CONF-720635--1

RECEIVED BY TIC JUL 12 1972

Construction of Miniature, Low-Level Proportional Counters*

Fred H. Kummer, R. W. Stoenner, and Raymond Davis Jr.

Chemistry Department, Brookhaven National Laboratory, Upton, New York 11973

MASTER

INTRODUCTION

This article describes the design and details of construction of some low level proportional detectors with very low background counting rates that have been found highly useful for counting very small amounts of argon-37, argon-39, argon-42, carbon-14, tritium, and radon-222. With these counters, the radioactive isotopes of argon and tritium have been measured in a large number of meteorites, particularly in fresh falls.¹ These same isotopes have been extracted from lunar samples from the Apollo Missions XI, XII, XIV, XV, and XVI.²⁻⁴ With the exception of radon-222, which is a product in the natural decay series of uranium-238, these isotopes are the result of interaction of high energy cosmic ray and solar flare particles with the material of the meteorite or the lunar surface. In the search for the solar neutrino, a tank containing 100,000 gallons of perchloroethylene (cleaning fluid) has been installed in the Homestake Gold Mine in South Dakota.⁵ Interaction of neutrinos with the chlorine atoms should yield argon-37, which is removed from the tank by sweeping with helium. The argon is separated from the stream of helium, and after purification is counted in a detector of the type we are going to describe.

These detectors have a number of design features which make them particularly useful in experiments where very small amounts of gaseous radioactivities are to be measured. The materials of construction are carefully

selected to be as free as possible from naturally occurring radioactive contaminants. The envelope is constructed from fused silica, optical grade, so as to avoid incorporating any potassium-40 in the body of the counter. The metals used for fabricating the cathode are of the highest purity available and in general are made from zone refined stock. The counter contains no plastics or porous surfaces where gas might be absorbed. It is readily bakeable and in fact is routinely baked at a temperature of 250°C while evacuating. We have designed the counter so that the total volume is quite accurately defined by two capillary columns of mercury. The inactive volume is kept to a minimum in order that the counting efficiency be as high as possible. Even the smallest of these miniature counters (0.2 cm^3) has a counting efficiency approaching that of conventional internal gas counters. In order that pulse height analysis may be used to enhance specificity and reduce background counting rates, the end plug is provided with a very thin window ($\sim .002''$) through which radiation of known energy may be directed for purposes of calibrating the energy scale of the pulse height analyzer. Finally, the same general design has been used with minor modifications to construct counters of widely varying sizes, from 0.2 cm^3 to 12 cm^3 .

CONSTRUCTION

The techniques found to be most successful in fabricating these counters follow.

Section A (Figure I)

The counter body consists of the silica envelope containing the cathode cylinder, center wire, insulating plug, metal ring and the end plug with a thin window. Depending on the size of the counter, a regular wall tube is selected (Figure IIa) (example 8 mm O.D. for a 0.5 cm^3 volume). With a standard tapered carbon reamer a female taper is formed at one end.

There should be no constriction or bulge between the conical and the cylindrical portion.

Using a 1 mm capillary tube a male taper is made to match the female taper (Figure IIa). In this process the opening at the bottom of the taper is constricted to 0.5 mm.

The tapers are then ground to mate. The bottom of the male taper is then cut off to coincide with the interface of the tube and the start of the female taper. Next a groove of about 0.5 mm wide and 0.5 mm deep is ground along the length of the male taper (Figure IIa). This is accomplished using a drill press and a handy grinder with a small carborundum wheel used as a grinding tool. The taper is held in the drill press, the handy grinder mounted in a drill vise. With the aid of a stop, against which the drill vise is resting, the carborundum wheel is lined up with the taper. With an up and down motion of the taper, at the same time applying slight pressure to the drill vise, the groove is ground.

After these operations, the male and female tapers are cleaned with hydrofluoric acid and rinsed with distilled water. When dry, the male is inserted into the female and they are fused together under vacuum (Figure IIb). Fusing is done progressively starting from the top. As one approaches the edges of the groove and the lower end of the tapers, the vacuum is disconnected to avoid sucking into the groove or sucking in below the male member. After cooling one cuts off the top of the fused tapers exposing the two openings (Figure IIc). To each of these a 3 mm x 1 mm tube is sealed.

In preparing the insulating plug and the end plug, attention should be given to the close fit of both plugs in the envelope containing the cathode. The insulating plug (Figure IIc) is made of 0.5 mm capillary tubing and cut to a length of 3 mm to 5 mm depending on the size of the counter.

The end plug (Figure IIe) is faced off with a window ~ 0.002 " thick. In fabricating this end plug, the following method is used. A standard wall tubing of approximately six inches long is ground to fit the envelope. One end is cut straight and cleaned. To make the thin window, a large bubble is blown, having a thickness of approximately 0.002 ". To measure this thickness, openings are provided in the bulb to make possible the use of a ball micrometer. It might become necessary to repeat this procedure to obtain the desired thickness. As the next step, the end plug tube is heated at the cleaned end and tacked to the selected portion of the thin bubble. The excess glass around the tube is then burned and picked off. Finally the edge of the window is carefully fused. To improve the physical strength of the window in respect to pressure differences on the two sides of the window, it is given a slight concave geometry. Before the end plug is cut off to the indicated length, it is tested for leaks on a helium leak detector.

