De Vries and Barendsen used carbon dioxide as counting gas[2]. According to their report if carbon dioxide were mixed UP with 10^{-3} mmHg of oxygen, the counter would be in bad condition. But their method is much complicated. We use a simple way by driving- away carbon dioxide many times until the trace of impurity is completely replaced.

II; COUNTING APPARATUS

1. Counter

A sketch of the counter system is shown in Fig. 1. The center counter is made of copper tube, the inside diameter of which is 4.4 cm and the length is 60cm. The total volume of the center counter is approximately 912 cm^3. The center wire is 0.005 cm. diameter of stainless steel. Vacuum tightness is particularly important, since no trace of air or water vapor can be tolerated in the filling gas.

2. Shielding

The outer counter is made of stainless steel pipe of 12.8 cm in diameter, 75 cm long, with 20 stainless steel wires which are supported by acryl resin rings, and it forms the anticoincidence shield. The center counter is put inside the outer counter. The whole counting system is shielded by iron plates of 25 cm thickness, as reported in the previous work.

3. High-Voltage Power Supply

The power supply delivers an electronically regulated voltage variable from 1 kv to 6 kv and the ripple is about 0.1 mv.. 0- to 6 kv voltmeter is used to

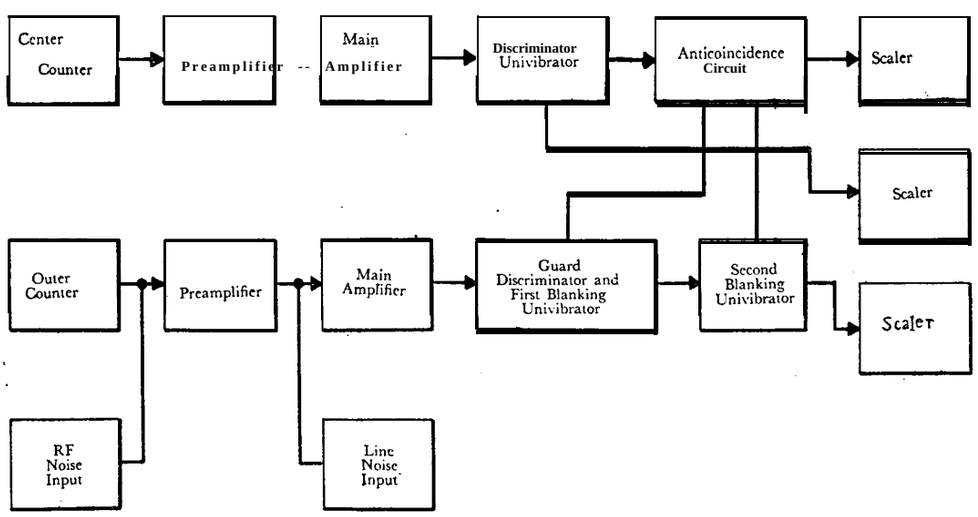

Fig. 2. Block diagram of the electronic system

(2) HI. de Vries and G. W. Barendsen, Physica, 19, 987 (1953).

indicate the voltage supplied to the counter.

4. Electronic Equipment

As shown in Fig. 2, the pulses from the center counter are sent to amplifying circuit of gain of 10,000, which amplifies the signal pulses up to 12 v. The pulses from the outer, after being amplified 10,000 times by the amplifier, are fed to the guard discriminator and the first blanking univibrator which triggers at about 0.5 mv referred to the preamplifier input. The first blanking univibrator feeds a 600 microsecond, 8 volts pulse to the anticoincidence circuit and to the second blanking univibrator while the second blanking univibrator feeds a 400 microsecond, 8 volt blanking pulse to the anticoincidence circuit and also to the guard output. Thus, for each cosmic ray or noise pulse in the guard channel, both counters are blanked for 600 microsecond +400 microsecond, that is, 1 msec in total. Activation of the anticoincidence circuit by the center counter pulses is delayed by 300 μsec with respect to the arrival of a coincident guard discriminator and a first blanking univibrator pulse. The second blanking univibrator pulse keeps the anticoincidence circuits blanked out while the guard discriminator and first blanking univibrator recover. This prevents sneak thrugh of closely spaced cosmic ray or noise pules.

