

WIRE AGING OF HYDROCARBON GASES WITH TMAE ADDITIONS

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ABSTRACT

We report on experimental results describing wire aging properties of the CRID/RICH detectors¹ in the TMAE gas environment. This test used 7 μm and 33 μm diameter carbon fibers, and 20 μm diameter gold-plated tungsten wires as sense wires, and operated in gas mixtures of either CH_4 or C_2H_6 at 1 atm, together with 0.6 Torr of the photoionizing vapor TMAE (tetrakis dimethylamino ethylene), which serves as a photocathode in the CRID detectors. The wires were damaged by the Fe^{55} X-rays. This paper is based on irradiation of 20 wires, each being a separate test. The gas purity was consistent with requirements of CRID detectors (< 1 ppm of oxygen, < 1 ppm of water).² The test explored the aging as a function of wire diameter, wire material, gas flow, gas temperature, rate of radiation and gas type. Several chemical analyses of the deposit samples were performed (FTIR, ESCA and GC-MS techniques).

1. MOTIVATION FOR THIS PROJECT

The TMAE molecule is very fragile³ and therefore one would expect a large production of radicals in the plasma environment of an avalanche.⁴ In addition, it does not contain an oxygen atom which is known to impede the polymerization process in the plasma chemistry (in our field, compare for instance with molecules of alcohol or DME which contain the oxygen atom and are known to slow down the polymerization process). The way the oxygen helps is that the oxygen atoms react with the radicals and form mostly volatile and more stable products such as CO , CO_2 , H_2O , etc.⁴ Unfortunately, sometimes the oxygen reacts with certain materials of some wires and this affects adversely the aging rate.^{5,6} Finally, additional factors influencing the wire aging might be the use of very small anode wires,⁴ high electric field of about 900 kV/cm on their surface (creating more energetic electrons in the avalanche), as well as its carbon material.

All these factors created an interest to perform a sequence of tests to improve our understanding of the aging behavior of TMAE laden gases operating in the CRID environment.

2. EXPERIMENTAL SETUP

Special care was taken in preparing the test. This is because the TMAE molecule is known to react with oxygen and water even in minute quantities, and products of these reactions are electronegative.² The detector vessel was made of stainless steel (304 type) with gold-plated soft copper gasket (CONFLAT type flange), and all gas fittings and feedthroughs were of either stainless steel or ceramic. The whole structure was baked at 400°C after the machining. The exception was the detector PC-board made of G-10 (nonflammable) and the 0.0005-inch thick stainless steel window which was glued to the vessel. The test detector simulated the geometry of the CRID detector,⁷ with the exception that we did not use "blinding" wires to prevent a photon feedback. Figure 1 shows the test detector geometry. The Fe^{55} source could move in a slot perpendicular to the wire. The source irradiated the wire along 5 mm of its length. The cathode was made of nickel-plated aluminum, the anode wire was either of 7 μm or 33 μm carbon fibers, or 20 μm gold-plated tungsten wires. The carbon wires are difficult to solder and therefore we have adopted the following procedure to support them. After stretching we secured the wire temporarily with a tiny drop of five-minute epoxy (Hardman) at four spots along the wire. We then used a conducting epoxy (EPO-TEK H2O-E, cured at 90°C for two hours)

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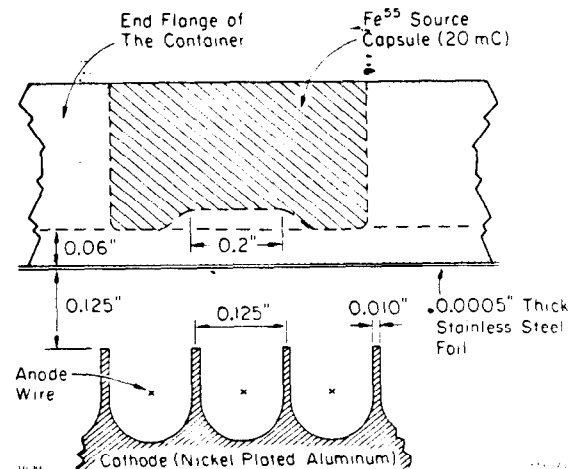


Fig. 1. Geometry of the test detector. The source slot runs perpendicular to the wires. Five mm of wire is irradiated on the average.

to make the electrical contact to the PC-board trace. We then covered the previous epoxies with a new coat of epoxy (Shell Epon 815+Versamid 140, cured at 50°C for 12 hours), which secured the wire and prevented the TMAE vapor from a direct contact with the above mentioned epoxies. We mention the wire support procedure exactly because it may have an effect on the aging due to its use of epoxies.

