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HIGH ENERGY NUCLEAR INTERACTIONS WITH MATTER AND NUCLEAR PROCESSES IN NATURE

Annual Report for AEC Contract No. AT(11-1)3080
for the period July 1, 1972 to June 30, 1973

O. A. Schaeffer, Principal Investigator

Abstract:

Proton cross sections for the production of stable rare gases in uranium and vanadium have been extended to higher energies. The ^{39}Ar and ^{36}Cl content of the Benthullen meteorite has been measured in order to determine its terrestrial age. Experimental procedure of the activity extraction from the FeNi-phase and the stone-phase is described. The preliminary result of 75 years is much younger than expected. A low level proportional counting system for $^{37,39}Ar$ and ^{36}Cl has been set up. Background counting rates of 3 to 10 counts per day could be achieved. The activities are released from meteorites and targets by either vacuum melting or dissolving in new designed vacuum apparatus.

DISCLAIMER

A.E.C. Contract AT(11-1)3080

Annual report for the period July 1, 1972 to June 30, 1973.

TITLE: High Energy Nuclear Interactions with Matter and Nuclear Processes in Nature

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INTRODUCTION:

During the past year, studies were continued on proton production cross sections. The cross section for uranium, and vanadium were investigated.

Work was started on cosmic ray produced radioactivity and stable rare gases in meteorites. A possible old meteorite observed fall, Benthullen was studied and found to be relatively young. In the thesis of A. Chang, it is suggested that cosmic rays are not constant during the last million years as shown by comparison of $^{36}Cl/^{39}Ar$ ratio in meteorites and iron targets. The work is being extended to other meteorites.

PROTON PRODUCTION CROSS SECTIONS

Spallation of vanadium and uranium with protons.

This work is described in the accompanying reprint and preprint.

NUCLEAR PROCESSES IN NATURE

The terrestrial age of the Benthullen meteorite.

So far, the oldest observed meteorite fall for which a piece still remains in the Ensisheim fall, 1492, with a recovered mass of 127 kg. E. Chladni reports in his book "Die Feuer - Meteore" a meteorite fall

in 1368 near Oldenburg in Germany. Professor P. Ramdohr suggested that a meteorite belonging to that fall is the H - chondrite Benthullen. The best way to prove this suggestion is to measure the terrestrial age of Benthullen. ^{39}Ar with a half life of 269 years is the most suitable radionuclide to measure a terrestrial age of about 600 years. During the travel in space as a small body, the meteorites are bombarded by high energy particles of the cosmic rays. Nuclear reactions induced by these particles produce stable and radioactive isotopes different from the target elements present in the meteorites. After an irradiation time of several half lifes, the activity of a radionuclide reaches a saturation value at which as much of a nuclide decays as is produced. It can be assumed that the ^{39}Ar activity in Benthullen has its equilibration value, because cosmic ray exposure ages of less than one million years are very rarely found. The youngest so far found is about 10,000 years old. When a meteorite is captured by the earth, it becomes shielded against cosmic rays by the earth's atmosphere, and the produced radionuclides start to decay.

The main target elements for the production of ^{39}Ar are iron, potassium and calcium. The saturation value for the H - chondrites can be taken from the numerous measurements in the literature. Fourteen H - chondrites investigated by different authors have a mean ^{39}Ar activity of 8.6 dpm/kg at the time of fall. The highest value is 11.7 dpm/kg, the lowest 6.7 dpm/kg. Omitting the highest value, the mean is 8.4 dpm/kg and the spread is reduced to + 26% and - 20%. Production rate differences due to varying chemical compositions can be ruled out by measuring the ^{39}Ar content of the FeNi - phase alone.

^{39}Ar activity values of the FeNi - phase from 20 chondrites taken from the literature center around 22.5 dpm/kg with no deviations larger than + - 14 %. The production rate depends also on the unknown depth of the examined sample within that body. However, the ratio of two radionuclide concentrations, which are produced from the same target elements by energetically similar reactions, should be independent of size and depth. The most favorable radionuclide pair in our case is ^{39}Ar - ^{36}Cl in the FeNi - phase. Because of the much longer half-life of 3.1×10^5 years, the ^{36}Cl concentration can be assumed to still have its saturation value. Six chondrites compiled by Begemann and Vilcsek[1] show ^{39}Ar - ^{36}Cl ratios which scatter around 0.91+ 0.08 - 0.11, similar to the ratios found in iron meteorites: 0.96; the iron phase of stony iron meteorites: 1.09; and target measurements: 1.06; all the values are taken from a compilation by A. Cheng[2].