III. PREPARATION OF CARBON DIOXIDE

1. Preparation of Dead Carbon and Sample

Extraneous materials are removed manually from the sample, and the sample is converted to carbon dioxide by acid treatment if it is a carbonate, or by combustion if it is an organic. The carbon dioxide is absorbed in ammonium hydroxide, then pure calcium carbonate is precipitated by the addition of a warm calcium chloride solution. The precipitate is washed until free from the chloride ion, and filtered off and then dried. Silver nitrate has to be used to test the chloride ion. Approximately 50 gr of the precipitate of calcium chloride is washed with 30 *l* of boiling water. Then approximately 15 gr of the pure calcium carbonate is placed in quartz tube which is kept at a temperature of 500 °C. The sampling system, as shown in Fig. 3, is evacuated to 10^{-3} mmHg for five hours. Then the carbon dioxide is liberated by raising the temperature to 750°C, and the water vapor is removed from the gas by passing through a dry ice trap. During an evacuation time about 10 minutes, the water and oxygen are effectively removed from the system with a little loss of sample. The pure carbon dioxide is then evolved from the calcium carbonate by raising the temperature up to about 900 °C. The gas is collected under continuous evacuation in a liquid air trap. After collection, the carbon dioxide is allowed to vaporize from the trap into the bulb. Before introducing all the gas into the bulb, the bulb is washed three

times by the gas itself. After introducing the carbon dioxide, the sample bulb which is maintained at liquid air and the previously evacuated counter are both re-evacuated to a pressure less than 10^{-5} mm Hg. Thus we can assure the complete removal of the residual gases. The sample bulb and the counter are then closed off from the vacuum, and then the carbon dioxide is allowed to vaporize into the counter.

2. Preparation of Modern Carbon Standard

We dissolve 30 gr NBS oxalic acid standard into 300 ml water in a flask and mix it with 160 cc of 6N sulfuric acid. Then we let the oxygen gas flowed into the solution with dripping 550 cc of 4% potassium permanganate at the same time. The carbon dioxide evolved is absorbed in ammonium hydroxide. The remaining procedure for preparing the carbon dioxide is the same as described in the above paragraph.

IV. RESULTS AND DISCUSSION

1. Characteristics of the Counter Filled with Pure Carbon Dioxide

Through the present experiment, the operating voltage was set at 3.8 kv under one atmospheric pressure. The starting voltage of the plateau is

$$V = (2100 + 2P) \text{ volts}, \quad 360 > P > 860,$$

where P is in units of mmHg. The characteristic curves of the center counter

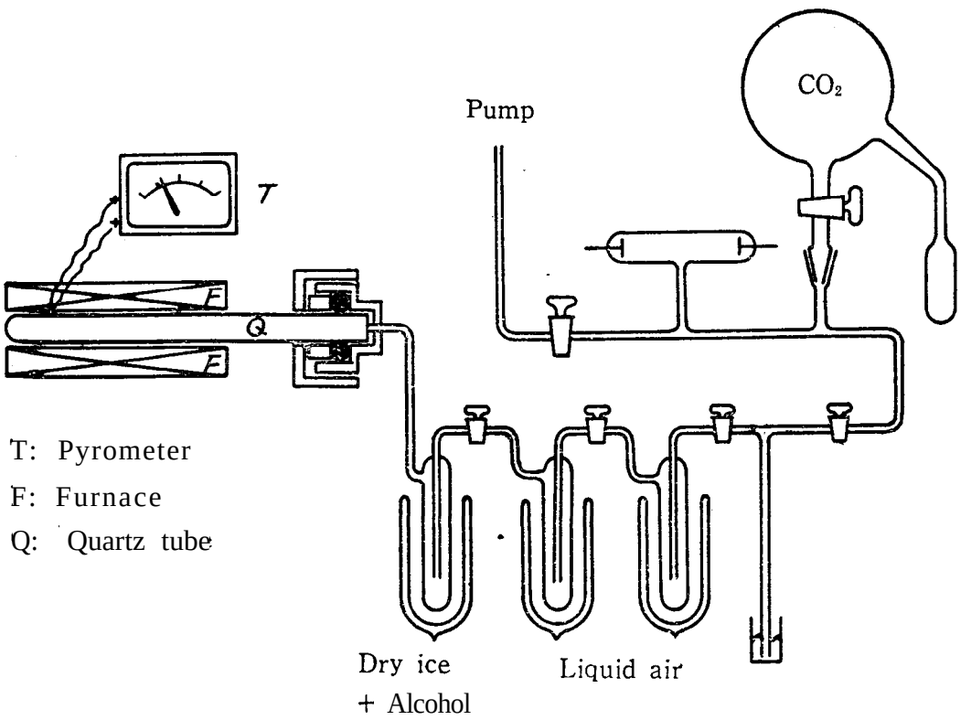

Fig. 3. Apparatus for preparation of carbon dioxide

and the outer counter for the pure carbon dioxide, methane and propane gas are shownin Fig. 4.

