formaldehyde.

TABLE 5

Additive	Molecular Structure	Dipole Moment
Water	$H - O - H$	1.85 D *
Alcohols	$R - O - H$ ($R = CH_3, CH_3CH_2, \text{etc.}$)	1.66 - 1.7 D
Ethers	$R - O - R'$ (DME = $R = R' = CH_3$)	1.29 D
Methylal	$R - O - R' - O - R$ ($R = CH_3, R' = CH_2$)	

* D = Debye units

TABLE 6

Examples of Industrial Use of Halogens

Ethyl Chloride	CH_3CH_2Cl	Gasoline Production
Vinyl Chloride	$H_2C = CHCl$	Raincoat Production
Tetrachloroethylene	$Cl_2C = Cl_2$	Cleaning of Clothing
Tetrafluoroethylene	$F_2C = CF_2$	Teflon Production, Plasma Etching
Methylchloride	CH_2Cl_2	Paints and Varnish
Freon 11	$FCCl_3$	Refrigerant
Freon 12	Cl_2CF_2	Refrigerant
Freon 13-B1	CF_3Br	

Nevertheless, these molecules are fragile. For instance, the dimethyl ether molecule can be destroyed with only 3.2 eV! One can destroy these additive molecules successively to produce, for instance, formaldehyde (see Fig. 11) which can easily polymerize to produce paraformaldehyde $(\text{CH}_2\text{O})_n$. But still, the rate of this process tends to be lower than the polymerization rate of ordinary hydrocarbon molecules and therefore the addition of H_2O , alcohols, dimethylether, and methylal can be considered beneficial. As one can see in Table 10 (under "Conclusions") many of the experiments which reached at least 0.1 C/cm radiation dose, are using one or a combination of these additives. In addition, alcohols, DME, and methylal are used in many tests and experiments currently underway.^[27,48,54] (see also Table 11).

Water may have an additional value, *i.e.*, it tends to increase conductivity of deposits which are otherwise poorly conductive and as a result will prolong chamber lifetime.^[15,16,17,18,43,45,50,53]

5.1.3 Good additives - Hydrogen

It is believed that the addition of hydrogen would help to restore radicals to their original molecular form. For instance CH_2 radicals would get restored to CH_4 . The final outcome of this operation will depend on the competition of the polymerization rate of radicals with the rate of restoring them to the original molecular form by the hydrogen. As we said earlier, one should tune this balance. Because this is not generally done in our field, results of adding hydrogen are confusing. For instance, H. Sipilä and M. Järvinen^[19] have improved the life of a permanently sealed counter operating at very low gain (200) approximately twenty times by adding 0.1% H_2 to 90% Ar + 10% CH_4 . However, one should say that permanently sealed counters have very poor lifetimes nominally ($\sim 10^{-3}$ C/cm). Since 1983, H_2 has been used as an additive in commercial permanently sealed proportional counters to increase their lifetimes when hydrocarbons are used as a quenching gas.^[20] However, others tried hydrogen (usually 1%) and generally there was no observed improvement.^[18,21,22,23] Perhaps the problem is that tuning of hydrogen content was not attempted. The halogen impurities might also be a significant problem because they will react with the hydrogen in plasma and nullify its effect.

5.2 BAD IMPURITIES

In this section we will discuss several examples of undesirable impurities which generally accelerate chamber aging. An examination of analysis of deposits^[2,5,6,11,18,25,26,27,55,56] reveals that the *dominating* species usually found are carbon, carbon based polymers, silicon based species, halogen and sulphur elements,^[66] or their combination. It depends on a particular experimental setup whether a given element is found. For instance, F. Villa pointed out that most of the elements found on the wire could also be seen in the analysis of various materials used for building the chamber (delrin, O-rings, *etc.*).^[25]

5.2.1 Bad impurities - Halogens

Why should we worry about halogen contamination? Table 6 indicates that they are in widespread industrial use and therefore there is a good chance that our bottles have

been contaminated. This contamination level might be regionally dependent; in the San Francisco Bay Area it appears to be a problem.

As we discussed earlier the halogens are very reactive in the atomic form, because of their high electronegativity—see Fig. 6. In addition, they can form only single bonds (one electron pair), which are generally weaker than the double bonds. Finally, for instance C-Cl, and C-Br bonds are weaker than C-H bonds. Therefore, as we can see in Table 7 it takes more energy to dissociate, say CH₄ into free radicals than the halogenated hydrocarbons such as CF₂Cl₂, CH₃Cl, C₂H₃Cl, C₂H₃F, etc. This would mean, according to our earlier naive model, some halogenated hydrocarbons create a higher density of free radicals in the plasma and will have a greater tendency to form polymers. This is in fact observed in plasma chemistry.^[24] Figure 12 shows that in the plasma chemistry environment a small addition of halogenated hydrocarbons can vastly increase the polymerization rate of CH₄, C₂H₂ and C₂H₆ gases.

