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ALPHA MEASUREMENTS AND AUTORAMP

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ABSTRACT

AUTORAMP, designed and developed at the Environmental Measurements Laboratory (EML), is a completely automatic, field deployable system that performs sampling and gamma-ray spectrometry analyses of filter cartridges. However, for certain measurement applications such as airborne plutonium, gamma-ray analysis alone does not suffice. Plutonium cannot be detected by gamma-ray analysis, but is detectable with alpha analysis. This paper describes the initial research towards giving the AUTORAMP the dual capability of both alpha and gamma measurements. Because of the geometry used in the AUTORAMP cartridge, the only compatible alpha analysis involves the measurement of the ionization produced by an alpha particle as its energy is adsorbed in air. Although this type of alpha measurement has been done before, the associated technology is difficult and requires many of the techniques used in the measurement of currents at the fempto-amp (10^{-15}) level. Fortunately, EML pioneered MOSFET (metal oxide semiconductor field effect transistor) technology at currents in the atto-amp (10^{-18}) region, and is, therefore, uniquely qualified for this research. The paper presents the results obtained from the initial chambers and electronic circuits for gross alpha counting and for alpha spectrometry analysis. The techniques employed to eliminate noise are presented, as is the design of the proposed chamber to accommodate the filter cartridge and a study to shift the frequency spectrum of the collected ionization pulses away from the noise.

INTRODUCTION

Since 1963, the Environmental Measurements Laboratory (EML) has maintained a worldwide network of surface air sampling stations for its Surface Air Sampling Program (SASP) to monitor and inventory radionuclides. At these stations, airborne particles are collected by drawing about 2500 m^3 of air per day through filters. After collection, these filters are sent to EML where they are analyzed for gamma-ray emitting radionuclides. Logistical problems at some remote sites prevented the timely analysis of the filters for the relatively short half-life of the radionuclide of interest, ^{7}Be, and for other anthropogenic radionuclides. This led to the development of the Remote Atmospheric Measurements Program (RAMP). Through telemetry with the ARGOS or the GOES satellite systems, the results of the on-site gamma-ray analyses are now received by EML within hours. Fourteen such systems have been successfully deployed worldwide, with some in continuous operation since 1987.

However, these systems still require an on-site operator to first load the filter in the aerosol collector, and, after the sampling period, transfer it to the gamma-ray detector, and initiate the counting and data transmission process. The AUTORAMP system is a major advance which eliminates this manual handling with a robotic system (1).

The AUTORAMP system, see Fig. 1, operates in a completely automatic mode and performs all of the manual functions mentioned with additional enhancements. For example, the standard SASP air pump was replaced with a higher volume turbine blower, resulting in a flow rate of over 12,000 m^3 per day, which is an increase of over 450%. This air volume flows through a pleated cylindrical filter cartridge that was designed by EML. By placing this cylindrical filter cartridge around a 35% high purity germanium (HPGe) gamma-ray detector, the counting efficiency obtained is about 25% higher than that obtained with the aforementioned systems. These filter cartridges have an outer diameter of 12.5 cm, an inner diameter of 7.5 cm, and are 8.9 cm high and contain 1580 cm^2 of HEPA filter medium. They are capable of sampling over 15,000 m^3 of air with negligible loading, and achieving a collection efficiency of greater than 99% for submicron particles. In addition, a projection bar code reader in the AUTORAMP reads the label on each cartridge thereby providing accurate identification. The robotic system can move these filter cartridges within a volume of 91.4 cm x 58.8 cm x 36.8 cm (L, W, H). Located at the lower end of the vertical robotic axis, see Fig. 1, is an electro-mechanical "pickup" mechanism that grips and holds the cartridges while they are being transported. The tri-axial robotic arm performs all the mechanical movements required for automatic operation, including:

- opening the detector shield and transporting the analyzed filter cartridge back to the sample tray,
- opening the sample chamber and transferring the sampled cartridge to the detector shield,
- transporting a new filter cartridge from the sample tray to the sample chamber, and
- closing the detector shield and sample chamber and then initiating sampling and counting.