To get the most reliable terrestrial age of Benthullen, it was decided to measure the ^{39}Ar and ^{36}Cl content of the iron phase and ^{39}Ar activity of the whole rock.

In order to separate the relevant target elements--and the activities of ^{39}Ar and ^{36}Cl contained therein--as effectively as possible, the FeNi - phase was separated from the ground whole rock by repeated magnetic separation. A fractional dissolution technique described by Begemann and Vilcsek[1] was used. A 6.8 g iron phase separated from 80 g of the Benthullen meteorite was treated with a 10 % solution of $(NH_4)_2S_2O_8$ in 4 $\underline{N}$ H_2SO_4 in a vacuum dissolving apparatus. The released argon was swept out by flushing with He that had passed through

a charcoal trap at liquid nitrogen temperature to remove argon. ^{36}Ar and ^{37}Cl carriers were added to the reaction for determining the chemical yields. Isotopic enriched carriers were used to distinguish them at the end of the chemical separation from native Ar and Cl by mass spectrometry.

The helium and carrier argon entered the acid - solution through a frit and passed then through a condenser and an alumina desiccant to remove water vapor, then through a charcoal trap at liquid nitrogen temperature to trap the argon. Further purification of the trapped gas was done by releasing it from the charcoal at - 70° C and gettering by titanium at 950° C. The purified argon was transferred together with 10 % methane into a proportional counter of 0.5 cc volume of a design similar to that used by Davis et al[3]. Transportion of the gas was done by a Toepler pump.

After centrifuging and filtering the acid solution, the chlorine was precipitated as AgCl. The precipitation was recycled with ammonia and nitric acid. AgCl was collected on a quartz filter, which was placed into a quartz tube, and hydrogen was passed over the filter at 700° C. Hydrogen chloride was swept by the hydrogen gas into dilute ammonia. The ammonium chloride solution was evaporated to dryness in a test tube furnished with a grind joint. After adding diethyleneglycol as a solvent and methyl-para-toluenesulfonate, the test tube was attached to a vacuum line. At a He pressure of about 160 mm Hg, the synthesis of chloromethane was started by heating the solution to about 80° C. After the ammonium chloride had disappeared,

the heating bath was removed and the synthesised chloromethane was allowed to diffuse for about eight hours through a helium column with an ice acetone trap, and a trap at liquid nitrogen on the opposite end of the test tube. With the ice acetone column, impurities were trapped out, and in the liquid nitrogen trap, CH_3Cl was frozen out. The diffusion through the helium yielded a separation of the chloromethane from olefine impurities. The chloromethane was released at ice acetone temperature into the evacuated line and transferred with a Toepler pump into a proportional counter of the same kind used for the ^{39}Ar counting.

The insoluble residue of the dissolved iron phase was washed, dried, weighed and added to the stone phase (whole rock, iron phase removed). The argon was extracted from the stone phase by vacuum melting in a newly designed extraction apparatus. The apparatus consists of a molybdenum crucible lined inside with magnesia, a titanium sponge furnace, a removable charcoal trap, a sample tube, pirani, and ionisation gauges, and pump system. The Benthullen samples were individually wrapped in aluminum, predegassed at 150° C and then dropped into the crucible. Here, they were melted by radio frequency induction in the presence of ^{36}Ar carrier gas. After heating up the samples to 1600° C for about one hour, the charcoal was opened to trap all gases except He and Ne at - 176°C. For purification of the gas, the charcoal was transferred to the vacuum line described above, and the gas was released at dry ice temperature. The gas was now treated in the same way as the argon from the dissolving line.

The counting is done in a low level counting system which will be described below.

The measured ^{39}Ar activity of the stone phase combined with the iron phase activity gives the ^{39}Ar content of the whole rock.

Preliminary results of the iron phase ^{39}Ar activity seem to show that the meteorite Benthullen belongs to a recent fall and does not originate from the fall in 1368.

Table 1 shows counting results of the Benthullen FeNi-phase and a Bruderheim whole rock sample. The ^{39}Ar released from the Bruderheim sample by vacuum melting was used for a preliminary determination of the counting efficiency.

Meteorite	Bruderheim whole rock	Benthullen Fe-phase
sample weight (g)	14,0	5,72
extraction yield (%)	78,4	98,5
total counting rate (c/d)	86,0 ± 3,4	80,8 ± 3,1
background counting rate (c/d)	7,2 ± 0,8	7,5 ± 0,7
sample counting rate (c/d)	78,8 ± 3,5	73,3 ± 3,2
specific activity (dpm/kg)	10,4*	18,8 ± 0,9

*Mean value of 5 different measurements taken from literature.