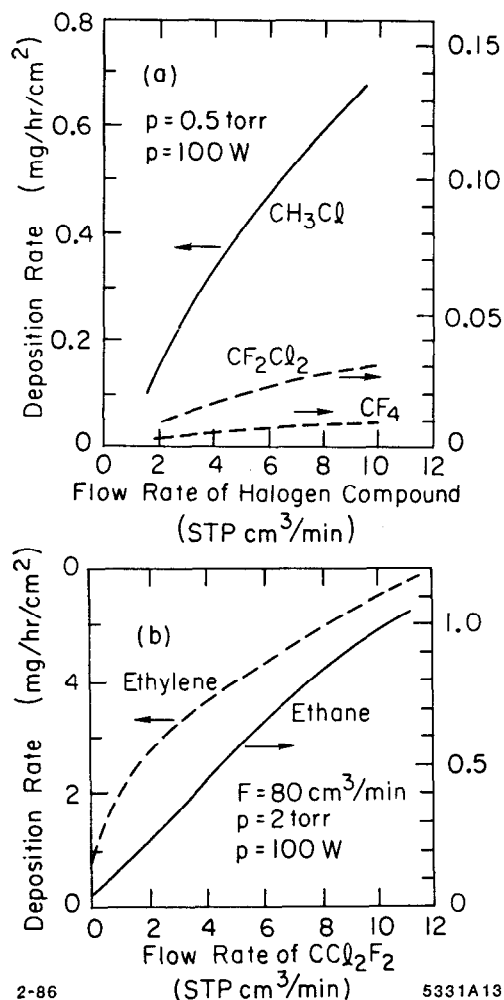


Fig. 12. An influence of halogens on the polymerization rate of methane, acetylene and ethane in a plasma chemistry environment.^[72]

TABLE 7

Energy Needed to Destroy Some Molecules [69]

(a)	$e^- + CH_4 \rightarrow CH_2 + H_2 + e^-$	4.6 eV
	$\rightarrow CH_3 + H + e^-$	4.5 eV
versus		
(b)	$e^- + CF_4 \rightarrow \boxed{CF_2} + F_2$	7.8 eV
	$e^- + CF_4 \rightarrow CF_3 + F$	5.2 eV
	$e^- + CF_2Cl_2 \rightarrow \boxed{CF_2} + Cl_2 + e^-$	3.2 eV
	$e^- + CH_3Cl \rightarrow \boxed{CH_2} + HCl + e^-$	3.9 eV
	$e^- + C_2H_3Cl \rightarrow \boxed{C_2H_2} + HCl + e^-$	1.1 eV
	$e^- + C_2H_3F \rightarrow \boxed{C_2H_2} + HF + e^-$	0.8 eV

TABLE 8

Wire Aging for Different Cathode Structures [32]

Type of Cathode	Lifetime Limit
(1) ϕ 100 μ m Cu-Be Wires	$\sim 10^6/\text{cm}^2$
(2) Stainless Steel Mesh	$\sim 10^7 - 10^8/\text{cm}^2$
(3) Continuous Sheet	$\sim 10^{10}/\text{cm}^2$
(4) As (3), But Add Methylal	$\sim 3 - 5 \times 10^{10}/\text{cm}^2$
(5) As (1), But Add Isopropyl Alcohol (1%)	$\sim 3 \times 10^{10}/\text{cm}^2$
(6) As (1), But Add 4% Methylal	$\sim 3 \times 10^{10}/\text{cm}^2$

However, not all halogens are as fragile. For instance, CF_4 is more stable (bond strength is 5.2 eV — see Table 7) and as one can see in Fig. 12, it has a lower rate of polymerization. Apparently just a few eV in the bond strength makes a large difference in the polymerization rate. In fact, it has been used in wire chambers.^[60] It also has a wide industrial use etching metals in the plasma environment.

In our field I would like to quote J. Kadyk's measurement with *DME* gas.^[23] For no halogen contamination, he measured a gain drop of $\sim 10\%/C.cm.$ A gas bottle containing 10–20 ppm of Freon 11 gave a result more than fifty times worse. In addition, the preliminary results from a test with 2 and 4 ppm of Freon 11 contamination (specified by manufacturer) were 10–20 times worse compared to the reference (“pure”) bottle. Does this mean that we have to worry about 1 ppm contamination level? At that level we probably have many other types of halogens present! F. Villa^[25] and H. Spinka^[26] have also reported a good efficiency of transfer of halogen impurities on the chamber electrodes.

5.2.2 Bad impurities – Silicon

Silicon is the most abundant element on earth. It is present in gas bottles, in a form of a fine dust or gas impurities (silane SiH_4 , tetrafluorosilane SiF_4 , etc.). In addition, there are many products containing silicon (G-10, RTV, various oils, molecular sieves,^[57] etc.).

As we discussed earlier, Fig. 8 shows that the valence shell of silicon is the same as that of carbon. One therefore expects some similar features. Silicon can polymerize both

with hydrocarbons and oxygen to form a polysilicone $\left[\begin{array}{c} R \\ -Si - O- \\ R \end{array} \right]_n$, ($R = \text{CH}_3$) and a silicate $\left[\begin{array}{c} O \\ -Si - O- \\ O \end{array} \right]_n$. As discussed earlier, many products of oxygen and hydrocarbon

radicals are volatile and can be removed by a gas flow. This is not so much the case with the silicon species because they are heavier. In this sense, the silicon contamination is probably a more serious problem.