Communication with the AUTORAMP is via a two-way modem link. The AUTORAMP will call to announce the start of a count, to send data, or to report an unusual condition. The receiving station may call AUTORAMP at any time for a status check, to examine current or previous data, or to change the system's operating parameters.

WHY ALPHA MEASUREMENTS?

At some DOE facilities, alpha measurements of airborne radioactive contaminants may be more sensitive than gamma-ray analysis. This is especially true where the contaminants of concern are depleted uranium or plutonium since the long-lived isotopes of both of these elements decay primarily with alpha emissions. These isotopes can be quantified by gross alpha analysis or by alpha spectrometry and computer deconvolution if some degree of energy discrimination can be achieved.

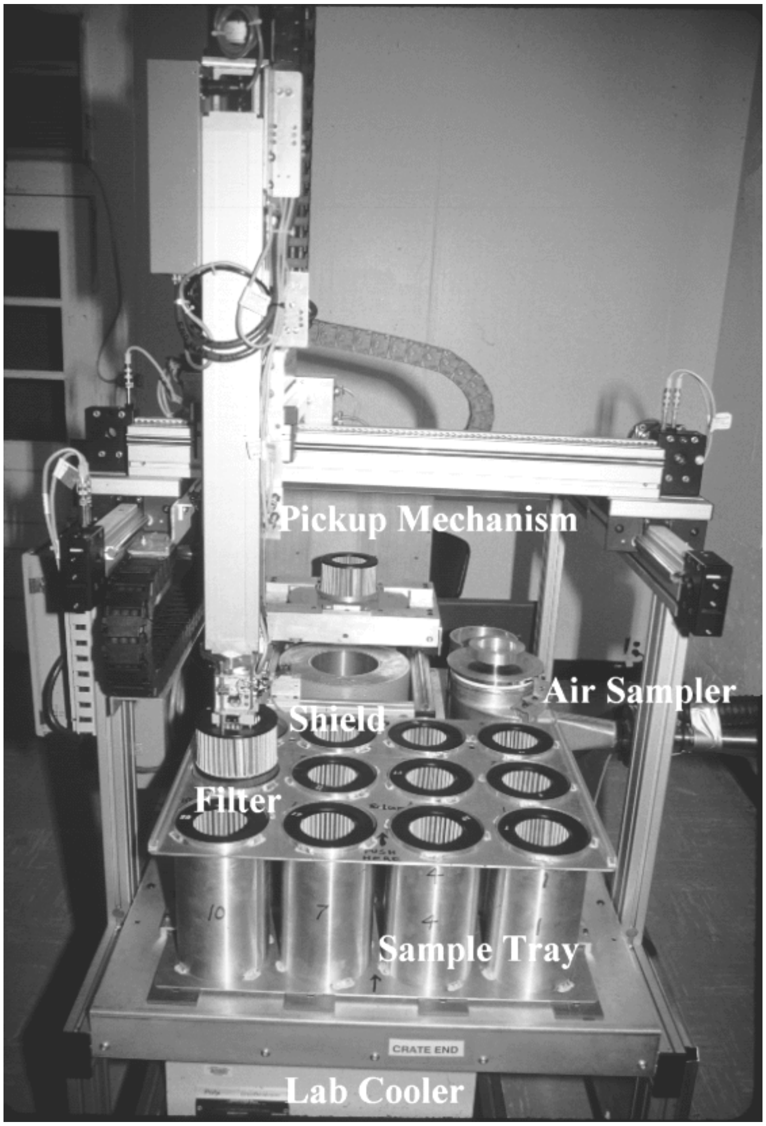

Fig. 1. EML's AUTORAMP system. The sample tray with the filter cartridges is in the foreground.