The deduced ^{39}Ar activity of Benthullen corresponds to a terrestrial age of 75 ± 20 years. Taking into account the spread of the ^{39}Ar contents in the FeNi-phase of different chondrites increases the uncertainty to ± 60 years. The counting efficiency measurement will be repeated with a calibrated ^{39}Ar standard.

Low Level Counting System For Proportional Counter

Six low level proportional counters have been constructed according to the directions given by F. Kummer et al[4]. Two have an active volume of 0.2 cc and four 0.5 cc. The cathodes consist of zone refined iron to ensure very low radioactive contamination. The center wire is .00125 mm and .0025 mm tungsten respectively. The envelope is constructed from fused silica, optical grade, so as to avoid incorporating any potassium - 40 in the body of the counter. The end plugs are provided with a very thin window (0.04 mm) through which radiation of a ^{55}Fe source can be directed for purposes of energy calibration. The counters are either filled up to 2 atm. with P - 10 (90 % Ar, 10 % CH_4) or 0.5 - 1 atm. CH_3Cl.

For counting, the proportional counter tubes are put in 120° segment formed copper cases with removable lids (Figure 1). The preamplifier (Figure 2) is placed in front of the case supplied with high voltage, + 12 V and the signal output through the front plate. The counter and the preamplifier are separated by a copper plate which acts with the whole copper case as an electrostatic shield. The end of the counter is held in a hole through which the ^{55}Fe source can be put in and placed before the thin quartz window. The very short distance between the counter and the preamplifier helps to keep the noise level as low as possible. Three of the copper cases slide from each side into the inner tube of a mercury shield, so that up to six counters can be in operation at the same time. The shield arrangement was described by