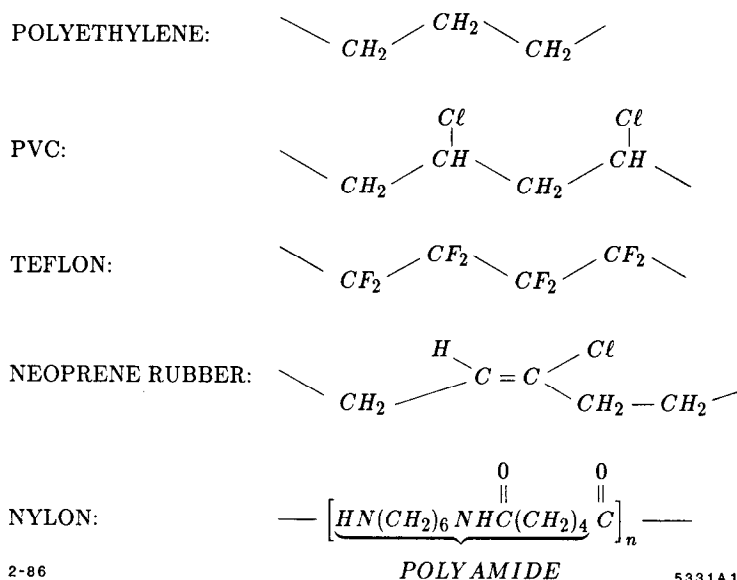
Many people reported problems with silicon based deposits—H. Hilke^[18], H. Spinka^[26], etc. As in case of the halogen contamination, the electrode coating by silicone products is very efficient. F. Villa suggested that this point could be utilized by removing them in a primary recirculation loop. D. Hess pointed out that the CF_4 gas could be used to remove silicon based products (to create, for instance, a volatile SiF_4) similar to the way oxygen might remove hydrocarbon based deposits. Certainly good filters should be used to prevent dust from getting into the active volumes of the chambers.^[58,28]

5.3 GAS TUBING

Figure 13 shows a polymer structure of several gas tubing materials. One can see that *PVC*, teflon and neoprene rubber contain halogen atoms in the molecular chains. It is conceivable that some parts of these chains will be broken off and they will contaminate the active volume of the wire chamber, *i.e.*, we will get halogen contamination. This may cause problems as discussed in Section 5.2.1. Kotthaus^[27] has measured the effect of *PVC*

tubing on the rate of polymerization. As one can see in Fig. 14, the introduction of PVC tubing initiated aging and this continued even after the tubing was replaced with the original stainless steel tubing. This indicates a potentially serious problem. Once aging is initiated, it will continue even if we introduce correct conditions to prevent aging. There has been good experience with *RILSAN* tubing which is a form of nylon.^[38] As we can see in Fig. 13, the molecular chain of this material doesn't contain halogens. Copper tubing can trap oxygen, react with some gas mixtures, and one should be aware that silicon oils^[6] are frequently used during a drawing process. Clearly, the safest way is to use stainless steel tubing.

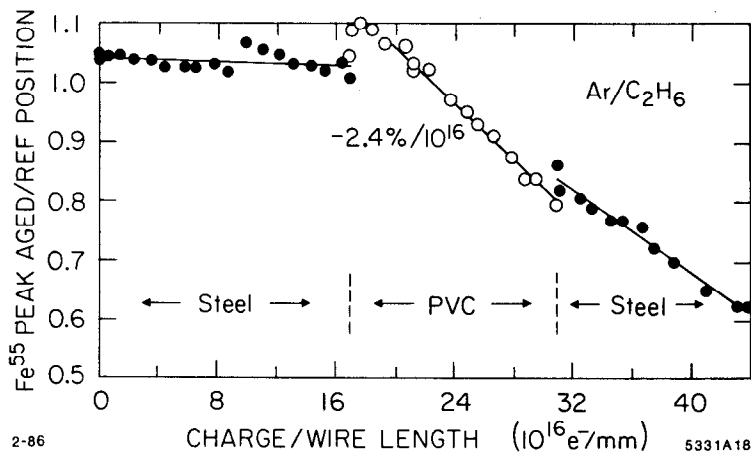
GAS TUBING



2-86

5331A14

Fig. 13. Examples of chemical composition of some typical gas testing material (notice halogens).



2-86

5331A18

Fig. 14. The observed effect of the PVC tubing on the chamber aging (notice that aging continues even after switching to steel tubing).^[27]

5.4 GAS FLOW RATE

This is one of the most important variables to control in plasma chemistry whenever polymerization or etching is performed (Figs. 15 & 9 show typical examples of both types of behavior). The rate of polymerization (Fig. 15) is low for both a very high and very low flow rate. In plasma chemistry it is argued that at low flow rate there is not enough of the primary polymerization material (original hydrocarbon molecules), and at high flow rate one reduces the concentration of radicals by supplying more of the original hydrocarbon molecules and heavy positively charged polymers are swept away before they can reach the cathode.