Section B (Figure I)

The requirements for this section of the counter consist of: two spring loaded 1.5 mm bore filling stopcocks, two graded seals 5 mm O.D., quartz to Corning #7720, and two beaded tungsten sealthroughs. The intermediate glasses used in the graded seals are: Corning #723-720-724-7740 and 5420. Due to the high radioactivity, Corning glass #3320 (uranium) should not be used in the graded seal. The two graded seal tubes are identified as the anode leg on the left and the cathode leg on the right. Employing conventional glass blowing, these parts are then joined into Section B, as shown in Figure I.

Cathode

A number of different metals have successfully been used as cathodes. Considerations which must be made in the choice of cathode material concern

the purity of available metals, the manner in which the cathode lead may be attached, and the gas which is to be counted in the completed detector. A large variety of zone refined metals of 99.999% purity in rod form are now available. We have constructed counters with cathodes of aluminum, titanium, iron and copper. If tritium is to be measured in the counter, a metal must be chosen which does not form a hydride and in which the solubility of hydrogen is very low. Copper has been used quite satisfactorily for this purpose, while iron on the other hand would be a very poor choice.

The cathode is machined from rod stock. The cathode cylinder is first drilled to the approximate internal diameter, after which the final I.D. is obtained by using an appropriate reamer. If necessary, the internal surface can be polished with emery. The outside diameter is determined individually for each silica envelope after allowance for cathode expansion upon heating to 250°C. A washer or ring, which is used to anchor one end of the center wire, is machined from the same stock to the same outside diameter. The diameter of the hole in the washer is not critical, but must be larger than the hole in the silica insulating spacer.

Assembly

Prior to assembly all parts are thoroughly cleaned. The metal pieces are washed with hexane to remove any cutting oils used in machining, after which they are immersed in a suitable etchant (dilute acid) for a sufficient time to dissolve away the outer surfaces. Following this they are rinsed with distilled water, then ethyl alcohol, and dried in a rapid stream of nitrogen. The silica parts are likewise washed with hexane to remove any oil coating, and then are immersed in concentrated hydrofluoric acid for a few minutes, rinsed thoroughly with distilled water, and dried in a drying oven. The sealthroughs are obtained commercially and consist of a 2.5 cm

length of .025" tungsten rod, to one end of which is butt-welded a 3 cm length nickel rod of the same diameter, while the other end has attached a length of stranded copper wire. The sealthrough is prepared by fusing on a 1 cm portion of the tungsten rod a layer of Corning No. 7720 glass. No cleaning of the sealthroughs is required.

The cathode is installed in the envelope first. Connection from the cathode cylinder to the sealthrough is generally made with .003" stainless steel wire, a generous length of which is passed through the cathode leg, through the counter body and out the end. The nickel portion of the sealthrough is shortened to about 0.5 cm and the end is flattened to provide a knob. The stainless steel wire is secured to the seal-through by a simple knot pulled taut against this knob, while just beyond the knot the wire is spot welded to the nickel rod and the excess is trimmed to this point. Connection to the cathode has been made in a number of ways, and is determined in part by the cathode material. An iron cathode presents the easiest case in that the stainless steel wire is trimmed to the proper length and by advancing the lead assembly into the cathode leg as far as it will go, the end of the wire protrudes from the counter body so that it may be spot welded to a flat filed in the outer surface of the cathode cylinder. The entire cathode assembly is drawn the reverse direction into the counter body and the glass bead should now be in proper position for sealing in the cathode leg. Connections to aluminum, titanium, and copper cathodes are not readily spot welded, so other means must be employed. One method which has been used involves spot welding the stainless steel wire to a small tab (2 x 10 mm) of .002" iron foil slightly arched to provide a spring action. This tab is placed in between the cathode and the silica envelope, and serves adequately as electrical contact. When this method is used, proper allowance must be made when machining the outer diameter of the cathode.

If the counter is to be used to measure tritium activity, a copper cathode is used, and it is best to eliminate the use of ferrous metal as well as other materials which exhibit a significant hydrogen solubility. A .001" tungsten wire is used in place of the stainless steel. The copper cathode has drilled in the cylinder wall longitudinally a 1/32" hole 1/4" deep. From the outside of the cylinder a hole is drilled to meet the bottom of the first hole. Connection to the cathode is made by threading the tungsten wire in the end hole, out the side, back through once more, and finally the wire is tied with the knot concealed in the hole in the outside of the cathode cylinder.