ALPHA MEASUREMENTS IN AIR

Our initial effort to count alpha emissions consisted of a small chamber containing an alpha source and a collecting electrode. The signal from the collecting electrode was coupled to an amplifier contained in a shielded box. To collect the ions from the alpha source, a bias voltage of 300 V was applied between the walls of the chamber and the collecting electrode. However, building vibrations, walking, talking, etc., cause small mechanical movements (microphonics) between the chamber and the amplifier. The net effect of this is to generate low frequency noise which completely obscures the alpha signal. It quickly became apparent that many of the same techniques that were used in the development of low current MOSFET (metal oxide semiconductor field effect transistor) technology were also required in this application. EML was uniquely qualified to initiate this study because it pioneered the low current MOSFET technology, which Keithley Instruments, (Cleveland, OH) incorporates in the world's most sensitive current instrument, and because it invented the temperature-compensated-electrometer (2) used worldwide in nuclear power plant monitoring. Paramount among these low current techniques is to rigidly couple the amplifier to the chamber, that is, avoid connectors. To establish an overall rigid system without connectors, an existing chamber with solid 1 inch walls was used. The amplifier was then securely bolted on top of this chamber. This arrangement was tested by placing an alpha source inside the chamber and using a collection voltage of 300 V. When the waveforms are viewed on an oscilloscope, the alpha pulses are clearly seen but the baseline remains too noisy for alpha spectrometry.

Thus, the AUTORAMP can easily be adapted for gross alpha counting by using a suitable chamber with a rigidly attached amplifier. Moreover, the existing filter could probably be used without any changes. This will be confirmed once the ionization chamber is built.

ALPHA SPECTROMETRY IN AIR

Pulse counting ionization chambers are usually filled with rare gases (e.g., P-10, 90% argon and 10% methane), which have a low electron affinity and do not produce negative ions by electron attachment. This allows the ionization from the alpha particle to be collected as electrons, which yields a fast pulse signal that is easily measured electronically. However, in atmospheric air, the electrons from the alpha-particle ionization quickly form negative ions, principally with oxygen, so the pulse signal collected is now much slower and more difficult to measure. The principle reason for the difficulty is that the frequency content of the signal is below 1 kHz, which places it right in the middle of low frequency noise such as microphonics, and other low frequency noise sources.

PRESENT ALPHA SPECTROMETRY TECHNIQUES

The literature contains several references to methods that employ alpha spectrometry in atmospheric air and two of these describe instruments that are used commercially; all of these instruments are used to measure ^{222}Rn. In Japan, Katase et al. (3) describe a chamber using layers of multiwire electrodes in a plane geometry chamber configuration. This chamber measures 60 cm by 50 cm and is 17 cm high. Two multiwire electrode layers are used, with the wires made of 0.2 mm nylon coated with a conductive layer. Nylon is used so that the characteristic mechanical frequency is above the frequency range of the signal. Even with these precautions to further reduce low frequency noise, this instrument has to be externally suspended with an elastic rope for meaningful results.

In Sweden, Baltzer et al. (4) describe an instrument that is sold commercially as the "Atmos-10". This instrument employs a cylindrical chamber with spirally wound multi-wire electrodes to produce a field gap of 1 cm. The active volume of the chamber is 0.6 l and the electrode wires are stainless steel. This chamber also requires an elastic mount, which is integrated in the instrument, and also uses a dehydrator for drying the inlet air.

In Germany, Genrich (5) describes an instrument using a cylindrical chamber with a center electrode that is sold commercially as the "Alphaguard". This instrument employs high speed digital signal processing (DSP) that allows rapid execution of cross correlation routines to aid in the extraction of the signal from the noise.

ALPHA SPECTROMETRY AT EML

As a result of our initial work and the present techniques in use, it became apparent that changes are required in the AUTORAMP to accommodate alpha spectrometry. The pleated filter cartridge must be replaced with a simple cylindrical filter in which the filter media has alpha spectrometry capability. Because of the small pore size associated with this type of media, the present sample pump used in the AUTORAMP must also be replaced. However, these changes can be made without altering anything in the robotic assembly. Of more immediate concern is what technique should be used for alpha spectrometry. Because of the problems associated with using counting gases in a remote instrument, our present thinking is to use atmospheric air. While all of the above mentioned techniques extract the signal from the noise, our focus is to reduce the microphonics and other low frequency noise as much as possible, and to try and shift the frequency spectra of the signal to higher frequencies away from the low frequency noise.