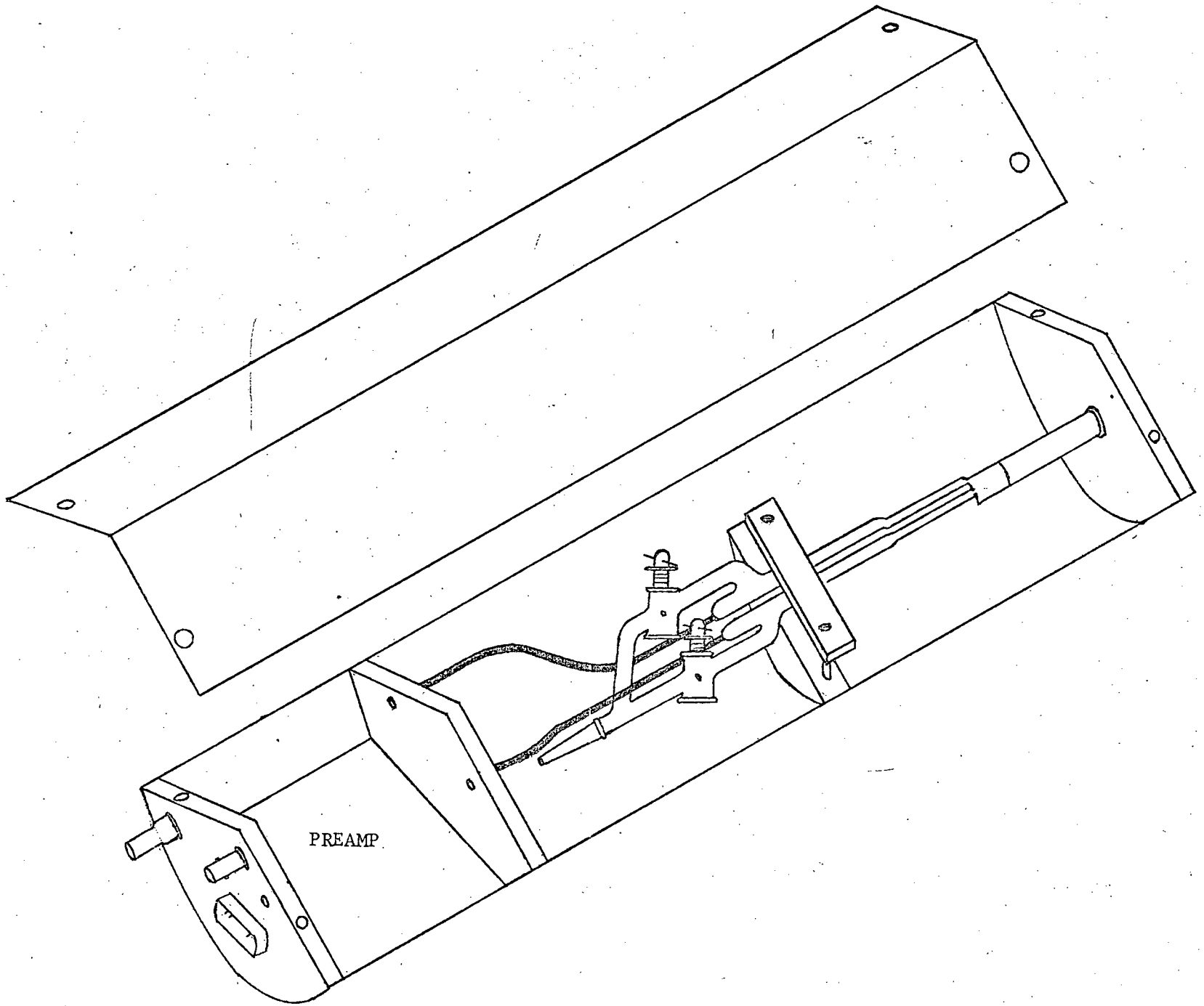

Figure 1: Proportional counter in electrostatic shield case with preamplifier integrated.