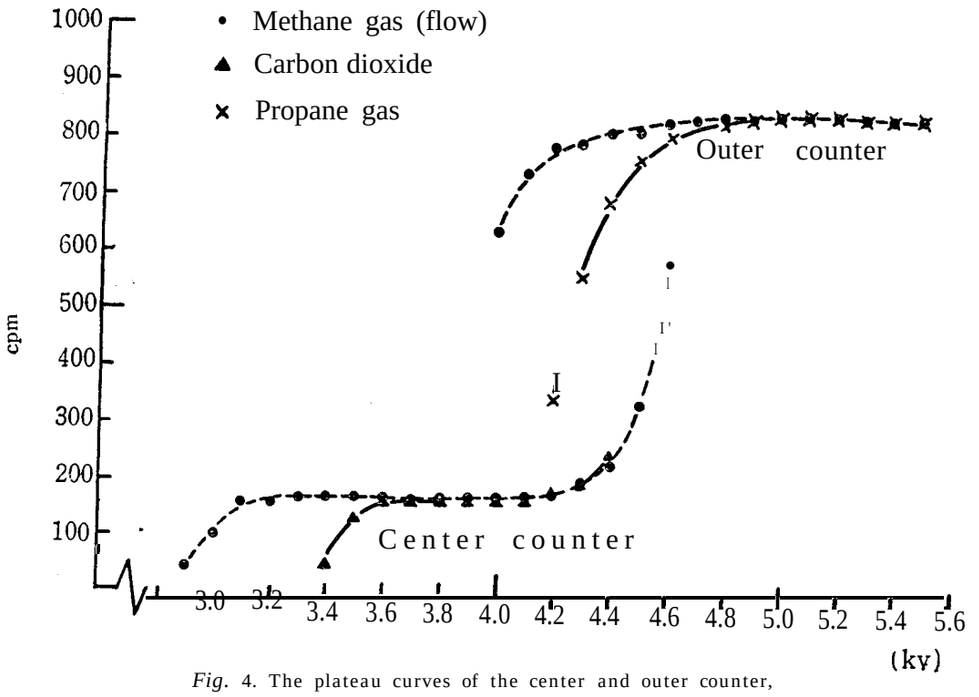

Fig. 4. The plateau curves of the center and outer counter,

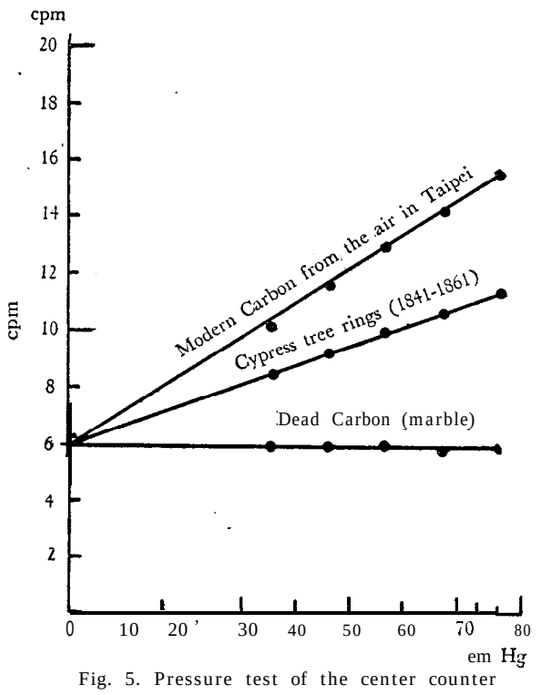

Fig. 5. Pressure test of the center counter

Fig. 5 shows the number of counts per minute for the different amounts of carbon dioxide of the modern and dead carbon. There is no change in the number of counts per minute in the case of the dead carbon. This indicates that the carbon dioxide produced from the dead carbon has no activity. The extrapolated values of counts per minute to the zero pressure for the modern and dead carbon show the same value.