We needed a rather high intensity Fe^{55} source (20 mCi), because of low Z of all gases used in this test. We run all tests with +1.7 kV on the 7 μm diameter sense wire (the cathode is grounded in this test), which is rather close to a typical operating point of the CRID detectors.¹ The Fe^{55} source gave typically about 220 nA at that operating point with CH_4 +TMAE (27°C). All other operating points were chosen to give similar anode current (see Fig. 2 for the gain behavior as a function of voltage for different diameters of wires). We prefer not to exceed this current to prevent lowering of the effective gain due to space charge effects.^{4,8}

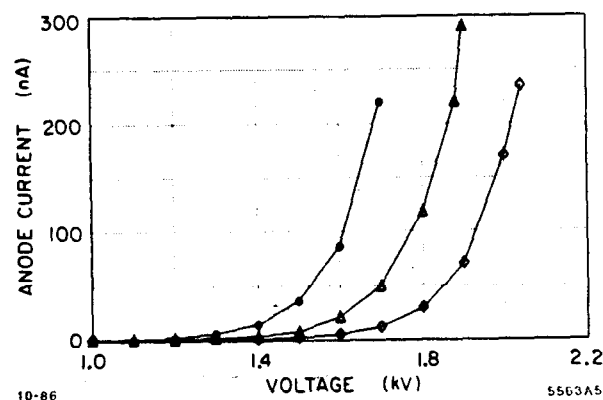


Fig. 2. The gain as a function of voltage for different diameters of wires as measured using the high intensity Fe^{55} source; CH_4 +TMAE (27°C), detector temperature at 45°C.

The gas used in this test was provided by the CRID gas system. It uses electropolished stainless steel tubing throughout and all connections are welded with the exception of VCR disconnectable fittings. The gas is purified by a molecular sieve and an oxysorb.¹² The TMAE vapor is added to the carrier gas in the bubbler operating at the appropriate temperature (in this test either 27°C or 12°C). All gas tubing as well as the detector vessel were heated to a temperature higher than

the TMAE bubbler temperature to prevent a condensation (in this test either 45°C, 38°C or 58°C). The oxygen impurity was measured directly after the detector vessel and was typically 1 ppm (Teledyne Co. instrument). If the oxygen level was higher, we would assume that something was wrong and would investigate the problem. Since the similar amount of oxygen impurity was also measured upstream of this test, we assume that the test was adding less than 1 ppm of oxygen. The gas passed the silicon diffusion pump oil¹³ bubbler downstream of this test, as is presently customary in the CRID group. It is not known if that is adding some effect to the reported aging.⁴

This test measured the anode current as a function of time. We interpret the drop in this current as a loss of gain on the wire. Periodically we used a low intensity Fe⁵⁵ source to measure the absolute wire gain (typically the total gain was about 2×10^5 on a fresh new wire). In addition, we monitored the temperature of the TMAE bubbler and the detector vessel and outside barometric pressure. However, since the aging in the TMAE gases is very rapid, the corrections for the barometric pressure changes were not necessary in the end.

EXPERIMENTAL RESULTS

We found that the wire aging in all our tests is surprisingly fast. The results correspond to the worst aging results in the wire chamber field.⁴ Typically, the wires would lose gain by a factor of two within ten minutes for anode currents of about 50–200 nA for an exposure over 5 mm of wire length. To put it differently, we see a factor of two gain loss after a total accumulated charge of about 10^{-4} C/cm. The rate of aging is slowing down by a factor of 5 to 10 after the initial steep gain drop, however, at that point, the gain is at 10–20 % of its original value. When the chamber was opened to air (after three days of flushing with a gas without TMAE), indeed, we found deposits on the wire in a spot corresponding to a source position. In addition, the corresponding region on the cathode was discolored to a light brown color. The anode wire deposits were initially colorless and continuously coating the wire (see Fig. 3). However, they would change into droplets within ten minutes to several hours if exposed to air (see Fig. 4). The droplets would change color from colorless to dark brown over a period of several days, indicating a continuous reaction with the oxygen and/or water from the air. In this form, they would stay for months at room temperature. We do not have any information about what the deposits look like before they are exposed to air. However, we can say that the initial very large gain drop is caused by a very thin layer small compared to



Fig. 3. Wire deposits in CH₄+TMAE (27°C) on 7 μm diameter carbon wire after ten minutes in air.