How is it in our field? First of all, the gas pressure is typically ~ 1000 times higher and we usually find ourselves on a descending part of the Fig. 15 curve. In fact, the permanently sealed counters have some of the worst lifetimes measured. For instance, A. Smith and M. Turner have measured lifetimes of $\sim 2 \times 10^{-4}$ C/cm (90% Ar + 10% CH₄, 1.1 atm, 50 μ m anode diameter) and $\sim 5 \times 10^{-6}$ C/cm (90% Xe + 10% CH₄, 1.4 atm, 20 μ m anode diameter).^[29] J. Va'vra has measured a lifetime of about 4×10^{-4} C/cm in the micro-jet chamber (90% Ar + 10% C₄H₁₀, 6.1 atm, 7.8 μ m anode diameter).^[39] One criticism of such tests is, of course, possible confusion with an outgassing effect. It would be more direct to actually measure the lifetime as a function of flow. J. Kadyk^[23] has measured, for instance, a gain drop of $\sim 33\%/C.cm$ at a flow rate 105 cm³/min, and $\sim 69\%/C.cm$ at 50 cm³/min (50% Ar + 50% C₂H₆). He has reached similar conclusions in other gas flow tests.

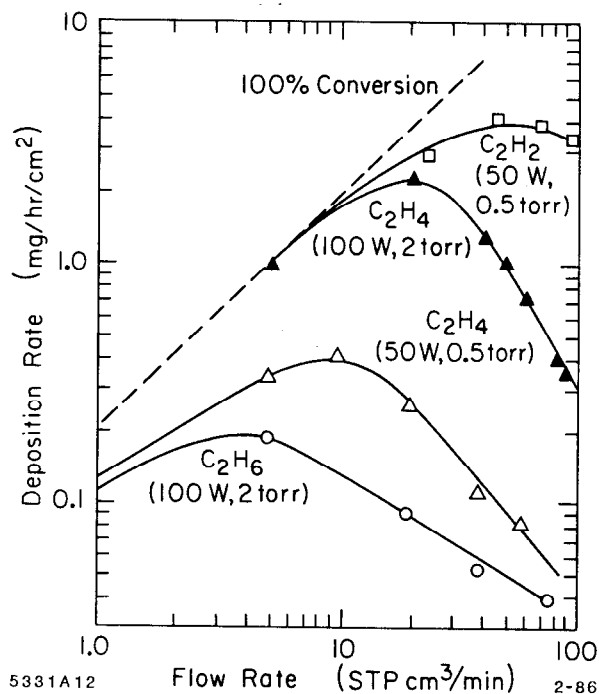


Fig. 15. An effect of a gas flow rate on the polymerization rate of acetylene, ethylene and ethane in a plasma chemistry environment.^[73]

5.5 ANODE-CATHODE DISTANCE

According to our naive model, if the cathode is very close to the anode and the gas flow is not sufficient, the large positively charged polymerized species will have a higher probability of reaching the cathode before the gas flow could remove them. In addition, the attenuation length ($1/e$) of photons which are capable of knocking out electrons from the cathode surface could be rather long. For instance, M. Atac^[12] has measured this length to be $\sim 480 \mu\text{m}$ in 50% Ar + 50% C_2H_6 and $\sim 160 \mu\text{m}$ when $\sim 1.5\%$ ethanol is added. This may be important for very closely spaced wires like pickup wires or grids.

An example of such a measurement is P. Drell's lifetime tests.^[30] With a grid structure 1.7 mm away from the anode plane the measured lifetime was $\sim 0.03\text{--}0.04 \text{ C/cm}$ with 92% CO_2 + 8% C_4H_{10} . With a structure where the grid was 6 mm away the test obtained $\sim 0.15\text{--}0.4 \text{ C/cm}$. What is needed is to check if larger gas flow would improve the first measurement.

5.6 TYPE OF CATHODE (WIRE VERSUS CONTINUOUS)

The larger the cathode surface, the longer it takes to accumulate a certain thickness of insulating material. This material, as we said, is mostly produced at the anode and brought to the cathode by electrostatic forces (either positive charge or dipole moment). Once we accumulate a certain thickness of the insulating material, a continuous cathode will trigger the Malter effect^[31] later than the wire cathode, because it has a generally lower electric field at the surface—Table 8 shows experimental results supporting this mechanism (see Fig. 16). The measurements by G. Charpak et al^[32] were done with a magic gas mixture and a high gain. One can see that the continuous cathode is substantially better, from the aging point of view, than the wire cathode. However, this test also indicates that