Generally the center wire is 0.001" tungsten, however counters have been made with 0.0005" tungsten wire. As was done with the cathode wire, a generous length of center wire stock is passed through the anode leg, through the body of the detector and out the end being very careful that it is not kinked in the process. As shown in Fig. 1, a molybdenum spring is spot welded to the nickel butt of the anode seal-through, and the center wire is secured by tying and spot welding to the hooked end of the spring. The spring serves to keep the center wire straight and taut and thus the resolution of the detector is maintained at the maximum level. After this attachment has been made the silica insulating spacer is threaded on the center wire protruding from the counter body and inserted in the envelope in position against the cathode cylinder. With the seal-through assembly in proper position for final sealing the wire is kinked where it exits the insulating spacer, for it is at this point that the center wire is secured to the metal washer which is used to anchor this end of the center wire. Attachment as before is by tying a knot around the washer and spot welding the free end close behind the knot, after which the excess wire is trimmed

away. The center wire assembly is then drawn back into the counter body with the washer lying flat against the insulating spacer, and the end plug is inserted in place. The counter is now ready for final sealing.

To facilitate sealing the beaded seal-through into the anode and cathode legs the counter is held in a vertical position with the seal-through hanging down. Care must be taken to keep the anode center wire taut while sealing, and to accomplish this a small weight is suspended from the anode lead during this operation. With a hand torch, using a small tip, the seals are then made to the previously beaded leads.

To complete the assembly, the final seal of the end plug to the counter envelope is made. The end plug, when inserted, extends approximately 1 mm beyond the end of the envelope to enable one to make the usual Dewar seal. The end plug is first lightly tacked after which the clearance between the plug and the metal washer is checked. The clearance required depends on the cathode metal and the length of the cathode, and should be sufficient to permit heating the counter to 250°C, but should not be excessive since this would increase the inactive volume of the counter and lower unnecessarily the counting efficiency. For a counter of the dimensions shown in Fig. 1, and with an iron cathode, a clearance of about 0.3 mm is customarily used. Since the plug is not tightly sealed as yet, this clearance can be adjusted as necessary. The final Dewar seal is then completed using a small flame and at the same time keeping ^{the}counter body cool by wrapping with ^{asbestos}wet/paper to prevent the oxidation of the washer and cathode cylinder. An atmosphere of forming gas (90% N₂, 10% H₂) can also be used. After completion, a final leak test with a helium leak detector is made.

After a thorough baking and evacuation the counter is ready for filling. These detectors have been used with a variety of counting mixtures. Argon

is counted with the addition of a small amount of methane (0.5 to 10%). Radon is counted with P-10 (90% Ar, 10% CH₄). Tritium is counted in a mixture of P-10 and hydrogen. Carbon-14 is counted as carbon dioxide with nothing added. Filling pressures may be as low as 0.5 atmosphere and routinely go as high as two atmospheres. Employing conventional vacuum line techniques, the filling gas is pushed into the counter with mercury to the point where both the anode and cathode legs are filled with mercury up to where the capillaries enter the counter body. Details of performance, counting efficiencies, and resolution will appear in a subsequent publication.

REFERENCES

- * This work was carried out under the auspices of the U.S. Atomic Energy Commission.
- ¹ Forman, M. A., Stoenner, R. W., and Davis, R., Jr. Cosmic Ray Gradient Measured by the Argon-37/Argon-39 Ratio in the Lost City Meteorite. J. Geophys. Res., 76, 4109-20 (1971).
 - ² Stoenner, R. W., Lyman, W. J., and Davis, R., Jr. Cosmic Ray Production of Rare Gas Radioactivities and Tritium in Lunar Material. Proc. Apollo 11 Lunar Science Conf. 2, 1583-94 (1970).
 - ³ Stoenner, R. W., Lyman, W., and Davis, R., Jr. Radioactive Rare Gases and Tritium in Lunar Rocks and in the Sample Return Container. Proc. 2nd Lunar Science Conf. 2, 1813-23 (1971).
 - ⁴ Stoenner, R. W., Lindstrom, R. M., Lyman, W., Davis, R., Jr. Argon, Radon, and Tritium Radioactivities in the Sample Return Container and the Lunar Surface. Geochim. et Cosmochim. Acta Suppl. In press.
 - ⁵ Davis, R., Jr., Harmer, D. S., Hoffman, K. C. A Search for Neutrinos from the Sun. Phys. Rev. Lett. 20, 1205-1209 (1968).

ACKNOWLEDGMENT

The authors would especially like to acknowledge the many contributions of Mr. Karl Walther in the work described here. In the course of the development of the counters he has advanced many useful suggestions toward the design and construction techniques.

NOTICE

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Atomic Energy Commission, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

FIGURE CAPTIONS

Fig. I. Counter assembly for 0.5 cm^3 counter.

Fig. II. Construction details for 0.5 cm^3 counter.

COUNTER ASSEMBLY FOR 0.5 cm³ COUNTER

CONSTRUCTION DETAILS FOR 0.5 cm³ COUNTER

FABRICATION OF COUNTER BODY

CONSTRUCTION OF THE INSULATING AND WINDOW PLUG