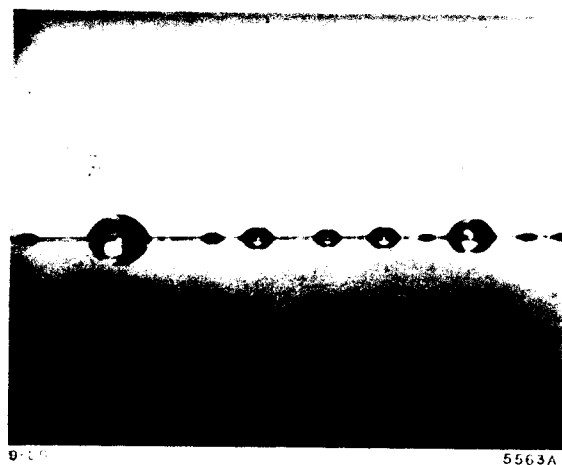


Fig. 4. Wire deposits in CH₄+TMAE (27°C) on 7 μm diameter carbon wire (different to Fig. 3) after several hours in air. The deposits obtained after a total charge dose of about 6×10^{-3} C/cm.

the diameter of the wire (one can easily miss it; it is only obvious after the droplets develop). This layer may be insulating and prevents the electrons to bleed away, thus reducing the electric field around the wire. This mechanism is somewhat similar reported by M. Atac⁵ for aluminum wires which oxidized and formed an insulating thin layer. We will discuss the analysis of the deposits in the last section. Now we will discuss the wire aging as a function of various variables.

Aging as a function of wire diameter and material (Fig. 5)

We can see that 33 μm diameter carbon wire aged about ten times slower than 7 μm diameter wire of the same material, if we use a gain loss of factor 2 as a criterium for comparison. This result is qualitatively expected.⁴ However, the surprising result is that the gold-plated tungsten wire is aging also fast and the rate of its aging correlates with its diameter rather than its material. This means that the carbon material does not contribute to the poor TMAE results.

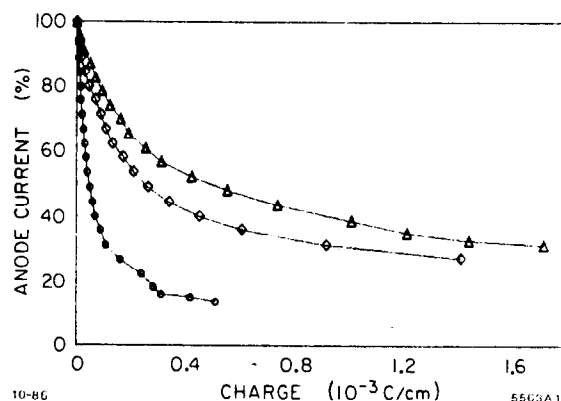


Fig. 5. Wire gain loss as a function of total charge for various wire types and diameters; (○) 7 μm carbon wire, (◇) 20 μm gold-plated tungsten wire, (△) 33 μm carbon wire. Starting current 220 nA, CH₄+TMAE (27°C) gas, 45°C detector temperature, 100 cc/min gas flow.

Aging as a function of gas flow (Fig. 6)

We have not seen any substantial dependence on the gas flow. This is not expected⁴ and is presumably caused by the fact that the TMAE aging is too rapid and our range of gas flow changes was too small. We varied the gas flow from 100 cc/min to 460 cc/min, corresponding to gas volume change every 25 and five minutes, respectively.