PREAMPLIFIER CONSIDERATIONS

The preamplifier or charge-sensitive amplifier is critical and establishes the overall performance (signal-to-noise ratio). Several circuit configurations were tried. The field effect transistor (FET) approach is basically a low-noise FET followed by a gain (operational amplifier) element with a current-to-voltage resistor in the feedback loop. Right now, this is the type of circuit being used. Some designers prefer a switching integrator element for the preamplifier, so this was also tried. But using the integrator (Burr-Brown ACF2101, Tucson, AZ) showed no significant improvement over the FET approach. It was thought that the leakage current of the Burr-Brown integrator $\sim 10^{-13}$ A was too high, so a MOSFET integrator was also tried. Although this EML designed integrator-electrometer has a leakage current of 10^{-16} A, which is far superior to the Burr-Brown device, it was designed for d.c. work and tended to be unstable with pulses. In addition, the integration process itself tends to make pulse height analysis more cumbersome.

Besides the integrator approach, there are also several promising products in a class of design known as 'transimpedance' amplifiers. Some examples of these are the OPA128BM (Burr-Brown) and the LMC6001 (National Semiconductor, Santa Clara, CA). We plan to evaluate these 'transimpedance' amplifiers as time allows. Also planned is a study of the EML integrator-electrometer with the objective of improving its frequency response.

FREQUENCY ANALYSIS

To limit the low frequency noise as much as possible, it is first necessary to know the exact amplitude distribution, or spectra, of this noise. Most spectrum analyzers deal with frequencies in the mega-hertz range, but we were able to acquire a low frequency spectrum analyzer suitable for this investigation. This allows a precise identification of the frequency(s) causing the most difficulty. Knowing this, specific corrective action can be taken, e.g., additional mechanical isolation to further reduce the effect of building vibrations, or a different FET if the spectra reveals noise that is inversely related to frequency (so called 1/f or 'popcorn' noise).

SPECTRAL SHIFTING OF THE SIGNAL

As previously mentioned, one objective is to determine if the signal can be shifted to higher frequencies, away from the principal noise at low frequencies. Stated differently, how can the ionization from the alpha particle be collected more quickly? An obvious answer is to increase the intensity of the electric field used to collect the ionization. Under ideal conditions, the maximum electric field below the region where charge multiplication (breakdown) starts is about 30 kV/cm. While this might be attempted in a laboratory environment, a more practical number for remote equipment is well below this figure. Baltzer et al. (4) have determined that a reasonable electric field intensity is about 10 kV/cm and even with this reduced value, the air had to be dried. In addition, because of the wire grid they used, the 10 kV/cm field value is very close to the wires, while the average electric field is about 1 kV/cm. For their configuration, the authors observe a risetime of 0.5 ms for the collected ionization.

The risetime can be estimated from $d = KEt$, where d is the distance traveled in cm, K is the mobility of the ion and has units of cm^2 per V-s, E is the electric field in V/cm, and t is time in seconds. The mobility, K, has been reported ranging from 1.4 to 1.8 (4,6), although the value of 1.8 appears to be specifically for ^{218}Po. Since this calculation depends on geometry, consider the AUTORAMP alpha filter which will be a simple cylinder. If the maximum energy of the alpha particle is about 5 MeV, then the distance this particle will travel in ordinary atmospheric air is about 4 cm. With a cylindrical collecting electrode diameter of 21 cm, a filter diameter of 13 cm, and a maximum electric field of 10 kV/cm, the average electric field, E, is about 8.5 kV/cm. Using a value of 1.4 for K, and d = 4, the collection time, which is a fair measure of the risetime, is found to be 0.3 ms.

While collecting the ionization from the alpha particle in 0.3 ms would put the signal away from much of the low frequency noise, the voltage required is rather high. The voltage is calculated by using the relationship $V = E\{r[\ln(b/a)]\}$; where r = a = 6.5 cm, b = 10.5 cm, and E = 10 kV/cm. This results in a voltage of 30 kV, which is of considerable concern for remote equipment, thus, prompting further investigation.

One way of reducing the voltage is to reduce the distance between the filter and the collecting electrode. This in turn requires that the distance traveled by the alpha particles also be reduced by increasing the density of the air. If, for example, the density was increased by a factor of two, the 4 cm spacing would be reduced to 2 cm, and the required voltage would be just under 10 kV. Initially, one might think that this would also be beneficial to the collection time because the distance has been halved from 4 to 2 cm. However, the ion mobility, K, is inversely proportional to the collision frequency, which is directly proportional to density. Therefore, K is also halved and the collection time remains unchanged. Also, as it requires a compressor and air pressure seals, increasing the density is not readily done.