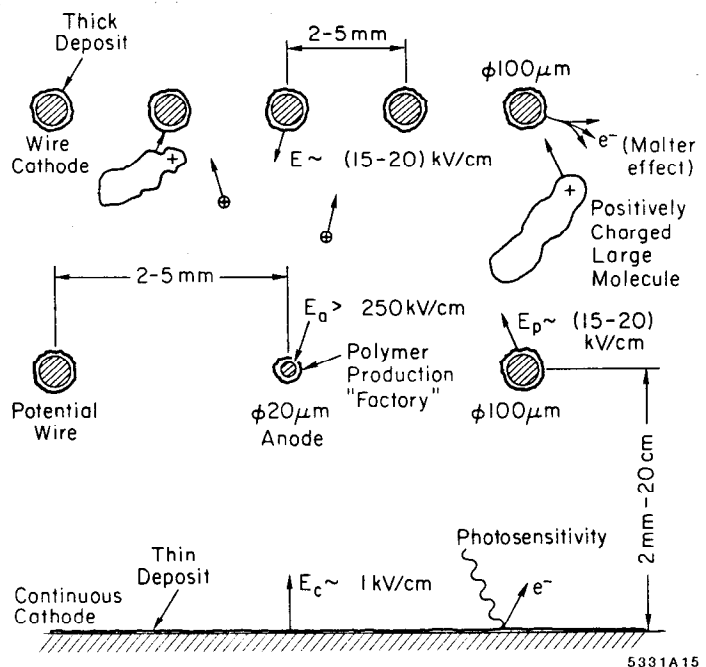


Fig. 16. Typical wire chamber structures, distances, electric fields on the surface and basic aging processes.

the continuous cathode + additive is equivalent, as far as aging, to the wire cathode + additive, where the additive is either methylal or isopropyl alcohol.

5.7 PRESSURE DEPENDENCE

This is certainly one of the least understood variables affecting aging. The aging is probably a delicate balance between two basic effects:

- (a) average electron energy $\sim E/P$, and
- (b) hydrocarbon molecule concentration $\sim P$.

For a fixed voltage, as one increases the pressure, the E/P value gets smaller, and the average electron energy is also reduced and therefore the production rate of free radicals is also reduced. This effect is offset by a higher concentration of hydrocarbon molecules (*i.e.*, the primary building material). However, one should mention that the pressure increase will help to reduce the photon attenuation length and therefore reduce the photo effect processes at the cathode surfaces. H. Boenig^[35] predicts that in the plasma chemistry

environment the final outcome will be a slow increase in the polymerization rate (for example ethylene polymerization).

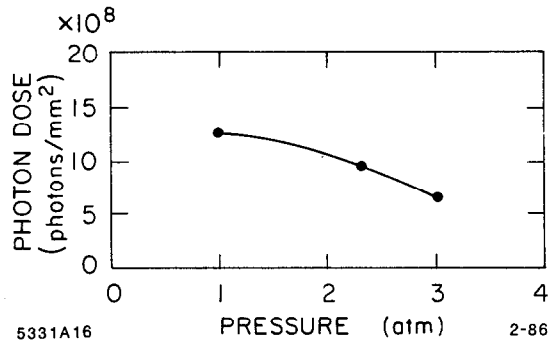


Fig. 17. The pressure dependence of aging (a photon dose required to reduce the plateau^[36] by 50%) in 70% Ar + 30% CO₂, voltage constant.

It is difficult to extrapolate this effect to our range of variables. Faruqi^[36] measured this dependence (see Fig. 17) and it appears to agree with H. Boenig's conclusion. A. Wagner^[43] reported on JADE experience. Innermost wires might have dropped the gain by about $\sim 11\%$ for a total charge dose of about 0.01 C/cm and in 88.8% Ar + 8.9% CH₄ + 2.6% C₄H₁₀ at 4 atm. However, since this experiment is a five year effort and many variables were changed, one cannot necessarily use it as an indication of the pressure effect.

5.8 TEMPERATURE

This is an even more confusing parameter. Nevertheless, one can find a number of examples in plasma chemistry where the polymerization rate decreases with increasing temperature. For instance, Fig. 18 shows this effect for a case of polymerization of C₂F₄.^[37]

In our field there is not much data, except H. Hilke^[18] reported preliminary data indicating agreement with an observation of the plasma chemistry people. If this correlation is proven true one might find a number of possible applications. For instance, aging in the *CRID counters*, which presently use *TMAE* and for which the remedies recommended in the previous sections might not apply (except a high gas flow rate). According to M. Atac, reporting on D. Anderson's preliminary measurements at 20 Torr of pressure, the gases containing *TMAE* have a rather poor lifetime ($\lesssim 0.1$ C/cm).

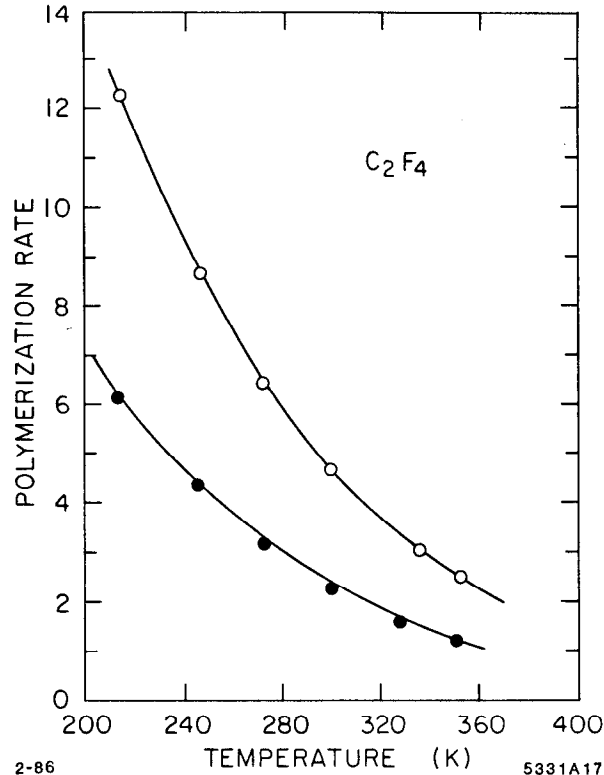


Fig. 18. Temperature dependence of the polymerization rate of C_2F_4 gas.^[37]