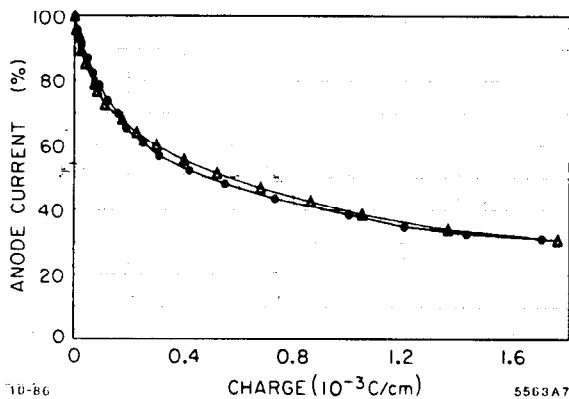


Fig. 6. Wire gain loss as a function of total charge for two different gas flows, (o) 100 cc/min, (Δ) 460 cc/min. Starting current 220 nA, CH₄+TMAE (27°C) gas, 45°C detector temperature, 33 μ m detector carbon fiber.

Aging as a function of gas type (Fig. 7)

We compared the aging rate in three different gases, CH₄+TMAE (27°C), C₂H₆+TMAE (27°C), and CH₄ without TMAE. One can see that the rate of aging in CH₄ gas is slower, however after about 2×10^{-4} C/cm, the chamber goes into a continuous discharge if the source is present (without the source there is no current). This can be immediately corrected if we switch into a gas with TMAE. After opening, we see no visible deposits either on the wire or the cathode. We interpret this as an indication that the cathode has become photosensitive by an extremely thin layer of deposits which effectively lowers its work function, and one needs TMAE to quench these photons. The CH₄ gas aging has been checked twice. In addition, Fig. 7 indicates that the aging in CH₄+TMAE gas is about three times faster than in C₂H₆+TMAE gas.

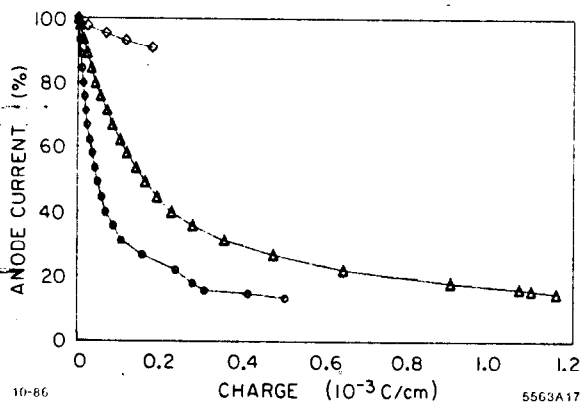


Fig. 7. Wire gain loss as a function of total charge for different gases, (o) CH₄+TMAE (27°C), (\diamond) CH₄ only, (Δ) C₂H₆+TMAE (27°C). Starting current 220 nA, 7 μ m carbon fiber, 45°C detector temperature, 100 cc/min gas flow.

Aging as a function of temperature (Fig. 8)

During this test we have varied the temperature of the TMAE bubbler and the temperature of the detector vessel. Since there was a good thermal contact between the detector and the vessel, we assume that quoted temperature is also a temperature of the wire. We do not see a strong temperature dependence of the wire aging.

Aging as a function of source intensity (Fig. 9)

We have performed this measurement to make sure that our results are not affected by space charge effects.^{4,8} The source intensity was varied by shims while the voltage was kept constant. We do not see a strong dependence of the wire aging on the source intensity in a range of initial anode currents between 39 and 225 nA.

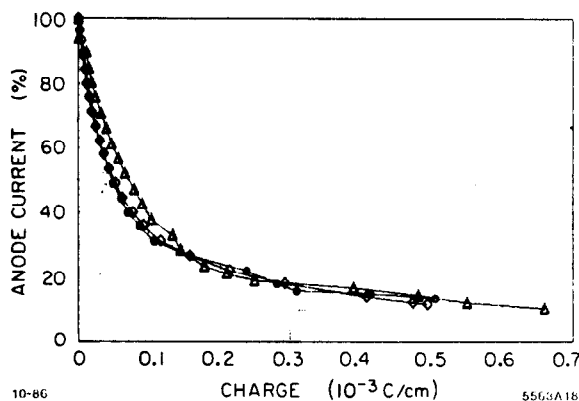


Fig. 8. Wire gain loss as a function of total charge for different temperatures, (o) TMAE bubbler at 27°C and detector vessel at 45°C, (Δ) TMAE bubbler at 12°C and the detector vessel at 38°C, and (\diamond) TMAE bubbler at 27°C and the detector at 58°C; CH₄+TMAE gas, 7 μ m carbon fiber, starting current 220 nA, 100 cc/min gas flow.