At this time, the approach under consideration is a series of concentric multi-wire electrodes between the filter and the outer solid electrode 4 cm away. Alternate electrodes are set to a positive voltage potential, while the remaining electrodes are at zero or ground potential. In effect, the collection space with one pair of electrodes is now subdivided into several. The volume of the wire electrodes is made small compared to the total volume so that the chance of the alpha particle striking a wire electrode is small. This geometry could lead to a significant reduction in the collecting voltage and faster collection times. A more detailed explanation appears in the following sections.

PRELIMINARY ALPHA SPECTROMETRY STUDIES

The first test chamber using wires was designed with four layers of wire electrodes. The layers are spaced 1 cm apart and the spacing of the wires on each layer is also 1 cm. The diameter of the wires is 1 mm. Designating the layers, starting at the bottom, from 1 to 4, layers 1 and 3 are joined and connect to the collection voltage. Layers 2 and 4 are also joined and connect to the preamplifier, which, due to the virtual-ground of a feedback amplifier, is at 0 V (ground). With this arrangement, the maximum distance traveled by the ions is 0.5 cm.

To evaluate the performance of this chamber, an alpha source was placed below a small hole in the bottom plate. The hole is used to collimate the emitted alpha particles so that most of them will travel in a direction normal to the lower plate towards the upper plate. The assembly was then placed in a larger chamber and connected to the preamplifier and a collection voltage. The results were far too noisy. The noise was traced to the way the layers were connected to the collection voltage and preamplifier and also the current leakage across the insulator blocks supporting the wire electrode layers. While the connection problems could have been addressed with a more rigid mechanical arrangement, the leakage-current problem requires yet another low current technique known as'guarding'. To implement 'guarding' necessitates the elimination of the insulator blocks. As this would require a major design effort, in the interim, another method that does not use wires was investigated. In this experiment, the wire layers were replaced with perforated metal sheets to achieve the multi-electrode geometry. While this is suitable for development, the open space area is considerably less than that obtained with wire layers, causing losses when the alpha particle impacts on the perforated metal web. However, the wire layers are very tedious to assemble as each wire in the layer must be installed one at a time. In addition, the wires must be tightly held to reduce microphonics, and all of the wires on each layer must be electrically connected together. Since all this effort is not required with perforated metal, we plan some additional evaluation and perhaps using hexagon perforated metal which has higher values of open space.

For this configuration, although an electrical connector is used, there is a heavy metal bracket joining the chamber containing the perforated metal electrodes to the amplifier housing, preventing any mechanical movement of the connector and insuring a low level of microphonic noise. To isolate the assembly from building vibrations, it is placed on top of layers of bubble wrap, a heavy steel block, and soft foam. To further minimize electrical noise, the measurements are made in an electrically shielded room (Faraday cage). For these experiments, the source of alpha particles was obtained from the decay of ^{222}Rn gas that was injected into the test chamber. Fig. 2 shows the frequency spectrum over the range of 0 to 50 kHz using a logarithmic vertical scale. The series of evenly spaced spikes are harmonics from the switching regulator used in the high voltage power supply, and indicates still another noise source that may be problematic. As can be seen in Fig. 2, the signal from the collected ionization is at lower frequencies. Fig. 3 is the same as Fig. 2, but taken over a lower frequency range of 0 to 5 kHz. The two traces, which are barely visible in Fig. 2, are now clearly discernable. The upper trace was taken with a higher electric field intensity than the lower trace, and shows that the signal is starting to shift to higher frequencies. There is a pronounced peak at approximately 500 Hz, whose origin is not from microphonics, building noise or signal content, but, if necessary, this spike can be eliminated by filtering (e.g., ‘notch filter’).