5.9 ANODE WIRE DIAMETER

For a general deposit on the anode, the gain behavior can be quite erratic. However, in many instances, the anode deposits can be quite uniform (oil or powder), and in that case the gain drop will be $G/G_o \simeq 1 - \alpha \epsilon/a$, where a is the anode radius, ϵ is the deposit thickness and α is a geometry and gas dependent constant (typically^[29] $\alpha \simeq 5-10$). If we express the gain drop in terms of a volume per unit length of the deposited material (W), we get a formula $G/G_o \simeq 1 - \alpha W/2\pi a^2$, i.e., the smaller the anode wire diameter, the larger the gain drop, assuming a constant W .

5.10 ANODE AND CATHODE WIRE MATERIAL

There could be a complex chemical process going on either on the anode or cathode surface. For instance, if the anode wire is made of a complex alloy, which is not gold plated, some elements of the alloy might react with the gas or some of its additives (respectively, their radicals) and rapid aging may occur. As an example, J. Kadyk^[23] has compared the gain drop due to aging with two types of anode wires and 49.3% Ar + 49.3% C_2H_6 + 1.4% Ethanol. The Au/W wire gave a gain drop of $\sim 1.3\%/C.cm$ as compared to the Stabelohm wire (70% Ni, 25% Cr, 5% Al + Cu) giving $\sim 800\%/C.cm$. D. Hess suggested that Al and Cu might oxidize utilizing the oxygen from a destroyed ethanol molecule. In the second example, H. Nelson^[21] has reported a loss of thin Al coating in aluminized mylar straw tubes, when exposed to $\sim 0.25 C/cm$ of ^{55}Fe source in the 50% Ar+50% CO_2 gas (Al was simply etched away). To indicate the complexity of the details of the aging, we

mention a test of P. Le Dû *et al.*,^[40] where they markedly improved the chamber lifetime by coating the mylar cathode with a graphite conducting paint (as compared to a bare Al surface). Clearly, the details of "sticking" (electrostatic attraction?) the polymers to the cathode surface are far from clear and will require more study.

6. How Should the Aging Test Be Done?

A basic message in this section is that it is far from easy to perform an accelerated aging test correctly. To name a few problems associated with such tests:

- (a) The most significant problem is that they typically do not test " $\sim 1\%$ tail effects" which are certainly very visible in a system with a large number of wires.
- (b) The gas flow, which is a very important parameter in plasma chemistry and probably also in our field, is usually much larger and more efficient in a small test compared to a typical large device.
- (c) The gas impurities are usually completely different in the small tests as compared to a real detector environment.
- (d) There is the usual question: "clean" versus "dirty (but realistic)" environment. If "clean," how clean? Is 10–20 ppm impurity level significant?

Our point is that if there is to be any chance of disentangling a many variable problem, we have to be able to separate the variables. It is better to introduce 10 ppm of O₂, H₂O or freon into the system controllably, rather than have all of them at the same time. This has, however, one basic problem: how do we apply results of such "controlled" tests to a real environment where one has 15 variables at the same time? My answer would be that if we find exact conditions for extremely good lifetimes, we might try to reproduce them in the real detector.

In the next section we will discuss the question of ultra pure gases. It is obvious that it doesn't make sense to put the ultra pure gas into a dirty gas system. Therefore, these tests should also be made in an ultra-pure gas system, which allows recirculation with removal of oxygen, water, dust and other contaminants.

6.1 ULTRA PURE GASES?

There is clear evidence that one should buy the highest possible purity of gases. For instance, Hartman^[41] reports a stabilization of aging in the TASSO central detector after ethane (50% Ar + 50% C₂H₆) was cleaned from oil contaminants and gas filters were installed. Kotthaus^[27] reached 0.7% C/cm with 50% Ar + 50% C₂H₆ (no additives) in a very clean environment (MPI tests in Table 11). J. Kadyk^[23] has done several such comparisons, for instance the gain drop was 15 %/C.cm versus 147%/C.cm for "pure" and "regular" purity of 89% Ar + 10% CO₂ + 1% CH₄ gas, and -6%/C.cm versus 63%/C.cm in the case of 80% Ar + 20% CH₄.

6.2 HIGH VERSUS LOW INTENSITY?

In a low source intensity test, each avalanche is realistic and such a test simulates an experiment correctly. However, in a high intensity test we start getting a modification of the avalanche strength due to several effects:

- (a) pile up of two ionization events on the same spot of wire,
- (b) saturation of ionization within the same event, and
- (c) modification of field due to a space charge.

These effects will generally tend to reduce the electron density (total charge is reduced) in each individual avalanche and to make up an overall integrated current, we have to either increase the intensity of the source or increase the voltage on the chamber (this causes the avalanche to spread, reducing the electron density). If we reduce the electron density in the avalanche, we expect, according to our wire model, to reduce the density of free radicals and therefore improve chamber life.