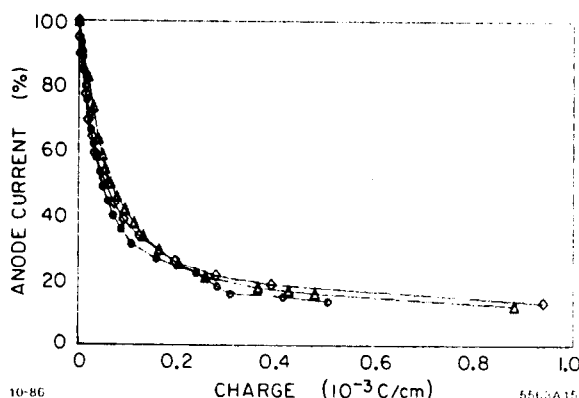


Fig. 9. Wire gain loss as a function of total charge for different Fe⁵⁵ source intensities and constant voltage, (o) starting current 225 nA, (\diamond) starting current 120 nA, (Δ) starting current 39 nA. CH₄+TMAE (27°C) gas, 100 cc/min gas flow, 7 μ m carbon fiber and 1.7 voltage in all three cases.

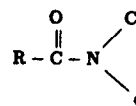
Is there a slow recovery of aged wires?

This is an interesting question. We have checked every wire after its aging was finished. But, based on 24- to 48-hour time delays, we have not seen any recovery. However, this should be checked over a longer period of time and as a function of temperature of the wire.

ANALYSIS OF THE DEPOSITS

The wire deposits and the cathode coating were analyzed by three methods, FTIR⁹, ESCA¹⁰, and GC-MS¹¹. Even though these are powerful techniques, we have obtained only sketchy information about the deposits.

The FTIR technique is based on the fact that molecular bonds vibrate at characteristic frequencies when exposed to infrared radiation. Specific pieces of molecules can be determined. The analysis was performed on the wire deposit sample (after being exposed to air for several days), clean TMAE sample, and TMAE sample exposed to air for ten minutes. Figure 10 shows the results. The conclusions of this study are that the deposits are not oxidized TMAE, and that they are instead a tertiary amide



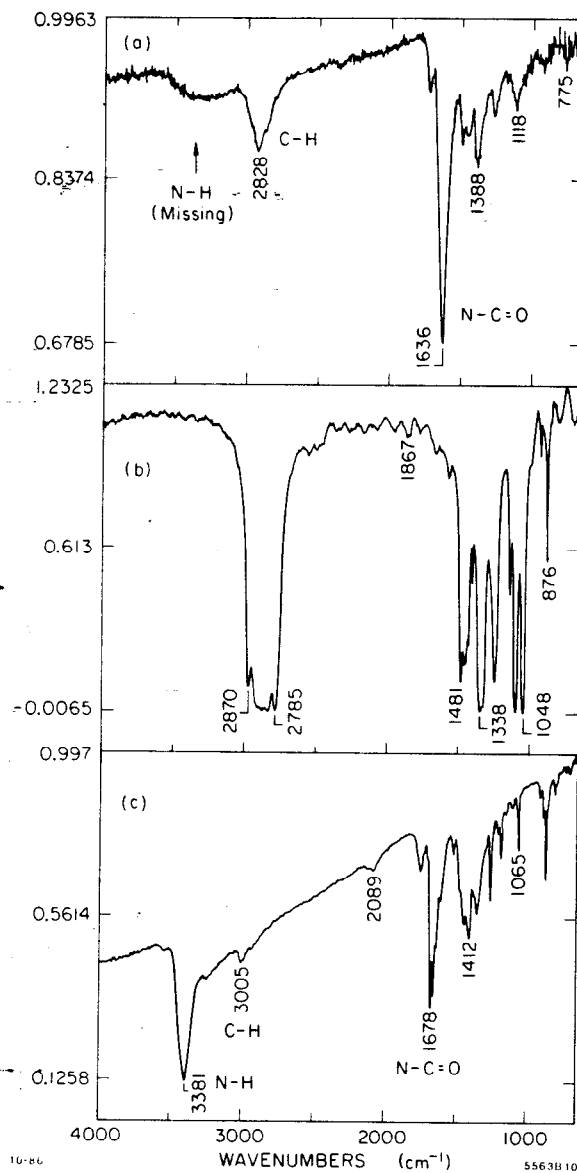


Fig. 10. FTIR⁹ spectrum on three samples, (a) this test wire deposits, (b) clean² TMAE not exposed to air, and (c) the same TMAE exposed for ten minutes to air.