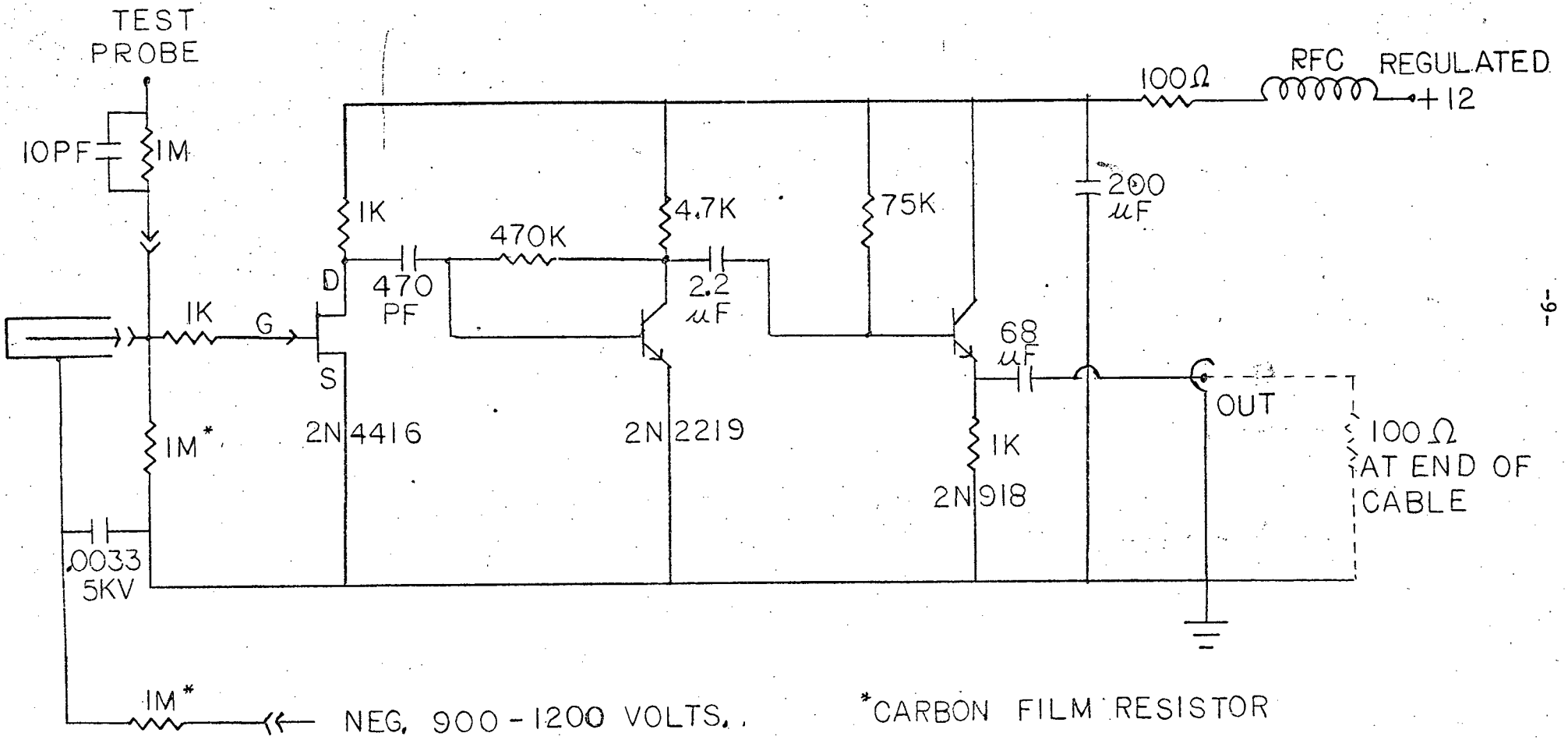

Figure 2: Low level charge preamplifier.

A. Cheng[2]. Figure 3 shows the block diagram of the electronics.

With the router of the pulse heigh analyser (PHA), signals of three different counters can be recorded at the same time. Each spectrum is stored in 128 channels. The stability of the whole counting system is checked twice a week with the ^{55}Fe source and a pulser. The peak location of the 5.9 KeV X-ray from ^{55}Fe on the pulse heigh analyser controls the counter and its associated electronics, and the pulser controls the electronics alone. Background counting rates of the individual counter range from 3 to 10 counts per day. The counting efficiency for ^{39}Ar is about 50%.

The main advantage of proportional counting as against Geiger counting is that electron captive radionuclides can be resolved from β - decaying isotopes; so that, for example, the activities of the very useful ^{37}Ar (T 1/2 = 35d) and ^{39}Ar isotopes can be measured at the same time.

The system was set up to measure $^{37,39}Ar$ and ^{36}Cl in meteorites, and $^{37,39}Ar$ in terrestrial surface material induced mainly by muon capture.

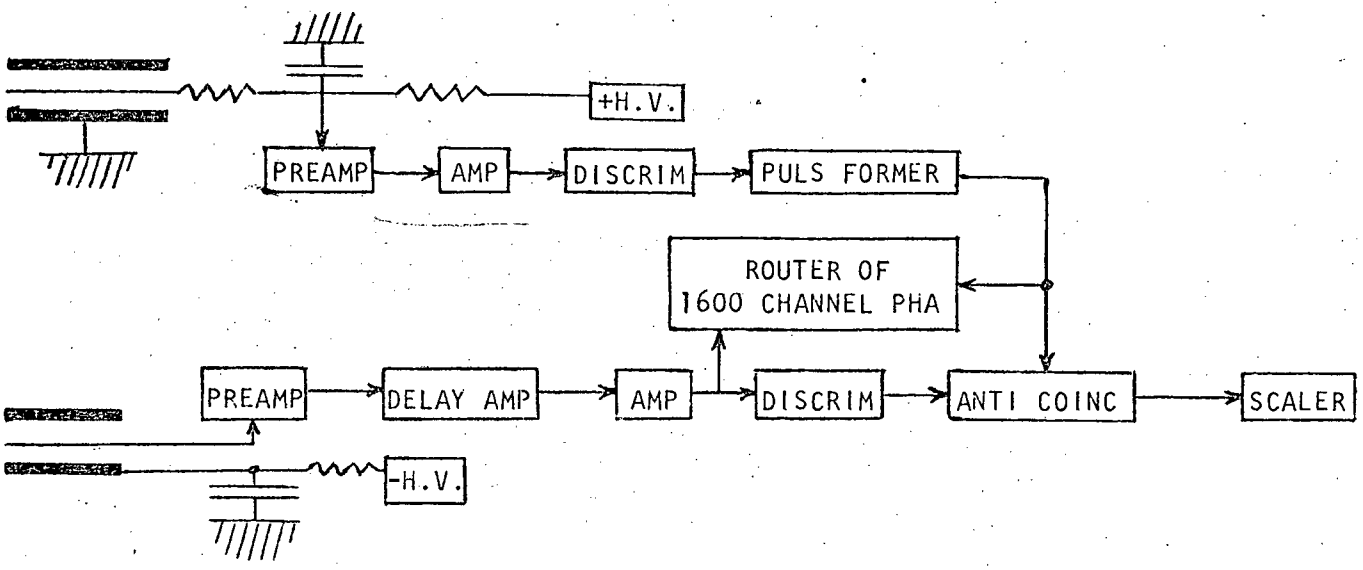

Fig. 3: Block diagram of electronics

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