To calculate the gain drop due to the space charge, one either has to make a proper solution of Poisson equations in cylindrical coordinates, including the Z -coordinate dependence,^[43] or to get a feeling for the problem, one can neglect the Z -dependence and assume a Gauss law and a simple gain dependence $G \simeq \exp \{ \gamma (V - V_T) \}$. One obtains a formula for gain drop $G/G_o \simeq \exp [-\gamma r_o^2 \ln(r_o/r_a) (\rho^+ / 2\epsilon_o)]$, where r_o is the cathode radius, r_a is the anode radius and ρ^+ is the density of positive ions. For instance, for J. Kadyk's geometry^[23] one obtains the result that the anode current should be limited to ~ 50 nA/cm if one desires a gain drop in the center of the radiation flux to be less than 10%.^[44] J. Kadyk^[23] has done two types of tests addressing the intensity question:

1. Keeping the chamber voltage constant and varying the source strength ($A\ell$ shims) and using the same bottle of high purity 50% Ar + 50% C₂H₆ he obtained a gain drop of $\sim 23\%/C.cm$ for the anode current 61 nA and $\sim 6\%/C.cm$ for the current ~ 392 nA.
2. Keeping the source strength constant and varying the chamber voltage, he obtained the following results: $\sim 99\%/C.cm$ for 95 nA, $\sim 66\%/C.cm$ for 372 nA and $\sim 34\%/C.cm$ for 1495 nA. In fact, all his ~ 1600 nA runs have rather good lifetimes.

Similar results have been reported by Kotthaus.^[27] Table 11 (under "Conclusions") shows results in typical tests where rather large radiation doses have been achieved. As one can see, many results were obtained at rather large current and therefore one needs to perform additional measurements at low source intensities before the test results can be extrapolated to large experiments. This emphasizes the point that aging tests are difficult to do. In order to see a dependence, we have to make an accelerated aging test. However, we need to normalize the final result with a low intensity test.

6.3 DELIBERATE SPARKING TESTS

One thing we cannot avoid is accidental sparking in the chamber due to beam losses. Each spark is a potential danger from the aging point of view because it can "prime" the

wire for subsequent deposit buildup. Therefore it makes sense to perform special tests under very high current conditions.^[12,34,45] One should point out that these are special tests where the plasma conditions are generally different compared to usual aging due to typical avalanche conditions. For instance, B. Foster concludes that gas mixtures containing even small quantities of hydrocarbon (ethane, for instance) are very prone to whiskers or deposits.^[34] Adding ethanol vapor inhibits but does not prevent this process. Ar/CO₂ mixtures generally do not grow whiskers and can rehabilitate chambers where whisker growth has started. H. Sadrozinski has performed similar tests with 89% Ar + 10% CO₂ + 1% CH₄ mixture and breakdown currents of > 0.1 μ A/cm. If the content of CH₄ exceeded 5% in this mixture, the whiskers were grown on the cathode at a rate of \sim 1mm/5min.^[45]

7. A Cookbook of NO, NO ...

Table 9 shows a summary of various variables which may affect aging adversely. Clearly, the basic conclusion is that the chamber structure should be made of metals, ceramic and glass, perhaps with a possibility of baking (applicable to some designs).

8. Conclusions on Gases

- A. If we obtain regular purity gases, a basic conclusion of the workshop is that *Noble gas + hydrocarbon* mixture should not be trusted for more than^[11,18,21,46] 0.01–0.05 C/cm. The *Noble gas + CO₂* mixture appears to behave about ten times better.^[11,21,46]
- B. If we obtain the “highest possible” purity of a gas, there is a clear evidence that a considerable improvement can be gained. For instance, at least a factor of ten can be gained in the case of the *Noble gas + hydrocarbon* mixtures.^[23,27,41]
- C. There is a clear evidence that the oxygen containing additives (*H₂O, alcohols, etc.*) can considerably improve the lifetime.^[12] — see also Tables 10 and 11.
- D. *DME* gas looks very attractive, but one should check for halogen impurities.^[23,25]

Conclusions

1. Chamber aging is clearly a very complex subject. If we are to make any significant headway in this field, we will have to consider it as a serious subject for scientific investigation and treat it correspondingly.
2. The gas impurities may be the most critical variable to control.
3. We should always tune the variables, such as gas flow, additive fraction, gas type, gain on the wire, impurities level, temperature, *etc.*
4. Perhaps one should look, as D. Nygren suggested, for a way to increase a conductivity of some of these insulating deposits.^[16]
5. One should limit the total radiation dose by limiting the gain on the wire (aging and cost of electronics are inversely proportional) and/or limit the primary amount of ionization through suitable geometry (small cells^[62] or restrictive curtains^[61]).