We get the reproducible results at operating voltage 3.8 kv with the background of 5.87± 0.01 cpm and a counting time of 48 hours. In our experiment, carbon dioxide of modern carbon stored more than

three weeks is filled in the center counter and its characteristic curve is determined. We do not find radon effect. According to Hamada[3], the center counter increased the number of counts by 0.1 cpm on each time of charging the counting gas. In our case, however, any increase was not found.

Fig. 6 shows the counting rate of the center counter with and without the anticoincidence when the applied voltage of the center counter is varied, if operating voltage of the outer counter is fixed at 5.2 kv. Fig. 7 shows the counting rate of the center counter with anticoincidence when the applied voltage of the outer counter is varied, if operating voltage of the center counter is fixed at 3.8 kv. They show that the operating voltages of the center and outer counter should be fixed at 3.8 kv and 5.2 kv respectively.

2. Measurement of Modern Carbon

It has been pointed out by several authors[4,5,6] that the atmospheric radio-

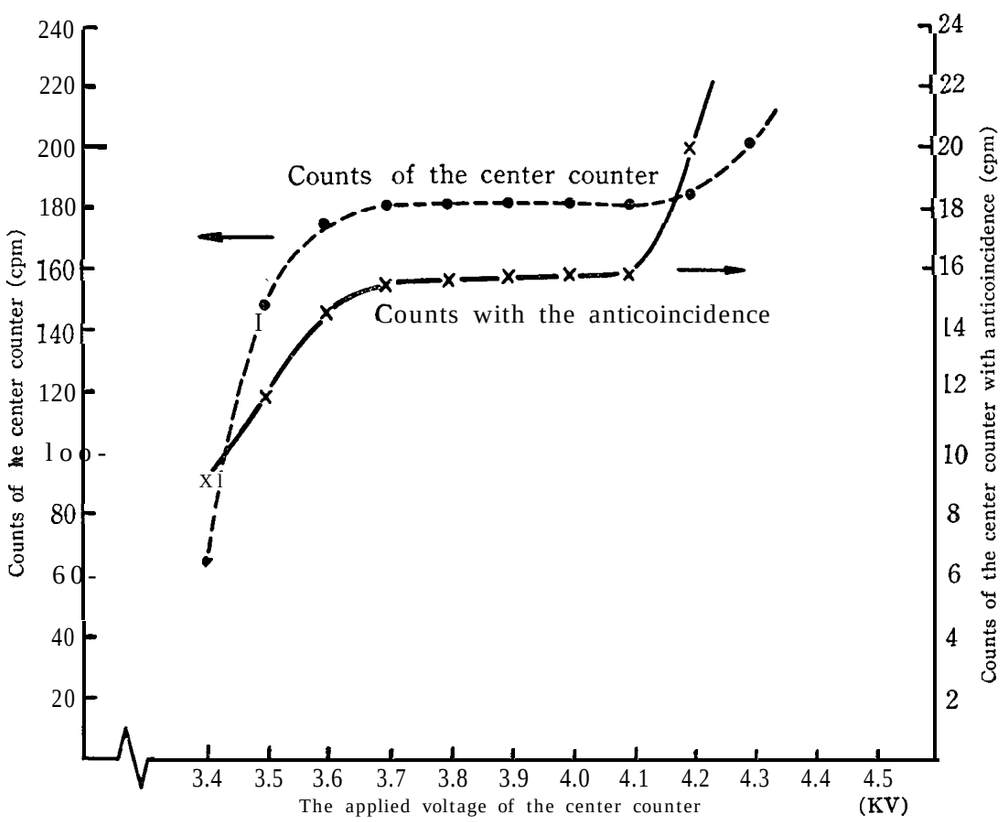

Fig. 6. Counts of the center with and without the anticoincidnce when the applied voltage is varied

(3) T. Hamada and C. Fujiyama, Rikenhōkoku (理研報告) **50,** 309 (1964).
(4) W. S. Broecker and A. Walton, Science, **130,** 309 (1959).
(5) Hl. de Vries, Science, **128,** 250 (1958).
(6) T. A. Rafter and G. J. Fergusson, Science, **126,** 557 (1957).

carbon is increasing because of nuclear tests and decreasing by Suess effect.[7,8] The value of 95% of the NBS contenporary standard for carbon-14 dating was taken as a standard of dating work. It was the mean value of radio-carbon concentration in the tree which grew in Europe in the nineteenth century.