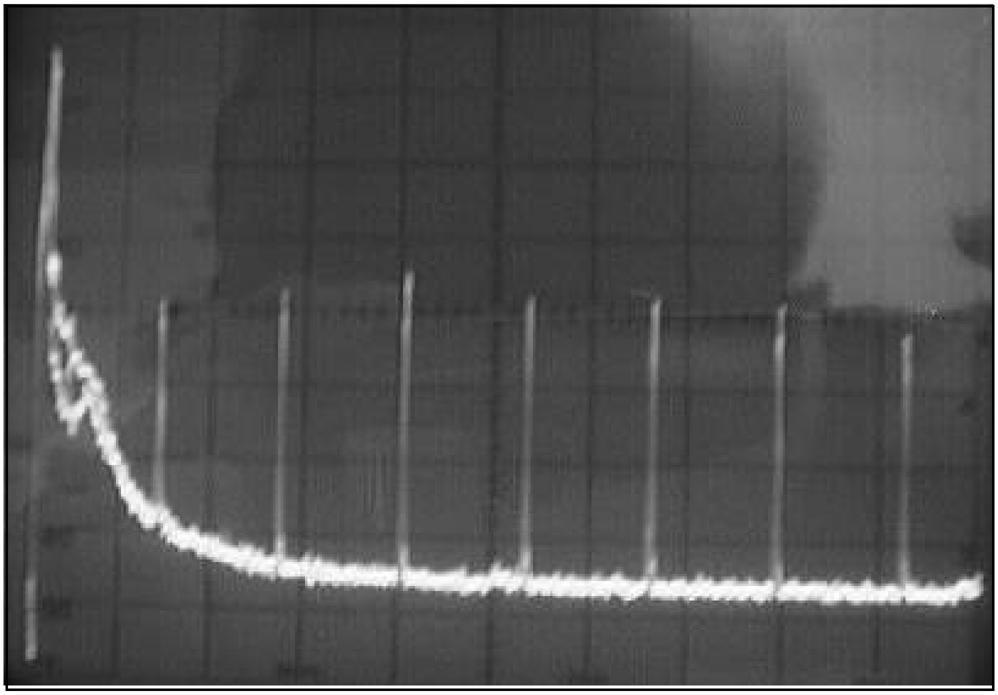

Fig. 2. Frequency spectra from 0-50 kHz. Note the evenly spaced harmonics from the d.c.- d.c. converter of the high voltage supply.

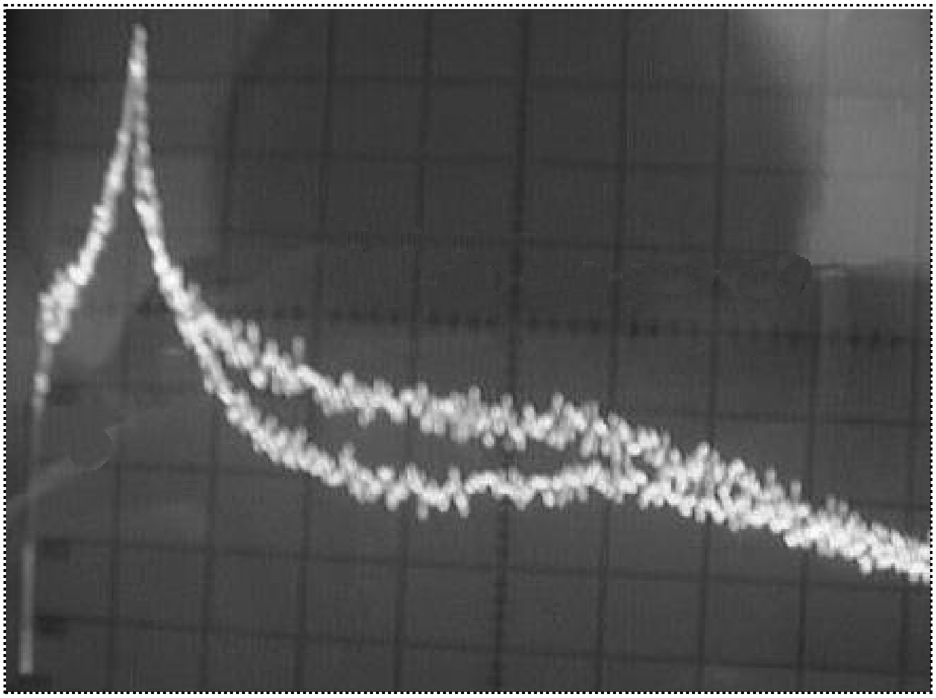

Fig. 3**.** This is the same spectra as Fig. 2, but the frequency range is 0-5 kHz. The upper trace shows some shifting to higher frequencies