TABLE 9
List of NO, NO

- No halogens in the gas [23,25]
- No oil traces in the gas [23,41]
- No oil bubblers [27,38]
- No silicon rubber (RTV) to seal the chamber [23,47]
- No rubber O-rings [66, Fig. 14]
- No polyurethane adhesive [47]
- No PVC or teflon tubing [27]
- Avoid Cu-tubing [6]
- No soft epoxies or adhesives [47]
- No aggressive solder
- Avoid sparking with large capacitance in very early testing stages [12,34,45]
- Avoid unknown organic materials
- Avoid a large amount of G-10 (S_i) [18]
- During an entire operation of the chamber do not allow even a single case of bad gas! [63]
- Methanol and isopropanol attack dielectrics, methylal attacks mylar [12]
- No insulators on wires (lubricating oil, fingerprints, oxydized regions, etc.) [5]

TABLE 10

Experiments (Accept only those which accumulated more than 0.1 C/cm)

Gas	Charge Dose	Observed Change in Operation	Person and Detector
65% Ar + 35% C ₄ H ₁₀ + 1.5% Isopropanol	≲ 0.1 C/cm	Some (Traces of Deposits)	Beusch and Omega [38] CERN
50% Ar + 50% C ₂ H ₆ + Ethanol (2°C)	~ 0.2 – 0.4 C/cm	Initially: Yes Now: No	Hartman and Tasso Central Detector [17]
<p>Note: Initially observed aging. At that point add ethanol, and later filters in gas line, remove oil from ethane. No further deterioration seen and running well for two years. Alcohol added, not because shown necessary, but mainly in attempt to reduce anticipated high background currents.</p>			
95% Ar + 5% CO ₂ + H ₂ O + Ethanol (4°C)	~ 0.25 C/cm	Yes	Binnie, Tasso Vertex Ch. [17]
<p>Note: Onset of the Malter effect observed already at ~ 0.02 C/cm. Cured by adding H₂O and ethanol.</p>			
53% Ar + 40% C ₄ H ₁₀ + 7% Methylal	~ 0.2 C/cm	Some (~ 1 μm Thick Deposits)	Ullaland SFM, CERN [38]
76% Ar + 20% C ₄ H ₁₀ + 4% Methylal	~ 0.2 C/cm	No	Pile, BNL Hypernuclear Spectrometer [47]
<p>Note: However troubles seen in a chamber with a different construction</p>			

TABLE 11

Tests (Accept only those which accumulated more than 0.1 C/cm)

NOTE: Many results done at large currents

⇒ Absolute Lifetime Suspect!!

Gas	Charge Dose C/cm	Observed Change in Operation	Anode Current (cm ⁻¹)	Person
50% Ar + 50% C ₂ H ₆ (with and without 0.7% H ₂ O)	~ 0.7	No	~ 1 μA	Kotthaus [27] (MPI Test)
89% Ar + 10% CO ₂ + 1% CH ₄ (with and without 0.7% H ₂ O)	~ 0.7	Yes	~ 1 μA	Kotthaus [27] (MPI Test)
50% Ar + 50% C ₂ H ₆	~ 0.15	10% Gain Drop	~ 1 μA	Kotthaus [27] (KEK Test)
89% Ar + 10% CO ₂ + 1% CH ₄	~ 0.3	10% Gain Drop	~ 1 μA	Kotthaus [27] (KEK Test)
50% Ar + 50% CO ₂ 50% Ar + 40% CO ₂ + 10% X _e 49% Ar + 50% CO ₂ + 1% CH ₄	~ 0.3 – 0.5	Yes	~ 1 μA (20 kHz)	Nelson [21]
25% Ar + 50% CO ₂ + 25% n-Pentane	~ 1.0	Yes		Iaroci [49]
27% Ar + 73% C ₄ H ₁₀ + ≥ 0.1% H ₂ O	~ 0.1	Yes		Seiden [50]
50% Ar + 50% C ₂ H ₆ + 1.4% Ethanol (Very Pure Gases)	~ 1.0	< 2% Gain Drop	~ 400 nA	J. Kadyk [23]
89% Ar + 10% CO ₂ + 1% CH ₄ (Very Pure Gases)	~ 1.0	9% Gain Drop	~ 400 nA	J. Kadyk [23]
DME (No Freon 11)	~ 1.0	< 10% Gain Drop	~ 400 nA	J. Kadyk [23]
DME	~ 4.0	Some	~ 5 μA	F. Villa [25]
50% Ar + 50% C ₂ H ₆ + 0.5% Ethanol	~ 1.5	No	~ 440 nA	M. Atac [12]
DME	~ 1.0	Some	~ 3 μA	Majewski [6]
DME	~ 2.0	Some	~ 2 μA	Godfrey [46]
89% Ar + 10% CO ₂ + 1% CH ₄	~ 0.3	50% Gain Drop	~ 50 μA/25 cm	Sadrozinski [45]
93% Ar + 3% CO ₂ + 4% CH ₄	~ 1.9	50% Gain Drop	~ 50 μA/25 cm	Sadrozinski [45]
92% CO ₂ + 8% C ₄ H ₁₀	~ 0.15 – 0.4	Yes	~ 260 nA	Drell [30]

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57. Linde Molecular Sieve Type 3A has a chemical composition:

$$\text{K}_9\text{Na}_3[(\text{AlO}_2)_{12}(\text{SiO}_2)_{12}] \times \text{H}_2\text{O}.$$
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