For the check, we have measured the radio-carbon concentrations in the tree ring samples which grew in Taiwan. Table 1 shows the ratio of the radio-carbon concentrations in the wood sample to that of the NBS standard. Hamada et *al.*[3] have pointed that the atmospheric radio-carbon is decreasing when the island is surrounded by the ocean. It is obvious from the table that the radio-carbon concentration in Taiwan district is higher than the average result in Europe.

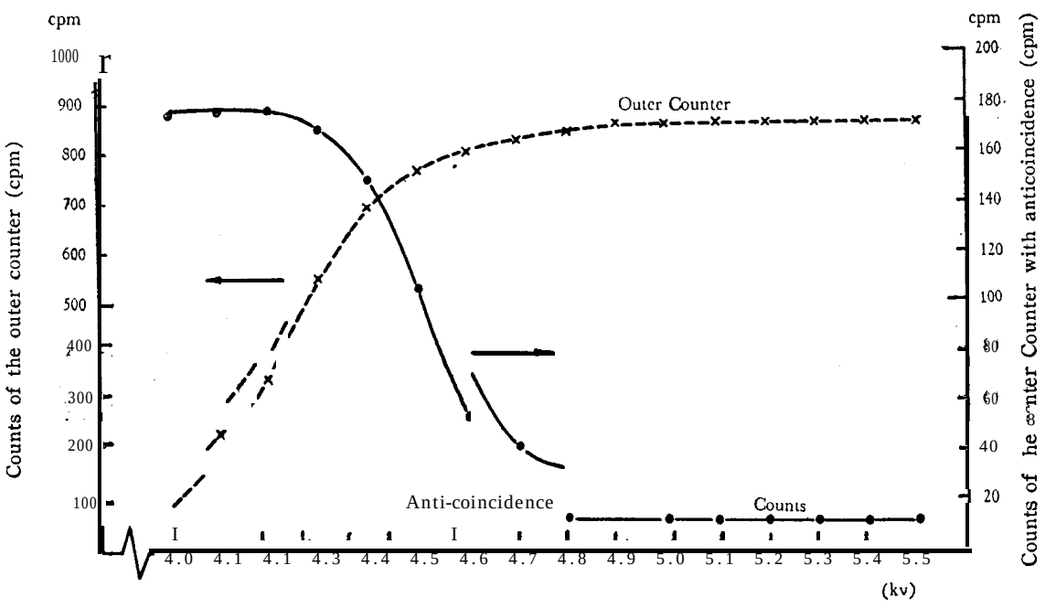

Fig. 7. Counts of the center counter with anticoincidence when the voltage of the outer counter is varied

Table I The ratio of the number of counting of the contemporary wood sample to that of NBS

Intervals of grown of tree ring	the ratio of the number of counting of the contemporary wood sample to that of NBS (%)
X41-1861	96
1861-1891	96
1911-1941	96
1949	99

(7) R. Revelle and H. E. Suess, Tellus, 9, 18 (1957).
(8) G. J. Fergusson. Proc. Roy' Soc. A243, 561 (1958).

3. Determination of Age

The method for determinining age is as follows:

(i) After reading the temperature and the pressure when the gas is charged, the pressure P_0 at temperature 0 °C is calculated. During the measurement, if a difference in temperature and pressure is found the average was taken.

(ii) A series of background tests was made of dead carbon and the NBS standard under different pressures as shown in Fig. 5. After assuring that the relation between counts and pressure is on a straight line, the background counts and the NBS counts under the pressure P_0 were determined to be N_b and N_0 from Fig. 5.

(iii) If the counting rate of the sample is N_s under the pressure P_0, the age t will be calculated as follow

$$N_s - N_b = 0.95 N_0 \left(\frac{1}{2}\right)^{t/T}$$

where T is the half life of carbon-14, which is taken as 5730 ± 40 years.[9]

(iv) To determine the statistical error, the errors caused by pressure and temperature must be taken into consideration, in addition to the counting error, that is, error of 1% of N_b, 0.5 % of N_0 and 0.5 % of $N_s - N_b$ are added

The maximum age, that can be measured is defined as the age for which the sample count is three times the statistical deviation for a counting time of 24 hours. A maximum age of 20,000 years is the practical limit at the present work. In the near future we want to make a counter of 2.5 liters, which will decrease the deviation and increase the upper limit.

V. SAMPLES DESCRIPTION

The age indicated as follows represents the one before 1950, and the name represents the place where the shell sample obtained.