PLANNED SPECTROMETRY STUDIES

The assembly that uses layers of wire electrodes and incorporates 'guarding' to eliminate insulator leakage is complete and is shown in Fig. 4 with most of the wires removed for clarity. Each layer of wire is separated with oversized phenolic insulators that help to prevent electrical breakdown at high values of the electric field. In addition, above and below these oversized insulators are 'guard' electrodes fabricated from copper clad circuit material. All of the leakage current across the phenolic insulators, flows through these 'guard' electrodes and, therefore, is totally eliminated from the ionization current collected by the wires. Once the preamplifier is attached to this assembly, it will be evaluated in the same manner used for the previously discussed assembly (no 'guarding'). These tests will help establish, the collection voltage and electrode arrangement. It is also anticipated that computer modeling of the electric field between the electrodes will be required to optimize the wire diameter and spacing.

The filter media also remains to be selected. Preliminary investigations suggest that Nuclepore with a 0.4 micron pore size or Millipore type: AW06 or FHLP, both with a pore size of 0.5 microns, may be suitable (7). When the filter media is selected, the specifications for the new air pump can be determined.

The final phase of this work will be the design and integration of the pulse ionization chamber with the AUTORAMP robotic assembly.

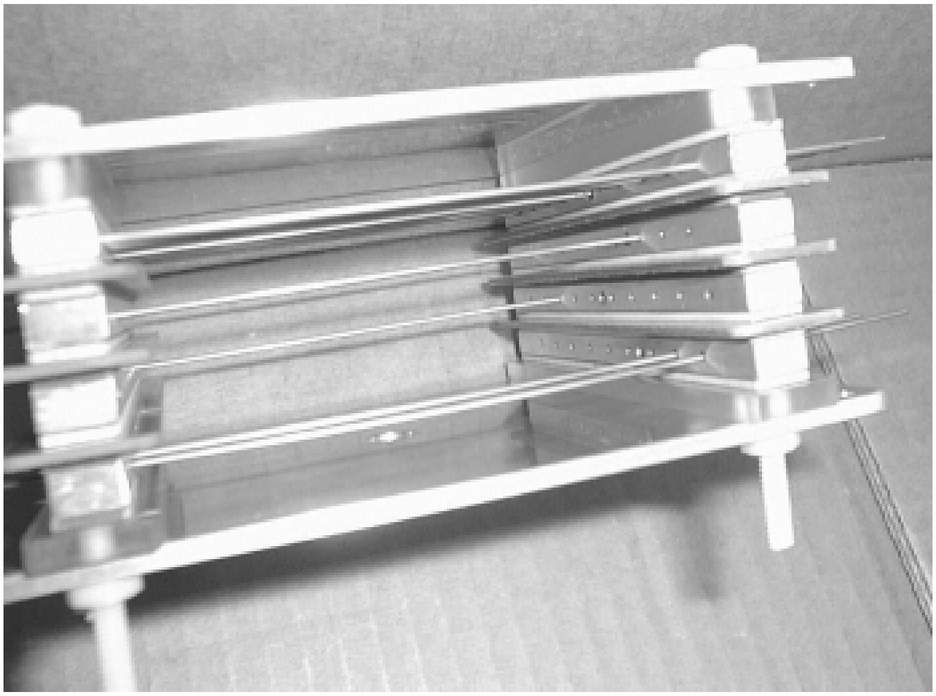

Fig. 4. This assembly uses 'guarding' which eliminates insulator leakage current from the ionization signal. The 'guard' electrodes are placed above and below the oversized insulators.

CONCLUSION

The preliminary results, along with the work of others who have obtained alpha spectrometry with air ionization chambers, are encouraging and show that this project can be successfully completed. As discussed, the assembly of the wire electrode layers is difficult, making the perforated metal approach attractive, provided that the associated signal loss is acceptable. Some observed shifting of the signal to higher frequencies is not nearly sufficient to relax the precautions required by the low frequency noise, but perhaps the proposed electrode arrangement and computer study of the electric field will lead to significant strides in this area.

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