(tertiary stands for three carbons attached to a nitrogen atom). A search of available reference material revealed three compounds with similar IR spectrum: tetramethylurea, dimethyl acetamide, and dimethyl formamide. However, the real question is what was the original molecule which reacted with oxygen to create the tertiary amide.

The next technique GC-MS combines the gas chromatography with the mass analysis. The wire sample was transferred this time into a glass container in a glove box (no reaction with oxygen or water this time) and the deposits were dissolved in methylene-chloride. Unfortunately, we did not have enough deposits in this particular test. Nevertheless, the results indicate that the deposits contain at least three different compounds with the molecular weight over charge ratio (m/Z) 202, 100 and 100.

The ESCA technique was applied to the cathode deposits. The method uses 1.487 keV X-rays which strike the sample, and one measures the energy of liberated electrons. The method can identify not only the elemental composition, but also the bonding structure in the first 100 Å of deposits (~ 20 atomic layers). One operates the sample in a 10⁻⁸ Torr vacuum. The basic conclusions of the ESCA analysis of the irradiated region were as follows: (a) the elemental structure of the deposits (atomic percentages) was 83% C, 8.2% O, 5.9% N, 2.7% S; and 0% N; (this means that the deposits are thicker than 100 Å), (b) carbon occurred mostly in a form of graphite (we used 7 μm carbon wires) and in smaller fraction in amides (N - C = O), silicone (- [R₂SiO]_n -), and amines (NR₃, R=C,H), (c) silicon was present in a form of silicone (probably coming from the oil bubbler), (d) oxygen occurred either in the silicone or in C-O bonds, (e) nitrogen was present either in the amide or the amine. In any case, the cathode was coated by many types of organic species and the total thickness of deposits was more than 100 Å (for the total charge dose of less than 5 × 10⁻³ C/cm).

CONCLUSIONS

The wire aging in the TMAE laden hydrocarbon gases is a complex phenomenon and we do not expect that this paper represents the end of an effort to understand it. On the contrary, this paper should encourage interest in this area. We need to invent additives or some curing procedure to reduce the rate of wire aging. Otherwise, we do not see that this kind of Cerenkov counter in a version as is now envisaged can be used in a high radiation environment like SSC. One expects trouble even on some present accelerators (a worry is that the aging is nonuniform, creating regions of inefficiency). If our results are correct, one would expect to lose a factor of two in gain for about 10⁹ single photoelectrons arriving on 1 cm of 7 μm carbon wires operating with a gain of about 2 × 10⁵ and CH₄+TMAE (27° C) gas. This corresponds to a rate of less than 50 Hz/cm of wire length in one year, if one assumes that the problem is linear and that there is no slow recovery due to evaporation. We improve the aging performance by a factor of 3 if we replace CH₄ by C₂H₆ gas for 7 μm carbon wires. We gain a factor of 10 if we increase the wire diameter from 7 μm to 33 μm in CH₄+TMAE gas, and a factor of 5 if we increase it to 20 μm gold-plated tungsten wire. We have seen no strong dependence on the gas temperature, on the gas flow, or on the source intensity. Both anode and cathode are coated by a variety of organic material.

We would like also to stress also that a proper accounting of anode currents down to the nA range is required to estimate projected problems. The detectors should be designed so that they can be removed and cleaned without tremendous difficulty. We believe that the detector should have also a gating scheme to be able to reject unwanted ionisation before it is amplified on the wire. In any case, it is obvious that one will have to be unusually "gentle" with this type of detector.

The aging study of TMAE gases will continue in the near future.

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9. FTIR stands for Fourier Transform Infrared Analysis and was performed by Ch. Brisko, SSL, Mountain View, CA 94043.
10. ESCA stands for Electron Spectroscopy for Chemical Analysis and was performed by R. Lipari, SSL, Mountain View, CA 94043.
11. GC-MS stands for Gas Chromatography coupled with Mass Spectroscopy and was performed by R. T. Rewick, SRI, Menlo Park, CA.
12. Made by Messer Griesheim GMBH, W. Germany (3.3 l volume).
13. Made by Dow Corning, U.S.A. (#704 type).