NTU-1: Lungkang 226 ± 192

Mollusks from Lungkang (24°38′N Lat, 120°45′E Long), Miaoli, Taiwan, imbedded in the sand dune in gravel bed at Lungkang.

NTU-2 Lungkang 2 8,415 ± 433
6,465 B. C.

Mollusks from the level 10 m below the maine terrace surface at Lungkang, Miaoli.

NTU-3 Lungkang 3 6,822 ± 308
4,872 B. C.

Mollusks from the level 8.4 m below the marine tarrace surface at Lungkang,

(9) Harry Godwin, Nature, 195, 984 (1962).

Miaoli.

NTU-4 Arkuntien 7,532 ± 482
5,582 B. C.

Corals from Arkuntien Coral Reef at Arkuntien (23°00′*N* Lat, 120°13′*E* Long) near Tainan, Taiwan.

NTU-5 Milung 3,991 ± 269
2,041 B. C.

Corals from Milung Coral Reef at Milung (23°58′*N* Lat, 121°37′*E* Long) near Hwalien, Taiwan.

NTU-6 Taipei 4,429 ± 281
2,299 B. C.

Peat from Taipei Peat Formation, Taipei (25°02′*N* Lat, 121°31′*E* Long)

VI. ACKNOWLEDGMENTS

We are much indebted to Prof. Ting-Ying H. Ma of Department of Geology, National Taiwan University, for supplying shell samples. Also we wish to express our gratitudes to Mr. T. H. Lin for help in the preparation of the samples. This work was supported by the National Council on Science Development.

Low Background Counter for Carbon-14 Dating

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A proportional counting system for radiocarbon dating and precedures for producing carbon dioxide are described in this paper. The center counter was filled with carbon dioxide at one atmospheric pressure. The total background is 5.87f0.01 cpm. The value of 95% of the NBS contemporary standard for carbon-14 dating was taken as a standard of dating work.

I. INTRODUCTION

IN our previous report[1], we took the acetylene gas as a counting gas for the measurement of radiocarbon. Though the acetylene gas has two atoms of carbon and has a good counting characteristics, the procedure is some-what trouble-some for continuous measurement. Therefore, in this experiment, we try to use the carbon dioxide as thk counting gas.

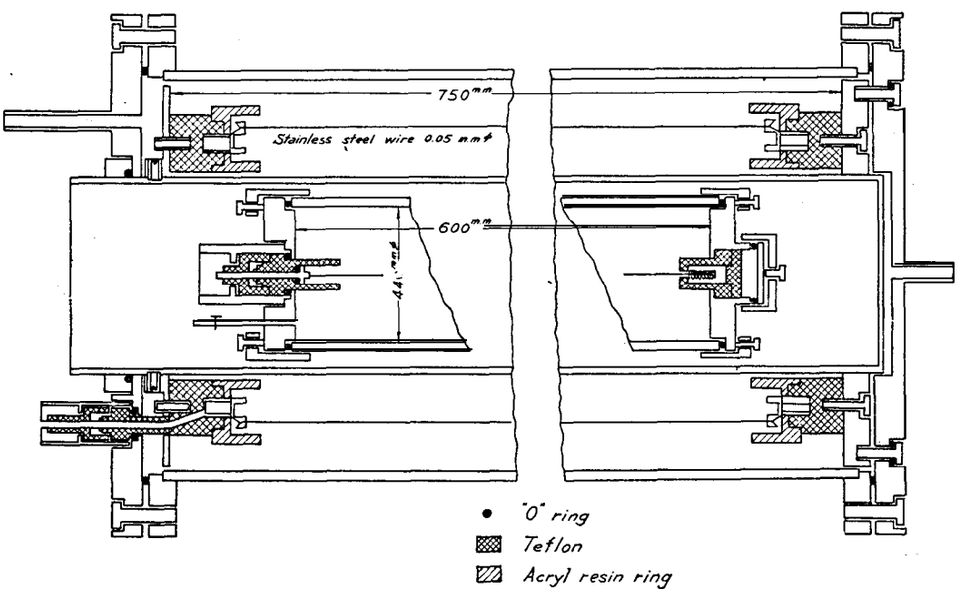

Fig. 1. Center counter and outer counter

(1) S.C. Lu, Y.K. Tai and Y. C. Hsu, Chin. J. Phys. 2, 1 (1964).